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Interaction of weak pulses with a low-frequency high-intensity wave in a dispersive medium

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We consider three-frequency nonstationary interaction of waves in a quadratically nonlinear medium. The low-frequency high-initial-intensity pump wave is not subject to decay instability, so that the nonlinear interaction regime can be described in the given-pump-field approximation. The cases of excitation of a wave at the sum-frequency by a long pump pulse and a short signal pulse, and the converse situation, are discussed. Analytic and numerical methods are used. Effects of nonlinear disperse spreading are described, as is also the breakup of the excited pulse into subpulses.

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1. INTRODUCTION

The study of synchronous (resonant) interactions in dispersive media plays a fundamental role in various branches of physics, such as plasma physics, nonlinear optics, or hydrodynamics. In the last decade, much progress was made in the development of the theory of nonstationary interactions of modulated waves (wave packets) (see, e.g., Refs. 1–3). The most advanced is the description of the interaction of pulses in first-order approximation of dispersion theory, which takes into account the difference between the group velocities. The method of solving the inverse scattering problem yielded in this case a general analytic solution of the system of three equations for the complex amplitudes.[3] However, by virtue of the complicated form of the solution[3] at arbitrary boundary (or initial) conditions, it cannot always be used in the analysis of the concrete situations. Therefore, in addition to the approach developed by Belavin and Zakharov,[3] use is made also of other methods of solving the equations (for example, the given-field method, or asymptotic methods), and the numerical experiments are used more and more extensively.

Until recently, most attention was paid in the theory of nonlinear three-frequency interactions of pulses to the analysis of parametric processes (decay instability)[4,5] and second-harmonic generation by short pulses.[6-8] The nonstationary interaction of another type, wherein a high-intensity low-frequency wave (pump) is mixed with a weak signal of another frequency, resulting in production of a wave at the sum or difference frequency, remain practically uninvestigated. Wave generation at difference and sum frequencies plays an important role in nonlinear optics.[9] To describe the excitation of picosecond and subpicosecond pulses it is necessary to develop a nonstationary theory that takes into account the specifics of this problem. Of principal nontrivial interest in this case is the development of a theory of the nonlinear frequency-conversion regimes. In the present article we have attempted to fill this gap.

We consider nonstationary interaction of three pulses propagating in a general case with different group velocities. Since the powerful pump pulse is not subject to decay instability, the nonlinear frequency-conversion regimes are well described in the given-pump-field approximation. We discuss in the paper the physics of the interaction of a short signal pulse with a long pump

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pulse and vice versa. By combining analytic methods with numerical experiments we were able to draw the complete picture of nonstationary excitation of a short pulse at the sum (difference) frequency. The article reports such effects as nonlinear-disperse spreading and the breakup of the excited pulse into subpulses.

2. SHORTENED EQUATIONS. SOLUTION METHODS

We consider the interaction of three modulated waves with average frequencies $\omega_3 = \omega_1 + \omega_2$ in a medium with nonlinearity. In first-order approximation of dispersion theory, the slow changes of the complex amplitudes of the waves $A_j(t,z)$ are described by the reduced equations[1-3]

$$\frac{\partial A_1}{\partial z} = -i\gamma_1 A_3 A_2^* e^{-i\Delta kz}, \tag{1}$$

$$\frac{\partial A_2}{\partial z} + \nu_{21}\frac{\partial A_2}{\partial \eta_1} = -i\gamma_2 A_3 A_1^* e^{-i\Delta kz}, \tag{2}$$

$$\frac{\partial A_3}{\partial z} + \nu_{31}\frac{\partial A_3}{\partial \eta_1} = -i\gamma_3 A_1 A_2 e^{i\Delta kz}, \tag{3}$$

where $\eta_j = t - z/u_j$, $\nu_{j1} = u_j^{-1} - u_1^{-1}$, z is the coordinate, t is the time, u_j is the group velocity, γ_j is the nonlinearity coefficient, and $\Delta k = k_3 - k_1 - k_2$.

At $z=0$, we specify the boundary conditions:

$$A_j(t,\ 0) = E_j(t). \tag{4}$$

We are interested in the case when the low-frequency pump wave, say of frequency ω_1, has an intensity greatly exceeding the intensities of the two other waves:

$$\max|E_1(t)| \gg \max|E_{2,3}(t)|. \tag{5}$$

To simplify the exposition that follows it is advantageous to set the initial amplitude of one of the weak waves equal to zero. We consider the case of generation of a wave at the sum frequency ω_3, putting

$$A_3(t,\ 0) = E_3(t) = 0. \tag{6}$$

The excitation of a wave at the difference frequency ω_2, when $E_2(t) = 0$, is analyzed by the same procedure.

Equations (1)–(3) with boundary conditions (4)–(6) were solved by us for Gaussian pulses $E_j = E_{0j}\exp(-t^2/\tau_j^2)$ by numerical methods[10]; the results are shown below (see Figs. 2 and 4) in the form of plots. At the same time, taking into account the specifics of the problem, we can propose effective approximate analytic methods of solving the system (1)–(3). In fact, at the chosen relations (5) between the initial amplitudes no decay (parametric) instability of the pump wave develops in the medium, i.e., the nonlinear distortions of the envelope of the high-intensity pulse of frequency ω_1 are quite negligible, $A_1(t,z) \approx E_1(\eta_1)$. In other words, it can be assumed with high degree of accuracy that the right-hand side of (1) is equal to zero, so that the initial system of equations can be linearized:

$$\frac{\partial A_2}{\partial z} + \nu_{21}\frac{\partial A_2}{\partial \eta_1} = -i\gamma_2 E_1^*(\eta_1) A_3 e^{-i\Delta kz}, \tag{7}$$

$$\frac{\partial A_3}{\partial z} + \nu_{31}\frac{\partial A_3}{\partial \eta_1} = -i\gamma_3 E_1(\eta_1) A_2 e^{i\Delta kz}. \tag{8}$$

The solution of Eqs. (7), (8) can be written in integral form[11-13]:

$$A_3(\eta_3, z) = -i\gamma_3 \int_0^z dy\, E_2(\eta_3 + \nu_{32}y) E_1(\eta_3 + \nu_{31}y) \bar{R} e^{i\Delta ky}, \tag{9}$$

where R is the normalized Riemann function (see the Appendix). The concrete form of the Riemann function depends on the amplitude–phase modulation of the pump $E_1(t)$ and on the ratio of the detunings of the group velocities ν_{21} and ν_{31} (see the Appendix). At the upper and lower limits of integration at $y=0$ and $y=z$ we have $\bar{R}=1$. We note that the linear frequency conversion regime ($\gamma_2 = 0$) corresponds to $\bar{R} \equiv 1$.

With the aid of (9) it is particularly convenient to analyze the conversion of the frequency of an ultrashort delta pulse in the field of a given pump pulse, when the dispersion effects (or the group-delay effects) begin to manifest themselves ahead of the nonlinear ones (see Sec. 3.1). If the ratio of the nonlinear and dispersion effects is reversed, it is better to use the diffusion approximation (see Sec. 3.2). On the other hand, if the pump pulse has the shortest duration, then in the case of group-velocity mismatch in all the waves, many features of the process of the nonlinear interaction are described by stationary solutions of the system (7), (8) (see 4.2) and in the case of group synchronism with the pump they are described by asymptotic expressions that follow from (9) (see Sec. 4.2).

We consider next typical cases of interaction of pulses with different durations.

3. QUASICONTINUOUS PUMP WAVE AND SHORT SIGNAL PULSE

Let the initial signal $E_2(t)$ have a duration τ_2. As the result of the group-delay effects, the durations of the weak waves $A_2(t)$ and $A_3(t)$ can increase over the distance z to a value $\tau_2 + |\nu_{21}|z$ [this follows from a simple analysis of Eqs. (1)–(3) in terms of the characteristic variables]. If the complex amplitude of the pump wave $E_1(t)$ does not change significantly over the time interval $\tau_2 + |\nu_{21}|z$, then it can be regarded formally as constant when Eqs. (7) and (8) are integrated. In the absence of nonstationary effects in the pump wave, the Riemann function is expressed in terms of the Bessel function of zeroth order $J_0(x)$ [see (A.1)] and the solution (9) can be written in the form

$$A_3 = -i\gamma_3 E_1(\eta_1) \int_0^z dy\, E_2(\eta_3 + \nu_{32}y) J_0\left\{\frac{2}{l_{nl}}(y(z-y))^{1/2}\right\} \tag{10}$$

Here $l_{nl} = (\gamma_2\gamma_3 E_{10}^2)^{-1/2}$ is the nonlinear length.

In (10) the nonlinear effects due to the reaction of the excited wave A_2 on the input signal are described in terms of a Bessel function and manifest themselves over lengths $z \gtrsim l_{nl}$, while the nonstationary effects due to detuning of the group velocities of the weak waves ν_{32} are described in terms of the initial amplitude of the input signal with retarded argument $E_2(\eta_3 + \nu_{32}y)$ and manifest themselves over distances that exceed the group-delay length $l_{\nu 32} = \tau_2/|\nu_{32}|$. Depending on the ratio of the characteristic lengths l_{nl} and $l_{\nu 32}$ we can separate six different wave-interaction regimes (Fig. 1).

3.1. Weakly nonlinear process of frequency conversion

This case is characterized by the fact that the nonstationary effects begin to manifest themselves ahead of

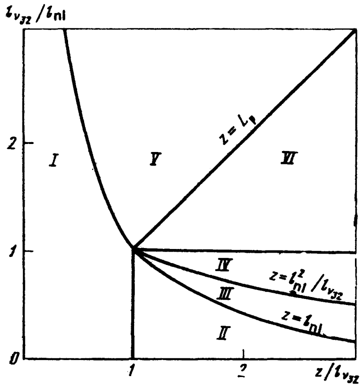

FIG. 1. Regions of existence of different frequency-conversion regime in interaction of a short signal pulse and a long pump pulse. I and II—linear regimes, III–IV—nonlinear, I and V—quasistationary, II–IV and VI—nonstationary.

the nonlinear ones, $l_{\nu 32} < l_{nl}$. In the case of a weakly nonlinear interaction we can separate the following frequency-conversion regimes.

I. The regime $z < l_{\nu 32} < l_{nl}$–the well known quasistationary linear regime,[1] for which we put in (10) $J_0 \approx 1$ and $E_2(\eta_3 + \nu_{32} y) \approx E_2(\eta_3)$. Taking this into account we have

$$A_3 = -i\gamma_3 z E_1(\eta_1) E_2(\eta_3), \tag{11}$$

i.e., the waveform of the pulse at the sum frequency ω_3 duplicates the waveform of the initial signal of frequency ω_2, while the amplitude increases in proportion to the distance.

II. The regime $l_{\nu 32} < z < l_{nl}$ is likewise a well known nonstationary linear regime,[1] in the description of which we can, putting $J_0 \approx 1$ in (10), obtain the equation

$$A_3 = -i\gamma_3 E_1(\eta_1) \int_0^z dy\, E_2(\eta_3 + \nu_{32} y). \tag{12}$$

Over lengths exceeding the group length $z > l_{\nu 32}$, the sum-frequency pulse acquires an almost rectangular form; its duration increases like $\tau_3(z) \approx |\nu_{32}| z$, and the peak amplitude saturates:

$$\max|A_3| = \frac{\gamma_3 E_{10}}{|\nu_{32}|} \int_{-\infty}^{\infty} E_2(y)\, dy.$$

Since $\tau_3(z) > \tau_2$, the input pulse $E_2(t)$ can be regarded as a delta pulse. It is interesting to note that if

$$\int_{-\infty}^{\infty} E_2(y)\, dy = 0,$$

then the pulse at the sum frequency is not excited effectively in this regime.

III. The regime $l_{nl} < z < l_{nl}^2 / l_{\nu 32}$–the first stage of nonstationary weakly nonlinear interaction.[13] In this region, the input signal can be regarded, as before, as a delta function in the integrand of (10), so that the Bessel function can be taken outside the integral sign at the value of the argument $y = -\eta_3/\nu_{32}$. Therefore the envelope of the pulse at the summary frequency is described by a Bessel function

$$A_3 = -i\frac{\gamma_3}{|\nu_{32}|} E_1(\eta_1) J_0(x) \int_{-\infty}^{\infty} dy\, E_2(y), \quad x = \frac{2(-\eta_1\eta_3)^{1/2}}{|\nu_{32}| l_{nl}}. \tag{13}$$

The argument of the Bessel function has a maximum value $x_{max} = z/l_{nl}$ at the center of the pulse at $\eta_1 = \eta_c = (\nu_{21} + \nu_{31}) z/2$ and is equal to zero at the edges of the pulses at $\eta_1 = \nu_{31} z$ and $\eta_1 = \nu_{21} z$. It is seen thus that a distance $z \gtrsim l_{nl}$ the rectangular shape of the pulse A_3 is distorted.

During the initial stage of these distortions we can expand the Bessel function in a series

$$A_3 = -i\frac{\gamma_3}{|\nu_{32}|} E_1(\eta_1) \left\{1 - \frac{z^2}{l_{nl}^2} + \frac{4(\eta_1 - \eta_c)^2}{\nu_{32}^2 l_{nl}^2}\right\} \int_{-\infty}^{\infty} E_2(y)\, dy. \tag{14}$$

It follows from (14) that when the nonlinear regime is reached an intensity dip is produced at the center of the pulse $\eta_1 = \eta_c$, where $A_3 \sim (1 - z^2/l_{nl}^2)$. Next, at $z \gtrsim l_{nl}$, the distortions become stronger, the pulse breaks up into subpulses, and on the edges (on the leading and trailing edges, $\eta_3 = 0$ and $\eta_3 = \nu_{32} z$) two principal subpulses of largest amplitude are formed.

The number of subpulses increases as the wave propagates and the duration of the principal subpulses decreases. When the length of the latter becomes comparable with the initial duration of the input signal τ_2 (this takes place over a length $z \sim l_{nl}^2/l_{\nu 32}$), the second stage of the nonstationary weakly nonlinear interaction sets in.

IV. The regime $z > l_{nl}^2/l_{\nu 32}$. During the second stage of the weakly nonlinear regime, the Bessel function in the integrand of (10) varies more rapidly at the edges of the pulse A_3 than the initial amplitude E_2 of the input signal, as a result of which the envelope of the pulse at the sum frequency assumes a complicated irregular shape.

3.2. Nonlinearly disperse spreading

Assume that in the course of the wave interaction the nonlinear effects that manifest themselves ahead of the dispersion effects, $l_{nl} < l_{\nu 32}$ (Fig. 1). Although the effects produced thereby are described as before by Eq. (10), the analysis of this equation becomes difficult. It is expedient here to use the method of slowly varying amplitude. We reduce Eqs. (7) and (8) to a single equation (the pump amplitude A_1 will be assumed constant as before):

$$\frac{\partial^2 A_3}{\partial z^2} - \frac{\nu_{32}^2}{4}\frac{\partial^2 A_3}{\partial \eta_{av}^2} + \frac{A_3}{l_{nl}^2} = 0,$$
$$\eta_{av} = {}^1/_2(\nu_{21} + \nu_{31}) z + \eta_1 = {}^1/_2(\eta_2 + \eta_3). \tag{15}$$

We seek the solution of (15) in the form

$$A_3 = C_+(\eta_{av}, z)\exp(iz/l_{nl}) + C_-(\eta_{av}, z)\exp(-iz/l_{nl}). \tag{16}$$

In the absence of dispersion effects, $\nu_{32} = 0$, the partial amplitudes do not depend on the distance transversed (the waveform of the pulse is preserved) and are determined completely from the boundary conditions: $C_+ = C_- = (\gamma_3/4\gamma_2)^{1/2} E_2(\eta_2)$. In the presence of group-velocity detunings ν_{32}, the amplitude profiles $C_\pm$ become distorted. Since we assume the dispersion effects to be weaker than the nonlinear effects, we can assume that the amplitudes $C_\pm$ vary slowly compared with the exponential factors $\exp(\pm iz/l_{nl})$. This gives grounds for employing the known method of slowly varying amplitude.

Substituting (16) in (15) and discarding the second derivative $\partial^2 C_\pm/\partial z^2$, we obtain the parabolic equation

$$\partial C_\pm/\partial z = \mp iD\partial^2 C_\pm/\partial\eta_{av}^2,\quad D = {}^1/_8\nu_{32}^2 l_{nl}. \tag{17}$$

Obviously, the detunings ν_{32} of the group velocities lead to nonlinearly diffuse spreading of the pulse. The boundary conditions for the partial amplitudes are

$$C_+(\eta_1, 0) = C_-(\eta_1, 0) = (\gamma_3/4\gamma_2)^{1/2} E_2(\eta_1). \tag{18}$$

Solving the diffusion equation (17) with the boundary conditions (18), we obtain from (16) the general expression for the amplitude

$$A_3 = -i(\gamma_3/4\pi Dz\gamma_2)^{1/2}\int_{-\infty}^{\infty} dy\, E_2(y)\sin[z/l_{nl} - (y-\eta_{av})^2/4Dz]. \tag{19}$$

In particular, if a Gaussian pulse of frequency ω_2 is applied at the input to the medium:

$$E_2(t) = E_{20}\exp(-t^2/\tau_2^2), \tag{20}$$

then, integrating (19) and taking (20) into account, we obtain the waveform of the pulse excited at the frequency ω_3:

$$A_3 = -i(\gamma_3\tau_2/\gamma_2\tau_3(z))^{1/2}E_{20}\exp[-\eta_{av}^2/\tau_3^2(z)] \times \sin\{z/l_{nl} + z\eta_{av}^2/L_{spr}\tau_3^2(z) - \mathrm{arctg}(z/L_{spr})\}. \tag{21}$$

The duration of the generated pulse increases here with distance:

$$\tau_3(z) = \tau_2(1 + z^2/L_{spr}^2)^{1/2}, \tag{22}$$

where the length of the nonlineraly disperse spreading is equal to

$$L_{spr} = 2l_{\nu 32}^2/l_{nl}. \tag{23}$$

Equations (19) and (21) describe the excitation of the pulse F_3 in the regions I, V, and VI (Fig. 1). A consequence of the strong nonlinear interactions is the partial suppression of the dispersion spreading of the excited pulse: first, broadening of the pulse A_3 begins not at the group length $z = l_{\nu 32}$, as in the linear regime II, but later, at the length $L_{spr} > l_{\nu 32}$; second, the nonlinearly dispersed spreading is slower than in the linear regime II [see (12)], where $\tau_3 \approx \tau_2 z/l_{\nu 32}$ [cf. (22)]. Thus, the group delay effects with respect to the weak waves become substantially weaker in the strong pump field.

The waveforms of the generated pulse during different stages of the conversion are shown in Fig. 2.

4. SHORT PUMP-WAVE PULSE

Assume that a short pulse pump and a long signal pulse $E_2(t) \approx$ const. are applied to the input of the nonlinear medium. Now the nonstationary effects are characterized by two group lengths: $l_{\nu 31} = \tau_1/|\nu_{31}|$ and $l_{\nu 21} = \tau_1/|\nu_{21}|$, while the nonlinear effects are characterized as before by the nonlinear length $l_{nl} = (\gamma_2\gamma_3 E_{10}^2)^{-1/2}$ (E_{10} is the peak amplitude of the pump pulse). Depending on the ratio of these lengths we can distinguish between six frequency-conversion regimes (see Fig. 3), where we have put for the sake of argument $l_{\nu 21} > l_{\nu 31}$. The classification of the regimes at $l_{\nu 31} < l_{\nu 21}$ or at equal group lengths $l_{\nu 31} = l_{\nu 21}$ or in the case of group synchronism with the pump $l_{\nu 31}, l_{\nu 21} \to \infty$, can be easily obtained in analogy with Fig. 3.

The waveforms of the sum-frequency pulse A_3 at the different conversion stages, observed in the numerical experiments, are shown in Fig. 4.

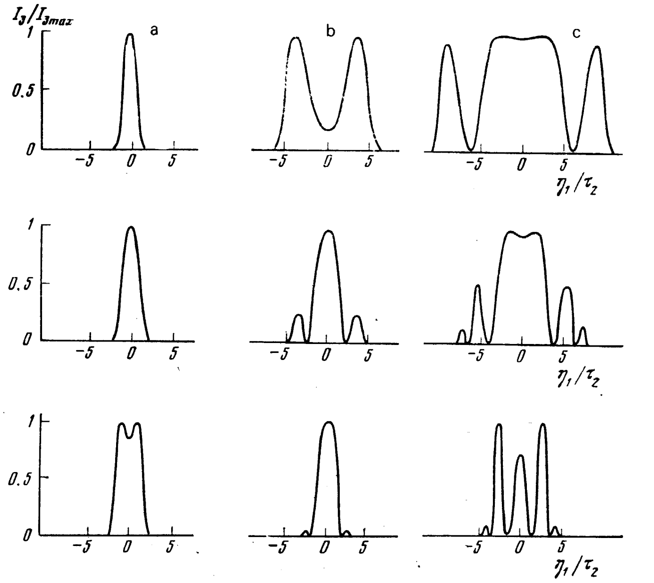

FIG. 2. Results of numerical experiments for an input signal pulse of Gaussian shape $E_2 = E_{20}\exp(-t^2/\tau_2^2)$ and for a monochromatic pump wave $E_1 = E_{10} =$ const. The distortion of the waveform of the exciting pulse as it propagates in the nonlinear medium is shown ($z/l_{\nu 32} = 2$—a, 10—b, 20—c) at various levels of the pump amplitude ($l_{\nu 32}/l_{nl} = 0.2$—upper row, 1—middle, 3—low row).

4.1. Linear frequency-conversion regimes

In the linear regimes I–III, which correspond to a Riemann function $R = 1$, the waveform of the input signal A_2 is not distorted, and the amplitude of the excited pulse is [see (9) and cf. (12)]

$$A_3 = -i\gamma_3 E_2\int_0^z dy\, E_3(\eta_3 - \nu_{31}y). \tag{24}$$

I. The regime $z < l_{\nu 31} < l_{\nu 21}$ is quasistationary; the group-delay effect does not manifest itself, and the waveform of the excited pulse duplicates the waveform

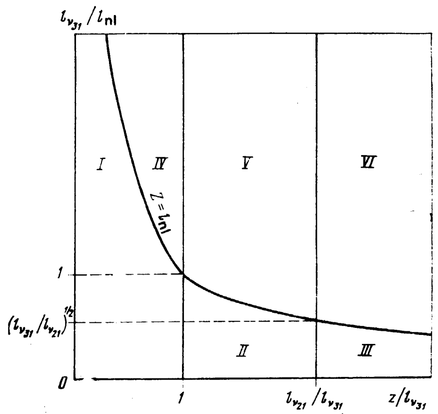

FIG. 3. Regions of existence of different regimes of frequency conversion in the interaction of a short pump pulse and a long signal pulse. I–III—linear regimes, IV–VI—nonlinear, I, IV—quasistationary, II, III, V, VI—nonstationary. It is assumed that $l_{\nu 21} > l_{\nu 31}$.

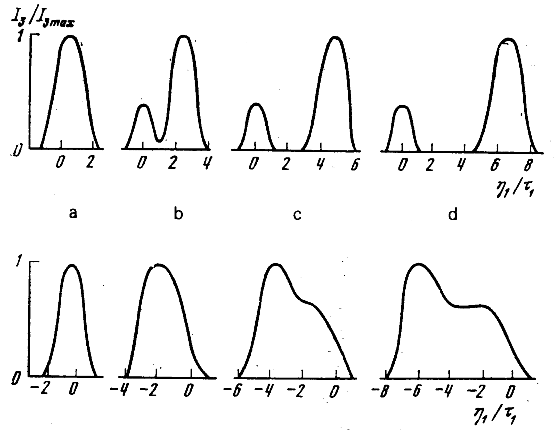

FIG. 4. Result of numerical experiments for a short Gaussian pump pulse $E_1=E_{10}\exp(-t^2/\tau_1^2)$ and a monochromatic input signal $E_2=E_{20}=\text{const}$. The distortion of the waveform of the excited pulse in the course of its propagation is shown ($z/l_{\nu31}$ = 1—a, 3—b, 5—c, 7—d) at different levels of the peak pump amplitude and at different relations, between the group velocities ($l_{\nu31}/l_{nl}=1.25$, $l_{\nu31}/l_{\nu21}=0.5$, $\nu_{21}\nu_{31}>0$—upper row and $l_{\nu31}/l_{nl}=0.71$, $l_{\nu31}=l_{\nu21}$, $\nu_{21}\nu_{31}<0$—lower row).

of the pump pulse $A_3\approx -i\gamma_3E_2E_3(\eta_3)z$ [cf. (11)]. If the pump intensity is large, $l_{nl}<l_{\nu31}$, then at a distance $z=l_{nl}$ the linear regime I goes over into the nonlinear quasistationary regime IV. At lower intensities, $l_{nl}>l_{\nu31}$, the conversion regime remains linear with increasing z, but beyond the group lengths $z>l_{\nu31}$ the process of wave interaction becomes nonstationary.

II. The regime $l_{\nu31}<z<l_{\nu21}$, $l_{nl}>l_{\nu31}$ is nonstationary and is due to the influence of only one of the detunings of the group velocities (in our case, ν_{31}). In this region, the pulse A_3 acquires a rectangular shape with amplitude

$$A_3\sim(\gamma_3E_2/\nu_{31})\int_{-\infty}^{\infty}dy\,E_1(y)$$

and with duration $\tau_3\sim|\nu_{31}|z$ that increases with the distance [cf. (12)]. Since the group dispersion effects are stronger than the nonlinear ones, $l_{\nu31}<l_{nl}$, the latter can become substantially weakened or become completely suppressed. At moderate intensities $l_{\nu31}<l_{nl}<(l_{\nu31}l_{\nu21})^{1/2}$ the linear nonstationary regime II gives way to the linear regime V, but this does not occur over the nonlinear length (as in the transition from I to IV), but at a larger distance $z\approx l_{nl}^2/l_{\nu31}$. If the amplitude of the pump pulse is relatively small, $l_{nl}>(l_{\nu31}l_{\nu21})^{1/2}$, then the dispersion group effects suppress the nonlinear interaction to such an extent that in region III, which follows region II, the linear process of frequency mixing continues at all distances (no reaction by A_3 on A_2 appears).

III. The regime $z>l_{\nu21}>l_{\nu31}$ is nonstationary; the pulse A_3 continues to broaden, $\tau_3\sim|\nu_{31}|z$, but its edges acquire stationary forms.

4.2. Nonlinear conversion regimes

When the pump pulse intensity exceeds a certain threshold, $l_{nl}<(l_{\nu31}l_{\nu21})^{1/2}$, the linear regimes I and II go over at a definite distance into the nonlinear regimes IV and VI, which are characterized by the fact that the waveform of the input signal A_2 is distorted by the opposing reaction of the excited pulse A_3. In regions IV–VI the Riemann functions cannot be set identically equal to unity, and its concrete form must be taken into account. We examine now the characteristic features of the nonlinear regimes.

IV. The regime $l_{nl}<z<l_{\nu31}$ is quasistationary. The group velocities of all three waves can be assumed equal: $\nu_{21}'=\nu_{31}=0, \eta_1=\eta_2=\eta_3$. In this approximation it is easy to obtain directly from (7) and (8) the solution

$$A_3=-i(\gamma_3/\gamma_2)^{1/2}E_2e^{i\varphi_2}\sin((\gamma_2\gamma_3)^{1/2}|E_1(\eta_1)|z). \qquad (25)$$

The amplitude A_3 begins to oscillate with distance at $z>l_{nl}$, and if the pump pulse $E_1(\eta_1)$ has a waveform other than homogeneous and rectangular, for example Gaussian, then the excited pulse A_3 breaks up into subpulses. The total duration τ_3 does not exceed the duration τ_1 of the pump pulse.

V. The regime $l_{\nu31}<z<l_{\nu32}, l_{nl}<(zl_{\nu31})^{1/2}$ is nonstationary in the case of group synchronism for one of the weak waves ($\nu_{31}=0$ in the situation of Fig. 3). The Riemann function is expressed in terms of a Bessel function [see (A.2)]. In the case of group synchronism of the excited pulse with the pump, $\nu_{31}=0$, we have

$$A_3=-i\gamma_3E_1(\eta_1)E_2\int_0^z dy\,J_0\left\{2\left[\gamma_2\gamma_3(z-y)\int_0^y|E_1(\eta_1-\nu_{21}y_1)|^2dy_1\right]^{1/2}\right\}. \qquad (26)$$

The pulse E_3 does not broaden but in contrast of the complete synchronism case (25) it is always chopped up under nonlinear frequency conversion even in the field of a rectangular homogeneous pump pulse.

Beyond the group length $z>l_{\nu21}$, the waveform of the pulse is described by an asymptotic expression that follows from (26):

$$A_3=-i\gamma_3E_1(\eta_3)E_2zJ_1(g)/g, \quad g=2\left[\frac{\gamma_2\gamma_3z}{|\nu_{21}|}\int_{-\infty}^{\eta}dy|E_1(y)|^2\right]^{1/2}.$$

In the region V the argument $g>1$ and a nonlinear conversion regime sets in, in the course of which the pulse A_3 acquires a choppy asymmetrical form. The largest amplitude takes place on that front which is overtaken by the long pulse of the input signal. We have observed this picture qualitatively in the numerical experiments.

If group synchronism is obtained between the signal wave and the pump pulse, $\nu_{21}=0, u_1=u_2, \eta_1=\eta_2$, then the pulse A_3 in the region V broadens as a result of the group-delay effect, and becomes strongly chopped up as a result of the nonlinear character of the conversion.

VI. The regime $z>l_{\nu31}, l_{\nu21}, l_{nl}<(l_{\nu31}l_{\nu21})^{1/2}$. Qualitatively, the picture of the interaction of the pulses is as follows. The leading (trailing) edge of the excited pulse A_3 moves with velocity $u_3>u_1$ ($u_3<u_1$), and the other edge with the pump velocity u_1; the total pulse duration ν_3 increases with distance. The leading (trailing) part of the pulse duration $\Delta\tau=\tau_1|\nu_{32}|/|\nu_{21}|$, which propagates with velocity u_3, is excited over the initial distances $z<l_{\nu21}$ and takes on the waveform typical of regime V. The succeeding part of the pulse A_3 is excited beyond the group lengths $z>l_{\nu31}, l_{\nu21}$ in the steady state, i.e., the part of the pulse adjacent to the pump pulse has a stationary form.

It is convenient to calculate the stationary part of the pulse A_3 directly from (7) and (8), putting $\partial A_j/\partial z=0$. Depending on the ratios of the group velocities of the three pulses, one must distinguish here between two regimes: $\nu_{21}\nu_{31}>0$ and $\nu_{21}\nu_{31}<0$. We consider them separately.

In the regime $\nu_{21}\nu_{31}>0$, the pump pulse travels more rapidly ($u_1>u_2,u_3$) or more slowly ($u_1<u_2,u_3$) than the two weak waves; in the former case, in the calculation of the stationary profile it is necessary to specify the conditions $A_3(\eta_1=\infty)=0, A_2(\eta_1=\infty)=E_2$, while in the latter case, the conditions are $A_3(\eta_1=-\infty)=0, A_2(\eta_1=-\infty)=E_2$. As a result of simple calculations we obtain ($u_1<u_2,u_3$):

$$A_3=-i(\gamma_3|\nu_{31}|/\gamma_2|\nu_{21}|)^{1/2}E_2\sin G;$$
$$G(\eta_1)=(\gamma_2\gamma_3/|\nu_{31}\nu_{21}|)^{1/2}\int_{-\infty}^{\eta_1}dy\,E_1(y). \qquad (27)$$

In the case $u_1>u_2,u_3$ it is necessary to reverse the sign of the lower limit of the integration.

It follows from an analysis of (27) that if $G(\infty)=n\pi$ (n is an integer), then the amplitude A_3 outside the pump pulse, $\eta_1>\tau_1$, is equal to zero—the excited pulse is blocked, so to speak, inside the pump pulse (to be sure, A_3 has a trailing edge $\Delta\tau=\tau_1|\nu_{31}|/|\nu_{21}|$ that is excited in the region V). The energy of such a pulse does not increase with increasing z. On the other hand if $G(\infty)=(2n+1)\pi/2$, then the amplitude A_3 has outside the pump pulse a constant maximum values that does not depend on the intensity $|A_1|^2$, $|A_3|=(\gamma_3|\nu_{31}|/\gamma_2|\nu_{21}|)^{1/2}E_2$. The pulse has a practically rectangular waveform (with complicated fronts); the duration of the pulse and the energy increase in proportion to the length z.

Thus, in the nonlinear regime VI, if the dispersion of the group velocities is such that $\nu_{21}\nu_{31}>0$, it is possible to alter radically the waveform of the excited pulse and its energy, by varying the pump intensity [the parameter $G(\infty)$].

We note that in the particular case $\nu_{21}=\nu_{31}, u_2=u_3$ there exists a simple exact solution of Eqs. (7) and (8):

$$A_3=-i\left(\frac{\gamma_3}{\gamma_2}\right)^{1/2}E_2\sin\left[\frac{(\gamma_2\gamma_3)^{1/2}}{|\nu_{31}|}\int_{\eta_1-\nu_{31}z}^{\eta_1}|E_1(y)|dy\right]. \qquad (28)$$

In the region VI we have $z>l_{\nu 21}$ and $|\eta_1|<\tau_1 z$, and formula (28) goes over into (27). This, as well as numerical experiments, confirms the validity of the preceding statements that the exciting pulse has a stationary part.

In the regime $\nu_{21}\nu_{31}<0$, the pump pulse propagates with intermediate group velocity $u_2<u_1<u_3$ or $u_3<u_1<u_2$. In the former case stationary pulses correspond to the conditions $A_3(\eta_1=-\infty)=0$ and $A_2(\eta_1=\infty)=E_2$. The stationary pulses now have a qualitatively different waveform than in the regime $\nu_{21}\nu_{31}>0$, namely ($u_2<u_1<u_3$).

$$A_3=(\gamma_3|\nu_{31}|/\gamma_2|\nu_{21}|)^{1/2}E_2\,\mathrm{sh}\,G(\eta_1)/\mathrm{ch}\,G(\infty);$$
$$G=(\gamma_2\gamma_3/|\nu_{21}\nu_{31}|)^{1/2}\int_{-\infty}^{\eta_1}dy\,E_1(y). \qquad (29)$$

When the weak waves move away from the pump pulse in opposite directions after the interaction then, as seen from (29), the stationary part of the excited pulse A_3 has a smooth form [cf. the oscillatory regime at $\nu_{21}\nu_{31}>0$ (27)]. If $G(\infty)\gg 1$, then saturation of the stationary amplitude is observed:

$$|A_3|_{\mathrm{sat}}=(\gamma_3|\nu_{31}|/\gamma_2|\nu_{21}|)^{1/2}E_2.$$

The duration and energy of the pulse A_3 increase in proportion to the distance z.

APPENDIX

1. In the case of monochromatic pumping, $E_1(t)=E_{10}=\mathrm{const}$, and in the case of arbitrary group-velocity mismatches ν_{21} and ν_{31}, the Riemann function is expressed in terms of a Bessel function of zero order

$$\bar{R}=J_0\{2(\gamma_2\gamma_3)^{1/2}E_{10}(y(z-y))^{1/2}\}. \qquad (A.1)$$

2. In the case of arbitrary modulation of the pump and group synchronisms for the principal and excited waves, $u_1=u_3$ ($\nu_{31}=0$),

$$\bar{R}=J_0\left\{2\left[\gamma_2\gamma_3(z-y)\int_0^y dy_1|E_1(\eta_1-\nu_{21}y_1)|^2\right]^{1/2}\right\}. \qquad (A.2)$$

On the other hand, if group synchronism obtains between the pump and the single pulse, $u_1=u_2$ ($\nu_{21}=0$), then we have

$$\bar{R}=J_0\left\{2\left[\gamma_2\gamma_3(z-y)\int_0^y dy_1|E_1(\eta_1-\nu_{31}y_1)|^2\right]^{1/2}\right\}. \qquad (A.3)$$

3. For a bell-shaped pump pulse $E_1(T)=E_{10}\cosh^{-1}(t/\tau_1)$ and for arbitrary mismatches ν_{21} and ν_{31}, the Riemann function is expressed in terms of the Gauss hypergeometric function

$$\bar{R}=F(n,-n;1;y), \qquad (A.4)$$

where

$$n=(\gamma_2\gamma_3E_{10}^2\tau_1^2/\nu_{21}\nu_{31})^{1/2},$$
$$y=\mathrm{sh}(\nu_{21}y/\tau_1)\,\mathrm{sh}(\nu_{31}(z-y)/\tau_1)\,\mathrm{ch}^{-1}(\eta_1/\tau_1)\,\mathrm{ch}^{-1}((\eta_2-(\nu_{31}-\nu_{21})y)/\tau_1).$$

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Exciton luminescence on anthracene-gold boundary

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The luminescence spectra of thin anthracene single crystals in optical contact with quartz and gold were measured for the first time ever at low temperatures (1.7–100 K). It is shown that the presence of the metal layer leads to metallic quenching of the Frenkel excitons on the interface. Questions involved in the quenching of coherent and noncoherent excitons are discussed, as well as the peculiarities of the influence of a metallic substrate on the processes of exciton-phonon interaction in anthracene crystals.

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INTRODUCTION

A number of recent studies are devoted to the peculiarities in the behavior of excitons near the surface of the crystal, especially if the latter is in contact with another medium. For molecular crystals, such as anthracene, these investigations were carried out in Refs. 1–6. In addition to the singularities of exciton reflection and luminescence spectra, studies are made also of luminescence quenching, which is important for the determination of the behavior of Frenkel excitons on a free surface or in the case of contact between the crystal and another medium. The quenching of excitonic luminescence of a single crystal of anthracene was measured in Refs. 3 and 4 at room temperature as a function of the thickness of the dielectric interlayer between the crystal and the metal. It was concluded that the quenching is due to annihilation of the excitons into electrons and holes on the separation boundary.

Theoretical investigations of the behavior of Frenkel excitons on a boundary with a metal[1,5,6] show that the phenomena depend substantially on whether the excitons that come into play are coherent or noncoherent. Thus, for example, if the excitons are noncoherent then the presence of a metal can produce forces that drag the excitons to or from the metal, and metallic quenching of the excitons should be observed. To identify the type of excitons that manifest themselves in the spectra, it is necessary to carry out the measurements at various temperatures, especially at low temperatures, when the excitons can be coherent.[7] There are still no published reports of investigations of Frenkel-exciton quenching on an interface with a metal at low temperatures.

We report here the results of investigations of the characteristics of the luminescence spectra of thin anthracene single crystals which are freely mounted and in optical contact with quartz and gold, as functions of the temperature in the interval from 1.7 to 100 K. We prove experimentally, for the first time ever, the existence of metallic quenching of excitons on the anthracene-gold interface, and that the quenching depends substantially on the temperature, An analysis of the dependence of the parameters of the electron band of the exciton luminescence (position and half-width) on the temperature and on the type of the substrate has made it possible to identify the different effects of metallic and dielectric substrates on the exciton-phonon interaction in anthracene.

EXPERIMENT

The investigated anthracene single crystals were less than 1 μm thick and were grown by sublimation in an inert atmosphere from zone-purified (~100 zones) of material. No impurity bands whatever were observed in the luminescence spectra of these crystals at low temperatures. One part of the anthracene crystal was in optical contact with fused quartz and the other with a semitransparent gold electrode. For comparison, we investigated also freely supported single crystals of anthracene approximately 10 μm, with a gold semitransparent layer evaporated on half of their surface.

The luminescence spectra were measured in the temperature interval 1.7–100 K. Stabilization and monitoring of the temperature were by means of two germanium thermoresistors of the KGG type (which measured the temperature in the range from room to helium and below). The accuracy of the temperature determination was 0.05° in the interval 1.7–4.2 K and 0.1° in the interval 4.2–100 K. The spectra of the exciton luminescence of the anthracene were measured with a DFS-12 spectrometer. The luminescence was excited at an angle <10° to the crystal by a DRSh-250-2 mercury lamp through a UFS-6 filter (λ_m = 365 nm). The anthracene emission spectra were registered from the side of the free surface of the crystal, with account taken of the light lost in the various substrates on the other side of the crystal. Since the crystal thickness was less than

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