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Optical pumping and optical detection of the magnetic resonance of alkali-metal atoms in superfluid helium

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By D_1-line optical pumping of alkali-metal atoms (Cs and Rb) in superfluid helium (He II), we could achieve large spin polarization of the atoms in the ground states, more than 50% for both electron and nuclear spins, especially for the Cs atoms. The alkali-metal atoms were implanted into He II by laser sputtering. We detected optically the magnetic resonances between the ground-state Zeeman sublevels, and found that the g values and hyperfine constants in He II were the same as in vacuum within the experimental error. We discuss possible applications of these spin-polarized atoms in He II.

The method for implantation of various kinds of foreign atoms and molecules into superfluid helium (He II) was developed by a group at Heidelberg.[1] Recently we have developed a laser sputtering method,[2] and have successfully detected the signals of alkali-metal atoms (Rb and Cs) in He II (Ref. 3) in spite of the previous null results.[1,4] So far most of the studies have been limited to those on the optical spectra of the impurities influenced by the surrounding helium. In this paper we report results of optical pumping and optical detection of the ground-state magnetic resonance[5] of alkali-metal atoms (^{133}Cs, ^{85}Rb, and ^{87}Rb) in He II. Using the circularly polarized pumping beam tuned to the D_1 line, we have achieved large spin polarization of the atoms in the ground states, more than 50% for both electron and nuclear spins for Cs atoms. We observed the magnetic resonances between the ground-state Zeeman sublevels through monitoring the D_1 fluorescence by means of the optical-rf double resonance technique. These experiments are possible since D_1- and D_2-excitation spectra are well-resolved even in He II.[3,6]

The experimental apparatus and procedure are similar to the ones used for the optical spectroscopy of alkali-metal and alkaline earth atoms in He II.[2,3] The experimental setup is illustrated schematically in Fig. 1(a). The experiment was performed at the helium temperature of about 1.6 K at the saturated vapor pressure. We injected alkali-metal atoms (Rb and Cs) into He II by laser sputtering (density $\sim 10^{8\sim 10}$ atoms/cm^3). A sputtering pulse laser beam from a Q-switched Nd: YLF (YLiF$_4$) laser (wavelength: 523 nm, pulse energy: 200 μJ, repetition rate: 1 KHz) was focused onto the metal samples mounted in He II. A cw circularly polarized pumping light beam from a Ti:sapphire laser was incident parallel to the external static magnetic field H_0 (≤ 100 G). The power of this pumping beam was varied from 100 to 700 mW in front of the glass dewar. The wavelength was tuned to 778 nm for Rb or 876 nm for Cs which correspond to the peak position of the atomic D_1-excitation spectrum in He II.[3] The pumping and sputtering beams were crossed about 1 cm away from the metal sample surface to avoid the vapor region due to sputtering near the sample. The pumping beam was focused (beam diameter ~ 500 μm) at the crossing region (≤ 1 mm^3) located at the center of the six-turn rf coil (length ~ 20 mm, diameter ~ 10 mm). The direction of the rf magnetic field $H_1(t)$ was perpendicular to that of the static magnetic field. The frequency of the rf field was fixed and the external magnetic field H_0 was swept slowly, the typical sweep rate being about 0.1 G/sec. The laser-induced

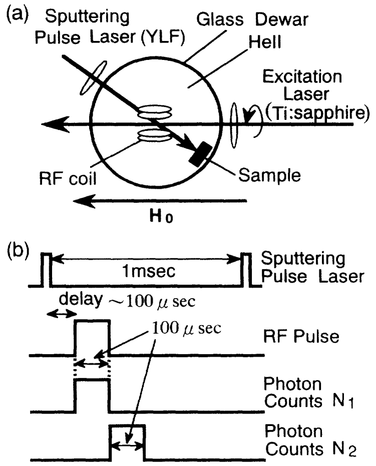

FIG. 1. (a) Experimental setup. The alkali-metal atoms were injected into He II by a sputtering beam from a Q-switched laser focused onto the metal sample mounted in He II. A cw circularly polarized pumping beam from a Ti:sapphire laser propagated parallel to the external static magnetic field H_0. The laser power was varied from 100 to 700 mW in front of the glass dewar. The crossing region of the sputtering and pumping beams was set in the middle of the rf coil [$H_1(t) \perp H_0$]. The photon counting system of the laser-induced fluorescence (LIF) from the crossing region is not shown in the figure. (b) Typical timing chart of sputtering, monitoring, and rf pulses. Between the sputtering and rf pulses the atoms were optically pumped. During the rf pulse the LIF signals were up-counted (N_1), and then down-counted (N_2) during the following 100 μsec. Thus we detected only the change of the LIF counts ($N_1 - N_2$) due to the rf field.

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fluorescence (LIF) from the crossing region was focused onto a monochromator and was detected by a photon counting system. The wavelength of the monochromator was set to 793.5 nm for Rb or 892.5 nm for Cs which corresponded to the peak position of the atomic D_1-emission spectrum in He II.[3]

The count rates of the LIF signals were as high as 10^5 sec^{-1} and the S/N ratio was excellent (typically $10^{2\sim3}$). The count rates fluctuated severely in time mainly due to the change of the surface conditions in the course of sputtering. Thus the following procedure were required to obtain a good S/N ratio for the magnetic resonance signals. Figure 1(b) shows a typical experimental timing chart. The rf pulse of about 100 μsec width was applied after a sputtering pulse with a delay time of about 100 μsec. Between these two pulses the atoms were optically pumped. During the rf pulse of 100 μsec width, the LIF signals were monitored. The LIF signal increased due to the repopulation in case of the resonance. The LIF signals were then down-counted during the following 100 μsec for optical pumping. The difference between the photon counts during the two spans (N_1-N_2) was detected. Repeating this sequence about 10^3 times, we integrated the signal counts and then recorded them as one datum.

Figure 2 shows the magnetic resonance signals of Cs atoms observed in the cases of (a) σ^+ pumping and (b) σ^- pumping, respectively. The frequency of the rf field is 18.5740 MHz. Figure 3 shows the magnetic resonance signal of ^{85}Rb and ^{87}Rb atoms in the cases of (a) σ^+ pumping and (b) σ^- pumping, respectively. The frequency of the rf field is 23.1740 MHz. Resonance signals were not detected when the pumping light was linearly polarized. The observed linewidth of about 0.5 G (FWHM) was much broader than the electron spin resonance (ESR) width (half-width at maximum derivative) of 5 mG for e^- in He II.[5] The present linewidth was found to be mainly due to rf power broadening from the rf amplitude estimated in the transient nutation experiment. We applied a relatively strong rf field in order to obtain the magnetic resonance signal with a good S/N ratio. Inhomogeneity of H_0 was negligible because the observation region was very small (≤ 1 mm^3). The optical pumping time, the rf pulse duration, and the residence time of the atoms within the beam (≥ 1 msec) are too long to give the observed width.

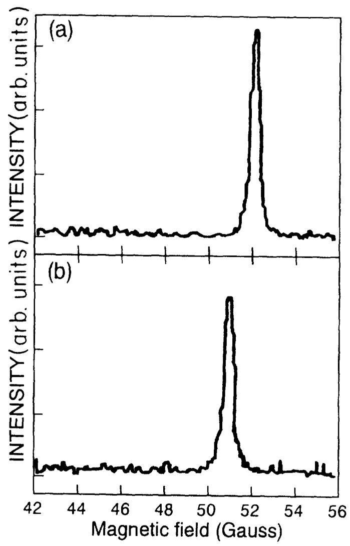

FIG. 2. Optical-rf double resonance signals for Cs atoms in He II in the cases of (a) σ^+ and (b) σ^- pumping. These signals were obtained by the excitation at 876 nm (about 500 mW in front of the glass dewar) and by detecting the emission line at 892.5 nm. The frquency of the rf field was fixed at 18.5740 MHz.

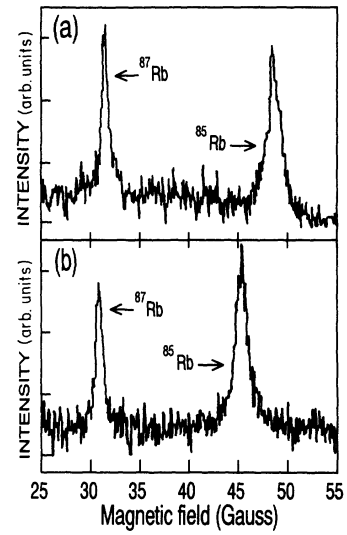

FIG. 3. Optical-rf double resonance signals for ^{85}Rb and ^{87}Rb atoms in He II in the cases of (a) σ^+ and (b) σ^- pumping. These signals were obtained by the excitation at 778 nm (about 500 mW in front of the glass dewar) and by detecting the emission line at 793.5 nm. The frequency of rf field was 23.1740 MHz.

The observed resonance frequencies or magnetic fields were the same within the experimental error as those of the free atoms if the observed signals corresponded to the transition from $|F,M_F=F>$ to $|F,F-1>$ for σ^+ pumping and that from $|F,-F>$ to $|F,-F+1>$ for σ^- ($F=4$ for Cs, 3 for ^{85}Rb, and 2 for ^{87}Rb). Thus we believe that these assignments are correct indeed.

We could obtain the g_J values and the hyperfine constants A_J $(J=\frac{1}{2})$ of the ground states of the atoms in He II by using the Breit-Rabi formula.[7] Here we assumed that the formula remained valued for the atoms in He II, and only the values of g_J and A_J were changed. At first the resonance data were fitted using the Breit-Rabi formula for the $|F,F>$ to $|F,F-1>$ transition or for the $|F,-F>$ to $|F,-F+1>$ transition. We obtained $g_J=2.10\pm0.08$ for Cs, 2.12 ± 0.02 for ^{85}Rb, and 2.13 ± 0.04 for ^{87}Rb atoms from the least-squares fitting. The accuracy was limited mainly by the uncertainty of the value of the static magnetic field H_0, including an error of a few percent mainly due to stray magnetic fields. Considering the systematic error of several percent, these values are the same as those of free atoms within the experimental error, which indicates that the symmetry of the ground-state electron orbital is not significantly affected by the surrounding helium atoms. Because of the weak magnetic fields (≤ 100 G) used in this experiment the fitting was much less sensitive for the hyperfine constants A_J. However, the difference of the resonance magnetic fields in the cases of σ^+ and σ^- pumping (ΔH_0) is more sensitive to the A_J value, and is, of course, insensitive to the stray field. Using the expression for ΔH_0 as a function of the frequency obtained by solving the Breit-Rabi formula and fixing the g_J value to that of free atoms, we obtained that $A_J=2.37\pm0.11$ GHz for Cs, 1.13 ± 0.06 GHz for ^{85}Rb, and 3.25 ± 0.02 GHz for ^{87}Rb atoms from the least-squares fitting. Here we neglected the nuclear Zeeman term, which is three orders of magnitude smaller than the electron Zeeman term. These values are again not so different from those of free ones, which indicates that the ground-state electron-nucleus interaction is not so largely affected by surrounding helium atoms.

Here we estimate the influence of the surrounding helium on the hyperfine constant of the ground state in terms of the simple spherical square-well bubble model[8] which has been found to explain qualitatively the optical spectra of the alkali-metal atoms.[3] In this model the surrounding helium atoms are found only outside a sphere of radius R which we shall call the bubble radius. The electrons of the atoms are assumed to feel the pseudopotential of 1.02 eV depth outside the bubble radius. We calculated the equilibrium bubble radius R_{eq} which gives a minimum of the total energy $E_{tot}(R)$ of the atomic bubble. The total energy $E_{tot}(R)$ is expressed as

$$E_{tot}(R)=E_{atom}(R)+4\pi R^2\sigma+\tfrac{4}{3}\pi R^3 P\ , \qquad (1)$$

where $E_{atom}(R)$ is an atomic energy with the bubble radius R, the second term is the surface energy, and the third term is the pressure-volume work. The value of the surface tension σ is taken to be 0.354 erg/cm^2, and the helium pressure P in the present case is 5.6 Torr. For Cs atoms R_{eq} becomes 7.24 A, and the probability of finding an electron at the nucleus, $|\phi(0)|^2$, for this bubble radius increased about 0.6% from that of free Cs atoms. The hyperfine constant is expected to be increased by this same factor. Quite recently we observed directly the hyperfine transition for Cs atoms with a microwave field and found A_J was increased by about 0.5%. The details will be reported elsewhere.

In the case of Cs atoms, we also observed the resonance signals under relatively large H_0 (~80 G), which showed again symmetrical line shapes as seen in Fig. 2 and did not show apparent structured profiles due to the transitions other than those from $|F,\pm F>$ to $|F,\pm(F-1)>$. This suggested a sizable polarization was achieved. In fact, in our quite recent experiment with Cs atoms we observed only the hyperfine transition from $|4,-4>$ to $|3,-3>$ or from $|4,4>$ to $|3,3>$, indicating that nearly perfect polarization was achieved for Cs atoms. However, from the ratio of the LIF counts in the presence of the saturating rf field to those without the rf field, we estimated the polarization to be more than 50% for Cs atoms. We have not yet fully understood this discrepancy about the value of polarization, but it is apparent that we must take into consideration that the atoms in the outer region of the beam should contribute to the photon counts, but not to the magnetic resonance signal since these atoms would not be optically pumped due to the low intensity of the pumping beam. On the other hand, in the case of ^{85}Rb and ^{87}Rb atoms, the spin polarization was not perfect (at most ~20% deduced from LIF count ratio). The relatively small polarization of Rb atoms compared with Cs ones would be due to the slight overlap of the D_1- and D_2-excitation spectra.[3]

We also measured the pumping time T_P for Cs atoms. The static magnetic field and the frequency of the rf field were fixed at the resonance condition ($H_0=41.9$ G and $f=15.1600$ MHz). The gate pulse (10 μsec width) for the photon counting was delayed from the rf pulse. Figure 4 shows the change of the LIF intensity plotted against the delay time. We omitted the data at longer delay time in Fig. 4 because of their poor S/N ratio. Thus we obtained $T_P=21\pm1$ μsec and 110 ± 30 μsec for 700 and 100 mW excitations, respectively. We estimated T_P of the Cs atoms in He II using the assumptions that both electron and nuclear spins were randomized in the excited state within the radiative lifetime of 34 nsec (Ref. 9) and that the oscillator strength was not affected by the surrounding helium. Using these assumptions and taking account of the observed width of the D_1-excitation spectrum,[3] we obtained $T_P\simeq20$ and 150 μsec for 700 and 100 mW excitations, respectively.

The spin polarized atoms in He II have unique features of long residence time, long relaxation time, high sensitivity due to optical detection, and large spin-polarization due to optical pumping. Such an atomic system is applicable to various fundamental researches. First we discuss the possibility of searching for a permanent electric dipole moment (EDM) of paramagnetic atoms in He II which has recently been proposed by Arndt *et al.*[10] They considered Au atoms to be the prime candidate for the experiment and investigated their recombination fluorescence in He II. We think that Cs atoms may be better for this purpose because efficient optical pumping and the optical detection of magnetic resonance was really possible as demonstrated in the present work. The excitation spectrum of the D_1 line of Au atoms, not yet observed, is

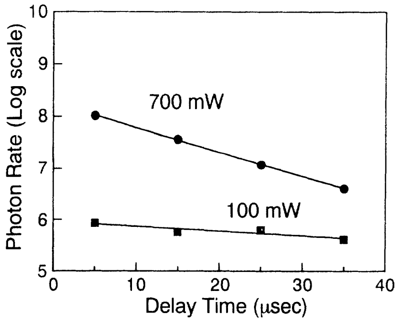

FIG. 4. Change of the LIF intensity (log scale) of Cs atoms in He II as a function of the delay time of the monitoring pulse from the rf pulse. From the fitting, the pumping time T_P was measured to be 21 μsec for 700 mW excitation and 110 μsec for 100 mW excitation.

expected to be in the uv region, which may cause some difficulties. Second, an application to nuclear physics is expected. Radiation-detected optical pumping[11] can be performed in superfluid helium. The values of g factors and hyperfine constants of unstable nuclei may be systematically investigated, and thus studies of the hyperfine anomaly might be carried out. By using various techniques of spin echoes[12] in inhomogeneous magnetic fields, the motions of neutral impurity atoms can be also studied which gives information on He II and the structures of impurities as well as the homogeneous linewidth.

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[6] We also tried to observe the magnetic resonances of Ba atoms in the metastable triplet states ^3D_J $(J=1,2,3)$ in He II with a similar experimental system. However, we could not detect the signals. That is, a sizable population difference could not be produced by optical pumping. This may be because the spin relaxation time T_1 of a non S state becomes quite short through the perturbed spin-orbit interaction.

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[9] This may be appropriate because we measured radiative lifetimes of triplet states ^3P_J $(J=0,1,2)$ of Ba atoms in He II and found that they were about the same as those in vacuum.

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