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On the nonlinear optics of extremely short pulses

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A brief scientific-methodological review of theoretical works and approaches created in the nonlinear optics of extremely short pulses from the 1970s to the present is presented. Important features are emphasized that distinguish the nonlinear optics of extremely short pulses from the nonlinear optics of quasi-monochromatic signals.

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Short and ultrashort pulses

With the advent of pulsed lasers in the 1960s, one of the main trends in the development of nonlinear optics and laser physics was the creation of light pulses of ever shorter durations under laboratory conditions. In turn, this caused the rapid development of the theory of the interaction of short laser pulses with matter.

The development of Q-switched and mode-locked lasers led to the generation of nanosecond optical pulses. Such pulses began to be called as short pulses. The point here is that the durations of these pulses turned out to be comparable or even shorter than the characteristic times of irreversible relaxation of the medium, which determine the rates of irreversible losses of the energy acquired by the medium. Therefore, it became possible to study fast intraatomic processes. As a result, such a resonant effect as self-induced transparency (SIT) [1,2] was discovered. The resonance here is that the carrier frequency of the optical pulse is very close to the frequency of one of the quantum transitions of the medium. The SIT effect is interesting, among other things, because it is accompanied by the propagation of a resonant optical soliton i.e. a stable solitary-pulse. It was the first optical soliton that was observed under experimental conditions and was described theoretically.

In theoretical studies for the electric field of a laser pulse, the wave equation is used, which follows from the Maxwell equations:

$$\nabla^2\mathbf{E} - \frac{1}{c^2}\frac{\partial^2\mathbf{E}}{\partial t^2} - \nabla(\nabla\cdot\mathbf{E}) = \frac{4\pi}{c^2}\frac{\partial^2\mathbf{P}}{\partial t^2}. \quad (1)$$

Here c is speed of light in vacuum, $\mathbf{P}$ is polarization response of the medium.

For the response $\mathbf{P}$, constitutive equations are written. Their appearance depends on the choice of a particular model of the medium, which in each particular case can be different and is determined by physical considerations.

Equation (1) and constitutive equations for the response of the medium constitute a self-consistent nonlinear system that describes the dynamics of the medium and the pulse propagating in it.

For a purely transverse wave, equation (1) can be simplified by considering in it

$$(\nabla\cdot\mathbf{E}) = 0. \quad (2)$$

Below we will return to this condition, which is consistent with the Gauss theorem at zero density of free and bound charges.

The carrier frequency of a short optical pulse in the visible range is $\omega \sim 10^{15}\,\mathrm{s}^{-1}$. Moreover, its duration is $\tau_p \sim 10^{-9}$ s. The number of light vibrations that such a pulse can contain is of the order of $N \sim \omega\tau_p \sim 10^6$. This circumstance allows us to introduce a small parameter

$$\delta_1 = \frac{1}{N} \sim \frac{1}{\omega\tau_p} \ll 1. \quad (3)$$

Owing to this small parameter, it is possible to determine, in the general case, the complex slowly-varying envelope (SVE) ψ of the electric field of a linearly polarized pulse:

$$\mathbf{E}(\mathbf{r},t) = \mathbf{e}E = \mathbf{e}\psi(\mathbf{r},t)e^{i(\omega t - kz)} + c.c. \quad (4)$$

Here $\mathbf{e}$ is the unit vector in the direction of the pulse polarization, $\mathbf{r}$ is the radius vector of the observation point inside the medium, z is axis along which the pulse propagates, k is projection of the wave vector onto the z axis.

The SVE approximation corresponds to the fact that the envelope ψ noticeably changes at times of the order of τ_p, which significantly exceed the period of optical oscillations

$T \sim 1/\omega$, which can be formally written as (3) . This allows us to neglect the second derivatives of ψ with respect to z and t on the left side of the wave equation (1). As a result, the wave equation for ψ is of the first order with respect to the derivatives with respect to z and t. This greatly facilitates theoretical studies. In turn, the neglect in constitutive equations (including for the polarization response of the medium) of rapidly oscillating terms at frequencies of the order of ω in comparison with terms that change noticeably at times of the order of τ also significantly simplifies the constitutive equations .

The resonant nature of the SIT effect makes it possible, with good accuracy, to take into account only one quantum transition of the medium, which is in resonance with the laser pulse (two-level medium approximation). The system of wave and material equations describing the SIT is called the Maxwell–Bloch (MB) [3,4] system. If the carrier frequency of the laser pulse exactly coincides with the frequency of the excited quantum transition, the MB system in the one-dimensional case reduces to the sine–Gordon (SG) equation for „the area“ of the pulse — the time integral of the envelope of its electric field, $\theta \sim \int\limits_{-\infty}^{t} \psi dt$ [3,4]:

$$\frac{\partial^2 \theta}{\partial z \partial \tau} = -\alpha \sin\theta. \quad (5)$$

Here, the running time $\tau = t - z/v_g$, v_g is the linear group velocity of light corresponding to the carrier frequency of the pulse, α is a constant coefficient determined by the parameters environment.

Equation (5) has a well-known soliton solution, as well as multisoliton solutions, which describe elastic interactions between solitons of different amplitudes, durations and velocities [3]. In addition, the SG equation has breather solutions. A breather can be considered as a bound state of two solitons with the same group but different phase velocities. As a result, the breather is a localized pulse propagating at a constant speed, the profile of which periodically changes („breathes“) in the comoving coordinate system.

What has been said above about the solutions of the SG equation can also be fully applied to the MB system. Both SG and MB are integrable nonlinear systems in the sense that boundary problems can be analytically solved for them, and not only many exact solutions can be found. This is a non-trivial fact for nonlinear systems.

It is important to note that the velocities and amplitudes of the SG and MB solitons increase with a continuous shortening of their time duration τ_p.

In the 1970s, one taked a leap in the field of picosecond laser optics. Accordingly, the intensity of the generated signals, called ultrashort pulses, increased. Therefore, there appeared a real possibility of strong excitation of the medium not only by resonant, but also by nonresonant pulses. It was a jump-start to the development of nonlinear nonresonant optics. For picosecond pulses, the parameter δ_1 is still small, on the order of 10^{-3}. Such intense non-resonant signals lead to a nonlinear modification of the refraction index of the medium [5,6]

$$n \to n + n_2|\psi|^2, \quad (6)$$

where n_2 is so-called non-linear refraction index.

For $n_2 > 0$ the nonlinearity is focusing, otherwise is defocusing one.

The envelope ψ of such pulses obeys the nonlinear Schrödinger equation (NSE) [5,6]

$$i\frac{\partial \psi}{\partial z} = -\frac{k_2}{2}\frac{\partial^2 \psi}{\partial \tau^2} + a_1|\psi|^2\psi, \quad (7)$$

where $k_2 = \partial^k/\partial\omega^2 = \partial v_g^{-1}/\partial\omega$ is parameter group velocity dispersion (GVD) of the second order, the nonlinearity coefficient a_1 is proportional to n_2.

The nonresonant cubic nonlinearity in (7) is often called as „Kerr nonlinearity“.

NSE, like SG and MB, turns out to be integrable, having soliton solutions [3] for $k_2 a_1 < 0$. Moreover, in contrast to SG and MB solitons, the velocity of NSE solitons is equal to the linear group velocity of light v_g and does not depend on their amplitude and duration.

In the case of a focusing nonlinearity ($a_1 > 0$), NSE solitons are formed in the spectral range where the GVD is negative (anomalous). And vice versa.

By the 1980s, pulses with a duration of several tens of femtoseconds had been generated. Such pulses are called as ultrashort pulses. For them $\delta_1 \sim 10^{-2}$–10^{-1}. Despite the fact that this parameter is still small, it is nevertheless much larger than in the case of short and ultrashort pulses. Therefore, equation (7) has to be modified by adding higher-order derivatives to it [7,8]:

$$i\frac{\partial \psi}{\partial z} = -\frac{k_2}{2}\frac{\partial^2 \psi}{\partial \tau^2} - i\frac{k_3}{3!}\frac{\partial^3 \psi}{\partial \tau^3} + i\frac{k_4}{4!}\frac{\partial^4 \psi}{\partial \tau^4} + i\frac{k_5}{5!}\frac{\partial^5 \psi}{\partial \tau^5} + a_1|\psi|^2\psi + ia_2\frac{\partial}{\partial \tau}\left(|\psi|^2\psi\right) - a_3\frac{\partial^2}{\partial \tau^2}\left(|\psi|^2\psi\right) - ia_4\frac{\partial^3}{\partial \tau^3}\left(|\psi|^2\psi\right) + \ldots \quad (8)$$

Here k_j and a_j are respectively the parameters of the GVD and j-order nonlinear dispersion ($j = 2, 3, 4, \ldots$).

Each higher order derivative in (8) is a subsequent expansion term in the still small parameter δ_1.

It is important to note once again that for short, ultrashort, and ultrashort pulses, the number of oscillations satisfies the condition $N \gg 1$. Consequently, the spectra of these pulses are rather narrow, since the inequality $\delta\omega \ll \omega$ is valid for their widths $\delta\omega \sim 1/\tau_p$. Thus, these signals in the spectral sense are quasi-monochromatic pulses (QMP).

With a further shortening of the laser pulse durations, in (8) it is necessary to take into account the linear and nonlinear dispersions of ever higher orders In addition, as the intensities of the generated signals increase, higher-order nonlinearities become significant. All this leads to a significant complication of equation (8) and suggests the need to use a fundamentally new approach.

Extremely Short Pulses

For pulses consisting of one period of optical oscillations, the value of the parameter δ_1 becomes about unity. In this case δ_1 is no longer a small parameter, and the expansion in its powers becomes incorrect. Therefore, adding new terms to equation (8) will not lead to an adequate description of the pulse dynamics in a nonlinear medium. Here it becomes necessary to search for a fundamentally new theoretical approach.

Pulses containing the order of one period of electromagnetic oscillations in the domestic literature are called Extremely Short Pulses (ESP) [9]. Recently, the term „low-cycle pulses“ [10] has been applied to such signals. The term „few-cycle pulses“ [11,12] has firmly taken root in the English literature. The duration of a ESP in the optical range is on the order of a few femtoseconds.

At the turn of the 1980s and 1990s, successful experiments were carried out on the generation of ESPs in laboratory conditions. Although, two even earlier experimental works on the generation of ESPs in the infrared [13] and subterahertz ranges [14] by optical rectification should be noted. In this connection, we will apply the term ESP to all single-period pulses, regardless of their absolute duration. As a rule, the duration of such signals lies in the range from pico- to units of femtoseconds. Attosecond pulses that cause ionization processes in a nonlinear medium will not be discussed here. Below, the focus of our attention will be on the interaction of ESPs with nonconducting dielectric media.

To construct theoretical schemes and methods that make it possible to describe the propagation of USPs in dielectrics, it is necessary to return to the wave equation (1), abandoning the SVE approximation. In the constitutive equations, one must also abandon the concept of envelopes of the components of the polarization response.

As usual, theoretical studies are ahead of experimental ones, sometimes anticipating them by several decades.

The refusal to use the SVE approximation was made in the early 70s of the last century. We note the theoretical works [15–17], where this failure was made when describing the SIT effect. Let us emphasize the approximation of unidirectional propagation (UP), with the help of which the wave equation is reduced from the second order to the first one [16]. Let us illustrate the use of the UP approximation by the example of equation (1) with allowance (2). For simplicity, let's restrict ourselves „to the scalar“ case, assuming the field to be linearly polarized. Therefore, from vectors **E** and **P**, let's move on to scalar quantities E and P. Then, having singled out the predominant momentum propagation along the z axis, we rewrite (1) in the form

$$\left(\frac{\partial}{\partial z}-\frac{1}{c}\frac{\partial}{\partial t}\right)\left(\frac{\partial E}{\partial z}+\frac{1}{c}\frac{\partial E}{\partial t}\right)=\frac{4\pi}{c^2}\frac{\partial^2 P}{\partial t^2}-\nabla_\perp^2 E. \quad (9)$$

We will assume that the right side in (9) is relatively small. This is possible if both terms on the right-hand side are small. The smallness of the first term corresponds to a low concentration of atoms actively interacting with the laser pulse field. In turn, this means that the value of the refractive index of the medium is close to unity:

$$\delta_2 = |n-1| \ll 1. \quad (10)$$

The second term is small if we use the paraxial approximation to describe the momentum dynamics in directions transverse to the z axis.

Let us neglect the right side in (9) in the zeroth approximation and take into account only the wave propagating along the z axis, discarding the wave propagating in the opposite direction. Then from (9) in the zero approximation we have $\frac{\partial E}{\partial z}+\frac{1}{c}\frac{\partial E}{\partial t}=0$ or $\frac{\partial}{\partial t}=-\frac{1}{c}\frac{\partial}{\partial t}$. Using this approximate equality in the first left bracket of equation (9), we write

$$\frac{\partial}{\partial t}\left(\frac{\partial E}{\partial z}+\frac{1}{c}\frac{\partial E}{\partial t}\right)=-\frac{2\pi}{c}\frac{\partial^2 P}{\partial t^2}+\frac{c}{2}\nabla_\perp^2 E,$$

where $\nabla_\perp^2$ is the transverse Laplacian.

Integrating over time, we have

$$\frac{\partial E}{\partial z}+\frac{1}{c}\frac{\partial E}{\partial t}=-\frac{2\pi}{c}\frac{\partial P}{\partial t}+\frac{c}{2}\nabla_\perp^2\int_{-\infty}^{t} E\,dt'. \quad (11)$$

Equation (11) describes the propagation of a laser pulse in a rarefied medium (see (10)) along the z axis at a velocity close to the speed of light in vacuum, taking into account its dynamics in transverse directions in the paraxial approximation. It is important to note that the reduction of the wave equation from the second order to the first occurred due to the use of the UP approximation, and not the SVE. Therefore, equation (11) is valid both for QMP and for pulses with an arbitrarily small number of optical oscillations, including ESP. It would be worth emphasizing that such a reduction was used back in the 60s of the last century when solving problems in nonlinear acoustics [18,19], where the role of the speed of light was played by the linear speed of sound. In those problems, the UP approximation was called the slowly varying profile approximation (SVP), which should not be confused with the SVE approximation. The origin of this name is easy to understand if once again we take into account that the reduction of the wave equation to the first order is possible under the assumption that the wave velocity is very close to the linear velocity. Therefore, in the comoving reference frame, the profile of this wave (not the envelope!) changes very slowly.

In the work [16] one-dimensional version of equation (11) was considered when $\nabla_\perp^2 = 0$. Supplementing (11) with constitutive Bloch-type equations for a medium of two-level atoms led to a new nonlinear integrable system called the reduced Maxwell–Bloch system (RMB).

The RMB system has both soliton (unipolar) and breather solutions for the electric field E of the pulse [4]. In this case, unipolar solitons have one continuous free parameter, for which one can choose, for example, the time duration τ_p. Breathers are two-parameter solutions. Taking for them τ_p and the center frequency ω of the spectrum as free parameters, one can analyze the situation for various ratios between these parameters. For example, at $\omega\tau_p \gg 1$ the breather solution transforms into a soliton of the SIT envelope. However, it should be remembered that the speed of such a soliton is close to the speed of light in vacuum. At the same time, the speed of the SIT soliton can be hundreds and thousands of times less than this speed. Thus, the fact that the UP approximation makes it possible to write the first-order wave equation for the electric field itself, and not for its envelope, is an indisputable advantage of this approximation over the SVE approximation. On the other hand, the SVE approximation can be used to describe resonant SIT solitons, whose speed can be much less than the speed of light in vacuum The UP approximation does not allow this here.

If we put $\omega\tau_p \sim 1$ in the breather solution of the RMB system, then we will arrive at a ESP with a periodically changing profile in the accompanying coordinate system.

In the early 70s of the last century, theoretical works [15,16] were mainly of academic interest. An urgent need for such studies arose at the turn of the 1980s and 1990s, when ESPs were generated under experimental conditions [20–23].

Here one should single out the theoretical works [24–26], in which both the SVE (3) approximation and the approximation of a low medium density (10) were not used. Instead of them, in the nonlinear optics of ESP, it was proposed to use the approximations of sudden perturbations (SPs)

$$\delta_3 = \omega_0\tau_p \ll 1 \qquad (12)$$

and optical transparency (OT)

$$\delta_4 = (\omega_0\tau_p)^{-1} \ll 1. \qquad (13)$$

Note that the (12) approximation was proposed by A.B. Migdal [27] when solving problems related to particle collisions in atomic physics. As applied to optics, condition (12) means that the pulse spectrum overlaps the involved quantum transition with a frequency ω_0. Indeed, in this case the spectral pulse width is $\delta\omega \sim 1/\tau_p \gg \omega_0$. Therefore, one should expect a strong excitation of the medium, accompanied by significant changes in the populations of stationary quantum states. This, in turn, means a strong manifestation of the nonlinear optical properties of the medium. In addition, with such short pulsed impacts, the role of dispersion is very large. Therefore, favorable conditions are created here for the formation of solitons.

As shown in [24], under condition (12) the ESP dynamics is described by an SG equation of the form (5). Only here, in contrast to QMP, the truncated „area“ θ is the integral not of the envelope ψ, but of the electric field E of the pulse itself: $\theta \sim \int_{-\infty}^{t} E dt'$. Therefore, mathematically similar solutions are now written not for the envelope, but for the pulse field. Here there are both solutions in the form of unipolar solitons and breathers [3].

Note that condition (12) was used in a number of problems related to the effects of ESP on various media [28–33]. At the same time, attention was focused on the fact that in this approximation, it is the electric area of the pulse $S = \int_{-\infty}^{\infty} E dt \sim \theta_{t\to\infty}$ that determines the result of its action on various quantum objects. The electric area of a single-period ESP is equal to zero. Therefore, the result of its impact on the environment also turns out to be zero. In order for the action to be nonzero, the pulse must have the properties of a unipolar signal, for which $S \neq 0$.

In contrast to (12), condition (13) corresponds to weak excitation of the medium and weak dispersion. In this case, the ESP dynamics is described by the modified Korteweg-de Vries (MKdV) equation [25,26]

$$\frac{\partial E}{\partial z} + \frac{n}{c}\frac{\partial E}{\partial t} - aE^2\frac{\partial E}{\partial t} - b\frac{\partial^3 E}{\partial t^3} = 0. \qquad (14)$$

We emphasize once again that here the refractive index n, generally speaking, does not satisfy condition (10).

Equation (14), like the SG equation, has solutions in the form of unipolar solitons and breathers [3].

Note that the existence of one-dimensional solutions of the type of unipolar solitons was also discussed in [34].

The model of a two-level medium used in the derivation of the SG and MKdV equations is generally unsatisfactory. As mentioned above, under condition (12), the ESP spectrum significantly overlaps the involved quantum transition. Therefore, this spectrum should cover other quantum transitions as well. To prevent this from happening, it should be assumed that the two quantum states under consideration should be significantly removed on the energy scale from other stationary states.

Under condition (13), the two-level model implies a uniquely negative value of the nonlinear refractive index n_2, determined according to (6). At the same time, this value is usually positive in the spectral region of the transparency of solids. The Kerr nonlinearity in the two-level model appears solely due to the change in the populations of the stationary quantum states. In the general case, this is clearly not enough. This is another shortcoming of the two-level environment model.

Equation (14) can be obtained under condition (13) using a classical oscillator with cubic non-linearity (Duffing oscillator) [35–38] as a constitutive equation. As was shown in the work [39], this model generally does not correspond to the experimental dispersion law of the nonlinear refractive index. This is especially true for spectral regions near resonances. Therefore, instead of the Duffing oscillator, a classical empirical model of two nonlinearly coupled oscillators was proposed in [39], which satisfactorily describes the noted dispersion law. In the work [40] this model was used to simulate the electron-optical nonlinearity of femtosecond MPSR in wide-gap dielectrics. Since the characteristic values of the eigenfrequencies of electron-optical transitions are of the order of $\omega_0 \sim 10^{16}\,\mathrm{s}^{-1}$, for a ESP of duration $\tau_p \sim 10^{-15}$ s condition (13) is satisfied. For the vibrational optical modes participating in the interaction with MPCP at the sites of the crystal lattice, we used the usual model of the classical Lorentz oscillator satisfying condition (12), since here the characteristic eigenfrequencies $\omega_0 \sim 10^{13}\ \mathrm{s}^{-1}$. As a result, for the electric field MPCP an equation of the form [40]

$$\frac{\partial E}{\partial z} + \frac{n}{c}\frac{\partial E}{\partial t} + aE^2\frac{\partial E}{\partial t} - b\frac{\partial^3 E}{\partial t^3} + g\int\limits_{-\infty}^{t} E dt' = \frac{c}{2n}\nabla_\perp^2 \int\limits_{-\infty}^{t} E dt'. \qquad (15)$$

Note that all coefficients in (15) are positive, which corresponds to the parameters of wide-gap dielectrics.

Neglecting the response of the vibrational modes of the crystal, when $g = 0$, (15) in the one-dimensional case ($\nabla_\perp^2 = 0$) passes into an MKdV with a focusing nonlinearity and a normal GVD ($a, g > 0$). In this case, one cannot speak of the formation of solitons or breathers. Let us make a reservation right away that here we are talking only about „bright" solitons, the field of which vanishes at infinity. Dark solitons [6] will not be discussed here. The situation can be changed only by the presence of vibrational modes ($g \neq 0$). Numerical experiments carried out with equation (15) in the one-dimensional case show that it has solutions in the form of a breather-type MPCP with a duration of approximately one and a half periods of optical oscillations [40]. However, it has no solutions in the form of unipolar solitons [40].

If the pulse spectrum lies closer to the frequencies of the vibrational infrared modes of the crystal than to the visible frequencies of electron-optical transitions, then the dispersion created by these transitions can be neglected by putting in (15) $b\partial^3 E/\partial t^3 = 0$. Then in the one-dimensional case from (15) we arrive at the Shäfer−Wayne equation [41]

$$\frac{\partial^2 E}{\partial z \partial t} + \frac{n}{c}\frac{\partial^2 E}{\partial t^2} + \frac{a}{3}\frac{\partial^2}{\partial t^2}\left(E^3\right) + gE = 0. \qquad (16)$$

In [42,43] it is shown that the equation (16) is integrable and has breather solutions of the MPCP type.

In the works [44,45], a two-component model of a medium was considered, containing two kinds of two-level atoms with strongly different transition frequencies, which satisfy conditions (12) and (13), respectively. As a result, a generalization of equation (15) is obtained, which consists in replacing $g\int_{-\infty}^{t} E dt' \to (g/\mu)\sin\left(\mu\int_{-\infty}^{t} E dt'\right)$, where the constant μ is determined by the characteristics of the quantum transition that satisfies the BB (12) condition. Obviously, in the $\mu \to 0$ limit, we have a transition to equation (15), when the nonlinearity of the response of the transition overlapped by the MPCP spectrum can be neglected.

The two-component model of the medium considered in [44,45] describes, for example, electron-optical (satisfying (13)) quantum transitions and tunneling (satisfying (13)) transitions remote from them.

Accounting for the partial capture of states remote to tunnel transitions in the interaction with MPCP was made in the works [46,47]. As a result, a generalization of the SG equation is obtained:

$$\frac{\partial^2 \theta}{\partial z \partial t} + \left(\frac{n}{c} - 4\beta\sin^2\frac{\theta}{2}\right)\frac{\partial^2\theta}{\partial t^2} = -\left[\alpha - \beta\left(\frac{\partial\theta}{\partial t}\right)^2\right]\sin\theta. \qquad (17)$$

Here β is the coefficient describing the admixture of distant quantum states to the overlapped MPCP spectrum.

Equation (17) turned out to be integrable [47]. It has both unipolar soliton solutions and breather-type solutions [47]. If $\beta < 0$, the unipolar solitons of equation (17) look taller and sharper than the solitons of the SG equation. Otherwise, the solitons, on the contrary, are blunted. As the positive value β increases, the profile of a unipolar soliton tends to a rectangular shape with an extremely short duration $\tau_p^{\min} = 2\pi\sqrt{\beta/\alpha}$ [46]. The breathers in this case have the form of localized profiles with oscillations not of a sinusoidal, but of a rectangular type [47].

Above, attention was paid to the propagation of MPCP in optically isotropic media; the polarization response of the medium consists of expansion only in odd powers of the pulse electric field. In this connection, one should note the works [48–50], where the soliton dynamics of unipolar and single-period MPCP, including the generation of harmonics [50], was studied. In the work [51] a vector system of two equations was obtained for the ordinary and extraordinary components of a MPCP in a uniaxial crystal interacting nonlinearly with each other. This system is a direct generalization of equation (15) and makes it possible, in particular, to study the generation of terahertz radiation in a square-nonlinear crystal using a femtosecond optical pulse. In this connection, one should return to experimental works [13,14], where terahertz MPCP were generated in quadratically nonlinear crystals by the method of optical rectification. To do this, an optical picosecond QMP was applied to a uniaxial crystal. Due to the quadratic nonlinearity in the crystal, among other things, the optical pulse was rectified, accompanied by the generation of a broadband terahertz signal consisting of only one oscillation period. The profile of the electric field E_T of this signal is determined by the profile of the envelope ψ of the optical pulse. From a theoretical perspective the interest here also lies in the fact that the SVE approximation is used for the optical pulse, and the UP approximation for the terahertz pulse. In the simplest case, the self-consistent dynamics of the above process is described by a nonlinear integrable system of Yajima–Oikawa equations [51,52]

$$i\left(\frac{\partial\psi}{\partial z}+\frac{1}{v_g}\frac{\partial\psi}{\partial t}\right)=-\frac{k_2}{2}\frac{\partial^2\psi}{\partial t^2}+\sigma E_T\psi, \tag{18}$$

$$\frac{\partial E_T}{\partial z}+\frac{n_T}{c}\frac{\partial E_T}{\partial t}=-q\frac{\partial}{\partial t}\left(|\psi|^2\right). \tag{19}$$

Here n_T is the terahertz refractive index, σ and q are coefficients proportional to the components of the second-order nonlinear optical susceptibility.

It was noted above that the concept of MPCP is applicable to pulses of various absolute durations. It is important that they contain about one period of electromagnetic oscillations. It is this condition that the generated terahertz component E_T measures up.

Generation is most efficient when the equality $v_g = c/n_T$, which is called as „Zakharov–Benney (ZB) [4] condition" in the theory of nonlinear waves, is satisfied. It is very difficult to satisfy this condition in real crystals, since usually $v_g > c/n_T$. Therefore, one has to use optical pulses with oblique wave fronts [53–57]. The phase and group wave fronts of such pulses are not parallel to each other, but form an angle φ between them. Then the ZB- condition takes the form of a Cherenkov-type equality [55]: $v_g \cos\varphi = c/n_T$. The energy efficiency of generation in this way reaches the values $\sim 10^{-4}$–10^{-2}. In [58,59], systems of equations are obtained and analyzed that generalize system (18), (19) to the case of tilted wave fronts of an optical pulse.

System (18), (19) has a soliton solution describing the generation process. In this case, the generated terahertz component is a unipolar one-dimensional soliton [51,52].

A noteworthy point of this solution is that the carrier frequency of the optical pulse is shifted to the red region: $\omega \to \omega - \Omega$, where the value Ω belongs to the terahertz range [51,52]. This fact can be interpreted in such a way that during the generation process, each optical photon gives up part of its energy to the generated terahertz photon. As a result, the frequency spectrum of the optical pulse as a whole shifts to the red region [51]. Such a phenomenon was observed under experimental conditions [54].

At present, significant progress has been made in the generation of both broadband and quasi-monochromatic terahertz signals. Their intensities are such that it is time to talk about the need to develop nonlinear „terahertz optics" [46,60]. In [60] it is shown that the nonlinear refractive index in the terahertz frequency range can be six orders of magnitude higher than this index in the visible frequency range. In this case, the nonlinearity is due to the anharmonicity of the optical vibrational mode of the crystal nodes, and the value of n_2 is expressed in terms of the experimentally measured parameter i.e.- the thermal expansion coefficient of the substance under consideration [61]. Giant values n_2 indicate that nonlinear effects in the terahertz range can manifest themselves at pulsed intensities that are millions of times lower than the corresponding intensities in the visible range.

On the Gauss theorem, electric area and diffraction

Relation (2) is the Gauss theorem at zero densities of free and bound charges. As mentioned above, we do not consider processes associated with ionization here. In addition, the media are assumed to be non-conductive. Therefore, the Gauss theorem for the cases under consideration has the form

$$\nabla\cdot(\mathbf{E}+4\pi\mathbf{P})=0. \tag{20}$$

The density of the bound charge, as is known, is determined by the expression $\rho_b = -(\nabla\cdot\mathbf{P})$.

A nonzero bound charge density arises from the spatial separation of positive and negative charges in the direction of the $\mathbf{E}$ field of the laser pulse (across its propagation). Obviously, $(\nabla\cdot\mathbf{E}) \sim E/D$, where D is the characteristic transverse size (aperture) of the pulse.

In the case of a quasi-monochromatic pulse with a slowly varying envelope in the pulse propagation direction transverse to $\mathbf{E}$, the induced bound charge density changes sign on the wavelength scales λ. In this case $\partial E/\partial z \sim E/\lambda$. As a result, we have $(\nabla\cdot\mathbf{E})/(\partial E/\partial z) \sim \lambda/D$. Thus, relation (2) can be considered satisfied under the condition

$$\frac{\lambda}{D}\ll 1. \tag{21}$$

If the transverse size of the pulse (for example, during self-focusing) becomes of the order of the wavelength, then $\nabla\cdot\mathbf{E}\neq 0$. In this case it is necessary to use the Gauss theorem in the form (20). Here the condition of paraxiality is already violated [62].

In the case of MPCP, as well as unipolar (half-wave) pulses, condition (21) is generalized in an obvious way:

$$\frac{l_\parallel}{D}\ll 1, \tag{22}$$

where $l_\parallel$ is the characteristic size of the pulse in the direction of its propagation.

Thus, the Gauss theorem in the form (2) is also valid for unipolar pulses if their longitudinal dimensions are much smaller than their transverse dimensions. In particular, equation (2) is exactly valid for one-dimensional and two-dimensional pulses when the transverse dimensions are equal to infinity. For linearly polarized pulses in these cases, one can write $E = E_x(z,t)$ and $E = E_x(z,y,t)$, respectively. In the general (three-dimensional) case, equation (2) can be considered true as long as condition (22) is satisfied.

Note that the nonparaxial diffraction regime of MPCP, in a nonlinear medium was studied, for example, in [63]. It is shown, in particular, that at the non-paraxial stage, the pulse self-separates into separate bunches of light energy.

The issue of using the Gauss theorem is closely related to the dynamics of the electric area $\mathbf{S} \equiv \int_{-\infty}^{+\infty} \mathbf{E} dt$. As shown in [64,65], Maxwell's equations for the electric area of solitary pulses imply the equation

$$\nabla \times \mathbf{S} = 0. \quad (23)$$

That is, the field $\mathbf{S}$ is irrotational. In the one-dimensional case, when $\mathbf{S} = \mathbf{S}(z)$, this equation has the form [64] $d\mathbf{S}/dz = 0$. Thus, in the one-dimensional case, the rule of conservation of the electric area of the transverse momentum, $\mathbf{S} = \text{const}$ [65], is satisfied. This rule was noticed and discussed in [66]. However, only in [64] it was rigorously proved, and it was done regardless of the form of the constitutive equations

In the general case, a vector field is uniquely determined by specifying its curl and divergence at each point of the considered region of space. Integrating over time (20), we arrive at the equation

$$(\nabla \cdot \mathbf{S}) = 4\pi Q, \quad (24)$$

where the charge „associated with the electric area of the pulse" Q is defined by the expression

$$Q = -\nabla \int_{-\infty}^{+\infty} \mathbf{P} dt = \int_{-\infty}^{+\infty} \rho_b dt. \quad (25)$$

Thus, the electric area of a solitary pulse obeys electrostatic type equations (23), (24).

The irrotational nature of the vector field $\mathbf{S}$ allows us to introduce „the potential" Φ of the electric area, defined as $\mathbf{S} = -\nabla Q$. Then (24) takes the form of the Poisson equation

$$\nabla^2 \Phi = -4\pi Q. \quad (26)$$

In the paraxial approximation (see (22)) we can set $Q = 0$. Then (26) becomes the Laplace equation $\nabla^2 \Phi = 0$.

The Gauss theorem is a kind of test for the physical correctness of the obtained solutions of the wave and material equations in the form of a MPCP. It is possible that some solutions here may be „redundant", not satisfying the Gauss theorem. Particular care must be taken here with respect to solutions such as unipolar pulses in three-dimensional space.

Let us illustrate what has been said above using the example of the diffraction of MPCP.in free space by setting $P = 0$ in (11). Let the distribution of the electric field be given on the plane $z = 0$: $E|_{z=0} = E(0, t, \mathbf{r}_\perp)$, where the transverse radius vector $\mathbf{r}_\perp$ defines coordinates across the z axis. Then it is easy to show that for the half-space $z > 0$ the solution of equation (11) has the form

$$E(z, \mathbf{r}_\perp, t) = \frac{1}{2\pi c z} \times \frac{\partial}{\partial t} \int E(0, \mathbf{r}'_\perp, t - z/c - |\mathbf{r}_\perp - \mathbf{r}'_\perp|^2/2cz) d^2\mathbf{r}'_\perp. \quad (27)$$

Here the integration is carried out over the entire plane $z = 0$.

Assuming that the integral in (27) takes a finite value, we obtain for the electric area of any pulse localized in time $S = \infty_{-\infty}^{+\infty} E dt = 0$.

Thus, the area of MPCP subjected to diffraction in free three-dimensional space is equal to zero. This is true even if the MPCP is unipolar on the plane $z = 0$. This circumstance can be easily explained using the expression for the diffraction length $l_d \sim D^2/\lambda \sim \omega D^2/c$. This shows that for zero frequency, this length is zero. Therefore, due to diffraction, the zero frequency „is immediately washed out" from the pulse spectrum. As a result, its electric area becomes zero.

Conclusion

The brief, far from complete, scientific and methodological review carried out in this work shows that the approaches to theoretical studies of the nonlinear dynamics of MPCP in various media are very diverse. A wide range of MPCP indicates the need to take into account a large number of degrees of freedom of the medium that can be involved in the interaction. At the same time, taking into account a large number of degrees of freedom and quantum transitions in constitutive equations can greatly complicate the study. Therefore, the process of derivation or empirical search for material equations requires the researcher to have the professional skills of a theoretical physicist, intuition and, if you like, art.

It is intuitively clear that for pulses with a duration of only a few periods of electromagnetic oscillations, the concept of an envelope is inapplicable. In fact, it turns out that everything is not so simple here either. For example, in the work [67] for such pulses, the concept of an envelope is nevertheless used and a general equation containing an integral operator is derived. The expansion of this operator into a series in terms of higher time derivatives leads to an equation of type (8). Here it is useful to note one of the results of the work [68], where it is shown that for a single-period MPCP in a medium with Kerr nonlinearity, instead of the third harmonic of the central frequency of its spectrum (as happens in the case of (QMP), the fourth harmonic is generated. An increase in the number of oscillation periods from one to two leads to the generation of the third harmonic, as for conventional QMP. A similar situation takes place in a medium with a quadratic nonlinearity, where in the case of a single-period MPCP, not the second, but the third harmonic [69] is generated. If the pulse is two-period, then, as in the case of QMP, the second harmonic is generated. This transition between the generated harmonics cannot be described using the electric field envelope. At the same time, starting from the work [67], it is possible to assume that the use of the envelope is correct for impulses containing only up to two oscillations. For single-period MPCP, the concept of an envelope is no longer correct; therefore, it is necessary to use the electric field of the pulse itself in the material and wave equations.

Numerous papers are beyond the scope of our analysis, where the effects of ionization under the action of MPCP on matter and the formation of laser plasma are considered. Here already the Gauss theorem in the form (20) is not valid. The right side in these cases should contain the density of free charges. The study of these processes is now most relevant for intense attosecond pulses. Plasma generation here can also contribute to the formation of solitons [70–72].

There is no doubt that in the near future the nonlinear optics of MPCP will present both theorists and experimenters with many more new mysteries and surprises.

Conflict of interest

The author declares that he has no conflict of interest.

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