Nonlinear refractive index near points of zero absorption and in the dead zone

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Cross focusing (defocusing), determined by the probe Kerr coefficient, which is defined as the derivative of the probe refractive index with respect to the pump intensity, is studied at points of probe nonabsorption and in the dead zone. Spatial control of the probe can be achieved not only by the probe Kerr coefficient itself but also by its strong variation with pump intensity at points of probe nonabsorption. This strong variation allows the pump beam to be used as a waveguide for the probe.

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I. INTRODUCTION

As noted by Scully and co-workers, a variety of driven three- and four-level systems exhibit an extremum (minimum or maximum) in their probe dispersion spectrum at points of zero probe absorption [1,2]. Moreover, the much simpler, resonantly or near-resonantly driven two-level system displays the same behavior when interrogated by a weak or moderately strong probe [3–5]. The probe absorption and dispersion spectra for two-level and V-shaped three-level systems have recently been measured experimentally for nuclear magnetic resonance transitions by Wei *et al.* [6], with excellent agreement between experiment and theory. When the pump detuning $\Delta_L=\omega_0-\omega_L$ from the resonance frequency ω_0 is zero [see Fig. 1(a)], the point of zero absorption in a two-level system occurs at a pump-probe detuning $\delta=\omega_P-\omega_L$ such that

$$\delta=\pm 2V_L, \qquad (1)$$

where $2V_L=\mu E_L/\hbar$ is the pump Rabi frequency. Two additional points of zero absorption are situated near $\delta=0$ but at these points the refractive index is near its vacuum value. For $|\delta|>2V_L$, the probe is absorbed and for $0<|\delta|<2V_L$, the probe experiences gain. In addition, an increase of absorption of the probe is accompanied by a decrease of pump absorption and reciprocally, probe gain is accompanied by pump absorption beyond its saturation value in the absence of the probe [7,8]. At the point of zero probe absorption, pump absorption reverts to its value in the absence of the probe. Disregarding the linear effect of probe absorption leading to additional excitation to an already saturated two-level system, the correlation between the pump and probe absorption can be understood by two-photon and higher-order processes which consists of absorption of a pump and simulated emission of a probe photon or vice versa. An excess probability of probe stimulated emission over pump stimulated emission in these nonlinear processes leads to probe gain and higher pump absorption. Similarly, when $|\delta|>2V_L$, an excess of probe absorption over pump absorption leads to overall probe absorption. These energy-transfer processes between pump and probe lasers lead to fluctuations in the probe intensity. As shown in the noise spectrum [9,10], these fluctuations are largest at the points of zero absorption. Thus, the fact that extrema of the refractive index normally occur at frequencies where the absorption is large is not contradicted here: probe nonabsorption simply arises from the cancellation of large absorption by large stimulated emission.

In the presence of a Doppler-broadening width D, larger than the pump generalized Rabi frequency $(\Delta_L^2+4V_L^2)^{1/2}$, the absorption no longer vanishes at the points defined by Eq. (1). Instead a zone of almost zero probe absorption appears between these points. This is the so-called dead zone described by Baklanov and Chebotaev [11] and later by Khitrova, Berman, and Sargent [12]. Recently, there has been renewed interest in the dead zone, triggered by Scully's work on large values of the refractive index in the absence of absorption [1]. It was found that the probe refractive index is much less affected by Doppler broadening than is the probe absorption [5,13]. We may ascribe this, as we did for points of zero absorption, to the fact that the dead zone is due to compensation of stimulated probe emission by absorption and reciprocal energy transfer between the pump and probe beams. This is again confirmed by the quantum noise spectrum of the probe which shows that the noise does not vanish in the dead zone [14].

In the present paper, the large value of the refractive index at points of zero probe absorption or in the dead zone is exploited to study nonlinear effects in the refractive index. The nonlinear refractive index n is usually written in the form

$$n=n_0+n_2I, \qquad (2)$$

where n_2 is called the Kerr coefficient. This coefficient determines the spatial behavior of the beam. Thus a beam whose transverse intensity profile decreases monotonically from the center to the periphery will be focused if $n_2>0$ and defocused if $n_2<0$. This conclusion remains valid if n_2 is replaced by dn/dI which is obvious if Eq. (2) holds. However, Eq. (2) is only valid in the perturbation limit. At higher pump intensity dn/dI will determine the spatial behavior of the probe just as n_2 does at lower pump intensity. This can be seen by application of Huygens's principle. We shall show that the linear dependence of the refractive index on I breaks down near the points of nonabsorption, defined by Eq. (1), and that dn/dI changes sign near the three-photon scattering frequency (TPS) which corresponds to one of the Rabi sidebands:

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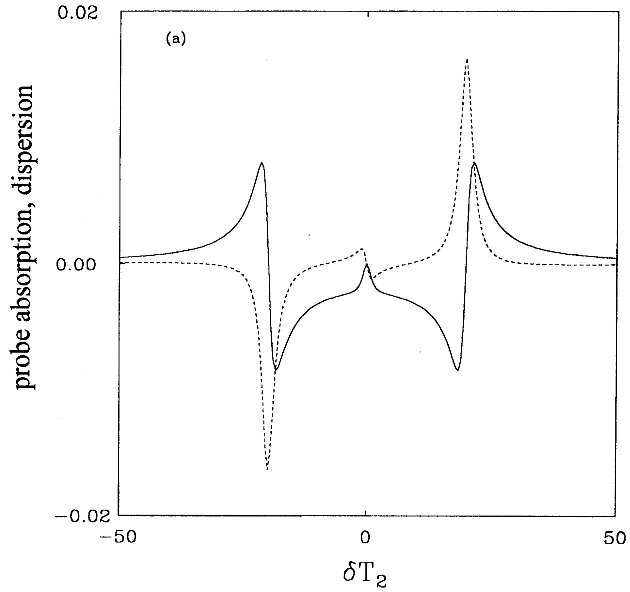

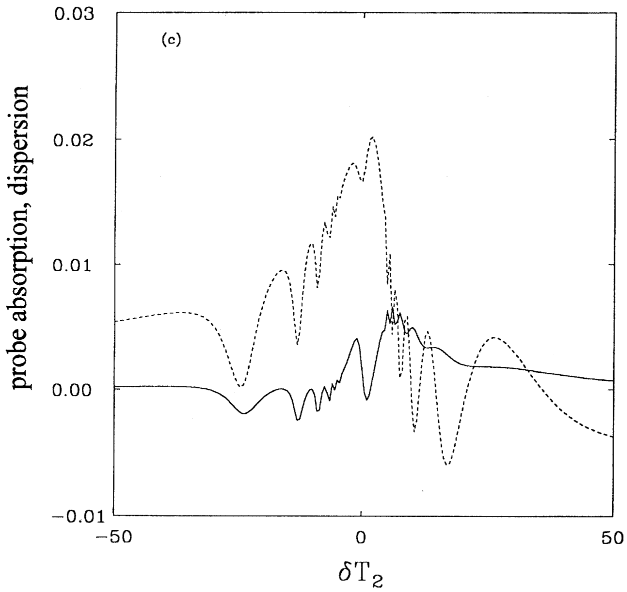

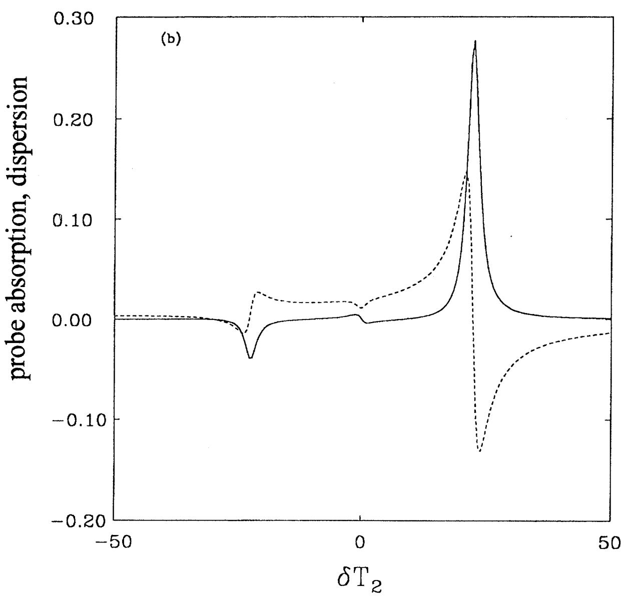

FIG. 1. Probe absorption $[\mathrm{Im}\rho_{ba}(\omega_P)/V_PT_2,—]$ and dispersion $[\mathrm{Re}\rho_{ba}(\omega_P)/V_PT_2,---]$ versus δT_2 for (a) a resonant strong pump $\Delta_LT_2=0$, $V_LT_2=10$, and a weak probe; (b) a detuned strong pump $\Delta_LT_2=10$, $V_LT_2=10$, and (c) a detuned strong pump $\Delta_LT_2=10$, $V_LT_2=10$, and a strong probe $V_PT_2=10$.

$$\delta=\pm(\Delta_L^2+4V_L^2)^{1/2}. \tag{3}$$

Note that, although Eqs. (1) and (3) both refer to TPS, Eq. (1) corresponds to points of probe nonabsorption whereas Eq. (3) corresponds to probe amplification. There are, however, four values of δ where the probe absorption is zero [see Fig. 1(b)]. In contrast to the situation discussed above where $\Delta_L=0$, points of zero absorption near $\delta=0$ may coincide with relatively large values of the refractive index. Moreover at none of these points does the refractive index assume a maximum value and therefore the Kerr coefficient dn/dI does not change sign at these points. In addition, we consider Doppler broadening and describe the nonlinearity of the refractive index in the dead zone.

In a previous commutation [15], we analyzed the nonlinearity of both the pump refractive index n_L and probe refractive index n_P by plotting them as a function of δ for two different pump or probe intensities. In this way, we were able to distinguish between induced self-focusing, dn_L/dI_L and dn_P/dI_P, and cross focusing, dn_L/dI_P and dn_P/dI_L. These considerations allow us to propose qualitative explanations for a number of anomalous effects recently observed in conical emission [16]. The procedure we adopted previously [15] only gives correct indications concerning the generalized Kerr coefficient dn/dI for intensity regions where this coefficient is constant. Otherwise, only the procedure adopted in the present paper, where the dependence of the refractive index on the square of the Rabi frequency is considered in detail, gives correct results. Note that Doppler broadening and dead zones were not considered in Ref. [15].

We consider here only cross focusing of the probe beam induced by the transverse intensity profile of the pump beam, that is dn_P/dI_L, rather than the self-focusing of the probe beam dn_P/dI_P, induced by its own transverse intensity profile. The self-focusing is important only for large probe intensities and has been discussed for a number of multilevel schemes [2] and also for a driven two-level system [5,15].

Whereas dead zones are usually discussed in the context of systems interacting with two copropagating fields, Moseley *et al.* [17] and Gea-Banacloche *et al.* [18] have recently considered a Doppler-broadened cascade three-level system whose upper transition interacts resonantly with a strong pump while the lower transition interacts with a weak counterpropagating probe. Due to Rabi splitting induced by the pump, the middle level splits into two levels equally displaced from its unperturbed energy value. Thus the system becomes almost transparent to the probe in a region centered at the unperturbed lower transition frequency with a half-width equal to the pump Rabi frequency. This effect has been called electromagnetically induced transparency (EIT) by Harris and co-workers [19,20]. It provides an additional model for the study of strong dispersion [20] and nonlinear properties of the refractive index such as cross focusing dn_p/dI_L [17] in the presence of almost vanishing absorption. Note that since the pump interacts with the upper transition, its absorption is also almost vanishing. In this respect, EIT is similar to the case of probe absorption in the strongly driven Λ-shaped three-level system where, at a point of zero probe absorption, the pump is also not absorbed due to population trapping [21].

II. THEORY

The Bloch equations for a two-level system interacting with a pump beam at frequency ω_L with Rabi frequency $2V_L$ and an arbitrarily strong probe beam at frequency ω_P with Rabi frequency $2V_P$ have been solved in a previous publication [8]. The pump and probe refractive indices (n_L and n_P) and absorption coefficients (α_L and α_P) are given by

$$n_{L,P}=1+(N\mu_{ba}/\varepsilon_0 E_{L,P})\mathrm{Re}\rho_{ba}(\omega_{L,P}) \quad (4)$$

and

$$\alpha_{L,P}=(N\mu_{ba}\omega_{L,P}/\varepsilon_0 c E_{L,P})\mathrm{Im}\rho_{ba}(\omega_{L,P}), \quad (5)$$

where N is the atomic number density and $\rho_{ba}(\omega_{L,P})$ can be calculated from

$$\rho_{ba}(\omega_L)=\frac{V_L+V_P Z_1}{\Delta_L-i/T_2}\,\frac{i/T_2}{P_0-2i\ \mathrm{Im}R_0 V_L^* V_P Z_1} \quad (6)$$

and

$$\rho_{ba}(\omega_P)=\frac{V_P+V_L Z_1^*}{\Delta_P-i/T_2}\,\frac{i/T_2}{P_0-2i\ \mathrm{Im}R_0 V_L^* V_P Z_1}\,. \quad (7)$$

The continued fraction Z_1 is given by

$$Z_1=\cfrac{U_1 V_L V_P^*}{1-\cfrac{S_1|V_L|^2|V_P|^2}{1-\cfrac{S_2|V_L|^2|V_P|^2}{1-\cdots}}}, \quad (8)$$

where

$$U_n=\frac{Q_n}{P_n}, \quad S_n=\frac{R_n}{P_n}U_{n+1}, \quad (9)$$

$$P_n=-n\delta+i/T_1 -4(n\delta-i/T_2)\{|V_L|^2[\Delta_L^2-(n\delta-i/T_2)^2]^{-1} +|V_P|^2[\Delta_P^2-(n\delta-i/T_2)^2]^{-1}\}, \quad (10)$$

$$Q_n=2[(2n-1)\delta-2i/T_2][(\Delta_L-n\delta+i/T_2) \times(\Delta_P+n\delta-i/T_2)]^{-1}, \quad (11)$$

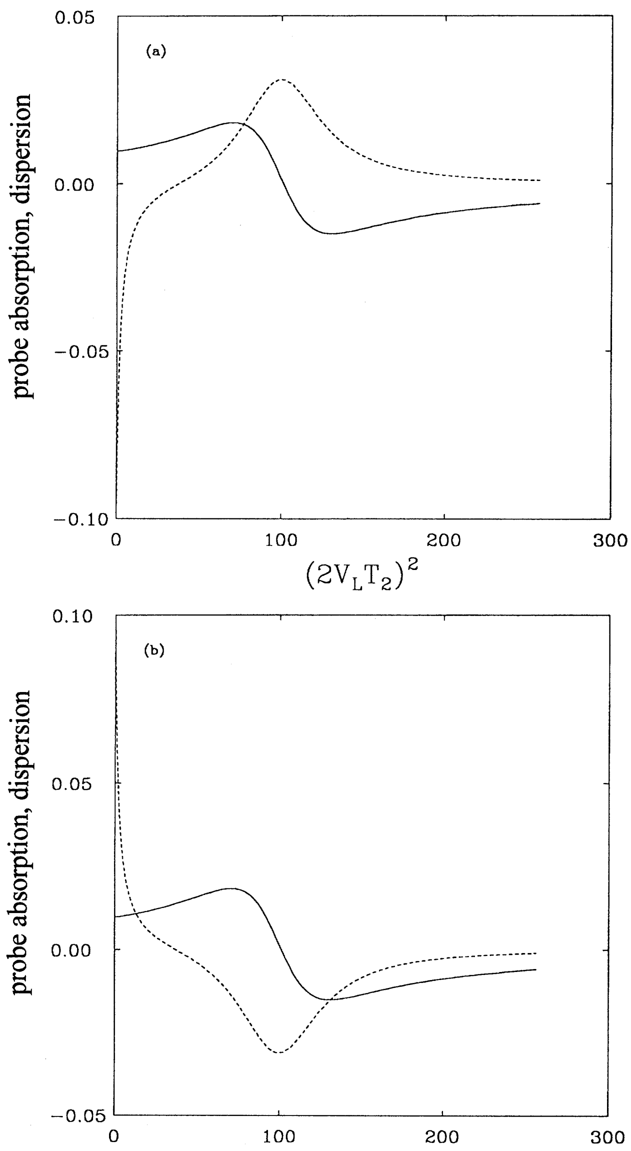

FIG. 2. Probe absorption [$\mathrm{Im}\rho_{ba}(\omega_P)/V_PT_2$,—] and dispersion [$\mathrm{Re}\rho_{ba}(\omega_P)/V_PT_2$,---] versus $(2V_LT_2)^2$, beginning at $V_LT_2=0.01$, for a resonant strong pump $\Delta_LT_2=0$, $V_LT_2=10$, and a weak probe for (a) $\delta T_2=10$ and (b) $\delta T_2=-10$.

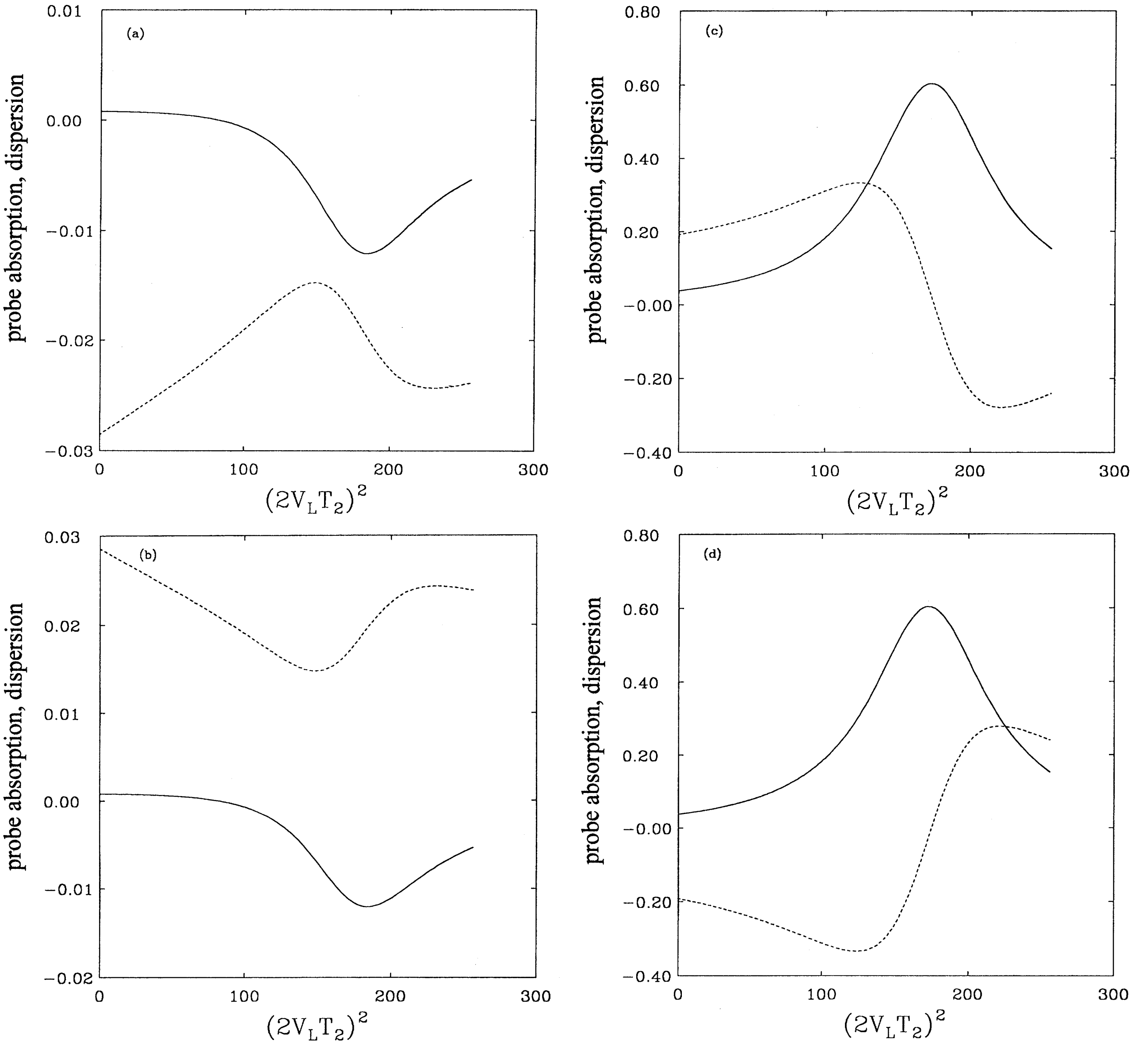

FIG. 3. Probe absorption $[\mathrm{Im}\rho_{ba}(\omega_P)/V_PT_2,$—] and dispersion $[\mathrm{Re}\rho_{ba}(\omega_P)/V_PT_2,$---] versus $(2V_LT_2)^2$, beginning at $V_LT_2=0.01$, for a detuned strong pump and a weak probe for (a) $\Delta_LT_2=-15$, $\delta T_2=20$; (b) $\Delta_LT_2=15$, $\delta T_2=-20$; (c) $\Delta_LT_2=-15$, $\delta T_2=-20$, and (d) $\Delta_LT_2=15$, $\delta T_2=20$.

and

$$R_n=2[(2n+1)\delta-2i/T_2][(\Delta_L+n\delta-i/T_2)\times(\Delta_P-n\delta+i/T_2)]^{-1}. \tag{12}$$

In these equations, $\delta=\omega_P-\omega_L$ is the pump-probe detuning, $\Delta_P=\omega_0-\omega_P$ is the detuning of the probe from the resonance frequency, and T_1 and T_2 are the longitudinal and transverse lifetimes.

For brevity, we refer in the figures to the quantity,

$$\mathrm{Im}\rho_{ba}(\omega_{L,P})/V_{L,P}T_2=2\hbar\varepsilon_0\omega_{L,P}\alpha_{L,P}/cN\mu_{ba}^2T_2, \tag{13}$$

as the "absorption" and to

$$\mathrm{Re}\rho_{ba}(\omega_{L,P})/V_{L,P}T_2=\hbar\varepsilon_0(n_{L,P}-1)/N\mu_{ba}^2T_2, \tag{14}$$

as the "dispersion."

When Doppler broadening is included for a copropagating bichromatic field, $\rho_{ba}(\omega_{L,P})$ should be replaced by $\rho_{ba}^D(\omega_{L,P})$ defined by

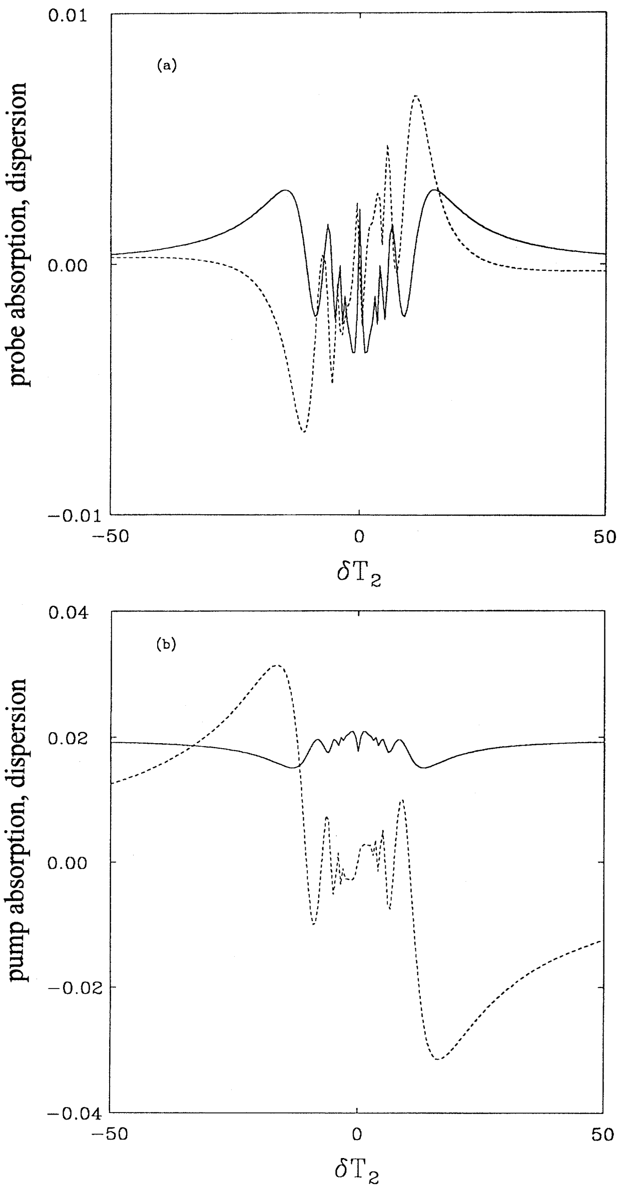

FIG. 4. Correlation between (a) probe absorption [$\mathrm{Im}\rho_{ba}(\omega_P)/V_PT_2$,—] and dispersion [$\mathrm{Re}\rho_{ba}(\omega_P)/V_PT_2$,---] versus δT_2 and (b) pump absorption [$\mathrm{Im}\rho_{ba}(\omega_L)/V_LT_2$,—] and dispersion [$\mathrm{Re}\rho_{ba}(\omega_L)/V_LT_2$,---] versus δT_2 for a resonant pump $\Delta_LT_2=0$, and almost equal pump and probe Rabi frequencies $V_LT_2=5$, $V_PT_2=4$.

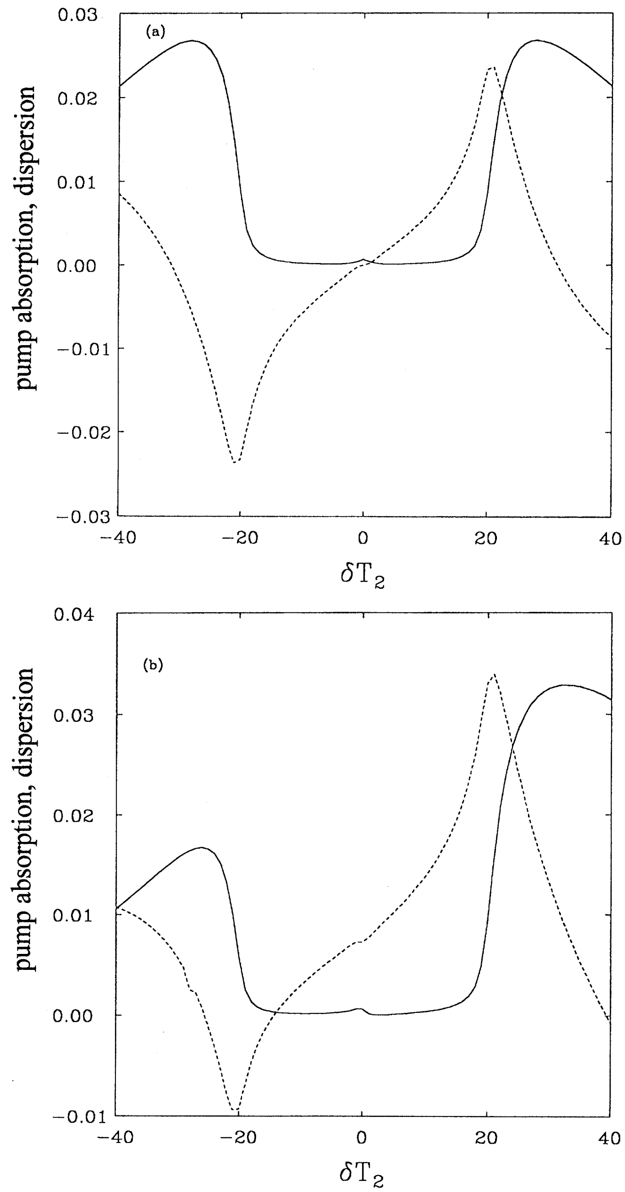

FIG. 5. Doppler-broadened probe absorption [$\mathrm{Im}\rho_{ba}(\omega_P)/V_PT_2$,—] and dispersion [$\mathrm{Re}\rho_{ba}(\omega_P)/V_PT_2$,---] versus δT_2 for a strong pump $V_LT_2=10$, and a weak probe for (a) a resonant pump $\Delta_LT_2=0$ and (b) a detuned pump $\Delta_LT_2=20$. Doppler width $DT_2=50$.

$$\rho^D_{ba}(\omega_{L,P})=\rho^D_{ba}(\Delta_L,\Delta_P,\delta)=\rho^D_{ba}(\Delta_L,\Delta_P=\Delta_L-\delta,\delta)$$
$$=(1/\pi D^2)^{1/2}\int_{-\infty}^{\infty}\rho_{ba}(\Delta_L',\Delta_P'=\Delta_L'-\delta,\delta)$$
$$\times\exp[-(\Delta_L-\Delta_L')^2/D^2]d\Delta_L'. \quad (15)$$

Perturbation limit

In the perturbation limit where V_P is small, Eq. (7) reduces to

$$\rho_{ba}(\omega_P)=\frac{V_P(\rho_{bb}-\rho_{aa})^{\mathrm{dc}}}{-\Delta_P+i/T_2}+\frac{V_L(\rho_{bb}-\rho_{aa})^{\delta}}{-\Delta_P+i/T_2}, \quad (16)$$

where the population pulsation term $(\rho_{bb}-\rho_{aa})^{\delta}$ is given by

$$(\rho_{bb}-\rho_{aa})^{\delta}$$
$$=\frac{V_LV_P(\delta+\Delta_L+i/T_2)(\delta+2i/T_2)(\rho_{bb}-\rho_{aa})^{\mathrm{dc}}}{(\Delta_L+i/T_2)D(\omega_P)}, \quad (17)$$

with

$$D(\omega_P) = -2|V_L|^2(\delta + i/T_2) - (1/2)(\Delta_P + i/T_2) \times(\Delta_L + \delta + i/T_2)(\delta + i/T_1). \quad (18)$$

If the pump laser is sufficiently weak, Eq. (16) becomes

$$\rho_{ba}(\omega_P) = \frac{V_P \rho_{aa}^{\text{eq}}}{\Delta_P - i/T_2}\left\{1 - \frac{2|V_L|^2}{(\Delta_L + i/T_2)(\Delta_P + i/T_2)} \times\left[1 + \frac{2i/T_2 - i/T_1}{\delta + i/T_1}\right]\right\}, \quad (19)$$

which shows that the nonlinear part of the probe refractive index is determined by the part proportional to $|V_L|^2$.

III. RESULTS AND DISCUSSION

In the present section, we apply the general theory of Sec. II to a number of particular cases. All calculations are carried out in the collisionless regime and the value of the probe Rabi frequency is only given in the case where the probe is sufficiently strong so that the perturbation theory results are invalid. We have verified that the essential features derived for weak probes persist up to $V_P T_2 \approx 1$ [4]. Figure 1(a) relates to a two-level system interacting resonantly with an intense pump laser and a weaker probe. The probe "absorption" and "dispersion" [see Eqs. (13) and (14)] are plotted versus the probe-probe detuning δ. The figure shows that there are four nonabsorption points, two of which are very near $\delta = 0$. At these two points, the dispersion is almost equal to zero. The other two nonabsorption points, defined by Eq. (1), are more interesting since they correspond to a maximum and minimum value of the refractive index. Figure 1(b) depicts the situation where $\Delta_L > 0$. Inspection of this figure again reveals four points of nonabsorption. The dispersion differs appreciably from zero at three of these points. In Fig. 1(c), we also have $\Delta_L > 0$ but now the probe is also strong so that, due to the appearance of Rabi subharmonics [8], there are more than four nonabsorption points accompanied by appreciable dispersion.

In Fig. 2(a), the probe absorption and dispersion are plotted versus the squared Rabi frequency of the pump which is proportional to its intensity, for a resonant pump and $\delta > 0$. The figure shows that the dispersion has its maximum value at $2V_L = \delta$ and at this point, the probe absorption is zero. For $2V_L < \delta$, $dn_P/dI_L > 0$ leading to focusing of the probe beam, and for $2V_L > \delta$, $dn_P/dI_L < 0$, leading to defocusing. We can also envisage a special experimental setup where the transverse pump profile is chosen such that $2V_L > \delta$ near the beam axis and decreases monotonically to values $2V_L < \delta$ away from the beam axis. Then the beam will behave as a waveguide for the probe laser, leading to focusing of the peripheral region of the probe towards the pump axis and to defocusing of the axial region towards the periphery. The beam emerging from the nonlinear medium will consist of a cone surrounding a narrow axial beam.

In Fig. 2(b), we take $\delta < 0$ and obtain a minimum for the refractive index at $2V_L = |\delta|$ which again corresponds to a point of zero absorption. We can therefore again obtain focusing or defocusing depending on the pump intensity.

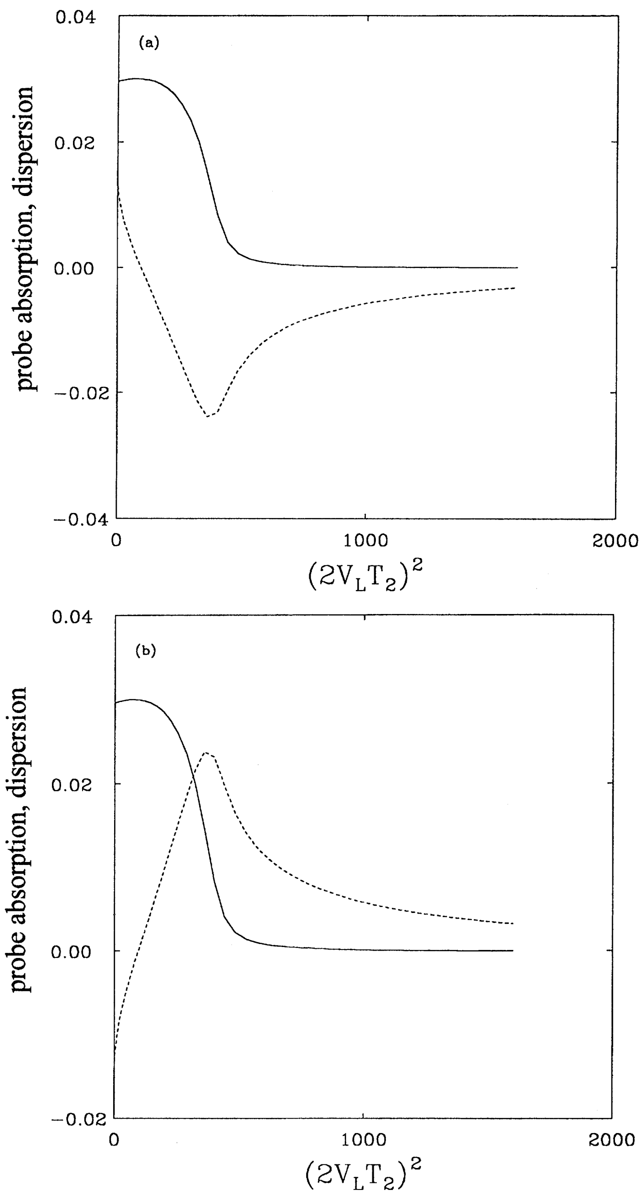

FIG. 6. Doppler-broadened probe absorption [$\text{Im}\rho_{ba}(\omega_P)/V_PT_2$,—] and dispersion [$\text{Re}\rho_{ba}(\omega_P)/V_PT_2$,---] versus $(2V_LT_2)^2$, beginning at $V_LT_2 = 0.01$, for a resonant strong pump $\Delta_LT_2 = 0$ and a weak probe with Doppler width $DT_2 = 50$, for (a) $\delta T_2 = -20$ and (b) $\delta T_2 = 20$.

When, as above, the Rabi frequency $2V_L$ is larger than $|\delta|$ near the beam axis and decreases monotonically to $2V_L < |\delta|$ away from the beam axis, the axial part of the probe beam will be focused and the peripheral part defocused. Under these conditions, the pump beam will not act as a waveguide. We predict again that the probe beam will consist of a cone surrounding a narrow axial beam.

When $\Delta_L \neq 0$, the behavior is more complex. As shown in Fig. 1(b), the absorption spectrum as a function of δ has a dispersive shape centered at $\delta = 0$ which is a point of nonab-

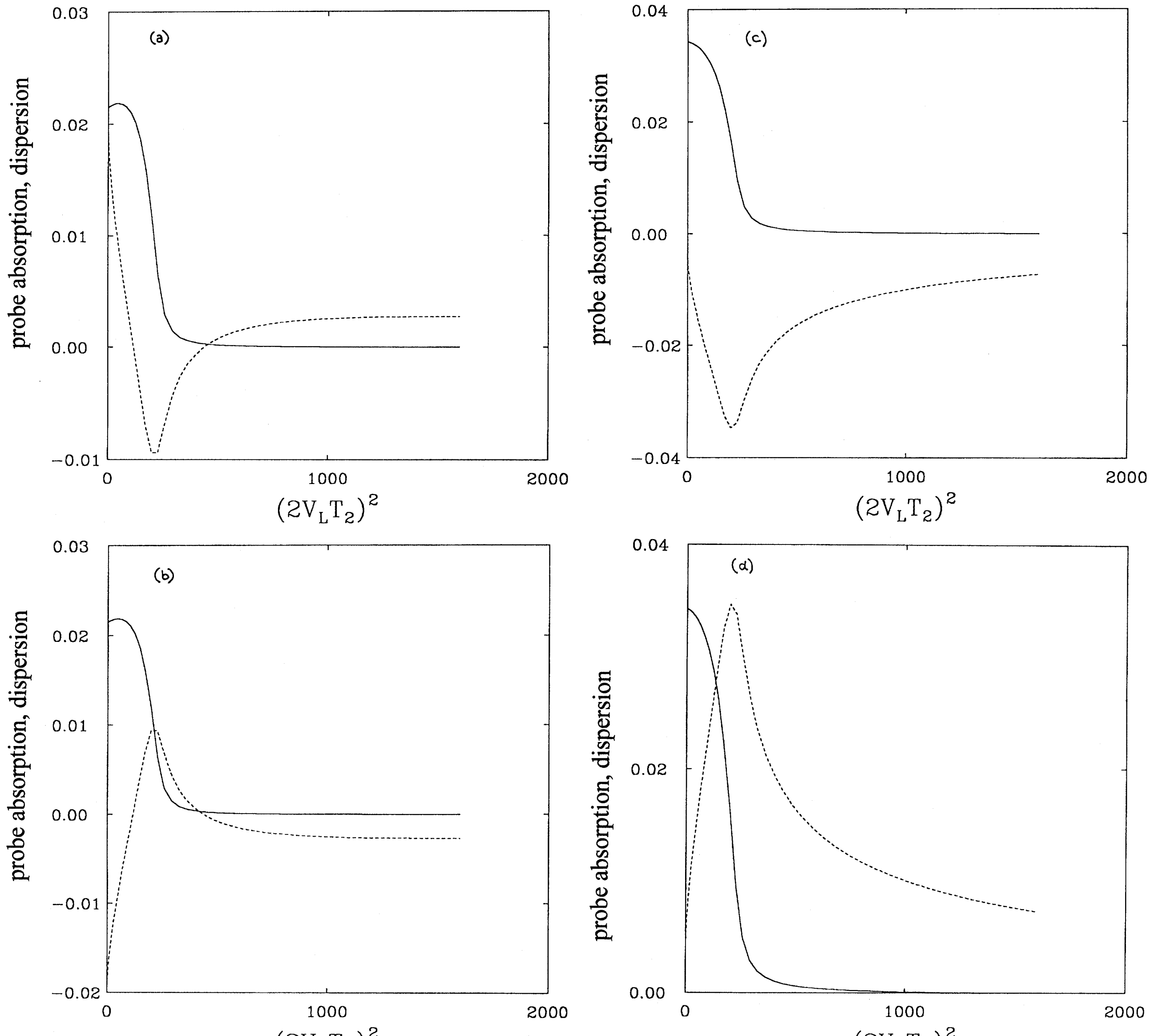

FIG. 7. Doppler-broadened probe absorption $[\mathrm{Im}\rho_{ba}(\omega_P)/V_PT_2,$—] and dispersion $[\mathrm{Re}\rho_{ba}(\omega_P)/V_PT_2,$---] versus $(2V_LT_2)^2$, beginning at $V_LT_2=0.01$, for a detuned strong pump and a weak probe, with Doppler width $DT_2=50$, for (a) $\Delta_LT_2=-15$, $\delta T_2=20$; (b) $\Delta_LT_2=15$, $\delta T_2=-20$; (c) $\Delta_LT_2=-15$, $\delta T_2=-20$ and (d) $\Delta_LT_2=15$, $\delta T_2=20$.

sorption. This is an extra resonant feature which results either from dephasing collisions [22,23], or at low pressure, as considered here, from nonlinear effects in the pump intensity [23,24]. The refractive index does not vanish at $\delta=0$ [see Fig. 1(b)]. The reason for this is similar to that given in the Introduction for the case where $\Delta_L=0$ since the correlation between pump and probe absorption also exists for the case of off-resonant pumps [25].

Figure 3(a) shows that for $\Delta_L<0$ and $\delta>-\Delta_L$, the maximum of the refractive index appears at a pump Rabi frequency such that $(\Delta_L^2+4V_L^2)\approx\delta^2$ which corresponds to the TPS frequency. This is similar to Fig. 2(a) apart from the fact that the maximum of the refractive index does not appear at a point of probe nonabsorption. The point of nonabsorption appears at smaller values of the pump Rabi frequency and there the Kerr coefficient $dn_P/dI_L>0$. Figure 3(b), where $\Delta_L>0$ and $\delta<-\Delta_L$, exhibits a minimum in the refractive index when $(\Delta_L^2+4V_L^2)\approx\delta^2$ which again corresponds to the TPS frequency. Here, $dn_P/dI_L<0$ at the point of probe nonabsorption.

In Fig. 3(c) where $\Delta_L<0$ and $\delta<\Delta_L$ and in Fig. 3(d) where $\Delta_L>0$ and $\delta>\Delta_L$, the dispersion has a dispersive line shape centered near the Rabi-shifted resonance frequency given by $\Delta_L^2+4V_L^2=\delta^2$. Thus when $\Delta_L^2+4V_L^2\approx\delta^2$, the probe is defocused for blueshifted pump frequencies and focused for redshifted pump frequencies. In Figs. 4(a) and 4(b), we compare the probe and pump absorption and dispersion at zero pump detuning, when the probe and pump Rabi frequencies are almost equal. We see that the correlation between the probe and pump absorption discussed in the Introduction is accompanied by the correlation between probe and pump refractive indices.

Figures 5–7 relate to Doppler-broadened systems. In Fig. 5(a), $\Delta_L = 0$ and the "absorption" and "dispersion" are plotted against δ. The absorption spectrum exhibits a dead zone which sets in abruptly, whereas the dispersion varies smoothly. This shows that appreciable values of the refractive index can be obtained with vanishing absorption. In Fig. 5(b), the situation where $\Delta_L \neq 0$ is considered. Again the half-width of the dead zone about the point $\delta = 0$ equals the pump Rabi frequency $2V_L$ [12] just as in Fig. 5(a). However, the absorption spectrum is asymmetrical in the sense that there is more probe absorption at those values of δ, outside the dead zone, where the probe frequency is closer to the resonance frequency ω_0. Thus the dead zone, centered at the extra resonance frequency $\delta = 0$, is mainly, although not exclusively, due to atoms with velocities such that the effective pump and probe frequencies they experience are within the dead zone of Fig. 5(a), where $\Delta_L = 0$.

In Figs. 6 and 7, we plot the Doppler-broadened absorption and dispersion as a function of the square of the pump Rabi frequency for various values of δ and Δ_L. In these plots, the dead zone appears as an abrupt decrease of absorption when $2V_L \approx |\delta|$ whereas the variation in the refractive index is smooth.

These figures show that spatial control of the probe beam is obtained by varying the intensity of the pump beam. For example, Fig. 6(a) shows that in the dead zone $dn_P/d(4V_L^2) > 0$ when $\delta < 0$; this corresponds to focusing. Since the focusing of the probe is due here to the pump intensity, we call it "cross focusing" [15]. The control of the spatial behavior of the probe beam by the pump beam becomes apparent by noting that in the absence of the pump, the probe would be self-defocused for $\delta < 0$. Thus cross focusing can be applied to the construction of a lens for weak probe beams. The abrupt decrease of probe absorption at $2V_L \approx |\delta|$ can be used as an optical switch [8].

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