OPTICALLY INDUCED COHERENT SPIN TRANSIENTS IN AN ATOMIC GROUND STATE

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It is well known that transient coherence between atomic sublevels can be created by a resonant light pulse in a Raman-type process. However, up to now most experimental and theoretical work on these optically induced sublevel coherences or spin transients was restricted to the case of optical broadband excitation and impact excitation of the sublevel coherence[1].

In this contribution, we report on new studies of the optical creation and detection of spin nutation, free induction decay and spin echoes in an atomic ground state. Optically resonant laser pulses are used to excite the spin coherence and the spin transients are observed with a cw optical probe beam using polarization selective detection. Our present studies include the limit of long pulse excitation and also allow for the investigation of saturation phenomena and dispersive effects; as a result, novel signal features are observed that have remained elusive so far[2].

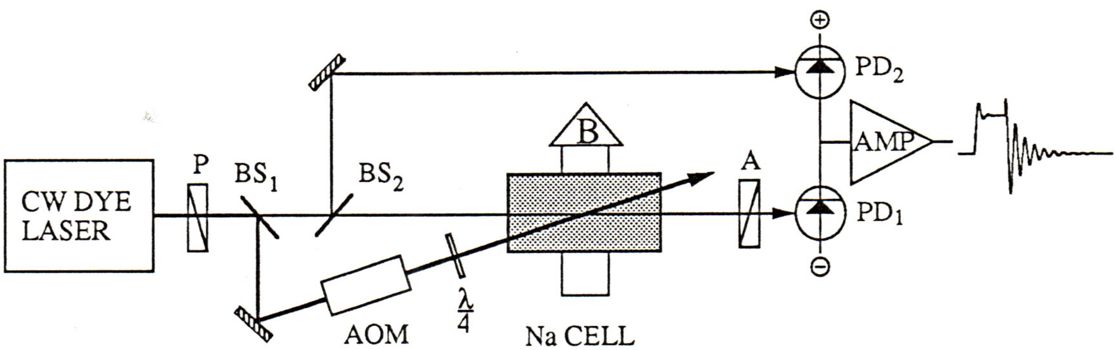

Fig. 1a): Schematic representation of our experimental setup. P=polarizer, BS=beam splitter, A=analyzer, PD=photodiode, B=static magnetic field.

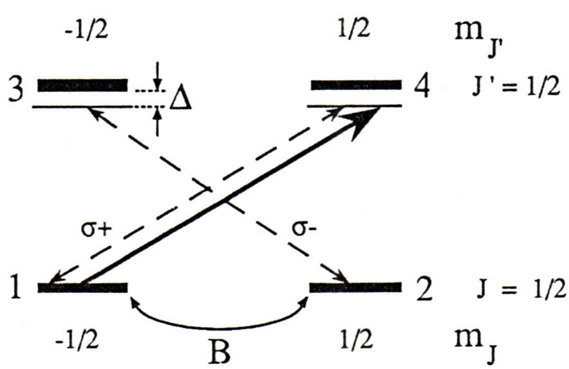

Fig. 1b): Simplified energy level scheme for the Na ground state. B=static magnetic field, Δ=optical resonance offset.

Our experiments were performed on Zeeman sublevels of the $^2S_{1/2}$ ground state of atomic sodium in an argon atmosphere[2]. The experimental setup together with the simplified level scheme of Na is shown in Fig. 1; we note that the quantization axis is chosen along the direction of laser beam propagation. The D_1 line was used for optical excitation with circularly polarized pulses which were derived from a cw single mode dye laser via an acoustooptic modulator; this allows us to vary the pulse length, pulse shape and pulse sequence. Under our experimental conditions the population differences and sublevel coherences in the $^2P_{1/2}$ excited state are rapidly destroyed by collisions; the full width of the pressure broadened optical line is 4.2 GHz. Consequently, the dynamic response of the sample to the pump pulses is essentially governed by the $^2S_{1/2}$ ground state[3]; here the static transverse magnetic field B makes the spins, or the magnetization, precess around the B-field direction. Thus the magnetic field induces Zeeman coherence and drives transitions among Zeeman levels thereby redistributing the optically pumped sublevel populations until a stationary state is reached. A cw weak optical probe beam in combination with polarization selective detection is used to monitor the spin dynamics. Here various detection geometries are possible; in our experiments, either differences in the absorption[4] or in the dispersion of the two circularly polarized probe beam components were measured[2]. In this case, the detector signal directly reflects the time evolution of the ground state magnetization component parallel to the optical axis.

A typical result for the dynamic response of the sample to an optical pump pulse is shown in Fig. 2 together with the excitation pulse. Clearly three different signal regions can be distinguished. The initial transient displays an oscillatory behaviour that can be interpreted as an optically driven spin nutation. The damping of this oscillation strongly depends on the pump pulse intensity. For a nonzero optical detuning ($\Delta \neq 0$) this spin nutation shows pronounced light shift contributions: Here the off-resonant laser pulse induces in the ground state a dc magnetization that is oriented parallel to the optical axis, i.e. perpendicular to the static transverse B-field. As a consequence, for $\Delta = 0$, the spin nutation frequency always exceeds the Larmor frequency and also depends on pump pulse intensity[4].

After the initial transient is damped out, a stationary state is reached (see Fig. 2) corresponding to an optically pumped time-independent spin polarization in the Na ground state. This situation represents the limit of long pulse excitation as the pump pulse clearly exceeds the duration of the initial transient. Finally, when the pulse is switched off, a free induction decay (FID) signal is observed with the precession frequency being determined by the Larmor frequency. Our experimental results clearly show that not only the amplitude but also the phase of the FID strongly depend on pulse intensity; in Fig. 3 the measured dependence of the FID amplitude and phase on the pump pulse power is displayed for various values of the ground state Larmor frequency.

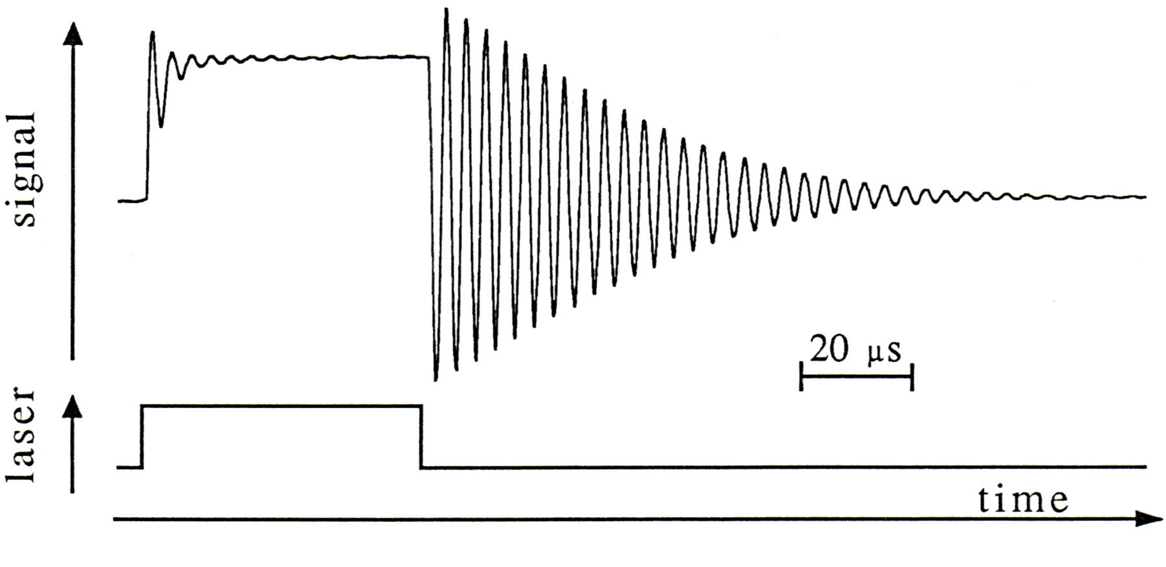

Fig. 2): The upper trace shows the differential absorption of the Na sample which is optically punped by a circularly polarized laser pulse. The lower trace shows the intensity of the laser field.

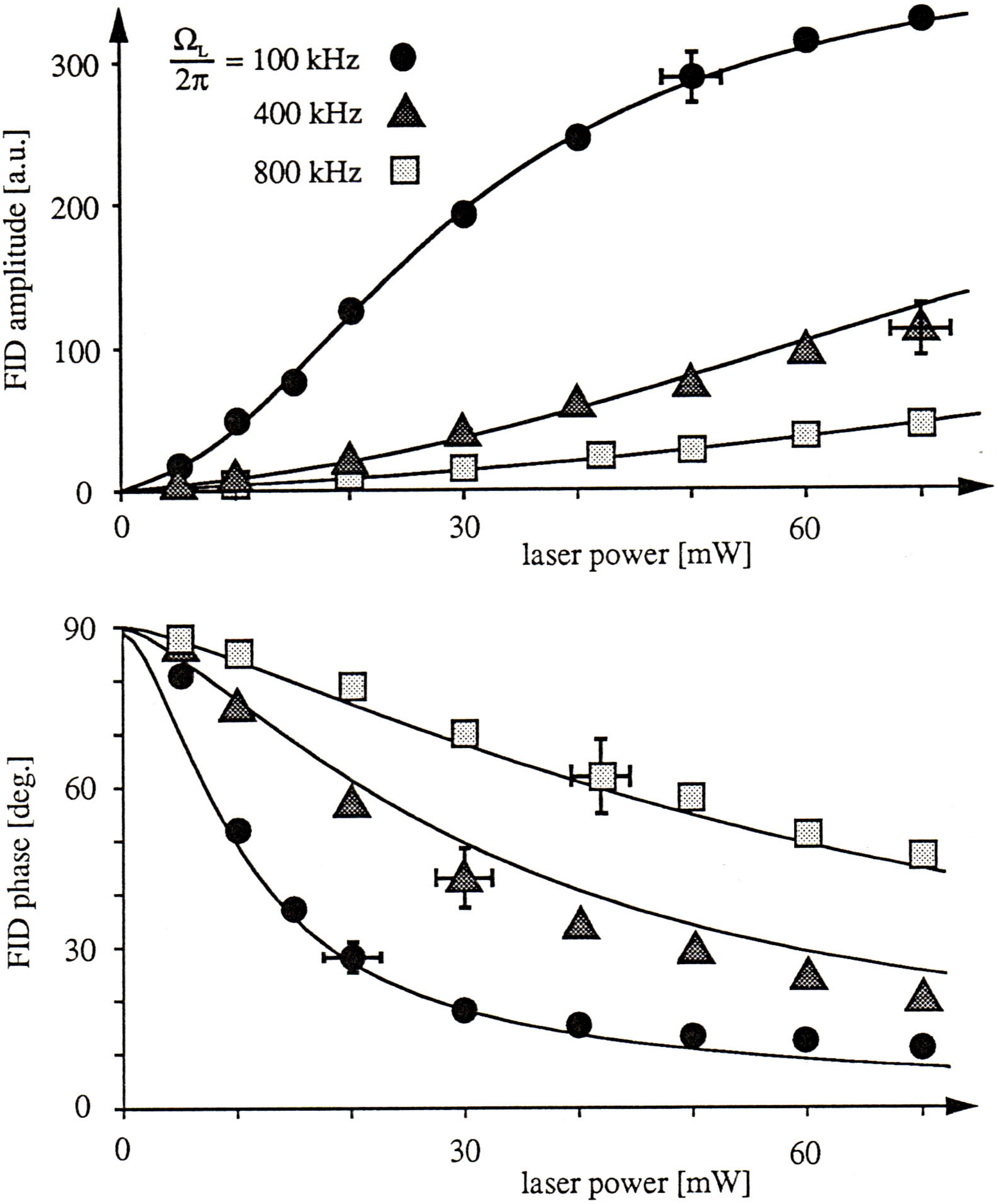

Fig. 3): Amplitude (top) and phase (bottom) of the optically induced FID as a function of laser intensity, for different Larmor frequencies. The experimental results are shown together with a least squares fit to the theoretically predicted behaviour.

The measured FIDs are related to previous theoretical studies on laser saturation effects in quantum beats from an excited atomic state[5] where a regeneration of oscillating sublevel coherence through saturation was predicted in the limit of long excitation pulses; no observation of this phenomenon was reported so far, probably because long and high power laser pulses are needed to observe this effect in short-lived excited atomic states. In contrast, our studies on the saturation behaviour of spin coherence in a long-lived atomic ground state are much more favorable with respect to laser power requirements. In fact, the saturation intensity for power broadening of the sublevel coherence in the ground state is decreased roughly by the factor τ/T_1 with respect to similar effects in optically excited atomic states; here T_1 is the excited state lifetime and t is the ground state relaxation time which, in our situation, is limited by atomic diffusion processes through the laser beam ($\tau \cong 100$ msec). Here we just note that the nonlinear increase of the FID amplitude with pump pulse intensity (see Fig. 3) can be interpreted as a regeneration of sublevel coherence through intense light fields under steady-state conditions ("long pulse limit")[6].

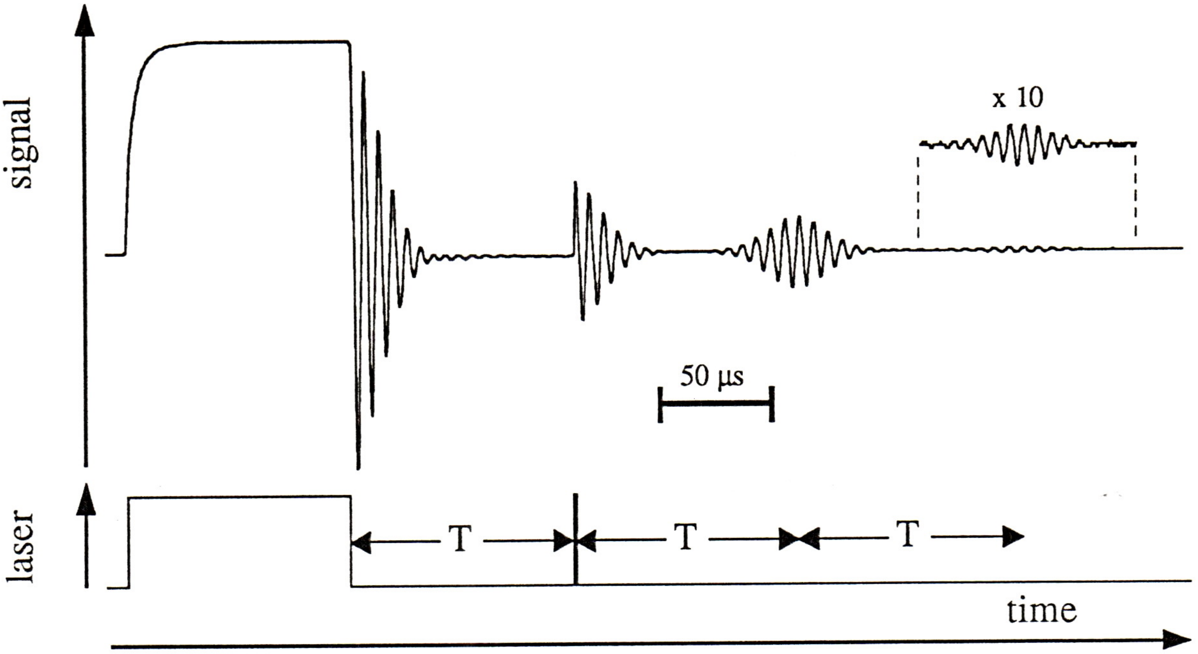

Fig. 4: Differential dispersion signal (top) for a sequence of two optically off-resonant laser pulses (bottom) .

Our experimental arrangement also offers the possibility to apply a sequence of two optical pulses. If a small artificial inhomogeneity of the static magnetic field is introduced, a spin echo can be observed under appropriate conditions (see Fig. 4): Here, a second laser pulse is applied after the observable coherence has decayed in the inhomogeneous magnetic field. This pulse not only excites a second FID, but also leads to a refocussing of the spin packets resulting in an echo at time 2T after the end of the first pulse, where T is the delay between the two pulses. As can be seen from Fig. 4, additional spin echoes can occur at times t = 3T, 4T ... . In contrast to previous work[7] we find that strong spin echoes occur only for off-resonant optical excitation. This is shown in Fig. 5 where the echo intensity is shown as a function of the optical detuning Δ. For this measurement, the sample was placed between crossed polarizers; with this arrangement, the detector measures differential absorption thereby ensuring detection sensitivity on resonance.

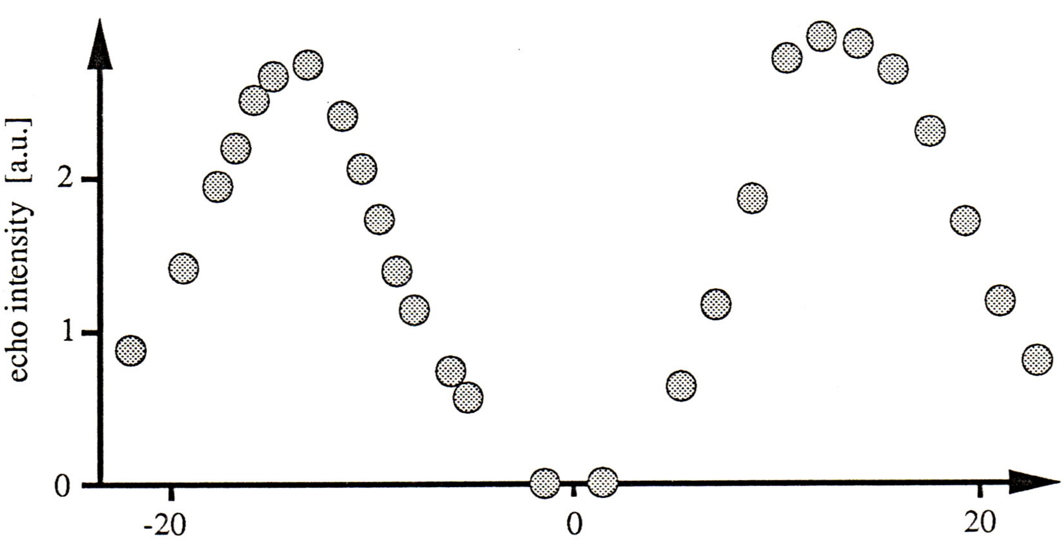

Fig. 5): Intensity of the optically induced spin echo after a sequence of two pulses as a function of the detuning of the laser frequency.

A simple theory based on a Bloch-type equation enables us to calculate the dynamic response of the sodium vapor under rather general conditions with respect to pulse length, pulse intensity and optical detuning; this allows for a detailed comparison of experiment and theory. It turns out, e.g., that the rephasing process for the first spin echo can occur only if the second light pulse induces a light shift in the ground state 2. This light shift acts like an additional dc magnetic field along the beam axis and is essential for the echo formation by tilting the overall precession axis away from the transverse magnetic field direction. Thus on optical resonance where the light shift vanishes, no echoes should show up in accordance with our measurements (see Fig. 5).

The origin of the multiple spin echoes is not yet completely understood. It might be due to higher order Zeeman coherences which are not included in our simplified atomic level scheme (see Fig. 1) or to optical pumping effects. Model calculations show that optical pumping during the second pulse can introduce a frequency-dependent modulation of the spin polarization; this modulation can lead to additional echoes at times which are multiples of the delay time T between the two excitation pulses. Clearly further experimental studies are necessary to better characterize these surprising multiple spin echoes.

It is obvious that a detailed understanding of optically induced sublevel transients is a prerequisite for pulsed magnetic resonance experiments with laser fields. Due to their high sensitivity compared to pure radiofrequency spectroscopy, optical techniques may be of special interest for the study of magnetic resonance phenomena in dilute samples, at interfaces and even on surfaces. We also note that optically induced spin transients like spin echoes may prove useful for the investigation of magnetic resonances in optically excited states of solids.

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