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Transverse optical pumping with polarization-modulated light

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Optical pumping in a transverse magnetic field can be significantly enhanced if the polarization of the pump beam is modulated at a frequency near the Larmor frequency of the atomic system or a submultiple thereof. We give a theoretical analysis of the associated dynamics and present experimental results from the ground state of atomic sodium.

1. Introduction

One of the most versatile and convenient systems for the investigation of radiatively coupled multilevel systems consists of atomic vapours in a magnetic field. The magnetic field lifts the degeneracy of the sublevels within the hyperfine multiplets and makes it possible to adjust the energy level splittings so that, e.g., the associated Bohr frequencies fall into the desired range which may be limited by theoretical or experimental considerations. The interaction of such multilevel atomic system with laser radiation leads in general to the establishment of a non-thermal population and coherences between the different sublevels of the electronic ground state. Due to the long lifetimes of these ground state sublevels, such optical pumping effects lead to interesting nonlinear dynamics, even at relatively low laser intensity.

Here, we are specifically interested in the case where a magnetic field is applied perpendicular to the laser beam. A typical experimental setup for this type of experiments, such as the one shown in fig. 1, uses a circularly polarized laser beam for optical pumping. The absorption of photons from this pump beam creates magnetization in the electronic ground state which is forced into Larmor precession by the transverse magnetic field. The precessing polarization can be monitored with a second, linearly polarized laser beam, the probe beam, if polarization-selective detection is used after transmission through the sample cell [1].

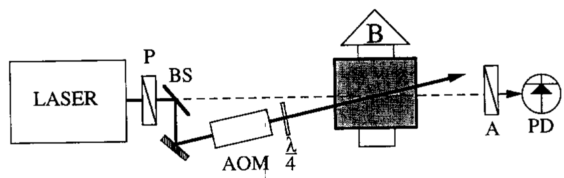

Fig. 1. Experimental setup for pump-probe experiments in a transverse magnetic field. P = polarizer, BS = beam splitter, AOM = acoustooptic modulator, A = analyzer, PD = photodiode, AMP = amplifier.

In order to efficiently polarize such an atomic system by optical pumping, the rate at which the system absorbs angular momentum from angular momentum from the laser beam must be large compared to the rates of all competing processes that tend to rearrange this order, such as the Larmor precession. In the case of strong magnetic fields, when the Larmor precession exceeds a value of the order of, say, 1 MHz, this requirement becomes difficult to fulfill with cw lasers, especially if buffer gas is used to eliminate the Doppler broadening, and/or the frequency of the laser field is far from resonance. Under these conditions, the system can no longer be polarized efficiently [2,3]. This decrease in sublevel polarization at higher Larmor frequencies can be understood as a cancellation process of polarization-packets that are produced at different times: the Larmor precession leads to an accumulation of phase, starting at the time of generation of the packet and extending over its lifetime $\tau \sim 1/P$ where P is the optical pump rate. Packets created at different times interfere therefore destructively if the Larmor frequency Ω_L exceeds the optical pump rate, $\Omega_L > P$.

This destructive interference can be reversed, at least partly, by modulating the intensity of the optical field at a frequency near the Larmor frequency [4–9]. In this case, the polarization of the system can reach values near unity, if the modulation frequency is close to the Larmor frequency: the pump rate need no longer compete with the Larmor frequency, but only with the mismatch between the modulation frequency which can be made arbitrarily small. This amplitude modulation can be achieved either by using a mode-locked laser [8,9] or by external modulation of a cw light source [4–7].

While this intensity-modulation does allow an efficient excitation of sublevel coherence even at relatively high Larmor frequencies [10], it is certainly not the only possibility. One alternative, which offers certain advantages, is polarization-modulation. In this case, the laser intensity remains fixed, but the polarization is modulated between opposite circular polarizations at a frequency near the Larmor frequency of the system. As a result, the polarization that is generated in the atomic system changes sign twice during one modulation period. This leads again to a resonant increase in the polarization of the system if the modulation frequency is equal to the Larmor frequency. The atomic polarization that can be achieved with this scheme is somewhat higher than in the case of sinusoidal amplitude modulation. In addition, the alteration between opposite circular polarizations provides a higher symmetry than the amplitude-modulation of a single sense of circular polarization, which may be desirable in certain experimental situations.

2. Theory

In order to calculate the requirements for the implementation of the polarization-modulation scheme, we have to specify the experimental setup. Specifically, we shall assume that the pump laser beam passes through an electrooptic modulator whose index of refraction is modulated in one direction by a sinusoidal electric field. At this stage, the polarization of the incident beam is left arbitrary, but fixed. We write the (complex) electrical field amplitude $\tilde{E}(t)$ of the optical field as

$$\tilde{E}(t) = (E_{x0}, E_{y0}, 0)\exp[\mathrm{i}(\omega t + kz)] , \tag{1}$$

where the z-axis is the direction of propagation. Behind the modulator of length l, the field is therefore

$$\tilde{E}(t,l) = (E_{x0}\exp(\mathrm{i}k_0 n_x l), E_{y0}\exp(\mathrm{i}k_0 n_y l), 0) \times\exp(\mathrm{i}\omega t) , \tag{2}$$

where k_0 is the vacuum-wavevector and $n_{x,y}$ the indices of refraction of the principal axes. For simplicity, we assume that the time-harmonic electric field with frequency ω_m modulates the index of refraction of the x-direction

$$n_x = n_{x0} + n_{x1}\cos(\omega_m t) . \tag{3}$$

If propagation effects can be neglected, the amplitude of the light behind the modulator is

$$\tilde{E}(t,l) = (E_{x0}\exp\{\mathrm{i}k_0 l n_{x1}[\cos(\omega_m t)]\}, E_{y0}, 0) , \tag{4}$$

where we have assumed $n_{x0} = n_y$ (i.e. no static birefringence).

The quantity that determines the dynamics of the sublevel polarization is the difference between the intensities of the circularly polarized components of the resulting beam [7,11]. For the general polarization assumed here, the intensity-difference can be written as

$$2E_{y0}E_{x0}^{\dagger}\Big(\mathrm{i}J_0(k_0 l n_{x1}) + 2\mathrm{i}\sum_{i=1}^{\infty} J_{2i}(k_0 l n_{x1})\cos(2i\omega_m t) + 2\sum_{i=1}^{\infty} J_{2i-1}(k_0 l n_{x1})\cos[(2i-1)\omega_m t]\Big) + \text{c.c.} , \tag{5}$$

where the J_i represent Bessel functions. This form suggests two possible polarizations of the input beam: it can be linearly polarized at $\pm 45°$ with respect to the principal axes of the modulator, in which case the even order Bessel functions cancel and the intensity difference is an odd function of time, or it can be circularly polarized, in which case the time-dependence is even and only the even harmonics of the modulation frequency occur. In both cases, the efficiency of the excitation of sublevel coherences is determined by the Bessel functions. In the simplest case of linear input polarization and modulation at the Larmor frequency, the efficiency is therefore maximized at the first maximum of J_1, i.e. for $n_{x1} \approx 1.84/(k_0 l)$, while for circular input polarization, the maximum of the second harmonic is reached at for $n_{x1} \approx 3.05/(k_0 l)$, i.e. at a field strength that is approximately 2/3 higher.

3. Experimental results

The experimental setup used to verify these predictions is similar to the one shown in fig. 1, except that the $\lambda/4$ retardation plate was replaced by an electrooptic modulator (Lasermetrics, model 3030FV). A Lyot-compensator was used to correct for the static birefringence of the ADP-crystals. The electrical field for the modulation of the index of refraction was derived from a commercial class A amplifier with a maximum output voltage of 165 V.

The atomic vapour (Na) was contained in a glass cell in the presence of 200 mbar of Ar as a buffer gas at a temperature of 120°C. The laser beams used in the experiment were derived from a single-mode cw ring dye laser (short term linewidth ⩽500 kHz). The laser frequency was set on resonance with the Na-D_1 line (λ=589.6 nm). The pump beam was chopped with an acoustooptic modulator and the pulses were passed through the electrooptic modulator. The linearly polarized probe beam (intensity = 1 μW/mm^2) overlapped with the pump beam in the probe region, with an angle of intersection of less than 1°. Behind the sample, the pump beam was blocked and the circular dichroism was measured, which yields a signal proportional to the ground state polarization component parallel to the laser beam.

If the pump beam is linearly polarized before the EOM, we expect the pumping to be most efficient if the Larmor frequency of the system is an odd multiple of the modulation frequency. This is confirmed by the measurements shown in fig. 2: the solid lines (Bessel functions) represent the calculated ground state polarization as a function of the voltage over the EOM. The scaling of the theoretical curves in the vertical direction was calculated from the laser intensity and the measured decay rate of the polarization which is due to diffusion of the atom out of

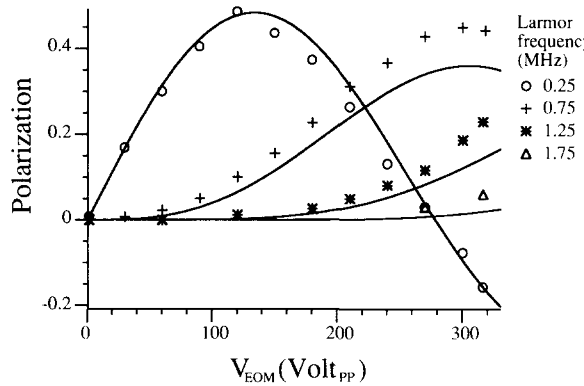

Fig. 2. Relative polarization of the atomic system during excitation with polarization-modulated light for various magnetic field strengths. The solid lines represent the theoretical behaviour, while the symbols correspond to the experimental points measured with four different field strengths. The light incident on the EOM was polarized at 45° with respect to the principal axes of the modulator and the intensity was 1 mW/mm^2.

the laser beam. The experimental data, which were recorded at four different field strengths agree reasonably well with the theoretical functions. The discrepancy in the polarization at higher field strength appears to be due to drift of the laser. For experimental applications, the first order harmonic will in most cases be used, but the higher order harmonics may also be useful, if the desired frequency range cannot be reached directly.

If the pump beam is circularly polarized before the EOM, we expect that pumping will be most efficient if the Larmor frequency of the system is an even multiple of the modulation frequency. This is shown in fig. 3, where the same data as in fig. 2 are shown for the case of circular polarization. In both cases, the pump laser intensity was kept quite low (0.5 and 1 mW/mm^2, respectively), not only to demonstrate that the modulation scheme allows an efficient polarization even with low laser power, but also to make certain that the effect is really resonant and only the nearest harmonic of the excitation is important. Under these conditions, the excitation efficiency drops off quite rapidly if the modulation frequency is deviated from resonance; the half width of the excitation bandwidth is of the order of 5 kHz.

While the modulation frequency of these demonstration experiments is quite low, higher frequencies can readily be achieved. Using rf electronics and

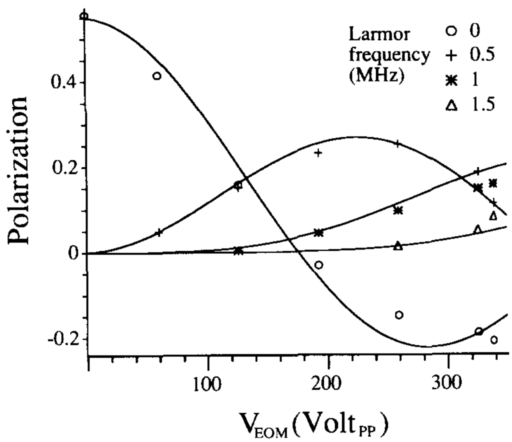

Fig. 3. Same as fig. 2, but with the light incident on the EOM circularly polarized. The intensity of the pump laser beam was 0.5 mW/mm^2.

tuned resonance circuits, we have recorded spectra in the MHz-region, and even higher frequencies should be possible if travelling wave modulators are used.

4. Summary and conclusion

We have shown that polarization-modulated light is an efficient means to optically pump an atomic system in a transverse magnetic field. In this method, the polarization of the resonant light is cycled between opposite circular polarizations at a rate that leads to a resonant enhancement of the ground state polarization produced in the atomic system. Similar to amplitude-modulation schemes [7], this improves not only the polarization of the system, but also enhances other effects of the laser–atom interaction, such as the light-shift, whose effect on the dynamics of the system is small if the splitting of the sublevels is large. As in these cases, the modulation introduces additional degrees of freedom into the experiment, such as the frequency and the phase of the modulation, which may be harnessed for new types of spectroscopic experiments [10].

For the atomic system investigated here, the polarization-modulation is most efficient when it is performed between left- and right circularly polarized light. This is, however, not the only possibility and other experimental situations may profit from different schemes, where the polarization may be cycled between different linear orientations or even more general circuits on the Poincaré sphere. It has been shown that such experiments lead, in general, to geometrical effects ("Berry's phases") which depend on the trajectory on the Poincaré sphere. Since the atomic system follows the polarization of the light adiabatically, these effects should be observable in the resonant medium.

In the experiment demonstrated here, the laser radiation is simultaneously resonant with two electronic transitions. While the modulation creates sidebands which may be well outside the spectral width of the laser, their separation is, under our experimental conditions, still small compared to the homogeneous width of the optical transition. The efficiency of the excitation of the sublevel coherences is therefore not associated with a change in the optical resonance, but only with the modification of the dynamics within the electronic ground state. Like other modulation methods (see, e.g. ref. [9]), it provides therefore the high resolution of rf-spectroscopy combined with the high sensitivity of optical spectroscopy. The method could in principle be applied even to systems with sublevel splittings in the GHz range, if travelling wave electrooptic modulators are used; however, in this case the associated optical transitions may no longer be within the same homogeneous linewidth and the simple theoretical analysis given above is no longer applicable.

Acknowledgements

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