

# Amplification in a nonthreshold parametric x-ray ( $\gamma$ -ray) laser

V. I. Vysotskiĭ and V. I. Vorontsov

Kiev State University

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A theory of coherent parametric nonthreshold x-ray ( $\gamma$ -ray) amplification is developed. It is shown, for the first time, that exponential x-ray ( $\gamma$ -ray) amplification with a high growth rate ( $\sim 1 \text{ cm}^{-1}$ ) is possible in a medium with nonlinear susceptibility and inversion at the idler-wave frequency in the optical region. At the frequency of the amplified x-ray ( $\gamma$ -ray) signal, this process is independent of dissipative parameters of the medium, i.e., the absorption coefficient, linewidth, and so on. A theory is constructed for the nonlinear parametric three-frequency interaction in a highly dispersive nonlinear medium (with linear and nonlinear susceptibility dispersion) in the field of a spatially and temporally noncoherent pump. It is shown that three-frequency nonthreshold parametric amplification is possible in a resonant three-level optically inverted medium with electronic x-ray or nuclear Mössbauer transitions. When the Mössbauer  $\gamma$ -transitions are used, it is realistic to expect a gain of about 100 when the thickness of the nonlinear medium is about 7–8 cm and the pump is in the form of a conventional isotropic laboratory source of Mössbauer  $\gamma$ -rays with an activity of about 1 Ci.

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## 1. INTRODUCTION

The basic difficulties in developing the x-ray ( $\gamma$ -ray) laser (including the question of the active medium, the resonating cavities, and the properties of x-ray and  $\gamma$ -ray kinetics<sup>[1–3]</sup>) are associated in the first instance with the necessity of satisfying at least the threshold amplification condition in the active medium in which inversion at the frequency of the resonance transition has been produced, i.e., it is necessary to ensure that the gain exceeds the losses at the frequency of the amplified high-frequency signal. Analysis shows that, to satisfy this condition, one must use a high-frequency pump, i.e., a flux of  $\gamma$  rays or of thermal neutrons with  $N_{\text{th}} \sim 10^{23} - 10^{25} \text{ sec}^{-1} \cdot \text{cm}^{-2}$  (for shortlived isomers with  $T \lesssim 10^{-6} \text{ sec}$ ), or look for methods whereby the  $\gamma$ -ray linewidths can be reduced by six to ten orders of magnitude for reasonable values of the excitation parameters (for longlived isomers with  $T \gtrsim 10^5 \text{ sec}$ ). Analysis of isomers with lifetimes lying between these limits results in the elimination of some of the difficulties and in the appearance of other difficulties.<sup>[8]</sup> One of the theoretically possible solutions is the usual parametric amplification. However, the threshold condition

$$N_{\text{th}} > \delta_s \delta_b / k_s k_b \chi_s^{NL} \chi_b^{*NL} \hbar \omega_\gamma$$

(where  $\delta_n$ ,  $k_n$ ,  $\chi_n^{NL}$  are the absorption coefficient, wave number, and nonlinear susceptibility at the frequencies of the blank and signal waves) leads to still more stringent conditions to be satisfied by the pump even in the absence of absorption ( $\delta_p = 0$ ).

None of the methods suggested in the literature for producing the  $\gamma$  laser (amplifier) is anywhere near experimental verification because of the unrealistic conditions which they have to satisfy, and this, in turn, impedes further development of the theory of the effect. It is important to note that the high flux densities of  $\gamma$  rays or neutrons that are necessary to produce the inverted state are not directly related to the experimental detection of the amplification effect, which can be

achieved provided only the excitation parameters exceed their threshold values. The difference between the applied excitation and the threshold determines the actual growth of the signal along the amplifying medium, and may be smaller than the threshold itself by several orders of magnitude. It is clear that the above stringent requirements imposed on the threshold conditions for amplification are the result of the present state of the theory in which existing traditional schemes, and methods of quantum electronics as applied to the optical and microwave bands (the principle of the pulsed solid-state laser), are directly transferred to the new region of x rays and  $\gamma$  rays.

We now propose a new and fundamentally different method for the coherent amplification of x-ray ( $\gamma$ -ray) radiation which is distinguished by the nonthreshold character (subject to the validity of the initial classical equations) of the parameters of the high-frequency pump and the amplified signal. In principle, this should result in the amplification of x-ray ( $\gamma$ -ray) radiation under the usual laboratory conditions in which the source of the pump radiation is a conventional Mössbauer or other source of about 1 Ci, i.e., the strength of the source is lower by 13–15 orders of magnitude as compared with the strength of the sources proposed previously. In the new method, the amplification effect relies on three-frequency parametric interaction in a nonlinear medium in which inversion has been produced at the frequency of the idler wave (for example, in the optical band<sup>[9]</sup>). The problem is solved for the case of a realistic nonlinear dispersive medium in which the signal and the pump radiation are spatially and temporally noncoherent.

## 2. NONTHRESHOLD PARAMETRIC INTERACTION (AMPLIFICATION) OF PLANE MONOCHROMATIC WAVES

The standard set of equations for the three-frequency parametric interaction in a nonlinear dispersive medi-

um is as follows:

$$\begin{aligned} & [\nabla[\nabla\mathbf{E}_{1,2}(\mathbf{r},t)]] + \frac{\varepsilon_{1,2}}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E}_{1,2}(\mathbf{r},t) \\ &= \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{P}_{1,2}^{NL}(\mathbf{r},t), \\ & [\nabla[\nabla\mathbf{E}_3(\mathbf{r},t)]] + \frac{\varepsilon_3}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{E}_3(\mathbf{r},t) = 0, \end{aligned} \quad (1)$$

where  $\mathbf{P}_{1,2}^{NL}(\mathbf{r},t) \equiv \mathbf{P}_{1,2}^{NL}(\mathbf{E}_{1,2}^*(\mathbf{r},t)\mathbf{E}_3(\mathbf{r},t))$  is the nonlinear polarization vector.

Consider the nonlinear problem for a semiinfinite medium ( $z > 0$ ). To simplify our analysis, we shall omit the polarization indices which are unimportant in this context. Transforming to the equations for slowly-varying amplitudes

$$\mathbf{E}_n(\mathbf{r},t) = \mathbf{A}_n(z) \exp[i(\mathbf{k}_n' \cdot \mathbf{r} - \omega_n t) - \delta_n z]$$

and confining our attention to narrow-band parametric pumping

$$A_3(\omega_3) = A_{30} \delta(\omega_3 - \omega_{30})$$

in an isotropic medium, we obtain the following set of equations:

$$\begin{aligned} \frac{\partial A_1(z)}{\partial z} &= \frac{2\pi\omega_1^2}{ik_1 c^2 \cos \theta_1} \chi_1^{NL}(\omega_1, \omega_2 = \omega_{30} - \omega_1) \\ &\quad \times A_{30} A_2^*(z) \exp[-(i\Delta k - \delta_1 + \delta_2 + \delta_3)z], \\ \frac{\partial A_2(z)}{\partial z} &= \frac{2\pi\omega_2^2}{ik_2 c^2 \cos \theta_2} \chi_2^{NL}(\omega_1 = \omega_{30} - \omega_2, \omega_2) \\ &\quad \times A_{30} A_1^*(z) \exp[-(i\Delta k + \delta_1 - \delta_2 + \delta_3)z], \end{aligned}$$

where

$$\begin{aligned} k_n &= \omega_n \sqrt{\varepsilon_n} / c, \quad \Delta k = k_1' \cos \theta_1 + k_2' \cos \theta_2 - k_3' \cos \theta_3, \\ k_n' &= \text{Re } k_n, \quad \delta_n = \text{Im } k_n / \cos \theta_n, \quad \theta_n = \angle(\mathbf{k}_n, \mathbf{e}_z). \end{aligned}$$

The solution of the above set of equations must satisfy the usual boundary conditions:

$$A_{1,2}(z=0) = A_{(1,2)0}, \quad \left. \frac{\partial A_{1,2}}{\partial z} \right|_{z=0} = \frac{2\pi\omega_{1,2}^2}{ik_{1,2} c^2 \cos \theta_{1,2}} \chi_{1,2}^{NL} A_{(2,1)0}^* \quad (2)$$

Eliminating the amplitude  $A_2$  from the above equations, we obtain (for simplicity, we substitute  $\chi_n^{NL} \rightarrow \chi_n$ )

$$\frac{\partial^2 A_1}{\partial z^2} + (i\Delta k - \delta_1 + \delta_2 + \delta_3) \frac{\partial A_1}{\partial z} - \left( \frac{2\pi\omega_1\omega_2}{c^2} \right)^2 \frac{\chi_1 \chi_2^* |A_{30}|^2}{k_1 k_2 \cos \theta_1 \cos \theta_2} e^{-2\delta_3 z} A_1 = 0.$$

If we now substitute  $\exp(-\delta_3 z) = \tilde{z}$ , we obtain the Bessel equation<sup>[10]</sup> with the solution

$$A_1 = \exp[-1/2(i\Delta k - \delta_1 + \delta_2 + \delta_3)z] \{ C_1 J_\nu(x_0 e^{-\delta_3 z}) + C_2 J_{-\nu}(x_0 e^{-\delta_3 z}) \},$$

where

$$\begin{aligned} \nu &= -\frac{i\Delta k - \delta_1 + \delta_2 + \delta_3}{2\delta_3}, \\ x_0 &= -i \frac{2\pi\omega_1\omega_2}{\delta_3 c^2} \left( \frac{\chi_1 \chi_2^* |A_{30}|^2}{k_1 k_2 \cos \theta_1 \cos \theta_2} \right)^{1/2}. \end{aligned}$$

To satisfy the boundary conditions given by (2), we must have

$$\begin{aligned} A_1 &= \frac{\exp[-1/2(i\Delta k - \delta_1 + \delta_2 + \delta_3)z]}{J_{(\nu+1)}(x_0) J_{-\nu}(x_0) + J_{-(\nu+1)}(x_0) J_\nu(x_0)} \\ &\quad \times \left\{ A_{10} [J_{(\nu+1)}(x_0) J_{-\nu}(x_0 e^{-\delta_3 z}) + J_{-(\nu+1)}(x_0) J_\nu(x_0 e^{-\delta_3 z})] \right. \\ &\quad \left. + \frac{\partial A_1}{\partial z} \Big|_{z=0} [J_{-\nu}(x_0) J_\nu(x_0 e^{-\delta_3 z}) - J_\nu(x_0) J_{-\nu}(x_0 e^{-\delta_3 z})] \frac{1}{x_0 \delta_3} \right\}. \end{aligned} \quad (3)$$

Since  $A_1$  and  $A_2$  appear in (1) in a completely symmetric fashion, the solution for  $A_2$  can be obtained from (3) simply by interchanging the subscripts 1  $\leftrightarrow$  2. The solution given by (3) is the most general solution of the above set of truncated equations for arbitrary amplification coefficients  $\delta_n$ , velocities  $v_n = c/\text{Re}\sqrt{\varepsilon_n}$  of the interacting waves, and pump amplitude  $A_{30}$ . This solution seems to be unknown in traditional nonlinear optics (there are solutions with  $\delta_1 = \delta_2 = \delta_3$ , with  $\delta_3 = 0$  and arbitrary  $\delta_{1,2}$ , and with  $\delta_n = 0$  and arbitrary velocities of interacting waves, etc.<sup>[11-14]</sup>). This has given rise to a gap in the theory of parametric amplification and nonlinear transformations, especially in dispersive absorbing media.

Analysis of (3) can be carried out quite simply in each particular case. The problem can be formulated as follows: suppose that the nonlinear medium under consideration is inverted at the idler wave frequency  $\omega_2 = \omega_{30} - \omega_1$ , which may be, for example, in the optical band:

$$\delta_2 < 0, \quad \text{Im } \sqrt{\varepsilon_2} < 0.$$

A relatively weak pump wave  $\mathbf{E}_3$  of frequency  $\omega_3$  propagates in the direction of the positive  $z$  axis along the wave vector  $\mathbf{k}_3'$  and is received by the crystal face in the  $z = 0$  plane together with the input signal  $\mathbf{E}_{10}$  (in the direction of  $\mathbf{k}_1'$ ) which is to be amplified. The pump wave is weak in the above sense, i.e., it is produced by a laboratory  $\gamma$ -ray source, but its amplitude is much greater than the amplitudes of the waves  $\mathbf{E}_{1,2}$ . It is readily verified that, for this particular pump, the arguments of the Bessel function must satisfy the condition  $|x_0| \ll 1$ , and (3) can be expanded into a series in powers of this argument with the aid of the expansion<sup>[10]</sup> for the product  $J_\nu(a) J_\mu(b)$ . Retaining only the first two terms in the expansion, we obtain

$$\begin{aligned} A_1(z) &= A_{10} \left\{ 1 - \frac{4\pi^2 \omega_1^2 \omega_2^2 \chi_1 \chi_2^* |A_{30}|^2}{k_1 k_2 c^2 \cos \theta_1 \cos \theta_2} \right. \\ &\quad \times \left[ \frac{1 - \exp[-(i\Delta k - \delta_1 + \delta_2 + \delta_3)z]}{(i\Delta k - \delta_1 + \delta_2)^2 - \delta_3^2} - \frac{1 - \exp(-2\delta_3 z)}{2\delta_3(i\Delta k - \delta_1 + \delta_2 - \delta_3)} \right] \Big\} \\ &\quad + A_{20} \frac{2\pi\omega_1^2 \chi_1 A_{30}}{ik_1 c^2} \left[ \frac{1 - \exp[-(i\Delta k - \delta_1 + \delta_2 + \delta_3)z]}{i\Delta k - \delta_1 + \delta_2 + \delta_3} \right]. \end{aligned}$$

Since

$$E_1 \sim A_1(z) e^{-\delta_1 z},$$

it is clear that, when the pump intensity is relatively low, any nonlinearly transformed wave in a nonlinear medium is a superposition of several waves of the same frequency  $\omega_n$  but with different wave vectors ( $\mathbf{k}_n'$  and  $\mathbf{k}_n' + \mathbf{e}_z \Delta k$ ) and different attenuation coefficients (the former depend on the spectral parameters of the nonlinear medium at the given frequency, i.e.,  $\delta_n$  and  $\delta_n + \delta_3$ , and the latter depend only on the analogous parameters at the complementary frequencies among the set of frequencies satisfying the relation  $\omega_1 + \omega_2 = \omega_3$ ). An increase in the pump intensity leads to the "mixing" of the fields which thereby lose their individual characteristics and the rate of development of each of them becomes a function of all three spectral characteristics and the pump field, so that the problem reduces to a situation typical for traditional nonlinear optics. In its turn, the

reduction in the pump intensity will, in the limit, lead to linear optics.

If there are no strong resonance absorption lines in the neighborhood of  $\omega_3$  then, for the usual inversion densities encountered in quantum electronics, we have  $|\delta_2| > \delta_3$ , i. e.,  $-(\delta_2 + \delta_3) > 0$  for arbitrary  $\delta_1$ . The result is that, for depths  $z \gg \delta_3^{-1}$ , the field is given by

$$E_1 = \left\{ -E_{10} \frac{4\pi^2 \omega_1^2 \omega_2^2 \chi_1 \chi_2^* |E_{30}|^2}{k_1 k_2 c^4 [(i\Delta k - \delta_1 + \delta_2)^2 - \delta_3^2] \cos \theta_1 \cos \theta_2} \right. \\ \left. + E_{20} \frac{2\pi i \omega_1^2 \chi_1 E_{30}}{k_1 c^2 (i\Delta k - \delta_1 + \delta_2 + \delta_3) \cos \theta_1} \right\} \exp[i(\mathbf{k}, \mathbf{r} - \omega_1 t) - (i\Delta k + \delta_2 + \delta_3)z] \quad (4)$$

This formula leads to the condition for the nonthreshold parametric amplification of  $\gamma$ -radiation (in the case of narrow-band pumping and inversion at the frequency of the blank wave:  $\delta_2 + \delta_3 < 0$  or

$$\omega_2 \operatorname{Im} \sqrt{\varepsilon_2 + \omega_2} \operatorname{Im} \sqrt{\varepsilon_3} < 0.$$

It is clear from the above solution that the proposed model of the parametric  $\gamma$  amplifier does have a solution describing nonthreshold (i. e., independent of the absorption coefficient  $\delta_1$  at the frequency  $\omega_1$  of the amplified signal) coherent exponential amplification with a very high growth rate  $-(\delta_2 + \delta_3) \gtrsim 1 \text{ cm}^{-1}$ . The first term in (4) describes the direct amplification effect and the second is nonzero for  $E_{20}^* \neq 0$  and represents one-sided parametric transformation.

The transition from the general solution given by (3) to the approximation with  $|x_0| \ll 1$  corresponds physically to the situation with only one nonlinear transformation of each of the waves  $\mathbf{E}_1(\omega_3 - \omega_1 - \omega_2)$  and  $\mathbf{E}_2(\omega_3 - \omega_2 - \omega_1)$ , i. e., the input signal  $\mathbf{E}_{10}$  is transformed in the nonlinear medium into the wave  $\mathbf{E}_2$  of frequency  $\omega_2$  which is amplified with a growth rate  $-\delta_2 > 0$  (the medium is inverted at  $\omega_2$ ), and by interacting with the pump wave  $\mathbf{E}_3$  is transformed into  $\mathbf{E}_1$  of frequency  $\omega_1$ . Since the amplification of the wave  $\mathbf{E}_2$  can be very large, the resulting amplitude of the parametrically amplified wave  $\mathbf{E}_1$  at the exit from the system may exceed the amplitude of the input wave  $\mathbf{E}_{10}$  despite the fact that the nonlinear transformation coefficient is small. The subsequent terms in the expansion of (3) represent a large number of transformations, and the general solution describes the dynamics of the process, including all the transformations.

The difference between the above expressions for a nonlinear amplifying medium and the solution for the usual parametric amplifier is that the growth rate at the frequency  $\omega_1$  is independent of both  $\delta_1$  and of the synchronization parameter  $\Delta k$ , which are present only in the amplitude factor. This is so because the parameters of the medium (including the dissipative parameters) at the frequency of the incoming signal do not influence the amplification coefficient at the idler frequency  $\omega_2$  (and, in the case of the second, reverse, transformation, the resulting amplification at  $\omega_1$  is also independent of these parameters), and determine only the efficiency of the transformation. This remark also applies to  $\Delta k$  because the parametric amplification itself is connected not only with the accumulation of the results of nonlinear interactions, just as in ordinary non-

linear optics, but also (and this is most important) with amplification in the inverted medium at the idler-wave frequency. Nevertheless, it is important to note the very strong dependence of the transformation amplitude on the synchronization parameter  $[E_1 \sim (\Delta k)^{-2}$  for  $|\Delta k| \gg \delta_n]$ . The latter ensures that the maximum amplification and transformation occur for  $\Delta k = 0$ . We also note that the interaction that we are considering is essentially different from the usual parametric interaction in which the total number of quanta is always conserved (in a nonabsorbing medium) or reduced (in an absorbing medium). In fact, the number of quanta increases substantially (due to the idler wave), i. e., the Manley-Rowe relation is not satisfied.

### 3. NONTHRESHOLD PARAMETRIC $\gamma$ AMPLIFICATION IN THE SPATIALLY NONCOHERENT FIELD OF A $\gamma$ PUMP

It is shown above that the case of amplification in the field of a plane x-ray ( $\gamma$ -ray) pump wave can be used to obtain the complete solution and to provide a physical interpretation of the problem. The formulation of the problem is, however, somewhat idealized. This is so because most  $\gamma$ -ray sources are spatially noncoherent, and have a large angular divergence, whereas x-ray pump sources are characterized by temporal noncoherence. We shall now analyze the conditions for amplification in the field of a spatially noncoherent pump. For the sake of simplicity, we shall neglect spatial dispersion of the nonlinear susceptibility and, bearing in mind the amplification problem, we shall suppose that  $E_{20} = 0$ .

Expanding  $\mathbf{E}_n(\mathbf{r}, t)$  over transverse waves, we obtain

$$E_n^i(\mathbf{r}, t) = \int E_n^i(\mathbf{k}_{n\perp}, z) \exp[i(\mathbf{k}_{n\perp} \mathbf{r}_\perp - \omega_n t)] d\mathbf{k}_{n\perp},$$

where

$$E_n^i(\mathbf{k}_{n\perp}, z) = A_n^i(\mathbf{k}_{n\perp}, z) \exp(ik_{nz}z), \\ \mathbf{k}_n = \omega_n \sqrt{\varepsilon_n} / c, \quad k_{nz} = (k_n^2 - k_{n\perp}^2)^{1/2}, \quad \mathbf{b}_\perp = \mathbf{e}_x b_x + \mathbf{e}_y b_y, \quad E_n = \{E_n^i\}.$$

Passing now to the equations for the slowly-varying amplitudes  $A_n^i$ , we can readily show that

$$A_1^i(\mathbf{k}_{1\perp}, z) = \frac{4\pi^2 \omega_1^2 \omega_2^2}{k_1^2 k_2^2 c^4 \cos \theta_1} \int_0^z dz_1 \int d\xi \int \frac{\chi_1^{\mu\nu} \chi_2^{\mu'\nu'}}{\cos \theta_2} A_1^{i'}(\mathbf{k}_{1\perp}' - \mathbf{k}_{2\perp} + \mathbf{k}_{1\perp}, \xi) \\ \times A_{30}^{\mu\nu}(\mathbf{k}_{3\perp}) A_{30}^{\mu'\nu'}(\mathbf{k}_{3\perp}') \exp[i(k_{1z} + k_{2z} - k_{3z})\xi - i(k_{1z} + k_{2z} - k_{3z})z_1] d\mathbf{k}_{3\perp} d\mathbf{k}_{3\perp}'. \quad (5)$$

The statistical characteristics of the spatially noncoherent spontaneous emission of real macroscopic Mössbauer  $\gamma$ -ray sources can be represented with a high degree of accuracy by the spectral correlation function

$$G_3^{\mu\nu}(\mathbf{k}_{3\perp}, \mathbf{k}_{3\perp}') = \langle A_{30}^{\mu\nu}(\mathbf{k}_{3\perp}) A_{30}^{\mu'\nu'}(\mathbf{k}_{3\perp}') \rangle = 2\pi S^{\mu\nu}(\mathbf{k}_{3\perp}) \delta(\mathbf{k}_{3\perp} - \mathbf{k}_{3\perp}') \quad (6)$$

It was shown above that, when experiments with real laboratory sources are considered, it is sufficient to confine our attention to the first terms in the expansion of the general solution (3), and it is readily verified that this is equivalent to the substitution

$$A_1^i(\mathbf{k}_{1\perp}, \xi) \rightarrow E_{10}^i(\mathbf{k}_{1\perp})$$

i. e., the replacement of  $A_1$  by the input signal in the integrand in (5). This follows from the second equation in (1) because, in this particular approximation, the

source of the polarization  $P_2^{NL}$  at the frequency of the idler wave  $\omega_2$  is the input signal  $\mathbf{E}_{10}$  together with the pump. This and the previous approximation mean that the weakening of the input signal  $\mathbf{E}_{10}$  due to absorption in the medium (dissipation) with the growth rate  $\delta_1 = \text{Im}k_{1z}$  is much more important than its reduction resulting from the transformation into the idler wave. This situation is a direct consequence of the fact that the transformation coefficient which is connected with the nonlinear susceptibility and the pump is small. If we take the solution given by (5) subject to this assumption as the first approximation, we can obtain the second and higher-order approximations by iteration.

Using (5) and the spectral correlation function for the input signal  $G_{10}(\mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{1\perp})$ , we obtain the following expression for the average bilinear combination:

$$\langle E_i(\mathbf{k}_{1\perp}, z) E_i^*(\boldsymbol{\kappa}_{1\perp}, z) \rangle = G_i(\mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{1\perp}, z).$$

Assuming that the source of spatially noncoherent pump radiation is Gaussian, and using the equation

$$\langle a_i a_j a_k a_l \rangle = \sum_{i,j,k,l} \langle a_i a_j \rangle \langle a_k a_l \rangle,$$

we finally obtain (omitting, for simplicity, the obvious polarization indices)

$$G_i(\mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{1\perp}, z) = \left( \frac{2\pi\omega_1\omega_2}{c^2} \right)^2 \frac{|\chi_1|^2 |\chi_2|^2 4\pi^2}{k_1^2 k_2^2} \exp[i(k_{1z} - \kappa_{1z})z] \\ \times \left\{ G_{10}(\mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{1\perp}) \left| \int \frac{\exp[-i(k_{1z} + k_{2z} - k_{3z})z] S_3(\mathbf{k}_{3\perp})}{(k_{1z} + k_{2z} - k_{3z})(k_{1z} + k_{2z} - k_{3z}) \cos \theta_1 \cos \theta_2} d\mathbf{k}_{3\perp} \right|^2 \right. \\ \left. + \int \int G_{10}(\boldsymbol{\kappa}_{3\perp} - \mathbf{k}_{3\perp} + \mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{3\perp} - \mathbf{k}_{3\perp} + \boldsymbol{\kappa}_{1\perp}) \frac{S_3(\mathbf{k}_{3\perp}) S_3(\boldsymbol{\kappa}_{3\perp})}{\cos^2 \theta_1 \cos^2 \theta_2} (7) \right. \\ \left. \times \frac{\exp[-i(k_{1z} - k_{1z}' + k_{2z}' - k_{2z} - k_{3z} + k_{3z}')z] d\mathbf{k}_{3\perp} d\boldsymbol{\kappa}_{3\perp}}{(k_{1z} + k_{2z}' - k_{3z}') (k_{1z} + k_{2z} - k_{3z}) (\kappa_{1z}' + k_{2z} - k_{3z}')} \right\}.$$

The total intensity of the amplified signal is

$$I_1(z) = (2\pi)^{-2} \iint G_i(\mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{1\perp}, z) d\mathbf{k}_{1\perp} d\boldsymbol{\kappa}_{1\perp}.$$

The terms retained in (7), whose dependence on  $z$  is of the form  $\exp[-2(\delta_2 + \delta_3)z]$ , ensure the amplification effect. It follows from (7) that part of the spectral correlation function  $G_1(\mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{1\perp}, z)$  of the amplified signal coincides with the correlation function for the input signal (first component and the second for the diagonal terms with  $\mathbf{k}_{3\perp} = \boldsymbol{\kappa}_{3\perp}$ ) and corresponds to the nonthreshold amplification without change in the spatial characteristics in the nonlinear medium with the pump given by (6). The other part is very different and describes amplification with a change in these characteristics. Moreover, it is clear from Fig. 1 that, for the usual input signal,

$$G_{10}(\mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{1\perp}) = 2\pi S_1(\mathbf{k}_{1\perp}) \delta(\mathbf{k}_{1\perp} - \boldsymbol{\kappa}_{1\perp}),$$

each spectral component  $S_1(\mathbf{k}_{1\perp})$  interacts (i.e., is parametrically amplified) without change in direction with a large spatial set of spectral pump components  $S_3(\mathbf{k}_{3\perp})$  distributed uniformly near the direction of synchronism within the solid angle  $\Omega_0 \approx (k_2/k_3)^2$ , and may be amplified by taking up quanta from this "spatial pump reservoir." The system that we are considering is, in principle, very efficient because amplification in a given direction

in an infinitely small spatial interval occurs because of the removal of quanta from a finite interval (the low efficiency in the present problem is connected with the fact that the particular parameters are small). The amplification mechanism is parametric, as before. Each component  $S_1(\mathbf{k}_{1\perp})$  interacts with the set  $S_3(\mathbf{k}_{3\perp})$  and excites a set of waves of nonlinear polarization at frequency  $\omega_2$ , and these waves are sources of idler waves distributed over all directions in the left hemisphere. The idler waves are amplified in the inverted medium with a growth rate  $-\delta_2 > 0$  and interact with the spatially distributed pump, transforming it into the amplified signal for the same fixed value of  $k_{1z}$ . The second part of the spectral correlation function  $G_1(\mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{1\perp}, z)$  of the amplified signal in (7) is connected with the "nondiagonal" interaction (the direct and converse transformations take place with the participation of different  $\mathbf{k}_{3\perp}$ ) and leads to the spreading of the angular spectrum of the amplified signal.

As an example, we take the special form of (7) corresponding to a plane-wave input signal incident on a nonlinear medium at angle  $\theta_n = \chi(\mathbf{k}_n, \mathbf{e}_z)$ :

$$G_{10}(\mathbf{k}_{1\perp}, \boldsymbol{\kappa}_{1\perp}) = 2\pi S_1 \delta(\mathbf{k}_{1\perp} - \boldsymbol{\kappa}_{1\perp}) \delta(\mathbf{k}_{1\perp} - \boldsymbol{\kappa}_{1\perp}).$$

Since for  $\theta_n < \pi/2$  we can always write

$$k_{nz} = k_{nz}' + i\delta_{nz},$$

it is readily shown that, for  $\mathbf{k}_{1\perp} \parallel \mathbf{e}_z$ ,

$$I_1(z) \approx I_{10} |K(z)|^2 \exp(-2(\delta_2 + \delta_3)z),$$

where

$$|K(z)| = \left( \frac{2\pi\omega_1\omega_2}{c^2} \right)^2 \frac{2\pi |\chi_1 \chi_2|^2}{k_1 |\delta_1 - \delta_2|} \frac{I_3}{k_2^2 \Delta \Omega} \left\{ \frac{1}{z^2 (\delta_1 - \delta_2)^2} + 1 \right\}^{1/2}. \quad (8)$$

The first term in this expression corresponds to amplification without a change in the spatial properties, and the second with a change in these properties. It is assumed in the solution that the spectrum  $G_3(\mathbf{k}_{3\perp})$  is distributed uniformly within the solid angle  $\Delta \Omega$ , symmetrically relative to the direction of synchronism. We note that, for real nuclear and optical parameters, the

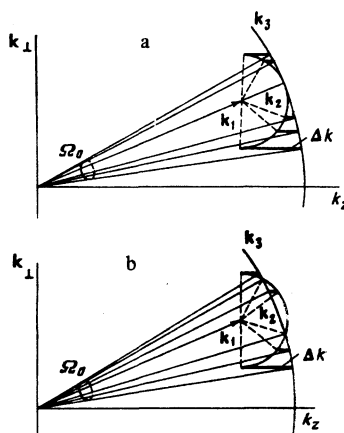


FIG. 1. Paramagnetic interaction of a spectral component of the amplified signal with the spatially noncoherent pump  $S_3(\mathbf{k}_{3\perp})$ : a—scalar synchronism, b—vector synchronism.

directional spreading [second term in (7) and (8)] is no more than a few minutes of arc.

#### 4. BROAD-BAND PARAMETRIC PUMP

The case of a monochromatic pump corresponds approximately to the amplification of Mössbauer  $\gamma$  radiation (provided the spectral width of the  $\gamma$  pump is small in comparison with the characteristic spectral widths of the dispersion curves of the nonlinear susceptibilities  $\chi^{NL}$  or, which amounts to the same, for negligible dispersion of  $\chi^{NL}$  within the limits of the spectral width of  $I_3$ ). It is useful to note that there is practically no information in the literature on the effects of nonlinear optics for random fields in dispersive media (including the dispersion of nonlinear susceptibility, and not merely that due to the linear response<sup>[15]</sup>). This is understandable in the case of the optical band, where the spectral width of the dispersion curves of nonlinear susceptibilities is usually much greater than the bandwidth of the interacting radiation (usually laser radiation) excluding, possibly, picosecond pulses which have not been extensively investigated. However, it becomes a serious impediment to the study of problems in nonlinear  $\gamma$ -ray optics where the very presence of the Mössbauer effect leads to dispersion curves of natural width in linear and nonlinear susceptibilities and equally narrow-band spontaneous emission of the pump source as well as a possible variation within certain limits of the ratio between them.<sup>[6,16]</sup> There is considerable interest in this connection in the analysis of the problem of amplification in a broad-band parametric x-ray and  $\gamma$ -ray pump in an inverted, highly dispersive, nonlinear real medium.

We shall confine our attention to the case where the spectrum of the random broad-band pump is broader than the width of the dispersion curves of the nonlinear susceptibilities. This condition is satisfied in the case of x-ray (bremsstrahlung and characteristic) pump radiation, ordinary (non-Mössbauer)  $\gamma$  radiation, and, in a number of cases (depending on the particular ratio of parameters), the Mössbauer  $\gamma$  radiation. A pump of this kind can be approximately described as delta-correlated in time:

$$G_s^{ij}(t, t') = \langle A_{30}^{ij}(t) A_{30}^{ij}(t') \rangle = 2\pi S_3^{ij} \delta(t - t'), \quad (9)$$

provided the width  $\Delta\omega$  of the pump spectrum is greater than the width  $\Gamma$  of the dispersion curves. At the same time,  $S_3 \approx I_3/\Delta\omega$ , where  $S_3$  is the spectral density and  $I_3$  the intensity of the pump. Transforming from (1) to the equations for the slowly-varying amplitudes

$$E_n^i(r, t) = \int A_n^i(\omega_n, z) \exp[i(k_n r - \omega_n t)] d\omega_n$$

and omitting the polarization indices, we obtain

$$\begin{aligned} \frac{\partial A_1(\omega_1, z)}{\partial z} &= \frac{2\pi\omega_1^2}{ik_1 c^2 \cos \theta_1} \iint \chi_1(\omega_1, \omega_2, \omega_3) \delta(\omega_1 + \omega_2 - \omega_3) A_{30}(\omega_3) A_{20}^*(\omega_2) \\ &\times \exp[-i(k_{1z} + k_{2z} - k_{3z})z] d\omega_2 d\omega_3 + \frac{4\pi^2\omega_1^2}{k_1 c^4 \cos \theta_1 \cos \theta_2} \int d\xi \iiint \chi_1(\omega_1, \omega_2, \omega_3) \\ &\times \chi_2^*(\bar{\omega}_1, \omega_2, \bar{\omega}_3) \delta(\omega_1 + \omega_2 - \omega_3) \delta(\bar{\omega}_1 + \omega_2 - \bar{\omega}_3) A_1(\bar{\omega}_1, \xi) A_{30}(\omega_3) A_{30}^*(\bar{\omega}_3) \\ &\times \exp[-i(k_{1z} + k_{2z} - k_{3z})z + i(k_{1z} + k_{2z} - k_{3z})\xi] \frac{\omega_2^2}{k_2^2} d\bar{\omega}_1 d\omega_2 d\omega_3 d\bar{\omega}_3, \end{aligned}$$

where  $\bar{k}_n = k(\bar{\omega}_n)$ .

In analyzing the amplification problem, we shall assume that  $E_{20} = 0$ . Let us confine our attention to the approximation of a moderately weak pump source and substitute  $A_1(\bar{\omega}_1, \xi) \rightarrow E_{10}(\bar{\omega}_1)$  is the integrand in the last expression. Using (9), this leads to the equation for the mean amplitudes  $\langle A_n \rangle = \bar{A}_n$  for scalar synchronism and  $k_n \parallel e_z$ :

$$\begin{aligned} \frac{\partial \bar{A}_1(\omega_1, z)}{\partial z} &= \frac{4\pi^2\omega_1^2 S_3}{c^3 k_1} \int_0^z d\xi \int \chi_1(\omega_1, \omega_2, \omega_1 + \omega_2) \chi_2^*(\omega_1, \omega_2, \omega_1 + \omega_2) \frac{\omega_2}{n_2} \\ &\times \exp\left[-\left(\frac{\omega_1}{c} n_1 + \frac{\omega_2}{c} n_2 - \frac{\omega_1 + \omega_2}{c} n_3'\right)(z - \xi) - \frac{\omega_1 + \omega_2}{c} n_3''(z + \xi)\right] E_{10}(\omega_1) d\omega_2, \end{aligned}$$

where  $n_p = \sqrt{\epsilon_p} = n_p' + in_p''$ ,  $n_p = n(\omega_p)$ .

We now introduce the natural assumption that there are no strong resonance absorption lines within the bandwidth  $\Delta\omega$  of the pump, i. e., resonance absorption at the frequency  $\omega_1 + \omega_2$  is small in comparison with the nonresonance absorption which can be arbitrary. The problem can be solved even without this assumption, but the solution is then very unwieldy. The validity and usefulness of this assumption will be examined below.

To calculate the inner integral in (10), let us extend the limits of integration with respect to  $\omega_2$  to  $\pm\infty$  and close the contour of integration. Since, as  $|\omega_2| \rightarrow \infty$  (i. e., at frequencies much greater than the resonance frequencies),

$$n_2' - n_3' \approx \text{Re}(e^{i\omega_2} - e^{i(\omega_1 + \omega_2)}) > 0,$$

the condition of Jordan's lemma is satisfied in the lower half-plane of the complex values of  $\omega_2$ . Near resonance

$$\epsilon_2 = \epsilon_{20} + p_2/(\omega_2 - \omega_{20} + i\Gamma_2),$$

where  $\epsilon_{20}$  is the nonresonance part of  $\epsilon_2$ , and  $p_2 = \pm |p_2|$  corresponds to absorption or amplification of the idler wave. The singularities of  $n_2^*$  lie in the upper half-plane of  $\omega_2$ . Hence, it follows that nontrivial solutions of the problem are possible only when the product

$$\chi_1(\omega_1, \omega_2, \omega_1 + \omega_2) \chi_2^*(\omega_1, \omega_2, \omega_1 + \omega_2)$$

has poles in the lower half-plane. Moreover, since we are interested in solutions corresponding to a rapid increase in the idler-wave amplitude (i. e., in the amplified signal  $E_1$ , as well) for  $p_2 = -|p_2|$ , such singularities should occur for  $\text{Re}\omega_2 \approx \omega_{20}$ , i. e., for the same values as they occur in the case of  $\epsilon_2$ .

In accordance with the foregoing, we shall write the product of the nonlinear susceptibilities in the form

$$\begin{aligned} &\chi_1(\omega_1, \omega_2, \omega_1 + \omega_2) \chi_2^*(\omega_1, \omega_2, \omega_1 + \omega_2) \\ &= \frac{\beta_1(\omega_1, \omega_2, \omega_1 + \omega_2)}{\omega_2 - \omega_{2k} + i\Gamma_{2k}} + \beta_2(\omega_1, \omega_2, \omega_1 + \omega_2). \end{aligned} \quad (11)$$

where  $\beta_{1,2}(\omega_1, \omega_2, \omega_1 + \omega_2)$  has no singularities in the lower half-plane, and  $\omega_{2k}$  and  $\Gamma_{2k}$  are not, in general, equal to  $\omega_{20}$  and  $\Gamma_2$ . We shall show below that (11) is

the only possible representation for real two- and three-level quantum systems.

We now find the residue at  $\omega_2 = \omega_{2k} - i\Gamma_{2k}$  and, by evaluating the space integrals, we finally obtain

$$\bar{E}_1 = -E_{10} K_\omega \exp \left\{ \left[ i \left( k_1' - \Delta\bar{k} + \frac{\Gamma_{2k}}{c} n_{2k}'' \right) - \delta_2 - \delta_3 \right] z - \omega_1 t \right\} \quad (12)$$

where

$$K_\omega = \frac{4\pi^2 \omega_1^2 \omega_{2k}^2 \beta_1}{k_1 k_2 c^4} \frac{2\pi \Gamma_{2k} S_2}{i \Gamma_{2k} (i \Delta\bar{k} - \delta_1 + \delta_2)^2 - \delta_3^2}$$

$$n_{2k} = n_2 (\omega_{2k} - i\Gamma_{2k}), \quad n_{3k} = n_3 (\omega_1 + \omega_{2k} - i\Gamma_{2k}),$$

$$\delta_2 = [\Gamma_{2k} (n_{2k}' - n_{2k}'') + \omega_{2k} n_{2k}''] / c, \quad \delta_3 = (\omega_1 + \omega_{2k}) n_{3k}'' / c.$$

$$\Delta\bar{k} = [\omega_1 n_1' + \omega_{2k} n_{2k}' - (\omega_1 + \omega_{2k}) n_{3k}' - \Gamma_{2k} n_{2k}''] / c.$$

The results given by (12) can be rewritten in the form

$$\bar{E}_1 = -E_{10} \exp \left[ i \left( k_1' - \Delta\bar{k} - \frac{\Gamma_{2k}}{c} n_{2k}'' \right) z - \omega_1 t \right] - (\delta_2 + \delta_3) (z - l).$$

The real exponential condition begins for  $z > l'$  when  $|E_1/E_{10}| > 1$ , where

$$\exp [(\delta_2 + \delta_3)l] = K_\omega. \quad (13)$$

Despite the fact that the representation of the real pump by a delta-correlated process formally leads to an infinite integrated pump intensity

$$I_{32} \sim \int S_3 d\omega_3,$$

the solution of the problem for a highly dispersive, nonlinear, real medium removes this defect because the solution shows that the process of parametric amplification involves the participation of only photons from a spectral interval of width  $\sim \Gamma_{2k} < \Delta\omega$ , and the effective integrated pump intensity is  $2\pi \Gamma_{2k} S_3$ . This point was noted in<sup>[17]</sup> in the case of the ordinary parametric trans-

formation. The condition for nonthreshold parametric amplification in the case of a broad-band x-ray or  $\gamma$ -ray noise pump follows from (12):

$$(\delta_2 + \delta_3) = [\omega_{2k} n_{2k}'' + \Gamma_{2k} (n_{2k}' - n_{2k}'') + (\omega_1 + \omega_{2k}) n_{3k}''] / c < 0, \quad (14)$$

and this can, of course, be satisfied only for  $n_{2k}'' < 0$ . Let us compare (14) with the amplification condition for the monochromatic pump

$$(\delta_2 + \delta_3) = (\omega_2 n_2'' + (\omega_1 + \omega_2) n_3'') / c < 0. \quad (15)$$

Since the average attenuation (growth) of the wave at distance  $\lambda$  is very small, i. e.,

$$\operatorname{Re} \sqrt{\bar{\epsilon}_{2k}} \gg \operatorname{Im} \sqrt{\bar{\epsilon}_{2k}}, \quad \operatorname{Im} [p_2 / (\omega_{2k} - \omega_{20} - i(\Gamma_2 + \Gamma_{2k}))] \ll 1,$$

we have, by expanding  $\sqrt{\bar{\epsilon}_{2k}}$ ,

$$\left[ \omega_{2k} \left( \operatorname{Im} \sqrt{\bar{\epsilon}_{2k}} + \frac{p_2/2 \operatorname{Re} \sqrt{\bar{\epsilon}_{2k}}}{\omega_{2k} - \omega_{20} - i(\Gamma_2 + \Gamma_{2k})} \right) + \Gamma_{2k} \operatorname{Re} (\sqrt{\bar{\epsilon}_{2k}} - \bar{\nu}_{2k}) + (\omega_1 + \omega_{2k}) \operatorname{Im} \bar{\nu}_{2k} \right] \frac{1}{c} < 0, \quad (14a)$$

$$\omega_{20} \left[ \operatorname{Im} \sqrt{\bar{\epsilon}_{20}} + \frac{p_2/2 \operatorname{Re} \sqrt{\bar{\epsilon}_{20}}}{\Gamma_2} \right] + (\omega_1 + \omega_{20}) \operatorname{Im} \bar{\nu}_{20} \frac{1}{c} < 0. \quad (15a)$$

Comparison of the last two expressions will show that the maximum possible amplification growth rate in the field of the broad-band noise pump (for  $\omega_{2k} = \omega_{20}$ ) in the

limiting case when  $\Gamma_{2k} \ll \Gamma_2$  is equal to the resonance value of the coherent amplification growth rate in the case of the monochromatic pump. We note that this is accompanied by a sharp reduction in the effectively utilized part of the pump  $2\pi \Gamma_{2k} S_3$  and, as a consequence, by a reduction in the coefficient of nonlinear transformation  $K_\omega$ . When  $\Gamma_{2k} = \Gamma_2$ , the maximum value is

$$(\delta_2 + \delta_3)_{\max} = 1/2 (\delta_2 + \delta_3)$$

and corresponds to small values of  $\Gamma_2$ . The quantity  $p_2$  can easily be obtained from the identity

$$|p_{\text{res}}| = |p_2| \omega_{20} / \Gamma_2 c,$$

where  $\bar{n}$  and  $T$  are, respectively, the inversion population and the lifetime of atoms excited at the frequency of the idler wave,  $\sigma_{\text{res}}$  is the resonance value of the cross section for stimulated emission, and  $g$  is the degeneracy factor for the transition. The final expression is

$$|p_2| = c^3 |\bar{n}| g / 4\pi^2 \omega_{20}^2 T.$$

As shown below, it is sometimes more convenient to use a pump at the frequency  $\omega_1 = \omega_3 - \omega_2$ . The analytic singularities of the product

$$\chi_2(\omega_3 - \omega_2, \omega_2, \omega_3) \chi_3(\omega_3 - \omega_2, \omega_2, \omega_3),$$

which appears in the course of solution of the problem, now lie in the upper half-plane of  $\omega_2$  at the point  $\omega_{2k} + i\Gamma_{2k}$ , and the singularities of  $n_2 = \sqrt{\bar{\epsilon}_2}$  lie in the lower half-plane. By closing the contour of integration in the upper half-plane, we obtain, after the appropriate evaluations,

$$\bar{E}_3 = -E_{30} \frac{4\pi^2 \omega_{2k}^2 \omega_3^2 \beta_1}{k_{2k} k_3 c^4} \frac{2\pi \Gamma_{2k} S_1}{i \Gamma_{2k} (i \Delta\bar{k} - \delta_2 + \delta_1)^2 - \delta_1^2} \times \exp \left\{ \left[ i \left( k_3' + \Delta\bar{k} - \frac{\Gamma_{2k}}{c} n_{1k}'' \right) - \delta_1 - \delta_2 \right] z - \omega_3 t \right\}, \quad (16)$$

where

$$\langle A_{10}(t) A_{10}^*(t') \rangle = 2\pi S_1 \delta(t - t').$$

$$n_{1k} = n_1 (\omega_3 - \omega_{2k} - i\Gamma_{2k}), \quad n_{2k} = n_2 (\omega_{2k} + i\Gamma_{2k}).$$

$$\delta_1 = (\omega_3 - \omega_{2k}) n_{1k}'' / c, \quad \delta_2 = [-\Gamma_{2k} (n_{2k}' - n_{1k}') + \omega_{2k} n_{2k}''] / c.$$

$$\Delta\bar{k} = [(\omega_3 - \omega_{2k}) n_{1k}' + \omega_{2k} n_{2k}' - \omega_3 n_3' + \Gamma_{2k} n_{2k}''] / c.$$

## 5. THE PROBLEM OF NONLINEAR SUSCEPTIBILITY AND NUMERICAL ESTIMATES

The final analysis of the conditions for the realization of a nonthreshold parametric amplification of x-ray or  $\gamma$ -ray radiation must take into account the particular expressions for the nonlinear susceptibilities. Since the final aim is to obtain the conditions for the maximization of  $\chi^{NL}$  for given amplification (this is obvious because  $\chi^{NL}$  increases sharply near the resonances, but there is an attendant increase in absorption), we shall consider the resonance susceptibility. The nonlinear susceptibility of the three-level system for a real, i. e., relatively weak (without saturation effects) intensity of the interacting waves, is given by<sup>[18]</sup> ( $V$  is the volume of the nonlinear medium and  $N$  is the number of resonance centers)

$$\chi_{mk} = (N/V) d_{kn} d_{nm} d_{mk} \lambda_{mk}^{(2)} / \hbar^2.$$

$$\lambda_{23}^{(2)} \equiv \lambda_1^{(2)} = \frac{\rho_{33} - \rho_{11}}{(\omega_3 - \omega_{30} + i\Gamma_{31})(\omega_1 - \omega_{10} + i\Gamma_{32})} + \frac{\rho_{11} - \rho_{22}}{(\omega_3 - \omega_{20} + i\Gamma_{21})(\omega_1 - \omega_{10} + i\Gamma_{32})},$$

$$\lambda_{21}^{(2)} \equiv \lambda_2^{(2)} = \frac{\rho_{33} - \rho_{11}}{(\omega_2 - \omega_{20} + i\Gamma_{21})(\omega_3 - \omega_{30} + i\Gamma_{31})} + \frac{\rho_{33} - \rho_{22}}{(\omega_2 - \omega_{20} + i\Gamma_{21})(\omega_1 - \omega_{10} - i\Gamma_{32})} \quad (17)$$

$$\lambda_{31}^{(2)} \equiv \lambda_3^{(2)} = \frac{\rho_{11} - \rho_{22}}{(\omega_2 - \omega_{20} + i\Gamma_{21})(\omega_3 - \omega_{30} + i\Gamma_{31})} + \frac{\rho_{22} - \rho_{33}}{(\omega_1 - \omega_{10} + i\Gamma_{32})(\omega_3 - \omega_{30} + i\Gamma_{31})}$$

where

$$\Gamma_{mn} = \Gamma_m + \Gamma_n, \quad \omega_{10} = \omega_{32}, \quad \omega_{20} = \omega_{21}, \quad \omega_{30} = \omega_{31}.$$

If  $\omega_n - \omega_{n0} \gg \Gamma_{nk}$ , (17) becomes identical with the expression for the two-level system. Using the explicit expressions for  $\chi_{mk} \equiv \chi_n$ , we can readily verify that the product  $\chi_1 \chi_2^*$  in (10) has only one pole in the lower half-plane of complex  $\omega_2$ , namely,

$$\omega_1 = \omega_{30} - \omega_1 - i\Gamma_{32},$$

and, in the case of pumping at the frequency  $\omega_1 = \omega_3 - \omega_2$ , the product  $\chi_2 \chi_3$  has a pole at

$$\omega_2 = \omega_3 - \omega_{10} + i\Gamma_{32},$$

i. e., (11) is, in fact, valid and

$$\beta_1(\omega_1, \omega_2) = \left( \frac{N d_{12} d_{23} d_{31}}{V \hbar^2} \right)^2 \frac{\rho_{11} - \rho_{33}}{(\omega_1 - \omega_{10} + i\Gamma_{32})(\omega_2 - \omega_{20} - i\Gamma_{21})},$$

$$\times \left[ \frac{\rho_{11} - \rho_{33}}{(\omega_1 + \omega_2 - \omega_{30} - i\Gamma_{31})} - \frac{\rho_{22} - \rho_{33}}{(\omega_1 - \omega_{10} + i\Gamma_{32})} \right],$$

$$\beta_1(\omega_2, \omega_3) = \left( \frac{N d_{12} d_{23} d_{31}}{V \hbar^2} \right)^2 \frac{\rho_{22} - \rho_{33}}{(\omega_2 - \omega_{20} + i\Gamma_{21})(\omega_3 - \omega_{30} + i\Gamma_{31})},$$

$$\times \left[ \frac{\rho_{11} - \rho_{22}}{(\omega_2 - \omega_{20} + i\Gamma_{21})} - \frac{\rho_{22} - \rho_{33}}{(\omega_3 - \omega_{30} - \omega_{10} + i\Gamma_{32})} \right].$$

Let us now consider some concrete models of resonance nonlinear systems suitable for nonthreshold parametric amplification.

**A. Coherent transformation of x radiation.** As the nonlinear system for the x-ray band, it is convenient to take the idealized three-level system from the atomic spectrum, comprising the zeroth level 1, the excited level 2 corresponding to the optical transition of frequency  $\omega_{21} = \omega_{20}$ , and one of the x-ray levels of the same atom. The transition matrix element  $d_{nm}$  can be related to the known lifetime  $T$  of the given level for radiative decay:

$$d_{nm} = (\hbar c^3 / 16\pi^2 \omega_{nm}^3 T_{nm})^{1/2}.$$

Since, in the x-ray band,

$$\Gamma_3 \gg \Gamma_1, \quad \Gamma_1 \approx 0, \quad \Gamma_{32} \approx \Gamma_{31} \approx \Gamma_3, \quad \rho_{33} \approx 0, \quad \rho_{11} \approx \rho_{22} \approx 1/2$$

(but  $\rho_{22} - \rho_{11} > 0$ ), the resonance values of the nonlinear susceptibilities (17) are given by

$$\chi_1 \approx \frac{(N/V) (c^3 / \omega_1 \omega_2 \omega_3)^{1/2}}{2\sqrt{\hbar} (4\pi)^3 (T_{32} T_{31} T_{21})^{1/2} \Gamma_3 (\Gamma_2 + \Gamma_3)} \quad (18)$$

$$\chi_2 \approx \frac{(N/V) (c^3 / \omega_1 \omega_2 \omega_3)^{1/2}}{2\sqrt{\hbar} (4\pi)^3 (T_{32} T_{31} T_{21})^{1/2} \Gamma_2 \Gamma_3}.$$

When  $c/\omega_3 = \lambda_3 \sim \lambda_1 \sim 10^{-8}$  cm,  $\lambda_2 \sim 10^{-4}$  cm,  $N/V = 10^{21}$  cm<sup>-3</sup>,  $\Gamma_3^{-1} \approx T_{31} \approx T_{32} \approx 10^{-15}$  sec,  $T_{21} \sim 10^{-4}$  sec,  $\Gamma_2 \sim 10^{10}$  sec<sup>-1</sup>, we have  $\chi_1 \approx 10^{-11}$  cgs esu,  $\chi_2 \approx 10^{-6}$  cgs esu, and,

when  $\lambda_3 \sim \lambda_1 \sim 10^{-9}$  cm, we obtain  $\chi_1 \approx 10^{-13}$  cgs esu and  $\chi_2 \approx 10^{-8}$  cgs esu. The integrated pump intensity

$$2\pi\Gamma_3 S = |E_{p \text{ eff}}|^2$$

is given by

$$|E_{p \text{ eff}}| = [4\pi\hbar\omega_3 N_3 1.6 \cdot 10^{-12} / c]^{1/2} \quad (\text{cgs esu/cm}),$$

where  $N_3$  is the integrated flux of pump photons in the spectral interval  $\sim \Gamma_{31}$  (the quantities  $\hbar\omega_3$  and  $N_3$  are expressed in electron volts and cm<sup>-2</sup> · sec<sup>-1</sup>, respectively). When  $\lambda_3 \sim \lambda_1 \sim 10^{-8}$  cm and  $N_3 = 3 \times 10^{10}$  cm<sup>-2</sup> · sec<sup>-1</sup> ( $\sim 1$  Ci), we have  $2\pi\Gamma_3 S \approx 10^{-7}$  (cgs esu/cm)<sup>2</sup>, and at  $\lambda_3 \sim \lambda_1 \sim 10^{-9}$  cm we have  $2\pi\Gamma_3 S \approx 10^{-6}$  (cgs esu/cm)<sup>2</sup>. For the above parameter values, the nonlinear transformation coefficient  $K_\omega$  given by (13) in the direction of synchronism is equal to  $10^{-11}$  in the first case and  $10^{-13}$  in the second. An actual increase in the pump intensity by three or four orders of magnitude results in an increase in  $K_\omega$  by a comparable factor.

The problem is solved on the assumption that resonance absorption at the frequency  $\omega_3$  is small in comparison with the nonresonance absorption. This is necessary for the development of real devices. This condition can be achieved in practice for a low concentration of absorbing centers, which corresponds to the typical situation in experimental quantum electronics in which the centers form an impurity. As an example, we consider nonlinear parametric amplification in crystals containing uranium ions with a concentration of 0.1–1.0% in a suitable nonlinear host (for example, optical amplifiers using CaFe:V<sup>3+</sup>). For typical optical transition parameters of uranium  $\lambda_2 \approx 10^{-4}$  cm,  $\Gamma_2 \approx 10^{11}$  sec<sup>-1</sup>,  $T_2 \approx 10^{-4}$  sec,  $n = N(\rho_{22} - \rho_{11})/V \approx 10^{20}$  cm<sup>-3</sup>,  $p_2 \approx 10^9$  cgs esu, and resultant absorption coefficient from (14a) of the order of 1 cm<sup>-1</sup>, amplification is possible for  $\Gamma_3 \lesssim 10^{13}$  sec<sup>-1</sup>, which is difficult to achieve at present (there are no data on such narrow-band x-ray transitions). The amplification condition can be satisfied by increasing the concentration of active centers and the degree of inversion of the latter, or by choosing a medium with better parameters.

**B. Coherent transformation of Mössbauer  $\gamma$  radiation.** The three-level system necessary for increasing the efficiency of coherent transformation in the  $\gamma$  band can be simulated with the aid of resonance optical and  $\gamma$  transitions, as follows. It is known<sup>[19]</sup> that the interaction between  $\gamma$  rays and nuclei (i. e., with atoms and molecules) is accompanied both by purely nuclear transitions and by mixed electron-nuclear transitions due to, for example, the recoil effect (motion of the nucleus in the center-of-mass system of the atom or molecule leads to transitions in the electron subsystem). The matrix element of such a nondiagonal (in nuclear and electron quantum numbers) transition is

$$d \approx d_{\text{nuc}1, e1} (m_e \omega_\gamma / Mec),$$

where  $d_{\text{nuc}1, e1}$  is the matrix element for purely nuclear or purely electron transitions,  $m_e$ ,  $e$ ,  $M$ , and  $\omega_\gamma$  are, respectively, the mass of the electron, the charge of the electron, the mass of the atom (molecule), and the frequency of the  $\gamma$ -radiation. For typical parameters

in the Mössbauer range and intermediate atomic weights ( $Z = 40$ ),

$$m_e \omega_p / Mec \approx 10^{14}.$$

Typically,  $d_{01} \sim 10^{-17}$  cgs esu. In that case,

$$d = \eta d_{\text{nuc}} \approx 10^{-3} d_{\text{nuc}}. \quad (19)$$

Out of the set of nuclear, electron, and mixed transitions, it is practically always possible to select an appropriate three-level system suitable for efficient nonlinear transformation. This choice can be made on the basis of the following considerations. Since the coefficient of nonlinear parametric  $\gamma$ -amplification is independent of the absorption coefficient at the frequency  $\omega_s$  of the amplified signal, and depends on the corresponding quantity at the pump frequency  $\omega_p$ , it is reasonable to choose the frequency  $\omega_s$  to be equal to the frequency of the purely nuclear transition, and take the frequency of the pump signal equal to the frequency of the nuclear-electron transition [i. e., take  $d \equiv d_{13}$  in (19)]. Because of (19), resonance absorption at the pump frequency is then strongly suppressed. If we suppose that the  $1 \rightarrow 2$  transition is optical with frequency  $\omega_2$ , and the  $1 \rightarrow 3$  and  $2 \rightarrow 3$  transitions lie in the  $\gamma$ -band and are such that  $\omega_{31} = \omega_{32} + \omega_{21}$ , then it is more convenient in the achievement of our aim to use pump frequency  $\omega_1 = \omega_3 - \omega_2$  and the solution given by (16) or, in the case of a nonplane pump wave, the solution given by (6) with the substitution  $F_1 \rightarrow F_3$ . In fact, it turns out that the resonance absorption coefficient at the frequency  $\omega_p = \omega_1$  depends on the matrix element (19), which facilitates the suppression of resonance absorption at this frequency without a change in the nonlinear transformation coefficient. The frequency  $\omega_2$  should coincide with the optical transition frequency of the Mössbauer atom and be equal to the frequency at which the optical amplification takes place (i. e., the atom should have a Mössbauer nucleus and electron transitions suitable for optical amplification).

There is a large number of such atoms (Sm<sup>149</sup>, Eu<sup>151</sup>, Eu<sup>153</sup>, Dy<sup>160</sup>, Dy<sup>161</sup>), and many Mössbauer experiments have been carried out on the nuclei of these atoms. Their electron transitions have been used to produce amplification and generation in the optical band. Moreover, a similar situation can be realized for a much broader class of materials if we recall that the nucleus-electron interaction effect can be realized not only between the nucleus and electrons in a given atom, but also between the nucleus and the electrons of different atoms comprising a molecule, i. e., the molecule can contain a Mössbauer atom and an atom with an inverted optical transition. The efficiency of such transitions can be of the same order as for intraatomic transitions. For typical Mössbauer  $\gamma$ -transitions, we have  $10^{-6}$  sec  $\geq T_{31,32} \geq 10^{-8}$  sec. It follows from (17) that the resonance values of those parts of the nonlinear susceptibilities that enter into the expression for  $\beta_1$  (we shall denote them by  $\tilde{\chi}_n$ ) are given by

$$|\tilde{\chi}_1|_{\omega_1} \approx a_0 \left| \frac{\rho_{11} - \rho_{33}}{\Gamma_2(\Gamma_2 + \Gamma_3)} \right|, \quad |\tilde{\chi}_2|_{\omega_1} \approx a_0 \left| \frac{\rho_{11} - \rho_{33}}{\Gamma_2 \Gamma_3} + \frac{\rho_{22} - \rho_{33}}{\Gamma_2(\Gamma_2 + \Gamma_3)} \right| \quad (20)$$

for pump radiation at frequency  $\omega_3 = \omega_1 + \omega_2$  and, correspondingly,

$$|\tilde{\chi}_2|_{\omega_1} \approx a_0 \left| \frac{\rho_{22} - \rho_{33}}{\Gamma_2(\Gamma_2 + \Gamma_3)} \right|, \quad |\tilde{\chi}_3|_{\omega_1} \approx a_0 \left| \frac{\rho_{11} - \rho_{22}}{\Gamma_2 \Gamma_3} + \frac{\rho_{22} - \rho_{33}}{\Gamma_3(\Gamma_2 + \Gamma_3)} \right| \quad (21)$$

for pump frequency  $\omega_1 = \omega_3 - \omega_2$ , where

$$a_0 = \eta \frac{N}{V} \frac{(\lambda_1 \lambda_2 \lambda_3)^{3/2}}{\sqrt{\hbar} (4\pi)^2 (T_{31} T_{32} T_{21})^{1/2}}.$$

For a typical linewidth of an optical transition in a solid,  $\Gamma_2 \sim 10^{11} - 10^{10}$  sec and  $\rho_{11} \sim \rho_{22} \sim 1/2$  (but  $\rho_{22} - \rho_{11} > 0$ ),  $\rho_{33} \approx 0$ ,  $\Gamma_{31} \approx \Gamma_3 \approx T_{31}^{-1}$ ,  $\Gamma_{32} \approx \Gamma_2$ ,  $\Gamma_{21} \approx \Gamma_2$ , all the  $|\tilde{\chi}_n|$  are equal to

$$|\tilde{\chi}_n|_{\omega_1} \approx \eta N (\lambda_1 \lambda_2 \lambda_3)^{3/2} / 2V \sqrt{\hbar} (4\pi)^2 T_{21}^{-1} \Gamma_2. \quad (22)$$

For  $\lambda_1 \sim \lambda_3 \sim 10^{-8}$  cm,  $\lambda_2 \sim 10^{-4}$  cm,  $N/V = 10^{21}$  cm<sup>-3</sup>,  $T_{21} \sim 10^{-4}$  sec, and  $\Gamma_2 \sim 10^{10}$  sec<sup>-1</sup>, we have  $|\tilde{\chi}_n| \approx 10^{-9}$  cgs esu and  $\beta_1/\Gamma_{2k} \approx 10^{-18}$  cgs esu<sup>2</sup>, whereas, for  $\lambda_1 \sim \lambda_3 \sim 10^{-9}$  cm, we have  $|\tilde{\chi}_n| \approx 10^{-11}$  cgs esu and  $\beta_1/\Gamma_{2k} \approx 10^{-22}$  cgs esu<sup>2</sup>. The typical value of the resonance absorption coefficient in a host material with a moderate atomic number is  $\delta_p \sim 1$  cm<sup>-1</sup>.<sup>[11]</sup> Exponential  $\gamma$ -amplification is then possible for a narrow-band  $\gamma$ -pump if the growth rate of the optical system is  $-\delta_2 > \delta_p$ , which can be achieved for lasers or Q-switched optical amplifiers. The transformation coefficient  $K_k(z)$  given by (8) for  $\lambda_1 \sim \lambda_3 \sim 10^{-8}$  cm,  $\delta_1 \leq 10^2$  cm<sup>-1</sup> (concentration of nonlinear Mössbauer atoms  $\sim 1\%$ ),  $\lambda_2 \sim 0.6$   $\mu$ , and pump intensity  $\sim 1$  Ci is

$$K_k(z) \approx 10^{-22} (1 + 10^{-4} z^{-2}) / \Delta\Omega.$$

Under these conditions, for example, with  $-\delta_2 \approx 8$  cm<sup>-1</sup> (which, evidently, does not yet produce superradiance) and, in the case of an isotropic  $\gamma$ -source ( $\Delta\Omega = 4\pi$ ) and  $z \sim 7-8$  cm, the amplification is  $I_1/I_{10} \approx 10^2$ . Analysis of (22) then shows that, to increase  $\chi^{NL}$ , we can use, in addition to the well-known recombinations (increase in  $\lambda_n$ ,  $N/V$ ), the following interesting result: the product  $T_{21}^{1/2} \Gamma_2$  must be reduced and the increase in  $\Gamma_2^{-1}$  gives a more important effect than the use of transitions with lower  $T_{21}$ . This result is essentially different from the requirement of minimum  $\Gamma T$  in the theory of the "usual" laser,<sup>[11-51]</sup> and this product cannot, of course, be less than unity. The reduction in  $T_{21}^{1/2} \Gamma_2$  is not, at least in principle, limited provided  $d_{nm} \sim T_{nm}^{-1/2}$  and  $\Gamma_2 > \Gamma_3$ . In the ideal limit with  $\Gamma_2 = T_{21}^{-1}$ , we have  $T_{21}^{1/2} \Gamma_2 = 1/T_{21}^{1/2}$ , i. e., the above product decreases monotonically with increasing  $T_{21}$ , so that we can increase  $\chi^{NL}$  and hence rapidly increase the overall efficiency of transformation and amplification. When  $\Gamma_2 < \Gamma_3$ , analysis of (20) and (21) leads to a requirement for  $\Gamma_3$  to be reduced. The more efficient transformation then corresponds to pump frequency  $\omega_1 = \omega_3 - \omega_2$  (21). We note that, when the inversion ( $\rho_{22} - \rho_{11}$ ) is increased, pumping at frequency  $\omega_1$  is also more effective.

The above brief discussion of the problem of nonlinear susceptibilities in the  $\gamma$  band shows that it is, in principle, possible to achieve  $\chi^{NL} \sim 10^{-5}$  cgs esu (by reducing the luminescence linewidth<sup>[21]</sup>). The corresponding nonlinear transformation coefficient for an isotropic source of  $\sim 1$  Ci is  $K_k \sim 10^{-14}$  and the amplification length is  $l \sim 4-5$  cm. When a more anisotropic pump is employed ( $\Delta\Omega \sim 10^5$ ), the amplification length is  $l \sim 3$  cm.

It is important to note that the increase in  $\chi^{NL}$ , the attainment of the highest possible growth rates for the



idler wave, and spatial concentration of the pump radiation make the problem physically more realistic and rigorous because the achievement of high  $\gamma$  amplification over small thicknesses justifies the above approximation of given amplitude (which should not be too low) and the validity of the classical equations in the present context, which may not be correct in the case of large thicknesses for the real pump-wave absorption coefficients  $\delta_p$ .

In the case of the noise pump, the growth rate of the  $\gamma$  wave is described by (14a). Since, in the case of the Mössbauer  $\gamma$  rays and the usual solid-state medium  $\Gamma_2 \gg \Gamma_{2k} = \Gamma_3$ , the true growth rate for pumping at frequency  $\omega_3$  is

$$\delta_{ik} = \delta_{i0} - \Gamma_3 \operatorname{Re}(\epsilon_{20}^{1/2} - \epsilon_{30}^{1/2})/c,$$

i. e., amplification is possible if

$$\Gamma_2 \operatorname{Re}(\epsilon_{20}^{1/2} - \epsilon_{30}^{1/2})/c < 1 \text{ cm}^{-1}$$

which can always be satisfied for Mössbauer radiation. When the pump frequency is  $\omega_1$ , the quantity  $\Gamma_{2k} = \Gamma_3 + \Gamma_2$  and the maximum growth rate are lower by a factor of two as compared with the last case, and the following condition must be satisfied:

$$(\Gamma_2 + \Gamma_3) \operatorname{Re}(\epsilon_{20}^{1/2} - \epsilon_{30}^{1/2})/c < 1 \text{ cm}^{-1} .$$

We note in conclusion that an increase by several orders of magnitude in  $\chi_1 \sim T_{21}^{-1/2}$  and  $\chi_2 \sim T_{21}^{-1/2} \Gamma_2^{-1}$  is also possible in the x-ray region.

The above analysis of coherent nonthreshold parametric amplification shows that the problem of the  $\gamma$  laser (amplifier) has a realistic solution in the Mössbauer range and that this solution provides reasonable criteria for the parameters of the problem in the x-ray region. The models that we have considered, and the numerical estimates that we have quoted, show that there is a consistent set of nuclear and electron parameters that will ensure a real exponential growth of the wave in a host with nonlinear, optically inverted, Mössbauer atoms. Analysis of various types of spatial and frequency statistics of high-frequency pumping shows that the amplification conditions are satisfied in a broad range (from the monochromatic and plane to the delta-correlated

waves) with a  $\gamma$ -wave growth rate that is not very dependent on the type of statistics.

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