

# Evolution of ultrashort light pulses in a two-level medium visualized with the finite-difference time domain technique

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**Abstract:** The finite-difference time-domain technique is employed to examine the evolution of the amplitude, duration, waveform, and phase of ultrashort light pulses propagating in a medium of two-level atoms or molecules. The results of these numerical simulations agree reasonably well with predictions of the McCall–Hahn analysis for the evolution of the amplitude and the phase of short pulses in a two-level medium until the pulse duration becomes less than the duration of a single optical cycle. Noticeable deviations from the McCall–Hahn scenario were observed for pulses with durations shorter than the duration of a single field cycle.

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## 1 Introduction

An impressive progress achieved recently in the generation of ultrashort light pulses with a duration corresponding to a few cycles of the optical field [1, 2] allowed unique measurements with an unprecedentedly high resolution to be performed and stimulated many applications [3, 4]. Several methods of generating subfemtosecond and attosecond pulses using high-order harmonic generation [5 – 7] and coherent Raman sidebands [8 – 10] are now extensively discussed. The methods of theoretical analysis of the generation and propagation of ultrashort light pulses have to meet the challenges of the rapidly progressing experimental technique. In particular, many useful physical results obtained within the framework of slowly varying envelope approximation have to be tested very carefully with the use of numerical methods. As a part of this program, in this paper, we integrate numerically the Maxwell equations to test the general predictions of McCall and Hahn [11, 12] for the evolution of light pulses propagating through a two-level medium. According to these predictions, pulses whose areas are multiple of  $\pi$  propagate in a two-level medium with no changes in their area, while pulses with other values of the area change their area during the propagation in a two-level medium until their area reaches a value multiple of  $\pi$  [11, 12]. The pulse area then remains stable for even multiples of  $\pi$  and is unstable for odd multiples of  $\pi$ .

Much analytical and numerical work has been done recently to extend this analysis, including the classical pulse area theorem, to short pulses. Eberly [13] has rederived the area theorem for the case of short light pulses, modifying this theorem to include pulse chirping and homogeneous damping and obtaining a new equation for the evolution of the pulse phase. Ziolkowski et al. [14] applied the finite-difference time-domain (FDTD) technique [15] to numerically solve the semiclassical Maxwell-Bloch equations. This approach revealed several important features of short-pulse propagation in a two-level medium and allowed a more detailed analysis of self-induced transparency effects. Hughes [16, 17] has recently employed the FDTD approach to demonstrate the possibility of generating subfemtosecond transients in a two-level medium.

In this paper, we apply an FDTD-based algorithm solving Maxwell and Schrödinger equations to model the interaction of ultrashort laser pulses with an ensemble of two-level atoms or molecules in the sharp-line case. Since no SVEA assumptions are employed in this case, such an approach seems to offer especially much promise for the analysis of the propagation of ultrashort pulses through two-level media. We shall employ the FDTD technique to examine the evolution of the amplitude, duration, waveform, and phase of short pulses propagating in a two level medium and compare the results of these simulations with predictions of the McCall-Hahn analysis. In addition to the findings earlier reported by Hughes [17], we will also demonstrate that noticeable deviations from the McCall-Hahn scenario are observed for pulses with durations shorter than the duration of a single field cycle and explore the physical origin of these deviations.

## 2 The FDTD procedure solving Maxwell and Schrödinger equations for a two-level medium

We start with the extension of the standard FDTD procedure to the case of short pulses propagating in a two-level medium, when the Maxwell equation for the fields and the Schrödinger equation for the wave functions should be solved without any assumptions that are usually employed in the SVEA approach. In the one-dimensional case, the FDTD algorithm involves step-by-step integration of two curl Maxwell equations for the  $E_z$  and  $H_y$  components of the electric and magnetic fields

$$\frac{\partial D_z(x,t)}{\partial t} = \frac{\partial H_y(x,t)}{\partial x}, \quad \frac{\partial H_y(x,t)}{\partial t} = \frac{\partial E_z(x,t)}{\partial x}, \quad (1)$$

where  $D_z$  is the  $z$ -component of the electric displacement vector.

To perform this integration, we have to define the relation between the components of the electromagnetic induction and the electromagnetic field. This can be done through the

equation for the polarization of the medium. In our case of a two-level medium, this involves the solution of the Schrödinger equation for the wave functions of the energy levels.

We will consider an ensemble of noninteracting two-level atoms or molecules whose wave functions can be represented as superpositions of two basis states 1 and 2:

$$\psi(t) = a(x, t)\psi_1 + b(x, t)\psi_2, \quad (2)$$

where  $\psi_1$  and  $\psi_2$  are the eigenfunctions of an unperturbed system corresponding to the states with energies  $E_1$  and  $E_2$  (we assume for definiteness that  $E_1 > E_2$ ), respectively, and  $a(x, t)$  and  $b(x, t)$  are complex coefficients. Then, the Schrödinger equation for the wave function yields the following set of differential equations:

$$i\hbar \frac{da(x, t)}{dt} = E_1 a(x, t) - \mu E_z(x, t) b(x, t), \quad i\hbar \frac{db(x, t)}{dt} = E_2 b(x, t) - \mu E_z(x, t) a(x, t), \quad (3)$$

where  $\mu$  is the dipole moment of transition between the levels 1 and 2.

Our approach to the joint solution of Eqs. (1) – (3) involves the use of the FDTD algorithm [15], which uses a difference approximation of time and spatial derivatives appearing in Maxwell equations [Eq. (1)]:

$$D_{z,i}^{n+1} = D_{z,i}^n + \frac{\Delta t}{\Delta x} (H_{y,i+1/2}^{n+1/2} - H_{y,i-1/2}^{n+1/2}), \quad H_{y,i+1/2}^{n+3/2} = H_{y,i+1/2}^{n+1/2} + \frac{\Delta t}{\Delta x} (E_{z,i+1}^{n+1} - E_{z,i}^{n+1}), \quad (4)$$

where  $i$  and  $n$  indicate the values of discrete spatial and temporal variables, respectively,  $x = i\Delta x$ ,  $t = n\Delta t$ ,  $\Delta x$  and  $\Delta t$  are the steps of discretization in spatial and temporal variables. Relating the components of the electric displacement vector to the components of the electric field through the polarization of the medium, as mentioned above (see also [15]), we arrive at a closed algorithm simulating the propagation of an ultrashort laser pulse in a medium of two-level atoms (molecules).

### 3 Testing McCall–Hahn predictions for very short pulses

To test the FDTD-based procedure of simulations described in Section 2, we will model the propagation of light pulses

$$E(x, t) = A(x, t)e^{i\phi + ikz - i\omega t} + \text{c.c.}, \quad (5)$$

where  $A(x, t)$  is the pulse envelope,  $\phi$  is the phase, in a two-level medium in the sharp-line case and compare the results of these simulations with the predictions of the McCall–Hahn analysis for the evolution of a pulse envelope in a two-level medium [12].

The SVEA equation for the pulse envelope  $A(x, t)$  can be represented after McCall and Hahn [11, 12] as

$$\frac{\partial A(x, t)}{\partial x} + \frac{n}{c} \frac{\partial A(x, t)}{\partial t} = -\frac{2\pi\omega\mu N}{nc} \sin(\theta(x, t)), \quad (6)$$

where  $n$  is the refractive index of the medium,  $N$  is the number density of resonant species,

$$\theta(x, t) = \int_{-\infty}^t \Omega(x, \tau) d\tau, \quad \Omega(x, t) = 2\mu / \hbar A(x, t) \text{ is the real Rabi frequency.}$$

The celebrated solution to Eq. (6) is a pulse with a hyperbolic-secant shape, which propagates in a resonant two-level medium with no changes in its envelope:

$$A(x, t) = \frac{\hbar}{\mu\tau} \operatorname{sech} h \left( \frac{t - x/V}{\tau} \right), \quad (7)$$

where  $\tau$  is the pulse duration,  $V$  is the pulse velocity in the medium,

$$V = \left[ \frac{4\pi\mu^2\tau^2\omega N}{\hbar nc} + \frac{n}{c} \right]^{-1} = \left[ \frac{4\pi\hbar\omega N}{E_0^2 nc} + \frac{n}{c} \right]^{-1}. \quad (8)$$

As can be seen from Eq. (8), the group velocity  $V$  of the pulse propagating in a two-level medium differs from its phase velocity, which is equal to  $c/n$ . Thus, the time required for an arbitrary point on the pulse envelope to cover a distance  $X$  is  $X/V$ . The change in the phase of the pulse corresponding to this period of time is then given by

$$\varphi = kX - \omega X/V = \omega X(1/v - 1/V) = -4\pi\omega^2\mu^2\tau^2 NX / (\hbar nc). \quad (9)$$

Therefore, the characteristic length corresponding to the phase shift of the pulse equal to  $\pi$  is

$$L = (\hbar nc) / (4\omega^2\mu^2\tau^2 N) = (E_0^2 nc) / (4\omega^2 N \hbar). \quad (10)$$

The results of FDTD simulations for the propagation of light pulses in a two-level medium agree very well with general predictions of McCall and Hahn [11, 12] until the pulse duration becomes less than the duration  $T$  of a single optical cycle (Figs. 1a – 1d).

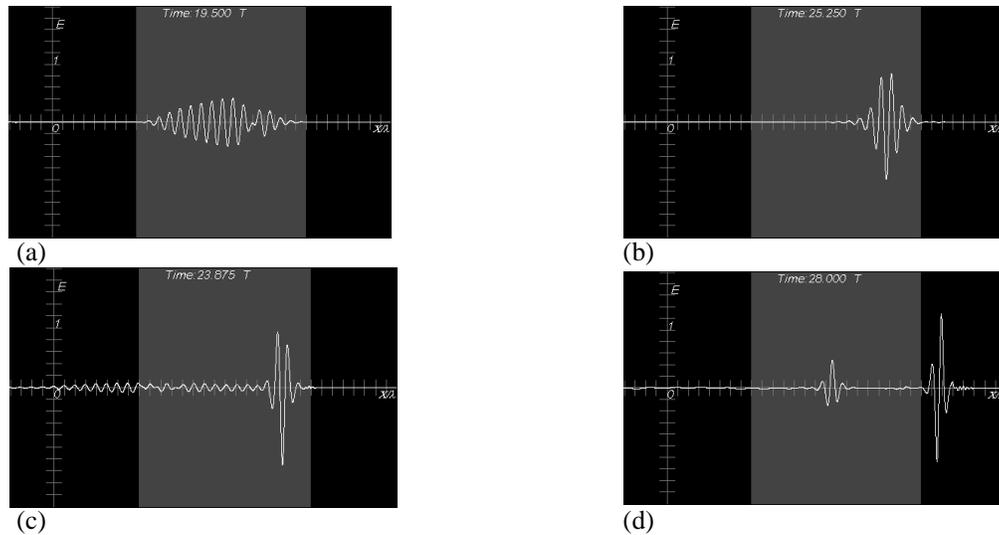


Fig. 1. Evolution of single-cycle (a)  $\pi$  (0.98 MB), (b)  $2\pi$  (1.81 MB), (c)  $2.9\pi$  (1.88 MB), and (d)  $4\pi$  (2.14 MB) pulses in a two-level medium.

In particular, pulses with areas  $\theta(x) = \int_{-\infty}^{\infty} \Omega(x, \tau) d\tau$  less than  $\pi$  ( $\tau = T$ ,  $2\mu/\hbar E_0 \pi \tau = \pi$ ,

and  $\mu N/E_0 = 0.0004$ ) are completely absorbed by a two-level medium (see Fig. 1a). As shown in Fig. 1b, a  $2\pi$  pulse with a duration  $\tau = T$ ,  $2\mu/\hbar E_0 \pi \tau = 2\pi$ , and  $\mu N/E_0 = 0.0008$  propagates through a two-level medium in the soliton regime. Pulses whose areas range from  $\pi$  to  $3\pi$  transform into  $2\pi$  pulses (Fig. 1c,  $\tau = T$ ,  $2\mu/\hbar E_0 \pi \tau = 2.9\pi$ , and  $\mu N/E_0 = 0.0016$ ). Pulses with areas exceeding  $3\pi$  experience splitting into several  $2\pi$  pulses (Fig. 1d,  $\tau = T$ ,  $2\mu/\hbar E_0 \pi \tau = 4\pi$ , and  $\mu N/E_0 = 0.0016$ ). In particular, pulses with areas exactly equal to  $4\pi$ ,  $6\pi$ , ... decay into a sequence of  $2\pi$  pulses having different amplitudes and durations, and, consequently, possessing different group velocities (see Fig. 1d).

Along with the testing of the reliability of our numerical algorithm, these simulations, as can be seen from Figs. 1a–1d, reveal important features of the evolution of very short light pulses and formation of  $2\pi$  solitons in a two-level medium. In particular, detailed information

regarding the changes in the phase and the waveform of the light pulse can be extracted from the results of FDTD simulations. Figure 1c illustrates these opportunities of FDTD simulations by showing how a  $2.9\pi$  pulse with  $\tau = T$ ,  $2\mu/\hbar E_0 \pi \tau = 2.9\pi$ ,  $\mu N/E_0 = 0.00116$  evolves while propagating in a two-level medium. In agreement with general predictions of McCall and Hahn, this pulse is transformed until its area becomes equal to  $2\pi$ . Figure 1c shows the details of this process. In particular, as can be seen from this animation, the peak amplitude of the pulse increases by a factor of 1.31 under these conditions, while its duration decreases down to  $0.55T$ .

Figure 1d illustrates the evolution of a  $4\pi$  pulse with  $\tau = T$ ,  $2\mu/\hbar E_0 \pi \tau = 4\pi$ , and  $\mu N/E_0 = 0.0016$ . This pulse decays into two solitons under these conditions. The amplitude of the shorter of these pulses thus produced is higher than the amplitude of the longer pulse by a factor of 1.52. The duration of the shorter pulse is equal to  $0.36T$ , i.e., this pulse is three times shorter than the initial pulse. Since the group velocity of a  $2\pi$  pulse, in accordance with Eq. (8), is a quadratic function of the pulse amplitude, the shorter pulse, having a higher amplitude, propagates faster than the longer pulse, which has a lower amplitude (Fig. 1d). The evolution of the phase of a  $2\pi$  pulse with a duration  $\tau = T$ ,  $2\mu/\hbar E_0 \pi \tau = 2\pi$ , and  $\mu N/E_0 = 0.0008$ , can be seen from Fig. 1b. The characteristic length of phase evolution estimated for such a pulse with the use of Eq. (10) is  $L = 8\lambda$ . This estimate agrees perfectly well (with an error less than  $10^{-3}$ ) with the results of FDTD simulations for  $2\pi$  pulses whose durations are no less than two optical cycles. For a single-cycle pulse shown in Fig. 1b, the characteristic lengths corresponding to the phase shift (9) equal to  $\pi$  estimated on the basis of FDTD simulations was  $8.2\lambda$ , which slightly differs from the estimate for  $L$  obtained from Eq. (10).

#### 4 Deviations from the McCall–Hahn scenario

Noticeable deviations from the McCall–Hahn regime were observed for pulses with durations shorter than the duration of a single field cycle.

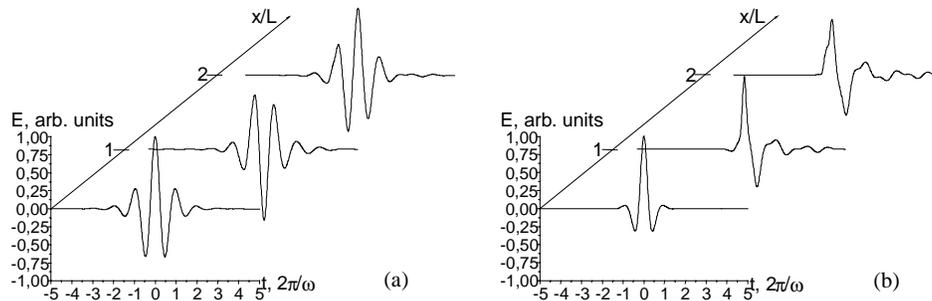


Fig. 2. (a) Evolution of a half-cycle  $2\pi$  pulse in a two-level medium:  $\tau = 0.5 T$ ,  $2\mu/\hbar E_0 \pi \tau = 2\pi$ , and  $\mu N/E_0 = 0.0016$ , (b) Evolution of a quarter-cycle  $2\pi$  pulse in a two-level medium:  $\tau = 0.25 T$ ,  $2\mu/\hbar E_0 \pi \tau = 2\pi$ , and  $\mu N/E_0 = 0.0032$

In particular, half-cycle  $2\pi$  pulses become asymmetric as they propagate through a two-level medium (Fig. 2a), and the characteristic length corresponding to the phase shift (9) equal to  $\pi$  estimated on the basis of FDTD simulations for such pulses was equal to  $9.4\lambda$ , which appreciably differs from the estimate for  $L$  obtained from Eq. (10). We failed to find the soliton regime for a quarter-cycle  $2\pi$  pulse, which displayed noticeable distortions and lengthening in the process of propagation through a two-level medium (Fig. 2b).

Deviations observed in the behavior of very short  $2\pi$  pulses from the McCall–Hahn scenario are due to the fact that, although, formally, such pulses have an area of  $2\pi$ , the cycle of interaction between light and a two-level system remains incomplete in this case, as the pulses do not even contain a full cycle of the field (Fig. 2). As a result, such pulses leave

some excitation in a two-level medium (Fig. 3b) instead of switching excited-state population back to the ground state, as in the case of longer  $2\pi$  pulses (Fig. 3a).

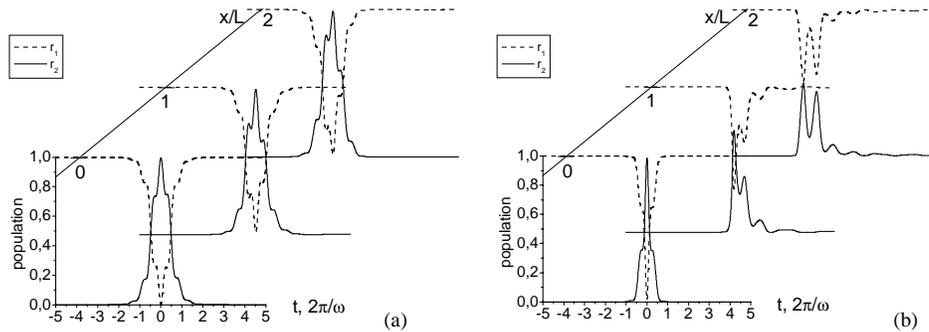


Fig. 3. Evolution of the excited- and ground-state populations in a two-level medium under the action of (a) a half-cycle  $2\pi$  pulse and (b) a quarter-cycle  $2\pi$  pulse: (dashed line) ground-state population  $r_1$  and (solid line) excited-state population  $r_2$ .

The amplitude of the leading edge of the pulse becomes higher than the amplitude of its trailing edge, and the pulse waveform becomes noticeably asymmetric (Fig. 2a). The group velocity of such very short pulses increases due to this incompleteness of the light–two-level-system interaction cycle, leading to a discrepancy between the estimates for the characteristic lengths corresponding to the phase shift (9) equal to  $\pi$  estimated from Eq. (10) and FDTD simulations. The residual population in the medium and the asymmetry of the pulse waveform increase with pulse shortening.

## 5 Conclusion

The results of finite-difference time-domain simulations performed in this paper show that the general predictions of McCall and Hahn for the evolution of the amplitude and the phase of short pulses in a two-level medium generally agree reasonably well with the results of numerical simulations until the pulse duration becomes less than the duration of a single optical cycle. Our numerical analysis has revealed several interesting physical features in the formation of  $2\pi$  solitons produced as a result of splitting of single-cycle pulses propagating in a two-level medium. In particular, the resulting pulses are shown to have different amplitudes, durations, and group velocities, allowing the formation of subfemtosecond pulses and slowing down of the light in two-level media. Noticeable deviations from the McCall–Hahn regime were observed for pulses with durations shorter than the duration of a single field cycle. Half-cycle  $2\pi$  pulses become asymmetric as they propagate through a two-level medium, while quarter-cycle  $2\pi$  pulse display considerable distortions and lengthening in the process of propagation through a two-level medium. We failed to find the soliton regime for such pulses. Deviations observed in the behavior of very short  $2\pi$  pulses from the McCall–Hahn scenario are due to the fact that the cycle of interaction between light and a two-level system remains incomplete in this case, and light pulses leave some excitation in a two-level medium instead of switching excited-state population back to the ground state.

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