Doppler-free nonlinear processes in strong optical fields

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It is shown that compensation of Doppler broadening of two-photon transitions in oppositely traveling waves disappears with increasing pump intensity. On the other hand, a sharp Doppler-free resonances appears possible in a radiation field with two different frequencies, not only in the two-photon absorption scheme but also in the Raman scattering scheme. Since we can select the pump frequencies so that detuning from the intermediate level is small, an additional possiblity is found for raising the probability of Doppler-free transitions. Conditions are formulated for obtaining sharp Doppler-free resonances in fields of different frequencies. Possibilities are shown for a sharp increase in the nonlinear susceptibility for resonant four-photon processes due to this phenomenon. Estimates are made on the conditions for the observation of the predicted phenomenon in sodium vapor. The phenomenon can be observed in moderate pump intensities of the order of 1kW/cm^2.

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INTRODUCTION

The concepts of stepwise and multiphoton processes play an important role in modern physics. These ideas were introduced based on perturbation theory. In strong fields the frequency-correlation properties of radiative processes that discriminate the various processes undergo changes that are accompanied by variation in the Doppler widths of the emission and absorption lines.[1] In particular, it was shown in Ref. 1 that Doppler-free processes are possible in strong fields even in those cases when they are impossible in weak fields. This pertains, in particular, to processes of the Raman-scattering type. The conditions of Doppler-free spectroscopy in weak fields were subsequently considered in Refs. 2 and 3 (also see Ref. 4 for Bibliography). Variation in the line shape of the absorption and transmission of the weak field in the presence of a strong field was studied in Ref. 1. The development and generalization of the results to the case when both fields can strongly perturb the atomic system are given

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in this paper. It is shown that the conditions of Doppler-free absorption and scattering of the radiation can vary substantially. Doppler-free transitions in strong fields are of considerable interest for the physics of selective action on a material, for stimulated-Raman lasers, and with optical pumping. The role of these phenomena in resonant four-photon parametric interactions in gases is studied. Possibilities are shown for increasing the nonlinear susceptibility and radiation conversion factor by several orders.

BASIC EXPRESSIONS

Let us consider the quasi-resonant interaction of two strong monochromatic fields with a three-level system [see Fig. 1(a)] by assuming that each field interacts only with one transition. For a Raman-type transition scheme [see Fig. 1(b)] the results can be obtained by simple substitutions in the final expressions for the scheme of Fig. 1(a).

The set of equations for the elements of the density matrix reduces to a set of algebraic equations in the stationary approximation

$$\left.\begin{aligned}P_{gm}r_{gm}&=iG^{*}\Delta_{gm}+ir_{gn}G_{\mu},\\ P_{mn}r_{mn}&=iG_{\mu}^{*}\Delta_{mn}-iGr_{gn},\\ P_{gn}r_{gn}&=-iG^{*}r_{mn}+ir_{gm}G_{\mu}^{*},\\ \Gamma_{m}r_{m}&=2\,\mathrm{Re}\,(ir_{mn}G_{\mu}-ir_{gm}G),\\ \Gamma_{n}r_{n}&=-2\,\mathrm{Re}\,(iG_{\mu}r_{mn}).\end{aligned}\right\}\qquad(1)$$

Here r_{ij} are amplitudes of the density matrix elements ρ_{ij} in the interaction representation [for example, $\rho_{gn}=r_{gn}\exp\{i[\Omega_{gn}t-(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{r}]\}$, $\mathbf{k}_j$ are wave vectors of the radiation, $\Omega_{gn}=\Omega_{mn}+\Omega_{gm}=(\omega_2-\omega_{mn})+(\omega_1-\omega_{ng})$], Γ_i are relaxation rates of the populations, Γ_{ij} are half-widths of transitions $P_{ij}=\Gamma_{ij}+i\Omega'_{ij}$; $\Omega'_{gn}=\Omega_{gn}-(\mathbf{k}_1+\mathbf{k}_2)\cdot\mathbf{v}$; $\Omega'_{mn}=\Omega_{mn}-\mathbf{k}_2\cdot\mathbf{v}$; $\Omega'_{gn}=\Omega_{gn}-\mathbf{k}_1\cdot\mathbf{v}$; $\mathbf{v}$ is the velocity of the atom; $\Delta_{ij}=r_i-r_j$ are population differences; $G_{\mu}=-\mathbf{E}_2\cdot\mathbf{d}_{mn}/2\hbar$; $G=-\mathbf{E}_1\cdot\mathbf{d}_{mg}/2\hbar$.

We obtain from set (1)

$$\left.\begin{aligned}r_{gm}&=\frac{iG^{*}(|G_{\mu}|^2\Delta_{mn}+(P_{mn}P_{gn}+|G|^2)\Delta_{gm})}{P_{gm}P_{mn}\tilde{P}_{gn}},\\ r_{mn}&=\frac{iG_{\mu}^{*}(|G|^2\Delta_{gm}+(P_{gm}P_{gn}+|G_{\mu}|^2)\Delta_{mn})}{P_{gn}P_{mn}\tilde{P}_{gn}},\end{aligned}\right\}\qquad(2)$$

where

$$\tilde{P}_{gn}=\left(\Gamma_{gn}+\frac{|G|^2}{\Omega_{\mu}'^{2}}\Gamma_{mn}+\frac{|G_{\mu}|^2}{\Omega'^{2}}\Gamma_{gm}\right)+i\left(\Omega'_{gn}-\frac{|G|^2}{\Omega'_{\mu}}-\frac{|G_{\mu}|^2}{\Omega'}\right)=\tilde{\Gamma}_{gn}+i\tilde{\Omega}'_{gn}.$$

Using Eqs. (2) and the last two equations of set (1) we find the solution for the populations of levels n and m in the field $\mathbf{E}_1$ and $\mathbf{E}_2$ for the case in which $|\Omega|$, $|\Omega_{\mu}|$ $\gg|\Omega_{gn}|$, Γ_{ij}, $k_{1,2}\bar{v}$, where $\bar{v}$ is the most probable velocity

$$\left.\begin{aligned}r_n&=\left\langle\frac{a\varkappa}{\tilde{\Omega}_{gn}'^{2}+\tilde{\Gamma}_{gn}^{2}(1+\varkappa)}\right\rangle,\\ r_m&=\left\langle\frac{b\varkappa}{\tilde{\Omega}_{gn}'^{2}+\tilde{\Gamma}_{gn}^{2}(1+\varkappa)}\right.\\ &\left.-\frac{\dfrac{\Omega_{\mu}}{\Omega}\dfrac{\varkappa}{|G_{\mu}|^2}\tilde{\Gamma}_{gn}\Gamma_{gm}\left(\Gamma_n+2\dfrac{|G_{\mu}|^2}{\Omega_{\mu}^2}\Gamma_{mn}\right)}{\Gamma_m-2\Gamma_n-\dfrac{\Omega}{\Omega_{\mu}}\Gamma_m-\left(1+2\dfrac{\Omega_{\mu}}{\Omega}+\dfrac{\Omega}{\Omega_{\mu}}\right)\Gamma_n}\,\frac{\tilde{\Omega}_{gn}'^{2}+\tilde{\Gamma}_{gn}^{2}}{\tilde{\Omega}_{gn}'^{2}+\tilde{\Gamma}_{gn}^{2}(1+\varkappa)}\right\rangle.\end{aligned}\right\}\qquad(3)$$

Here the angle brackets denote averaging with a Maxwellian distribution over the velocities of the atoms

$$\left.\begin{aligned}a&=\tilde{\Gamma}_{gn}^{2}\Gamma_m\left[\Gamma_m-2\Gamma_n-\frac{\Omega}{\Omega_{\mu}}\Gamma_m-\left(1+2\frac{\Omega_{\mu}}{\Omega}+\frac{\Omega}{\Omega_{\mu}}\right)\Gamma_n\right]^{-1},\\ b&=-\tilde{\Gamma}_{gn}^{2}\left(\Gamma_n+\frac{\Omega_{\mu}}{\Omega}\Gamma_n\right)\left[\Gamma_m-2\Gamma_n-\frac{\Omega}{\Omega_{\mu}}\Gamma_m-\left(1+2\frac{\Omega_{\mu}}{\Omega}+\frac{\Omega}{\Omega_{\mu}}\right)\Gamma_n\right]^{-1},\\ \tilde{\Gamma}_m&=\Gamma_m-2\left(\frac{|G|^2}{\Omega\Omega_{\mu}}\Gamma_{gm}+\frac{|G_{\mu}|^2}{\Omega\Omega_{\mu}}\Gamma_{mn}\right),\\ \tilde{\Gamma}_n&=\Gamma_n+2\left(\frac{|G|^2}{\Omega\Omega_{\mu}}\Gamma_{gm}+\frac{|G_{\mu}|^2}{\Omega\Omega_{\mu}}\Gamma_{mn}\right),\\ \varkappa&=-2\frac{|GG_{\mu}|^2}{\Omega\Omega_{\mu}\tilde{\Gamma}_{gn}}\,\frac{\Gamma_m-2\Gamma_n-\dfrac{\Omega}{\Omega_{\mu}}\Gamma_m-\left(1+2\dfrac{\Omega_{\mu}}{\Omega}+\dfrac{\Omega}{\Omega_{\mu}}\right)\Gamma_n}{\Gamma_m\Gamma_n+2\dfrac{|G_{\mu}|^2}{\Omega_{\mu}^2}\Gamma_{mn}(\Gamma_m+\Gamma_n)+4\dfrac{|G|^2}{\Omega^2}\Gamma_{gm}\left(\Gamma_n+3\dfrac{|G_{\mu}|^2}{\Omega_{\mu}^2}\Gamma_{mn}\right)}.\end{aligned}\right\}\qquad(4)$$

As Γ_m tends to ∞, we derive from Eqs. (3) an expression for r_n when the population of the intermediate level r_m is negligibly small. Assuming $|G|$, $|G_{\mu}|\ll|\Omega|$, $|\Omega_{\mu}|$, $\Omega_{\mu}/\Omega\approx-1$, we obtain the normal expression for two-photon absorption in weak fields and the condition when the Doppler effect does not appear: $\mathbf{k}_1=-\mathbf{k}_2$.

DOPPLER-FREE TRANSITIONS IN STRONG FIELDS

Atoms moving with different velocities experience different field perturbation of two-photon resonance. We analyze the expression for the resonance dominator $\tilde{P}_{gn}$

$$\tilde{P}_{gn}=\tilde{\Gamma}_{gn}+i\left(\Omega'_{gn}-\frac{|G|^2}{\Omega'_{\mu}}-\frac{|G_{\mu}|^2}{\Omega'}\right).\qquad(5)$$

Taking into account Doppler shifts in the first nonvanishing approximation and expanding in $\mathbf{k}_1\cdot\mathbf{v}/\Omega$ and $\mathbf{k}_2\cdot\mathbf{v}/\Omega_{\mu}$ accurate up to first order, we obtain

$$\begin{aligned}\tilde{P}_{gn}&=\tilde{\Gamma}_{gn}+i\,[\tilde{\Omega}_{gn}-(\tilde{\mathbf{k}}_1+\tilde{\mathbf{k}}_2)\cdot\mathbf{v}]=\tilde{\Gamma}_{gn}+i\tilde{\Omega}_{gn}\\ &-i\left[\left(1+\frac{|G_{\mu}|^2}{\Omega^2}\right)\mathbf{k}_1+\left(1+\frac{|G|^2}{\Omega_{\mu}^2}\right)\mathbf{k}_2\right]\cdot\mathbf{v},\end{aligned}\qquad(6)$$

where

$$\tilde{\mathbf{k}}_1=\left(1+\frac{|G_{\mu}|^2}{\Omega^2}\right)\mathbf{k}_1,\quad\tilde{\mathbf{k}}_2=\left(1+\frac{|G|^2}{\Omega_{\mu}^2}\right)\mathbf{k}_2.$$

An important conclusion thus follows that if field shifts of the energy levels become commensurate with the Doppler widths of the transitions, the condition of Doppler-free transitions depends on the intensity of the interacting waves. This condition has the form $\mathbf{k}_1=-\mathbf{k}_2$ for absorption.

For scattering of radiation when the energy of level n is less than that of level m [see Fig. 1(b)], the formula for r_n is derived from Eqs. (3) and (4) by the substitutions $\Omega'_{\mu}\to-\Omega'_{\mu}$, $\mathbf{k}_2\to-\mathbf{k}_2$. In particular, the condition of Doppler-free transition $g\to n$ in this case acquires

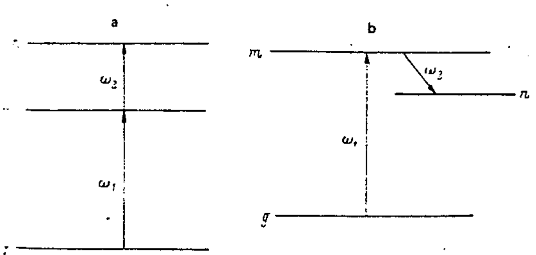

FIG. 1.

the form $\mathbf{k}_1 = \mathbf{k}_2$. In general cases, the following condition on the moduli of wave vectors must be satisfied:

$$k_2 = \frac{1 \pm \dfrac{|G_\mu|^2}{\Omega^2}}{1 \pm \dfrac{|G|^2}{\Omega_\mu^2}} k_1. \tag{7}$$

It is clear from Eq. (6) that the condition for Doppler-free transition can be satisfied with greatly different frequencies ω_1 and ω_2 if $|G| \sim |\Omega_\mu|$ or $|G_\mu| \sim |\Omega|$. With satisfaction of Eq. (7), atoms with any velocities will participate equally in transitions to level n, while the transition probability acquires a Lorentzian shape with width Γ_{gn} as a function of $\tilde{\Omega}_{gn}$. If Eq. (7) is not satisfied, by averaging over velocities (3), we obtain the following formula for the transition probability W to level n per unit time:

$$\left.\begin{aligned} W = \langle \Gamma_n r_n \rangle = \frac{a \varkappa \Gamma_n}{\bar{\Gamma}_{gn} \sqrt{1 + \varkappa}} \operatorname{Re} \left\{ \frac{\sqrt{\pi}}{|\bar{\mathbf{k}}_1 + \bar{\mathbf{k}}_2| \bar{v}} e^{p^2} [1 - \Phi(p)] \right\}, \\ p = [\bar{\Gamma}_{gn} \sqrt{1 + \varkappa} + i \tilde{\Omega}_{gn}] / |\bar{\mathbf{k}}_1 + \bar{\mathbf{k}}_2| \bar{v}. \end{aligned}\right\} \tag{8}$$

$\Phi(z)$ is the probability integral. If $|\bar{\mathbf{k}}_1 + \bar{\mathbf{k}}_2| \bar{v} \gg \bar{\Gamma}_{gn} \sqrt{1 + \kappa}$,

$$W = \frac{a \varkappa \Gamma_n}{\bar{\Gamma}_{gn} \sqrt{1 + \varkappa}} \frac{\sqrt{\pi}}{|\bar{\mathbf{k}}_1 + \bar{\mathbf{k}}_2| \bar{v}} e^{-(\tilde{\Omega}_{gn} / |\bar{\mathbf{k}}_1 + \bar{\mathbf{k}}_2| \bar{v})^2}. \tag{9}$$

Equation (7) generalizes the condition of Doppler-free absorption to the case of strong fields. In this expression the smallness of the quantities $|G/\Omega|$ and $|G_\mu/\Omega_\mu|$ is not assumed.

In weak fields for Raman-type processes, complete compensation of Doppler broadening is impossible since $\mathbf{k}_2 \neq \mathbf{k}_1$, while it is necessary for two-photon absorption that $\mathbf{k}_2 = -\mathbf{k}_1$. In the last case, the yield of the intermediate resonance will be high in a system with unequally spaced levels [see Fig. 1(a)], and this lowers the efficiency of the two-photon excitation process. Equation (7) shows that due to field perturbations, two-photon Doppler-free transition in quasi-resonant conditions for the intermediate level is possible in such a system by an appropriate choice of the field quantities despite substantial departure of ω_1 from ω_2 (and, consequently, $\mathbf{k}_1$ from $\mathbf{k}_2$).

SHARP DOPPLER-FREE RESONANCE IN A STRONG FIELD

If both fields are equally strong, an increase in the rate of two-photon transitions is accompanied by significant broadening of the two-photon resonance.

We analyze conditions under which the relation $\tilde{\Omega}_{gn} = 0$ and the equality (7) with auxiliary requirement $\Gamma_{gn}^2 (1 + \kappa) \sim \Gamma^2 \ll (k_1 + k_2) \bar{v}$, where Γ are typical intrinsic transition widths, can be satisfied simultaneously. The last requirement is equivalent to the condition $|GG_\mu|^2 / |\Omega\Omega_\mu| \sim \Gamma^2$. On the other hand, to obtain a significant difference between k_1 and k_2 [see Eq. (7)], it is necessary that $|G| \sim |\Omega_\mu|$ (or $|G_\mu| \sim |\Omega|$). It follows from the expressions obtained that these conditions are consistent if $|G| \sim |\Omega_\mu|$, $|G_\mu| \sim \Gamma$ (or $|G_\mu| \sim |\Omega|$, $|G| \sim \Gamma$). If $\omega_{mn} < \omega_{mg}$, $k_2 < k_1$ under quasi-resonant conditions with respect to the intermediate level. As is clear from Eq. (7), it is necessary to select just the field $\mathbf{E}_1$ to be the stronger one. If $\omega_{nm} > \omega_{mg}$, the situation is reversed.

We can show that if the frequency ω_1 of the stronger field is given, $\omega_{nm} < \omega_{mg}$, $|\Omega|$, $|\Omega_\mu| < \omega_{mg}, \omega_{mn}$, the peak of Doppler-free absorption is reached for this range of field intensities at the following values of the frequency ω_2 and intensity of the field $\mathbf{E}_1$:

$$\Omega_\mu = -\Omega \frac{\omega_{mn}}{2\omega_{nm} - \omega_{mg}}; \quad |G|^2 = \Omega^2 \frac{\omega_{nm} (\omega_{mg} - \omega_{nm})}{(\omega_{mg} - \omega_{nm})^2}. \tag{10}$$

If the frequency ω_2 is given, we have for the frequency and of the field $\mathbf{E}_1$

$$\Omega = -\Omega_\mu \frac{2\omega_{mn} + 2\Omega_\mu - \omega_{mg}}{\omega_{nm}}; \quad |G|^2 = \Omega_\mu^2 \frac{\omega_{mg} - \omega_{nm} - 2\Omega_\mu}{\omega_{nm}}. \tag{11}$$

Let us illustrate the conclusions drawn from estimates for the $3s - 3p - 4s$ transition of sodium. We select the frequency of the weaker field $\nu_2 = 8823$ cm^{-1}, and this corresponds to going out of resonance with the $3p$–$4s$ transition, $\Delta\nu_\mu = \nu_2 - \nu_{nm} = 50$ cm^{-1}. Since $|d_{nm}| \approx |d_{mg}| \simeq 10$ dB, $\Gamma \sim 10^9$ sec^{-1}, the condition $|G_\mu| \sim \Gamma$ is satisfied for weak field intensities $I_\mu \simeq 5$ W/cm^2. We then obtain from Eq. (11) $\nu_1 = 16{,}963$ cm^{-1} and $|G| = 48$ cm^{-1}, i.e., the required intensity of the strong field corresponds to $I \approx 10$ MW/cm^2. If $\Delta\nu_\mu \approx 1$ cm^{-1}, the required intensity value decreases, $I \approx 5$ kW/cm^2.

The example considered illustrates the possibilities of the existence of Doppler-free transition under conditions of two-photon resonance and quasi-resonance with respect to the intermediate level with substantial frequency difference and moderate field intensities participating in the process. On the contrary, another important conclusion follows from Eq. (7) that for a single-frequency pump with $\omega_1 = \omega_2$, $\mathbf{k}_1 = -\mathbf{k}_2$, if $|d_{mn}| \neq |d_{mg}|$, compensation of Doppler broadening in two-photon transition disappears with increasing pump intensity, and field broadening of the resonance increases simultaneously.

ABSORPTION LINE SHAPE FOR FIXED FREQUENCY OF ONE OF THE FIELDS

Let us consider the absorption line shape of field $\mathbf{E}_2$ in the presence of field $\mathbf{E}_1$ with fixed frequency ω_1. We find from Eq. (5) the expression for the resonance frequency

$$\Omega_\mu^{1,2} = -\frac{1}{2}\left(\Omega - \frac{|G_\mu|^2}{\Omega}\right) \pm \frac{1}{2}\sqrt{\left(\Omega - \frac{|G_\mu|^2}{\Omega}\right)^2 + 4|G|^2}. \tag{12}$$

Thus, in the prescribed $\mathbf{E}_1$ field, the absorption line shape of field $\mathbf{E}_2$ has the doublet form, one of the components of which corresponds to perturbed two-photon transition and the other, the perturbed stepwise transition. The conditions of Doppler-free absorption at the peaks of these components have the form

$$\mathbf{k}_2 = M_1 \mathbf{k}_1, \quad \mathbf{k}_2 = M_2 \mathbf{k}_1,$$

where

$$M_{1,2} = -\frac{1}{2}\left(1 + \frac{|G_\mu|^2}{\Omega^2}\right)\left[1 \pm \frac{\Omega - \dfrac{|G_\mu|^2}{\Omega}}{\sqrt{\left(\Omega - \dfrac{|G_\mu|^2}{\Omega}\right)^2 + 4|G|^2}}\right]. \tag{13}$$

In the case of weak fields $|G|$, $|G_\mu| \ll |\Omega|$; $M_1 \approx -1$, $-M_2 \ll 1$, and the condition of Doppler-free two-photon absorption assumes the well-known form of $\mathbf{k}_2 = -\mathbf{k}_1$. Equations (12) and (13) are the generalization of corresponding expressions derived in Ref. 1 for a strong field $\mathbf{E}_1$ and a weak field $\mathbf{E}_2$ to the case of two strong fields and reflect variation in the frequency–correlation properties of radiative processes in strong fields. The functions $M_{1,2}$ are correlation factors (storage size) between frequencies ω_1 and ω_2 in resonances (12), since $M_{1,2} = \partial\Omega_\mu^{1,2}/\partial\Omega$. Thus, in weak fields, the single resonance at ω_2 is completely correlated with the frequency ω_1 ($M_1 = -1$, $\Omega_\mu = -\Omega$). It corresponds to a two-photon process. The other resonance is totally uncorrelated with the frequency ω_1 ($M_2 = 0$, $\Omega_\mu = 0$). It corresponds to stepwise transition (absorption from level m).

It follows from above that the two factors M_1 and M_2 become different from both zero and -1 with increasing intensities of the fields $\mathbf{E}_1$ and $\mathbf{E}_2$. Thus, a separation of radiative processes corresponding to the two components of the spectrum into stepwise and two-photon processes becomes physically meaningless.

DOPPLER-FREE RESONANT FOUR-PHOTON PARAMETRIC PROCESSES

Doppler-free transitions can be quite evident in resonant four-photon frequency mixing of the type $\omega_s = \omega_1 + \omega_2 - \omega_3$ and $\omega_s = \omega_1 - \omega_2 \pm \omega_3$ in gases, when $\omega_1 + \omega_2$ or $\omega_1 - \omega_2$ correspond to two-photon resonance with the transition ng. In strong fields at frequencies ω_1 and ω_2 a resonance dominator of the $\tilde{P}_{ng}$ type arises in the nonlinear susceptibility. For conditions and estimates similar to those given above, Doppler broadening of this transition disappears, and at exact resonance a factor $\tilde{\Gamma}_{ng}$ appears in the denominator of the nonlinear susceptibility averaged over velocities instead of the factor $|\mathbf{k}_1 \pm \mathbf{k}_2|\bar{v}$. Thus the nonlinear susceptibility increases by $\beta = |\mathbf{k}_1 \pm \mathbf{k}_2|\bar{v}_2/\tilde{\Gamma}_{ng}$ times, and the radiation power generated at the sum frequency, by β^2 times. The value of β^2 may be 10^4–10^6. Since we can achieve the condition for Doppler-free resonance in strong fields when $\omega_1 \neq \omega_2$, i.e., under conditions of quasi-resonance to the intermediate level, the additional possibility of raising the power of generated radiation appears besides the factor β^2.

In addition to a sharp increase in the conversion factor of radiation, this phenomenon can be used for generation spectroscopy of forbidden transitions and a study of their field broadening.

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