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Broadband Instability of Electromagnetic Waves in Nonlinear Media

Sergey Vlasov, Elena Kuposova and Alexey Babin
*Institute of Applied Physics, Russian Academy of Science
Russia*

1. Introduction

Wave propagation in nonlinear media is usually accompanied by variation of their space-time spectra (V.I. Bespalov & Talanov, 1966; Lighthill, 1965; Litvak & Talanov, 1967; Benjamin & Feir, 1967; Zakharov & Ostrovsky, 2009; Bejot et al., 2011). The character of this variation is frequently explained in terms of modulation instability which occurs, in particular, for electromagnetic waves in cubic media, where polarization is approximately equal to the cube of electric field intensity. Modulation instability manifests itself in partitioning of the originally uniform packet into separate beams and pulses. It has been studied in ample detail for the perturbations whose frequencies and propagation directions slightly differ from those of intense waves (pump waves), i.e., temporal and spatial spectra of the waves participating in the process are rather narrow (paraxial approximation) (V.I. Bespalov & Talanov, 1966; Litvak & Talanov, 1967; Agraval, 1995; Talanov & Vlasov, 1997). In this case, modulation instability is described by the nonlinear parabolic equation for a wave train envelope.

The instability is quite different, if the nonlinearity is higher than the third order of magnitude. It was shown in (Talanov & Vlasov, 1994; Kuposova & Vlasov, 2007) that the wave propagating in such a medium may be unstable relative to collinear perturbations at frequencies so high that not only wave packet envelope but the structure of each wave in the packet may change too. The parabolic equation does not hold for description of such phenomena; hence, the methods for solution of wave equations in a wide frequency band developed in (Talanov & Vlasov, 1995; Brabec & Krausz, 1997; V.G. Bespalov et al., 1999; Kolesic & Moloney, 2004; Ferrando et al., 2005; Kuposova et al., 2006) are employed. One of such methods, namely, the technique of pseudodifferential operators (Kuposova et al., 2006) is used in the current paper.

The first part of the paper that is an extension of (Talanov & Vlasov, 1994) is concerned with the instability of perturbation waves at combination frequencies noncollinear to pump for the case of nonlinear polarization represented as a polynomial of arbitrary but finite degree. Further, application of the theory to air, for which dielectric permittivity may be represented in the form of a polynomial, is addressed (Loriot et al., 2009). Finally, methods of finding amplitude and phase distribution of electric field in an ultrashort wave packet by analyzing signals from intensity autocorrelator traditionally used for measuring duration of ultrashort laser pulses are considered.

2. Plane wave instability in media with polynomial nonlinearity

Consider a linearly polarized wave packet propagating along the z -axis. We will make use of the equations for field E in this beam:

$$\frac{\partial E}{\partial z} + i \sqrt{-\frac{1}{c^2} \frac{\partial^2}{\partial t^2} n^2(-i \frac{\partial}{\partial t}) + \Delta_{\perp}} E = i \frac{1}{2 \sqrt{-\frac{1}{c^2} \frac{\partial^2}{\partial t^2} n^2(-i \frac{\partial}{\partial t}) + \Delta_{\perp}}} \frac{4\pi}{c^2} \frac{\partial^2 P_{NL}}{\partial t^2}, \quad (1)$$

obtained in (Koposova et al., 2006). In Eq. (1), $n(-i \frac{\partial}{\partial t})$ is the operator describing linear dispersion of the medium; for the processes $\sim \exp[i\omega t]$ stationary in time, $n(\omega)$ is the index of refraction at circular frequency ω ; Δ_{\perp} is Laplace operator in \vec{r}_{\perp} coordinates transverse to the propagation direction; P_{NL} is nonlinear polarization; and c is the velocity of light. Assuming the angular difference between the directions of the interacting waves to be small, we will neglect dispersion of the nonlinear polarization coefficient and the Laplace operator in the right-hand side of (1) and suppose that

$$\frac{1}{\sqrt{-\frac{1}{c^2} \frac{\partial^2}{\partial t^2} n^2(-i \frac{\partial}{\partial t}) + \Delta_{\perp}}} \approx \frac{c}{\omega \sqrt{\varepsilon(\omega)}},$$

where $\varepsilon(\omega) = n^2(\omega)$ is the dielectric permittivity at the same frequency.

We will describe the polarization by the polynomial dependence on the magnitude of electric field:

$$P_{NL} = \sum_{i=1}^S \chi^{(2N+1)} E^{2N+1}, \quad (2)$$

where $\chi^{(2N+1)}$ are $(2N+1)$ -th order susceptibilities, and $2S+1$ is the highest degree polarization taken into consideration.

To the first approximation of the asymptotic theory of nonlinear oscillations (Bogolyubov & Mitropol'skii, 1958), a steady-state plane wave

$$E(t, \vec{r}) = \frac{A_0 \exp[i\omega_0 t - ih_0 z] + A_0^* \exp[-i\omega_0 t + ih_0 z]}{2} \quad (3)$$

having amplitude A_0 , frequency ω_0 , and propagation constant $h_0 = k_0(1 + \frac{\delta\varepsilon_{NL}}{2})$ (where

$k_0 = \frac{\omega_0}{c} n(\omega_0)$ is the propagation constant of the wave of the same frequency in a linear medium and $\delta\varepsilon_{NL}$ is a nonlinear additive to permittivity) may propagate in the medium with permittivity

(2). The nonlinear additive is represented as a sum $\delta\varepsilon_{NL} = \sum_{N=1}^S \delta\varepsilon_{2N+1, NL}$ of nonlinear additives

$$\delta\varepsilon_{2N+1, NL} = \frac{4\pi}{\varepsilon(\omega_0)} \frac{(2N+1)!}{N!(N+1)! 2^{2N}} \chi^{(2N+1)} |A_0|^{2N} = \varepsilon'_{2N+1} |A_0|^{2N} \quad (4)$$

generated by each term of the series (2) and related to the nonlinearity of degree $2N + 1$, the nonlinearity coefficient ε'_{2N+1} in (4) is of the $(2N + 1) - th$ order of magnitude.

We will study stability of the solution relative to two-frequency perturbations of the form $u_1 \exp[i(\omega_1 t - hz) - i\vec{k}_\perp \vec{r}_\perp] + u_2 \exp[i(\omega_2 t - hz) + i\vec{k}_\perp \vec{r}_\perp]$, \vec{k}_\perp being their transverse wave number. Wave frequencies ω_1 and ω_2 and their propagation constant h will be found from solution of dispersion equations for perturbations.

We will seek solution of Eq. (1) in the form

$$E = \frac{A_0 \exp(i\varphi_0) + u_1 \exp[i\varphi_1 - i\vec{k}_\perp \vec{r}_\perp] + u_2 \exp[i\varphi_2 + i\vec{k}_\perp \vec{r}_\perp] + c.c.}{2}, \quad (5)$$

where $\varphi_0 = \omega_0 t - h_0 z$, $\varphi_1 = \omega_1 t - hz$, $\varphi_2 = \omega_2 t - hz$. Among the polarization terms with the perturbation of the first degree u_1 and u_2^* we will select the terms of identical degrees A_0 and A_0^* :

$$\sim |A_0|^{2N} u_1 \exp[i\varphi_1 - i\vec{k}_\perp \vec{r}_\perp] \quad (6)$$

and the terms whose difference of degrees A_0 and A_0^* is equal to the even integer $2M \leq 2N$:

$$\sim A_0^{N+M} A_0^{N-M} \exp[i(N+M)\varphi_0 - i(N-M)\varphi_0] u_2^* \exp[-i\varphi_2 - i\vec{k}_\perp \vec{r}_\perp]. \quad (7)$$

If the conditions

$$2M\omega_0 = \omega_1 + \omega_2, \quad Mh_0 = h \quad (8)$$

are fulfilled, for definite values of $\omega_{1,2}$ and h the terms (6) and (7) will be synchronous, i.e., they will have identical frequencies and propagation constants. Sets of frequencies $\omega_{1,2}$ and propagation constants h will be different for $M=1$, $M=2$, and so on. In other words, solution of the form (5) may have M branches, with the characteristics of branch M depending on nonlinear polarization terms with indices $M \leq N \leq S$. For $M=1$, we have perturbations near carrier frequency - the well-known modulation instability describing variations of the wave packet envelope that are slow compared to the carrier.

Making use of the equality (8) we will express the frequencies $\omega_1 = M\omega_0 + \Omega$, $\omega_2 = M\omega_0 - \Omega$ through their difference $\Omega = \frac{\omega_1 - \omega_2}{2}$ and frequency $M\omega_0$. Assuming that $k(\omega) = \frac{\omega n(\omega)}{c}$ is real in the frequency range under consideration, from Eq. (1) we obtain for the functions u_1 and u_2^* the following system of equations

$$\begin{aligned} \frac{\partial \{u_1\}}{\partial z} &= -i\beta_{M+} u_1 - i\gamma_{M+} u_2^* \\ \frac{\partial \{u_2^*\}}{\partial z} &= i\beta_{M-} u_2^* + i\gamma_{M-} u_1, \end{aligned} \quad (9)$$

where

$$\beta_{M\pm} = \sqrt{k^2(M\omega_0 \pm \Omega) - k_\perp^2} - Mh_0 + k_0 \frac{(M\omega_0 \pm \Omega)n(\omega_0)}{\omega_0 n(M\omega_0 \pm \Omega)} \sum_{N=1}^S \frac{N+1}{2} \delta \varepsilon_{2N+1, NL},$$

$$\gamma_{M\pm} = k_0 \frac{(M\omega_0 \pm \Omega)n(\omega_0)}{\omega_0 n(M\omega_0 \pm \Omega)} \frac{A_0^{\pm M}}{A_0^{*\pm M}} \sum_{N=M}^S \frac{\delta \varepsilon_{NL,2N+1} N!(N+1)!}{2(N-M)!(N+M)!} .$$

Solutions to (9) are sought in the form

$$u_1 = A_1 \exp[-iH_1 z] ,$$

$$u_2^* = A_2^* \exp[-iH_1 z] ,$$

constants A_1 and A_2^* are found from the system of linear equations with constant coefficients the determinant of which is a characteristic equation for the corrections H_M to the propagation constants $Mh_0 = h$. The determinant has the form

$$\{H_M - \beta_{M+}\}\{H_M + \beta_{M-}\} + \gamma_{M+}\gamma_{M-} = 0 . \quad (10)$$

For the corrections H_M to the propagation constants we have

$$H_M = \frac{\beta_{M+} - \beta_{M-}}{2} \pm \sqrt{\frac{(\beta_{M+} + \beta_{M-})^2}{4} - \gamma_{M+}\gamma_{M-}} . \quad (11)$$

By wave of illustration we present in Fig. 1 the diagram of wave vectors $\vec{k}(\omega_1)$, $\vec{k}(\omega_2)$, $\vec{k}(\omega_0)$ for the considered effects in the case $M=2$, when the inequality $k(2\omega_0) = \frac{2\omega_0}{c}n(2\omega_0) > 2k(\omega_0) = \frac{2\omega_0}{c}n(\omega_0)$ or $n(2\omega_0) > n(\omega_0)$ is met.

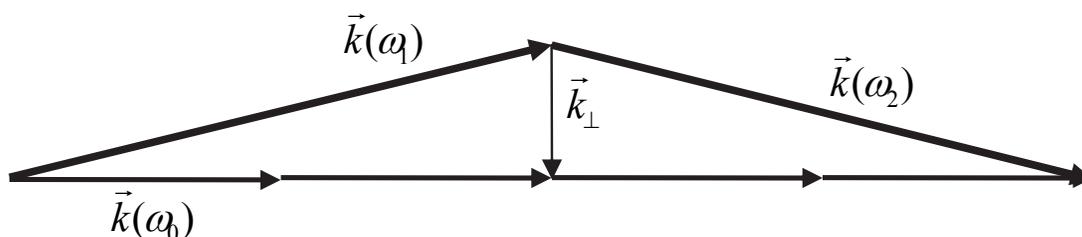


Fig. 1. Wave vector diagram of converting frequency ω_0 to the frequency close to the second harmonic for modulation instability and condition $|k(\omega_1)| > 2|k(\omega_0)|$.

The latter inequality is satisfied if the frequencies ω_0 and $2\omega_0$ are in one transparency band of the substance (Born & Volf, 1964). The wave vector diagram shows that in this case the waves at frequencies ω_0 and $2\omega_0$ may be synchronized during propagation in the direction of vector \vec{k}_0 , which is accompanied by transformation of four vectors (quanta) of frequency ω_0 into two vectors (quanta) of frequency $2\omega_0$.

Let us study (12) in more detail in the quasiparaxial approximation, omitting Ω everywhere except the expression $\sqrt{k^2(M\omega_0 \pm \Omega) - k_\perp^2}$. Let us designate mismatches as

$$\Delta k_M = \frac{\sqrt{k^2(M\omega_0 + \Omega) - k_\perp^2} + \sqrt{k^2(M\omega_0 - \Omega) - k_\perp^2}}{2k_0} - M .$$

For $M = 1$ (modulation instability) the radicand (11) is rewritten in the form

$$D = (\Delta k)^2 + (\Delta k)\Delta\varepsilon \tag{12}$$

In (12) we have

$$\Delta\varepsilon = \sum_{N=1}^s N\delta\varepsilon_{2N+1,NL} = \sum_{N=1}^s I \frac{d}{dl} \delta\varepsilon_{2N+1,NL} = I \frac{d}{dl} \delta\varepsilon_{NL}$$

that is equal to the product of the derivative of the nonlinear additive to intensity and intensity. In the case of cubic nonlinearity, this expression is equal to the additive.

The modulation instability occurs (Litvak & Talanov, 1967) at $D < 0$. Its behavior changes as a function of the signs of Δk and $\Delta\varepsilon$. For $\Delta\varepsilon > 0, \Delta k < 0$, perturbations with spatial scale are more pronounced (“self-focusing” instability); whereas for $\Delta\varepsilon < 0, \Delta k > 0$, perturbations with temporal scales come to the forefront. Note that the modulation instability increments turn to zero at $(\Delta k) = 0$ and

$$\Delta\varepsilon = I \frac{d}{dl} \delta\varepsilon_{NL} = \sum_{N=1}^s N\delta\varepsilon_{2N+1,NL} = 0 \tag{13}$$

From the latter condition it follows that the increments produced by different terms in the expansion (2) may “obliterate” each other under certain conditions.

For $M \geq 2$, in the paraxial approximation we obtain

$$H_M = k_0(\Delta k_M \pm \sqrt{(\Delta k_M + \Delta\varepsilon)^2 - (\Delta\bar{\varepsilon})^2}), \tag{14}$$

where

$$\Delta\bar{\varepsilon} = \frac{M}{2} \sum_{N=M}^s \frac{\delta\varepsilon_{NL,2N+1} N!(N-1)!}{(N-M)!(N+M)!}$$

The propagation constant H_M will be a complex one, and instability will occur, given $|\Delta k_M + \Delta\varepsilon| < |\Delta\bar{\varepsilon}|$. The increment reaches its maximum near zeros of the expression

$$\Delta k_M + \Delta\varepsilon = 0, \tag{15}$$

with the increment being of order

$$H_M = k_0 \Delta\bar{\varepsilon} \tag{16}$$

The expression may become zero at an arbitrary sign of nonlinear additive due to appearance of a transverse component of the perturbation wave number on a certain curve

$$\Delta k_M + \Delta\varepsilon \approx M \left[\frac{n(M\omega_0)}{n(\omega_0)} - 1 \right] + \frac{\Omega^2}{2k_0} \frac{\partial^2 k}{\partial \omega^2} \Big|_{\omega=M\omega_0} - \frac{k_{\perp}^2}{2k_0^2} + \Delta\varepsilon = 0 \tag{17}$$

on the ω, k_{\perp} -plane. At normal dispersion $\frac{\partial^2 k}{\partial \omega^2} \Big|_{\omega=M\omega_0} > 0$ and frequencies $M\omega_0$ and ω_0

located in one transparency band of the substance, the equality (15) is fulfilled for the hyperbolae (17). There exists in this case a minimal value of the transverse wave number

$$\frac{k_{\perp}}{k_0} \approx \sqrt{M \left[\frac{n(M\omega_0)}{n(\omega_0)} - 1 \right]}, \quad M \geq 2 \quad (18)$$

at which instability occurs at arbitrary weak nonlinearity. Fulfillment of the equality (18) indicates the presence of conic radiation.

3. Intense plane wave instability in air

Consider the effects in air. The dependence at normal pressure of the nonlinear index of

refraction $n_{NL} = \frac{\sqrt{\epsilon_0}}{2} \sum_{N=1}^S \epsilon'_{2N+1} |A_0|^{2N}$ on intensity $I = \frac{c}{8\pi} |A_0|^2$:

$$n_{NL} = \sum_{N=1}^S n_{NL,N} I^N \quad (19)$$

taken from (Loriot et al., 2009). is shown in Fig. 2. Curve 1 is plotted taking into consideration four (all known from (Loriot et al., 2009)) terms, curve 2 taking into consideration two terms, and curve 3 taking into consideration only the first term, when purely cubic nonlinearity occurs. Note that for curve 1 there exist unstable branches at $M=1,2,3,4$, and for curve 2, despite its qualitative coincidence with curve 3 (one maximum), the instability branches exist only at $M=1,2$; for curve 3 instability known as modulation (self-focusing) instability occurs at $M=1$.

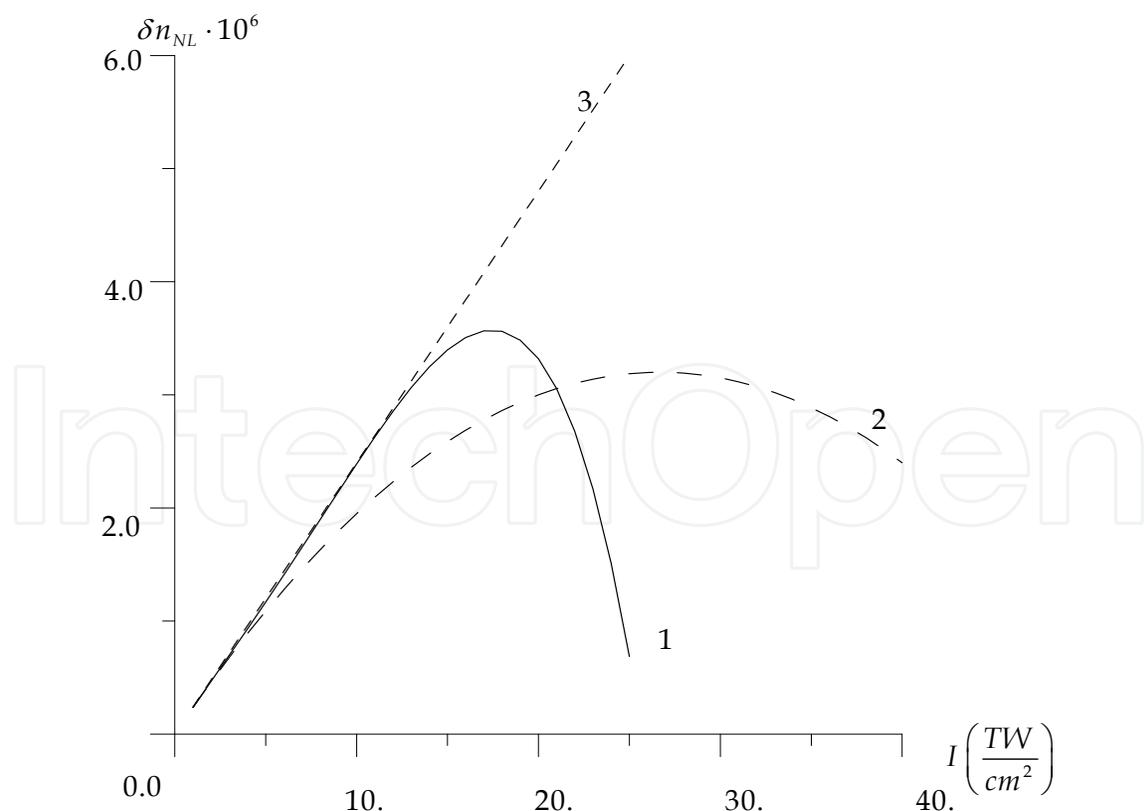


Fig. 2. Nonlinear additive to the index of refraction of air as a function of intensity 1- four-term approximation, 2- two-term approximation, 3- one-term approximation.

The instability regions on the plane of transverse wave numbers and frequencies within the 5-octave band for air, where the frequency dependence of refractive index is known (Grigor'ev & Melikhov, 1991), are shown in Fig.3 for two values of intensity: $I = 15 \text{ TW} / \text{cm}^2$ and $I = 19 \text{ TW} / \text{cm}^2$ for $\omega_0 = 7.85 \cdot 10^{14} \text{ s}^{-1}$, which corresponds to the radiation wavelength $\lambda_0 = \frac{2\pi}{k_0} \approx 0.8 \mu\text{m}$. The shadow density is proportional to the value of the normalized increment $\bar{H}_M = \frac{H_M}{k_0}$. In the first case, the intensity is smaller than its value at maximum nonlinear additive to permittivity and $\Delta\varepsilon > 0$. In the second case, the intensity is larger than its value at maximum nonlinear additive to permittivity and $\Delta\varepsilon < 0$.

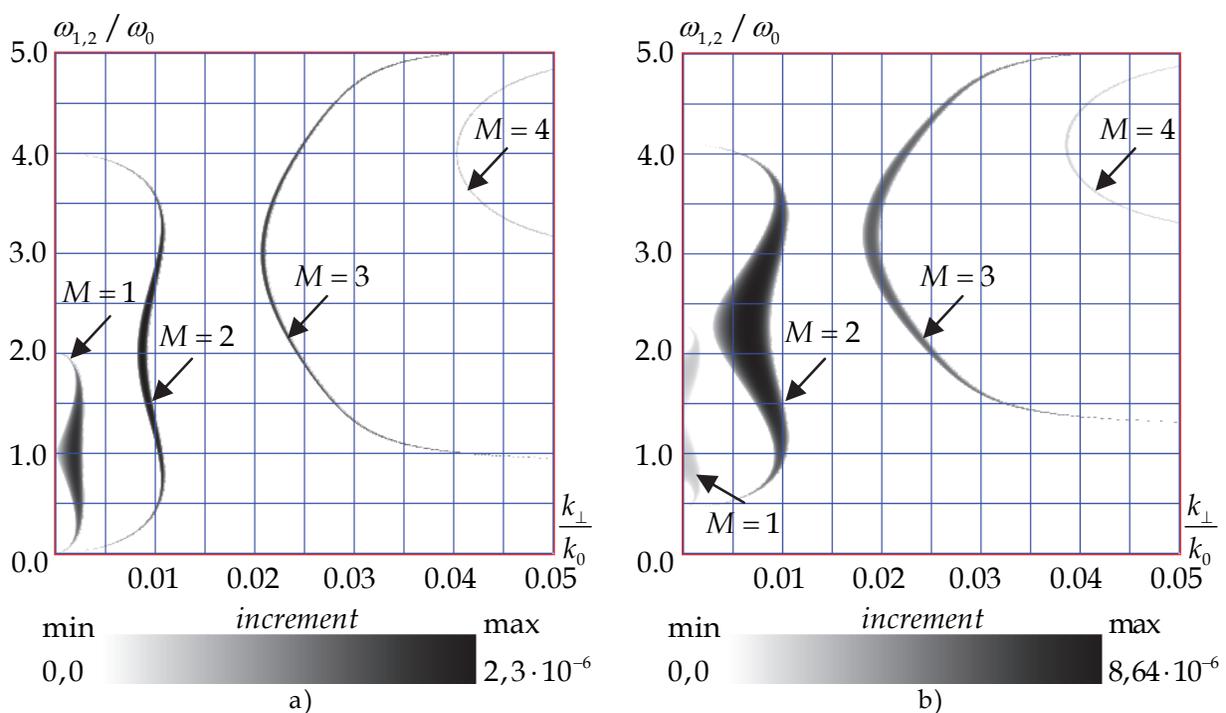


Fig. 3. Instability increment \bar{H}_M in air on the plane of parameters $k_{\perp} / k_0, \Omega / \omega_0$ for $I = 15 \text{ TW} / \text{cm}^2$ (a) and $I = 19 \text{ TW} / \text{cm}^2$ (b) at $\lambda_0 = 0.8 \mu\text{m}$ for different instability branches $M = 1, 2, 3, 4$.

For the values of intensities given above, the increments are different for all branches. At small intensities, $I < 15 \text{ TW} / \text{cm}^2$, the increment is largest for $M = 1$ (modulation instability); at large intensities it is largest for $M = 2$. The increment for perturbations with $M = 2, 3$ attains its maximum near the frequencies $2\omega_0$ and $3\omega_0$, respectively, for the values of κ_{\perp} satisfying the equality (15). It should be born in mind that accuracy of estimates becomes worse near the boundaries of the frequency interval $\omega \ll \omega_0, \omega \sim 5\omega_0$.

Fig. 4, that supplements Fig. 3., demonstrates maximal increments as a function of intensity, with the maximum attained for each branch and each value of intensity at definite values of frequency and transverse wave number that are also functions of intensity and number of the branch. For the branch $M = 1$, the increment becomes small when $\Delta\varepsilon$ vanishes to zero; for the other branches, the increments grow with increasing intensity in the $I > 10 \text{ TW} / \text{cm}^2$ region.

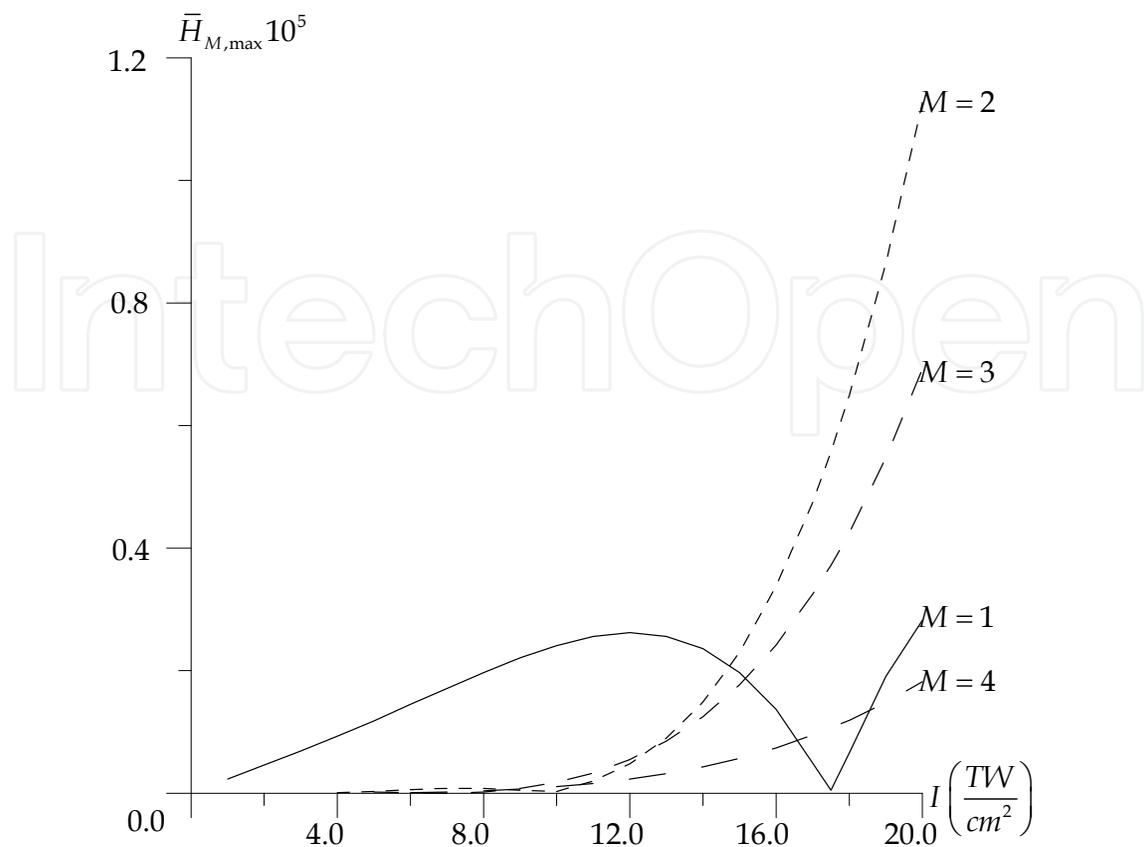


Fig. 4. Maximum value of increment $\bar{H}_{M,\max}$ versus intensity.

The studied instability may be one of the mechanisms of conic beam generation (Nibbering et al., 1996; Dormidontov et al., 2010).

4. Analysis of characteristics of ultrashort wave packet phase modulation by means of autocorrelation intensity function

It has been demonstrated above that the originally spectrum-limited powerful short wave packet is phase modulated during propagation in a nonlinear medium. In this Section we will show that some characteristics of the acquired phase modulation may be measured by analyzing the intensity autocorrelator signal usually used for measuring duration of ultrashort laser pulses. In addition, it is possible to qualitatively assess by the shape of this signal the presence of phase modulation in the studied light signal. Basically, solution of this problem allows retrieving amplitude and phase distribution of electric field in an ultrashort wave packet.

Methods for retrieving the total field of an optical pulse may conventionally be divided into three groups. The first group includes techniques based on interference measurements. They have a long history and, consequently, have been developed most comprehensively. It is clear that, based on the interference of an ultrashort pulse in the temporal or spectral domain, one can in principle derive information on the phase distribution of the studied field. A great number of publications, from which we cite only the key papers (Kuznetsova, 1968; Verevkin et al., 1971; Sala et al., 1980; Diels et al., 1985;

Naganuma et al., 1989; Iaconis & Walmsley, 1998), are devoted to interference measurements. Another group of fairly recent publications (Naganuma et al., 1989; Iaconis & Walmsley, 1998; DeLong et al., 1996; Koumans & Yariv, 1999; Kane, 1999) take advantage of the mathematically proven fact that a multi-dimensional (noninterference) auto- or cross-correlation function allows retrieving the phase distribution of a short pulse. A two-dimensional signal distribution on the frequency-delay plane is usually measured in experiment, and then an iterative processing algorithm yields the desired parameters. The most advanced to-date technique of this group is referred to as FROG in the literature (Kane, 1999). The third group of methods (Naganuma et al., 1989; Kane, 1999; Peatross et al., 1998; Nicholson et al., 1999), that are based on the results of simultaneous spectral and correlation energy measurements and some iterative algorithm, also make it possible to determine phase and amplitude characteristics of an optical pulse.

The interference methods of retrieving ultrashort wave packet field parameters are simpler for implementation and in terms of processing than the methods of the second group. However, despite the fact that the speed of obtaining qualitative information for the latter techniques often does not meet the needs of experiment, such techniques are clearer, since the form of phase modulation of an optical pulse can qualitatively be deduced immediately from a two-dimensional distribution of the obtained signal. In this respect, methods of the third group are much faster, as the corresponding iterative retrieval procedure is based on operations with one-dimensional data files. However, the problem of accuracy of retrieval of the desired parameters remains open for such methods. In this part we demonstrate that in certain cases, the interference methods can also ensure clear, simple, and fast acquisition of information on the amplitude and phase of an optical pulse. In our opinion, the present part is methodical to a significant extent, although it has practical applications related to measuring dispersion characteristics of optical materials. It can easily be shown that the output signal from the photodetector of an interferometric intensity autocorrelator, whose scheme can be found in a number of publications (e.g., Krukov, 2008), has the form:

$$U(\tau) \approx \int_{-\infty}^{\infty} \left\{ \rho^2(t) + \rho^2(t+\tau) + 2\rho(t) * \rho(t+\tau) * \cos[\phi(t) - \phi(t+\tau)] \right\}^2 dt \quad (20)$$

where $\rho(t)$ and $\phi(t)$, respectively, are the slowly varying amplitude (envelope) and phase of the optical-pulse field

$$E(t) = \rho(t) * \exp[j\phi(t)] + c.c. \quad (21)$$

Equation (20) is known (Akhmanov, Vysloukh & Chirkin, 1988) to be valid as long as the inequality $\omega_0\tau_0 > 1$ is satisfied, i.e., if the envelope $\rho(t)$ contains at least a few optical cycles. Here, ω_0 and τ_0 are the central frequency and duration of the wave packet envelope, respectively. Note that, strictly speaking, the signal (20) contains the intensity autocorrelation function

$$G(\tau) = \int_{-\infty}^{\infty} \rho^2(t) * \rho^2(t+\tau) dt \quad (22)$$

but is not equal to it. In addition to an informative component, Eq. (20) for $U(\tau)$ comprises a constant background which usually impedes experimental measurement of temporal characteristics. This well-known fact was extensively discussed in the literature (Kuznetsova, 1968; Krukov, 2008). It is not difficult to design the scheme of the so-called background-free autocorrelator for which the output signal is free of this background. However, it follows from Eq. (20) that in this case, information on the phase structure of a pulse is completely lost. Nevertheless, such devices for measuring temporal parameters of ultrashort pulses are widely used in experiments, as they yield fairly reliable information on the wave packet envelope duration.

It follows from Eq. (20) that the signal $U(\tau)$ contains information on the time dependence of the envelope amplitude and phase. Typically of inverse problems, it is impossible to retrieve parameters of the field $E(t)$ in the general case, since the integral equation (20) is ill-posed. However, in certain particular cases of practical importance, $\rho(t)$ and $\phi(t)$ can be found from this expression. Consider this possibility in more detail.

Assume that the slowly varying envelope $\rho(t)$ has a Gaussian profile, while the phase $\phi(t)$ can be described by a cubic polynomial:

$$\rho(t) = \rho_0 \cdot \exp\left(-\frac{t^2}{2\tau_0^2}\right); \quad \phi(t) = \omega_0 \cdot t + t^2 \cdot \frac{\alpha}{2} + t^3 \cdot \frac{\beta}{3}. \quad (23)$$

where the coefficients α and β refer to the linear and quadratic frequency chirps, respectively. Within the framework of this assumption, the temporal distribution of the wave packet envelope is determined by one parameter, the envelope duration τ_0 , and the phase distribution by two coefficients α and β . Representation of the phase in the form given by Eq. (23) corresponds to a Taylor expansion in which any higher-order term is much less than the previous lower-order one. It usually suffices to use such a phase expansion for an ultrashort wave packet, for which Eq. (21) is valid, propagated in a substance with weak dispersion, i.e., in the transparency band. Let us firstly put $\beta = 0$, i.e., allow for only a linear chirp of an input optical pulse. Then Eq. (20) for the autocorrelator output signal $U(\tau)$ takes the form

$$U(\tau) \approx 1 + 2G(\tau) + [G(\tau)]^L \cos 2\omega_0\tau + 4[G(\tau)]^{(2+L)/4} \cos \omega_0\tau \cdot \cos\left[\sqrt{L^2-1} \cdot \tau^2 / 4\tau_0^2\right], \quad (24)$$

Here, L is the temporal-compression ratio of an initial phase modulated pulse with envelope duration τ_0 due to compensation for its quadratic phase (Akhmanov, Vysloukh & Chirkin, 1988):

$$L^2 = \left(\frac{\Delta\omega}{\Delta\omega_0}\right)^2 = 1 + (\alpha\tau_0^2)^2, \quad (25)$$

$\Delta\omega$ is the spectrum width of an input optical signal, and $\Delta\omega_0 = 1/\tau_0$ is the spectrum width of a transform limited pulse for which $\alpha = 0$ and, therefore, $L = 1$. Note that Eq. (24) was derived in (Sala, Kenney-Wallace & Hall, 1980; Diels, Fontane, McMichel & Simoni, 1985). However, the authors of (Sala, Kenney-Wallace & Hall, 1980). did not reduce it to such a clear form, which seemingly impeded its further analysis, while the approximation of the upper and lower branches of the envelope of the signal $U(\tau)$ used in (Diels, Fontane, McMichel & Simoni, 1985) is not sufficiently accurate for retrieval of the parameter α , especially for an analysis of few-optical-cycle pulses.

Let us analyze the obtained expression. In fact, the signal $U(\tau)$ is not equal to the intensity autocorrelation function (22) even for a transform limited optical signal and contains several characteristic temporal scales. The largest one, $\tau_1 \sim \sqrt{2} \tau_0$, is related to the duration of the envelope $\rho(t)$ and is determined by the function $G(\tau) = \exp[-\tau^2 / (2\tau_0^2)]$. The next, shorter-term scales

$$\tau_2 \sim \frac{2\tau_0}{(L^2 - 1)^{1/4}}; \quad \tau_3 \sim \frac{2\sqrt{2}\tau_0}{(2 + L^2)^{1/2}}; \quad \tau_4 \sim \frac{2\tau_0}{L}. \quad (26)$$

are determined by the third and fourth terms on the right-hand side of Eq. (24). Finally, the minimum temporal scale $\tau_5 \sim 1 / \omega_0$ in Eq. (24) is determined by the optical cycle of a pulse. If $L > 1$, then the hierarchy of these temporal scales is as follows: $\tau_1 > \tau_2 > \tau_3 > \tau_4 > \tau_5$. This means that the signal $U(\tau)$ in the presence of a linear chirp should have an oscillatory component near $\tau = 0$, which is primarily determined by the last term on the right-hand side of Eq. (24), and smooth wings with shape determined by the function $G(\tau)$. In this case, the larger the value of L , i.e., the greater deviations of an optical pulse from a transform limited one, the more prominent the localization of the oscillatory component of such a signal in the vicinity of $\tau = 0$. If the averaging over the shortest scale τ_5 is performed in the operation of a detecting system, then the signal $U(\tau) \sim 1 + 2G(\tau)$ does not contain information on the optical pulse phase. This is also a well-known fact (Diels, Fontane, McMichel & Simoni, 1985; Krukov, 2008).

The function $U(\tau)$ calculated using Eq. (24) for various values of the pulse duration τ_0 and the parameter L is plotted in Fig. 5. It follows from analysis of these plots that, despite the fact that it is impossible to obtain the actual autocorrelation function from the signal of an interferometric intensity autocorrelator, the duration of a transform limited optical pulse can be determined with experimentally plausible accuracy using the upper branch $(7G(\tau) + 1)$ of the oscillatory-component envelope of the signal. This fact is quite pleasant, as namely such a technique for measuring durations of ultrashort optical pulses is used by virtually all researchers. The appearance of smooth signal wings without any periodic modulation is indicative of a quadratic phase modulation of the studied wave packet. Hence, the form of the measured function $U(\tau)$ provides information on the presence of a linear frequency chirp in the optical band. It follows from Eq. (25) that for finding the numerical value of the coefficient a one should determine L and τ_0 . In what follows, when discussing the experimental verification of the calculations, we describe in detail a procedure for determining these quantities. Here, we only note that the sign of the coefficient α , as is seen from Eq. (25), cannot be specified by this method; its determination requires either some *a priori* information or an additional experiment.

Let us now turn to analysis of the effect of the nonlinear frequency chirp, i.e., the cubic additive to the phase in Eq. (23) on the signal profile $U(\tau)$. For this, we put $\alpha = 0$ in Eq. (23). This can be done experimentally, if the quadratic phase is pre-compensated using, e.g., a prism dispersion compensator. With allowance for this assumption, we can also find an analytical formula for the output signal of an interferometric intensity autocorrelator

$$V(\tau) \approx 1 + 2G(\tau) + G(\tau) * \sqrt[4]{4F^2(\tau) - 3} \cos \Phi(\tau) + 4[G(\tau)]^{1-1/4F^2(\tau)} * \cos \Psi(\tau) * 1 / \sqrt{F(\tau)} \quad (27)$$

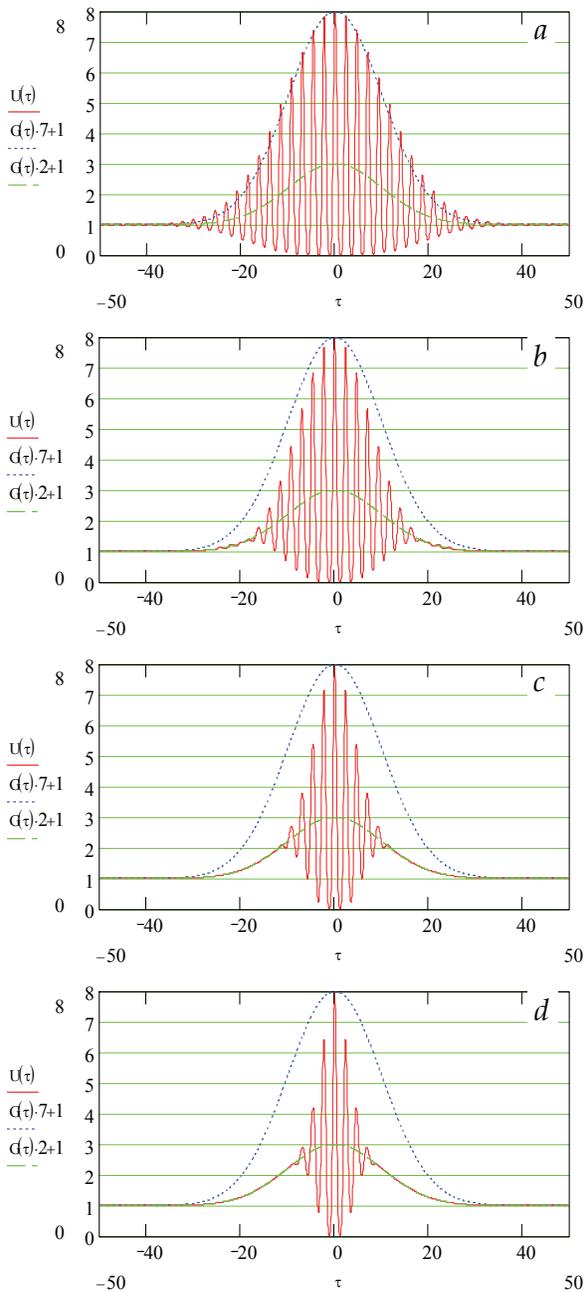


Fig. 5. Profile of the signal $U(\tau)$ for $L = 1$ (a), $L = 2$ (b), $L = 4$ (c), and $L = 6$ (d) in the case of a pulse of duration $\tau_0 = 10\text{ fs}$ with quadratic phase modulation.

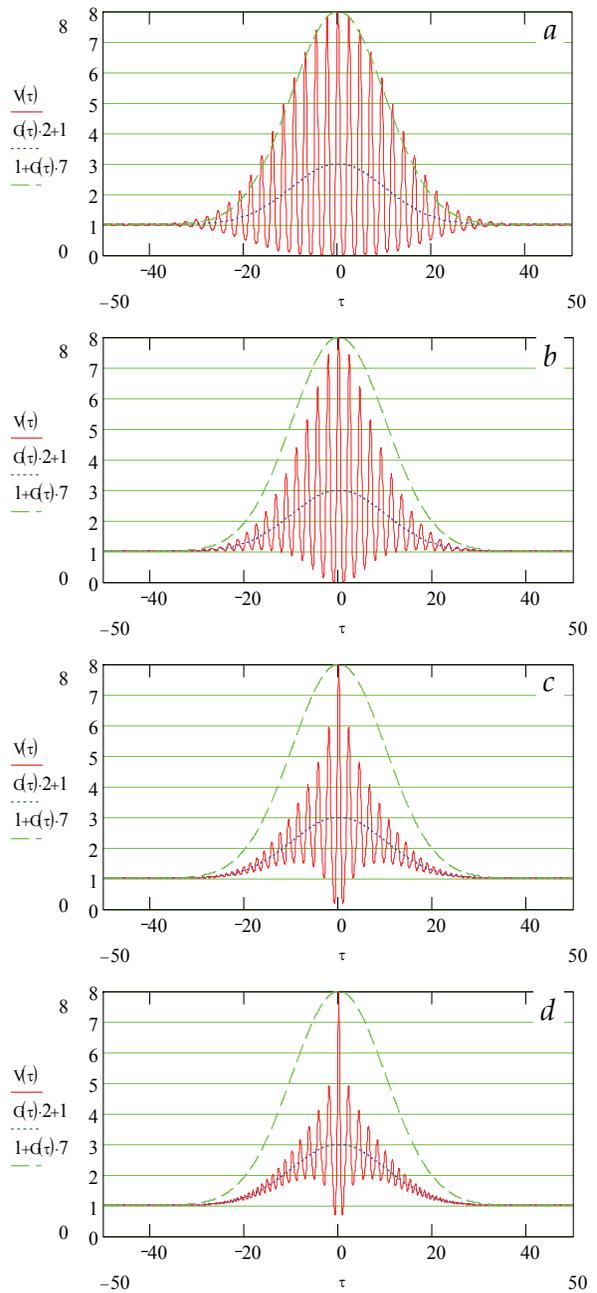


Fig. 6. Profile of the signal $V(\tau)$ for $K = 1$ (a), $K = 5$ (b), $K = 20$ (c), and $K = 50$ (d) in the case of a pulse of duration $\tau_0 = 10\text{ fs}$ with cubic phase modulation.

where

$$F(\tau) = \sqrt{1 + (K^2 - 1) * (\tau / 2\tau_0)^2} , \tag{28a}$$

$$\Phi(\tau) = 2\omega_0\tau + 0.5a \tan \left[(\tau / \tau_0) \sqrt{K^2 - 1} \right] + (\tau / \tau_0)^3 (\sqrt{K^2 - 1}) / 6 \tag{28b}$$

$$\Psi(\tau) = \omega_0 \tau + 0.5a \tan\left((\tau / 2\tau_0)\sqrt{K^2 - 1}\right) + (1 / 16F^2(\tau) + 1 / 12)(\tau / \tau_0)^3 \sqrt{K^2 - 1} \quad (28c)$$

$$K^2 = 1 + (\beta\tau_0^3)^2. \quad (28d)$$

It follows from Eq. (27) that the formula for $V(\tau)$ is analogous to Eq. (24). The first two terms in these formulas are identical, while the third and fourth terms are oscillatory additives. The physical meaning of the parameter K is similar to that of the parameter L specified above. Note that in the considered case, localization of the oscillatory component of a signal near $\tau = 0$ is less prominent than for a quadratic phase since the exponent of the function $G(\tau)$ in the last term on the right-hand side of Eq. (27), which determines this localization, tends to unity with increasing K . Moreover, it follows from Eqs. (27) and (28) that the output signal modulation becomes much more aperiodic than in the case of a linear frequency chirp, and the aperiodicity of the observed oscillations is the stronger, the larger the parameter K . Therefore, the presence of a linear frequency chirp in a signal can be determined from broadening of the profiles of the harmonics of the signal $V(\tau)$ at the frequencies ω_0 and $2\omega_0$.

The signal profiles $V(\tau)$ for various K are plotted in Fig. 6. Analysis of these plots shows that, as the behavior of the curves $V(\tau)$, especially for small K impedes finding any features entirely determined by τ_0 or β , the procedure of determining the parameters τ_0 and β is more difficult and complicated in this case than in the case of a linear frequency chirp. If the phase modulation is relatively weak ($K < 5$), then, in contrast to the case of a quadratic phase, the obtained dependences are close to the signal $V(\tau)$ for $K = 1$. Of course, this makes retrieval of the desired quantities from experimental data more difficult. The difference in the functions $V(\tau)$ for different K is quite measurable for $N \geq 5$, so that the above mentioned features or specific components can already be pointed out. Therefore, the parameters τ_0 or β can quite easily be determined in this case. However, such large values of K can hardly be realized in practice. In the limiting case of $K \gg 1$, the signal $V(\tau)$ has a smooth shape, determined by the first two terms in Eq. (27), with a very narrow, the so-called coherence peak in the vicinity of $\tau = 0$, because $V(0) = 8$ in any case. Determination of the sign of β , as in the case of a quadratic phase modulation, is impossible. This is explained by the physical principle of operation of this correlator. Therefore, the possibility of experimental measuring of a cubic phase of an ultrashort optical pulse using an ordinary interferometric intensity autocorrelator seems very problematic, except probably for some special cases.

If the studied optical signal comprises both quadratic and cubic phase modulation, then the output signal of an interferometric intensity autocorrelator can also be found analytically assuming a Gaussian signal envelope. The resulting formula is similar to the functions $U(\tau)$ and $V(\tau)$ obtained above, but is much more cumbersome and, correspondingly, far less illustrative than the formula for $U(\tau)$.

To check the results of calculation, we performed an experiment in which the signal from an interferometric intensity autocorrelator was obtained for a phase-modulated ultrashort optical pulse. The femtosecond ring laser described in (Babin, Kiselev, Kirsanov & Stepanov, 2002) was used as the radiation source. An external prism dispersion compensator mounted immediately after the output mirror was used to compensate for the dispersion of the

substrate material of this mirror to ensure that a transform limited wave packet is input to the autocorrelator. The total duration $2\tau_0$ of the compensated pulse measured using its interference autocorrelation function (Fig. 7.) amounted to about 20 fs at the e^{-1} -folding level of the maximum intensity. The measured value of the product $\Delta\omega\tau_0$ was equal to 1.3, which is 30% larger than the corresponding value for a transform limited Gaussian pulse. This is known (Rousseau, McCarthy & Piche, 2000) to be related to a slight deviation of the generated spectrum from the Gaussian one, as is the case for the considered experiment. Note that hardware averaging of the autocorrelation function shown in Fig. 7 over fast oscillations yields the same value of τ_0 , which, according to Eqs. (24) and (27), is indicative of absence of phase modulation of the optical pulse at the compensator output.

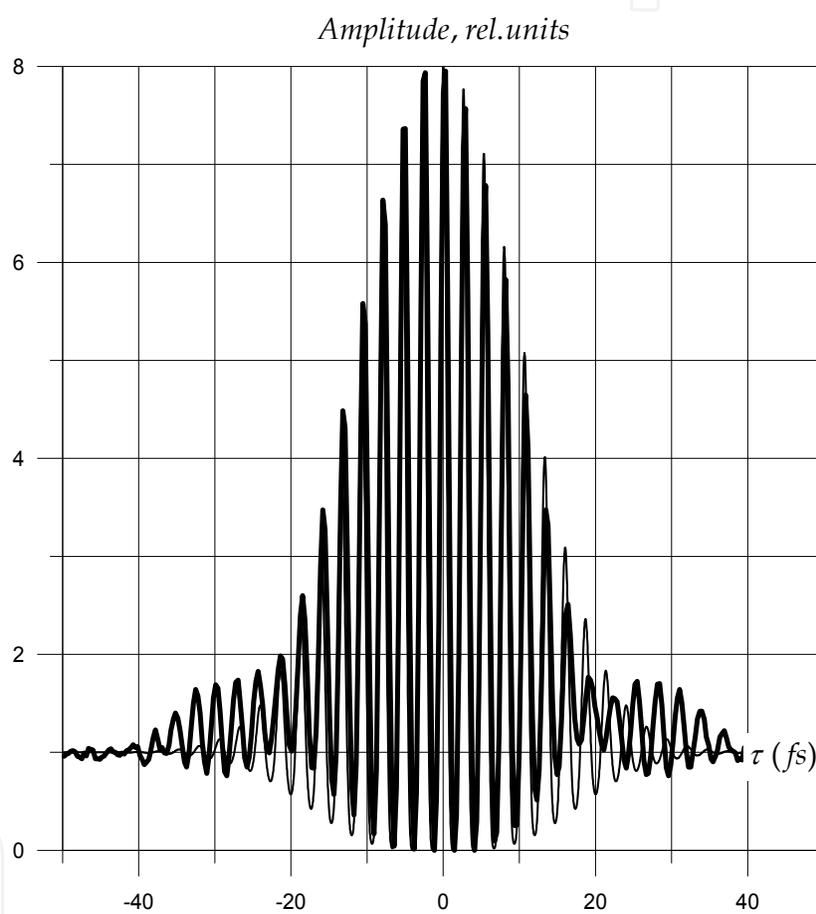


Fig. 7. Output signal of interferometric intensity autocorrelator for a transform limited wave packet. The bold and fine curves are for the experiment and the calculation using Eq. (24) for $L = 1$, respectively.

The idea of the experiment is fairly simple: to introduce a chirp in an initial transform limited laser pulse and to check the resulting output signal of the autocorrelator. It is known (Akhmanov, Vysloukh & Chirkin, 1988) that an ultrashort optical pulse can easily be phase modulated upon propagation through a linear dispersive medium. To realize this experimentally, we mount plane-parallel plates made of various materials immediately before the autocorrelator input.

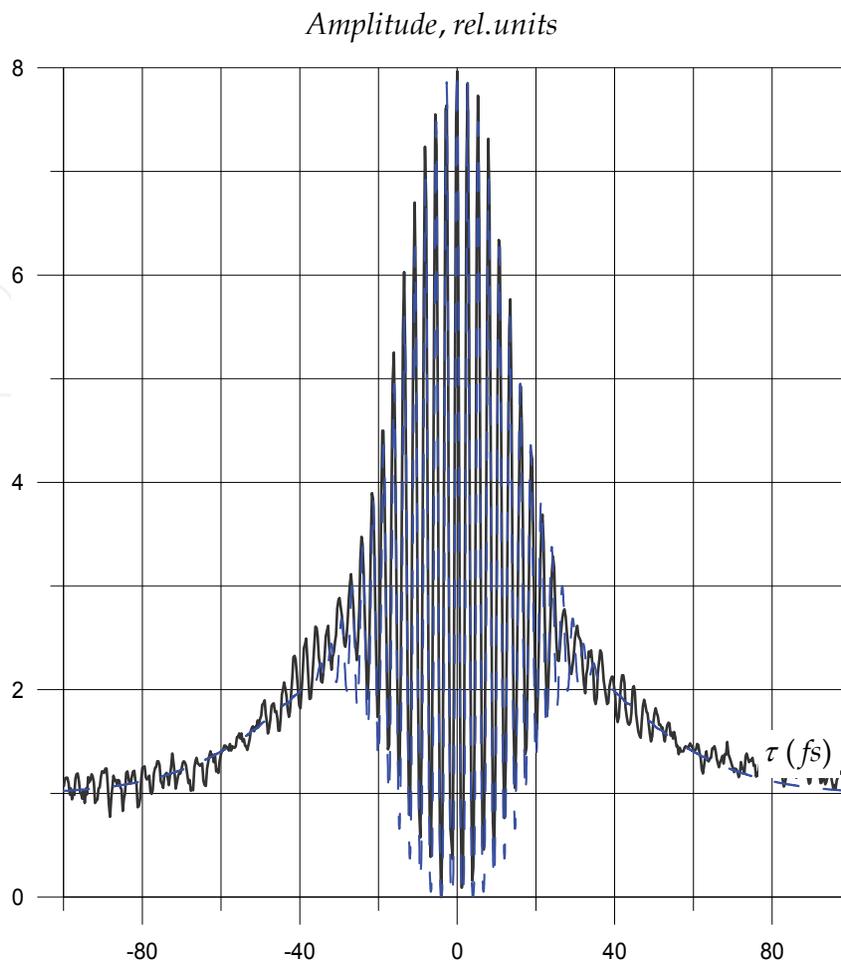


Fig. 8. Output signal of interferometric intensity autocorrelator for a phase modulated wave packet. The initial transform limited signal passed through a 3.25 mm thick ZnSe plate. The solid and dashed curves are, respectively, for the experiment and the calculation using Eq. (24) for $\tau_0 = 33.5$ fs and $L = 4.47$.

Fig. 8. shows a typical profile of the output signal obtained in this case. It follows from this figure that the observed picture fully agrees with the theoretical analysis for the case of a quadratic phase. Note that a similar profile was obtained in (Sala, Kenney-Wallace & Hall, 1980) for a linearly chirped pulse of about 13 ps in duration. Excess noise in the signal is due to a nonoptimal frequency band of the amplifier used in our experiment. Let us find the parameters of a phase modulated optical pulse input to the correlator assuming that a substance gives only a quadratic additive to the phase. The envelope duration τ_0 can most easily be derived from the profile of the wings of the signal $U(\tau)$. According to Eq. (24), the shape of the signal wings is described by the formula $U(\tau) \sim 1 + 2G(\tau)$. Therefore, the parameter τ_0 is readily determined for a wave packet with Gaussian envelope. The next step is to appropriately choose the parameter L to ensure the best agreement between the oscillating parts of experimental and theoretical functions $U(\tau)$. Note that one more parameter, ω_0 , entering Eq. (24), can easily be found either from the period of oscillations in Fig. 7 or from the measured average frequency of the spectrum of the analyzed pulse. Hence, the formulated problem is solved completely in this approximation, i.e., we have

measured the parameter τ_0 and the coefficient α of the quadratic phase using the output signal of an interferometric intensity autocorrelator. A cubic phase additive is known to be much less than the quadratic one for transparent optical materials. Therefore, in our case, this additive has almost no impact on the increase in the envelope duration τ_0 . This means that the parameter K is close to unity and, according to the above theoretical analysis, it is impossible to measure the parameter β in the experiment. In this experiment, using a scanning intensity autocorrelator, we actually determined the averaged parameters of an ultrashort wave packet, as the analyzed signal had the shape of a femtosecond pulse train with a repetition rate of about 100 MHz. In principle, a similar interference autocorrelator can also be realized for rarely repeated or even single optical pulses if, as was proposed in (Brun, Georges & LeSaux, 1991), the standard scheme of a single-pulse correlator (Brun, Georges & LeSaux, 1991) including a nonlinear crystal with tangential (superwide angular) synchronism is used.

5. Conclusion

To conclude we enumerate the principal results of the work. It was shown that in media with nonlinearity described by a finite-degree polynomial, instability may develop at frequencies greatly exceeding the carrier frequency. Increments of these frequencies are found. Methods of measuring temporal characteristics of femtosecond pulses were analyzed. Retrieval of amplitude and phase modulated wave packets by means of interference intensity autocorrelator were demonstrated theoretically and experimentally.

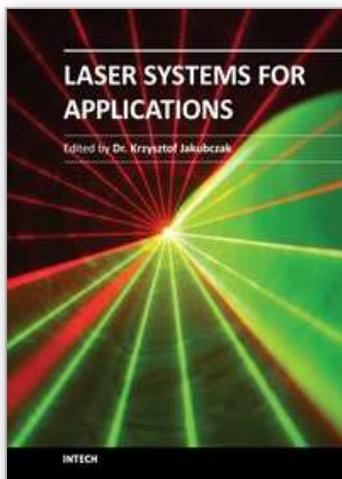
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This book addresses topics related to various laser systems intended for the applications in science and various industries. Some of them are very recent achievements in laser physics (e.g. laser pulse cleaning), while others face their renaissance in industrial applications (e.g. CO₂ lasers). This book has been divided into four different sections: (1) Laser and terahertz sources, (2) Laser beam manipulation, (3) Intense pulse propagation phenomena, and (4) Metrology. The book addresses such topics like: Q-switching, mode-locking, various laser systems, terahertz source driven by lasers, micro-lasers, fiber lasers, pulse and beam shaping techniques, pulse contrast metrology, and improvement techniques. This book is a great starting point for newcomers to laser physics.

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