Breakdown of the Area Theorem: Carrier-Wave Rabi Flopping of Femtosecond Optical Pulses

S. Hughes*
Department of Physics, Washington State University, Pullman, Washington 99164-2814
(Received 31 March 1998)

By solving Maxwell's curl equations coupled to a two-level atom, a theoretical study of carrier-wave Rabi flopping of femtosecond optical pulses of only several carrier-cycles time duration is reported. For pulse areas of 2π, the usual self-induced transparency regime is essentially recovered. However, for larger pulse areas, carrier-wave Rabi flopping occurs that manifests in local carrier reshaping and subsequently to the production of significantly higher spectral components on the propagating pulse. These new features are not predicted by employing the area theorem or a *slowly-varying-envelope approximation* for the amplitude and phase terms—which is the usual approach. [S0031-9007(98)07419-5]

PACS numbers: 42.50.Md, 42.50.Gy, 42.65.–k

The development of high-intensity ultrashort optical pulses has led to a number of fascinating nonlinear optical propagation studies including Rabi flopping [1], self-induced transparency [2], photon echo [3], and optical shock formation [4], to name but a few. Common to the description of these effects is that their propagation phenomena can be described quite adequately by employing the appropriate coupled matter-Maxwell equations within the slowly-varying-envelope approximation (SVEA), i.e., the envelopes of the electromagnetic field and polarization are assumed to vary little over an optical period and wavelength. However, recently, a number of theoretical recent works have demonstrated the limitations of the SVEA that adopts slowly-varying phase and amplitude components [5–10]. These *non-slowly-varying* Maxwell-Bloch studies are both timely and necessary since on one hand device sizes and absorption lengths can now be reduced to subwavelength scales, and on the other hand several-cycle fs optical pulses are readily available [11]. Single-cycle and several-cycle THz pulses have also been receiving a lot of interest recently [5].

Nonlinear optical pulse propagation studies in the single-cycle regime [12] have recently been investigated by using improved envelope equations (IEE's). The IEE's can be employed within the slowly-evolving-wave approximation [12] where the amplitude and phase terms do not vary much over a wavelength (no emergence of a backwards propagating wave), but, however, the assumption that the amplitude and phase terms vary slowly over an optical period is not made; a perturbative expansion of small population transfer was then applied to describe ultrashort pulse propagation in a Kerr medium. However, for our study, such a technique cannot be applied (i) because of the large population transfer that occurs during Rabi flopping and (ii) because the source of our interesting carrier-wave effects (see below) arises from the fast oscillations in the material equations; the neglect of these oscillations is equivalent to the rotating-wave approximation (RWA). The RWA breakdown in the strong coupling regime is widely discussed in the literature beginning with the pioneering work reported in Ref. [13], though no connection has been made to Rabi flopping.

When a resonant excitation field drives the excitation from the ground state to the excited state and back again, this is termed Rabi flopping (RF) [14–16]. The usual analysis of RF assumes that the optical-frequency components of the energy density do not contribute to the nonlinearity. In this Letter, we return to the problem of RF in a two-level atom (TLA) and present a quantitative Maxwell-Bloch analysis beyond the SVEA to model pulses of only a few optical-cycles time duration. Several new featues arise in the full Maxwell solution that are absent in the standard SVEA models. This work is motivated in part as an intriguing theoretical study and in part because of recent advances in the compression of optical pulses using self-phase modulation [17]. Our predictions employ realistic material and laser parameters and should therefore be experimentally verifiable.

We model pulse propagation of various $2l\pi$ optical pulses (where l is an integer) of 18 fs time duration (FWHM irradiance). For a pulse area (the integral of its Rabi frequency over time) of 2π the standard self-induced transparency (SIT) results are essentially recovered in agreement with the work presented in Ref. [7]. However, the standard results for higher area pulses do not hold because of a strong reshaping of the individual optical carriers. We predict that electric field time-derivative effects will lead to carrier-wave Rabi flopping (CWRF) and subsequently to the formation of higher spectral components on the propagating pulse. The propagation distances required to induce strong carrier reshaping are moderate and are ultimately determined by the TLA-material parameters. Electric field time-derivative effects were previously emphasized in Ref. [7]. The current effort pushes the boundary of understanding and the importance of these effects further.

We employ a standard finite-difference time-domain [FDTD] [7,8,18] approach for solving the full-wave

0031-9007/98/81(16)/3363(4)$15.00

Maxwell equations, and a fourth-order Runge-Kutta method to solve the Bloch equations. A plane-wave pulse normally incident upon a TLA material that is unbounded in the transverse direction is considered. Assuming linear and nonlinear polarization, Maxwell's equations can therefore be written $\dot{B}_y = \partial E_x / \partial z$ and $\dot{D}_x = \partial H_y / \partial z$, with $B_y = \mu_0 H_y$, $D_x = \epsilon_0 E_x + P_x$, and $P_x = \epsilon_0 (n_0^2 - 1) E_y + P_{\mathrm{nl}}$ (n_0 is the background refractive index). The nonlinear polarization $P_{\mathrm{nl}} = 2Nd\,\mathrm{Re}[\rho_{12}]$, where N is the density of TLA's, and ρ_{12} is the off-diagonal density matrix element obtained from the optical Bloch equations $\dot{\rho}_{12} = i\Omega n - (\Gamma_2 + i\omega_{12})\rho_{12}$ and $\dot{n} = i2\Omega(\rho_{12} - \rho_{12}^*) - \Gamma_1(n - 1)$, with $\Omega = dE/\hbar$ the Rabi frequency, $n = (\rho_{11} - \rho_{22})$ the population difference between the lower and upper states, ω_{12} the transition frequency, and d the field-direction dipole moment. The phenomenological population and polarization relaxation rates are given by Γ_1 and Γ_2, respectively. The initial field $E_x(z = 0, t) = \hat{\mathbf{n}}_x E_0 \,\mathrm{sech}(-\frac{t - \tau_{\mathrm{off}}}{\tau_0}) \sin(\omega t)$, where E_0 is the peak input electric field, τ_{off} is the offset position of the pulse center (at $t = 0$), $\tau_p = 2\,\mathrm{arcosh}(1/\sqrt{0.5})\tau_0$ is the FWHM of the pulse irradiance profile, $\hat{\mathbf{n}}_x$ is a unit vector perpendicular to the direction of propagation, and $\omega = 2\pi c/\lambda_0$ is the central pulse frequency.

Within the SVEA, it is well established that when the envelope of the pulse has an area that is an integer number of 2π, then lossless propagation is possible. For a 2π pulse, the rotating dipoles are exactly returned to their initial state while maintaining the shape of the excitation pulse. Moreover, the hyperbolic secant solution propagates without change at a velocity which can be substantially slower than the speed of light. This will occur when $E_0^{2\pi} = 2\hbar/d\tau_0$. Additionally, by virtue of the area theorem [1], $4\pi, 6\pi, \ldots$ pulses are also asymtotic solutions to the coherent propagation problem, although such pulses are not stable and will split up into $2\times, 3\times, \ldots\ 2\pi$ sech pulses, as a consequence of multiple RF. One essential feature of RF is that coherence must be maintained in the system. Hence we choose relaxation times much longer than the input pulse duration and adopt the following material and laser parameters: $\omega = \omega_{12} = 0.6 \times 10^{15}$ rads^{-1}, $\tau_p = 18$ fs, $\Gamma_1^{-1} = \Gamma_2^{-1} = 1$ ns, $n_0 = 1$, $d = 2.65$ eÅ, $N = 2 \times 10^{18}$ cm^{-3}, and t_{off} is chosen appropriately to propagate the pulse in time from outside the computational domain (see below). The results to follow can of course be scaled to various laser and material parameters.

The peak amplitude of the necessary pulse to achieve a 2π envelope area is approximately 0.5 GV/m. Figure 1(a) shows an example of a propagating 2π pulse through the TLA medium. The pulse initially propagates in the free-space region, and thereafter enters the two-level medium at 20 μm; the pulse subsequently propagates the nonlinear medium and finally exits into the free-space region again at 100 μm. The total simulation region is 120 μm. The 2π pulse simulation approximately recovers the well known analytic results in agreement with Ref. [7], i.e., the excitation drives a complete transition of the TLA from its ground state to its excited state and back to its ground state while maintaining its shape. For comparison, Fig. 1(b) shows the corresponding temporal development of the inversion n (solid curve), electric field E_x (chain curve), and the polarizarion component Re$[\rho_{12}]$ (dotted curve), at the fixed position of $z = 21$ μm that is near the input surface of the nonlinear medium. Although the medium is completely inverted and returned to its initial state, oscillation features at the zero points of the pulse arise due to the time-derivative behavior of the input field (see also Ref. [7]). For longer propagation distances, these transient features cause local carrier modification though the envelope of the input pulse is essentially unchanged. Maxwell's curl equations also account for backwards propagating fields which do not occur in the present study since the linear refractive index is unity and also because the absorption length is too large (thus the SVEA in space is valid here).

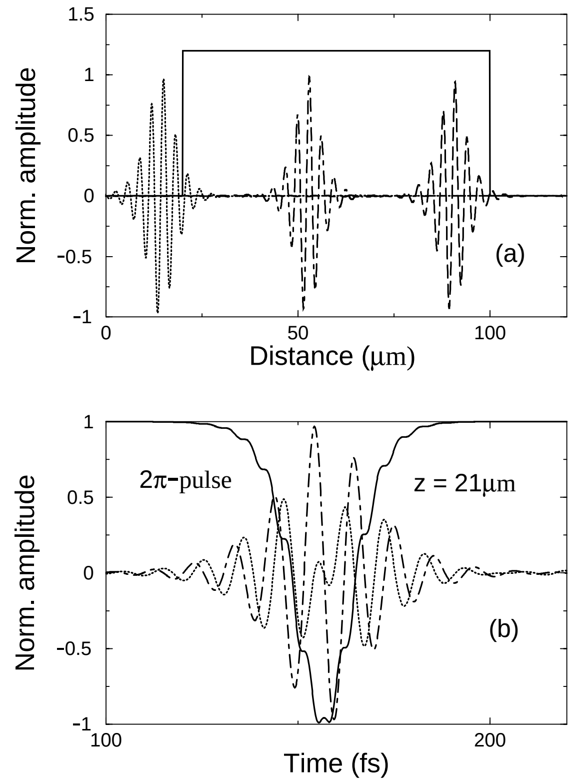

FIG. 1. (a) 2π-pulse propagation through the two-level system. The normalized electric field is shown at the respective times of 140 fs (dotted curve), 275 fs (chain curve), and 415 fs (dashed curve). (b) Normalized field (chain curve), inversion n (solid curve), and Re$[\rho_{12}]$ (dotted curve), near the front face of the two-level material ($z = 21$ μm).

Next we investigate 4π-pulse excitation, and to emphasize the effects of propagation and to avoid long computations we have used, respectively, a nonlinear medium length of 140 μm and $N = 4 \times 10^{18}$ cm^{-3}. Figure 2(a) depicts the electric field profile at the respective propagation times of 180, 350, and 525 fs. The driven density shows the expected two symmetric transversals between the ground and excited states. As a consequence of driving two complete Rabi flops [see Fig. 2(b)], the propagating pulse evolves into two separate pulses with differing spectral profiles. Again this is in agreement with the standard SVEA results. However, extraneous transient features arise once more due to the time-derivative nature of the input field, i.e., because we are resolving the carriers.

Now, because the duration of the input pulse covers only several optical cycles, the perplexing question now arises: What happens when larger input areas are injected into the material? By large, we mean large enough so that the area under the individual carriers may themselves cause RF. The left-hand side of Fig. 3 shows the time-dependent inversion, E_x, at $z = 21$ μm, for pulse areas of 6π, 8π, 10π, 12π, and 14π. The individual carriers now have a profound effect. First, it is noted that incomplete Rabi flops occur instead of the anticipated integer number. For example, for the 10π-pulse case, 4.5 Rabi flops occur instead of 5. Complete RF is very difficult to achieve because of the transient features in the Bloch equations beyond the RWA. Second, local CWRF is clearly discerned. One finds that $\mathrm{Re}[\rho_{12}]$ follows E_x instantaneously so that its peak occurs at the peak in the E_x time derivative. Longer tail effects result from the fact that the medium still has energy residing in the $\mathrm{Re}[\rho_{12}]$, which oscillates at ω.

As a consequence of CWRF, carrier-wave reshaping is expected. To investigate this, the right-hand side of Fig. 3 displays the pulse time profile after the sample exit face ($z = 180$ μm). Strong carrier-wave reshaping is indeed found in addition to a significant interaction with the free-induction decay (FID) of the material. FID interferes with the latter part of the propagating pulse because of incomplete RF and the relatively long polarization decay. For the chosen pulse and material parameters, it is not possible to achieve complete symmetric inversion for areas of and above 6π-pulse excitation, and less than 100% inversion is obtained in each swing of the pulse since the medium is responding rapidly to the variations in the pulse shape and its time derivative. For the 10π pulse, numerically it is found that an 11.4π pulse approximately returns the inversion to the ground state while driving five density flops; however, these flops are far from symmetrical. We have also verified that for the chosen material parameters, no backwards wave emerges and hence the SVEA in space is not violated. The source for the CWRF is due to fast oscillations in the polarization equations outside the RWA.

An experimental signature of such an effect could be seen, for example, on the output spectrum of the propagated pulse. Figures 4(a) and 4(b) show, in comparison to the input spectrum, the output irradiance of 0π and 10π pulses, respectively. Figure 4(a) reproduces the

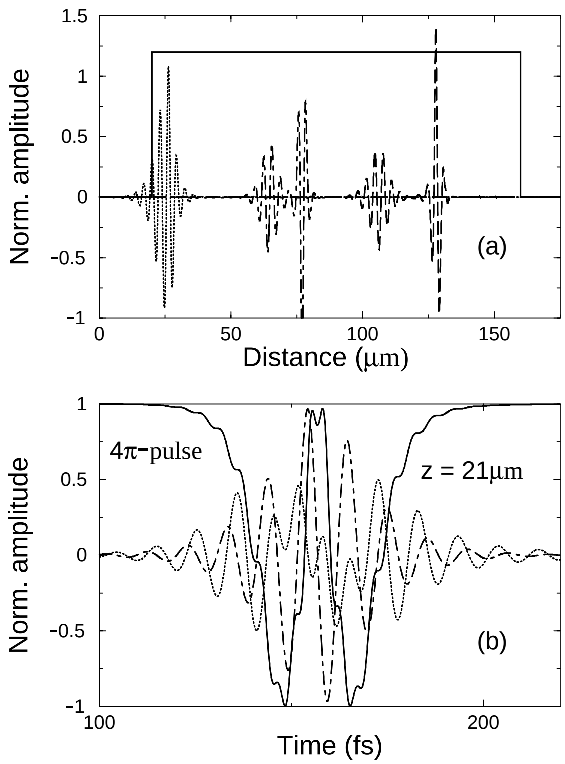

FIG. 2. (a) As in Fig. 1(a) but for 4π-pulse propagation at the respective times of 180, 350, and 525 fs. (b) As in Fig. 1(b) but for 4π-pulse propagation.

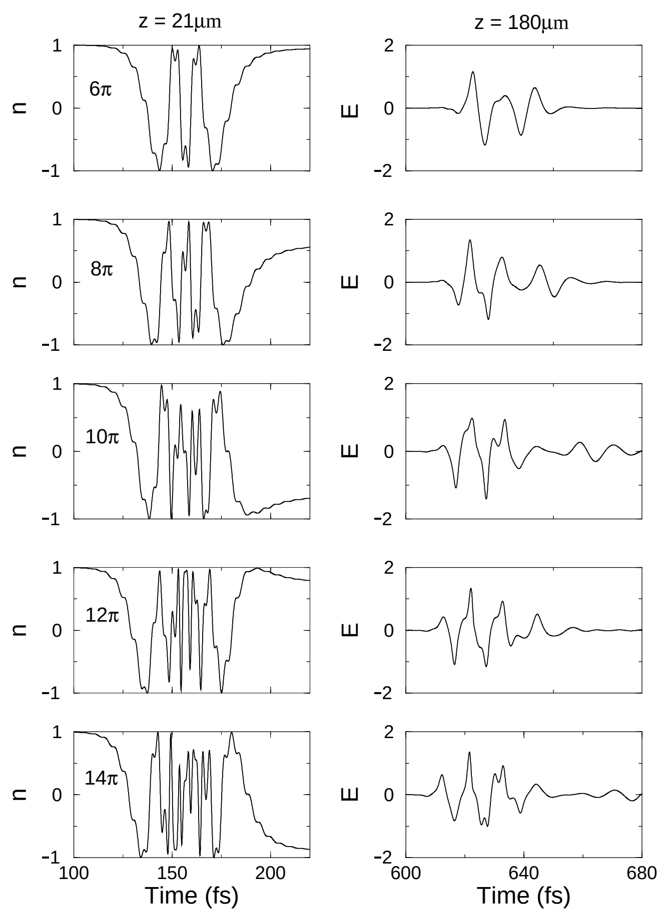

FIG. 3. The left-side graphs show $6\pi - 14\pi$ pulse-induced population difference near the front face of the two-level material ($z = 21$ μm). The right-side graphs display the subsequently propagated pulse (normalized electric field) at $z = 180$ μm.

familiar 0π-pulse scenario in the resonant medium, showing the characteristic spectral hole resulting from a beating structure in the time dependence; the asymmetry is due to dephasing [19]. However, Fig. 4(b) clearly shows the formation of higher (and some lower) spectral components due to CWRF. We have verified computationally that, if the input carrier frequency is increased by a factor of 10, then five symmetric transversals occur between the ground and excited states and there is no evidence for time-derivative features and no generation of these higher spectral features. This, of course, is also true if one increases the time duration of the exciting pulses since sufficient optical carriers will again be present to allow the validity of the SVEA in time. However, for increasing irradiances the area theorem will again break down and, indeed, experimental evidence for similar oscillations may already have been seen using much longer pulses and higher input irradiances [20]. Further, all the results obtained for the 18 fs pulse scale to much longer pulses (and spectrally more narrow two-level systems) assuming the irradiances increase or the carrier frequency changes accordingly. For example, high intensity several-cycle THz pulses are commonly used from free-electron lasers and solid-state sources, and even in the linear regime, propagation of these several-cycle pulses necessitates a Maxwell's curl approach [5]. Moreover, optical pulses with areas of 100π and more are currently being employed to investigate RF [20]; in this regime, even for several-hundred fs optical pulses, the SVEA in time will break down.

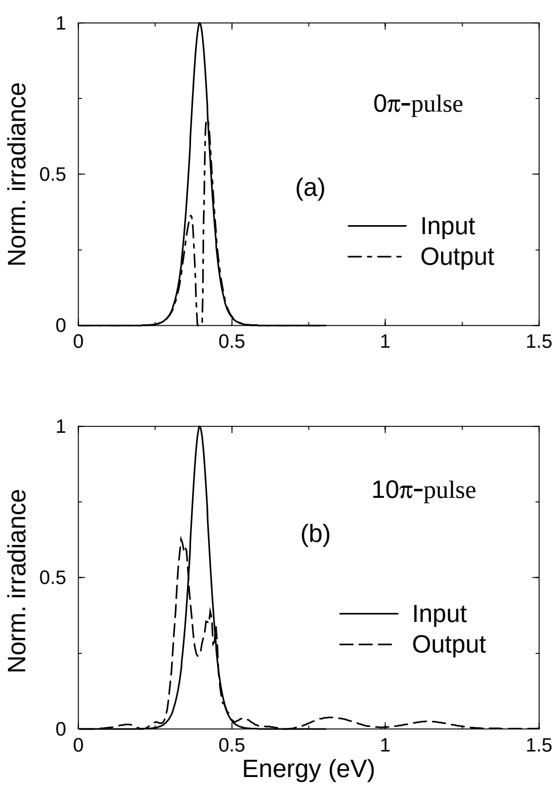

FIG. 4. (a) Input and output pulse irradiance spectra for 0π-pulse propagation. (b) As in (a) but for 10π-pulse propagation.

In conclusion, a first principles approach that utilizes the optical Maxwell-Bloch system coupled to Maxwell's curl equations with a TLA model for the polarization has been utilized to describe carrier-wave RF. The model incorporates all propagation effects such as nonlinearity, dispersion, absorption, stimulated emission, and more. It is shown that the nonlinear behavior is dependent not only on the electric field envelope but also on its propagating time-derivative effects. Standard SIT 2π and 4π pulses are essentially reproduced with minor modifications due to local carrier effects in agreement with Ref. [7]. However, for higher pulse areas, it is found that carrier-wave effects become predominant, and a new, novel type of CWRF is demonstrated—whose experimental signature will appear via the production of significantly higher frequency components on the propagating pulse. These features are absent in the standard area theorem and envelope-type models.

The author thanks D. M. Sullivan and W. Forysiak for useful discussions regarding the FDTD Maxwell computation. Stimulating comments from D. S. Citrin, A. Maslov, A. Schülzgen, H. Giessen, and A. Knorr are gratefully acknowledged. This work was supported in part by the Japanese Society for the Promotion of Science, the National Science Foundation by Grant No. DMR9705403, and the Office of Naval Research.

*Email address: shughes@wsu.edu

[1] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1995).
[2] S. L. McCall and E. L. Hahn, Phys. Rev. Lett. **18**, 908 (1967); G. L. Lamb, Jr., Rev. Mod. Phys. **43**, 99 (1971).
[3] I. D. Abella *et al.*, Phys. Rev. **141**, 391 (1966).
[4] F. Demartinii *et al.*, Phys. Rev. **164**, 312 (1967).
[5] J. Bromage *et al.*, Opt. Lett. **22**, 627 (1997).
[6] A. Schülzgen *et al.*, Phys. Status Solidi B **206**, 125 (1998).
[7] R. W. Ziolkowski *et al.*, Phys. Rev. A **52**, 3082 (1995).
[8] S. A. Basinger and D. J. Brady, J. Opt. Soc. Am. B. **11**, 1504 (1994).
[9] W. Forysiak *et al.*, Phys. Rev. Lett. **76**, 3695 (1996).
[10] R. G. Flesch *et al.*, Phys. Rev. Lett. **76**, 2488 (1996).
[11] See, for example, W. H. Knox *et al.*, Appl. Phys. Lett. **46**, 1120 (1985); A. Baltuska *et al.*, Opt. Lett. **22**, 1344 (1997); see also [17], and references therein.
[12] T. Brabec and F. Krausz, Phys. Rev. Lett. **78**, 3282 (1997).
[13] S. Swain, J. Phys. A **6**, 192 (1973).
[14] I. I. Rabi, Phys. Rev. **51**, 652 (1937).
[15] I. I. Rabi *et al.*, Phys. Rev. **53**, 318 (1938).
[16] S. T. Cundiff *et al.*, Phys. Rev. Lett. **73**, 1178 (1994).
[17] *Ultrashort Laser Pulses*, edited by W. Kaiser, Topics Applied Physics Vol. 60 (Springer, Berlin, Heidelberg, 1993), 2nd ed.
[18] D. M. Sullivan, IEEE Trans. Microwave Theory Tech. **43**, 676 (1995).
[19] Note: for numerical convenience in calculating the Fourier transform of the rapidly-varying time-dependent field, Γ_2 was increased to $1\,\mathrm{ps}^{-1}$ for this simulation.
[20] H. Giessen (private communication).