Many-photon parametric resonant frequency up-conversion in a nonmonochromatic pump field

N. A. Iskanderov, V. A. Kudryashov, I. N. Matveev, and N. D. Ustinov

(Submitted 15 April 1981)

Pis'ma Zh. Eksp. Teor. Fiz. **33**, No. 10, 542–545 (20 May 1981)

Photoinduced relaxation in a nonmonochromatic pump field should lead to the decay of the coherent response of the medium and to an increase in the saturation intensity, thereby strongly affecting the conversion efficiency.

PACS numbers: 42.65.Cq, 42.65.Gv

1. The behavior of the higher-order optical nonlinearities under resonant conditions is attracting interest because of its importance to spectroscopy and broad fields of application.

In this paper we use the model of a two-level medium to derive a theory for the frequency up-conversion of a signal through the use of a nonlinear susceptibility of arbitrary order at a many-photon resonance of a given nonmonochromatic pump field. The dynamics of the populations, the time-varying response of the medium, the Stark shift of the levels, and the width of the field frequency spectrum are all taken into account.

This approach is mandated by the trend toward frequency conversion through the use of nonlinear susceptibilities of progressively higher order in the field of a randomly modulated pump.[2–5]

 0021-3640/81/10525-04$00.60 © 1981 American Institute of Physics

2. The self-consistent semiclassical system of equations for a two-level medium[6] has been simplified to derive equations for the population difference η, the off-diagonal density matrix element σ_{12}, and the complex amplitude of the converted light, A_l, which arises in the parametric conversion of the signal field A_{l-1} by virtue of the generalized susceptibility of order $l-1$:

$$\frac{\partial \eta}{\partial t}+\frac{\eta-\eta_o}{T_1}=-4\,\mathrm{Im}\,[\sigma_{12}\,(r_k\prod_{i=1}^{k}A_i^*+q_{lk}\hbar^{-1}A_l\,A_{l-1}^*\ldots A_{l-k}^*)],$$

$$\frac{\partial \sigma_{12}}{\partial t}+\left(\frac{1}{T_2}+i\delta\right)\sigma_{12}=i\eta\,(r_k\prod_{i=1}^{k}A_i+q_{lk}\hbar^{-1}A_l\,A_{l-1}^*\ldots A_{l-k}^*),$$

$$\frac{\partial A_l}{\partial z}+\frac{1}{u}\frac{\partial A_l}{\partial t}=i\gamma_l\,q_{lk}\sigma_{12}\prod_{i=k+1}^{i=l-1}A_i\,. \tag{1}$$

Here A_i is the complex amplitude of the pump field ($i\neq l-1$); $T_{1,2}$ are relaxation time; r_k is the composite matrix element of the k-photon transition: q_{lk} is the composite matrix element of the Raman-like transition; the frequency difference from resonance is $\delta=\omega_{12}-\Sigma_{i=1}^{k}\omega_i+\Omega$, $\Omega=\Sigma_{i=1}^{l}p_i|A_i|^2$ is the Stark shift; $\gamma=2\pi\omega_l N/\eta c$; ω_l is the frequency of the converted light; N is the concentration of particles; η_l is the linear part of the refractive index at the frequency ω_l; and u is the wave velocity.

These equations are the starting point for an analysis of many-photon frequency up-conversion of a signal for the case in which the pump field is not monochromatic and the conversion is a time-varying process.

3. Using the procedure of Ref. 7 for taking the average of stochastic differential equations, we find from (1) some equations describing the time evolution of the average conversion efficiency, $\langle\kappa\rangle=\langle\Pi_{i=k+1}^{i=l-2}A_iA_i^*\sigma_{12}\sigma_{21}\rangle$, for the case in which the pump field is applied instantaneously with a diffusing phase. The corresponding time evolution is shown in Fig. 1, from which it is clear that the broadening of the frequency spectrum of the pump field causes a qualitative change in the conversion.

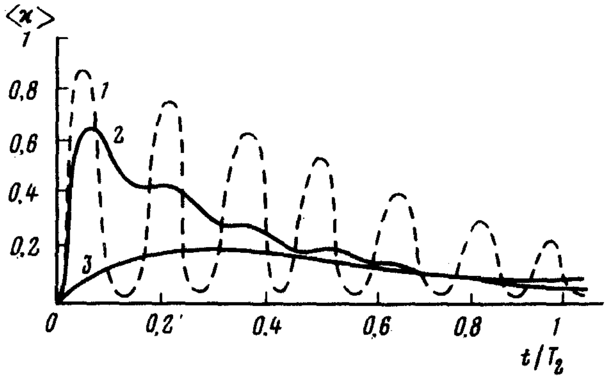

FIG. 1. Time evolution of the average conversion efficiency for various widths of the field frequency spectrum, D_i • 1–$D_1=0$; 2–$D_2>D_1$; 3–$D_3>D_2$.

While the resonant response in a monochromatic field is coherent (curve 1), the photoinduced relaxation in a nonmonochromatic field leads to a decay of the coherent interaction and to a transition to incoherent conversion (curves 2 and 3). The threshold pump intensity at which nutation of the average conversion efficiency arises can be estimated from $I_{\text{nut}} \sim \sqrt[k]{k^4 D^2 T_1 T_2}$, where k is the order of the resonance, and D is the half-width of the field frequency spectrum, which has a Lorentzian shape.

4. In steady-state conversion for a frequency-degenerate pump field with $\delta = 0$, the average normalized conversion efficiency is

$$\langle \kappa_k \rangle = \frac{\alpha_2 I^{l-2}}{\alpha_3 (1 + \alpha_2 I^k/\alpha_3)(1 + \beta I^k/\alpha_4)}, \tag{2}$$

where I is the pump intensity normalized to the saturation intensity $I_s = (1/4T_1 T_2 r_k^2) = (1/4T_1 T_2 r_k^2)^{1/k}$, $\alpha_{1,2} = T_{1,2}^{-1}$, $\alpha_3 = \alpha_2 + k^2 D$, $\alpha_4 = \alpha_1 + \alpha_2 + k^2 D$, $\alpha_5 = \alpha_2 + 2k^2 D$, and $\beta = \alpha_2 + \alpha_1/2 + \alpha_1\alpha_2/2\alpha_5$..

It follows from (2) that under the condition $l - 2 > k$ the maximum average conversion efficiency in a nonmonochromatic field exceeds the maximum conversion efficiency in a monochromatic field,

$$\frac{\langle \kappa_k^{max} \rangle}{\kappa_k^{max}(D=0)} \sim \sqrt[k]{[(l-2-k)\alpha_3/\alpha_2]^{l-2-k}} \quad (k < l-2 < 2k)$$

$$\frac{\langle \kappa_k^{max} \rangle}{\kappa_k^{max}(D=0)} = \frac{\alpha_1 + \alpha_2 + k^2 D}{\alpha_2 + \alpha_1/2 + \alpha_1\alpha_2/2\alpha_5} \quad (l - 2 \geqslant 2k), \tag{3}$$

but this maximum efficiency is reached at a higher pump intensity,

$$I_{\text{opt}} \sim I_{\text{opt}}(D=0)\sqrt[k]{\alpha_3/\alpha_2}. \tag{4}$$

5. In summary, in parametric frequency conversion by virtue of a nonlinear susceptibility of arbitrary order, accompanied by the excitation of a many-photon resonance by a nonmonochromatic pump field, the dynamic gain in the average conversion efficiency due to photoinduced relaxation can be expected to decrease significantly or vanish altogether in the field of picosecond or nanosecond pump pulses. In the steady state, the average conversion efficiency can reach a much higher value than in a monochromatic field, because of an increase in the saturation intensity in a nonmonochromatic pump field.

1. S. A. Akhmanov, Opticheskie nelineĭnosti vysshikh poryadkov, Kn. Nelineĭnaya spektroskopiya ("Higher-order optical nonlinearities,") in: Nonlinear Spectroscopy (Russ. transl. Mir, M., 1979, p. 323).
2. K. N. Drabovich, A. I. Kovrigin, S. M. Pershin, N. M. Sinyavskiĭ, and A. L. Surovegin, Pis'ma Zh. Eksp. Teor. Fiz. 32, 72 (1980) [JETP Lett. 32, 162 (1980)].

3. K. N. Drabovich, M. Igntavichyus, R. Kupris, A. Matsulyavichus, S. M. Pershin, N. M. Sinyavskiĭ, V. Smil'gyavichus, and A. L. Surovegin, Tezisy dokladov X Vsesoyuznoĭ konferentsii po kogerentnoĭ i nelineinoĭ optike (Abstracts of Reports, Tenth All-Union Conference on Coherent and Nonlinear Optics), Part 1, Moscow, 1980, p. 327.
4. Yu. E. D'yakov, N. A. Iskanderov, and V. A. Nekhaenko, in: Nelineĭnoe rezonansnoe preobrazovanie chastoty lazernogo izlucheniya (Nonlinear Resonant Frequency Conversion of Laser Beams), Izd. FAN, Tashkent, 1979, p. 49.
5. N. A. Iskanderov, V. A. Kudryashov, I. N. Matveev, and N. D. Ustinov, Tezisy dokladov X Vsesyuznoĭ konferentsii po kogerentnoĭ i nelineinoĭ optike (Abstracts of Reports, Tenth All-Union Conference on Coherentand Nonlinear Optics), Part 2, Moscow, 1980, p. 210.
6. V. S. Butylkin, Yu. G. Khronopulo, and E. I. Yakubovich, Zh. Eksp. Teor. Fiz. **71**, 1712 (1976) [Sov. Phys. JETP **44**, 897 (1976)].
7. Yu. E. D'yakov, Kratkie Soobshcheniya po Fizike, FIAN **7**, 49 (1971).

Translated by Dave Parsons
Edited by S. J. Amoretty