Millihertz magnetic resonance spectroscopy of Cs atoms in body-centered-cubic ^{4}He

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We report the results of electron-spin-resonance studies of Cs atoms trapped in the body-centered-cubic phase of a solid ^{4}He matrix. Free-induction-decay times longer than 100 ms have been measured in spin-transient experiments. In optical-rf double-resonance experiments we have obtained linewidths of less than 20 Hz (full width at half maximum). The magnetometric sensitivity of our sample is 26 nG/$\sqrt{\text{Hz}}$. We have also demonstrated that magnetic resonance in such samples can be detected with high sensitivity using forward-scattered optical radiation. [S1050-2947(96)50108-3]

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The optical spectroscopy of foreign atoms in condensed ^{4}He is a new branch of matrix isolation spectroscopy that has rapidly developed in the past decade. Early investigations were focused on the study of optical properties of atoms in superfluid ^{4}He (He II). For the past few years the further development of laser sputtering techniques used to implant atoms into He II [1–3] has allowed us to extend these studies to solid ^{4}He matrices.

The main interest of our group has been the application of matrix isolated paramagnetic species to high-resolution spin physics experiments [4]. In particular we investigate whether such samples can be used to search for permanent electric dipole moments whose existence is forbidden by the conservation of the discrete symmetries P (parity) and T (time reversal) [5]. The high degree of spherical symmetry of the local trapping site of foreign atoms in condensed ^{4}He allows the efficient creation of spin polarization of implanted atoms by the technique of optical pumping. Optical pumping has been demonstrated so far on Rb and Cs atoms in He II [6] and on Cs atoms in body-centered-cubic (bcc) and in hexagonal-close-packed (hcp) solid ^{4}He [4,7,8]. In the bcc phase of the He matrix we observed longitudinal electronic spin relaxation times T_1 on the order of 1–2 s [7]. These relaxation times were essentially independent of the strength of the holding magnetic field B_z in the region $B_z > 10$ mG. This was a first indication that the transverse spin relaxation time T_2 for such samples could reach the same order of magnitude, and therefore extremely narrow electron-spin-resonance (ESR) lines should be observable. In the experiments reported here we have imposed a lower bound of 0.1 s on the intrinsic T_2 time for Cs atoms in a bcc ^{4}He matrix. ESR linewidths as narrow as 15 Hz were measured, the widths being mainly determined by optical power broadening. The center frequency of the resonance can be determined with a precision of better than 10 mHz/$\sqrt{\text{Hz}}$.

So far all studies of impurity atoms in condensed ^{4}He matrices have been performed by detecting laser-induced fluorescence (LIF). The observation of optical signals in absorption is hindered by the small optical thickness of the samples (typically 10^{-4}–10^{-5}) and by a large optical linewidth of several nanometers. Since the LIF detection method is not a universal one, for example, it cannot be used for light alkali-metal atoms, where a radiationless quenching of the excited states is believed to occur [9,10], the development of a technique allowing the observation of atomic resonances in the transmitted light is of great interest. In this paper we also report an observation of resonance signals from He-matrix isolated atoms in a transmission experiment and compare the results to those from fluorescence experiments.

Cs atoms were implanted in a ^{4}He crystal grown in a pressurized cell at 1.6 K using the laser ablation technique described in [11]. The main loss mechanism for the implanted atoms is the formation of dimers and clusters. These dimers and clusters were destroyed by sending a focused beam of frequency-doubled Nd:YAG (neodymium-doped yttrium aluminum garnet) laser radiation (10 ns, 10 mJ, 1 Hz repetition rate) through the sample volume, thus keeping the average atomic concentration constant during the measurements.

Several changes were implemented in the cryogenic part of our apparatus as compared to previous experiments [7]. The size of the pressure cell is significantly increased, its interior has a cubic shape with a side length of 5.5 cm. Laboratory magnetic fields are suppressed to the level of several mG with a single μ-metal (CRYOPERM) shield placed in the He bath. The residual fields are further compensated to the μG level by a set of three superconducting Helmholtz coils. Two orthogonal pairs of rectangular rf coils with low inductivity are located inside of the cell. Besides conventional ESR studies these coils are also used to rapidly apply a transverse dc-magnetic field in the free-induction-decay (FID) experiments.

The sample was polarized by the optical pumping of Cs atoms on the $6S_{1/2} \rightarrow 6P_{1/2}$ D_1 resonance line with circularly polarized 851-nm radiation from a free-running diode laser. Typically 0.5 mW of unfocused laser power were used. Fluorescence at 886 nm was separated from stray laser light by an interference bandpass filter and detected at right angle with a cooled photomultiplier with a 4×10-mm^2 GaAs photocathode.

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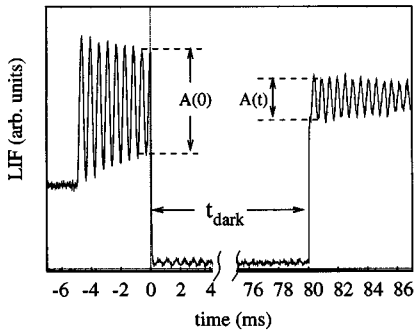

FIG. 1. FID signal obtained for a dark period of 80 ms (see the text).

In a first series of experiments we determined the transverse spin relaxation time T_2 using the free-induction-decay technique. The sample was initially polarized in a weak magnetic field of 1 mG along the direction of the laser beam $\hat{\mathbf{k}}$. The nonadiabatic switching on of a $\sim$ 10-mG transverse magnetic field causes the sample polarization $\boldsymbol{P}$ to precess, and a corresponding modulation of the LIF signal S_{LIF} appears at the Larmor frequency ω_L: $S_{LIF} \propto 1-\xi \boldsymbol{P}\cdot\hat{\boldsymbol{k}} = 1-\xi P\cos\omega_L t$. Here ξ is a constant that depends on parameters of the atomic transition and on the degree of light polarization. The amplitude of the modulated component of the signal (FID signal) is thus a measure of the sample polarization, and its time dependence gives information about the decay time T_2 of the ground-state spin coherences. Figure 1 shows an example of such a FID signal. The transverse magnetic field was applied at time $t=-5$ ms and the FID signal was recorded up to $t=0$. In order to avoid sample depolarization by the probing light the latter was switched off at $t=0$, thus allowing the transverse polarization to precess and decay in the dark for a period t_{dark}. The amplitude of the FID signal $A(t)$ immediately after switching the probing light back on is then a measure of the sample polarization that has survived the dark period. In Fig. 2 these amplitudes normalized to the initial amplitude $A(0)$ are plotted as a function of the duration of the dark period. By fitting a single exponential to these data we infer a value for the transverse relaxation time of our sample of $T_2=108(3)$ ms. As we cannot exclude the fact that this value is affected by residual B-field inhomogeneities (the quoted T_2 is equivalent to a field inhomogeneity δB of 4.2 μG) the quoted value is to be

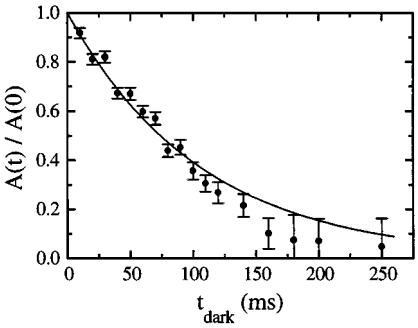

FIG. 2. FID amplitude as a function of the duration of the dark period. The solid line is a fitted single exponential.

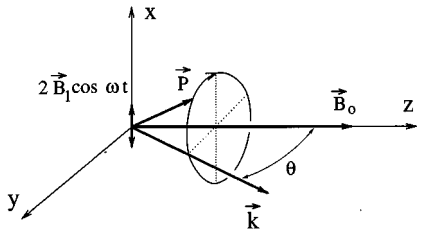

FIG. 3. B-field geometry in the driven precession experiments.

understood as a lower bound for the *intrinsic* transverse spin relaxation time for paramagnetic atoms in the bcc phase of a ^{4}He matrix.

Due to the long transverse spin relaxation time T_2 observed in the FID experiments, extremely narrow magnetic resonance lines can be obtained with this type of sample. As we have mentioned earlier, one of our motivations to study paramagnetic atoms isolated in ^{4}He matrices was the hope that such a sample could be used in the experiments searching for permanent atomic electric dipole moments (EDM's) [3]. An indication for the existence of such EDM's would be, for example, the observation of a linear Stark effect in the atomic ground state or, equivalently, a precession of the atomic spin polarization induced by a static electric field. A prerequisite for such experiments is a high magnetometric sensitivity [4]. We have measured our current magnetometric sensitivity in the following experiment using the technique of driven spin precession under transverse optical pumping.

The sample was placed in a static magnetic field $\boldsymbol{B}_0$ oriented at an angle θ with respect to the wave vector $\boldsymbol{k}$ of the circularly polarized laser beam (Fig. 3). At the same time a linearly polarized oscillating magnetic field $\boldsymbol{B}_1$ was applied to the sample. We shall briefly explain the origin of the observed signals in the limit of a weak rf field ($B_1 \ll B_0$). An analysis of arbitrary rf field strengths can be found in [12].

The evolution of the sample polarization $\boldsymbol{P}$ is described by the system of phenomenological Bloch equations:

$$\boldsymbol{P}' + \boldsymbol{\Omega}\times\boldsymbol{P} + \frac{\boldsymbol{P}}{\tau^*} = \frac{\boldsymbol{P}_0}{\tau^*}. \tag{1}$$

where

$$\boldsymbol{\Omega} = \left(\frac{2g\mu_B}{\hbar}B_1\cos\omega t,\ 0, \frac{g\mu_B}{\hbar}B_0\right) \equiv (2\omega_1\cos\omega t,\ 0, \omega_0),$$
$$\tau^* \equiv \{\tau_2, \tau_2, \tau_1\}$$

and the optical pumping is taken into account as an additional relaxation mechanism towards an equilibrium polarization $\boldsymbol{P}_0=(0, P_0\sin\theta, P_0\cos\theta)$ at a rate Γ_{pump}. The effective spin relaxation times are then given by $1/\tau_{1,2} = 1/T_{1,2} + \Gamma_{pump}$.

We introduce the notation $F = P_x + iP_y$ and search for the steady-state solution of Eq. (1) in the form of Fourier series $F = \Sigma f_n \exp(in\omega t)$ and $P_z = \Sigma p_{n'} \exp(in'\omega t)$. By retaining in the resulting system only resonant terms (p_0, f_{-1}), which is equivalent to the rotating-wave approximation, we obtain

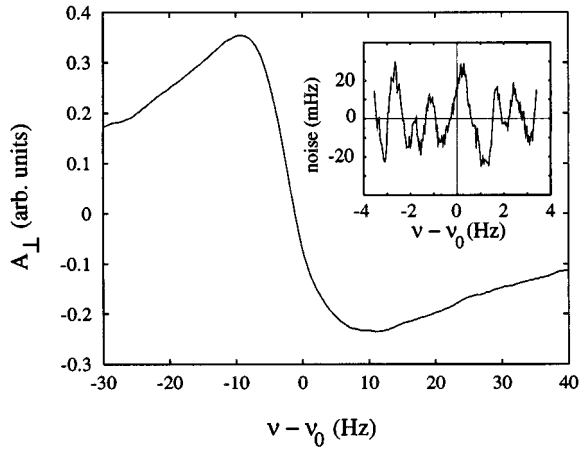

FIG. 4. Magnetic resonance spectrum recorded with LIF detection by scanning the driving radio frequency ν over the resonance at $\nu_0 \approx 40$ kHz (lock-in time constant 30 ms). The digital integration time was 1 s per sampling point. The inset shows the residuals of a fit with a fifth-order polynomial as discussed in the text.

$$p_0 = P_0 \cos\theta \frac{1+(\Delta\omega\tau_2)^2}{1+(\Delta\omega\tau_2)^2+\omega_1^2\tau_1\tau_2},$$

$$f_{-1} = p_0 \frac{i\omega_1\tau_2}{1+i\Delta\omega\tau_2},$$

where $\Delta\omega = \omega_0 - \omega$. This solution describes a precession of the sample polarization at the frequency ω around the constant magnetic field $\boldsymbol{B_0}$ with a phase determined by the detuning from the resonance $\Delta\omega$. For the absorption of the polarized sample A $\propto 1 - \xi \boldsymbol{P}\cdot\hat{\boldsymbol{k}}$ one gets

$$A \propto 1 - \xi p_0 \cos\theta - \xi |f_{-1}| \sin\theta \cos(\omega t + \phi),$$

$$\cos\phi = \frac{\Delta\omega\tau_2}{\sqrt{1+(\Delta\omega\tau_2)^2}}. \tag{2}$$

The last term in Eq. (2) oscillates at the driving radio frequency ω and is proportional to the amplitude $P_\perp$ of the transverse component of the polarization. The in-phase component $A_\perp$ of this signal has a dispersionlike dependence on the detuning from resonance:

$$A_\perp \propto \sin 2\theta \frac{\omega_1 \Delta\omega \tau_2^2}{1+\omega_1^2\tau_1\tau_2+(\Delta\omega\tau_2)^2}.$$

This $A_\perp$ signal was extracted from the total fluorescence signal with a phase-sensitive amplifier locked to the driving radio frequency ω. A very important advantage of this technique is the possibility of its performing a phase-sensitive detection at a high frequency, where the apparatus noise is significantly suppressed. Figure 4 shows the resulting magnetic resonance spectrum recorded in a digital storage oscilloscope by scanning the radio frequency over the resonance at $\omega_0/2\pi \approx 40$ kHz. In order to determine the magnetometric sensitivity δB of our sample we fit the central part of the

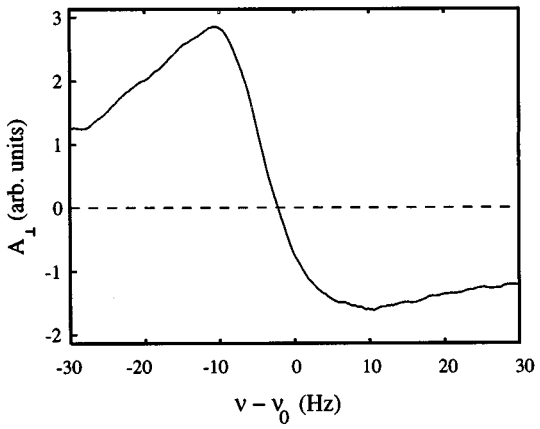

FIG. 5. Spectrum of magnetic resonance obtained by monitoring the transmitted light intensity.

resonance ($|\Delta\nu| \leq 4$ Hz, $\nu \equiv \omega/2\pi$) with a fifth-order polynomial $\Sigma_0^5 a_n (\Delta\nu - \Delta\nu_0)^n$. The coefficient a_1 gives the slope of the signal $\partial A_\perp / \partial \nu$ near the center of the resonance, and the sensitivity is found by dividing the rms value $\langle \delta A_\perp \rangle_{rms}$ of the residuals from the fit by this slope:

$$\delta B = \frac{\langle \delta A_\perp \rangle_{rms}}{a_1} = 26 \text{ nG},$$

which corresponds to $\delta\nu = 9.1$ mHz with an integration time of 1 s. A closer look at the residuals of the fit shown in the inset to Fig. 4 reveals an oscillatory structure, which we attribute to slow variations of system parameters, such as magnetic fields, atomic concentration, and light intensity inside the cell. The pure statistical noise is at least one order of magnitude smaller than the quoted value.

Being encouraged by the high signal-to-noise ratio of the magnetic resonance observed with the driven spin precession magnetometric technique and the LIF detection, we have made an attempt to detect the resonance in a transmission experiment. The experiment was performed in the same way as discussed above but this time the output of a photodiode measuring the transmitted light intensity was analyzed by the lock-in amplifier. The resulting magnetic resonance spectrum is shown in Fig. 5. The observed signal-to-noise ratio is quite comparable to that obtained with the LIF detection technique. From the absolute value of the $A_\perp$ signal we estimate the total absorption in our sample to be $\approx 10^{-4}$ cm^{-1}. This is an observation of an optical signal from He-matrix isolated atoms obtained by monitoring transmitted (forward-scattered) radiation.

We have demonstrated that paramagnetic atoms implanted in the isotropic bcc phase of a ^{4}He matrix yield transverse spin relaxation times exceeding 100 ms. Extremely narrow magnetic resonance lines can be obtained with these atoms. The magnetometric sensitivity of our sample was measured to be 26 nG with a 1-s integration time. A driven precession technique allows the efficient detection of the magnetic resonance with the monitoring of the forward scattered radiation.

The high resolution of the ESR spectroscopy demon-

strated in this work should allow among others a precise measurement of the electronic g factors of the implanted atoms. Such investigations are currently underway and should bring more detailed information about the symmetry of the local trapping sites for the atoms implanted in the helium crystal.

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