PHYSICAL REVIEW A VOLUME 42, NUMBER 3 1 AUGUST 1990

Light-shift-induced spin echoes in a $J = \frac{1}{2}$ atomic ground state

M. Rosatzin, D. Suter, and J. Mlynek*
Institute of Quantum Electronics, Swiss Federal Institute of Technology (ETH), CH-8093 Zürich, Switzerland
(Received 29 January 1990)

We report on a spin-echo phenomenon that is induced between Zeeman sublevels by two laser pulses close to an optical resonance. The echo formation is due to a Zeeman light shift during the second off-resonant pulse; this light pulse creates a fictitious magnetic field that leads to the phase reversal of the spins. Our experimental results on the $^2S_{1/2}$ sodium ground state are in good qualitative agreement with theoretical predictions.

Coherent transient sublevel phenomena such as spin echoes have been widely studied in magnetic-resonance spectroscopy; here pulsed radio-frequency or microwave fields are used to drive the sublevel coherence.[1] It is also well known that transient sublevel coherence can be created by a light pulse in a resonant Raman-type process.[2] In this context, previous studies have demonstrated the occurrence of sublevel or spin echoes in atomic ground states by applying a sequence of two resonant light pulses.[3,4] So far, however, most experimental and theoretical work was restricted to the case of optical broadband excitation and impact excitation of the spin coherence.[3] Due to these limitations, the mechanism of the echo formation in the presence of a near-resonant light field could not be studied in greater detail.

In this Rapid Communication we report results on the optical creation and detection of spin echoes in the $J = \frac{1}{2}$ ground state of atomic Na in the presence of an inhomogeneous transverse magnetic field B (see Fig. 1). We find that this spin echo is due to a *Zeeman light shift* during the second off-resonant optical pulse. In this respect, our present investigations also differ from previous work on Raman echoes[5] where light-induced level shifts have not been considered so far. Our experimental results are well described by a theory that is valid for arbitrary pulse lengths and that includes the dependence on the optical-resonance detuning.

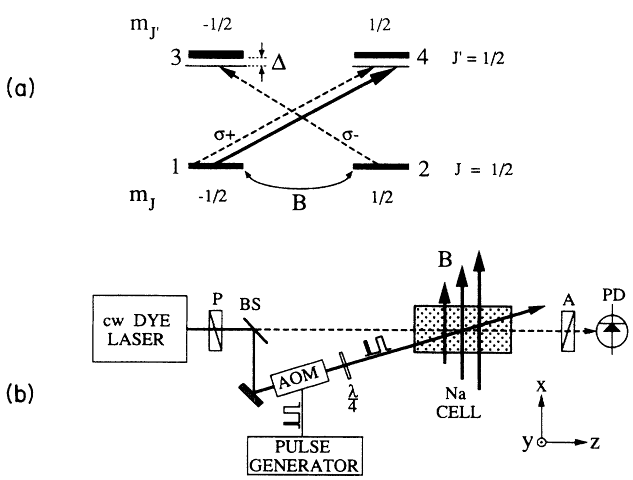

FIG. 1. (a) Schematic representation of the $J = \frac{1}{2} \rightarrow J' = \frac{1}{2}$ system coupled to the optical fields. With the quantization axis parallel to the laser beam a transverse magnetic field B induces transitions between the Zeeman substates. (b) Schematic experimental setup. BS, beam splitter; AOM, acousto-optic modulator; $\lambda/4$, retardation plate; P, polarizer; A, analyzer; PD, photodiode; and B, inhomogeneous magnetic field.

Let us start with a simple qualitative explanation of the phenomenon. We consider the atomic-model system depicted in Fig. 1(a); here the ground and excited state are both $J = \frac{1}{2}$ states. We have chosen the propagation direction of the laser beam as the quantization axis. For simplicity, we will neglect excited-state effects. The first circularly polarized light pulse (solid line) induces a population difference between the two magnetic substates of the ground state corresponding to a magnetization along the laser-beam axis. In the presence of a static transverse magnetic field, this magnetization precesses around the field direction creating coherence between the Zeeman substates. If the transverse magnetic field B is inhomogeneous, the observable spin coherence decays rapidly after the pulse. However, this inhomogeneous decay can be reversed by a second circularly polarized optical pulse: If this pulse excites the atoms off-resonance ($\Delta \neq 0$), a Zeeman light shift occurs. This light shift corresponds to a *fictitious magnetic field* in laser-beam direction,[6] i.e., perpendicular to the external field B. It introduces the required phase reversal and is thus a prerequisite for the refocusing of the spin packets that gives rise to a spin echo. The transient spin polarization in the ground state leads to different absorption and dispersion for left- and right-circularly polarized light and can therefore be detected by a linearly polarized probe beam (dashed line).[7]

Our experimental setup is shown schematically in Fig. 1(b); it has been described in detail before.[4,8] The measurements were performed on Zeeman sublevels of the $3s\,^2S_{1/2}$ sodium ground state; the D_1 line ($\lambda = 589.6$ nm) was used for optical excitation. The circularly polarized light pulses were derived from a single-mode-ring dye laser (short-term linewidth $\lesssim 500$ kHz) by an acousto-optic modulator (AOM). The transients induced by the pulses were observed with a weak linearly polarized optical probe beam in combination with polarization selective detection. The diameter of both beams within the sample was about 0.5 mm and the angle between the two beams was less than 1°. After transmission through the polarization analyzer A, the probe beam was detected on a fast p-i-n photodiode (PD).

The sodium vapor was contained in an 18-mm-diam ceramic tube; the length of the heated zone was 60 mm and sodium number densities of about 10^{10} cm^{-3} were used. Argon buffer gas was added to the sodium vapor at a pressure of 210 hPa; the small signal absorption at the optical line center was typically 30%. The measured width of the pressure-broadened D_1 line was $\Gamma/2\pi = 2.1$ GHz [half-width at half maximum (HWHM)] and thus masked the hyperfine splitting (1.8 GHz) as well as the Doppler broadening (0.85-GHz HWHM). Under these experimental conditions, excited-state effects can be neglected and the dynamic response of the sample to the laser pulses is essentially determined by the ground state,[8,9] which decays on a time scale of some 100 μs due to diffusive motion of the atoms out of the laser beams. Thus, to a first approximation, we can model the D_1 line as a homogeneously broadened optical $J=\frac{1}{2}$ to $J'=\frac{1}{2}$ transition as shown in Fig. 1(a).

Three pairs of Helmholtz coils were used to apply a transverse magnetic field in the x direction and to compensate for the earth magnetic field. In addition, two parallel straight wires in the y direction provided a well-defined magnetic-field inhomogeneity in the test region with the field gradient along the optical beam axis. The mean value of the B field was typically 25 μT; the full width of the distribution of B values was about 5 μT. Depending on the B-field values, the corresponding distribution of Larmor frequencies Ω_L of the Na ground state is determined by $\Omega_L = 2\pi \times 7.0 \times B$ MHz/mT.

The optical detuning Δ from the line center was measured with a Na reference cell (without buffer gas) and a scanning Fabry-Pérot was used to calibrate the laser-frequency scan range; positive Δ corresponds to blue detuning. In this way the optical detunings could be determined with an accuracy of less than $2\pi \times 100$ MHz. With the present setup the optical detuning of the two pump pulses are identical.[10] The intensity of the probe beam (<200 mW/cm^2) was low enough to avoid any perturbation of the probe beam on the polarization of the sample.

A typical experimental result of the observed spin transients is shown in Fig. 2 together with the optical-pulse sequence. Here differential dispersion is measured with an angle of 45° between polarizer and analyzer. A first pulse is used to create a steady-state spin polarization.[8] After the end of the pulse a free-induction-decay (FID) signal occurs; its decay time is related to the inhomogeneous broadening of the spin transition. A second optical pulse of duration $t_2 = 0.7$ μs is applied a time $T = 100$ μs later, giving rise to another FID. Finally, at a time T after the second pulse, a pronounced spin-echo signal appears.

The measured echo signal as a function of the normalized optical detuning $\bar{\Delta} = \Delta/\Gamma$ is displayed in Fig. 3. Here crossed polarizers were used to ensure detection sensitivity on the optical line center. Our experimental results clearly demonstrate that the spin echo *disappears* on optical resonance ($\Delta = 0$). With increasing detuning the echo intensity reaches a maximum on both sides of the resonance and then rapidly drops off for detunings Δ large compared to the optical linewidth Γ.

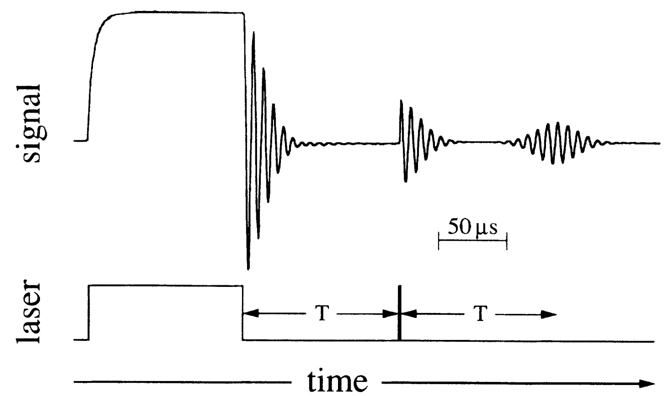

FIG. 2. Typical experimental spin-echo signal as a function of time. The experimental parameters are $\Omega_L = 2\pi \times 142$ kHz, $\Delta = 2\pi \times 13.5$ GHz, pump-pulse power = 210 mW, and $t_2 = 0.7$ μs. The lower trace represents the laser-pulse sequence.

For a quantitative description of the experiment the corresponding Maxwell-Bloch equations for our $J=\frac{1}{2} \rightarrow J'=\frac{1}{2}$ atomic-model system have to be solved. As has been shown earlier, the dynamic response of the atoms to a light field can be described by a simple Bloch-type equation for the ground-state magnetization $\mathbf{m} = (m_x, m_y, m_z)$ (Ref. 9):

$$\dot{\mathbf{m}} = \mathbf{\Omega} \times \mathbf{m} - \gamma_{\text{eff}} \mathbf{m} + \mathbf{P}, \tag{1}$$

with

$$\mathbf{\Omega} = (\Omega_L, 0, \bar{\Delta} P_+), \quad \mathbf{P} = (0, 0, P_+),$$

$$\gamma_{\text{eff}} = \gamma + P_+, \quad P_+ = \frac{|\beta_+|^2}{\Gamma(1+\bar{\Delta}^2)}.$$

These equations have been discussed in detail before:[8,9] P_+ denotes a pump rate induced by the σ^+ light and β_+ is the corresponding Rabi frequency. Note that for off-resonant excitation ($\bar{\Delta} \neq 0$), the precession vector $\mathbf{\Omega}$ is not

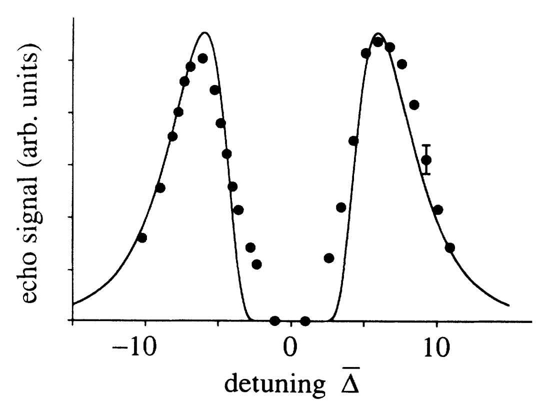

FIG. 3. Measured echo signal as a function of the optical detuning $\bar{\Delta} = \Delta/\Gamma$ together with a least-squares fit (solid line) of our theory. The experimental parameters are $\Omega_L = 2\pi \times 142$ kHz, pump-pulse power = 160 mW, $t_1 = 100$ μs, and $t_2 = 1.0$ μs.

parallel to the transverse magnetic field but has an additional light-shift component $\bar{\Delta}P_+$ in the direction of the laser beam. With our detection scheme (Fig. 1), we only monitor the component m_z of $\mathbf{m}(t)$, i.e., the population difference between the ground-state sublevels.[8]

We used Eq. (1) to calculate the time evolution of $m_z(t)$ during a double pulse sequence. The result was integrated over an inhomogeneous magnetic field. The maximum of the resulting echo envelope at time $t=T$ after the second pulse is then given by

$$\bar{m}_z(t=T)=m_{z1}\sin^2\eta_2\sin^2(\Theta_2/2)\times\exp(-\gamma_{\text{eff}}t_2)\exp(-\gamma 2T)\,, \qquad (2)$$

with

$$\eta_2=\arctan(\bar{\Delta}P_+/\Omega_L),\;\; \Theta_2=t_2\Omega=t_2[\Omega_L^2+(\bar{\Delta}P_+)^2]^{1/2}.$$

m_{z1} describes the spin polarization created by the first laser pulse (for details see Ref. 8). η_2 is the tilt angle between the effective field $\mathbf{\Omega}$ during the second laser pulse and the direction of the transverse magnetic field. Θ_2 can be interpreted as a pulse area and describes the flip angle of the spins during the second pulse of length t_2. The last two terms in Eq. (2) are damping terms describing the phase relaxation after the end of the first pulse. During the second pulse the relaxation is determined by γ_{eff}, which accounts for destruction of coherence by optical pumping.

On the basis of Eq. (2), the spin-echo phenomenon can easily be interpreted: The spin polarization m_{z1} created by the first laser pulse dephases rapidly since the spin transition is inhomogeneously broadened and the average polarization of the spin ensemble vanishes. However, a partial rephasing can be induced by a rotation of the spins around any axis that is not parallel to the direction of the transverse magnetic field. The light-shift component $\bar{\Delta}P_+$ acts as an additional longitudinal magnetic field and tilts the rotation axis away from the direction of the external magnetic field. Therefore, an *off-resonant* second laser pulse ($\eta_2\neq 0$) can refocus the spin packet leading to a spin-echo signal; on resonance ($\eta_2=0$), the echo signal vanishes.

A least-squares fit of the measured echo signal as a function of the normalized optical detuning is shown in Fig. 3 (solid line). As free parameters we only adjusted the conversion factor of pump-pulse intensity to pump rate P_+ and the overall scaling factor which accounts for the detector efficiency. We note that the maxima of the echo signal in Fig. 3 are not at $\bar{\Delta}=\pm 1$ where the light shift is largest: At high intensities of the second pulse the echo signal near resonance is strongly decreased due to optical pumping [see Eq. (2)] and thus the echo maxima are shifted farther away from resonance. The small deviation of the measured points from the theoretical curve very close to resonance is due to the Gaussian laser-beam profile: For small laser intensities the optical pumping of the second pulse is negligible; as a consequence, nonvanishing echo signals also occur close to resonance. Apart from these small discrepancies, our experimental results are in good quantitative agreement with our theory. We therefore interpret the behavior of the echo signal near resonance as evidence for a *light-shift-induced* phenomenon.

As another test of our theory, we also studied the echo amplitude as a function of the duration t_2 of the second optical pulse, keeping the length t_1 of the first pulse constant. We observe a damped oscillatory behavior in agreement with Eq. (2): If Θ_2 is an odd multiple of π, the rephasing of the spin ensemble is efficient leading to a large echo amplitude. Moreover, we observed additional weaker echoes at times $2T$ and $3T$ after the second pulse; however, their formation mechanism is not yet understood.

In conclusion, we have reported results on optically induced spin echoes in the $3s\,^2S_{1/2}$ ground state of atomic sodium. We could clearly assign the observed echo to a *light-shift effect* during the second light pulse. From a more general point of view, our experiments represent another demonstration for the manipulation of atomic and molecular multipole moments by light-induced fictitious fields. In contrast to previous work,[11,12] the light shift is used to refocus spins of an inhomogeneously broadened sublevel transition. Let us point out that the occurrence of light-shift-induced spin echoes is neither restricted to homogeneously broadened optical transitions nor to ground-state sublevels as discussed here. These echoes should also be present for inhomogeneously (e.g., Doppler) broadened optical lines as well as for sublevels of excited electronic states.

We gratefully acknowledge financial support by the Schweizerische Nationalfonds.

*Present address: Fakultät für Physik, Universität Konstanz, D-7750 Konstanz, Federal Republic of Germany.

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