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Influence of the order parameter on the dynamics of ultrashort pulses in an environment with carbon nanotubes

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In this study, we address the influence of the order parameter on the three-dimensional dynamics of extremely short optical pulses in a nonlinear media made of carbon nanotubes creating a heterogeneous distribution of electrons. We obtained the effective nonlinear wave equation, which allowed us to analyze the dependence of the shape of three-dimensional ultrashort optical pulses on the relaxation rate of the order parameter, as well as on its equilibrium value. *Published by AIP Publishing.* [http://dx.doi.org/10.1063/1.4977011]

I. INTRODUCTION

In recent years, researchers have devoted significant attention to the study of strongly nonlinear media—including composite ones—due to advances in the synthesis of a variety of materials.[1–3] Among all the variety of newly investigated environments, there are two classes standing out: (i) materials undergoing phase transitions, which can be characterized in general by an order parameter quantifying its macroscopic level of ordering and (ii) media containing carbon nanotubes (CNTs). Environments with CNTs are of interest primarily for their unique nonlinear properties.[4–6] In particular, by virtue of the unique structure of CNTs, they can withstand very intense electric fields. At the same time, these substances are extremely sensitive to the orientation of the electric field because of the anisotropy of the structure of CNTs. Media capable of undergoing phase transitions—i.e., whose order parameter significantly changes following variations of a control parameter in the parameters space, are of paramount importance with regard to a myriad of novel practical applications.[7–9] Suffice it to say, that all ferroelectric and ferromagnetic materials belong to this type of materials. It is also worth mentioning that some important studies of carbon fibers doped with CNTs, which offer an array of promising practical applications.[10–12]

Despite several decades of intense theoretical and experimental activity on the subject of phase transitions in condensed matter physics, there is still a number of open questions, which continue to attract the attention of researchers. One such example is the nonequilibrium dynamics of the order parameter, especially in the presence of external alternating fields. The study of these issues remain to be fully understood from the theoretical point of view. In particular, the relaxation dynamics of the order parameter toward its equilibrium value is extremely important for practical applications. On the other hand, media containing CNTs can withstand extremely strong electromagnetic field and, as shown previously, they allow for the propagation of stable solitary states of the electromagnetic field.[13–24] Such localized wave packets that are localized in space and that can travel while retaining their spatiotemporal shape—in spite of diffraction and dispersion effects—are referred to as light bullets. It is worth adding that light bullets can be used to carry out the spectroscopy of such media and processes occurring therein.

Given all the above, the question of the interplay between phase transition properties and spectroscopic ones naturally arises for nonlinear media containing CNTs. There are several crucial questions in this regard, which can readily be addressed by means of a theoretical analysis. For instance, a specific dynamics of the order parameter may potentially lead to the collapse of light bullets due to an imbalance between dispersive and nonlinear effects. On the other hand, even if a light bullet is stable, there is a possibility that it is insensitive to the order parameter—i.e., insensitive to the level of ordering or to its relaxation dynamics. In other words, it is important to focus on both the stability of a light bullet, and its sensitivity to the order parameter. In practice, the order parameter can be a scalar, a vector, or any higher order representation depending on the nature of the problem. Here, we start with the simplest case of a scalar order parameter. However, one should note that the generalization to more complicated cases might not be trivial or straightforward.

This study is devoted to the analysis of possible scenarios for the influence of the order parameter on the dynamics of three-dimensional (3D) ultrashort optical pulses in an array of CNTs.

II. FORMULATION OF THE PROBLEM AND GENERAL EQUATIONS

Let us consider, for definiteness, the dynamics of a *scalar* order parameter in a nonlinear medium containing CNTs. To obtain the corresponding governing equation of motion for the order parameter, we exploit the approach developed in Refs. 7–9 such that

$$\frac{dP}{dt} = -\Gamma \frac{\delta \Phi}{\delta P}, \tag{1}$$

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where the functional derivative in the r.h.s. reflects the fact that P is in general a function of spatial and temporal coordinates, Γ is the kinetic coefficient, which determines the relaxation rate; P is the order parameter; Φ is the density of free energy functional. Indeed, according to the classical nonequilibrium thermodynamics, the rate of change of P should be proportional to the thermodynamic driving. In other words, the order parameter evolves such as to tend to its local free energy minimum. Note that in our framework, the order parameter can describe the different physical systems. For instance, in the case of ferroelectric (resp. ferromagnetic) materials, the order parameter P would then be the polarization (resp. the magnetization). Accordingly, the governing equation [see Eq. (2)] is able to describe the dynamics of a wide range of physical systems—this is one of the key features of the phenomenological theory developed by Patashinskii and Pokrovskii.[9] Furthermore, given the specificity of the problem at hand, it will be assumed that the scalar order parameter P is related to the electric field directed along the nanotube axis. For the sake of definiteness and clarity, we consider a ferroelectric medium with the polarization axis coinciding with the axis of the nanotube.

Based on the Ginzburg–Landau theory,[7–9] the free energy density Φ of a ferroelectric material, in the presence of an electric field and applied stress may be written as a Taylor expansion in terms of the order parameter P

$$\Phi = \Phi_0 + \alpha P^2 + \beta P^4 - \chi E P, \tag{2}$$

where Φ_0 corresponds to the origin of energy for a free unpolarized ($P=0$) and unstrained medium, and E is the applied electric field. Furthermore, one needs to take into account the fact that electrons of carbon nanotubes are subjected to action from both the electromagnetic field of the pulse and from the field of the environment. The latter appears due to the emergence of a nonzero order parameter, and is defined as

$$E_s = \frac{\delta\Phi}{\delta P}. \tag{3}$$

Let us consider the propagation of 3D extremely short electromagnetic pulses through an array of CNTs. The electric field of the pulse is assumed to be directed along the axis of the nanotubes, i.e., $\mathbf{E} = \{0,\ 0,\ E(\mathbf{r},\ t)\}$. The dispersion law for *zigzag* nanotubes $(m, 0)$ reads[24,25]

$$\epsilon_s(p) = \pm\gamma\left\{1 + 4\cos(ap)\cos\left(\pi\frac{s}{m}\right) + 4\cos^2\left(\pi\frac{s}{m}\right)\right\}^{1/2}, \tag{4}$$

where $s = 1,\ 2,\ldots m$, $\gamma \approx 2.7\,\text{eV}$, $a = 3b/2\hbar$, $b = 0.142\,\text{nm}$ is the distance between the neighboring carbon atoms.

The vector potential $\mathbf{A}$ and the current density $\mathbf{j}$ are assumed to have the following form $\mathbf{A} = \{0,\ 0,\ A(\mathbf{r},\ t)\}$ and $\mathbf{j} = \{0,\ 0,\ j(\mathbf{r},\ t)\}$, respectively. Thus, using the particular choice of Coulomb's gauge, $\mathbf{E} = -\frac{1}{c}\partial\mathbf{A}/\partial t$, one can follow the formalism developed in Ref. 17 to obtain the following expression for the current density:

$$j = -ean_0\gamma\sum_k D_k \sin\left(\frac{ke}{c}A(t)\right),$$
$$D_k = \sum_{s=1}^{m}\int_{-\pi/a}^{\pi/a} dp A_{ks}\cos(kp)\frac{\exp\left(-\epsilon_s(p)/k_BT\right)}{1+\exp\left(-\epsilon_s(p)/k_BT\right)}, \tag{5}$$

where n_0 is the equilibrium electron concentration in CNTs, ω_0 is the frequency of the external electric field E_0, k_B is the Boltzmann constant, T is the temperature, and c is the speed of light in vacuum. The coefficients A_{ks} arise from the expansion of the charge carriers velocity in Fourier series. With account for Coulomb's gauge and Eq. (5), the corresponding Maxwell's equation in a cylindrical coordinates system reads as

$$\frac{\partial^2\mathcal{A}}{\partial t'^2} = \frac{1}{r'}\frac{\partial}{\partial r'}\left(r'\frac{\partial\mathcal{A}}{\partial r'}\right) + \frac{\partial^2\mathcal{A}}{\partial z'^2} + \frac{1}{r'^2}\frac{\partial^2\mathcal{A}}{\partial\varphi^2} + \sin(\mathcal{A}+\mathcal{A}_s) + \sum_{k=2}^{\infty} D_k\sin(k\mathcal{A}+k\mathcal{A}_s), \tag{6}$$

where $\mathcal{A} = eaA/c$ is the single component of the dimensionless vector potential of the pulse field, and $\mathcal{A}_s = eaA_s/c$ corresponds to the electric field of the medium, $\mathbf{E}_s = -\frac{1}{c}\partial\mathbf{A}_s/\partial t$ [see also Eq. (3)]. We define the dimensionless coordinates as follows:

$$r' = r\frac{ea}{c}\sqrt{4\pi_0\gamma}, \quad z' = z\frac{ea}{c}\sqrt{4\pi_0\gamma}, \quad t' = tea\sqrt{4\pi_0\gamma}. \tag{7}$$

Here and thereafter, we assume the cylindrical symmetry, so that $\partial/\partial\varphi \equiv 0$. The latter is a simplifying approximation since we consider an array of nanotubes. However, previous estimates (see, e.g., Refs. 21–24) indicate that the resulting error can be considered to be negligible (less than 1%). It is worth noting that due to the field inhomogeneity along certain directions (e.g., the field is nonuniform along the z-axis, the current is also not uniform. The heterogeneity of the current causes an accumulation of charges in some areas that can be assessed from the charge conservation law

$$\frac{d\rho}{dt} + \frac{dj}{dz} = 0, \tag{8}$$

$$\rho \propto \tau\frac{j}{l_z}. \tag{9}$$

Here ρ is the charge density, j is the current density along the z-axis, τ is the pulse duration, and l_z is the characteristic length for the electric field change along the z-axis. Equation (9) allows us to conclude that the duration of a short pulse has a significant impact on the accumulated charge. Our estimates show that the accumulated charge is about 1%–2% of the charge, which contributes to the current. The latter allows one to neglect the charge accumulation effect for femtosecond pulses. This approximation has been validated by other numerical experiments for the case of CNTs and a pulse duration of tens of femtoseconds.[21–23]

III. RESULTS AND DISCUSSION

Equation (7) is solved numerically.[26] Steps in time and coordinates are determined from the stability conditions. They were decreased until the solution is not changed in the eighth decimal place. The initial condition is chosen in the form

$$A(z,r,t=0) = Q\exp\left\{-\left(\frac{z-z_0}{\gamma_z}\right)^2\right\}\exp\left(-\frac{r^2}{\gamma_r^2}\right), \quad (10)$$

$$\frac{d}{dt}A(z,r,t=0) = 2Qv_z\frac{z-z_0}{\gamma_z^2}\exp\left\{-\left(\frac{z-z_0}{\gamma_z}\right)^2\right\}\exp\left(-\frac{r^2}{\gamma_r^2}\right), \quad (11)$$

where Q is the field amplitude, γ_z and γ_r determine the pulse width in corresponding directions, v_z is the initial pulse velocity along the z-axis.

The evolution of the electromagnetic pulse as it propagates through the sample is shown in Fig. 1. Parameters χ and Γ were chosen in a range of values typical of ferroelectric materials; β determines T_c and does not actually affect the results of our study; α is governed by the distance from the critical point, $T - T_c$. One can observe a clear broadening of the ultrashort optical pulse during its propagation through the sample, thereby reducing the amplitude of the electric field associated with the pulse. This behavior is caused by the interaction of the current flowing through the carbon nanotubes with the subsystem described by the order parameter. The dynamics of this subsystem has a relaxation nature, which leads to a decrease in the electric field of the pulse.

The influence of relaxation rate of the order parameter Γ, on the process of propagation of ultrashort optical pulse is shown in Fig. 2. An increase in the relaxation rate Γ reduces the peak amplitude of the pulse along with an increase in the "tail" following the main pulse. This, in turn, shows the mechanism of reducing the amplitude of the pulse, which is related to the relaxation dynamics of the order parameter.

The dependence of the pulse shape on the parameter α is shown in Figure 3. It is important to note that in the Ginzburg–Landau theory of phase transitions, the magnitude of the parameter α is directly related to the distance of the

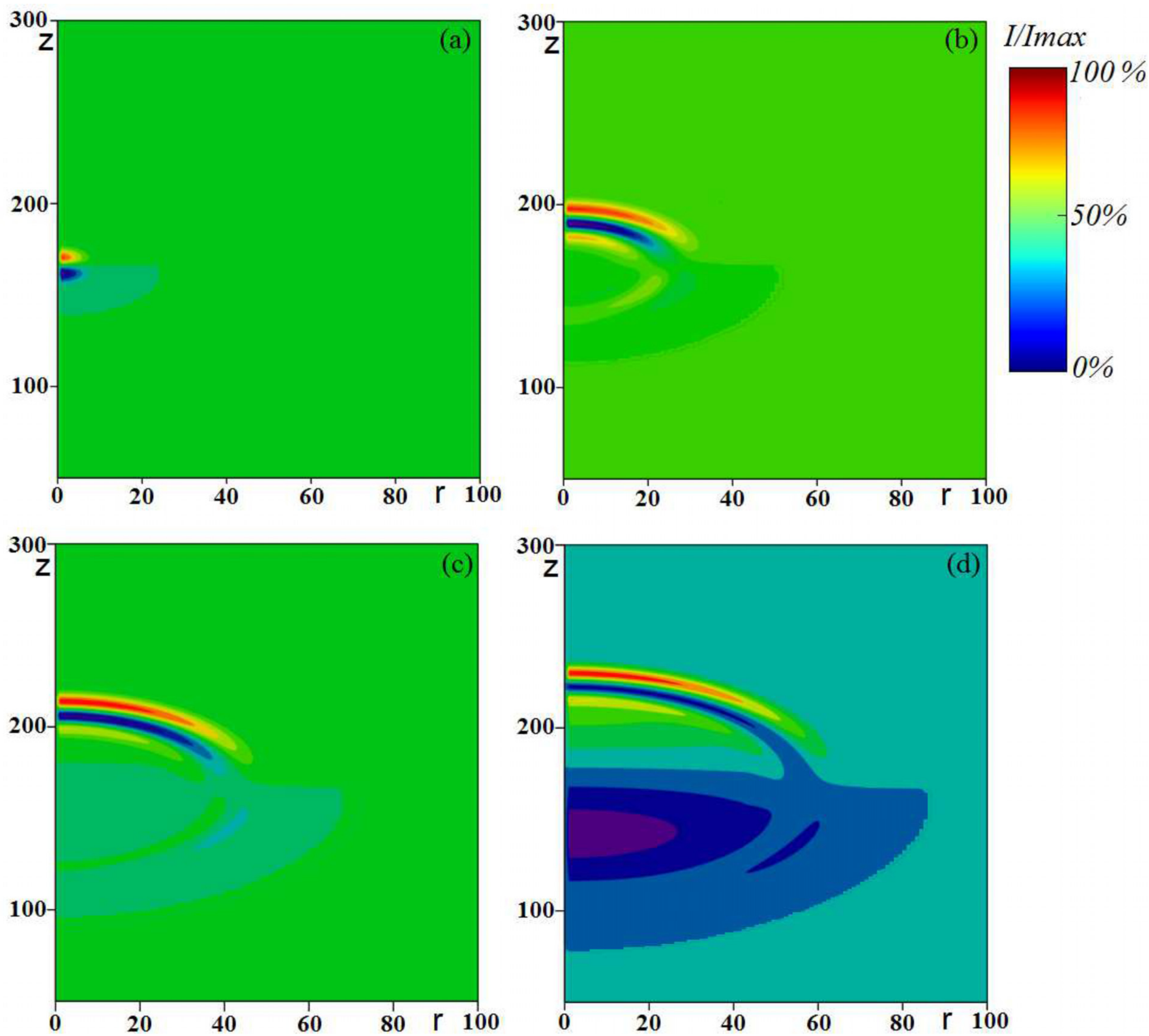

FIG. 1. Three-dimensional electromagnetic pulse intensity $I(r, z, t) = E^2(r, z, t)$ at different instants of time ($\alpha = 0.005$, $\beta = -1$, $\chi = 0.1$, $\Gamma = 0.1$): (a) initial pulse; (b) $t = 2 \times 10^{-13}$ s; (c) $t = 5 \times 10^{-13}$ s; and (d) $t = 7 \times 10^{-13}$ s.

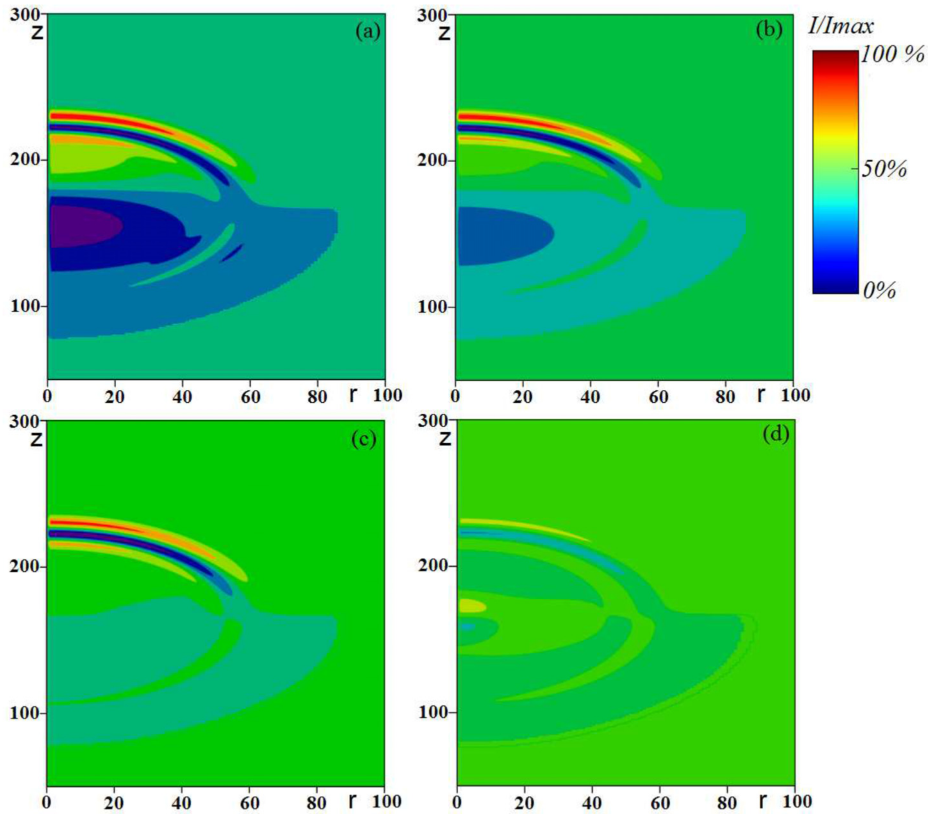

FIG. 2. Three-dimensional electromagnetic pulse intensity $I(r, z, t) = E^2(r, z, t)$ for different values of the relaxation rate Γ ($t = 7 \times 10^{-13}$ s, $\alpha = 1$, $\beta = -1$): (a) $\Gamma = 0.01$; (b) $\Gamma = 0.02$; (c) $\Gamma = 0.05$; and (d) $\Gamma = 0.1$.

critical point of phase transition, namely, $\alpha \propto T_c - T$, where T_c is the critical temperature, and T is the current temperature. The graphs in Fig. 3 show that the shape of the pulse is directly determined by the distance to the critical point. That important observation can in practice be used to identify experimentally the critical point. Thus, this makes it possible to investigate the dynamics of the order parameter with the help of ultrashort optical pulses.

Note that although pulses are experiencing some level of broadening due to inherently present dispersive effects on

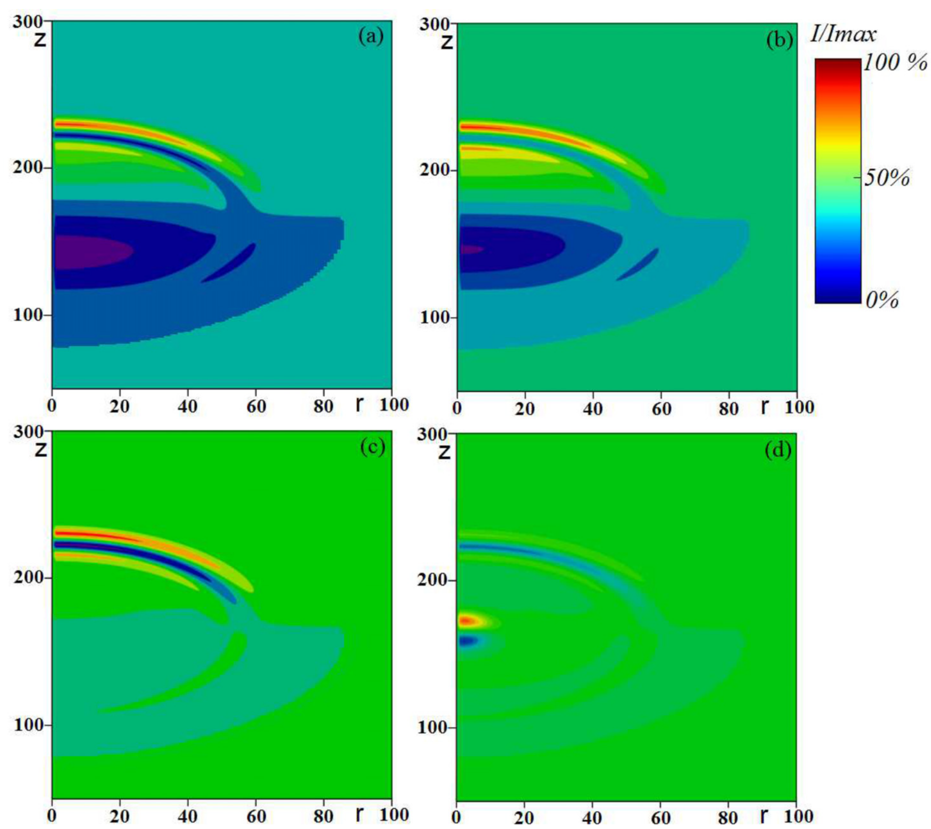

FIG. 3. Three-dimensional electromagnetic pulse intensity $I(r, z, t) = E^2(r, z, t)$ for different values of the parameter α ($t = 7 \times 10^{-13}$ s, $\Gamma = 0.1$): (a) $\alpha = 0.0$; (b) $\alpha = 0.1$; (c) $\alpha = 0.5$; and (d) $\alpha = 1.0$.

the medium, the pulse remains localized in space. Indeed, the main part of the pulse energy is still concentrated in a limited region of space; in that sense, the pulse propagation is stable. Furthermore, we would like to draw attention to the fact that the pulse propagation is not accompanied with a secondary wave radiation, which also speaks in favor of the stability of light bullets.

IV. CONCLUSIONS

As a result of our study, the following conclusions can be made:

(i) We derived the effective equation describing the dynamics of multidimensional extremely short optical pulses in an array of CNTs in a medium that can undergo phase transitions when subjected to variations of an order parameter.
(ii) Ultrashort optical pulses propagate with decaying amplitude as a consequence of the relaxation dynamics of the order parameter.
(iii) The possibility of carrying out spectroscopy by means of varying the order parameter while probing the medium with ultrashort pulses is demonstrated.

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