Resonance effect of attosecond transient absorption spectroscopy in graphene

Fulong Dong, Jie Liu∗
Graduate School, China Academy of Engineering Physics, Beijing 100193, China
(Dated: July 14, 2022)

We investigate the attosecond transient absorption spectroscopy (ATAS) in graphene numerically and analytically. In contrast to the velocity gauge, we process the mechanism analysis based on the numerical results of the density-matrix equations of the length gauge. We simplify the band structure of graphene to a two-electron model in a one-dimensional band structure, based on which our analytical results are qualitatively consistent with the numerical results. Inspired by the analytical theories, we divide the fishbone structure of ATAS into the zeroth-order and first-order stripes, and clearly define the opening angle of the V-shaped structure of the first-order stripes. Our analytical results reveal that the resonance effect is the main generation mechanism of absorption spectroscopy. The periodic variation of the first-order stripes at twice the infrared laser frequency can be clearly explained with our analytical theory. Our analysis reveals the underlying mechanism that the opening angles of V-shaped structure increase with increasing the infrared laser frequency. The analytical results indicate that the first-order stripe is brighter for the smaller effective mass of electrons in Van Hove singularities. These results may be generalized to other periodic materials and shed light on the complex generation mechanism of the ATAS.

I. INTRODUCTION

Recent progress in laser technology has made it possible to produce isolated attosecond pulses [1, 2], which accelerates the study of electron dynamics on ultrashort time scales [3]. One of the promising approaches to investigate subfemtosecond dynamics in electronic systems is attosecond transient-absorption spectroscopy (ATAS) [4, 5], which offers an all-optical approach to light-matter interactions, with the high temporal resolution of the attosecond pulse and the high energy resolution characteristic of absorption spectroscopy. Over the past few years, ATAS has been used to study electronic dynamics of atoms and molecules [6–13], in which the quantum interference [14] and a nonresonant AC Stark shift [15] and a resonant Autler-Townes splitting [16] phenomena have been directly observed.

Recently, the attosecond time-resolved technology has been applied to solids [6, 17–22] and some two-dimensional materials [23–24]. For these periodic materials, the special fishbone structure presented in the ATAS has attracted much attention [18, 24]. The previous studies have attributed the generation mechanism of ATAS to the dynamical Franz-Keldysh effect which is related to the intraband motion of electrons [18, 19, 22, 25, 26], however, the explanation for microscopic origin of the absorption spectroscopy is still unclear.

Graphene is a simple but specific two-dimensional material, in which there are only two carbon atoms per unit cell and the atoms are orderly arranged in a periodic hexagonal lattice. It has important applications because of its unusual optical properties [27, 28]. The ATAS of graphene are of natural concern, and the corresponding computational method have been proposed in Ref. [23].

In our work, we reproduce the ATAS of graphene by using density-matrix equations in length and velocity gauges. The absorption spectroscopy near K point originates from interband transition of electrons [28], and we mainly pay attention to ATAS around M and Γ points of graphene. We propose a simple two-electron model to replace the two-dimensional four-band model of graphene, which is feasible because the numerical results of both methods are qualitatively consistent. In particular, based on the two-electron model, we have derived analytical results of absorption spectroscopy, which agree well with the numerical results. According to the analytical results, we can further divide fishbone structure of the absorption spectroscopy into zeroth-order and first-order stripes. Our analytical results reveal that the opening angles of V-shaped structure increases with the IR laser frequency as observed in [18]. Our analysis explains the phenomena that the periodic variation of the first-order stripes at twice the laser frequency [18, 24]. In addition, the analytical results indicate that the first-order stripe is brighter for the smaller effective mass of electrons in Van Hove singularities.

This paper is organized as follows. We describe our calculation models of the ATAS for graphene in Sec. II, Section III presents our main numerical results, and the analytical explanation of the ATAS are discussed in Sec. IV. Finally, Sec. V presents our conclusion. Throughout the paper the atomic units are used if not specified.

II. MODEL

Graphene is a two-dimensional single layer of carbon atoms arranged in a honeycomb lattice [23], and its reciprocal space is a hexagonal lattice structure. In this work, we consider four energy bands of graphene consisting of two core bands (g1 and g2), which arise from the two 1s orbitals of the two carbon atoms in a unit cell, as well as the valence (v) and conduction (c) bands, which arise from the π orbitals orthogonal to the monolayer. The two core bands are degenerate and have constant energy of −280 eV over the k space.
The tight-binding Hamiltonian $H_0$ arising from π orbitals in graphene has the form
\[
H_0 = \begin{pmatrix} 0 & \gamma_0 f(k) \\ \gamma_0 f^*(k) & 0 \end{pmatrix}
\]
in which electrons can only hop to nearest-neighbor atoms with hopping energy $\gamma_0 = 0.1$ a.u. and $f(k) = e^{ik_d/2} - e^{-ik_d/2}$. The diagonalization of $H_0$ matrix can yield energy eigenvalues, which describe the dispersion relation of $v$ and $c$ bands $\varepsilon_v(k) = -\varepsilon_c(k) = \gamma_0 f(k) = \gamma_0 \sqrt{3 + 2 \cos(3k_y d) + 4 \cos(3k_x d)/2} \cos(3k_y d)/2$.

### A. length gauge

We numerically simulate the ATAS of graphene in the length gauge by using the density-matrix equations in which the four energy bands have been included.\[23\] Within the dipole approximation, it reads
\[ i \frac{\partial}{\partial t} \rho_{mn}(k, t, t_d) = [\varepsilon_n(k) - \varepsilon_m(k) - i\Gamma_{mn}] \rho_{mn}(k, t, t_d) 
+ iE_I(t, t_d) \frac{\partial}{\partial k} \rho_{mn}(k, t, t_d) 
+ [E_I(t, t_d) + E_X(t)] \cdot [\hat{D}, \rho]_{mn} \]

(1)

where $\Gamma_{mn}$ are the relaxation parameters. The core-hole lifetime is set to 6.1 fs (corresponding relaxation parameters are 0.04 a.u.) and other relaxation parameters are zero.

For the two-dimensional four-band structure of graphene in Fig. 1(b), the interband transitions includes $D_{\alpha\iota}(k) = \iota(u_{\alpha\iota}(r)|\nabla|u_{\alpha\iota}(r))$ where $u_{\alpha\iota}(r)$ is the periodic part of the Bloch wavefunction for the conduction (valence) band of graphene.\[23\] [30].

At the initial moment $t = 0$, electrons populate in two core bands and valence band, thus $\rho_{\alpha\iota}(k, t = 0, t_d) = \rho_{\alpha\iota}(k, t = 0, t_d) = \rho_{\alpha\iota}(k, t = 0, t_d) = 1$ and other terms of the density matrix elements are zero.

$E_X(t) = E_X f_X(t) \cos(\omega_X t) \mathbf{e}_z$ is the electric field of X-ray pulse, in which $f_X(t) = e^{-4(\ln 2/t/\pi)^2}$ has a full width at half maximum (FWHM) of $\tau_X = 80$ as and laser intensity is $1 \times 10^{10}$ W/cm$^2$. The frequency of X-ray is $\omega_X = 280$ eV, which is equal to energy gap between Fermi energy and the core bands. $\mathbf{e}_z$ is the polarization direction which is perpendicular to the graphene monolayer.

$A_I(t, t_d) = A_{I0} f_I(t + t_d) \cos(\omega_I t + \omega_I t_d) \mathbf{e}$ is the vector potential of an infrared (IR) laser field. $f_I(t) = \cos^2(\omega_I t/2n)$ is an envelope with $n = 10$ and the amplitude of $A_{I0}$ corresponds to laser intensity of $1 \times 10^{11}$ W/cm$^2$. $\omega_I$ is the frequency of the IR laser field corresponding to the wavelength of $\lambda = 3000$ nm. $\mathbf{e}$ is the unit vector along the the $\Gamma - M$ direction of graphene. $t_d = t_X - t_{IR}$ is time delay between X-ray pulse and IR laser field as shown in Fig. 1(a). The electric field of the IR laser is calculated by $\hat{E}_I(t, t_d) = -\partial A_I(t, t_d)/\partial t$.

The X-ray response intensity is calculated by\[3\]
\[ S(\omega, t_d) = 2 \text{Im} [\hat{\mu}(\omega, t_d) \hat{E}_X(\omega)], \]

(2)

where $\hat{E}_X(\omega)$ is Fourier transform of the electric field of the X-ray pulse, and $\hat{\mu}(\omega, t_d)$ is Fourier transform of the dipole response of the system which calculated by
\[ \mu(t, t_d) = \sum_{\mathbf{k}} \sum_{i, g} [r_z \rho_{ig}(k, t, t_d) + c.c.], \]

(3)

where $g$ represents the core bands ($g_1$ or $g_2$) and $i$ represents $v$ or $c$ band. $r_z = \int dz \psi_{1s}(z) \psi_{2p_z}(z) = 0.041 \lambda_{\text{IR}}^2$, where $\psi_{1s}(z)$ and $\psi_{2p_z}(z)$ are the wavefunctions of 1$s$ and 2$p_z$ orbits along the $z$ direction.

The ATAS can be calculated by
\[ \Delta S(\omega, t_d) = S(\omega, t_d) - S^X(\omega), \]

(4)

where $S^X(\omega)$ and $S(\omega, t_d)$ are X-ray response intensities of graphene without and with IR laser field of $t_d$, respectively.

### B. velocity gauge

We also calculate the ATAS of graphene in the velocity gauge. Within the dipole approximation, the corresponding density-matrix equations are
\[ i \frac{\partial}{\partial t} \rho_{mn}(k, t, t_d) = |\varepsilon_n(k) - \varepsilon_m(k) - i\Gamma_{mn}] \rho_{mn}(k, t, t_d) 
+ [A_I(t, t_d) + A_X(t)] \cdot [\hat{D}, \rho]_{mn}, \]

(5)

where $A_X(t) = -\int E_X(t') dt'$ is the electric field of X-ray pulse. The momentum matrix $\hat{p}(k)$ consists of the intraband dipole elements $p_{\alpha\iota}(k) = \nabla \varepsilon_{\alpha\iota}(k) = -p_{\alpha\iota}(k)$, $p_{g_1g_1}(k) = p_{g_2g_2}(k) = 0$ and the interband dipole elements $p_{\bar{m}n}(k) = i(\varepsilon_{m}(k) - \varepsilon_{\bar{m}}(k)) D_{\bar{m}n}(k)$ where $\bar{m}$ represent the index of the band that is not the $m$ band. The other parameters and the calculation methods of the ATAS are the same as the length gauge.
III. THE NUMERICAL RESULTS OF ATAS

The top panel of Fig. 2(a) shows the X-ray response intensities of graphene, which are calculated by Eq. (2) based on the density-matrix equations in the length gauge. The corresponding ATAS evaluated by Eq. (4) is presented in the bottom panel of Fig. 2(a). Figures 2(b) show the ATAS as a function of time delay in units of optical cycles of IR laser. The purple and orange lines mark the centres of the zeroth and first order stripes, respectively. As the time delay changes, the first-order stripes oscillate at twice the IR laser frequency while the zero-order stripes do not exhibit similar characteristics. The black solid arrows indicate the V-shaped structure of the first order stripes and \( \alpha \) defines the opening angle. Figures 2(c) and 2(d) are the same as Figs. 2(a) and 2(b), respectively, but the results are calculated within the velocity gauge framework.

The ATAS of graphene mainly consists of three regions which are around the K point (at 0 eV), M point (near 2.72 eV), and \( \Gamma \) point (about 8.16 eV). For the both gauges, one can obtain that the ATAS results near K and M points are qualitatively consistent, however, there exist observable difference for that around \( \Gamma \) point. The discrepancy might arise from that the velocity gauge requires the solution of dynamics equations on the basis including many virtual states (conduction and valence bands), which are not physically occupied. Therefore, we investigate the underlying mechanism of the ATAS based on the numerical results of the length gauge in the following.

We first study the effects of intraband and interband transitions on the ATAS, respectively. In the two-dimensional four-band model of Fig. 1(b), we block the interband transition between conduction and valence bands as shown in Fig. 3(a), i.e., we artificially set \( D_{cv}(k) \) to be 0 in the numerical calculation. The corresponding ATAS is presented in Fig. 3(b). By comparing Fig. 3(b) and Fig. 2(b), it can be obtained that interband transition have a significant effect on the ATAS near K point, however, it plays a little role on that of \( \Gamma \) and M points. The underlying mechanism is that for the model of Fig. 1(b), the electrons near the K point can easily
transition from the valence band to the conduction band excited by the IR laser field due to small energy gap. As a result, X-ray absorption capability of the valence band is enhanced while that of the conduction band is weakened. Therefore, compared with Fig. 3(b), the ATAS around K point in Fig. 2(b) is significantly brighter. At M and Γ points, however, the wider energy gaps block interband transition process caused by the IR laser, and the generation mechanism of the ATAS arises from intraband dynamics of electrons driven by the IR laser. In the following work, we mainly investigate the generation mechanism of the ATAS near M and Γ points.

We further simplify our two-dimensional (2D) model of Fig. 3(a) to a one-dimensional (1D) model of Fig. 3(c) by taking a section along the Γ – M direction of the 2D band structure. The dispersion relation of the one-dimensional bands are \( \epsilon_c(k) = -\epsilon_v(k) = \epsilon_c(k = k_x, k_y = 0) = -\gamma_0 \sqrt{\frac{1}{2} + 4 \cos(3kd/2)} \) and \( \epsilon_g = \epsilon_{g1}(k) = \epsilon_{g2}(k) = -280 \) eV. Based on this model, the corresponding ATAS is presented in Fig. 3(d). Compared with Fig. 3(b), the absorption spectroscopy near the K point disappears in Fig. 3(d), while around the M and Γ points the results are consistent with Fig. 3(b). Therefore, we can exploit the the one-dimensional model of Fig. 3(c) to study ATAS near the M and Γ points.

Furthermore, in the model of Fig. 3(c), the electrons in the \( g_1 \) and \( v \) bands can not jump to the \( g_2 \) or \( c \) bands due to \( D_{g_1c}(k) = D_{vc}(k) = D_{g_1g_2}(k) = D_{vg_2}(k) = 0 \). On the other hand, although \( D_{g_1v}(k) \neq 0 \), the transition between \( g_1 \) and \( v \) bands is forbidden due to \( \rho_{g_1v}(k, t = 0) = \rho_{vc}(k, t = 0) = 1 \). Therefore, the one-dimensional four-band model of Fig. 3(c) is equivalent to the one-dimensional two-band structure of Fig. 3(e) consisting of \( g_2 \) and \( c \) bands. We propose a two-electron model as shown in Fig. 3(e), in which the lattice momentums of the two electrons are \( k = 0 \) (Γ point) and \( k \approx 0.78 \) a.u. (M point), and the corresponding two-electron ATAS is shown in Fig. 3(f). Compared with Fig. 3(d), the main features, such as the V-shaped structure, the periodicity of the stripes, etc., have been reproduced. Next, based on this two-electron model in one-dimensional two-band structure, we investigate the underlying mechanism of the ATAS, analytically.

\section*{IV. THEORETICAL EXPLANATION OF RESONANCE EFFECT IN ATAS}

When the IR laser is absent, for the two-electron model in one-dimension two-band structure of Fig. 3(e), the response intensity around Γ or M points can be expressed by

\[
S_k^X(\omega) \propto \frac{\Gamma_0}{\Gamma_0^2 + (\omega - \epsilon_c(k))^2},
\]

(see Appendix A for the detailed derivation).

When the IR laser is on and the time delay is \( t_d = 0 \), the response intensity of the electron of Γ or M points can be evaluated by

\[
S_k(\omega, t_d = 0) \propto J_0(b_{0k})[L[\omega, \epsilon_{0s}(k)] + J_1(b_{0k}) \cdot L[\omega, \epsilon_{0s}(k) + 2\omega I] - J_1(b_{0k})L[\omega, \epsilon_{0s}(k) - 2\omega I]],
\]

where \( L(\omega, x) = \frac{\Gamma_0}{\Gamma_0^2 + (\omega - x)^2} \) is Lorentzian line shape centered in \( x \). \( \epsilon_{0s}(k) = \epsilon_c(k) + A_{10}^2/(4m^*_s) \) where the second term on the right hand side is the energy shift. \( b_{0k} = A_{10}^2/(8\omega_I m^*_s) \) where \( m^*_s = 1/\sqrt{2}\epsilon_c(k) \) is effective mass. \( J_n(x) \) is the \( n \)th order Bessel function.

For the more general situations of \( t_d \neq 0 \), the ATAS of the electron near Γ or M points can be approximated by

\[
\Delta S_k(\omega, t_d) = S_k(\omega, t_d) - S_k^X(\omega) \]

\[
\propto J_0(b_{0k})[L[\omega, \epsilon_{s}(k)] - L[\omega, \epsilon_c(k)] + J_1(b_{0k}) \cdot L[\omega, \epsilon_{s}(k) + 2\omega I] \cos(2\omega I t_d)
+ J_1(b_{0k})F[\omega, \epsilon_{s}(k) + 2\omega I] \sin(2\omega I t_d)
- J_1(b_{0k})L[\omega, \epsilon_{s}(k) - 2\omega I] \cos(2\omega I t_d)
+ J_1(b_{0k})F[\omega, \epsilon_{s}(k) - 2\omega I] \sin(2\omega I t_d)],
\]

(8)
where $F(\omega, x) = \frac{\omega - x}{\Gamma_0 + (\omega - x)^2}$ corresponds to Fano line shape centered at $x$. $\epsilon_x(k) = \epsilon_x(k) + A_0^2f_{1}^2(t_d)/(4m^*_c)$ and $b_k = A_0^2f_{1}^2(t_d)/(8\omega_1m^*_c)$.

In Fig. 4(a), the solid lines are the numerical results of response intensity without (black) and with (red) IR laser of $t_d = 0$ for the two-electron model in the one-dimensional two-band structure of Fig. 3(e). The dotted lines are the corresponding analytical results calculated by Eq. (6) (black) and Eq. (7) (red), which agree well with the numerical results. According to Eq. (6), for the two-electron model, when the IR laser is absent, the response intensity presents two Lorentzian line shapes centered at $\epsilon_x(k_M) \approx 2.72$ eV and $\epsilon_x(k_F) \approx 8.16$ eV. As the IR laser of $t_d = 0$ is on, according to Eq. (7), the centre of the main peak related to $J_0(\theta_{\text{IR}})$ (the zeroth order peak) is shifted by $\Delta J_0(4m^*_c)$. For the electron of M point (Γ point) whose effective mass $m^*_c$ is 0.3 a.u. (~0.9 a.u.), the shift energy $A^2f_{0}^2(4m^*_c)$ is 0.27 eV (~0.09 eV). In addition, Eq. (7) tells that for the response intensity of each electron, two additional Lorentzian line shapes (the first order peak) appear with the amplitude of $|J_1(b_{\text{IR}})|$. Because $|m^*_c| < |m^*_c|$, one can obtain $|J_1(b_{\text{IR}})| > |J_1(b_{\text{IR}})|$ due to $b_{\text{IR}} > b_{\text{IR}}$. The energy interval between centres of zeroth and first order peaks is $2\omega_1$ as shown in Fig. 4(a).

The numerical results of the two-electron ATAS presented in Fig. 4(b) qualitatively agree with analytical results calculated by Eq. (8) as shown in Fig. 4(c). The vertical black and red lines mark the time delays of $t_d = 0$ and $t_d = -T/8$, and the marked ATAS are also presented in Fig. 4(d). In Fig. 4(c), the horizontal purple dotted lines and orange dashed lines indicate the centres of the zeroth order and first order stripes of the ATAS, corresponding to the same type of vertical lines in Fig. 4(d).

In Fig. 4(d), we choose a typical energy range of ATAS, which is marked by the blue dashed rectangle, to discuss. The absorption spectroscopy of this energy range is associated with the term $-J_1(b_{\text{IR}})L[\omega, \epsilon_x(k_M) - 2\omega_1 \cos(2\omega_1 t_d)] + J_1(b_{\text{IR}})F[\omega, \epsilon_x(k_M) - 2\omega_1 \sin(2\omega_1 t_d)]$ of Eq. (8). For the time delay of $t_d = 0$, this term is reduced to $-J_1(b_{\text{IR}})L[\omega, \epsilon_x(k_M) - 2\omega_1]$ which is the Lorentzian line shape corresponding to the black lines. While for the time delay of $t_d = -T/8$, this term is $J_1(b_{\text{IR}})F[\omega, \epsilon_x(k_M) - 2\omega_1]$ which is Fano line shape corresponding to the red lines. As the time delay varies continuously, the two line shapes alternately forming first-order stripes of ATAS, which vary at twice the IR laser frequency (corresponding to $2\omega_1 T_d$ of Eq. (8)) as presented in Figs. 3(f) and 2(b) [18, 19, 24, 25]. In addition, one can obtain $|J_1(b_{\text{IR}})| > |J_1(b_{\text{IR}})|$ due to $|b_{\text{IR}}| > |b_{\text{IR}}|$ with $|m^*_c| < |m^*_c|$. It implies that for smaller absolute value of the effective electron mass, the first-order stripes of ATAS are brighter, which can be observed in Figs. 3(f) and 2(b).

In Figs. 4(b), 4(c) and 2(b), we show the opening angle $\alpha$ of V-shaped structure of first-order stripes. The angle is defined by

$$\tan(\alpha) \approx \frac{2\omega_1}{3T/8} = \frac{8\omega_1}{3\pi}.$$  

where $2\omega_1$ is the energy interval between the zeroth and first order peaks of response intensity as shown in the red lines of Fig. 4(a), and $3T/8$ is the time interval between a Lorentzian line shape and its next-adjacent Fano line shape as shown in Figs. 4(b) and 4(c). Equation (9) implies that the opening angle increases as the increase of the IR laser frequency [18].

In Fig. 5, we compare our main analytical conclusions and the corresponding numerical results of the ATAS in Fig. 2(b). The two purple solid curves ($E(M^*_c)$...
the maximum energy shifts of the zeroth and first stripes of the ATAS near M point in Fig. 2(b), and the dotted lines ($\epsilon_0(M_M)$, $\epsilon_1(M_M)$) and $\epsilon_0(M_M) + 2\omega_I$ are the corresponding theory prediction results. In (c), the black line is calculated by $E(M_1^+) - E(M_0^+)$ and the black dotted line marks the energy of $2\omega_I$. In (d), the black line is the ATAS of Fig. 2(b) as a function of time delays at the specific frequency of $\omega = \epsilon_0(M_M) + A_{10}^2/(4m^*_k)$, and the red line is the corresponding analytical results calculated by Eq. (B4).

Furthermore, the numerical results of Figs. 5(a) and 5(b) indicate that in contrast to the theory prediction, the maximum energy shifts of the zeroth and first stripes of the ATAS near M point (about 2.72 eV) in Fig. 2(b) are offset by about 0.52 laser cycle (about 5 fs). In Fig. 5(d), we further check the offset by comparing the ATAS of Fig. 2(b) as a function of time delays at the specific frequency of $\omega = (\epsilon_0(M_M) + A_{10}^2/(4m^*_k)$ with our corresponding theoretical predictions of the two-electron model. In Fig. 5(c), the black solid line is the energy difference between $E(M_1^+)$ and $E(M_0^+)$. And the black dotted line marking $2\omega_I$ is the corresponding theory prediction. By comparing analytical and numerical results shown in Figs. 5(a), 5(b) and 5(c), one can conclude that both are qualitatively consistent.

V. CONCLUSION

In summary, we have investigated the underlying generation mechanism of ATAS in graphene. Based on the four-band model, we numerically calculated the ATAS by using the density-matrix equations of the length and velocity gauges, respectively. The numerical results show that the ATAS near M and K points are qualitatively consistent between two gauges, however, the ATAS around $\Gamma$ point present observable difference. This might be partly due to that the velocity gauge requires the solution of dynamics equations on the basis including many virtual states which are not physically occupied.

We propose a two-electron model, based on which we qualitatively reproduce the numerical results of the ATAS near M and $\Gamma$ points in the length gauge. According to the analytical results corresponding to the two-electron model, we divide the fishbone structure of the ATAS into the zeroth and first order stripes, and clearly define the opening angle of the V-shaped structure of the first order stripes. The periodic variation of the first-order stripes at twice the infrared laser frequency can be clearly explained using our analytical theory. Our theories also reveal the underlying mechanism that the opening angles of V-shaped structure increase with increasing the IR laser frequency. These results may be generalized to other periodic materials and shed light on the complex generation mechanism of the ATAS.

ACKNOWLEDGMENTS

This work is supported by NSAF (Grant No. U1930403). We acknowledge valuable discussions with Professor Difa Ye.

Appendix A: Analytical results of ATAS based on the two-electron model

We consider the one-dimensional two-band structure consisting of a core ($g$) band and a conduction ($c$) band as shown in Fig. 3(e). In this model, we approximate the X-ray pulse to be a $\delta$ function $E_X(t) = E_X\delta(t)$ because it is relatively short and weak, and therefore we can consider that the X-ray pulse instantaneously populates the $c$ band at the moment of $t = 0$. According to the perturbation theory, the density matrix elements change from $\rho_{gg}(\mathbf{k}, t < 0^-) = 1$, $\rho_{cc}(\mathbf{k}, t < 0^-) = 0$, $\rho_{cg}(\mathbf{k}, t < 0^-) = 0$ to $\rho_{gg}(\mathbf{k}, t = 0^+) \approx 1$, $\rho_{cc}(\mathbf{k}, t = 0^+) \approx 0$, $\rho_{cg}(\mathbf{k}, t = 0^+) \approx -iE_Xr_z$ with $r_z = (g(k)|c(k))$. Next, the IR laser induces Bloch oscillation of electrons and holes, i.e., $\rho_{cg}(\mathbf{k}, t > 0^+, t_d) = -iE_Xr_ze^{-i\int_0^t c(k+\mathbf{A}(t,t_d)-c(k))dt'}e^{-1/4t_d}$. According to Eq.
(3), when \( t < 0 ^ { - } \), the time-dependent dipole is \( \mu_k (t, t_d) = 0 \); and for \( t > 0 ^ { + } \), it is

\[
\mu_k (t, t_d) = -2E_X r_z^2 \sin \left[ \int_0^t \left( \epsilon_c (k + A_1 (t', t_d)) - \epsilon_g \right) dt' \right] \gamma \omega^{-1} .
\]  

(A1)

The response intensity is calculated by

\[
S^X (\omega, t_d) = \sum \left[ S_k (\omega, t_d) \right] \text{and} \quad S_k (\omega, t_d) = 2 \left( \mu_k (\omega, t_d) \times \hat{E}_X (\omega) \right) \propto \text{Im} [\hat{\mu}_k (\omega, t_d)].
\]

When the IR laser is on, the time-dependent dipole is

\[
\mu_k^X (t) = -2E_X r_z^2 \sin (\epsilon_c (k) - \epsilon_g) \gamma e^{-\gamma \omega t}.
\]

For both \( \Gamma \) and \( M \) points in the one-dimension two-band structure, \( \nabla \varepsilon (\omega, t_d) = 0, \quad \nabla_\omega \varepsilon (\omega, t_d) = 0 \), and we ignore higher order terms in Eq. (A3). In the integral with respect to time in Eq. (A3), for simplicity, we consider the vector potential \( A_1 (t, t_d) = A_{10} f_{11} (t + t_d) \cos (\omega t + \omega t d) \approx A_{10} f_{11} (t_d) \cos (\omega t + \phi) \) with \( \phi = \omega t d \), we can approximate the time-dependent dipole as

\[
\mu_k (t, t_d) \approx -2E_X r_z^2 \sin (\epsilon_c (k) t - t_d) + \frac{1}{2} \nabla^2 k \varepsilon (\omega, t_d)
\]

\[
= -2E_X r_z^2 \left[ \frac{1}{2} \nabla^2 k \varepsilon (\omega, t_d) \right] e^{-\gamma \omega t}.
\]

(A4)

where \( \phi = -b_k \sin (2 \omega t) \), \( a_k = \epsilon_c (k) - \epsilon_g + A_{10} f_1^2 (t_d) / (4 m^* k) \) and \( b_k = A_{10} f_1^2 (t_d) / (8 m^* k) \) with the effective mass \( m^* = 1 / (\nabla^2 k \varepsilon (k)) \) for lattice momentum \( k \).

Utilizing Jacobi-Anger expansion formulas and product to sum formulas, Eq. (A4) can be further reduced to be

\[
\mu_k (t, t_d) = -2E_X r_z^2 \sin (\epsilon_c (k) t + \phi) J_0 (b_k) e^{-\gamma \omega t}
\]

\[
-2E_X r_z^2 \sum_{m=1}^{\infty} J_{2m} (b_k) \sin \left[ \left( a_k + 4m \omega t \right) t + (\phi + 4m \phi) \right] e^{-\gamma \omega t}
\]

\[
-2E_X r_z^2 \sum_{m=1}^{\infty} J_{2m-1} (b_k) \sin \left[ \left( a_k - 4m \omega t \right) t + (\phi - 4m \phi) \right] e^{-\gamma \omega t}
\]

\[
-2E_X r_z^2 \sum_{m=1}^{\infty} J_{2m-1} (b_k) \sin \left[ (4m - 2) \omega t + \phi \right] t + (4m - 2) \phi) e^{-\gamma \omega t}
\]

(A5)

where \( J_n (x) \) is the \( n \)th order Bessel function.

The response intensity is evaluated by

\[
S_k (\omega, t_d) \propto \text{Im} [\hat{\mu}_k (\omega, t_d)] = \text{Im} \left[ \int_0^\infty \mu_k (t, t_d) e^{-i \omega t} dt \right]
\]

\[
= J_0 (b) \left[ L (\omega, a_k) \cos (\phi) + F (\omega, a_k) \sin (\phi) \right]
\]

\[
+ \sum_{m=1}^{\infty} J_{2m-1} (b_k) L (\omega, a_k + (4m - 2) \omega t) \cos (\phi + (4m - 2) \phi)
\]

\[
+ \sum_{m=1}^{\infty} J_{2m-1} (b_k) F (\omega, a_k + (4m - 2) \omega t) \sin (\phi + (4m - 2) \phi)
\]

\[
- \sum_{m=1}^{\infty} J_{2m-1} (b_k) L (\omega, a_k - (4m - 2) \omega t) \cos (\phi - (4m - 2) \phi)
\]

\[
- \sum_{m=1}^{\infty} J_{2m-1} (b_k) F (\omega, a_k - (4m - 2) \omega t) \sin (\phi - (4m - 2) \phi)
\]

\[
+ \sum_{m=1}^{\infty} J_{2m} (b_k) L (\omega, a_k + 4m \omega t) \cos (\phi + 4m \phi)
\]

\[
+ \sum_{m=1}^{\infty} J_{2m} (b_k) F (\omega, a_k + 4m \omega t) \sin (\phi + 4m \phi)
\]

\[
+ \sum_{m=1}^{\infty} J_{2m} (b_k) L (\omega, a_k - 4m \omega t) \cos (\phi - 4m \phi)
\]

\[
+ \sum_{m=1}^{\infty} J_{2m} (b_k) F (\omega, a_k - 4m \omega t) \sin (\phi - 4m \phi),
\]

(A6)

where \( L (\omega, x) = \frac{\Gamma_0}{\Gamma_0^2 + (\omega - x)^2} \) and \( F (\omega, x) = \frac{\omega - x}{\Gamma_0^2 + (\omega - x)^2} \) are Lorentzian and Fano line shapes centred at \( x \), respectively.
For a special time delay of $t_d = 0$, we can obtain $\varphi = 0$ and $\phi = 0$. When the laser intensity is $1 \times 10^{11}$ W/cm², $b_{ok} = A_{I0}^2/(8\omega_1 m_k^* c) = -0.11$ and $b_{okt} = A_{I0}^2/(8\omega_1 m_{k0}^*) = 0.33$. Wherefore we consider that high-order terms of Bessel function $J_{n\geq 1}(b_{ok})$ are approximately equal to zero for both $\Gamma$ and $M$ points. The response intensity is

\[
S_k(\omega, t_d = 0) \simeq J_0(b_{ok}) L(\omega, a_{ok}) + J_1(b_{ok}) \cdot L(\omega, a_{ok} + 2\omega_I) - J_1(b_{ok}) L(\omega, a_{ok} - 2\omega_I), \quad (A7)
\]

where $a_{ok} = \epsilon_e(k) - \epsilon_g + A_{I0}^2/(4m_k^*)$.

For general situations of $t_d \neq 0$, because $|b_k| \leq 0.33$ for $\Gamma$ and $M$ points, we do the approximations that $\cos(\phi) = \cos(-b_k \sin(2\phi)) \approx J_0(b_k) \approx 1$ and $\sin(\phi) = \sin(-b_k \sin(2\phi)) \approx 0$. According to Eq. (A6), the ATAS is evaluated by

\[
\Delta S_k(\omega, t_d) = S_k(\omega, t_d) - S_k^X(\omega) \\
\approx J_0(b_k) L(\omega, a_k) - L(\omega, \epsilon_e(k) - \epsilon_g) + J_1(b_k) L(\omega, a_k + 2\omega_I) \cos(2\omega_I t_d) + J_1(b_k) F(\omega, a_k + 2\omega_I) \sin(2\omega_I t_d) - J_1(b_k) L(\omega, a_k - 2\omega_I) \cos(2\omega_I t_d) + J_1(b_k) F(\omega, a_k - 2\omega_I) \sin(2\omega_I t_d). \quad (A8)
\]

### Appendix B: The ATAS as a function of time delay at a specific frequency

We deduce the ATAS as a function of $t_d$ at a specific frequency. Considering the vector potential $A_I(t, t_d) = A_{I0} f_I(t + t_d) \cos(\omega_I t + \varphi)$ with $\varphi = \omega_I t_d$, the time-dependent dipole of Eq. (A3) can be reduced to be

\[
\mu_{ok}(t, t_d) = -2EX r_k^2 \sin(\epsilon_c(k)t) - \epsilon_g t + \frac{1}{2} \nabla_k^2 \epsilon_c(k) A_{I0}^2 \\
\int_0^t (1 + \beta) \cos(2\omega_I t' + 2\varphi) + \frac{1}{2} dt'[e^{-\Gamma_{rt} t} \\
= -2EX r_k^2 \sin(a_{ok} t + b_{ok} \sin(2\omega_I t + 2\varphi) + c(t, t_d)) e^{-\Gamma_{rt} t} \quad (B1)
\]

where $\beta = f_I(t' + t_d) - 1$, $a_{ok} = \epsilon_e(k) - \epsilon_g + A_{I0}^2/(4m_k^*)$, $b_{ok} = A_{I0}^2/(8\omega_1 m_k^*)$, $c(t, t_d) = a_{ok} \int_0^t \beta[\cos(2\omega_I t' + 2\varphi) + 1] dt' + \phi_0$ with $\phi_0 = -b_{ok} \sin(2\phi)$.

We expand Eq. (B1) using Jacobi-Anger expansion formulas and ignore the high-order terms of Bessel function $J_{n \geq 2}(b_{ok})$. The time-dependent dipole is deduced to be

\[
\mu_{ok}(t, t_d) \approx -2EX r_k^2 J_0(b_{ok}) \sin(a_{ok} t + c(t, t_d)) e^{-\Gamma_{rt} t} \\
-2EX r_k^2 J_0(b_{ok}) \sin(c(t, t_d)) e^{-\Gamma_{rt} t} \\
+4EX r_k^2 J_1(b_{ok}) \sin(a_{ok} t) \sin(2\omega_I t + 2\varphi) \sin(c(t, t_d)) e^{-\Gamma_{rt} t} \\
+4EX r_k^2 J_1(b_{ok}) \sin(a_{ok} t) \sin(2\omega_I t + 2\varphi) \sin(c(t, t_d)) e^{-\Gamma_{rt} t} \quad (B2)
\]

We focus on the Fourier transform of Eq. (B2) at $\omega = a_{ok}$, which can be classified into slow and fast oscillating terms and those fast oscillating term will be averaged out after long time integration, so

\[
\tilde{\mu}_{ok}(\omega = a_{ok}, t_d) = \int_0^{\infty} \mu_{ok}(t, t_d) e^{-i\omega t} dt \\
\approx -2EX r_k^2 J_0(b_{ok}) \int_0^{\infty} [\sin(a_{ok} t) \cos(c(t, t_d)) + \cos(a_{ok} t) \sin(c(t, t_d))] e^{-\Gamma_{rt} t} e^{-i\omega t} dt \\
\approx iEX r_k^2 J_0(b_{ok}) \int_0^{\infty} \cos(c(t, t_d)) e^{-\Gamma_{rt} t} dt \\
-EX r_k^2 J_0(b_{ok}) \int_0^{\infty} \sin(c(t, t_d)) e^{-\Gamma_{rt} t} dt. \quad (B3)
\]

The ATAS at $\omega = a_{ok}$ is evaluated by

\[
\Delta S_k(\omega = a_{ok}, t_d) = S_k(\omega = a_{ok}, t_d) - S_k^X(\omega = a_{ok}) \\
= \text{Im}[\tilde{\mu}_{ok}(\omega = a_{ok}, t_d)] - \text{Im}[\tilde{\mu}_{ok}^X(\omega = a_{ok})] \\
= EX r_k^2 J_0(b_{ok}) \int_0^{\infty} \cos(c(t, t_d)) e^{-\Gamma_{rt} t} dt, \quad (B4)
\]

where we have considered $S_k^X(\omega = a_{ok}) = \text{Im}[\tilde{\mu}_{ok}^X(\omega = a_{ok})] = 0$.

---

[1] F. Krausz and M. Ivanov, *Attosecond physics*, Rev. Mod. Phys. 81, 163 (2009).
[2] M. Hentschel, R. Kienberger, Ch. Spielmann, G. A. Reider, N. Milosevic, T. Brabec, P. Corkum, U. Heinzmann, M. Drescher, and F. Krausz, *Attosecond metrology*, Nature 414, 509 (2001).
[3] A. L. Cavalieri, N. Mueller, T. Uphues, V. S. Yakovlev, A. Baltuska, B. Horvath, B. Schmidt, L. Bluemel, R. Holzwarth, S. Hendel, M. Drescher, U. Kleineberg, P. M. Echenique, R. Kienberger, F. Krausz, and U. Heinzmann, Attosecond metrology, Attosecond spectroscopy in condensed matter, Nature 449, 1029 (2007).

[4] E. Goulielmakis, Zhi-Heng Loh, A. Wirth, R. Santra, N. Rohringer, V. S. Yakovlev, S. Zherebtsov, T. Pfeifer, A. M. Azzeer, M. F. Kling, S. R. Leone, and F. Krausz. Real-time observation of valence electron motion, Nature 466, 739 (2010).

[5] M. B. Gaarde, C. Buth, J. L. Tate, and K. J. Schafer, Transient absorption and reshaping of ultrafast XUV light by laser-dressed helium, Phys. Rev. A 83, 013419 (2011).

[6] M. Wu, S. Chen, S. Camp, K. J. Schafer, and M. B. Gaarde, Theory of strong-field attosecond transient absorption, J. Phys. B 49, 062003 (2016).

[7] A. R. Beck, D. M. Neumark, and S. R. Leone, Probing ultrafast dynamics with attosecond transient absorption, Chem. Phys. Lett. 624, 119 (2015).

[8] P. M. Kraus, M. Zürich, S. K. Cushing, D. M. Neumark, and S. R. Leone, The ultrafast X-ray spectroscopic revolution in chemical dynamics, Nat. Rev. Chem. 2, 82 (2018).

[9] R. Geneux, H. J. B. Marroux, A. Guggenmos, D. M. Neumark, and S. R. Leone, Transient absorption spectroscopy using high harmonic generation: A review of ultrafast x-ray dynamics in molecules and solids, Phil. Trans. R. Soc. A 377, 20170463 (2019).

[10] H. Wang, M. Chini, S. Chen, C.-H. Zhang, F. He, Y. Cheng, Y. Wu, U. Thumm, and Z. Chang, Attosecond time-resolved autoionization of Argon, Phys. Rev. Lett. 105, 143002 (2010).

[11] M. Holler, F. Schapper, L. Gallmann, U. Keller, Attosecond electron wave-packet interference observed by transient absorption, Phys. Rev. Lett. 106, 123601 (2011).

[12] Z. Q. Yang, D. F. Ye, T. Ding, T. Pfeifer, and L. B. Fu, Attosecond XUV absorption spectroscopy of doubly excited states in helium atoms dressed by a time-delayed femtosecond infrared laser, Phys. Rev. A 91, 013414 (2015).

[13] J. E. Barkhøj and L. B. Madsen, Attosecond transient-absorption spectroscopy on aligned molecules, Phys. Rev. A 94, 043416 (2016).

[14] S. Chen, M. Wu, M. B. Gaarde, and K. J. Schafer, Quantum interference in attosecond transient absorption of laser-dressed helium atoms, Phys. Rev. A 87, 033408 (2013).

[15] M. Chini, B. Zhao, H. Wang, Y. Cheng, S. X. Hu, and Z. Chang, Subcycle ac Stark shift of Helium excited states probed with isolated attosecond pulses, Phys. Rev. Lett. 109, 073601 (2012).

[16] X. Wu, Z. Yang, S. Zhang, X. Ma, J. Liu, and D. Ye, Buildup time of Autler-Townes splitting in attosecond transient absorption spectroscopy, Phys. Rev. A 103, L061102 (2021).

[17] M. Volkov, S. A. Sato, F. Schlaepfer, L. Kasmi, N. Hartmann, M. Lucchini, L. Gallmann, A. Rubio, and U. Keller, Attosecond screening dynamics mediated by electron localization in transition metals, Nat. Phys. 15, 1145 (2019).

[18] M. Lucchini, S. A. Sato, A. Ludwig, J. Herrmann, M. Volkov, L. Kasmi, Y. Shinozaka, K. Yabana, L. Gallmann, and U. Keller, Attosecond dynamical Franz-Keldysh effect in polycrystalline diamond, Science 353, 916 (2016).

[19] F. Schlaepfer, M. Lucchini, S. A. Sato, M. Volkov, L. Kasmi, N. Hartmann, A. Rubio, L. Gallmann, and U. Keller, Attosecond optical-field-enhanced carrier injection into the GaAs conduction band, Nat. Phys. 14, 560 (2018).

[20] T. Otobe, Attosecond electro-optic effect in zinc sulfide induced by a laser field, Phys. Rev. A 100, 033401 (2019).

[21] M. Schultze, K. Ramesh, C. D. Premaratne, S. A. Sato, D. Whitmore, A. Gandman, J. S. Prell, L. J. Borja, D. Prendergast, and K. Yabana, Attosecond band-gap dynamics in silicon, Science 346, 1348 (2014).

[22] M. Lucchini, S. A. Sato, G. D. Lucarelli, B. Moio, G. Inzani, R. Borrego-Varillas, F. Frassetto, L. Poletto, H. Hübener, U. De Giovannini, A. Rubio, and M. Nisoli, Unravelling the intertwined atomic and bulk nature of localised excitons by attosecond spectroscopy, Nat. Commun. 12, 1021 (2020).

[23] G. Cistaro, L. Plaja, F. Martín, and A. Picón, Attosecond x-ray transient absorption spectroscopy in graphene, Phys. Rev. Research 3, 013144 (2021).

[24] S. A. Sato, H. Hübener, U. De Giovannini, and A. Rubio, Ab initio simulation of attosecond transient absorption spectroscopy in two-dimensional materials, Appl. Sci. 8, 1777 (2018).

[25] T. Otobe, Y. Shinozaka, S. A. Sato, and K. Yabana, Femtosecond time-resolved dynamical Franz-Keldysh effect, Phys. Rev. B 93, 045124 (2016).

[26] N. T. Dejean, M. A. Sentef, and A. Rubio, Ultrafast transient absorption spectroscopy of the charge-transfer insulator NiO: Beyond the dynamical Franz-Keldysh effect, Phys. Rev. B 102, 115106 (2020).

[27] A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim, The electronic properties of graphene, Rev. Mod. Phys. 81, 109 (2009).

[28] P. R. Wallace, The band theory of graphite, Phys. Rev. 71, 622 (1947).

[29] G. Vampa, C. R. McDonald, G. Orlando, D. D. Klug, P. B. Corkum, and T. Brabec, Theoretical analysis of high-harmonic generation in solid materials, Phys. Rev. A 113, 073001 (2014).

[30] S. C. Jiang, H. Wei, J. G. Chen, C. Yu, R. F. Lu, and C. D. Lin, Effect of transition dipole phase on high-order harmonic generation in solid materials, Phys. Rev. A 96, 053850 (2017).

[*] jliu@gscaep.ac.cn