Modeling of nonlinear optical activity in propagation of ultrashort elliptically polarized laser pulses

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We propose a general model of third-order nonlinear optical susceptibility of isotropic gyrotropic medium with frequency and spatial dispersion. Our model allows for the description of the propagation of ultrashort (several oscillations) elliptically polarized laser pulses in such a medium and does not require smallness of the characteristic nonlocality dimension, unlike the conventional phenomenological model. We implemented our model numerically by means of a modified finite-difference time-domain method with an auxiliary differential equation. We have validated the correctness of our model by the comparison of the results obtained in our numerical simulations with generally known effects observed experimentally and described earlier theoretically for the monochromatic radiation or within the slowly varying envelope approach. We investigated effects accompanying the propagation of ultrashort (several oscillations) light pulses in nonlinear isotropic gyrotropic medium with frequency and spatial dispersion of cubic nonlinearity.

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I. INTRODUCTION

The effect of nonlinear optical activity, being understood as the intensity-dependent polarization plane rotation for the linearly polarized light in a medium with cubic nonlinearity, was predicted almost 50 years ago by Akhmanov and Zharikov [1]. Initially it was explained solely by the spatial dispersion of the nonlinear optical response of the medium. Later it was shown [2] that it can be caused also by the anisotropic nonlinear dissipation in a crystal, i.e., by the dependence of the nonlinear absorption on the mutual orientation of the crystal symmetry axes and the polarization plane of the propagating light.

At the earlier stage the phenomenon of nonlinear optical activity was unjustifiably opposed to the self-rotation of the polarization ellipse [3], which became stronger with the increase of the polarization ellipse ellipticity degree in the incident plane wave, and completely disappears for the linearly polarized light (zero ellipticity). Both these effects, responsible for the polarization ellipse rotation and deformation in a plane wave approximation, are described by the nonlinear cubic susceptibility tensor $\tilde{\chi}^{(3)}_{ijmn}(\omega,\mathbf{k};\omega,\mathbf{k},\omega,\mathbf{k},-\omega,-\mathbf{k})$ which in the first-order approximation on the spatial dispersion parameter d/λ (where d is the characteristic scale of the nonlocality of optical response of a medium; ω, $\mathbf{k}$, and λ are the frequency, the wave vector, and the wavelength of the propagating radiation) can be presented as follows:

$$\tilde{\chi}^{(3)}_{ijmn}(\omega,\mathbf{k};\omega,\mathbf{k},\omega,\mathbf{k},-\omega,-\mathbf{k}) \approx \chi^{(3)}_{ijmn}(\omega;\omega,\omega,-\omega) + i\gamma^{(3)}_{ijmnp}(\omega;\omega,\omega,-\omega)k_p, \quad (1)$$

where the fourth-rank tensor $\hat{\chi}^{(3)}$ is responsible for the local contribution and the fifth-rank tensor $\hat{\gamma}^{(3)}$ is responsible for the nonlocal contribution of the medium.

In the middle of 70 s the first experimental evidence of the nonlinear optical activity [4] came to light, giving the impulse to the development of the corresponding phenomenological theory [5–9]. The subsequent theoretical and experimental investigations assure that the polarization self-action and interaction of waves are fine and widespread phenomena in nonlinear optics. Owing to the appearance of metamaterials in optics (artificial media with high-efficiency nonlinearity and optical activity, and also media with negative refraction), many fascinating phenomena which earlier required complicated experimental setups to be detected, now can be readily reproduced and studied. In particular, recently giant nonlinear optical activity was experimentally observed in a plasmonic metamaterial [10].

However, all results of [5–9] were obtained within the framework of slowly varying envelope approximation (SVEA), which is not suitable for the description of ultrashort pulses. In this case the evolution of the electric field strength vector can be described in terms of the changes of its modulus and orientation in space. In the description of ultrashort pulses the Stokes parameters have no physical meaning, as well, as the orientation and the ellipticity degree of the polarization ellipse well known in "conventional" nonlinear polarization optics. In this case, the description of the polarization state of the ultrashort pulse can be given in the frequency spectrum domain, when the polarization state can be attributed to each Fourier harmonic with certain frequency value [11], or one should directly consider the hodograph of the electric field vector. However, in this case it is impossible to discuss any changes of polarization along the pulse in the spatiotemporal domain, although sometimes it could be tempting to compare its behavior with one of the longer pulses.

The finite-difference time-domain method (FDTD method) [12–17] is currently the most popular and probably the most efficient tool for the direct integration of the Maxwell equations along with material equations of an arbitrary type for the propagation of ultrashort pulses. In Ref. [18] it was the first time, when the FDTD method with the auxiliary differential equation (ADE) [17] was used for the description of the propagation of ultrashort laser pulses in linear media, possessing both frequency and spatial dispersion. It was shown that the changes of the direction of the electric field vector oscillations in the course of the propagation of the ultrashort pulse in a medium with spatial dispersion is far from the well-known linear polarization plane rotation for the long pulse.

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In the present work the modification of the FDTD method with ADE was used in our study of the propagation of the ultrashort elliptically polarized pulse in a medium with frequency dispersion and spatial dispersion of cubic nonlinearity. The results obtained for the ultrashort pulses are essentially different from those predicted by the analytical formulas for the evolution of the intensity-dependent ellipticity degree and the orientation of the polarization ellipse within the framework of SVEA.

II. FORMULATION OF THE PROBLEM: MAIN EQUATIONS

A. Physical model

Let us consider plane electromagnetic wave propagating along the z axis of the coordinate system in a medium with frequency dispersion and spatial dispersion of cubic nonlinearity. In this case Maxwell equations and the material equations connecting the x and y components of the strength and the induction of the electric [$\mathbf{E}(z,t)$ and $\mathbf{D}(z,t)$] and magnetic [$\mathbf{H}(z,t)$ and $\mathbf{B}(z,t)$] fields can be written as follows:

$$\begin{aligned} \frac{1}{c}\frac{\partial B_x}{\partial t} &= \frac{\partial E_y}{\partial z}, \quad -\frac{1}{c}\frac{\partial B_y}{\partial t} = \frac{\partial E_x}{\partial z}, \\ \frac{1}{c}\frac{\partial D_x}{\partial t} &= -\frac{\partial H_y}{\partial z}, \quad \frac{1}{c}\frac{\partial D_y}{\partial t} = \frac{\partial H_x}{\partial z}, \end{aligned} \tag{2}$$

$$D_i = E_i + 4\pi\left(P_i^L + P_i^{\mathrm{NL}}\right), \quad B_i \equiv H_i. \tag{3}$$

Here c is the velocity of light in vacuum, $i,j = x,y$. The most general relations between electric field strength $\mathbf{E}(z,t)$ and linear $\mathbf{P}^L$ and nonlinear $\mathbf{P}^{\mathrm{NL}}$ polarization of the medium are the following (see [19]):

$$P_i^L(t,\mathbf{r}) = \int d\mathbf{r}_1 \int_0^\infty \chi_{ij}^{(1)}(t_1,\mathbf{r},\mathbf{r}_1)E_j(t-t_1,\mathbf{r}_1)dt_1, \tag{4}$$

$$\begin{aligned} P_i^{\mathrm{NL}}(t,\mathbf{r}) = \iiint d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 \int_0^\infty \int_0^\infty \int_0^\infty \\ \times \chi_{ijmn}^{(3)}(t_1,t_2,t_3,\mathbf{r},\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)E_j(t-t_1,\mathbf{r}_1) \\ \times E_m(t-t_2,\mathbf{r}_2)E_n(t-t_3,\mathbf{r}_3)dt_1dt_2dt_3, \end{aligned} \tag{5}$$

where $\int d\mathbf{r}_1$ and $\iiint d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3$ indicates the integration on the whole space by one (x_1,y_1,z_1) or three coordination sets. The explicit views of $\chi_{ij}^{(1)}(t_1,\mathbf{r},\mathbf{r}_1)$ and $\chi_{ijkl}^{(3)}(t_1,t_2,t_3,\mathbf{r},\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)$ describing the dielectric properties of the medium remain rather unknown yet. For infinite homogeneous medium possessing arbitrary symmetry, these tensors should depend on the difference $\mathbf{r}-\mathbf{r}_{1,2,3}$ (instead of $\mathbf{r},\mathbf{r}_{1,2,3}$) and rapidly decrease to zero with the increase of $|\mathbf{r}-\mathbf{r}_{1,2,3}|$. After we proceed to the spatial and temporal Fourier representations of electric induction, strength, and medium polarization in Eqs. (3)–(5), when the explicit forms of $\chi_{ij}^{(1)}(t_1,\mathbf{r},\mathbf{r}_1)$ and $\chi_{ijkl}^{(3)}(t_1,t_2,t_3,\mathbf{r},\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)$ within the first-order approximation on d/λ are expected to provide the following expression (very well known in nonlinear optics [5,6,9,19]):

$$\begin{aligned} &D_i(\omega = \omega_1+\omega_2+\omega_3, \mathbf{k} = \mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3) \\ &= \left[\varepsilon_{ij}(\omega) + i\gamma_{ijm}^{(1)}(\omega)k_m + \cdots\right]E_j(\omega,\mathbf{k}) \\ &\quad + 4\pi\left[\chi_{ijmn}^{(3)}(\omega;\omega_1,\omega_2,\omega_3) + i\gamma_{ijmnp}^{(3,1)}(\omega;\omega_1,\omega_2,\omega_3)k_{1p}\right. \\ &\quad + i\gamma_{ijmnp}^{(3,2)}(\omega;\omega_1,\omega_2,\omega_3)k_{2p} \\ &\quad \left.+ i\gamma_{ijmnp}^{(3,3)}(\omega;\omega_1,\omega_2,\omega_3)k_{3p} + \cdots\right] \\ &\quad \times E_j(\omega_1,\mathbf{k}_1)E_m(\omega_2,\mathbf{k}_2)E_n(\omega_3,\mathbf{k}_3). \end{aligned} \tag{6}$$

Here and further we assume the summation by the indices, which appear twice in each term in the right part of the expression. At the same time each tensor in Eq. (6) has to possess "correct" internal and external permutation symmetry (which is in agreement with the symmetry of the medium and physical meaning of the tensor). In the case of the propagation of linearly polarized light (if we assume that the polarization remains unchanged) in the absence of spatial dispersion, tensor $\chi_{ijkl}^{(3)}(t_1,t_2,t_3,\mathbf{r},\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)$ must be in accordance with the well-known model for the nonlinear (third order) optical response of the medium [17], accounting for instant electron response, as well as for the effects of Raman scattering. And, finally, after the substitution of the explicit form of $\chi_{ijkl}^{(3)}(t_1,t_2,t_3,\mathbf{r},\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3)$ in Eq. (5), the expression for the $P_i^{\mathrm{NL}}(t,\mathbf{r})$ should remain unchanged after simultaneous permutations of indices $j \leftrightarrow k$ and arguments $\mathbf{r}_1 \leftrightarrow \mathbf{r}_2$, $t_1 \leftrightarrow t_2$ (or indices $j \leftrightarrow l$ and arguments $\mathbf{r}_1 \leftrightarrow \mathbf{r}_3$, $t_1 \leftrightarrow t_3$; indices $k \leftrightarrow l$ and arguments $\mathbf{r}_2 \leftrightarrow \mathbf{r}_3$, $t_2 \leftrightarrow t_3$).

When considering the plane electromagnetic wave (one-dimensional case) propagating along the z axis, expressions (4) and (5) become simpler:

$$P_i^L(t,z) = \int_{-\infty}^{\infty} dz_1 \int_0^\infty \chi_{ij}^{(1)}(t_1,z,z_1)E_j(t-t_1,z_1)dt_1, \tag{7}$$

$$\begin{aligned} P_i^{\mathrm{NL}}(t,z) = \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} dz_1dz_2dz_3 \int_0^\infty \int_0^\infty \int_0^\infty \\ \times \chi_{ijmn}^{(3)}(t_1,t_2,t_3,z,z_1,z_2,z_3)E_j(t-t_1,z_1) \\ \times E_m(t-t_2,z_2)E_n(t-t_3,z_3)dt_1dt_2dt_3. \end{aligned} \tag{8}$$

If we assume the medium to be in a half-space $z > 0$ and to possess symmetry group $\infty\infty$ (isotropic gyrotropic medium), when the following form of the tensor will satisfy to all the abovementioned requirements for $z_1 > 0$:

$$\begin{aligned} \chi_{ij}^{(1)}(t_1,z,z_1) = g^{(1)}(t_1)[\delta_{ij} + \gamma_1(\delta_{xi}\delta_{yj} - \delta_{xj}\delta_{yi})(z-z_1)] \\ \times \exp\left[-(z-z_1)^2/d_1^2\right]\big/(\sqrt{\pi}d_1). \end{aligned} \tag{9}$$

Here δ_{ij} is the Kronecker symbol and γ_1 and d_1 are the parameters characterizing linear gyrotropy of the medium. Here [in Eq. (9)] and further $i,j = x,y$. When the frequency dispersion is one of the Lorentz type [17], then

$$\begin{aligned} g^{(1)}(t_1) = \left[(\varepsilon_\infty - 1)\delta(t_1) + \omega_0^2(\varepsilon_S - \varepsilon_\infty)\left(\omega_0^2 - \delta_0^2\right)^{-1/2}\right. \\ \left.\times \exp(-\delta_0 t_1)\sin\left(\left[\omega_0^2 - \delta_0^2\right]^{1/2}t_1\right)\right]\big/4\pi, \end{aligned} \tag{10}$$

where $\varepsilon_S, \varepsilon_\infty, \omega_0, \delta_0$ are constants, and $\delta(t_1)$ is a delta function. For $z_1 < 0$, $\chi_{ij}^{(1)}(t_1,z,z_1) \equiv 0$. The substantiation of choice of such a form of $\chi_{ij}^{(1)}(t_1,z,z_1)$ is given in Ref. [18], where the expressions (7), (9), and (10) were used for the study of the propagation of ultrashort (about several oscillations of the light field) laser pulses in a linear medium with both frequency and spatial dispersion by means of integration of Maxwell equations by the FDTD method with ADE.

The reasonable generalization of this model for the nonlinear isotropic gyrotropic medium with frequency dispersion and spatial dispersion of cubic nonlinearity $\chi_{ijkl}^{(3)}(t_1,t_2,t_3,z,z_1,z_2,z_3)$ could be the following ($z_{1,2,3} > 0$):

$$\begin{aligned}\chi_{ijkl}^{(3)}(t_1,t_2,t_3,z,z_1,z_2,z_3) &= a\delta(t_1)\delta(t_2)\delta(t_3)\delta(z_1)\delta(z_2)\delta(z_3)[\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}]\\ &+\delta(t_1)\delta(t_2-t_3)g_3(t_3)\delta(z-z_1)\delta(z_2-z_3)[b\delta_{ij}\delta_{kl}+c(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})\\ &+\gamma_3(z-z_3)(\delta_{xi}\delta_{yj}-\delta_{yi}\delta_{xj})\delta_{kl}]\exp\left[-(z-z_3)^2/d_3^2\right]/(\sqrt{\pi}d_3)\\ &+\delta(t_2)\delta(t_1-t_3)g_3(t_1)\delta(z-z_2)\delta(z_1-z_3)[b\delta_{ik}\delta_{jl}+c(\delta_{ij}\delta_{kl}+\delta_{il}\delta_{jk})\\ &+\gamma_3(z-z_1)(\delta_{xi}\delta_{yk}-\delta_{yi}\delta_{xk})\delta_{jl}]\exp\left[-(z-z_1)^2/d_3^2\right]/(\sqrt{\pi}d_3)\\ &+\delta(t_3)\delta(t_1-t_2)g_3(t_2)\delta(z-z_3)\delta(z_1-z_2)[b\delta_{il}\delta_{jk}+c(\delta_{ij}\delta_{kl}+\delta_{ik}\delta_{jl})\\ &+\gamma_3(z-z_2)(\delta_{xi}\delta_{yl}-\delta_{yi}\delta_{xl})\delta_{jk}]\exp\left[-(z-z_2)^2/d_3^2\right]/(\sqrt{\pi}d_3),\end{aligned} \quad (11)$$

and $\chi_{ijkl}^{(3)}(t_1,t_2,t_3,z,z_1,z_2,z_3) \equiv 0$ at $z_{1,2,3} < 0$. Here a, b, and c determine the cubic nonlinearity of the medium, and γ_3 and d_3 determine its spatial dispersion. Indices i, j, k, and l take values of x and y. Such a choice of the coordinate dependence (exponential) is prescribed only by the requirement of rapid decay of $\chi_{ijkl}^{(3)}(t_1,t_2,t_3,z,z_1,z_2,z_3)$ to zero with the increase of $|z-z_{1,2,3}|$ and also by its similarity with $\chi_{ij}^{(1)}(t_1,z,z_1)$. Terms $g^{(3)}(t_{1,2,3})$ in Eq. (11) we take as

$$g^{(3)}(\tilde{t}) = \left(\tau_1^2+\tau_2^2\right)\left(\tau_1\tau_2^2\right)^{-1}\exp(-\tilde{t}/\tau_2)\sin(\tilde{t}/\tau_1), \quad (12)$$

which was thoroughly substantiated in Ref. [19] and references therein. Here $\tau_{1,2}$ are the relaxation time constants, b/a and c/a determine the relative contributions of Kerr-type and Raman-type nonlinearities. Substituting (9)–(12) into (3), (7), and (8) with $\gamma_{1,3} = d_{1,3} = 0$ (in the absence of spatial dispersion) it is easy to obtain the following:

$$\begin{aligned}&D_i(\omega=\omega_1+\omega_2+\omega_3,\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}_3)\\ &\quad=\varepsilon_0(\omega)\delta_{ij}E_j(\omega,\mathbf{k})+4\pi(\tilde{\alpha}_1(\omega_1,\omega_2,\omega_3)\delta_{ij}\delta_{mn}\\ &\qquad+\tilde{\alpha}_2(\omega_1,\omega_2,\omega_3)\delta_{im}\delta_{jn}+\tilde{\alpha}_3(\omega_1,\omega_2,\omega_3)\delta_{in}\delta_{jm})\\ &\qquad\times E_j(\omega_1,\mathbf{k}_1)E_m(\omega_2,\mathbf{k}_2)E_n(\omega_3,\mathbf{k}_3),\end{aligned} \quad (13)$$

which also follows from (6) in the case of $\infty\infty m$ medium symmetry (isotropic nongyrotropic medium). Constants ε_0 and $\tilde{\alpha}_{1,2,3}$ in Eq. (13) can be expressed through the parameters of our model by the following way:

$$\begin{aligned}\tilde{a}_{1,2,3} &= a+b\tilde{g}_3(\omega_{2,1,1}+\omega_{3,3,2})+c(\tilde{g}_3(\omega_{1,2,1}+\omega_{3,3,3})\\ &\quad+\tilde{g}_3(\omega_{1,1,2}+\omega_{2,2,3})),\\ \varepsilon_0(\omega) &= \varepsilon_\infty-\omega_0^2(\varepsilon_s-\varepsilon_\infty)/\left(\omega^2-\omega_0^2+2i\delta_0\omega\right),\end{aligned}$$

where $\tilde{g}_3(\omega) = -(\tau_1^{-2}+\tau_2^{-2})(\omega^2-\tau_1^{-2}-\tau_2^{-2}+2i\omega/\tau_2)^{-1}$. Dependencies (9)–(11) not only satisfy all the abovementioned requirements for $\chi_{ij}^{(1)}(t_1,z,z_1)$ and $\chi_{ijkl}^{(3)}(t_1,t_2,t_3,z,z_1,z_2,z_3)$, but also allow for a thin (of the order of $\max\{d_1,d_3\}$) surface layer with transient dielectric properties. Actually, the border of the medium represents not an abrupt but, rather, a smooth (although fast enough) change in optical properties (from one medium to another). This smooth change can be treated also as an additional layer on the medium surface with transient properties (see, for example, [20]). This fact can be neglected in many problems, but if one is considering nonlocal spatial response, this may cause additional features of radiation-matter interaction [20], because in this case the thickness of such a layer should not be less than the characteristic nonlocality dimension.

However, one should take into account not only the fact that the response of the *homogeneous medium with sharp boundary* will be different near the surface of the medium and in its bulk, but also that the properties of the medium (the dielectric permittivity and the nonlinear susceptibility) are different near the surface of the medium and in its bulk. Our model does not account for this. Apart from that, we do not consider a number of complicated phenomena on the border of the medium with spatial dispersion in the vicinity of frequency resonances [21], e.g., appearance of so-called additional waves.

It is also worth noticing that the models of frequency and spatial dispersion of linear and nonlinear optical susceptibilities of the medium we use in our work, indeed, do not account for a number of effects, which are described by a Debye model. However, in our work we prefer to use another model, which represents itself the generalization (for the case of spatial dispersion) of a widely used and efficiently working dispersion model.

In the case of the propagation of the tightly focused ultrashort light pulse with broad frequency and spatial spectra, there become possible the second-order processes [sum-frequency generation, difference frequency generation (SFG), second harmonic generation (SHG)] of nonlinear interaction between its noncollinear components with different frequencies, which may result in considerable modification of the frequency and spatial spectrum of the propagating pulse.

In our work we do not take into account second-order nonlinear processes. Therefore, it would be correct to consider our results under conditions of absence or smallness of the second-order response, i.e., in centrosymmetric medium with spatial dispersion of cubic nonlinearity, or when the effective quadratic optical susceptibility value is negligibly small, or when the phase-matching condition cannot be satisfied, for example, in the case of the plane-wave propagation in the isotorpic gyrotropic medium.

B. Auxiliary definitions and difference approximation of expressions

Numerical model. After substitution of (9)–(12), formulas (7) and (8) can be written as follows:

$$P_x^L(t,z) = \int_0^\infty g_1(t')f_1(t-t',z)dt' + \zeta_x\gamma_1\int_0^\infty g_1(t')f_4(t-t',z)dt', \quad (14a)$$

$$P_y^L(t,z) = \int_0^\infty g_1(t')f_2(t-t',z)dt' + \zeta_y\gamma_1\int_0^\infty g_1(t')f_3(t-t',z)dt', \quad (14b)$$

$$\begin{aligned} P_x^{\rm NL}(t,z) &= 3aE_x(t,z)\big(E_x^2(t,z)+E_y^2(t,z)\big) \\ &+6cE_x(t,z)\int_0^\infty g_3(t')f_5(t-t',z)dt' \\ &+6cE_y(t,z)\int_0^\infty g_3(t')f_7(t-t',z)dt' \\ &+3bE_x(t,z)\int_0^\infty g_3(t')[f_5(t-t',z)+f_6(t-t',z)]dt' \\ &+3\zeta_x\gamma_3E_y(t,z)\int_0^\infty g_3(t')f_8(t-t',z)dt', \end{aligned} \quad (15a)$$

$$\begin{aligned} P_y^{\rm NL}(t,z) &= 3aE_y(t,z)\big(E_x^2(t,z)+E_y^2(t,z)\big) \\ &+6cE_y(t,z)\int_0^\infty g_3(t')f_6(t-t',z)dt' \\ &+6cE_x(t,z)\int_0^\infty g_3(t')f_7(t-t',z)dt' \\ &+3bE_y(t,z)\int_0^\infty g_3(t')[f_5(t-t',z)+f_6(t-t',z)]dt' \\ &+3\zeta_y\gamma_3E_x(t,z)\int_0^\infty g_3(t')f_8(t-t',z)dt', \end{aligned} \quad (15b)$$

where $\zeta_x = 1$, $\zeta_y = -1$, and

$$f_1(t,z) = \int_{-\infty}^\infty \exp\left[-z_1^2/d_1^2\right]/(\sqrt{\pi}d_1)E_x(t,z-z_1)dz_1, \quad (16a)$$

$$f_2(t,z) = \int_{-\infty}^\infty \exp\left[-z_1^2/d_1^2\right]/(\sqrt{\pi}d_1)E_y(t,z-z_1)dz_1, \quad (16b)$$

$$f_3(t,z) = \int_{-\infty}^\infty z_1\exp\left[-z_1^2/d_1^2\right]/(\sqrt{\pi}d_1)E_x(t,z-z_1)dz_1, \quad (17a)$$

$$f_4(t,z) = \int_{-\infty}^\infty z_1\exp\left[-z_1^2/d_1^2\right]/(\sqrt{\pi}d_1)E_y(t,z-z_1)dz_1, \quad (17b)$$

$$f_5(t,z) = \int_{-\infty}^\infty \exp\left[-z_1^2/d_3^2\right]/(\sqrt{\pi}d_3)E_x^2(t,z-z_1)dz_1, \quad (18a)$$

$$f_6(t,z) = \int_{-\infty}^\infty \exp\left[-z_1^2/d_3^2\right]/(\sqrt{\pi}d_3)E_y^2(t,z-z_1)dz_1, \quad (18b)$$

$$f_7(t,z) = \int_{-\infty}^\infty \exp\left[-z_1^2/d_3^2\right]/(\sqrt{\pi}d_3)E_x(t,z-z_1) \times E_y(t,z-z_1)dz_1, \quad (19)$$

$$f_8(t,z) = \int_{-\infty}^\infty z_1\exp\left[-z_1^2/d_3^2\right]/(\sqrt{\pi}d_3)\big(E_x^2(t,z-z_1) + E_y^2(t,z-z_1)\big)dz_1. \quad (20)$$

Here functions

$$F_s(t,z) = \frac{\omega_0^2(\varepsilon_S-\varepsilon_\infty)}{\left(\omega_0^2-\delta_0^2\right)^{1/2}}\int_0^\infty \exp(-\delta_0 t_1) \times \sin\left(\left[\omega_0^2-\delta_0^2\right]^{1/2}t_1\right)f_s(t-t_1,z)dt_1, \quad (21)$$

$$F_h(t,z) = \frac{\tau_1^2+\tau_2^2}{\tau_1\tau_2^2}\int_0^\infty \exp(-t_1/\tau_2)\sin(t_1/\tau_1)f_h(t-t_1,z)dt_1, \quad (22)$$

($s = 1,2,3,4$, and $h = 5,6,7,8$) satisfy the system of ordinary differential equations,

$$\frac{d^2}{dt^2}F_s + 2\delta_0\frac{d}{dt}F_s + \omega_0^2F_s = \omega_0^2\left(\varepsilon_s-\varepsilon_\infty\right)f_s, \quad (23)$$

$$\frac{d^2}{dt^2}F_h + \frac{2}{\tau_2}\frac{d}{dt}F_h + \left(\frac{1}{\tau_1^2}+\frac{1}{\tau_2^2}\right)F_h = \left(\frac{1}{\tau_1^2}+\frac{1}{\tau_2^2}\right)f_h, \quad (24)$$

with zero initial conditions. Furthermore they can be used in the new form of the material equation (3):

$$\begin{aligned} D_x &= E_x + (\varepsilon_\infty-1)(f_1+\gamma_1\zeta_xf_4) + (F_1+\gamma_1\zeta_xF_4) \\ &+ 12\pi\big\{aE_x(t,z)\big[E_x^2(t,z)+E_y^2(t,z)\big] \\ &+ bE_x(t,z)(F_5+F_6) + 2cE_x(t,z)F_5 + 2cE_y(t,z)F_7 \\ &+ \gamma_3\zeta_xE_y(t,z)F_8\big\}. \end{aligned} \quad (25a)$$

$$\begin{aligned} D_y &= E_y + (\varepsilon_\infty-1)(f_2+\gamma_1\zeta_yf_3) + (F_2+\gamma_1\zeta_yF_3) \\ &+ 12\pi\big\{aE_y(t,z)\big[E_x^2(t,z)+E_y^2(t,z)\big] \\ &+ bE_y(t,z)(F_5+F_6) + 2cE_y(t,z)F_6 \\ &+ 2cE_x(t,z)F_7 + \gamma_3\zeta_yE_x(t,z)F_8\big\}. \end{aligned} \quad (25b)$$

For numerical solution of (2) and (23)–(25) we used uniform grid $z_m = m\Delta z$, $t_n = n\Delta t$, where $m = 0,1,2,\ldots,M-1$, $n = 0,1,2,\ldots,N-1$. Values of M and N were determined by the distance traversed in a medium by the pulse, and by the interval of time it took for a pulse to propagate. Functions $f_{1,2,\ldots,8}$ in Eqs. (21), (22), and (25), and also in difference schemes of (23) and (24), were taken as the following difference expressions:

$$f_{1,2}(t_n,z_m) = \sum_{l=0}^{M-1} \frac{\exp\left[-(m-l)^2(\Delta z)^2/d_1^2\right]}{\sqrt{\pi}d_1} \times E_{x,y}(n\Delta t,l\Delta z)\Delta z = f_{1,2}^{n,m}, \quad (26)$$

$$f_{3,4}(t_n,z_m) = \sum_{l=0}^{M-1} \frac{\exp\left[-(m-l)^2(\Delta z)^2/d_1^2\right]}{\sqrt{\pi}d_1} \times E_{x,y}(n\Delta t,l\Delta z)(m-l)(\Delta z)^2 = f_{3,4}^{n,m}, \quad (27)$$

$$f_{5,6}(t_n,z_m) = \sum_{l=0}^{M-1} \frac{\exp\left[-(m-l)^2(\Delta z)^2/d_3^2\right]}{\sqrt{\pi}d_3} \times E_{x,y}^2(n\Delta t,l\Delta z)\Delta z = f_{5,6}^{n,m}, \quad (28)$$

$$f_7(t_n,z_m) = \sum_{l=0}^{M-1} \frac{\exp\left[-(m-l)^2(\Delta z)^2/d_3^2\right]}{\sqrt{\pi}d_3} \times E_x(n\Delta t,l\Delta z)E_y(n\Delta t,l\Delta z)\Delta z = f_7^{n,m}, \quad (29)$$

$$f_8(t_n,z_m) = \sum_{l=0}^{M-1} \frac{\exp\left[-(m-l)^2(\Delta z)^2/d_3^2\right]}{\sqrt{\pi}d_3}\left[E_x^2(n\Delta t,l\Delta z) + E_y^2(n\Delta t,l\Delta z)\right](m-l)(\Delta z)^2 = f_8^{n,m}. \quad (30)$$

The substitution of these difference approximations into (23) and (24) allows one to find the values of $F_{1,2,\ldots,8}(t_{n+1},z_m)$, and, finally, to obtain the system of $2M$ nonlinear equations for the electric field strength vector components $E_{x,y}(t_{n+1},z_m) = E_{x,y}^{n+1,m}$ from (25):

$$D_{x,y}^{n+1,m} = D_{x,y}(t_{n+1},z_m) = \Phi_{x,y}^m\left(E_x^{n+1,0},E_x^{n+1,1},\ldots,E_x^{n+1,s},\ldots,E_x^{n+1,M-1}, E_y^{n+1,0},E_y^{n+1,1},\ldots,E_y^{n+1,s},\ldots,E_y^{n+1,M-1}\right). \quad (31)$$

In Eq. (31) and further index s takes values $0,1,2,\ldots, m,\ldots,M-1$, and the overall look of functions $\Phi_{x,y}^m$ is determined from (25) for each m [also taking into account (26)–(30)].

In this algorithm in each step at the first stage one determines the strength of the magnetic field at the time step $t_n + \Delta t/2$ from the strength of the electric field at the previous time step t_n, and then the induction of the electric field $D_{x,y}^{n+1,m}$ at the time step $t_n + \Delta t$. At the final stage we solve the equation system (31) by the iteration method of the Newton type, in which at each step we find the next element of $\{E_{x,y}^{n+1,m}\}_k$ sequence, converging to the electric field strength $E_{x,y}^{n+1,m}$. Numerical value of each $\{E_{x,y}^{n+1,m}\}_k$ was found as a result of the solution of the following system of linear equations:

$$J_{ij}^{m,l}\left[\{E_j^{n+1,l}\}_k - \{E_j^{n+1,l}\}_{k-1}\right] = D_i^{n+1,m} - \Phi_i^m\left(\{E_x^{n+1,0}\}_{k-1},\{E_x^{n+1,1}\}_{k-1},\ldots, \{E_x^{n+1,s}\}_{k-1},\ldots,\{E_x^{n+1,M-1}\}_{k-1},\{E_y^{n+1,0}\}_{k-1}, \{E_y^{n+1,1}\}_{k-1},\ldots,\{E_y^{n+1,s}\}_{k-1},\ldots,\{E_y^{n+1,M-1}\}_{k-1}\right). \quad (32)$$

In Eq. (32) $i,j = x,y$, and we assume the summation by the indices j and $l = 0,1,2,\ldots,m,\ldots,M-1$ which appear twice in this expression. The elements of block matrix $J_{ij}^{m,l} = \partial\Phi_i^m/\partial E_j^{n+1,l}$ having dimension $2M \times 2M$ are calculated for $E_j^{n+1,l} = \{E_j^{n+1,l}\}_{k-1}$. If the medium parameters $d_{1,3}$ are such that $\max\{d_1,d_3\}$ is much less than the propagation path of the pulse L, then $J_{ij}^{m,l}$ is a sparse matrix, in which the nonzero elements number is of the order of $\max\{d_1,d_3\}L/(\Delta z)^2$. In this case for the solution of the linear algebraic equations (32) and (33) we can use the generalized minimal residual method (GMRES) [22].

C. Representation of results

It is well known that the polarization state of the monochromatic radiation can be wholly described by a set of four independent quantities [23]. Apart from the Stokes parameters, one is able to use the intensity $\tilde{I} = A_x^2 + A_y^2$, the ellipticity degree of the polarization ellipse $\tilde{M} = 2A_xA_y\sin\Delta/(A_x^2 + A_y^2)$, the angle of orientation of the polarization ellipse $\tilde{\Psi} = 0{,}5\,\mathrm{arctg}[2A_xA_ys\cos\Delta/(A_x^2 - A_y^2)]$, and the parameter, characterizing the orientation of the electric field vector at fixed timing. In the formulas above $A_{x,y}$ are the real-valued amplitudes of harmonic (sinusoidal) oscillating Cartesian components of the electric field strength vector; Δ is the difference of their oscillation phases. Now different modifications of the SVEA are widely used for the description of the propagation of long pulse. In this method $A_{x,y}$ are treated as slowly varying quantities with respect to changes of z and/or t. In this case for the fixed time value, the spatial (or temporal) distribution of the electric field in a pulse is attributed to the set of a relatively big amount of the polarization ellipses characterizing the polarization state of radiation in different points in space (or at different moments in time). The ellipticity degree $M'(z,t)$ and the angle of orientation $\Psi'(z,t)$ for each of these ellipses at certain z can be found by the same formulas with $A_{x,y}(z,t)$ instead of $A_{x,y}$ and $\Delta(z,t)$ instead of Δ. The analysis of $M'(z,t)$ and $\Psi'(z,t)$ gives us the information about the polarization changes during pulse propagation.

When proceeding to the shorter pulses (including those we call *ultrashort*, thus indicating several-oscillation pulses) the ellipticity degree and the angle of orientation of the polarization ellipse defined as above have no physical meaning. This also concerns any other sets of four parameters, describing the intensity and the polarization of the propagating radiation in such a case, because we cannot average along the set of almost identical ellipses as earlier. In the domain of ultrashort pulses it would be correct to speak in terms of the changes of modulus of the electric field strength vector $I_1^{1/2}(z,t) = (E_x^2(z,t) + E_y^2(z,t))^{1/2}$ and the angle

$\Psi(z,t) = \text{arctg}(E_y/E_x)$ it composes with the x axis of the coordinate system. These two values wholly characterize the propagating pulse. Additionally, one can define quantities $M(z,t)$ and $\Psi(z,t)$, which, at certain degree, can be treated as $M'(z,t)$ and $\Psi'(z,t)$ defined for the slowly varying envelope and will carry the information on the predominant orientation of the electric field vector. It is necessary to provide that these newly defined quantities $M(z,t)$ and $\Psi(z,t)$ would tend to $M'(z,t)$ and $\Psi'(z,t)$ correspondingly when increasing the duration of a pulse, and turned into M and Ψ in the extreme case of the plane monochromatic wave. The need for definition of $M(z,t)$ and $\Psi(z,t)$ is justified solely by the necessity of the comparison of results obtained numerically in the present work as $E_{x,y}(z,t)$ with intensity and polarization data obtained within the framework of SVEA for long pulses.

In the course of the solution of the one-dimensional problem of pulse propagation in a medium with spatial dispersion of cubic nonlinearity by the FDTD method it is reasonable to define $M(z,t)$ and $\Psi(z,t)$ as the following discrete functions satisfying the abovementioned conditions:

$$|M(\tilde{z}_m,t_n)| = \frac{2^{1/2} I_1^{1/2}(\tilde{z}_m,t_n)\cdot[I_1(\bar{z}_m,t_n)+I_1(\bar{z}_{m+1},t_n)]^{1/2}}{I_1(\tilde{z}_m,t_n)+[I_1(\bar{z}_m,t_n)+I_1(\bar{z}_{m+1},t_n)]/2}, \tag{33}$$

$$\Psi(\tilde{z}_m,t_n) = -\text{arctg}[E_x(\tilde{z}_m,t_n)/E_y(\tilde{z}_m,t_n)]. \tag{34}$$

These dependencies are defined only in a number of points $z = \tilde{z}_m$, where $I_1^{1/2}(z_m,t_n)$ achieves local maximum value. In Eqs. (33) and (34) $\bar{z}_m$ are the points of local minima of $I_1^{1/2}(z_m,t_n)$, numbered in such a way that $\bar{z}_m \leqslant \tilde{z}_m \leqslant \bar{z}_{m+1}$. The sign of $M(\tilde{z}_m,t_n)$ is determined by the direction of rotation of the electric field vector. In the case of a relatively long pulse the interpolation of $M(\tilde{z}_m,t_n)$ and $\Psi(\tilde{z}_m,t_n)$ gives $M'(z,t)$ and $\Psi'(z,t)$. The definitions (33) and (34) also have some limitation: it is meaningful to use them until the oscillations of the electric field Cartesian components still can be considered as something similar to harmonic oscillations (even for several-oscillation pulses). Some examples will be discussed below.

III. THE DISCUSSION OF RESULTS

Let us consider elliptically polarized light pulse with the Gaussian envelope, which propagates in vacuum towards the plane boundary $z = 0$ of the medium with frequency dispersion and spatial dispersion of cubic nonlinearity (medium symmetry group $\infty\infty$). We assume that at $t = 0$ the Cartesian components of the electric field in the pulse do not depend on x and y and are expressed by the following formulas:

$$E_x(z,t=0) = \left(\frac{PI_0}{2}\left(1-\sqrt{1-M_0^2}\right)\right)^{1/2}\exp\left(-\frac{(z-z_0)^2}{w_0^2}\right) \times \text{sign}(M_0)\cdot\sin\left(\frac{2\pi(z-z_0)}{\lambda}\right), \tag{35}$$

$$E_y(z,t=0) = \left(\frac{PI_0}{2}\left(1+\sqrt{1-M_0^2}\right)\right)^{1/2}\exp\left(-\frac{(z-z_0)^2}{w_0^2}\right) \cdot\cos\left(\frac{2\pi(z-z_0)}{\lambda}\right), \tag{36}$$

where λ is the wavelength in vacuum corresponding to the central frequency of the spectrum of the incident pulse. For relatively big values of w_0 in $E_{x,y}(z,t)$, the terms standing before sine in Eq. (35) and cosine in Eq. (36) can be treated as slowly varying envelopes. In this case the dimensionless intensity $I = (E_x^2 + E_y^2)/I_0$ achieves its maximum value P in $z = z_0$. Using (33) and (34) it is easy to show that for all values of z and t the incident pulse has the ellipticity degree of the polarization ellipse, which is equal to M_0, and the main axis of the polarization ellipse is parallel to the y axis (we always can choose the coordinate axes in such a way for a medium with $\infty\infty$ symmetry).

A. Medium with local optical response

Comparison with SVEA for the long pulses. As in Refs. [17,18], we consider a value of ω equal to 8.61×10^{14} rad s^{-1} ($\lambda \approx 2.19\,\mu$m), which means that for $w_0 = 100\lambda$ the effective duration of the pulse in vacuum is ≈730 fs. Let us assume that at $t = 0$ the maximum of the intensity of the elliptically polarized pulse ($M_0 = 0.1$) is located at 400 wavelength distance from the medium border, and the medium is described by the following parameters: $\varepsilon_s = 5.25$, $\varepsilon_\infty = 2.25$, $\delta_0 = 1.64 \times 10^{-5}\omega$, $\omega_0 = 0.46\omega$, $\tau_1 = 10.5/\omega$ (≈12.2 fs), $\tau_2 = 27.6/\omega$ (≈32 fs), which are analogous to those used in Refs. [17,18]. Within the SVEA the self-action of the long pulse, in which the spectrum does not cover the resonance frequencies of the isotropic nonlinear medium response, will be affected only by two components of tensor $\hat{\chi}^{(3)}$ (see, for example, [5]; p. 1362): $\chi^{(3)}_{xyxy} = a + b\tilde{g}_3(0) + c(\tilde{g}_3(0) + \tilde{g}_3(2\omega))$ and $\chi^{(3)}_{xxyy} = a + b\tilde{g}_3(2\omega) + 2c\tilde{g}_3(0)$. For purely Kerr nonlinearity ($b = c = 0$) the following takes place $\chi^{(3)}_{xyxy} = \chi^{(3)}_{xxyy}$. And for a purely Raman nonlinear response ($a = 0$), $\chi^{(3)}_{xyxy}/\chi^{(3)}_{xxyy}$ depends on b, c, τ_1, τ_2, and ω. Different mechanisms of nonlinearity (Kerr or Raman) identically affect propagation of the pulse in terms of SVEA, if they yield the same values of $\chi^{(3)}_{xyxy}$ and $\chi^{(3)}_{xxyy}$. Let us check whether the same statement is true within the framework of our model.

Figure 1 shows the z dependencies of the intensity (a) and the intensity-dependent angle of rotation of the polarization ellipse (b) for the pulse traversed about 200 wavelengths in a medium without spatial dispersion ($\gamma_{1,3} = 0$, $d_{1,3} = 0$), which were calculated in the present work by means of the algorithm described above. This distance is much less than the dispersion length of the pulse under consideration for the chosen values of linear optical response parameters. Therefore, the influence of linear optical response on the intensity profile of the propagating pulse in this case can be neglected.

Solid lines in Fig. 1 correspond to $a = 2 \times 10^{-4} \times I_0^{-1}$, $b = 0$, $c = 0$, dashed lines correspond to $a = 0$, $b = 1 \times 10^{-4} \times I_0^{-1}$, $c = 1 \times 10^{-4} \times I_0^{-1}$. These values of a, b, and c characterizing the cubic nonlinearity of the medium are chosen in such a way that their substitution in Eq. (11) and subsequent calculation of $\chi^{(3)}_{ijmn}(\omega;\omega_1,\omega_2,\omega_3)$ [which presents in Eq. (6)] for given τ_1 and τ_2 (chosen above) would yield equal values for $\chi^{(3)}_{xxyy}(\omega;-\omega,\omega,\omega)$ and $\chi^{(3)}_{xyxy}(\omega;-\omega,\omega,\omega)$. But in the first case this will be achieved solely by the Kerr contribution to the nonlinear response, while in the second case this will be a purely Raman (retarded) response without

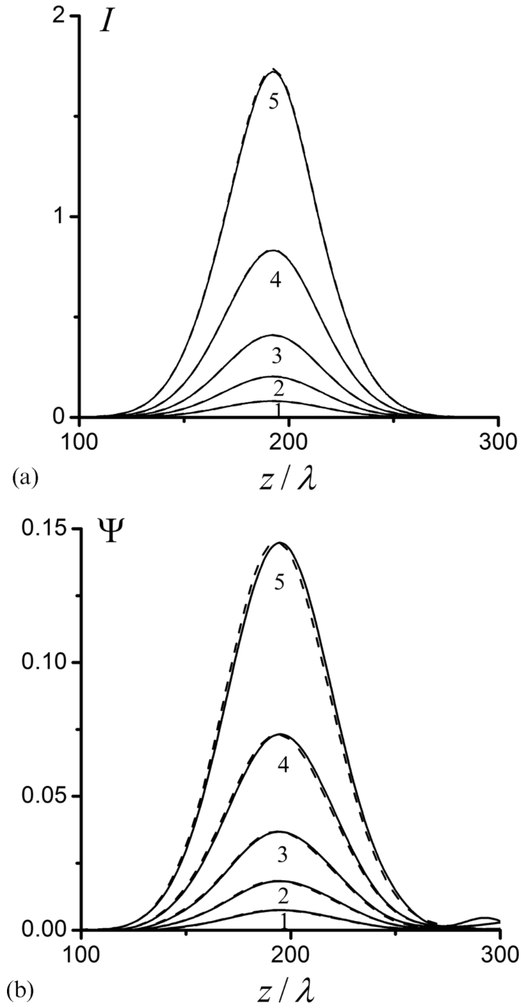

FIG. 1. The dependencies of the intensity I (a) and the angle of polarization ellipse rotation Ψ (b) on the propagation coordinate z/λ for the pulse traversed approximately 200 wavelengths in the nonlinear medium. The medium parameters are as follows: $\varepsilon_s = 5.25$, $\varepsilon_\infty = 2.25$, $\gamma_1 = 0$, $d_1 = 0$, $\delta_0 = 1.64 \times 10^{-5}\omega$, $\omega_0 = 0.46\omega$, $\gamma_{1,3} = 0$, $d_{1,3} = 0$, $\tau_1 = 10.5/\omega$, $\tau_2 = 27.6/\omega$; $a = 2 \times 10^{-4} \times I_0^{-1}$, $b = 0$, $c = 0$ (for solid-line dependencies) and $a = 0$, $b = 1 \times 10^{-4} \times I_0^{-1}$, $c = 1 \times 10^{-4} \times I_0^{-1}$ (for dashed-line dependencies). The parameters of the incident pulse in accordance with the formulas (35) and (36) are as follows: $w_0 = 100\lambda$, $M_0 = 0.1$, $P = 0.1$ (1); $P = 0.25$ (2); $P = 0.5$ (3); $P = 1$ (4); $P = 2$ (5).

"instant" nonlinearity. Lines 1–5 in this figure correspond to the values of $P = 0.1, 0.25, 0.5, 1, 2$.

The almost complete coincidence of solid and dashed lines in Fig. 1 testifies to the weakness of the influence of frequency dispersion of cubic nonlinearity on the effect of polarization ellipse self-rotation for given values of intensity and duration of the incident pulse. After traversing some hundreds of wavelengths in a medium, the shape of the propagating pulse practically does not change, the ellipticity degree of its polarization ellipse is constant, and the z dependence of the angle of rotation of the polarization ellipse [Fig. 1(b)] replicates the shape of the intensity profile $I(z/\lambda)$ [Fig. 1(a)] (the difference between these two dependencies becomes visible for $P = 2$ and higher input intensities). This testifies direct proportionality of $\Psi(z/\lambda)$ to $I(z/\lambda)$ in this case.

In order to validate our model of nonlocal and nonlinear response of the medium, we have performed a number of numerical simulations in a broad range of parameters of the nonlinear medium for the long pulses (hundreds of wavelengths) and for the small values of the intensity of the incident radiation. Changing the parameters I_0 and M_0, we have tracked the value of the rotation angle of the polarization ellipse in the peak of the propagating pulse. We established that these values of the rotation angle are directly proportional to I_0 and M_0. At that, the rotation does not occur if $M_0 = 0$. Such a dependence of Ψ on M_0 and I_0 is wholly in accordance with the plane-wave polarization ellipse rotation in isotropic medium with cubic nonlinearity predicted in Ref. [3]. In the latter case the angle of rotation was found to behave as the following: $\Psi \sim \chi^{(3)}_{xxyy}(\omega; -\omega, \omega, \omega) M_0 P I_0 z$.

For the bigger intensities of the incident radiation, unlike in Fig. 1, there occurs change of shape of the temporal envelope of the propagating laser pulse owing to the nonlinear optical response of the medium. In addition, the polarization ellipse rotation angle dependence on the propagation coordinate changes its character.

In Fig. 2 we compare the difference between the instant (Kerr) and retarded (Raman) nonlinear response in the same way as in Figs. 1(a) and 1(b), but for bigger values of intensity. For $P \approx 10$, Kerr [Fig. 2(a)] and Raman [Fig. 2(b)] nonlinearity mechanisms yielding the same values of $\chi^{(3)}_{xyxy}$ and $\chi^{(3)}_{xxyy}$, give different z dependencies of the intensity (solid lines), ellipticity degree (dashed lines), and the angle of rotation of the polarization ellipse (dotted lines) in the laser pulse traversed about 200 wavelengths in a medium. For such big intensities this difference is caused by the strong broadening of the spectrum of the pulse, which also makes unjustified the SVEA in this case (predicting similar behavior). In addition, the abrupt changes of I, M, and Ψ indicate that the SVEA may give wrong results even in the case of propagation of relatively long pulses.

B. Medium with nonlocality of nonlinear optical response

Comparison with SVEA for the long pulses. The effects caused by the nonlocality of the *linear* optical response were discussed in Ref. [18] in detail. In the present work, in order to demonstrate how the polarization plane rotation occurs, caused solely by the spatial dispersion of cubic nonlinearity, we use $\gamma_1 = 0$ and $d_1 = 0$ for the example shown in Fig. 3. There one can see the z dependencies of the intensity (solid lines), the ellipticity degree (dashed lines), and the angle of orientation of the polarization ellipse (dotted lines) for the long pulse traversed in a medium about 600 wavelengths. The curve $\Psi(z/\lambda)$ practically replicates $I(z/\lambda)$. The maximum of $\Psi(z/\lambda)$ appears to be shifted in the direction of pulse propagation [relatively to the intensity maximum—Fig. 3(a)]. This is due to the fact that for the given period of time the front part of the pulse traverses in a medium more than the backward part of the pulse (which enters the medium some periods of oscillation later), thus experiencing stronger polarization rotation than the backward part. The increase of the distance traversed in a medium makes closer the maxima

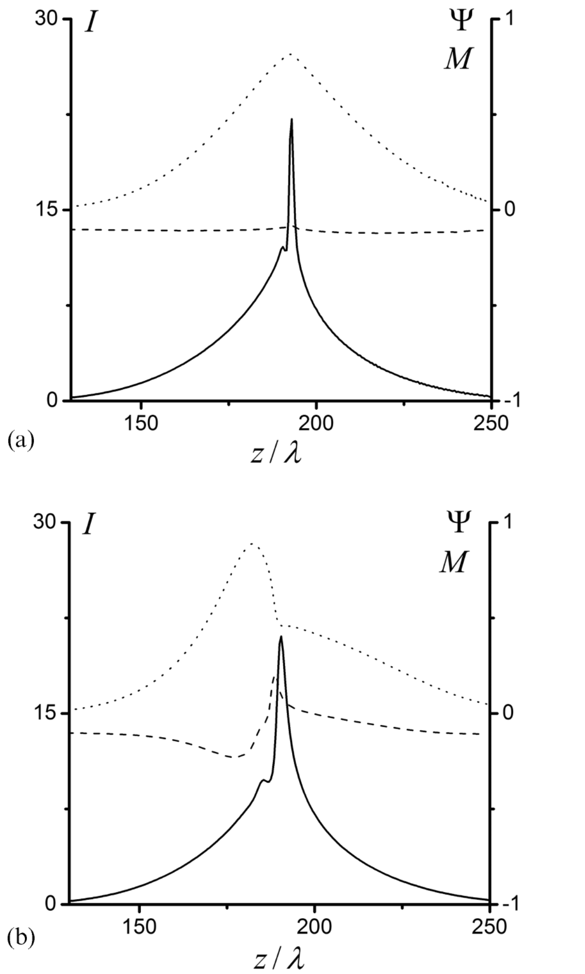

FIG. 2. The dependencies of the intensity $I(z/\lambda)$ (solid lines), the ellipticity degree $M(z/\lambda)$ (dashed lines), and the angle of rotation of the polarization ellipse $\Psi(z/\lambda)$ (dotted lines) on the propagation coordinate z/λ for the pulse traversed approximately 200 wavelengths in the nonlinear medium, $P = 10$ and $a = 2 \times 10^{-4} \times I_0^{-1}$, $b = 0$, $c = 0$ (a); $a = 0$, $b = 1 \times 10^{-4} \times I_0^{-1}$, $c = 1 \times 10^{-4} \times I_0^{-1}$ (b). The other parameters of the medium and the pulse are the same as in Fig. 1.

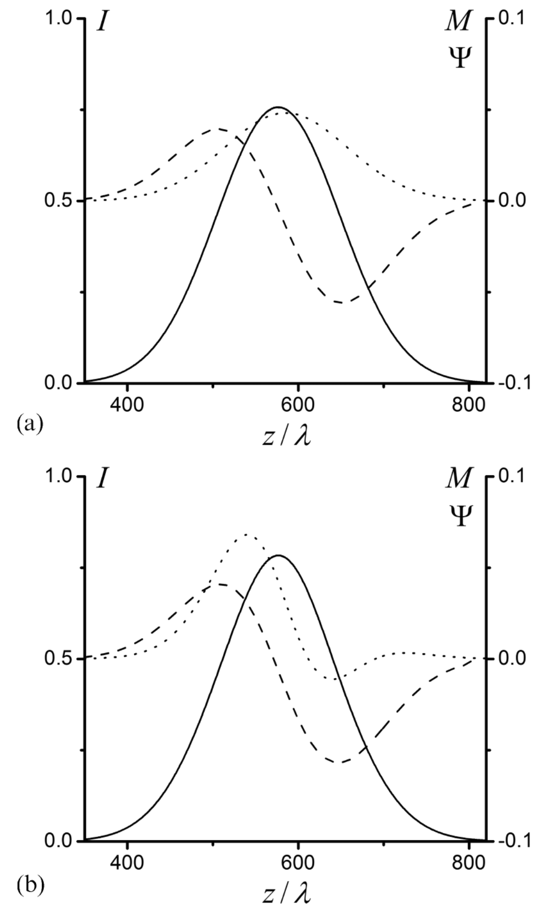

FIG. 3. The dependencies of the intensity $I(z/\lambda)$ (solid lines), the ellipticity degree $M(z/\lambda)$ (dashed lines), and the angle of rotation of the polarization ellipse $\Psi(z/\lambda)$ (dotted lines) on the propagation coordinate z/λ for the pulse traversed approximately 600 wavelengths in nonlinear medium. The intensity of the incident pulse is $P = 1$, its longitudinal half width (corresponding to the effective duration) is $w_0 = 300\lambda$, and the ellipticity degree is $M_0 = 0$. The nonlinear medium parameters are $\gamma_3 = 0.1 \times (\lambda I_0)^{-1}$, $d_3 = 0.05\lambda$, $a = 0$, $b = c = 2 \times 10^{-6} \times I_0^{-1}$ (a) and $b = c = 2 \times 10^{-4} \times I_0^{-1}$ (b); the other parameters of the medium and the pulse are the same as in Fig. 1. The increase of the $\chi^{(3)}_{xxyy}$ (at fixed γ_3) in (b) (by means of the increase of b and c) results in the asymmetric dependence of $\Psi(z/\lambda)$.

of the intensity and the polarization plane rotation angle. The pulse, which was linearly polarized at the beginning, now becomes elliptically polarized, and its front and backward parts have the ellipticity degree of opposite signs (opposite sense of polarization rotation). This is caused by the opposite signs of the intensity-dependent contributions to the effective refraction indices for the circularly polarized components of the electric field in a medium with spatial dispersion. In particular, this results in the difference of the group velocities of the circularly polarized components. If $\chi^{(3)}_{xxyy}$ is small, then the sense of rotation is the same in the front and in the backward parts of the pulse [see Fig. 3(a)]. The increase of the $\chi^{(3)}_{xxyy}$ (at fixed γ_3) results in the asymmetric dependence of $\Psi(z/\lambda)$ relatively to the center of the propagating long pulse [see Fig. 3(b)]. In this case its front and backward parts have the opposite sense of rotation of the electric field vector. When shortening the duration of the incident pulse, the dependencies $I(z/\lambda)$, $M(z/\lambda)$, and $\Psi(z/\lambda)$ become even more sophisticated. This effect is especially strong if w_0 is of the order of $c\tau_2$. The value of the angle of rotation of the polarization ellipse in the peak of the propagating pulse appears to be directly proportional to P (initial intensity) in a broad range of parameters of the radiation and medium with spatial dispersion of cubic nonlinearity, which is in accordance with the "classic" theory [1]. And, as was expected, the rotation of the polarization plane does not take place if $\gamma_3 = 0$ or $d_3 = 0$.

C. Propagation of ultrashort pulses in a medium with nonlocality of nonlinear optical response

When considering the ultrashort (several-oscillation) pulses the dispersion length calculated from the parameters of the linear response of the medium is about tens of wavelengths. Owing to the nonlinearity of the medium and to the nonlocality of its linear and nonlinear response, the propagating pulse,

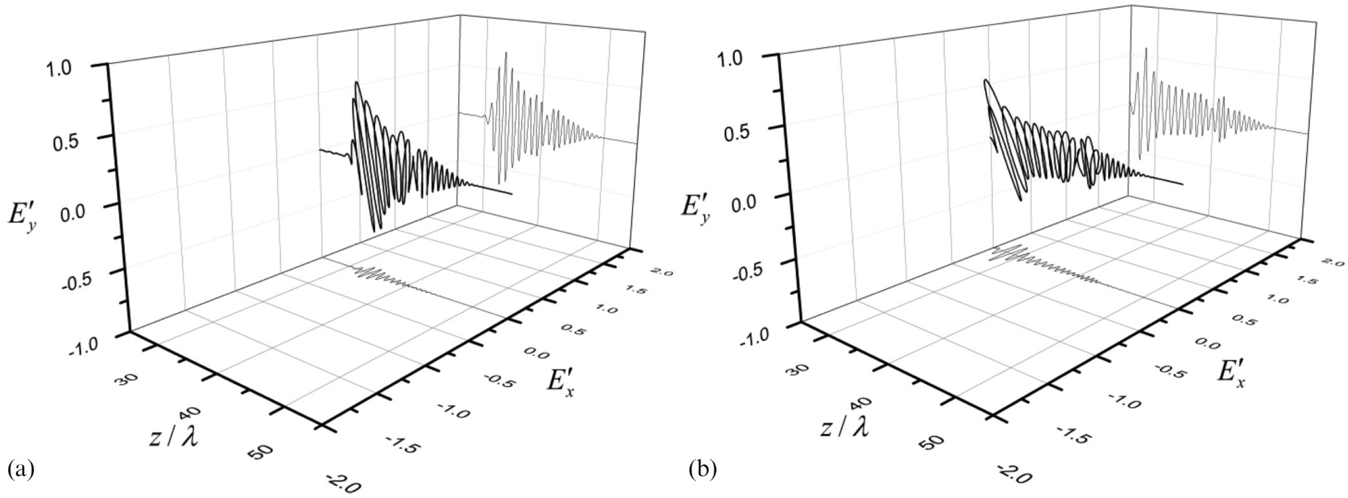

FIG. 4. The hodograph of the electric field strength vector for the pulse traversed 40 wavelengths in the nonlinear medium, $P = 2.5$ (a) and $P = 5$ (b). The half width of the incident pulse is five wavelengths. The nonlinear medium parameters are $\gamma_3 = -0.1 \times (\lambda I_0)^{-1}$, $d_3 = 0.05\lambda$, $a = 0$, $b = c = 10^{-3} \times I_0^{-1}$; the other parameters are the same as in Fig. 1.

which was linearly polarized at the medium border acquires an orthogonal polarization component during the propagation. In this case it is more convenient to represent the changes of the E_x and E_y by the hodograph of the electric field vector rather than by the conventionally defined in Eqs. (33) and (34) $M(z/\lambda)$ and $\Psi(z/\lambda)$ quantities, because in some fragments of the propagating pulse the changes of its Cartesian components cannot be treated as similar (even roughly) to the harmonic oscillations. The hodograph can be represented as a curve in the space of parameters $E'_x = E_x/(PI_0)^{1/2}$, $E'_y = E_y/(PI_0)^{1/2}$, and z, traced by the end of the electric field vector. The example of such a hodograph is shown in Fig. 4 for the pulse with initial half width of five wavelengths after traversing in a medium 40 wavelengths with $P = 2.5$ (a) and $P = 5$ (b). The line projected to the plane $E'_x = \text{const}$ represents the dependence $E'_y(z/\lambda)$, and the line projected to the plane $E'_y = \text{const}$ represents the dependence $E'_x(z/\lambda)$. It can be seen that the increase of the initial intensity results in the increase of the orthogonal polarization component. The hodograph looks like a deformed helix with changing radius, and its axis coincides with the z axis. In this case it is not possible to discuss the polarization state of light.

In a number of cases the helicity of the hodograph changes along the pulse (for example, from right to left, or vice versa), which reflects the change of the sense of rotation of the electric field vector. Such a case is well illustrated in Fig. 5, where the fragmentation of the pulse into two parts occurred, and the senses of rotation of the electric field vector within these parts are opposite.

If one considers the medium without spatial dispersion, when the hodographs of the electric field vector in the propagating pulses with initial values of polarization ellipse ellipticity degree $-M_0$ and M_0, as it was expected, are absolutely reflection symmetric with respect to one another relatively to the plane yz, because the optical properties of such a medium are identical for right- and left-handed circularly polarized components of light. In the presence of nonlocality of the nonlinear optical response of the medium ultrashort pulse with initial ellipticity degree M_0 of the same sign as $-\gamma_3\chi^{(3)}_{xxyy}(\omega; -\omega,\omega,\omega)$, rotates faster than in vacuum, and the pulse with the opposite sign of the ellipticity degree $-M_0$ rotates slower. Thus, various regimes of pulse propagation can take place depending on the relations between the quantities M_0, a, b, c, and γ_3. The hodographs of two pulses with initial ellipticities M_0 and $-M_0$ may rotate either in the same direction or in opposite directions with different rates of rotation (see Fig. 6) depending on the parameters of medium. Under certain conditions there can be realized the situation, when the hodograph of one pulse is rotating, and the hodograph of another almost does not rotate.

Our numerical investigations have shown that the pulse possessing initially circular polarization maintains its polarization state during the propagation in a medium with spatial

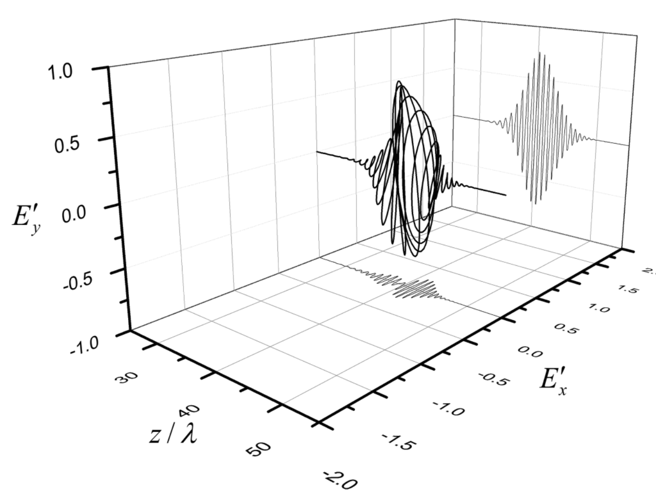

FIG. 5. The hodograph of the electric field strength vector for the pulse with peak intensity $P = 5$ and half width of five wavelengths after traversing 40 wavelengths in a medium with spatial dispersion of nonlinearity. Here $b = c = 10^{-4} \times I_0^{-1}$; the other parameters are the same as in Fig. 4.

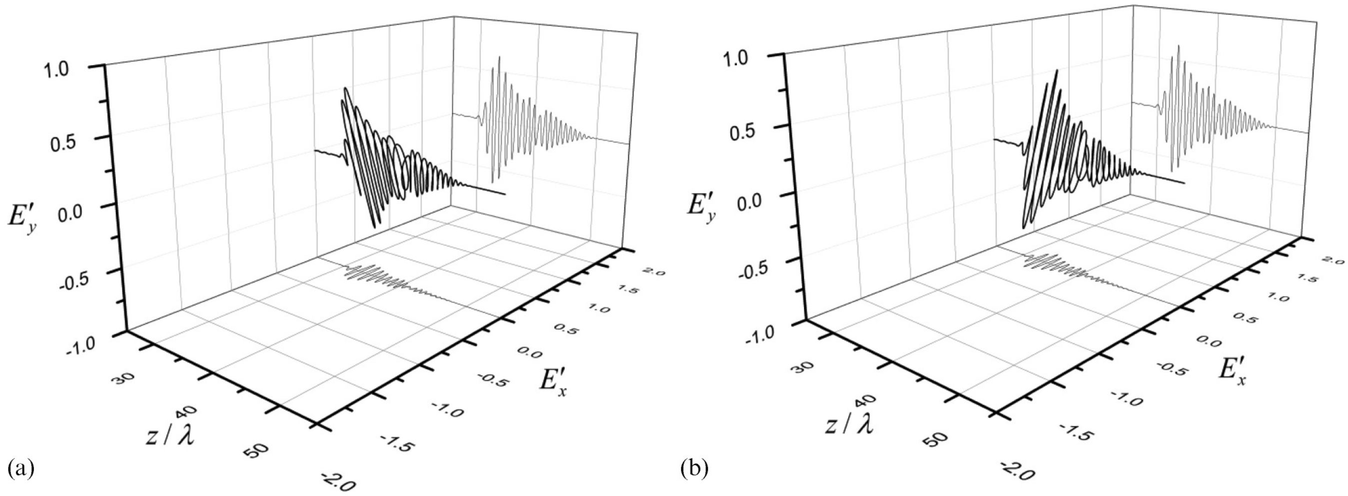

FIG. 6. The hodograph of the electric field strength vector for the pulse with peak intensity $P = 1$, half width of five wavelengths and the ellipticity degree $M_0 = 0.25$ (a) and $M_0 = -0.25$ (b) after traversing 40 wavelengths in a medium with spatial dispersion of nonlinearity. Here $b = c = 0.0025 \times I_0^{-1}$, $\gamma_3 = 0$, and $d_3 = 0$; the other parameters are the same as in Fig. 3.

dispersion of cubic nonlinearity. The same result is given by the slowly varying envelopes method. The nonzero value of γ_3 provides nonlinear circular dichroism, also predicted by the slowly varying envelopes method, which is the intensity-dependent difference of the absorption lengths for the circularly polarized components of the light field with opposite rotation directions. The latter is illustrated by Fig. 7, where the hodographs of the electric field vectors for pulses with $M_0 = 1$ (a) and $M_0 = -1$ (b) are shown. It can be seen that for a distance of 40 wavelengths in a medium with spatial dispersion the absorption for the pulse with $M_0 = -1$ (b) is remarkably stronger than that for a pulse with $M_0 = 1$ (a).

IV. CONCLUSION

In the present work, we propose the model of the nonlocal and nonlinear optical response of the medium with frequency dispersion, allowing one to formulate the material equations without the requirement of smallness of characteristic dimension scale of the nonlocality and without the limitations on the duration of the propagating pulse. The modification of FDTD with ADE was used for the description of the propagation of elliptically polarized pulses of arbitrary duration in such a medium. For relatively long pulses the results of the numerical analysis coincide with those obtained analogously within the SVEA. For the ultrashort pulses (containing about 10 or less oscillations of the electric field vector) the results of our numerical simulations significantly differ from those predicted by the SVEA, particularly, from the analytic expressions for the intensity-dependent ellipticity degree and the angle of rotation of the polarization ellipse in the case of propagation of monochromatic radiation. For this case we define new conventional conceptions of the ellipticity degree and the angle of rotation of the polarization ellipse, since the conception

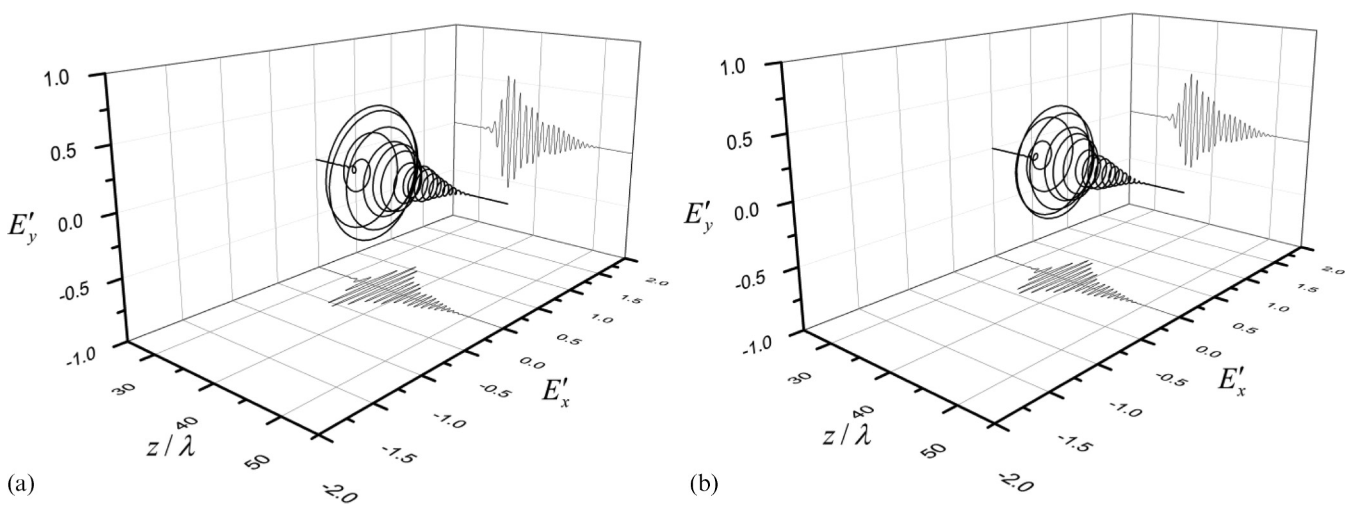

FIG. 7. The hodograph of the electric field strength vector for the circularly polarized pulse with peak intensity $P = 2.5$, half width of five wavelengths, and the ellipticity degree $M_0 = 1$ (a) and $M_0 = -1$ (b), after traversing 40 wavelengths in a medium with spatial dispersion of nonlinearity. The medium parameters are the same as in Fig. 4.

of the polarization state is only valid for monochromatic or quasimonochromatic (long enough) pulses. However, in certain cases, when the changes of the Cartesian components of the electric field in the propagating ultrashort pulse cannot be approximated (even roughly) by the harmonic oscillations, then it is rather correct to discuss the changes of the modulus of the electric field strength vector and its orientation. It is more convenient to analyze E_x and E_y by means of the hodograph of the electric field vector. The relations between the polarization parameters of the incident pulse and the medium parameters determine the scenario of the pulse evolution.

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