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Propagation of ultrashort pulses through a nonresonance quadratically nonlinear medium in the unidirectional wave approximation

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Abstract. The propagation and interaction of ultrashort pulses in a nonresonance quadratically nonlinear medium is considered. A stationary solution was found analytically. It was demonstrated numerically that pulses with an energy much lower than the energy of a stationary pulse decay under the influence of dispersion during propagation, whereas pulses with an energy higher than the energy of a stationary pulse disintegrate into a series of pulses moving like stationary ones. The effect of additive and multiplicative amplitude modulation on the pulse propagation was investigated. Stationary pulses were shown to be stable upon weak modulation and in collisions with each other.

1. Introduction

The last decade has seen a vigorous mastering of the range of femtosecond electromagnetic radiation pulses [1-11]. One way to attain these pulse durations is to compress the initial pulse employing fibre-grating compressors [1-3]. By this means, for instance, 6-fs pulses were obtained [2]. Another way to produce femtosecond pulses is to generate them directly in laser systems [4-8]. Sartania et al. [4] succeeded in generating 20-fs pulses with an energy of 1.5 mJ and a repetition rate of 1 kHz. The subsequent compression of these pulses using a fibre-prism compressor yielded 5-fs pulses with an energy of 0.5 mJ. Jung et al. [6] demonstrated the generation of 6.5-fs pulses with an average power of 200 mW and a repetition rate of 86 MHz by a Ti: sapphire laser. The parametric wave interaction, the self-focusing and the self-modulation, and also the coherent transient processes in the field of femtosecond pulses were considered in Refs[9-11].

Achievements in the field of generation of ultrashort radiation pulses (USPs), make the analysis of their propagation in nonlinear dispersion media in the context of different medium models a topical problem. A natural foundation for all the theories are Maxwell's equations. They are complemented with either the constitutive equations that determine the evolution of radiation-induced polarisation and currents in the medium [12–17] or with the Schrödinger equation for the electrons that interact with the external electromagnetic

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field [18, 19]. Because the explicit analytical results can be obtained only rarely, different approximations are widely used, which permit the problem to be simplified and analytical expressions to be derived.

Among the numerous models of a nonlinear medium invoked to investigate the propagation of an USP pulse, we mention the medium with a cubic nonlinearity [20, 21], in which the nonlinear response was determined using the Duffing model. As noted in Ref. [22], for a more precise description of the dispersion of the nonlinear refractive index it is necessary to use a model of at least two coupled oscillators. In subsequent papers, the interaction of USPs with dielectric media was considered using models of two [23, 24] and three [25] coupled oscillators. Note also the paper by Belenov et al. [26], who pointed out the role of the electronic-vibrational (Raman) interaction in the formation of the nonlinear response of the medium to the action of USPs.

Therefore, within the framework of classical physics, the nonlinear properties of an isotropic dielectric can be described by the Duffing model (or an anharmonic oscillator with a cubic anharmonicity) for coupled electrons and by the Placzek model [27] (or the Bloembergen – Shen model [28]) used to describe Raman scattering, like in Refs [23 – 26]. The next step in the generalisation of these models is taking into account the vector nature of electromagnetic radiation and the passage to the model of an anharmonic oscillator with two degrees of freedom [29 – 31].

In anisotropic nonlinear media, the potential energy of bound electrons is not an even function of the electron displacement from the equilibrium position. Therefore, the Duffing model should be replaced by a model of an anharmonic oscillator with a quadratic nonlinearity (the quadratic Duffing model). This model was employed in Refs [31, 32] to describe the propagation of an USP of arbitrarily polarised radiation in a quadratically nonlinear medium with or without dispersion. Like in media with a cubic nonlinearity, one would expect here a generalisation of a purely electronic model by taking into consideration the Raman-type interaction (like in Refs [23–28]) with the inclusion of the medium anisotropy and the vector nature of the electromagnetic field.

An important and yet simple approximation can be obtained by assuming that electromagnetic waves propagate only in one of the possible directions [33]. The condition of unidirectional waves reduces the order of the wave equation without introducing limitations on the pulse length, which was shown in detail in Refs [12, 20, 33, and 34]. It is important that this approximation does not use the concept of a quasiharmonic nature of this wave. (Naturally, there exist sit-

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uations where this approximation is known to be inappropriate, e.g., waves in periodic or scattering media.)

In this paper, we study the propagation and interaction of ultrashort pulses of linearly polarised electromagnetic radiation, which have one or several oscillations of the electric field intensity in a medium characterised by a nonlinear response and dispersion.

2. Basic equations of the model

Under the assumption that the polarisation vector retains its direction, the propagation of a linearly polarised plane electromagnetic wave is described by the scalar wave equation

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2 E}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 P}{\partial t^2},\tag{1}$$

where P is the polarisation of the medium. The problem is simplified if the USP propagation is considered in the unidirectional wave approximation [12, 33, and 34]. In this case, instead of Eqn. (1), the simpler equation

$$\frac{\partial E}{\partial z} + \frac{1}{c} \frac{\partial E}{\partial t} = -\frac{2\pi}{c} \frac{\partial P}{\partial t}$$
 (2)

is used.

To calculate the polarisation P, the model of the medium should be adopted. We will use the anharmonic oscillator model — the quadratic Duffing model, which was considered in the description of parametric processes in quadratically nonlinear media [35–37]. It is assumed that electrons are located in a potential well and oscillate with the frequency ω_0 about its equilibrium position under the action of the external field. Let X be the displacement from the equilibrium position averaged over the ensemble of all bound electrons. Then, following Bloembergen [37], the equation of motion can be written as

$$\frac{\partial^2 X}{\partial t^2} + \omega_0^2 X + \kappa_2 X^2 = \frac{e}{m_{\text{ef}}} E(z, t),$$
 (3)

where κ_2 is the anharmonicity constant and $m_{\rm ef} = 3m/(\varepsilon+2)$ is the effective electron mass. The oscillation damping is neglected here, assuming the duration of the USP-electron system interaction to be much shorter than the relaxation time of the system. The polarisation of the unit volume of the medium is defined as $P = n_{\rm a} e X$, where $n_{\rm a}$ is the atomic number density.

It is convenient to pass to new dimensionless variables $\zeta = x/L$ and $\tau = \omega_0(t-z/c)$ and fields $\mathscr{E} = E/A_0$ and $q = X/X_0$ by using the normalisation parameters

$$A_0 = \frac{m_{\rm ef}\omega_0^4}{e|\kappa_2|}, \ X_0 = \frac{\omega_0^2}{|\kappa_2|}, \ L^{-1} = \frac{2\pi n_{\rm a}e^2}{m_{\rm ef}c\omega_0} = \frac{\omega_{\rm p}^2}{2c\omega_0},$$

where $\omega_{\rm p} = (4\pi n_{\rm a}e^2/m_{\rm ef})^{-1/2}$ is the plasma frequency. In the dimensionless variables, Eqns (1) and (2) take the form

$$\frac{\partial \mathscr{E}}{\partial \zeta} = -\frac{\partial q}{\partial \tau}, \quad \frac{\partial^2 q}{\partial \tau^2} + q + q^2 = \mathscr{E}. \tag{4}$$

The passage from the wave equation (1) to the approximate equation (2) has been described in a number of pa-

pers [12, 20, 33, and 34]. However, there is good reason to discuss once again the condition for the validity of this passage. Let us introduce the characteristic variables T=t-z/c and Z=t+z/c. For P=0 (or for $P=\chi E$, where the susceptibility χ is a constant), the wave equation (1) has solutions in the form of the waves traveling either along the characteristic T= const or along Z= const (or along the characteristics T=t-z/c' and Z=t+z/c', where c' is the velocity of light in a dispersion-free medium with the susceptibility χ). This picture breaks down when P is nonzero (or the medium is dispersive and/or is nonlinear). Let us make the formal substitution $P\to \varepsilon P$ and expand E, B, and P in powers of ε assuming this parameter to be small:

$$E = E^{(0)}(T) + \varepsilon E^{(1)}(T, Z) + \varepsilon^2 E^{(2)}(T, Z) + \dots,$$
(5)

$$P = P^{(0)}(T) + \varepsilon P^{(1)}(T, Z) + \varepsilon^2 P^{(2)}(T, Z) + \dots$$

By substituting these expansions into the wave equation (1) and collecting the terms of the same order of smallness in ε , we can easily see that equation (2) arises in the first order in ε , i. e., the unidirectional wave approximation is equivalent to replacing expansion (5) by the expression

$$E(T,Z) = E^{(0)}(T) + \varepsilon E^{(1)}(T,Z). \tag{6}$$

The polarisation of the medium results in a change in the characteristic of a stationary wave but does not rule out the propagation of variable-profile waves, i.e., the transient waves. Here, the concept of a quasiharmonic wave is not invoked: no limitations are imposed on the rate of variation of the electric field strength E(T,Z)=E(t,z). The criterion for the validity of approximation (6) is the smallness of the parameter ε (which is a measure of smallness of the polarisation effect). To explicitly determine this parameter in the case under consideration, we may proceed as follows. Like in Eqns (4), we pass to variables $\xi=\omega_0z/c$ and $\vartheta=\omega_0t$ and to fields $\mathscr E$ and g. Then, the system of equations (1) and (3) takes the form

$$\frac{\partial^2 \mathscr{E}}{\partial \xi^2} - \frac{\partial^2 \mathscr{E}}{\partial \theta^2} = \left(\frac{\omega_{\rm p}}{\omega_{\rm 0}}\right)^2 \frac{\partial^2 q}{\partial \theta^2}, \quad \frac{\partial^2 q}{\partial \theta^2} + q + q^2 = \mathscr{E}. \tag{7}$$

It is now evident that the $(\omega_p/\omega_0)^2$ ratio serves as a parameter which is a measure of the influence exerted by polarisation on the wave propagation. Therefore, the parameter ε in expansion (5) may be defined as $2\varepsilon = (\omega_p/\omega_0)^2$. Upon reducing expansion (5) to expression (6) and reverting to the initial dimensional variables, we obtain the system of equations (2) and (3). Hence, the condition for the validity of the unidirectional wave approximation is the requirement that $(\omega_p/\omega_0)^2 \ll 1$. In what follows this condition is assumed to be fulfilled and we investigate the solutions of the system of equation (4).

3. Stationary solitary wave

As a simple example of solving this system, we consider the solution that describes the propagation of a stationary solitary wave. Let $\mathscr E$ and q depend only on the variable $\eta=\tau-\zeta/\alpha=\omega_0(t-z/V)$, where V is the propagation velocity of a stationary electromagnetic pulse, and

$$\frac{1}{V} = \frac{1}{c} \left[1 + \frac{1}{2\alpha} \left(\frac{\omega_{\rm p}}{\omega_{\rm 0}} \right)^2 \right]. \tag{8}$$

One can see that α numbers the members of the family of stationary solutions of Eqns (4). This system of equations can be solved, taking into account that the relation

$$\mathscr{E} = \alpha q \tag{9}$$

follows from the first of Eqns (4). The second equation takes the form

$$\frac{d^2q}{d\eta^2} + (1 - \alpha)q + q^2 = 0. {10}$$

Solutions of this equation that satisfy the boundary conditions

$$q \to 0$$
 and $\frac{dq}{d\eta} \to 0$ for $\eta \to \pm \infty$, (11)

are possible only for $\alpha > 1$. A solution of this kind is

$$q = \frac{3}{2}(\alpha - 1)\mathrm{sech}^{2}\left[\frac{1}{2}(\alpha - 1)^{1/2}\eta\right]. \tag{12}$$

By using (9), we obtain

$$\mathscr{E} = \frac{3}{2}\alpha(\alpha - 1)\mathrm{sech}^2 \left[\frac{1}{2}(\alpha - 1)^{1/2} \eta \right]. \tag{13}$$

A similar stationary solution of the system (1) and (3) can be found for comparison without using the unidirectional wave approximation. This exact solution is found in the same way as (12) and (13). Assuming that $\mathscr E$ and q depend exclusively on the variable $\eta = \omega_0(t-z/V)$, we can rewrite Eqn (1) in the form of an ordinary second-order differential equation. By integrating this equation twice taking the boundary conditions (11) into account, we obtain

$$\mathscr{E}(\eta) = \tilde{\alpha} g(\eta),\tag{14}$$

where $\tilde{\alpha} = V^2(\omega_{\rm p}/\omega_0)^2(c^2-V^2)^{-1}$. This relationship is similar to (9). Therefore, the final solution of the system of equations (1) and (3) determines the stationary pulse of the electromagnetic field

$$\mathscr{E} = \frac{3}{2}\tilde{\alpha}(\tilde{\alpha} - 1)\mathrm{sech}^2\left[\frac{1}{2}(\tilde{\alpha} - 1)^{1/2}\eta\right],\tag{15}$$

which differs from expression (13) in only the definition of the parameter α . Attempts to analytically find more general solutions have not met with success. Subsequent investigations of the propagation and interaction of waves in a quadratically nonlinear medium using the model under consideration in the unidirectional wave approximation were carried out numerically.

4. Numerical treatment of the propagation of electromagnetic radiation pulses

To study the solutions of system (4) numerically, we will write it in the form:

$$\frac{\partial \mathscr{E}}{\partial \zeta} = -p,\tag{16.1}$$

$$\frac{\partial q}{\partial \tau} = p, \quad \frac{\partial p}{\partial \tau} = \mathscr{E} - q - q^2. \tag{16.2}$$

Since Eqn (16.1) contains only the derivative with respect to the ζ coordinate, while Eqn (16.2) only with respect to τ , we can use any methods of numerical integration of ordinary differential equations by applying it in turn to Eqn (16.1) and to Eqn (16.2). Here, we used the predictor-corrector method to solve Eqn (16.2) over the entire τ -axis and the fourth-order Runge–Kutt method to integrate Eqn (16.1) by one step in the ζ -axis.

We adopted the following initial and boundary conditions:

$$\mathscr{E}(\zeta=0,\tau)=\mathscr{E}_0(\tau),\ \ \lim_{|\tau|\to\infty}q(\zeta,\tau)=\lim_{|\tau|\to\infty}p(\zeta,\tau)=0.$$

With these boundary conditions, two integrals can be found:

$$I_0 = \int_{-\infty}^{\infty} \mathscr{E}(\zeta, \tau) \mathrm{d} \tau, \quad I_1 = \int_{-\infty}^{\infty} \mathscr{E}^2(\zeta, \tau) \mathrm{d} \tau.$$

When the stationary solution (13) characterised by the parameter $\alpha = 5$ (i.e. $\mathcal{E}_0(\tau) = 30 \text{sech}^2(\tau)$) was selected as the initial condition for \mathcal{E} , this pulse was shown to propagate without distortion up to the coordinate value $\zeta = 130$, which would be expected. This may also serve as a test of the code for numerical solution of the system of equations (16).

The solutions of system (16) for small-amplitude initial pulses exhibit a dispersion spreading of the initial pulse accompanied by the formation of harmonic waves. If a pulse with an energy exceeding that of a stationary pulse enters the medium, it will break down into a series of pulses, each propagating, like stationary pulses, with a velocity of its own.

Completely integrable evolutionary nonlinear equations are characterised by the elastic interaction of stationary solitary waves, which are solitons in this case. The system (16) under consideration is unlikely to belong to the class of completely integrable systems. Nevertheless, a collision of two stationary pulses having different propagation velocities has shown them to be rather stable in collisions. We also considered a collision of two pulses produced in the decay of a pulse with the initial field profile $\mathcal{E}_0(\tau) = 30 \mathrm{sech}^2(\tau/2)$. A pair of solitary waves originated after the collision. The propagation velocities of the waves were changed but their amplitudes and lengths were conserved within the limits of computational error.

Of considerable interest is the stability of nonlinear waves with respect to continuous perturbations, e.g., to a regular modulation of the envelope. It is known that solitons with this modulation can transform back to pulses with a smooth envelope. However, when the modulation is deep enough, a soliton may decay and turn into spreading wave packets.

We considered two types of amplitude modulation by a harmonic wave — the additive and multiplicative ones. In the former case, a stationary pulse was found to propagate against the background of a continuous harmonic wave and to preserve its shape. Because of the dispersion and nonlinearity of the medium, a harmonic wave decays into low-amplitude wave packets, which spread out during their propagation. A stationary pulse of the higher intensity remains unmodulated and propagates independently of weak harmonic wave packets. A consideration of the evolution of the Fourier spectrum of a modulated stationary pulse reveals that the low-frequency part of the spectrum, which corresponds to the smooth stationary pulse, is very weakly

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changed. The peak in the spectrum at the frequency of the additive harmonic wave is somewhat broadened and gradually decreases with the distance travelled through the medium. Fig. 1 shows the evolution of a modulated signal and its spectrum for the initial profile:

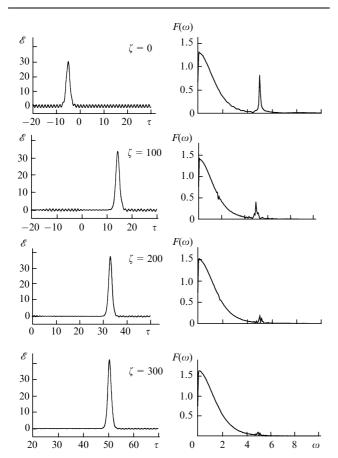


Figure 1. Evolution of a stationary pulse and its spectrum upon the additive modulation of the pulse amplitude by a harmonic wave.

$$\mathcal{E}_0(\tau) = 30 \text{sech}^2(\tau + 5) + \sin(5\tau).$$

A pulse with an energy twice as high as that of a stationary pulse was previously found to decay into two pulses that behave like stationary ones. It turned out that the additive modulation by a harmonic wave did not prevent the decay. Note that the evolution of the spectrum of a modulated powerful pulse is more complex. Apart from the separate peak corresponding to the harmonic wave, the low-frequency part of the spectrum acquires modulation. This modulation is caused by the interference of the Fourier transforms $F_{1,2}(\omega t_{1,2})$ of the two signals with durations t_1 and t_2 produced upon the decay of the powerful initial pulse, and propagating with velocities v_1 and v_2 , respectively. The Fourier transform of the sum of these pulses is

$$F(\omega,\zeta) = t_1 F_1(\omega t_1) \exp(i\omega \zeta/v_1) + t_2 F_2(\omega t_2) \exp(i\omega \zeta/v_2).$$

The power spectrum is proportional to $|F(\omega)|^2$, which results in the appearance of an interference term proportional to $\cos[\omega(v_1^{-1}-v_2^{-1})\zeta]$. Fig. 2 shows the decay of the pulse with the initial profile

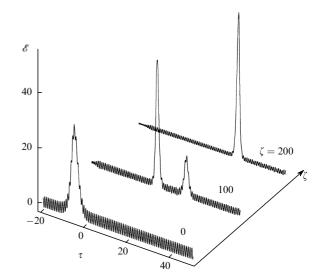


Figure 2. Decay of a pulse, having an energy exceeding that of a stationary pulse, into separate pulses.

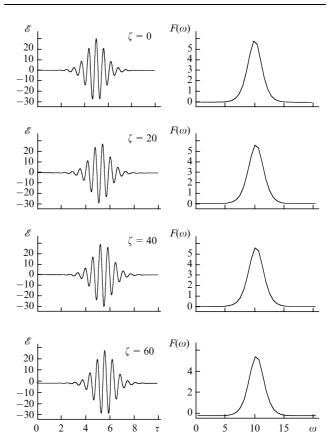


Figure 3. Stability of the envelope shape and of the spectrum of a pulse with the initial profile $\mathscr{E}_0(\tau) = 30 \mathrm{sech}^2(\tau - 5) \cos(10\tau)$.

$$\mathcal{E}_0(\tau) = 30 \operatorname{sech}^2((\tau + 5)/2) + 2 \cos(15\tau)$$

upon the additive modulation.

The case of multiplicative modulation of a stationary pulse was considered by the example of solution of the system of equations (16) with the initial profile of the form

$$\mathscr{E}_0(\tau) = 30 \operatorname{sech}^2(\tau) \cos(\Omega \tau),$$

where Ω is the normalised modulation frequency whose values were selected in the range from 10 to 30. An initial pulse of this type was found to behave like a stationary one; however, it remains to be modulated. Its spectrum is localised in the vicinity of the modulation frequency, as in the case of a quasiharmonic signal (Fig. 3).

Fig. 4 shows the evolution of a weak signal with the initial profile

$$\mathcal{E}_0(\tau) = 30 \operatorname{sech}^2(\tau) \sin(2.5\tau - 5)$$
,

modulated by a harmonic wave.

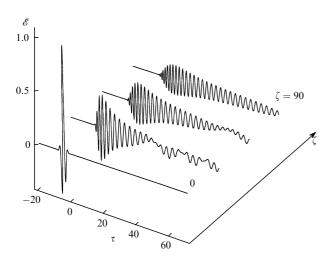


Figure 4. Decay of a weak pulse modulated by a harmonic wave.

During its propagation, a modulated weak signal experiences dispersion spreading (as does an unmodulated signal having an energy lower than that of a stationary pulse) and turns into a quasiharmonic solitary wave with a slowly varying envelope.

5. Discussion

We have considered the simplest model of a nonlinear medium which was used previously to determine the response of a quadratically nonlinear medium [37, 38] (see also Refs [31, 35, and 36]). Within the framework of this model, Garrett and Robinson [38] and Miller [39] defined a quantity referred to as the Miller index, which allows one to express the nonlinear susceptibilities in terms of the linear ones. For several materials (KDP, CdS, Te, GaAs, etc.), the Miller index varies within an order of magnitude [39], and its average value can be calculated using the anharmonic oscillator model (2) for a specific choice of the anharmonicity constant (Yariv [35] found that $\kappa_2 \approx -1.64 \times 10^{41}$ m⁻¹ s⁻²). This justifies the choice of the model discussed above to describe the evolution of electromagnetic pulses in the media where the main contribution to the nonlinear response is due to electrons. A natural generalisation of the model should be analysis of the electron-vibrational interaction [23, 26] and consideration of the evolution of the polarisation vector of the electromagnetic wave [31].

The unidirectional wave approximation used here greatly simplifies the problem from the point of view of numerical solution of the system of equations for the pulse propagation, but at the same time it imposes limitations on the range of validity of the results. The unidirectional-wave criterion discussed in the Introduction is fulfilled (as already noted in Refs [33, 34]) in low-density media. Therefore, the results obtained do not apply in the case of dielectrics and semiconductors. The role of appropriate media may be played by impurities in a dielectric matrix, a molecular layer on the surface of a dielectric, or a system of microcrystallites (quantum dots), provided that their spatial density is low. The anisotropy required for the quadratic nonlinearity of this gas-like medium may be caused by either an external field (electric, magnetic) or a substrate, when surface electromagnetic waves are considered. Quasi-one-dimensional dielectrics and semiconductors (e.g., polyacetylene) that contain low-density impurities can serve as media in which the unidirectional-wave criterion is fulfilled.

An investigation of the simple model of short-pulse propagation through a quadratic medium, performed in the unidirectional-wave approximation, showed the existence of a one-parameter family of stationary solitary waves corresponding to USPs. A numerical simulation of the propagation revealed that collisions of two stationary pulses do not lead to their decay. Pulses of higher intensity (different from the stationary ones) were found to decay into several separate pulses traveling with velocities of their own. These pulses retain their individuality throughout the distance in which numerical integration of the system of equation (16) was performed.

Collisions of two pulses, both stationary ones and those produced in the decay of one high-intensity pulse, are associated with a phase shift of each of the colliding pulses. The summary phase shift is not an integral of motion, as would be the case with true solitons. Moreover, the pulse propagation velocities are changed upon the interaction, even if only slightly. Therefore, the stationary pulses discovered in this work are not solitons in the rigorous sense of the word, even though they exhibit a rather high degree of stability. It is valid to say that we have arrived at just one more example of so-called robust solitons discussed in Ref. [40].

In the context of the model under consideration, one would expect the solitary waves in quadratically nonlinear media to behave in this manner when the following circumstances are taken into account. From Eqns (4) it follows that there exists a 'potential' $\phi(\zeta,\tau)$ such that

$$\mathscr{E}(\zeta,\tau) = \frac{\partial \phi}{\partial \tau}, \quad q(\zeta,\tau) = -\frac{\partial \phi}{\partial \zeta}. \tag{17}$$

Substitution of these expressions into the second equation (4) gives

$$\frac{\partial \phi}{\partial \tau} + \frac{\partial \phi}{\partial \zeta} \pm \left(\frac{\partial \phi}{\partial \zeta}\right)^2 + \frac{\partial^3 \phi}{\partial \tau^2 \partial \zeta} = 0. \tag{18}$$

From Eqns (17) and (18) follows the equation for the normalised coordinate of an anharmonic oscillator:

$$\frac{\partial q}{\partial \tau} + \frac{\partial q}{\partial \zeta} \pm 2q \frac{\partial q}{\partial \zeta} + \frac{\partial^3 q}{\partial \tau^2 \partial \zeta} = 0. \tag{19}$$

This equation resembles the known Korteweg-de Vries (KdV) equation [41]

$$\frac{\partial q}{\partial \tau} + \frac{\partial q}{\partial \zeta} \pm 6q \frac{\partial q}{\partial \zeta} + \frac{\partial^3 q}{\partial \zeta^3} = 0, \tag{20}$$

but differs from the latter by the last term and, as a con-

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sequence, by the linear dispersion law. The stationary solution of the system of equations (16) coincides in form with the soliton solution of the KdV equation while the I_0 and I_1 integrals of motion coincide with its first two integrals. Therefore, it is likely that the solutions of the system (16) are close to true solitons while Eqn (19) forms the basis for the development of the perturbation theory, which will permit to approximately describe the evolution of USPs in nonlinear media of the type considered in this work. However, it should be borne in mind that the evolution proceeds in the ζ -variable, unlike the case of a regularised equation for long waves [42–44]

$$\frac{\partial q}{\partial \tau} + \frac{\partial q}{\partial \zeta} \pm 2q \frac{\partial q}{\partial \zeta} - \frac{\partial^3 q}{\partial \tau^2 \partial \zeta} = 0, \tag{21}$$

and the case of the KdV equation (20), where the role of ζ is played by the τ variable.

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