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Measurement of a Long Electronic Spin Relaxation Time of Cesium Atoms in Superfluid Helium

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The longitudinal electronic spin relaxation time of Cs atoms optically polarized in superfluid helium (He II, 1.5 K) has been measured with special care to cope with a serious decrease in the number of Cs atoms in the observation region. This decrease, mainly caused by helium convection in introducing the atoms into He II by laser sputtering, was significantly reduced using a new atom implantation method. Combined with a careful correction for the number of atoms, we have determined the relaxation time to be 2.24(19) s or longer, roughly twice as long as that in solid He.

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The study of spin-polarized atoms in condensed helium matrices is a new, fascinating subject in the long history of investigations of foreign particles isolated in the quantum matrices [1–4]. Long longitudinal and transverse spin relaxation times and hence narrow magnetic resonance lines are expected in such matrices because of the low temperature, small polarizability, and spinless nature of the surrounding He atoms. Studies motivated by this expectation are, for example, the search for T -violating permanent electric dipole moments [5,6], and the investigation of the properties of superfluid and quantum solid [7]. Our practical interest in such spin properties is the study of the electromagnetic moments of short-lived nuclei using liquid He as a host matrix through magnetic resonance spectroscopy by optical detection [8] or by β -decay asymmetry measurement [9].

The pioneering work for studying the spin properties is the achievement of optical pumping for Rb and Cs atoms in He II [5] using the laser sputtering technique to introduce the atoms into He II [10]. The study was further developed in the solid He matrix of the body-centered-cubic (bcc) or the hexagonal close-packed (hcp) phase [11]. The long longitudinal and transverse electronic spin relaxation times and the narrow magnetic resonance lines were confirmed experimentally [12,13]. Furthermore, the microscopic structure of He in the solid phase was successfully investigated by magnetic resonance spectroscopy [7]. Principally in the same manner as in the solid phase case, liquid He, which has rich properties such as superfluidity and quantized vortices, can also be studied microscopically through the spin-polarized atoms immersed in He II. The study can be extended to the helium nanodroplet system, whose unique properties are currently of great interest [14]. Moreover, for our primary interest in applications to nuclear physics studies [8], a deep understanding of the spin properties of impurity atoms in liquid He is necessary. However, since the work of Ref. [5], the study of spin-polarized atoms in liquid He has not progressed so much as in solid He. The imperfect polarization in Ref. [5] (50% or more) suggests that the spin relaxation time of Cs atoms in

He II may be much shorter than that in solid He. Therefore, the relaxation time in He II should be measured directly, but has not been successful so far. This is presumably because of the difficulty in the experiment, specifically the rapid disappearance of Cs atoms under observation after their implantation by laser sputtering. In this well-established sputtering method, a sample material was irradiated by a laser beam in He II. Although the high densities of atoms (10^8 – 10^{10} cm⁻³) were achieved, the introduced atoms disappeared very quickly from the optical detection region within a few tens of milliseconds mostly because of the convection flow [10].

In this Letter we report the first successful measurement of a long longitudinal spin relaxation time (≥ 2 s) of Cs atoms in He II by observing laser induced fluorescence (LIF) from the atoms relaxed in the dark [relaxation in the dark method [15]]. This success is essentially due to (i) significant reduction of the convection flow in introducing the Cs atoms into superfluid helium, and (ii) a precise correction for the number of Cs atoms still decreasing in the observation region. The measured relaxation time is almost twice as long as that in the bcc phase of solid He and by orders of magnitude longer than that in the hcp phase. This result suggests the difference in microscopic configurations of He atoms around the impurity atoms.

In our new method to introduce Cs atoms into He II, Cs clusters were implanted into He II from the laser sputtered Cs sample placed *above* the liquid helium surface. The Cs clusters then diffused to the observation region and were dissociated with a femtosecond pulsed laser. This method was very effective in introducing as many Cs atoms as the previous laser sputtering method *in* He II without a serious convection flow. It should be pointed out that we succeeded in introducing a substantial number of Cs atoms without any electric field applied due to the effective dissociation by the femtosecond pulsed laser compared to the similar method in which sputtered ions were introduced into He II from outside with an applied electric field [6]. Figure 1 shows the experimental setup. In a helium cryostat at a temperature of approximately 1.5 K, an open-topped

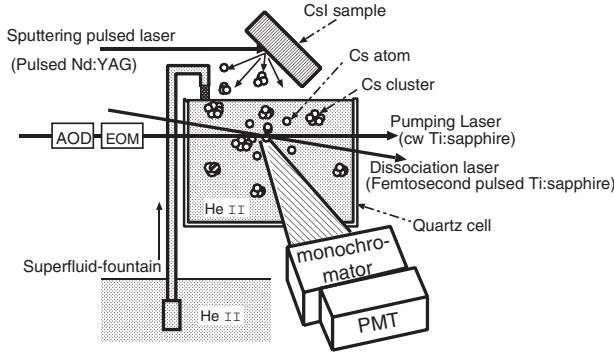


FIG. 1. Schematic of the experimental setup in a helium cryostat (helium temperature: ~ 1.5 K). A magnetic field of ~ 5 G is applied in the direction of the pumping laser beam.

quartz cubic cell ($7 \times 7 \times 7$ cm³) was filled with superfluid helium pumped up from a pool of He II through a thin tube where the fountain effect worked. A Cs sample (CsI crystal), placed approximately 1 cm above the helium surface, was sputtered by a pulsed Nd:YAG laser (wavelength: 355 nm, pulse energy: 3 mJ, repetition rate: 10 Hz, pulse width: 10 ns). The sputtered materials, most of which were clusters [10], fell into the liquid and diffused to the observation region about 1 cm below the liquid surface. The implanted clusters were dissociated with a femtosecond pulsed Ti:sapphire laser (wavelength: 790 nm, pulse energy: $150 \mu\text{J}$, repetition rate: 500 Hz, pulse width: 200 fs) focused to the observation region. The introduced Cs atoms in the observation region were irradiated with a cw Ti:sapphire laser tuned to the Cs D_1 excitation line [876 nm in He II [16]] to pump the Cs atoms. A magnetic field of ~ 5 G was applied in the direction of the pumping laser. LIF photons from the Cs atoms were focused through the glass windows of the cryostat on a monochromator which transmitted the D_1 emission line [892.5 nm in He II [16]] to a cooled photomultiplier tube (PMT).

The LIF intensity in the present case is proportional to $N_{\text{Cs}}(1 - \sigma P_z)$, where N_{Cs} is the number of Cs atoms in the observation region, σ the polarization of the pumping laser, and P_z the electronic spin polarization of Cs atoms. By observing the LIF intensity as irradiating the linearly polarized ($\sigma = 0$) pumping laser, we measured the number of Cs atoms in the observation region, which was decreased by convection, diffusion, and recombination of Cs atoms. Figure 2 shows the measured LIF intensity as a function of time after the sputtering and dissociation lasers were both turned off. Note that the measured photon count rate includes 2–3 kcps of the stray laser light scattered from the walls of the quartz cell and the windows of the cryostat. In Fig. 2, the background count rate, measured separately, has been subtracted. It was found that the atomic density achieved with this new method was as high as that in the previous method [10], where a Cs sample was sputtered in the liquid. It is seen in Fig. 2 that the LIF intensity, namely, the number of Cs atoms, decreases rap-

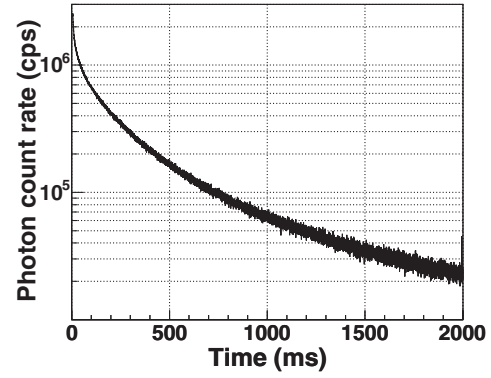


FIG. 2. Typical LIF signal from Cs atoms as a function of time after the termination of irradiation with the sputtering and dissociation lasers. The background count rate of 2–3 kcps due to stray laser light has been subtracted.

idly in the first 100 ms, and later the decrease becomes slower. The rapid decrease is due to the rapid flushing of the Cs atoms out of the observation region. The flushing is mainly caused by the convection flow induced by the dissociation laser. The gradual decrease is due to the remaining weak convection flow, diffusion, and recombination of the Cs atoms. A residence time constant at 500 ms after the Cs introduction was estimated to be ~ 440 ms, which is over 10 times longer than that in the previous method [10]. Even at 2000 ms after the Cs introduction, the LIF count rate is higher than 10^4 cps, which is large enough for long relaxation time measurement.

The residence time achieved in our method is actually still comparable with the relaxation time, so that the decrease of the number of Cs atoms is to be corrected for precisely. For this purpose, we made use of the fact that the linearly polarized pumping laser light makes $P_z = 0$, and that the LIF intensity with the circularly polarized laser just after the irradiation of the linearly polarized laser is thus proportional to only N_{Cs} . Figure 3 shows the time sequence of the laser shots and the measurements of photon signals. After the sputtering and dissociation lasers are shot for 2 s,

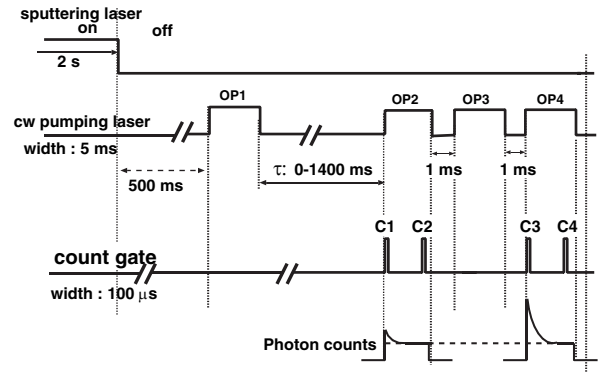


FIG. 3. Time sequence of the laser shots and the measurements of photon signals. Schematic time evolutions of the photon count rate are shown at the bottom.

we wait for 500 ms before starting the relaxation in the dark method to avoid the influence by the large convection flow. The first σ^+ optical pumping (“OP1,” duration of 5 ms) polarizes Cs atoms almost completely. The pumping laser is quickly switched on or off with an acousto-optic deflector (AOD in Fig. 1). After OP1, the electronic spin polarization of the atoms relaxes due to the interaction with the surrounding He atoms for a certain period (dark period, τ). The second σ^+ pumping (“OP2,” 5 ms) follows the dark period ($\tau = 0$ –1400 ms) and the measurements of photon signals are performed during OP2. A schematic time evolution of the photon count rate is shown at the bottom of Fig. 3. As partly depolarized Cs atoms are polarized again, the intensity decreases rapidly. The photon count rates $I_1(\tau)$ and $I_2(\tau)$ are obtained in the count gates C_1 and C_2 (width: 100 μ s), respectively. The larger the spin relaxation is, namely, the smaller the polarization is, the higher the count rate is expected in C_1 . One millisecond after the OP2 period, the polarization of the pumping laser is switched from circular to linear with an electro-optic modulator (EOM in Fig. 1), and the electronic spin polarization of the Cs atoms is extinguished during this period (“OP3,” 5 ms). The last σ^+ pumping (“OP4,” 5 ms) is used to measure the photon count rates $I_3(\tau)$ and $I_4(\tau)$ in C_3 and C_4 , respectively, from the atoms of $P_z = 0$ for the correction of the number of Cs atoms. It should be noted that this normalization was accurate enough because the period from OP2 to OP4 (12 ms) was short compared with the observed residence time of N_{Cs} (~ 440 ms). The above sequence was repeated about 500 times for each τ . Figure 4 shows examples of the time evolution of measured photon count rate during the OP2 (solid lines) and OP4 (dotted lines) periods. The count rate at the beginning of OP2

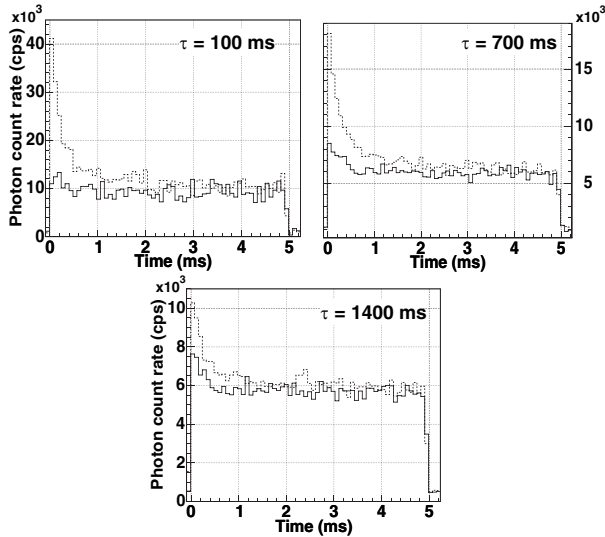


FIG. 4. Typical time evolutions of the measured photon count rate during σ^+ pumping. The solid and dotted lines are the count rates observed during OP2 and OP4, respectively. The constant rates after ~ 2 ms include LIF from the atoms which are not polarized and the background counts from stray laser light.

(solid lines) increases with increasing τ , showing the effect of spin relaxation.

In order to obtain a reliable relaxation time, the waiting time of 500 ms after the sputtering laser shot was adopted by examining the waiting time dependence of the polarization $P_z(\tau)$. When the waiting time was much shorter than 500 ms, we observed rapid decrease of $P_z(\tau)$ with increasing τ , prior to slower decrease. The rapid relaxation was probably due to the effect by the convection flow, which made unpolarized Cs atoms outside the pumping region intrude into the observation region during the dark period. Special care was also paid to the pumping laser power, because the pumping laser may dissociate the Cs clusters, and as a result, N_{Cs} may be increased. The laser power was set to be as low as 150 mW with the spot size of 1 mm² so that the effect of dissociation was negligible while the signal to noise ratio was acceptable.

In our measurement, approximately 90% electronic spin polarization of Cs was achieved. The achieved polarization was estimated by comparing the saturated photon intensity in OP2 (the circularly polarized laser) with that in OP3 (the linearly polarized laser). The imperfect polarization is considered to originate from the pumping laser polarization, which was not completely made circular by the EOM and further deteriorated due to mirrors and cryostat windows. The imperfect polarization, as well as the background count rate from stray laser light, must be taken into account for deducing the relaxation time from the measured photon count rates. The observed photon count rates are described as

$$\begin{aligned} I_1 &= \alpha N_{\text{Cs}}[1 - \sigma P_z(\tau)] + I_{\text{bg}}, \\ I_3 &= \alpha N_{\text{Cs}}[1 - \sigma P_z^{\text{depol}}] + I_{\text{bg}}, \\ I_2 &= I_4 = \alpha N_{\text{Cs}}[1 - \sigma P_z^{\text{pol}}] + I_{\text{bg}}, \end{aligned} \quad (1)$$

where α is a coefficient for overall efficiencies of photon emission and detection, I_{bg} the photon count rate from the stray laser light, and P_z^{pol} and P_z^{depol} the electronic spin polarization of the Cs atoms after the pumping with circularly and linearly polarized lasers, respectively. From Eq. (1), $P_z(\tau)$ is deduced as

$$\frac{P_z(\tau)}{P_z^{\text{pol}}} = 1 - \frac{I_1(\tau) - I_{2,4}(\tau)}{I_3(\tau) - I_{2,4}(\tau)} \left(1 - \frac{P_z^{\text{depol}}}{P_z^{\text{pol}}} \right). \quad (2)$$

Because the photon count rates I_2 and I_4 are the same in principle, they are averaged as $I_{2,4}(\tau) = [I_2(\tau) + I_4(\tau)]/2$. Note that $P_z^{\text{depol}} \neq 0$ due to the imperfection of laser polarization. To determine the ratio of the polarization $P_z^{\text{depol}}/P_z^{\text{pol}}$, we measured the photon count rates I_1 – I_4 when the first pumping laser (OP1) was not irradiated [hence $P_z(\tau) = 0$]. From Eq. (2), we determined the ratio $P_z^{\text{depol}}/P_z^{\text{pol}} = -0.12(11)$.

Figure 5 shows $P_z(\tau)$ normalized to P_z^{pol} for various τ . The errors are only statistical. Although the measurement is limited up to the dark time of 1400 ms due to low count

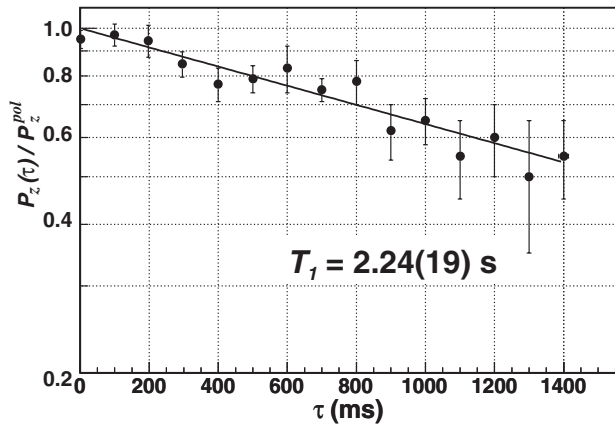


FIG. 5. Relative polarization $P_z(\tau)/P_z^{\text{pol}}$ as a function of dark period τ . The errors are only statistical. The solid line represents the best fit with a single exponential.

rates at large τ , a clear exponential decrease is observed. From a single component exponential fit, the spin relaxation time was determined to be $T_1 = 2.24(19)$ s. In the present analysis, the intrusion of unpolarized Cs atoms due to the remaining weak convection flow and/or diffusion was not taken into account. The effect of intrusion, if any, leads to a spuriously shorter relaxation time. Our result is thus to be taken as a lower limit.

As demonstrated by the much shorter relaxation times in the uniaxially crystallized hcp phase (a few ms) than those in the isotropic bcc phase (1.0–1.8 s) [11] the spin relaxation is strongly dependent on the configuration of surrounding He atoms. The relaxation time in He II is close to but apparently longer than that in the bcc phase of solid He. It suggests that the configuration of He atoms around the Cs atom is as highly symmetric as that in the bcc phase, although there are differences between them. The longer relaxation time in He II would be understood qualitatively as follows. (1) The smaller He density and lower pressure in He II than those in solid lead to weaker perturbations to the impurity Cs atoms. (2) The more flexible structure of liquid around the Cs atom causes less deviation from the spherical shape of the trapping site [“atomic bubble” [1]]. Future investigations of the relaxation mechanism under various He pressures and temperatures would provide more precise information on the differences between liquid and solid He than optical spectroscopic studies, in which even the spectroscopy of Tm atoms with very narrow spectra (~ 0.1 nm) did not find any clear differences between them [17].

In conclusion, we have successfully measured the longitudinal spin relaxation time of Cs atoms in He II for the first time, by employing the new atom implantation method

and the precise correction of the number of Cs atoms. The measured relaxation time is $T_1 = 2.24(19)$ s (lower limit) at 1.5 K. Considering the results of well-studied Cs atoms in solid He, our result suggests that the microscopic configuration of He around the impurity Cs atom is as highly symmetric as or may even be more symmetric than that in the bcc phase. Further systematic investigation of the relaxation time under various conditions, together with magnetic resonance studies, will be useful for better understanding of the spin relaxation mechanism and the microscopic properties of the cold, condensed helium matrices. Finally, this quite long relaxation time encourages us to search for other optically polarizable atoms in He II for the study of various unstable nuclei in addition to alkalis.

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