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Propagation of light in a $J=1/2 \leftrightarrow J'=1/2$ resonant medium

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The propagation of light in a resonant medium with multiple sublevels depends on the populations of and coherences between theses sublevels. We analyze the case of a $J=1/2 \leftrightarrow J'=1/2$ transition with arbitrary ground-state polarization in the limit of a weak probe beam. It is found that transverse components of the ground-state orientation lead to a longitudinal component in the eigenpolarizations; a laser beam propagating through an optically pumped medium should therefore be displaced laterally.

1. Introduction

Polarized light propagation through a resonant medium induces in general not only optical polarizations in the medium, but in the frequently encountered case of multiple substates of the electronic ground state also population differences and coherence between the sublevels. These sublevel coherences give rise to many interesting magneto-optical and nonlinear optical effects; more recently they have been used in optical cooling experiments [1]. The population differences and coherences induced in the atomic medium, on the other hand, also influence the optical properties of the medium. Since the effect occurs even for arbitrarily weak optical fields, which have a negligible effect on the system, it is possible to probe the sublevel dynamics by a weak laser beam, using polarization-selective detection of the transmitted beam [2].

The creation of order among a manifold of sublevels occurs usually by some variation of an optical pumping process. The basic requirements are that the absorption of photons from the laser field depends on the internal state of the atom and that the probabilities for stimulated absorption and spontaneous emission are different. Apart from giving rise to sublevel coherence, the optical field also influences their dynamics: absorption (i.e. optical pumping) leads to a loss of phase memory while dispersive effects lead to virtual level shifts via the so-called light-shift effect [3]. All these processes can in principle be observed via an optical probe beam and indeed have been [4].

Several theoretical analyses exist [5] for the case of axial symmetry, taking ground-state polarization parallel to the direction of propagation of the laser beam into account. If we quantize the atomic system along this axis, the longitudinal ground-state polarization corresponds to a population difference between the ground state sublevels. If the cylindrical symmetry is broken, e.g. by a magnetic field perpendicular to the laser beam, or by the presence of multiple beams, the ground state polarization is in general not parallel to the laser beam, and the influence of transverse components must also be taken into account. This article gives a detailed analysis of the propagation of light of arbitrary polarization in a resonant medium with a $J=1/2 \leftrightarrow J'=1/2$ electronic transition and arbitrary ground state polarization.

2. Anisotropic optical susceptibility

The atomic model system used for the calculation is represented schematically in fig. 1: the atom is assumed to interact with the radiation field only by two sets of eigenstates of the free-atom hamiltonian which are separated roughly by the optical photon energy. The ground state and the electronically excited state are both $J=1/2$ states; important practical examples for such transitions are the D_1 resonance lines of alkali atoms. For this figure, the quantization axis is

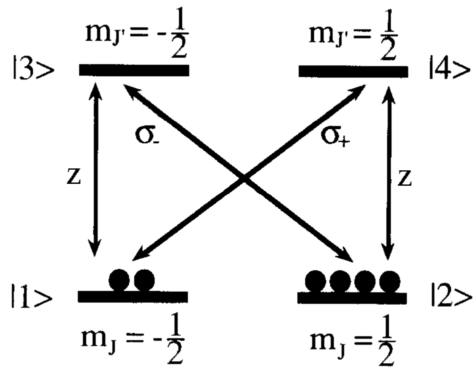

Fig. 1. Schematic representation of the atomic level system interacting with the optical field. Ground- and excited state are both assumed to have an angular momentum $J=\hbar/2$. The quantization axis is chosen parallel to the k-vector of the travelling wave. The transitions labelled $\sigma_\pm$ interact with circularly polarized light while the transitions labelled "z" couple to the longitudinal modes.

chosen parallel to the k-vector of the travelling wave. The transitions labelled $\sigma_\pm$ correspond to the interaction with circularly polarized light while the transitions labelled "z" couple to the longitudinal modes of the field.

We are interested in the case where only the ground state is populated and a weak electromagnetic wave with a frequency near the resonance frequency of the optical transition travels through the medium. The experiments that we have in mind are pump and probe experiments where the medium is polarized by a strong pump beam and a weak probe beam is used to observe the resulting dynamics of the coherences between the different ground state sublevels [2]. The intensity of the probe beam is then chosen such that it does not significantly affect the sublevel dynamics.

In the absence of the optical proble beam, the system can be described by three real parameters. A convenient choice of these parameters is to make them proportional to the cartesian component of the ground state magnetization:

$$\boldsymbol{m}(m_x, m_y, m_z) = (\rho_{12}+\rho_{21}, -\mathrm{i}(\rho_{12}-\rho_{21}), \rho_{22}-\rho_{11}) , \qquad (1)$$

where ρ_{ij} represent the density operator elements and the substates are labelled as shown in fig. 1. The density operator components are then

$$\rho_{11}=(1-m_z)/2 , \quad \rho_{22}=(1+m_z)/2 ,$$
$$\rho_{12}=\rho_{21}^*=(m_x+\mathrm{i}m_y)/2 . \qquad (2)$$

With the quantization axis parallel to the k-vector of the field, m_z represents therefore a ground-state polarization parallel to the direction of propagation, while m_x and m_y are the orthogonal components. The hamiltonian includes only the interaction between the atom and the applied laser field. With the conventional choice of phases for the individual states [6], the matrix representation of the electric dipole moment operator is

$$\mu_x=\mu_\mathrm{E}\begin{pmatrix}0&0&0&1\\0&0&1&0\\0&1&0&0\\1&0&0&0\end{pmatrix}, \quad \mu_y=\mu_\mathrm{E}\begin{pmatrix}0&0&0&\mathrm{i}\\0&0&-\mathrm{i}&0\\0&\mathrm{i}&0&0\\-\mathrm{i}&0&0&0\end{pmatrix},$$

$$\mu_z=\mu_\mathrm{E}\begin{pmatrix}0&0&-1&0\\0&0&0&1\\-1&0&0&0\\0&1&0&0\end{pmatrix}, \qquad (3)$$

where μ_E represents the reduced electric dipole moment. The atom interacts with a time-harmonic field of arbitrary direction, $\boldsymbol{E}=\{E_x, E_y, E_z\}$. The elements of the hamiltonian in the rotating wave approximation are then

$$\mathscr{H}=\begin{pmatrix}0&0&-\beta_0&\beta_+\\0&0&\beta_-&\beta_0\\-\beta_0^\dagger&\beta_-^\dagger&\delta&0\\\beta_+^\dagger&\beta_0^\dagger&0&\delta\end{pmatrix}, \qquad (4)$$

with $\beta_\pm=\mu_\mathrm{E}(E_x\pm\mathrm{i}E_y)$, $\beta_0=\mu_\mathrm{E}E_z$, and δ represents the detuning of the optical frequency ω above the atomic resonance frequency ω_0. We calculate now the derivative of the equilibrium density operator as

$$\dot\rho=-\mathrm{i}[H,\dot\rho] \qquad (5)$$

which evaluates to

$$\dot\rho_{13}=(\mathrm{i}/2)[\beta_- m_+ -\beta_0(1-m_z)] ,$$
$$\dot\rho_{14}=(\mathrm{i}/2)[\beta_+(1-m_z)+\beta_0 m_+] ,$$
$$\dot\rho_{23}=(\mathrm{i}/2)[\beta_-(1+m_z)-\beta_0 m_-] ,$$
$$\dot\rho_{24}=(\mathrm{i}/2)[\beta_+ m_- +\beta_0(1+m_z)] , \qquad (6)$$

where $m_\pm=m_x\pm\mathrm{i}m_y$. Only the optical coherences are affected in this approximation, while the derivatives of the populations and the Zeeman coherences vanish. The main effect of the population difference m_z is an apparent change of the number of atoms. As is well known [5], this leads to circular dichroism and circular birefringence for a wave propagating parallel to the quantization axis. The main effect of the sublevel coherence $m_\pm$, which is omitted in most treatments, is a coupling between the different field modes: in the presence of sublevel coherences, the transverse field components E_x, E_y induce polarization not only in the σ-transitions $1 \leftrightarrow 4$ and $2 \leftrightarrow 3$, but also in the π-transitions $1 \rightarrow 3$ and $2 \leftrightarrow 4$, while the z-component of the field also couples to the σ-transitions.

In order to calculate the steady-state polarization, we have to include also the effects of relaxation and the detuning of the laser frequency from optical resonance. Within the approximations made, all four optical coherences are affected in the same way by these effects:

$$\dot{\rho}_{ij} = (\pm \mathrm{i}\delta - \Gamma)\rho_{ij} , \qquad (7)$$

where δ represents the resonance detuning and Γ the optical dephasing rate; the plus-sign applies above the diagonal, the minus-sign below. To first order in the optical field, the coherences evolve then towards the steady-state values

$$\begin{aligned} \rho_{13\infty} &= (1/2)[\beta_- m_+ - \beta_0(1-m_z)]g(\delta) , \\ \rho_{14\infty} &= (1/2)[\beta_+(1-m_z) + \beta_0 m_+]g(\delta) , \\ \rho_{23\infty} &= (1/2)[\beta_-(1+m_z) - \beta_0 m_-]g(\delta) , \\ \rho_{24\infty} &= (1/2)[\beta_+ m_- + \beta_0(1+m_z)]g(\delta) , \end{aligned} \qquad (8)$$

where $g(\delta)$ represents the lineshape function

$$g(\delta) = (-\delta + \mathrm{i}\Gamma)/(\delta^2 + \Gamma^2) . \qquad (9)$$

In cartesian coordinates, the optical polarization per atom is therefore

$$\boldsymbol{P} = \mathrm{Tr}(\boldsymbol{\mu}_\mathrm{E}\rho_\infty) = \epsilon_0\chi_0(\delta)\boldsymbol{E} + \mathrm{i}\epsilon_0\chi_0(\delta)\boldsymbol{m}\times\boldsymbol{E} , \qquad (10)$$

where $\chi_0(\delta) = 2\mu_\mathrm{E}^2 g(\delta)/\epsilon_0$ represents the susceptibility of the unpolarized medium ($\boldsymbol{m}=0$). The effect of the sublevel polarization $\boldsymbol{m}$, represented by the last term in eq. (10) is therefore a rotation of the optical polarization with respect to the electric field. In tensor notation, the optical susceptibility is then $\boldsymbol{P} = \epsilon_0\chi(\delta)\boldsymbol{E}$ with

$$\chi(\delta) = \chi_0(\delta)\begin{pmatrix} 1 & -\mathrm{i}m_z & \mathrm{i}m_y \\ \mathrm{i}m_z & 1 & -\mathrm{i}m_x \\ -\mathrm{i}m_y & \mathrm{i}m_x & 1 \end{pmatrix} . \qquad (11)$$

3. Eigenpolarizations of plane waves

Using this susceptibility tensor, we may now solve Maxwell's equations for the propagation of electromagnetic waves in the anisotropic medium. We look for harmonic plane-wave solutions with the wavevector parallel to the z-direction

$$\begin{aligned} \boldsymbol{E} &= \mathrm{Re}[\{E_x, E_y, E_z\}\exp[\mathrm{i}(\omega t - k_z z)]] , \\ \boldsymbol{H} &= \mathrm{Re}[\{H_x, H_y, H_z\}\exp[\mathrm{i}(\omega t - k_z z)]] , \\ \boldsymbol{D} &= \epsilon_0(1+\chi)\cdot\boldsymbol{E} , \quad \boldsymbol{B} = \mu_0\boldsymbol{H} , \end{aligned} \qquad (12)$$

where we have assumed that the magnetic properties of the medium can be neglected ($\mu = 1$). Starting from the "curl"-Maxwell equations

$$\nabla\times\boldsymbol{H} = \frac{\partial}{\partial t}\boldsymbol{D} , \quad \nabla\times\boldsymbol{E} = -\frac{\partial}{\partial t}\boldsymbol{B} , \qquad (13)$$

we eliminate the magnetic field

$$\begin{pmatrix} H_x \\ H_y \\ H_z \end{pmatrix} = \frac{k_z}{\mu_0\omega}\begin{pmatrix} -E_y \\ E_x \\ 0 \end{pmatrix} \qquad (14)$$

to get the following equation for the electric field amplitudes:

$$\begin{pmatrix} E_x \\ E_y \\ 0 \end{pmatrix}\left(\frac{ck_z}{\omega}\right)^2 = \boldsymbol{E}(1+\chi_0) - \mathrm{i}\chi_0\boldsymbol{E}\times\boldsymbol{m} . \qquad (15)$$

This equation has two independent solutions

$$\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} = E_0 \times \begin{bmatrix} 1+\chi_0\left(1+\mathrm{i}\dfrac{m_x m_y}{m_z}\right) \\ \mathrm{i}\left(\pm r_1 - \chi_0 \dfrac{m_x^2 - m_y^2}{2m_z}\right) \\ \dfrac{\chi_0}{1+\chi_0}\left(\pm m_x r_1 + \mathrm{i} m_y (1+\chi_0) - \chi_0 m_x \dfrac{m_x^2 + m_y^2}{2m_z}\right) \end{bmatrix} \tag{16}$$

where E_0 represents the (arbitrary) amplitude. The constant r_1 is defined as

$$r_1 = \left[(1+\chi_0)^2 + \chi_0^2 \left(\frac{m_x^2 + m_y^2}{2m_z}\right)^2 \right]^{1/2}, \tag{17}$$

and the wavevectors corresponding to the eigenpolarizations are

$$k_z^2 = \left(\frac{\omega}{2}\right)^2 \left[1+\chi_0 + \frac{\chi_0}{1+\chi_0}\left(\pm m_z r_1 - \chi_0 \frac{m_x^2 + m_y^2}{2}\right)\right]. \tag{18}$$

In the typical case of a low-density atomic gas where $\chi_0 \ll 1$, these formulas can be simplified considerably by neglecting terms of order >1 in χ_0:

$$\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}_{\pm} \approx \begin{pmatrix} 1 \\ \mathrm{i}(\pm 1 - \chi_0 m_{\pm}^2 / 2m_z) \\ \pm \chi_0 m_{\pm} \end{pmatrix},$$

$$k_z \approx \frac{\omega}{c}\left(1+\chi_0 \frac{1 \pm m_z}{2}\right), \tag{19}$$

or

$$\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix}_{\pm} \approx \begin{bmatrix} 1+\chi_0\left(1+\mathrm{i}\dfrac{m_x m_y}{m_z}\right) \\ \mathrm{i}\left[\pm 1 + \chi_0\left(\pm 1 - \dfrac{m_x^2 - m_y^2}{2m_z}\right)\right] \\ \pm \chi_0 m_{\pm} \end{bmatrix}, \tag{19'}$$

where the overall amplitude differs by a fraction of order χ_0. Both forms are equivalent; which form is used depends on the particular problem at hand.

4. Discussion

In the case of cylindrical symmetry, i.e. $m_{\pm} = 0$, the above formulas reduce to the earlier results [5], where the eigenpolarizations are circular and the index of refraction depends linearly on the population difference m_z of the ground state sublevels. If the medium is not cylindrically symmetric ($m_{\pm} \neq 0$), the polarizations become slightly elliptical and the longitudinal component E_z becomes nonzero; both effects scale with χ_0 and the transverse magnetization component $m_{\pm}$. The Poynting-vector is then no longer parallel to the k-vector and a laser beam propagation through such a medium is laterally displaced.

For a brief investigation of the size of this effect and the possibility to observe the associated beam displacement, we calculate the Poynting vector in the linear regime as

$$\frac{4\pi}{c}\boldsymbol{S} = \boldsymbol{E} \times \boldsymbol{H} = \frac{k_z}{\mu_0 \omega}\begin{pmatrix} E_x \\ E_y \\ E_z \end{pmatrix} \times \begin{pmatrix} -E_y \\ E_x \\ 0 \end{pmatrix}$$

$$= \frac{k_z}{\mu_0 \omega}\begin{pmatrix} -E_x E_z \\ -E_y E_z \\ E_x^2 + E_y^2 \end{pmatrix} \approx \frac{k_z}{\mu_0 \omega}\begin{pmatrix} \mp \chi_0 m_{\pm} \\ \mp \mathrm{i} \chi_0 m_{\pm} \\ 2 \end{pmatrix}. \tag{20}$$

The direction of propagation deviates therefore from the z-axis by an angle of the order of $\chi_0 m_{\pm}$, i.e. proportional to the atomic density, the transition strength and the transverse ground-state orientation. A laser beam propagating through a completely polarized medium of length l would therefore be displaced by an amount of the order of $\chi_0 l$. Since the beam is attenuated in the medium by $\approx \exp[-\mathrm{Im}(\chi_0) l/\lambda]$, a measurement of the beam displacement requires $\mathrm{Im}(\chi_0) \sim \lambda / l$ so that the observable beam displacement Δ is limited to $\Delta \leqslant (\lambda / l [\mathrm{Re}(\chi_0)/\mathrm{Im}(\chi_0)]$.

In summary, we have shown that the propagation of light in a resonant medium can be strongly affected by the order present among the sublevels of the atomic ground state, even for arbitrarily weak light. In the practically important case of a $J=1/2 \leftrightarrow J'=1/2$ atomic transition and arbitrary ground state polarization, analytical solutions of the Maxwell equations have been obtained in the limit of low irradiation. The analysis does not assume axial symmetry and is therefore of special interest in cases where this symmetry is broken, e.g. by the presence of transverse magnetic fields [1,4], multiple laser beams [1] or broken symmetry of the medium itself, e.g. by the presence of a dielectric interface [7]. In these cases, the analysis sheds some light on the dynamics of the atomic system coupled to the resonant light field and shows how an optical probe beam can be used to monitor them.

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References

[1] See, e.g., A. Aspect, E. Arimondo, R. Kaiser, N. Vansteenkiste and C. Cohen-Tannoudji, Phys. Rev. Lett. 61 (1988) 826; S.Q. Shang, B. Sheehy, H. Metcalf, P. van der Straten and G. Nienhuis, submitted to Phys. Rev. Lett. (1991).
[2] J. Mlynek and W. Lange, Optics Comm. 30 (1979) 337; D. Grison, B. Lounis, C. Salomon, J.Y. Courtois and G. Grynberg, Europhys. Lett. 15 (1991) 149.
[3] C.Cohen-Tannoudji and J. Dupont-Roc, Phys. Rev. A 5 (1972) 968.
[4] See, e.g., D. Suter and J. Mlynek, Adv. Magn. Opt. Reson. 16 (1991), and references therein.
[5] S. Nakayama, J. Phys. Soc. Jap. 50 (1981) 608; A.W. McCord and R.J. Ballagh, J. Opt. Soc. Am. B 7 (1990) 73.
[6] M. Weissbluth, Atoms and molecules (Academic Press, San Diego, 1988).
[7] D. Suter, J. Äbersold and J. Mlynek, Optics Comm. 84 (1991) 269.