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Absolute phase control of spectra effects in a two-level medium driven by two-color ultrashort laser pulses

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Abstract

Using a ω–3ω combination scenario, we investigate the absolute phase control of the spectra effects for ultrashort laser pulses propagating in a two-level medium. It is found that the higher spectral components can be controlled by the absolute phases. In particular, different absolute phase combinations can lead to the buildup or split of the even harmonics.

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Keywords: Absolute phase effect; Ultrashort laser pulse; Higher spectral component

1. Introduction

The development of high-intense laser pulses has led to a number of significant nonlinear optical propagation studies for the past decade such as Rabi flopping, self-induced transparency (SIT) etc. [1–3]. Common to the description of these effects is that their propagation phenomena can be beautifully described by using the coupled Maxwell–Bloch equations within the slowly-varying-envelope approximation (SVEA) and the rotating-wave approximation (RWA). However, the limitation of SVEA and RWA for the case of few-cycle pulse has been revealed by Ziolkowski et al. [4,5]. They have found that the nonlinear behavior is dependent both on the electric field envelope and on its propagating time-derivative effects. For the nonperturbative resonant excitation of an intense few-cycle pulse in a semiconductor, Mücke and coworkers have reported the different Rabi sideband interference around twice the laser center frequency when the Rabi frequency becomes comparable to the light frequency [6]. In addition, a new, novel type of carrier-wave Rabi flopping, which is caused by the electric field time-derivative effects, has been theoretically studied by Hughes [7] and experimentally demonstrated by Mücke et al. [8,9].

In the past few years, encouraged by the developments in the engineering of intense ultrashort laser pulses with precisely reproducible electric and magnetic field [10–12] or of single-cycle pulse [13], the use of the relative phase difference between two laser pulses to coherently control excitation in atoms or molecules has received considerable attention [14–19]. The dependence of the harmonic generation on the relative phase in two-color excitation has been investigated numerically [20–22]. The occurrence of even harmonics, which is due to the breakdown of the symmetry of the multiphoton pathway, are reported in the system driven by a fundamental laser field with its much weaker even harmonic field [20,22]. In addition, a generalized RWA has been developed to theoretically describe the relative phase control of the multiphoton resonant excitations in a two-level system with two-color laser pulses [17,18,23]. Recently, a

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relative phase-related coherent control technique for four-wave mixing nonlinear interactions with two-color ω–3ω femtosecond laser pulses has been reported by Serrat [14,15]. The competition or interference between different quantum paths in the multiphoton transitions proves to be very fruitful for understanding the relative phase effects [14,15,18,23,24]. Moreover, Bouchene has presented a theoretical analysis of the pulse-to-pulse phase control of dispersion effects for an ultrashort pulse train propagating in a resonant dense medium [25].

In the coherent control of the extreme nonlinear optics, the carrier-envelope phase Φ_{CE} (defined as the phase of the optical carrier with respect to the pulse peak [26]), i.e. the absolute phase, is a very important factor to be considered. The absolute phase effects have attracted a great deal of interest in extremely nonlinear optical processes, such as high-order harmonic generation [27], photoionization [26,28], photo emission [29], nonperturbative resonant nonlinear process [6] and carrier-envelope phase-controlled quantum interference [30] in a semiconductor, and carrier-envelope phase-sensitive inversion [31,32], etc. Brown et al. have investigated the absolute phase effects of the much weaker and longer probe pulse in the pump-probe excitation of dipolar molecules [24,33], where the pump pulse is tuned close to the transition frequency and requires an ultrashort duration and/or rise and fall times while the probe carrier frequency is extremely small. They have found that the final state populations are dependent on the probe field's absolute phase but are independent of that of the pump field. The probe laser absolute phase effect arises through the coherent excitation of multiple coherent pathways from the initial to the final state containing one pump photon and a varying number of probe photons. They argue that the absolute phase effects are negligible for nondipolar systems [24,33]. Gallagher and coworkers have examined the phase dependent multiphoton excitation of potassium-atom Rydberg states using much weaker radio-frequency fields which have appreciable rise and fall times [34]. Very recently, we have demonstrated that the absolute phase of few-cycle laser pulse dramatically changes due to the near dipole–dipole interactions during propagating in a dense two-level medium [35].

In this Letter, for the first time to our knowledge, the effects of absolute phases on the higher spectral components in the multiphoton excitation of a two-level system with two-color, intense and ultrashort laser pulses are investigated. By solving the full Maxwell–Bloch equations beyond the slowly-varying-envelope approximation and the rotating-wave approximation with an iterative predictor–corrector finite-difference time domain method (FDTD) [4,36], we find that the absolute phases have a crucially influence on the higher spectral components if the durations of the pulses become comparable to the optical period. The yields of higher spectral components sensitively depend on the absolute phases. Surprising, in the case of zero relative phase, for some special absolute phases, the even harmonics are significantly built up. While for some other absolute phases, the even harmonics split into two parts. Thus strong even harmonic pulses can be obtained by optimizing the absolute phases. These phenomena can be understood as a result of the local carrier-wave effect, sideband interference and multiphoton ac Stark effect [37,38].

This Letter is organized as follows: in Section 2, a theoretical model for a two-level system interacting with two-color ultrashort laser pulses is presented. The absolute phase control of the higher spectral components is studied and discussed in detail in Section 3. Finally, a summary of our results for this coherent control is presented in Section 4.

2. Theoretical model

We consider a system shown in Fig. 1, the two-color excitation of a two-level medium can be modelled by means of the full Maxwell–Bloch equations beyond the standard approximations SVEA and RWA [1,4]. The equations are written as:

$$\partial_t H_y = -\frac{1}{\mu_0}\partial_z E_x, \tag{1a}$$

$$\partial_t E_x = -\frac{1}{\varepsilon_0}\partial_z H_y - \frac{1}{\varepsilon_0}\partial_t P_x, \tag{1b}$$

$$\partial_t u = -\omega_0 \upsilon - \gamma_1 u, \tag{1c}$$

$$\partial_t \upsilon = \omega_0 u + 2\Omega w - \gamma_1 \upsilon, \tag{1d}$$

$$\partial_t w = -2\Omega \upsilon - \gamma_2 (w - w_0), \tag{1e}$$

where E_x and H_y are the electric and magnetic fields propagating along the z direction, respectively. $\Omega = dE_x/\hbar$ is the Rabi frequency, d is the electric dipole coupling coefficient, μ_0 and ε_0 are the magnetic permeability and electric permittivity of the free space, γ_1 and γ_2 are the polarization and population relaxation constant, ω_0 is the transition frequency of the two-level medium, N is the density of the polarizable atoms. The macroscopic polarization $P_x = N d u$ connects with the off-diagonal density matrix element $\rho_{ab} = (u + i\upsilon)/2$ and the population difference $w = \rho_{bb} - \rho_{aa}$ between the upper and lower states, which are determined by the Bloch equations, w_0 represents the initial value of the population difference.

We employ an iterative predictor–corrector finite-difference time-domain method to solve the full Maxwell–Bloch equations [4,36]. Mur absorbing boundary conditions [36] are incorporated with FDTD discretization at the left and right boundaries to avoid the influence of the finite space computational

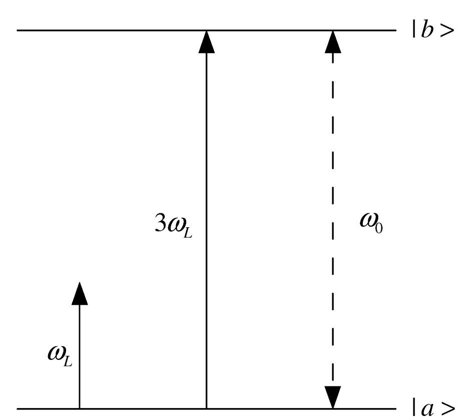

Fig. 1. Schematic representation of a two-level system interacting with two-color ω–3ω laser pulses.

domain. The time and space steps Δt and Δz are chosen to ensure the Courant condition: $c\Delta t \leqslant \Delta z$. For all of the simulations discussed below, we begin the external field propagating into the FDTD mesh at the location $z_0 = 15$ μm in the left air zone with the initial time history

$$E_x(z_0, t) = E_0 \operatorname{sech}\big[1.76(t - t_0)/\tau_p\big]\cos\big[\omega_L(t - t_0) + \Phi_1\big] + E_0 \operatorname{sech}\big[1.76(t - t_0)/\tau_p\big]\cos\big[3\omega_L(t - t_0) + \Phi_2\big], \quad (2)$$

where ω_L is the fundamental pulse frequency. The fundamental and third harmonic components have the same maximum amplitude E_0, the same duration τ_p, i.e. the full width at half maximum (FWHM) of the intensity, and the same delay t_0. The choice of t_0 ensures that the field at $t = 0$ is negligible. The phases Φ_1 and Φ_2 are the absolute phases of the fundamental and third harmonic pulses, respectively. Clearly, the incident field is equivalent to the case in Ref. [4] but has a different exciting position. We adopt the following field and material parameters [16]: $\omega_L = 0.6$ fs^{-1}, $\omega_0 = 1.8$ fs^{-1}, $d = 2.65$ eÅ, $N = 2 \times 10^{24}$ m^{-3}, $\gamma_1^{-1} = \gamma_2^{-1} = 1$ ns. The system is initialized with $u = 0, \upsilon = 0$ and $w_0 = -1$ at $t = 0$. The Rabi frequency $\Omega_0 = 1$ fs^{-1} corresponds to the electric field of $E_0 = 2.5 \times 10^9$ V/m or an intensity of $I = 1.7 \times 10^{12}$ W/cm^2. For such laser intensity, recent experiments on semiconductors have demonstrated that a description in terms of two-level systems has been able to reproduce the experimental results amazingly well [6,39,40]. Moreover, a few significant works that have studied this model can also be found in Refs. [4,7,41]. Very recently, Song et al. have investigated the influence of ionization on the propagation and spectral effects of a few-cycle laser pulse in an open two-level system [42]. They found that the carrier-wave Rabi flopping is still clearly discerned and the production of higher spectral components is almost no different if the fractional ionization is weak during the interaction. So, when the laser intensity is of the order of 10^{12} W/cm^2, the simple two-level system can provide an adequate description despite a little ionization [6]. The pulses initially propagate in the air region, and thereafter inject into the medium at $z = 20$ μm. They subsequently propagate through the nonlinear medium and finally exit into the right air zone at $z = 70$ μm. The total simulation region is taken to be 75 μm.

3. Numerical results and discussion

In this section, we present the numerical results in detail by exactly solving Eqs. (1) and all the higher spectral components normalized by the peak of the spectrum of the Rabi frequency corresponding to the incident field are obtained at $z = 71$ μm. Here, we investigate the $(N_{\omega_L}, N_{3\omega_L})$-photon combined excitation corresponding to the harmonic frequency $\omega_h = N_{\omega_L}\omega_L + N_{3\omega_L}3\omega_L$, where either N_{ω_L} or $N_{3\omega_L}$ can be positive or negative integers, or one of them zero. Usually, only small N ($N = |N_{\omega_L}| + |N_{3\omega_L}|$) combinations are dominant because the harmonic outcome of a given frequency decrease very fast when N increases [18,33]. One example, there are only two primary combinations $(2, 1)$ and $(-1, 2)$ contributing to the fifth harmonic $\omega_h = 5\omega_L$. As the cases in Refs. [18,19,23,24], during the ω–3ω pulsed laser excitation, the magnitudes of the higher spectral components depend on the relative phase $\Phi = \Phi_2 - 3\Phi_1$ defined in a principal value range from $-\pi$ to π.

We investigate the absolute phase effects on the higher spectral components in the two-level medium interacting with two-color ultrashort pulses of various durations and areas. When the pulses have more than five optical cycles in their envelopes, even for relatively large pulse areas, the conversions to the higher spectral components are determined by the relative phase Φ due to the quantum interference or competition during the multiphoton processes [14,15,18,19,23,24] rather than by the absolute phases Φ_1 and Φ_2. As an example, for five-cycle pulses, i.e. $\tau_p = 53$ fs, even they have pulse areas of 8π, the magnitudes of the higher spectral components are almost independent of the absolute phases. In the case of $\Phi = 0$, the same higher spectral components are produced for the absolute phase (Φ_1, Φ_2) combinations: $(0, 0)$ and $(\pi/2, -\pi/2)$. Similarly, the higher spectral components of $(0, \pi)$ and $(\pi/2, \pi/2)$ are identical since they have the same relative phase $\Phi = \pi$. In addition, the magnitudes of the higher spectral components are larger in the case in phase ($\Phi = 0$). Clearly, the interference between the two quantum path is constructive when the pulses are in phase, while it is destructive in the case out of phase ($\Phi = \pi$) [14].

Next, we fix the pulse area at 8π and focus on the absolute phase effects with the decrease of the pulse durations. For three-cycle pulses, a subtle difference appears in the higher spectral components for different absolute phases. For shorter pulses, the individual optical carriers may themselves cause Rabi flopping, i.e. so-called carrier wave Rabi flopping (CWRF) [7]. As shown in Fig. 2, for two-cycle pulses, the change of the absolute phases obviously modifies the higher spectral components. When $\Phi = 0$, the even harmonic components of $(\pi/2, -\pi/2)$ are slightly weaker than those of $(0, 0)$ due to the sideband interference. While when $\Phi = \pi$, in the case of $(0, \pi)$, the fifth harmonic is stronger and the seventh harmonic is weaker relative to the case of $(\pi/2, \pi/2)$. As the durations of two pulses decrease, the absolute phase effects become more and more predominant. For only single optical cycle ($\tau_p = 11$ fs) and 8π pulse area corresponding to the Rabi frequency $\Omega_0 = 1.3$ fs^{-1} or an intensity of $I = 2.7 \times 10^{12}$ W/cm^2, the time-derivative behavior of the local electric field leads to a very strong carrier-wave reshaping and the individual CWRF has a very profound effect [7]. The incomplete Rabi flops occur and local CWRF is considerable. Moreover, the absolute phase-sensitive sideband interference [6] and ac Stark effect [37,38,43], which is closely relates to the Rabi frequency, also contribute to the higher spectral components. As a consequence, the higher spectral components crucially depend upon the absolute phases. As clearly demonstrated by Fig. 3(a), in the case of $(\pi/2, -\pi/2)$, the even harmonics at $6\omega_L$, $8\omega_L$ and $10\omega_L$ are enhanced by a factor of 2.7–5.0 relative to the case $(0, 0)$. Furthermore, the harmonic at $12\omega_L$ is relative strong for $(\pi/2, -\pi/2)$ but vanishes for $(0, 0)$. So a strong coupling to the even harmonics can be achieved by optimizing the absolute phases. For absolute phase combination $(0, 0)$, dips appear at the center of the even harmonics and cause them to split into two parts. The ac Stark

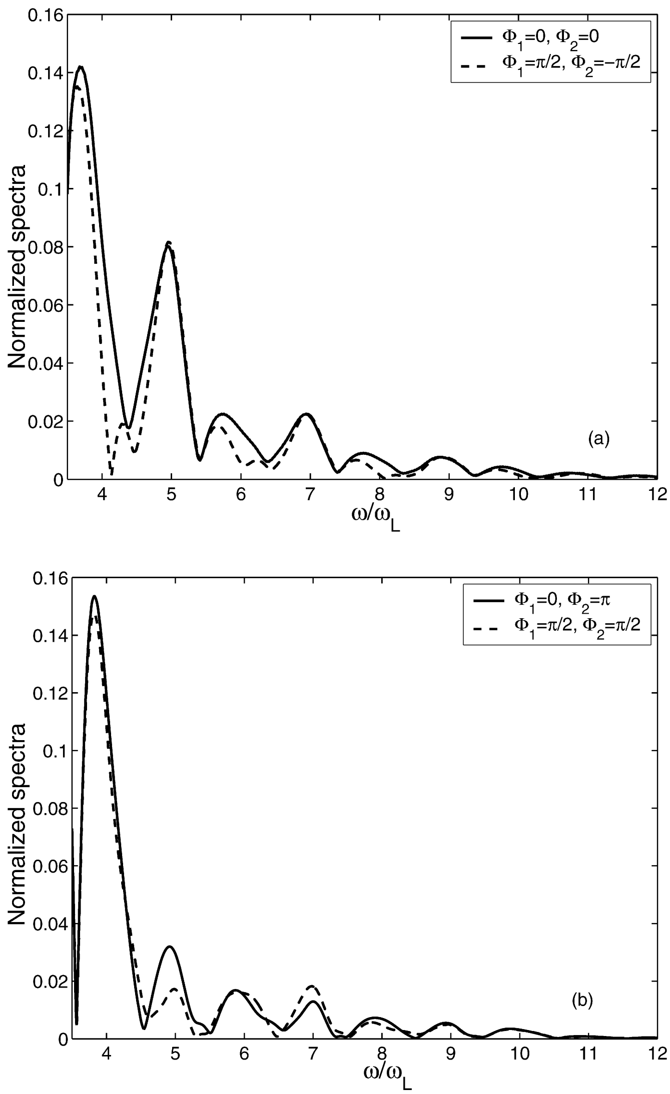

Fig. 2. Normalized higher spectral components for different relative phases and absolute phases at $z = 71$ μm for $\tau_p = 21$ fs. (a) $\Phi = 0$, (b) $\Phi = \pi$.

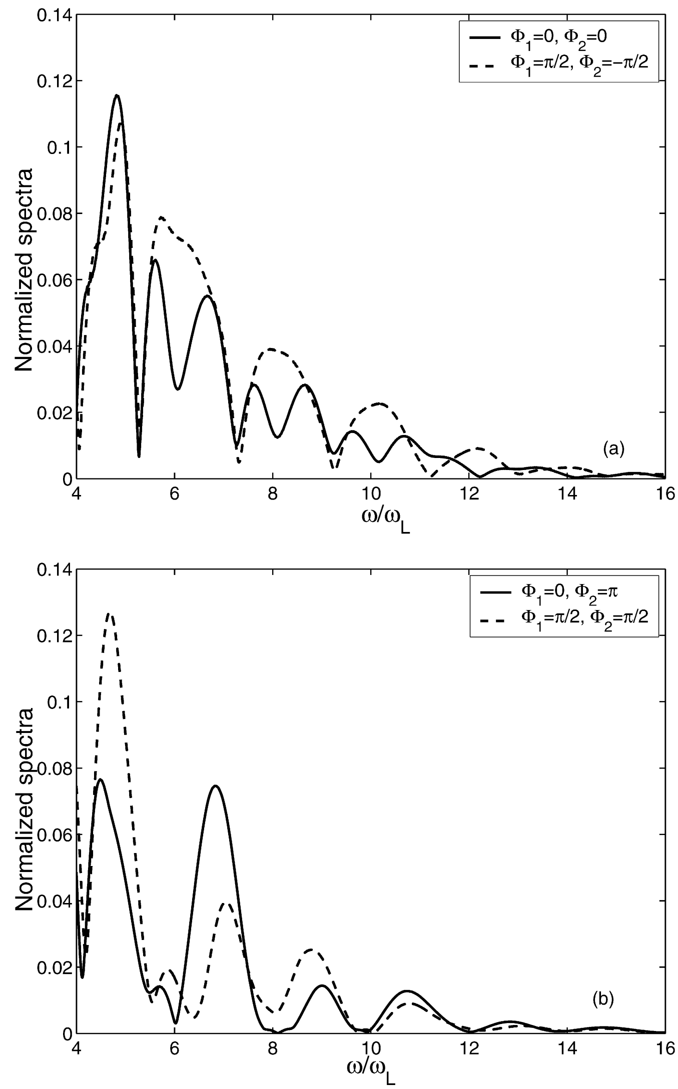

Fig. 3. As in Fig. 2 but for $\Omega_0 = 1.3\ \text{fs}^{-1}$ and $\tau_p = 11$ fs. (a) $\Phi = 0$, (b) $\Phi = \pi$.

effect accounts for the emergence of the even harmonic spectral components. The enhancement and the dips at the centers of the even harmonics are attributed to the sideband interference. Whereas when $\Phi = \pi$, the even harmonics are almost suppressed and only the odd harmonics are produced efficiently. However, for different absolute phase combinations, the yields of the odd harmonics not only are different but also exhibit a regular change. Here we draw a comparison between the combinations $(0, \pi)$ and $(\pi/2, \pi/2)$. One can find that the 5th and 9th harmonics in the case of $(\pi/2, \pi/2)$ are much stronger than those of $(0, \pi)$ while the 7th and 11th harmonics in the former are weaker (see Fig. 3(b)). These spectral features suggest that one can realize pre-selected harmonics by proper choosing absolute phase combination. For $(\pi/2, \pi/2)$ comparing with the case of $(0, \pi)$, except for the substantially changed magnitudes of the higher spectral components, the centers of gravity of the harmonics also shift: obvious blueshifts are found at the 5th and 7th harmonics and a tiny redshift at the 9th one appears. These phenomena mentioned above can be qualitatively interpreted as the combination of CWRF, the sideband interference and the ac Stark effect. For other fixed relative phase Φ but different absolute phase combinations, the absolute phase-sensitive higher spectral components are also investigated. On the other hand, the absolute phase effects are limited by the pulse area. For small pulse area, the relative phase-dependent higher spectral components are reproduced in agreement with Refs. [14–16] because the carrier-wave effect and the sideband interference are negligible.

4. Conclusion

In summary, we have studied the absolute phase effects on the higher spectral components in the two-color and ultrafast excitation of the two-level medium by exactly solving the full Maxwell–Bloch equations. For few-cycle, relatively large area pulses, it is demonstrated that the absolute phases have an essential influence on the higher spectral components: the outcomes of harmonics can be controlled by the absolute phases. Moreover, the centers of gravity of the harmonics shift with the absolute phases. More important, for $\Phi = 0$, a significantly enhanced even harmonic yield can be achieved through the control of the absolute phase combination. In contrast, the harmonics split into two parts for some other absolute phase combinations. Based on these absolute phase-dependent phenomena, a coherent control scheme can be contemplated to optimize the ultrafast nonlinear optical spectroscopy techniques.

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References

[1] L. Allen, J.H. Eberly, Optical Resonance and Two-Level Atoms, Wiley, New York, 1975.
[2] S.L. McCall, E.L. Hahn, Phys. Rev. Lett. 18 (1967) 908.
[3] S.L. McCall, E.L. Hahn, Phys. Rev. 183 (1969) 457.
[4] R.W. Ziolkowski, J.M. Arnold, D.M. Gogny, Phys. Rev. A 52 (1995) 3082.
[5] T. Brabec, F. Krausz, Rev. Mod. Phys. 72 (2000) 545.
[6] O.D. Mücke, T. Tritschler, M. Wegener, U. Morgner, F.X. Kärtner, Phys. Rev. Lett. 89 (2002) 127401.
[7] S. Hughes, Phys. Rev. Lett. 81 (1998) 3363.
[8] O.D. Mücke, T. Tritschler, M. Wegener, U. Morgner, F.X. Kärtner, Phys. Rev. Lett. 87 (2001) 057401.
[9] O.D. Mücke, T. Tritschler, M. Wegener, U. Morgner, F.X. Kärtner, G. Khitrova, H.M. Gibbs, Opt. Lett. 29 (2004) 2160.
[10] A. Baltuška, T. Fuji, T. Kobayashi, Phys. Rev. Lett. 88 (2002) 133901.
[11] D.J. Jones, S.A. Diddams, J.K. Ranka, A. Stentz, R.S. Windeler, J.L. Hall, S.T. Cundiff, Science 288 (2000) 635.
[12] A.V. Sokolov, D.R. Walker, D.D. Yavuz, G.Y. Yin, S.E. Harris, Phys. Rev. Lett. 87 (2001) 033402.
[13] M.Y. Shverdin, D.R. Walker, D.D. Yavuz, G.Y. Yin, S.E. Harris, Phys. Rev. Lett. 94 (2005) 033904.
[14] C. Serrat, Phys. Rev. A 72 (2005) 023808.
[15] C. Serrat, Phys. Rev. A 72 (2005) 051803.
[16] X.H. Song, S.Q. Gong, S.Q. Jin, Z.Z. Xu, Phys. Lett. A 319 (2003) 150.
[17] A.E. Kondo, V.M. Blokker, W.J. Meath, J. Chem. Phys. 96 (1992) 2544.
[18] A. Brown, W.J. Meath, P. Tran, Phys. Rev. A 65 (2002) 063401.
[19] A. Brown, W.J. Meath, Chem. Phys. 198 (1995) 91.
[20] C.P. Liu, S.Q. Gong, R.X. Li, Z.Z. Xu, Phys. Rev. A 69 (2004) 023406.
[21] M. Protopapas, A. Sanpera, P.L. Knight, K. Burnett, Phys. Rev. A 52 (1995) R2527.
[22] T. Zou, A.D. Bandrauk, M. Ivanov, P.B. Corkum, Phys. Rev. A 51 (1995) 3991.
[23] C. Chen, Y. Yin, D.S. Elliott, Phys. Rev. Lett. 64 (1990) 507.
[24] A. Brown, Phys. Rev. A 66 (2002) 053404.
[25] M.A. Bouchene, Phys. Rev. A 66 (2002) 065801.
[26] G.G. Paulus, F. Grasbon, H. Walther, P. Vllloresl, M. Nlsoll, S. Staglra, E. Prlorl, S. De Sllvestrl, Nature 414 (2001) 182.
[27] A. de Bohan, P. Antoine, D.B. Milošević, B. Piraux, Phys. Rev. Lett. 81 (1998) 1837.
[28] X. Liu, C. Figueira de Morisson Faria, Phys. Rev. Lett. 92 (2004) 133006.
[29] A. Apolonski, P. Dombi, G.G. Paulus, M. Kakehata, R. Holzwarth, T. Udem, C. Lemell, K. Torizuka, J. Burgdörfer, T.W. Hänsch, F. Krausz, Phys. Rev. Lett. 92 (2004) 073902.
[30] T.M. Fortier, P.A. Roos, D.J. Jones, S.T. Cundiff, R.D.R. Bhat, J.E. Sipe, Phys. Rev. Lett. 92 (2004) 147403.
[31] A. Brown, W.J. Meath, J. Chem. Phys. 109 (1998) 9351.
[32] C. Jirauschek, L. Duan, O.D. Mücke, F.X. Kärtner, M. Wegener, U. Morgner, J. Opt. Soc. Am. B 22 (2005) 2065.
[33] A. Brown, W.J. Meath, A.E. Kondo, Phys. Rev. A 65 (2002) 060702(R).
[34] W.M. Griffith, M.W. Noel, T.F. Gallagher, Phys. Rev. A 57 (1998) 3698.
[35] K.Y. Xia, S.Q. Gong, C.P. Liu, X.H. Song, Y.P. Niu, Opt. Express 13 (2005) 5913.
[36] A. Taflove, Computational Electrodynamics: The Finite-Difference Time-Domain Method, Artech House, Boston, 1995.
[37] T.G. Rudolph, H.S. Freedhoff, Z. Ficek, Phys. Rev. A 58 (1998) 1296.
[38] G.N. Gibson, Phys. Rev. A 67 (2003) 043401.
[39] T. Tritschler, O.D. Mücke, M. Wegener, Phys. Rev. A 68 (2003) 033404.
[40] T. Tritschler, O.D. Mücke, M. Wegener, U. Morgner, F.X. Kärtner, Phys. Rev. Lett. 90 (2003) 217404.
[41] V.P. Kalosha, J. Herrmann, Phys. Rev. Lett. 83 (1999) 544.
[42] X.H. Song, S.Q. Gong, R.X. Li, Z.Z. Xu, Phys. Rev. A 72 (2005) 043820.
[43] M.O. Scully, M.S. Zubairy, Quantum Optics, Cambridge Univ. Press, Cambridge, 1997.