RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 48, NUMBER 6 DECEMBER 1993

Nonlinear effects of radiation trapping in ground-state oriented sodium vapor

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(Received 10 September 1993)

Effects of radiation transport in the interaction of a laser beam with sodium vapor in an argon atmosphere are reported. Investigations of the temporal and spatial evolution of ground-state orientation, which is produced by optical pumping with a circularly polarized near-resonant laser beam, reveal a strong deviation from the expected pure diffusive behavior. Transverse profiles are narrowed considerably with increasing sodium density as well as with increasing pump intensity. The temporal decay of orientation is also drastically altered. A simplified nonlinear and nonlocal theoretical model of radiation trapping gives excellent qualitative agreement with the observations and allows an intuitive interpretation.

PACS number(s): 32.80.−t

The process of radiation trapping has been studied extensively in different subfields of physics like astrophysics, plasma physics, and atomic spectroscopy, but its role in the interaction of an intense laser beam with an atomic sample has been left largely unexplored, with a few theoretical exceptions only [1,2]. The saturation phenomena occurring in this interaction are obviously enhanced by the increase of the population of the excited state due to radiation trapping. Also the dynamics of the system will be changed, since the decay of the population may be slowed down by orders of magnitude. However, especially in the resonant or near-resonant case optical pumping phenomena in the ground state may be an essential feature of the interaction. They can be used for several purposes, such as producing polarized atomic sources or targets, and they can give rise to nonlinear optical phenomena, even at very low laser intensities.

Recently the existence of a destructive action of radiation trapping on the orientation produced by optical pumping in the ground state has been demonstrated [3–6]. In this paper we present results of a detailed experimental study of the influence of radiation trapping on the spatial distribution and on the temporal evolution of the orientation created by a circularly polarized light beam in sodium vapor and we find dramatic effects. These include a counterintuitive narrowing of the spatial distribution of orientation with increasing intensity and a strong influence of the laser intensity on the temporal behavior of the orientation. These phenomena may be regarded as basically nonlinear. We also present a simplified theoretical model which gives an excellent qualitative description of the observations.

The experimental setup is illustrated in Fig. 1. Sodium vapor in an argon atmosphere is contained in a stainless steel tube of 2 cm diameter, sealed off with glass windows. A segment of 3 cm length is heated to temperatures in the range between 180 and 280 °C. We determine sodium densities from measurements of the linear Faraday effect in gases [7], yielding values between 5×10^{11} and 5×10^{13} cm^{-3}, with a residual uncertainty of 15%. To ensure large homogeneous broadening the buffer gas pressure is typically adjusted to about 300 hPa, resulting in a linewidth of 3.7 GHz (full width at half maximum). A static longitudinal magnetic field of about 1 mT is applied to suppress any coherence between Zeeman substates of the atomic ground state, i.e., any transverse components of the spin expectation value. As a light source we use a cw ring dye laser which provides a maximum output power of 90 mW at the cell entrance in the spectral region of the sodium D_1 line, which was exclusively used in the experiments. The frequency detuning was adjusted to establish a pump beam absorption of about 10–30%. Typically this was achieved by detunings of about 10 to 20 GHz. The laser beam is split into counterpropagating beams of suitable polarization, which act as a pump and a probe beam, respectively, with a typical power ratio of 10^6. The lateral distance between the beams can be varied over the full cell diameter. Both beams are Gaussian (TEM_{00}) with $1/e^2$ radii of 460 and 260 μm, respectively.

The continuous excitation by the pump beam, which is chosen to be circularly polarized, should create a stationary spatial distribution of ground-state-oriented sodium atoms with cylindrical symmetry. This distribution is probed by measuring the transmission for right- and left-hand circularly polarized probe beams. Thus, a quantitative measure of the orientation, averaged over the length of the interaction region, as a function of the lateral distance from the pump beam can be derived.

A typical dependence of the stationary radial distribution of the orientation on the sodium density is shown in Fig. 2. With increasing sodium density there is not only

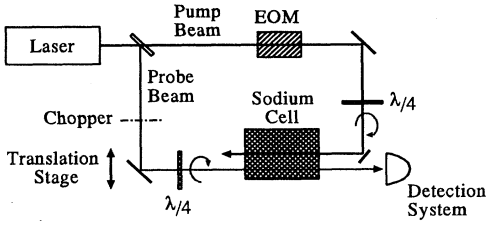

FIG. 1. Schematic of the experimental setup. EOM: electro-optic modulator (Pockels cell); λ_4: quarter-wave plate.

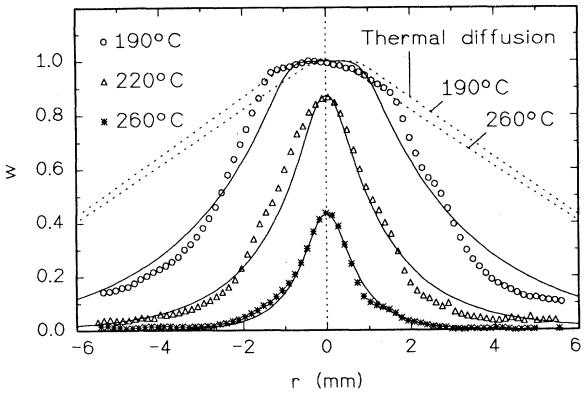

FIG. 2. Measured steady-state spatial profiles of ground-state orientation for different densities of sodium atoms compared with calculated profiles. Pump power, 90 mW; argon pressure, 300 hPa; 190 °C corresponds to a sodium density of 3.5×10^{11} cm^{-3}, 220 °C to 2.6×10^{12} cm^{-3}, and 260 °C to 2.0×10^{13} cm^{-3}. Calculations with particle diffusion only (dashed lines) and including radiation trapping (solid lines).

a decrease of the maximum obtainable orientation, but a considerable *narrowing* of the ground-state profiles also occurs. In Fig. 3 the dependence of the orientation profiles on pump laser intensity is displayed. Surprisingly enough the orientation is found to fall off more rapidly

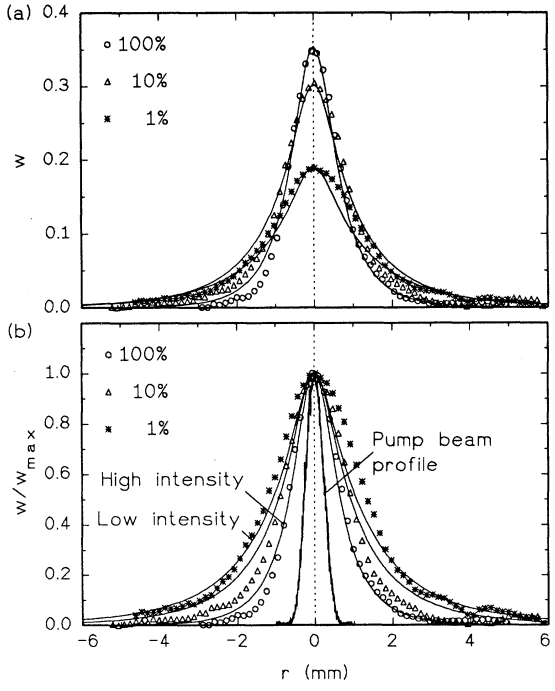

FIG. 3. Measured steady-state spatial profiles of ground-state orientation for different pump intensities; (b) normalized representation of (a). Temperature, 270 °C corresponding to a density of 3.1×10^{13} cm^{-3}; frequency detuning, 10.3 GHz; argon pressure, 300 hPa; 100% corresponds to 90 mW pump power. Solid lines, theoretical curves (assumed pump intensity ratios: 100%:40%:15%).

in an intense pump beam.

In order to extend the investigations to the dynamical behavior of ground-state orientation, we measured the temporal evolution of the probe beam transmission following a sudden switch-off of the pump light from a large stationary value. The obtainable temporal resolution was limited by the switch-off time of the Pockels cell to about 20 ns. A typical result for the on-axis transmission is shown in Fig. 4; the displayed curve is an average over 200 experimental runs. Two different time scales are evident: The "slow" one is in the order of a few microseconds while the "rapid" one is in the 100-ns range. A similar behavior has already been reported in Ref. [4].

In order to understand the observations by means of a theoretical model, we start from rate equations for an atomic spin-degenerate two-level system as shown in Fig. 5. The medium is described by the diagonal elements ρ_{ii} of the density matrix. Under the conditions of the experiment it seems reasonable to neglect the Doppler effect and the hyperfine structure. Any population differences between sublevels of the excited state are rapidly destroyed by collisions with buffer gas atoms, i.e., $\rho_{33} = \rho_{44}$ is a reasonable approximation. Under these assumptions only the "orientation" $w = \rho_{22} - \rho_{11}$ and the total population of the excited state $s = \rho_{33} + \rho_{44}$ have to be considered. Introducing D_T as the thermal particle diffusion coefficient [8], Γ_1 as the decay rate of $s = s(\mathbf{r}, t)$ due to spontaneous emission, and $P_+ = P_+(\mathbf{r}, t)$ as the pump rate for circularly polarized light (σ_+), the following set of rate equations can be derived [9]:

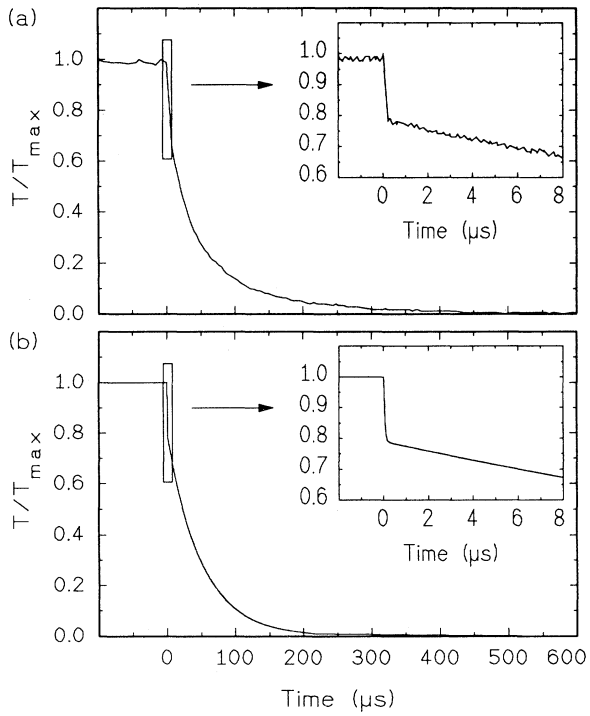

FIG. 4. Temporal evolution of the on-axis probe beam transmission after switching off the pump source. Temperature, 250 °C corresponding to a density of 1.6×10^{13} cm^{-3}; frequency detuning, 10 GHz; argon pressure, 300 hPa; (a) experimental, (b) calculated.

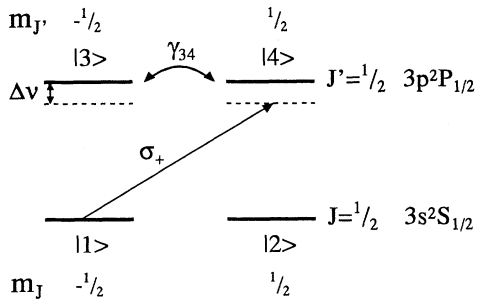

FIG. 5. Four-level scheme for the $3s\,^2S_{1/2}$ ground state and the $3p\,^2P_{1/2}$ excited state of sodium. Any excited-state population difference is assumed to be rapidly destroyed ($\gamma_{34} \gg \Gamma_1$).

$$\dot{w} = P_+(1 - w - 2s) + D_T\nabla^2 w - P_F\, w, \tag{1a}$$

$$\dot{s} = P_+(1 - w - 2s) - \Gamma_1\, s + P_F\,(1 - 2s). \tag{1b}$$

The terms proportional to $P_F = P_F(\mathbf{r}, t)$ account for the absorption of randomly polarized fluorescent light that has been emitted at any point $\mathbf{r}'$ in the considered volume and is reabsorbed at point $\mathbf{r}$. In the case of Eq. (1a) this leads to a destruction of orientation and in Eq. (1b) to an increase of excited-state population. The high buffer gas pressure used in our experiment leads to a considerable population in the excited state via a collisionally assisted excitation process, even when the laser is detuned far from the resonance [10]. Under these conditions the emission line profile is proportional to the absorption line profile (complete frequency redistribution [11]).

For simplicity we assume that the absorption coefficient $\bar{\alpha}$ of the medium does not depend on s, w, and the direction of polarization. We also introduce a mean free path $1/\bar{\alpha}$ for the fluorescence photons [12,13]. Though this approach is by no means allowed in general (cf. Ref. [14] and references therein), it should be suitable in the case of our experiment, due to the low optical density. Moreover, we use a one-dimensional calculation, since it has proved to be successful for radiation trapping problems including only the excited state [12]. Defining the radial coordinate x we finally obtain the incoherent pump rate

$$P_F(x,t) = \bar{\alpha}\,\frac{\Gamma_1}{2}\int_{-R}^{R} s(x',t)\, e^{-\bar{\alpha}\,|x'-x|}\, dx'. \tag{2}$$

Thus, with $P_F(\mathbf{r}, t)$ described by a spatial integral, the set of equations (1) reveals itself to be a set of nonlinear nonlocal equations. The interesting feature in these equations is that even outside the pump beam volume a nonlinear coupling between the atomic variables w and s is introduced by the reabsorption process. Typical steady-state distributions of the atomic orientation $w(x,t)$ obtained from Eqs. (1) were calculated under the boundary conditions of w and s vanishing at the cell walls, i.e., $w(R) = s(R) = 0$.

In the calculations the value of the pump rate was used as a fit parameter. As a consequence of the one-dimensional calculation of P_F we had to reduce the mean absorption coefficient $\bar{\alpha}$ for homogeneous broadening from Ref. [13] by a factor of about 4. This correction has been confirmed by a small series of fully three-dimensional calculations neglecting pump beam depletion.

As can be seen in Figs. 2–4 the simple model presented here describes the experimental observations surprisingly well. We conclude that the mechanism responsible for the observations is the following: Via P_F, radiation trapping introduces an additional destruction mechanism for orientation even outside the pump beam volume where $P_+ = 0$. The influence of this term can now be enhanced by increasing the density of excited atoms either via the pump intensity or by increasing the sodium density, leading to a reduction of the mean free path for photons. Both lead in effect to a narrowing of the orientation profiles, as observed in the experiments.

The temporal behavior displayed in Fig. 4 may be interpreted in the following way: Immediately after the switch-off, there is a large value of P_F that practically decays, with a time constant determined by the lifetime of s which is prolonged from the natural lifetime (16.4 ns) into the 100-ns range by radiation trapping. Due to the structure of Eq. (1a) there is, for $P_+ = 0$, a fast decrease of w, as long as $P_F \neq 0$. Later there is a diffusion of particles only, and therefore we observe a slow decay. The diffusion process, however, depends on the initial spatial profile of w, which in turn is strongly influenced by radiation trapping. As shown before, the slope of w becomes steeper with increased sodium density or laser intensity, thus leading to a faster diffusive decay of the orientation.

It has to be emphasized that the massive influence of radiation trapping on the dynamics and steady-state distributions of Zeeman pumped atomic ensembles presumably has consequences for many experiments relying on Zeeman pumping. This has very recently been shown for the dynamics of sodium-filled resonators [15], and degenerate four-wave mixing [1,16]. Studies of transverse optical effects in the interaction of a laser beam with sodium vapor reveal very clearly that radiation trapping can efficiently counteract the smoothing effect of particle diffusion which tends to suppress many spatial effects if relaxation processes are absent [16]. Finally, we would like to point out that the influence of radiation trapping or, more generally, of the diffusion of radiation on the population of the excited state should also be taken into account in the quantitative interpretation of experiments, even though we generally expect less spectacular effects than the one discussed here.

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