# Modulational instability at sum and difference frequencies in media with a high-order nonlinearity

S. N. Vlasov and V. I. Talanov[1)]
*Institute of Applied Physics, Russian Academy of Sciences, 603600 Nizhniĭ Novgorod, Russia*

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As electromagnetic waves of frequency $\omega_0$ propagate through a medium with a nonlinear polarization of order $2N+1$, an instability can occur at sum and difference frequencies, $\Omega_1$ and $\Omega_2$, which satisfy the condition $2M\omega_0=\Omega_1+\Omega_2$, where $M$ is an integer $\leqslant N$. The conditions for the onset of this instability are determined. © *1994 American Institute of Physics.*

As waves propagate through nonlinear dispersive media, a modulational instability of the envelopes of the waves can occur under certain conditions.[1] In particular, electromagnetic waves of frequency $\omega_0$ in an insulator with dispersion and an inertialess cubic polarization $P=\alpha_3 E^3$, where $E$ is the electric field, are unstable with respect to perturbations at the frequencies

$$\omega_0 \pm \Omega, \quad |\Omega| \ll \omega_0. \tag{1}$$

Here $\Omega^2 \simeq \delta\epsilon_3 \omega_b^2$, $1/\omega_b^2 = -(\partial^2 k/\partial\omega^2)/k$, $\delta\epsilon_3 \simeq 12\pi\alpha_3|E|^2$, $k=\frac{\omega}{c}\sqrt{\epsilon_d}$ is the magnitude of the wave vector, $c$ is the velocity of light, and $\epsilon_d$ is the linear part of the dielectric constant if the dispersion, which we describe by means of the quantity $(\partial^2 k/\partial\omega^2)/k = -1/\omega_b^2$, and the nonlinearity coefficient $\alpha_3$ differ in sign. We show in this letter that if the polarization includes terms of higher order, $P \simeq \alpha_N E^{2N+1}$, the instability can occur at sum and difference frequencies $\Omega_1$ and $\Omega_2$ which satisfy the condition

$$2M\omega_0 = \Omega_1 + \Omega_2, \quad \text{where } M \text{ is an integer } \leqslant N. \tag{2}$$

The wave frequencies $\Omega_1$ and $\Omega_2$ may be very different from each other. The efficiency of their interaction with the pump wave depends on the dispersion and the nonlinearity of the medium. If $\Omega_1$ and $\Omega_2$ differ only slightly from the frequencies $\omega_M = M\omega_0$, i.e., if

$$\Omega_1 = M\omega_0 + \Omega_p, \quad \Omega_2 = M\omega_0 - \Omega_p, \tag{3}$$

where

$$|\Omega_p| \ll M\omega_0, \tag{4}$$

then we can use the method of slowly varying amplitudes for the analysis. In this case the conditions for the onset of the modulational instability at the sum and difference frequencies can be written in a simpler way.

We consider the propagation of a scalar wave in a nonlinear medium described by the equation

$$\partial^2 E/\partial z^2 - 1/c^2(\partial^2 E/\partial t^2 + 4\pi \partial^2 P/\partial t^2) = 0, \tag{5}$$

where the polarization of the medium is $P = \alpha_d E + \alpha_N E^{2N+1}$. In a medium of this sort, with a fairly strong dispersion, and with the direct generation of harmonics being negligible, a wave $E = U = V e^{i(\omega_0 t - hz)} + \text{c.c.}$ can propagate with an amplitude $V$ and a propagation constant $h^2 = k_d^2(1 + \delta\epsilon_N)$. The quantity $\delta\epsilon_N = 4\pi\alpha_N |V|^{2N} C_N^{2N+1}/\epsilon_d$, where $C_N^{2N+1} = (2N+1)!/[N!(N+1)!]$, describes the change in the dielectric constant at the carrier frequency, and c.c. means the complex conjugate. Below we assume that the amplitude $V$ is real.

We seek a perturbed solution of (5) in the form $E = U + u$, where we assume that the perturbation $u$ is small. We assume $u = u_M e^{i(M\omega_0 t - hz)} + \text{c.c.}$, where $u_M$ are functions which vary slowly in accordance with (4). For $u_M$ we write the parabolic equations

$$-2ih_M \partial u_M/\partial z - (k_{dM}\partial^2 k_{dM}/\partial\omega^2)\partial^2 u_M/\partial\tau^2 + (k_{dM}^2 - h_M^2)u_M + A_p u_M + B_p u_M^* = 0, \tag{6}$$

where $h_M = Mh$, $k_{dM}$ is the wave vector in the linear medium at a frequency $\omega_M$, $A_p = 4\pi\alpha_N \omega_M^2 V^{2N}(2N+1)!/(N!)^2$, $B_p = 4\pi\alpha_N\omega_M^2 V^{2M}(2N+1)!/[(N-M)! \times (N+M)!]$, $\tau = t - z/v_M$, and $v_M$ is the group velocity at the frequency $\omega_M$. We seek a solution of Eq. (6) in the form $u_M = w_m e^{i\Omega_p \tau + Hz}$. To determine their propagation constants $G = H/h_M$ we have the dispersion relation

$$4G^2 + (\Delta + \Omega^2 + A)^2 - B^2 = 0, \tag{7}$$

where $\Delta = k_{dM}^2/h_M^2 - 1$ is the detuning at the frequency $\omega_M$, $\Omega^2 = \Omega_p^2(k_{dM}\partial^2 k_{dM}/\partial\omega^2)/h_M^2$ is the square of the dimensionless frequency, $A = A_p/h_M^2 = (N+1)\delta\epsilon_N/(\epsilon_d[1+\delta\epsilon_N])$, and $B = B_p/h_M^2 = [N!(N+1)!/(N-M)! \times (N+M)!]\delta\epsilon_N/\{\epsilon_d[1+\delta\epsilon_N]\}$. If, in the detuning $\Delta = [k_{dM}^2(M\omega) - M^2 k_d^2(\omega) - M^2 k_d^2 \delta\epsilon_N]/h_M^2$, the nonlinear term $M^2 k_d^2 \delta\epsilon_N$ is substantially greater than the linear term $k_{dM}^2(M\omega) - M^2 k_d^2(\omega)$, and if the dispersion $(\partial^2 k/\partial\omega^2)/k$ and the nonlinearity $\alpha_N$ have different signs, there is an instability near the frequency $\omega_M$ with a maximum growth rate

$$H = h_M B/2 \simeq MhB/2 \tag{8}$$

at the detuning frequency

$$\Omega_p \approx (N\delta\epsilon_N \Omega_{0M})^{1/2},$$

where $\omega_{0M}$ is the value of the quantity $[(k_{dM}\partial^2 k_{dM}/\partial\omega^2)/h_M^2]^{-1/2}$ at the frequency $\omega_M$.

With increasing value of the exponent $N$, the growth rates and frequency bands increase. The number of frequencies $M$ near which the instability occurs is equal to $N$ according to the quantum interpretation of Eq. (2) concerning the generation, from $2N$ photons of frequency $\omega_0$, of photons with frequencies $2M\omega_0 + \Omega_p$ and $2M\omega_0$, respectively. The growth rates at higher frequencies may be larger than those at lower frequencies because of the factor $M$ in (8).

For an arbitrary field dependence of the polarization, in contrast with a power-law dependence, we need to expand the polarization in a power series in the field $E$ in analyzing the instability, and we need to allow for the circumstance that all terms with $N>M$ affect the instability near the frequency $\omega_M$.

Let us assume that the nonlinearity is described by two terms with nonlinearities differing in sign:

$$P=\alpha_3 E^3-\alpha_N E^{2N+1}. \tag{9}$$

Near the carrier frequency we then have

$$\delta\epsilon=3\alpha_3|E|^2-\frac{(2N+1)!}{N!(N+1)!}\alpha_N|E|^{2N}=\delta\epsilon_3-\delta\epsilon_N. \tag{10}$$

$$A=2\delta\epsilon_3-(N+1)\delta\epsilon_N,\quad B=\delta\epsilon_3-Nd\epsilon_N. \tag{11}$$

At $N\gg 1$, the function in (9) describes the rapid change in $\epsilon(|E|^2)$ which is characteristic of multiphoton processes.[2] The signs of $\delta\epsilon$ from (10) and that of the coefficient $A$ from (11) may differ because of the factor $N+1$ in the second term in the expression for $A$. Consequently, the modulational instability at the fundamental frequency may occur even under the condition $\delta\epsilon(\partial^2 k/\partial\omega^2)>0$. This could not happen in the case with a cubic nonlinearity. For the other sum and difference frequencies near $\omega_M$ with $M\geqslant 2$, the coefficients $A$ and $B$ are determined by exclusively the term with $E^{2N+1}$.

We can use the theory outlined above to interpret the ultrabroadening of spectra which has been observed during the self-focusing of short, light pulses in solids.[3-9] This broadening may result from the onset of a modulational instability at sum and difference frequencies in the case of a nonlinearity of the type in (9). If the coefficient $\alpha_N$ is sufficiently small, and the relation $N\gg 1$ holds, then the cubic nonlinearity will be predominant over a substantial range of the intensity, and we will observe a self-focusing of the beam. The sign of the dispersion in glasses is such that the modulational instability does not occur in the case of a cubic nonlinearity. For nonlinearity (9), however, the field at the focal point remains bounded with increasing intensity, the sign of the derivative of the nonlinearity with respect to $|E|^2$ changes, and a modulational instability occurs at sum and difference frequencies with the growth rate in (8) and with characteristic detunings from the frequencies $\omega_M$:

$$\Omega^2/(M\omega_0)^2\approx\delta\epsilon N/\{M\omega_0[2dn(\omega)/d\omega+\omega d^2n/d\omega^2]\}. \tag{12}$$

Here $dn(\omega)/d\omega$ and $d^2n(\omega)/d\omega^2$ are respectively the first and second derivatives of the linear part of the refractive index $n=\sqrt{\epsilon_d}$ with respect to the frequency $\omega_M$. We are ignoring the dispersion of the nonlinear coefficients under the assumption $\delta\epsilon\approx|\delta\epsilon_3|\approx|\delta\epsilon_N|\ll 1$. In transparent glasses, $dn(\omega)/d\omega$ and $d^2n/d\omega^2$ are on the order of $dn(\omega)/d\omega\approx 0.05/\omega$ and $d^2n/d\omega^2=0.01/\omega^2$ (Ref. 9). Under these conditions, the relative detuning is $\Omega^2/\omega^2\approx 16\delta\epsilon NM^2$; we thus see that for values $\delta\epsilon\approx 10^{-2}$, even at small values $N\geqslant 3$, the detuning frequencies are on the order of the carrier frequency. It follows from expression (8) for the growth rate that for a beam width $2a_c\approx 10\lambda$ in the high-intensity region near the collapse point, with a size $L\approx k_d a_c^2\approx 150\lambda$ for this region, the overall growth rate reaches values required for the growth of harmonic fields to levels at which they can be distinguished experimentally from noise.

In summary, the assumption of a nonlinearity of the type in (9), along with boundedness of the field at the focal point, leads to the conclusion that emission occurs over a substantial part of the visible range.

We note in conclusion that a study of collapse with various nonlinearity powers also requires consideration of the modulational instability which occurs near frequencies $\omega_M$ if the nonlinearity is of high power.[11]

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[1)]e-mail: TALAN@APPL.NNOV.SU

[1] V. I. Bespalov *et al.*, in: *Nonlinear Optics* [in Russian], Nauka, Novosibirsk, 1968).
[2] N. B. Delone, *Interaction of Laser Light with Matter* [in Russian] (Nauka, Moscow, 1989).
[3] N. G. Bondarenko *et al.*, JETP Lett. **12**, 85 (1970).
[4] R. R. Alfano and S. L. Shapiro, Phys. Rev. Lett. **24**, 592 (1970).
[5] A. Penzkofer and W. Kaizer, Opt. Quant. Electron. **9**, 315 (1977).
[6] V. N. Lugovoĭ and A. M. Prokhorov, Usp. Fiz. Nauk **111**, 203 (1973) [Sov. Phys. Usp. **16**, 658 (1973)].
[7] V. A. Petrishchev and V. I. Talanov, in: *Quantum Electronics*[in Russian] (Sov. Radio, Moscow, 1971), No. 6, 35.
[8] N. Blombergen, Opt. Commun. **8**, 235 (1973).
[9] N. G. Bondarenko *et al.*, in: *Quantum Electronics* [in Russian] (Naukova Dumka, Kiev, 1987), No. 33, 89.
[10] M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1964).
[11] K. Rypdal and J. J. Rusmussen, Phys. Scr. **33**, 481 (1986).

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