Local field effect as a function of pulse duration

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In this brief report we give semiclassical consideration to the role of pulse duration in the observation of local field effects in the regime of optical switching. We show that the main parameter governing local field influence is the ratio of peak Rabi frequency corresponding to medium inversion and Lorentz frequency of the medium. To obtain significant local field effect, this parameter should be near unity that is valid only for long enough pulses. We also discuss the role of relaxation and pulse shape in this process.

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The concept of local field was introduced in the second half of the nineteenth century by Hendrik Antoon Lorentz and Ludvig Valentin Lorenz [1]. They demonstrated that the microscopic (local) electric field $\mathbf{E}_L$ acting on atoms or molecules of the medium is different from the macroscopic applied field $\mathbf{E}$. This difference is due to polarization of the medium $\mathbf{P}$ and describes the near dipole-dipole (NDD) interactions between atoms or molecules. The well-known expression for the local field in the case of isotropic homogeneous media is

$$\mathbf{E}_L = \mathbf{E} + \frac{4\pi}{3}\mathbf{P}. \tag{1}$$

Utilization of this relation leads to the classic Clausius-Mossotti equation between microscopic (molecular polarizability) and macroscopic (dielectric permittivity) parameters of the medium [2]. Local field correction (1) is a good approximation in the case of nonresonant dense gases, liquids, and solids. Moreover, it can be used to determine the refractive index even for such quantum medium as Bose-Einstein condensate [3–5] including the effects of atomic correlations.

It turned out that it leads to some fundamental effects if one considers radiation interacting with a dense collection of resonant two-level atoms. This system known as a dense resonant medium should contain many atoms within a cubic resonant wavelength [6]. The strength of NDD interactions between atoms of this medium is measured by value of the Lorentz frequency $\omega_L = 4\pi\mu^2 C/3\hbar$, where μ is a transition dipole moment, C is the atom concentration per unit volume, and $\hbar$ is the Planck constant. The most studied effect induced by the presence of the local field is the intrinsic optical bistability which results in two-valued dependence of the population difference between the ground and excited states on light intensity in a stationary regime. This effect was predicted theoretically [7] and then observed experimentally [8]. The condition of bistability existence can be formulated as an inequality $b = \omega_L T_2 > 4$ [9], where T_2 is the transverse relaxation time. Realistic estimates of value of b show that, as a rule, it does not exceed several units, that is, $b \leqslant 10$. For example, for gaseous media with typical parameters $\mu^2 = 10^{-38}$ erg cm^3, $T_2 = 10^{-9}$ s^{-1}, and $C = 10^{20}$ cm^{-3} [10], we have $b \approx 4$. Condensed matter, for example, excitonic media, possesses substantially greater dipole moments, but they are compensated by relatively small atomic concentrations [11].

Local field correction results in some remarkable effects on pulse propagation in resonant medium. Some of them are connected with new solitary wave properties, such as distinctions of soliton form from the standard hyperbolic-secant envelope and its area from 2π [12]. NDD interactions play a crucial role in the generation of the so-called "incoherent" solitons [11]. They influence soliton formation in the so-called resonantly absorbing Bragg reflectors [13], pattern formation in lasers [14], and ultrashort, few-cycle pulse propagation in dense resonant medium [15,16]. Crenshaw *et al.* [17] considered ultrafast optical switching of the medium between ground and excited states due to the action of a coherent pulse, that is, a pulse whose duration is much less than the relaxation times of the medium, $t_p \ll T_1, T_2$. Switching was obtained for the pulses with peak Rabi frequencies $\Omega_p = \mu E_p/\hbar$ (E_p is the peak amplitude of the electric field) approximately equal to the Lorentz frequency of the medium, that is, $\Omega_p/\omega_L \approx 1$. This is valid independent of the pulse area; however, if the pulse is very short, it contains only a small fraction of π and, obviously, cannot excite the medium. Therefore, this switching effect holds true only for pulses long enough, namely, for $\omega_L t_p > 1$. This condition can be rewritten as $bt_p/T_2 > 1$ and, taking into account pulse coherence, gives $b \gg 1$, which seems not to be realistic. In the next article [18], a more moderate condition was considered, $\omega_L t_p \leqslant 1$, together with taking into account propagation effects. The results of that work were obtained for pulses of picosecond durations. On the other hand, in a femtosecond regime, the influence of NDD interactions on pulse propagation was reported to be negligible, at least for realistic values of b and ω_L [19].

In this brief report we carefully examine the role of pulse duration in the appearance of local field effects. We assume the pulse to be intensive enough to excite the medium; that is, the regime of ultrafast switching is considered. Therefore the main dimensionless parameter of our research is $\psi = \Omega_0/\omega_L$, where Ω_0 is the characteristic peak Rabi frequency corresponding to the pulse that switches the medium. Ω_0 can be found due to the conception of the pulse area. Indeed, if we take the equality

$$2\frac{\mu}{\hbar}\int_{-\infty}^{\infty} E dt = 2\pi \tag{2}$$

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and assume the pulse to have Gaussian shape $E = E_0 \exp(-t^2/2t_p^2)$, we obtain

$$\Omega_0 = \sqrt{\frac{\pi}{2}}\frac{1}{t_p}. \tag{3}$$

So, the main parameter is

$$\psi = \frac{\Omega_0}{\omega_L} = \sqrt{\frac{\pi}{2}}\frac{1}{\omega_L t_p}. \tag{4}$$

This value allows us to say whether the local field correction is significant or not. It is seen that ψ is dependent on pulse duration.

Our main thesis is that local field effects can be observed when the ratio ψ is near unity (relatively long pulses), while they can be neglected in the case of $\psi \gg 1$ (short pulses). Further we prove this statement directly by numerical simulations of pulse propagation inside a dense two-level medium. The model used is based on the semiclassical Maxwell-Bloch system for population difference W, microscopic polarization R, and electric field amplitude $\Omega' = \Omega/\omega = (\mu/\hbar\omega)E$ (in dimensionless form) [6,19,20]:

$$\frac{dR}{d\tau} = i\Omega' W + iR(\delta + \epsilon W) - \gamma_2 R, \tag{5}$$

$$\frac{dW}{d\tau} = 2i(\Omega'^* R - R^*\Omega') - \gamma_1(W - 1), \tag{6}$$

$$\frac{\partial^2\Omega'}{\partial\xi^2} - \frac{\partial^2\Omega'}{\partial\tau^2} + 2i\frac{\partial\Omega'}{\partial\xi} + 2i\frac{\partial\Omega'}{\partial\tau} = 3\epsilon\left(\frac{\partial^2 R}{\partial\tau^2} - 2i\frac{\partial R}{\partial\tau} - R\right), \tag{7}$$

where $\tau = \omega t$ and $\xi = kz$ are dimensionless arguments; $\delta = \Delta\omega/\omega$ is the normalized detuning of the field carrier (central) frequency ω from the atomic resonance; $\gamma_1 = (\omega T_1)^{-1}$ and $\gamma_2 = (\omega T_2)^{-1}$ are the rates of longitudinal and transverse relaxation, respectively; $\epsilon = \omega_L/\omega$ is the normalized Lorentz frequency; $k = \omega/c$ is the wave number, and c is the light speed in vacuum. Here we assume that the background dielectric permittivity of the medium is unity (two-level atoms in vacuum). Equations (5) and (6) are derived in the framework of the rotating wave approximation (RWA) which requires $\Omega' \ll 1$ [21]. This condition is satisfied throughout the article. In Eq. (7) we do not use slowly varying envelope approximation (SVEA) which cannot hold true even for thin films of the medium as noted in Ref. [18]. Description based on Eqs. (5) and (6) does not take into account such processes as multiple scattering, radiation reabsorption, and spontaneous emission which result in quantum corrections of the Lorentz-Lorenz relation [22,23]. However, many usual effects of light propagation such as self-induced transparency can be correctly treated in semiclassical approximation [21].

In our calculations we use Gaussian pulses with peak amplitudes (3) and central wavelength $\lambda = 0.5$ μm. We consider the case of strict resonance, that is, $\delta = 0$. Initially (before pulse incidence) the medium is in the ground state, that is, $W = 1$ and $R = 0$. Thickness of the layer of the medium is $L = 5\lambda$. NDD interactions between two-level atoms provide Lorentz frequency $\omega_L = 10^{11}$ s (note, that $\omega_L \ll \omega$). This value is believed to be high enough according to the typical parameters described previously.

First, we consider the case of coherent pulses; that is, for the phenomenological relaxation terms in Eqs. (5) and (6), we assume $\gamma_1 = \gamma_2 = 0$. This allows us to study the pure effect of the local field without any side effects connected with relaxation. As one can see in all parts of Figs. 1(i) and 1(ii), the influence of NDD interactions on the dynamics of pulses with durations $t_p = 0.1$ and 1 ps ($\psi = 125$ and 12.5, respectively) is negligible. For shorter (femtosecond) pulses this is valid as well, in accordance with the results of Ref. [19]. Such pulses act as usual 2π ones, first inverting the medium and then returning it exactly into the ground state. Transmitted pulses demonstrate shape transformation resulting in pulse compression [19], while reflected radiation is almost absent. This situation can be treated as a self-induced transparency (SIT) regime.

When we further make pulse duration greater, for $t_p = 5$ and 10 ps [$\psi = 2.5$ and 1.25, all parts of Figs. 1(iii) and 1(iv)], local field effects become apparent. Inversion of the medium is reached later in comparison with the case of the absence of NDD interactions. At the same time, the transmitted pulse is decreasing, while the reflected one is getting more intensive. At $t_p = 10$ ps almost the entire initial energy of radiation is transformed into the reflected pulse. Perhaps, this is connected with the effect of coherent internal reflection which was studied in the stationary regime earlier [24,25]. However, the local field results in larger transmittance as compared with the case when it is absent [see Fig. 1b(iv)].

Now let us add phenomenological relaxation. We take typical parameters $T_1 = 1000$ ps and $T_2 = 100$ ps, so that the NDD interaction parameter is $b = \omega_L T_2 = 1$. For pulse durations $t_p = 0.1$ and 1 ps the results are almost the same as those in the relaxation-free case [see all parts of Figs. 1(i) and 1(ii)]. But for longer pulses we have to take into account relaxation. It is seen in Fig. 2a(i) that for $t_p = 5$ ps relaxation results in energy conservation inside the medium for a long time (the population difference does not reach unity) and, hence, the output (transmitted and reflected) radiation is only a small fraction of the incident one [compare with Fig. 1(iii)]. For the pulse with $t_p = 10$ ps [Fig. 2a(ii)], relaxation of the population difference on the entrance of the medium is slow, too. However, this results in strong reflection rather than trapping of pulse energy. The time shift of both population difference and peak of reflected radiation in the case of local field correction is seen as well. Therefore, one can say that local field effects appear in the regime of internal reflection rather than in the regime of self-induced transparency.

Finally, we should discuss the question of pulse shape. One can see in Fig. 1a(iv) that the behavior of the population difference for a long pulse with $t_p = 10$ ps is different from that in Figs. 1a(i) and 1a(ii) even in the case when the local field correction is absent. This is due to the Gaussian shape of such a long pulse. For comparison we take the invariant pulse with hyperbolic secant shape, $E = E_0 \mathrm{sech}(t/t_p)$. The condition (2) leads in this case to the peak Rabi frequency

$$\Omega_0 = \frac{1}{t_p}. \tag{8}$$

Figure 3 demonstrates that the curve of the population difference for hyperbolic secant pulse with peak amplitude (8) and duration $t_p = 10$ ps is really less deformed as compared with Gaussian pulse of the same duration. However, all other

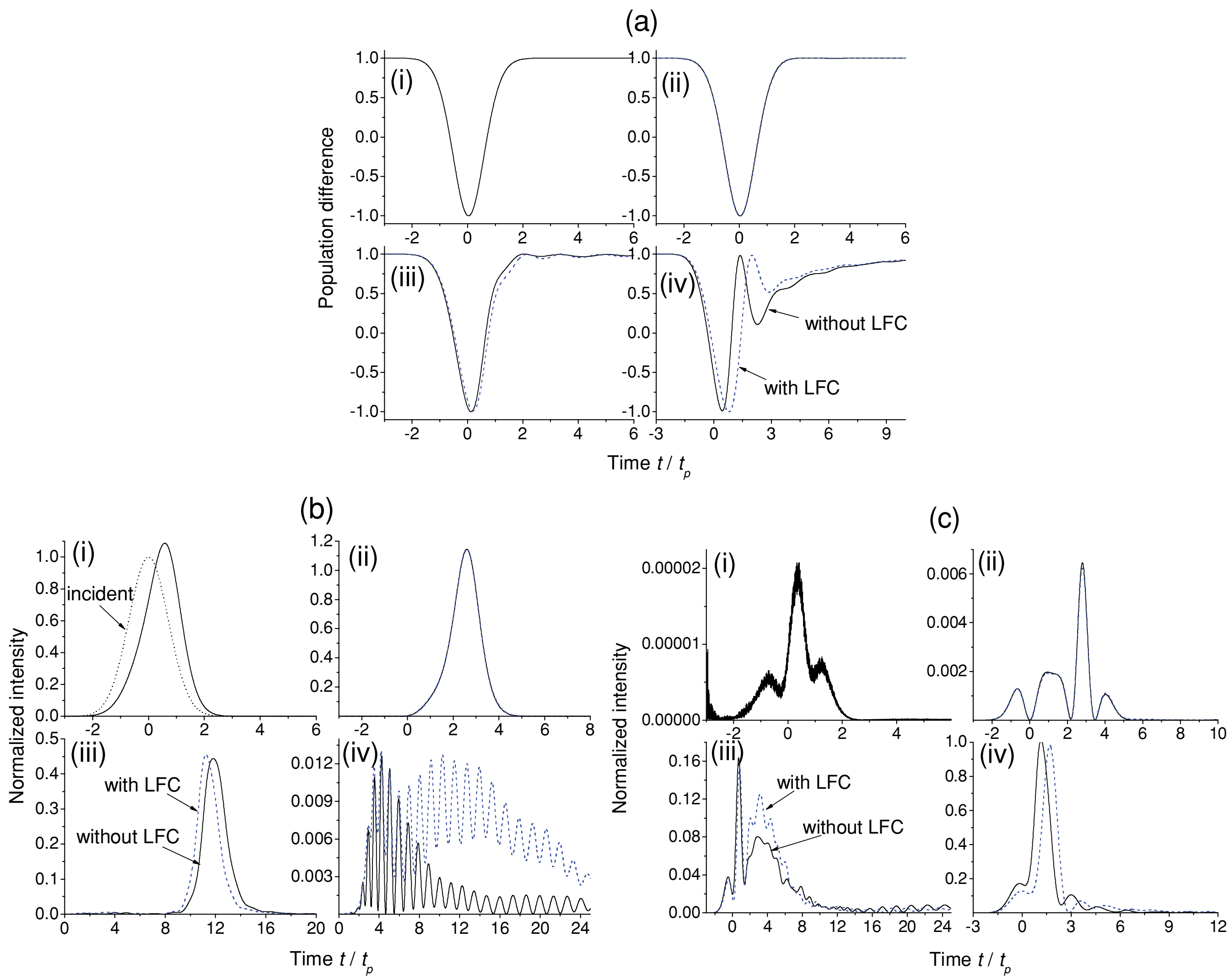

FIG. 1. (Color online) (a) Population difference on the entrance of the medium, (b) transmitted and (c) reflected radiation at different pulse durations: (i) $t_p = 0.1$ ps, (ii) $t_p = 1$ ps, (iii) $t_p = 5$ ps, and (iv) $t_p = 10$ ps. Relaxation is absent. Results correspond to calculations without local field correction (LFC) (solid lines) and with it (dashed lines) in Eq. (5).

peculiarities (e.g., predominant reflection) are still valid in this case. The same statement is true for qualitative properties of the effect of the local field correction on pulse propagation in the dense two-level medium considered.

In conclusion, in this note we considered the case of pulse propagation in a dense two-level medium in the regime of optical switching. It is clearly demonstrated by direct numerical calculations that the local field effect on pulse

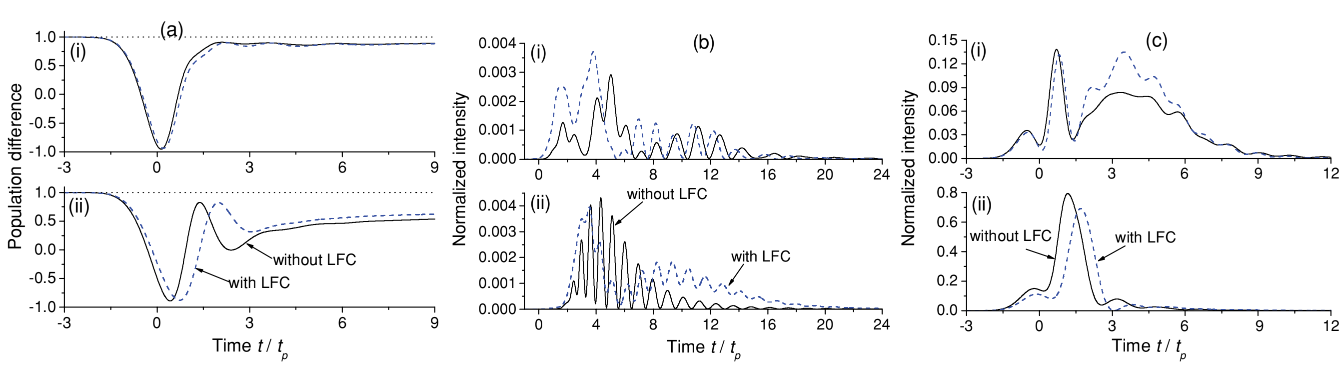

FIG. 2. (Color online) (a) Population difference on the entrance of the medium, (b) transmitted, and (c) reflected radiation at different pulse durations: (i) $t_p = 5$ ps, (ii) $t_p = 10$ ps. Relaxation times $T_1 = 1000$ ps and $T_2 = 100$ ps. Results correspond to calculations without local field correction (LFC) (solid lines) and with it (dashed lines) in Eq. (5).

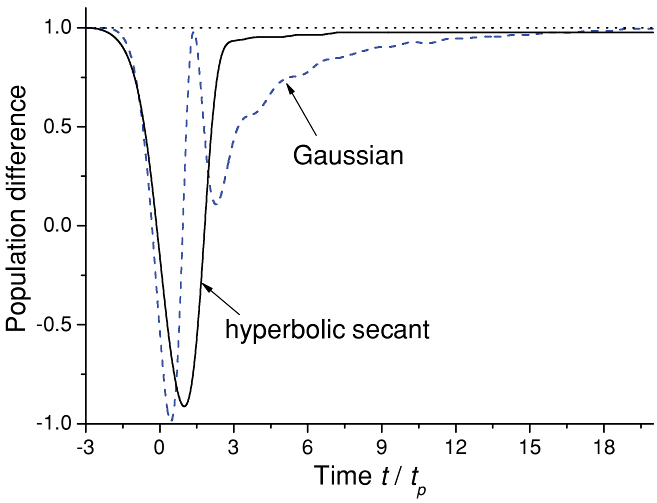

FIG. 3. (Color online) Population difference on the entrance of the medium for different pulse shapes: hyperbolic secant and Gaussian. Pulse duration $t_p = 10$ ps. Relaxation and local field correction are absent.

propagation in such media is dependent on pulse duration. The governing parameter ψ is the ratio of peak Rabi frequency (characteristic for medium switching) and Lorentz frequency of the medium. For short (femtosecond) pulses this ratio is large, and we have the regime of self-induced transparency without any significant influence of local field. In other words, as pulse duration is decreasing, one needs to have much greater Lorentz frequencies (that seems not to be realistic) to obtain any local field effect. On the other hand, when Lorentz frequency is increasing as medium is getting more dense, one has to take into account the processes of multiple scattering (and, hence, radiation trapping), which was ignored in our study. For long (picosecond) pulses, such that $\psi \sim 1$, the influence of the local field becomes apparent, while the SIT regime transforms into the regime of coherent internal reflection. On the other hand, the relaxation processes (just as the pulse shape) can be sufficient in the case of long pulses. The results obtained may be used for proper choice of the parameters of experiments dealing with local field observation (at least, in some special experimental geometries).

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