Spin Manipulation Using the Light-Shift Effect in Rubidium Atoms

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Optical manipulation of spin coherence in rubidium atoms is studied. The effect of off-resonant and circularly polarized light on optically induced magnetization is investigated. The change in precession frequency caused by the light-shift effect is verified. Absorption-free phase control of spin precession and pure spin rotation about an arbitrary axis are demonstrated. A theory of precession frequency shift that includes the effect of absorption is considered by using the density matrix and the experimental results are in agreement with the predictions of the theory. Thus, we show that it is possible to carry out off-resonant control of spin coherence and all-optical manipulation of spins.

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Recently, there has been growing interest in quantum information processing, and trapped ions or atoms have played an important role in this regard [1]. Entanglement and quantum gate operations have been realized [2]. A register of atomic qubits can be served by the array of atoms [3]. The two substates in the ground states of the atoms can be modeled as spin $1/2$ particles [4]. In trapped-atom systems, optical manipulation of spins is one of the essential tools for the implementation of the processing. The optical manipulation of spins is also important for verifying the nonadiabatic quantum phases [5], because it has been proposed that the geometric phase in electron spin systems can be detected by using Faraday rotation spectroscopy [6]. Geometric phases depend only on the geometry of the path executed, and are therefore resilient to certain types of errors. This suggests the possibility of an intrinsically fault-tolerant way of performing quantum gate operations [7].

Quantum computing in spin systems has been demonstrated by manipulating nuclear spins using the nuclear magnetic resonance technique [8]. If the spin can be manipulated voluntarily by an all-optical method, more rapid control of the spin system and broadband responses can be achieved. Ultrafast spin dynamics can be observed in the gigahertz and terahertz regions if ultrashort laser pulses are used [9,10]. Several studies have demonstrated ultrafast optical manipulation of spins [11–13]. However, pure spin rotation about an arbitrary axis and phase control of spin precession, which are free from absorption effects, have not yet been reported.

In order to achieve optical manipulation of spin coherence, it is required that the magnetization is generated, detected, phase controlled, and rotated about an arbitrary axis by optical means. One of the most promising approaches for achieving this is off-resonant manipulation of spins by using the light-shift effect. By this effect, off-resonant radiation causes an apparent shift in the energy levels of the atoms [14,15]. Under appropriate experimental conditions, the light shift has a similar effect on the spin dynamics as magnetic fields. Selected energy levels as well as the dynamics of sublevel coherences are affected by the light shift [16–18].

From the viewpoint of spatial control of level shift and atomic potential, the light-shift effect is useful as a tool to be used in laser cooling techniques and trapping of neutral atoms [19]. The concept of fictitious magnetic field was already pointed out at the beginning [14,20]. However, very few studies have been conducted on spin manipulation using fictitious magnetic fields that are generated due to the light-shift effect. Ultrafast manipulation of electron spin coherence in semiconductor quantum wells was studied by Gupta *et al.* [21]. They reported that optical "tipping" pulses can enact substantial rotations of electron spins through a mechanism dependent on the optical Stark effect, also known as the light-shift effect. However, in their experiment, the amplitude of the spin-precession signal was reduced by the absorption effect of the tipping pulse, and the observed rotation angle approached $\pi/2$ as the intensity of the tipping pulse increased. A rotation angle greater than $\pi/2$ was not observed.

In our previous paper [22], we studied optically induced spin echoes in rubidium atoms, where on- and off-resonant manipulations of spins were investigated theoretically and experimentally. Optically induced magnetization generated by a generation pulse was manipulated by a short control pulse parallel to the induced magnetization. In the present study, we investigate the optical manipulation of spin coherence in rubidium atoms by studying the effect of control pulses on the optically induced magnetization, where the direction of the control pulse is perpendicular to the induced magnetization and its duration is relatively long. We verify the generation of fictitious magnetic fields and control the change in the precession frequency. Absorption-free phase control of spin precession and pure spin rotation about the direction of the control pulse with a rotation angle of greater than 2π are demonstrated. We consider a theory for the precession frequency shift in the optically induced magnetization that includes absorption effects, by using the density matrix. The dependence of the precession frequency shift on detuning frequency is compared with the predictions of the theory. In this study, we show that it is possible to carry out all-optical manipulation of spin coherence.

The experimental setup is similar to that used for off-resonant manipulation in our previous study [22], except for the direction and beam profile of the control pulse. The generation and probe pulses are provided by a laser diode, and the control pulse by a Ti:sapphire laser. The beam directions of the three pulses are shown in the inset of Fig. 1. The generation and control pulses are circularly polarized by $\lambda/4$ plates. The magnetization generated by optical pumping is detected as the change in the polarization of the linearly polarized probe pulse. The beam diameter of the generation and probe pulses at the sample is approximately 1 mm. For the control pulse, spatial homogeneities of circular polarization and light intensity in the laser beam are very important. A zeroth order $\lambda/4$ plate made of quartz crystal is used to achieve a homogeneous circular polarization of the control pulse. The beam diameter of the control pulse is extended to approximately 2 cm using a lens and the central portion, i.e., a diameter of approximately 1 cm, is used to obtain homogeneous light intensity. The control beam, which is perpendicular to the direction of the collinear generation and probe beams, is loosely focused on the two overlapping beams using a cylindrical lens. Typical intensities of the generation, control, and probe pulses are 2, 50, and 0.3 mW, respectively.

In the experiment with a fictitious magnetic field, the sample cell is enclosed in μ-metal plates, which shield the cell from residual magnetic fields. A typical experimental result of the observed spin precession around a fictitious magnetic field in the absence of an external magnetic field is shown in Fig. 1, where the generation and probe beams are tuned to the resonance line of ^{87}Rb ($F = 2 \rightarrow F' = 1$) and the detuning frequency $\Delta/2\pi$ of the control beam is -4.25 GHz. The generation pulse is turned off at $t = 0$. The black curve shows the signal of the optically induced magnetization under the influence of the control pulse. The gray curve shows the signal without the influence of the control pulse; this is the free induction decay (FID) of the optically induced magnetization. The dotted curve shows the control pulse. The damped oscillations under the influence of the control pulse represent the precession of the magnetization around the fictitious magnetic field generated by the control pulse, where the precession angle is greater than 2π. When the control pulse is off, the signal damps without any oscillation.

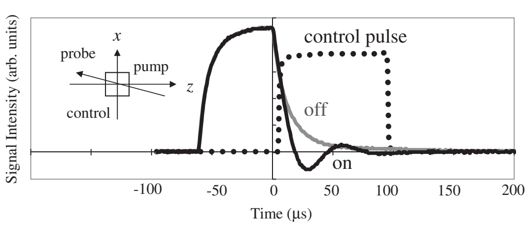

FIG. 1. Typical experimental result of observed spin precession around fictitious magnetic field, where the detuning frequency $\Delta/2\pi$ is -4.25 GHz. The inset shows the beam directions of the three pulses.

The precession frequency is derived from the Fourier transform of the normalized signal $A\cos(\delta t)\exp(-\gamma t)$, which is obtained by dividing the magnetization signal under the influence of the control pulse by the magnetization signal without the influence of the control pulse. δ denotes the light shift, i.e., the angular frequency of precession induced by the optical field of the control pulse, and γ denotes the relaxation rate. The detuning-frequency dependence of the precession frequency caused by the light-shift effect is shown in Fig. 2, where the horizontal axis represents the detuning frequency, i.e., the difference between the transition frequency of atoms and the control pulse frequency, and the vertical axis represents the precession frequency of magnetization. The precession frequency is high at the off-resonant parts of the probed line; however, the signal at zero detuning frequency does not show precession.

The dependence of the precession frequency on the intensity of the control pulse is observed at detuning frequencies of -1, -2, -3, -4, and -5 GHz. At a zero detuning frequency, precession of magnetization is not observed, because of large damping. The dependence for the detuning frequency $\Delta/2\pi = -1$ GHz is shown in the inset of Fig. 2. It is observed that the precession frequency is proportional to the light intensity of the control pulse for all values of detuning frequencies.

In order to demonstrate frequency and phase control of the spin precession, we applied an external magnetic field and observed the influence of the control pulse on the precession of the magnetization. The observed frequency and phase control of the optically induced magnetization is shown in Fig. 3. The direction of the external magnetic field is parallel to that of the control pulse. The optically

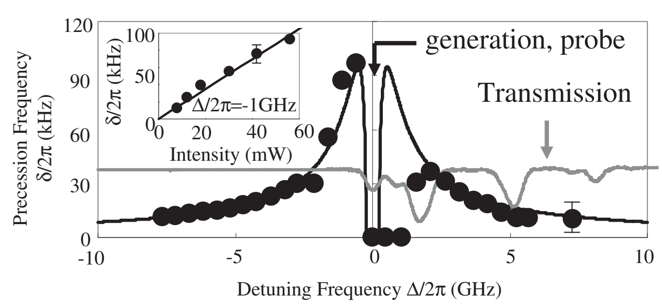

FIG. 2. Detuning-frequency dependence of precession frequency. The horizontal axis represents the detuning frequency of the control pulse, and the vertical axis the precession frequency of magnetization. The intensity of the control pulse is 60 mW. The solid line represents the theoretical detuning-frequency dependence of the precession frequency calculated from Eq. (4). Intensity dependence of precession frequency for the detuning frequency $\Delta/2\pi = -1$ GHz is shown in the inset.

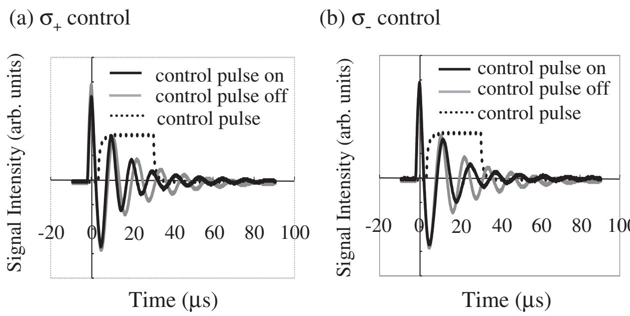

FIG. 3. Effect of σ_+ and σ_- polarizations of control pulse on optically induced magnetization in an external magnetic field of 0.14 Oe, where the detuning frequency $\Delta/2\pi$ is -6.5 GHz.

induced magnetization precesses at the Larmor frequency in the magnetic field, which is the vector sum of the external and fictitious magnetic fields. For circular polarization σ_+ of the control pulse, the precession frequency is increased over the duration of the control pulse. On the other hand, for circular polarization σ_-, the precession frequency is decreased. In the case of σ_+ polarization, the phase of precession proceeds after the end of the control and delays in the case of σ_- polarization. This result suggests that the phase of the spin precession can be controlled by applying an appropriate short control pulse.

The experimental result of optically induced spin echoes using spin rotation [22] is shown in Fig. 4. The rotation axis is determined by the direction of the control pulse. The observed spin-echo signals for the parallel and perpendicular manipulations of the control pulse are shown in Figs. 4(a) and 4(b), respectively, where the direction of the control pulse is parallel (z axis) and perpendicular (x axis) to that (z axis) of the magnetization generated by the generation pulse. The spin-echo signal for parallel manipulation shown in Fig. 4(a) is in-phase with the FID signals of the generation and control pulses, which implies that the spins are refocused along the $+z$ axis. This can be understood from the vector model for the off-resonant manipulation [22]. On the other hand, the spin-echo signal for perpendicular manipulation shown in Fig. 4(b) is out of phase with the FID signals. This implies that the control pulse rotates the spins around the x axis, and the spins are refocused along the $-z$ axis. This can also be understood from the vector model, where the direction of the rotation axis is changed from the z axis to the x axis. This result of the spin-echo signals suggests that spin rotation about an arbitrary axis can be achieved by the control pulse. The rotation axis is determined by the beam direction of the control pulse, and the rotation angle can be adjusted by adjusting the intensity or duration of the control pulse.

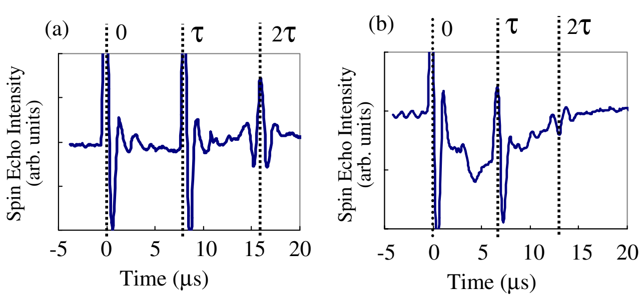

FIG. 4 (color online). Optically induced spin-echo signals using spin rotation. The direction of the control pulse is (a) parallel (z axis) and (b) perpendicular (x axis) to that (z axis) of the magnetization generated by the generation pulse. An inhomogeneous external magnetic field, whose inhomogeneous width is 1 Oe centered at 0.8 Oe, is applied along the y axis.

In the experiments on spin rotation and phase control, it is shown that the fictitious magnetic field caused by the light-shift effect has a similar effect as a real magnetic field. To explain the effect of the control polarization, we consider four-level atoms having sublevels $|+1/2\rangle$ and $|-1/2\rangle$ in the excited and ground states, respectively. The degeneracies of energy levels are removed by applying an external magnetic field. The optically induced magnetization in the ground state processes at the Larmor frequency. When the redshifted control pulse with σ_+ or σ_- polarization is applied, energy shifts, which increase the optical transition frequency, occur only in the case of energy levels involved in allowed transitions. As a result, when a pulse with the σ_+ (σ_-) circular polarization is applied, splitting between the sublevels is increased (decreased) due to the light-shift effect. Therefore, the precession frequency of magnetization changes during the control pulse, and the precession phase can be controlled by the control pulse with circular polarization. The results obtained from our experiment are well understood by the fact that σ_+ and σ_- polarized beams work as fictitious magnetic fields in opposite directions.

The expression for the light shift is given as [23]

$$\delta = \frac{\chi^2 \Delta}{4(\Delta^2 + \Gamma_2^2)}, \tag{1}$$

where δ denotes the light shift, Δ the detuning of laser frequency from the optical resonance, χ the optical Rabi frequency, and Γ_2 the dephasing of optical coherence. In this experiment, the component of the magnetization along the probe direction is detected, whereas the direction of rotation of the magnetization is not detected. Therefore, the absolute value of Eq. (1) should be compared with the observed precession frequency. Equation (1) explains well the observed precession frequency at the off-resonant frequencies in Fig. 2. However, at resonance, the precession signal disappears because of the absorption effect. Here, Eq. (1) does not hold.

In order to explain the observed detuning-frequency dependence of the precession frequency shown in Fig. 2, we consider a theory of frequency shift that includes the effect of absorption by using the density matrix. In order to examine the effect of the control pulse on the spin dynamics, we consider an ensemble of three-level atoms whose ground state is a degenerated doublet, $|1\rangle$ and $|2\rangle$, and the excited state is $|3\rangle$. After the adiabatic elimination of the optical coherence [23], four differential equations for the population (ρ_{11}, ρ_{22}) and sublevel coherence (ρ_{12}, ρ_{21}) in the ground state are derived, as shown in Eqs. (7) of Ref. [21]. The generation pulse constructs the initial density matrix ρ^0 where $\rho_{11}^0 = 1 - \sigma$, $\rho_{22}^0 = \sigma$, and $\rho_{12}^0 = \rho_{21}^0 = 0$. The magnetization signal $S(t)$ is proportional to $\rho_{11} - \rho_{22}$. When the direction of the control pulse is perpendicular to that of the generation pulse, a simple result is obtained from the differential equations:

$$S(t) \propto (1 - 2\sigma) \exp\left\{-\frac{\chi^2 \Gamma_2 t}{4(\Delta^2 + \Gamma_2^2)}\right\} \cos\left\{\frac{\chi^2 \Delta t}{4(\Delta^2 + \Gamma_2^2)}\right\}. \tag{2}$$

This equation is interpreted as $S(t) = A \exp(-\gamma t) \cos(\delta t)$, where the precession frequency δ is the same as the light-shift frequency in Eq. (1).

The spectral density $S(\omega)$ for the spin precession is obtained from the Fourier transform of $S(t)$:

$$|S(\omega)| = \sqrt{\frac{A^2(\gamma^2 + \omega^2)}{(\gamma^2 + \delta^2 - 2\delta\omega + \omega^2)(\gamma^2 + \delta^2 + 2\delta\omega + \omega^2)}}. \tag{3}$$

The value of ω that yields the spectral peak is derived from equation $d|S(\omega)|/d\omega = 0$ yielding five solutions. We select only real and non-negative solutions of ω. Then, the value of ω for the spectral peak is represented by

$$\omega = \begin{cases} 0 & (|\Delta/\Gamma_2| < a) \\ \frac{\chi^2 \sqrt{|\Delta| \sqrt{\Delta^2 + 4\Gamma_2^2} - \Gamma_2^2}}{4(\Delta^2 + \Gamma_2^2)} & (|\Delta/\Gamma_2| \geq a) \end{cases} \tag{4}$$

where a is a positive constant ($a^2 = \sqrt{5} - 2$).

The solid curve shown in Fig. 2 indicates the theoretical detuning-frequency dependence of the precession frequency ω calculated from Eq. (4), where the fitting parameters are $\chi = 480$ MHz and $\Gamma_2/2\pi = 80$ MHz. The theoretical curve matches with the observed precession frequencies in the whole red-detuning side and in the off-resonant blue-detuning tail. The deviation in the precession frequencies from the theoretical curve around the neighboring absorption line ^{87}Rb ($F = 2 \rightarrow F' = 2$) on the blue-detuning side is considered to be caused by the effect of optical pumping, because the line ^{87}Rb ($F = 2 \rightarrow F' = 1$) excited by the generation pulse and the neighboring line have the same lower level.

The precession frequency δ is proportional to χ^2. Since χ is proportional to the field amplitude of the control pulse, the precession frequency is expected to be proportional to the intensity of the control pulse. The observed linear dependence of the precession frequency on the intensity of the control pulse corresponds to the dependence of δ on χ^2 in Eq. (1).

In conclusion, we studied optical manipulation of spin coherence in rubidium atoms. The generation of fictitious magnetic fields and the frequency control of precession frequency were verified experimentally. We demonstrated absorption-free phase control of spin precession and pure spin rotation. The observed precession frequency shift was explained well by the theory considered by using the density matrix. We showed that it is possible to carry out all-optical manipulation of spins including off-resonant control of spin coherence.

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