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Nonlinear saturated absorption in resonant media: Level-degeneracy-induced polarization effects

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The effect of level degeneracy on saturated absorption in a resonant medium is considered using a simple but realistic three-level system to model $J=1\rightarrow J=0$ atomic transition. Assuming homogeneous broadening and exact resonance, an analytic nonperturbative expression for the medium susceptibility is obtained and used to study propagation of an arbitrarily polarized optical field. Optical saturation affects considerably Zeeman–coherence-induced circular dichroism. The analysis may find application in high-resolution spectroscopy and other branches of nonlinear optics.

Since the advent of the laser, the nonlinear interaction of an optical field with resonant atomic media has been a subject of considerable interest.[1–16] Of particular interest are the saturation effects that lead to reduced optical absorption when the field intensity exceeds a threshold value determined by the atomic relaxation rates and the dipole moment associated with the single-photon transition. A widely employed model[1,2] considers a single optical mode interacting with a two-level system and has been highly successful in various fields such as nonlinear spectrcosopy,[3] optical bistability,[4] and phase conjugation.[5] Such a two-level model is based on the assumption of nondegenerate energy states and ignores beam-polarization effects.

A realistic description of atom-radiation interaction requires that level degeneracy related to angular momenta of the atomic states should be incorporated. For arbitrary angular momenta, complexity of the problem necessitates a perturbation approach that excludes the saturation regime.[6] The simplest case of $J=1\rightarrow J=0$ transition permits a nonperturbative approach and has been extensively studied in the context of three-level spectroscopy.[3,7–13] In the transition scheme shown in Fig. 1 the right (σ_+) and left (σ_-) circularly polarized components of the incident beam interact with $m=\pm 1$ Zeeman sublevels of the ground state. Zeeman coupling between the σ_+ and σ_- modes leads to polarization effects such as circular dichroism and gyrotropic birefringence[14] and has found application in polarization spectroscopy.[15] Nonlinear absorption of an optical beam in such a Λ-type three-level system depends on its intensity as well as its state of polarization. However, no systematic study of the beam-polarization effects appears to have been performed in the saturation regime. A partial reason may be that a simple analytic expression for the medium susceptibility is not available for a three-level system interacting with two intense optical modes.[10–12]

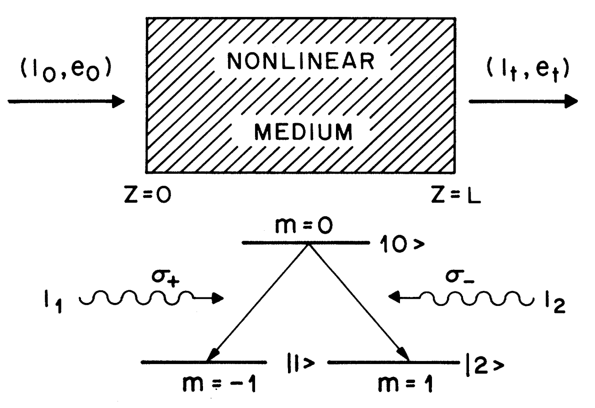

FIG. 1. Schematic illustration of the geometry, notation, and transition scheme for propagation of an elliptically polarized optical field in a three-level system, modeling $J=1\rightarrow J=0$ transition.

The purpose of this Rapid Communication is twofold. An analytical nonperturbative expression for the three-level susceptibility is presented assuming homogeneous broadening and exact resonance. It clearly manifests the role and the relative importance of self-saturation, cross saturation, two-photon saturation, and Zeeman coherence. The susceptibility is then used to study propagation of an arbitrarily polarized optical field inside a nonlinear medium. In general, polarization changes on propagation by an amount that depends on the beam intensity and is largest in the saturation regime. This is a direct consequence of level degeneracy since a two-level system exhibits no polarization effects.

The steady-state susceptibility is obtained by solving a set of nine density-matrix equations with phenomenological relaxation terms.[12,13] No severe restrictions are imposed on the population (longitudinal) decay rates γ_i and the atomic-coherence (transverse) decay rates γ_{ij} ($i,j=0$, 1, or 2) except for assuming that $\gamma_1=\gamma_2$ and $\gamma_{01}=\gamma_{02}$ because of sublevel degeneracy. In particular, the transverse decay rates

$$\gamma_{ij}=(\gamma_1+\gamma_2)/2+\gamma_{ij}^{\text{ph}} \tag{1}$$

are allowed to have a contribution γ_{ij}^{ph} from phase-interrupting collisons. In contrast to a previous three-level model,[10] this model allows for the effective population decay of the ground state owing to its finite interaction time. This is often the limiting factor in a typical experimental situation since atomic motion or collisional diffusion tends to move atoms outside the beam cross section.[16] Furthermore, this happens on a faster time scale (usually $\sim$few μs) than a slow collisional population transfer between Zeeman sublevels. Assuming homogeneous broadening and exact resonance, the susceptibility is found to be[13]

$$\chi_n=\frac{i\alpha_0}{k}\left(\frac{(1+\tilde{p}I_{3-n})}{1+(1+q)(I_1+I_2)+\tilde{p}(1+2q)I_1I_2}\right) , \tag{2}$$

where $k=\omega/c$, ω is the angular frequency of the incident light, α_0 is the small-signal absorption coefficient, and

$$\tilde{p} = p[1+p+(I_1+I_2)/2]^{-1} \tag{3}$$

is itself intensity dependent. The subscripts n equals 1 or 2 for σ_+ and σ_- polarizations, respectively. Various decay rates enter through two dimensionless parameters

$$p=\gamma_{12}^{\text{ph}}/\gamma_1, \quad q=\gamma_1/\gamma_0 \ . \tag{4}$$

The dimensionless intensity $I_n=|E_n|^2/I_s$, $I_s=2qI_{s0}$, and $I_{s0}=(\hbar^2\gamma_0\gamma_{01})/\mu^2$ is the two-level saturation intensity, μ being the transition dipole moment. Since $q \ll 1$ owing to the nonradiative nature of the ground-state decay, optical pumping reduces the three-level saturation intensity I_s by several orders of magnitude[16] in comparison to I_{s0}.

The intensity-dependent susceptibility χ_n clearly shows the coupling of σ_+ and σ_- polarization modes that leads to circular dichroism. It represents a generalization of the well-known two-level susceptibility[1-3] to which it reduces when only one mode is present. Several features of χ_n are worth noting. The saturation denominator in Eq. (2) has contributions from self-saturation, cross saturation, and two-photon saturation. Optical-pumping effects are manifested by the parameter $p=\gamma_{12}^{\text{ph}}/\gamma_1$ that governs the collisional relaxation of Zeeman coherence. The mode coupling decreases with an increase in p and vanishes for $p \rightarrow \infty$.

As an application of Eq. (2), consider propagation of an elliptically polarized optical field. After making the plane-wave and the paraxial approximations, the wave equation can be used to yield[1]

$$dE_n/dz=(ik/2)\chi_n(E_1,E_2)E_n \ . \tag{5}$$

In the absence of dispersive effects ($\text{Re}\chi_n=0$) owing to the assumption of exact resonance, the phase of E_n does not change during propagation (no birefringence). Using $I_n=|E_n|^2/I_s$ and Eq. (2) with $q \ll 1$, the coupled intensity equations are

$$\frac{dI_n}{dz}=-\frac{\alpha_0(I_n+\tilde{p}I_1I_2)}{1+I_1+I_2+\tilde{p}I_1I_2} \tag{6}$$

for $n=1,2$. The circular components I_1 and I_2 are related to the total intensity I and the ellipticity e of the polarization ellipse by[17]

$$I_{1,2}=\tfrac{1}{2}I[1 \pm 2e/(1+e^2)] \ . \tag{7}$$

Equations (6) can be used to obtain the transmitted intensity I_t and the ellipticity e_t at a distance L in terms of their boundary values I_0 and e_0 at $z=0$. The role of Zeeman coherence is evident since absorption depends on its collisional decay governed by the parameter p [see Eq. (4)]. The mode coupling is strongest when $p=0$. In this specific case Eqs. (6) can be solved analytically. Since the ratio I_1/I_2 remains constant, the ellipticity e does not change on propagation. The interesting point to note is that for finite values of p, the beam polarization changes on propagation and the amount of change depends on e_0. This is shown in Fig. 2 where I_t and e_t are plotted as a function of e_0 for several values of p at a fixed input intensity. In the limit $p \rightarrow \infty$, Zeeman coherence vanishes and the two polarization modes are uncoupled. Note that the extent of circular dichroism increases with p. For circularly polarized light

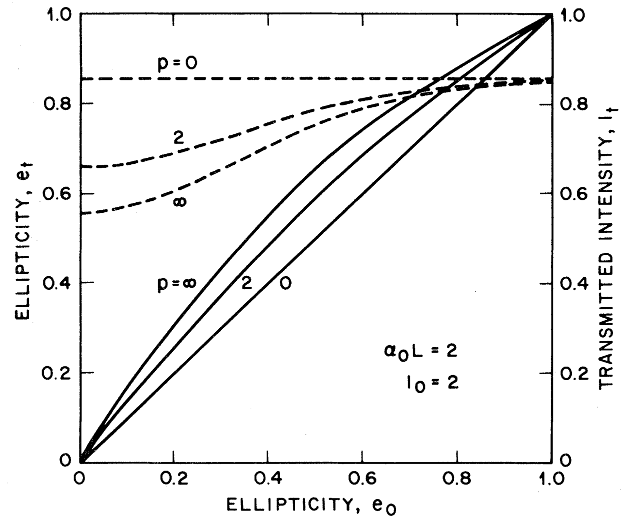

FIG. 2. Variation of the transmitted intensity I_t (dashed line) and the ellipticity e_t (full line) with the input ellipticity e_0 for three values of the parameter p that is a measure of the collisional decay of Zeeman coherence.

($e_0=1$), I_t and e_t are p independent since the three-level system reduces to a two-level system (Fig. 1). For linearly polarized light ($e_0=0$), Zeeman-coherence effects lead to p-dependent absorption. The transmitted light, however, remains linearly polarized because of an inherent symmetry with respect to the mode intensities [see Eq. (2)].

The effect of optical saturation on circular dichroism is illustrated in Fig. 3 where the transmittivity $T=I_t/I_0$ is plotted as a function of the incident intensity I_0 for different ellipticities e_0. For any state of polarization, the general behavior is reminiscent of a two-level system. In the linear regime ($I_0 \ll 1$), T is constant. In the saturation regime

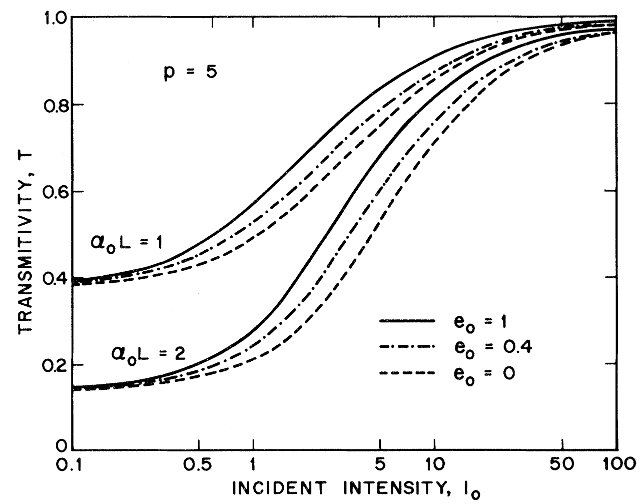

FIG. 3. Transmittivity $T=I_t/I_0$ as a function of the incident intensity I_0 (normalized to the saturation intensity) for three values of the ellipticity e_0. The cases $e_0=0$ and $e_0=1$ correspond to linear and right-circular polarizations, respectively.

($I_0 \gtrsim 1$), absorption is reduced so that T increases nonlinearly and eventually approaches its limiting value of unity when the medium is bleached. However, at a given I_0 the transmittivity T depends on the state of beam polarization and increases with e_0. Linearly polarized light suffers greater absorption since the Zeeman-coherence-induced mode coupling leads to cross saturation and two-photon saturation. The consideration of the output ellipticity e_t shows that it changes significantly (as much as by 40%) in the saturation regime ($I_0 \gtrsim 1$). This clearly implies that optical saturation plays an important role in circular dichroism.

For simplicity of discussion, we have assumed exact one-photon resonance. The present analysis can be extended to include dispersive effects that arise when ground-state degeneracy is removed by applying an external magnetic field. It is still possible to obtain an analytical expression for the medium susceptibility.[13] A new feature is that the σ_+ and σ_- polarization modes now generally have different refractive indices (gyrotropic birefringence). This leads to a rotation of the polarization ellipse together with the ellipticity change during beam propagation. Another distinctive feature is that nonlinear absorption as a function of the Larmor frequency Ω exhibits a narrow central dip in the Lorentzian profile that would be expected for a decoupled two-level system. This so-called nonabsorption resonance[10] is well known in three-level spectroscopy.[3] It arises from transverse optical pumping[10] that leads to coherent population trapping for $\Omega = 0$.

A major assumption of this work is related to homogeneous broadening, made to obtain a closed-form expression for the susceptibility given by Eq. (2). Strictly speaking, the analysis applies to atomic-beam experiments. The results presented here are, however, expected to remain qualitatively valid even for Doppler-broadened media since in folded three-level systems Doppler broadening plays a relatively minor role[3] for copropagating modes.

In conclusion, the effect of level degeneracy on saturated absroption in a resonant medium is investigated using a simple but realistic three-level model. In general, nonlinear absorption depends on the intensity as well as the polarization of the incident light. Zeeman-coherence-induced circular dichroism is analyzed and discussed. The results suggest that nonlinear polarimetry may provide a useful tool to investigate atom-radiation interaction. An experimental verification of these results would be of considerable interest. A suitable atomic medium is samarium with $6s^2\,{}^7F_1 \rightarrow 6s6p\,{}^7F_0$ transition at 570.7 nm. The present work is useful in accessing the importance of polarization effects during wave propagation in atomic media. In particular, it may find application in high-resolution spectroscopy.[3,8] Our analytical approach should also be useful in other branches of nonlinear optics such as optical bistability and phase conjugation.[13]

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