Hybrid Optical Pumping of Optically Dense Alkali-Metal Vapor without Quenching Gas

M. V. Romalis
Department of Physics, Princeton University, Princeton, New Jersey 08544, USA

(Received 25 September 2010; published 7 December 2010)

Optical pumping of an optically thick atomic vapor typically requires a quenching buffer gas, such as N_2, to prevent radiation trapping of unpolarized photons which would depolarize the atoms. We show that optical pumping of a trace contamination of Rb present in K metal results in a 4.5 times higher polarization of K than direct optical pumping of K in the absence of N_2. Such spin-exchange polarization transfer from optically thin species is useful in a variety of areas, including spin-polarized nuclear scattering targets and electron beams, quantum-nondemolition spin measurements, and ultrasensitive magnetometry.

DOI: 10.1103/PhysRevLett.105.243001 PACS numbers: 32.80.Xx, 32.50.+d

Optical pumping is an extremely versatile technique that can be used to achieve nearly complete atomic polarization [1]. However, in an optically thick medium, radiation trapping imposes a limit on the maximum achievable polarization, because spontaneously emitted photons do not have the same polarization and wavelength as does the optical pumping light. Multiple reabsorption of these photons causes depolarization and for a high optical density completely prevents optical pumping [2]. It was realized soon after the development of optical pumping that molecular buffer gases can be used to quench excited atomic states and to prevent emission of spontaneous radiation. As a result, many applications of optical pumping, particularly for alkali-metal atoms, use N_2 buffer gas to prevent radiation trapping [3]. However, there are several situations where the presence of 5–50 torr of N_2 gas is detrimental. One example is polarization of H and D nuclear spins by spin exchange with alkali metals for use as nuclear targets in electron scattering experiments [4,5]. A large magnetic field is used in this case to reduce the effect of radiation trapping [6,7] even though it also reduces the efficiency of spin exchange, particularly for D [8]. Another example where the quenching gas is detrimental is for polarization of an electron beam using optically pumped alkali-metal atoms [9,10].

Spin-exchange collisions can be used to transfer spin polarization between different species, as was first demonstrated by Dehmelt [11]. Recently, “hybrid” optical pumping using Rb and K atoms has been explored for spin-exchange optical pumping of ^{3}He gas [12,13]. Optical pumping of Rb atoms with readily available lasers creates spin polarization in K, which is then transferred to ^{3}He with a higher overall efficiency than by direct Rb-^{3}He spin exchange [14]. Here we point out that hybrid optical pumping can be used in a wider set of circumstances when the optical density of one of the alkali-metal atoms is kept small. This allows optical pumping in the absence of N_2 gas. In addition to the examples mentioned above, we point out that the absence of N_2 improves the effective optical depth in quantum nondemolition spin measurements using Faraday rotation [15].

Small optical density is also beneficial for spin-exchange relaxation free magnetometers [16]. In this case the limiting factor is not the presence of N_2 gas but the attenuation of the pumping beam. In steady-state operation the magnetometer has optimal sensitivity when the optical pumping rate is equal to the spin relaxation rate, a condition that cannot be maintained throughout an optically thick vapor. It is possible to use a far-detuned pump laser to reduce absorption, but this causes unwanted light shifts [17]. Optical pumping of a low-density alkali metal and Faraday rotation measurements on the high-density metal will achieve optimal sensitivity throughout the sensor.

We experimentally demonstrate spin polarization of optically dense K vapor without buffer gas by spin exchange with optically pumped Rb. The spin polarization of K obtained in this way is a factor of 4.5 higher than by direct optical pumping of K. Interestingly, the Rb metal was not intentionally introduced into the cell but was present in the vapor at approximately 0.2% due to contamination of the K metal. We also develop a density matrix model for optical pumping in the presence of a high spin-exchange rate but without any buffer gas and point out particular sensitivity to the circular polarization of the pumping light in this situation.

Consider two alkali-metal species undergoing spin-exchange collisions, one much more abundant than the other. For definiteness we consider K and ^{85}Rb, with a small Rb fraction $f = n_{\mathrm{Rb}}/(n_{\mathrm{Rb}} + n_{\mathrm{K}}) \ll 1$. Rb atoms are being optically pumped with an average absorption rate R for an unpolarized atom, and both atoms are undergoing electron spin randomization collisions at a rate Γ. The thermally averaged spin-exchange rate constant $\langle \sigma_{\mathrm{ex}} v \rangle$ for different alkali-metal atoms as well as their mutual spin-exchange rates are nearly the same [18,19]. Thus, the spin-exchange rate of atom i with atom j is given by $X_{ij} = \langle \sigma_{\mathrm{ex}} v \rangle n_j$, and the total spin-exchange rate $X = \langle \sigma_{\mathrm{ex}} v \rangle (n_{\mathrm{Rb}} + n_{\mathrm{K}})$. If we make a simplifying assumption of fast electron spin randomization in the excited state, for example, due to collisions with helium buffer gas, it is particularly easy to derive a simple equation for the spin polarization of K atoms due to optical pumping of Rb [20]. For $f \ll 1$ we get

$$P_{\mathrm{K}} = 1/[1 + \Gamma(1/R + 1/X)/f]. \tag{1}$$

Thus, two conditions need to be satisfied to achieve a high K polarization: $fR \gg \Gamma$ and $fX \gg \Gamma$. The first condition ensures that the overall input rate of angular momentum from the pump beam is sufficient to polarize all atoms, and the second condition ensures that the spin-exchange rate of K with Rb exceeds its own spin relaxation rate.

A more detailed analysis is necessary in the absence of buffer gas, when there is no electron randomization in the excited state and the depopulation optical pumping is determined by spontaneous emission. For this case we calculate the equilibrium density matrix in the presence of spin exchange, ground state electron spin relaxation, and optical pumping in vacuum. We assume the laser is tuned to the peak of $F = 2 \rightarrow F' \rightarrow 3$ D_1 transition in ^{85}Rb. Figure 1 shows a contour plot of P_{K} as a function of fR/Γ and fX/Γ. The plot is made for $f = 0.01$, but the results are virtually identical for $f = 0.1$–0.001. One can see that Eq. (1), plotted with dashed lines, gives a good approximation to the full density matrix calculation.

In the absence of a buffer gas, one typically uses an antirelaxation surface coating to reduce spin relaxation of alkali-metal atoms on cell walls [21]. The spin relaxation rate is then given by $\Gamma \simeq \bar{v}/lN_e$, where $\bar{v}$ is the thermal velocity of atoms, l is the characteristic dimension of the cell, determined by the volume to surface ratio, and N_e is the number of bounces that the coating allows on average before electron spin relaxation. In order to avoid radiation trapping in optical pumping of Rb, we need $\sigma_0 n_{\mathrm{Rb}} l < 1$. The K optical density $\mathrm{OD_K} = \sigma_0 n_{\mathrm{K}} l$, where σ_0 is the peak photon absorption cross section, including the effects of Doppler broadening. Thus, to avoid radiation trapping we need $f \simeq 1/\mathrm{OD_K}$. It follows that $fX/\Gamma = N_e \sigma_{\mathrm{ex}}/\sigma_0$. The peak Doppler broadened absorption cross section in alkali-metal atoms is about $\sigma_0 = 7 \times 10^{-12}$ cm^2, while the spin-exchange cross section is $\sigma_{\mathrm{ex}} = 2 \times 10^{-14}$ cm^2. Hence we need $N_e \gg 300$ in order to achieve high spin polarization. Paraffin has long been used as an antirelaxation coating with an operating temperature up to 50 °C. It typically allows about 10 000 surface bounces before relaxation of the atomic polarization. The relaxation is predominantly due to electron spin randomization [22], so by taking into account the nuclear slowing-down factor it corresponds to $N_e \sim 1700$ for an alkali-metal atom with $I = 3/2$. More recently octadecyltrichlorosilane coating with $N_e > 300$ has been shown to operate at temperatures up to 170 °C [23]. Alkane coatings recently reported in Refs. [24,25] allow over $N_e \gg 10^5$ bounces and can operate up to 100 °C. A higher temperature results in a higher density of the more abundant alkali metal, which is advantageous for most applications.

We experimentally explore spin-exchange optical pumping in a 1.9-inch-diameter evacuated cell with K metal and an octadecyltrichlorosilane surface coating. Previously, it was found that in the absence of N_2 quenching the maximum K polarization achieved with optical pumping of K was limited to 2%–3% [23]. It has been known anecdotally that commercial alkali-metal samples are often cross-contaminated, so a small impurity of Rb could be present in the cells. We have found that Rb vapor density was about 2×10^{-3} of the K density and also observed trace amounts of Cs in the K cell. An absorption spectrum of Rb and K at 160 °C is shown in Fig. 2. It can be seen that, while K vapor is optically thick, the optical density of Rb is significantly

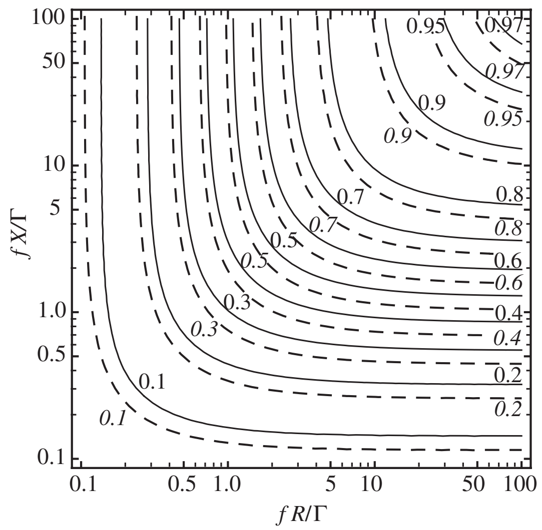

FIG. 1. Contour plot of maximum polarization of K as a function of the Rb pumping rate R, the spin-exchange rate X, and the Rb fraction f. The solid lines and upright numbers correspond to the density matrix calculations, while the dashed lines and italic numbers correspond to Eq. (1).

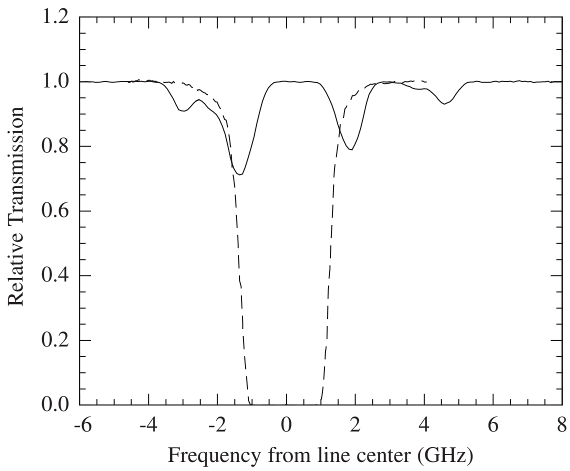

FIG. 2. Absorption spectrum of K (dashed line) and trace amounts of Rb (solid line) at 160 °C in a K cell.

less than 1, ideal for optical pumping in the absence of quenching gas.

Figure 3 shows measurements of the K spin polarization with paramagnetic optical rotation of an off-resonant linearly polarized laser detuned by 51 GHz from the K D_1 line. The K vapor is optically pumped by a copropagating circularly polarized laser tuned to the Rb or to the K D_1 line and parallel to a holding magnetic field. The maximum polarization obtained when pumping on Rb is about 4.5 times larger than when pumping directly on K. The inset shows the power dependence of the maximum rotation signal as a function of K pump laser power, demonstrating the saturation and eventual decrease of the polarization due to radiation trapping. In contrast, for Rb pumping the K polarization continues to increase with available laser power. For the data in Fig. 3, the K density is determined from the spin-exchange relaxation rate to be $n_{\rm K} = 9.2 \times 10^{12}$ cm^{-3}. The optical rotation corresponds to K polarization obtained with Rb pumping of 13.7% vs 3.0% for direct pumping on K. The time constant for the transient decay of spin polarization in the dark is $T_1 = 62$ ms, which corresponds to $N_e = 120$, taking into account the nuclear slowing-down factor for K. We find that both Eq. (1) and the density matrix model predict the K polarization with Rb pumping to be 10%–15%, in agreement with experimental measurements within uncertainties of the input parameters.

The dependence of the K spin polarization on the Rb pump laser frequency is not trivial, as shown in Fig. 4. At a low optical pumping rate, the spectrum of K spin polarization, shown with solid triangles, resembles the Rb absorption spectrum. However, if the optical pumping rate is larger than the spin-exchange rate, the polarization exhibits a more complicated profile that is very sensitive to the degree of light polarization. As can be seen in Fig. 4, the measured polarization spectrum is very different from predictions for perfectly circularly polarized light but follows closely the model assuming light polarization of 80%. This is consistent with about 20% photon reabsorption probability due to Rb optical density. The polarization sensitivity is due to different degrees of hyperfine optical pumping. For perfectly circularly polarized light, the $F = 3, M = 3$ state in ^{85}Rb is a dark state and Rb atoms can reach 100% polarization at high optical pumping intensity. However, if the light polarization is not perfectly circular, hyperfine pumping into the $F = 2$ state takes place when the optical pumping rate exceeds the spin-exchange rate. This results in the reversal of the electron spin polarization of Rb as well as K atoms near the $F = 3$ ^{85}Rb line. Similar behavior has been observed in Ref. [26].

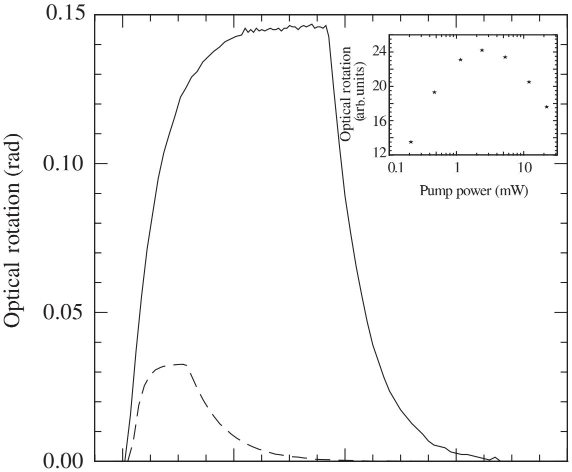

FIG. 3. Optical rotation transient in K vapor generated by optical pumping of optically dense K (dashed line) or trace amounts of Rb (solid line). The pump light is abruptly turned on and off. The inset shows the maximum polarization of K as a function of K pumping laser power.

Below, we give two examples beyond those already explored in the literature where hybrid pumping with low optical density of one species is particularly useful. In quantum-nondemolition (QND) measurements using paramagnetic Faraday rotation [15], the intensity of the probe beam is usually limited by its absorption rate. In the regime of far detuning of the probe laser, the wing of the Voight absorption profile is dominated by the Lorentzian width Γ_L, which is the sum of the natural atomic decay rate and pressure broadening linewidth. The optical rotation angle is given by $\phi = nlr_ecf_{\rm osc}(\nu - \nu_0)/[(\nu - \nu_0)^2 + \Gamma_L^2]/2$, while the absorption cross section is $\sigma = r_ecf_{\rm osc}\Gamma_L/[(\nu - \nu_0)^2 + \Gamma_L^2]$. If the noise in measurements of Faraday

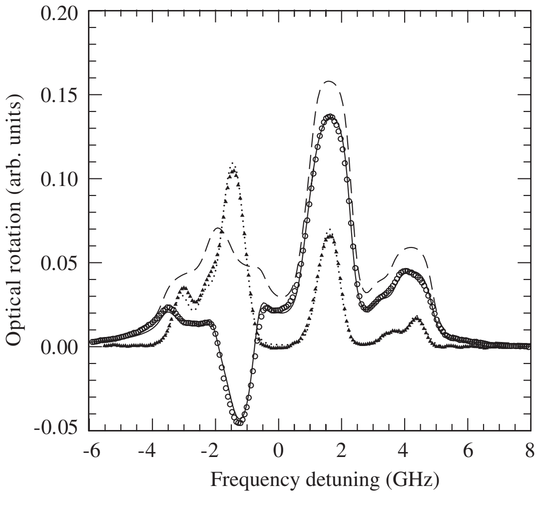

FIG. 4. Optical rotation in K vapor as a function of Rb pump light frequency. Open circles show K polarization measurements for a Rb optical pumping rate much larger than the spin-exchange rate. Triangles show the K polarization for a Rb pumping rate much smaller than the spin-exchange rate, with data scaled up by a factor of 14. Note the reversal of the polarization sign for pump light near $F = 3$ ^{85}Rb lines due to hyperfine optical pumping. The theoretical prediction for a low pumping rate is shown with dots, for a high pumping rate and perfect circular polarization with a dashed line, and for 80% circular polarization with a solid line.

rotation is dominated by photon shot noise, and spin relaxation of the atoms is limited by the probe beam photon absorption rate, one can show that the signal-to-noise ratio in one atomic spin relaxation time is equal to $\phi/\delta\phi = \sqrt{N_{at}\mathrm{OD}_L}$, where $\mathrm{OD}_L = r_e c f_{\mathrm{osc}} n l/\Gamma_L$. Hence, the atom spin can be read out with a signal-to-noise ratio that exceeds atom shot noise $\sqrt{N_a}$ by a factor of $\sqrt{\mathrm{OD}_L}$, thus allowing QND measurements that follow atomic spin evolution. The OD_L that enters here corresponds to the Lorentzian linewidth, not to the actual observed optical depth on resonance, which is dominated by the Doppler effect for hot atoms. In fact, for large probe laser detuning, atom cooling does not offer any advantages in QND measurements. On the other hand, the presence of even a few torr of N_2 gas, which is necessary for optical pumping of optically dense vapor, dramatically increases the Lorentzian linewidth while having little effect on the overall absorption profile. For example, 10 torr of N_2, which is the minimum typically required for quenching [10], increases the Lorentzian half-width in Rb by a factor of 39 from its value in vacuum (3 MHz). In the absence of N_2, the effective OD_L for QND measurements is on the order of 5×10^4 for a 5-cm-long cell with an atom density of 10^{13} cm^{-3}. Since one wins only relatively slowly with OD in quantum measurements, such large optical densities are crucial to realizing a significant increase in sensitivity from quantum entanglement.

Different considerations apply for the use of hybrid optical pumping in spin-exchange relaxation free magnetometers [16]. When the magnetometer is operated in the steady-state regime, the response to a magnetic field B is given by $S = \gamma BR/(R + \Gamma)^2$. Hence the largest signal is obtained when $R = \Gamma$, corresponding to atomic spin polarization of 50%. Since the atoms are not fully polarized, the circularly polarized pump laser is significantly absorbed. This limits the maximum density of the alkali metal so that the optical depth in the direction of the pump laser is $\mathrm{OD}_p \sim 2$. Using hybrid optical pumping on a different alkali metal with much smaller density eliminates this problem. For light alkali metals, such as K and Rb, the spin destruction cross sections are several orders of magnitude smaller than the spin-exchange cross section. Therefore, spin polarization can be maintained with minor alkali-metal fractions of $f = 10^{-2}$–10^{-3}, increasing operating alkali-metal density or the size of the cell by a large factor. We have explored this technique in a ^{21}Ne-Rb comagnetometer [27], by using a Rb-K mixture with $f_{\mathrm{K}} = 0.005$ and optical pumping on K. We obtained an order of magnitude higher polarization of ^{21}Ne than was possible by using a single alkali metal [28], opening a new potential for sensitive nuclear spin gyroscopes [29].

In conclusion, we describe a simple technique for spin polarization of optically thick alkali-metal atoms using spin exchange with an optically thin species. This approach, in combination with high quality antirelaxation surface coatings, opens the possibility of creating very optically dense spin-polarized alkali-metal vapors without any quenching gas. Such vapors can be used to transfer polarization to other species, such as atomic hydrogen or electrons. They are also useful for spin quantum nondemolition measurements. Even in the presence of buffer gas, hybrid optical pumping allows one to independently control absorption of optical pumping light, which is beneficial in ultrasensitive atomic magnetometers and other optical pumping experiments.

We thank Scott Seltzer for fabrication of the alkali-metal cells and Ivanna Dimitrova for discussions. This work was supported by the ONR MURI.

[1] W. Happer, Rev. Mod. Phys. **44**, 169 (1972).
[2] D. Tupa and L. W. Anderson, Phys. Rev. A **36**, 2142 (1987).
[3] B. Lancor and T. G. Walker, Phys. Rev. A **82**, 043417 (2010).
[4] K. P. Coulter *et al.* Phys. Rev. Lett. **68**, 174 (1992).
[5] B. Clasie *et al.*, Phys. Rev. A **73**, 020703 (2006).
[6] D. Tupa, L. W. Anderson, D. L. Huber, and J. E. Lawler, Phys. Rev. A **33**, 1045 (1986).
[7] L. W. Anderson and T. Walker, Nucl. Instrum. Methods Phys. Res., Sect. A **316**, 123 (1992).
[8] M. Poelker *et al.*, Phys. Rev. A **50**, 2450 (1994).
[9] H. Batelaan, A. S. Green, B. A. Hitt, and T. J. Gay, Phys. Rev. Lett. **82**, 4216 (1999).
[10] M. A. Rosenberry, J. P. Reyes, D. Tupa, and T. J. Gay, Phys. Rev. A **75**, 023401 (2007).
[11] H. G. Dehmelt, Phys. Rev. **109**, 381 (1958).
[12] E. Babcock *et al.*, Phys. Rev. Lett. **91**, 123003 (2003).
[13] W. C. Chen, T. R. Gentile, T. G. Walker, and E. Babcock, Phys. Rev. A **75**, 013416 (2007).
[14] A. B. Baranga *et al.*, Phys. Rev. Lett. **80**, 2801 (1998).
[15] Y. Takahashi *et al.*, Phys. Rev. A **60**, 4974 (1999).
[16] I. K. Kominis, T. W. Kornack, J. C. Allred, and M. V. Romalis, Nature (London) **422**, 596 (2003).
[17] V. Shah and M. V. Romalis, Phys. Rev. A **80**, 013416 (2009).
[18] H. M. Gibbs and R. J. Hull, Phys. Rev. **153**, 132 (1967).
[19] N. W. Ressler, R. H. Sands, and T. E. Stark, Phys. Rev. **184**, 102 (1969).
[20] S. Appelt *et al.*, Phys. Rev. A **58**, 1412 (1998).
[21] H. G. Robinson, E. S. Ensberg, and H. G. Dehmelt, Bull. Am. Phys. Soc. **3**, 9 (1958).
[22] D. Budker *et al.*, Phys. Rev. A **71**, 012903 (2005).
[23] S. J. Seltzer, M. V. Romalis, J. Appl. Phys. **106**, 114905 (2009).
[24] M. V. Balabas *et al.*, Opt. Express **18**, 5825 (2010).
[25] M. V. Balabas, T. Karaulanov, M. P. Ledbetter, and D. Budker, Phys. Rev. Lett. **105**, 070801 (2010).
[26] E. B. Norrgard, D. Tupa, J. M. Dreiling, and T. J. Gay, Phys. Rev. A **82**, 033408 (2010).
[27] R. K. Ghosh and M. V. Romalis, Phys. Rev. A **81**, 043415 (2010).
[28] L. W. Cheuk, Senior thesis, Princeton University, 2010.
[29] T. W. Kornack, R. K. Ghosh, and M. V. Romalis, Phys. Rev. Lett. **95**, 230801 (2005).