Two- and three-photon resonant frequency mixing and the interference of quantum transitions

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A theoretical study is done of the role of quantum interference in dynamic level splitting during frequency mixing in conditions of two- and three-photon resonances when the field interacting with the two-photon transition is strong. It is demonstrated that near the unperturbed three-photon resonance the interference of quantum transitions may lead to a considerable decrease in the absorption of the lased radiation (induced transparency sets in), with a simultaneous increase in the nonlinear atomic polarization. These effects enable raising the transformation efficiency by several orders of magnitude in comparison to the case of weak fields. Effective mixing is found to be possible in these conditions for intensities of the strong field exceeding the intensity of saturation of the population difference on the two-photon transition.

1. INTRODUCTION

The nonlinear susceptibility of matter, which is responsible for frequency-mixing processes, increases sharply at one-photon and multiphoton resonances.[1] This makes it possible to lower the intensity of pumping radiation to levels characteristic of continuous-wave lasers.[2] However, in the presence of one-photon and three-photon resonances on the transitions from the ground state to the excited states the absorption of the pumping and output radiations grows sharply and phase-matching problems occur. This leads to a limit in the concentration of atoms of the nonlinear medium and, as a result, to a lowering of the transformation efficiency. With a two-photon resonance, two-photon absorption is usually much lower than the absorption accompanying a one- or three-photon resonance. In this case the nonlinear susceptibility can be considerably increased without loss of optical power. But at high enough power, two-photon absorption becomes important (the saturation effect) and limits the conversion efficiency.[1,3] Under the conditions of a two-photon resonance, interference of quantum transitions may strongly affect the process of nonlinear optical frequency mixing. For instance, under certain conditions the interference of two-photon and Raman transitions suppresses two-photon absorption and, as a result, stops the parametric frequency conversion.[4,5] The effect is often called parametric bleaching,[6] and has been observed in experiments.[7–9]

Recently experiments have demonstrated the possibility of transforming an opaque (optically thick) medium into a transparent medium when three-level atoms interact in a resonant manner with two laser fields, one of which is a strong field (electromagnetically induced transparency, or EMIT).[10] This effect was studied theoretically in the 1970s (see, e.g., Refs. 11 and 12). Its physical nature lies in the dynamic splitting of levels in the strong field and the interference of quantum transitions in the absorption of the weak field. On the basis of this phenomenon, Harris, Field, and Imamoglu[13] suggested a method for dramatically increasing the conversion efficiency in four-wave mixing in conditions of a three-photon resonance, when one of the fields being mixed is strong and resonantly couples a metastable (two-photon excited) state and a higher-lying state. The results of experimental realization of this method are given in Refs. 14 and 15. Another possibility for inducing transparency at the frequency of the resulting radiation is to use an additional strong field that is in resonance with the transition between the upper level and a level to which the transition from the ground level is dipole-forbidden (its energy may be either lower or higher than that of the upper level).[16]

There have proved to be other variants of frequency mixing that use the EMIT phenomenon. The aim of the present investigation is to analyze the role of quantum interference in dynamic level splitting when frequency mixing occurs in the presence of two- and three-photon resonances, with the field resonantly interacting with the two-photon transition being strong. This conversion scheme has been studied by many researchers without allowing for quantum coherence effects (see, e.g., Refs. 1 and 3 and the references therein). It will be shown that in strong fields, allowing for the interference of quantum transitions leads to a considerable alteration of nonlinear susceptibility and absorption of the resulting radiation. Quantum interference is manifested differently in absorption and in nonlinear susceptibility. As a result there is the possibility of selecting conditions in which the decrease in absorption of the resulting radiation is accompanied by a dramatic increase in atomic nonlinear susceptibility. The refractive index at the frequency of the resulting radiation also decreases, which is important for phase matching. These effects make it possible to increase the conversion efficiency dramatically by raising the intensity of the strong field, which can considerably exceed the saturating intensity for the two-photon transition.

2. BASIC EQUATIONS

Two plane-polarized monochromatic waves with amplitudes E_1 and E_2 and frequencies ω_1 and ω_2 propagate along

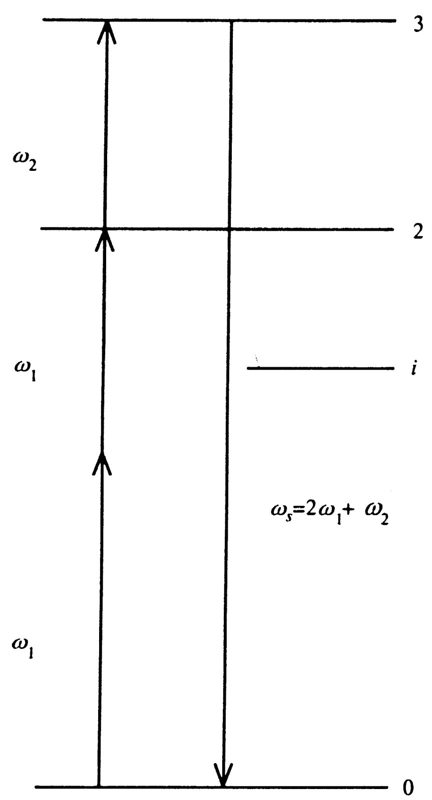

FIG. 1. The energy level diagram for four-wave frequency mixing with two- and three-photon resonances: 0 is the ground state; i, 2, and 3 are the excited states; ω_s is the frequency of the generated radiation; and ω_1 and ω_2 are the frequencies of the mixed strong and weak radiations.

the z axis and interact with each other. A third wave is produced at the sum sum frequency $\omega_s = 2\omega_1 + \omega_2$ via a nonlinear interaction. The energy level diagram is shown in Fig. 1. The $|2\rangle - |0\rangle$ transition is dipole-forbidden. The strong coherent optical radiation couples the levels $|0\rangle$ and $|2\rangle$ in a two-photon resonant manner, but its frequency is not in resonance with the $|i\rangle - |0\rangle$ transition. The fields E_2 and E_s, with the latter generated at the frequency ω_s, are assumed weak (they do not change the population of the levels) and resonantly interact with the $|3\rangle - |2\rangle$ and $|3\rangle - |0\rangle$ transitions, respectively. Initially only the lower (ground) level is assumed to be populated.

At the frequency ω_s of the generated wave, the absorption and refractive index are determined, respectively, by the imaginary and real parts of the atomic susceptibility $\chi_s(-\omega_s;\omega_s)$, and the nonlinear polarization, responsible for generating radiation at the sum frequency, is determined by the atomic third-order nonlinear susceptibility $\chi^{(3)} \times(-\omega_s;\omega_1,\omega_1,\omega_3)$. In a strong field these susceptibilities are functions of the field's amplitude and can be calculated by solving the system of equations for the density matrix exactly in the strong field, and in the first approximation in the weak fields. In the steady-state case for plane waves, the equations for the density matrix take the form

$$\Delta_{30}\rho_{30} = -i[G_{30}\rho_0 + G_{32}\rho_{20} - \rho_{32}V_{20}], \tag{1}$$

$$\Delta_{32}\rho_{32} = -i[G_{32}\rho_2 + G_{30}\rho_{02} - \rho_{30}V_{02}], \tag{2}$$

$$\Delta_{20}\rho_{20} = -iV_{20}(\rho_0 - \rho_2), \tag{3}$$

$$\Gamma_2\rho_2 = -i(V_{20}\rho_{02} - \rho_{20}V_{02}), \quad \rho_1 + \rho_2 = 1, \tag{4}$$

where ρ_{ij} $(i \neq j)$and ρ_i and the off-diagonal and diagonal elements of the density matrix; $\Delta_{30} = -i\Omega_s + \Gamma_{30}$; $\Delta_{32} = -i\Omega_2 + \Gamma_{32}$; and $\Delta_{20} = -i\Omega_{20} + \Gamma_{20}$; $\Omega_s = \omega_s - \omega_{30}$, $\Omega_2 = \omega_2 - \omega_{32}$, and $\Omega_{20} = 2\omega_1 - \omega_{20}$ are the frequency offsets; ω_{ij} and Γ_{ij} are the frequencies and halfwidths of the respective transition lines; Γ_2 is the population relaxation rate of level $|2\rangle$; $G_{30} = -d_{30}E_s/2\hbar$ and $G_{32} = -d_{32}E_2/2\hbar$ are the matrix elements of the interaction with the fields E_s and E_2, $V_{20} = (4\hbar)^{-2}\Sigma_i d_{2i}d_{i0}E_1^2/(\omega_1 - \omega_{i0})$ is the composite effective matrix element of the two-photon transition $|2\rangle - |0\rangle$ (see, e.g., Ref. 3); $V_{20} = V_{02}^*$ (an asterisk stands for complex conjugation); and the d_{ij} are the dipole moments of the transitions.

To allow for level splitting in a strong field and the coherence of the quantum transitions, the system of equations (1)–(4) was first solved exactly in the strong field, assuming that both E_2 and E_s are zero. The solution was then used as the zeroth approximation for solving Eqs. (1) and (2) simultaneously to first order in the weak fields E_2 and E_s.

The solution for ρ_{30} has the form

$$\rho_{30} = -iG_{30}\frac{\Delta_{32}\rho_0 + i\rho_{02}V_{20}}{\Delta_{30}\Delta_{32} + |V_{20}|^2} - i\frac{G_{32}\rho_{20} + iG_{32}V_{20}\rho_2}{\Delta_{30}\Delta_{32} + |V_{20}|^2}, \tag{5}$$

$$\rho_{20} = -iV_{20}(\rho_0 - \rho_2)/\Delta_{20}, \quad \rho_2 = 0.5\kappa/[\Omega_{20}^2 + \Gamma_{20}(\kappa + 1),$$

$$\rho_0 = 1 - \rho_2, \tag{6}$$

where $\kappa = 4|V_{20}|^2/\Gamma_{20}\Gamma_2$ is the saturation parameter.

The first term on the right-hand side of Eq. (5) describes the ordinary linear atomic polarization modified by the strong field. The second term in the numerator of that expression is due to a nonlinear interference effect (in the terminology of Ref. 12). The second term on the right-hand side of Eq. (5), the nonlinear (cubic) atomic polarization also modified by the strong field, consists of two parts: one is proportional to the off-diagonal element ρ_{20} of the density matrix (this term is always present in perturbation theory), and the other is proportional to the population ρ_2. The atomic polarization is established by the standard method.[1,3] As a result, the linear and cubic susceptibilities $\chi(\omega_s)$ and $\chi^{(3)}(\omega_s)$ can be written as

$$\chi(\omega_s) = -i\chi_0\Gamma_{30}\frac{\Delta_{32}\rho_0 - |V_{20}|^2\Delta_{20}^{-1}(\rho_0 - \rho_2)}{\Delta_{30}\Delta_{32} + |V_{20}|^2}, \tag{7}$$

$$\chi^{(3)}(\omega_s) = -\chi_0^{(3)}\Gamma_{30}\Gamma_{20}\frac{(\rho_0 - \rho_2)\Delta_{32}\Delta_{20}^{-1} - \rho_2}{\Delta_{30}\Delta_{32} + |V_{20}|^2}, \tag{8}$$

where χ_0 and $\chi_0^{(3)}$ are the respective resonant susceptibilities $(\Omega_{20} = \Omega_s = 0)$ as $V_{20} \to 0$. Equations (7) and (8) assume homogeneous broadening of the resonant transitions.

For purposes of analysis, we write Eqs. (7) and (8) as

$$\chi(\omega_s) = \chi_1 + \chi_2, \quad \chi^{(3)}(\omega_s) = \chi_1^{(3)} + \chi_2^{(3)},$$

where

$$\chi_1 = -i\chi_0\Gamma_{30}\Delta_{32}\rho_0 d^{-1},$$

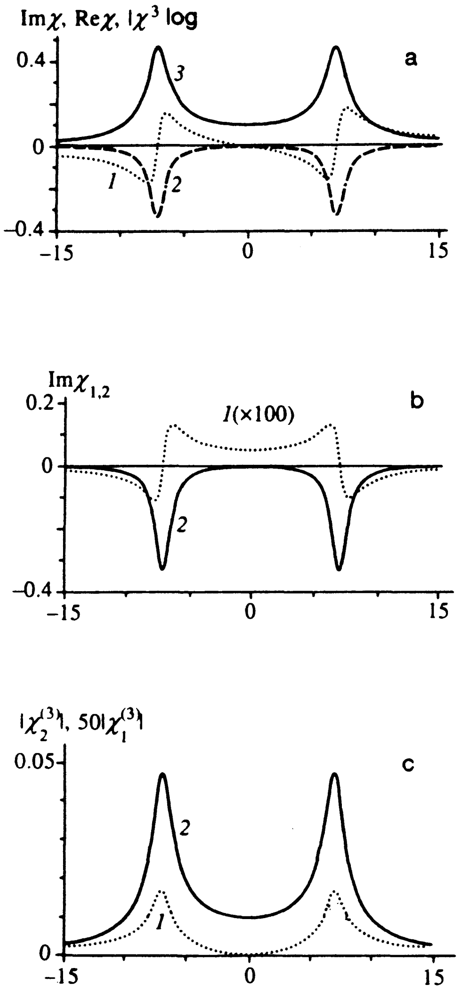

FIG. 2. Normalized curves illustrating the dependence on the normalized offset from the three-photon resonance, $x_s=(\omega_s-\omega_{30})/\Gamma_{30}$, at $g=10$, $\Gamma_{20}/\Gamma_{32}=2$, $\Gamma_{32}/\Gamma_2=5$, and $\Gamma_{20}/\Gamma_{30}=1$ of (a) dispersion Re χ (curve *1*), absorption Imχ (curve *2*), and nonlinear atomic polarization $P\propto\chi^{(3)}g$ (curve *3*); (b) Imχ_2 (curve *1*) and Imχ_1 (curve *2*); and (c) $|\chi_1^{(3)}|$ (curve *1*) and $|\chi_2^{(3)}|$ (curve *2*).

$$\chi_2=i\chi_0\Gamma_{30}|V_{20}|^2\Delta_{20}(\rho_0-\rho_2)d^{-1}, \tag{9}$$

$$\chi_1^{(3)}=-\chi_0^{(3)}\Gamma_{30}\Gamma_{20}(\rho_0-\rho_2)\Delta_{32}\Delta_{20}^{-1}d^{-1}, \tag{10}$$

$$\chi_2^{(3)}=\chi_0^{(3)}\Gamma_{30}\Gamma_{20}\rho_2 d^{-1}, \quad d=\Delta_{30}\Delta_{32}+|V_{20}|^2. \tag{11}$$

3. ANALYSIS OF RESULTS

The dependence of the absorption (Imχ), dispersion (Reχ), and nonlinear atomic polarization ($P\propto\chi^{(3)}g$), as well as Im$\chi_{1,2}$ and $|\chi_{1,2}^{(3)}|$, on ω_s is shown in Fig. 2. For $g=|V_{20}|/(\Gamma_{30}\Gamma_{32})^{1/2}\gg 1$, the contribution of nonlinear interference effects to absorption is small (Fig. 2b), and the cubic susceptibility $\chi^{(3)}$ is determined by the second term in (8) proportional to the population ρ_2 (Fig. 2c), since ρ_{20} is small due to the saturation of populations on the $|0\rangle-|2\rangle$ transition. Note that near an unperturbed resonance Reχ tends to zero, which, on the one hand, facilitates phase matching at high atomic concentrations and, on the other, makes it possible to control the sign of the dispersion (this is important for signal generation under focusing conditions). Curves *1* and *2* in Fig. (2a) Im χ and $|\chi^{(3)}|$ experience a dip at the center, and each dip becomes deeper as V_{20} grows. The effect is due to the dynamic splitting of the frequency of the $|3\rangle-|0\rangle$ transition in the strong two-photon resonant field E_1. The interference of quantum transitions plays an important role here and shows updifferently in absorption nonlinear susceptibility. This can be demonstrated by expanding the denominator in Eqs. (7) and (8), which is quadratic in Ω_s, in prime factors. As a result, Eqs. (7) and (8) can be conveniently written in the following form:

$$\chi(\omega_s)=-i\chi_0\,\frac{\Gamma_{30}[\Delta_{32}\rho_0-|V_{20}|^2\Delta_{20}^{-1}(\rho_0-\rho_2)]}{z_1-z_2}\times\left\{\frac{1}{\Omega_s-z_1}+\frac{(-1)}{\Omega_s-z_2}\right\}, \tag{12}$$

$$\chi^{(3)}(\omega_s)=-\chi_0^{(3)}\,\frac{\Gamma_{20}\Gamma_{30}[(\rho_0-\rho_2)\Delta_{32}\Delta_{20}^{-1}-\rho_2)]}{z_1-z_2}\times\left\{\frac{1}{\Omega_s-z_1}+\frac{(-1)}{\Omega_s-z_2}\right\}, \tag{13}$$

where $z_{1,2}=z'_{1,2}+iz''_{1,2}$ are the roots of the denominator (quasilevels or quasienergies[5]),

$$z_{1,2}=-0.5i\{(i\Omega_{20}+\Gamma_{30}+\Gamma_{32})\mp[(i\Omega_{20}+\Gamma_{30}-\Gamma_{32})^2-4|V_{20}|^2]^{1/2}\},$$

$$z'_{1,2}=\mathrm{Re}\,z_{1,2},\quad z''_{1,2}=\mathrm{Im}\,z_{1,2}.$$

The quantities $z'_{1,2}$ and $z''_{1,2}$ determine the position and half-widths of the new resonances (quasilevels). The presence of two terms within the braces in Eqs. (12) and (13) reflects the effects of level splitting in a strong two-photon resonant field and of quantum interference.

We now write explicitly the interference contributions in Imχ, Reχ, and $|\chi^{(3)}|$:

$$\mathrm{Im}\,\chi(\omega_s)=\chi_0\Gamma_{30}\,\mathrm{Im}\{-i[\Delta_{32}\rho_2-|V_{20}|^2\Delta_{20}^{-1}(\rho_0-\rho_2)]\times(\Omega_s-z_1^*)(\Omega_s-z_2^*)\}D, \tag{14}$$

$$\mathrm{Re}\,\chi(\omega_s)=\chi_0\Gamma_{30}\,\mathrm{Re}\{-i[\Delta_{32}\rho_2-|V_{20}|^2\Delta_{20}^{-1}(\rho_0-\rho_2)]\times(\Omega_s-z_1^*)(\Omega_s-z_2^*)\}D, \tag{15}$$

$$|\chi^{(3)}(\omega_s)|=\chi_0^{(3)}\Gamma_{30}\Gamma_{20}|[(\rho_0-\rho_2)\Delta_{32}\Delta_{20}^{-1}-\rho_2](\Omega_s-z_1^*)(\Omega_s-z_2^*)|D, \tag{16}$$

$$D=\left\{\frac{1}{|\Omega_s-z_1|^2}+\frac{1}{|\Omega_s-z_2|^2}-2\,\mathrm{Re}\left(\frac{1}{(\Omega_s-z_1)(\Omega_s-z_2^*)}\right)\right\}|z_1-z_2|^{-2}. \tag{17}$$

The third terms in braces in Eqs. (14)–(16) describe the interference of quantum transitions through the quasilevels that emerged as a result of dynamic level splitting. The minus sign preceding these terms signifies that the interference is constructive when $\Omega_s-z'_1$ and $\Omega_s-z'_2$ have opposite signs (i.e., when the frequency lies region between the quasi-

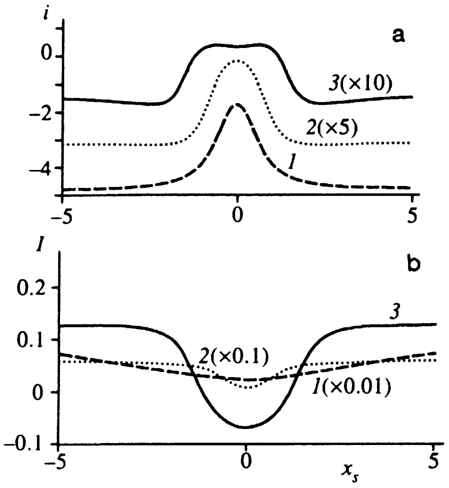

FIG. 3. The dependence of the interference terms i and I on the normalized offset from the three-photon resonance, $x_s = (\omega_s - \omega_{30})/\Gamma_{30}$, for various magnitudes of the strong field $g=|V_{20}|/(\Gamma_{30}\Gamma_{32})^{1/2}$ at $g=0.5$ (curve 1), $g=1$ (curve 2), and $g=2$ (curve 3). The other parameters are the same as in Fig. 2.

levels). We have introduced the notation $\mathrm{Im}\,\chi=s-i$ and $|\chi^{(3)}|=S-I$, where s and S are the sums of the first two terms (without allowing for interference) in Eqs. (14) and (16), respectively, and i and I are the interference terms (the third terms). Figure 3 shows the ω_s-curves for i and I at different values of the strong-field amplitude. Clearly, the interference can be either destructive or constructive, depending on the frequency and the strong-field amplitude. Figure 4 shows similar curves for s, i, $\mathrm{Im}\,\chi$, S, I, and $|\chi^{(3)}|$ for a given value of the strong-field amplitude.

The curves reveal that for $g>1$, the relative contribution of the interference part to the absorption is smaller than to the nonlinear susceptibility. As a result the dynamic level

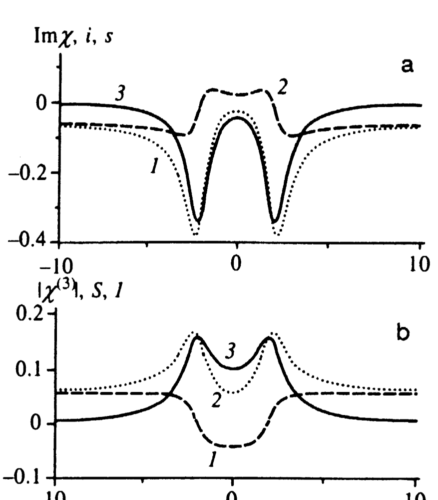

FIG. 4. Curves illustrating the dependence on the normalized detuning from the three-photon resonance, $x_s=(\omega_s-\omega_{30})/\Gamma_{30}$, at $g=|V_{20}|/(\Gamma_{30}\Gamma_{32})^{1/2}=3$ of (a) s (curve 1), i (curve 2), and $\mathrm{Im}\,\chi$ (curve 3); and (b) I (curve 1), S (curve 2), and $|\chi^{(3)}|$ (curve 3). The other parameters are the same as in Fig. 2.

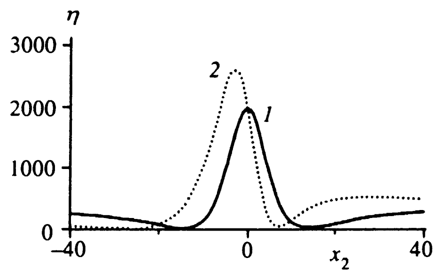

FIG. 5. η as a function of the normalized offset from the three-photon resonance, $x_2=(\omega_2-\omega_{32})/\Gamma_{32}$, at $\Omega_{20}=0$ (curve 1) and $\Omega_{20}=5$ (curve 2) with $g=|V_{20}|/(\Gamma_{30}\Gamma_{32})=10$; $\Gamma_{20}/\Gamma_{32}=4$; $\Gamma_{32}/\Gamma_{20}=5$; $\Gamma_{20}/\Gamma_{30}=2$.

splitting in a strong field changes the absorption to a much greater extent than it does the nonlinear polarization. As the strong-field amplitude grows, the contribution of the interference part diminishes, and this effect is more pronounced in the absorption. Here near an unperturbed resonance the nonlinear (cubic) atomic polarization remains fairly large (much larger than in a weak field). For a finite offset from the two-photon resonance, the symmetry of the peaks breaks down (one peak becomes larger than the other), and the zero in Re χ gets shifted. The direction of this shift is determined by the sign of the offset from the two-photon resonance.

When the generated radiation is heavily absorbed, the conversionefficiency becomes independent of the atomic concentration and is determined by the factor $\eta=(|\chi^{(3)}|g/\mathrm{Im}\,\chi)^2$ (see Ref. 3). Figure 5 shows the ω_s-dependence of η at $\Omega_{20}=0$ and $\Omega_{20}=5$, which illustrate the possibility of conversion efficiency being considerably enhanced under conditions of induced transparency. Such an enhancement results from absorption of the generated radiation. Estimates show that observing these effects requires a strong field whose intensity of of order 10^8-10^9 W cm^{-2}.

In the event of Doppler broadening of the resonant transitions, averaging over velocities becomes obligatory. When $|V_{20}|$ exceeds the Doppler width, the effects just discussed are still present.

4. CONCLUSION

We have studied the role of quantum interference in the dynamic level splitting in the process of nonlinear frequency summing when two- and three-photon resonances are present and when the radiation coupling the two-photon transition is strong. We see that near an unperturbed three-photon resonance, considerable suppression of absorption of the generated radiation becomes possible (induced transparency is said to set in), with a simultaneous increase in the atomic nonlinear polarization and a decrease in the refractive index at the frequency of the generated radiation. Under such conditions, the conversion efficiency increases by several orders of magnitude beyond that in weak fields. We also see that under specified conditions, effective mixing can occur at strong-field intensities exceeding the saturation intensity of the two-photon transition. The results can easily be generalized to incorporate the case of linear mixing with frequency subtraction.

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