Quantum interference and Manley–Rowe relations in resonant four-wave frequency mixing in an optically thick Doppler-broadened medium

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It is shown that under resonant interaction conditions certain notions in nonlinear optics which are based on the Manley–Rowe relations no longer hold because of the interference of elementary quantum-mechanical processes. This conclusion is illustrated by numerical examples corresponding to the experiments performed. © *1999 American Institute of Physics.* [S0021-3640(99)00512-5]

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Coherent quantum processes accompanying the interaction of laser radiation with multilevel systems are attracting a great deal of interest because of the possibility of using such processes to manipulate nonlinear-optical responses, the populations of energy levels, refractive indices, and absorption in resonant media.[1,2] Such processes include resonant four-wave processes (RFPs). However, under resonant conditions, together with a giant increase in the nonlinear susceptibilities, many other attendant processes start to play a principal role. This can lead to qualitative contradictions with conventional ideas that hold in limiting cases. The present letter is devoted to this question.

Relatively few experiments have been performed to investigate quantum interference in RFPs in a continuous-wave monochromatic radiation field. Such experiments are difficult because three quite powerful single-frequency lasers with frequency tuning near the resonances must be used simultaneously. One possible solution is to use a Raman transition scheme. Then only two tunable radiations are required in order to obtain RFP by generation of a third radiation on an adjacent transition in the optical pump field. In addition, if molecules with many closely spaced levels are used instead of atoms, then the characteristics of the interacting transitions can be varied and generation can be tuned over a wide frequency range. This possibility was recently realized using the double-Λ transition scheme in sodium dimers (Fig. 1).[3] Similar possibilities have been demonstrated for iodine molecules. For this reason we take as an example the transition scheme

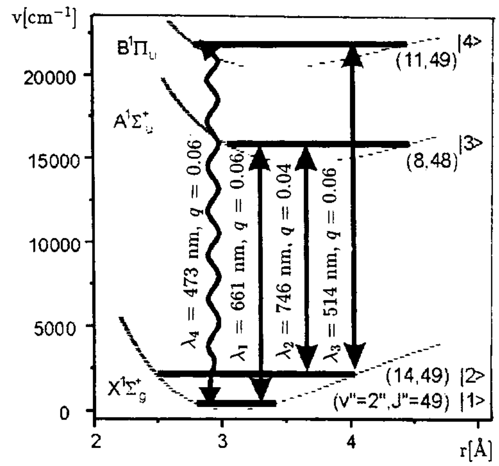

FIG. 1. Scheme of energy levels and transitions in the Na_2 molecule which were used in the experiment of Ref. 3 (the Raman laser runs on the transitions 1-3-2).

corresponding to Fig. 1. The system of wave equations for the slow complex field amplitudes is

$$\frac{dE_{4,2}(z)}{dz}=i\sigma_{4,2}(z)E_{4,2}+i\tilde{\sigma}_{4,2}(z)E_1E_3E^*_{2,4}, \tag{1}$$

$$\frac{dE_{1,3}(z)}{dz}=i\sigma_{1,3}(z)E_{1,3}+i\tilde{\sigma}_{1,3}(z)E_4E_2E^*_{3,1}. \tag{2}$$

Here $\sigma_j(z)=-2\pi k_j\chi_j(z)=\delta k_j(z)+i\alpha_j(z)/2$, δk_j and α_j are the intensity-dependent resonant components of the wave numbers and absorption coefficients, $\tilde{\sigma}_4(z)=-2\pi k_4\tilde{\chi}_4(z)$ and so on are intensity-dependent complex nonlinear-coupling parameters for four-wave mixing, and χ_j and $\tilde{\chi}_j$ are the corresponding susceptibilities. The condition $\omega_4+\omega_2=\omega_1+\omega_3$ is assumed to hold. Switching to real amplitudes and phases, we obtain

$$dA_{4,2}/dz=-\alpha_{4,2}A_{4,2}/2-(\tilde{\sigma}''_{4,2}\cos\Theta+\tilde{\sigma}'_{4,2}\sin\Theta)A_1A_{2,4}A_3, \tag{3}$$

$$A_{4,2}d\phi_{4,2}/dz=\delta k_{4,2}A_{4,2}-(\tilde{\sigma}''_{4,2}\sin\Theta-\tilde{\sigma}'_{4,2}\cos\Theta)A_1A_{2,4}A_3, \tag{4}$$

$$dA_{1,3}/dz=-\alpha_{1,3}A_{1,3}/2-(\tilde{\sigma}''_{1,3}\cos\Theta-\tilde{\sigma}'_{1,3}\sin\Theta)A_2A_{3,1}A_4, \tag{5}$$

$$A_{1,3}d\phi_{1,3}/dz=\delta k_{1,3}A_{1,3}+(\tilde{\sigma}''_{1,3}\sin\Theta+\tilde{\sigma}'_{1,3}\cos\Theta)A_2A_{3,1}A_4, \tag{6}$$

where $\Theta=\phi_1-\phi_2+\phi_3-\phi_4-\Delta kz$ and $\Delta k=k_1-k_2+k_3-k_4$.

If several strong fields interact with a multilevel system, then the simultaneously occurring elementary processes and interfering quantum paths together lead to a complicated dependence of the optical characteristics of the medium on the frequency and

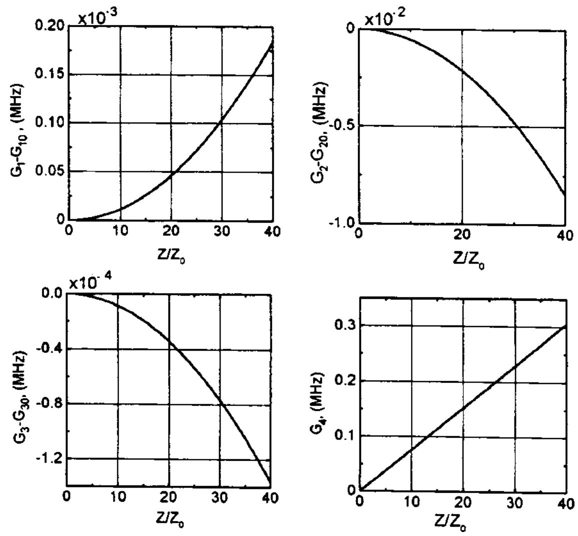

FIG. 2. Completely resonant four-wave conversion on Doppler-broadened transitions. All other accompanying processes are neglected. G_i — interaction parameters (Rabi frequencies, in MHz), G_{i0} — values at the entrance into the medium, Z — thickness of the medium, Z_0 — resonant absorption length of the generated radiation (at the frequency ω_4) in zero fields. $G_{10}=50$, $G_{20}=1, G_{30}=40$ (in MHz). These and the relaxation parameters correspond to the experimental parameters.

intensity of the radiation as well as on all the relaxation parameters by which individual processes can be discriminated.[4] In the limit of weak nonperturbing radiations the expressions for the susceptibilities $\tilde{\chi}_i$ become

$$\tilde{\chi}_2=\frac{iK}{d_2}\left[\frac{1}{P_{43}^*}\left(\frac{\Delta n_4}{P_4^*}+\frac{\Delta n_3}{P_3}\right)+\frac{1}{P_{41}^*}\left(\frac{\Delta n_1}{P_1}+\frac{\Delta n_4}{P_4^*}\right)\right], \tag{7}$$

$$\tilde{\chi}_4=\frac{iK}{d_4}\left[\frac{1}{P_{12}}\left(\frac{\Delta n_1}{P_1}+\frac{\Delta n_2}{P_2^*}\right)+\frac{1}{P_{32}}\left(\frac{\Delta n_2}{P_2^*}+\frac{\Delta n_3}{P_3}\right)\right], \tag{8}$$

$$\tilde{\chi}_1=\frac{iK}{d_1}\left[\frac{1}{P_{43}}\left(\frac{\Delta n_4}{P_4}+\frac{\Delta n_3}{P_3^*}\right)+\frac{1}{P_{32}^*}\left(\frac{\Delta n_2}{P_2}+\frac{\Delta n_3}{P_3^*}\right)\right], \tag{9}$$

$$\tilde{\chi}_3=\frac{iK}{d_3}\left[\frac{1}{P_{12}^*}\left(\frac{\Delta n_1}{P_1^*}+\frac{\Delta n_2}{P_2}\right)+\frac{1}{P_{41}}\left(\frac{\Delta n_1}{P_1^*}+\frac{\Delta n_4}{P_4}\right)\right]. \tag{10}$$

where $P_j=\Gamma_j+i\Omega_j$ are resonant denominators for the corresponding radiations (for example, $P_4=\Gamma_4+i\Omega_4$, $P_{43}=\Gamma_{43}+i(\Omega_4-\Omega_3)$, and so on); $d_2=\Gamma_2+i(\Omega_1+\Omega_3-\Omega_4)$, $d_4=\Gamma_4+i(\Omega_1-\Omega_2+\Omega_3)$, $d_1=\Gamma_1+i(\Omega_4-\Omega_3+\Omega_2)$, $d_3=\Gamma_3+i(\Omega_4-\Omega_1+\Omega_2)$; $\Delta n_4=n_1-n_4$, Γ_4 is the homogeneous width of the transition 1–4, and so on; n_j are the level populations; $K=d_{13}d_{32}d_{24}d_{41}/4\hbar^3$; and, d_{ij} are the electric-dipole transition moments (see Fig. 1).

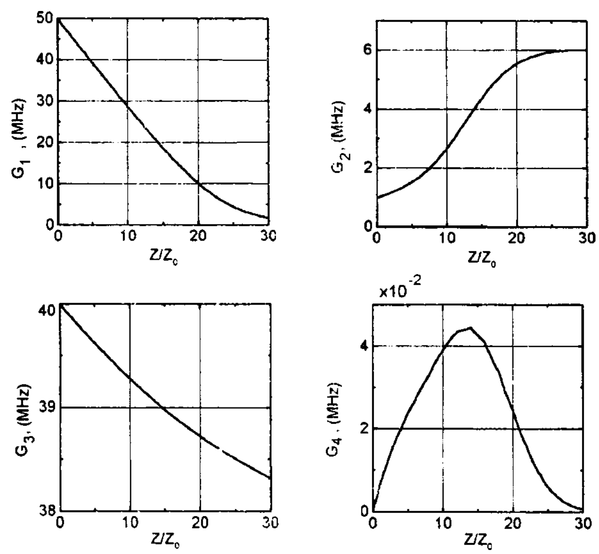

FIG. 3. Same as in Fig. 2 but with allowance for the Raman amplification at the frequency ω_2 and the radiation-perturbed absorption at all other frequencies. The parameters and notation employed are the same as in Fig. 2.

If only the lower level is populated and the departures from all resonances are much greater than the widths of the resonances, all susceptibilities are the same: $\tilde{\chi}_1=\tilde{\chi}_2=\tilde{\chi}_3=\tilde{\chi}_4=\tilde{\chi}=-Kn_1/\Omega_1\Omega_{12}\Omega_4$. The imaginary parts of the susceptibilities (including absorption) can be neglected in comparison with the real parts. We shall also assume phase matching $\Delta k=0$. Then the equation for the phase becomes

$$d\Theta/dz=\tilde{\sigma}A_1A_2A_3\cos\Theta/A_4\,.$$

Hence it follows that the phase $\Theta=\pi/2$ is stable, and according to Eqs. (1)–(6) the waves E_1 and E_3 become weaker, while E_2 and E_4 grow. The number of photons $\hbar\omega_1$ and $\hbar\omega_3$ which have vanished is equal to the number of photons $\hbar\omega_2$ and $\hbar\omega_4$ which are generated, and the numbers of each are also equal. At first glance the same thing should happen for the parametric part of the interaction in the resonant case also. However, in this case the susceptibilities become purely imaginary, and their magnitudes and signs differ and their dependences on the radiation intensities are different.[4]

Inhomogeneous broadening due to the variance of the Doppler frequency shifts of individual molecules can also have a large effect on the resonant nonlinear-optical interactions, leading to qualitative effects. In experiments appreciable conversion is ordinarily obtained through the use of optically thick media in which the radiation intensities vary along the medium. For this reason, using the analytical expressions in Ref. 4 to solve the problem posed, we shall illustrate the main results for the resonant case by numerical experiments using an interactive computational program which we developed for this purpose. The perturbation of the medium by the radiation, the Doppler broadening, and effects due to the propagation of the initial and generated radiations in an optically thick medium are taken into account. In accordance with the experiment, the model employed assumes that each level is perturbed by only a single strong field, i.e., the fields E_1 and E_3 can be arbitrarily strong while all other fields are weak.[4]

The results of the numerical simulation of completely resonant conversion in a Doppler-broadened medium, neglecting the attendant absorption processes, are presented in Fig. 2, and the analogous results obtained with these processes taken into account are presented in Fig. 3. As follows from Fig. 2, the relations obtained on the basis of this approach are in qualitative disagreement with the Manley–Rowe relations (the number of photons $\hbar\omega_1$ increases, while the number of photons $\hbar\omega_2$ decreases). Conversely, the curves in Fig. 3, which were obtained with allowance for the attendant multiphoton absorption and Raman amplification processes, completely agree with the notions concerning the conversion of radiation in absorbing (amplifying) media.

In summary, the main result of this work is that under resonant conditions, in contrast to nonresonant conditions, parametric conversion and absorption of photons cannot be treated independently. This result is also confirmed by a direct analysis of the expressions obtained in Ref. 4, the general forms of which are excessively complicated. Some preliminary results of this work have been presented in Ref. 5.

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