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To cite this article: Shicheng Jiang et al 2017 J. Phys.: Condens. Matter 29 275702

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Quantum-trajectory analysis for charge transfer in solid materials induced by strong laser fields

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Received 6 February 2017, revised 6 May 2017
Accepted for publication 8 May 2017
Published 6 June 2017

Abstract

We investigate the dependence of charge transfer on the intensity of driving laser field when SiO₂ crystal is irradiated by an 800 nm laser. It is surprising that the direction of charge transfer undergoes a sudden reversal when the driving laser intensity exceeds critical values with different carrier–envelope phases. By applying quantum-trajectory analysis, we find that the Bloch oscillation plays an important role in charge transfer in solids. Also, we study the interaction of a strong laser with gallium nitride (GaN), which is widely used in optoelectronics. A pump–probe scheme is applied to control the quantum trajectories of the electrons in the conduction band. The signal of charge transfer is controlled successfully by means of a theoretically proposed approach.

Keywords: laser, ultrafast current, charge transfer, SBEs

(Some figures may appear in colour only in the online journal)

1. Introduction

It is a common sense that the minimum time window required to change the electronic signal determines the rate of information exchange. The metal–oxide–semiconductor field-effect transistors (MOSFETs) are the basic devices in modern telecommunication technology [1]. The cutoff-frequencies of the MOSFETs can be higher than 100 GHz in theory [2–4]. While the maximum speed of the processor has been limited to ~3 GHz for a long time because of two main factors: the charging time of the interconnected wires and heat dissipation [5]. Fortunately, laser technology provides an alternative strategy to control the electric signals. In 2011, Ghimire et al first observed non-perturbative high-order harmonics generation (HHG) in ZnO crystal [6]. More recently, the spectral plateau has been extended to the extreme ultraviolet (EUV) range [7, 8]. These works demonstrated that it is possible to control the electric current on a subpicosecond timescale. In 2013, Krausz and his team revealed an ultrafast ‘turning on’ and ‘turning off’ of the current [9, 10]. The conductivity and, consequently, current can be switched on and off in a dielectric using optical fields on a timescale of the order of or less than 1 fs. Since the current signals are switched so fast that the current measurement techniques do not allow for detecting such currents in real time. The transferred charges induced by the driving laser become the signal used to investigate the response of the materials to the driving laser. It has been demonstrated in experiment that there is charge transfer along the direction of the driving few-cycle laser [9]. Besides, the signals of charge transfer show oscillation behavior with changing the carrier–envelope phase (CEP) of the laser [9, 11]. These characteristics will not be changed for different materials [12]. The CEP-dependent charge transfer can also be used to detect the CEP of few-cycle laser pulses under ambient conditions [11]. In theory [13], CEP control of the electric current was interpreted as a result of quantum-mechanical interference of
multiphoton excitation channels. For such important potential applications, laser-induced charge transfer has attracted much attention in recent years [9–14].

In this work, the laser-induced charge transfer is investigated theoretically by solving semiconductor Bloch equations (SBEs) [15–20]. In our previous study, the shapes of k-space-dependent dipole moments have been demonstrated to play an important role in harmonic generation [21]. It is necessary to perform calculations with accurate band structure and k-space-dependent dipole moments. Hereafter, all the band structures and k-dependent dipole moments of the target materials were obtained from high-level first-principles calculations using the Vienna ab initio simulation package (VASP) [22, 23]. As an insulator, α-quartz SiO2 has been widely used in recent experiments [7, 9–11]. To test whether the SBEs is valid to investigate laser-induced charge transfer, theoretical results and the first experimental data [9] are compared in appendix A. Using the same target, i.e. SiO2, we study the dependence of charge transfer on laser intensity in the first main part of this work. As the intensity of the driving field is tuned from 1 × 10^13 W cm^{-2} to 6 × 10^13 W cm^{-2}, the direction of the charge transfer undergoes a sudden reversal when the driving laser intensity continues increasing. A classical method called quantum-trajectory analysis is employed to seek the mechanism underlying this phenomenon. It is found that the quantum trajectories of the electron wavepackets in the conduction band can be controlled by the driving field. In the second main part of this work, a pump–probe scheme is applied to control the quantum trajectories of the excited electrons. Actually, the highly nonlinear current control for large bandgap materials is difficult to realize in experiment. Significant current signal should be generated at the expense of very high intensity fields. This can be a limitation for the practical application of the proposed theoretical scheme. Thus, the widely used material in optoelectronics, gallium nitride (GaN) with a medium bandgap, is chosen as the target to implement the pump–probe scheme. The signal of the charge transfer is successfully controlled by tuning the delay time between two different pulses.

2. Theoretical model

The first target we studied is SiO2 crystal in α-quartz structure with P3121 symmetry group. To model the laser–crystal interaction, we start with the time-dependent Schrödinger equation (TDSE) in length gauge under single electron approximation:

$$i\hbar \frac{\partial \psi(r, t)}{\partial t} = \left( \frac{p^2}{2m} + v(r) + r \cdot E(t) \right) \psi(r, t).$$

(1)

Using the field-free Bloch function basis, the wavefunction \( \psi(r, t) \) of the above equation can be expressed as:

$$\psi(r, t) = \sum_m \int_{\text{BZ}} a_m(k, t) \phi_{m, k}(r) dk,$$

(2)

where \( \phi_{m, k}(r) \) is the Bloch eigenstates with energies \( \varepsilon_m(k) \). Based on the Bloch theorem, \( \phi_{m, k}(r) \) can be expressed as the product of a plane wavefunction \( e^{ik \cdot r} \) and a periodic function \( u_{m, k}(r) \), e.g. \( \phi_{m, k}(r) = u_{m, k}(r)e^{ik \cdot r} \). Inserting equation (2) into (1), equation (1) becomes

$$i\hbar \frac{\partial u_m(k, t)}{\partial t} = \varepsilon_m(k) u_m(k, t) + E(t) \sum_k \int \phi_{m, k}(r) \cdot r \cdot \phi_{m, k}(r) \times \partial u_m(k, t).$$

(3)

We define the microscopic polarization \( P_k^{mn} = a_m(k, t) a_n^*(k, t) \), and electron density \( \vec{f}_k \). When only valence and conduction bands are taken into account, the two-band SBEs describing the laser–crystal interaction is deduced:

$$i\hbar \frac{\partial \vec{P}_k}{\partial t} = \left( \varepsilon_c(k) - \varepsilon_v(k) - i\frac{1}{T_2} \right) \vec{P}_k + \hbar \vec{f}_k, \quad P_k^{mn} + iE(t) \nabla \epsilon \vec{P}_k,$$

(4)

$$\hbar \frac{\partial \vec{f}_k}{\partial t} = -2im \left[ \vec{h} \varepsilon(k) E(t) \vec{P}_k^* \right] + E(t) \nabla \epsilon \vec{f}_k,$$

(5)

where \( \vec{f}_k \) is the occupation of electron (hole) in the lowest conduction band (highest valence band). \( P_k \) is the microscopic polarization between the conduction and valence band. \( \varepsilon_c(k) \) and \( \varepsilon_v(k) \) are the k-dependent energy bands of the conduction band and valence band, respectively. \( d(k) \) is the k-dependent transition dipole moment. In the equation above, \( \frac{1}{T_2} \) represents the dephasing effect in the solid materials. \( T_2 \) is set to be 1.0 fs for SiO2 [7]. Since the driving laser in our work is linearly polarized, a one-dimension calculation is sufficient. So, the variable \( k \) in equations (4) and (5) is scalar. Here, the \( \Gamma-M \) direction in reciprocal space is chosen. What we should emphasize is that, in the derivation, the relation \( \int \phi_{m, k}^*(r) \phi_{n, k}(r) = i \nabla_k \delta_{m, n} + d_{m, n}(k) \), where \( d_{m, n}(k) = \{ u_{n, k}(r) \} \nabla_k \{ u_{m, k}(r) \} \) is used. Under the infinite volume, nonvanishing boundary conditions,

$$\{ u_{n, k}(r) \} \nabla_k \{ u_{m, k}(r) \} = -\frac{\{ u_{n, k}(r) \}}{\varepsilon_n(k) - \varepsilon_m(k)} \{ u_{m, k}(r) \}.$$

(6)

This formula is correct only when \( m = n \). For the case of \( m \neq n \), the expression is much more complex. The diagonal term \( d_{m, m}(k) \) can be regarded as just a small correction of the band structure which is usually ignored.

The wavefunction \( u_{n, k}(r) \) and energy bands \( \varepsilon_m(k) \) can be obtained by using first-principles calculations. Here, the VASP code is used [22, 23] to calculate the electronic properties with the exchange-correlation functional of Perdew–Burke–Ernzerhof [24] within the generalized gradient approximation. The energy cutoff was set to be 500 eV. Geometric structures were fully relaxed and numerical convergence was achieved with thresholds of 10^{-5} eV in energy and 10^{-2} eV Å^{-1} in force with cutoff energy of 500 eV. A Monkhorst–Pack mesh of 11 × 11 × 9 k points was used in the Brillouin zone. The band structures and dipole moments of SiO2 can be found in our previous work [21]. Once the electrons in the valence
band are excited to the conduction band, the electrons in the conduction band and holes in the valence band will be driven by the laser. The total currents induced by the driving field contains three parts: intraband electric currents in the conduction band $j_c(t)$ and the valence band $j_v(t)$, and $j_{sv}(t)$ induced by macroscopic interband polarization. These three items are given by

$$j_{sv}(t) = \int_{BZ} e \frac{\partial \epsilon_{cv}(k)}{\partial k} f_{k}^{(v)}(t) dk,$$

$$j_c(t) = \int_{BZ} \left[ d(k) \frac{\partial p_t(k)}{\partial t} + c.c. \right] dk,$$

where $e = +1$ for the valence band and $e = -1$ for the conduction band. The transferred charge induced by the driving laser is

$$Q(t) = \int_{-\infty}^{t} \left( j_{c}(t') + j_{v}(t') \right) dt'.$$

The component of $j_c(t)$ is negligible for the low mobility of holes in the valence band. Besides, $j_{sv}$ mainly describes the macroscopic polarization response which will vanish when the laser is over [18]. So, we restrict our analysis to the transferred charge induced by $j_c(t)$ in this work. As far as we know, the signals of charge transfer calculated by solving SEBs have not been compared with available experimental data. To provide more evidences for that, the SEBs can be used to calculate the signal of charge transfer, we compare our theoretical results with the first experimental data [9] in appendix A.

3. Results and discussion

3.1. Quantum-trajectory analysis for laser-induced charge transfer in SiO$_2$

In figure 1(a), the dependence of transferred charge on the intensity of laser with CEP $= 0.4\pi$, wavelength $\lambda = 780$ nm is displayed. The black squares represent the values of transferred charge. The red dots represent the values of $C \int_{-\infty}^{t} \tilde{A}(t')dt'$, where $A(t)$ is the vector potential of the driving laser, and $C$ is an adjustable parameter. As already demonstrated [25], the charge can be obtained by $Q(t) \sim \int_{-\infty}^{t} \tilde{A}(t')dt' + \ldots$ when the laser intensity is weak. Whereas we observe that as the intensity of the laser is increased, the direction of the charge transfer undergoes a sudden reversal, and the charge transfer will not follow $\int_{-\infty}^{t} \tilde{A}(t')dt'$ any more. The critical intensity can be seen to be $4.4 \times 10^{13}$ W cm$^{-2}$ (1.84 V Å$^{-1}$) for CEP $= 0.4\pi$. In figure 1(b), we also present the dependence of transferred charge in SiO$_2$ on CEP for laser intensities of $3.6 \times 10^{13}$ W cm$^{-2}$, $4.0 \times 10^{13}$ W cm$^{-2}$, $4.8 \times 10^{13}$ W cm$^{-2}$, and $5.0 \times 10^{13}$ W cm$^{-2}$, respectively. It is found that there exist three main quantum trajectories labeled by trajectory 1, 2 and 3. Like the ‘long or short quantum trajectory’ of HHG from atoms and molecules, the electrons ‘born’ in the conduction band at different times will experience different trajectories, leading to different contribution to the charge transfer. The solid lines in figures 2(a) and (b) represent the ‘semiclassical trajectories’ born at different times. The ‘semiclassical trajectories’ can be expressed as follows:

$$k(t) = k_0 - \int_{t_0}^{t} E(t) dt,$$

where $k_0$ is the initial position of the excited electron, $t_0$ is the time when the electron is excited from the valence band to the conduction band. In our calculations, $k_0$ is set to be $-0.045$, and $t_0$ is set to be 6.44 fs, 6.65 fs and 6.83 fs for these three trajectories, respectively. By comparing figures 2(a) and (b), we can see that as the intensity of the laser increases, the quantum trajectories will be changed, especially for the trajectory 1. This phenomenon will shed light on the charge transfer reversal. In a semiclassical picture, the displacement of the electron in the conduction band driven by the laser can be expressed as:

![Figure 1](image-url)
\[
D = \int_{t_{\text{min}}}^{t_{\text{max}}} v_c(k(t))dt, \quad (11)
\]
where \(v_c = \frac{\partial \psi(k)}{\partial k}\). The positive/negative \(D\) will contribute to the negative/positive transferred charge. We then investigate the dependence of \(D\) on laser intensities for these three semiclassical trajectories, as presented in figure 3. It is clearly seen that the semiclassical displacement for trajectory 1 changes from positive values to negative values as the intensity of the laser is increased. As a result, the whole transferred charge will be changed from negative to positive as the laser intensity reaches a threshold value.

The quantum-trajectory analysis has been applied to explain the reversal of the charge transfer successfully above. Now, we know that the signal of charge transfer is determined by the quantum trajectory of the electronic wave packets on the conduction band. Based on the analysis above, two important aspects should be kept in mind. First, the electrons excited from the valence band to the conduction band at different times would travel different quantum trajectories. Second, all these quantum trajectories would contribute to the charge transfer signal. The origin on signal reversal is the well-known Bloch oscillation. Specifically, since the group velocities for two opposite directions of the reduced Brillouin zone (RBZ) have opposite values, when the electronic wave packets are driven by laser field to exceed the boundary of the RBZ and reach the opposite direction, i.e. Bloch oscillation happens, there will be opposite current. As the intensity of the driving laser is increased, the electronic wave packets will exceed the boundary of the RBZ and then experience longer excursion with opposite group velocity. As we can see from figure 2, when the laser intensity is \(3.6 \times 10^{13} \text{ W cm}^{-2}\), electrons of...
trajectory $i$ almost always stay in the positive region of the RBZ. If the laser intensity is increased, e.g. $4.8 \times 10^{13}$ W cm$^{-2}$, these electrons will almost always stay in the negative part of the RBZ resulting in a reversal of charge transfer.

3.2. Quantum-trajectory control of charge transfer in GaN by the pump–probe scheme

Based on the analysis above, a pump–probe scheme can be utilized to control the signal of charge transfer. An ultraviolet pulse can pump the electrons from the valence band to the conduction band. The other mid-infrared pulse can then be used to drive the electrons on the conduction band. It should be noted that the highly nonlinear laser-induced current control for large bandgap materials is very difficult in experiment. For SiO$_2$ with a bandgap of $\sim$9 eV, significant current signal should be generated at the expense of a very high intensity laser, which is in fact a limitation for the practical application of the proposed theoretical scheme. Furthermore, if the intensity of the pump laser is too high, the driving process would be influenced strongly by the pump pulse. Thus, GaN with a bandgap of $\sim$3.4 eV [27], is a better choice to implement the pump–probe scheme. Also, GaN materials have been previously taken as samples to investigate the current control in semiconductors by the CEP of the laser [28].

In this work, a 1.68 fs/337 nm ultraviolet pulse is used to excite the electron from the valence band to the conduction band. Then, a 15 fs/2500 nm mid-infrared pulse drives the electron in the conduction band. As the delay time between these two pulses is altered, the quantum trajectory of the electron in the conduction band will be controlled. The signal of the charge transfer is successfully controlled by tuning the delay time between two pulses. The sketch of the pump–probe scheme is presented in figure 4(a).

The accurate band structure and $k$-space-dependent dipole moments of GaN obtained from high-level first-principles calculations can be found in appendix B. The intensities of the ultraviolet and mid-infrared pulses are set to be $5 \times 10^{11}$ W cm$^{-2}$ and $5 \times 10^{10}$ W cm$^{-2}$, respectively, so that the ultraviolet pulse could pump the electron efficiently, but due to its short wavelength, it has little influence on the quantum trajectories when they are driven by the mid-infrared pulse. The mid-infrared pulse will hardly contribute to the excitation process for its low intensity and long wavelength. The black squares in figure 4(b) show the dependence of the charge transfer on the delay time. The charge transfer is controlled successfully by altering the time delay between the ultraviolet pulse and mid-infrared pulse. The blue line in figure 4(b) is the vector potential of the mid-infrared laser with CEP = 0.

We find that the charge transfer highly depends on the value of the vector potential of the laser when the electrons are excited into the conduction band. Next, we will explain this finding. Assuming that the charge transfer is mostly induced by the intraband moving of the electrons on the conduction band, the transferred charge $Q$ will read as:

$$Q = \int_{t_b}^{t_a} (-v_x(k(t))f_0)dt,$$

where $v_x(k) = \frac{\partial \epsilon(k)}{\partial k}$, $f_0$ is the density of the electrons excited to the conduction band by ultraviolet pulse. The excitation is assumed to occur at time $t_b$, and $f_0$ will not change in the following time. Since the energy bands can be expressed as equation (B.1) (appendix B), the odd function $v_x(k)$ can be expanded as:
which is the value of the vector potential of the mid-infrared laser when the electrons are excited from the valence band to the conduction band. For example, the absolute values of the vector potential marked for S1 is slightly larger than that for S2.

The first part in the right side of equation (16) will contribute little to $Q$. So, we can understand that the charge transfer will highly depend on $A(b_0)$, which is the value of the vector potential of the mid-infrared laser when the electrons are excited into the conduction band. Of course, charge transfer will also depend on $b_0$ which is the time when the electrons are excited from the valence band to the conduction band. For example, although the absolute values of the vector potential marked by red circles are the same in figure 4(b), $|Q|$ for S1 is slightly larger than that for S2.

### 4. Conclusions

In summary, the SBEs have been solved to calculate the laser-induced charge transfer. In general, the theoretical results match the early experimental data very well, so it is appropriate by using this approach to investigate the phenomenon of laser-induced charge transfer in semiconductors. The direction of laser-induced charge transfer will undergo a sudden reversal when the driving laser intensity exceeds critical values in SiO$_2$ crystal irradiated by the 800 nm laser. By applying quantum-trajectory analysis, we address that the electrons excited from the valence band to the conduction band at different times would travel different quantum trajectories. All these quantum trajectories would contribute to the charge transfer signal. As laser intensity is increased, one of these trajectories will travel beyond the first Brillouin zone, and Bloch oscillation occurs. Consequently, the signal of charge transfer will reverse its direction. Further, a pump–probe scheme is proposed to control the signal of charge transfer in GaN semiconductors. As the delayed time between these two pulses is altered, the quantum trajectories of the electrons on the conduction band will be tuned; hence the signal of charge transfer is controlled successfully. What is more, the charge transfer is found to highly depend on $A(b_0)$ which is the value of the vector potential of the mid-infrared laser when the electrons are excited into the conduction band. We expect this theoretical work will provide useful insights into the potential utilization of optical field controlled ultrafast electronics using dielectrics or semiconductors.

### Acknowledgments

This work was supported by NSF of China Grant No. 21373113, the Fundamental Research Funds for the Central Universities (No. 30920140111008 and 30916011105).

### Appendix A. Theoretical results versus experimental data

In this part, the results calculated by solving SBEs are compared with the first experimental data for SiO$_2$ crystal [9]. The parameters of the laser are set to be the same as in the experiment. A 4 fs/780 nm laser pulse is used. The dependence of the maximum amplitude of the transferred charge on the electric field strength is presented in figure 5(a) in red stars. What we should emphasize is that all the calculated results are multiplied by 3500 to match the experimental data. Since the effective surface area is difficult to determine experimentally, it is reasonable that the theoretical results are multiplied by a scale factor for comparing the experimental data. In figure 5(b), CEP dependence of charge transfer is shown when the electric field strength is about 1.7 V Å$^{-1}$. In order to simulate the experimental environment as much as possible, the screening-field-induced laser is taken into account [29]. While in the main text, the screening field is not taken into account for simplicity. In general, our calculated results by solving SBEs match the experimental data very well.

### Appendix B. Band structure and dipole moments for GaN

Only the lowest conduction band and the highest valence band are considered in calculations. The polarization of the laser is along Γ–A. Therefore, we merely present the dispersions of these two bands for GaN in figure 6. The valence and conduction bands are indicated by black squares and red dots, respectively. The blue line is the
dipole moments between the highest valence band and the lowest conduction band. The $k$-dependent band structures can be expanded as:

$$E_{c}(k) = \sum_{j=0}^{+\infty} a_{c0}(i) \cos((i - 1) \cdot k \cdot b),$$  \hspace{1cm} (B.1)

where $b$ (5.166 Å) is the optimized lattice constant along Γ–A that is close to the reported value [27]. $a_{c0}(i)$ are expansion coefficients. These coefficients can be found in table 1.

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