Harmonic generation without phase matching

É. M. Belenov and A. V. Nazarkin
P. N. Lebedev Physics Institute, Academy of Sciences of the USSR, Moscow

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Harmonic generation in an unmatched wave interaction in a nonlinear medium is analyzed. The conversion into the harmonic may be highly efficient when impurities which resonantly retard the ultrashort pump pulse are added to the host nonlinear medium.

Efficient harmonic generation in a nonlinear medium requires phase matching, i.e., that the refractive indices of the medium be the same at the frequency of the pump and at the frequency of the field that is generated.[1] In crystalline media this condition is usually satisfied by means of birefringence, but in several nonlinear media the birefringence is either absent or inadequate in the frequency range of interest.[2] If matching is not achieved, energy is transferred from the fundamental wave at the frequency ω into the harmonic $n\omega$ over the matching distance $L_c = \pi/\Delta k$, where Δk is the distance between wave vectors, and then the energy of the harmonic is returned to the pump wave.

We wish to call attention to a pertinent point. Let us assume that the group velocities of the pump pulse and the harmonic pulse differ by an amount such that at a distance L_c the pump pulse lags behind the generated harmonic pulse by a distance greater than its spatial dimension. In this case there will be no inverse conversion from the harmonic to the pump, since this conversion occurs only in the presence of the pump field.

If this situation is to be achieved, the group retardation of the pump pulse must be substantial, while the pulse itself must be quite short. We propose arranging this situation by adding to the nonlinear medium an impurity that is at resonance with the pump frequency. If the length of the pump pulse is shorter than the phase relaxation time T_2, a sufficiently intense pulse that interacts coherently with the impurities will break up into several 2π pulses, which will move through the medium in a situation of self-induced transparency at a velocity well below the velocity of light.[3]

For definiteness, we consider second-harmonic generation in a nonlinear crystal that contains impurities of this type.

We write the fields of the pump pulse and of the harmonic, both propagating along the z axis, as

$$e_1(z,t) = \frac{1}{2}\left(E_1(z,t)e^{i(\omega t - k_1 z)} + \text{c.c.}\right),$$

$$e_2(z,t) = \frac{1}{2}\left(E_2(z,t)e^{i(2\omega t - k_2 z)} + \text{c.c.}\right),$$

where E_1 and E_2 are "slow" complex amplitudes, and k_1 and k_2 are the wave vectors of the corresponding fields. The harmonic generation is described by the system of equations

$$\frac{\partial E_1}{\partial z} + \frac{1}{v_1}\frac{\partial E_1}{\partial t} = -\frac{i\omega\gamma}{cn_1} E_2 E_1^* e^{-i\Delta kz} + i\frac{4\pi\omega N\mu}{cn_1} P,$$

$$\frac{\partial E_2}{\partial z} + \frac{1}{v_2}\frac{\partial E_2}{\partial t} = -\frac{i\omega\gamma}{cn_2} E_1^2 e^{i\Delta kz},$$

$$\frac{dP}{dt} + i(\omega-\omega_{21})P = -\frac{i}{2\hbar}\mu n E_1, \tag{1}$$

$$\frac{dn}{dt} = \frac{i}{\hbar}\mu P^* E_1 + \text{c.c.}$$

Here $\Delta k = k_2 - 2k_1 = (2\omega/c)(n_1 - n_2)$, n_1, n_2 are the refractive indices of the crystal at the pump and harmonic frequencies, v_1 and v_2 are the group velocities of the pulses, which are coupled by the linear dispersion, $\gamma = 4\pi\chi^{(2)}$ is the second-order susceptibility, which is responsible for the second-harmonic generation, P is the polarization of the resonant transition in the impurity, n is the difference between the transition populations, μ is the dipole moment, ω_{21} is the transition frequency, and N is the concentration of the impurity particles.

Let us assume that a steady-state 2π pulse of length $\tau_0 \ll T_2$ is propagating through a crystal under conditions such that there is an exact resonance with a transition in the impurity ($\omega = \omega_{21}$). The envelope and velocity (v_0) of this steady-state pulse are given by[3]

$$E_1(z,t) = \frac{2\hbar}{\mu\tau_0}\,\mathrm{sech}\left(\frac{t-(z/v_0)}{\tau_0}\right), \quad \frac{1}{v_0} - \frac{1}{v_1} = \frac{2\pi\omega N\mu^2\tau_0^2}{cn_1\hbar}. \tag{2}$$

We are interested in the situation in which the distance over which the pulse is converted into the harmonic in the absence of impurities is $L_{nl} = \mu\tau_0 cn_2/2\hbar\omega\gamma \gg L_c$, i.e., a situation in which there can be no substantial energy transfer to the harmonic. We assume that the impurity concentration N is such that the velocity of the 2π pulse satisfies $v_0 \ll v_1, v_2$, so that the spatial dimension of this pulse satisfies $L_{2\pi} = v_0\tau_0 < L_c$. In the approximation of a given pump field (2), we find the following expression from the second equation of system (1):

$$E_2(z,\tau) = i\frac{\tau_0\omega\gamma}{bcn_2}\left(\frac{2\hbar}{\mu\tau_0}\right)^2\left\{\tanh\left(\frac{\tau - zb}{\tau_0}\right) - \tanh\left(\frac{\tau}{\tau_0}\right)\right\}\exp(i\Delta k\,\tau b^{-1}), \tag{3}$$

where $\tau = t - z/v_2$, and $b = 1/v_0 - 1/v_2$. It follows from (3) that the harmonic is continuously generated in the forward direction by the slowly moving pump pulse. The wave difference Δk does not prevent the transfer of energy to the harmonic; it simply causes a temporal modulation of the phase of the harmonic.

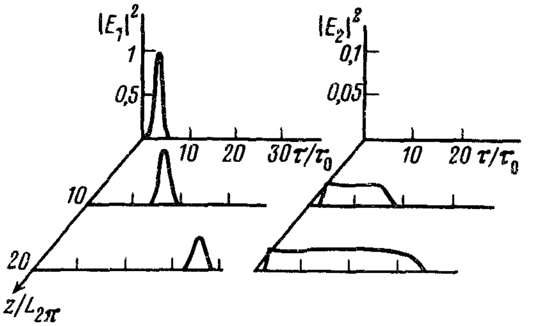

FIG. 1. Evolution of a 2π pump pulse $|E_1|^2$ and of the second-harmonic pulse $|E_2|^2$.

Equations (1) have been integrated numerically in an effort to study the evolution of the pulses, with allowance for pump depletion. We found that the transfer of energy from the pump pulse to the second harmonic can be significant. The reason is that during the generation of the harmonic the "area" under the pump pulse, $\theta_0 = \mu\hbar^{-1}\int |E_1| dt$, manages to adiabatically "track" the value of 2π, so that the energy loss due to resonant absorption in the impurity turns out to be extremely slight. This behavior of the pulse is apparently due to a general stability of solitons.[3] Figures 1 and 2 show results calculated for the values $L_{nl} = 4.5L_{2\pi}$ and $L_c = 1.5L_{2\pi}$. Over a distance $z \approx 60L_{2\pi}$, 70% of the energy of the pulse is transferred to the harmonic, whereas in the absence of impurities the transfer does not exceed 5%. Since the area under the pulse remains approximately equal to 2π, while the energy of the pulse decreases, the length of the pulse increases, and its velocity decreases (Fig. 1). Our study shows that the conversion efficiency remains high in the general case of $2\pi n$ pump pulses.

Let us look at some numerical estimates of the parameters that would be required of the pulse and of the impurity concentration. In crystalline or liquid media, pulses of

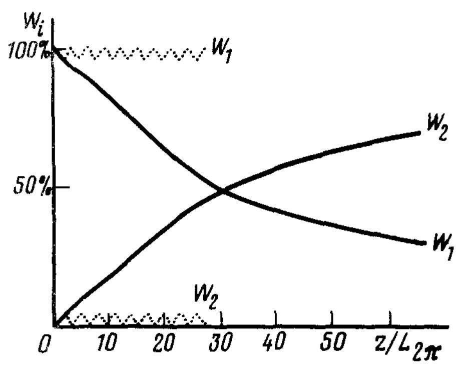

FIG. 2. Conversion of the pump pulse energy W_1 into the energy of the second harmonic, W_2. Dotted curve—Without resonant impurities; solid curve—with resonant impurities.

length $\tau_0 = 10^{-11}$ s will be coherent. With $\mu \sim 10^{-18}$ esu, the power density of the 2π pulse is $q \approx 10^8$ W/cm^2. At pump wavelengths $\lambda_1 \sim 1$ μm, the matching length characteristic of GaAs, $LiNbO_3$, and other nonlinear materials would be $L_c \gtrsim 10^{-3}$ cm. From the condition $L_{2\pi} < L_c$ we find that the impurity concentration would have to be $N \sim 10^{18}$ cm^{-3}.

Apparently the simplest way to implement this scheme for conversion in liquid and gaseous media would be to mix a nonlinear host medium with a resonant "retarder" of relatively low concentration or to coat the walls of an optical waveguide made of a nonlinear material with thin resonant films.[4] The method proposed here is also effective in the generation of higher harmonics in the general case of parametric frequency conversion.[1)]

[1)]In several cases, an analysis of frequency conversion in media with resonant impurities must incorporate the dipole field of the impurity in the near zone,[5] but this measure causes no qualitative changes in the effects discussed here, so we will defer a discussion of it to a future, more detailed paper.

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