RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 49, NUMBER 3 MARCH 1994

Role of radiation trapping in degenerate four-wave-mixing experiments

M. Schiffer, G. Ankerhold, E. Cruse, and W. Lange
Institut für Angewandte Physik, Westfälische Wilhelms-Universität Münster, Corrensstrasse 2/4, D-48149 Münster, Germany

(Received 23 November 1993)

The process of degenerate four-wave mixing is studied experimentally in dense sodium vapor in a rare-gas atmosphere. The influence of trapped fluorescence light on the ground-state orientation is shown to be responsible for the observed strong reduction of saturation phenomena with increased sodium density. The interpretation is based on a simple model and is supported by results obtained by suppressing the fluorescence.

PACS number(s): 42.65.Hw, 32.80.−t

Sodium vapor under a rare-gas atmosphere has been a model system for degenerate four-wave mixing (DFWM) for years. It has played an important role in the discussion of pressure-induced processes [1,2] and is well understood for small sodium densities. In the present paper, however, we study DFWM under conditions of high sodium densities and focus on the dependence of the signals on a transverse magnetic field. Though it is anticipated that the behavior observed in similar experiments in less dense vapors [1,3] will be modified under the present conditions, it may be unexpected that the observations reveal very clearly that a mechanism has come into play which has not been taken into account before. We identify the new mechanism as the influence of radiation trapping on the atomic ground state and use a simplified theoretical model which describes the observations surprisingly well. Further evidence of our interpretation is given by presenting experimental results obtained after suppressing the resonance fluorescence.

The process of radiation trapping is well known to be responsible for an increase of the lifetime of the excited-state population in an atomic ensemble beyond the natural lifetime of isolated atoms [4]. Its consequences for the propagation of a laser beam [5] and very recently for DFWM in an ensemble of nondegenerate two-level atoms [6] have been studied theoretically. It is also known that the process is responsible for the destruction of long-lived spin orientation in the atomic ground state [7], but the consequences for nonlinear optical processes have remained largely unexplored. Very recently, experimental and theoretical studies of the role of radiation trapping in the dynamics of a sodium-filled resonator [8] and of its impact on the spatial distribution of ground-state orientation which determines the nonlinear optical properties of a sample [9,10] have been performed. In this paper we demonstrate that DFWM signals can qualitatively be changed by radiation trapping and thus provide a striking example of its potential importance in nonlinear optical processes.

In our experiment we use a standard DFWM geometry, as shown in Fig. 1. Sodium vapor in an argon or a nitrogen buffer-gas atmosphere of typically 100 to 300 hPa is contained in a stainless steel tube with a heated zone of 5 cm length and a cell diameter of 2 cm. The cell temperature can be varied from 200 to 320 °C, corresponding to a sodium density of 5×10^{12} to 2×10^{14} cm^{-3}. Particle densities were measured using the Faraday effect [10], and it was confirmed that sodium densities were independent of the kind of buffer gas used.

Helmholtz-type coils in three dimensions allow a well-defined magnetic field to be produced in the interaction region. The earth magnetic field and magnetic stray fields were carefully compensated. Two circularly polarized (σ_+) beams E_1 and E_2 from a cw ring dye laser tuned near to the D_1-resonance line, with a power of approximately 70 mW each, intersect in the vapor with an angle of 2 mrad. These beams are regarded as pump beams. The backward beam E_B of 8 mW, derived from the same laser, has the same circular polarization (σ_+). Both pump beams E_1 and E_2 can be switched on using a fast Pockels cell with a switch-on time of about 20 ns.

Under these experimental conditions the basic mechanism leading to the DFWM signal can be well understood in terms of an atomic spin-degenerate two-level system as schematically depicted in Fig. 2. Since a high buffer-gas pressure is used it can be justified to neglect the Doppler effect and the hyperfine splitting [11]. Any excited-state population difference is assumed to be destroyed during the excited-state lifetime. Under these assumptions only the "ground-state orientation," represented by $w = \rho_{22} - \rho_{11}$, and the total population of

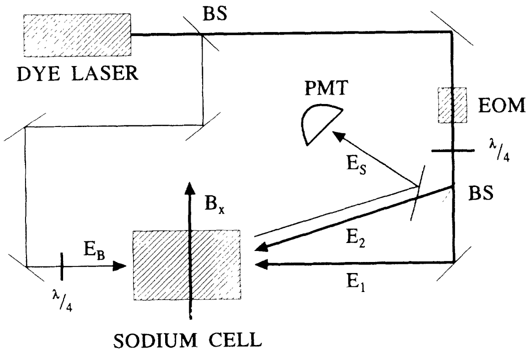

FIG. 1. Schematic of the experimental setup. All input beams (E_1, E_2, and E_B) as well as the DFWM output signal E_S are circularly polarized. B_x denotes the transverse component of an external magnetic field. BS, beam splitter; EOM, electro-optic modulator (Pockels cell); $\lambda/4$, quarter-wave plate; PMT, photomultiplier.

1050-2947/94/49(3)/1558(4)/$06.00 49 R1558

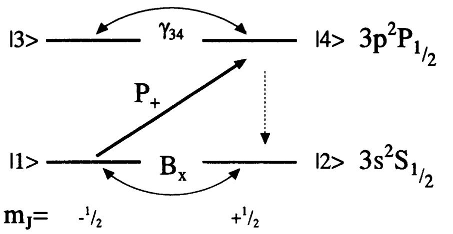

FIG. 2. Spin-degenerate two-level scheme used for a simplified description of the $3s\,^2S_{1/2}$ ground state and the $3p\,^2P_{1/2}$ excited state of sodium. The pump rate P_+ is induced by the circularly polarized pump beams. The transverse magnetic field B_x couples the Zeeman sublevels.

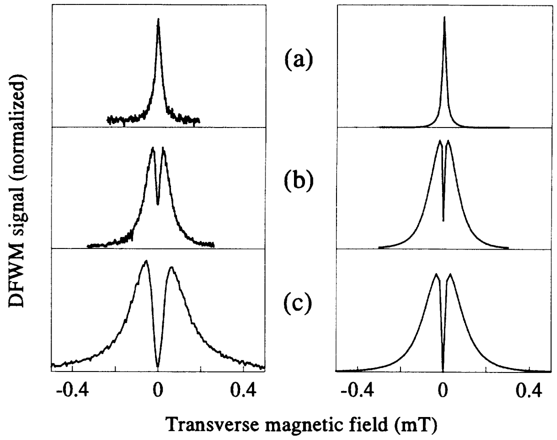

FIG. 3. Magnetic-field dependence of the DFWM signal with argon as a buffer gas for different temperatures. Left column, experimental. Buffer-gas pressure is 100 hPa. Temperatures are (a) 260 °C, (b) 210 °C, (c) 170 °C. Right column, theory. Reabsorption probabilities are (a) $\mathcal{R} = 0.8$, (b) $\mathcal{R} = 0.2$, (c) $\mathcal{R} = 0$. The fit parameter was the absorption coefficient.

the excited state, represented by $s = \rho_{33} + \rho_{44}$, have to be considered in a treatment using the density matrix formalism.

The interference between the two pump beams in the interaction region results in a spatial modulation of the refractive index. The backward beam is scattered from this index grating, leading to a DFWM signal I_S proportional to the square of the modulation amplitude [12]. In case of negligible excited-state population s, only the spatial modulation of the orientation contributes to the refractive-index grating. Assuming sinusoidal spatial modulation of the orientation grating, the DFWM signal then is proportional to the square of the modulation amplitude Δw of the orientation:

$$I_S \sim |\Delta w|^2. \tag{1}$$

The modulation of w, which may be regarded as the grating contrast, vanishes if thermal diffusion of oriented atoms from the maxima to the minima of the orientation leads to a washout of the grating. However, in very small magnetic fields the contrast can be enhanced by applying a transverse component of the magnetic field, which causes transitions between the Zeeman sublevels of the ground state and thus reduces the orientation in the minima of the grating, while the nearly saturated orientation in the maxima is not severely affected by weak transverse magnetic fields. Finally, when the Larmor frequency in the transverse field is increased and becomes comparable to the pump rate P_+ as defined in Ref. [11], the orientation decreases even in the maxima of the grating. Scanning the transverse field thus typically leads to Hanle-type spectra with a saturation dip centered around zero-magnetic field (cf. Refs. [1,3]).

Generally the mechanisms considered to be responsible for the destruction of orientation are collisions with buffer-gas atoms and collisions with the cell walls [13]. The cross section σ for the destruction of ground-state orientation by collisions with argon atoms is well known to be very small ($\sigma_{\mathrm{Ar}} = 8.8 \times 10^{-27}\,\mathrm{m}^2$ [14]). Moreover, the thermal diffusion of oriented atoms, responsible for wall collisions, is slow in a buffer-gas pressure of a few 100 hPa and corresponds to a decay rate of a few kilohertz at best. On the other hand, the laser intensity used in the experiment leads to a pump rate of about 1 MHz. Therefore it should be expected that saturation effects are observable over the entire temperature range.

Experiments with argon as buffer gas, however, revealed that the zero-magnetic-field saturation dip vanishes at temperatures above 240 °C, leading to a Lorentzian-type behavior. Typical dependencies of the DFWM signal output on the transverse magnetic field for different temperatures are shown in the left column of Fig. 3.

An explanation for the disappearance of the saturation effects with increasing temperature is provided by considering the additional destructive influence of radiation trapping on the orientation. With increasing sodium density the excited-state population accumulates due to radiation trapping and leads to a nearly unpolarized and incoherent but resonant background radiation field which is present in the entire interaction zone. Oriented atoms can absorb this radiation and may be disoriented efficiently, as was shown in Refs. [7,9].

Radiation trapping is the dominant loss mechanism for orientation, if collisions with buffer-gas atoms reduce the thermal motion of oriented atoms to the cell walls. It reduces the obtainable orientation and enhances the absorption of pump and probe beams, since it counteracts the saturation of the medium. At higher temperatures also absorption of the DFWM signal beam has a strong influence on the shape of the Hanle signal: The absorption of the light beams is determined by the value of w and vanishes if the medium is saturated. Scanning the transverse magnetic field B_x leads to a decrease of orientation and thus to enhanced absorption. At higher temperatures even a small reduction of the orientation due to the transverse magnetic field results in a dramatic decrease of the transmitted signal. This should be especially significant if the orientation is additionally diminished by radiation trapping, and a strong narrowing of the Hanle signals with increasing temperature is expected.

A second consequence of radiation trapping is its influence on spatial ground-state orientation distributions, as was pointed out in Refs. [9,10]. Due to its destructive action, radiation trapping counteracts the diffusive transport of spin orientation. In the case of near saturation, this loss mechanism has the largest impact in regions of small intensity, thus preserving the shape of the spatial structures of orientation imposed on the sample by the pump beam profile. In the case of DFWM this leads to an increase of the grating contrast under saturation conditions, and in turn to an increase of the DFWM signal in the zero-magnetic-field region ($B_x = 0$) despite thermal washout.

Further insight into the mechanisms involved in the DFWM process could be achieved by measuring the temporal behavior of the signal after switching on both pump beams simultaneously. A typical experimental result is shown in Fig. 4(a). The pump beams first create a grating with high contrast, which saturates after a few microseconds. Thermal diffusion of oriented atoms now leads to a washout of the grating contrast, with a time constant depending on pump intensity, thermal diffusion constant, and the grating period determined by the angle included by the pump beams. Finally, an equilibrium state is achieved by the simultaneous effect of radiation trapping, which destroys ground-state orientation in the minima of the grating and thus preserves the grating contrast, and particle diffusion, which would otherwise cause complete washout. In the zero-magnetic-field region radiation trapping acts now as the mechanism solely responsible for the occurrence of a DFWM signal. This is in contrast to the role of radiation trapping in two-level media, where a decrease of the DFWM signal for all parameter regimes is expected, as was shown theoretically in Ref. [6].

For a simplified theoretical treatment radiation trapping is included in the density matrix formalism following the argumentation of Refs. [8–10]. The fluorescence field is assumed to be proportional to the decay of the excited-state density ($\Gamma_1 s$) and the optical depth of the sample is taken into account by the reabsorption probability $\mathcal{R}$, which increases with sodium density. Further we assume that the background radiation field is spatially constant inside the grating region [6]. This assumption provides a very rough description of the complex problem of radiation trapping processes, but keeps the problem tractable for a numeric solution. The elements of the density matrix are calculated in a space- and time-dependent formulation, taking into account thermal diffusion inside the grating region and introducing an exponential decay rate describing the overall loss due to the diffusion of oriented atoms to the cell walls. The refractive-index modulation can then be calculated and the intensity of the DFWM signal I_S is adapted using Eq. (1). We further assume undepleted pump beams, but take into account the absorption of the DFWM signal wave. A series of numerical results for the magnetic-field dependence is presented in the right column of Fig. 3, while results for the time dependence are shown in the right column of Fig. 4. Good qualitative agreement with all experimental results could be achieved with this simplified treatment.

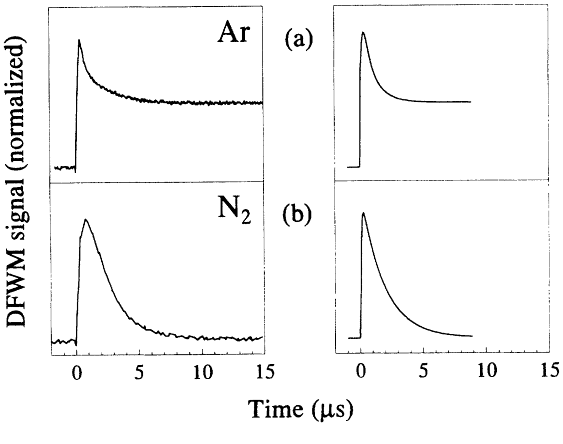

FIG. 4. Temporal evolution of the output signal after switching on the pump beams: (a) argon buffer gas, (b) nitrogen buffer gas. Left column, experimental. Temperature is 250 °C; buffer-gas pressure, 100 hPa; transverse magnetic field, $B_x = 0$. Right column, theoretical calculations; (a) $\mathcal{R} = 0.6$, (b) $\mathcal{R} = 0$.

A direct experimental test of these considerations on the role of radiation trapping could be provided, if there were a means of switching it off, with all other parameters left unchanged. This can indeed be achieved by employing a buffer gas with a high quenching cross section for the excited state. Nitrogen serves well for this purpose, having a quenching cross section for the sodium excited $3p\,^2P_{1/2}$ state of 5×10^{-15} cm^2 and inducing radiationless transitions to the ground state, whereas ground-state orientation is not affected [15]. It was also verified experimentally that the shape of the D_1 line is not considerably altered if argon is replaced by nitrogen of the same pressure in the 100-hPa range. In the absence of radiation trapping the nonlinear susceptibility of the sample should be proportional to the particle density and we can expect strong DFWM signals at high temperatures. On the other hand, in a DFWM experiment in zero-magnetic field, due to the lack of an additional destructive mechanism, strong saturation should occur because the diffusion of oriented atoms into the minima of the grating provides an efficient washout of grating contrast.

We repeated the experiments under nitrogen buffer gas and found that under conditions of similar sodium densities, frequency detuning, and buffer-gas pressure the behavior is indeed changed completely. A series of experiments within the temperature range from 180 to 310 °C revealed that the saturation dip was always observable (Fig. 5). Also the temporal behavior of the DFWM signal, which shows the time dependence of the grating buildup in the zero-magnetic-field region, shows saturation behavior with an equilibrium value of almost zero signal intensity [Fig. 4(b)].

The washout of the grating contrast by particle diffusion does not occur, if the circular polarization of the pump beams is replaced by mutually orthogonal linear polarizations, i.e., if the intensity grating is replaced by a polarization grating [1]. In this case the suppression of radiation trapping allows strong saturation and thus yields an enhancement of the nonlinearity. In the ex-

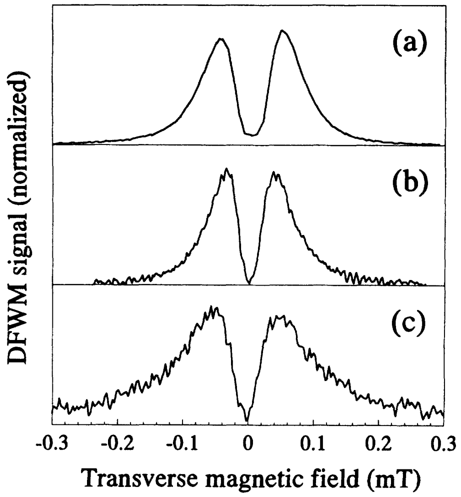

FIG. 5. Magnetic-field dependence of the DFWM signal for nitrogen buffer gas at different cell temperatures. Buffer-gas pressure is 100 hPa. Temperature (a) 310 °C; (b) 210 °C; (c) 180 °C.

periment this gives rise to diffracted beams of up to the eighth order.

We would like to point out that in dense vapors an increase of the pump intensity does not automatically lead to a recovery of saturation phenomena, since, according to the considerations presented here, the background fluorescence typical for radiation trapping situations is increased simultaneously. Thus, with respect to ground-state orientation, radiation trapping may be regarded as an intensity-dependent loss mechanism. This intensity dependence adds a qualitatively new type of nonlinearity to atomic vapors [9], whose consequences have not yet been fully explored.

In the present work the crucial role of radiation trapping was experimentally demonstrated by eliminating it by means of a quenching buffer gas. In doing so we observed details of the DFWM process, which were obstructed by the effect of radiation trapping and will be described in a forthcoming paper [16]. On the other hand, the comparison between Figs. 4(a) and 4(b) and between Figs. 3 and 5 which is used here to elucidate the role of radiation trapping in nonlinear optical processes, also demonstrates a pronounced influence of quenching collisions on DFWM. It is mediated by their influence on radiation trapping and is thus based on a more sophisticated mechanism than the reduction of the excited-state population described recently [17].

We gratefully acknowledge the contributions of U. Haupt in parts of the experimental work.

[1] Y. H. Zou and N. Bloembergen, Phys. Rev. A **33**, 1730 (1986).
[2] G. S. Agarwal, Adv. Atom. Mol. Opt. Phys. **29**, 113 (1991), and references therein.
[3] E. Köster, J. Mlynek, and W. Lange, Opt. Commun. **53**, 53 (1985).
[4] T. Colbert and J. Huennekens, Phys. Rev. A **41**, 6145 (1990).
[5] A. W. McCord, R. J. Ballagh, and J. Cooper, Opt. Commun. **68**, 375 (1988).
[6] N. J. Mulgan and R. J. Ballagh, Phys. Rev. A. **48**, 3321 (1993).
[7] M. Tanaka *et al.*, Phys. Rev. A **41**, 1496 (1990).
[8] M. Möller and W. Lange (unpublished).
[9] G. Ankerhold, M. Schiffer, D. Mutschall, T. Scholz, and W. Lange, Phys. Rev. A **48**, R4031 (1993).
[10] G. Ankerhold, Ph.D. thesis, University of Münster, 1993.
[11] D. Suter, Phys. Rev. A **46**, 344 (1992).
[12] H. J. Eichler, P. Günter, and D. W. Pohl, *Laser-Induced Dynamic Gratings* (Springer-Verlag, Berlin, 1986).
[13] C. D. P. Levy and P. W. Schmor, J. Appl. Phys. **63**, 4819 (1988).
[14] A. T. Ramsey and L. W. Anderson, Nuovo Cimento **32**, 1151 (1964).
[15] A. C. Tam, J. Appl. Phys. **50**, 1171 (1979).
[16] M. Schiffer, E. Cruse, and W. Lange (unpublished).
[17] P. M. Danehy, E. J. Friedman-Hill, R. P. Lucht, and R. L. Farrow, Appl. Phys. B **57**, 243 (1993).