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Light-induced gain and directional energy flow with counterpropagating light beams in dense media

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Counterpropagation of two light beams in a resonant medium so dense that the local driving field differs from the external field is considered. It is shown that efficient energy transfer between the two beams occurs if there is some asymmetry between their propagation conditions due to, e.g., their frequency difference or Doppler detuning caused by the motion of the medium. The directional energy flow may predominate over absorption and lead to gain.

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Phenomena caused by local fields in optically dense atomic media are of growing interest because of their possible applications in optoelectronics and spectroscopy. Physically, the difference between an externally applied field and a local electric field, driving atoms or molecules in the medium, may be explained as the consequence of the dipole-dipole interaction between resonant centers. The difference can be calculated with the two-level approximation, which is reasonable for many optically susceptible media, including semiconductors [1]. This difference can be expressed via the Lorentz-Lorenz relation [2] for the local field $\mathbf{E}^L$,

$$\mathbf{E}^L = \mathbf{E} + \frac{4\pi}{3}\mathbf{P}, \tag{1}$$

where $\mathbf{E}$ is the externally applied electric field and $\mathbf{P}$ is the volume polarization. The dipole-dipole interaction can significantly affect the light-atom dynamics when the atomic density is sufficiently high. In the case of linear interaction of weak light beams with the medium, the main consequence of the local-field effect is a spectral shift of the absorption line [3]. The value of this shift,

$$\sigma = 4\pi\mu^2 N_0 / 3\hbar\gamma_2 \tag{2}$$

(μ and N_0 are the dipole moment and the density of resonant centers, respectively) in units of the transition linewidth γ_2, may be considered as the characteristic parameter of the influence of the local field. In experiments with vapors, σ cannot reach values higher than several units, because with the increase of atomic densities the collisional self-broadening of the line occurs ($\gamma_2 \propto N_0$). It might be possible to avoid this restriction by using an atomic beam technique. Media, where high values of σ should be available, seem to be Rydberg atoms, activated crystals, and semiconductors, because of their large dipole moments.

For arbitrarily strong light fields, the frequency shift due to the local field is proportional to the population difference between the resonant levels, which makes the Bloch equations nonlinear with respect to the atomic variables [4]. This fact is the cause of intrinsic optical bistability (IOB) [1,4], i.e., a bistable dependence of the atomic variables upon the applied external field intensity. Light propagation through a medium exhibiting IOB can result in a spatial first-order phase transition that separates regions of high and low excitation of the atomic medium [5–7]. The boundary between the two phases gives rise to nonzero reflectivity in backward phase conjugation, which might be useful for studying internal switching characteristics [6].

In this work we are studying the effect of IOB on the dynamics of two arbitrarily strong light beams counterpropagating in the dense medium. We predict the effect of light-induced gain, which occurs at a certain ratio between the beam intensities, when the modulation depth of the light interference pattern exceeds some threshold value. The direction of the energy transfer between the two fields is determined by the sign of the small frequency detuning between them or, equivalently, by a slow motion of the medium along the direction of light propagation. Such directional energy flow can be efficient enough to overcome absorption, which is particularly strong for media exhibiting the IOB. As the direction of the energy transfer is sensitive only to the sense of the motion of the medium, this phenomenon could find applications in mobility detectors and, possibly, in neutral atom trapping.

The Bloch equations for the polarization amplitude P and the population difference N governing the two-level atomic dynamics read, in the slowly-varying envelope approximation,

$$\frac{dP}{dt} + [\gamma_2 - i(\omega - \omega_0)]P = -i\frac{\mu^2}{\hbar}NE^L,$$

$$\frac{dN}{dt} + \gamma_1(N + N_0) = \frac{i}{2\hbar}(E^L P^* - E^{L*}P), \tag{3}$$

where ω and ω_0 are the field and transition frequencies, γ_1 and γ_2 are the population and coherence relaxation rates, respectively, and E^L is the amplitude of the local driving field.

After substitution of the Lorentz-Lorenz expression (1) into (3) we get the following dimensionless system:

$$\frac{d\mathscr{P}}{d\tau} + [1 - i(\delta - \sigma n)]\mathscr{P} = -n\mathscr{E},$$

$$\frac{dn}{d\tau} + b(n+1) = \frac{1}{2}(\mathscr{E}\mathscr{P}^* + \mathscr{E}^*\mathscr{P}), \tag{4}$$

where we have introduced the following substitutions:

$$P=i\mu N_0\mathscr{P}, \quad N=nN_0, \quad E=\frac{\hbar\gamma_2}{\mu}\mathscr{E},$$

$$\tau=\gamma_2 t, \quad \delta=(\omega-\omega_0)/\gamma_2, \quad b=\gamma_1/\gamma_2.$$

The steady-state ($d/d\tau=0$) solutions $\mathscr{P}_s, n_s$ of (4) may be found from the following expressions:

$$\mathscr{P}_s=\frac{-n_s\mathscr{E}}{1-i\Delta}, \quad n_s=-\frac{1+\Delta^2}{1+\Delta^2+|\mathscr{E}|^2/b}, \quad \Delta=\delta-\sigma n_s. \tag{5}$$

The expressions (5) have the same form as those of the standard Bloch equations without the local-field correction. The only, but very important, difference is that δ is replaced by the renormalized detuning Δ, which depends on the population difference n_s. Thus the solutions (5) exhibit a bistable dependence upon the intensity $I=|\mathscr{E}|^2$ of the external field, which is illustrated in Fig. 1(c) The change of the field intensity I with propagation distance due to the presence of absorption can produce a switch from the upper branch of the S-shaped bistability curve to the lower one [see Fig. 1(c)], resulting in the formation of two coexisting high- and low-excitation domains. In Ref. [5] this effect was considered as a spatial first-order phase transition.

If the net field $\mathscr{E}(z)$ consists of two components propagating in opposite directions $\pm z$, the resulting intensity interference pattern $|\mathscr{E}(z)|^2$ [Fig. 1(a)] should lead to the establishment of the population and polarization gratings $n_s(z),\mathscr{P}_s(z)$ with the same period π/k, where k is the light wave number [Fig. 1(b)]. If the interval $[I_{min};I_{max}]$ between the minimal and maximal intensities in this interference pattern contains one or two switching points $I_{up,down}$ of the bistability characteristic, i.e., of the light-intensity dependence of a given atomic variable, e.g., population difference n_s as in Fig. 1(c), the resulting grating should also have a bistable shape as shown in Fig. 1(b). In a specific experiment, the choice between the two (upper and lower) stable branches is governed by the system's previous states, and an adequate description should necessarily consider the temporal dynamics leading to the steady state. On the other hand, the uncertainity in the shape of the grating [$n_s(z)$ in Fig. 1(b)] could be avoided by relative motion of the medium with respect to the light interference pattern, i.e., along the z axis. Then atoms moving adiabatically along z choose their switch-up and switch-down points according to the direction of their relative velocity [see Fig. 1(b)].

To account for the atomic movement, we assume $\mathscr{E}$ to be a sum of two counterpropagating cw waves with slightly different frequencies:

$$\mathscr{E}e^{-i\omega t}=(\mathscr{E}_+e^{ikz+i\epsilon t}+\mathscr{E}_-e^{-ikz})e^{-i\omega t}. \tag{6}$$

The small relative frequency shift ϵ causes movement of the grating along z, or, equivalently, if both waves have the same frequency it can reflect the motion of the medium, since ϵ is related to the Doppler shift kv as $\epsilon=2kv$. The requirement of adiabaticity of the motion means that the time taken by an atom to pass through one period of the interference pattern should be longer than the shortest of the medium relaxation

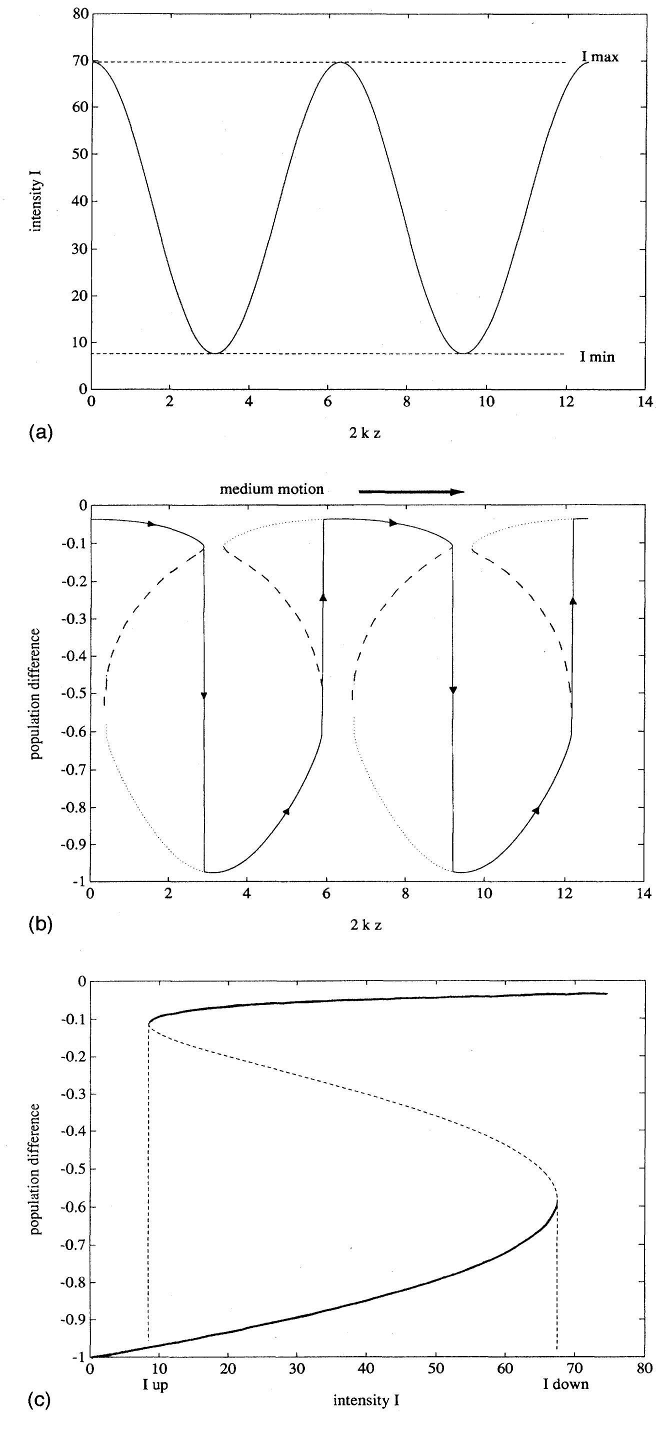

FIG. 1. Light intensity (a) and population difference (b) spatial distributions over two periods of the grating $2\pi/k=\lambda$, and the bistable dependence of the population difference upon the light intensity (c). The dashed lines in (b) and (c) represent the unstable states. The dotted line in (b) corresponds to the stable but not occupied state. The direction of the relative motion of the medium is shown. The solid line with arrows in (b) shows the resulting shape of the population grating.

times $\pi/kv \gg \gamma_2^{-1}$. This condition also ensures that the relative frequency shift ϵ is small in comparison to the linewidth γ_2, which is necessary if effects of phase self-modulation are to be neglected. The value $\sim 10^{11}\ \mathrm{s}^{-1}$ for the linewidth sets the upper limit on the velocity of the medium, $v<10^5$ cm/s. The present steady-state consideration does not allow a lower limit for the velocity to be determined, but from intuitive arguments it is clear that π/kv should not exceed the time constants of any possible intensity variations of the external fields.

The asymmetry of the grating profile in Fig. 1(b) with respect to the point $z=\pi/2k$, which is absent in the external field intensity distribution [Fig. 1(a)], reflects the spatial manifestation of the hysteresis of n_s and results in the appearance of the odd spatial components in the induced grating. As will be shown below, it is this shape of the grating that is responsible for the directional energy flow.

Because of the nonlinear dependence of the polarization $\mathscr{P}$ on light intensity,

$$|\mathscr{E}(z)|^2=|\mathscr{E}_+|^2+|\mathscr{E}_-|^2+|\mathscr{E}_+\mathscr{E}_-|\cos[2kz+(\Phi_+-\Phi_-)],$$

where $\Phi_\pm$ are the phases of field components $\mathscr{E}_\pm=|\mathscr{E}_\pm|\exp(i\Phi_\pm)$, the function $D(z)=\mathscr{P}/\mathscr{E}$, related to the medium susceptibility, should be expanded into a Fourier series with the basic spatial frequency $2k$. The lowest-order contributions to the susceptibility of the medium to the counterpropagating beams then read

$$D(z)=D_0+D_c\cos[2kz+(\Phi_+-\Phi_-)]+D_s\sin[2kz+(\Phi_+-\Phi_-)]+\cdots, \qquad (7)$$

where the complex Fourier coefficients $D_{0,c,s}$ are expressed as follows:

$$D_0=D_0^r+iD_0^i=\frac{k}{\pi}\int_0^{\pi/k}D(z)dz,$$

$$D_c=D_c^r+iD_c^i=\frac{2k}{\pi}\int_0^{\pi/k}D(z)\cos(2kz+\Phi_+-\Phi_-)dz,$$

$$D_s=D_s^r+iD_s^i=\frac{2k}{\pi}\int_0^{\pi/k}D(z)\sin(2kz+\Phi_+-\Phi_-)dz.$$

It should be pointed out that the appearance of the finite odd sine term in expansion (7) is a direct consequence of IOB, which is manifested on a scale of $\lambda/2$. Using the terminology introduced in [5], the spatial first-order phase transitions are organized in a one-dimensional array, whose spatial phase shift with respect to the light field intensity distribution leads to a nonmutuality, i.e., directionality of the Bragg reflection of one of the light waves. Selection of terms $\mathscr{P}_\pm$ in polarization P corresponding to $\mathscr{E}_\pm\exp(\pm ikz)$ results in the following expressions:

$$\mathscr{P}_\pm=\left[D_0+\frac{|\mathscr{E}_\mp|}{2|\mathscr{E}_\pm|}(D_c\mp iD_s)\right]\mathscr{E}_\pm e^{\pm ikz}\equiv\chi_\pm\mathscr{E}_\pm e^{\pm ikz}. \qquad (8)$$

After substitution of expressions (8) into the wave equation, two coupled equations for the amplitudes of the counterpropagating fields $\mathscr{E}_\pm$ are obtained:

$$\pm\frac{d}{dz}\mathscr{E}_\pm=-\kappa_0\chi_\pm\mathscr{E}_\pm, \qquad (9)$$

where κ_0 is the linear amplitude absorption coefficient in the line center.

It follows from (8) and (9) that the real part of the susceptibilities $\chi_\pm$, which is responsible for the energy wave balance ($\mathrm{Im}\chi_\pm$ contributes to the refractive index), can be divided into two parts. Linear and nonlinear absorption are given by the term $D_0^r+D_c^r|\mathscr{E}_\mp/2\mathscr{E}_\pm|$, and it is the imaginary part D_s^i of the sine component D_s that is responsible for the directional energy flow.

Analysis of (8) shows that the shape of the grating, as displayed in Fig. 1, is ensured by the motion of atoms in a positive z direction or, equivalently, by the positive sign of the detuning ϵ. The corresponding distribution of the value $\mathrm{Im}D(z)$ has a negative Fourier sine component $\mathrm{Im}D_s<0$, which means that the energy is transferred from $\mathscr{E}_-$ to $\mathscr{E}_+$. Consequently, even if $|\mathscr{E}_+|\geq|\mathscr{E}_-|$, $\mathrm{Re}\chi_+<\mathrm{Re}\chi_-$, and under certain conditions the energy flow can dominate the absorption, given by D_0^r and D_c^r; hence wave $\mathscr{E}_+$, which propagates in the direction of motion of the medium, can be amplified. This flow occurs only when $D_s^i\neq 0$, which requires the following inequalities for the amplitudes of the counterpropagating beams:

$$(|\mathscr{E}_+|-|\mathscr{E}_-|)^2<I_{down}, \quad (|\mathscr{E}_+|+|\mathscr{E}_-|)^2>I_{up}, \qquad (10)$$

where I_{down} and I_{up} are defined as the switch-down and switch-up points on the bistable curve (see Fig. 1). In other words, the depth of the intensity modulation of the spatial interference pattern should include the bistability interval $[I_{down};I_{up}]$.

The range of amplification of wave $\mathscr{E}_+$ ($\mathscr{E}_-$ for $\epsilon<0$) defined by conditions (10) is shown in Fig. 2. The asymmetry with respect to the line $|E_+|=|E_-|$ appears to be due to the cross effect.

To solve the propagation problem (9), we calculated numerically the distributions of susceptibilities $\chi_\pm$ on the plane $\{\mathscr{E}_+;\mathscr{E}_-\}$ for different sets of the atomic parameters. The amplitudes of the beams, which entered the medium with the boundary values $\mathscr{E}_+(z=0)=I_+^{1/2}$ and $\mathscr{E}_-(z=L)=I_-^{1/2}$, were changed because of the absorption, and eventually could satisfy (10). This seriously affected the counterpropagation dynamics, resulting in the occurrence of spatial first-order phase transitions. A typical distribution of the beams' intensities along the propagation direction z is shown in Fig. 3. This figure clearly exhibits the presence of the amplification regime for beam $\mathscr{E}_+$, caused by the directional energy flow. The discontinuity in spatial derivatives of the field can also be considered as the spatial phase transition between regions with different propagation properties. In contrast to the situation described in Ref. [6], this phase transition indicates a change in the profiles of the polarization and the population grating; in other words, in the character of the distributed feedback. This means that the transition zone should be thicker than one wavelength of the illuminating light.

Naturally, a question about the validity of the approximation of amplitudes slowly varying in space could arise if we remind ourselves that the value of the local-field coefficient σ is of the order of linear on-line ($\omega=\omega_0$) absorption losses

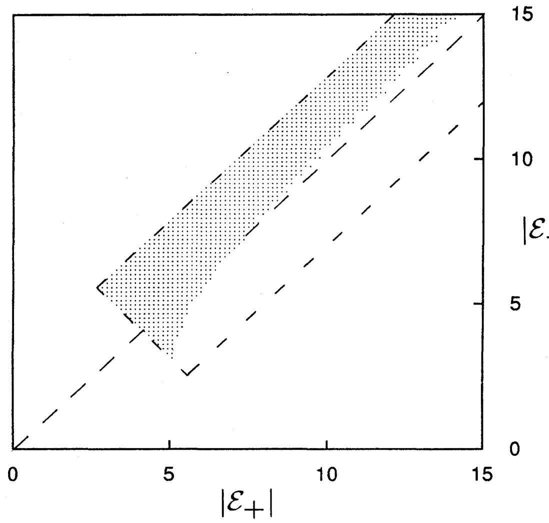

FIG. 2. Diagram displaying the range of amplification of the wave $\mathscr{E}_+$ (shaded area) at $\delta=-2$ and $\sigma=20$. The dashed line encloses the region of the energy transfer.

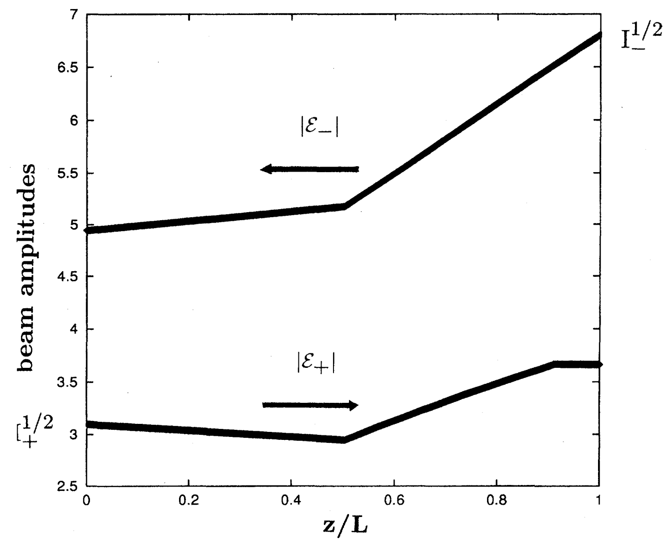

FIG. 3. Longitudinal distribution of the field amplitudes $|\mathscr{E}_+|$ and $|\mathscr{E}|_-$ in the medium for $\delta=-2$, $\sigma=20$. The gain for the beam $\mathscr{E}_+$ occurs in the middle part of the plot.

per wavelength $\kappa_0\lambda$ [4]. This approximation is correct, provided the field amplitudes $\mathscr{E}_\pm$ are not significantly affected over one grating period. Referring to Fig. 2, one can see that self-induced gain is obtained in the region of sufficient and unequal field intensities, which ensures strong saturation of the nonlinear absorption coefficient for all points along z. Even in the most critical case, $|\mathscr{E}_+|=|\mathscr{E}_-|$, when the total intensity in nodes of the interference pattern is zero, the smallness of the absorption coefficient in comparison to κ_0 should be ensured by the local-field linear frequency shift, reducing the linear on-line absorption by a factor of σ^2, as can be seen from (5).

It should be noted that directional energy flow as a consequence of IOB can occur not only with counterpropagating beams but also in any other geometry of the interacting waves in which light-induced gratings could be created. For sufficiently dense samples, such energy transfer should affect many nonlinear processes, e.g., forward two-wave interaction, forward and backward four-wave mixing, and nonlinear spectroscopy signals. IOB creates a spatial hysteresis in the gratings' profiles, which is eliminated by relative motion of the light interference pattern and the medium, producing an asymmetric shape in the grating profiles, and the nonreciprocal Bragg scattering of light beams on them results in an intense energy flow, which could compensate a nonlinear absorption. The sense of the energy transfer depends strongly on the mutual orientation of the beams' propagation direction and on the direction of the relative light-medium motion. This phenomenon could find an application in sensitive motion detectors, allowing motion of the medium to be detected at speeds much slower than $\lambda\gamma_2$.

We also wish to point out that energy transfer of the kind described above is related to a momentum transfer, i.e., with the exertion of mechanical force. Attenuation of one of the counterpropagating beams together with enhancement of the other lead to a strong stimulated optical force acting upon atoms in a direction determined by the relative detuning of the two beams, or by the atomic motion with respect to the light interference pattern. This effect could be very useful in optical trapping of neutral atoms. Most exciting seems to be the possibility of strong atomic compression by overcoming the repulsive forces arising from radiation trapping [8], since the force we discuss here occurs only at sufficiently high densities.

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