

Optically polarized ^3He

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This article reviews the physics and technology of producing large quantities of highly spin-polarized ^3He nuclei using spin-exchange (SEOP) and metastability-exchange (MEOP) optical pumping. Both technical developments and deeper understanding of the physical processes involved have led to substantial improvements in the capabilities of both methods. For SEOP, the use of spectrally narrowed lasers and K-Rb mixtures has substantially increased the achievable polarization and polarizing rate. For MEOP nearly lossless compression allows for rapid production of polarized ^3He and operation in high magnetic fields has likewise significantly increased the pressure at which this method can be performed, and revealed new phenomena. Both methods have benefitted from development of storage methods that allow for spin-relaxation times of hundreds of hours, and specialized precision methods for polarimetry. SEOP and MEOP are now widely applied for spin-polarized targets, neutron spin filters, magnetic resonance imaging, and precision measurements.

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I. INTRODUCTION

A. Overview and road map

Gases of nuclear spin-polarized ^3He , highly or “hyper-polarized” to close to 100% in quantities on the order of a standard liter, have extensive scientific applications. As a target for nuclear and particle physics with charged particle and photon beams (see Sec. VI), polarized ^3He provides a reasonable approximation to a polarized free neutron target. Neutron spin filters (Sec. VII) can polarize neutron beams because of the large spin dependence of the cross section for the absorption of neutrons by ^3He . For hyperpolarized magnetic resonance imaging (MRI) (Sec. VIII), long relaxation times *in vivo* due to the chemical inertness of He and the absence of an electric quadrupole moment, along with a large magnetic moment, yield the highest resolution imaging of

human air spaces. Relaxation times of hundreds of hours (Sec. IV) make polarized ^3He extremely stable and sensitive for precision measurements (Sec. IX).

These applications are enabled by specialized optical pumping methods that polarize large volumes of ^3He nuclei to polarizations approaching unity. This review focuses on the significant developments in the theory, practice, and applications of these methods in the last two to three decades.

Because of the small hyperfine splitting in the ^3He $1s2p$ state and resulting long hyperfine mixing time relative to the short excited-state lifetime, it is not possible to directly optically pump ^3He to a useful polarization level using the $1s^2-1s2p$ transition, even if the required 58 nm radiation were conveniently available. Hence two indirect optical pumping methods, spin-exchange (SEOP) and metastability-exchange (MEOP), are employed to hyperpolarize ^3He nuclei.

In the SEOP method (Sec. II), electronic polarization produced in alkali-metal atoms by optical pumping at bar-scale pressures is slowly transferred to ^3He nuclei during collisions via the Fermi-contact hyperfine interaction between the alkali electron and the ^3He nucleus. The MEOP method (Sec. III) rapidly produces nuclear polarization in metastable ^3He atoms at mbar-scale pressures by a combination of optical pumping and hyperfine mixing. The metastable nuclear spin polarization is then rapidly transferred to the ground-state population via metastability-exchange collisions. Typically the gas is then compressed for use in applications.

Brief overviews of the two techniques are presented in Secs. I.B and I.C, with a comparison of their relative merits in Sec. I.D. Although limited by different issues, the polarizations achievable by each method for applications have remained roughly comparable, with the current achievable time-averaged values between 55% and 85%, depending on conditions. The polarizations and polarizing rates and thus the capabilities for applications have dramatically improved with the advent of new lasers and their continual progress in capability and convenience. In both methods, increased capabilities have led to deeper examinations of their respective physical limits.

For SEOP (Sec. II) the discovery of an unexpected ^3He relaxation mechanism proportional to the alkali-metal density has modified our view of the maximum attainable polarization, but the limit is still not completely clear. Motivated by the potential of lower alkali-metal spin-relaxation rates, studies of optical pumping with K and “hybrid” K-Rb mixtures have been performed with hybrid K-Rb emerging as the favored approach at this time. With the elucidation of the physical processes that increase the laser power demand, the sophistication of SEOP models has increased but they are still not fully mature and verified.

Major thrusts for MEOP (Sec. III) have been the parallel development of different compression approaches and extensions to high-pressure (~ 0.1 bar), high-field (~ 1 T) operation. Large-scale piston compressors are now used with mbar-scale MEOP to rapidly produce highly polarized gas at pressures of up to several bar. For MRI applications, compact inexpensive peristaltic pumps are being employed that exploit the reduced compression ratio requirements allowed by high-pressure, high-field MEOP. For MEOP itself, there have been advances in the theory of this complex process. Recent studies of the

maximum attainable polarization have revealed a light-induced relaxation mechanism that is particularly important at high pressures.

Spin relaxation remains a key issue for practical application of polarized ^3He (Sec. IV). Despite much technical progress, with relaxation times in the hundreds of hours now quite common, new unexplained aspects of wall relaxation have been observed and the subject remains poorly understood from a fundamental perspective. Extensive work has been done on the storage of polarized ^3He in glass containers of various types and with a variety of coatings. Magnetostatic cavities allow long ^3He relaxation times to be maintained even in the large magnetic-field gradients of neutron scattering experiments. At cryogenic temperatures cesium has been employed to inhibit wall relaxation and superfluid film flow.

Accurate absolute polarization metrology (Sec. V) methods are crucial for target and neutron spin-filter applications. Techniques now include water-calibrated NMR, neutron transmission, and magnetometry. Method-specific approaches include alkali-metal electron paramagnetic resonance (EPR) polarimetry for SEOP, and calibrated fluorescence and light absorption polarimetry for MEOP optical pumping cells.

All of these improvements have greatly enhanced applications of polarized ^3He . As an example, order-of-magnitude increases in the luminosity of polarized ^3He targets for nuclear and particle physics with charged particle and photon beams (Sec. VI) have enabled high precision studies of topics such as nucleon electric and magnetic form factors, spin structure functions, and three-body nuclear physics.

The large spin dependence of the cross section for absorption of neutrons by ^3He allows polarization of neutron beams (Sec. VII). Such “neutron spin filters” (NSFs) are being developed worldwide and applications to neutron scattering are growing rapidly. Examples of topics and materials under study include magnetic ordering, magnetic multilayers, and magnetic nanoparticles. Applications of NSFs to fundamental neutron physics include studies of neutron beta decay, measurements of parity-violating asymmetries, and measurements of the spin dependence of the neutron- ^3He scattering length.

A variety of polarized ^3He apparatuses are being employed for medical studies of human-lung airways (Sec. VIII). Although the range of imaging studies with ^{129}Xe is larger because it dissolves in water and fat, the larger magnetic moment and generally higher attainable polarizations for ^3He generally makes it the preferred choice for the highest resolution lung images.

Precision spectroscopy on ^3He (Sec. IX) has resulted in, for example, new magnetometry methods, searches for violations of fundamental symmetries, and searches for interactions mediated by axionlike particles.

^3He has been used for other applications that we will not consider further in this review. Newbury *et al.* (1991) studied polarized muonic He by capturing muons on polarized ^3He . The macroscopic behavior of spin-polarized ^3He fluids is modified at low temperature by polarization of the ^3He nuclei (Castaing and Nozieres, 1979; Lhuillier and Laloë, 1979, 1982a, 1982b; Owers-Bradley, 1997). Nonlinear spin dynamics (Akimoto *et al.*, 2000; Desvaux, 2013) and NMR time reversal (Baudin *et al.*, 2008) have been studied in ^3He - ^4He mixtures.

We conclude the review by discussing future trends in Sec. X.

B. Overview of SEOP physics and apparatus

In the SEOP method, electronic polarization produced in alkali-metal atoms by optical pumping is slowly transferred to ^3He nuclei during collisions via the Fermi-contact hyperfine interaction between the alkali electron and the ^3He nucleus (Bouchiat, Carver, and Varnum, 1960; Walker and Happer, 1997). ^3He gas at typical pressures¹ between 1 and 10 bar is contained in sealed glass cells along with on the order of 0.1 g of alkali metal, usually rubidium or a rubidium-potassium mixture. Pure Rb or K-Rb cells are typically heated to 170 or 220 °C, respectively, to establish an alkali-metal density around $3 \times 10^{14} \text{ cm}^{-3}$. Because of the short diffusion length of the alkali-metal atoms, the entire volume of the cell must be immersed in circularly polarized laser light, typically provided by diode laser arrays with output power on the order of 100 W, an air wavelength of 794.7 nm, and a typical linewidth of 0.25 nm. A number of factors such as absorption by optically thick alkali-metal vapor, focusing and/or distortion of the laser light by nonuniform blown glass cells, and in some cases poor spatial mode quality make it difficult to realize the goal of a uniform optical pumping rate at all points in the cell, which can be partially addressed by pumping from opposing directions (Chann *et al.*, 2003; Chen, Gentile, Erwin *et al.*, 2014). The absorption width of the Rb vapor is determined by pressure broadening of 0.040 nm/amg (Romalis, Miron, and Cates, 1997; Kluttz, Averett, and Wolin, 2013), so the atomic pressure-broadened width is comparable to the laser linewidth for high density² (10 amg) targets but substantially smaller than the laser linewidth for typical neutron spin filters (1.5 amg). The degree of the circular polarization (99% or better is usually attained with commercial wave plates) is not highly critical because the relatively high alkali-metal density strongly absorbs the undesired photon spin state (Bhaskar *et al.*, 1979; Chann *et al.*, 2002b). To suppress radiation trapping from radiative decay of the excited alkali-metal atoms, on the order of 0.1 amg of nitrogen gas is added to provide rapid collisional deexcitation (Walker and Happer, 1997; Lancor and Walker, 2010).

The alkali-metal polarization P_A is determined by the ratio of the electronic spin-relaxation rate to the optical pumping rate. Rubidium spin-relaxation rates are typically a few hundred s^{-1} whereas optical pumping rates are much higher, hence P_A near unity is established on a very short time scale. The temperature must be maintained at a low enough value so that absorption of the laser light by the optically thick alkali-metal vapor does not yield too low an optical pumping rate in the interior of the cell. Alkali-metal spin relaxation results primarily from alkali-alkali collisions (dominant at low ^3He gas pressures) and alkali- ^3He collisions (dominant for high ^3He gas pressures), with some contribution from alkali-nitrogen collisions (Ben-Amar Baranga *et al.*, 1998). For the same spin-exchange rate,

¹All pressures refer to room temperature unless otherwise noted.

²1 amgat (amg) = $2.69 \times 10^{19} \text{ cm}^{-3}$ is the density of an ideal gas at standard temperature and pressure.

the spin relaxation for potassium is typically about 5 times lower than that of rubidium; hence the use of K-Rb mixtures or pure K increases the efficiency of SEOP (Babcock *et al.*, 2003). In practice K-Rb mixtures are typically employed because of greater laser availability at 795 nm for Rb pumping as compared to K pumping at 770 nm (Chen, Gentile *et al.*, 2007).

The ^3He polarization is determined by the ratio of ^3He nuclear spin relaxation to the spin-exchange rate. The spin-exchange rate is typically on the order of 0.1 h^{-1} , hence a day is required to approach the maximum polarization. This slow time scale makes long ^3He relaxation times critical. Sealed cells made from fully blown aluminosilicate glass are typically employed for charged particle and photon scattering and neutron spin filters, whereas borosilicate glass and/or open systems are more common for polarized gas MRI and other applications. Aluminosilicate glass has low ^3He permeability and is alkali-metal resistant, and for neutron applications GE180³ is particularly desirable because it is boron free. The use of fully blown glass (Newbury *et al.*, 1993; Chen *et al.*, 2011) and the presence of the alkali metal (Heil *et al.*, 1995) are both important for achieving the longest relaxation times. ^3He cells with room temperature relaxation times on the order of 100 h or longer (Chen *et al.*, 2011) ostensibly make wall relaxation a minor contributor to limiting ^3He polarization, but a strongly temperature-dependent relaxation mechanism has been found to limit the achievable ^3He polarization (Babcock *et al.*, 2006). For poorer cells, short wall-relaxation times can in principle be overcome by increasing the alkali density (and using hybrid pumping) and thus increasing the spin-exchange rate, but this is possible only to the extent that sufficient laser power is available.

A typical SEOP apparatus (Fig. 1) consists of a nonmagnetic oven to heat a ^3He cell, a uniform magnetic field provided by Helmholtz coils, and a spectrally narrowed high-power diode array laser with suitable optics for producing circular polarization and focusing and steering the laser beam. Hot air is typically used for heating but nonmagnetic electrical heating has also been employed (Tong *et al.*, 2010; Babcock *et al.*, 2016). Although Helmholtz coils are the most common, four-coil systems, compensated solenoids, and magnetostatic cavities have also been used. Diode laser bars are spectrally narrowed with diffraction gratings (Babcock, Chann *et al.*, 2005) or volume holographic gratings (VHG) (Nikolaou *et al.*, 2013; Chen, Gentile, Ye *et al.*, 2014; Liu *et al.*, 2015). ^3He polarization is monitored using either adiabatic fast passage (AFP) or free induction decay (FID) NMR. AFP typically requires a pair of drive coils large enough to immerse the cell in a reasonably uniform radio-frequency (rf) field and pickup coils to detect the precessing magnetization. FID can also be performed in this scheme but more often a small surface coil that both transmits a short rf pulse and detects the small

³GE Lighting Component Sales, Cleveland, OH. Certain trade names and company products are mentioned in the text or identified in an illustration in order to adequately specify the experimental procedure and equipment used. In no case does such identification imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the products are necessarily the best available for the purpose.

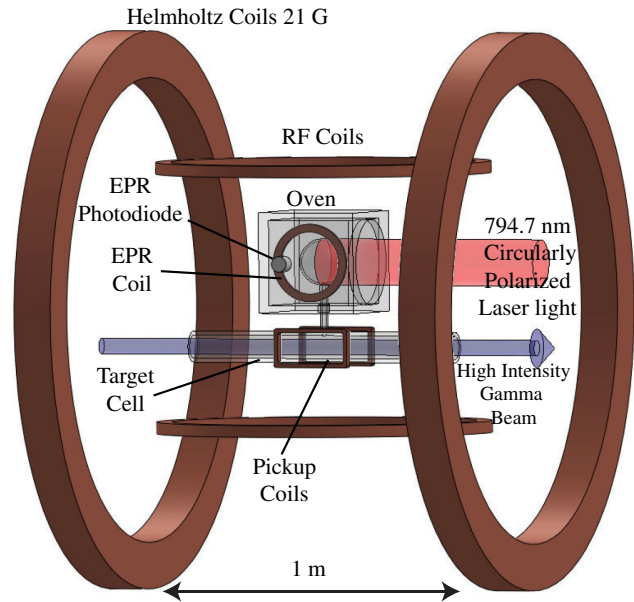


FIG. 1. An example of an apparatus for spin-exchange optical pumping (SEOP). This apparatus has been employed for photon scattering experiments. Whereas SEOP is typically performed in single cells for neutron spin-filter and magnetic resonance imaging applications, both electron and photon scattering applications employ a double cell configuration in which a target cell (TC) is linked to an optical pumping cell (OPC) through a connecting tube. Electron paramagnetic resonance and nuclear magnetic resonance are employed to measure the ^3He polarization in the OPC and TC, respectively. Adapted from Ye *et al.*, 2010.

precessing transverse magnetization is employed. SEOP apparatuses may also employ other diagnostics such as EPR (Romalis and Cates, 1998), Faraday rotation (Vliegen *et al.*, 2001), and transmission spectrum monitoring with a small diffraction grating spectrometer. Other characteristics of SEOP apparatuses are discussed under individual applications.

C. Overview of MEOP physics and apparatus

In the MEOP method, nuclear polarization is produced in metastable ^3He atoms by a combination of optical pumping and hyperfine mixing, and then rapidly transferred to the ground-state population via metastability-exchange collisions (Colegrove, Scheerer, and Walters, 1963; Batz, Nacher, and Tastevin, 2011). Traditional low-field MEOP is performed in pure ^3He or ^3He - ^4He mixtures (Stoltz, Meyerhoff *et al.*, 1996) at pressures on the order of 1 mbar, whereas high-field MEOP has been performed at pressures up to a few hundred millibar (Nikiel-Osuchowska *et al.*, 2013). An electrical discharge produced by external electrodes is employed to produce metastable densities on the order of 10^{10} cm^{-3} . Low gas-phase impurity levels are required because metastable atoms have 20 eV of energy, which is sufficient to ionize most common impurities, resulting in destruction of the metastable atoms. In practice, highly pure source gas and baked and discharge-cleaned glass walls are required to achieve an adequately high metastable density. A hand-held spectrometer provides a simple and convenient method to evaluate the

purity of the ^3He gas, since the light emitted from a MEOP cell should not show broadband background and/or impurity emission lines.

Because of the relatively low metastable density the gas is optically thin, hence long cells can be employed to absorb more laser light and thus yield larger quantities of hyper-polarized gas. However, due to the weak absorption, the degree of circular polarization is more critical than for SEOP. The spatial profile of the light somewhat underfills the pumping cell, approximately matching the metastable density profile that vanishes at the cell walls. Radiation trapping is not a major issue since the diameter of the cells is typically less than ≈ 7 cm to avoid this consideration. Optical pumping is typically performed with Yb fiber lasers with output power on the order of 10 W, an air wavelength of 1083 nm, and a typical linewidth of 2 GHz to match the Doppler-broadened absorption width. The polarizations of the ground state and metastable populations are strongly coupled (Nacher and Leduc, 1985) and evolve together on typical time scales of seconds to minutes, depending on the cell size and laser power (Gentile and McKeown, 1993). The ^3He ground-state relaxation time is generally dominated by the discharge and ranges from a fraction of a minute to several minutes. Stronger discharges yield higher metastable densities and thus higher polarizing rates, but also increased relaxation. In many applications the polarized gas is compressed to obtain pressures of between 1 and 4 bar at polarizing rates of a few amg-L/h, with a typical time scale of a few hours or less to fill a cell (Batz *et al.*, 2005; Lelievre-Berna, 2007). Storage cells are typically made of fused quartz or aluminosilicate glass and coated with alkali metal (usually cesium) to reduce relaxation (Heil *et al.*, 1995).

A typical MEOP apparatus (Fig. 2) consists of a radio-frequency discharge, a uniform magnetic field provided by a set of coils or solenoid, and a Yb fiber laser with suitable optics for producing circular polarization and focusing and steering the laser beam (Andersen *et al.*, 2005). Although Helmholtz coils are common, multiple-coil systems are also

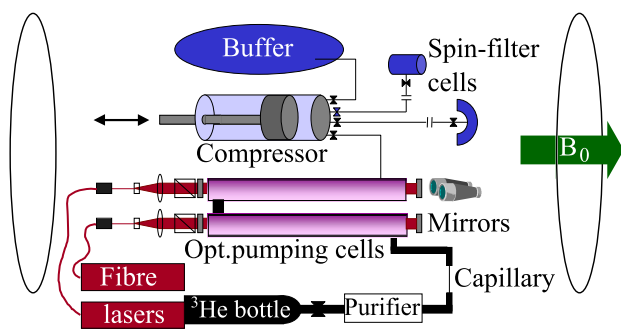


FIG. 2. An example of an apparatus for metastability-exchange optical pumping. This apparatus has been employed for compressing gas into neutron spin-filter cells. Nine 2 m diameter coils provide the uniform magnetic field for the 2.3 m long OPCs. ^3He gas is purified, polarized in the OPCs, compressed in two stages with an intermediate buffer cell, and dispensed into detachable neutron spin-filter cells. The capillary serves to control the flow rate and restrict diffusion for the typical 1 mbar pressure in the OPCs. Adapted from Andersen *et al.*, 2005.

used, in particular, for large compression apparatuses. In the optical pumping cell, ^3He polarization is monitored by measuring the degree of circular polarization from the 668 nm ^3He emission line or absorption of a probe laser at the pumping wavelength. Compression apparatuses range from large-scale piston compressors for a range of applications to small-scale peristaltic pumps typically employed for polarized gas MRI (Nacher *et al.*, 1999; Nikiel *et al.*, 2007). In these small-scale systems high-field pumping in the bore of the MRI magnet can be employed. After compression, either AFP or FID NMR is used. Other characteristics of MEOP apparatuses are discussed under individual applications.

D. SEOP and MEOP compare and contrast

Although SEOP and MEOP address the problem of polarizing ^3He nuclei quite differently, the needs of applications are met by both methods, with comparable practical results. Figure 3 shows representative performance of the two methods as the advances discussed in this review occurred.

The key feature of SEOP is the ability to polarize ^3He directly at a wide range of pressures (typically between 0.5 and 13 bar), which is required for most applications. In contrast MEOP is typically performed at pressures of the order of 1 mbar, thus requiring polarization preserving compression. The key feature of MEOP is the ability to produce polarized ^3He at rates of a few amg-L/h (Batz *et al.*, 2005), typically an order of magnitude faster than most SEOP apparatuses. Most applications have been approached by both methods, but in some cases one may be preferred, e.g., dual species masers, in which both ^3He and ^{129}Xe can be simultaneously polarized by SEOP, and internal targets, for which the low operating pressure and high polarizing rate are well matched to MEOP.

A comparison of the key parameters of each method reveals how their practical performance is comparable. MEOP starts off with a rate constant nearly 9 orders of magnitude higher than SEOP, primarily because of the inherent weakness of the spin-exchange cross section between alkali-metal atoms and ^3He nuclei as compared to the nearly gas-kinetic cross section for metastability exchange. However, this advantage is dramatically reduced because the typical alkali-metal density is about 4 orders of magnitude higher than the typical metastable density thus resulting in a difference of nearly 5 orders of magnitude in the polarizing time constant, e.g., 15 h for pure Rb SEOP and 1 s for low-field MEOP. SEOP is performed at pressures 10^3 to 10^4 times higher than low-field MEOP; hence overall the polarizing rate for MEOP is about an order of magnitude faster than SEOP (see Fig. 3). The operational approach of each method reflects exploitation of their respective strengths: SEOP is slow but compact and can operate unattended, hence one operates continuously on charged particle beam lines and/or makes use of overnight operation with multiple polarizing stations to remotely polarize cells for neutron beam lines. MEOP is fast but compression requires greater attention and the speed is maximized with large optical pumping volumes, hence one operates with replaceable cells that are rapidly filled with polarized gas by a remote compressor.

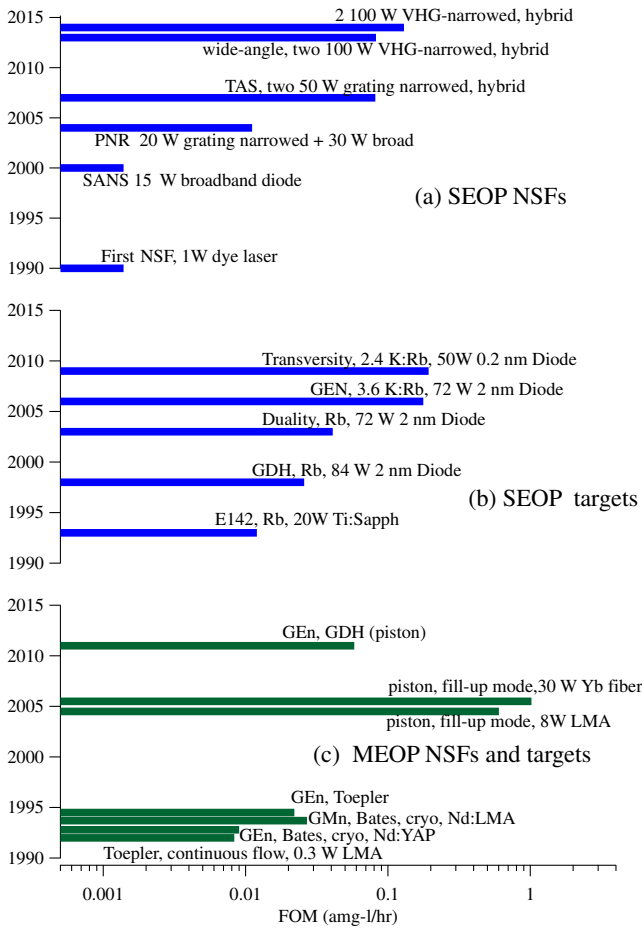


FIG. 3. Development of NSFs and spin-polarized ^3He targets, illustrated by representative devices and experiments. In each case, key parameters are listed to show how advances for each method improved performance, e.g., spectrally narrowed lasers and K-Rb mixtures for SEOP, and filling of cells with piston compressors and high-power laser sources for MEOP. See Secs. VI and VII for definitions of identifiers. The figure of merit (FOM) is defined to be $P_{\text{He}}^2 N / \tau$ for SEOP NSFs and targets as well as MEOP cryogenic targets, where P_{He} is the ^3He polarization, N is the total number of atoms in the cell in units of amg-L, and τ is the time constant for polarizing the cell. (Reminder: to reach 90% of the maximum polarization requires 2.3 time constants.) For compression-based MEOP NSFs and targets, τ is replaced by T , where T is the time to refresh the gas for continuous flow or the time between cell exchange for remotely operated compression. (a) SEOP NSFs: First NSF (Coulter *et al.*, 1990), SANS (Gentile *et al.*, 2000; Jones *et al.*, 2000), PNR (Chen *et al.*, 2004), TAS (Chen, Armstrong *et al.*, 2007), wide angle (Ye *et al.*, 2013), and VHG narrowed (Chen, Gentile, Erwin *et al.*, 2014). (b) SEOP targets for electron-scattering experiments. Here the FOM is equivalent to the potential effective luminosity of Singh *et al.* (2015). Data provided by J. Singh. E142 (Anthony *et al.*, 1993), GDH (Amarian *et al.*, 2002), GEN (Riordan *et al.*, 2010), duality (Solvignon *et al.*, 2008), and transversity (Qian *et al.*, 2011). (c) MEOP NSFs and targets: GEN (Bates, cryogenic, Nd:YAP) (Jones *et al.*, 1993), GMn (Bates, cryogenic, Nd:LMA) (Gao *et al.*, 1994), GEN (Toepler pump) (Meyerhoff *et al.*, 1994), GEN, GDH (piston) (Krimmer *et al.*, 2009, 2011; Schlimme *et al.*, 2013), (Toepler pump) (Eckert *et al.*, 1992), and piston fill-up mode (Batz *et al.*, 2005).

In recent years the limitations of each method are being addressed by new methods. More compact piston compression apparatuses have been developed (Beecham *et al.*, 2011; Mrozik *et al.*, 2011; Kraft *et al.*, 2014) and for MRI applications MEOP has been extended to optical pumping pressures up to hundreds of millibar at high magnetic fields (Nikiel-Osuchowska *et al.*, 2013). The use of K-Rb mixtures has increased the polarizing rate for SEOP (Chen, Gentile *et al.*, 2007; Singh *et al.*, 2015) and large-scale production of polarized ^3He via SEOP is under development (Hersman *et al.*, 2013).

II. SPIN-EXCHANGE OPTICAL PUMPING

Spin-exchange optical pumping of ^3He was pioneered by Bouchiat, Carver, and Varnum (1960) using lamps. With some exceptions (Gamblin and Carver, 1965; Fitzsimmons, Tankersley, and Walters, 1969; Grover, 1978) the topic was largely ignored until the proposal and initial demonstration of Chupp *et al.* (1987) for producing high density polarized targets using SEOP with tunable lasers. Subsequent experimental developments are described in detail throughout this review. Basic descriptions of SEOP physics (Chupp *et al.*, 1987; Wagshul and Chupp, 1994; Walker and Happer, 1997) have been presented previously, as well as much more detailed discussions (Appelt *et al.*, 1998; Happer, Jau, and Walker, 2009). In this section we present the current understanding of SEOP physics, with particular emphasis on new understanding obtained since the RMP colloquium of Walker and Happer (1997).

A. Spin-exchange collisions

Spin-exchange optical pumping of ^3He transfers spin polarization between alkali-metal electron spins (A) and ^3He nuclear spins during binary collisions:

$$A \uparrow + {}^3\text{He} \downarrow \leftrightarrow A \downarrow + {}^3\text{He} \uparrow \quad (1)$$

The dominant interactions experienced during A-He collisions are

$$V(\xi) = V_0(\xi) + \alpha(\xi) \mathbf{S} \cdot \mathbf{I}_{\text{He}} + \gamma(\xi) \mathbf{N} \cdot \mathbf{S}, \quad (2)$$

where $V_0(\xi)$ is the spin-independent interaction potential between the two atoms separated by a distance ξ and $\alpha(\xi)$ is the strength of the Fermi-contact hyperfine interaction between the alkali electron spin \mathbf{S} and the ^3He nuclear spin \mathbf{I}_{He} . Recent theoretical calculations (Partridge, Stallcop, and Levin, 2001; Tscherbul *et al.*, 2011) for K- ^3He are shown in Fig. 4. The extremely weak attractive portion of V_0 , supporting at most one bound state (Kleinekathöfer, Lewerenz, and Mladenović, 1999), is imperceptible on the scale of typical thermal collision energies so that the dominant spin transfer occurs at the inner turning points of binary collisions. The spin-rotation interaction $\gamma(\xi) \mathbf{N} \cdot \mathbf{S}$, which couples the electron spin to the rotational angular momentum \mathbf{N} of the alkali-metal-He pair, is a major source of angular momentum loss in the system and governs the maximum possible efficiency with which spin exchange can occur.

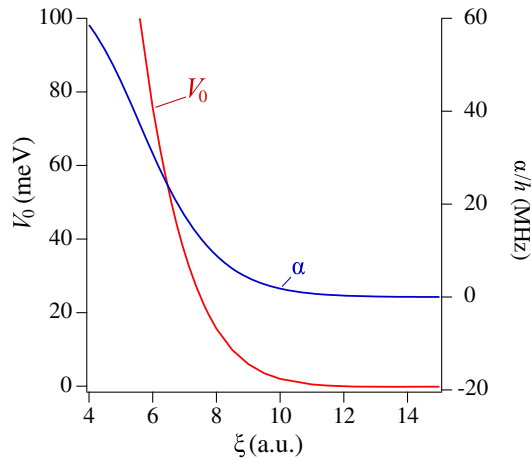


FIG. 4. Calculated spin-independent ($V_0(\xi)$) (Partridge, Stallcop, and Levin, 2001) and Fermi-contact [$\alpha(\xi)$] (Tscherbil *et al.*, 2011) potentials for $\text{K-}^3\text{He}$ molecules, as a function of interatomic separation ξ in atomic units.

The curves of Fig. 4 can be used with time-dependent perturbation theory to estimate the spin-exchange rate coefficient $k_{\text{SE}} \approx v\sigma(\alpha\tau/\hbar)^2$ to be within a factor of 2 or so of the measured spin-exchange rate coefficients, around $6 \times 10^{-20} \text{ cm}^3/\text{s}$. Here $v\sigma$ is the gas-kinetic rate coefficient and τ the collision time. The probability of spin exchange during a single collision $(\alpha\tau/\hbar)^2$ is on the order of 10^{-10} . With alkali-metal densities $[A]$ in the range of 10^{14} – 10^{15} cm^{-3} , this implies a spin-exchange time constant $(k_{\text{SE}}[A])^{-1}$ of 5–50 h. These numbers illustrate why the technical development of storage containers with ^3He lifetimes of hundreds of hours (Sec. IV) is vital to attaining polarizations that approach unity.

The ^3He polarization dynamics due to spin exchange and other sources of relaxation (rate Γ_w , usually dominated by wall collisions) can under most conditions be accurately modeled as

$$\frac{d}{dt}P_{\text{He}} = k_{\text{SE}}[A](P_{\text{A}} - P_{\text{He}}) - \Gamma_w P_{\text{He}}, \quad (3)$$

where the alkali-metal electron polarization P_{A} , due to its subsecond relaxation times, is essentially constant compared to the hour-scale variations of P_{He} . Thus the noble-gas polarization builds up to a steady-state value

$$P_{\text{He}\infty} = P_{\text{A}} \frac{k_{\text{SE}}[A]}{k_{\text{SE}}[A] + \Gamma_w}, \quad (4)$$

with a time constant τ obeying

$$\frac{1}{\tau} = k_{\text{SE}}[A] + \Gamma_w. \quad (5)$$

This latter relation seems to imply a simple method for measuring k_{SE} , namely, measure the time constant as a function of the alkali density $[A]$. This method does not work, however, since for reasons not yet understood the wall-relaxation rate Γ_w in spin-exchange cells is observed to

increase rapidly with temperature. This issue is discussed in detail in Sec. II.E.2.

1. Measurements of spin-exchange collision rates

Several wall-independent methods have been used to determine k_{SE} for different species. The repolarization method (Ben-Amar Baranga *et al.*, 1998; Chann *et al.*, 2002a) measures the alkali polarization produced by spin exchange in the absence of optical pumping,

$$P_{\text{A}}^{\text{re}} = k_{\text{SE}}[\text{He}]P_{\text{He}}/\Gamma_{\text{A}}, \quad (6)$$

where Γ_{A} is the measured alkali spin-relaxation rate. The rate balance method (Chann *et al.*, 2002a) measures $P_{\text{He}\infty}$, τ , P_{A} , and the alkali density to deduce k_{SE} from Eq. (4). A combination of these two methods, measuring the time rate of change of the repolarization signal, was used by Borel *et al.* (2003). For alkali atoms with small Γ_{A} , the spin-exchange rate can also be deduced by measuring the difference between Γ_{A} for ^3He and for ^4He , with a correction for the reduced-mass scaling of the spin-relaxation contribution (Walker, Nelson, and Kadlecck, 2010). Most recently, Singh *et al.* (2015) used absolute alkali polarimetry and density measurements, combined with the initial slope from Eq. (3), to infer k_{SE} . Table I shows the status of wall-independent spin-exchange rate coefficient measurements for the various alkali-metal atoms. For potassium, the recent result of Singh *et al.* (2015) is 30% higher than the weighted average of three prior measurements. They speculated that this difference may be due to their operation at substantially higher temperature, but the origin of the disagreement has not been established.

TABLE I. Spin-exchange rate coefficient (k_{SE}) measurements, in units of $10^{-20} \text{ cm}^3/\text{s}$, using wall-independent methods, with EPR frequency shift enhancement factors κ_0 and $d\kappa_0/dT$ (units K^{-1} ; see Sec. II.F). Throughout this review, numbers in parentheses represent the uncertainty in the last digit(s).

	Na	K	Rb
k_{SE}	6.1(6) ^b	6.1(4) ^a 5.5(2) ^c 6.1(7) ^e 7.5(5) ^g	6.7(6) ^d 6.8(2) ^f
κ_0	4.72(09) ^h	6.01(11) ⁱ	6.15(09) ^j
$d\kappa_0/dT$	0.009 14(56) ^k	0.0086(20) ^l	0.009 34(14) ^m 0.009 16(26) ⁿ

^aBabcock *et al.* (2003).

^bBorel *et al.* (2003).

^cBabcock (2005).

^dBen-Amar Baranga *et al.* (1998).

^eWalker, Nelson, and Kadlecck (2010).

^fChann *et al.* (2002a).

^gSingh *et al.* (2015).

^hBabcock, Nelson *et al.* (2005), 200 °C.

ⁱBabcock, Nelson *et al.* (2005), 200 °C.

^jRomalis and Cates (1998), 175 °C.

^kBabcock, Nelson *et al.* (2005), 210–350 °C.

^lBabcock, Nelson *et al.* (2005), 150–220 °C.

^mRomalis and Cates (1998), 110–172 °C.

ⁿBabcock, Nelson *et al.* (2005), 170–350 °C.

2. Spin-exchange efficiency

Despite the very slow time constants associated with spin-exchange pumping of ^3He , the efficiency of the transfer of angular momentum from the photons in the optical pumping light to the nuclei can, under ideal conditions, be surprisingly high. If fully polarized alkali-metal atoms are assumed not to scatter optical pumping light (an assumption to be examined in Sec. II.B.3), the loss of angular momentum by $[A]V$ polarized alkali-metal atoms in volume V occurs at a rate $[A]V\Gamma_A P_A$. Comparing this to the rate $V[\text{He}]dP_{\text{He}}/dt$ at which angular momentum is added to the noble-gas nuclei, gives the collisional efficiency

$$\eta(P_{\text{He}}) = \frac{V[\text{He}]dP_{\text{He}}/dt}{[A]V\Gamma_A P_A} = \frac{k_{\text{SE}}[\text{He}]}{\Gamma_A} \left(1 - \frac{P_{\text{He}}}{P_{\text{He}\infty}}\right). \quad (7)$$

The collisional efficiency is maximum at low He polarizations, then decreases at higher polarizations as significant amounts of angular momentum are returned to the alkali-metal atoms via spin-exchange collisions from the polarized He nuclei. Direct measurements of the efficiency (Ben-Amar Baranga *et al.*, 1998) were made using Eq. (6) rewritten as $\eta(0) = P_A^{\text{re}}/P_{\text{He}}$. Variations from atom to atom in Γ_A are the primary determining factor in the efficiency.

There are many contributing processes to the alkali spin-relaxation rate Γ_A (Sec. II.B.2), but at a given $[\text{He}]$ the minimum relaxation rate is $\Gamma_A = (k_{\text{SR}} + k_{\text{SE}})[\text{He}]$, where k_{SR} is the rate coefficient for relaxation due to the spin-rotation interaction. Thus when relaxation due to He atoms dominates the alkali-metal spin relaxation, the spin-exchange limited efficiency is

$$\eta_{\text{SE}} = \frac{k_{\text{SE}}}{k_{\text{SE}} + k_{\text{SR}}}. \quad (8)$$

Measurements of the spin-exchange efficiency for Rb and K (Ben-Amar Baranga *et al.*, 1998), shown in Fig. 5, obtained $\eta_{\text{SE}} \sim 1/50$ and $1/6$ under typical conditions. For Na the spin-exchange efficiency is consistent with 1 (Borel *et al.*, 2003). The unmeasured Cs spin-exchange efficiency is predicted to be about 7 times lower than Rb using estimates from Happer, Jau, and Walker (2009).

B. Optical pumping

In order to produce substantial quantities of highly polarized ^3He by SEOP, it is necessary to spin polarize large volumes of high density alkali-metal vapor interacting with ^3He gas at densities ranging from 0.5 amg for neutron spin filters to 10 amg for targets. The optical pumping is typically done using 100 W scale lasers whose spectral linewidths are comparable to or broader than the atomic lines. These extreme conditions raise a number of issues that are typically not encountered in other optical pumping contexts.⁴

⁴Recent treatments of optical pumping include Happer, Jau, and Walker (2009) and Auzinsh, Budker, and Rochester (2010). The classic review of the subject is Happer (1972), and a good introduction is Happer and Van Wijngaarden (1987).

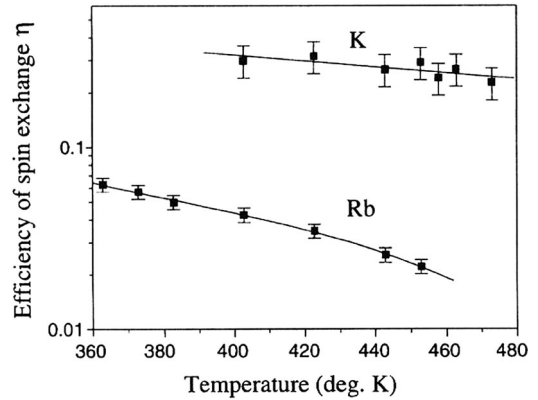


FIG. 5. Spin-exchange efficiency (η_{SE}) measurement for K and Rb. From Ben-Amar Baranga *et al.*, 1998.

The breakdown of free-atom light selection rules, light propagation and spectral evolution in optically thick conditions, and dissipation of heat are examples of issues that are key to understanding the SEOP process. These and other effects are discussed in this section.

1. High-pressure optical pumping

Figure 6 shows the energy levels and photon absorption rates relevant for optical pumping of alkali-metal atoms in the $S_{1/2}$ ground state using circularly polarized light tuned to the $P_{1/2}$ “D1” resonance. In the presence of He atoms, there is collisional mixing of the $P_{1/2}$ and $P_{3/2}$ levels, so that the $P_{1/2}$ level acquires some $P_{3/2}$ character, indicated by the dashed Zeeman sublevels in the figure. The relative absorption probability, parametrized by P_∞ , for atoms in the $m_S = -1/2$ Zeeman sublevel is $1 + P_\infty$, much greater than the relative absorption probability $1 - P_\infty$ for those in the $m_S = 1/2$ sublevel. Thus atoms in the $-1/2$ sublevel are selectively

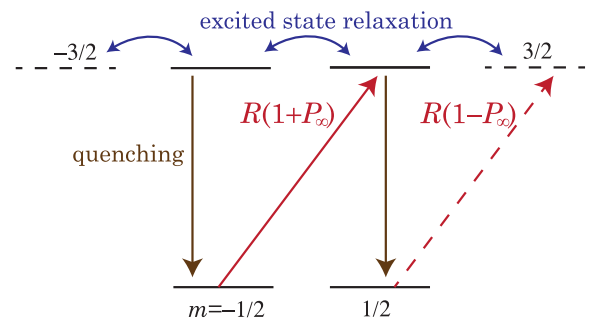


FIG. 6. Key elements of the optical pumping cycle for light tuned to the alkali-metal $S_{1/2}$ - $P_{1/2}$ resonance in the presence of high-pressure He gas. Collisions with the He atoms mix the $P_{1/2}$ and $P_{3/2}$ levels, altering the selection rules for light absorption. Atoms in the $m_S = \pm 1/2$ Zeeman ground-state sublevels absorb photons with relative probabilities $1 \mp P_\infty$. Once excited, rapid collisional spin-relaxation occurs. Quenching collisions with N_2 molecules randomly repopulates the ground-state sublevels. The atoms accumulate in the $m_S = 1/2$ sublevel, acquiring a steady-state spin polarization of P_∞ in the absence of ground-state spin relaxation.

excited by the light. The excited atoms experience rapid spin relaxation in collisions with the He and N_2 buffer gases, randomizing populations among the $P_{1/2}$ and, to a significant extent, $P_{3/2}$ sublevels. Quenching collisions with N_2 molecules then resonantly transfer the P-state energy to excited N_2 vibrational levels, returning the alkali atoms to the ground $S_{1/2}$ state. Atoms that return to the $m_S = 1/2$ state only rarely absorb the polarized light, while those that return to $m_S = -1/2$ will be efficiently reexcited. In this manner atoms will preferentially populate the $m_S = 1/2$ state, reaching a steady-state population of P_∞ (B. Lancor *et al.*, 2010).

Rapid quenching of the excited atoms by N_2 molecules plays an essential role in this process (Lancor and Walker, 2010). Since SEOP typically takes place in extremely optically thick cells, the reabsorption probability for photons emitted by the excited alkali atoms can be considered to be nearly unity. Since such photons constitute an essentially unpolarized background of resonant light, they act as a relaxation mechanism and decrease the efficiency of optical pumping.

In the limit of large pressure broadening and short excited-state quenching times, the alkali-metal nuclear spin I_A can be considered to be conserved in the optical pumping cycle, an excellent approximation for SEOP in the 5 to 10 amg He density range. At lower densities, however, as the electrons are rapidly relaxed in the excited state, hyperfine coupling with the nuclei causes some nuclear spin relaxation, reducing the optical pumping efficiency and requiring more photons per atom to reach full polarization (Lancor and Walker, 2010).

Under the high alkali density conditions of SEOP, spin-exchange collisions between the alkali-metal atoms are by far the dominant spin-dependent collision process. Since alkali-metal spin-exchange collisions conserve total angular momentum, they mainly serve, in concert with the hyperfine coupling between the nuclei and electrons, to reach a spin-temperature equilibrium where the fraction of atoms in any total angular momentum state F, m_F is $\rho(F, m_F) \propto e^{\beta m_F}$ (Anderson, Pipkin, and Baird, 1959; Happer, 1972; Appelt *et al.*, 1998). The spin-temperature parameter β is related to the alkali-metal electron spin polarization by $P_A = \tanh(\beta/2)$, and either of these numbers (β, P_A) is sufficient to completely describe the states of the alkali-metal spins. It is often convenient to account for the alkali-metal nuclei by the “slowing-down factor” $q = \langle F_z \rangle / \langle S_z \rangle$ which is approximately $2I_A + 1$ (isotopic average of 5.4 for Rb) under the usual high polarization conditions of SEOP (Appelt *et al.*, 1998). Sometimes it is useful to note that, at small magnetic fields where F is a good quantum number, the electron spin polarization arises solely from the populations of the two states $F = I_A + 1/2, m_F = \pm(I_A + 1/2)$, i.e., $P_A = \rho(F, F) - \rho(F, -F)$.

Since the rapid alkali-alkali spin-exchange collisions keep the electronic and nuclear spins in spin-temperature equilibrium, regardless of the collision mechanism we can write the optical pumping process using conservation of angular momentum:

$$\frac{d\langle F_z \rangle}{dt} = \frac{1}{2} [R(\mathbf{r})(P_\infty - P_A) - \Gamma_A P_A], \quad (9)$$

where Γ_A is an effective electron spin-relaxation rate and $R(\mathbf{r})$ is the local photon absorption rate (Fig. 6) for unpolarized atoms at position \mathbf{r} in the cell. The local steady-state polarization in the bulk of the cell is therefore

$$P_A(\mathbf{r}) = P_\infty \frac{R(\mathbf{r})}{R(\mathbf{r}) + \Gamma_A}. \quad (10)$$

The photon scattering rate is

$$\begin{aligned} \mathcal{A}(\mathbf{r}) &= R(\mathbf{r})[1 - P_\infty P_A(\mathbf{r})] \\ &= \Gamma_A P_\infty P_A(\mathbf{r}) + R(\mathbf{r})(1 - P_\infty^2). \end{aligned} \quad (11)$$

The first term is the scattering rate required to make up for spin-relaxation collisions, while the second term represents the scattering rate from a maximally polarized atom and would be zero for idealized D1 pumping (Happer and Van Wijngaarden, 1987; Happer, Jau, and Walker, 2009; B. Lancor *et al.*, 2010).

2. Alkali spin relaxation

There are many collision processes that can relax the alkali-metal spin polarization in SEOP cells. We already mentioned (Secs. II.A and II.A.2) the relaxation due to the spin-rotation interaction $V_{\gamma N} = \gamma(\xi)\mathbf{S} \cdot \mathbf{N}$ that couples the electron spin to the rotational angular momentum \mathbf{N} of a colliding alkali-He pair at distance ξ (Walker, Thywissen, and Happer, 1997). $V_{\gamma N}$ arises from spin-orbit interactions induced by s - p mixing during collisions and is therefore proportional to the spin-orbit splitting in the alkali excited state. This is a primary motivation for using K, with its smaller fine-structure interaction, over Rb as a preferred spin-exchange partner. This collision process has a strong T^4 temperature dependence (Ben-Amar Baranga *et al.*, 1998). Analogous interactions are presumably responsible for relaxation in alkali- N_2 collisions.

Also of practical importance is the small nonconservation of spin angular momentum in alkali-metal-alkali-metal interactions. This relaxation arises from the spin-axis interaction $V_{SS} = 2\lambda(\xi)\mathbf{S} \cdot (\hat{\xi}\hat{\xi} - 1/3) \cdot \mathbf{S}$, with about 1/2 of the relaxation coming from binary collisions (Kadlecek, Walker *et al.*, 2001) and 1/2 from the formation of triplet molecules (Kadlecek, Anderson, and Walker, 1998; Erickson *et al.*, 2000). The molecular contribution can be isolated by magnetic decoupling with 0.1 T-scale magnetic fields and has a surprising and as yet unexplained persistence at high He pressures. The low-field rate coefficients are 1.0×10^{-18} cm³/s for K-K collisions and 9.3×10^{-18} cm³/s for Rb-Rb. Again, the smaller rate for K relaxation makes it attractive for ^3He SEOP. At very low pressures, a few tens of mbar, relaxation from Rb₂ singlet molecules becomes important (Kadlecek, Anderson *et al.*, 2001); this is a minor contribution for most SEOP situations but has a similar pressure dependence to diffusion and care has to be taken to account for both in relaxation experiments.

Spin-relaxation rates and optical pumping rates are generally much larger than the characteristic diffusion rates of the alkali atoms to the cell walls. Thus throughout most of the cell the alkali polarization varies slowly (cm length scales) as the pumping

light is attenuated during propagation through the cell. Near the cell walls, however, there is a thin “diffusion layer” of unpolarized atoms. The alkali polarization is nearly zero at the walls, so to a good approximation the polarization at a distance z from the wall is modified from Eq. (10) to $P_A(\mathbf{r})[1 - \exp(-z/\Lambda)]$, where the diffusion layer length scale is approximately $\Lambda = \sqrt{qD/R}$ at the entrance to the cell (Wagshul and Chupp, 1994; Walker and Happer, 1997; Appelt *et al.*, 1998). At 3 amg, $D \approx 0.15 \text{ cm}^2/\text{s}$ (Nelson and Walker, 2001), and assuming $R = 100\Gamma_A = 40\,000 \text{ s}^{-1}$ gives $\Lambda \sim 60 \mu\text{m}$.

3. Light propagation and circular dichroism

As already noted, the slow spin-exchange rates for SEOP require optical pumping of high density alkali-metal vapors. For unpolarized atoms, the optical depth OD_0 is typically of the order of 100, i.e., the transmission of light through the cell is e^{-100} . For uniformly polarized atoms, the optical depth of circularly polarized light is $OD = OD_0(1 - P_A P_\infty)$. Thus the light can only significantly penetrate the cell if $1 - P_A P_\infty \ll 1$, so the atoms must be polarized to nearly 100% ($P_A \sim 1$) and they must become transparent when maximally polarized ($P_\infty \sim 1$). These conditions are met only for pumping light tuned to the $P_{1/2}$ D1 resonance.

Under the high density conditions of SEOP, absorption of light during alkali-metal-He (or N_2) collisions does not obey the selection rules for isolated atoms (B. Lancor *et al.*, 2010; Brian Lancor *et al.*, 2010). This is because the fine-structure states are mixed during collisions with He atoms, as illustrated in Fig. 7. The normal D1 atomic selection rules are slightly violated, allowing absorption of circularly polarized light by fully polarized atoms. Thus in the presence of He gas the alkali atoms do not become fully spin polarized in the limit of high pumping rates; they instead acquire a maximum polarization $P_\infty < 1$.

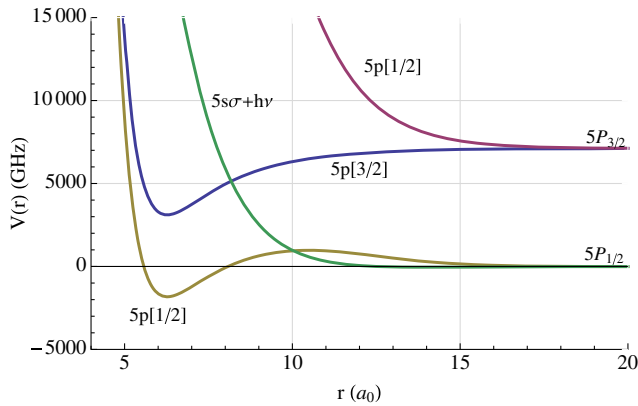


FIG. 7. Energy levels of RbHe molecules in the presence of optical pumping light (Pascale, 1983; B. Lancor *et al.*, 2010). The green curve, the ground-state potential energy plus one photon, crosses two excited-state potentials at two different interatomic separations. The $5p[M=3/2]$ curve is of purely $P_{3/2}$ nature while the $5p[1/2]$ is of mixed $P_{1/2}$ - $P_{3/2}$ character. The projection of the electronic angular momentum along the interatomic axis is M . For both crossings the colliding atom pair can absorb the circularly polarized optical pumping light even when each is fully spin polarized. From B. Lancor *et al.*, 2010.

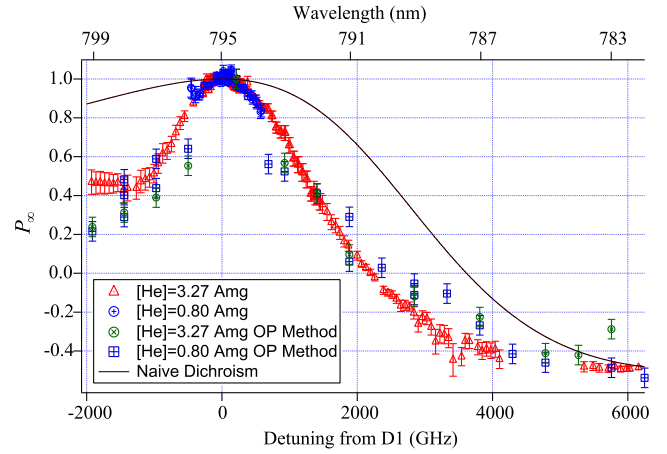


FIG. 8. Circular dichroism of Rb atoms in the presence of He gas. Near the D1 line, the dichroism approaches 1, reaching $-1/2$ for the D2 line. The solid line shows the dichroism neglecting He collisions. The very significant reduction near the D1 line is responsible for excess photon absorption under SEOP conditions. Adapted from B. Lancor *et al.*, 2010.

Figure 8 shows measurements of the circular dichroism P_∞ of maximally polarized Rb atoms in the presence of ^3He gas (B. Lancor *et al.*, 2010; Brian Lancor *et al.*, 2010). While the dichroism peaks at very close to 1 right on resonance, at only 1 nm (475 GHz) detuning from the line center, the dichroism has already dropped to 0.9. The strong reduction of the circular dichroism for off-resonant pumping implies that narrow-band lasers will not only yield more efficient optical pumping, but also higher maximum polarization.

For narrow-band pumping in the bulk of the cell, the photon flux density $I(\mathbf{r})$ ($\text{cm}^{-2} \text{ s}^{-1}$) obeys (suppressing from now on the implied spatial dependence of I , P_A , and \mathcal{A})

$$\frac{dI}{dz} = -\mathcal{A}[A] = -[A]\Gamma_A P_\infty P_A - [A]\sigma I(1 - P_\infty^2), \quad (12)$$

where the pumping rate and the photon flux density are related, $R = \sigma I$, by the optical absorption cross section σ for unpolarized atoms. For pumping by resonant, narrow-band light the first term, which leads to a linear decay $I(z) = I(0) - [A]\Gamma_A P_\infty^2 z$ (Bhaskar *et al.*, 1979; Walker and Happer, 1997), dominates. The light is attenuated purely to compensate for loss of angular momentum in ground-state collisions. The transparency of nearly fully polarized atoms allows the light to be only weakly attenuated despite the great optical thickness of SEOP cells.

When there are significant off-resonant light components, however, the second term becomes important. For high optical pumping rates $P_A \approx P_\infty$ and Eq. (12) leads to exponential attenuation of the flux density in the usual fashion $I = I(0) \exp[-[A]\sigma(1 - P_\infty^2)z]$. The absorption length of the light is increased by $1/(1 - P_\infty^2)$ over what it would be for unpolarized atoms, but can still be smaller than the cell length when P_∞ deviates from 1.

The excess photon scattering due to the circular dichroism effect is exacerbated by pumping with un-narrowed diode array bars with linewidths on the order of 2 nm (1000 GHz).

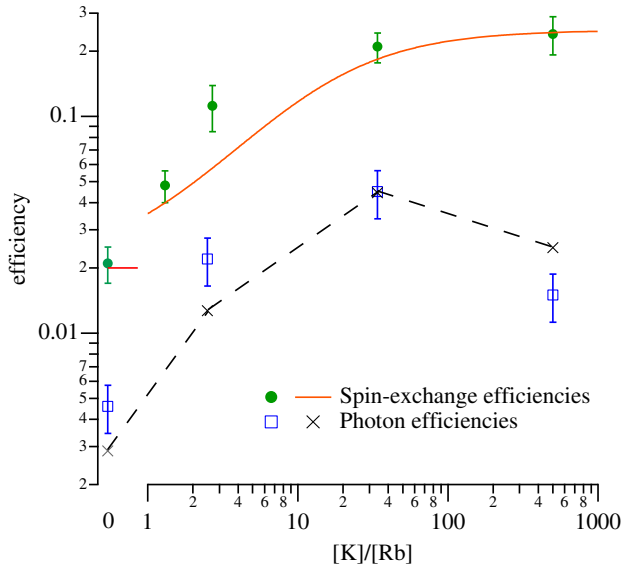


FIG. 9. Measured efficiencies at 190 °C as a function of density ratio $\mathcal{D} = [\text{K}]/[\text{Rb}]$. The spin-exchange efficiency η_{SE} , which is the maximum possible efficiency with which the angular momentum of the pumping light can be transferred to the nuclei, shows the clear increases predicted by Eq. (16), solid line, as the vapor approaches pure K. Measured and modeled photon efficiencies η_γ are much smaller, thanks to dichroism effects; see Sec. II.B.3. From Babcock *et al.*, 2003.

One can define a photon efficiency analogous to the ideal spin-exchange efficiency of Eq. (7), namely, the ratio of the rate of production of polarized ^3He to the photon scattering rate:

$$\eta_\gamma = \frac{V[\text{He}]dP_{\text{He}}/dt}{[A]V\mathcal{A}} = \eta_{\text{SE}} \frac{\Gamma_A P_A}{\mathcal{A}}. \quad (13)$$

For ideal circular dichroism, $P_\infty = 1$, $\eta_\gamma = \eta_{\text{SE}}$. At high optical pumping rates,

$$\eta_\gamma \approx \eta_{\text{SE}} \frac{\Gamma_A P_\infty}{R(1 - P_\infty^2)}, \quad (14)$$

which, since $R \gg \Gamma_A$, can be much less than η_{SE} . Measured photon efficiencies for pumping with a particularly poor laser are shown in Fig. 9.

4. Laser linewidth issues

The early laser-based SEOP experiments (late 1980s, early 1990s) with ^3He used Ar^+ -pumped tunable dye lasers (Chupp *et al.*, 1987; Coulter *et al.*, 1990) or Ti:sapphire lasers (Larson, Häusser *et al.*, 1991) to optically pump Rb vapor. With the notable exception of the early SLAC⁵ ^3He targets that used up to six Ti:sapphire lasers (Johnson *et al.*, 1995), these large, expensive, and often unreliable sources were generally limited to <10 W of pumping light. The first application to SEOP

with diode lasers was performed with relatively low power individual diodes (Wagshul and Chupp, 1989).

With the introduction of inexpensive, high-power laser diode array bars in the early 1990s, SEOP experiments rapidly converted to these new sources that were readily available at a fraction of the cost (Cummings *et al.*, 1995). The drawback of laser diode array bars is their relatively broad spectral profile (typically 2 nm or 900 GHz) that is not well matched to the 20 GHz linewidth of Rb atoms at 1 amg of He density. Thus only a small fraction of the laser light is actually available for resonant pumping of the Rb atoms. It thus became common practice to use higher He densities (between 3 and 10 amg) and rely on high optical thickness of the SEOP cells to absorb a significant fraction of the broad spectral profile of the pumping light (Driehuys *et al.*, 1996). Such pressures were well matched to high luminosity targets for electron scattering. For MRI pressure of a few bar is convenient for gas delivery; for this reason and for optical pumping efficiency high He densities were also employed (Middleton *et al.*, 1995; MacFall *et al.*, 1996).

In practice, although these high-power lasers were able to polarize larger volumes of ^3He , the attainable ^3He polarizations using these lasers were generally found to be limited to 50% or less for these applications. Examination of Fig. 8 shows that a polarization reduction could be at least qualitatively explained because of optical pumping by off-resonant light. In optically thick SEOP cells this effect is exacerbated because the central core of the spectral profile of the light is depleted in the front portion of the cell, so that in the back of the cell the atoms are generally being pumped by off-resonant light with a substantially reduced value of P_∞ . The broad linewidth of diode lasers was a particularly bad match to neutron spin filters. Although pressures of 3 bar were employed in the early development of SEOP-based NSFs (Gentile *et al.*, 2000; Jones *et al.*, 2000), simpler construction and long lifetimes made pressures of closer to 1 bar preferable (Rich, Gentile *et al.*, 2002).

Attainable ^3He polarizations increased with the introduction of narrower diode laser sources, first using long external cavities (Chann *et al.*, 2003; Babcock, Chann *et al.*, 2005) similar to those used in pulsed dye lasers and later using compact VHGs (Volodin *et al.*, 2004; Chen, Gentile, Ye *et al.*, 2014). Both narrowing methods generally reduce the laser linewidth to 0.2 nm (90 GHz), thus giving a much better match to the atomic absorption spectrum and also increasing the value of P_∞ . SEOP experiments utilizing such frequency narrowed lasers generally produce ^3He polarizations of 70% or greater, and recently have demonstrated 85% (Chen, Gentile, Ye *et al.*, 2014). Scaling of narrowing techniques to stacks of diode laser bars (Zhu, Ruset, and Hersman, 2005) has reached kilowatt levels (Hersman *et al.*, 2016).

Full simulations of propagation of broad- and narrow-band laser light due to competing spin relaxation and circular dichroism effects have been made for SEOP in 1 to 3 bar spin-filter-type cells (Brian Lancor *et al.*, 2010), and for 8–10 bar cells (Singh *et al.*, 2015). Such models generalize Eq. (12) to account for both spectral and spatial evolution of the optical pumping light and do not yet include dual-sided pumping. They also generally attempt to account for heating effects

⁵Stanford Linear Accelerator Center, Menlo Park, CA.

(Sec. II.B.5) in a simplified manner. Such models generally predict higher polarizations than are observed experimentally, although they do semiquantitatively explain the much higher polarizations achieved with narrow-band pumping over broadband pumping. An important gap in the literature is a quantitative comparison of such models with three-dimensional alkali-metal polarization mapping techniques using EPR spectroscopy (Young *et al.*, 1997).

5. Heating

With the common use of 100 W scale diode lasers for SEOP, it is natural and important to ask about how this energy is dissipated in the vapor. The first work addressing this topic was Walter, Griffith, and Happer (2001). They used Raman spectroscopy to measure the rotational and vibrational spectra of the N_2 molecules under SEOP conditions. The picture painted by this study is as follows. The N_2 quenching collisions that are so important for preventing radiation trapping leave the N_2 molecules with 1.5 eV of excitation contained in five or six vibrational quanta. The vibrational energy relaxes slowly, while the rotational degrees of freedom rapidly thermalize with the local translational temperature. Therefore the rotational spectroscopy can be used to infer the internal temperature of the vapor. The spatial dependence of the internal temperature was inconsistent with conductive heat transfer, indicating convective heat transport inside the cell. While the convection tends to reduce temperature gradients, striking high-temperature increases were observed. For example, a 95 °C internal temperature increase with respect to the wall temperature occurred with only 22 W of deposited light power in an 8.4 bar cell.

Parnell *et al.* (2010) measured the temperature rise in a 2.3 bar ^3He SEOP cell under illumination by a 100 W narrowed diode array bar. Using a gradient spin-echo sequence, they were able to measure the spatial profile of the local diffusion coefficient and observed an increase in diffusion at the center of a cell consistent with a 30 K temperature rise. Double cells provide a means for measuring the temperature rise during SEOP by the resulting drop in gas density in the SEOP cell and a consequent increase in the non-SEOP cell. Singh *et al.* (2015) compared ^3He NMR signals with and without pumping light to deduce 20 to 50 K temperature increases for high-pressure electron-scattering cells. Normand *et al.* (2016) used neutron transmission to observe the reduction in the ^3He gas density in the SEOP cell under optical pumping conditions. Their results indicated a remarkable 135 K temperature increase in a 1 bar SEOP cell with 200 W of laser illumination. Systematic studies using these kinds of techniques on cells with varied nitrogen and ^3He pressures, hybrid mixtures, and laser illumination would fill in an important gap in our understanding of the interior conditions of SEOP cells.

C. SEOP with pure K or Na

As compared to traditional SEOP with rubidium, the efficiency of SEOP should be much greater for potassium due to its lower spin destruction rate (Walker, Thywissen, and Happer, 1997; Ben-Amar Baranga *et al.*, 1998). Increased

spin-exchange efficiency for K, up to a factor of 10 above that of Rb for the same temperature, was observed (Ben-Amar Baranga *et al.*, 1998). Enthusiasm for exploiting this substantial advantage has been tempered by potential pumping of the D2 line due to the small (3.4 nm) fine-structure splitting of the $4p$ state. The first reported SEOP application yielded 46% in a midsized double cell target, using a Ti:sapphire laser (Wang, Shao, and Hughes, 2003) operating at the potassium 770 nm D1 transition wavelength. 770 nm diode lasers have had limited availability and lower power compared to their 795 nm counterparts for pumping Rb. Thus hybrid SEOP with K-Rb mixtures has remained the preferred approach. The single study of 770 nm pumping (Chen, Gentile *et al.*, 2007), which focused on NSF applications, did find increased efficiency as compared to pure Rb SEOP but also observed that the excess laser power demand and decline of alkali-metal polarization with increasing spin-exchange rate was similar to pure Rb cells, with poorer performance relative to K-Rb hybrid pumping. These observations are not well understood and merit future investigations. Although 770 nm pumping was found to be preferable for the special case of high mixture ratio hybrid cells (see Sec. II.D), it is generally desirable to use the same laser for all cells and so hybrid SEOP has remained favored. In principle, SEOP with Na would yield even higher efficiency (Borel *et al.*, 2003) but practical application is hampered by cell browning at the required temperatures of ≥ 300 °C and the absence of convenient laser sources at 590 nm.

D. Hybrid spin exchange

The idea of hybrid SEOP is to optically pump Rb in the presence of a higher density vapor of K. Rapid spin-exchange collisions between the Rb and K atoms lead to spin-temperature equilibrium between them; thus the K atoms become collisionally polarized to a level equal to that of the Rb atoms. The K-rich K-Rb vapor loses less angular momentum per atom due to spin relaxation as compared to a pure Rb vapor, so that the fraction of the angular momentum of the light that gets transferred to ^3He increases. The spin-relaxation rate of the Rb atoms in the presence of K is increased due to spin-exchange collisions with the K atoms. The rapid K-Rb spin exchange thereby causes the effective Rb spin-relaxation rate to increase from its K-free value Γ_{Rb} to

$$\Gamma'_{\text{Rb}} = \Gamma_{\text{Rb}} + \mathcal{D}\Gamma_{\text{K}}, \quad (15)$$

where $\mathcal{D} = [\text{K}]/[\text{Rb}]$ and the total K relaxation rate is Γ_{K} . The spin-exchange efficiency becomes

$$\eta_{\text{SE}}^{\text{KRb}} = \frac{(k_{\text{SE}}^{\text{Rb}} + \mathcal{D}k_{\text{SE}}^{\text{K}})[^3\text{He}]}{\Gamma_{\text{Rb}} + \mathcal{D}\Gamma_{\text{K}}} \quad (16)$$

which approaches $\eta_{\text{SE}}^{\text{K}}$ at large \mathcal{D} . Measurements of the spin-exchange efficiency as a function of \mathcal{D} are shown in Fig. 9.

The earliest experiments indicated that the maximum attainable alkali polarization drops at high \mathcal{D} (Fig. 10). This comes from off-resonant pumping of the K atoms by the resonant Rb light (Lancor and Walker, 2011) and limits the useful density ratios to $\mathcal{D} < 10$. Chen, Gentile *et al.*, 2007

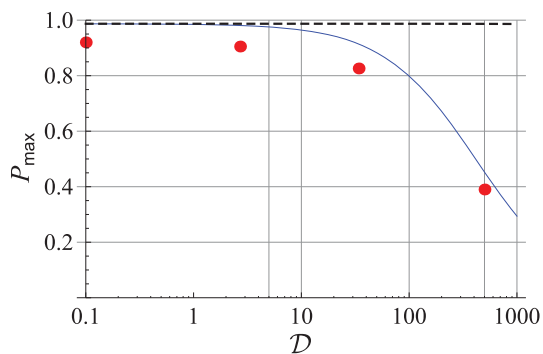


FIG. 10. Alkali polarization as a function of K-Rb density ratio. The solid line shows the density limit for a narrow-band pumping laser, using the measured K absorption cross section at 795 nm. The dots are experimental measurements using a broad pumping laser whose maximum polarization is limited by the dichroism effect to 0.92. The dashed line shows expected polarization limits predicted from a naive line-broadening model. From [Lancor and Walker, 2011](#).

observed similar results and found that the polarization decline was not observed if hybrid cells were optically pumped with 770 nm light.

The reduced collisional loss per atom for hybrid pumping means that for a given laser power the volume of ^3He can be increased or the alkali density and thus the polarizing rate can be increased. Practical application of hybrid SEOP has been investigated for neutron spin filters ([Chen, Gentile *et al.*, 2007](#); [Chen *et al.*, 2011](#)) and targets for electron scattering ([Ye *et al.*, 2013](#); [Singh *et al.*, 2015](#)). Vapor mixture ratios \mathcal{D} between 2 and 7 were found to yield the best results. Because of the difference in vapor pressures for a given temperature, ≈ 25 times more condensed phase K than Rb is required to yield $\mathcal{D} = 4$. [Chen, Gentile *et al.* \(2007\)](#) and [Chen *et al.* \(2011\)](#) described individual Rb and K distillation whereas [Singh *et al.* \(2015\)](#) described premixing Rb and K in a glove box. Variations in \mathcal{D} occur in both methods but can be minimized; in the individual approach \mathcal{D} can be checked before the cell is sealed off and in the premix approach experimental feedback on the premix ratio to account for fractional distillation improves the reproducibility. For NSF cells \mathcal{D} has been determined by white light absorption and the pumping rate for a given temperature ([Chen, Gentile *et al.*, 2007](#); [Chen *et al.*, 2011](#)) and for targets by laser light absorption and Faraday rotation ([Singh *et al.*, 2015](#)). After a newly filled cell has been heated 1 or more times, the mixture ratio may increase, perhaps due to curing effects ([Ma *et al.*, 2009](#)).

Although Fig. 9 indicates 6 times higher efficiency for hybrid SEOP in the optimum regime, hybrid cells are operated at typically 40°C higher temperature than pure Rb cells in order to obtain the same spin-exchange rate. For NSF cells at pressures near 1 bar, alkali-metal spin destruction from alkali-alkali collisions dominates, which decreases the efficiency and/or rate gain. In this regime, a resulting efficiency or rate gain approaching 3 or $\sqrt{3}$, respectively, has been calculated and observed. For high-pressure targets, alkali-metal spin destruction from alkali- ^3He collisions dominates, and thus both the efficiency and rate gain are expected to be closer to

that shown in Fig. 9. An additional benefit observed for hybrid cells is an observed slower decline of the alkali-metal polarization with increasing spin-exchange rates. Whereas this result was found to be in agreement with modeling, a steeper decline was observed for both pure Rb and pure K pumping in disagreement with modeling ([Chen, Gentile *et al.*, 2007](#)).

Hybrid SEOP has substantially increased the quantity and production rate of polarized gas for both NSFs and polarized targets. For NSFs cells 85% ^3He polarization in cells approaching 1 L in volume with pumping time constants of between 4 and 8 h ([Chen, Gentile, Erwin *et al.*, 2014](#)) has been achieved and for double cell polarized targets up to 70% has been reached with similar time constants ([Singh *et al.*, 2015](#)).

E. Limits to ^3He polarization

Combining the capability of polarizing high density alkali vapors to near 100% polarization, the developments of glass cells with hundreds of hour room temperature wall-relaxation times, and rapid spin exchange with hybrid pumping, the attainable ^3He polarizations should be nearly 100%. The issues limiting the polarization are not yet fully understood. We briefly review here what is known about this issue. More details on most of these topics are discussed in other sections of this review.

1. Anisotropic spin exchange

[Walter, Happer, and Walker \(1998\)](#) pointed out that the long-range portion of the ^3He nuclear magnetic field causes an additional term in the alkali- ^3He Hamiltonian:

$$V_\beta = \beta(\xi)\mathbf{S} \cdot (3\hat{\xi}\hat{\xi} - 1) \cdot \mathbf{I}_{\text{He}}. \quad (17)$$

This anisotropic spin-exchange interaction tends to polarize the ^3He nuclei toward $P_{\text{He}} = -P_A/2$, and so serves to limit the maximum ^3He polarization to

$$P_{\text{He}\infty} \leq 1 - \frac{3k_\beta}{2k_\alpha}, \quad (18)$$

where k_α and k_β are the respective rate coefficients for the (isotropic) Fermi-contact interaction and the anisotropic interaction in Eq. (17). A theoretical estimate of the effect of anisotropic spin-exchange yielded $P_{\text{He}\infty} = 0.95$ for SEOP with either Rb or K ([Walter, Happer, and Walker, 1998](#)), and more recent calculations yielded a similar value of 0.96 ([Tscherbul *et al.*, 2011](#)).

There are no definitive experimental measurements of anisotropic spin exchange. Wall-independent techniques (Sec. II.A.1) for measuring spin-exchange rates are sensitive to the combination $k_\alpha - k_\beta/2$. As discussed next (Sec. II.E.2), spin-exchange transients are generally sensitive to $k_{\text{SE}}[A] + \Gamma_w$, where $k_{\text{SE}} = k_\alpha + k_\beta$, but the strong temperature dependence of Γ_w makes it difficult to isolate k_β from Γ_w . However, [Walker, Nelson, and Kadlecik \(2010\)](#) pointed out that comparison of spin-relaxation rates of K in ^3He and ^4He , when combined with k_{SE} measurements, allow k_β to be

isolated without any assumptions about wall relaxation. A first attempt gave $P_{\text{He}\infty} = 0.90 \pm 0.11$.

An upper limit on anisotropic spin exchange can be deduced from absolute ^3He polarimetry. The highest ^3He polarization obtained to date is 0.88 ± 0.03 (Chen, 2016) in a $\mathcal{D} = 4$ K-Rb hybrid cell, giving $k_{\beta} \leq (0.09 \pm 0.02)k_{\alpha}$.

2. X factor

Studies of the time constants for spin exchange obey the phenomenological relation (Chann *et al.*, 2002a, 2003; Babcock *et al.*, 2006; Chen, Gentile *et al.*, 2007; Walker, 2011; Chen, Gentile, Ye *et al.*, 2014; Singh *et al.*, 2015)

$$\frac{1}{\tau} = k_{\text{SE}}(1 + X)[A] + \Gamma_r, \quad (19)$$

where Γ_r is the room temperature relaxation rate, taken to be independent of temperature. This relation, experimentally verified for both Rb and K-Rb hybrid cells, indicates that the wall-relaxation rate has an exponential increase with temperature that mimics the variation of alkali vapor pressure with temperature. This limits the ^3He polarization to

$$P_{\text{He}\infty} \leq \frac{1}{1 + X} \quad (20)$$

for 100% Rb polarization and negligible Γ_r . That the ‘‘X factor’’ originates from a temperature dependence of the wall-relaxation rate is suggested by the tremendous variations in measured values of X for many different cells. Figure 11 shows a sample of such data. If it is a wall-relaxation effect, $X = \chi S/V$ should hold, where S/V is the cell surface-to-volume ratio and the relaxivity χ is a poorly controlled parameter. Indeed, there are greater fluctuations in X for small cells (large S/V) than for large cells (Walker, 2011). For large cells (e.g., $S/V \leq 1 \text{ cm}^{-1}$) X was found to be typically ≈ 0.3 , thus limiting the maximum ^3He polarization to between 75% and 80%. Direct determinations of X by measurements of

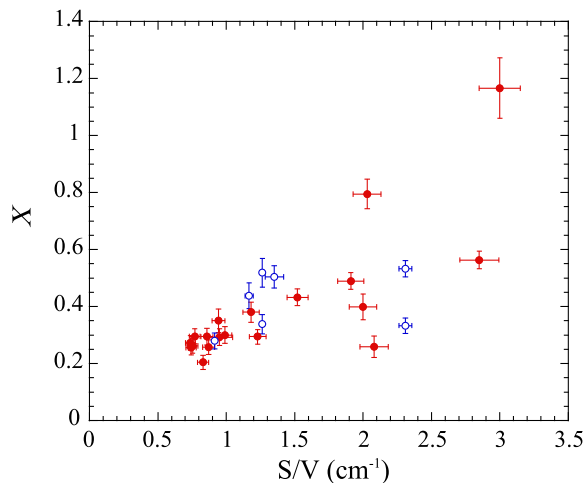


FIG. 11. NIST measurements of the X factor, deduced from ^3He polarization limits at high temperature, for both blown (solid, red) and flat-windowed (open, blue) neutron spin-filter cells with a range of surface-to-volume ratios S/V . Adapted from Babcock *et al.*, 2006.

the relaxation of heated cells were found to be consistent with measurements of the maximum ^3He polarization.

Further determinations of X in spin-filter cells (Parnell *et al.*, 2009) and double cells for ^3He targets (Ye *et al.*, 2010; Singh *et al.*, 2015) have been reported. The temperature dependence of relaxation in quartz cells has also been studied with deviation from the characteristic linear dependence on alkali-metal density (Ino and Muto, 2007) and larger X values observed (Ye *et al.*, 2013). For this reason quartz presents issues for achieving the best ^3He polarization values for SEOP, even if a long relaxation time at room temperature can be obtained.

Recently, the polarization of NSFs (Chen, Gentile, Ye *et al.*, 2014) pumped by 100 W VHG-narrowed lasers has found polarization limits over 85%, compared to the 75% to 80% found in earlier studies (Babcock *et al.*, 2006; Chen, Gentile *et al.*, 2007). These polarization increases were also observed in cells that were studied over a decade time scale. The polarization increases with time (or perhaps equivalently, narrow-band laser power) suggest that at least some of the polarization limits attributed to the X factor may be insufficiently polarized Rb vapor in those early experiments. However, the Rb polarizations measured in those experiments using EPR spectroscopy were found to be conservatively in excess of 95%, generally 98% or above (Babcock, 2005).

3. Rb polarization limits

When the spin-exchange rates dominate over any other relaxation mechanisms, the ^3He polarization can never exceed the spatial average of the alkali polarization.

The first rather obvious implication of this statement is that due to the high pressures used in SEOP, so that there is little alkali polarization diffusion, care must be taken to provide sufficient pumping light to all regions of the cell, including ensuring that the light exiting the cell is not fully depleted of useful pumping light. Because most SEOP experiments use lasers with spectral linewidths greater than the pressure-broadened linewidth of the atoms, it is essential to measure the spectral profile of the transmitted light, checking that the useful resonant portion of the light is not fully depleted.

Because of the imperfect dichroism of Rb pumped in the presence of high-pressure ^3He (Sec. II.B.3), spectrally broad lasers have been observed to have 10% reduction in their maximum Rb polarization, even under low Rb density conditions (Babcock *et al.*, 2003). This effect is exacerbated as the light propagates through the optically thick cell and the resonant portion is depleted, further lowering the dichroism. These effects are now understood to be largely responsible for the 20% increases in polarization observed when experimenters first used frequency narrowed diode array bars (Chann *et al.*, 2003; Gentile, Chen *et al.*, 2005).

Because of the locally enhanced spin relaxation of alkali atoms within a diffusion length of the wall, correspondingly large light absorption occurs at the entrance to the cell, leading to reduced pumping rates and Rb polarizations within the bulk of the cell. For single-frequency pumping with dye or Ti: sapphire lasers, where the spectrum of the light is much narrower than that of the atoms, this effect can be somewhat mitigated by purposely tuning the laser off resonance (Wagshul and Chupp, 1994). For broad or frequency narrowed

diode array bars, typically up to 10% of the useful light can be absorbed by this layer, again reducing pumping rates and hence alkali polarizations within the bulk of the cell.

Finally, even for narrow-band pumping hybrid K:Rb mixtures exhibit reduced maximum polarizations at high \mathcal{D} due to off-resonant absorption of the Rb pumping light by the K atoms (Lancor and Walker, 2011); see Fig. 10. This effect limits the useful values of $\mathcal{D} < 10$.

4. EPR spectroscopy

It is often useful to complement ^3He polarimetry with diagnostics to measure and even image the Rb polarization. In the simplest version, one can monitor the transmission of the optical pumping light. Assuming the pump light is approximately circularly polarized, the light transmission is minimum when the laser is tuned directly on resonance. Then as the circular polarization of the light is adjusted, the transmission will be a maximum when the pumping light is maximally circularly polarized. The sensitivity of this technique is enhanced by monitoring the transmitted pumping light with a diffraction grating spectrometer. Then the greater polarization sensitivity of the on-resonant spectral components is easy to observe. Such a diagnostic can however identify the alkali polarization only as “high.” For a more sensitive alkali polarization monitor, a linearly polarized Faraday probe, copropagating with the pumping light but tuned near the D2 optical resonance (so it can be distinguished from the pumping light by a spectrometer or interference filter), can be used. The high optical depth in SEOP makes the polarization rotations large ($\sim\text{rad}$) even for light tuned a number of linewidths off resonance. The Faraday rotation is proportional to $P_A[A]$, so absolute calibration of P_A is difficult without a precise alkali number density measurement. Indeed, assuming $P_A \approx 1$, the Faraday rotation can be used to measure $[A]$ (Chann *et al.*, 2002a).

For a more quantitative alkali polarization assessment, EPR spectra can be obtained by monitoring the transmitted intensity of the pumping light, Faraday rotation of a copropagating probe laser, or residual cell fluorescence as a transverse oscillating magnetic field is swept through the alkali Zeeman resonances, slightly changing the alkali polarization (Romalis and Cates, 1998; Chann *et al.*, 2002a; Kramer *et al.*, 2007). These detection modalities are all proportional to the longitudinal alkali spin polarization, so the “longitudinal EPR” signal is proportional to the square of the oscillating magnetic-field strength. At small magnetic fields, the splitting between adjacent EPR lines arises largely from the second-order Zeeman effect and is $q(\mu_B B)^2/h^2 \delta\nu$, where μ_B is the Bohr magneton, $\delta\nu$ is the hyperfine splitting, h is Planck’s constant, and $q = 2/9$ for ^{85}Rb with nuclear spin $I_A = 5/2$ and $q = 1/2$ for the $I_A = 3/2$ species ^{87}Rb and K. This neglects the nuclear Zeeman interaction, which slightly splits the rf resonances for the two hyperfine levels. Under the strong pumping conditions typical for SEOP, the two primary EPR lines are $m = F_{\max} \rightarrow F_{\max} - 1$ and $F_{\max} - 1 \rightarrow F_{\max} - 2$, where the maximum angular momentum of the atom is $F_{\max} = I_A + 1/2$. The F_{\max} line is narrowed due to having little spin-exchange broadening, while the $F_{\max} - 1$ line experiences substantial spin-exchange broadening

(Appelt *et al.*, 1999). The Rb polarization is simply related to the areas of the rf resonances. In ^{85}Rb , for example, assuming spin-temperature equilibrium one gets

$$P_{\text{Rb}} = \frac{7A_3 - 3A_2}{7A_3 + 3A_2}, \quad (21)$$

where A_m is the area of the EPR peak originating from $m_F = m$. This is valid when the $F, m = 3, 2$ and $2, 2$ lines are not resolved.

A more sensitive technique for EPR alkali polarimetry, and the first to be demonstrated, is to directly detect the transverse alkali polarization induced by the oscillating magnetic field (Young *et al.*, 1997; Ben-Amar Baranga *et al.*, 1998). This is done by demodulating the Faraday rotation of a transversely propagating probe laser. The transverse polarization is linear in the oscillating magnetic-field amplitude, so weaker oscillating fields are required to observe these “transverse EPR” signals. Again, ratios of the areas of resonance peaks allow the alkali polarization to be measured. Lancor (2011) compared simultaneous longitudinal and transverse EPR signals using Faraday rotation of a skew off-resonant laser. He observed that the spatially averaged alkali polarizations deduced by the two methods differed, with the longitudinal EPR generally yielding 0%–10% higher alkali polarization estimates than transverse EPR. The origin of this discrepancy is not known, although both transverse and longitudinal methods gave distributions consistent with a spin temperature. Thus earlier studies relying on longitudinal EPR for alkali polarimetry may have overestimated alkali polarizations.

Young *et al.* (1997) and Ben-Amar Baranga *et al.* (1998) also demonstrated the use of EPR spectroscopy for alkali polarization imaging. In this case the most convenient approach is to work at low fields where the individual EPR lines are not resolved and apply a longitudinal magnetic-field gradient $\partial B_z/\partial x$ so that the Faraday rotation of the probe beam, propagating along the x direction, arises solely from a small voxel of atoms that satisfy the EPR resonance condition. Sweeping the longitudinal field B_z then produces a spatial map of the Rb polarization along the x direction. Moving the probe laser allows the polarization to be mapped in three dimensions.

F. EPR frequency shift

The spin-polarized alkali-metal atoms that are present in the SEOP cell can serve as a sensitive *in situ* magnetometer, detecting the magnetic field produced by the polarized ^3He . The magnetic field at position \mathbf{x} with respect to a spin-polarized nucleus with magnetic moment \mathbf{m} is the sum of the classical dipole field and a contact term $\mathbf{B}(\mathbf{x}) = [3\mathbf{x}\mathbf{x} \cdot \mathbf{m} - x^2\mathbf{m}]/|\mathbf{x}|^{-5} + 8\pi\mathbf{m}\delta(\mathbf{x})/3$ (Jackson, 1999). The average field experienced by the alkali atoms due to the polarized ^3He is (Schaefer *et al.*, 1989; Barton *et al.*, 1994; Romalis and Cates, 1998)

$$B_{\text{He}} = B_{\text{cl}} + \frac{8\pi\mu_{\text{He}}}{3}\kappa_0[\text{He}]P_{\text{He}} \quad (22)$$

$$= \frac{8\pi\mu_{\text{He}}}{3}\kappa[\text{He}]P_{\text{He}}, \quad (23)$$

where μ_{He} is the magnetic moment of ^3He , and κ_0 is a frequency shift enhancement factor whose value is proportional to the average alkali electron spin density at the ^3He nucleus. For a gas that is uniformly polarized inside a spherical cell, the spatial average of B_{cl} vanishes so that only the contact term, responsible for the hyperfine interaction [second term of Eq. (2)], contributes $\kappa = \kappa_0$. The field B_{He} produces an EPR frequency shift $\delta\nu = \gamma_A B_{\text{He}} = \gamma_S B_{\text{He}} / (2I_A + 1)$, where $\gamma_S \approx 28$ MHz/mT is the electron gyromagnetic ratio. For the most common ^{85}Rb isotope, the numerical value of the shift is $1.13\kappa_0$ kHz for fully polarized ^3He at a density of 1 amg.

Using two orientations of a long cylindrical cell, for which B_{cl} can be accurately calculated, Romalis and Cates (1998) isolated the classical and contact contributions, thus enabling a precision measurement of κ_0 for Rb- ^3He with an uncertainty of 1.5%. This result agreed with a prior calibrated absolute NMR polarimetry measurement (Newbury *et al.*, 1993) and forms the basis of precision absolute polarimetry of electron scattering targets. EPR frequency shift polarimetry has also been indirectly tested using neutron transmission (Ye *et al.*, 2013).

The resulting alkali EPR frequency shifts $\delta\nu$ (Schaefer *et al.*, 1989) are typically tens of kHz for high density ^3He SEOP and are easily measured using any of the EPR methods. AFP is often used to briefly reverse the ^3He polarization with respect to the magnetic field and hence isolate the EPR frequency shift. The results are improved when the bias external magnetic field is stabilized using an auxiliary magnetometer (flux gate or atomic). Since the temperature dependence of κ_0 yields a typical change in the EPR frequency shift of $0.14\%/^{\circ}\text{C}$, the gas temperature must be carefully determined in order to retain the full precision in the absolute polarimetry. The effect of internal heating (Sec. II.B.5) has not generally been investigated as a source of error and uncertainty. If unaccounted for a 10°C temperature rise of the gas sampled for the EPR measurement would yield a 1.4% error, which would be comparable to the uncertainty in current values for κ_0 .

Using the carefully measured Rb- ^3He enhancement factor as a reference, the enhancement factors for K and Na have also been measured, and the temperature range for Rb- ^3He extended as needed for hybrid pumping (Babcock, Nelson *et al.*, 2005). The values of $\kappa_0(T)$ for the various alkali-metal atoms are given in Table I. The temperature dependence $d\kappa_0/dT$, also given in Table I, is sufficiently large so that 10% variations are seen over common SEOP temperature ranges.

III. METASTABILITY-EXCHANGE OPTICAL PUMPING

Metastability-exchange optical pumping mainly involves two processes: optical pumping on the optically closed $2^3\text{S}-2^3\text{P}$ transition of He at 1083 nm, described in Sec. III.C, and nuclear orientation transfer to the ground state of ^3He through metastability-exchange (ME) collisions, described in Sec. III.B. MEOP is usually performed in pure ^3He gas, and it is often sufficient to consider these two processes for ^3He atoms only. However, the addition of ^4He to ^3He gas can lead to higher nuclear polarizations and faster buildup rates, with potential applications whenever the

admixture of ^4He atoms has no adverse effect (e.g., for neutron spin filters). In isotopic mixtures, optical pumping advantageously operates on ^4He atoms (Stoltz, Meyerhoff *et al.*, 1996). From another point of view, traces of ^4He are often found in ^3He cells, resulting for instance from cell preparation or from permeation through glass walls. The impact of even sub-% ^4He fractions on MEOP efficiency or optical measurement of polarization can be significant (Batz, 2011; Talbot *et al.*, 2011). Therefore knowledge of features and processes relevant for MEOP may be needed and they will be examined for both helium isotopes in this section, starting with the relevant He level structures in Sec. III.A.

A. Atomic levels involved in MEOP

Figure 12 schematically displays the optical pumping and ME collision processes and the most relevant atomic levels for both He isotopes. The 2^3S state of ^4He ($J = S = 1$) has three magnetic sublevels ($m_S = -1, 0, \text{and } 1$), linearly split at all values of the applied magnetic field B by the Zeeman energy. They are named Y_1 to Y_3 (for all sets of Zeeman sublevels indices increase with increasing energies). The 2^3P state of ^4He has three fine-structure levels with $J = 0, 1$ and 2 , and hence nine Zeeman sublevels (Z_1 to Z_9). Because of its two nuclear

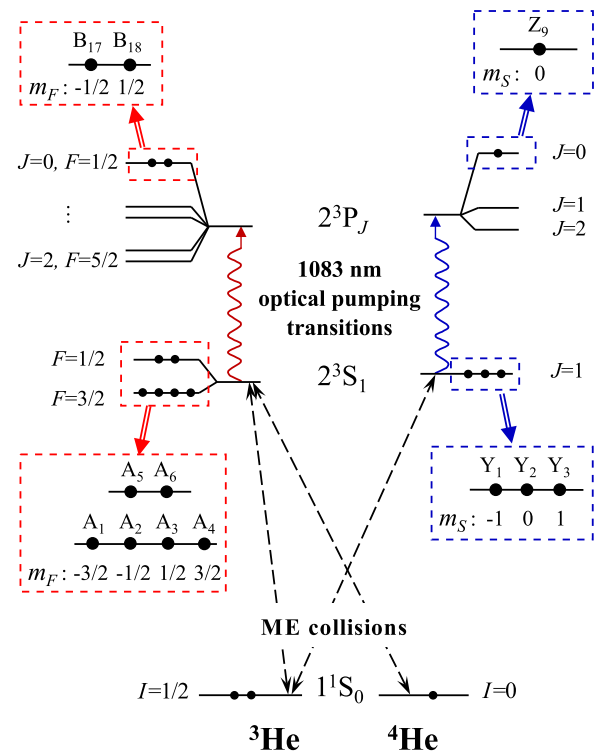


FIG. 12. Fine and hyperfine structures of the atomic states of He involved in the metastability-exchange optical pumping process, for the ^3He (left) and ^4He (right) isotopes, in low magnetic field (for negligible magnetic Zeeman energies, i.e., below a few mT). The values of the total angular momenta (J for ^4He , F for ^3He) are indicated. Details and names of the magnetic sublevels of the 2^3S and 2^3P_0 states are given in blown-up boxes [with notations of Courtade *et al.* (2002)]. The shifts and splittings are not displayed to scale.

spin states ^3He has twice as many Zeeman sublevels: six in the 2^3S state (A_1 to A_6) and 18 in the 2^3P state (B_1 to B_{18}). The magnetic sublevels A_i can be written using the decoupled basis states $|m_S, m_I\rangle$. $A_1 = |-1, -\rangle$ and $A_4 = |1, +\rangle$ are pure states while the states for which $m_F = \pm 1/2$ involve two mixing parameters θ_{\pm} (Courtade *et al.*, 2002):

$$\begin{aligned} A_2 &= \cos \theta_- | -1, +\rangle + \sin \theta_- | 0, -\rangle, \\ A_3 &= \cos \theta_+ | 0, +\rangle + \sin \theta_+ | 1, -\rangle, \\ A_5 &= \cos \theta_- | 0, -\rangle - \sin \theta_- | -1, +\rangle, \\ A_6 &= \cos \theta_+ | 1, -\rangle - \sin \theta_+ | 0, +\rangle. \end{aligned} \quad (24)$$

For simplicity, only the highest-lying Zeeman sublevels of the 2^3P_0 states are displayed in Fig. 12.

At low magnetic field, the $F = 3/2$ and $F = 1/2$ hyperfine levels of the 2^3S state of ^3He are well resolved and split by 6.74 GHz. Strong mixing of electronic and nuclear angular momenta yields large values of the mixing parameters θ_{\pm} : $\sin^2 \theta_- = 2/3$ and $\sin^2 \theta_+ = 1/3$. The 2^3P state levels extend over 32.91 GHz for ^4He and 32.60 GHz for ^3He . The isotope shifts of the energy levels result in an increased optical pumping transition energy (by 33.67 GHz) and a higher 2^3S state energy (by 259.6 GHz, i.e., 12 K in temperature units) for ^4He (Drake, Nörtershäuser, and Yan, 2005).

At high magnetic field, the Zeeman energy is no longer a small correction to the fine- and hyperfine-structure energy contributions and the angular momentum structures of the 2^3S and 2^3P levels are deeply modified. For instance, the mixing parameters in Eq. (24) have small and almost equal values $\theta_{\pm} \approx 0.11/B$, with the field strength B in T. This has a strong impact on ME and optical pumping mechanisms alike. Therefore, in each of the following sections, the low-field and high-field situations will be separately discussed as limiting cases. Traditional low field corresponds to field strengths $B \leq 10$ mT, with θ_{\pm} decreasing by less than 3% and Zeeman shifts significantly lower than Doppler widths at room temperature. Conversely, $B \geq 1$ T will be considered a high-field situation with small mixing parameters θ_{\pm} and a strongly modified level structure (see Sec. III.C.3).

B. Metastability-exchange collisions

1. Treatment of the metastability-exchange collision process

Metastability-exchange collisions are binary collisions in which the electronic excitation is transferred from a He^* atom in the metastable 2^3S state to one in the ground state. For a gas confined in a cell, the quantum states of the atoms can be statistically described by density operators depending on atomic positions and velocities. The position-dependent parameters include the local plasma characteristics and the optical pumping light intensity. This intensity has a spatial dependence because of the pumping beam transverse shape and divergence (if any) and of atomic absorption. The velocity dependence mainly results from the velocity-selective character of optical pumping with a narrow-band laser and will be addressed in Sec. III.C.2. Using a standard approach (Partridge and Series, 1966), the effect of a ME collision

between He^* and He atoms characterized by density operators ρ_m and ρ_g can be described as

$$\begin{array}{cccc} \text{He}^* & + & \text{He} & \rightarrow & \text{He} & + & \text{He}^* \\ \rho_m & & \rho_g & & \rho'_g & & \rho'_m. \end{array} \quad (25)$$

The ground-state density operator of ^3He is directly related to the populations of the two spin states $|+\rangle$ and $|-\rangle$, which are functions of the nuclear polarization P_{He} :

$$\rho_g = \frac{1 + P_{\text{He}}}{2} |+\rangle\langle +| + \frac{1 - P_{\text{He}}}{2} |-\rangle\langle -|, \quad (26)$$

while the He^* density operator can be written as

$$\rho_m = \sum_1^3 y_i |Y_i\rangle\langle Y_i| \quad \text{or} \quad \rho_m = \sum_1^6 a_i |A_i\rangle\langle A_i| \quad (27)$$

for $^4\text{He}^*$ or $^3\text{He}^*$, respectively. The relative populations of the 2^3S state sublevels Y_i and A_i are noted y_i and a_i , respectively, with $\sum y_i = \sum a_i = 1$. The local number density of 2^3S state atoms in the Zeeman sublevels are thus $n_m^{(4)} y_i$ and $n_m^{(3)} a_i$, where $n_m^{(4)}$ and $n_m^{(3)}$ are the local number densities of 2^3S state atoms of each isotope. Their ratio depends on the gas composition and temperature, due to the lower energy of the $^3\text{He}^*$ state (Zhitnikov *et al.*, 1975; Nacher, 1985; Courtade *et al.*, 2002). At room temperature ($T \gg 12$ K), $n_m^{(4)}/n_m^{(3)} = N_g^{(4)}/N_g^{(3)}$ where $N_g^{(4)}$ and $N_g^{(3)}$ are the number densities of ground-state atoms of the isotopes. Number densities are simply noted n_m and N_g in a pure gas.

For an outgoing ^3He atom,

$$\rho'_g = \text{Tr}_e \rho_m \quad (28)$$

is simply obtained using the trace operator Tr_e over the electronic variables of the incoming $^3\text{He}^*$ atom. For the outgoing He^* atom in ^3He - $^3\text{He}^*$ collisions the density operator is written as

$$\rho'_m = \sum_F P_F \rho_m'' P_F \quad \text{with} \quad \rho_m'' = \rho_g \otimes \text{Tr}_n \rho_m. \quad (29)$$

The tensor product ρ_m'' is the density operator immediately after the collision. Off-diagonal terms (coherences) between the $F = 1/2$ and $F = 3/2$ hyperfine sublevels are created during metastability-exchange collisions, but they can be neglected for usual MEOP conditions, at low enough pressure or high enough field (Courtade *et al.*, 2002). Therefore the restricted expression of ρ'_m can be used [Eq. (29)], where P_F is the projection operator onto the F hyperfine substate.

Note that off-diagonal elements can also be created if coherent light is used for pumping with a V-type or Λ -type scheme, with two optical transitions addressing at least one common sublevel in the lower state or the upper state, respectively. However, such schemes are avoided in MEOP since well-polarized light of given helicity is used.

For isotopic He mixtures, Eq. (25) generically refers to three different ME processes depending on the isotopic nature of the colliding atoms (see Fig. 12, dashed lines). Two additional equations, similar to Eq. (29), are used for collisions between atoms of different isotopes (Courtade *et al.*, 2002). The fourth type of collisions, between $^4\text{He}^*$ and ^4He atoms, plays no role in the evolution of nuclear or electronic spin variables, and therefore no direct link is sketched in Fig. 12.

The initial approach of Partridge and Series was subsequently improved to quantitatively link the time evolution of selected atomic observables with ME collision cross sections in weak pumping and low polarization conditions (Dupont-Roc, Leduc, and Laloë, 1971, 1973; Pinard and Laloë, 1980). It was then used in models suitable for the description of MEOP with lasers, in which the evolution of all Zeeman sublevel populations is evaluated for arbitrary pumping conditions (Nacher and Leduc, 1985; Batz, Nacher, and Tastevin, 2011). Rate equations are derived from Eq. (29) for the populations in the 2^3S state. They explicitly depend linearly on P_{He} due to the linear dependence of ρ_g on P_{He} in Eq. (26). For pure ^3He they are written as

$$\left. \frac{da_i}{dt} \right|_{\text{ME}} = \gamma_e \left[-a_i + \sum_{k=1}^6 (E_{ik}^{(3)} + P_{\text{He}} F_{ik}^{(3)}) a_k \right]. \quad (30)$$

The ME collision rate $\gamma_e = N_g k_{\text{ME}}$ is proportional to the number density of atoms in the ground state and depends on temperature through the ME rate coefficient k_{ME} (Sec. III.B.2). The matrices $E^{(3)}$ and $F^{(3)}$ involve B -dependent parameters (Courtade *et al.*, 2002, Tables 16 and 17). For the ground state, the contribution of ME collisions to the rate equation describing the evolution of P_{He} is obtained by computing $\text{Tr}_n \rho_g I_z$ using Eqs. (27) and (28):

$$\left. \frac{dP_{\text{He}}}{dt} \right|_{\text{ME}} = \gamma_e \int_{\text{cell}} \frac{d^3 \mathbf{r}}{V_{\text{cell}}} \frac{n_m}{N_g} (P_{\text{He}^*} - P_{\text{He}}), \quad (31)$$

where

$$P_{\text{He}^*} = \sum_{k=1}^6 L_k a_k. \quad (32)$$

The nuclear polarization is usually uniform in the ground state since the diffusion rate in a low-pressure gas is much larger than typical rates of evolution for P_{He} . On the contrary, both the nuclear polarization P_{He^*} and the density n_m of atoms in the 2^3S state depend on local pumping light and discharge intensities and may strongly vary with position. Hence Eq. (31) involves a spatial average over the cell volume V_{cell} . The metastable nuclear polarization P_{He^*} is directly derived from the set of populations using the field-dependent parameters L_k (Courtade *et al.*, 2002, Table 13).

This formalism has been extended to mixtures of ^3He and ^4He , and the corresponding rate equations have been established (Courtade *et al.*, 2002). If isotopic mixtures are optically pumped using a ^4He atomic transition, the electronic polarization created in the 2^3S state of ^4He atoms is first transferred to the 2^3S state of ^3He atoms by ME collisions with ground-state ^3He atoms. The nuclear polarization which subsequently

develops in $^3\text{He}^*$ atoms due to hyperfine interactions is then transferred to the ground state of ^3He atoms via further ME collisions. The larger light absorption probability of ^4He atoms (see Sec. III.C.2) contributes to make this indirect process more efficient (Stoltz, Meyerhoff *et al.*, 1996).

2. Temperature dependence of metastability-exchange collision rates

Early studies of the magnetic resonance linewidth in the 2^3S state of ^3He have shown that ME collision cross sections and collision rate coefficients strongly decrease with decreasing temperatures between 4.2 and 550 K (Colegrove, Scheerer, and Walters, 1964). These experimental data were found to be consistent with computations using empirically determined potential parameters (Fitzsimmons, Lane, and Walters, 1968), but the values inferred for the cross sections need to be corrected to take into account the partial character of the loss of orientation in ME collisions (Dupont-Roc, Leduc, and Laloë, 1971). These data and the results of more accurate measurements performed at low temperatures are compiled in Fig. 13 together with the results of *ab initio* calculations of different rates for collisions between 2^3S or 2^3P and ground-state He atoms (Vrinceanu and Sadeghpour, 2010). The first set of data points in Fig. 13 is evaluated from the plot of reduced linewidths $\Delta\nu/N$ in the temperature range 100 to 550 K (Colegrove, Scheerer, and Walters, 1964, Fig. 7) using the correcting relation $k_{\text{ME}} = 9/4 \times \pi \Delta\nu/N$.⁶ Other experimental data for the exchange rate coefficients k_{ME} are derived from published values of linewidth $\Delta\nu_{\text{ex}}$ (Rosner and Pipkin, 1972), exchange rate $1/\tau$ (Dupont-Roc, Leduc, and Laloë, 1971), cross section σ_{ex} (Zhitnikov *et al.*, 1975; Barbé, 1976), and number densities N using

$$k_{\text{ME}} = \pi \Delta\nu_{\text{ex}}/N, \quad k_{\text{ME}} = 1/N\tau, \quad \text{or} \quad k_{\text{ME}} = \sigma_{\text{ex}} \bar{v}_{\text{rel}}, \quad (33)$$

where $\bar{v}_{\text{rel}} = 4\sqrt{k_B T/\pi M}$ is the average relative velocity of colliding He atoms at temperature T (k_B is the Boltzmann constant, M is the ^3He atomic mass). The experimental and calculated values of ME rate coefficients are in fair agreement (the expected effect of isotope mass difference through \bar{v}_{rel} is of the order of a symbol size). For convenience, values of k_{ME} and of their local temperature variations inferred from the compiled experimental data are listed in Table II.

An interesting feature of the *ab initio* calculated rates in Fig. 13 is the comparatively high value of the exchange rate between ground-state and 2^3P state atoms, approximately 10 times larger than the ME rate at room temperature and more than 1000 times larger at a few Kelvin (the dashed curves labeled P and S, respectively). In spite of this, this coupling channel is traditionally not considered in the transfer of angular momentum to the ground state, due to the

⁶The total number density N and the ground-state number density N_g are almost equal in He gas discharges under MEOP conditions, with at most a few ppm of the atoms in an excited state. N is used instead of N_g whenever necessary or convenient.

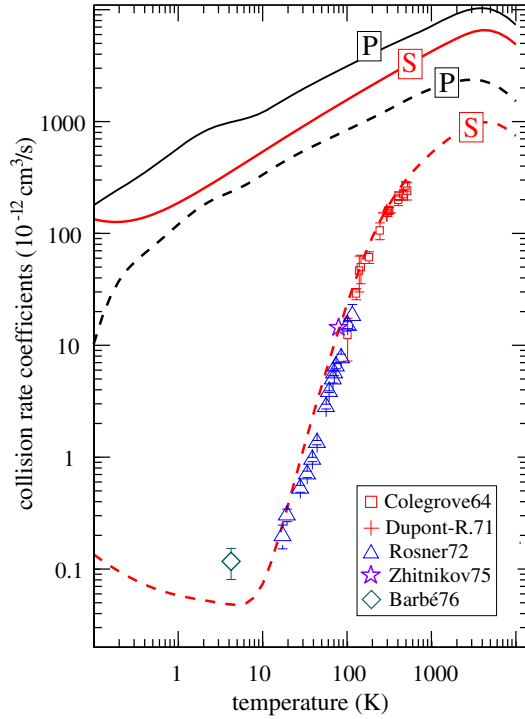


FIG. 13. Temperature dependence of calculated and experimentally assessed collision rate coefficients in He. *Ab initio* calculated rates are plotted for excitation transfer (dashed lines) and total (solid lines) rates of collisions for the 2^3S state (red curves, labeled S) and the 2^3P state (black curves, labeled P). Experimental k_{ME} data (symbols, see the legend) are derived from published values of linewidths or ME cross sections in ^3He (see text). Three calculated rate coefficients slowly decrease with decreasing temperature and thermal velocities, but the rate coefficients k_{ME} for ME collisions (the dashed red line and symbols) abruptly decrease below room temperature. This is attributed to a weak repulsive barrier at large distance in the He^*-He interaction potential. Adapted from Vrinceanu and Sadeghpour, 2010, Fig. 3 for the calculated curves.

comparatively low number density usually achieved in the 2^3P state.

3. Metastability exchange and spin-temperature distribution

An important property of ME collisions in helium is the negligible depolarization that occurs during the very short collisions (10^{-12} s), due to the fact that all involved angular momenta are spins (Pinard and Laloë, 1980). This accounts for the efficient transfer of angular momentum in MEOP from

TABLE II. Metastability-exchange rate coefficients k_{ME} and corresponding typical rates $1/\tau^{\text{yp}} = N^{\text{yp}}k_{\text{ME}}$ for a gas number density $N^{\text{yp}} = 2.687 \times 10^{16} \text{ cm}^{-3}$, i.e., 10^{-3} amg, which is a typical value in metastability-exchange optical pumping experiments (1 mbar at 273 K).

T (K)	k_{ME} ($10^{-12} \text{ cm}^3/\text{s}$)	$1/\tau^{\text{yp}}$ (10^6 s^{-1})
Around 300	$154 \times (T/300)^{1.09}$	$4.14 \times (T/300)^{1.09}$
Around 77	$6.6 \times (T/77)^{2.91}$	$0.177 \times (T/77)^{2.91}$
4.2	0.12	0.32×10^{-2}

the absorbed polarized light to the nuclear orientation. This also enforces a Boltzmann-like distribution in angular momentum for the relative populations of all sublevels coupled by ME collisions: a so-called spin-temperature distribution. Anderson *et al.* proposed that such a distribution is enforced by fast spin exchange (Anderson, Pipkin, and Baird, 1959; Happer, 1972). For He, whenever the effects of pumping light and relaxation can be neglected, the spin-temperature distribution can be derived from ME equations on density operators (Courtade *et al.*, 2002). Defining $e^\beta = (1 + P_{\text{He}})/(1 - P_{\text{He}})$ as the ratio of the populations in the ground state with nuclear polarization P_{He} ($1/\beta$ plays the role of a spin temperature), one finds that the ME-driven ratios of populations in the 2^3S state of ^3He are field independent and given by $e^{\beta\Delta m_F}$ for any two sublevels with a magnetic quantum number difference Δm_F . Similarly, in an isotopic mixture, the ME-driven ratios of populations for the three magnetic sublevels in the 2^3S state of ^4He are given by $e^{\beta\Delta m_S}$. Therefore, one may write for populations a_i^{st} and y_i^{st} , where the upper index indicates spin-temperature distribution values:

$$a_1^{\text{st}}/a_2^{\text{st}} = a_2^{\text{st}}/a_3^{\text{st}} = a_3^{\text{st}}/a_4^{\text{st}} = a_5^{\text{st}}/a_6^{\text{st}} = e^\beta, \quad (34)$$

$$y_1^{\text{st}}/y_2^{\text{st}} = y_2^{\text{st}}/y_3^{\text{st}} = e^\beta. \quad (35)$$

This yields the following explicit expressions:

$$a_i^{\text{st}} = e^{\beta m_F} / (e^{3\beta/2} + 2e^{\beta/2} + 2e^{-\beta/2} + e^{-3\beta/2}), \quad (36)$$

$$y_i^{\text{st}} = e^{\beta m_S} / (e^\beta + 1 + e^{-\beta}). \quad (37)$$

The spin-temperature values of populations of Eq. (36) can be checked to yield, using Eq. (32), a nuclear polarization in the 2^3S state of $P_{\text{He}^*} = P_{\text{He}}$, as expected.

These distributions of populations have a strong impact on the absorption of light on the $2^3\text{S}-2^3\text{P}$ optical pumping transition. The consequences on the efficiency of MEOP at high nuclear polarization are discussed in Sec. III.C and the resulting features of polarimetry using the optical pumping transition in Sec. III.D.

C. Optical pumping of the $2^3\text{S}-2^3\text{P}$ transition

Besides the ME collisions described, two kinds of processes jointly affect the populations in the 2^3S and 2^3P states: population transfers between Zeeman sublevels within the 2^3S and 2^3P states, usually modeled as relaxation processes, and optical pumping that combines absorption and spontaneous or induced emission of light on the closed $2^3\text{S}-2^3\text{P}$ transition. They are successively considered in the following.

1. Relaxation in the 2^3S and 2^3P states

Population transfers between sublevels occur with very different rates for the two states (Batz, Nacher, and Tastevin, 2011).

In the 2^3S state, the $L = 0$ orbital angular momentum is not affected by collisions: relaxation is slow, with rates γ_i^{S} typically of the order of 10^3 s^{-1} . It is attributed to excitation quenching (e.g., at cell wall or in three-body dimer-forming

collisions) and reexcitation in the gas discharge. Its effect on populations is simply written as

$$\left. \frac{da_i}{dt} \right|_r = \gamma_r^S \left(\frac{1}{6} - a_i \right). \quad (38)$$

In the 2^3P state, J -changing collisions occur at a much faster rate, proportional to gas pressure and of the order of a few $10^7 \text{ s}^{-1}/\text{mbar}$ (Schearer, 1967; Vranceanu, Kotochigova, and Sadeghpour, 2004). These collisions may induce significant population transfer during the radiative lifetime, which is phenomenologically described in MEOP models using a single rate γ_r^P in rate equations ruling the evolutions of the populations b_j of the sublevels B_j :

$$\left. \frac{db_j}{dt} \right|_r = \gamma_r^P \left(\frac{\sum_{k=1}^{18} b_k}{18} - b_j \right). \quad (39)$$

For convenience, the b_j 's are defined so that the number density of atoms in each sublevel B_j of the 2^3P state is $n_m b_j$. Consequently they are not true populations (i.e., diagonal elements of a trace-1 density matrix), and $\sum b_k < 1$ depends on the pumping light intensity. When isotopic mixtures are considered, additional phenomenological equations similar to Eqs. (38) and (39) are used, with possibly different rates for different isotopes due to the thermal velocity difference.

A more realistic treatment of the effects of collisions in the 2^3P state, in particular, at high magnetic field, remains to be implemented and tested in MEOP models (Batz, Nacher, and Tastevin, 2011).

2. Traditional low-field optical pumping

Optical pumping selectively promotes atoms from a subset of Zeeman sublevels of the 2^3S state to corresponding sublevels of the 2^3P state according to selection rules depending on the frequency and polarization of the pumping light. Figure 14(a) displays typical absorption spectra for ^3He and ^4He at 1083 nm.

The action of optical pumping in pure ^4He is quite straightforward: each of the two strong low-energy components of the pumping transition [partly resolved in Fig. 14(a)] can be used to efficiently obtain a high *electronic* polarization in the 2^3S state by depopulating, for instance, the Y_1 and Y_2 sublevels, whereas using the D_0 component depopulates only one sublevel and therefore yields a maximum polarization of 0.5 (Giberson *et al.*, 1982; Wallace *et al.*, 1995). In a similar way, optical pumping can efficiently depopulate selected sublevels of $^3\text{He}^*$ atoms whenever ME collisions have a negligible rate, for instance at low temperature (see Fig. 13) or in atomic beams (Stas *et al.*, 2004). Achieving full nuclear polarization would require depopulating all sublevels except A_1 or A_4 by suitably addressing both hyperfine levels [see Eq. (24) and Fig. 12].

The effect of pumping light in a gas of ^3He or in isotopic mixtures is quite different due to the strong coupling enforced by ME collisions between the Zeeman sublevels of the 2^3S state [Eq. (30)]. In a weak pumping limit, one may consider that $P_{\text{He}^*} \approx P_{\text{He}}$ and derive a simple phenomenological model

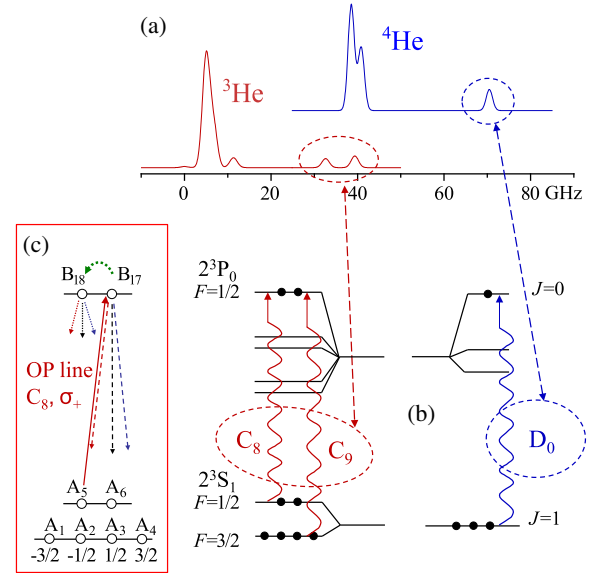


FIG. 14. Low-field metastability-exchange optical pumping transitions: (a) computed absorption spectra for the 2^3S - 2^3P transition for a low-pressure, optically thin gas at room temperature. The linewidths essentially arise from Doppler broadening. (b) Fine and hyperfine sublevels involved in the transition (see Fig. 12). The three transitions to the 2^3P_0 levels are schematically represented, and the names and positions of the corresponding line components in the spectra are highlighted. (c) The inset displays the optical transitions involved for C_8 , σ_+ pumping (straight arrows, see text). The curved arrow represents population transfer between Zeeman sublevels of the 2^3P state. Note that no *direct* transfer, corresponding to a nuclear spin flip, is expected to occur between these two sublevels.

at low P_{He} (Colegrove, Schearer, and Walters, 1963), or more generally consider the effect of optical pumping cycles on the P_{He} -dependent spin-temperature populations of Eqs. (36) and (37) for arbitrary P_{He} . In usual MEOP conditions, ME therefore not only transfers nuclear orientation to the ground-state atoms but also plays a key role in the way the pumping light is absorbed. In that case full polarization could in principle be achieved even if a single Zeeman sublevel is addressed by optical pumping. The actual limit for the polarization results from a balance of angular momentum gain from the pumping light and loss through relaxation. In practice, MEOP is usually performed by tuning the pumping light to one of the high-energy components of the spectra to selectively populate the 2^3P_0 level, using the D_0 component for ^4He or the C_8 or C_9 component for ^3He [see Fig. 14(b)]. These are the most efficient and widely used transitions for pumping with tunable lasers. In the early days of MEOP, pumping light was usually obtained from ^4He lamps to polarize ^3He gas with fair efficiency, thanks to the isotope shift (Colegrove, Schearer, and Walters, 1963).

Figure 14(c) displays in more detail the full optical pumping cycle process in the simple case of the C_8 component. Right-handed circular (σ_+) polarization excites atoms from the A_5 sublevel ($m_F = -1/2$) to the B_{17} sublevel ($m_F = 1/2$). Radiative decay brings atoms back to the 2^3S state by spontaneous and stimulated emissions with

well-defined branching ratios from the directly populated B_{17} sublevel and from any other sublevel that may have been indirectly populated by J -changing collisions, as described in Sec. III.C.1. There are two extreme situations for the impact of these collisions: low-pressure optical pumping (Kastler, 1957) for $\gamma_r^p \ll \gamma$, where $\gamma = 1.022 \times 10^7 \text{ s}^{-1}$ is the radiative decay rate of the 2^3P state, and depopulation optical pumping (Dehmelt, 1957), a high-pressure regime where all 2^3P populations are equalized, for $\gamma_r^p \gg \gamma$.

All radiative processes can be modeled by sets of coupled rate equations for the populations. For ^3He they are written as (Nacher and Leduc, 1985)

$$\left. \frac{da_i}{dt} \right|_{\text{OP}} = \gamma \sum_{j=1}^{18} T_{ij} b_j + \sum_{j=1}^{18} \gamma_{ij} (b_j - a_i), \quad (40)$$

$$\left. \frac{db_j}{dt} \right|_{\text{OP}} = -\gamma b_j + \sum_{i=1}^6 \gamma_{ij} (a_i - b_j), \quad (41)$$

where T_{ij} is the transition matrix element, and γ_{ij} is the optical pumping rate of the transition $A_i \rightarrow B_j$. γ_{ij} is proportional to the pump light intensity and to T_{ij} . On the right-hand sides of these equations, the first terms account for spontaneous emission and the second terms for the net difference between absorption and stimulated emission.

Adding up contributions of ME, relaxation, and radiative processes [Eqs. (30) and (38) to (41)], one obtains a set of coupled rate equations for the populations of all sublevels of the 2^3S and 2^3P states. Given that the rate parameters are in the range 10^3 to 10^7 s^{-1} in these equations, while the rate of change of the nuclear polarization is usually smaller than 1 s^{-1} , steady-state solutions for the populations are adiabatically computed taking P_{He} as a static input parameter.

Figure 15 displays examples of computed populations, pump absorption coefficients, and 2^3S state laser-induced overpolarization for parameters corresponding to typical MEOP experiments. Competition between metastability exchange and optical pumping is illustrated in Fig. 15(a), in which the steady-state populations of the 2^3S state are represented for different values of P_{He} and of the pumping rate γ_{ij} on the C_8 transition component. In that case, absorbed light directly depletes level A_5 while other populations are differently affected, which enforces systematic deviations from the spin-temperature distribution (the surrounding red boxes) that is obeyed in the absence of optical pumping and assumed in simple MEOP models. The effect of relaxation in the 2^3S state on populations is too weak to be seen in this figure.

Figure 15(b) displays the computed variation of the gas absorption coefficient with the reduced pumping rate $\gamma_{ij}/\gamma T_{ij}$ for both C_8 and C_9 components. A strong decrease of the absorption results from the modifications of populations in the 2^3S state as well as from stimulated emission of atoms promoted to the 2^3P state by optical pumping. Absorption also significantly decreases when nuclear polarization increases. It is therefore increasingly difficult to efficiently deposit angular momentum at increasing polarization and pumping intensity.

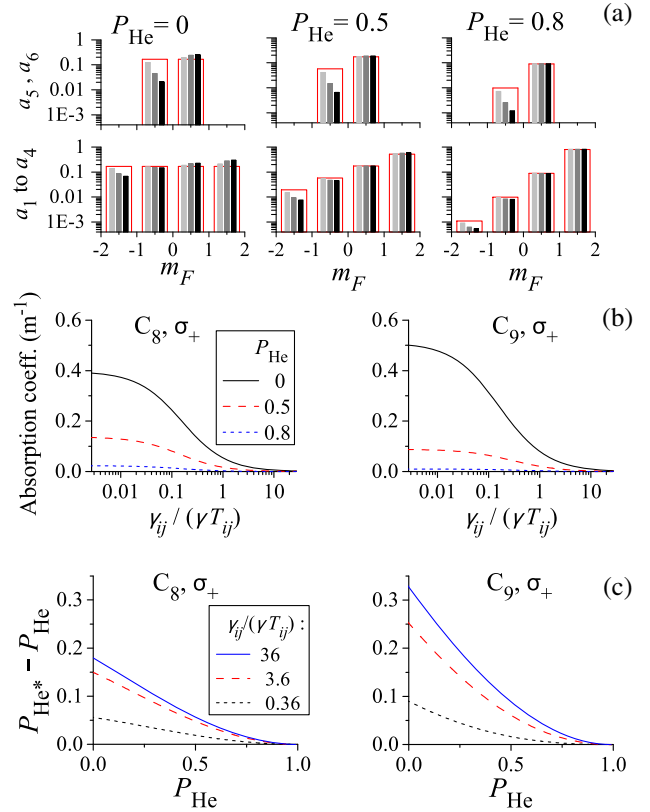


FIG. 15. Results of the MEOP rate equations obtained using the same computer code as in Batz, Nacher, and Tastevin (2011) for typical operating conditions: 0.5 mbar ^3He gas, $T = 293 \text{ K}$, $B = 1 \text{ mT}$, $n_m = 1.2 \times 10^{10} \text{ cm}^{-3}$, $\gamma_r^s = 10^3 \text{ s}^{-1}$, and $\gamma_r^p = 0.16 \times 10^7 \text{ s}^{-1}$. Atoms of all velocity classes are pumped with the same rates γ_{ij} (see text). (a) Distributions of the 2^3S populations a_i for C_8, σ_+ optical pumping and $\gamma_{ij} = \gamma/10, \gamma$, and 10γ (bars from left to right and from light gray to black in each group). Surrounding red boxes are the spin-temperature populations. (b) Computed absorption coefficients are plotted as functions of reduced pumping rates for C_8 and C_9 optical pumping and different nuclear polarizations (see the legend). For C_9 the two pumping rates for the two pumped sublevels jointly scale with the pumping light intensity, with a fixed ratio $T_{1,18}/T_{2,17} = 3$. Introducing T_{ij} in the horizontal scales makes the reduced pumping rates identically proportional to the pump intensity. (c) Differences between nuclear polarizations in the 2^3S and ground states are plotted as a functions of P_{He} for C_8 and C_9 pumping and different reduced pumping rates (see the legend).

Another meaningful quantity derived from the solutions of the rate equations is represented in Fig. 15(c): the nuclear overpolarization $P_{\text{He}^*} - P_{\text{He}}$, which acts as a driving term for the ground-state polarization P_{He} in Eq. (31). The parameters in Eq. (32) are $(L_k) = (-1, L_-, L_+, 1, -L_-, -L_+)$, with $L_{\pm} \approx \pm 1/3$ in low field. Therefore P_{He^*} is fairly sensitive to the modifications of all populations enforced by optical pumping in the 2^3S state. The decrease of this driving term with P_{He} and its saturating increase at high pumping rates mainly result from the decrease in pumping light absorption. Introducing the optical pumping photon efficiency η as the net change of atomic angular momentum projection m_F upon the absorption and emission of a photon, these correlated decreases can be

quantitatively linked (Batz, Nacher, and Tastevin, 2011). For C_8 , η has a fixed value ranging from 0.5 to 0.9 depending on γ_r^p , while for C_9 η also depends on P_{He} and the optical pumping rate, ranging from 0.5 to 1.25.

So far the pumping rates have not been explicitly related to the pumping light parameters. They depend on the local characteristics of the pumping light (intensity, direction of propagation, spectral characteristics) and on the atomic velocity projection v on the direction of propagation of the pumping light. Velocity-selective optical pumping, i.e., a strong correlation between v and populations or P_{He^*} , may occur if an intense narrow-band laser is used, with a spectral width L smaller than the Doppler width D (Pinard, Aminoff, and Laloč, 1979; Aminoff and Pinard, 1982). Velocity-dependent pumping rates $\gamma_{ij}(v, \mathbf{r})$ can be computed for a single-frequency pump laser (Courtade *et al.*, 2002). The maximum rate is experienced by atoms in the velocity class v^* that is resonant with the laser frequency and is given by

$$\gamma_{ij}(v^*, \mathbf{r}) = \frac{2\pi\alpha f}{m_e\omega\gamma^2} \frac{\gamma}{\Gamma'/2} I_{\text{las}}(\mathbf{r}) T_{ij}\gamma, \quad (42)$$

where $I_{\text{las}}(\mathbf{r})$ is the laser intensity, ω its angular frequency, α is the fine-structure constant, f is the oscillator strength of the transition, and m_e is the electron mass; the numerical value of the first fraction is $0.149 \text{ m}^2/\text{W}$. The total damping rate of the optical coherence of the transition $\Gamma'/2 = \gamma + \pi w$ results from the combined effects of the radiative decay rate γ and of the pressure-dependent collisional broadening w , with a value for w/p of the order of 20 MHz/mbar (Vrinceanu, Kotochigova, and Sadehpour, 2004) or 12 MHz/mbar (Nikiel-Osuchowska *et al.*, 2013) that remains to be confirmed. In the low-pressure limit ($w \ll \gamma$), $\gamma_{ij} = T_{ij}\gamma$ for $I_{\text{las}} = 6.7 \text{ W/m}^2$.

Pumping rates can also be evaluated for broadband lasers of known spectral intensity distribution. For instance, for a laser of Gaussian width L tuned to the center of the Doppler absorption profile, the rate is (Batz, Nacher, and Tastevin, 2011)

$$\gamma_{ij}(v, \mathbf{r}) = \frac{2\pi\alpha f}{m_e\omega\gamma^2} \frac{\gamma}{2\sqrt{\pi}L} \exp\left[-\left(\frac{Dv}{L\bar{v}}\right)^2\right] I_{\text{las}}(\mathbf{r}) T_{ij}\gamma, \quad (43)$$

where $\bar{v} = \sqrt{2k_B T/M}$ is the most probable speed and $D = (\omega/2\pi)\bar{v}/c$ is the associated Doppler width. Because of the second factor on the right-hand side, the rates are typically 100 to 1000 times smaller than in Eq. (42) for a given pumping intensity. This accounts for the much higher total power absorption for a pump laser with a suitable bandwidth $L \approx D$ (Tastevin *et al.*, 2004).

One may be tempted to make an exact treatment of velocity-selective optical pumping effects by explicitly keeping a dependence of populations and collision rates on the atomic velocity projection v . However, solving sets of rate equations for various velocity classes would be a difficult task since their populations are coupled by velocity changing and by ME collisions, with ill-known collision rates. Instead, a coarse description with only two broad velocity classes has been proposed to account for the Maxwell distribution of atomic velocities in the pumped gas: strongly pumped atoms, in the

center of the velocity distribution, and weakly pumped atoms, in the wings of the velocity distribution (Nacher and Leduc, 1985; Batz, Nacher, and Tastevin, 2011). Each class is pumped with an effective pumping rate depending on the pump spectral profile, and their populations are coupled by ME and velocity-changing collisions. This is a crude model; however, it is sufficient to capture key features of velocity-selective optical pumping effects with few free parameters and it usually provides meaningful quantitative results.

Velocity-independent optical pumping models can be reliably used in the following specific cases: if (i) broadband (“white”) pumping light is used, with L sufficiently larger than D , or if (ii) the rate of change of populations from Eqs. (40) and (41), $\gamma\gamma_{ij}/(\gamma + \gamma_{ij})$, is much smaller than ME and velocity-changing collision rates. Condition (ii) is automatically fulfilled if $\gamma \ll \gamma_e$. Because γ_e scales with gas pressure p , high-pressure optical pumping is immune to velocity-selective effects and a single set of populations can be used to locally characterize the effect of MEOP on the gas. In pure ^3He at room temperature, $\gamma_e = \gamma p/p^*$, where $p^* = 2.72 \text{ mbar}$ is the crossover pressure for which $\gamma_e = \gamma$ (from data in Sec. III.B.2). At lower pressure, a velocity-independent optical pumping regime is obtained only for laser intensities much smaller than a crossover value depending on pressure and laser linewidth, which can be evaluated using for instance a two-class model.

Indeed, during experiments where polarization decay occurs following the interruption of optical pumping, velocity-independent values of the populations are expected to be enforced by ME collisions as well.

3. High-field optical pumping

As mentioned above (Sec. III.A), the angular momentum structures of the 2^3S and 2^3P levels and the 1083 nm transition are deeply modified in high magnetic field. Figure 16 displays the absorption spectra and the energies of all Zeeman sublevels of ^3He for $B = 1.5 \text{ T}$, a field strength commonly met in MRI systems and thus of practical importance for applications. The six sublevels of the 2^3S state are organized in three pairs of states [Figs. 16(b) and 16(c), bottom graphs]. In each pair the sublevel energy is mostly determined by the common dominant value of m_S and the nuclear spin projections are almost opposite. Similarly, hyperfine coupling only weakly mixes levels of different m_I values in the 2^3P state [Figs. 16(b) and 16(c), upper graphs]. As a result, high-field spectra for a given light polarization comprise six main components clustered in two groups: a pair and a quartet, each group being unresolved at room temperature [Fig. 16(a)].

In high field, the parameters in Eq. (32) are $(L_k) = (-1, L_-, L_+, -L_-, 1, -L_+)$, with $L_{\pm} \approx 1$; the change in the order of levels results from crossings occurring around 0.16 and 4.7 T. Simplified models as well as full resolution of the rate equations of MEOP show that a large number of ME collisions, proportional to B^2 , are required to transfer a single unit of angular momentum between ground-state and 2^3S state atoms (Courtade *et al.*, 2002; Abboud *et al.*, 2004). For instance, at null ground-state polarization, strong pumping with the f_2^- pair of lines may depopulate the A_5 and A_6 sublevels while $a_1 = \dots = a_4 = 1/4$ if the spin temperature

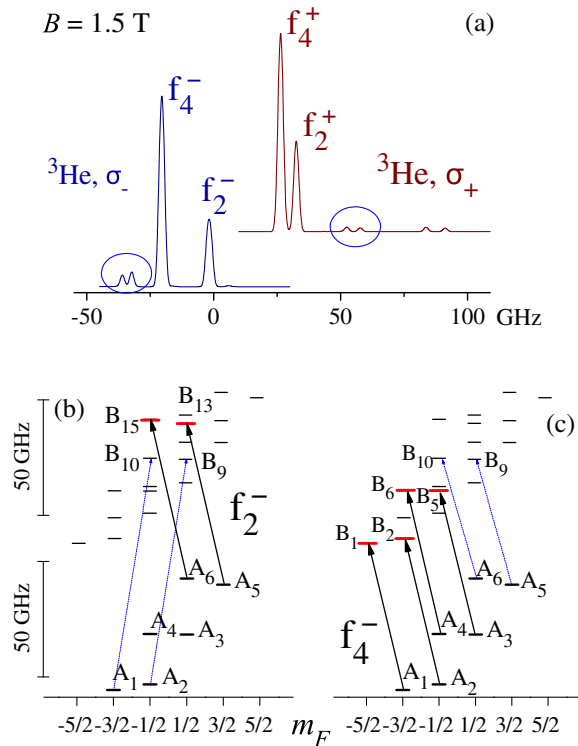


FIG. 16. High-field metastability-exchange optical pumping transitions: (a) Computed absorption spectra at $B = 1.5$ T for the $2^3\text{S}-2^3\text{P}$ transition (low-pressure, optically thin gas at room temperature) for both circular polarizations. The strong unresolved components in the spectra are labeled f_n^\pm , where $n = 2$ or 4 refers to the number of involved transitions and \pm to the sign of the circular polarization. Doublets of resolved weaker transitions of interest for optical detection purpose are highlighted. (b), (c) Energies of the ^3He sublevels at 1.5 T for the 2^3S and 2^3P states. The transitions induced by the σ_- -polarized pumps (thick lines) and the suitable probes (thin lines) are displayed [f_2^- pump, σ_+ probe in (b); f_4^- pump, σ_- probe in (c)].

is still obeyed for the unpumped sublevels. Hence $P_{\text{He}^*} = (L_+ - 1)/4 = -\sin^2\theta_+/2$ with $\sin^2\theta_+ = 5.62 \times 10^{-4}$ at 1.5 T. Similarly, strong f_4^- pumping depopulates A_1 to A_4 , so that $a_5 = a_6 = 1/2$ and $P_{\text{He}^*} = (1 - L_+)/2$. f_4^- pumping is thus expected to yield *positive* nuclear polarization in spite of its negative circular polarization, with a (twice) faster pump-up rate, in fair agreement with experimental observations (Abboud *et al.*, 2005; Nikiel *et al.*, 2007). Over a wide range of field strengths and gas pressures, σ_- pumping has been reported to be more efficient, possibly because the f_4^+ and f_2^+ groups are imperfectly resolved, especially at high pressure, and have opposite polarizing actions. The σ_- pumping schemes are thus detailed in Figs. 16(b) and 16(c), together with the corresponding probing transitions discussed in Sec. III.D.2.

The photon efficiency of these high-field optical pumping schemes can be as high as in low field ($\eta \approx 1$ for f_2^- pumping at high pressure) but this remains to be systematically studied (Batz, Nacher, and Tastevin, 2011; Nikiel-Osuchowska *et al.*, 2013). With modified He level structures and only weak hyperfine couplings remaining in the excited states, high-field

MEOP has two distinct features: highly absorbing lines can be used, with absorption coefficients ≈ 10 times larger than for C_8 or C_9 pumping at low field, and the transfer of angular momentum to the ground state is orders of magnitudes slower than in low field at a given pressure. For this last point, ME collisions fail to enforce a strong coupling between the 2^3S state populations, which is reminiscent of the low-field, low-temperature situation with its ME cross sections orders of magnitude lower than at room temperature.

It was initially argued that, due to the weak hyperfine coupling, MEOP would be inefficient in high field and early attempts were driven by applications and were limited to moderate field strengths: 0.1 T (Flowers, Petley, and Richards, 1990; Darrasse *et al.*, 1997) and 0.6 T (Flowers *et al.*, 1997). It was only later that a key benefit of high-field MEOP was recognized: its efficient operation at high gas pressures (Courtade *et al.*, 2000). More recently, high-field MEOP of ^3He at low pressure has been shown to be efficient as well (Abboud *et al.*, 2004; Nikiel *et al.*, 2014; Maxwell *et al.*, 2016). To date, no study of high-field MEOP in isotopic mixtures has been reported. Ultimately, the performance of MEOP for weakened ME coupling crucially depends on the balance between impeded polarization transfer and reduced polarization losses in the ground state, which will be discussed in Sec. III.E.

D. Optical methods of polarimetry

The measurement of the nuclear polarization of ^3He gas in optical pumping cells can be made using a direct magnetic measurement (see Sec. V). However, optical methods relying on polarimetry of the visible fluorescence light emitted by He gas or on 1083 nm light absorption measurements are more widely used since they are simple to perform, provide high sensitivity, and operate in the presence of the discharge sustained for MEOP.

1. Fluorescence light polarimetry

Nuclear spin polarization is conserved during collisions which excite atoms from the ground state and hyperfine interaction subsequently couples electronic and nuclear orientations in the excited states. The electronic orientation and therefore the degree of circular polarization of the fluorescence light emitted by atoms losing their excitation both reflect the value of the ground-state polarization (Pavlović and Laloë, 1970). The circular polarization P_{fluo} of the fluorescence light emitted along the direction of B is experimentally monitored using a rotating quarter-wave plate and a fixed linear polarizer (Pinard and van der Linde, 1974; Bigelow, Nacher, and Leduc, 1992; Lorenzon *et al.*, 1993), a two-channel polarimeter consisting of a static quarter-wave plate and a beam-splitting polarizer (Stoltz, Villard *et al.*, 1996), or an electromodulated polarimeter comprising a liquid crystal wave retarder (Maxwell, Epstein, and Milner, 2014). Lights of two spectral lines, at 668 and 588 nm, are significantly polarized and yield high signal-to-noise-ratio (SNR) measurements, with typical polarization ratios $P_{\text{fluo}}/P_{\text{He}} \approx 0.1$ (Stoltz, Villard *et al.*, 1996). The ratios significantly decrease with B above about 10 mT due to the weakening of hyperfine

coupling efficiency and decrease with gas pressure p . In spite of these variations and of its sensitivity to the discharge strength for the 588 nm line, fluorescence polarimetry is a convenient and accurate polarization measurement technique for low- p and low- B MEOP experiments. The two existing calibrations of the technique, performed at 668 nm using different methods (Bigelow, Nacher, and Leduc, 1992; Lorenzon *et al.*, 1993), agree within typical uncertainties of 2%.

2. Light absorption polarimetry

Since the early days of MEOP (Colegrove, Schearer, and Walters, 1963; Greenhow, 1964; Daniels and Timsit, 1971), 1083 nm light absorption measurements have been used to evaluate nuclear polarization in cells. The method is based on the fact that the ME-enforced spin-temperature distribution establishes a strong link between P_{He} and 2^3S state populations [Sec. III.B.3, Eqs. (36) and (37)]. Probing absorption by two suitable Zeeman sublevels, or combinations of sublevels, is sufficient to infer the spin temperature β , and hence P_{He} , from the ratio of measured absorption coefficients. Additionally, the 2^3S state density n_m integrated over the probe beam path can usually be inferred as well from such measurements.

At low B , two different probe beam polarizations are used to address different sublevels. For a longitudinal beam, propagating along \mathbf{B} , σ_+ and σ_- polarizations selectively probe a_5 and a_6 when tuned to C_8 or $a_1 + a_2/3$ and $a_4 + a_3/3$ when tuned to C_9 . For a transverse beam, the orthogonal linear polarizations ($\pi: \perp \mathbf{B}$ and $\sigma: \parallel \mathbf{B}$) provide independent combinations of populations only when tuned to C_9 . In isotopic mixtures the D_0 transition component can be used in both geometrical arrangements. For longitudinal probe polarimetry the ratios $r_i^{\parallel} = A_+/A_-$ of absorption coefficients for σ_+ and σ_- light (the subscript i is 8 or 9 for C_8 or C_9 , 0 for D_0) are

$$r_8^{\parallel} = \frac{1 - P_{\text{He}}}{1 + P_{\text{He}}}, \quad r_9^{\parallel} = \frac{2 - P_{\text{He}}}{2 + P_{\text{He}}} r_8^{\parallel 2}, \quad r_0^{\parallel} = r_8^{\parallel 2}. \quad (44)$$

This configuration offers the highest sensitivity at low polarizations, with linear coefficients -2 , -5 , and -4 , respectively. For transverse probe polarimetry the ratios $r_i^{\perp} = A_{\pi}/A_{\sigma}$ of absorption coefficients for π and σ light are

$$r_9^{\perp} = \frac{1 - P_{\text{He}}^2}{1 + 2P_{\text{He}}^2}, \quad r_0^{\perp} = \frac{1 - P_{\text{He}}^2}{1 + P_{\text{He}}^2}. \quad (45)$$

The transverse configuration has a weak sensitivity at low polarizations since it depends only on P_{He}^2 . This is actually a useful feature for measurements performed while an intense pumping light is applied: the method is almost insensitive to the potentially large pump-induced deviations of populations from their spin-temperature values, such as displayed in Fig. 15 (Talbot *et al.*, 2011).

As is done for fluorescence polarimetry, a mechanically or electrically modulated polarization or a two-channel static scheme can be used for light absorption polarimetry. The probe beams must be weak enough so as not to locally affect the measured populations (low intensity) nor globally deposit

angular momentum of the wrong sign (low absorbed power) (Talbot *et al.*, 2011). In static schemes, a small geometrical separation of the two probe beams should be used to avoid coherently addressing common levels with the two beams, which has been observed to induce sizable artifacts in the measurements (Talbot *et al.*, 2011).

At high B , all energy degeneracies of the Zeeman sublevels are lifted and two different probe beam frequencies are used to address different sublevels. Because of hyperfine decoupling, ME collisions imperfectly enforce the link between P_{He} and the 2^3S state populations, so that systematic measurement errors may appear even for moderate pump intensities. Fortunately, an adequate choice of optical transitions can be made to avoid such difficulties, by probing a pair of sublevels that are not addressed by the pump (Abboud *et al.*, 2004; Suchanek *et al.*, 2007; Nikiel-Osuchowska *et al.*, 2013). This is achieved using the convenient line doublets highlighted in Fig. 16(a): a periodic frequency sweep of the probe (at fixed circular polarization) sequentially yields values of two absorption coefficients that are used to infer both n_m and P_{He} .

In spite of the need for a probe laser at 1083 nm, absorption polarimetry is preferred for high-field or high-pressure situations, and for measurements in isotopic mixtures, since it yields accurate results if adequate care is taken.

E. Relaxation in MEOP cells and steady-state polarizations

1. Discharge-induced polarization decay

MEOP apparatuses and cells are designed so as to avoid significant polarization loss due to diffusion in field inhomogeneities or wall relaxation. When the discharge used to populate the 2^3S state is off, relaxation times of one to several hours are usually achieved. When the discharge is on and the pumping light is interrupted following polarization buildup, decay times $1/\Gamma_D$ of the order of 1 min are typical. As the discharge intensity is increased, the metastable density n_m and thus the polarizing rate increases but the achievable ^3He polarization usually decreases. These dependences vary with gas pressure and discharge frequency; Gentile and McKeown (1993) reported on studies with pressures between 0.13 and 6.5 mbar and frequencies between 0.1 and 10 MHz. Several other groups studied the influence of cell size and shape, configuration of external discharge electrodes, and frequency: over a wide range of pressures (up to hundreds of mbars) and fields (up to 4.7 T), n_m and Γ_D are found to positively correlate, with n_m increasing less rapidly than Γ_D with the discharge excitation power (Batz, 2011; Nikiel-Osuchowska *et al.*, 2013).

The effect of an applied field exceeding usual low holding fields depends on pressure. At low pressures ($p < 1$ mbar), the ratios Γ_D/n_m increase in the applied field (at 30 mT and above). At higher pressures, on the contrary, Γ_D/n_m decreases in the applied field, which is potentially beneficial for efficient MEOP. For instance, at $p = 2.45$ mbar, Γ_D is decreased by a factor of 2 to 4 at fixed n_m for $B = 30$ mT (Batz, 2011). At higher p (tens of mbar) and B (1.5 T) the decrease can exceed a factor of 10. However, at such high pressures, the radial distribution of n_m has an inverted distribution (with a

minimum on cell axis) correlated to a plasma localization near the cell wall (Dohnalik *et al.*, 2011). This distribution is sensitive to B , which makes it delicate to quantitatively compare Γ_D/n_m ratios between low and high B .

Part of the polarization decay is due to relaxation in the 2^3S state, which drives a flow of angular momentum from the ground-state reservoir with a rate Γ_{ME} . The remainder of the decay, with a rate Γ_g , is due to various relaxation processes directly affecting polarization in the ground state, for instance collisions with excited ionic or molecular He, or electronic excitation and loss of angular momentum by emission of polarized light in the radiative cascade (see Sec. III.D). Altogether, the decay rate is

$$\Gamma_D = \Gamma_{\text{ME}} + \Gamma_g. \quad (46)$$

Γ_{ME} scales as the volume average over the cell of $\gamma_r^S n_m / N_g$ with a P_{He}^2 -dependent computed factor equal to 11/3 in pure ^3He at low polarization (Batz, Nacher, and Tastevin, 2011). Moreover, n_m is experimentally found to depend on P_{He}^2 during polarization decays at fixed discharge excitation levels, changing by up to $\pm 20\%$. The density n_m depends on the local balance of excitation to the 2^3S state in the discharge (involving ionization by electron impact, recombination, and radiative cascade), local decay through various processes (e.g., quenching by chemical impurities in the helium gas, three-body conversion to metastable He_2 molecules, Penning ionizing collisions), and atomic diffusion in the helium gas combined with excitation loss at the cell wall. Optogalvanic effects may also play a role in the balance of processes occurring in the plasma. Therefore n_m may depend on P_{He}^2 and on MEOP conditions due, for example, to the influence of electronic orientation on the cross section of Penning collisions (Hill *et al.*, 1972; Fedichev *et al.*, 1996). In spite of the expected P_{He}^2 dependency of Γ_{ME} , decays are systematically observed to be accurately exponential with P_{He} -independent rates Γ_D over a wide range of gas pressures and field strengths, a so far unexplained observation (Batz, Nacher, and Tastevin, 2011; Nikiel-Osuchowska *et al.*, 2013).

2. Optical-pumping-induced polarization loss

When the pumping light is applied, the time evolution of P_{He} as well as its steady-state value $P_{\text{He}\infty}$ can be inferred from the balance between the inflow of angular momentum of Eq. (31) and the loss term $-\Gamma_g P_{\text{He}}$. This approach provides a detailed insight on the key relevant processes at play in MEOP, but it requires providing a trusted MEOP model with values for all physical and phenomenological parameters. A more pragmatic approach can be preferred for its robustness: a global angular momentum budget in which the rate of change of the ground-state nuclear polarization is written as the net balance between the angular momentum actually transferred through optical pumping cycles to the 2^3S atoms and an angular momentum loss directly associated with the ground-state atoms (Batz, Nacher, and Tastevin, 2011):

$$\frac{dP_{\text{He}}}{dt} = 2\eta \frac{W_{\text{abs}}}{N_g V_{\text{cell}} \hbar \omega} - \Gamma_R P_{\text{He}}, \quad (47)$$

where W_{abs} is the pumping light power absorbed by the gas. The global polarization loss rate Γ_R introduced in this approach is not a constant but may vary with P_{He} and MEOP conditions as does the photon efficiency η . Equation (47) involves known parameters and experimentally measurable quantities (P_{He} and W_{abs}), plus two unknown ones: η and Γ_R . The photon efficiency η can be evaluated using a MEOP model or directly inferred from transient buildup measurements at $P_{\text{He}} = 0$ for which the loss term in Eq. (47) vanishes. For C_8 or D_0 optical pumping in low field, where η does not vary with P_{He} , Γ_R can thus be fully inferred from experimentally determined quantities.

This pragmatic approach has been used to analyze experimental data for which the absorbed pump power was measured. Figure 17 displays selected results obtained at various pressures in low and high fields. Data obtained in steady state [once $P_{\text{He}\infty}$ has been reached, with a null left-hand side term in Eq. (47)] as well as data collected during polarization buildups toward $P_{\text{He}\infty}$ (three series of closely clustered symbols) are displayed. Despite the qualitatively different behavior of buildups, exponential approaches toward $P_{\text{He}\infty}$ at high fields (Nikiel-Osuchowska *et al.*, 2013) contrasting with nonexponential buildups having decreasing rates at high P_{He} (Gentile and McKeown, 1993; Batz, Nacher, and Tastevin, 2011), all corresponding data nicely collapse with the steady-state data. The decay rates Γ_D for the various experiments tend to increase with pressure, and are strongly reduced, at fixed p , in high field (inset in Fig. 17). The polarization loss rates Γ_R are found to significantly differ from Γ_D for most of the data points. The differences $\Gamma_R - \Gamma_D$

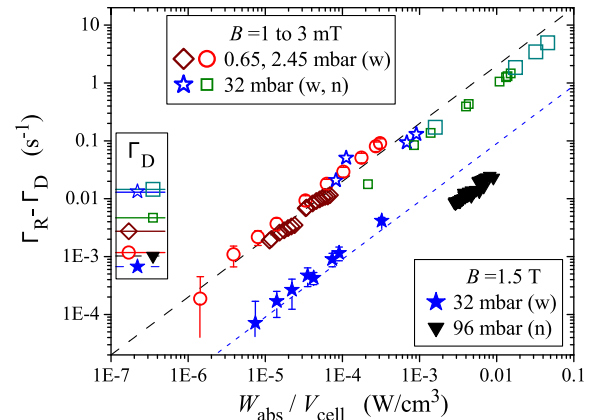


FIG. 17. Optical-pumping-induced additional loss rates vs absorbed pumping power per unit volume for various gas pressures and fields (see the legends) and cell diameters: wide (w: ≥ 5 cm) and narrow (n: 1.5 cm). Each of the two logarithmic scales spans six decades. The lines are guides for the eye corresponding to linear variations with linear coefficients $200 \text{ cm}^3/\text{J}$ (dashed line, low B) and $9 \text{ cm}^3/\text{J}$ (dotted line, 1.5 T). The ordinates of the horizontal lines in the box next to the left axis are the values of the pumping-free decay rates Γ_D for the different sets of data. They range from $0.67 \times 10^{-3} \text{ s}^{-1}$ (solid stars) to $14.5 \times 10^{-3} \text{ s}^{-1}$ (large open squares). The figure compiles data from Mainz (\diamond), Cracow (\blacktriangledown), and Paris (all other symbols); the small and large squares stand for weak and strong discharges in the same cell. Adapted from Batz, 2011, Fig. 6.62.

plotted in Fig. 17 for low-field data (open symbols) span over 4 orders of magnitude when experimental parameters are varied. The absorbed power W_{abs} is varied by changing the incident power or the absorption coefficient, through n_m (in steady state) or P_{He} (during buildup); different cell diameters yield different cell volumes V_{cell} . The pumping-induced additional relaxation rates reveal a consistent linearlike variation only when plotted versus the ratio $W_{\text{abs}}/V_{\text{cell}}$. The 1.5 T data display a similar pumping-enhanced relaxation behavior, but with a significantly reduced rate coefficient (the two lines in Fig. 17 correspond to linear laws with a ratio 30 of their slopes). Data obtained at 30 mT (Batz, 2011, Fig. 6.59) for p up to 2.45 mbar are identical to the corresponding low- B results. This field strength is sufficient to suppress the angular momentum loss which occurs in the radiative cascade and plays a key role in fluorescence polarimetry. The absence of effect of a 30 mT field on the data suggests that angular momentum loss in the radiative cascade plays no role in the pumping-induced polarization loss, but that larger hyperfine structures are involved.

The physical processes causing such strong pumping-enhanced polarization losses remain to be elucidated. Radiation trapping (i.e., reabsorption of 1083 nm fluorescence light) or plasma modification by light-enhanced creation of a relaxing long-lived species through the 2^3P state have been considered as possible origins of pumping-induced loss mechanisms, but they could not account for the observations (Batz, 2011).

3. Steady-state polarization limits

Since the development of powerful lasers for MEOP, it has been noted that the achievable steady-state polarizations $P_{\text{He}\infty}$ are in practice obtained at moderate laser intensities and that higher intensities only yield faster buildup rates. Moreover, very high $P_{\text{He}\infty} > 0.8$ can be obtained only in a narrow pressure range. Figure 18 displays a compilation of steady-state polarizations obtained at low fields (open symbols) and high fields (filled symbols, $B \geq 1$ T) over a wide range of pressures. A moderate field increase, just sufficient to impede angular momentum loss in the radiative cascade (30 mT, half-filled circles), reduces Γ_{D} and increases $P_{\text{He}\infty}$ for weak discharges, but the rapid decrease of $P_{\text{He}\infty}$ with pressure is still observed. At higher fields, on the contrary, large polarizations can be achieved at 10- to 100-fold higher pressures.

Using the steady-state solution of Eq. (47) to relate $P_{\text{He}\infty}$ to the absorbed power W_{abs} , one obtains strikingly different behaviors using for Γ_{R} an optical-pumping-independent loss rate, such as Γ_{D} , or the pumping-enhanced loss rate which has been consistently inferred from observations at high laser powers. The steep decrease of $P_{\text{He}\infty}$ with p above a few mbar is not reproduced by MEOP models assuming fixed losses. For instance, $P_{\text{He}\infty} > 0.8$ instead of 0.5 to 0.6 would be achieved for $p = 2.45$ mbar (Batz, 2011, Fig. 6.47). On the contrary, if the approximate scaling $\Gamma_{\text{R}} - \Gamma_{\text{D}} \propto W_{\text{abs}}/V_{\text{cell}}$ (the lines in Fig. 17) is substituted in Eq. (47) as a lower bound for Γ_{R} , an upper bound for $P_{\text{He}\infty}$ is found to be independent of W_{abs} , hence of the incident laser power, and to decrease as $1/p$ with pressure. The lines in Fig. 18 are upper bounds corresponding to the

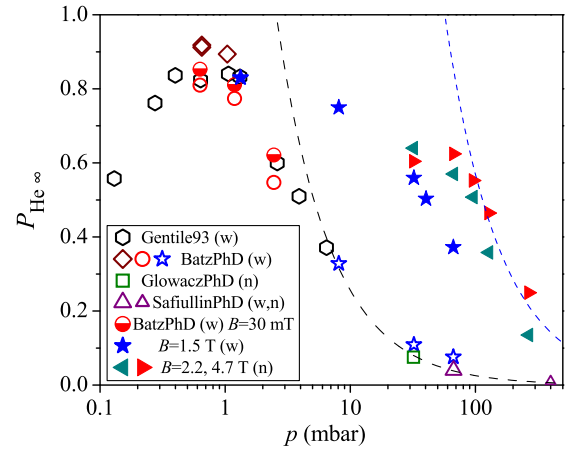


FIG. 18. Variation with pressure of highest steady-state polarizations achieved by various groups at low fields (open symbols, 1 to 3 mT) and high fields (filled symbols, see the legend). The two lines ($P_{\text{He}} = 2.56/p$ and $57/p$) are upper bounds derived from the lines in Fig. 17 (see text). The figure compiles data from Caltech (\circ), Mainz (\diamond), Cracow (\blacktriangleleft , \blacktriangleright), and Paris (all other symbols). Wide and narrow cells (w, n): see Fig. 17. Adapted from Batz, 2011, Fig. 6.48, with additional data from Glowacz, 2011 and Safiullin, 2011.

lines in Fig. 17 using $\eta = 1.25$ in Eq. (47). The angular momentum budget approach therefore provides a link between the observed pumping-enhanced polarization losses of the previous section and the limits in the experimentally achieved $P_{\text{He}\infty}$, especially at high pressures. Additionally, the spectacular increase in efficiency of MEOP at high pressure in high field can be attributed to the increasingly strong reduction of the pumping-enhanced loss for $B > 1$ T.

Achieving very high polarizations in low fields requires operating at a suitably low pressure with pumping light tuned to the most efficient transition component (usually C_8 or D_0) and carefully tailored to the needs as follows:

- (1) a bandwidth of the order of the Doppler width to avoid velocity-selective pumping and the associated optical saturation,
- (2) a spatial transverse profile matched to the pressure-dependent density profile of the 2^3S state atoms, and
- (3) a very high degree of circular polarization because sub-% fractions of an opposite polarization are more efficiently absorbed and thus have a strong adverse effect at high P_{He} (Leduc *et al.*, 2000).

At higher fields and pressures, stronger transition components are the most efficient [f_2^- or f_4^- with a slight detuning from the composite line center (Abboud, 2005, Chap. 6)] and the constraints 1 and 3 listed above can be somewhat relaxed thanks to collisional broadening and to large Zeeman splittings making the opposite polarization nonresonant. Recently, MEOP relying on the $2^3\text{S}-3^3\text{P}$ transition at 389 nm instead of the usual 1083-nm transition was demonstrated (Maeda *et al.*, 2010). It was found to yield much lower polarizations than standard MEOP for comparable laser powers; this may be due to the smaller line component splittings (Sulai *et al.*, 2008) and larger Doppler width for this transition which prevents selecting an isolated efficient component.

F. Lasers for MEOP

A succession of continually improving lasers has been used for MEOP. For traditional, low-field MEOP a match to the 2 GHz Doppler-broadened absorption width is ideal. The first work in laser optical pumping was performed with color center lasers (Nacher and Leduc, 1985) and argon-ion pumped neodymium-doped lanthanum magnesium hexaluminate (Nd:LMA) lasers (Eckert *et al.*, 1992). Velocity-selective optical pumping was an issue for narrow-band lasers, but increased efficiency was obtained by frequency modulation (Elbel *et al.*, 1990). Frequency stabilization to the helium absorption lines was demonstrated (Heil *et al.*, 1990). These early approaches were superseded in both performance and simplicity by the arc-lamp-pumped Nd:LMA laser, which was constructed by replacing the neodymium-doped yttrium aluminum garnet (Nd:YAG) crystal rod in a commercial arc-lamp-pumped laser and adding one or two etalons to produce a 2 GHz linewidth (Schearer and Tin, 1990; Aminoff *et al.*, 1991). Tuning the laser was accomplished by varying the temperature and/or angle of the etalons. These lasers produced an output of several watts, but performance was highly dependent on crystal quality. Although the spectrum was actually a series of narrow peaks with substantial jitter the influence of velocity-changing collisions yielded efficient optical pumping (Elbel *et al.*, 1990; Gentile, Hayden, and Barlow, 2003), resulting in a substantial improvement in polarization and pumping rate (Gentile and McKeown, 1993). More recently Nd:LMA has been superseded by Yb fiber lasers (Chernikov *et al.*, 1997; Mueller, 2001; Gentile, Hayden, and Barlow, 2003; Tastevin *et al.*, 2004). A Yb fiber oscillator and amplifier, properly constructed for MEOP, yields a fairly uniform spectrum with a ≈ 2 GHz spectral width. An alternative scheme is to use a diode laser oscillator and Yb fiber amplifier, which requires modulation for efficient optical pumping (Mueller, 2001). A comparable performance for a given power is obtained for Nd:LMA and both types of Yb lasers (Gentile, Hayden, and Barlow, 2003), but Yb lasers have higher available power as well as greater convenience and a smaller footprint. For small sealed cells, 1083 nm diode lasers (≈ 50 mW) have been used to produce up to 40% ^3He polarization in pure ^3He and 80% in ^3He - ^4He mixtures (Stoltz, Meyerhoff *et al.*, 1996).

G. Compression approaches

Application to polarized targets for charged particle and photon scattering, MRI, and neutron spin filters requires compressing ^3He gas polarized by MEOP. Following the path of an early attempt to use a Toepler pump (Timsit *et al.*, 1971), a successful compressor was developed for electron scattering at MAMI⁷ (Eckert *et al.*, 1992; Becker *et al.*, 1999). This compressor employed a mercury column to compress gas polarized by MEOP at a pressure of ≈ 1 mbar into a 100 cm³ target cell at ≈ 1 bar. For this device 50% ^3He polarization was achieved. At Mainz, this apparatus was replaced by a titanium piston compression apparatus (Becker *et al.*, 1994; Surkau

et al., 1997) that achieved pressures of a few bar. With subsequent improvements this compressor could produce between 60% and 80% ^3He polarization with corresponding polarizing rates of between 4 and 1 bar L/h (Batz *et al.*, 2005). For this apparatus, the best achievable polarization in the open compression system under static conditions was 84%, not far from the record high value of 91% achieved in a sealed cell. At Indiana University an aluminum piston compressor developed for neutron spin filters reached 50% ^3He polarization (Hussey *et al.*, 2005). Both the Mainz and Indiana University apparatuses employed two stages of compression with a buffer cell between the stages. In these and subsequent piston compressors, the polarization loss in the compressor itself was a few percent or less, but additional polarization losses are incurred due to gas flow and relaxation in the buffer cell, storage cell, and interconnecting tubing. A similar compressor was installed at the ILL,⁸ but eventually replaced by a new apparatus, denoted “Tyrex,” for filling neutron spin-filter cells. With further improvements, a typical value of 75% ^3He polarization has been obtained in NSF cells (Lelievre-Berna, 2007). A compression system denoted HELIOS was put into operation at the FRM II reactor in Munich (Hutanu, Masalovich *et al.*, 2007) and more recently a compression apparatus is also planned for use at ISIS (Beecham *et al.*, 2011).

In parallel with the development of these large-scale piston compression apparatuses, smaller scale compressors based on peristaltic (Nacher *et al.*, 1999) and diaphragm pumps (Gentile *et al.*, 2001) have been employed for polarized gas MRI. For the peristaltic approach with flow rates of 0.1 bar L/h and polarization preservation near unity, a polarization of between 30% and 40% could be achieved in the storage cell. Typically, 0.04 bar L was used routinely for MRI applications (Bidinosti *et al.*, 2003). More recently a similar apparatus was reported (Collier *et al.*, 2012). The diaphragm pump (Gentile *et al.*, 2001) method produced between 20% and 35% ^3He polarization using pure ^3He or between 35% and 50% polarization using ^3He - ^4He mixtures. The typical outlet pressure was 1 bar, polarization preservation was 0.75, and the polarizing rate was 0.4 bar L/h. However, further development of this approach has not been pursued.

IV. ^3He RELAXATION

A. Dipole-dipole interactions

Newbury *et al.* (1993) calculated that dipole-dipole interactions in bulk ^3He limit the longitudinal relaxation time of polarized ^3He to $T_1 = 807/p$ h (where p is the ^3He pressure in bar, for a cell temperature of 296 K), where the polarization decays as $\exp(-t/T_1)$ for ^3He polarized along the applied magnetic field. This expression was verified for ^3He pressure in the range between 4 and 12 bar in sealed SEOP cells made from aluminosilicate glass (Corning 1720).⁹ The observed limiting relaxation time T_1 was extended to lower pressures in borosilicate glass [$T_1 = 300$ h observed for $p = 2.5$ bar

⁷Mainz Microtron, Mainz, Germany.

⁸Institut Laue-Langevin, Grenoble, France.

⁹Corning Glass, Corning, NY.

(Smith *et al.*, 1998)] and sol-gel coated borosilicate glass (Corning Pyrex) [$T_1 = 344$ h observed for $p = 2.1$ bar (Hsu *et al.*, 2000)]. The development of neutron spin filters led to practical interest in pressures near 1 bar, in which relaxation times of several hundred hours have been observed (Rich, Gentile *et al.*, 2002; Parnell *et al.*, 2009; Chen *et al.*, 2011; Salhi *et al.*, 2014) in SEOP GE180 cells, corresponding to relaxation times of the order of several thousand hours from wall relaxation and other sources. All of these cells contained Rb or K-Rb mixtures for the SEOP process, films of which have been shown to suppress wall relaxation as compared to bare glass cells (Heil *et al.*, 1995).

B. Wall

1. Room temperature

In most practical situations, wall relaxation limits the achievable T_1 but is not well understood. Nearly a half century ago it was found that, for bare glass cells at room temperature, aluminosilicate glass yielded relaxation times of tens of hours as compared to a few hours for the borosilicate glass Pyrex (Fitzsimmons, Tankersley, and Walters, 1969). The temperature dependence of the relaxation times for both types of glass indicated that Pyrex relaxation at room temperature is dominated by permeation whereas adsorption dominates for aluminosilicate cells. In Pyrex, T_1 becomes shorter at high temperatures due to increased permeation of He into the glass. A detailed model of ^3He relaxation on the surface of borosilicate glass which accurately predicted observed relaxation rates and their temperature dependence was reported (Jacob, Driehuys, and Saam, 2003).

For bare aluminosilicate glasses, which have negligible permeation and adsorption energies of the order of 100 K, higher temperatures result in longer T_1 due to reduced adsorption. This discovery (Fitzsimmons, Tankersley, and Walters, 1969), plus the much better alkali-metal chemical resistance, led to the dominance of aluminosilicate glass for SEOP. Nearly complete suppression of wall relaxation was achieved in the study of dipole-dipole relaxation with SEOP cells made from aluminosilicate glass (Newbury *et al.*, 1993). It was found that cells made from fully blown glass yielded the best results and cleaning with nitric acid was suggested if a fully blown cell was not possible. The term “fully blown” indicates that all interior surfaces of the glass have been thoroughly melted; the resulting changes in these surfaces have generally been found to decrease ^3He relaxation. Wall-relaxation times of several thousand hours have also been observed in SEOP neutron spin-filter cells and the importance of fully blown glass verified (Rich, Gentile *et al.*, 2002; Parnell *et al.*, 2009; Chen *et al.*, 2011; Salhi *et al.*, 2014).

In SEOP cells, alkali-metal coatings are always present. In a series of measurements with various glasses and metal coatings, the importance of metal coatings for reducing wall relaxation was demonstrated by Heil *et al.* (1995). Relaxation times of 68 and 120 h were reported for cesium-coated Pyrex and cesium-coated aluminosilicate glass (Schott suprema^x),¹⁰ respectively. [It is interesting to note that Fitzsimmons,

Tankersley, and Walters (1969) reported a relaxation time of 250 h for a SEOP cell, which was substantially longer than they reported for cells without alkali metal. Whereas they recognized that the presence of Rb did not adversely affect the T_1 , the possible benefit of its presence did not appear to be recognized.] In a later study that also included alkali-metal oxide coatings, it was found that despite the substantial role of the coating, the longest relaxation times were still obtained with aluminosilicate glass (Deninger *et al.*, 2006). Hence the substrate still plays a role and it was speculated that this role could be associated with imperfections in the coating.

Whereas for both methods the longest relaxation times are usually obtained with aluminosilicate glasses, Pyrex SEOP cells have been employed for MRI applications (Jacob, Morgan, and Saam, 2002) and sol-gel coatings developed for SEOP cells (Hsu *et al.*, 2000). More recently this technique was employed for a ^3He target to avoid the substantial barium content in GE180 (Ye *et al.*, 2010). MEOP storage cells typically employ cesium coatings; long relaxation times have also been obtained in fused silica (Deninger *et al.*, 2006) and silicon-windowed Pyrex cells (Lelievre-Berna, 2007).

For compressor and target applications relaxation on various materials has been studied (Gamblin and Carver, 1965; Timsit, Daniels, and May, 1971; Jones *et al.*, 1993; Korsch *et al.*, 1997; Hussey *et al.*, 2005; Katabuchi *et al.*, 2005).

2. Magnetic-field and orientation effects

The advent of polarized gas MRI led to studies of relaxation in strong magnetic fields, which has revealed yet further unexplained aspects of ^3He relaxation. In the first study (Jacob *et al.*, 2001), it was found that the ^3He relaxation rate could be increased by a factor of between 2 and 20 solely by exposure of SEOP cells to a magnetic field of a few tenths of a Tesla (few kG). The original T_1 could be restored by degaussing the cell, leading to the term “ T_1 hysteresis” to describe the observed behavior. Soon thereafter a significant dependence of T_1 in SEOP cells due only to the physical orientation of the cell in a 3 mT (30 G) applied magnetic field was observed (Jacob *et al.*, 2004). The presence of both alkali metal and heating of the cells associated with the SEOP process were necessary to produce this low-field orientation dependence. Later studies showed that the relaxation time can depend on the direction and strength of the magnetic field (Chen *et al.*, 2011). More recently, the angular dependence of T_1 was measured and found to be characteristic of a dipolar effect (Boag *et al.*, 2014). Based on studies of boundary collisions of random walks, Bicout *et al.* (2013) concluded that the depolarizing effect of rare magnetic impurities may be much larger than expected and speculated that this enhancement could explain T_1 hysteresis. In a test on a pure Rb, sealed SEOP cell, the T_1 was observed to decrease by an order of magnitude for a field strength of only 40 mT (Chen *et al.*, 2011).

T_1 hysteresis was also observed in both Cs-coated and bare-glass, valved, MEOP cells (Hutanu, Rupp *et al.*, 2007). The variation of T_1 with magnetic field was measured and found to decrease exponentially with a constant of $(30 \text{ mT})^{-1}$ [$(300 \text{ G})^{-1}$]. Superconducting quantum interference device

¹⁰SCHOTT North America, Inc., Louisville, KY.

(SQUID) measurements revealed that most of the magnetization was associated with the glass valves, in particular, the plastic parts and O ring (Hutanu, Rupp, and Sander-Thömmes, 2007).

C. Field gradients

Relaxation due to static and oscillating magnetic-field gradients in a variety of regimes has been addressed in several references (Gamblin and Carver, 1965; Schearer and Walters, 1965; Cates, Schaefer, and Happer, 1988; Cates *et al.*, 1988; Hasson *et al.*, 1990; McGregor, 1990; Bohler and McGregor, 1994). At room temperature, a practical expression for the relaxation rate $1/T_1$ in a uniform field gradient is given by (McIver *et al.*, 2009)

$$\frac{1}{T_1^{\text{gradient}}} = \frac{6700}{p} \left(\frac{|\vec{\nabla}B_x|^2}{B_0^2} + \frac{|\vec{\nabla}B_y|^2}{B_0^2} \right) \text{h}^{-1}, \quad (48)$$

where p is the gas pressure in bar and $|\vec{\nabla}B_x/B_0|$ and $|\vec{\nabla}B_y/B_0|$ are the gradients in the transverse components of the magnetic field (i.e., the nuclear polarization is along the z axis) in units of cm^{-1} (Cates, Schaefer, and Happer, 1988). For a cell at a pressure of 1 bar in a uniform gradient

$$\left| \frac{\vec{\nabla}B_x}{B_0} \right| = \left| \frac{\vec{\nabla}B_y}{B_0} \right| = 3 \times 10^{-4} \text{ cm}^{-1}$$

the gradient-induced relaxation time is 830 h. We note that Eq. (48) is valid as long as the product of Larmor precession frequency and the collision time is small compared to unity, valid for SEOP and also for MEOP at 20 G and typical pressures of the order of 1 mbar. For MEOP at very low pressures and high fields corrections to this are necessary (Schearer and Walters, 1965).

In more recent work, Zheng, Gao *et al.* (2011) reported an approach based on calculating the autocorrelation function of spins to derive the magnetic-field gradient-induced transverse and longitudinal relaxation of spins undergoing restricted diffusion. Guigue *et al.* (2014) performed a theoretical analysis of spin relaxation, for a polarized gas of spin $1/2$ particles undergoing restricted adiabatic diffusive motion within a container of arbitrary shape, due to magnetic-field inhomogeneities of arbitrary form. This analysis provided a theoretical justification for the usual assumption that the relaxation rate is determined by the volume average of the relevant gradients. Studies of gradient-induced relaxation in the transfer of gas into high magnetic fields has also been reported (Zheng, Cleveland *et al.*, 2011; Maxwell, Epstein, and Milner, 2015).

D. Magnetostatic cavities

Neutron spin filters have motivated a variety of magnetostatic cavities to provide a uniform magnetic field for ^3He gas on neutron beam lines. For these applications, there may be space constraints and significant stray fields, as well as neutron spin transport fields to be matched (see Sec. VII.A). In early applications of NSFs on the IN20 triple-axis spectrometer at the ILL, an end-compensated solenoid surrounded by a μ -metal

box and a cubic iron box ($40 \times 40 \times 40 \text{ cm}^3$) was employed (Heil, Dreyer *et al.*, 1999). Soon thereafter a superconducting magnetostatic cavity (Cryopol) was developed for the D3 spectrometer that is capable of shielding an NSF from a nearby superconducting sample magnet operating at several Tesla (Dreyer *et al.*, 2000). For moderate stray fields a magnetic parallel plate capacitor was developed, which provided a uniform 1 mT field transverse to the neutron beam (known as a “magic box” because when first made, the performance was better than predicted by finite element calculations) (Petukhov *et al.*, 2006). This cavity consisted of a μ -metal box with sides that are magnetized by either coils or permanent magnets. The field lines enter the top plate and traverse the gap to the bottom plate. The original box was 80 cm along the beam line, but in a later, permanent-magnet design this distance was reduced to 40 cm (Hutanu *et al.*, 2008) and the field increased to 1.7 mT. An end-compensated design shortened the length along the beam line to 28.4 cm (McIver *et al.*, 2009), with field strengths up to 3.6 mT (Chen, Gentile, Erwin *et al.*, 2014). Magnetically shielded solenoids are also employed and typically provide greater protection from stray fields in space-constrained applications. Typical achievable gradients from magnetically shielded solenoids and magic boxes are between 2×10^{-4} and $6 \times 10^{-4} \text{ cm}^{-1}$, corresponding to gradient-induced relaxation times of between 3700 and 600 h, respectively (Chen, Gentile, Erwin *et al.*, 2014). Polarized gas MRI also provided a motivation for compact, light-weight, and inexpensive magnetized boxes for transporting polarized gas, in some cases via air freight (Hiebel *et al.*, 2010).

E. Ionizing radiation

Relaxation from 3 MeV protons for ^3He polarized at mbar pressures by MEOP was reported by Milner, McKeown, and Woodward (1987). Soon thereafter relaxation from 18 MeV ^4He alpha particles for ^3He polarized at 0.8 bar by SEOP was reported by Coulter *et al.* (1988). Theoretical studies (Bonin, Saltzberg, and Happer, 1988; Bonin, Walker, and Happer, 1988) showed that relaxation is caused by hyperfine coupling in $^3\text{He}^+$ and spin-rotation coupling in $^3\text{He}_2^+$. This relaxation source is greatly reduced in SEOP cells because nitrogen efficiently destroys these species. For application of MEOP cells to electron scattering, addition of a small ($[\text{N}_2]/[^3\text{He}] \approx 10^{-4}$) quantity of nitrogen after compression has been employed to quench these species (Meyerhoff *et al.*, 1994). Nevertheless, there is substantial relaxation induced by high current charged particle beams, thus motivating the use of hybrid SEOP and rapid exchange of gas in double cell targets (Dolph *et al.*, 2011; Singh *et al.*, 2015). Because of the much greater sensitivity of pure ^3He cells to ionization, large effects on ^3He relaxation have been observed in MEOP cells due to neutron beams (Petukhov, 2016). For *in situ* SEOP on neutron beam lines, beam-induced alkali-metal relaxation and other effects have been observed (see Sec. VII.E.3).

F. Low temperatures

Motivated by the need for dense polarized ^3He targets, as well as by studies of thermodynamic and transport properties

of polarized ^3He liquid, attempts to dynamically polarize liquid ^3He from optically pumped room temperature gas were made soon after the first MEOP experiments were reported. In this seminal work (McAdams, 1968), polarized ^3He gas was transported from a room temperature MEOP region through a connecting tube to a cold sample volume either by atomic diffusion or by transient gas flow during sample cool down. Most of the polarization was lost in the process, presumably due to fast wall relaxation in the cold part of the connecting tube or in the sample, with a maximum polarization of 0.15% achieved in a small liquid ^3He volume. This work triggered studies of wall relaxation of ^3He gas and liquid at low temperatures by several groups, using either Boltzmann-polarized or MEOP-polarized ^3He . It was confirmed that clean, bare Pyrex glass walls induce increasing relaxation rates $1/T_1$ at decreasing temperatures below ≈ 100 K (Fitzsimmons, Tankersley, and Walters, 1969; Lefevre-Seguin and Brossel, 1988), with $T_1 < 1$ s below 20 K for a low-density gas (Chapman and Richards, 1974; Barbé, Laloë, and Brossel, 1975).

In order to reduce the dwell time and polarization loss of ^3He atoms colliding with cold glass walls, weak-binding diamagnetic cryogenic coatings have been added onto cell walls: Ne (Chapman, 1975; Chapman and Bloom, 1976), H_2 (Barbé, Laloë, and Brossel, 1975), or liquid ^4He (Himbert *et al.*, 1983). The adsorption energies of ^3He atoms on these coated walls and the relaxation mechanisms of atoms or adsorbed layers on weakly relaxing substrates have been extensively studied and are well understood (Lefevre-Seguin *et al.*, 1985; Lefevre-Seguin and Brossel, 1988; Lusher, Secca, and Richards, 1988a, 1988b; Himbert and Dupont-Roc, 1989). Depending on experimental conditions, different relaxation regimes can be observed. For instance, for low-density adsorbed ^3He layers T_1 does not depend on the bulk gas density N and scales as $\exp(-2\Delta W/k_B T)$, where ΔW is the adsorption energy and $k_B T$ is the thermal energy. On the contrary, for high enough gas density or low enough temperature, a complete ^3He monolayer is condensed and $T_1 \propto N$ is essentially temperature independent. H_2 coatings, for which $\Delta W \approx 12$ K, yield long T_1 's (up to several days) at 4 K (for comparison, $\Delta W \approx 150$ K on bare glass). H_2 coatings are efficient up to ≈ 6 K where they desorb and down to ≈ 2 K where a ^3He monolayer is formed. ^4He films extend the temperature range over which polarized ^3He gas can be prepared or stored down to below 0.5 K (where the gas eventually liquefies if its pressure exceeds a few mbar).

Alternatively, cesium can be used to coat cell walls. With a ^3He adsorption energy as low as $\Delta W \approx 2.3$ K (Tastevin, 1992a) cesiated glass (i.e., glass that has been in contact with cesium) is a weakly relaxing material from room temperature down to hundreds of mK. This property is plausibly linked with the nonwetting of alkali-metal surfaces by ^4He liquid and films (Nacher and Dupont-Roc, 1991), and was initially demonstrated at low temperature before being assessed at room temperature (Chéron *et al.*, 1995).

Motivated by an experiment to measure the neutron electric dipole moment (EDM), new relaxation studies in liquid mixtures of ^3He and ^4He have been performed for the relevant

polymer-coated materials at cryogenic temperatures (Ye *et al.*, 2008, 2009).

With suitably coated walls, the way was open for reliable measurements of the bulk (dipole-dipole) relaxation in ^3He gas (Lusher, Secca, and Richards, 1988b) and ^3He - ^4He liquid mixtures, for which T_1 exceeding 10 h have been recorded at low ^3He concentration (Piegay and Tastevin, 2002). More importantly, McAdams's strategy to prepare polarized cold ^3He samples from optically polarized gas could be successfully applied. For instance, polarization was nearly fully preserved when transferred to a gas sample at 4.2 K, therefore exceeding 50% (Crampton *et al.*, 1984; Leduc *et al.*, 1984). Transient polarizations exceeding 40% were reported for liquid ^3He just after liquefaction (Tastevin *et al.*, 1988), subsequently decaying due to a bulk T_1 of ≈ 300 s. Thermally driven convective flow between the low- and high-temperature regions was used to sustain up to 56% nuclear polarization in steady state in ^3He - ^4He liquid mixtures (Candela, Hayden, and Nacher, 1994). Such high polarizations could not be achieved by MEOP directly performed at low temperature due to the highly reduced rate of ME collisions (see Sec. III.B.2), but room temperature OP with polarized gas transfer enabled a series of low-temperature studies of ^3He as a quantum fluid: characterization of spin waves (Nacher *et al.*, 1984; Tastevin *et al.*, 1985) and heat conduction changes (Leduc *et al.*, 1987; Larat *et al.*, 1990) in ^3He gas and of phase coexistence for liquid ^3He (Tastevin, 1992b; Candela, Hayden, and Nacher, 1994; Villard, Nacher, and Tastevin, 2000). A similar technique was used with refillable cells to repeatedly prepare larger samples of polarized liquid in which magnetic interactions play a key role in nonlinear NMR dynamics (Hayden *et al.*, 2007; Baudin *et al.*, 2008).

Let us also mention two alternative hyperpolarization techniques, not using MEOP but instead relying on nuclear relaxation and well-controlled phase transitions: rapid melting which yields up to 70% transient polarization in liquid ^3He (Bonfait *et al.*, 1984, 1987; Buu, Puech, and Wolf, 2000) and spin distillation which provides up to a sevenfold enhancement of the Boltzmann equilibrium polarization in the steady-state (Nacher *et al.*, 1991; Rodrigues and Vermeulen, 1997; van Steenberg *et al.*, 1998).

V. ^3He POLARIZATION METROLOGY AND CONTROL

Absolute polarization measurement methods for dense samples of polarized ^3He include water-calibrated NMR (Lorenzon *et al.*, 1993; Romalis *et al.*, 1998), EPR (Romalis and Cates, 1998; Babcock, Nelson *et al.*, 2005), neutron transmission (Jones *et al.*, 2000; Chupp *et al.*, 2007), and magnetometry (Noël *et al.*, 1996; Wilms *et al.*, 1997). In all of these methods except neutron transmission, the absolute ^3He polarization is determined by measuring both the ^3He magnetization and the ^3He density. The ^3He density is determined at the time a cell is filled with gas. For SEOP cells the density can also be determined after the cell is filled from the width and/or shift of the alkali-metal absorption lines (Romalis, Miron, and Cates, 1997; Kluttz, Averett, and Wolin, 2013). For SEOP electron-scattering targets, the two methods

of determining the density have been reported to agree within 2% (Romalis and Cates, 1998) and 1% (Singh *et al.*, 2015). Since SEOP cells contain both ^3He and N_2 gas, the pressure width and/or shift coefficients for both gases are required, although the shift and width is typically dominated by the ^3He gas. These coefficients were measured for the Rb D1 and D2 lines with a typical accuracy of 2% (Romalis, Miron, and Cates, 1997), and more recently for both Rb and K with a typical accuracy for the width coefficient of 1% (Kluttz, Averett, and Wolin, 2013). For neutron transmission, the density is replaced with the opacity, which is determined from the transmission through an unpolarized cell.

A. NMR

1. Adiabatic fast passage

In the technique of AFP NMR (Abragam, 1961), an rf magnetic field is applied transverse to the static magnetic field. If the magnetic field is swept such that the Larmor frequency passes through the rf frequency (or the rf frequency is swept so that it passes through the Larmor frequency), the ^3He polarization will invert. From a quantum-mechanical point of view, this process is essentially an avoided crossing due to perturbation from the rf field (Rubbmark *et al.*, 1981). The sweep rate and rf amplitude are adjusted to minimize losses resulting from polarization inversion. If a pickup coil transverse to both the static and rf magnetic fields is employed, a signal proportional to the magnetization is obtained. In order to minimize the large background from inadvertent pickup of the applied rf field, the pickup coil must be carefully adjusted to be orthogonal to the driving rf magnetic field. An absolute ^3He polarization measurement can be obtained if the response of the pickup coil and associated electronics is calibrated using a thermally polarized water cell with exactly the same size and geometry as the ^3He cell. Despite the extremely small thermal polarization of the order of 10^{-8} in typical holding fields of a few millitesla, these calibrations can be performed with uncertainties of a few percent. [In principle the magnitude of the AFP signal could be determined absolutely for a given apparatus, but in a study of this approach discrepancies of between 20% and 50% were observed with cell to cell variations (Chen *et al.*, 2011).] Further descriptions of this technique and its application to electron-scattering targets (Chupp *et al.*, 1987; Romalis *et al.*, 1998), polarimetry of low-pressure MEOP cells (Lorenzon *et al.*, 1993), and polarized gas MRI¹¹ have been reported.

Whereas losses of a few tenths of a percent are typically encountered for AFP, techniques have been developed to substantially reduce these losses. The primary motivation has been for neutron spin-filter cells that are not actively optically pumped on the beam line, but in which the ^3He polarization may be frequently inverted during use so as to invert the neutron polarization. Besides the usual optimization of rf magnetic-field strength and sweep rate, the rf field is modulated by a Gaussian envelope during the sweep (Petoukhov *et al.*, 2006; McKetterick *et al.*, 2011). Using this approach,

losses of 10^{-5} per flip have been obtained. In a compact rf solenoid with shielding to confine the rf field, loss as low as 0.03% was reported (Ye *et al.*, 2013).

2. Free induction decay

Monitoring of ^3He polarization can also be performed by FID (Bloch, 1946), in which an rf pulse tips the ^3He spins and a pickup coil detects the freely precessing transverse component of the magnetization following the tip. This approach avoids the need for orthogonal drive and pickup coils. The measurement can be nondestructive by using small tip angles for a coil of comparable size to either the cell and/or a small coil. The decay of the signal is given by the transverse relaxation time T_2^* , which is typically dominated by dephasing due to magnetic-field gradients and thus given in high-pressure cells by $T_2^* = (\gamma\Delta B)^{-1}$, where $\gamma/2\pi = 32.4$ kHz/mT (3.24 kHz/G) is the gyromagnetic ratio and ΔB is the variation in the magnetic field B over the gas volume sampled by the coil. Values of T_2^* in typical applications range from a few milliseconds to a few hundred milliseconds. For the typical low pressures employed for low-field MEOP, averaging of the field gradient [motional narrowing (Pines and Slichter, 1955; Cates *et al.*, 1988)] can increase T_2^* substantially above this value. The use of FID is discussed in several references (Lorenzon *et al.*, 1993; Parnell *et al.*, 2008; Krimmer *et al.*, 2009; Chen *et al.*, 2011). Whereas FID is typically used for relative measurements, Gentile *et al.* (2001) employed an FID system with large coils that was calibrated against fluorescence light polarimetry in an MEOP cell and then applied to determine absolute ^3He polarization in compressed gas.

3. Radiation damping issues and control

For pickup coils with a high filling factor, or dense samples with a high polarization, the nuclear magnetization can be significantly affected by radiation damping, i.e., the action of the resonant rf field generated by NMR-precession-driven current in the coils. For ^3He gas polarized in the low Zeeman energy state, the lifetime of the observed FID signals is decreased whereas for the high-energy state it is increased and unstable precession or maser operation can occur (Gentile *et al.*, 2001). During AFP magnetization reversal, radiation damping affects the applied rf field and therefore modifies line shapes and may increase losses, even if the unstable regime is avoided. Radiation damping is traditionally reduced using weakly coupled coils (at the expense of detected signal amplitudes) or applying a suitable field gradient to the sample (Romalis and Happer, 1999; Zheng, 2002). Active feedback schemes can also be used to control radiation damping without signal-to-noise penalty (Hoult, 1979; Baudin *et al.*, 2011).

B. Neutron transmission

^3He polarization measurement by neutron transmission relies on the simple equations governing a neutron spin filter (Coulter *et al.*, 1990; Jones *et al.*, 2000). The neutron polarization P_n is given by

¹¹Polarean Inc., Durham, NC.

$$P_n^2(\lambda) = 1 - \frac{T_0^2(\lambda)}{T_n^2(\lambda)}, \quad (49)$$

where λ is the neutron wavelength and $T_0(\lambda)$ and $T_n(\lambda)$ are the transmissions of unpolarized neutrons through the unpolarized and polarized ^3He cell, respectively. Hence P_n , which is typically what is of the greatest interest for spin-filter applications, can be accurately determined with only a relative transmission measurement. Using the relationship

$$P_n(\lambda) = \tanh [O(\lambda)P_{\text{He}}], \quad (50)$$

where $O(\lambda)$ is the opacity, the ^3He polarization P_{He} can be determined. The opacity is determined from $T_0 = T_e \exp[-O(\lambda)]$, where T_e is the transmission of the cell without ^3He gas. Hence to determine P_{He} from P_n requires an absolute measurement of $T_0(\lambda)$ and determination of T_e . The opacity is given by the product of the neutron absorption cross section (which typically increases linearly with neutron wavelength), the ^3He density, and the ^3He path length. Because of the dependence on neutron wavelength, neutron transmission measurements are typically performed with monochromatic beam lines at reactors or with the use of time-of-flight (TOF) analysis at pulsed neutron sources. Neutron transmission has been used to calibrate NMR-based polarization measurements for routine ^3He polarimetry on reactor beams for a wide variety of neutron spin-filter cells with typical accuracy of a few percent (Chen *et al.*, 2011). T_e can be determined by neutron transmission measurements before the cell is filled, but for routine applications is often determined from measured values for glass transmission (Chupp *et al.*, 2007) and estimated glass thickness; a typical value and uncertainty is $T_e = 0.88$ and 0.02, respectively. With TOF analysis, T_0 versus wavelength can be fit, allowing extraction of the opacity and T_e for a filled cell. Using this approach, determinations of P_{He} at pulsed sources with uncertainties of 2% have been reported (Chupp *et al.*, 2007; Tong *et al.*, 2012).

C. Detection by magnetometry

Accurate magnetometry has also been applied to measure ^3He polarization in a 5 bar MEOP-based target for electron scattering with accuracy of 3% (Krimmer *et al.*, 2009). This approach relies on the known dipolar field for a spherical sample of polarized gas. Since the field due to the gas is only ≈ 1 mG or 0.02% of the holding field, the change in field strength is determined by a fluxgate magnetometer upon inverting the ^3He polarization with low-loss AFP (Wilms *et al.*, 1997). In addition, a spherical phantom with equidistant current loops was employed to calibrate the exact location of the magnetic-field sensor.

VI. CHARGED PARTICLE AND PHOTON SCATTERING TARGETS

Polarized ^3He provides a reasonable approximation to a polarized neutron target because the proton spins are primarily paired off. Hence about 87% of the ^3He spin is carried by the

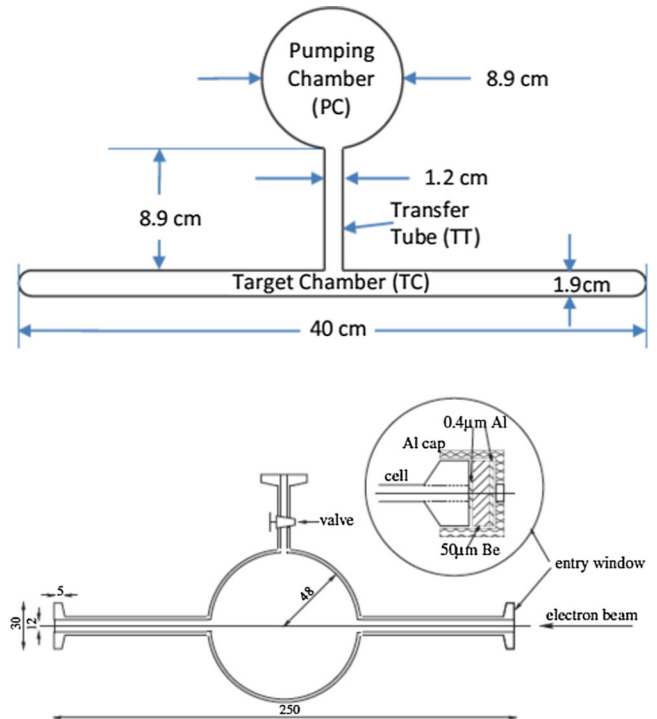


FIG. 19. Diagrams of current electron beam target designs. Top: Jefferson Laboratory. From Dolph *et al.*, 2011. Bottom: Mainz. Dimensions are in mm. From Krimmer *et al.*, 2009.

lone neutron (Friar *et al.*, 1990; Laskaris *et al.*, 2013, 2015). It is this property of the ^3He nucleus that yields its utility for studies of the spatial distribution of charge, magnetization (Bernauer, 2012), and spin in the neutron. ^3He targets for nuclear and particle physics with charged particle and photon beams fall into four groups: (1) continuously polarized SEOP external targets for electron (Singh *et al.*, 2015), photon (Ye *et al.*, 2010), proton (Häusser *et al.*, 1995; Katabuchi *et al.*, 2005), and pion (Larson *et al.*, 1991) scattering; (2) MEOP external targets that are polarized remotely and transported to electron (Krimmer *et al.*, 2009) and photon beam lines (Krimmer *et al.*, 2011); (3) MEOP internal targets for electron storage rings (DeSchepper *et al.*, 1998); and (4) continuously polarized MEOP external targets for electron scattering (Eckert *et al.*, 1992; Jones *et al.*, 1993). In addition, an MEOP-based polarized ^3He ion source is under development for RHIC¹² (Maxwell, Milner, and Epstein, 2014). Figure 19 shows two current target designs.

A. SEOP

1. TRIUMF, Bates LINAC, and SLAC

Early SEOP targets were constructed from Corning 1720 glass cells between 10 and 35 cm³ in volume, filled to pressures of several bar, and optically pumped with several watts of laser light from Ti-sapphire lasers. Typical ^3He polarization of 50% was obtained for pion (Larson *et al.*, 1991) and proton (Häusser *et al.*, 1995) scattering at

¹²Relativistic Heavy Ion Collider, Brookhaven, NY.

TRIUMF,¹³ and production of polarized muonic helium at LAMPF¹⁴ (Newbury *et al.*, 1991). Application to electron scattering followed soon thereafter but required the use of double cells (Chupp *et al.*, 1987) due to the strong ionization produced by electron beams with energies of several hundred MeV and several μA beam currents. In the double cell configuration, diffusion links the polarization produced in an optical pumping cell to the target cell via a transfer tube. The first double cell targets (Chupp *et al.*, 1992) were employed for quasielastic scattering of polarized electrons at the Bates Linear Accelerator Center, which provided some of the first double-polarization results for determination of the neutron's electric form factor (Thompson *et al.*, 1992). Using a few watts of laser light from a Ti-sapphire laser, an $\approx 50\text{ cm}^3$ cell was polarized up to 40% in tests and was maintained between 10% and 27% during the experiment. Double cell targets were then applied for deep inelastic scattering experiments (Johnson *et al.*, 1995) with $\approx 25\text{ GeV}$ energy electrons at SLAC for the first studies of the neutron's spin structure functions (Anthony *et al.*, 1993, 1996). In this experiment, ^3He polarization between 30% and 40% was maintained in a 9 bar, 200 cm^3 double cell with 20 W of laser light from five Ti-sapphire lasers, each pumped with a 20 W argon-ion laser (Johnson *et al.*, 1995). With improved polarimetry, further studies of spin structure functions were performed with this target (Abe *et al.*, 1997a, 1997b).

2. JLAB and TUNL targets

Double cell targets based on the design employed at SLAC were developed for electron-scattering experiments at Jefferson Laboratory with typical energies of $\approx 6\text{ GeV}$ and have steadily improved in performance and capability. As shown in Fig. 19, these targets typically have an optical pumping cell between 6 and 9 cm in diameter, a $\approx 2\text{ cm}$ diameter target cell between 25 and 40 cm long, end windows $\approx 120\ \mu\text{m}$ thick, densities of 7–12 amg, and relaxation times of 20 to 70 h (Alcorn *et al.*, 2004; Singh *et al.*, 2015). They have been constructed from Corning 1720 and, more recently, GE180 glass. Optical pumping is typically performed with $\approx 90\text{ W}$ of laser light from three fiber-coupled diode laser systems. The typical ^3He polarization achieved was 30% to 40% in the first experiments on electromagnetic form factors (Xu *et al.*, 2000; Xiong *et al.*, 2001) and neutron spin structure functions (Amarian *et al.*, 2002, 2004; Zheng *et al.*, 2004a, 2004b). With the use of hybrid SEOP, the ^3He polarization was increased to $\approx 45\%$ for a more recent measurement of the neutron's electric form factor (Riordan *et al.*, 2010). For hybrid SEOP with spectrally narrowed lasers, the polarization has been increased to between 50% and 55% for studies of asymmetries in scattering from transversely or vertically polarized targets generally aimed at improved understanding of the origin of the neutron's spin (Qian *et al.*, 2011; Huang *et al.*, 2012; Allada *et al.*, 2014; Katich *et al.*, 2014; Zhang *et al.*, 2014; Parno *et al.*, 2015; Zhao *et al.*, 2015). In a detailed study of these targets, a maximum offline value of 70% and a detailed analysis of achievable polarization

was reported (Singh *et al.*, 2015). Future experiments with even higher luminosity are expected to employ a new type of double cell in which the exchange of gas between the optical pumping and target cells is increased by convection (Dolph *et al.*, 2011). In this new design, there are two transfer tubes and gas circulation is produced by heating one tube.

Recently, two different SEOP targets have also been employed at TUNL.¹⁵ A target was developed for measurements of spin-correlation coefficients in $p + ^3\text{He}$ elastic scattering (Katabuchi *et al.*, 2005; Daniels *et al.*, 2010), in which the ^3He was polarized by SEOP at 8 bar and transferred through a plastic tube to spherical Pyrex cells with Kapton film covering the apertures for the beam and scattered particles. For application to gamma-ray beams, a sol-gel coated Pyrex double cell was employed to avoid background signals scattered off the barium present in GE180 glass (Ye *et al.*, 2010). The relaxation time in the 7 bar target was 35 h and a maximum offline ^3He polarization of 62% was obtained. In experiments to study three-body physics, $\approx 40\%$ ^3He polarization was maintained (Laskaris *et al.*, 2014, 2015).

B. MEOP

1. Early targets, Bates LINAC, and internal targets

The high efficiency of MEOP allowed for early targets based on optical pumping with lamps (Phillips *et al.*, 1962; Baker, McSherry, and Findley, 1969). The first laser-pumped targets were based on a double cell approach with a cryogenic target cell to increase the density (Milner, McKeown, and Woodward, 1989; Alley and Knutson, 1993). Such a target operated at a gas pressure of 2.6 mbar and a target cell temperature of 17 K was employed for quasielastic scattering of polarized electrons at the Bates Linear Accelerator Center, which provided some of the first double-polarization results to determine the neutron's electric form factor (Woodward *et al.*, 1990; Jones-Woodward *et al.*, 1991; Jones *et al.*, 1993). In this experiment, between 20% and 30% ^3He polarization was maintained using 0.3 W of laser light. The 10 cm long copper target cell was coated with frozen nitrogen to permit target cell relaxation times between 400 and 1000 s, depending on beam current. The same target design was employed in subsequent experiments to determine the neutron's magnetic form factor (Gao *et al.*, 1994) and to further study quasielastic scattering (Hansen *et al.*, 1995). Employing optical pumping with a few watts of laser light from a Nd:LMA laser (Gentile and McKeown, 1993) allowed for over 38% ^3He polarization at 2.5 times higher average electron beam current (Gao *et al.*, 1994).

Concurrently with these external targets, MEOP was applied to internal targets for storage rings. In these targets, gas flows from an optical pumping cell through a capillary to an open target cell. The first polarized ^3He internal target (Lee *et al.*, 1993) was employed at the Indiana University Cyclotron Facility for measurements of quasielastic scattering of polarized protons from polarized ^3He to study the ground-state spin structure of the ^3He nucleus (Lee *et al.*, 1993; Bloch *et al.*, 1995; Miller *et al.*, 1995). A Nd:LMA laser was used for optical

¹³Canada's national laboratory for particle and nuclear physics and accelerator-based science, Vancouver, BC.

¹⁴Los Alamos Clinton P. Anderson Meson Physics Facility, Los Alamos, NM.

¹⁵Triangle Universities Nuclear Laboratory, Durham, NC.

pumping and the average ^3He polarization in the target was 46% at a ^3He flow rate of 1.2×10^{17} atoms/s. Soon thereafter, an internal target with a cryogenic target cell (Kramer, DeSchepper *et al.*, 1995; Kramer, Kelsey *et al.*, 1995; Korsch *et al.*, 1997; DeSchepper *et al.*, 1998) was employed for spin-dependent deep inelastic scattering of 27.5 GeV polarized positrons at DESY¹⁶(Ackerstaff *et al.*, 1997; DeSchepper, 1998). For an ultrapure aluminum target cell cooled to 25 K, the ^3He polarization was 46% during the experiments. This target also used a variation of the fluorescence polarimetry method: rather than a discharge, high-energy positrons provided the required atomic excitation and the ^3He polarization was determined from the circular polarization of 492 nm light from the 4^1D-2^1P transition. An internal target at NIKHEF¹⁷ operating at a nominal atomic flow rate of $1 \times 10^{17} \text{ s}^{-1}$ yielded a nuclear polarization of 50% for a target thickness of $0.7 \times 10^{15} \text{ cm}^{-2}$ at a target temperature of 17 K (Poolman *et al.*, 2000).

2. MAMI

Early ^3He targets for electron scattering at MAMI employed a Toepler pump (Eckert *et al.*, 1992), in which a mercury column compressed gas polarized by MEOP at a pressure of ≈ 1 mbar into a 100 cm^3 target cell at ≈ 1 bar. The polarized gas was continuously recirculated from the target cell to the optical pumping cell for repolarization at a flow rate of 10^{18} atoms/s. Optical pumping with a few watts of laser light from a Nd:LMA laser (Eckert *et al.*, 1992) yielded an average ^3He polarization of 38% in the first MAMI experiment to determine the neutron's electric form factor (Meyerhoff *et al.*, 1994). In a later experiment on the same topic (Becker *et al.*, 1999), several improvements including a cesium-coated cell with a relaxation time of 6 h yielded 50% ^3He polarization. The continuous flow Toepler pump apparatus was replaced by remotely polarized cells filled using a piston compression apparatus (Becker *et al.*, 1994; Surkau *et al.*, 1997) to a pressure of 4 bar. This approach was employed for further measurements of the neutron's electric form factor (Bermuth *et al.*, 2003). More recently, an improved piston compression apparatus yielded 72% ^3He polarization in a 25 cm long, cesium-coated, quartz, 5 bar target cell with beryllium and aluminum foil windows (Krimmer *et al.*, 2009); see Fig. 19. Relaxation induced by the electron beam (see Sec. IV.E) yielded typical beam-on relaxation times between 30 and 40 h, hence the cells were replaced twice a day. A time-averaged ^3He polarization of 56% was maintained over the course of a three week measurement of the neutron electric to magnetic form factor ratio (Schlimme *et al.*, 2013). Similar targets (Krimmer *et al.*, 2011) have also been applied for the tagged photon beam facility at MAMI (Costanza *et al.*, 2014).

C. Summary of physics enabled

The complex and diverse subatomic physics studied with ^3He targets is beyond the scope of this review. Nevertheless, we briefly summarize the overall topics of the majority of

experiments. The charge and magnetization distributions are typically characterized by the electric and magnetic form factors of the neutron $G_E^n(Q^2)$ and $G_M^n(Q^2)$, respectively, which nonrelativistically can be considered to be the Fourier transforms of the charge and magnetization distributions. The first experiments with polarized ^3He focused on measuring these form factors and on testing the three-body nuclear physics calculations needed to quantitatively validate the approximation of a polarized ^3He target as a neutron target. Since the neutron is neutral overall, measuring the electric form factor is particularly difficult. In addition, measurements must be performed over a wide range of momentum transfer to test theoretical predictions. In the last 25 years, these measurements have greatly improved the knowledge of these form factors, yielding more precise tests of nucleon models (Qattan and Arrington, 2012). Whereas the uncertainty for the earliest measurements were larger than the values themselves and were performed at only a single value of relatively low momentum transfer, recent measurements have fractional uncertainties of 15% or better and cover a wide range of momentum transfer (Gentile and Crawford, 2011). Although $G_E^n(Q^2)$ is generally the more difficult and desired measurement, $G_M^n(Q^2)$ is also important because typically G_E^n is actually determined from a measurement of the ratio $G_E^n(Q^2)/G_M^n(Q^2)$ combined with separate measurements of $G_M^n(Q^2)$.

Analogous to the electric and magnetic form factors are the more complex spin structure functions that are related to the distribution of angular momentum in the neutron (Aidala *et al.*, 2013). These studies have been primarily motivated by understanding the origin of the spin of the neutron. Improvements in the polarized ^3He targets have allowed for a substantial improvement in the precision of tests of the fundamental sum rules for spin structure from quantum chromodynamics. Decades ago it was determined that, contrary to expectations, the intrinsic quark spins contribute only a small fraction of the nucleon spin. Despite many years of effort, studies of the origin of the spin of the nucleon are still incomplete. Continuing studies with polarized ^3He targets have focused on the possible contribution to the nucleon spin from the orbital angular momentum of the quarks.

VII. NEUTRON SPIN FILTERS

Neutron spin filters (Ioffe, Babcock, and Gutberlet, 2011) produce highly spin-polarized beams of low-energy neutrons, or analyze the neutron spin state, by passing the neutrons through a glass cell of polarized ^3He in a uniform magnetic field. The near 100% contrast in the spin dependence of the ^3He neutron absorption cross section (Coulter *et al.*, 1990) results in highly polarized neutron beams or very efficient spin analyzers. Although other devices such as supermirrors (Mezei, 1976) and Heusler alloy (Freund *et al.*, 1983) are employed for polarizing neutron beams (Williams, 1988), NSF's are advantageous for large area and large divergence beams, can be used for cold, thermal, and epithermal neutrons, and decouple polarization selection from wavelength selection. For both MEOP and SEOP, NSF cells are typically polarized offline, transported to the neutron beam line, and stored in various magnetostatic cavities. In addition, there has been increasing use of SEOP-based *in situ* NSF systems.

¹⁶Deutsches Elektronen Synchrotron, Hamburg, Germany.

¹⁷Dutch National Institute for Subatomic Physics, Amsterdam, Netherlands.

A. Principles

The large cross section for neutron absorption by ^3He arises from a broad, unbound, resonance that yields a proton and a triton. The strong spin dependence arises because absorption occurs only if the neutron and ^3He spins have their spins antiparallel. Whereas the cross section for capture of 25 meV neutrons with spin antiparallel to the ^3He nuclear spin is 10 666 b, the ratio of the absorption for neutrons with parallel and antiparallel spin has been determined experimentally to be less than a few percent and estimated theoretically to be less than 0.5% (Huber *et al.*, 2014), and the scattering cross section is only a few barns (Mughabghab, Divadeenam, and Holden, 1981). Hence for a sufficient opacity (product of gas density, cross section, and cell length) of 100% polarized ^3He , all neutrons with antiparallel spin would be absorbed, while nearly all neutrons with parallel spin would be transmitted, resulting in 100% neutron polarization and 50% transmission. Although imperfect ^3He polarization reduces the achievable neutron polarization and transmission for a given opacity, increasing the opacity allows any neutron polarization to be achieved at the expense of transmission. The absorption cross section is directly proportional to the wavelength, hence higher energy (shorter wavelength) neutrons require greater opacities. Typical room temperature pressure-length products vary between 4 and 25 bar cm for neutrons with energies between 2 and 80 meV (wavelengths between 0.6 and 0.1 nm). For a ^3He polarization of 75%, 90% neutron polarization with 28% neutron transmission can be obtained. These values include the transmission of 0.88 due to neutron scattering from a typical glass NSF cell. The basic equations governing the relationships between opacity, ^3He polarization, and neutron polarization and transmission appear in many publications (Coulter *et al.*, 1990; Jones *et al.*, 2000). These relationships allow both the neutron and ^3He polarization to be determined by neutron transmission measurements. NSFs are also used to analyze neutron polarization; in this case an analyzing power of 90% with a transmission for the desired spin state of 54% can be obtained. Figure 20 shows the variation of the neutron polarization and transmission with the pressure-length-wavelength product for ^3He polarizations of 0.5, 0.75, and 1. The ideal opacity factor depends on the type of experiment and its optimization has been addressed with different approaches (Tasset and Ressouche, 1995; Williams, 1999; Goossens and Cussen, 2002; Gentile *et al.*, 2005); a typical value is 3 bar-cm-nm.

The strong neutron absorption cross section for boron restricts the choice of materials for NSFs. For SEOP, GE180 is generally employed due to issues with ^3He permeation and temperature-dependent relaxation for quartz (Ino *et al.*, 2005; Ino and Muto, 2007; Ye *et al.*, 2013). Sapphire (Masuda *et al.*, 2005; Chen *et al.*, 2011) and silicon-windowed Pyrex cells (Chen *et al.*, 2011) have also been investigated for SEOP, but use has been limited due to relaxation issues. For MEOP cells, which do not need to be heated, both quartz and silicon-windowed Pyrex cells are routinely used (Hutanu and Rupp, 2005; Lelievre-Berna, 2007). Neutron transmission and scattering from glasses are important practical issues for NSFs and have been studied by various groups (Chen *et al.*, 2004;

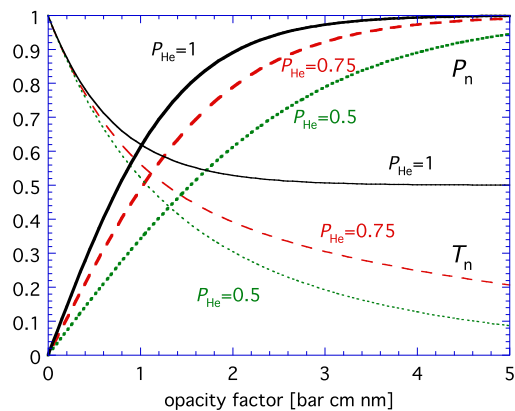


FIG. 20. The variation of the neutron polarization P_n and transmission T_n with the opacity factor, where the opacity factor is given by the pressure-length-wavelength product in bar-cm-nm, for ^3He polarizations (P_{He}) of 0.5, 0.75, and 1. The neutron transmissions shown do not include the transmission of 0.88 due to neutron scattering from a typical glass NSF cell.

Chupp *et al.*, 2007; Sakaguchi *et al.*, 2011b; Babcock *et al.*, 2014). A variety of MEOP and SEOP cells are shown in Fig. 21 and discussed below.

As discussed in Sec. IV.D, various magnetostatic cavities are employed to provide the highly homogeneous magnetic field required to maintain the ^3He polarization. Polarized neutron beam lines typically have magnetic fields to maintain the neutron polarization from the polarizer to the analyzer (“guide fields”) and fields for various types of neutron spin flippers. Often a sample under study may be immersed in a strong magnetic field provided by electromagnets or superconducting magnets; hence protecting the NSF from field gradients is an important practical consideration. In addition, maintaining the polarization of the neutron beam during transitions between the homogeneous magnetic field for the

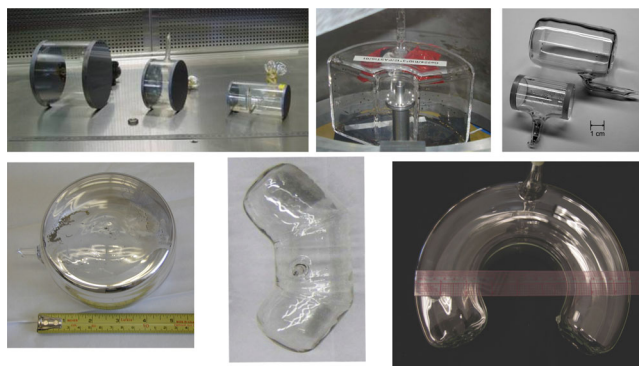


FIG. 21. Neutron spin-filter (NSF) cells. Clockwise from top left: MEOP silicon-windowed cells [largest cell is 14 cm diameter by 10 cm long (Lelievre-Berna, 2007)], MEOP wide-angle cell [6 cm inner diameter, 20 cm outer diameter, 12 cm tall (Andersen *et al.*, 2009)], cells for neutron interferometry [larger cell is 4 cm diameter by 6 cm long (Huber *et al.*, 2014)], horseshoe SEOP cell [9 cm inner diameter, 23 cm outer diameter, 7.5 cm tall (Chen, 2016)], SEOP wide-angle cell [14 cm inner diameter, 30 cm outer diameter, 8 cm tall (Ye *et al.*, 2013)], and typical SEOP NSF cell [12 cm diameter by 7 cm long (Chen *et al.*, 2011)].

^3He to the sample and/or neutron spin flipper fields can be difficult, in particular, on space-constrained apparatuses not originally designed for NSF. In some cases AFP NMR is used to flip the ^3He polarization (and thus the neutron polarization or analyzer direction) so as to avoid the need for traditional neutron spin flippers. For some applications, the polarizer NSF, sample, and analyzer NSF are all immersed in the same field, thus bypassing neutron spin transport issues but also precluding the use of strong sample fields (Ye *et al.*, 2013).

B. Neutron scattering implementation: SEOP

The first test of an NSF based on SEOP was performed at LANSCE¹⁸ (Coulter *et al.*, 1990) and applied to the search for parity violation in neutron resonances. A ^3He polarization of 70% was obtained in a 3 cm³ cell optically pumped with 1 W of laser light with a linewidth of 40 GHz that was provided by a standing-wave dye laser pumped by a krypton ion laser. A neutron polarization of 20% was produced in a beam of epithermal neutrons (energy of 0.734 eV, 0.03 nm wavelength). Soon thereafter an SEOP-based NSF was tested at the ILL (Tasset *et al.*, 1992), but the ILL program shifted to MEOP (see Sec. VII.C).

For neutron scattering, an ongoing program in SEOP-based NSFs was begun at the NCNR¹⁹ (Jones *et al.*, 2000), first demonstrated for small-angle neutron scattering (SANS) (Gentile *et al.*, 2000), and subsequently applied to polarized neutron reflectometry (PNR) (Chen *et al.*, 2004), triple-axis spectrometry (TAS) (Chen, Armstrong *et al.*, 2007), and wide-angle neutron polarization analysis (Ye *et al.*, 2013). For these applications, SEOP cells are polarized offline, transported to the neutron beam line, and stored in magnetostatic cavities. Typical NSF cells for SANS, PNR, and TAS are blown from GE180 glass, between 8 and 12 cm diameter, and filled to pressures between 1 and 2 bar. Cells for wide-angle analysis have been formed by optically sealing three sections of blown glass together so as to cover a 110° angular range (Ye *et al.*, 2013). More recently, fully blown cells for wide-angle analysis have been fabricated by first blowing a toroidal GE180 cell and then closing off a 270° section from the toroid (Chen *et al.*, 2016). The current state of the NCNR program was summarized recently (Chen, Gentile, Erwin *et al.*, 2014).

The relatively small size of the SEOP apparatus is well matched to *in situ* operation on neutron beam lines (Jones *et al.*, 2006). An *in situ* NSF has been operated routinely on the Magnetism Reflectometer at the SNS²⁰ (Tong *et al.*, 2012) and smaller *in situ* systems have been developed for the HB3 triple-axis spectrometer and the hybrid spectrometer (HYSPEC) at the SNS (Jiang *et al.*, 2014). At LENS,²¹ a continuously pumped analyzer was demonstrated for spin-echo small-angle neutron scattering (SESANS) (Parnell *et al.*, 2015). Compact NSFs with low-loss AFP NMR for on-beam

operation have been developed at KEK²² (Ino *et al.*, 2005, 2012). More recently, there has been spin-filter development at the Materials and Life Science Facility at J-PARC²³ and at the High-flux Advanced Neutron Application Reactor at KAERI²⁴ (Lee *et al.*, 2016). At J-PARC, *in situ* SEOP neutron spin filters have been demonstrated for SANS (Sakaguchi *et al.*, 2011a), PNR, and polarized neutron imaging (Hayashida *et al.*, 2016).

The SEOP programs at the Forschungszentrum Jülich (Babcock *et al.*, 2016) and the ISIS spallation source²⁵ (Beecham *et al.*, 2011) have also focused on *in situ* systems for continuously polarized ^3He . An apparatus was developed in which a ^3He polarization of 80% was maintained in a 100 cm³ cell by SEOP with two 100 W diode lasers, each spectrally narrowed to 0.2 nm bandwidth with an external cavity (Babcock, Mattauch, and Ioffe, 2011). It has been applied to PNR and SANS (Babcock *et al.*, 2016). Cells for wide-angle polarization analysis (Salhi *et al.*, 2016) fabricated from sections of fully blown doughnut-shaped GE180 cells (Salhi *et al.*, 2014) are under development.

C. Neutron scattering implementation: MEOP

Because of the large size of most compression apparatuses, NSFs based on MEOP are typically polarized offline and transported to neutron beam lines. The first demonstration of an MEOP-based NSF was performed at the University of Mainz with polarized ^3He from a piston compression system (Surkau *et al.*, 1997). A similar compressor was installed at the ILL, with the capability to produce between 50% and 55% ^3He polarization for a variety of cell designs (Heil, Dreyer *et al.*, 1999). The first application was on the IN20 triple-axis spectrometer (Kulda *et al.*, 1998). With the development of the superconducting magnetostatic cavity “Cryopol” (Dreyer *et al.*, 2000), an NSF was applied for spherical neutron polarimetry (SNP) with hot (0.08 nm wavelength) neutrons on the D3 instrument (Lelievre-Berna *et al.*, 2005). Large solid-angle polarization analysis at thermal neutron wavelengths was demonstrated on the two-axis diffractometer D1B using a cesium-coated quartz cell that analyzed a 90° range of scattering angles (Heil *et al.*, 2002). The 530 cm³ banana-shaped cell exhibited a relaxation time of 92 h. With improved optical pumping using a 30 W ytterbium fiber laser instead of an 8 W Nd:LMA laser, a spin filter with a ^3He polarization of up to 70% was demonstrated at the Mainz reactor (Batz *et al.*, 2005). Concurrently a new compressor, denoted Tyrex, was constructed at the ILL and employed to fill spin-filter cells with 65%–70% polarized ^3He gas at rates of 1 to 2 bar L/h (Petoukhov *et al.*, 2006). An NSF employing a 14 cm diameter silicon-windowed, valved, spin-filter cell was demonstrated on the D17 reflectometer, along with a “local filling” approach for refreshing the ^3He daily (Andersen *et al.*, 2006). With further improvements, a typical value of 75% ^3He polarization has been obtained in NSF cells, enabling a large

¹⁸Los Alamos Neutron Scattering Center, Los Alamos, NM.

¹⁹National Institute for Standards and Technology Center for Neutron Research, Gaithersburg, MD.

²⁰Spallation Neutron Source, Oak Ridge, TN.

²¹Low Energy Neutron Source, Bloomington, IN.

²²National Laboratory for High Energy Physics, Tsukuba, Japan.

²³Japan Proton Accelerator Research Complex, Tokai, Japan.

²⁴Korean Atomic Energy Research Institute, Daejeon, Korea.

²⁵Rutherford Appleton Laboratory, Oxfordshire, United Kingdom.

range of neutron scattering applications (Stewart *et al.*, 2006; Lelievre-Berna, 2007; Andersen *et al.*, 2009; Lelievre-Berna *et al.*, 2010). A similar compression system, denoted HELIOS, was put into operation at the FRM II reactor²⁶ (Hutanu, Masalovich *et al.*, 2007) and has been applied to SNP at the hot neutrons single crystal diffractometer POLI-HEiDI (Hutanu *et al.*, 2011). More recently, two compression apparatuses have been built at the ILL for use at the ISIS pulsed neutron and muon source (Beecham *et al.*, 2011) and the Australian Nuclear Science and Technology Organization (Lee and D'Adam, 2016).

D. Summary of neutron scattering physics enabled

The primary application of NSF in neutron scattering is in the study of magnetic materials. Neutron polarization analysis allows for separation of nuclear from magnetic scattering (Moon, Riste, and Koehler, 1969; Williams, 1988). SNP and TAS have been employed to study magnetic ordering in antiferromagnetic crystals (Hiess, Brown, Aso *et al.*, 2001; Hiess, Brown, Lelievre-Berna *et al.*, 2001; Blanco *et al.*, 2006; Zaharko *et al.*, 2006; Poole, Wills, and Lelievre-Berna, 2007), magnetization distributions (Brown *et al.*, 2002; Boehm *et al.*, 2003), magnetoelectric and multiferroic crystals and films (Brown, Forsyth, and Tasset, 2005; Lee *et al.*, 2008; Tian *et al.*, 2008; Cabrera *et al.*, 2009; Ratcliff *et al.*, 2011), and the interplay between magnetism and superconductivity (Schneider *et al.*, 2006; Chen *et al.*, 2008; Wang *et al.*, 2011). The ability of NSFs to analyze the polarization of diffusely reflected neutrons has been applied to magnetic multilayers (Nickel *et al.*, 2001), domain walls in magnetic thin films (Radu *et al.*, 2003, 2005), periodic magnetic rings (Ogrin *et al.*, 2007), and superlattices (Wildes, Björck, and Andersson, 2008). For SANS, NSFs allow for polarization analysis which has been implemented via SEOP at the NCNR (Gentile *et al.*, 2000; Krycka *et al.*, 2009), and via MEOP at the Hahn-Meitner Institute in Berlin, Germany (Wiedenmann, 2005; Keiderling *et al.*, 2008) and the ILL (Honecker *et al.*, 2010). At the NCNR, several recent studies have focused on understanding the structure of magnetic nanoparticles (Krycka *et al.*, 2009, 2010, 2011, 2013, 2014; Hasz *et al.*, 2014). SANS with NSFs has also been employed to study multiferroics (Ueland *et al.*, 2010; Ramazanoglu *et al.*, 2011), magnetostriction (Laver *et al.*, 2010), exchange bias (Dufour *et al.*, 2011), and nanowires (Pimpinella *et al.*, 2013). Polarization analysis also allows for separation of coherent from spin-incoherent scattering (Moon, Riste, and Koehler, 1969; Williams, 1988), which is relevant to biological samples due to the substantial spin-incoherent scattering from hydrogen. Applications of NSFs to this technique are emerging (Sakaguchi *et al.*, 2011a; Babcock *et al.*, 2016).

NSFs have advantages over neutron optical devices for polarized neutron imaging because they can provide a uniform analyzing power without optical distortions of the neutron beam. At the ILL, polarized neutron imaging using NSFs from the Tyrex compressor was demonstrated and applied to

visualize small-scale magnetic features and trapped magnetic flux (Dawson *et al.*, 2011). In addition, depolarization imaging has been used to image ferromagnetic phase separation in real space (Pfleiderer *et al.*, 2010).

E. Application to fundamental neutron physics

1. Accurate neutron polarimetry

For a monochromatic neutron beam, the neutron polarization produced by an NSF can be determined from the ratio of the transmission of unpolarized neutrons with the NSF polarized and unpolarized. If the NSF is used as a polarization analyzer, the analyzing power can be determined by this method and then employed to accurately measure the polarization of a neutron beam. This simple approach allows for highly accurate measurements of neutron polarization or analyzing power (Coulter *et al.*, 1990; Jones *et al.*, 2000). However, fundamental neutron physics experiments often require high neutron flux for statistics and thus polychromatic beams. With time-of-flight analysis, typically available at a pulsed neutron source, accurate polarimetry can be extended to polychromatic beams. At LANSCE neutron beam polarization was measured with an absolute accuracy of 0.3% in the neutron energy range from 40 meV to 10 eV (Rich *et al.*, 2002). More recently, Ino *et al.* (2011) reported a measurement of the near-unity average neutron beam polarization produced by supermirror benders at J-PARC with an absolute accuracy of 0.03% in the neutron wavelength range from 0.1 nm (81 meV) to 0.7 nm (1.7 meV). At continuous sources, one can use “opaque” spin filters, i.e., NSFs with sufficient opacity such that the analyzing power is near unity for the relevant wavelengths (Zimmer *et al.*, 1999). This approach has been applied to neutron beta decay (see Sec. VII.E.2).

2. Applications

The first application of NSFs to fundamental neutron physics was for measurements of the parity-violating neutron spin rotation in the 0.734 eV resonance in lanthanum. Measurements were performed at KEK in Japan (Sakai, 1997; Sakai *et al.*, 1998), LANSCE (Haseyama *et al.*, 2002), and the ILL (Heil, Humblot *et al.*, 1999). At LANSCE, SEOP with diode lasers was employed with 56% (polarizer) and 29% (analyzer) ^3He polarization obtained in 6 bar cells. At the ILL, the MEOP compression system was employed to obtain 50% ^3He polarization in a 5 cm diameter, 20 cm long NSF analyzer filled to 3 bar. More recently, a new experiment to search for time reversal violation in neutron transmission was proposed (Bowman and Gudkov, 2014).

A SEOP-based, large area NSF for long-term continuous operation (Chupp *et al.*, 2007) was employed for the study of parity violation in the absorption of cold neutrons by compound nuclei (Gericke *et al.*, 2006) and hydrogen (the “NPDGamma” experiment) (Gericke *et al.*, 2011). With two 30 W fiber-coupled diode lasers (between 1.5 and 2 nm bandwidth), 57% ^3He polarization was obtained in 11 cm diameter GE180 cells (Gentile, Chen *et al.*, 2005). The NSF was successfully operated for 1 year, but the ^3He polarization declined to $\approx 30\%$ due to long-term effects on the cell from the neutron beam (see Sec. VII.E.3.) The neutron

²⁶Forschungs-Neutronquelle Heinz Maier-Leibnitz, Munich, Germany.

polarization was determined with an accuracy of 0.1% with time-of-flight analysis.

Measurements of the electron (*A*), neutrino (*B*), and proton (*C*) asymmetries in polarized neutron beta decay provide accurate tests of the standard model of particle physics (Jackson, Treiman, and Wyld, 1957). These experiments require highly accurate determinations of the neutron polarization. For these experiments, the use of a series of opaque spin filters (see Sec. VII.E.1) was employed to determine neutron polarization to 0.1% accuracy, thus substantially reducing this contribution to the overall uncertainty budget (Schumann *et al.*, 2007, 2008; Mund *et al.*, 2013). An NSF was also used to determine the neutron polarization in a measurement of the neutron electric dipole moment via spin rotation in a non-centrosymmetric crystal (Fedorov *et al.*, 2010).

Polarized ^3He has also been used to determine the spin dependence of the neutron scattering length for ^3He , which is important for nuclear few body models. Two different methods were used. In the first method, the pseudomagnetic precession of an unpolarized neutron beam passing through a sample of polarized ^3He was detected using a neutron spin-echo spectrometer (Zimmer *et al.*, 2002). A 6 cm diameter, 10 cm long glass cell with flat, parallel, silicon windows was filled with polarized gas using the ILL MEOP compression system. In the second method, the spin dependence of the phase shift for polarized neutrons passing through a sample of polarized ^3He was determined by neutron interferometry (Huber *et al.*, 2014). A 2.4 cm diameter, 4.2 cm long glass cell with flat, parallel glass windows was polarized offline by SEOP and installed inside a neutron interferometer. The relaxation times observed during the experiments were between 75 and 120 h (Zimmer *et al.*, 2002) and between 135 and 150 h (Huber *et al.*, 2014). For the interferometry experiment, the polarization of the monochromatic neutron beam was determined with NSFs to better than 0.1% absolute accuracy. The two measurements differ by 2 standard deviations (Huber *et al.*, 2014).

3. Neutron beam effects

Effects of the neutron beam on the operation of *in situ* SEOP NSFs were first observed in the NPDGamma experiment (see Sec. VII.E.2) and soon thereafter studied at LANSCE (Sharma *et al.*, 2008). Further studies at the ILL revealed that at a neutron particle flux density of $4.7 \times 10^9 \text{ cm}^{-2} \text{ s}^{-1}$, the alkali-metal relaxation rate increased from 100 to 1000 s^{-1} (Babcock *et al.*, 2009). The relaxation rate was found to vary with the square root of the neutron flux, consistent with the source being the ionization produced by the energetic triton and proton produced in neutron absorption by ^3He . The alkali-metal relaxation rate was found to rapidly increase (time scale $< 1 \text{ s}$), followed by a further slow rise on a time scale of hundreds of seconds. Studies of the cell whitening that had been previously reported after long term exposure to the neutron beam (Chupp *et al.*, 2007) indicated that this problem may be reduced in K-Rb hybrid cells. The origin of this whitening is unknown, but speculated to be due to the production of RbH and/or alkali azides. It was demonstrated that a double cell configuration, an approach already long employed for electron scattering, is a practical method to bypass these neutron beam effects. In a later study,

Babcock *et al.* (2011) observed that the slow component of alkali-metal relaxation increases with increasing nitrogen density in the SEOP cell.

VIII. MAGNETIC RESONANCE IMAGING

A. Introduction and history

The application to MRI (Leawoods *et al.*, 2001; Möller *et al.*, 2002; Couch *et al.*, 2015) arguably provides the most visible and salient connection of hyperpolarized noble gases to commercial (in this case, medical) technologies. The two noble gases with nuclear spin 1/2, ^3He and ^{129}Xe , yield relatively long relaxation times due to the absence of a nuclear quadrupole moment that can interact with electric fields at surfaces. Conventional ^1H MRI of lung tissues is very challenging, mainly due to the very short (microseconds) NMR-signal lifetimes (Cuttillo *et al.*, 1991) resulting from the large magnetic susceptibility broadening in the lung microstructures (alveolar spaces and connecting bronchioles). Rapidly moving gases average away this broadening, but their low density relative to protons in water introduces a severe sensitivity problem. A hyperpolarized inert gas is potentially the ideal signal source for investigating lung function: it serves as a tracer of gas flow in an organ whose principal function is to move gas, it is not metabolized and interacts minimally with the body, and the enhanced magnetization overcomes the intrinsic sensitivity problem (Kastler, 1950; Cagnac, Brossel, and Kastler, 1958).

In the early 1980s, even as MRI itself was still being developed for clinical use, Bhaskar, Happer, and McClelland (1982) noted that about 11 $\text{bar cm}^3/\text{h}$ of highly polarized ^{129}Xe might be produced with 1 W of laser power, although at the time attention was focused on the ability this would afford to produce dense polarized targets. Some combination of the biological relevance of xenon (an anesthetic, which dissolves in lipid tissue), relatively fast spin-exchange rates (Zeng *et al.*, 1985), and the ability to freeze and transport xenon with minimal polarization loss (Cates *et al.*, 1990) led to the first rudimentary MR images of excised mouse lungs (Albert *et al.*, 1994). However, the tipping point for the technology was not so much the marriage itself of hyperpolarized noble gases to MRI, as it was two major improvements in related technology. For SEOP, the advent in the early 1990s of inexpensive high-power high-efficiency solid-state diode lasers increased the available photon flux from a few to many tens, even hundreds of watts (utilizing arrays of such lasers), at a fraction of the size and cost of Ti:sapphire lasers (Wagshul and Chupp, 1989). For MEOP, where the intrinsic efficiency of the process does not put a premium on high-power lasers, the principal problem solved was compression of ^3He gas polarized at very low pressure up to atmospheric pressure without substantial loss of polarization; this was accomplished at large scales through the use of titanium piston pumps (Becker *et al.*, 1994) and at smaller scales with peristaltic (Nacher *et al.*, 1999) and diaphragm (Gentile *et al.*, 2001) pumps.

The dramatic scaling up of production rates necessary for human-lung imaging occurred for both SEOP and MEOP at about the same time in the early 1990s, leading rapidly in both cases to the first human-lung MR images using hyperpolarized

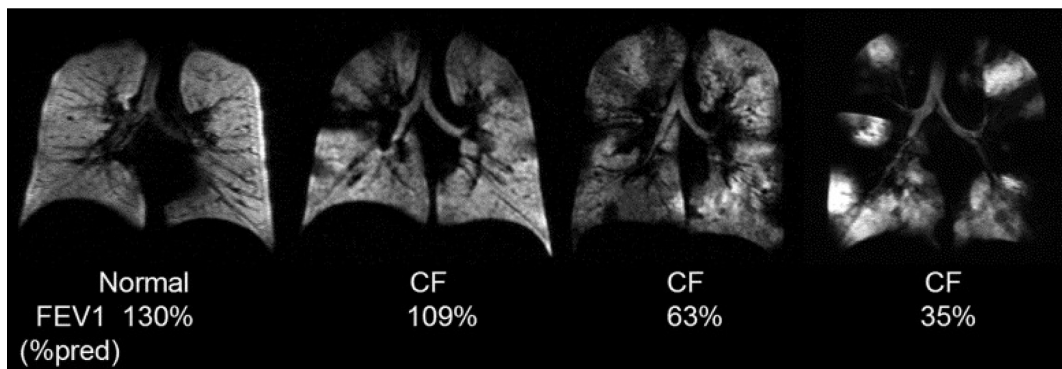


FIG. 22. Coronal (view from the front of the body) ^3He magnetic resonance images from a healthy subject (left) and three patients with cystic fibrosis (CF). The number of ventilation defects increases with worsening results of a standard global ventilation test FEV_1 (forced expiratory volume in 1 s). FEV_1 is shown as a percentage of the predicted value for a healthy subject. From Mentore *et al.*, 2005.

^3He , reported in 1996 (Ebert *et al.*, 1996; MacFall *et al.*, 1996). For the next decade or so, the field was dominated by imaging with ^3He , which was available for \$100/(bar-L) at near 100% isotopic abundance, has a large magnetic moment (75% of the proton's moment), and for which the physics (and hence the scale up) of SEOP and MEOP were generally better understood than for ^{129}Xe SEOP. In comparison, naturally abundant xenon (26% ^{129}Xe) costs \$50/(bar-L) (isotopically enriched samples cost about an order of magnitude more), and the ^{129}Xe nuclear moment is roughly one-quarter that of the proton. Commercial development of SEOP began²⁷ in 1996; currently two small companies²⁸ offer high volume ^3He and ^{129}Xe SEOP systems.

In small-animal work, MR microscopy with ^3He (Chen *et al.*, 1998) was developed to resolve airways down to the fifth generation of branching in the guinea pig lung. In humans, stunning 3D data sets of the lung with resolution of a few millimeters can now be acquired in a ten-second breath hold (Qing *et al.*, 2015). A variety of diseases have been studied, among them cystic fibrosis (Mentore *et al.*, 2005; Paulin *et al.*, 2015; Flors *et al.*, 2016) (see Fig. 22), asthma (Tustison *et al.*, 2010; Kruger *et al.*, 2014), and emphysema (Spector *et al.*, 2005; Quirk *et al.*, 2011; Kirby *et al.*, 2013).

B. Gas handling and delivery

Using SEOP, ^3He can be polarized in reusable valved glass cells at pressures of up to 10 bar prior to being released for use in imaging. Jacob, Morgan, and Saam (2002) provided a recipe for making such cells from inexpensive borosilicate (Corning Pyrex) glass, with a length of capillary tube separating the main cell volume from the wetted valve materials. The high pressure sets a limit on T_1 due to the ^3He - ^3He dipolar mechanism (Newbury *et al.*, 1993) and cells are frequently dispensed and refilled, so that using the low-permeability glasses that produce the longest wall-relaxation times (see Sec. IV.B.1) is not necessary. When polarized by MEOP for imaging, ^3He is typically compressed to a few bar

²⁷Magnetic Imaging Technologies, Inc., later acquired by GE Healthcare.

²⁸Polarean Inc. (Research Triangle Park, NC); Xemed LLC (Durham, NH).

and stored in separate long- T_1 storage cells (Heil *et al.*, 1995). Such cells have not only been used to supply ^3He for imaging experiments performed locally, but have also been transported by air in compact magnetized boxes, for use in MRI at distant sites around the world (Wild *et al.*, 2002; van Beek *et al.*, 2003; Thien *et al.*, 2008).

While more sophisticated ventilation systems have been employed, for both small-animal (Nouls *et al.*, 2011) and human (Guldner *et al.*, 2015) imaging, the most straightforward and widely used method to deliver hyperpolarized ^3He to human subjects has been the use of plastic-valved flexible bags made of and/or coated with one or more fluoropolymer materials; the most used among these is Tedlar.²⁹ Both the cell containing polarized ^3He and the bag are connected to a gas-handling manifold via a plastic valve built into the bag. Once the bag is filled with the requisite gas mixture, it is detached from the manifold and handed to the subject inside the magnet. We note that imaging protocols often call for ^3He to be mixed with nitrogen to conserve it in cases where there is plenty of signal intensity available. The use of ^3He for human-lung MRI is subject to country-specific regulations, such as FDA regulation in the United States, where it requires an exemption as an investigational new drug (IND).

C. Imaging modalities

Several important characteristics of hyperpolarized ^3He are of immediate consequence to MRI and necessitate rethinking of the pulse-sequence and signal-acquisition techniques commonly used for conventional ^1H MRI. First, there is no relevant thermal recovery of the magnetization: the thermally polarized signal is negligible and typically characterized by a long recovery time T_1 . Hence, while no (time-consuming) signal averaging is required, strategies are necessary to ration the magnetization inhaled in one breath to acquire the hundreds to thousands of data sets with different applied magnetic-field gradients needed to obtain an image. The imaging sequences in early work employed successive small-angle excitations (Kauczor *et al.*, 1997; de Lange *et al.*, 1999) or successions of rapidly refocused gradient echoes (Saam *et al.*, 1999).

²⁹Jensen Inert Products, Coral Springs, FL.

Strategies for variable-strength excitations (variable flip angle) were also explored to make more efficient use of the magnetization (Markstaller *et al.*, 2000; Santyr, Lam, and Ouriadov, 2008; Deppe and Wild, 2012). Thus, while total imaging time is ultimately limited by T_1 relaxation of the inhaled gas, the fact that there is no waiting period due to thermal recovery means that image acquisition speed is not T_1 limited; this was exploited to make frame-by-frame animations of gas motion during breathing (Saam *et al.*, 1999; Salerno *et al.*, 2001), pointing toward the use of ^3He to study lung function (van Beek *et al.*, 2004; Fain *et al.*, 2010) as well as structure. More recently, a wide range of more sophisticated pulse sequences and imaging protocols has been introduced (Salerno *et al.*, 2003; Ajraoui *et al.*, 2010; Horn *et al.*, 2017).

Second, the available ^3He polarization depends only on the process (SEOP or MEOP) and the subsequent prevention of T_1 relaxation in the stored sample prior to the start of image acquisition. Contrary to the case in conventional ^1H imaging, it does not depend on the applied magnetic field. Most human and animal MR imaging is ultimately dominated by conductive currents that produce Johnson noise in the sample and not in the probe. One can combine these facts to conclude that the SNR across a wide parameter space is approximately independent of the applied field, although Parra-Robles, Cross, and Santyr (2005) identified an optimal low field of ≈ 0.1 T, with a factor of 2 loss in SNR occurring near the edges of the range 0.01 to 1 T. In the lung, there is the added prospect of improving image fidelity at low field by reducing artifacts due to susceptibility broadening created by multiple air-water interfaces (Salerno *et al.*, 2005). Low-field MRI using ^3He has been developed and implemented both by Owers-Bradley *et al.* (2003) at 0.15 T and by Durand *et al.* (2002), the latter using a commercial 0.1 T MR imager to obtain images with higher SNR and greater fidelity than achieved at the more standard 1.5 T field. In principle, one should be able to image with ^3He at even lower fields (< 50 mT). In practice, the results have been mixed, as one has to contend with increased pickup and $1/f$ noise at lower Larmor frequencies, and concomitant gradients, i.e., the inability at very low fields to assume that gradients orthogonal to the applied-field direction are negligible (Yablonskiy, Sukstanskii, and Ackerman, 2005). There is often the need to develop dedicated pulse sequences, probes, gradient coils, and other components, since commercial MRI has developed for several decades almost exclusively at ever-increasing applied fields. Despite these limitations, ultra-low-field MRI with ^3He has been developed and implemented by Venkatesh *et al.* (2003) and Bidinosti *et al.* (2004), and by Tsai *et al.* (2008), who studied the orientation dependence of ventilation in the human lung with a custom-built 5-mT Helmholtz-coil pair in which a human subject could stand.

Third, paramagnetic molecular oxygen strongly relaxes ^3He [at a rate of about 0.5 s^{-1} for pure O_2 at 1 bar (Saam, Happer, and Middleton, 1995)], typically limiting imaging time *in vivo* to about 30 s for a single inhaled bolus of gas. Conventional MRI hardware is more than up to the task of using the magnetization during this time to acquire biologically relevant information, but one must be aware and account for the fact that a change in ^3He density cannot easily be discerned from a change in magnetization, potentially confounding image interpretation. Schemes

for continuous or quasicontinuous breathing of ^3He have been developed for humans and for small animals. This apparent limitation can also be turned around to quantify regional pulmonary oxygen pressure in human lungs (Deninger *et al.*, 1999; Miller *et al.*, 2010; Hamedani *et al.*, 2013).

While state-of-the-art spin-density-weighted ^3He lung images are quite spectacular in terms of brightness and resolution (see Fig. 22), there are several other mechanisms that can be used to generate MRI contrast. T_1 weighting via interaction with O_2 has already been discussed; highly mobile ^3He is also particularly suited to diffusion imaging (Schmidt *et al.*, 1997), where the attenuation of signal due to diffusion through applied magnetic-field gradients (Stejskal and Tanner, 1965) can be used to map the mean-squared displacement of spins during some characteristic time interval. This is particularly useful in the study of emphysema and related diseases, which are characterized by disintegration of the walls separating the many small ($\approx 300\text{ }\mu\text{m}$) alveolar (gas-exchange) spaces. The characteristic MRI diffusion time is easily chosen for ^3He to be significantly greater than the diffusion time across alveolar spaces in a healthy lung, but on the order of or less than the diffusion time across the larger spaces that are created as the disease destroys the alveolar walls. Bright regions are those where ^3He diffusion is restricted by many boundaries; diseased areas, where there is strong attenuation, show up darker, as shown in Fig. 23.

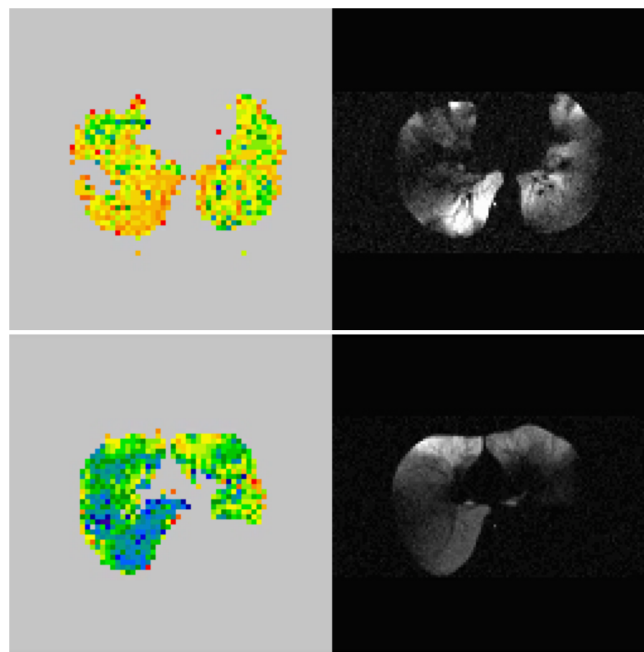


FIG. 23. Axial (perpendicular to the spine) ^3He magnetic resonance images of a cross section of the ventilated lung in two emphysema patients, top and bottom. Images on the right are spin-density weighted while those on the left are maps of apparent diffusion coefficient (ADC), where red represents the most restricted air spaces regions and blue the least restricted. The blue regions correspond to the most diseased tissue, where the alveolar walls have been destroyed. These regions do not necessarily correlate with the poorly ventilated regions seen in the spin-density weighted images, which demonstrate the potential for greater specificity with ADC mapping. From Conradi *et al.*, 2006.

The quantitative measure of mean-squared displacement mapped in such images is known as the apparent diffusion coefficient (ADC). Several groups developed and refined this technique (Fain *et al.*, 2005; Conradi *et al.*, 2006; Wang *et al.*, 2008). A quantitative model based on anisotropic ^3He diffusion and ADC measurement was developed by Yablonskiy *et al.* (2009) and supported through comparison to histologic sections of healthy and diseased tissue (Woods *et al.*, 2006). An alternate competing view of such modeling and its limitations was put forth by Parra-Robles and Wild (2014).

D. Current limitations and the future of ^3He MR imaging

While substantial progress has been made during the last two decades in improving image quality and applying various contrasts to study healthy and diseased lungs, these applications to date are generally termed “preclinical,” meaning that the technique has yet to find a specific application that warrants regular clinical use. One of the more promising avenues may be MR imaging of neonates (Krjukov *et al.*, 2007; Tkach *et al.*, 2014; Walkup *et al.*, 2015), where it is known that lung development is the limiting factor for survival of premature infants and where MRI is particularly attractive for longitudinal studies due to the absence of ionizing radiation.

Knowledge of the physics and correlated technological issues surrounding production of highly polarized ^{129}Xe has improved somewhat in the last decade (Nelson and Walker, 2001; Nikolaou *et al.*, 2013; Freeman, Emami, and Driehuys, 2014). Coupled with native interest in the more biologically active xenon gas as a signal source and with the scarcer (and much more expensive) availability of ^3He (Shea and Morgan, 2010), it would appear that, short of mining the Moon, with its higher isotopic fraction of ^3He or dedicated tritium production (Wittenberg *et al.*, 1992; Woods, 2013), future further development of hyperpolarized-gas MRI as a clinical tool will primarily focus on ^{129}Xe .

IX. PRECISION MEASUREMENTS

Although most applications of hyperpolarized ^3He exploit the large achievable magnetizations, ^3He has tremendous potential itself for precision spectroscopy due to both large magnetization and long coherence times (T_2). In a generic spectroscopy experiment, the uncertainty principle implies a measurement uncertainty $\sigma_\nu = 1/T_2^*$ for a single measurement on a single particle. For the $N = [\text{He}]V$ polarized nuclei, and repeating the measurement $m = t/T_2^*$ times, the quantum-limited precision of frequency measurement becomes $\sigma_\nu = T_2^{*-1}/\sqrt{Nm} = 1/\sqrt{T_2^*Nt}$. Defining the equivalent frequency noise spectrum $\delta\nu$ through $\sigma_\nu = \delta\nu/\sqrt{t}$ implies a quantum projection frequency noise of

$$\delta\nu = \sqrt{\frac{1}{[\text{He}]VT_2^*}} = 1.1 \times 10^{-13} \frac{\text{Hz}}{\sqrt{\text{Hz}}} \quad (51)$$

for a $V = 1 \text{ cm}^3$ volume if T_2 is limited by dipole-dipole relaxation (Sec. IV.A). The corresponding magnetic (Romalis,

2013) and rotation (Donley and Kitching, 2013; Walker and Larsen, 2016) sensitivities are $3 \times 10^{-21} \text{ T}/\sqrt{\text{Hz}}$ and $2.5 \times 10^{-9} \text{ deg}/\sqrt{\text{h}}$. As such samples are now realizable, the primary challenges are detection and, for nonmagnetic applications, compensation for environmental magnetic noise. Detection strategies to date include inductive pickup (Chupp *et al.*, 1988), SQUID detection (Greenberg, 1998; Savukov *et al.*, 2008; Gemmel *et al.*, 2010), external atomic magnetometers (Cohen-Tannoudji *et al.*, 1969; Kraft *et al.*, 2014; Koch *et al.*, 2015a, 2015b), fluxgate magnetometers (Wilms *et al.*, 1997; Guigue *et al.*, 2015), and, specific to SEOP implementations, using the embedded alkali atoms to detect the ^3He precession (Kornack and Romalis, 2002; Zou *et al.*, 2016).

The magnitude of the magnetic field produced by the precessing ^3He nuclei is of the order of

$$B_{\text{He}} = \kappa \frac{8\pi\mu_{\text{He}}}{3} [\text{He}]P_{\text{He}} = \kappa \times 0.2 \mu\text{T} \quad (52)$$

for 1 amg of hyperpolarized ^3He . The factor $\kappa \sim 6$ is, for imbedded alkali detection, the frequency shift enhancement factor exploited for EPR polarimetry (Sec. II.F), while for the other methods that detect the classical magnetic field produced by the nuclei it is a geometrical factor, typically less than 1, that depends on cell and detector geometry. Inside a multilayer magnetic shield made of high permeability metal, the magnetic noise can be $10^{-14} \text{ T}/\sqrt{\text{Hz}}$ or better, giving a potential SNR for the ^3He detection approaching 150 dB in a 1 Hz bandwidth. No experiment has yet attained this value, but a recent experiment (Gemmel *et al.*, 2010; Allmendinger *et al.*, 2014a) showed $5 \text{ nHz}/\sqrt{\text{Hz}}$ for a 2 mbar cell with $T_2^* = 60 \text{ h}$ in a He-Xe comagnetometer experiment, with the He contribution likely substantially better than this.

Both SEOP and MEOP involve spin-polarized paramagnetic species (alkali atoms, metastable He atoms, free electrons) that produce effective paramagnetic fields B_A that in turn cause NMR frequency shifts of the ^3He analogous to the EPR frequency shift $\delta\nu_A = b_A^{\text{He}}P_A$ of Eq. (23):

$$\delta\nu_{\text{He}} = -\gamma_{\text{He}}B_A = -\gamma_{\text{He}}b_A^{\text{He}}P_A. \quad (53)$$

The field $B_A = b_A^{\text{He}}P_A$ is 2 mG for fully polarized Rb at a density of 10^{14} cm^{-3} . This shift must be carefully managed in any precision measurement in which the ^3He is in the same region as the paramagnetic species. Most precision NMR experiments to date manage the problem by transporting the polarized ^3He to a region free of paramagnetic species, with a key exception being the alkali comagnetometer approach of Sec. IX.C.

A. Comagnetometry

Any use of ^3He beyond magnetometry (symmetry violations, rotation) must account for the inevitable magnetic-field fluctuations in any environment. Thus a second species is required to separate out magnetic from nonmagnetic interactions, generally by either locking one species to an atomic clock (Bear *et al.*, 1998), comparing the precession phase

directly (Chupp *et al.*, 1988; Allmendinger *et al.*, 2014a), or taking frequency ratios (Chupp *et al.*, 1988).

A special case to note is the proposed use of ^3He as a comagnetometer for neutron EDM experiments (Borisov *et al.*, 2000; Savukov *et al.*, 2008; Kim and Clayton, 2013; Tsentlovich, 2014; Chu and Peng, 2015). The cryogenic compatibility of ^3He is attractive to colocate with the neutrons, and the small 10% difference in the magnetic moments of ^3He and neutrons allows dressing techniques to give them effectively the same moment, thus allowing common-mode rejection of magnetic-field noise (Golub and Lamoreaux, 1994). Recent experiments have shown the viability of this approach (Chu *et al.*, 2011; Eckel *et al.*, 2012).

Dual ^3He - ^{129}Xe magnetometry was demonstrated by Gemmel *et al.* (2010) using SQUID detection of MEOP-produced ^3He and SEOP-produced ^{129}Xe . They demonstrated magnetic sensitivity of 1 fT in about 200 s of integration. Further developed versions of this approach, reaching coherence times of more than 100 h (Heil *et al.*, 2013), were used to set new limits on monopole-dipole interactions (Tullney *et al.*, 2013) and charge-parity-time (*CPT*) and Lorentz invariance (Allmendinger *et al.*, 2014a). In this latter experiment, an anomalous phase precession was attributed to interactions between the precessing nuclei due to the internal fields produced by the nuclei themselves, the magnitude of the effect being consistent with $\kappa_0 = 1$ [Eq. (23)], implying a significant contact interaction between the species (Romalis *et al.*, 2014) that would call into question the geometrical calibration of the EPR frequency shift (Romalis and Cates, 1998). This argument was disputed by Allmendinger *et al.* (2014b). Similar Xe-He techniques are being developed for new Xe EDM experiments (Heil *et al.*, 2013; Kuchler *et al.*, 2016).

He-Xe comagnetometry using the embedded magnetometer always available in SEOP was studied by Sheng, Kabcenell, and Romalis (2014). The alkali field shifts were nulled by strongly driving the Rb EPR resonance during free precession of the spins. Of particular emphasis was systematic shifts caused by magnetic field and temperature gradients.

B. Masers

The first demonstration of the potential of hyperpolarized ^3He for precision measurements was in a maser configuration, by Robinson and Myint (1964), who reported a statistical uncertainty of $500 \text{ nHz}/\sqrt{\text{Hz}}$ using a lamp-pumped MEOP setup. This experiment also pioneered the use of a dual cell system, with the MEOP process occurring in one bulb, connected by a few mm diameter tube to the spherical maser chamber. The use of a relatively low frequency (100 kHz) means that the resonant cavity used for most electron spin masers is replaced by an open *LRC* circuit. Nearly 25 years later, Richards *et al.* (1988) and Flowers, Petley, and Richards (1990) built similar systems for precision magnetometry at fields up to 0.1 T. Later, Gilles, Monfort, and Hamel (2003) built a laser-pumped MEOP maser for earth field measurements, comparing the performance to a metastable ^4He magnetometer and obtaining a field sensitivity of $20 \text{ pT}/\sqrt{\text{Hz}}$.

In 1994, dual species SEOP-pumped ^3He and ^{129}Xe masers were introduced (Chupp *et al.*, 1994). The maser bulb was again isolated from the pumping bulb, in order to suppress frequency shifts from optically pumped Rb. With dual species operation (shown in Fig. 24) (Stoner *et al.*, 1996; Bear *et al.*, 1998), the common-mode magnetic-field fluctuations were canceled to a high precision, enabling the dual species maser to be sensitive to nonmagnetic interactions such as permanent electric dipole moments. Using ^3He as a comagnetometer, Rosenberry and Chupp (2001) reported the most precise limits on a possible ^{129}Xe EDM to date. An earlier version demonstrated the development of molybdenum electrodes with long relaxation times (Oteiza, 1992). A dual species maser under development for future EDM tests was described by Funayama *et al.* (2015).

The dual species maser was used in a series of symmetry violation tests by Walsworth and co-workers. Bear *et al.* (2000) set new limits on Lorentz and *CPT* violations, further improved by Cane *et al.* (2004). These experiments searched for sidereal variation in the relative precession of the two nuclei as the laboratory rotates with respect to the distant stars (Walsworth *et al.*, 2000; Walsworth, 2006). Glenday *et al.* (2008) used the dual species maser and a nearby SEOP ^3He cell inside a separate magnetic shield to search for anomalous interactions between neutrons. Extensive modeling and details of the experimental implementation of the dual species maser,

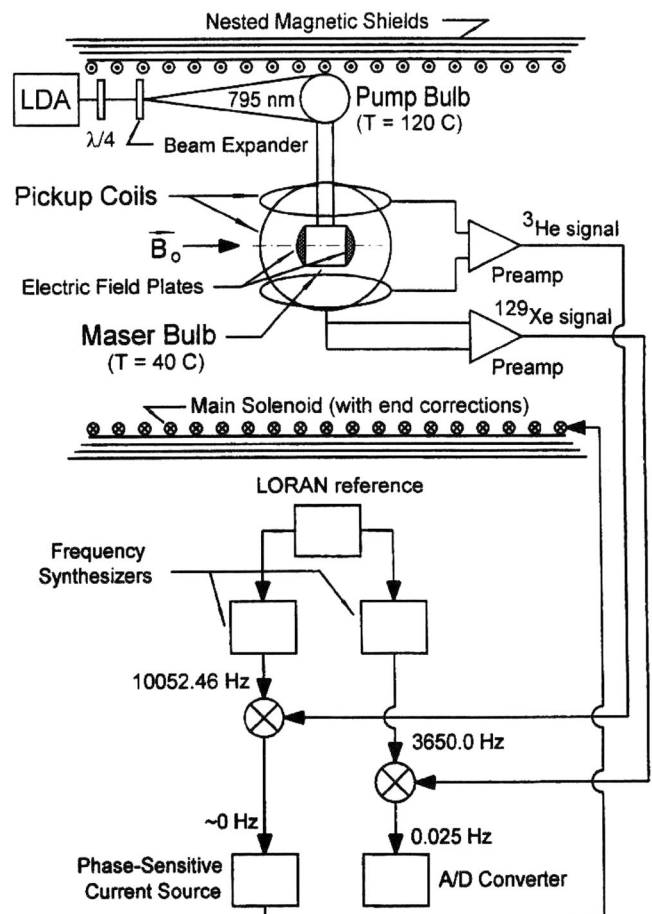


FIG. 24. Dual species maser. From Stoner *et al.*, 1996.

which reaches a precision of about 10 nHz, are given by Glenday (2010).

C. Alkali-He comagnetometer

The maser and FID approaches to precision spectroscopy and comagnetometry with ^3He avoid systematic shifts due to alkali spin-exchange fields by making measurements in the absence of the alkali atoms. Another approach, pioneered by Romalis and co-workers (Kornack and Romalis, 2002; Kornack, Ghosh, and Romalis, 2005), is to use the spin-exchange fields in an alkali- ^3He SEOP setup to null the magnetic sensitivity while maximizing sensitivity to non-magnetic interactions. Taking advantage of both the spin-exchange field B_{He} from the ^3He nuclei [Eq. (23)] and the analogous field $b_{\text{He}}^{\text{A}}P_{\text{A}}$ from the polarized alkali atoms, they showed that when $B_z + b_{\text{He}}^{\text{A}}P_{\text{A}} + b_{\text{A}}^{\text{He}}P_{\text{He}} = 0$ the alkali-metal atoms experience no response to transverse magnetic fields. Application of a transverse magnetic field causes precession of the ^3He whose spin-exchange field cancels the applied field. However, when a nonmagnetic interaction is present, the nuclei and alkali-metal atoms do not respond proportionately to their gyromagnetic ratios and the electrons then precess. The alkali atoms acquire a sensitivity to nonmagnetic interactions with the ^3He nucleus that is amplified by the factor $\gamma_{\text{S}}/\gamma_{\text{He}} = 617$.

The K- ^3He comagnetometer was used to demonstrate a sensitive gyroscope (Kornack, Ghosh, and Romalis, 2005), set new limits on anomalous nuclear spin-dependent forces (Vasilakis *et al.*, 2009), and set new limits on Lorentz and *CPT* violation (Brown *et al.*, 2010). These experiments exhibit a frequency sensitivity in the range of 18 pHz.

D. Magnetometry

Optically pumped metastable ^4He magnetometers have been extensively developed (Scheerer, 1985) and used for military, geomagnetic, and planetary magnetic-field measurement applications (Dunlop *et al.*, 1999).³⁰ Despite their greater potential sensitivity, ^3He NMR-based magnetometers have not been commercialized due to challenges of the NMR readout. Development has been primarily for specialized precision physics measurements, especially neutron EDM experiments. Borisov *et al.* (2000) pioneered a scheme where they measured the absolute ^3He polarization in a MEOP cell, let the gas expand into the measurement volume, applied a $\pi/2 - T - \pi/2$ Ramsey sequence, then recompressed the gas into the MEOP cell where the final polarization was measured. The magnetic-field measurement precision was 24 fT.

Extensive studies of ^3He -Cs magnetometers have been made for integration into the ultracold neutron EDM experiment being developed at PSI.³¹ The basic principle is to transport ^3He from a MEOP pumping setup inside the magnetic shield where the EDM experiment is located. They detect free induction decay of the ^3He with

lamp-pumped (Kraft *et al.*, 2014) or multiple laser-pumped (Koch *et al.*, 2015a, 2015b) Cs magnetometers.

For measurements of Tesla-scale magnetic fields, ^3He is attractive due to its immunity to systematic errors. Nikiel *et al.* (2014) demonstrated astounding relative accuracy of 10^{-12} at 1 T, with further advances promised from the use of essentially perfect spherical containers for the ^3He (Maul *et al.*, 2016).

E. Searches for axionlike interactions

Since ^3He has similar spin and electromagnetic properties as a free neutron, it can be used to investigate possible exotic spin-dependent interactions with matter. In particular, hypothetical axionlike particles would generate a *CP*-violating scalar-pseudoscalar coupling of strength $g_s g_p$ (Moody and Wilczek, 1984) with a Yukawa-like spatial dependence of length scale Λ , which is inversely proportional to the axionlike mass. The most straightforward approach is to search for a shift in the ^3He resonance frequency as the distance between a macroscopic mass and the ^3He gas is modulated (Tullney *et al.*, 2013). Another remarkable approach, proposed by Pokotilovski (2010), is to use the fact that spin relaxation of ^3He is sensitive to magnetic-field gradients, which in this case means gradients of the hypothetical Yukawa scalar-pseudoscalar coupling. Measurements of the transverse spin-relaxation rates are particularly sensitive to these nonmagnetic gradients and, in particular, the long coherence times of Gemmel *et al.* (2010) were used to set new limits (Petukhov *et al.*, 2010; Fu, Gentile, and Snow, 2011). For Λ smaller than the cell size, the classic transverse relaxation, analogous to Eq. (48), must be modified (Petukhov, Pignol, and Golub, 2011). Measurements of the ^3He transverse relaxation rate as a function of parameters such as bias magnetic field, density, and mass distribution then potentially reveal the existence of the scalar-pseudoscalar coupling. A series of experiments have been done along these lines (Fu, Gentile, and Snow, 2011; Chu *et al.*, 2013; Guigue *et al.*, 2015; Yan *et al.*, 2015). At the shortest distances $\Lambda < 20 \mu\text{m}$, this method is the most sensitive (Guigue *et al.*, 2015); for $\Lambda > 50 \mu\text{m}$ the ^3He -Xe comagnetometry prevails (Tullney *et al.*, 2013), and the small intermediate regime is limited by a dual isotope Xe comagnetometer (Bulatowicz *et al.*, 2013). A new approach for improving axion limits using a resonance method was proposed as well (Arvanitaki and Geraci, 2014).

X. FUTURE TRENDS

We close with some of the scientific and technical challenges in the theory and practice of SEOP and MEOP. The limiting polarization for SEOP is still not clear. Although the discovery of excess relaxation that scales with alkali-metal density has modified our understanding of this limit, the present analysis is purely phenomenological as the origin of this cell-dependent relaxation is not fundamentally understood. Indeed both its origin and magnetic-field effects on ^3He cells are new twists in the quest for understanding of wall relaxation, but perhaps they will turn out to be clues rather than simply a new source of confusion. The recent higher

³⁰Polatomic Inc., Richardson, TX.

³¹Paul Scherrer Institute, Villigen PSI, Switzerland.

polarizations from SEOP observed at the NCNR (Chen, Gentile, Erwin *et al.*, 2014) also suggest that there is more to the limit than excess relaxation. These results must be reproduced by other groups.

Hybrid SEOP has yielded a significant gain in the polarizing rate. A possibility for a further increase in rate is the operation in a ≈ 50 mT magnetic field, which has been shown to reduce alkali-alkali relaxation by a factor of ≈ 2 (Kadlecek, Anderson, and Walker, 1998; Kadlecek, 1999). This approach would be the most useful for the typical 1.5 bar pressure in NSFs, for which this contribution dominates over alkali-buffer gas relaxation. However, the detrimental effect of even such modest magnetic fields on ^3He wall relaxation (Jacob, 2003; Chen *et al.*, 2011) would have to be substantially reduced to employ this approach. Employing the very low relaxation rate for sodium (Borel *et al.*, 2003) is hampered by the browning of aluminosilicate glass at the high temperatures required (Babcock, 2005; Chen *et al.*, 2011). Although this issue could be addressed with the use of sapphire (Masuda *et al.*, 2005), practical construction of such cells for routine applications would be required. Again, we see that poorly understood wall-relaxation properties limit the performance of SEOP.

The continuing development of laser technology has greatly benefitted SEOP. Cost-effective laboratory systems can presently make use of chirped VHGs to narrow 100 W single diode bars to 0.25 nm bandwidth. However, fiber-coupled lasers are required or preferred for various *in situ* systems. If VHG-narrowed, fiber-coupled systems (Liu *et al.*, 2015) were commercialized and could reach this power level with similar bandwidth and comparable cost, the performance of targets and *in situ* NSFs could be improved. Similarly, ultranarrow lasers could yield benefits, but their utility has not been experimentally investigated. A study of the practical improvement in SEOP for narrowing high-power diode lasers from 2 to 0.2 nm was reported (Chann *et al.*, 2003), However, no such study has been reported for further narrowing from 0.2 to 0.04 nm (Gourevitch *et al.*, 2008), even though this is now commercially available.³² Another possible approach to future lasers is alkali-metal lasers (Zhdanov and Knize, 2012). If 770 nm lasers were to become as available as 795 nm lasers for the same cost, pure K pumping should be further investigated. Despite tremendous progress in the last two decades, a mature, common model for SEOP in both K-Rb hybrid and pure alkali-metal cells remains to be developed and tested.

The most significant new trend in MEOP is a high-field-high-pressure operation. To date the achievable polarization at high field has not exceeded that obtained at low field and low pressure. Whereas operation in low field is typically easier, operation at high field is under development for a polarized ^3He ion source at RHIC (Maxwell *et al.*, 2016) and for high-accuracy magnetic-field measurements (Nikiel *et al.*, 2014; Maul *et al.*, 2016). Recent studies revealed a light-induced relaxation mechanism that wastes most of the angular momentum deposited from the pumping light and limits the achievable polarization at higher pressures, but is not well understood. Whereas the lower polarizations currently

obtained at higher pressure are acceptable for MRI applications, NSF and electron-scattering applications typically demand the highest polarizations. However, the current remote mode of operating large compressors inherently introduces lower time-averaged polarization because of the need to replace cells or perform local refilling operations. If future work revealed the origin of this relaxation and a path to its elimination, there is the potential for simplifying and shrinking large-scale compression apparatuses for these applications and thus perhaps allowing for continuous operation, as well as for more efficient use of available pumping light and larger production rates for MRI applications.

The high luminosity planned for future experiments at JLAB will put new demands on SEOP targets for electron scattering. The high polarizing rates achievable with hybrid pumping make it possible to tolerate the ^3He relaxation produced by higher beam currents, but reducing the transfer time between the two volumes of the double cell is desirable. Since the ^3He polarimetry is typically done using EPR in the optical pumping cell, more rapid transfer increases the accuracy of the determination of the polarization in the target cell. Toward that end, targets employing convective transfer between the two volumes (see Fig. 25) are under development for these future experiments.

As NSFs are employed on an increasing range of neutron scattering instruments, there will be new challenges for both methods. For neutron scattering, NSFs have been typically polarized remotely and transferred to neutron beam lines, but it is becoming of increasing interest to operate NSFs continuously or pseudocontinuously so as to maximize the figure of merit and decrease polarization correction uncertainties. For SEOP, apparatuses that can fit within difficult space constraints and satisfy laser safety requirements are required. For MEOP, local filling is likely to be used more extensively. Increasing the range of applications will require NSFs with large and/or complex cells and/or greater tolerance to stray magnetic fields. Finally, some NSFs may provide the greatest impetus for achieving the highest possible ^3He polarizations

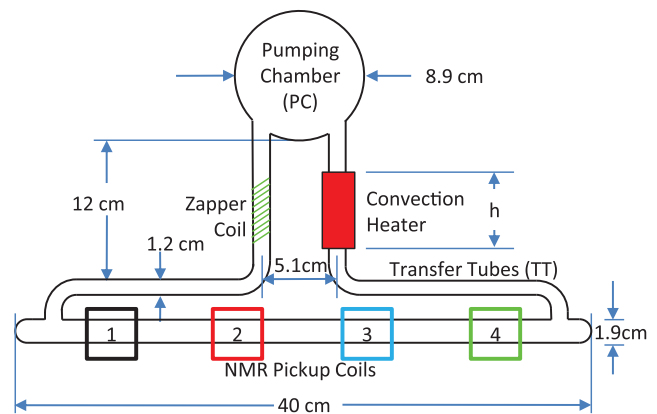


FIG. 25. Design of a convection cell for decreased transfer time between the two volumes of a double cell. The flow of gas was monitored using an NMR tagging technique, in which the zapper coil was used to depolarize a slug of gas and NMR signals were then detected at each of four locations along the target chamber (labeled 1, 2, 3, and 4). From Dolph *et al.*, 2011.

³²Optigrate Corp., Oviedo, FL.

for two reasons: (1) some NSF's are relatively simple, single, cells at pressures near 1 bar, for which the limiting polarization in a practical application can be close to that possible under ideal conditions; and (2) high neutron polarization and/or analyzing power are typically desirable for neutron scattering experiments in which a small component of magnetic scattering is separated from a much larger component of nuclear scattering (Gentile *et al.*, 2005). In contrast with the typical figure of merit proportional to P_{He}^2 for the running time of an experiment, such situations yield a stronger dependence that is closer to P_{He}^4 .

In the area of ^3He polarimetry, the high polarizations achievable in NSF's provide both the impetus and a methodology for tests and possible improvements. For example, if an accurate EPR apparatus were operated on an *in situ* NSF, a careful comparison of the two methods would enable a potential improvement in the determination of κ_0 . In the other direction, improved EPR, water-based NMR, and/or magnetometry could provide the means for a determination or better limit on the small cross section for neutron absorption by ^3He with parallel spins (Huber *et al.*, 2014). Finally if NSF's are to be used to polarize high-flux beams for fundamental neutron physics applications, double cells for neutron beams will have to be developed.

While the long-term future availability of ^3He for medical imaging remains uncertain, ^{129}Xe is naturally abundant and relatively inexpensive. The advancements in ^{129}Xe polarization technology and work comparing ^3He and ^{129}Xe support ^{129}Xe being a viable alternative to ^3He in many, but not all, instances. Furthermore, the unique physical properties of ^{129}Xe allow for dissolved-phase imaging capable of measuring biomarkers related to gas uptake and exchange. In patient populations where the slight dissolution of ^{129}Xe into blood is a potential issue, such as infants, ^3He may continue to be the preferred hyperpolarized gas.

Spin-polarized ^3He can be expected to be desirable for many applications in precision measurements. The insensitivity of ^3He to nonmagnetic interactions, plus its extremely small fundamental bandwidth, make it ideal for comagnetometry applications for symmetry tests like searches for electric dipole moments. ^3He is relatively immune to chemical shifts, so it can be considered a primary magnetic-field standard. In a synchronous SEOP configuration (Korver *et al.*, 2013, 2015) there is promise to avoid systematic shifts from the alkali fields for making absolute magnetic-field measurements or rotation measurements with the inherently high sensitivity of an imbedded alkali magnetometer.

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