

Three-dimensional real-space electron dynamics in graphene driven by strong laser fields

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We theoretically investigate the three-dimensional (3D) electron dynamics of graphene in real space under strong laser fields using time-dependent density functional theory (TDDFT). We successfully reproduce the reversal of current direction originating from the cancellation of two oppositely directed residual currents, as previously predicted by [Y. Morimoto *et al.*, *New J. Phys.* **24**, 033051 (2022)]. By distinguishing contributions from individual orbitals, our results validate the two-level system approximation and also emphasize that the first-principles approach agrees better with experimental results for light-driven residual current, especially in extremely strong fields. Furthermore, our 3D model reveals that the real-space atomic-scale current induced by strong laser fields is concentrated slightly above and below the graphene basal plane, rather than strictly within it. The two oppositely directed currents exhibit a pronounced height separation in the out-of-plane direction, indicating that the ring current is not confined to the graphene plane but forms a rotating 3D circulation loop, which is absent in the reduced-dimensional model.

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I. INTRODUCTION

Since its discovery, graphene [1] has attracted extensive attention due to its zero band gap, weak screening effect, high laser damage threshold, and ultrafast optical response [2,3]. These unique features make graphene one of the ideal materials for studying lightwave electronics, i.e., the ultrafast generation and manipulation of macroscopic electric currents through the waveform of ultrashort laser pulses.

In most studies, the electron dynamics in graphene (and solids in general) driven by strong laser fields is analyzed in the momentum (reciprocal) space [4–8]. The conduction and valence bands of graphene meet at the high-symmetry points K and K' , forming a characteristic Dirac cone. The interband transitions preferentially occur around these points, and the intraband electron dynamics follows the acceleration theorem, $\mathbf{k}(t) = \mathbf{k}_0 + e\mathbf{A}(t)/\hbar$, where $\mathbf{k}_0$ is the initial, field-free wave vector, e the elementary charge, $\mathbf{A}(t)$ the vector potential of the laser field, and $\hbar$ the reduced Planck constant.

The nonlinear interaction of laser pulses with low temporal symmetry can induce controllable currents in dielectrics [9–11]. Similar field-driven current phenomena have also been explored in gapless materials such as graphene. Ishikawa theoretically predicted the generation of a residual current in graphene under intense monocycle pulses [5]. Then, Higuchi *et al.* experimentally demonstrated that the residual electric current is generated by a spatially asymmetric few-cycle laser pulse because of the asymmetric carrier population in the reciprocal space induced via Landau-Zener-Stückelberg interference [12]. They also showed that its direction can be reversed by increasing the laser field [12–14].

On the other hand, real-space electron dynamics is emerging as a powerful approach for intuitively understanding ultrafast and nonlinear phenomena under strong light fields [15–19]. More recently, Morimoto *et al.* [20], including one of the present authors, theoretically studied the atomic-scale real-space distribution of the light-field-induced currents, by combining the nearest-neighbor tight-binding (TB) model and the semiconductor Bloch equation (SBE). It has been revealed that the atomic-scale currents flow dominantly through the network of the π bonds. However, surprisingly, when the residual current changes its sign with increasing laser intensity, this flow does not vanish uniformly, but the back current appears through the center of the hexagonal atomic framework, canceling the current along the hexagon. While the inclusion of the overlap integral between neighboring atomic orbitals, often neglected in the momentum-space analysis, was essential to properly describe the charge density between

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atomic sites, the same feature of the theoretical modeling led to slight violation ($\sim 10\%$) of the continuity equation for the current [20]. This concern motivates us to perform a verification using first-principles calculations.

The time-dependent density functional theory (TDDFT) is an *ab initio* framework successfully applied to ultrafast electron dynamics under intense laser fields in molecules [21–26] and solids [27–41]. These studies, often performed using real-time TDDFT frameworks, have demonstrated that the time propagation of Kohn-Sham orbitals provides a powerful tool to describe highly nonequilibrium electron dynamics under intense laser fields. In particular, previous real-time TDDFT studies have investigated ultrafast electron dynamics in graphene under short laser pulses, focusing mainly on occupation evolution and its momentum-space characteristics [42]. However, these analyses have primarily focused on macroscopic observables or reciprocal-space quantities, while the spatial structure of current density in real space remains largely unexplored. In the present study, we extend this previous work [20], expected to contain the essential physics in the simplest possible manner, to verify its findings with TDDFT. We systematically investigate graphene's real-space electron dynamics in three dimensions, which provides deeper insights into the spatial distribution of electron currents under intense

laser fields. Our numerical results well reproduce the measured dependence of the residual current on field strength and confirm the key results of Ref. [20]. Moreover, our calculation reveals the current distribution perpendicular to the graphene plane. In particular, the ring current reported in the previous work [20] is neither confined to the graphene plane nor even parallel to it, but instead forms a rotating three-dimensional (3D) circulation loop with pronounced out-of-plane height difference.

This paper is organized as follows. Section II briefly reviews TDDFT and how to evaluate the current. Section III shows the calculated ground state and linear response properties of graphene. In Sec. IV, we present and discuss our numerical results. Conclusions are given in Sec. V.

II. TIME-DEPENDENT DENSITY FUNCTIONAL THEORY

The quantum dynamics of a many-electron system subject to a laser field is calculated by solving the time-dependent Kohn-Sham (TDKS) equations for the time-dependent Bloch orbitals $u_{n,\mathbf{k}}(\mathbf{r}, t)$, with band index n and crystalline momentum $\mathbf{k}$ [29,43], which inherit the lattice periodicity of the system such that $u_{n,\mathbf{k}}(\mathbf{r} + \mathbf{R}, t) = u_{n,\mathbf{k}}(\mathbf{r}, t)$, where $\mathbf{R}$ is the lattice vector. The TDKS equation reads

$$i\hbar \frac{\partial}{\partial t} u_{n,\mathbf{k}}(\mathbf{r}, t) = \left\{ \frac{1}{2m} \left(-i\hbar \nabla + \hbar \mathbf{k} + \frac{e}{c} \mathbf{A}(t) \right)^2 - e\varphi(\mathbf{r}, t) + \hat{v}_{\text{NL}}^{\mathbf{k} + \frac{e}{\hbar c} \mathbf{A}(t)} + V_{\text{xc}}[n(\mathbf{r}, t)] \right\} u_{n,\mathbf{k}}(\mathbf{r}, t), \quad (1)$$

where m denotes the electron mass, c the vacuum velocity of light, and $V_{\text{xc}}[n(\mathbf{r}, t)]$ the exchange-correlation potential. The scalar potential $\varphi(\mathbf{r}, t)$ includes the Hartree potential and the local part of norm-conserving ionic pseudopotential [44,45]. The nonlocal part of the pseudopotential is defined as $\hat{v}_{\text{NL}}^{\mathbf{k}} = e^{-i\mathbf{k} \cdot \mathbf{r}} \hat{v}_{\text{NL}} e^{i\mathbf{k} \cdot \mathbf{r}}$, where $\hat{v}_{\text{NL}}$ can be separated from the norm-conserving pseudopotential.

The time-dependent electron density $n(\mathbf{r}, t)$ is given by

$$n(\mathbf{r}, t) = \frac{1}{N_k} \sum_{\mathbf{k}, n \in \text{occ}} |u_{n,\mathbf{k}}(\mathbf{r}, t)|^2, \quad (2)$$

where N_k and occ are the number of k points and the occupied orbitals, respectively. We adopt the Perdew-Zunger self-interaction-corrected local density approximation (LDA) functional for V_{xc} [46], which, for the case of graphene, yields accuracy comparable to the generalized gradient approximation [47] and, at the same time, can reduce the computational cost.

We evaluate the time-dependent electron current density distribution as

$$\mathbf{J}(\mathbf{r}, t) = -\frac{e}{mN_k} \text{Re} \sum_{\mathbf{k}, n \in \text{occ}} u_{n,\mathbf{k}}^*(\mathbf{r}, t) \times \left(-i\hbar \nabla + \hbar \mathbf{k} + \frac{e}{c} \mathbf{A}(t) \right) u_{n,\mathbf{k}}(\mathbf{r}, t), \quad (3)$$

and its volume average as

$$\mathbf{I}(t) = \frac{1}{\Omega} \int_{\Omega} \mathbf{J}(\mathbf{r}, t) d\mathbf{r} + \mathbf{I}_{\text{NL}}, \quad (4)$$

where Ω denotes the volume of the unit cell, and $\mathbf{I}_{\text{NL}}$ accounts for the contribution from the nonlocal pseudopotential in the unit cell, which can be calculated as [48]

$$\begin{aligned} \mathbf{I}_{\text{NL}}(t) = & -e \int_{\Omega} \frac{1}{N_k} \sum_{\mathbf{k}, n \in \text{occ}} u_{n,\mathbf{k}}^*(\mathbf{r}, t) \\ & \times \frac{1}{i\hbar} \left[\mathbf{r}, \hat{v}_{\text{NL}}^{\mathbf{k} + \frac{e}{\hbar c} \mathbf{A}(t)} \right] u_{n,\mathbf{k}}(\mathbf{r}, t) d\mathbf{r}. \end{aligned} \quad (5)$$

We define the time-dependent occupancy $\rho_{n,\mathbf{k}}(t)$ through the projection of the time-dependent Bloch orbitals $u_{n,\mathbf{k}}(t)$ onto the initial field-free orbitals $u_{n,\mathbf{k}}^{\text{GS}}$ as

$$\rho_{n,\mathbf{k}}(t) = \sum_{m \in \text{occ}} \left| \int_{\Omega} d\mathbf{r} u_{m,\mathbf{k}}^*(\mathbf{r}, t) u_{n,\mathbf{k}}^{\text{GS}}(\mathbf{r}) \right|^2. \quad (6)$$

Using the open-source package SALMON [26], our simulation is conducted on an orthogonal unit cell of graphene containing four carbon atoms, discretized into $24 \times 14 \times 85$ real-space and $45 \times 75 \times 1$ reciprocal-space grids. A vacuum layer with a total thickness of 1.5 nm (0.75 nm each side) perpendicular to the graphene plane is introduced. Periodic boundary conditions satisfied by the TDKS equations in the velocity gauge are used to model an infinitely extended graphene sheet, based on the Bloch theorem. This calculation domain corresponds to the orthogonal lattice parameters as $a = 0.426$ nm, $b = 0.246$ nm, $c = 1.5$ nm. Fourier interpolation is applied to smooth the real-space data for visualization. To compare our results with those in Ref. [20], we project the electron density and current density onto the graphene plane

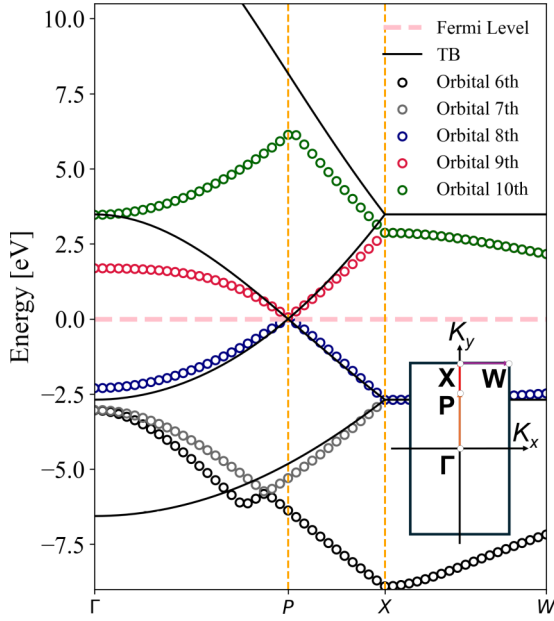


FIG. 1. Band structure of each orbital along high symmetry Γ - P - X - W direction. Open circles represent the DFT results while the solid line indicates the tight-binding results.

(xy plane) as $n_{2D-\hat{z}}(x, y) = \int_0^c n(\mathbf{r}) dz$, and $\mathbf{J}_{2D-\hat{z}}(x, y, t) = \int_0^c \mathbf{J}(\mathbf{r}, t) dz$. Similarly, to analyze the out-of-plane current distribution, we project $n(\mathbf{r}, t)$ and $\mathbf{J}(\mathbf{r}, t)$ onto the zx plane as $n_{2D-\hat{y}}(x, z) = \int_0^b n(\mathbf{r}) dy$ and $\mathbf{J}_{2D-\hat{y}}(x, z, t) = \int_0^b \mathbf{J}(\mathbf{r}, t) dy$.

III. GROUND STATE AND LINEAR RESPONSE

The initial state of the system is taken as the ground state of the field-free Hamiltonian. The calculated band structure is shown in Fig. 1, which represents the energy eigenvalues of the Bloch orbital functions. For comparison, the π^* and π band energies E_c and E_v , respectively [49],

$$E_c = \frac{t_0 - t_1 \sqrt{1 + 4 \cos^2\left(\frac{a}{2}k_y\right) \pm 4 \cos\left(\frac{\sqrt{3}a}{2}k_x\right) \cos\left(\frac{a}{2}k_y\right)}}{1 - s_1 \sqrt{1 + 4 \cos^2\left(\frac{a}{2}k_y\right) \pm 4 \cos\left(\frac{\sqrt{3}a}{2}k_x\right) \cos\left(\frac{a}{2}k_y\right)}},$$

$$E_v = \frac{t_0 + t_1 \sqrt{1 + 4 \cos^2\left(\frac{a}{2}k_y\right) \pm 4 \cos\left(\frac{\sqrt{3}a}{2}k_x\right) \cos\left(\frac{a}{2}k_y\right)}}{1 + s_1 \sqrt{1 + 4 \cos^2\left(\frac{a}{2}k_y\right) \pm 4 \cos\left(\frac{\sqrt{3}a}{2}k_x\right) \cos\left(\frac{a}{2}k_y\right)}}, \quad (7)$$

of the rectangular four-atom unit cell tight-binding model are also plotted, with the on-site hopping parameter $t_0 = 0$ eV, the adjacent hopping parameter $t_1 = -3.033$ eV and the overlap integral of $s_1 = 0.129$. We can clearly see the Dirac cone with zero band gap at the Fermi level and the P point. The tight-binding model and DFT results closely agree with each other around there. Since we use the orthogonal cell, the π^* (ninth orbital) local band separation at the Γ point is smaller than for the case of the primitive cell due to band-folding effects. The discrepancy between the DFT and TB π^* energies

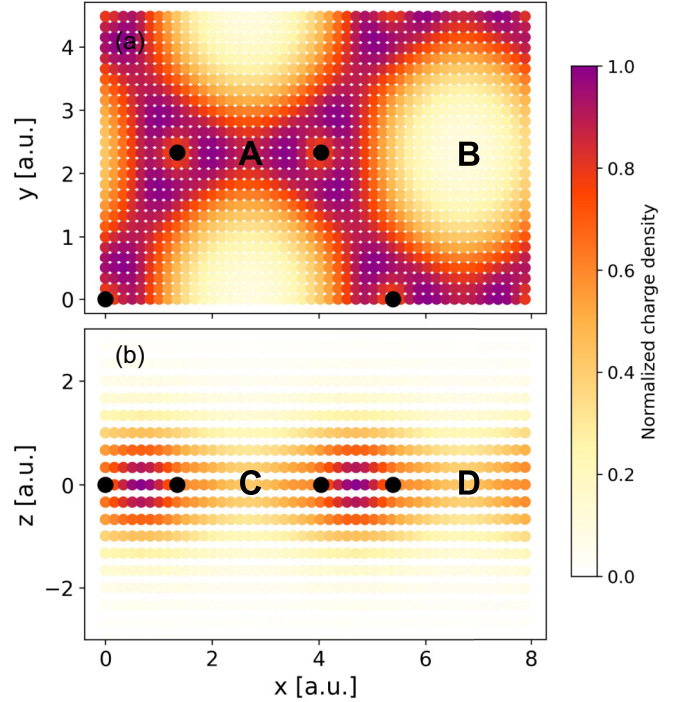


FIG. 2. (a) Top view and (b) side view of normalized charge density in real space projected onto the xy plane. A and B show position at the midpoint of a C-C bond and at the center of the hexagonal structure, respectively. C and D correspond to the midpoint of a C-C bond oriented at 0° and 60° with respect to the x axis, respectively. Black circles indicate carbon nuclei. All densities are normalized.

at Γ (TB: 6.17 eV; DFT: 3.99 eV) mainly originates from the absence of higher-order hopping terms in the TB model and the different treatment of the σ band (seventh orbital) contributions. In addition, near Γ , the σ^* band (tenth orbital) forms the shallow conduction-band minimum, whereas the π^* band lies at much higher energy. This implies that when electrons are excited into regions far away from the Dirac cone, the resulting carrier dynamics deviate from a two-level description and exhibit pronounced multiband character.

The charge density projected onto xy and zx planes in real space is displayed in Fig. 2. Electrons are concentrated in the hexagonal frame of graphene while the hexagonal center has the lowest charge density [Fig. 2(a)]. In Fig. 2(b), we see that the electron density is negligibly small at $z \sim \pm 2$ a.u., compared to that on the basal plane. This observation is consistent with the experimental results [50].

The linear response is obtained by time propagation after an impulsive momentum kick, realized by a steplike vector potential,

$$\mathbf{A}(t) = \begin{cases} -\mathbf{A}_0 & (t \geq 0), \\ 0 & (t < 0), \end{cases} \quad (8)$$

which corresponds to an impulsive electric field $\mathbf{E}(t) = \mathbf{A}_0 \delta(t)$. Since such an electric field has a constant power spectrum over all frequencies, the diagonal component of the

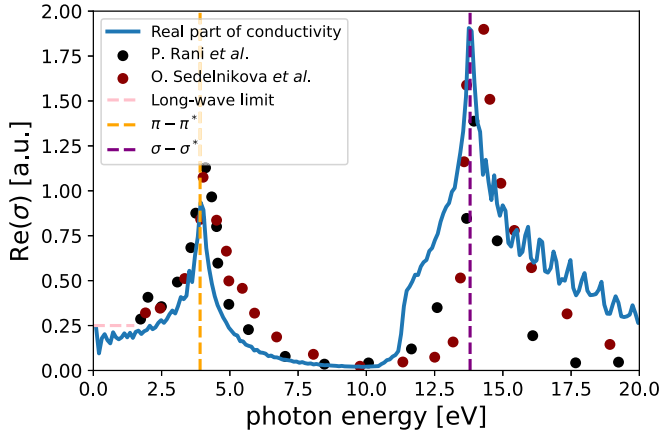


FIG. 3. Linear response of graphene computed with TDDFT. The blue line shows the real part of conductivity. Black and red dots are results from DFT calculations [51,52]. The horizontal pink dashed line indicates the zero-frequency conductivity (~ 0.25 a.u.). The orange and purple vertical lines mark the two peak positions, corresponding to the $\pi \rightarrow \pi^*$ and $\sigma \rightarrow \sigma^*$ transitions, respectively.

optical conductivity $\sigma_d(\omega)$ is calculated as

$$\sigma_d(\omega) = -\frac{e}{|\mathbf{A}_0|} I_d(\omega) \quad (d = x, y, z), \quad (9)$$

where $I_d(\omega)$ denotes the component along the polarization direction d of the temporal Fourier transform of the volume-averaged current density. The dielectric function $\varepsilon_d(\omega)$ is given by

$$\varepsilon_d(\omega) = 1 + 4\pi i \frac{\sigma_d(\omega)}{\omega}. \quad (10)$$

The off-diagonal components of the optical response tensors vanish due to the crystal symmetry of graphene.

Figure 3 shows the real part of thus calculated conductivity, in comparison with previous DFT calculations [51,52]. Two distinct absorption peaks are present, corresponding to transitions between the π and π^* bands and between the σ and σ^* bands. At the Γ point, the linear optical response is weak. In the long-wavelength limit ($\omega \rightarrow 0$), the conductivity approaches approximately 0.25 a.u., consistent with previous studies and experiments [53,54]. This agreement demonstrates the reliability of our TDDFT approach for linear optical properties.

IV. RESULTS AND DISCUSSION

We specifically consider a few-cycle carrier-envelope phase (CEP) laser pulse with the following form (see Fig. 4):

$$E(t) = F \sin^4\left(\frac{\pi t}{\tau}\right) \cos(\omega t + \varphi_{\text{CEP}}), \quad (11)$$

where we vary the peak field amplitude F up to 4 V/nm, within the reported damage threshold [55]. Following the previous studies [12,14,20], the pulse foot-to-foot duration is $\tau = 21$ fs, which corresponds to the full-width-at-half-maximum (FWHM) duration of 5.5 fs, and $\omega = 2\pi \times 0.375$ PHz (1.55 eV photon energy, 800 nm wavelength). A $\sin^4$ envelope is chosen to suppress high-frequency tails of the pulse

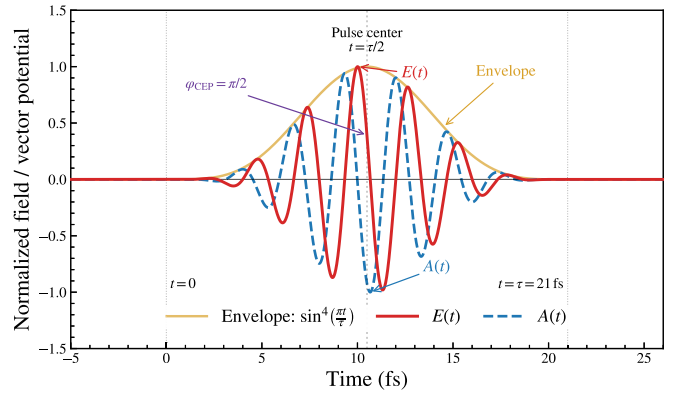


FIG. 4. Temporal profile of the laser field used in this work. The electric field $E(t)$ (red solid line) and the corresponding vector potential $A(t)$ (blue dashed line) are shown together with the envelope function (yellow), defined as $\sin^4(\pi t/\tau)$. The carrier-envelope phase is set to $\varphi_{\text{CEP}} = \pi/2$, and the pulse duration is $\tau = 21$ fs. The pulse center is indicated at $t = \tau/2$.

[33]. A representative carrier-envelope phase $\varphi_{\text{CEP}} = \pi/2$ is selected. The laser polarization is along a graphene C–C bond (taken as the $+x$ direction in our coordinates; see Fig. 2). The time step is set to be 0.02 a.u., and the total simulation time T is 100 fs.

Let us first focus on electron excitation in the reciprocal space after the laser pulse ($t = 21$ fs). Excited-state analysis is carried out by projecting the time-dependent Kohn-Sham orbitals onto Houston functions [56]. Figures 5(a)–5(h) show the calculated band occupation of varying laser field intensity. At low intensity, single-photon and then two-photon resonant excitation lead to carrier distribution symmetric with respect to the k_y axis, i.e., $k_x = 0$ [Figs. 5(a)–5(c)]. As the laser intensity increases, the region between the contour lines of single- and two-photon excitation starts to be populated [Fig. 5(d)], indicative of tunneling excitation, and asymmetry in excited carrier distribution appears in that region [Figs. 5(e) and 5(f)], consistent with the observations of Refs. [12,20].

At even stronger fields (3–4 V/nm) [Figs. 5(g) and 5(h)], higher-energy regions between the multiphoton resonant excitation contours are filled [see also Fig. 5(i)], which indicates increase in tunneling excitation. Simultaneously, the asymmetry extends to the higher-energy region. Figure 5(j) shows the asymmetry in carrier distribution between $k_x > 0$ and $k_x < 0$ defined as $\mathcal{A}(k_x) = \sum_{k_y > 0} [P(k_x, k_y) - P(-k_x, k_y)]$. At 4 V/nm, the asymmetry $\mathcal{A}(k_x)$ around $k_x = -0.02$ near the Dirac cone and $k_x = -0.13$ far from there, which contribute to the current in the negative direction, surpasses the positive component around $k_x = 0.04$, which leads to the negative total current.

In addition to the intraband residual current $I_i^{\text{intra-rd}}$ on band i ,

$$I_i^{\text{intra-rd}} = \begin{cases} \int_{\text{BZ}} \frac{\partial E_i}{\partial \mathbf{k}} \rho_i d\mathbf{k}, & i \in \text{conduction}, \\ \int_{\text{BZ}} \frac{\partial E_i}{\partial \mathbf{k}} (2 - \rho_i) d\mathbf{k}, & i \in \text{valence}, \end{cases} \quad (12)$$

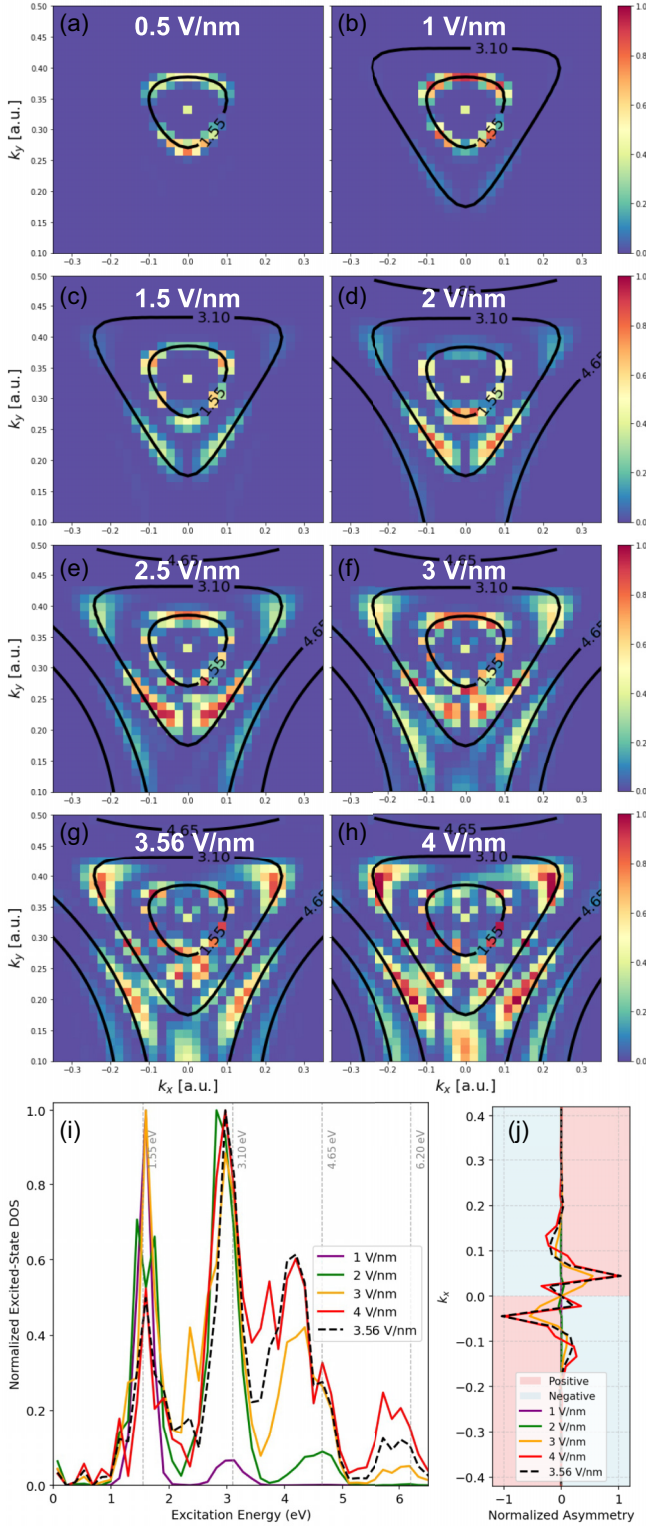


FIG. 5. The excited population $P(k_x, k_y)$ in the reciprocal space after the laser fields. (a)–(h) Population near Dirac cone on the lowest conduction orbital (ninth orbital) with contour lines at multiples of the photon energy under the peak field amplitudes from 0.5 to 4 V/nm. (i) Normalized density of states for the excited population on conduction band. (j) excited carrier distribution asymmetry $\mathcal{A}(k_x)$ normalized to its maximum value on 4 V/nm as a function of k_x . Positive (red) and negative (blue) background shadings indicate the sign of the asymmetry.

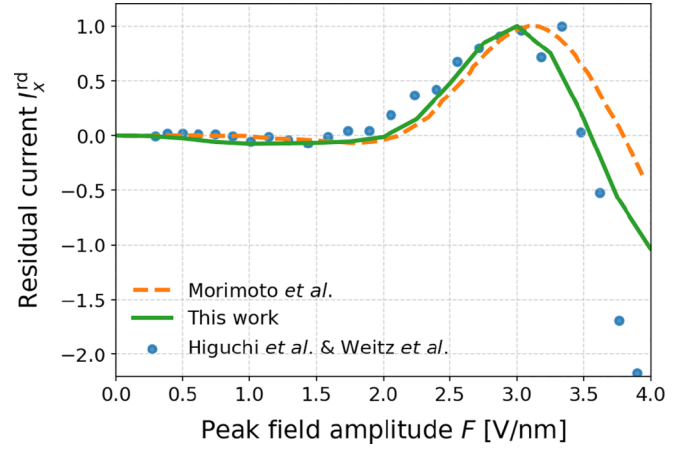


FIG. 6. Total residual current vs peak field amplitude F . Green solid line: our TDDFT results (this work); orange dashed: calculated results by Morimoto *et al.* [20]; blue circles: experimental data from Higuchi *et al.* [12] (≤ 3 V/nm) extended by Weitz *et al.* [14]. The plots are normalized by the maximum of the residual current.

we also evaluate the residual current by averaging the current after the pulse, which directly corresponds to the experimental measurement, as

$$I_d^{\text{rd}} = \frac{1}{T - \tau} \int_{\tau}^T I_d(t) dt, \quad (13)$$

where E_i and ρ_i denote the band energy and the electron occupation after the pulse [Eq. (6)], respectively, of band i . I_d^{rd} is the average residual current density in the d direction, and I_d is the component in the d direction of the volume average of current density $\mathbf{I}$. Similarly, the residual current distribution projected on xy and zx planes can be expressed as $\mathbf{J}_{2D-z}^{\text{rd}}(x, y) = \frac{1}{T - \tau} \int_{\tau}^T \mathbf{J}_{2D-z}(x, y, t) dt$ and $\mathbf{J}_{2D-y}^{\text{rd}}(x, z) = \frac{1}{T - \tau} \int_{\tau}^T \mathbf{J}_{2D-y}(x, z, t) dt$, respectively.

We have confirmed that our simulation time (100 fs) is sufficiently long that the contribution from the oscillating interband polarization is removed after the temporal integration in Eq. (13). As a result, the time-average residual current [Eq. (13)] becomes practically the same as the sum of the intraband residual currents [Eq. (12)] over all the orbitals, i.e., the remaining direct current (Fig. 7).

In Fig. 6 we show the residual current calculated with Eq. (13) as a function of field amplitude. Our TDDFT results agree very well with those calculated by the two-level SBE with the tight-binding model [20] and the experimental data [12,14]. In the strong-field regime ($F \geq 3$ V/nm), especially, our first-principles results lie closer to the experimental values than the SBE results do, which indicates that TDDFT describes nonlinear dynamics of highly excited electrons more accurately through the time-dependent effective Hamiltonian. The direction of the residual current is reversed at $F \approx 2$ and 3.56 V/nm, and the residual current reaches its maximum around 3 V/nm.

Figure 7 breaks down the intraband residual current by orbitals evaluated as Eq. (12). We find that the lowest conduction band (ninth orbital) and the highest valence band (eighth orbital) dominate the residual current. The contributions from

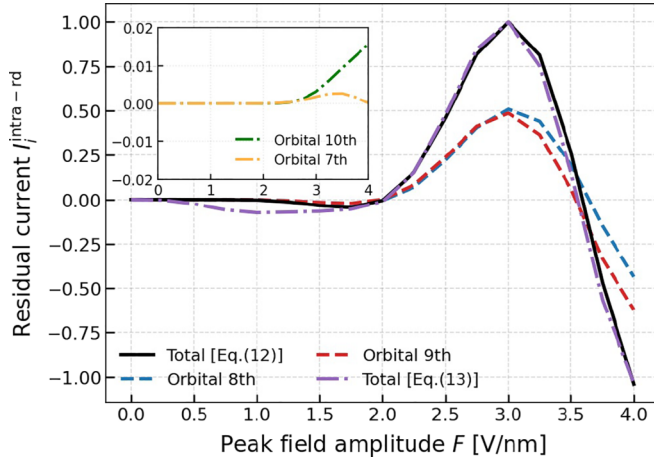


FIG. 7. Normalized intraband residual current contribution from each orbital of graphene. Black solid line: total current calculated by summing the currents evaluated as Eq. (12) over all the bands; purple dashed-dotted line: the total current calculated via Eq. (13). The contributions from the eighth (valence top, blue line) and ninth (conduction bottom, red line) orbitals are also plotted. Inset: contributions from the seventh (orange line) and tenth (green line) orbitals.

these two orbitals are nearly equal (symmetric) up to ≈ 3 V/nm field strength, indicating that excitation is confined within the Dirac cone. On the other hand, once the peak field exceeds 3 V/nm, some of electrons are driven into the energy regions beyond the pure Dirac cone, where the eighth orbital and the ninth orbital are not symmetric [also see Figs. 5(f)–5(h)], leading to the difference between the valence-band and conduction-band currents. At the same time, occupation of higher bands (for instance, the tenth orbital) becomes non-negligible, indicating that, when driven by strong fields, bands beyond the simple two-level model begin to contribute to the current.

In Fig. 8 we visualize the residual current distribution after the pulse in the real space, projected onto the graphene plane. At $F = 3$ V/nm, at which the total residual current reaches its maximum (see Fig. 6), the current flows along the hexagonal atomic framework, i.e., along the C–C bonds, especially along the laser polarization [Fig. 8(a)]. At $F = 3.56$ V/nm, where the total residual current is nearly zero; the microscopic local current does not vanish, but a return current appears at the center of each hexagon, forming two ring currents within a single hexagon. These observations are consistent with our previous study [20].

Let us further examine the residual current density at two representative positions: point A [atomic unit coordinates (2.68, 2.32)] at the midpoint of a C–C bond (aligned with the field polarization) and point B [atomic unit coordinates (6.71, 2.32)] at the center of a hexagon (see Fig. 8). Figure 9 plots the residual current density at these points as a function of peak field amplitude. Overall, the current densities at points A and B follow the same trend as the total residual current. If we look more closely, however, the local current at point B reverses its direction at a lower field (~ 3.3 V/nm) than the current at point A (~ 3.6 V/nm). As a consequence, the currents at the two points are in opposite directions at $F = 3.56$ V/nm, which leads to the ring currents we observed above in Fig. 8(b).

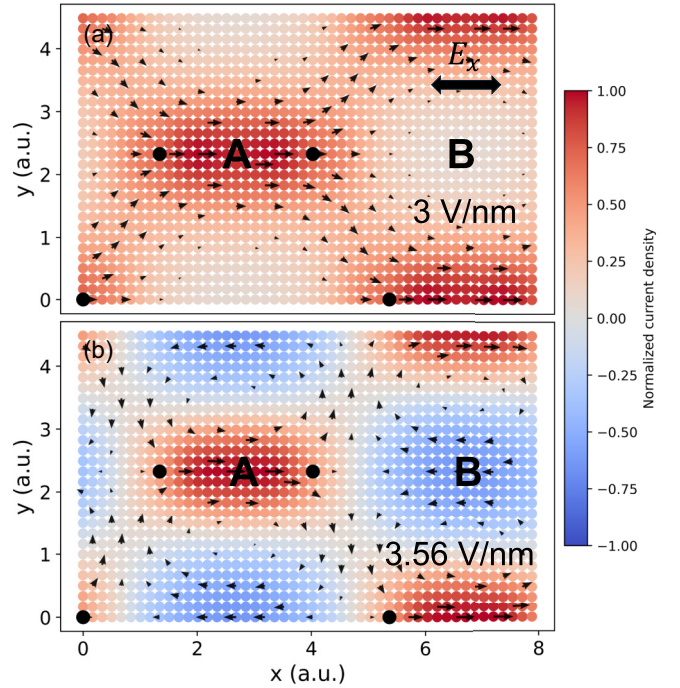


FIG. 8. Normalized real-space distribution of the x component of the local residual current projected onto the xy plane. (a) Peak field amplitude of 3 V/nm. (b) Peak field amplitude of 3.56 V/nm. Black arrows indicate the projected local current vectors, and black circles mark the carbon nuclei positions. (The nonlocal pseudopotential contribution to the current is not included in these plots.)

So far, we have discussed the current distribution projected onto the graphene plane. We now turn to the three-dimensional current distribution. Figure 10 presents the current density distribution projected onto the zx plane for $F = 3.56$ V/nm, which corresponds to Fig. 8(b). Out-of-plane circulating current is observed, with a positive current near the basal plane and an oppositely directed current emerging slightly away from the plane in Figs. 10(a) and 10(b). To clearly elucidate the 3D location of the return currents appearing in Fig. 8(b), we present in Fig. 10(c) the zx -plane slice of the three-dimensional residual current density, $J^{\text{rd}}_{zx}(x, z; y_0) = \frac{1}{T-t} \int_t^T \mathbf{J}(x, y_0, z, t) dt$, evaluated at $y_0 = 2.32$ a.u., which corresponds to the line connecting points A and B in Fig. 8(b). The opposite current components are visible, both slightly shifted away from the graphene basal plane but with a subtle height difference at the z axis. This trend is consistent with the distribution in Fig. 10(b), and corroborates that the current at point B in Fig. 8 resides at a comparatively more out-of-plane position. Notably, virtually no current flows exactly in the graphene plane ($z = 0$). Instead, the current is mainly concentrated approximately 1 a.u. above and below it, which coincides very well with the positions of the ground-state π and π^* orbitals [Figs. 10(d) and 10(e)]. This supports the electron dynamics being driven predominantly in the π and π^* orbitals, as was assumed in the previous studies [20], and the necessary degree of freedom for out-of-plane electron motion is also provided. From the 3D view, the ring current reported in the previous work [20] is neither confined to the graphene plane nor parallel to it,

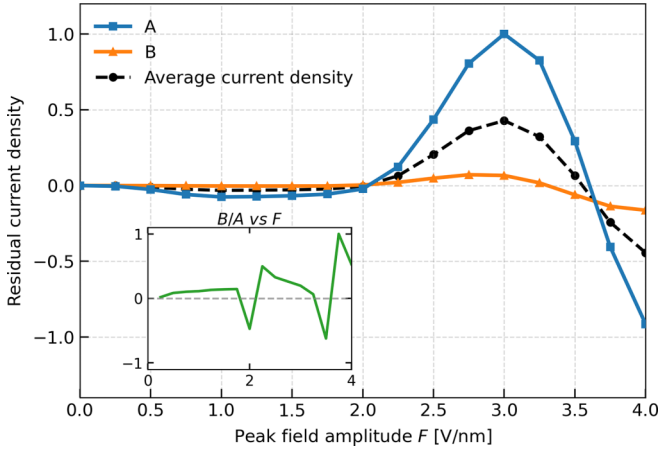


FIG. 9. Residual current density at two representative points in real space as a function of peak field amplitude: point A (midpoint of a C–C bond, along polarization) and point B (center of a hexagon). The blue curve is the current density at point A and the orange curve at point B. The black dashed line indicates the total residual current density (from Fig. 6) for reference. Inset: Normalized ratio of the residual current density at B and A as a function of the peak field amplitude F . The ratio takes negative values around $F \approx 2$ V/nm and $F \approx 3.56$ V/nm.

but forms a rotating 3D circulation loop with pronounced out-of-plane height difference. Furthermore, as demonstrated in Appendix A, the emergence of loop currents can be understood from a general perspective based on the Helmholtz decomposition. When current reversal occurs, the residual current becomes divergence-free and carries no net DC component, which constrains the current to a purely solenoidal form. Under this condition, the current must form closed circulation loops in real space. In this study, the center of the carbon hexagon (point B in Fig. 8) naturally serves as the return path of the current, completing the loop structure. As shown in Fig. 11, the out-of-plane electron density distributions at the hexagon center and the C–C bond midpoint exhibit clear differences. In particular, the electron density at the hexagon center is more localized at positions further away from the graphene basal plane. A pronounced shift in the peak positions is observed, with $\Delta z = 0.459$ a.u. This behavior is consistent with the spatial distribution of the residual current shown in Figs. 10(a) and 10(c), and indicates that the formation of the loop current is strongly governed by the real-space structure of the π orbitals. Therefore, the out-of-plane loop current can be understood as a consequence of CEP-induced excitation symmetry breaking in reciprocal space combined with the dominance of π and π^* orbitals that support out-of-plane motion.

V. CONCLUSIONS

We have performed first-principles real-time TDDFT simulations of graphene subject to intense few-cycle laser pulses. The calculated residual currents excellently reproduce the experimentally observed field-strength dependence, including the reversal of current direction. By resolving the contributions from individual orbitals, we have verified that the two-level description [12,20] around the Dirac cone provides

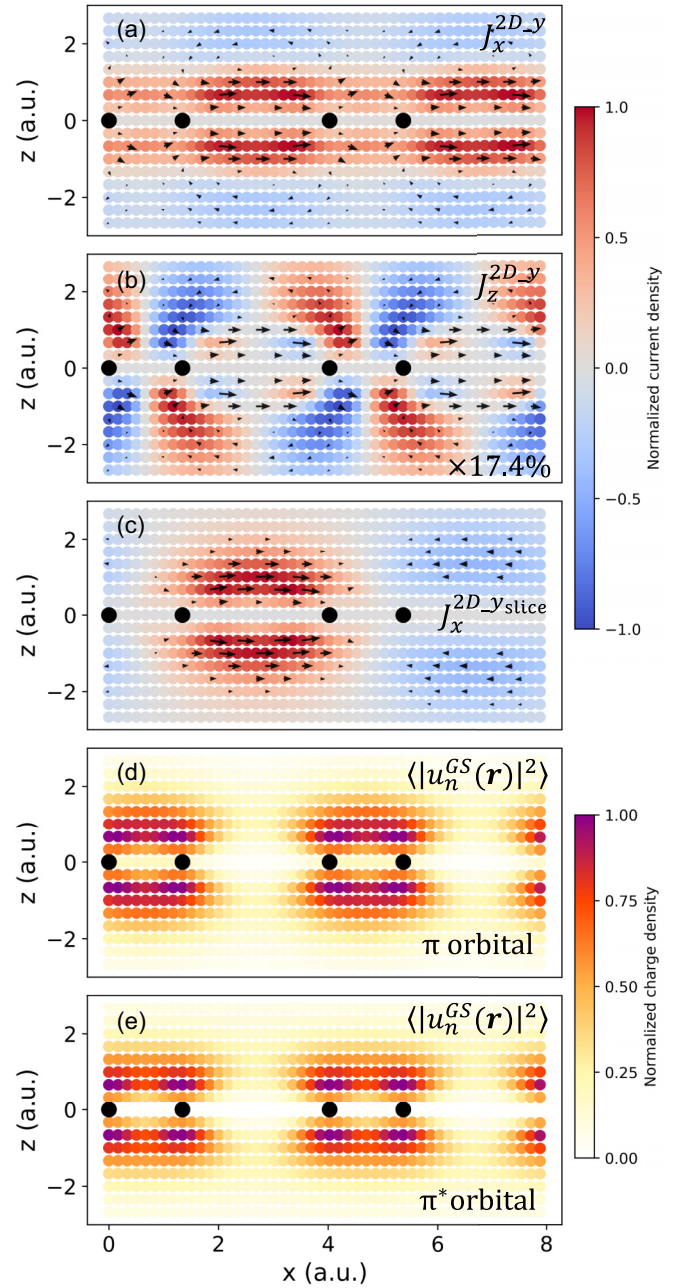


FIG. 10. Real-space distribution of the local residual current and ground-state orbital electron density projected onto the zx plane. (a) Color map: normalized real-space distribution of the x component of the local residual current projected onto the zx plane for a peak field amplitude of 3.56 V/nm. (b) Color map: normalized real-space distribution of the z component of the local residual current projected onto the zx plane under the same field. In panels (a) and (b), the black arrows represent the same total current vectors, while the color maps are used to visualize different components of the current density [x component in (a) and z component in (b)]. The scaling factor ($\times 17.4\%$) indicates that the color scale in (b) is normalized relative to the maximum value in (a). (c) Normalized cross-sectional slice at $y_0 = 2.32$ a.u. of (a) [aligned with the line connecting points A and B in Fig. 8(b)]. (d),(e) The normalized ground-state charge densities of π and π^* Bloch orbitals projected onto the zx plane. Black circles mark the carbon nuclei positions.

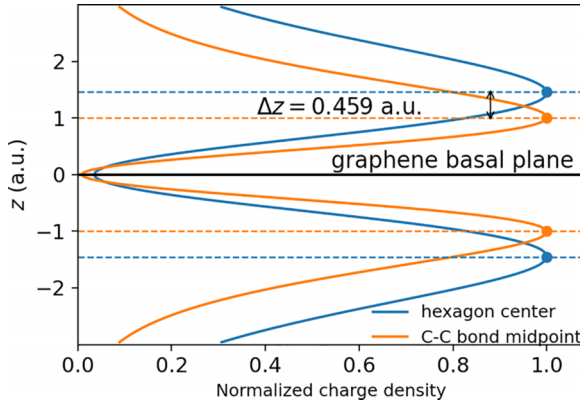


FIG. 11. Normalized π -orbital charge density along the out-of-plane (z) direction for two representative in-plane positions: the C-C bond midpoint (point A, orange) and the hexagon center (point B, blue). The vertical solid line indicates the graphene basal plane ($z = 0$). Dashed lines mark the positions of the maximum charge density for each curve, and symbols highlight the peak values. A clear spatial shift between the peak positions is observed, with $\Delta z = 0.459$ a.u., as indicated by the double arrow. The charge density profiles are smoothed using Fourier interpolation along the z direction.

an adequate picture at moderate fields, while additional bands start to be relevant in the strong-field regime.

Our *ab initio* results confirm and extend the scenario proposed in our previous work [20], in which current reversal arises from the cancellation of microscopic flows along the C-C bonds and at the center of the hexagon. The present TDDFT analysis not only validates this mechanism from *ab initio* perspective, but also reveals that the dominant residual current is carried by π and π^* orbitals slightly above and below the basal plane, rather than strictly within it. Further, rather than being confined or parallel to the graphene plane, the out-of-plane projection reveals that this vortexlike current forms a rotating three-dimensional circulation loop. These findings connect the reciprocal- and three-dimensional real-space viewpoints of strong-field dynamics in graphene. We note that recent theoretical studies suggest that real-space current distributions can be experimentally accessed [57,58]. Looking forward, we expect that the present real-space framework can be extended to more complex two-dimensional materials, where reduced symmetry, finite band gaps, and strong anisotropy may give rise to even richer out-of-plane electron dynamics and current topologies under strong laser fields. Our work thus provides microscopic insight useful for the development of ultrafast optoelectronics and light-wave electronics with a more realistic interpretation beyond 2D descriptions.

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The authors declare no competing financial interest.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: HELMHOLTZ DECOMPOSITION OF THE RESIDUAL CURRENT

Starting from the continuity equation,

$$\frac{\partial \rho(\mathbf{r}, t)}{\partial t} + \nabla \cdot \mathbf{J}(\mathbf{r}, t) = 0, \quad (\text{A1})$$

we average it over the after-pulse time interval (τ, T):

$$\frac{1}{T - \tau} \int_{\tau}^T \left[\frac{\partial \rho(\mathbf{r}, t)}{\partial t} + \nabla \cdot \mathbf{J}(\mathbf{r}, t) \right] dt = 0. \quad (\text{A2})$$

This gives

$$\frac{\rho(\mathbf{r}, T) - \rho(\mathbf{r}, \tau)}{T - \tau} + \nabla \cdot \mathbf{J}_{\text{rd}}(\mathbf{r}) = 0, \quad (\text{A3})$$

where

$$\mathbf{J}_{\text{rd}}(\mathbf{r}) = \frac{1}{T - \tau} \int_{\tau}^T \mathbf{J}(\mathbf{r}, t) dt \quad (\text{A4})$$

is the residual current density. Assuming a sufficiently long time-averaging window such that the system reaches a steady state, we have

$$\frac{\rho(\mathbf{r}, T) - \rho(\mathbf{r}, \tau)}{T - \tau} = 0, \quad (\text{A5})$$

which leads to

$$\nabla \cdot \mathbf{J}_{\text{rd}}(\mathbf{r}) = 0. \quad (\text{A6})$$

For a periodic vector field, the Helmholtz-Hodge decomposition can be written as [59]

$$\mathbf{J}_{\text{rd}}(\mathbf{r}) = \langle \mathbf{J}_{\text{rd}} \rangle + \nabla \chi(\mathbf{r}) + \nabla \times \mathbf{C}(\mathbf{r}), \quad (\text{A7})$$

where $\chi(\mathbf{r})$ is the scalar potential associated with the longitudinal part, and $\mathbf{C}(\mathbf{r})$ is a vector potential associated with

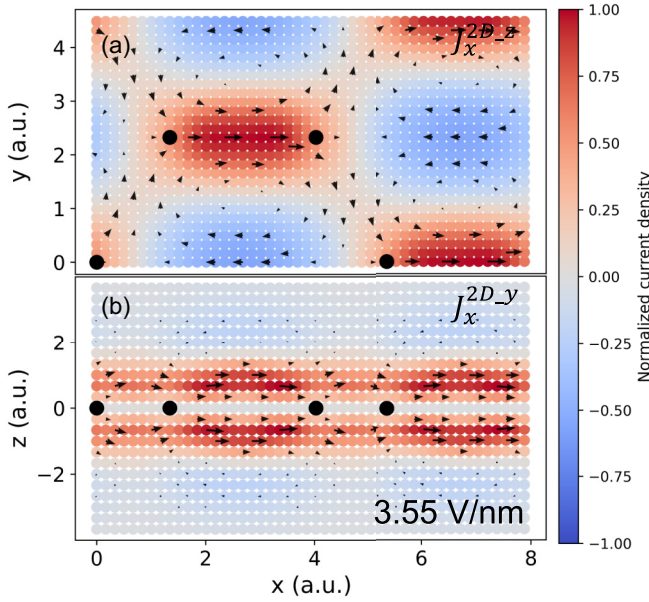


FIG. 12. Normalized real-space distribution of the x component of the local residual current projected onto the xy plane (a) and the zx plane (b). Here, the envelope of the electric field is changed to $\sin^2$.

the transverse part. $\langle \mathbf{J}_{\text{rd}} \rangle$ denotes the cell-averaged residual current, defined as

$$\langle \mathbf{J}_{\text{rd}} \rangle = \frac{1}{\Omega} \int_{\Omega} \mathbf{J}_{\text{rd}}(\mathbf{r}) d\mathbf{r}, \quad (\text{A8})$$

where Ω is the volume of the periodic unit cell. Taking the divergence of both sides gives

$$\nabla \cdot \mathbf{J}_{\text{rd}}(\mathbf{r}) = \nabla^2 \chi(\mathbf{r}). \quad (\text{A9})$$

Using $\nabla \cdot \mathbf{J}_{\text{rd}}(\mathbf{r}) = 0$, we obtain

$$\nabla^2 \chi(\mathbf{r}) = 0. \quad (\text{A10})$$

Under periodic boundary conditions, the only periodic solution of $\nabla^2 \chi(\mathbf{r}) = 0$ is $\chi(\mathbf{r}) = \text{const}$, so that

$$\nabla \chi(\mathbf{r}) = 0. \quad (\text{A11})$$

Furthermore, at the current-reversal point,

$$\langle \mathbf{J}_{\text{rd}} \rangle = 0. \quad (\text{A12})$$

Hence,

$$\mathbf{J}_{\text{rd}}(\mathbf{r}) = \nabla \times \mathbf{C}(\mathbf{r}). \quad (\text{A13})$$

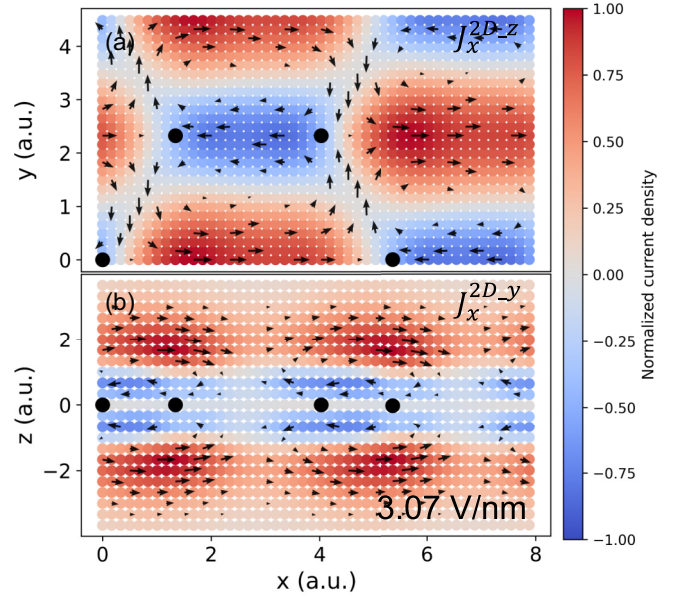


FIG. 13. Normalized real-space distribution of the x component of the local residual current projected onto the xy plane (a) and the zx plane (b). Here, the pulse duration of the electric field is changed to 18.7 fs.

Therefore, the residual current is solenoidal and is expected to form closed circulating flow patterns in real space, consistent with the observed loop-current pattern.

APPENDIX B: ROBUSTNESS OF THE 3D CIRCULATION LOOP

To verify the robustness of the 3D circulation loop, we examine the dependence on several representative parameters of the laser field. The envelope function is changed from $\sin^4$ to $\sin^2$ in Eq. (11). The corresponding peak field amplitude at which the current direction reversal occurs shifts slightly to 3.55 V/nm, as shown in Fig. 12. Similarly, when a shorter pulse duration is used ($\tau = 18.7$ fs, corresponding to a FWHM of 4.89 fs), the transition occurs at a field amplitude of 3.07 V/nm, as shown in Fig. 13. The results indicate that changes in the laser parameters can shift the field strength at which current reversal occurs. However, the key qualitative feature—the emergence of loop current patterns—remains robust.

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