Four-wave mixing under conditions of Doppler-free resonance induced by strong radiation

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The possibilities of enhancing the nonlinear optical response of a gaseous medium and the radiation conversion efficiency in four-wave mixing processes through the use of atomic coherence effects and the induced elimination of the Doppler broadening of resonances are investigated. Numerical illustrations of the effects are given for experiments in progress.
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1. A great deal of attention is being devoted to experimental and theoretical investigations of resonant coherent interaction of electromagnetic fields on quantum transitions (see, for example, Refs. 1 and 2). This is explained by the favorable possibilities of studying quantum interference phenomena that in turn form the basis for the manipulation of the nonlinear optical properties of atomic–molecular systems and resonant conversion of laser radiation. There are substantially fewer publications on the characteristic features of the coherent interaction at Doppler-broadened transitions, which strongly influence the manifestation of the processes indicated. Our objective in the present letter is to show that new resonances in the nonlinear susceptibility, which are free of Doppler broadening, can be produced using auxiliary strong fields and atomic coherence. A substantial increase in the quantum efficiency of conversion is achieved even though the absorption of the initial radiations increases at the same time. A model corresponding to the experimental conditions of Ref. 1 is used for numerical illustrations. The elementary physical processes on which the proposed method is based and which consist of a change in the frequency-correlation properties of multiphoton processes in strong electromagnetic fields are discussed in greater detail in Ref. 3.

2. Consider the interaction scheme presented in Fig. 1. The wave $\mathbf{E}_3(t,\mathbf{r})$, interacting with the transition 2–3, consists of two components, $E_3^+(t,\mathbf{r})$ and $E_3^-(t,\mathbf{r})$, having the same frequencies but propagating in opposite directions. The wave vector of the component E_3^+ and the wave vector of the component E_3^- are parallel and antiparallel, respectively, to the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. It is assumed that only the lower level is occupied, and the radiation field at the 0–1 transition is so weak that the change in the population of this level can be neglected. The component E_3^+ of the field at the frequency ω_3 is also

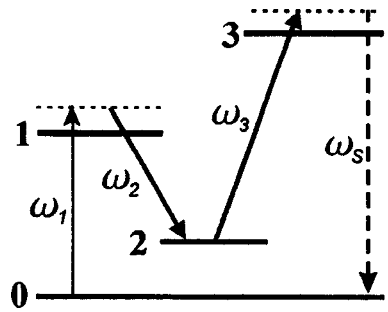

FIG. 1. Resonant four-wave interaction scheme.

weak, and the component E_3^- and the field E_2, interacting with the transition 1–2, can be of arbitrary intensity.

The system of equations for the density matrix in the interaction representation has the form

$$L_{03}\rho_{03}=i\{\rho_{00}V_{03}+\rho_{02}(V_{23}^{+}+V_{23}^{-})\},\quad L_{01}\rho_{01}=i\{\rho_{00}V_{01}+\rho_{02}V_{21}\},$$
$$L_{02}\rho_{02}=i\{\rho_{01}V_{12}+\rho_{03}(V_{32}^{+}+V_{32}^{-})\},\tag{1}$$

where

$$L_{ij}=\partial/\partial t+\mathbf{v}\cdot\boldsymbol{\nabla}+\Gamma_{ij},\quad V_{ij}=G_{ij}\exp\{i(\Omega_i t-k_i z)\},$$
$$V_{23}^{\pm}=G_{23}^{\pm}\exp\{i(\Omega_3 t\mp k_3 z)\},\quad G_{ij}=-E_j d_{ij}/2\hbar,\quad \text{and}\quad G_{23}^{\pm}=-E_3^{\pm}d_{23}/2\hbar$$

are the Rabi frequencies characterizing the interaction, Ω_i is the detuning from resonance for the corresponding resonant field (for example, $\Omega_1=\omega_1-\omega_{01}$), and Γ_{ij} are the homogeneous half-widths of the transitions.

Thus, the induced atomic coherence ρ_{02} is a source of new components in the polarizations responsible for the absorption and generation of radiation. We shall represent the off-diagonal elements ρ_{01}, ρ_{02}, and ρ_{03} as products of amplitude and phase factors:

$$\rho_{01}=r_{01}\exp\{i(\Omega_1-k_1 z)\}+r_{01}^{-}\exp\{i[\Omega_1 t-(k_2+k_3+k_4)z]\},$$
$$\rho_{02}=r_{02}\exp\{i[(\Omega_1-\Omega_2)t-(k_1-k_2)z]\}+r_{02}^{-}\exp\{i[(\Omega_1-\Omega_2)t-(k_3+k_4)z]\},$$
$$\rho_{03}=r_{03}\exp\{i[\Omega_4 t-k_4 z]+\tilde{r}_{03}^{+}\exp\{i[(\Omega_1-\Omega_2+\Omega_3)t-(k_1-k_2+k_3)z]\}$$
$$+\tilde{r}_{03}^{-}\exp\{i[(\Omega_1-\Omega_2+\Omega_3)t-(k_1-k_2-k_3)z]\}.$$

Substituting these expressions into Eq. (1) and solving the system of algebraic equations for the amplitudes of the off-diagonal elements of the density matrix, we obtain expressions for the off-diagonal element r_{03} characterizing absorption and refraction of the wave E_4 in the presence of the strong fields E_2 and E_3^- and for the off-diagonal element $\tilde{r}_{03}^{+}$ responsible for nonlinear polarization and susceptibility at the generated frequency $\omega_S=\omega_1-\omega_2+\omega_3$:

$$r_{03}=i\frac{G_{03}}{P_{03}}\frac{P_{01}^{-}P_{02}^{-}+|G_{12}|^2}{P_{01}^{-}\{P_{02}^{-}+|G_{23}^{-}|^2/P_{03}+|G_{12}|^2/P_{01}^{-}\}}, \tag{2}$$

$$\tilde{r}_{03}^{+}=r_{01}\frac{G_{12}G_{23}^{+}}{P_{02}P_{03}^{+}}\frac{1}{1+|G_{23}^{-}|^2/P_{03}^{-}P_{02}}\frac{1}{1+|G_{23}^{-}|^2/P_{03}^{+}P_{02}^{-}}, \tag{3}$$

where

$$P_{02}=\Gamma_{02}+i[\Omega_1-\Omega_2-(k_1-k_2)v],\quad P_{02}^{-}=\Gamma_{02}+i[\Omega_1-\Omega_2-(k_3+k_4)v],$$

$$P_{03}=\Gamma_{03}+i(\Omega_4-k_4v),$$

$$P_{03}^{-}=\Gamma_{03}+i(\Omega_1-\Omega_2+\Omega_3-k_S^{-}v),\quad P_{03}^{+}=\Gamma_{03}+i(\Omega_1-\Omega_2+\Omega_3-k_S^{+}v),$$

$$P_{01}^{-}=\Gamma_{01}+i[\Omega_1-(k_2+k_3+k_4)v],\quad k_S^{-}=k_1-k_2-k_3,\quad k_S^{+}=k_1-k_2+k_3,$$

and v is the projection of the velocity of the atom on the z direction.

The solution for the off-diagonal element r_{01}, characterizing absorption and refraction of the E_1 wave in the presence of the strong fields E_2 and E_3^{-}, has the form[4]

$$r_{01}=-i\frac{G_{01}}{P_{01}}\frac{P_{02}P_{03}^{-}+|G_{23}^{-}|^2}{P_{03}^{-}\{P_{02}+|G_{23}^{-}|^2/P_{03}^{-}+|G_{12}|^2/P_{01}\}}, \tag{4}$$

where $P_{01}=\Gamma_{01}+i(\Omega_1-k_1v)$. Using it, in lowest order of perturbation theory in G_{23}^{+} we obtain from Eq. (3) the solution for the off-diagonal element $\tilde{r}_{03}^{+}$:

$$\tilde{r}_{03}^{+}=\frac{i}{P_{01}}\frac{G_{01}G_{12}G_{23}^{+}}{(P_{02}+|G_{23}^{-}|^2/P_{03}^{-}+|G_{12}|^2/P_{01})(P_{03}^{+}+|G_{23}^{-}|^2/P_{02}^{+})}. \tag{5}$$

As the intensity of the interacting radiations increases, the nonlinear polarization at first increases and then saturates. The higher the intensity, the larger the detunings from the resonances and the higher the polarizations at which saturation occurs. Intensities corresponding to resonant or quasiresonant optimal conditions are characteristic for continuous-wave lasers. In gaseous media which possess sharper and stronger resonances than condensed media, on account of Doppler effects only a small fraction of the atoms is in one of the resonances.

We shall show that this limitation can be overcome by making use of the effects considered above. For detunings from the unperturbed resonances substantially greater than the corresponding Doppler widths, $|G|^2/P\approx|G|^2/p-ikv|G|^2/p^2$, where p are the corresponding P factors at $v=0$. Thus, together with a shift and broadening of the resonances, strong fields induce additional Doppler shifts.[3,5] Since Re$\{|G|^2/p^2\}<0$, $k_S^{-}<0$, it follows from Eq. (5) that there can be total compensation of the Doppler shifts and elimination of the inhomogeneous broadening of a two-photon resonance modified by strong fields. Since in the process all atoms, irrespective of their velocities, are simultaneously entrapped in the indicated resonance, a substantial increase of the nonlinear susceptibility and a simultaneous decrease of the saturation of nonlinear polarization can be expected. Using the indicated procedure, the cofactor, describing the modified two-photon resonance, in the denominator of Eq. (5) can be represented in the form

$$\tilde{P}_{02} \equiv P_{02} + |G_{12}|^2/P_{01} + |G_{23}^-|^2/P_{03}^-$$
$$\approx \tilde{\Gamma}_{02} + i\tilde{\Omega}_{02} - i\left\{ \left(1 + \frac{|G_{23}^-|^2}{\Omega_4^2} \right)(k_1 - k_2) + \frac{|G_{12}|^2}{\Omega_1^2} k_1 - \frac{|G_{23}^-|^2}{\Omega_4^2} k_3 \right\} v, \quad (6)$$

where $\tilde{\Gamma}_{02}$ and $\tilde{\Omega}_{02}$ are the half-width and the position of the resonance with the field-induced broadening and shift taken into account. The conditions for eliminating the dependence on v and therefore the Doppler broadening of the induced resonance follow from Eq. (6).

We underscore that all effects considered here and below are in no way due to the motion of populations, but rather they are a consequence of the appearance of coherent superpositions of quantum states as a result of the modulation of the wave functions in strong fields.[3] This is reflected in the appearance of traveling waves of coherence (described by the off-diagonal elements of the density matrix) on forbidden transitions and in the modification of their spectral dependences.

In contrast to the absorption and refraction processes studied in Ref. 3, where the relative directions of propagation of the interacting waves can be arbitrary, frequency-mixing processes require wave matching. This dictates only parallel directions of propagation of the parametrically interacting waves. The strong counterpropagating wave E_3^- does not participate directly in the photon conversion process. It only perturbs the quantum system. The effect can also be interpreted as a contribution of higher-order resonance processes, in which the number of absorbed and emitted counterpropagating photons is the same, so that this does not result in momentum nonconservation for the photons undergoing conversion. The resonances of the system and the degree of their inhomogeneous broadening can be manipulated by making use of the counterpropagating wave, and in this manner the conversion of radiations can be improved by making the correct choice of their intensity and frequency.

In contrast to cascade schemes, for transitions of the Raman scattering type k_1 is always greater than k_2. Induced elimination of the Doppler broadening of a quasi-two-photon transition by the strong field E_2 is impossible. The proposed method, which is based on the use of an additional counterpropagating wave E_3^-, makes it possible to overcome this limitation.

3. We shall illustrate the results of averaging with a Maxwellian velocity distribution and the effect of eliminating the Doppler broadening of the induced resonances on the absorption and the nonlinear susceptibility. For numerical analysis, we employ the parameters of the transitions of the sodium dimer molecule with the following wavelengths:[1] $\lambda_{01} = 661$ nm, $\lambda_{12} = 746$ nm, $\lambda_{23} = 514$ nm, and $\lambda_{03} = 473$ nm. The corresponding homogeneous half-widths of the transitions are 20.69, 23.08, 18.30, and 15.92 MHz, and the Doppler half widths are 0.678, 0.601, 0.873, and 0.948 GHz.

Figure 2 shows the normalized squared modulus of the velocity-averaged nonlinear susceptibility $(\tilde{\chi})$ that determines the four-wave interaction of the radiations versus the probe-field detuning normalized to the Doppler half-width $(\Delta\omega_{1D})$ of its transition. Normalization was performed for the value of the same quantity in the frequency range of interest in the absence of the counterpropagating wave. The strong field of the counterpropagating wave produces a substantial shift and narrowing of the resonance in the

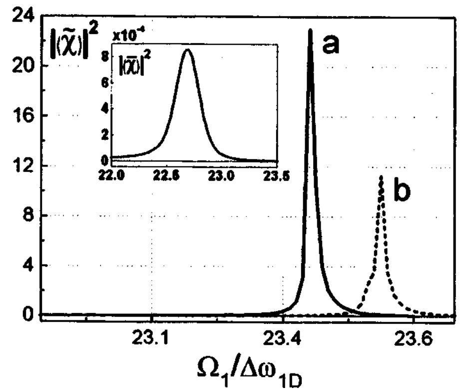

FIG. 2. Doppler-free resonance induced in the nonlinear susceptibility by the strong radiation of a counter-propagating wave. $\langle\tilde{\chi}\rangle$ is the velocity-averaged value of the normalized nonlinear susceptibility: a — under conditions of complete compensation of the Doppler broadening of the transition 0–2; b — under conditions of partial compensation, $\langle\overline{\chi}\rangle$ is the same, in the absence of the counterpropagating wave.

nonlinear susceptibility. This follows from a comparison of the curve in the inset, showing the spectral dependence of the same quantity in the absence of the counterpropagating wave and normalized to the corresponding value with simultaneous exact one- and multiphoton resonances in vanishingly weak fields. The Rabi frequency and detuning of the field E_2 are 2.35 GHz and 18.46 GHz ($27.2\cdot\Delta\omega_{1D}$), and the detuning of the field E_3^- is 1.83 GHz ($2.7\cdot\Delta\omega_{1D}$). Curve (a) corresponds to the optimal Rabi frequency, 0.89 GHz, of the field E_3^- in which Doppler broadening of the transition 0–2 is eliminated. For curve (b) the Rabi frequency, 1.06 GHz, is higher than the optimal value. The half-width of the resonance in the inset is approximately 80 MHz, which corresponds to a Doppler width of the unperturbed Raman transition of 7.8 MHz for resonance (a) and 8.4 MHz for resonance (b).

The formulas for the absorption coefficients have the form

$$\alpha_1(\Omega_1)=\alpha_{01}\,\mathrm{Re}\left\{\frac{\Gamma_{01}}{P_{01}}\,\frac{P_{02}P_{03}^-+|G_{23}^-|^2}{P_{03}^-\{P_{02}+|G_{23}^-|^2/P_{03}^-+|G_{12}|^2/P_{01}\}}\right\}, \tag{7}$$

$$\alpha_3(\Omega_S)=\alpha_{03}\,\mathrm{Re}\left\{\frac{\Gamma_{03}}{P_{03}}\,\frac{P_{01}^-P_{02}^-+|G_{12}|^2}{P_{01}^-\{P_{02}^-+|G_{23}^-|^2/P_{03}+|G_{12}|^2/P_{01}^-\}}\right\}. \tag{8}$$

Analysis of these velocity-averaged expressions shows that the maxima of the absorption and nonlinear susceptibility as a function of Ω_1 do not coincide with each other.

4. Let us consider the combined effect of induced Doppler-free resonances in the absorption and nonlinear polarization on the generation of radiation. We seek a solution in the form

$$E^j(z,t)=\mathrm{Re}\{E_j(z)\exp[i(\omega_j t-k_j z)]\}, \tag{9}$$

where k_j is the complex wave number at the corresponding frequency $k_j=k_j'-i\alpha_j/2$.

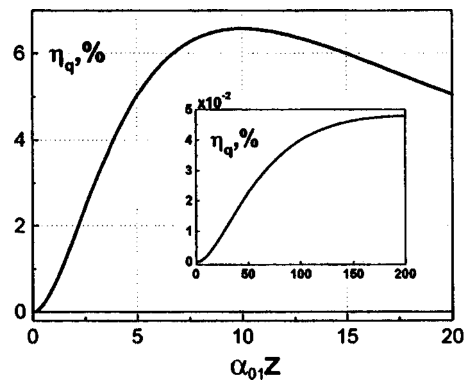

FIG. 3. Increase in the quantum efficiency of the conversion of E_1 into E_S radiation as a result of an induced Doppler-free resonance (α_{01z} is the optical thickness of the medium at frequency ω_{01}).

We shall confine our attention to the case of relatively small conversion factors, such that the fields E_2 and $E_3^{\pm}$ can be assumed to be constant throughout the medium, and only exponential absorption is taken into account for E_1. Then the truncated equation for $E_S(z)$ has the form

$$dE_S(z)/dz = i2\pi k_S' \chi_S^{(3)} E_1(0) E_2^* E_3 \exp(-i\Delta kz), \tag{10}$$

where $\chi_S^{(3)}$ is the effective nonlinear susceptibility, investigated above, for the four-wave parametric process $\omega_S = \omega_1 - \omega_2 + \omega_3$, $\Delta k = k_S - k_1 + k_2 - k_3$. The quantum conversion efficiency (QCE) for conversion of E_1 into E_S radiation at the exit from the medium is given by the formula

$$\eta_{\mathrm{q}} = (\omega_1/\omega_S)|E_S(z)/E_1(0)|^2 \exp(-\alpha_S z). \tag{11}$$

From Eq. (10) we obtain

$$\eta_{\mathrm{q}}(z) = (\omega_1/\omega_S)(|2\pi\chi_S^{(3)} E_2 E_3|^2/|\Delta k|^2)\exp(-\alpha_S z)|\exp(-i\Delta kz) - 1|^2. \tag{12}$$

Figure 3 shows the QCE versus the optical thickness of the medium with probe-field detuning corresponding to the peak of the nonlinear susceptibility under the conditions of phase matching ($\Delta k' = 0$) and compensation of Doppler broadening of the transition 0–2. The Franck–Condon factors for the transitions considered in the sodium dimer were used in the calculation. The inset shows the same dependence but with $E_3^- = 0$, in which case the conditions for the elimination of Doppler broadening are not satisfied. Comparing these curves shows a large increase in the conversion efficiency, which solves the problem posed in this work.

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[1] S. Babin, U. Hinze, E. Tiemann, and B. Wellegehausen, Opt. Lett. **21**, 1186 (1996).
[2] M. D. Lukin *et al.*, Phys. Rev. Lett. **81**, 2675 (1998).
[3] A. S. Baev and A. K. Popov, JETP Lett. **67**, 1018 (1998); A. K. Popov and V. M. Shalaev, Phys. Rev. A (1999), to be published; T. Ya. Popov, A. K. Popov, S. G. Rautian, and A. A. Feoktistov, Zh. Eksp. Teor. Fiz. **57**, 444 (1969) [Sov. Phys. JETP **30**, 243 (1970)].
[4] A. K. Popov, Izv. Ross. Akad. Nauk, Ser. Fiz. **60**, 99 (1996).
[5] C. Cohen-Tannoudji *et al.*, Opt. Commun. **27**, 71 (1978); S. Reynaud *et al.*, Phys. Rev. Lett. **42**, 756 (1979); S. Reynaud *et al.*, Opt. Commun. **42**, 39 (1982); G. Vemuri, G. S. Agarwal, and B. Rao, Phys. Rev. A **53**, 2842 (1996).

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