Optically Induced Coherence Transfer Echoes between Zeeman Substates

Dieter Suter and Martin Rosatzin

Institute of Quantum Electronics, Swiss Federal Institute of Technology (ETH) Zürich, CH-8093 Zürich, Switzerland

Jürgen Mlynek

Department of Physics, Universität Konstanz, D-7750 Konstanz, Germany

(Received 2 January 1991)

We report on optically induced spin-echo phenomena associated with Zeeman coherences in a sublevel manifold. The experiments on the ground state of sodium show multiple echoes occurring periodically after a double-pulse excitation sequence in an inhomogeneous magnetic field. The echo formation mechanism is attributed to a light-shift-induced transfer of sublevel coherence among the nondegenerate substates. This mechanism is verified experimentally, using a modulated excitation scheme with subsequent phase-sensitive detection.

PACS numbers: 32.60.+i, 32.80.−t, 42.50.Md

Echo phenomena are some of the most prominent features in coherent spectroscopy. After the first report on spin echoes in nuclear magnetic resonance by Hahn [1], this phenomenon has found numerous applications [2]. Echo phenomena were also observed in various other fields like electron spin resonance, microwave, and optical spectroscopy. The typical experimental procedure for an echo experiment uses a short pulse of intense radiation to create a coherent superposition between two or more eigenstates of the Hamiltonian. After the end of the pulse, the coherence is allowed to precess freely under the influence of an *inhomogeneous interaction* such as an inhomogeneous magnetic field, thereby destroying the overall phase coherence of the ensemble. A second pulse inverts the phase of the individual spins, so that the second free-precession period leads to a cancellation of the total phase, independent of the field strength experienced by the individual spins. As a result, the magnetic moments come back into phase and an echo is observed.

Spin-echo phenomena can be induced not only by radio-frequency pulses but also by a sequence of two pulses of polarized light [3,4]. As we have shown [4], fictitious magnetic fields due to the light-shift effect are responsible for the echo formation in this case. In the same context we noted that the double-pulse excitation sequence leads not only to a single echo, but to a sequence of echoes occurring at integer multiples of the pulse-separation time. This behavior is in obvious contradiction with the simple picture of echo formation described above and was not understood at that time. In this Letter we propose a mechanism for the echo formation and present additional experimental evidence supporting this hypothesis.

Our experimental setup is shown schematically in Fig. 1. The measurements were performed on the Zeeman sublevels of the $3s\,^2S_{1/2}$ ground state of atomic sodium. The beam of a single-mode cw ring dye laser was split into a circularly polarized pump beam (solid line in Fig. 1, intensity ~ 100 mW/mm^2) and a linearly polarized probe beam (dashed line, intensity ~ 100 μW/mm^2). The laser frequency was set near the Na D_1 line ($\lambda = 589.6$ nm) with a typical detuning $\Delta/2\pi \approx 5$ GHz. The pulses were generated with an acousto-optic modulator. Polarization-selective detection of the transmitted probe beam yielded a signal proportional to the magnetization component in the direction of the laser beam. Argon buffer gas was added to the sodium vapor, leading to a pressure broadening of the optical resonance of $\Gamma/2\pi = 4.2$ GHz (FWHM). Under these experimental conditions, excited-state effects can be neglected and the signal is determined by the ground-state coherences whose effective lifetime is determined by the diffusive motion of the atoms out of the laser beams with a half-life of ~ 100 μs. A magnetic field was applied perpendicular to the laser beam with a gradient parallel to the beam.

A typical example of the observed spin transients is shown in Fig. 2 together with the optical excitation sequence; for this experiment, pulses with constant amplitude were used. The experimental data show that the double-pulse excitation sequence excites a series of three

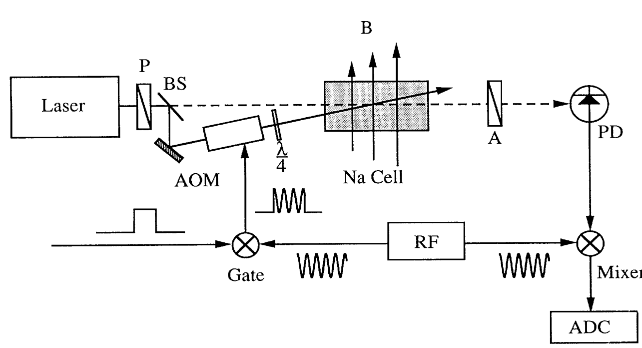

FIG. 1. Schematic experimental setup. P, polarizer; BS, beam splitter; λ/4, retardation plate; B, magnetic field; A, analyzer; AOM, acousto-optic modulator; PD, photodiode; RF, radio-frequency generator; and ADC, analog-digital converter.

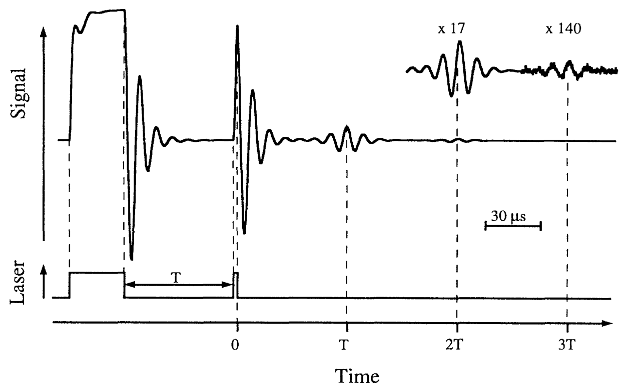

FIG. 2. Top, typical experimental signal measured at an optical detuning of $\Delta/2\pi = 5.5$ GHz with a laser power of 80 mW, duration of the refocusing pulse $= 1.6$ μs, pulse separation $T = 60$ μs, and Larmor frequency $\Omega_L/2\pi = 110$ kHz. The second and third echoes are expanded vertically. Bottom, optical pulse sequence.

well-resolved echoes occurring periodically at multiples of the pulse-separation time T after the second pulse. These multiple echoes persist under a wide range of experimental conditions and show a dependence on the experimental parameters such as laser intensity, laser detuning, pulse width, and magnetic-field strength that is similar to that of the first echo.

These multiple echoes are clearly incompatible with the simple echo formation mechanism given above. A possible resolution of this apparent paradox is the fact that this echo formation mechanism is strictly valid only for a spin-$\frac{1}{2}$ or other two-level system. While most experiments on optically induced spin transients can be explained by modeling the atoms as $J=\frac{1}{2}$ systems, this approximation apparently fails here. A more complete analysis therefore has to take the nuclear spin $I=\frac{3}{2}$ of Na into account. Its most important effect, in this context, is the increase in the number of available magnetic substates. While multiple-echo effects cannot occur in simple spin-$\frac{1}{2}$ systems [5], they have been observed in more complicated level systems [6]. Basically, the occurrence of multiple echoes in such systems is possible whenever the refocusing pulse transfers coherence between transitions whose Bohr frequencies have a different dependence on the inhomogeneous interaction, so that the refocusing time differs from the defocusing time. The occurrence of multiple echoes discussed here is, to the best of our knowledge, the first demonstration that such a transfer of coherence between different sublevel transitions can be initiated by the light-shift effect of a laser pulse.

For a more detailed analysis of the formation mechanism of coherence transfer echoes, we expand the density operator ρ of the system with total angular momentum F in terms of irreducible tensor operators ρ_m^l. The initial pulse prepares the system in a state [7]

$$\rho(0) = \sum_{l,m} c_m^l \rho_m^l , \tag{1}$$

with $l = 0, \ldots, 2F$ representing the rank of the tensor, $m = -l, \ldots, l$ the magnetic quantum number, and c_m^l the expansion coefficients. After the end of the preparation pulse, the system evolves freely in a magnetic field $\mathbf{B} = (0,0,B)$. We assume that the magnetic field is small enough, so that the effective Hamiltonian of the system contains only the linear Zeeman effect. The time evolution of the individual tensor components is then given by

$$\rho_m^l(t) = e^{-iI_z\Omega_L t}\rho_m^l(0)e^{+iI_z\Omega_L t} = e^{im\Omega_L t}\rho_m^l(0) , \tag{2}$$

where $\Omega_L = \mu_B g B/\hbar$ is the Larmor frequency of a spin $\frac{1}{2}$, I_z the z component of the angular momentum operator, g the Landé factor, and μ_B the Bohr magneton. The second equation follows directly from the tensor properties of the operators ρ_m^l [7]. Since ρ_m^l precesses with an angular frequency $m\Omega_L$, it is generally referred to as m-quantum coherence [2]. Neglecting any relaxation mechanisms, we write the density operator just before the second pulse as

$$\rho(T_-) = \sum_{l,m} c_m^l \rho_m^l e^{im\Omega_L T} , \tag{3}$$

where T is the pulse-separation time. As described elsewhere [4], the second pulse changes the evolution of the system via light shift and optical pumping. Neither effect preserves the rotational symmetry of the unperturbed Hamiltonian, so that the evolution during the pulse is not diagonal in the operator basis chosen here. We summarize the effect of the second pulse in a matrix η, so that the density operator after the pulse becomes

$$\rho(T_+) = \sum_{l',m'} c_{m'}^{l'}\rho_{m'}^{l'} = \sum_{l',m'}\sum_{l,m} \eta_{l'm'lm} c_m^l e^{im\Omega_L T}\rho_{m'}^{l'} . \tag{4}$$

After the second pulse, the system precesses freely again, so that at a time t after the second pulse, the density operator becomes

$$\rho(t) = \sum_{l',m'}\sum_{l,m} \eta_{l'm'lm} c_m^l \rho_{m'}^{l'} \exp[i\Omega_L(mT + m't)] . \tag{5}$$

The effect of the inhomogeneous magnetic field is a variation of the magnetic-field strength and therefore of the phase factors in Eq. (5). If the inhomogeneity $\Delta\Omega_L$ is large enough ($\Delta\Omega_L > 1/T$), the overall coherence vanishes unless the phase factor is identically zero. Echoes may therefore occur at $t = -Tm/m'$. With our detection geometry, the signal is proportional to the component of the magnetization parallel to the direction of the laser beam which transforms as a vector, so that m' is restricted to ± 1 and echoes are expected at $t = nT$ ($n = 0, \ldots, l$).

On the basis of this argument, the experimental results of Fig. 2 can be interpreted as follows: The simultaneous action of the laser field and the magnetic field during the first laser pulse creates m-quantum coherences with $-4 \leq m \leq 4$. After the end of the pulse, the coherences precess in the transverse magnetic field, each at its characteristic precession frequency $m\Omega_L$. The time evolution of the 1-quantum coherence leads to a free-induction-decay (FID) signal. The second pulse causes a redistribution of these coherences which subsequently evolve with a different frequency. The first "echo" is predicted to occur at $t=0$, right after the second pulse, and is therefore observed as a second free-induction decay. Also, the echoes with $n=1,\ldots,3$ are clearly seen in the experimental data of Fig. 2, while the fourth echo which is theoretically possible in the Na ground state was apparently too weak to be seen.

In order to confirm this interpretation of the multiple echoes, we developed an alternative excitation scheme which allows a distinction of different orders of coherence. As discussed in detail elsewhere [8], this scheme uses an excitation pulse whose intensity is modulated as $I(t)=I_0\sin(\omega t-\phi)$, where I_0 represents the peak intensity and the modulation frequency ω is set near the Larmor frequency of the system. This modulation introduces additional degrees of freedom into the experiment; the phase ϕ of the modulation, e.g., can be used as a label for the coherence created by the excitation pulse. In effect, the state of the system at the end of the pulse is modified by a rotation around the direction of the magnetic field, so that the initial density operator is now prepared into a state

$$\rho(0,\phi)=e^{-i\phi I_z}\rho(0,0)e^{i\phi I_z}=\sum_{l,m}c^l_m\rho^l_m e^{im\phi}. \tag{6}$$

It is therefore possible to label m-quantum coherences by phase factors proportional to m. As a scalar, this phase factor is not affected by the subsequent evolution, and can be used to track the coherence through the experiment. By systematic variation of the excitation phase ϕ, it is then possible to separate the different tensor components of the coherence, e.g., via Fourier analysis. For the experimental implementation of this idea, we derived the sinusoidal modulation from a gated function generator which allowed the selection of an arbitrary phase at the beginning of each pulse. The modulation phase of the first pulse was incremented systematically while the phase of the second pulse was kept constant. The resulting phases of both FIDs and of the first and the second echo are summarized in Fig. 3. The relatively small third echo was not observed in this experiment to reduce accumulation time.

Since our detection system is sensitive to a magnetization component, the only coherences that contribute to the observable signal are the tensor components $\rho^l_1+\rho^l_{-1}$. According to Eq. (6), the signal during and after the preparation pulse should therefore show directly the excitation phase ϕ, as found experimentally. The spin echoes occurring at times nT ($n=0,\ldots,l$) originate from coherence that was initially prepared as tensor component $\rho^l_{\pm n}$. Since an echo is formed only if the component is converted into ρ^l_{-1}, the signal during the nth echo should appear with a phase $-n\phi$. This is clearly corroborated by the experimental data, where the second FID corresponds to the echo with $n=0$ and the phases of the first and second echoes are directly proportional to the preparation phase, with the proportionality constants -1 and -2, respectively. The experimental result is therefore consistent with the assumption that these multiple echoes are due to multiple-quantum coherence generated during the first pulse and transferred into observable single-quantum coherence by the second pulse.

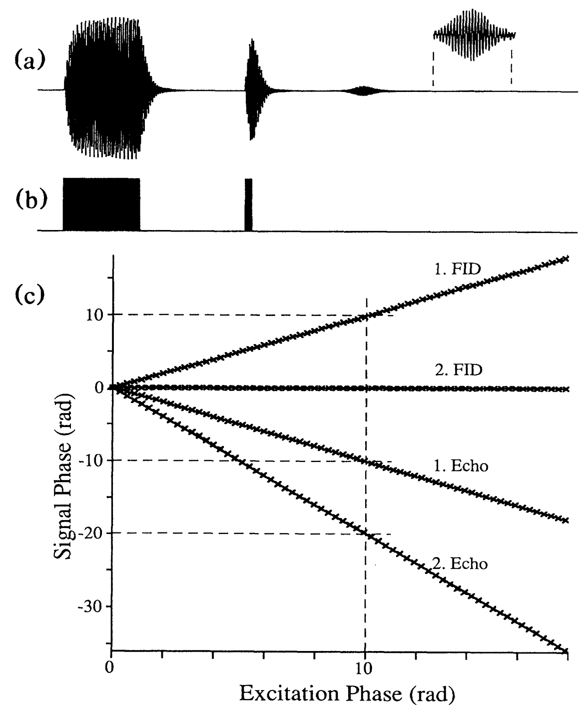

FIG. 3. (a) Typical experimental signal measured with intensity-modulated pulses. (b) Corresponding pump laser intensity. (c) Phases of the various signal components vs excitation phase; crosses represent experimental points, solid lines show the predicted behavior, and dashed lines indicate the slope of the different curves.

In addition to the experiments described here, we have also observed multiple echoes in the $^7F_1(4f^66s^2)$ state of Sm, using the $J=1 \rightarrow J'=0$ transition at $\lambda=570.675$ nm for optical excitation [9]. Since $F=J=1$, we expect coherence transfer echoes at nT with $n=0,1,2$. This was confirmed experimentally; a third echo could not be found in this case.

The process of optically induced coherence transfer between atomic sublevel transitions is a prime demonstration that the light-shift effect modifies not only the eigenvalues, but also the eigenvectors of the effective Hamiltonian. The experimental observation that the echoes vanish at the optical resonance [4] is a clear indication that the light shift is a necessary prerequisite for the occurrence of optically induced spin echoes. However, the details of the coherence transfer process, which we summarized in the matrix $\eta_{l'm'lm}$, are not yet understood. It depends not only on the light-shift effect but also on the optical pumping. It is remarkable that this process, which is usually considered a purely dissipative process, can actually be utilized for the transfer of coherence between different tensor components.

The proposed mechanism for the observed multiple echoes is in agreement with the extended form of the "theorem on coherent transients" by Schenzle, Wong, and Brewer [5]. It states that, in the case of Na, the signal should be restricted to $4T$ which, for an infinitely broadened spin transition, is obviously fulfilled. While there is a certain analogy between the echoes reported here and echo effects in magnetic resonance [6] and optics [10], the physical mechanisms driving the processes are radically different. The main difference is that in the case of the "trilevel echoes," the coherences are created and observed in optical transitions that are directly coupled to the radiation field. In our experiment, in contrast, the dephasing and rephasing of the coherences occur in sublevel transitions that do not interact with the laser beam. Since the optical pulses do not couple to the coherences, the refocusing process can be initiated only indirectly, via optical pumping and light-shift effects.

In recent years the relaxation of multipoles has been studied using techniques [11]. The coherence transfer echoes discussed here may well prove advantageous for such studies since the creation and relaxation of multipoles can be investigated directly: The multiple echoes lead to a separation of different components in the time domain. In addition, the new experimental technique introduced here can be used in general to distinguish between higher-order multipole components via their dependence on the phase of the modulated excitation.

In conclusion, we have reported the first observation of optically induced coherence transfer echoes in atomic substates. The experiments in the ground state of Na show multiple echoes occurring periodically after the optical double-pulse excitation sequence. The echo formation mechanism could be attributed to a transfer of coherence between different orders of Zeeman coherence by the second pulse. This new echo phenomenon may prove useful for the study of relaxation processes of atomic multipoles. In addition, optically induced coherence transfer processes may well become a useful tool for magnetic resonance spectroscopy in cases where the high sensitivity of optical methods is important. With regard to these potential applications, the phenomenon deserves further investigation, especially into the details of the mechanism of optically induced coherence transfer in multilevel systems.

This work was supported by the Schweizerischer Nationalfonds.

[1] E. L. Hahn, Phys. Rev. **80** 580 (1950).

[2] See, e.g., R. R. Ernst, G. Bodenhausen, and A. Wokaun, *Principles of Nuclear Magnetic Resonance in One and Two Dimensions* (Oxford Univ. Press, Oxford, 1987), and references therein.

[3] Y. Fukuda, K. Yamada, and T. Hashi, Opt. Commun. **44**, 297 (1983).

[4] M. Rosatzin, D. Suter, and J. Mlynek, Phys. Rev. A **42**, 1839 (1990).

[5] A. Schenzle, N. C. Wong, and R. G. Brewer, Phys. Rev. A **24**, 2250 (1981).

[6] I. Solomon, Phys. Rev. **110**, 61 (1958); A. A. Maudsley, A. Wokaun, and R. R. Ernst, Chem. Phys. Lett. **55**, 9 (1978).

[7] See, e.g., A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton Univ. Press, Princeton, 1974).

[8] D. Suter and J. Mlynek, Phys. Rev. A **43**, 6124 (1991).

[9] M. Rosatzin, Ph.D. thesis, ETH Zürich, 1990 (unpublished).

[10] See, e.g., T. Mossberg, A. Flusberg, R. Kachru, and S. R. Hartmann, Phys. Rev. Lett. **39**, 1523 (1977).

[11] A. P. Ghosh, C. D. Nabors, M. A. Attili, and J. E. Thomas, Phys. Rev. Lett. **54**, 1794 (1985); R. J. McLean, P. Hannaford, and R. M. Lowe, Phys. Rev. A **42**, 6616 (1990).