Lateral Displacement of a Laser Beam by a precessing magnetic dipole

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The transverse magnetization-components of an optically pumped atomic vapour induce a longitudinal component in a laser beam propagating through the medium. We show, how the resulting beam displacement is required for the conservation of angular momentum and present an experimental measurement of the beam displacement in Na vapour.

In 1950, Kastler suggested that it should be possible to use polarized light to produce population differences between angular momentum substates of atomic systems [1]. His prediction was based on a detailed analysis of the conservation of angular momentum during absorption of a photon by an atom [2]. While this early theoretical [1] and experimental [3] work dealt exclusively with the modification of the sublevel *populations*, it was shown later, in the 'quantum theory of optical pumping' [4], that also *coherences* between ground state sublevels are affected by the optical pumping process. Physically, these sublevels coherences correspond to angular momentum components perpendicular to the quantization axis. The effect of the pump field on the sublevel coherences is twofold: a damping process reduces their lifetime and a virtual level shift causes a precession of the angular momentum vector. Via the modification of the sublevel dynamics, the light changes therefore the atomic angular momentum. Since the total angular momentum is a conserved quantity, this change of the atomic contribution must be compensated by a change of the angular momentum of the radiation field. A detailed analysis of the situation shows, that this compensation leads to a lateral displacement of the laser beam: a transfer of one quantum of angular momentum in x (y) direction to every photon in the laser beam which propagates along the z-direction leads to a lateral displacement of the laser beam by a distance $\lambda/2\pi$ in -y (x) direction.

As the influence of the light on the dynamics of the atomic angular momentum includes contributions from the damping as well as from the light shift effect, it is also possible to distinguish two contributions to the beam displacement compensating for these effects. The damping of an atomic angular momentum oriented, e.g., along the y axis, should lead to a displacement in the x-direction. Like the damping effect, this beam displacement should have an absorption-like dependence on the detuning of the laser frequency from the optical resonance, independent of the polarization of the light. The light-shift effect, on the other hand, causes a rotation of the angular momentum around the direction of propagation of the laser beam. If the atomic angular momentum is initially oriented along the y-axis, the light-shift effect corresponds to a transfer of angular momentum in x-direction from the radiation field to the atomic medium and the laser beam should be displaced in y-direction. Like the light-shift itself, this displacement should be antisymmetric with respect to the laser detuning and change sign when the polarization of the light changes between opposite circular polarizations. These predictions are in agreement with a semiclassical analysis of the situation [5].

For an experimental verification, we have used the experimental setup of figure 1: the atomic medium, consisting of Na vapour in the presence of 200 mbar of Argon buffer gas, is contained in a glass cell which is heated to a temperature of 124 °C; at this temperature, the absorption at the centre of the resonance line is of the order of 40%. The Na vapour is polarized by irradiation with a pump laser beam in the presence of a transverse magnetic field. The polarization of the pump beam is modulated at a frequency close to the Larmor frequency of the Na ground state [6] to create a

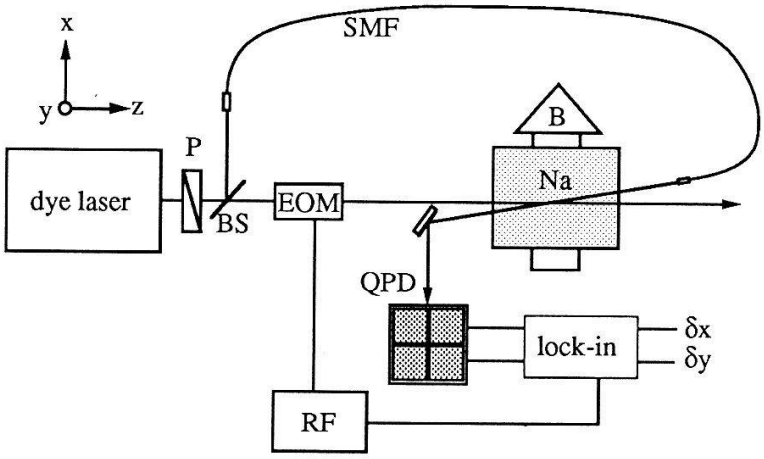

Figure 1: Schematic representation of the experimental setup used for measuring the beam displacement. P = polarizer, BS = beam splitter, EOM = electrooptic modulator, SMF = single-mode fiber, QPD = quadrant photodiode, B = magnetic field, RF = radio frequency synthesizer.

precessing atomic angular momentum. A second laser beam was used to measure the resulting beam displacement. This probe beam was passed through a single mode optical fiber in order to eliminate fluctuations of the beam position originating in the dye laser. For a precise measurement of the beam position, we used a quadrant photodiode with a home-built amplifier and focused the beam into the centre of the four diodes, so that the signal from the detector vanished if the sample was not optically pumped. The intensity of the probe beam was 0.9 mW/cm^2, the intensity of the pump beam 9 mW/cm^2. The two beams overlapped in the sample cell at an angle of ~0.5°.

The detector provides two signals which are proportional to the position of the laser beam. They are passed through a lock-in amplifier in order to extract the contributions from the transverse component of the atomic angular momentum. We measured the amplitude of the components which were out-of-phase with respect to the modulation as a function of the laser frequency in the vicinity of the Na D_1-transition, in order to get the amplitude of the beam displacement. The result is shown in figure 2: the amplitude of the displacement in y-direction, perpendicular to the magnetic field, shown on the left hand side, exhibits the predicted dispersion-like dependence on the laser frequency. The width of the resonance line is determined by the width of the pressure-broadened D_1-transition. Comparison of the upper and lower traces shows that if the polarization of the probe beam is changed between opposite circular polarizations, the displacement changes sign. The corresponding signals for the x-direction (parallel to the magnetic field) are shown on the right hand side. In this case, the displacement amplitude has an absorption-like dependence on the laser frequency detuning and is independent of the probe beam polarization, as expected. These experimental results represent therefore a verification of the predicted displacement of the laser beam.

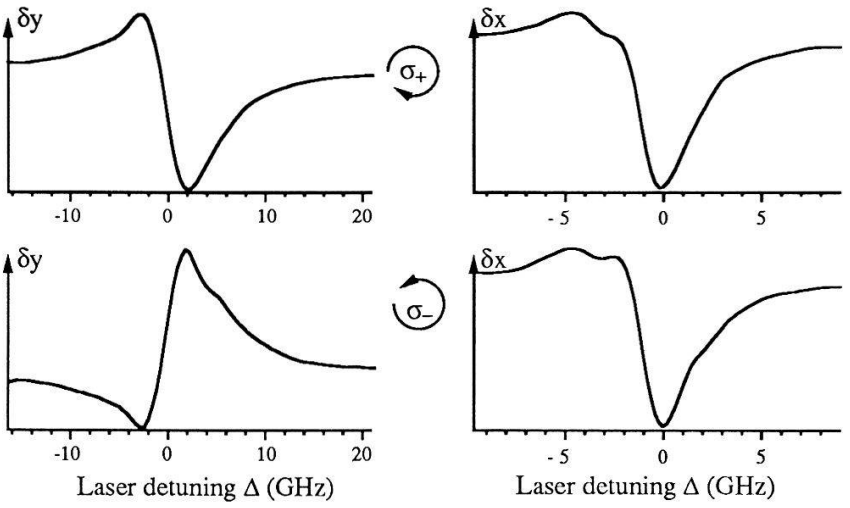

Figure 2: Experimentally measured beam displacement. Shown is the amplitude of the signal at the Larmor frequency, out of phase with respect to the modulation perpendicular (left) and parallel (right) to the magnetic field as a function of the laser frequency. The upper row was measured with a right circularly polarized probe beam, the lower row with a left circular polarization.

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Two-dimensional Spectroscopy of 'Forbidden' Raman Transitions by Laser-Induced Coherence Transfer

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We demonstrate a new spectroscopic technique that makes it possible to obtain information from 'forbidden' Raman transitions. The technique, which is based on time-resolved two-dimensional spectroscopy, uses laser-induced transfer of sublevel coherence from the Raman-inactive transition to an adjacent transition that can be observed in a Raman-process.

The interaction between atomic systems and radiation is often described in terms of a model that treats the atom as a system of two energy levels interacting with a classical electromagnetic wave [1]. In reality, however, most atomic systems have considerably more complicated level structures. The electronic ground state of atomic sodium, e.g., consists of eight energy levels grouped into two hyperfine multiplets. As is well known, this multiplicity leads to many interesting effects, like optical pumping, that cannot be explained in terms of the simple two-level model. We are interested primarily in the dynamics of such multilevel systems coupled to optical fields. Our experiments involve therefore primarily time-resolved experiments, in contrast to the more conventional steady-state experiments [2].

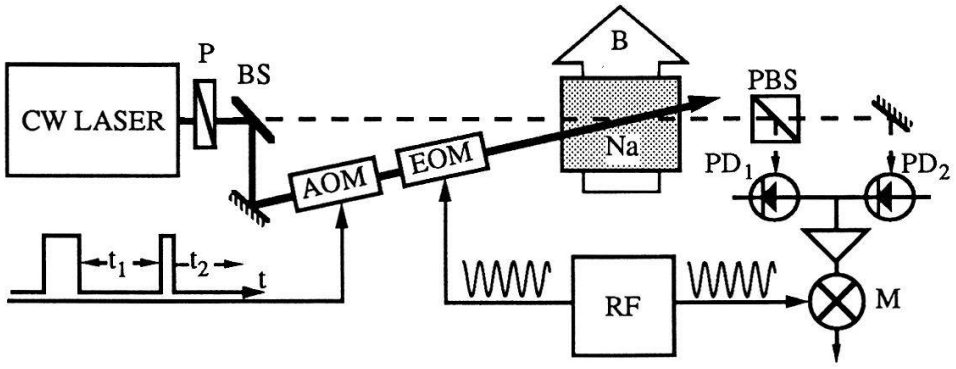

Figure 1: Experimental set-up used for time-resolved pump-probe experiments in atomic multilevel systems. P = polarizer, BS = beam splitter, AOM = acoustooptic modulator, EOM = electrooptic modulator, RF = radio-frequency synthesizer, B = magnetic field, PBS = polarizing beamsplitter, $PD_{1,2}$ = photodiodes, M = mixer.

The experimental setup used for this type of experiment is shown schematically in figure 1. The laser beam, which is derived from a single mode ring dye laser, is split into two parts, a pump beam with an intensity of the order of 100 mW/mm^2, and a probe beam with an intensity of the order of 1 mW/mm^2. The amplitude of the pump beam is controlled with an acoustooptic modulator (AOM) and its polarization can be modulated with an electrooptic modulator (EOM). For an efficient excitation of the atomic medium, which is located in a transverse magnetic field, the polarization of the pump laser beam is modulated between opposite circular polarizations at a frequency close to the Larmor frequency [3]. Pump and probe beam overlap in the sample region, where the metallic sodium is heated to a temperature of ~140°C in the presence of 200 mbar of Ar. The argon is used as a buffer gas to suppress the inhomogeneous Doppler broadening of the optical resonance line and to increase the time the atoms spend inside the laser beam. Behind the sample cell, the probe laser beam is passed into a polarization-selective detector . With the arrangement shown in figure 1 and the polarization of the incident probe beam rotated by 45° with respect to the axis of the polarizing beam splitter, the detector produces a signal proportional to the angular momentum component of the Na atoms parallel to the laser beam.

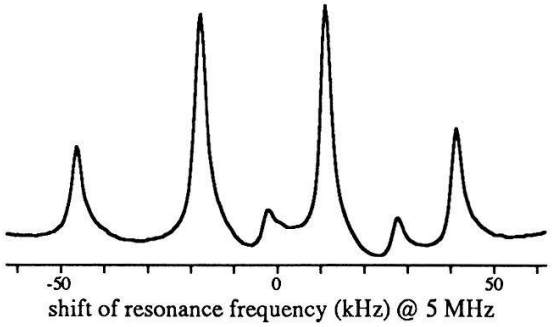

Figure 2: Raman spectrum of the Na ground state.

When a pump laser pulse is applied to the system, it initially induces transient oscillations. After a time of the order of 10 μsec, it settles into a stationary state and after the end of the pulse, the magnetization again starts to precess around the magnetic field; this precession is observed as a free induction decay (FID) signal. The interpretation of the signal can be simplified considerably if it is Fourier transformed.

The resulting spectrum, which is shown in figure 2, contains in general 6 distinct frequency components which can be assigned to the six possible Raman transitions between adjacent sublevels of the Na ground state, i.e. between sublevels whose magnetic quantum numbers m_F differ by $|\Delta m_F| = 1$.

The absence of transitions between sublevels whose magnetic quantum numbers differ by $|\Delta m_F|>1$ can be understood by considering a hypothetical Na atom with vanishing nuclear spin. The electronic ground state consists then of only two sublevels and therefore only a single transition with $|\Delta m_J| = 1$. In the limit of a weak hyperfine interaction, we expect a similar behaviour, i.e. to find only transitions with $|\Delta m_F| = 1$. This and other selection rules can be circumvented by two-dimensional time-resolved spectroscopy. This technique is used extensively in the field of nuclear magnetic resonance (NMR) [4] while applications to optical spectroscopy have not been demonstrated so far. In such an experiment, an initial laser pulse creates order in the form of population differences and coherences within the sublevel multiplets [5]. Since this laser pulse may be arbitrarily strong, multiple Raman transitions can excite coherences also in 'forbidden' transitions, e.g. in transitions with $|\Delta m_F|=2$ [6]. During the subsequent evolution period, these coherences are allowed to precess freely for a time t_1. This free precession period 'labels' the coherences with a phasefactor $\exp(i\,\omega_{ik}\,t_1)$. A second laser pulse causes an exchange of sublevel coherences between the various transitions. As a result, part of the coherence that evolved in transition ik may be transferred, together with the accumulated phase information, into a different transition. If this transition is Raman-active, it is then possible to observe the coherence and thereby measure the phase information associated with the forbidden Raman transition. The resulting signal depends then on the two independent time variables t_1 and t_2, where t_2 represents the time after the second laser pulse. Fourier transformation in both dimensions leads then to a two dimensional spectrum.

An example of a resulting spectrum is shown in figure 3 in contour plot representation. In this case, we suppressed all transitions except the $|\Delta m_F|=2$ transitions after the first pulse by using the different effect that phase shifts of the modulation frequency have on the different types of coherences [6] in order to simplify the spectrum. In the spectrum, we expect therefore along the vertical axis only frequencies corresponding to transitions between next-nearest neighbours. The corresponding frequencies are indicated in the form of a one-dimensional spectrum to the right of the two-dimensional spectrum. After the second pulse, only the 'natural' selection rule $|\Delta m_F|=1$ is active; as a result, the same frequencies as in the one-dimensional spectrum appear along the (horizontal) ω_2-axis. This experimental procedure allows therefore for the first time an observation of all possible transitions between Zeeman substates of the Na ground state.

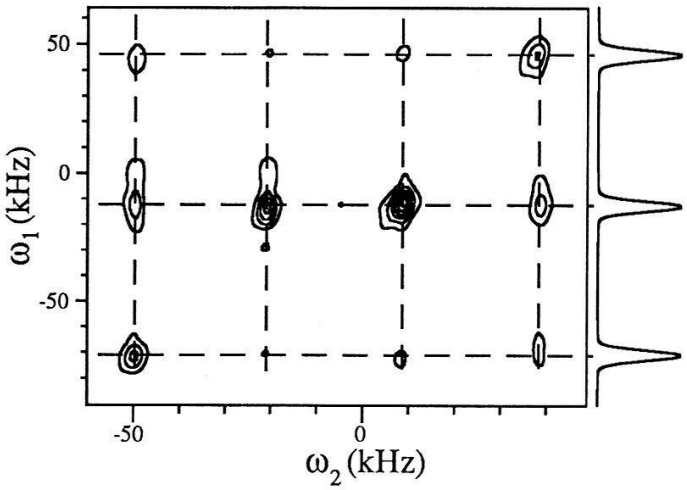

Figure 3: Example of a two dimensional spectrum demonstrating the possibility to use this method for observing 'forbidden' Raman transitions. The vertical axis represents the double quantum ($|\Delta m_F| = 2$) spectrum, while the horizontal axis corresponds to the single quantum spectrum.

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