Freezing light via hot atoms

Olga Kocharovskaya, ^{1,2} Yuri Rostovtsev, ¹ and Marlan O. Scully ^{1,3}

¹ Department of Physics and Institute for Quantum Studies, Texas A&M University,

College Station, Texas 77843-4242

² Institute of Applied Physics, RAS, Nizhny Novgorod, Russia

³ Max-Planck-Institut für Quantenoptik, D-85748 Garching, Germany

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We prove that it is possible to freeze a light pulse (i.e., to bring it to a full stop) or even to make its group velocity negative in a coherently driven Doppler broadened atomic medium via electromagnetically induced transparency (EIT). This remarkable phenomenon of the ultra-slow EIT polariton is based on the spatial dispersion of the refraction index $n(\omega, k)$, i.e., its wavenumber dependence, which is due to atomic motion and provides a negative contribution to the group velocity. This is related to, but qualitatively different from, the recently observed light slowing caused by large temporal (frequency) dispersion.

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Slow group velocity in coherently driven media [1] has been shown to provide new regimes of nonlinear interaction with highly increased efficiency even for very weak light fields, high precision spectroscopy and magnetometry [2]. It has been demonstrated [1,3,4] that EIT is accompanied by large frequency dispersion, $|\omega \partial n/\partial \omega| \gg 1$, and can slow the group velocity down to $10 - 10^2$ m/s.

In this paper we show that, using spatial dispersion due to atomic motion, it is possible to freeze the light, $v_g = 0$, or even to make its group velocity opposite to the wavevector, $v_g < 0$ (see Eq. (1)). We consider two different types of atomic media: (i) atomic beam or uniformly moving sample, and (ii) hot gas in a stationary cell.

Freezing of light in a stationary cell via a hot gas is especially intriguing (Fig. 1). The idea is to tune the driving field to resonance with the velocity group of atoms that moves in the direction opposite to the light pulse with velocity equal to the light group velocity that would be supported by this group of atoms if they were at rest.

The main result of the present paper is contained in Fig. 2 which shows that v_g can be zero, for a pulse in a hot gas, when the drive detuning $\Delta \omega_d$ is properly chosen.

As is well-known, in a medium possessing both temporal and spatial dispersion of the refraction index, $n(\omega, k) = 1 + 2\pi\chi(\omega, k)$, the group velocity of light contains two contributions,

$$v_g \equiv Re \frac{d\omega}{dk} = Re \frac{c - \omega \frac{\partial n(\omega, k)}{\partial k}}{n(\omega, k) + \omega \frac{\partial n(\omega, k)}{\partial \omega}} = \tilde{v}_g - v_s. \quad (1)$$

Eq. (1) is an immediate result of differentiating the dispersion equation $kc = \omega n(\omega, k)$, i.e., $c = v_g(n + \omega \partial n/\partial \omega) + \omega \partial n/\partial k$. The meaning of Eq. (1) becomes clear if one turns to the equation for a field amplitude

$$\left(c\frac{\partial}{\partial z} + \frac{\partial}{\partial t}\right)\mathcal{E} = 2\pi i\omega \int dz' dt' \chi(t-t',z-z')\mathcal{E}(t',z').$$

Using the convolution theorem to write the RHS as $\int d\bar{k}d\bar{\omega}\chi(\bar{\omega},\bar{k})\mathcal{E}(\bar{\omega},\bar{k})\exp i(\bar{\omega}t-\bar{k}z)$, expanding the susceptibility to the first order in \bar{k} , $\bar{\omega}$, noting that \bar{k} and $\bar{\omega}$ under the integral may be written in terms of $\partial/\partial z$ and $\partial/\partial t$ acting on $\mathcal{E}(t,z)$ and rearranging terms we have

$$\left(c - 2\pi\omega \frac{\partial \chi}{\partial k}\right) \frac{\partial \mathcal{E}}{\partial z} + \left(1 + 2\pi\omega \frac{\partial \chi}{\partial \omega}\right) \frac{\partial \mathcal{E}}{\partial t} = 2\pi i\omega \chi(\omega, k)\mathcal{E}$$

which implies the field equation with v_a given by Eq. (1),

$$\left(v_g \frac{\partial}{\partial z} + \frac{\partial}{\partial t}\right) \mathcal{E} = 2\pi i k \chi(\omega, k) \left[1 + 2\pi \omega \frac{\partial \chi}{\partial \omega}\right]^{-1} \mathcal{E}.$$

The first term in Eq. (1), $\tilde{v}_g = Re[c/(n + \omega \partial n/\partial \omega)],$ is due to frequency dispersion, and was discussed in recent papers [1–4]. The second term, $v_s =$ $Re[(\omega \partial n/\partial k)/(n+\omega \partial n/\partial \omega)]$, is due to the effect of spatial dispersion, i.e., nonlocal response of the medium to a probe field. We study dilute systems where the susceptibility is small, $|\chi(\omega, k)| \ll 1$, but $v_q \ll c$, as it is for all the EIT experiments carried out so far. As usual, we consider real-valued group velocities under the condition that imaginary part of $d\omega/dk$ is negligible. Otherwise group velocity looses its simple kinematic meaning and strong absorption governs or prevents propagation of the light pulse through the medium. The latter is the reason why the resonant interaction of light with a two-level medium never results in an ultra-slow polariton.

A mono-velocity atomic beam or uniformly moving sample corresponds to the simple case of spatial dispersion, so-called drift dispersion. In the co-moving frame atoms are at rest, there is no spatial dispersion and the group velocity is given by the first term of Eq. (1) alone, \tilde{v}_g . The Galilean transformation to the laboratory frame, $k = \tilde{k}, \quad \omega = \tilde{\omega} - \tilde{k}v$, where v is the atomic velocity, yields the group velocity $v_g = Re(d\omega/dk) = \tilde{v}_g - v$.

Eq. (1) yields the same result, since the susceptibility [5,6] depends only on the combination $\omega + kv$,

$$\chi_v(\omega, k) = \chi(\omega + kv) = \frac{i\mu_{ab}^2 N}{\hbar} \frac{n_{ab} \Gamma_{cb} + \Omega^2 n_{ca} / \Gamma_{ac}^*}{\Gamma_{ab} \Gamma_{cb} + \Omega^2}.$$
(2)

Here $n_{ab} = \rho_{aa} - \rho_{bb}$, $n_{ca} = \rho_{cc} - \rho_{aa}$, ρ_{ii} is the population of the ith level, γ and γ_{cb} are the relaxation rates of excited state and c-b coherence respectively $(\gamma \gg \gamma_{cb})$; ω_{ab} and ω_{cb} are the frequencies of the optical and low frequency transitions ($\omega_{ab} \gg \omega_{cb}$); ω_d , k_d and ω , k are the frequency and wavenumber of the driving and probe fields respectively, N is the atomic density, $\Omega = |\mu_{ac} E_d|/2\hbar$ is the Rabi frequency of drive field $(1/2)E_d \exp(i\omega_d t - ik_d z) + c.c.$, μ_{ac} and μ_{ab} are the dipole moments of a-c and a-b transitions respectively, $\Gamma_{ac} = \gamma + i(\Delta\omega_d + k_d v)$, $\Gamma_{ab} = \gamma + i(\Delta\omega + k v)$, $\Gamma_{cb} = \gamma_{cb} + i(\Delta\omega - \Delta\omega_d + \Delta kv), \ \Delta\omega_d = \omega_d - \omega_{ac},$ $\Delta\omega = \omega - \omega_{ab}$, $k_d = \omega_d/c$, $k = k_d + \omega_{cb}/c + \Delta k$. We use a standard model with incoherent pump and loss rates $(r_c = r_b = \gamma_{cb}/2)$, assuming time of flight broadening of b-c transition [6] (Fig. 1), so that in the absence of fields $\rho_{cc} = \rho_{bb} = 1/2$. According to Eqs. (1), (2), we again obtain $v_g = \tilde{v}_g - v$. The physical reason for this drifting of the pulse is that the field is basically "seized" by the atoms in the form of atomic polarization.

An important question is how to input the light pulse into the gas. There are different possibilities. One example uses a grid mirror that has the grid stripes of small area, so that atoms can freely fly through the mirror, and small spacing between the grid stripes as compared to the wavelength of light to provide efficient reflection, as in Fig. 1b. If atoms are at rest, the light would propagate in the forward direction. However, if the velocity of atoms is equal to (or larger than) \tilde{v}_g , one should see a frozen (or backward) pulse.

Depending on the mechanism of pulse input into the medium, one should look for the solution of the problem with initial (time), boundary (space), or mixed (timespace) conditions. In the case of the initial value problem, we solve the dispersion equation for $\omega = \omega(k)$, Fig. 3a. Galilean transformation ensures the same EIT half width, $\Delta k_{EIT} = \Omega^2/\gamma \tilde{v}_g$, as for the atoms at rest since $Im[\tilde{\omega}(\tilde{k})] = Im[\omega(k)]$. In the case of the boundary value problem, we find $k = k(\omega)$. The result shows narrowing of the EIT dip proportional to the kinematic factor $\alpha = (\tilde{v}_g - v)/\tilde{v}_g$. Indeed, in the accompanying frame the dispersion relation near EIT resonance can be decomposed in a form of a quadratic polynomial, $\Delta \tilde{k} = \Delta k_0 - i[\kappa_0 + \xi(\Delta \tilde{\omega} - \Delta \omega_d)^2] + (\Delta \tilde{\omega} - \Delta \omega_d)/\tilde{v}_g$. Its Galilean transformation to the laboratory frame yields

$$\Delta k = \Delta k_0 + \frac{1}{\alpha} \left[\frac{\delta \omega}{\tilde{v}_g} - i\kappa_0 - i\xi \left(\frac{\delta \omega}{\alpha} \right)^2 \right], \tag{3}$$

where $\delta\omega = \Delta\omega - \Delta\omega_d$. Coefficients in Eq. (3) can be easily deduced using Eq. (2). For example, for the case

of one-photon resonance $\Delta \omega_d = 0$ at $\Omega^2 \gg \gamma_{cb} \gamma$, we have $\tilde{v}_g = \hbar \Omega^2 / 2\pi \mu_{ab}^2 k_d N$, $\Delta k_0 = 0$, residual absorption coefficient at the center of EIT dip is $\kappa_0 = \gamma_{cb}/\tilde{v}_g$, and a coefficient determining the parabolic profile of absorption in the EIT dip is $\xi = \gamma/\Omega^2 \tilde{v}_g$. This approximation is valid if residual absorption is small, $\kappa_0 \xi \ll (1-v/\tilde{v}_g)^2/v^2$. Absorption increases twice as much as EIT minimum value at detuning $\delta \omega_{EIT} = |\tilde{v}_g - v| \Omega \sqrt{\gamma_{cb}/\gamma}/\tilde{v}_g$ that is much less than the EIT half width $\Delta \omega_{EIT} = \Delta k_{EIT} |\tilde{v}_g - v|$.

Eq. (3) shows that the absorption coefficient $Im\ k$ is increased and sharpened by a factor $(\tilde{v}_g - v)/\tilde{v}_g$ as compared to that in the co-moving frame. Since the spectrum of the pulse cannot be transformed on the stationary boundary, only those spectral components that are within the sharpened EIT dip penetrate deep into the medium. For drift velocity $v > \tilde{v}_g$, the backward EIT polariton can be excited from inside a cell (Fig. 1b).

In the case of an atomic beam with a moving boundary (or moving sample), i.e., for the mixed boundary-initial value problem, the spectrum (inverse duration) of the pulse shrinks at the moving boundary exactly in the same way as the EIT width in Eq. (3), $\Delta\omega = \Delta\tilde{\omega}(\tilde{v}_g - v)/\tilde{v}_g$. This is not a coincidence, but is necessary for consistency of viewing of the same process from different frames. The pulse within the EIT dip decays in time with the same rate independently of whether it propagates through atoms at rest or through a beam, since this decay is predetermined by atomic relaxation γ_{cb} , γ .

Atoms with a thermal velocity distribution. Let us consider a stationary cell of hot atoms. If the intensity of the drive is strong enough to provide EIT for the resonant group of atoms (see Fig. 4) but at the same time weak enough to avoid an interaction with off-resonant atoms, moving with "wrong" velocities, it is mainly this drifting beam that would support the ultra-slow EIT polariton with zero or even negative group velocity.

To prove this we calculate the dispersion law $\omega(k)$ for the EIT polariton in a hot gas in a cell at rest. The susceptibility is given by an average of the beam susceptibility over a velocity distribution F(v) of atoms in a gas with thermal velocity v_T , $\chi(\omega, k) = \int_{-\infty}^{+\infty} dv F(v) \chi_v(\omega, k)$. Instead of the Maxwellian thermal distribution we can use Lorentzian, $F(v) = v_T/[\pi(v_T^2 + v^2)]$, since the far-offresonant tails are not important. This allows us to obtain simple analytical results because an integration over velocities is reduced to a sum of a few residues in the simple poles, $v = v_i$. Only those poles count that lay in the lower half complex v-plane in the formal limit of infinitely large growth rate $Im\omega \to -\infty$. For a positive wavenumber detuning, $\Delta k > 0$, there are two such poles. One originates from Lorentzian, $v_1 = -iv_T$, and the other from the velocity dependent populations, $v_2 = -(i\gamma G + \Delta\omega_d)/k_d$. Here $\gamma G = \gamma \left(1 + \Omega^2/(\gamma_{cb}\gamma)\right)^{1/2}$ determines the velocity width of an effective drifting beam of atoms that are driven by an external field into a coherent "dark"

state [5,6], and, hence, responsible for the ultra-slow EIT polariton (see Fig. 4). For $\Delta k < 0$, there is an additional pole, $v_3 \propto 1/\Delta k$, originated from resonance $\Gamma_{ab}\Gamma_{cb} + \Omega^2 = 0$ in Eq. (2). However, near EIT resonance, i.e., for small detuning Δk , it enters the lower half plane from infinity, $v_3 \to -i\infty$, so that its contribution is negligible if $N \ll k_d^3(\gamma_{cb}/\gamma)\sqrt{k_d v_T/\Omega}$.

Calculation of the residues at poles v_1 and v_2 yields

$$\chi(\omega, k) = \frac{i\mu_{ab}^2 N}{2\hbar} \left[\frac{\eta_1}{\Omega^2 + \Gamma_{ab}^{(1)} \Gamma_{cb}^{(1)}} + \frac{\eta_2}{\Omega^2 + \Gamma_{ab}^{(2)} \Gamma_{cb}^{(2)}} \right], (4)$$

where $\eta_1 = [R_1 \Gamma_{ac}^{(1)} - \Gamma_{cb}^{(1)} (1 + 2\gamma R_1/\gamma_{cb})]/[1 + \gamma^2 (G^2 - 1)R_1/\Omega^2], \quad \eta_2 = k_d v_T R_2 [\Omega^2/(G - 1) - \Gamma_{cb}^{(2)} \gamma]/\gamma_{cb} \gamma G,$ $\Gamma_{ab}^{(1)} = \gamma + k v_T + i \Delta \omega , \quad \Gamma_{ab}^{(2)} = \gamma (1 + Gk/k_d) + i(\Delta \omega - k \Delta \omega_d/k_d), \quad \Gamma_{ac}^{(1)} = \gamma + k_d v_T + i \Delta \omega_d, \quad \Gamma_{cb}^{(1)} = \gamma_{cb} + |\Delta k| v_T + i(\Delta \omega - \Delta \omega_d), \quad \Gamma_{cb}^{(2)} = \gamma_{cb} + |\Delta k| \gamma G/k_d + i(\Delta \omega - \Delta \omega_d - \Delta k \Delta \omega_d/k_d), \quad R_1 = \Omega^2/[\gamma^2 + (\Delta \omega_d - ik_d v_T)^2],$ $R_2 = \Omega^2/[(k_d v_T)^2 + (\Delta \omega_d + i\gamma G)^2].$

The susceptibility (4) of a hot gas looks like the susceptibility of a medium consisting of just two mono-velocity components: (i) broad background with velocity v=0 and linewidth $\gamma+k_dv_T$, and (ii) a drifting beam with velocity $v_d=-\Delta\omega_d/k_d$ and power broadened linewidth $\gamma(1+G)$ (see Fig. 4). This interpretation becomes very accurate near EIT dip, $|\Delta\omega-\Delta\omega_d|\ll\gamma G$, at the conditions necessary for the existence of freezing ultra-slow EIT polariton: a) low-frequency coherence decay is much slower than optical decay $(\gamma_{cb}\ll\gamma)$; b) drifting beam width is less than Doppler broadening $(\gamma G\ll k_dv_T)$; c) detuning of driving and probe fields from one-photon resonance is large enough $(|\Delta\omega_d|\gg\gamma G)$ while two-photon resonance is maintained. Then, for the ultra-slow EIT polariton, the susceptibility is approximated as

$$\chi = \frac{\mu_{ab}^2 N'}{\hbar \gamma G} \left[\frac{\Omega^2}{\gamma (1+G)(\omega - \omega_k)} - i \right], \tag{5}$$

if we keep only resonant ω -dependence in denominators setting everywhere else $\Delta\omega=\Delta\omega_d$. Here $N'=N\gamma Gk_dv_T/[(k_dv_T)^2+\Delta\omega_d^2]\ll N$ is the density of atoms in the drifting beam. The resonant denominator, where $\omega_k=\omega_{ab}+\Delta\omega_dk/k_d+i\gamma_k,\ \gamma_k=\gamma_{cb}+\Omega^2/\gamma(1+G)+|\Delta k|\gamma G/k_d$, comes from the factor $\Omega^2+\Gamma_{ab}^{(2)}\Gamma_{cb}^{(2)}$ in Eq. (4). Thus, we explicitly find the frequency and the decay $(\omega_k,\ \gamma_k)$ of the EIT exciton coupling of which to the probe field produces the ultra-slow polariton.

For the boundary value problem, Eq. (5) yields a dispersion that is similar to that for the mono-velocity beam (3) with parameters $v=v_d,\ \tilde{v}_g'=[(k_dv_T)^2+\Delta\omega_d^2]\Omega^2\hbar/[\mu_{ab}^2N\gamma(1+G)k_d^2v_T],\ \kappa_0=\gamma_{cb}/\tilde{v}_g',\ \xi=1/\gamma_k\tilde{v}_g'.$

For the initial value problem, from the dispersion equation $kc = \omega(1 + 2\pi\chi)$ and Eq. (5), we find dispersion law

$$\Delta\omega = \Delta\omega_d - v_d \Delta k + i\gamma_k - \frac{\Omega^2}{\gamma(1+G)} \left[\frac{\hbar \gamma G \Delta k}{2\pi \mu_{ab}^2 k_d N'} + i \right]^{-1}$$
(6)

shown in Fig. 3b. The EIT half width is $\Delta k'_{EIT} = \gamma_k/\tilde{v}'_g$. For small detuning $|\Delta k| \ll \Delta k'_{EIT}$, Eq. (6) yields linear dispersion and parabolic decay profile, $\Delta \omega = \Delta \omega_d + \Delta k(\tilde{v}'_g - v_d) + i\gamma_{cb} + i\Delta k^2\tilde{v'}^2/\gamma_k$. Decay increases twice as much as EIT minimum value, $Im\Delta\omega = 2\gamma_{cb}$, at very small detuning $\delta k'_{EIT} = \sqrt{\gamma_{cb}\gamma_k}/\tilde{v}'_g \ll \Delta k'_{EIT}$. The group velocity describes pulse kinematics if $d\omega/dk$ has negligible imaginary part, i.e., near the center of the EIT dip where $|\Delta k| < |\tilde{v}_g - v_d|\gamma_k/\tilde{v}'_g^2$. The last inequality does not mean that the pulse cannot be stopped. It just means that when the pulse is frozen, $v_g = \tilde{v}'_g - v_d = 0$, its evolution is governed by the dispersion of absorption.

Fig. 3 clearly shows that the ultra-slow EIT polariton in a hot gas is similar to that in a mono-velocity beam, since detuning of driving field picks a beam with velocity $v_d = -\Delta \omega_d/k_d$. However, effective density of atoms supporting EIT polariton N' and EIT width $\Delta k'_{EIT} = \gamma_k/\tilde{v}'_g$ in a hot gas are different because of factors γG and F(v). As a result, the group velocity at the EIT resonance, according to Eq. (6), in terms of a critical density is

$$v_g = \frac{\beta N_{cr}}{NF(v_d)} - v_d, \quad N_{cr} = \frac{\hbar\Omega}{2\pi^2 \beta \mu_{ab}^2} \sqrt{\frac{\gamma_{cb}}{\gamma}}, \tag{7}$$

where $\beta = \max[v_d F(v_d)]$. For Lorentzian $F(v_d)$, we have $\beta=1/2\pi,$ and $v_g=(v_d-v_d^{(1)})(v_d-v_d^{(2)})N_{cr}/2Nv_T$ is a quadratic polynomial over v_d , i.e., the group velocity is zero for drive detunings $v_d^{(1,2)}=v_T[N/N_{cr}\pm$ $\sqrt{(N/N_{cr})^2-1}$ and negative between them for density higher than the critical value, $N > N_{cr}$, as is shown in Fig. 2. To achieve minimal group velocity, $\min v_g = -(v_T N/2 N_{cr})[1 - (N_{cr}/N)^2],$ one has to tune at $v_d = v_T N/N_{cr}$. The condition to freeze or reverse the light $(v_a \leq 0)$ means that the group velocity supported by the drifting beam with the density $N' = \pi N F(v_d) \gamma G/k_d$ should be equal to or less than the velocity of atoms in the beam, i.e., $\tilde{v}'_q = \tilde{v}_q N'/N \leq v_d$. If we compare a mono-velocity beam with a hot gas at $v_d = v$ and the same N' as the total density N in a beam to provide the same group velocity, $\tilde{v}_g = \tilde{v}_q'$, we find that the EIT width and the residual decay in a hot gas are $G \simeq \Omega/\sqrt{\gamma_{bc}\gamma}$ times less than in a beam. To minimize N_{cr} the drive intensity should be as low as possible to decrease \tilde{v}'_a due to power broadening effect and to avoid EIT contribution from the atoms with "wrong" (positive) velocities. That is, the drive intensity should be just above a threshold of the EIT effect at resonance, $\Omega^2 > \gamma_{cb}\gamma$. Under realistic parameters relevant to the experiments with ⁸⁷Rb vapor [4] and chozen in Figs. 2-5, the critical density is $N_{cr} \sim 10^{11} \text{ cm}^{-3}$.

Absorption or time variation of the drive field results in a spatial or time dependence of the group velocity in the cell. This allows us to control input and parameters of the pulse in the cell. According to geometrical optics, the parameters of the EIT polariton adiabatically follow the local properties of the driven atoms. Fig. 5 demonstrates how the ultra-slow pulse decelerates up to the point $v_g = 0$ where it becomes frozen.

The important conclusion is that the drifting beam provides large enough drift spatial dispersion $\partial n/\partial k$ (see Eq.(1)) to ensure $v_g \leq 0$. Although the density of drifting atoms is small $N' \ll N$, their resonant contribution dominates. This allows us to make the group velocity zero or even negative [7]. To observe freezing or backward light one can look, e.g., for a scattering, luminescence, delay, or enhanced nonlinear mixing caused by ultra-slow pulse.

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- FIG. 1. (a) Three-level atomic Λ -system. (b) Geometry of ultra-slow EIT pulse propagation in the gas of atoms.

- FIG. 2. Ultra-slow and negative group velocity of EIT polariton vs detuning of drive laser; $\Omega = 0.25\gamma$, $k_dv_T = 100\gamma$, $\gamma_{cb} = 0.001\gamma$, (a) $N = 0.6N_{cr}$; (b) $N = N_{cr}$; (c) $N = 1.5N_{cr}$.
- FIG. 3. Dispersion, $Re\Delta\omega = Re[\omega \omega_{ab}]$, and decay, $Im\Delta\omega$, spectra of the ultra-slow EIT polariton according to numerical solution of the dispersion equation for: (a) atomic beam $(N=1.1N_{cr}\pi F(v_d)\gamma G/k_d)$ with susceptibility (2); (b) stationary cell of hot gas $(N=1.1N_{cr})$ with exact susceptibility (4); $\Omega=0.25\gamma$, $v=v_d=v_T$, $k_dv_T=100\gamma$, $\gamma_{cb}=0.001\gamma$.
- FIG. 4. The velocity distribution of atoms in a cell (solid line). Effective drifting beam (dotted) selected by drive laser.
- FIG. 5. Kinematics of the deceleration of the ultra-slow pulse to the point of freezing $(v_g=0)$ along a cell with decreasing group velocity $v_g(z)$. Positions of pulse are shown at subsequent moments of time $t=m\tau$ ($\tau=3L/2v_g(0)$, m=0,1,2,3). $v_g(z)$ is calculated numerically according to decreasing drive intensity found from the wave equation for the same parameters as in Fig. 3(b), L=10 cm.