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The local-field corrective effect on Rabi oscillation of ultrashort pulse excitation in semiconductor GaAs

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The Rabi oscillation of the thin bulk semiconductor GaAs, which takes into account the effect of the local-field correction induced by the interacting excitons, is investigated by numerically solving the semiconductor Bloch equations. It is found, for a 2π few-cycle pulse excitation, that two incomplete Rabi-floppings emerge due to the competition between the Rabi frequency of the incident pulse and the internal-field matrices. Furthermore, for a sub-cycle 2π pulse excitation a complete Rabi-flopping can occur because of the absolute phase effect. We ascribe these characteristics of the Rabi oscillation to the renormalized Rabi frequency.

Keywords: Rabi oscillation; ultrashort pulse; absolute phase; exciton; local field

1. Introduction

In a two-level atomic system, Rabi oscillation generally refers to optical oscillations of level populations between the ground state and the excited state under the action of the driving electromagnetic field. The number of the Rabi oscillation depends on the pulse area Θ of the light pulse, which is defined by $\Theta = (d_{12}/\hbar) \int_{-\infty}^{+\infty} \tilde{E}(t) dt$, with $\tilde{E}(t)$ the electric-field envelope of the pulse and d_{12} the dipole transition moment. In a dilute medium, it is valid to assume that only atoms interact with the pulse separately from each other, and one complete Rabi-flopping is expected if the many-cycle pulse area is 2π . However, in a dense medium, because the atomic density is so high that there are many atoms within a cubic resonance wavelength, the near-dipole–dipole interaction (NDDI), which leads to Lorentz local-field correction (LFC), must be considered [1]. Some of us found, in a dense medium, that the solitary pulse area is evidently larger than 2π , but the area of the effective Rabi frequency, which equals the Rabi frequency of the incident pulse plus the product of the strength of the NDDI and the polarization, is consistent with the standard area theorem and keeps 2π [2].

In a semiconductor, Coulomb interaction between electrons and holes introduces a hydrogenic series of exciton bound states converging to the electron–hole

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continuum (Figure 1). Due to the excitonic effect, Rabi oscillation of the semiconductor shows a fundamental difference from that of the atomic systems, e.g. doubling the effective Rabi frequency of the applied field [3,4], low area fields density oscillation [5,6], etc. The exciton–exciton interaction in the semiconductor viewed as the NDDI can be considered as a LFC [7,8]. A system of interacting electrons and holes in the presence of an external field is somewhat analogous to a paramagnet or a dense dielectric medium. Arnaud and Alouani investigated local-field and excitonic effects in the calculated optical properties of semiconductors from first-principles [9]. Mutluay and Tanatar calculated the local-field factors and static correlation functions for electron and electron–hole double-wire systems [10]. They found that the interwire correlations become quite important for an electron–hole system. Slepian et al. investigated theoretically the influence of electron–hole dipole–dipole interaction on the excitonic Rabi oscillation in an isolated quantum dot (QD) [11]. They reported that for the Gaussian pulse, the final state of inversion as a function of the pulse peak strength demonstrates step-like transitions. Madureira et al. investigated the optical absorption spectra of semiconductor quantum wires and its dependence on the optical pumping power [12]. They demonstrated that the competing effects of the dynamical band-gap renormalization and the LFC leads to an almost cancellation of the red/blue shift energy. Recently, Paspalakis et al. studied theoretically the influence of local fields in the excitonic population dynamics of a quantum dot system under the action of either continuous-wave or pulsed and appropriately chirped electromagnetic fields [13]. They presented the results with emphasis given to the effects of local fields in excitonic Rabi oscillations and to the conditions that could give rise to population inversion in the system.

In a few-cycle pulse regime, the absolute phase, i.e. the carrier-envelope phase ϕ (defined as the phase of the optical carrier with respect to the pulse peak [14]), 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

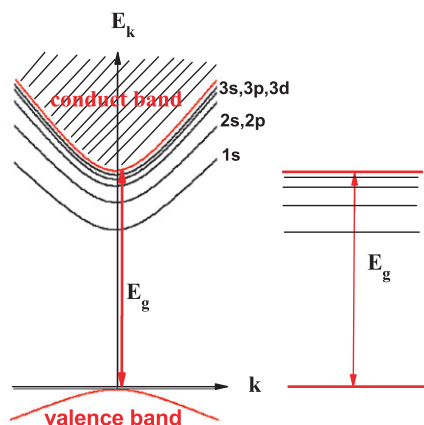


Figure 1. Scheme of the excitonic energy levels of the semiconductor. (The colour version of this figure is included in the online version of the journal.)

generation [15], photoionization [14,16], photoelectron emission [17], absolute phase controlled quantum interference in a semiconductor [18], and absolute phase sensitive inversion [19], etc. Mücke et al. investigated the role of absolute phase in nonperturbative resonant nonlinear optics [20]. They found that if the Rabi frequency becomes comparable to the light frequency, the different Rabi sidebands interfere around twice the laser center frequency, giving rise to a signal which strongly depends on the absolute phase. Gallagher and coworkers examined the phase dependent multi-photon excitation of potassium-atom Rydberg states using much weaker radio-frequency fields which have appreciable rise and fall times [21]. Recently, some of us demonstrated that the higher spectral components can be controlled by the absolute phases for ultrashort laser pulses propagating in a two-level medium [22].

Although extensive works have investigated the LFC effect in semiconductors [3–13], to the best of our knowledge, almost all of them are based on the continuous-wave or long pulse regime. In this paper, we investigate the Rabi oscillation of the thin bulk semiconductor GaAs under ultrashort pulse excitation by taking into account the LFC effect. Considering electron transition between the valence band and the conduction band, the typical parameters for GaAs at the temperature 300 K are [23–25]: the relative dielectric constant $\epsilon_r = 12.9$, the effective mass of the electron $m_e = 0.063m_0$ and hole $m_h = 0.51m_0$ (m_0 is free electron mass), the exciton Bohr radius $a_0 = 12.17$ nm, and binding energy $E_0 = 4.6$ meV, respectively. For a 2π few-cycle pulse, there are two incomplete Rabi-floppings. Furthermore, for a 2π sub-cycle pulse excitation, a complete Rabi-flopping can occur due to the absolute phase effect.

This paper is organized as follows. In Section 2, we present the model of the semiconductor Bloch equations (SBE) and the computational approach. In Section 3, the local-field corrective effect on Rabi oscillation of ultrashort pulse excitation in semiconductor GaAs is studied and discussed in detail. A short summary is presented in Section 4.

2. Theory model

In a two-band approximation including the direct Coulomb interactions under the Hartree–Fock approximation for laser-driven semiconductors, the SBE [26,27] are written as:

$$i\hbar \frac{\partial}{\partial t} P_k = \left(\epsilon_k - 2 \sum_{k'} V_{k-k'} f_{k'} \right) P_k - \left(\Omega(t) + \sum_{k'} V_{k-k'} P_{k'} \right) (1 - 2f_k) - i\gamma P_k, \quad (1a)$$

$$\hbar \frac{\partial}{\partial t} f_k = 2 \operatorname{Im} \left(\left(\Omega(t) + \sum_{k'} V_{k-k'} P_{k'} \right) P_k^* \right); \quad (1b)$$

here f_k denotes the carrier (electron or hole) density matrices, which is valid on a time scale where the carrier–carrier scattering process can be ignored, and P_k denotes the corresponding interband density matrices. $\epsilon_k = E_g + \hbar^2 k^2 / 2m_r$ is the energy difference between the unperturbed valence band and conduction band. E_g is the energy band gap and the reduce mass m_r : $1/m_r = 1/m_e + 1/m_h$ and γ is the phenomenological phase

relaxation rate. $\Omega(t) = d_{cv,k}E(t)/\hbar$ is the bare Rabi frequency, where $d_{cv,k}$ is the dipole matrix element for the electron and hole states at \mathbf{k} and $E(t)$ is the laser-driven field.

In Equations (1), the terms $\Sigma(\mathbf{k}) \equiv \sum_{k'} V_{k-k'} f_{k'}$ and $\sum_{k'} V_{k-k'} P_{k'}$ induced by Coulomb interaction are called self-energy matrices (Figure 2(a)) and internal-field matrices (Figure 2(b)), respectively. The self-energy describes the renormalization of the transition energy between the valence band and conduction band. In the thermal equilibrium, the self-energy [28] depends only on the wave vector \mathbf{k} of the particle and not upon its energy variable $i\omega$ or ω . The internal-field gives rise to excitonic effects and Coulomb enhancement by renormalized Rabi frequency $\Omega_R = \Omega(t) + (\sum_{k'} V_{k-k'} P_{k'})/\hbar$. The self-energy and internal-field modifications have a profound influence on the nonlinear response of semiconductors. In the following, we will study the effect of the renormalized Rabi frequency on the Rabi oscillation.

Considering the pulses as stable solitons, the incident femtosecond hyperbolic secant pulse exciting the semiconductors is

$$E(t) = \tilde{E}_m \sec h \left[\frac{1.76t}{t_p} \right] \cos(\omega_L t + \phi). \quad (2)$$

Here ω_L is the carrier frequency, ϕ is the absolute phase and t_p is the full width at half peak (FWHP), i.e. duration of the pulse. Correspondingly, $t_c = 2\pi/\omega_L$ is the carrier period and $n_c = t_p/t_c$ is the optical cycle number. For the semiconductor GaAs, the transition frequency between the valence band and conductor band is 1.424 eV, i.e. 2.16fs^{-1} ($\lambda = 871\text{ nm}$). Recent studies have shown that few- and sub-cycle pulses as short as femtosecond in the visible and the near UV can be generated by external compression of pulses from amplified or cavity-dumped low-repetition Ti:sapphire laser systems or by using molecular phase modulation [29–38].

Before presenting the results, we give some details of the numerical procedure. The LFC by using the assumption $\sum_{k'} V_{k-k'} P_{k'} = VP_k$ [7,8,39] is taken into account in this paper, which is equivalent to the statement that the total field is the sum of the electromagnetic field and the local field generated by the polarization of all the other pairs. We evaluate the SBE (1) for the thin bulk semiconductor GaAs by using a fourth-order Runge-Kutta procedure. We consider the case of the spatially isotropic system with only s wave scattering, which leads to all quantities only depending on the magnitude of the wave vector, and we integrate the Coulomb potential over the angle [26,28] between \mathbf{k} and \mathbf{k}' . The Rabi oscillation of the electron–hole density can be the sum of the moment of the density matrix, i.e. $f = 2 \sum_k f_k$. The factor of 2 accounts for the spin.

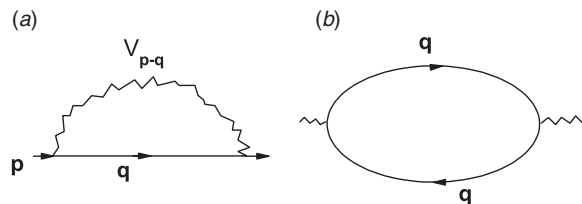


Figure 2. Schemes of (a) the self-energy and (b) internal-field. The solid and wavy lines indicate the carrier propagator and the carrier–carrier interaction, respectively.

The phase relaxation can be ignored since the pulse duration is much shorter than the phase relaxation time.

Since we are interested in the number of complete Rabi oscillations in the resonant condition, we define the dimensionless quantities $\beta = \Delta/\Omega$, $\Delta = \omega_g - \omega_x - \omega_L$. Here $\omega_x = E_0/\hbar$ is the exciton binding frequency, $\omega_g = E_g/\hbar$ is the transition frequency between the valence band and conductor band and Δ is a measure of the detuning of the optical carrier frequency ω_L from the fundamental exciton resonance.

3. Numerical results and discussion

In this section, we investigate the evolution of the Rabi oscillation in the thin bulk semiconductor interacting with the ultrashort pulse excitation for n_c range from 15 to 1 by exactly solving Equations (1). For convenience, we describe the duration of the incident pulse by the optical cycle number n_c . Figure 3 shows 2π incident pulses of $\phi = 0$ with different n_c : (a) 15, (b) 10, (c) 3 and (d) 1. Correspondingly, Rabi oscillations of the electron-hole density are shown in Figure 4. Obviously, there are two incomplete Rabi-floppings except for the case of $n_c = 1$, which is consistent with

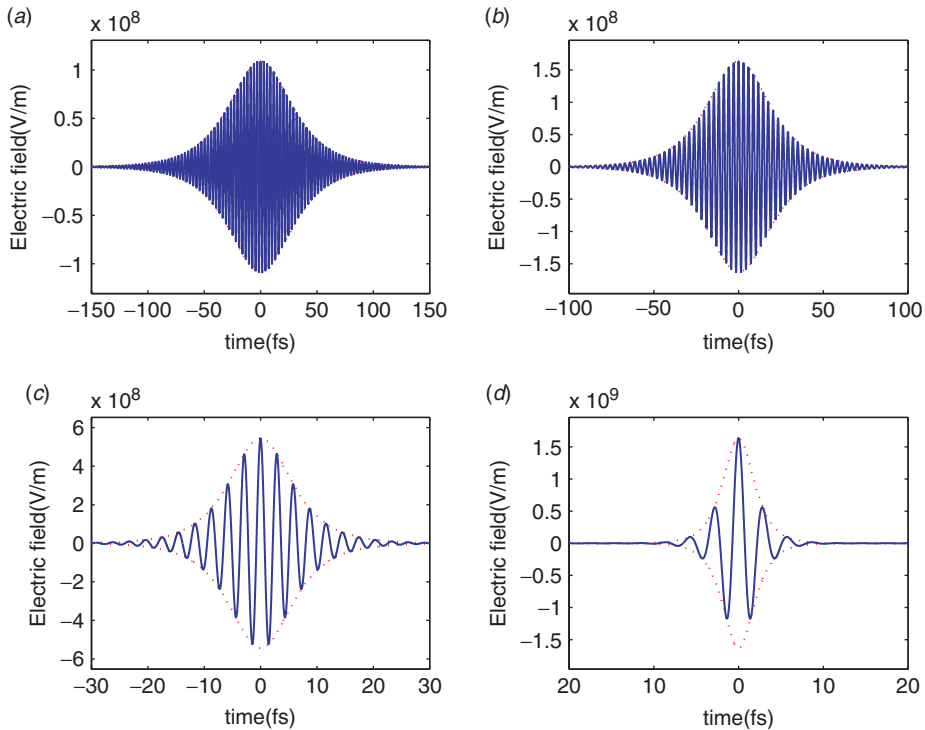


Figure 3. 2π incident pulse with various n_c : (a) 15, (b) 10, (c) 3 and (d) 1. Blue solid line: carrier of the incident pulse; red dot line: envelope of the incident pulse. (The colour version of this figure is included in the online version of the journal.)

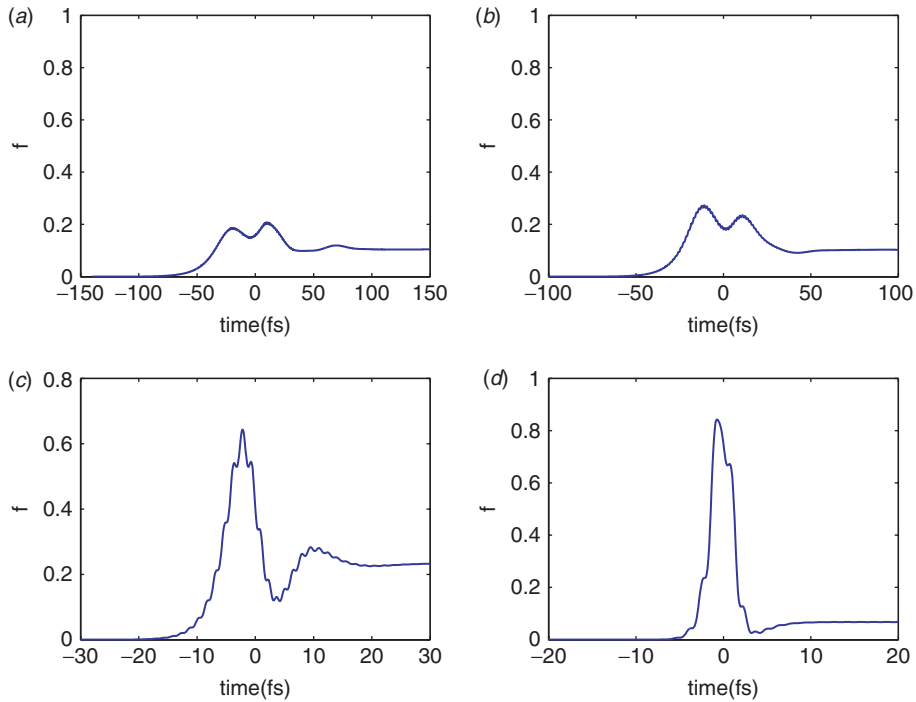


Figure 4. Rabi oscillation with LFC for the 2π incident pulse with various n_c : (a) 15, (b) 10, (c) 3 and (d) 1. (The colour version of this figure is included in the online version of the journal.)

the results of [3–5]. Moreover, the two incomplete Rabi-floppings compete against each other with the change of pulse duration. We find that when the optical cycle number n_c is about 12.5, the two peaks of the Rabi-flopping are equal. For the case of $n_c > 12.5$, the first peak of the Rabi-flopping is lower than the second one (see Figure 4(a) $n_c = 15$). In contrast, for $n_c < 12.5$, the peak of the first Rabi-flopping is higher than the second one (see Figures 4(b) and (c), $n_c = 10, 3$). For the case of $n_c = 1$, the Rabi-flopping evolves even more from two peaks into one peak (see Figure 4(d)). In addition, the peak of the first Rabi-flopping moves to positive time direction with decrease of the optical number, and so does the dip between the two peaks of the Rabi-flopping. These characteristics of the Rabi oscillation can be interpreted as follows: Rabi oscillation of electron–hole density results from the renormalized Rabi frequency $\Omega_R = \Omega(t) + (\sum_{k'} V_{k-k'} P_{k'})/\hbar$. Obviously, there is a phase lag between the two terms of the renormalized Rabi frequency. Moreover, the two terms of the renormalized Rabi frequency compete against each other. Figure 5 shows the renormalized Rabi frequency for different n_c : (a) 15, (b) 10, (c) 3 and (d) 1. Comparing Figure 4 with Figure 5, one can find that the evolution of the Rabi oscillation is almost the same as that of the envelope of the renormalized Rabi frequency.

Došlić investigated the Rabi oscillation in the case of a sub-cycle pulse driving a two-level atomic system [40]. He found a shortening of the Rabi inversion period and

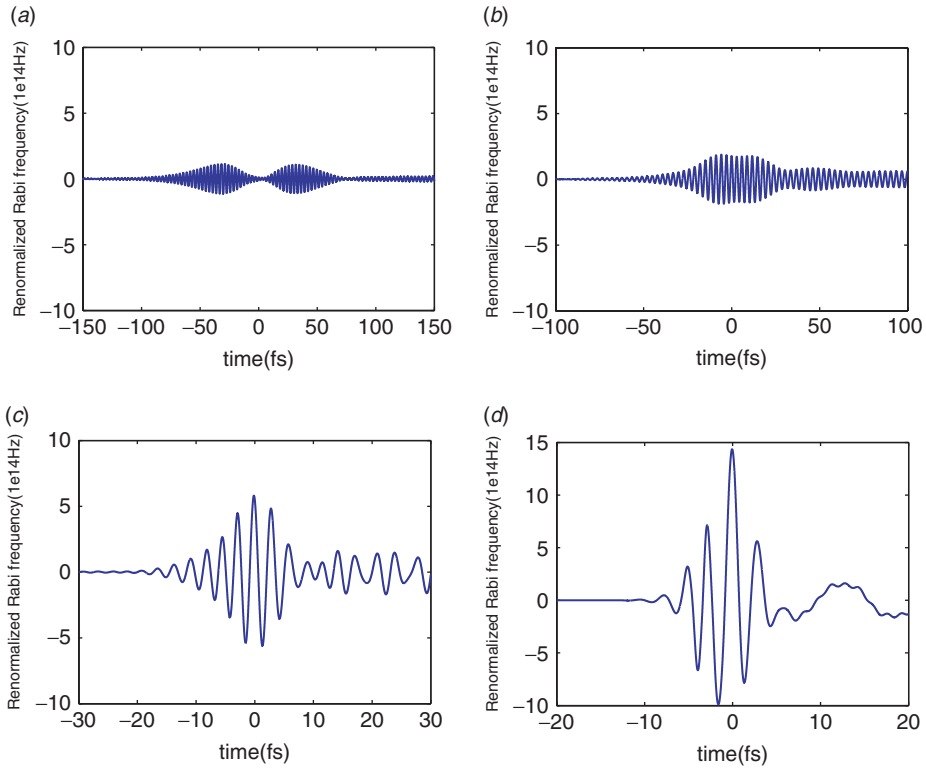


Figure 5. Renormalized Rabi frequency for the 2π incident pulse with various n_c : (a) 15, (b) 10, (c) 3 and (d) 1. (The colour version of this figure is included in the online version of the journal.)

showed that a complete inversion is unobtainable under resonant, ultrashort pulse conditions, and the efficiency of the population inversion is almost independent of the absolute phase in the nonresonant case. Here, we study the Rabi oscillation in the case of a few-cycle and even a sub-cycle pulse driving the semiconductor. We find due to the effect of the absolute phase, that a complete inversion may be obtainable under the resonant, one-cycle and sub-cycle condition. For a 2π pulse excitation, Figure 6 shows the Rabi oscillations for different absolute phase in the case of n_c : (a) 2, (b) 1 and (c) 0.8. For a few-cycle pulse case, the absolute phase hardly affects the Rabi oscillation (red line of Figure 6(a)). However, when the pulse duration reduces to one-cycle and sub-cycle, the magnitude of the Rabi oscillation enhances because of a $\pi/2$ absolute phase comparing with the case of zero absolute phase. As Figure 6(c) displays, in the case of $n_c = 0.8$, the magnitude of the Rabi oscillation reaches almost one, i.e. the complete Rabi-flopping emerges. The reason why the absolute phase enhances the magnitude of the Rabi oscillation is that the absolute phase changes strongly the renormalized Rabi frequency, especially for the sub-cycle pulse. Figure 7 shows the renormalized Rabi frequency for different absolute phase in the case n_c : (a) 2, (b) 1 and (c) 0.8. The amplitude of the envelope of renormalized Rabi frequency for a one-cycle

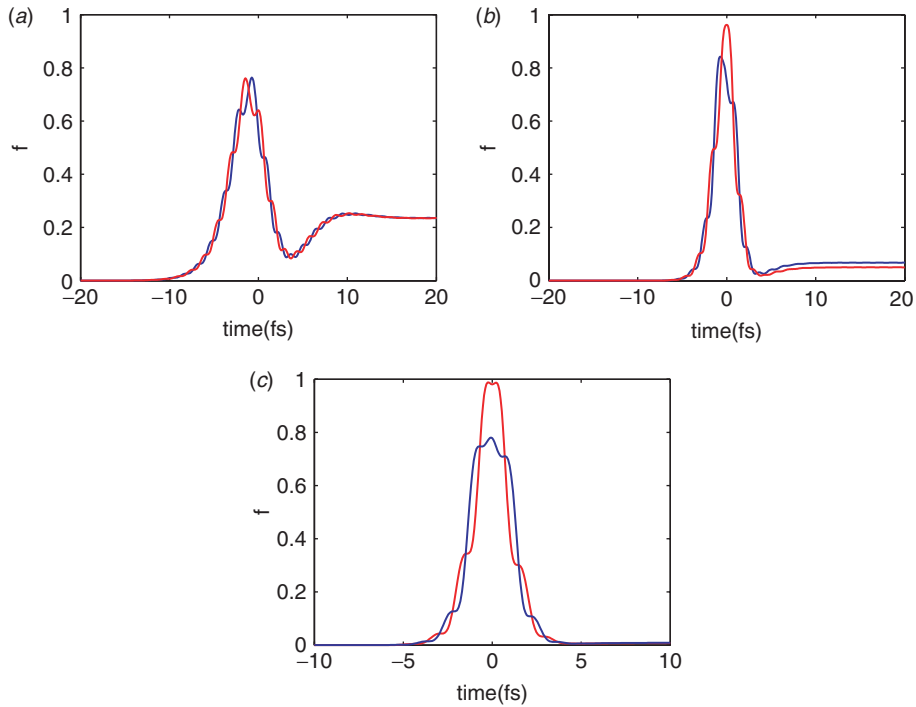


Figure 6. Rabi oscillation with LFC for the 2π incident pulse with various n_c : (a) 2, (b) 1.0 and (c) 0.8. Blue line: 0 absolute phase; red line: $\pi/2$ absolute phase. (The colour version of this figure is included in the online version of the journal.)

and a sub-cycle pulse was enhanced qualitatively due to the absolute phase. However, the peak position of the Rabi oscillation and the number of the Rabi-flopping are independent of the absolute phase. So, for the interaction between the one-cycle or sub-cycle pulse and the semiconductor, the magnitude of the Rabi oscillation may be controlled by adjusting the absolute phase of the incident pulse.

4. Summary

In conclusion, we have investigated Rabi oscillation of the electron-hole density by taking into account the LFC induced by the interacting excitons in the thin bulk semiconductor GaAs for 2π femtosecond pulse excitation. The result showed, for a few-cycle pulse, that there are two incomplete Rabi-floppings. For a one-cycle and a sub-cycle pulse, because of the absolute phase effects, the magnitude of the Rabi oscillation enhances, and even the complete Rabi-flopping emerges. The competition between the bare Rabi frequency and the internal-field matrices leads to these characteristics of the Rabi oscillation.

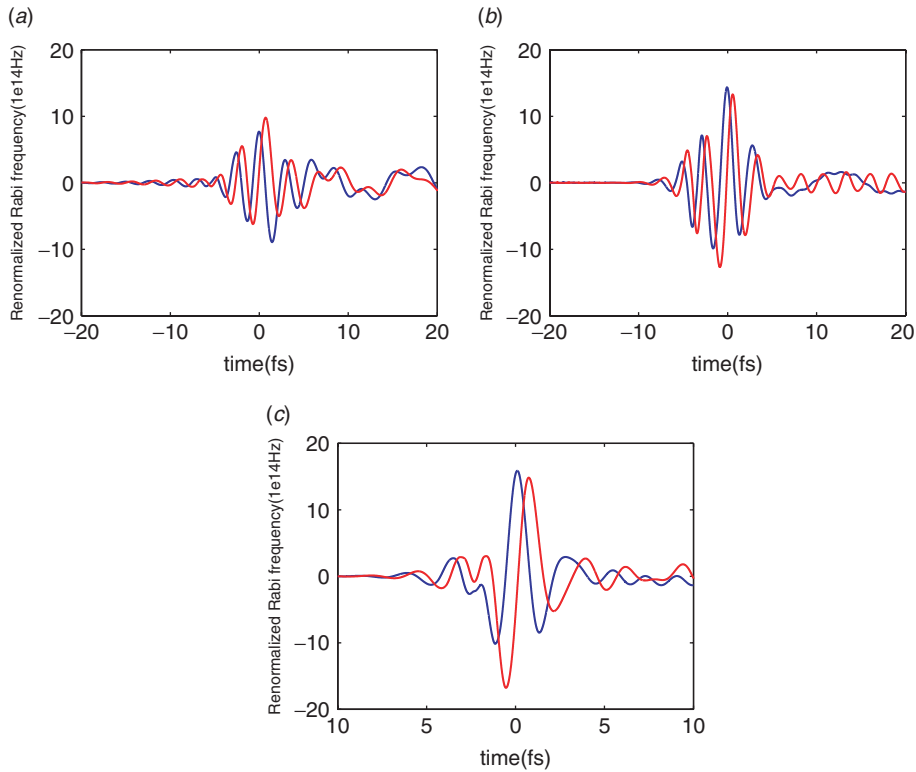


Figure 7. Renormalized Rabi frequency for the 2π incident pulse with various n_c : (a) 2, (b) 1 and (c) 0.8. Blue line: 0 absolute phase; red line: $\pi/2$ absolute phase. (The colour version of this figure is included in the online version of the journal.)

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