Laser bleaching modified by a local field

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A local field can radically change the shape of spectral lines which stem from an interference of quantum transitions. The effect is seen at particle densities on the order of 10^{17} cm^{-3}.

1. The interaction of optical radiation of various types with three-level systems can result in a splitting of resonances in an intense electromagnetic field, and it can give rise to a transparency at the frequency of an unperturbed resonance.[1] Furthermore, the resonant interference of quantum transitions leads to a difference between the interactions of probe radiation with absorbing and emitting atoms.[1] This circumstance may be accompanied by an amplification of the radiation without a population inversion.[1–3] Interference may lead to an increase in a nonlinear susceptibility and a simultaneous decrease in the absorption of short-wave radiation which is generated.[4,5] A splitting of optical transitions and a bleaching were observed experimentally in Refs. 6 (see also the papers cited there), while an amplification without a population inversion has been seen in several studies, cited in Ref. 7, among other places. Recent years have seen a sharp increase in interest in nonlinear interference effects in quantum optics, in connection with the goal of developing lasers which operate without a population inversion and in connection with experiments on the generation of short-wave radiation through the use of bleaching effects.[5] Over the past year, several experimental studies of amplification without a population inversion in the optical region have been published (e.g., Ref. 8).

In the present letter we use a simple example to show that an increase in the particle density under the conditions of these experiments may be accompanied by the onset of a qualitatively new spectral behavior because of the onset of a local field which differs from the external field. We also show that the local field at the frequency of a weak probe beam of electromagnetic radiation changes radically in the field of an intense auxiliary laser beam on an adjacent transition.

2. We know that an individual atom in a dense medium is acted upon by a field which is not the same as the electromagnetic radiation supplied by the external source.[9] This phenomenon is dealt with most simply by means of the standard (but approximate) concept of a local field (Ref. 9, for example), according to which the local field E_L and the external field E in an isotropic medium are related by the simple equation

$$E_L = E + P/3\epsilon_0, \tag{1}$$

where P is the polarization of the medium, which can be written $P = \epsilon_0 N \alpha E_L$ in the linear approximation; N is the density of atoms; α is the microscopic (atomic) polarizability; and ϵ_0 is the permittivity of free space.

One of the best known consequences of (1) is the Clausius–Mossotti equation,[9] which relates α to the permittivity of the medium, ϵ :

$$\epsilon = 1 + LN\alpha. \tag{2}$$

Here $L = (\epsilon+2)/3 = (1-\alpha N/3)^{-1}$ is a local-field factor, which tells us the factor by which the local field differs from the external field.

The local field plays an important role in linear and nonlinear optical phenomena (see, for example, Refs. 10–13). Although the derivation of (1) is only approximate, Maki *et al.*[10] have shown that this equation gives a good description of the linear and nonlinear optical responses of a dense atomic gas.

Let us consider the interaction of two optical beams with three-level systems of the cascade or Λ type. We assume that one of these fields is strong, and for simplicity we assume that it interacts with a transition between levels which are unpopulated or which have equal populations. According to the classification of resonant nonlinear processes,[1] only a splitting of the resonance for the probe field (bleaching by an electromagnetic field) will be exhibited in this case. For definiteness we consider a Λ scheme, in which the levels satisfy the conditions $E_3 > E_2 > E_1$. The strong field, with the frequency ω and the amplitude E, acts on the $2 \rightarrow 3$ transition. The weak (probe) field, with the frequency ω_μ and the amplitude E_μ, acts on the $1 \rightarrow 3$ transition. Levels 2 and 3 are unpopulated; level 1 is the ground state.

In the steady state, at an accuracy to first order in E_μ, the equations for the density matrix are $(\rho_1 = 1)$

$$\begin{aligned} \Delta_{31}\rho_{31} &= -iG_{\mu L} - iG_L\rho_{21}, \\ \Delta_{21}\rho_{21} &= -iG_L^*\rho_{31} + iG_{\mu L}\rho_{32}^*, \\ \Delta_{32}\,\rho_{32} &= -iG_{\mu L}\rho_{21}^*. \end{aligned} \tag{3}$$

Here $\Delta_{31} = \Gamma_{31} - i\delta_1$; $\delta_1 = \omega_\mu - \omega_{31}$; $\Delta_{32} = \Gamma_{32} - i\delta_2$; $\delta_2 = \omega - \omega_{32}$; $\Delta_{21} = \Gamma_{21} - i(\delta_1 - \delta_2)$; Γ_{ij} $(i, j = 1, 2, 3)$ are the transition half-widths; d_{31} and d_{32} are the transition dipole matrix elements; $G_L = -d_{32}E_L/\hbar$ and $G_{\mu L} = -d_{31}E_{\mu L}/\hbar$ are the Rabi frequencies incorporating the local fields; $E_L = E + P(\omega)/3\epsilon_0$; and $E_{\mu L} = E_\mu + P_\mu(\omega_\mu)/3\epsilon_0$. For simplicity we will incorporate below only the resonant parts of the polarizations $P(\omega)$ and $P_\mu(\omega_\mu)$ at the corresponding frequencies.

System (3) differs from the corresponding system in Ref. 1 only in that E_L is used in place of the external field E. This is a very common approach in local-field theory.[11,12]

We will be interested below in the absorption of the radiation at the frequency of the probe field, ω_μ. We accordingly write a solution for the element ρ_{31}, which is responsible for the absorption and the dispersion of the probe field:

$$\rho_{31} = -iG_{\mu L}\frac{\Delta_{21}}{\Delta_{31}\Delta_{21} + |G|^2}. \tag{4}$$

Here we have used $G_L = G = -d_{32}E/2\hbar$, $G_{\mu L} = G_\mu - d_{31}P_\mu(\omega_\mu)/3\epsilon_0\hbar$, and $G_\mu = -d_{31}E_\mu/\hbar$.

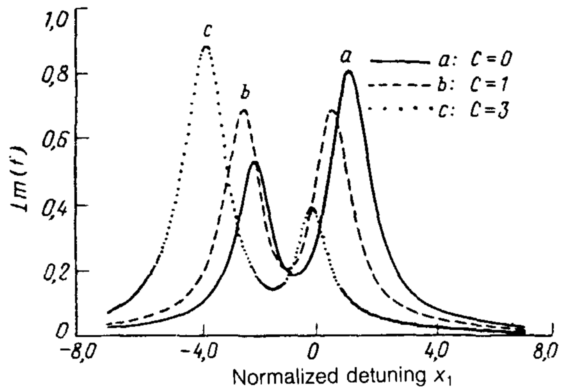

FIG. 1. Absorption of a probe field, Im(f), versus the deviation of the frequency of this field from resonance (in units of the line half-width: $x_1=\delta_1/\Gamma_{31}$) for $x_2=\delta_2/\Gamma_{21}=-2$, $q=\Gamma_{31}/\Gamma_{21}=2$, $G^2/(\Gamma_{31}\Gamma_{21})=4$, and various values of $C=\delta_{1L}/\Gamma_{31}$: a—0; b—1; c—3.

For the macroscopic complex polarization $P_\mu(\omega_\mu)=d_{13}\,\rho_{31}N$ we find the following expressions, using (4):

$$P_\mu(\omega_\mu)=\epsilon_0\chi_\mu(\omega_\mu)E_\mu,\quad \chi_\mu(\omega_\mu)=\chi_\mu^0\,f(\omega_\mu), \tag{5}$$

where

$$\chi_\mu^0=i\,\frac{N|d_{31}|^2}{\hbar\epsilon_0\Gamma_{31}},\quad f(\omega_\mu)=\frac{\Gamma_{31}\Delta_{21}}{(\Delta_{31}-i\delta_{1L})\Delta_{21}+|G|^2}, \tag{6}$$

χ_μ is the macroscopic susceptibility at the frequency of the probe field (ω_μ) in the presence of a strong field at the frequency ω, χ_μ^0 is the resonant value of this susceptibility in the absence of the strong field, and $f(\omega_\mu)$ is a form factor. The parameter

$$\delta_{1L}=|d_{31}|^2N/3\epsilon_0\,\hbar \tag{7}$$

arises as a frequency shift of the 3 → 1 transition caused by an increase in the density (caused by the local field). Significantly, the frequencies of the two-photon transition and of the strong-field transition do not change in the process. As a result, the effect of the local field does not reduce to a revision of the excursion of the weak field from resonance; it instead causes a qualitative change in the entire shape of the spectral line if this factor becomes comparable to the width of the resonance.

3. Let us calculate the parameter $C=\delta_{1L}/\Gamma_{31}$ for the case of a dense gas. With $d_{31}=1$ D and $N=10^{23}$ m^3 we estimate the shift to be $\delta_{1L}=8\times10^{11}$ s^{-1}. This shift may be comparable to the characteristic collisional widths of resonances. For example, if the collisional width is determined by a resonant exchange (a self-broadening), and if this width is much greater than the natural width, the expression for Γ_{31} becomes $\Gamma_{31}\approx|d_{31}|^2N/6\epsilon_0\hbar$ (see, for example, Ref. 11 and the papers cited there). It follows that the ratio δ_{1L}/Γ_{31} may approach 2 in this case.

4. Figures 1 and 2 show features of the local-field effect in the spectra of Im(f), which describes the absorption. The dip in Fig. 1 and the corresponding bleaching stem from a splitting of the 3–1 transition by the strong field.[1,2,6] The depth of this dip is determined by the value of the parameter G: the larger G, the weaker the absorption

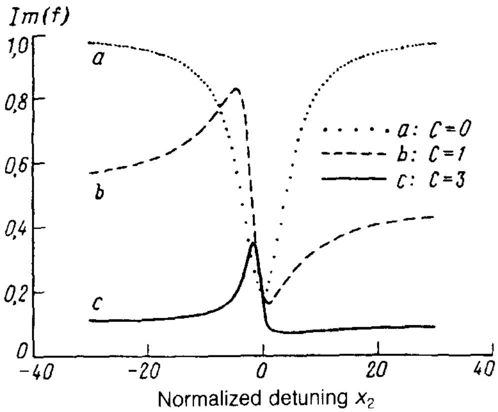

FIG. 2. Absorption of the probe field, Im(f), versus the detuning of the frequency of the strong field from resonance (in units of the line half-width: $x_2=\delta_2/\Gamma_{21}$) for $x_1=0$, $q=2$, $G^2/(\Gamma_{31}\Gamma_{21})=4$, and various values of the parameter C: a—0; b—1; c—3.

at the center of the dip. The increase in the local field with increasing density of absorbing particles leads to a qualitative change in the shape of the curves. Figure 2 demonstrates another characteristic dependence, which could easily be observed experimentally: a change in the absorption of the probe field at a fixed frequency upon a change in the frequency of the strong field. In this case the change in the shape of the curve is even more radical: from a symmetric, Lorentz-like absorption lineshape at $C=0$ to a dispersive lineshape at $C=1$ and then, with increasing C, to a Fano-like lineshape.[14] The local field thus causes pronounced changes (quantitative and qualitative) in the spectra of the absorption of the probe field in the presence of a strong field on an adjacent transition.

Let us consider the factor $L_\mu=E_{\mu L}/E_\mu$, which is a measure of the deviation of the local field from the external field in magnitude and phase:

$$L_\mu=1+iCf(\omega_\mu). \tag{8}$$

This difference also increases with increasing C.

Figure 3 shows L_μ as a function of the normalized frequency detuning of the probe field, x_1, for various values of the parameter G. The additional oscillations on

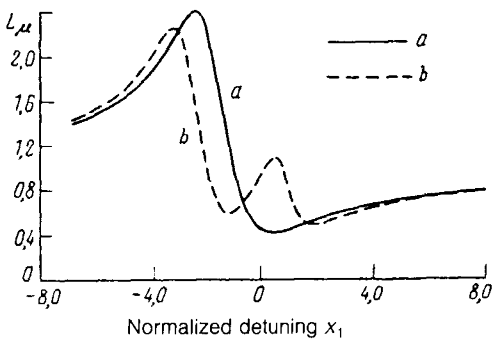

FIG. 3. Modulus of L_μ versus the normalized frequency detuning of the probe field, x_1, for various values of the parameter G. a—$G=0$, $C=2$; b— $G^2/(\Gamma_{31}\Gamma_{21})=4$, $C=2$, $q=2$.

curve b stem from a splitting of level 3 due to the strong field on the adjacent transition. It is thus possible to control the spectrum of the local field by varying the magnitude and frequency of the strong field.

Resonant exchange also leads to a shift of the frequency of the transition from the ground state to an excited state. This shift is also proportional to the density of atoms, but it is usually smaller than the broadening by a factor of 2 or 3 (Ref. 15). This shift can thus often be ignored.

The results derived here are quite general, applying to other interaction schemes also. In the case of a cascade scheme of transitions, if the populations of the levels involved in the strong-field transition are zero or equal, the results turn out to be a simple revision of the corresponding quantities.

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