Buffer-gas-assisted polarization spectroscopy of ^{6}Li

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We report on the demonstration of Doppler-free polarization spectroscopy of the D2 line of ^{6}Li atoms. Counterintuitively, the presence of an Ar buffer gas, in a certain pressure range, causes a drastic enhancement of the polarization rotation signal. The observed dependence of the signal amplitude on the Ar buffer pressure and the pump laser power is reproduced by calculations based on simple rate equations. We performed stable laser frequency locking using a dispersion signal obtained by polarization spectroscopy for laser cooling of ^{6}Li atoms.

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Since the first realization of Bose–Einstein condensation in a dilute atomic gas, extensive studies of quantum-degenerate gases have been carried out (see, for example, [1,2]). Recently, there has been increasing interest in quantum-degenerate fermions as a tool to explore the physics of strongly correlated electron systems [3]. Lithium-6 is one of the workhorses for the study of quantum-degenerate fermions since the strength of the interatomic interaction is widely tunable via a broad Feshbach resonance [4]. Lithium-6 is also attractive for the study of ultracold heteronuclear molecules since molecules composed of lithium, such as LiRb and LiCs, have relatively large electric dipole moments [5], which are beneficial for exploring novel quantum phases of ultracold molecules [6].

To date, quantum-degenerate samples of fermions have been produced by the combination of laser cooling and evaporative cooling [3]. For laser cooling, it is necessary to lock the frequency of the cooling laser to a specific resonance line. Frequency modulation spectroscopy is widely used to obtain an error signal for laser frequency locking [7]. Alternatively, modulation-free frequency stabilization techniques, such as dichroic atomic vapor laser lock [8], have been employed. Doppler-free polarization spectroscopy (DPS) [9] can also offer an dispersive error signal at the atomic resonances, and has been used for modulation-free laser frequency locking to the rubidium D2 line [10,11].

Surprisingly, there has so far been no report on DPS of lithium atoms in the literature. In this Letter, we demonstrate DPS of the D2 line of ^{6}Li atoms and prove that DPS provides dispersion signals suitable for laser frequency locking. For spectroscopy of Li atoms, an Ar buffer gas is usually introduced in the vapor cell to prevent the Li atoms from reaching to the viewports. We study the effect of the Ar buffer gas on the polarization spectrum, and find that the amplitude of the dispersion signal at the cooling transition is drastically increased by the help of the Ar buffer gas. The observed dependence of the signal amplitude on the Ar buffer pressure and the pump laser power is reproduced by calculations based on simple rate equations considering all the relevant magnetic sublevels. We also demonstrate laser cooling of ^{6}Li atoms with a laser that was frequency stabilized using a DPS dispersion signal enhanced by the Ar buffer gas.

Figure 1(a) shows the energy diagram of ^{6}Li. For laser cooling of ^{6}Li atoms, the cooling laser is tuned to the $2S_{1/2}$, $F = 3/2 \rightarrow 2P_{3/2}$, $F' = 5/2$ transition, whereas the repumping laser is tuned to the $2S_{1/2}$, $F = 1/2 \rightarrow 2P_{3/2}$, $F' = 3/2$ transition. The D2 line of ^{6}Li is unique in that the hyperfine-structure splittings of the $2P_{3/2}$ states are smaller than the natural linewidth of 5.9 MHz. Therefore, the atoms in the upper ($2S_{1/2}$, $F = 3/2$) hyperfine level are easily pumped to the lower ($2S_{1/2}$, $F = 1/2$) hyperfine level by the cooling laser via a few absorption-spontaneous emission cycles (hyperfine pumping).

Figure 1(b) shows the experimental setup. A 5 g chunk of ^{6}Li (enriched >95%) was installed in the middle of a 50-cm-long stainless steel tube ended by two ICF70 glass view ports. The tube had a valve to introduce an Ar buffer gas with a desirable pressure ranging from 0 to 100 mTorr. The central part (~20 cm) of the tube was magnetically shielded by winding a sheet of μ-metal and heated at 350 °C by a tape heater. A 671 nm laser beam was derived from an external-cavity diode laser using an antireflection-coated diode laser.

The optical setup for DPS of ^{6}Li atoms was basically the same as in [10]. A circularly polarized pump beam and a linearly polarized probe beam, both of which

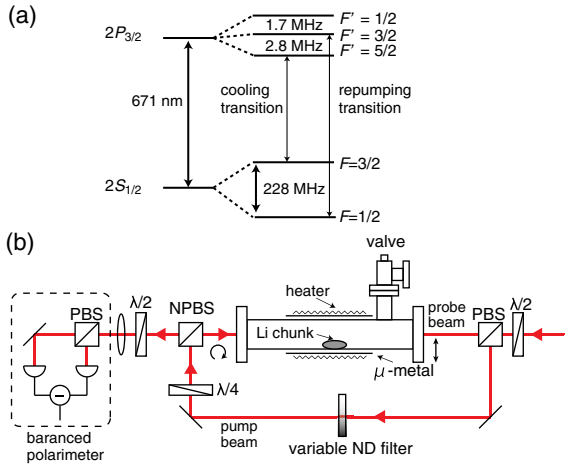

Fig. 1. (Color online) (a) Relevant ^{6}Li energy levels. (b) Schematic diagram of the experimental setup. PBS, polarization beam splitter; NPBS, nonpolarization beam splitter; $\lambda/2$, half-wave plate; $\lambda/4$, quarter-wave plate; ND filter, neutral density filter.

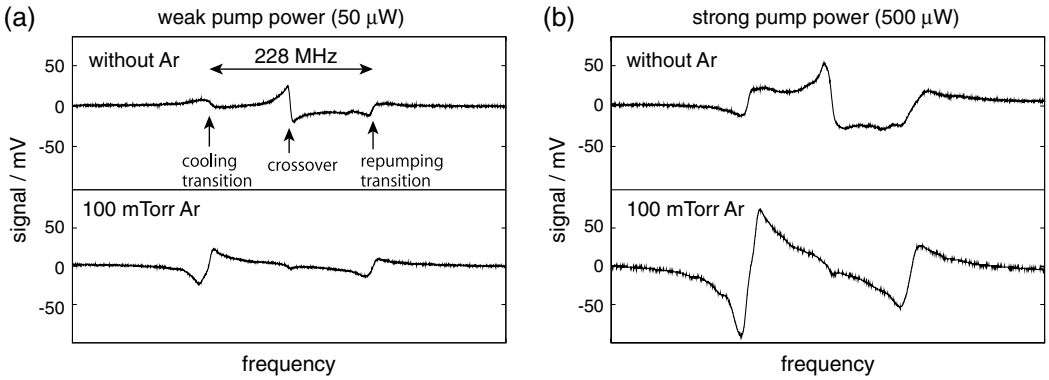

Fig. 2. Doppler-free polarization spectra of the $2S_{1/2} \rightarrow 2P_{3/2}$ transitions of ^{6}Li at zero and 100 mTorr Ar buffer gas pressures for (a) a weak (50 μW) and (b) a strong (500 μW) pump power.

had almost the same diameters (~2 mm), were sent to the vapor cell in a Doppler-free configuration. The power of the pump beam was varied from 0 to 500 μW using a neutral density filter, whereas that of the probe beam was fixed at 50 μW (a power of ~150 μW corresponds to the saturation intensity of 2.5 mW/cm^2). Circular birefringence induced by the pump beam was monitored by the probe beam as a rotation of the polarization axis. The angle of polarization rotation was then converted to an electronic signal using a balanced polarimeter [10].

We performed DPS with various pump powers and Ar buffer pressures. Figure 2 shows typical polarization spectra at zero and 100 mTorr Ar buffer pressures for a weak (50 μW) and a strong (500 μW) pump power. In the pressure range below 100 mTorr, the collision broadening due to the Ar buffer gas is negligible (<1 MHz). We observed three dispersion signals at the frequencies of the cooling, the crossover, and the repumping transitions, as indicated by the arrows in the figure. The $2P_{3/2}$ hyperfine structures were not resolved. Each dispersion signal displayed different behavior as we introduced an Ar buffer gas. The amplitude of the dispersion signal at the cooling (crossover) transition increased (decreased) with increasing the pressure of the Ar buffer gas. Note that the slope of the dispersion signal at the cooling transition changes its sign for a weak pump power, indicating that there are different physical origins of circular birefringence, as discussed below.

Figure 3 shows the dependence of the amplitude of the dispersion signal at the cooling transition on the pump power. The amplitudes of the dispersion signal with positive (negative) slopes are plotted in the positive (negative) direction of the vertical axis. Without an Ar buffer gas, the slope changes its sign at the pump power around 150 μW, and then the amplitude increases monotonically with increasing the pump power. With a 100 mTorr Ar buffer gas, the amplitudes are much larger than those without an Ar buffer gas for any pump power. We observed no such enhancement of the amplitude of the Lamb dips in usual saturated-absorption spectroscopy with an Ar buffer gas.

We compared the experimental data with calculations based on simple rate equations considering all the relevant magnetic sublevels as explored in [12], where the effect of velocity-changing collisions (VCCs) caused by the Ar buffer gas is included as a thermalization of the velocity distribution for each magnet sublevel (VCCs remove atoms from the zero velocity group out of resonance and bring atoms from other velocity groups into resonance). We also included the effect of hyperfine pumping due to the probe beam itself in the rate equations. The relaxation rate due to VCCs was set to a typical value of 10 MHz/Torr [13]. The optical density of the ^{6}Li gas and the diameter of the pump beam were chosen to fit the experimental data. The experimental data are in good agreement with the calculations assuming a pump beam diameter of 2.5 mm (the resultant transit-time broadening is 90 kHz), which is close to the actual pump beam diameter of ~2 mm.

Without an Ar buffer gas, the typical amplitude of the dispersion signal observed at the cooling transition corresponds to the polarization rotation angle of $\sim 10^{-3}$ rad. This rotation angle is about 1 order of magnitude smaller than those observed for Rb and Cs vapors with comparable optical densities [14]. For the D2 lines of Rb and Cs, the hyperfine-structure splittings of the excited states ($nP_{3/2}$) are much larger than the natural linewidth and the cooling transition is nearly closed. Therefore, the atoms are easily spin polarized by a relatively weak circularly polarized pump beam and exhibit circular

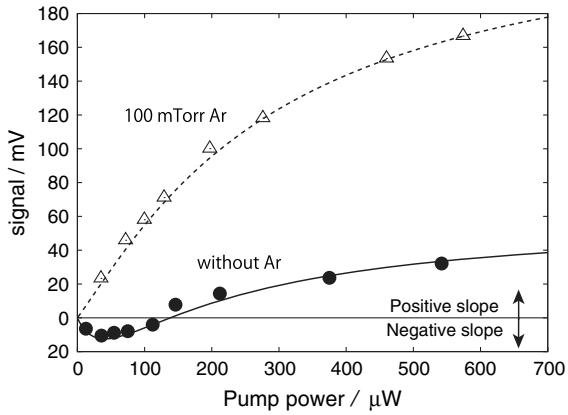

Fig. 3. Dependence of the dispersion signal at the cooling transition on the pump power. The vertical axis represents the peak-to-peak amplitude of the dispersion signal. The open triangles and the solid circles represent the experimental data at zero and 100 mTorr Ar buffer gas pressures, respectively. The solid curve and the dashed curve show calculations for each condition.

birefringence [14]. We call this birefringence due to spin polarization Type I. On the other hand, as mentioned above, ^{6}Li atoms are easily pumped to the lower hyperfine state by a circularly polarized pump beam, leading to a reduced efficacy of spin polarization. If we increase the pump power, another type of circular birefringence emerges: the σ^+ (σ^-) pump beam saturates the σ^+ (σ^-) transitions and weakens the strength of the interaction with the σ^+ (σ^-) component of the linearly polarized probe beam, resulting in circular birefringence. We call this saturation-induced birefringence Type II [9]. Unfortunately, the effects of these two types of birefringence cancel out for the cooling transition [15], resulting in a small dispersion signal for any pump power. The change of the sign seen in Fig. 3 is explained by a slight difference between the dependences of the two types of birefringence on the pump power.

The enhancement of the dispersion signal at the cooling transition by introducing an Ar buffer gas can be qualitatively explained as follows. VCCs due to the Ar buffer gas prevent spin polarization of the ^{6}Li atoms, leading to a suppression of Type I birefringence. On the other hand, Type II birefringence is not so affected by the presence of the Ar buffer gas since the population relaxation rate due to VCCs at Ar buffer pressures below ~100 mTorr is much smaller than the spontaneous decay rate [12]. As a consequence, the unfortunate cancellation of the two types of birefringence is broken and Type II birefringence manifests itself. Moreover, the reduction of the population of the zero velocity group due to hyperfine pumping by the probe beam is compensated to some extent by the help of VCCs. As a result, the dispersion signal at the cooling transition is significantly enhanced by the Ar buffer gas.

The origin of the crossover signal between ground-state hyperfine levels is a bump in the velocity distribution created by hyperfine pumping due to the pump beam. VCCs due to the Ar buffer gas, as mentioned above, smear the bump; therefore, the introduction of the Ar buffer gas leads to a diminishment of the crossover signal as seen in Fig. 2.

We performed laser frequency locking using a dispersion signal obtained by polarization spectroscopy for laser cooling of ^{6}Li atoms. The laser was locked with a stability of less than 1 MHz for a day. Using this frequency stabilized laser, we demonstrated magneto-optical trapping of ^{6}Li atoms.

In conclusion, we have demonstrated DPS of the D2 line of ^{6}Li atoms and studied the effect of an Ar buffer gas. We found that the dispersion signal at the cooling transition is drastically enhanced by an Ar buffer gas, which is explained by the two effects of VCCs with Ar atoms: the breaking of the cancellation between two types of birefringence and the compensation of hyperfine pumping by the probe beam. We performed stable laser frequency locking using a dispersion signal obtained by polarization spectroscopy for magneto-optical trapping of ^{6}Li atoms.

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