Slow Spin Relaxation of Rb Atoms Confined in Glass Cells Filled with Dense ^{4}He Gas at 1.85 K

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At temperatures below 2.1 K, long-lived gaseous Rb atoms in glass cells have been generated with a simple method: irradiating the cells, containing ^{4}He gas and Rb metal, with a cw laser. The obtained atomic Rb density ($\sim 10^8$ cm^{-3}) decreases with a $1/e$ time constant of about 10 s at 1.85 K. We have performed optical pumping of the Rb atoms and measured the longitudinal electronic spin relaxation time at 1.85 K as well. For processes (such as Rb-He collisions) which do not remove the atomic Rb from the vapor, this relaxation time is found to be about 60 ± 15 s.

We introduce, in this Letter, a new optical pumping environment for atomic Rb vapors: glass cells held at temperatures below 2.1 K, containing dense ^{4}He buffer gas in saturated equilibrium with ^{4}He films coating the walls. One of the principle motivations for this study is the efficacy of cryogenic He environments for the preservation of the spin polarization of embedded sample atoms or molecules. For example, large Rb and Cs polarizations have been produced in liquid He by optical pumping [1], very long relaxation times have been measured for Cs embedded in solid He [2], and high spin species such as Na quartet trimers have been studied using He cluster isolation spectroscopy [3]. For Rb atoms in He buffer gas, a strong temperature dependence has been observed for the spin relaxation rate [4], consistent with calculations of the Rb-He spin destruction mediated by the spin-rotation interaction [5]. The observed temperature dependence suggests that extraordinarily low collisional spin relaxation rates may pertain for cells at temperatures below 2.1 K.

Rb atomic vapors ($\sim 10^8$ cm^{-3}) were produced by directing a cw Ti:sapphire laser beam (~ 200 mW) into the cell. The obtained atomic Rb persisted for some time, decreasing with a time constant of about 10 s at 1.85 K. We optically polarized these Rb atoms and found that the longitudinal relaxation time of the electronic spin was much longer than the achieved relaxation times (~ 1 s) for alkali atoms in conventional cells at room temperature and above [6]. This cryogenic vapor cell could be a promising technique, in addition to the liquid or solid He matrix methods and the laser cooling and trapping [7], for applying cold atomic samples to spin physics studies.

The experimental setup is shown in Fig. 1(a). Rb vapor was produced and optically probed in a rectangular Pyrex cell ($2 \times 2 \times 3$ cm). About 10 mg of Rb metal of natural isotopic abundance was put into the small reservoir attached to the cell [see the schematic figure inside the circle in Fig. 1(a)]. A new method we developed for producing atomic Rb in the cell described below does not need Rb metal on the walls (such as Rb droplets) to be irradiated

with lasers. The cell was filled at liquid N$_2$ temperature (77 K) with ^{4}He gas at 730 torr and then sealed. It was mounted in a pumped liquid He cryostat with optical glass windows. The cell was submerged in liquid He to keep its temperature uniform and constant with various laser beams directed through the cell. We inferred the density of He gas in the cell from the cell temperature, as monitored by a calibrated sensor placed near the cell. The He density is almost constant (9×10^{19} cm^{-3}) above its condensation temperature T_c (~ 1.9 K) in this cell. Below T_c, a superfluid He film forms in equilibrium with its saturated vapor pressure [11]. The He density therefore decreases as the temperature falls below T_c (e.g., 2×10^{19} cm^{-3} at 1.5 K),
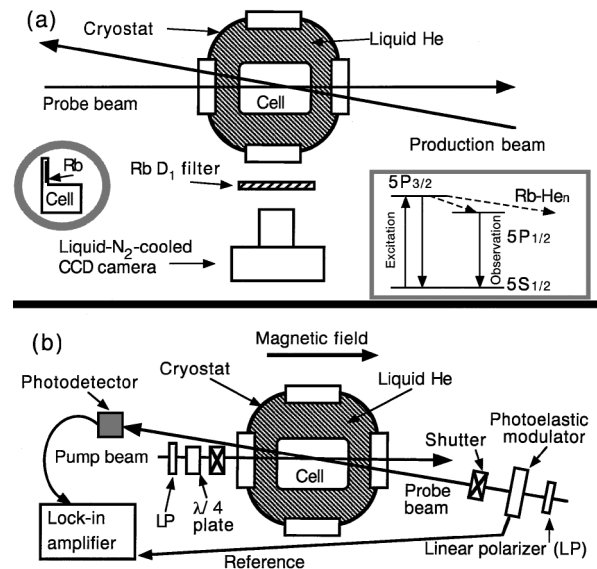

FIG. 1. Experimental setups (a) for monitoring Rb atoms by D_1 fluorescence imaging, and (b) for measuring the spin relaxation of optically polarized Rb atoms. A schematic figure of the cell is depicted inside circle in (a). The energy-level diagram of Rb related to D_1 fluorescence imaging is shown in the inset in (a). The production beam is not displayed in (b).

following the saturated vapor pressure of He [12]. The experiments were performed at temperatures of 1.4 to 2.1 K.

Rb vapor was produced by directing about 200 mW of Ti:sapphire laser radiation at 750 nm through the transparent walls of the cell, with the beam expanded to uniformly illuminate the cell volume. This new technique is particularly well suited to the spin relaxation measurements we present, in contrast with laser ablation techniques, which have been used to implant sample atoms into cold He gas as well [13,14]. Our attempts to ablatively load gas cells indicate that a large number of Rb clusters are formed and the produced Rb atoms are lost quickly ($\lesssim 1$ s).

The atomic Rb vapor present in the cell was monitored using an approximately 2 mW, 780 nm probe beam from a diode laser also expanded to uniformly illuminate the cell volume, which was used to excite a small fraction of the Rb atoms to the $5P_{3/2}$ state. The production of Rb atoms by the probe beam itself was negligible because of its low power. To suppress large backgrounds due to the scattered radiation from the probe beam, we actually observed fluorescence from the $5P_{1/2}$ state (the D_1 line at 795 nm) populated through Rb-He collisional transfer from the $5P_{3/2}$ state (see the inset in Fig. 1). The D_1 fluorescence from the cell was imaged by a liquid-N_2-cooled CCD camera through two bandpass filters [2 nm full width at half maximum (FWHM), centered on 795 nm]. These images were used to make rough estimations of the Rb density present in the cell by scaling the intensity of the D_1 fluorescence to that measured at room temperature. The Rb densities at room temperature in our cells were not always as large as the saturated vapor density of Rb metal [15], probably because the amounts of pure Rb metal were small [6,16]. The absolute density of Rb atoms at room temperature was inferred by the optical absorption measurements for the polarized Rb vapor using the polarization-modulated probe light (the details of these measurements are described in the optical pumping part of this Letter) [17], and it was typically 50% of the saturated vapor density. Scaling to lower temperatures requires corrections for the D_2 absorption linewidth and the branching ratio for D_1 fluorescence due to the collisional transfer, which are varied with temperature. The D_2 linewidth (FWHM) measured by monitoring the D_1 fluorescence intensity while scanning the probe laser wavelength was, for example, 60 GHz at 293 K (consistent with Ref. [18]), 12 GHz at 1.85 K, and 5 GHz at 1.5 K (about 10% accuracy), with the hyperfine structure unresolved. The branching ratios were measured by observing the D_1 and D_2 fluorescence simultaneously with a spectrometer coupled to a CCD detector [19]. The resultant density estimates have an uncertainty of about 30% altogether.

Figure 2 shows the time evolution of the density of atomic Rb averaged over the cell volume when the production beam irradiated the cell for 10 s. On the depicted density traces, the production beam is turned on at -10 s, and immediately begins driving Rb atomic vapor from the walls into the buffer gas with macroscopic flows (typically a few cm/s), which were observed by the CCD camera imaging. These flows exhibited a circulation of the Rb vapor reminiscent of convective transport. We observed a gradual increase (roughly a factor of 10) in the obtained density of Rb vapor by changing the wavelength of the production laser from 1000 nm down to 700 nm. The temperature dependence of the production was curious. As the temperature decreased, the production efficiency (Rb atoms produced/laser power) dramatically rose at about 1.9 K, roughly T_c for He gas in the cell, and below 1.9 K it dropped gradually. The dramatic change near T_c was also observed in a cell with a lower He density ($T_c \simeq 1.7$ K). The production efficiency also decreased with the repetitions of the production beam irradiation. The first irradiation after the cell was cooled from room temperature was most effective. We found that the production efficiency was recovered by increasing the cell temperature to roughly room temperature and then cooling again. Each curve in Fig. 2 was recorded when the cell was first irradiated by the production beam after the cell was cooled from 200 K. We consider that this production technique may contain interesting physics, and its mechanism will be presented elsewhere.

After the production beam is turned off at 0 s in Fig. 2, the density of the Rb atoms decreases. The atomic Rb loss rate was smaller at higher temperatures, with the highest achieved densities and longest lifetimes for the atomic Rb suitable for spin relaxation measurements being achieved at 1.8–1.9 K, where the Rb atoms decreased almost exponentially with a time constant of 10 s for the first 15 s, although the loss rate became smaller gradually (see also the inset in Fig. 2). The temperature dependence of the loss of the atomic Rb may be attributed to its movement to the walls due to diffusion (and due to the remaining flows caused by the production laser irradiation), which may be followed by the loss on the surface, because the diffusion should be faster with decreasing temperature due to the rapid decrease in the He vapor density below T_c
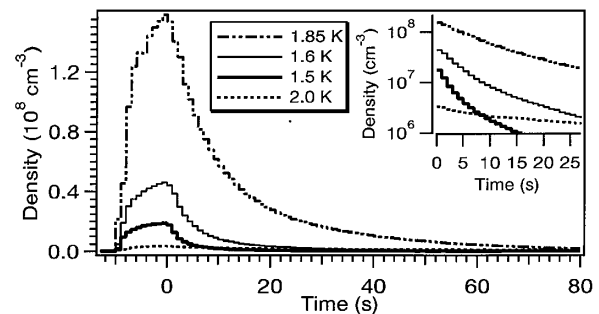

FIG. 2. The time evolution of the density of Rb atoms measured by CCD camera imaging when the production beam irradiates the cell between -10 and 0 s. The image is taken every 1 s. Inset: the initial density decay after the production is terminated (note logarithmic scale).

(and the observed flows were faster at lower temperatures). We calculated the diffusion coefficient D of Rb atoms in cold He gas in the same way as calculations have been performed for atomic hydrogen [21] using Patil's Rb-He potential [22], obtaining $D = 3 \times 10^{-3}$ cm^2/s at 1.85 K. This small diffusion coefficient suggests, in the limit of the small volume loss rates and slow gas flows in the cell, that atomic Rb loaded into the cryogenic cell could be expected to reside in the buffer gas for about 30 s even in the case of the rapid surface loss, estimated by solving a diffusion problem for $D = 3 \times 10^{-3}$ cm^2/s with a zero density boundary condition at the cell walls. Quite recently we have confirmed that larger cells and higher He pressure extend the lifetime of the produced Rb atoms, and, for one cell, have achieved a $1/e$ lifetime of 70 s at 2.1 K. The physical mechanisms by which atomic Rb is removed from the vapor phase in this cell could be complicated due to the gas flows and various loss processes in the gas and on the surface, and will hopefully be elucidated in subsequent studies.

We polarized Rb atoms in the cell at 1.85 K by optical pumping and observed the longitudinal electronic spin relaxation with the apparatus shown in Fig. 1(b). A circularly polarized pump beam (~ 5 mW) from a diode laser tuned to the D_1 line was applied to the cell along an about 3 G static magnetic field produced by a set of Helmholtz coils. The pump beam was expanded to polarize all the atoms in the cell. In order to observe the polarization of the Rb atoms, a probe beam (~ 10 μW) from another diode laser tuned to the D_1 line was applied to the cell almost parallel to the pump beam. Since the absorption of the probe beam was very small due to the low Rb density and the broad absorption line, we modulated the polarization of the probe beam with a photoelastic modulator (PEM-90 II/FS42, Hinds Instruments) to provide high sensitivity to the Rb polarization. The probe beam oscillated between σ^+ and σ^- polarized light, with elliptically polarized light between these extremes, after it passed through the PEM driven at a frequency of 42 kHz. The probe beam was also expanded and directed through the cell to a photodetector. The output of the photodetector was connected to a lock-in amplifier referenced to the PEM drive frequency, which amplified the difference in the absorption of the σ^+ and σ^- polarized probe beams and enabled us to detect the small polarization signal. For the weakly absorbed D_1 probe beam characteristic of our measurements, the polarization signal from the lock-in amplifier is proportional to the product of the mean electronic spin $\langle S_z \rangle$ of the Rb atoms and the Rb density. This technique exploits the fact that the photon absorption cross section σ for a Rb atom in a high density buffer gas is given by $\sigma = \sigma_0(1 - 2s_z\langle S_z \rangle)$, where σ_0 is the absorption cross section for an unpolarized Rb atom and s_z is the mean photon spin of the light propagating along the z direction [16,23].

For the probe power required to achieve appreciable polarization signals, the spin relaxation of the Rb atoms is completely dominated by the absorption of photons from the probe beam. Therefore, we used the "relaxation in the dark" method [24]. The inset in Fig. 3 shows the evolution of the polarization signal observed with this method, for the cell at 1.85 and 293 K. The pump beam is turned on at -0.5 s and polarizes the Rb atoms, with the polarization detected with the probe beam. The pump and probe beams are shut off at 0 s by mechanical shutters. At 1.85 K, the production beam is also turned off at 0 s after ten-second irradiation. Then only the probe beam is turned on at 5 s after a five-second dark period to measure the polarization decay in the dark. At room temperature, the polarization signal does not appear, since the dark period is much longer than the spin relaxation time, which was about 100 ms, while at 1.85 K the polarization remains after the dark period and the spin destruction by the probe beam is observed between 5 and 6 s. Then the pump beam is turned on again at 6 s and the polarization signal recovers, from which we measure the decrease of the Rb density. No decrease should be observed at room temperature. Note that the production of Rb atoms by the pump beam was negligible because of its low power and short irradiation time.

We measured the polarization decay and the decrease of the atomic Rb for five different dark periods up to 14 s according to the procedure mentioned above. In Fig. 3 we plot the means of these measured values with standard errors of the means, normalized to the measured values at 0 s. The loss rate of the atomic Rb plotted in Fig. 3 is consistent with that shown in Fig. 2. As seen in Fig. 3, the decay of the polarization signal, characterized qualitatively by a time constant of about 8.5 s if one tries a single exponential fitting, is dominated by the decrease of the Rb density (~ 10 s time constant), although the former is
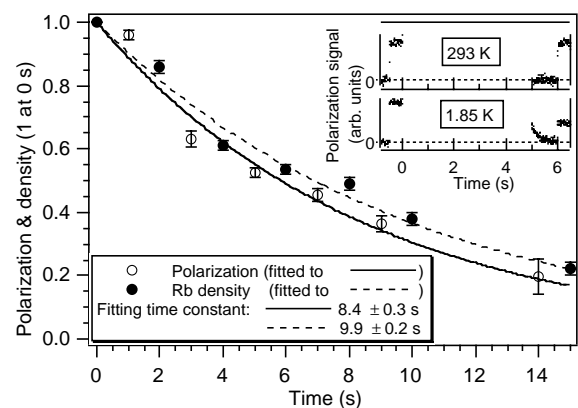

FIG. 3. The polarization and density decay data measured for various dark periods, normalized to the measured values at 0 s. The fitted single exponential curves to these data are also depicted. Inset: the temporal changes of the polarization signals from the lock-in amplifier at room temperature and at 1.85 K for the same dark period (5 s). The scales are the same for both temperatures. The measurement sequence is described in the text.

slightly faster than the latter. This indicates that the spin relaxation due to processes not causing the atomic Rb loss, such as Rb-He collisions, is much slower than the loss of the Rb atoms.

The small relaxation rate due to Rb-He collisions is consistent with calculations based on the spin-rotation interaction presented by Walker et al. [5], which yield longitudinal electronic depolarization times of about 10^5 s in cold He buffer gas [25]. In the limit of long atomic lifetimes, the spin relaxation time might be limited by interactions with cell walls, Rb clusters, or some other, as yet unidentified mechanisms, rather than Rb-He collisions. In our measurements this limiting relaxation time was about 60 ± 15 s, estimated by dividing the fitted exponential function to the polarization decay data by that to the density decay data in Fig. 3 [26]. A detailed understanding of the physical mechanisms underlying the observed Rb spin relaxation will require a systematic investigation of the limiting relaxation times as a function of the cell size, He gas density, and Rb density.

We have presented a simple technique to produce long-lived gaseous Rb atoms in cryogenic cells filled with He gas and demonstrated extraordinarily long spin relaxation times for these Rb atoms. This slow longitudinal relaxation suggests that transverse relaxation times might be made extremely long, opening the door to high precision spin physics studies such as the measurement of electronic permanent electric dipole moments in heavy alkali atoms [27].

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