Doppler-free transitions in four-photon parametric resonance processes

A. K. Popov and V. M. Shalaev

L. V. Kirenski Institute of Physics, Siberian Branch of the Academy of Sciences of the USSR, Krasnoyarsk

(Submitted August 20, 1979)
Kvantovaya Elektron. (Moscow) 7, 1362–1364 (June 1980)

It is shown that the resonance nonlinear susceptibility of four-photon parametric processes in gaseous media can be increased by several orders of magnitude. The increase in the susceptibility is due to suppression of the Doppler broadening of two-photon transitions in strong pump fields when the frequencies of the fields interacting with these transitions can be very different. Under these conditions there is an additional opportunity for increasing the susceptibility by reducing the frequency detuning relative to intermediate levels participating in two-photon transitions.

PACS numbers: 42.65.Cq

1. Four-photon parametric resonance processes in gaseous media are used widely in generation of infrared and ultraviolet radiation, transmission of weak infrared signals, and wavefront reversal. The nonlinear susceptibility of such processes increases resonantly when the sum or difference between two frequencies approaches the frequency of a two-photon transition. The corresponding frequency detuning from resonance, which occurs in the denominator of the expression for the nonlinear susceptibility, is replaced with the Doppler width of a two-photon transition.[1] We shall show that a suitable selection of the frequencies and intensities of the pump fields can ensure that the resonance denominator is governed not by the Doppler width but by a quantity close to the homogeneous width of the transitions occurring in a nonlinear process. This increases the resonance nonlinear susceptibility by 2-3 orders of magnitude and enhances correspondingly the conversion coefficient by 4-6 orders. Hence, it follows that forbidden transitions can be studied by laser spectroscopy.

2. We shall consider resonance processes of the $\omega_s = \omega_1 - \omega_2 + \omega_3$ (Fig. 1a) and $\omega_s = \omega_1 + \omega_2 - \omega_3$ (Fig. 1b) type, where $\omega_1 \pm \omega_2 \approx \omega_{20}$. We shall give the treatment for the case shown in Fig. 1a, but the results will apply also (subject to a simple modification) to the case shown in Fig. 1b.

Under steady-state conditions the system of equations for elements of the density matrix reduces to a system of algebraic equations

$$\left.\begin{array}{l} P_{01}r_{01}=iG_{01}\Delta_{01}+ir_{02}G_{21}, \\ P_{12}r_{12}=iG_{12}\Delta_{12}-iG_{10}r_{02}, \\ P_{02}r_{02}=-iG_{01}r_{12}+ir_{01}G_{12}+ir_{03}G_{32}, \\ P_{03}r_{03}=ir_{02}G_{23}, \\ \Gamma_2 r_2=-2\mathrm{Re}\{iG_{21}r_{12}\}, \\ \Gamma_1 r_1=2\mathrm{Re}\{ir_{12}G_{21}-ir_{01}G_{10}\}. \end{array}\right\} \quad (1)$$

Here, r_{ij} are the amplitudes of the elements of the density matrix ρ_{ij} considered in the interaction representation {for example, $\rho_{02}=r_{02}\exp i[\Omega_{02}t-(\mathbf{k}_1-\mathbf{k}_2)\mathbf{r}]$}; $\mathbf{k}_j$ are the wave vectors of the radiations involved; $\Omega_{02}+\Omega_{01}-\Omega_{12}=(\omega_1-\omega_{10})-(\omega_2-\omega_{12})$; Γ_j are the population-relaxation rates; Γ_{ij} are the half-widths of the transitions; $P_{ij}=\Gamma_{ij}+i\Omega_{ij}$; $\Omega'_{02}=\Omega_{02}-(\mathbf{k}_1-\mathbf{k}_2)\mathbf{v}$; $\Omega'_{01}-\Omega_{01}-\mathbf{k}_1\mathbf{v}$; $\mathbf{v}$ is the velocity of atoms; $\Delta_{ij}=r_i-r_j$ is the difference between the populations; $G_{ij}=-\mathbf{E}_j\mathbf{d}_{ij}/2\hbar$.

We shall introduce the following notation:

$$a=\widetilde{\Gamma}_{02}^2\bar{\Gamma}_1\left[\bar{\Gamma}_1-2\bar{\Gamma}_2+\Gamma_1\Omega_{01}/\Omega_{12}-\left(1-2\Omega_{12}/\Omega_{01}-\Omega_{01}/\Omega_{12}\right)\Gamma_2\right]^{-1};$$

$$b=-\widetilde{\Gamma}_{02}^2\left(\bar{\Gamma}_2-\Gamma_2\Omega_{12}/\Omega_{01}\right)\left[\bar{\Gamma}_1-2\bar{\Gamma}_2+\Gamma_1\Omega_{01}/\Omega_{12}-\left(1-2\Omega_{12}/\Omega_{01}-\Omega'_{01}/\Omega_{12}\right)\Gamma_2\right]^{-1};$$

$$\varkappa=2\frac{|G_{01}G_{12}|^2}{\Omega_{01}\Omega_{12}\widetilde{\Gamma}_{02}}\times\frac{\bar{\Gamma}_1-2\bar{\Gamma}_2+\Gamma_1\Omega_{01}/\Omega_{12}-(1-2\Omega_{12}/\Omega_{01}-\Omega_{01}/\Omega_{12})\Gamma_2}{\Gamma_1\Gamma_2+2|G_{12}|^2\Omega_{12}^{-2}\Gamma_{12}\left(\Gamma_1+\Gamma_2\right)+4|G_{01}|^2\Omega_{12}^{-2}\Gamma_{01}\left(\Gamma_2+3\Gamma_{12}|G_{12}|^2/\Omega_{12}^2\right)}, \quad (2)$$

where

$$\bar{\Gamma}_1=\Gamma_1+2[\Gamma_{01}|G_{01}|^2/(\Omega_{01}\Omega_{12})+\Gamma_{12}|G_{12}|^2/(\Omega_{01}\Omega_{12})];\quad \bar{\Gamma}_2=\Gamma_2-2[\Gamma_{01}|G_{01}|^2/(\Omega_{01}\Omega_{12})+\Gamma_{12}|G_{12}|^2/(\Omega_{01}\Omega_{12})].$$

Then, the solution of Eq. (1) can be represented in the form

$$r_{03}=iG_{01}G_{12}G_{23}(\Delta_{01}-\Delta_{12}P_{01}/P_{12})/P_{01}\widetilde{P}_{02}P_{03}. \quad (3)$$

Here,

$$r_2=\frac{a\varkappa}{\widetilde{\Omega}_{02}'^2+\widetilde{\Gamma}_{02}(1+\varkappa)};\quad r_1=\frac{b\varkappa}{\widetilde{\Omega}_{02}'^2+\widetilde{\Gamma}_{02}(1+\varkappa)}+\frac{(\Omega_{12}/\Omega_{01})\varkappa|G_{12}|^{-2}\widetilde{\Gamma}_{02}\Gamma_{02}\left(\Gamma_2+2|G_{12}|^2\Omega_{12}^{-2}\Gamma_{12}\right)}{\Gamma_1-2\Gamma_2+\Gamma_1\Omega_{01}/\Omega_{12}-\left(1-2\Omega_{12}/\Omega_{01}-\Omega_{01}/\Omega_{12}\right)\Gamma_2}\,\frac{\widetilde{\Omega}_{02}'^2+\widetilde{\Gamma}_{02}^2}{\widetilde{\Omega}_{02}'^2+\widetilde{\Gamma}_{02}^2(1+\varkappa)}; \quad (4)$$

$$\widetilde{P}_{02}=\left(\Gamma_{02}+\Gamma_{12}|G_{01}|^2/\Omega_{12}'^2+\Gamma_{01}|G_{12}|^2/\Omega_{01}'^2\right)+i\left(\Omega'_{02}+|G_{01}|^2/\Omega_{12}-|G_{12}|^2/\Omega_{01}\right)=\widetilde{\Gamma}_{02}+i\widetilde{\Omega}_{02}. \quad (5)$$

The expressions (2)–(5) are obtained on the assumption that $|\Omega_{03}|, |\Omega_{23}| \gg |G_{23}|$, but in Eqs. (3) and (4) it is not assumed that the ratios $|G_{01}/\Omega_{01}|$ and $|G_{12}/\Omega_{12}|$ are small. If $|\Omega_{01}| \gg k_1\bar{v}$ and $|\Omega_{12}| \gg k_2\bar{v}$ ($\bar{v}$ is the most probable velocity) and if the Doppler shifts are included in Eq. (5) in the first nonvanishing approximation, we obtain)

$$\widetilde{P}_{02}=\widetilde{\Gamma}_{02}+i\widetilde{\Omega}_{02}-i[(1+|G_{12}|^2/\Omega_{01}^2)\mathbf{k}_1-(1+|G_{01}|^2/\Omega_{12}^2)\mathbf{k}_2]\mathbf{v}=\widetilde{\Gamma}_{02}+i\widetilde{\Omega}_{02}-i\widetilde{\mathbf{k}}_1\mathbf{v}+i\widetilde{\mathbf{k}}_2\mathbf{v}, \quad (6)$$

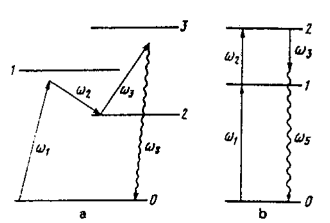

FIG. 1.

where

$\tilde{k}_1=(1+|G_{12}|^2/\Omega_{01}^2)k_1$; $\tilde{k}_2=(1+|G_{01}|^2/\Omega_{12}^2)k_2$.

3. Equation (6) readily yields the condition which must be satisfied by the wave-vector moduli to ensure that the Doppler shifts do not appear in $\tilde{P}_{02}$, i.e., $\tilde{\Omega}'_{02}=\tilde{\Omega}_{02}$,

$$k_2=(1+|G_{12}|^2/\Omega_{01}^2)k_1/(1+|G_{01}|^2/\Omega_{12}^2). \tag{7}$$

In the case of Fig. 1a we should have $\tilde{\mathbf{k}}_1=\tilde{\mathbf{k}}_2$, whereas in the case of Fig. 1b, we should have $\tilde{\mathbf{k}}_1=-\tilde{\mathbf{k}}_2$. It should be noted that if both fields have the same intensity, it follows from Eqs. (2)–(4) that the resonance suffers considerable field broadening. We shall analyze the conditions under which the relationship $\tilde{\Omega}_{02}=0$ applies and Eq. (7) is valid subject to the additional requirement $\tilde{\Gamma}_{02}^2(1+\varkappa)\sim\Gamma^2\ll(k_1+k_2)\bar{v}$, where Γ is the characteristic natural width of a transition. This last requirement is equivalent to the condition $|G_{01}G_{12}|^2/|\Omega_{01}\Omega_{12}|\sim\Gamma^2$. On the other hand, a considerable difference between k_1 and k_2 in Eq. (7) requires that $|G_{01}|\sim|\Omega_{12}|$ (or $|G_{12}|\sim|\Omega_{01}|$). It follows from the expressions obtained that these conditions are compatible if $|G_{01}|\sim|\Omega_{12}|, |G_{12}|\sim\Gamma$ (or $|G_{12}|\sim|\Omega_{01}|, |G_{01}|\sim\Gamma$). In the scheme of Fig. 1a, we have $k_1>k_2$. Then, it is clear from Eq. (7) that the stronger field should be $\mathbf{E}_1$, which we shall assume henceforth.

Then, we have $r_2\ll r_1$, and r_1 is given by the expression

$$r_1=\frac{2|G_{01}/\Omega_{01}|^2\Gamma_{01}}{\Gamma_2+4|G_{01}/\Omega_{01}|^2\Gamma_{01}}. \tag{8}$$

In this case the susceptibility χ has the form:

$$\chi=-\left\langle\frac{d_{01}d_{12}d_{23}d_{30}\,[1-(2-\Omega_{01}/\Omega_{12})\,2\,|G_{01}/\Omega_{01}|^2\,\Gamma_{01}/(\Gamma_2+4\,|G_{01}/\Omega_{01}|^2\Gamma_{01})]}{\Omega_{01}\Omega_{03}(\tilde{\Omega}'_{02}-i\tilde{\Gamma}_{02})}\right\rangle. \tag{9}$$

If $k_2>k_1$, it follows from Eq. (7) that E_2 should be taken as the stronger field. We then have $r_1\sim r_2\ll 1$ and the susceptibility simplifies to

$$\chi=-\left\langle\frac{d_{01}d_{12}d_{23}d_{30}}{\Omega_{01}\Omega_{03}(\tilde{\Omega}'_{02}-i\tilde{\Gamma}_{02})}\right\rangle. \tag{10}$$

The angular brackets in Eqs. (9) and (10) represent averaging over the atomic velocities. If $\tilde{\Omega}_{02}=0$, the susceptibility averaged over these velocities is inversely proportional to $(\tilde{k}_1-\tilde{k}_2)\bar{v}$, which is generally much greater than $\tilde{\Gamma}_{02}$. However, if the condition of Eq. (7) is satisfied, the Doppler broadening is suppressed and the susceptibility increases by a factor $\beta=(\tilde{k}_1-\tilde{k}_2)\bar{v}/\tilde{\Gamma}_{02}\approx 10^3-10^4$.

4. Estimates indicate that for $|\Omega_{12}|\sim 1\ \mathrm{cm}^{-1}$ and $|d_{01}|\sim 10$ D, the value $|G_{01}|\sim|\Omega_{12}|$ is obtained for moderate radiation intensities of ~1 kW/cm^2. For these intensities the coefficient in front of k_1 in Eq. (7) can differ significantly from unity.

In the case of weak fields, Eq. (7) reduces to the usual requirement that $k_1=k_2$. In other words, even in the case of a considerable difference between the frequencies of the photons interacting with a forbidden transition (and, consequently, in the case of a considerable difference between k_1 and k_2), we find—in the scheme of Fig. 1a—that the Doppler broadening can be suppressed by pumping at moderate rates.

Since ω_1 may differ considerably from ω_2, the scheme in Fig. 1b makes it possible to observe a nonlinear interaction with the Doppler broadening suppressed when conditions correspond to a quasiresonance relative to an intermediate level and this results in an additional increase in the nonlinear susceptibility. It should be noted that in a weak field the Doppler broadening can be suppressed only in the scheme of Fig. 1b if $\mathbf{k}_1=-\mathbf{k}_2$, which usually corresponds to a greater detuning from the intermediate resonance.

[1]P. P. Sorokin, J. J. Wynne, J. A. Armstrong, and R. T. Hodgson, Ann. Acad. Sci. N. Y. 267, 30 (1976).

Translated by A. Tybulewicz