Microscopic theory of ultrafast dynamics of carriers photoexcited by THz and near-infrared linearly polarized laser pulses in graphene

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Abstract. We investigate the dynamics of photoexcited carriers and non-equilibrium phonons in graphene under the linear energy dispersion approximation by solving the microscopic kinetic Bloch equations. The pump and drift effects from the laser field as well as the relevant scatterings (including Coulomb scattering with dynamic screening) are explicitly included. When the pump-photon energy is high enough (with the oscillation period much smaller than the pulse width and the scattering time), the influence of the drift term is shown to be negligible and the isotropic hot-electron Fermi distribution with separate conduction- and valence-band chemical potentials is established under scattering during the linearly polarized laser pulse investigated here. However, in the case with low pump-photon energy (with the oscillation period larger than the pulse width), the drift term is important and leads to a net momentum transfer from the electric field to the electrons. Owing to this net momentum and the dominant Coulomb scattering, a drifted Fermi distribution different from the one established under static electric field is found to be established in several hundred femtoseconds. We also show that the Auger process investigated in the literature involving only the diagonal terms of density matrices is forbidden by the dynamic screening. However, we propose an Auger process involving interband coherence and show that it contributes to the dynamics of carriers.

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when the pump-photon energy is low (comparable with the variation speed of the distribution). In addition, the anisotropically momentum-resolved hot-phonon temperatures due to the linearly polarized light are also investigated, with the underlying physics revealed.

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1. Introduction

Graphene is a promising material which has received intensive attention both theoretically and experimentally due to its excellent transport and optical properties [1–12]. In particular, its linear band structure and zero band gap lead to unique optical and electrical properties, making it suitable for various optoelectronic applications [1, 8]. A comprehensive understanding of the optical and electrical properties is one of the prerequisites for its potential applications.

Experimentally, the time-resolved pump–probe measurement is widely used to investigate the dynamics of carriers in graphene [13–30]. With this method, both the differential reflection and the differential transmission (DT) after pump are measured [13–30]. In the experiments with medium photoexcited electron density $N_e^p$ (around $10^{12}$ cm$^{-2}$) and probe-photon energy much higher than the equilibrium Fermi energy, positive DT is often observed with its fast relaxation of several hundred femtoseconds followed by a slower picosecond relaxation [13, 20, 21, 25]. The mechanism leading to these two distinct relaxations is attributed to the rapid carrier–phonon thermalization and the slow decay of hot phonons, respectively [20, 22, 31]. On the other hand, in recent years, experiments with higher photoexcited electron densities ($N_e^p > 10^{13}$ cm$^{-2}$) [23, 26, 29] show that a negative DT appears when the probe-photon energy ($>1500$ meV) is much higher than the equilibrium Fermi energy [23, 26]. Such negative DT was usually claimed to arise from the renormalization of the single-particle energy by the electron–electron Coulomb Hartree–Fock contribution [23, 26]. It is noted that although the observed DT with high probe-photon energy and low photoexcited electron density is usually positive, negative DT was also reported with $N_e^p = 3 \times 10^{11}$ cm$^{-2}$ by Plochocka et al [19] where the sample is 70–100 layer epitaxial graphene exhibiting the Dirac-type electron spectrum. In contrast to the negative DT with probe-photon energy much higher than equilibrium Fermi energy, other types of negative DTs with lower probe-photon energy have already been observed.
in previous works [14, 15]. Sun et al [14] reported the appearance of negative DT under weak pump intensity with the probe-photon energy between 528 and 697 meV, which is lower than twice the Fermi energy \( (E_F \sim 350 \text{ meV}) \). This negative DT was suggested to come from the weakening of the Pauli blocking due to heating of the electrons by the pump pulse [14]. However, this claim has not yet been verified by the microscopic theoretical approach. Soon after that negative DT was also observed by George and co-workers [15, 16] in the case when the probe-photon energy is as low as a few tens of meV (with the frequency about several terahertz). With such low probe-photon energy, the increase of the intraband absorption [15] and the heating of low-energy electrons [32] are assumed to be responsible for negative DT. In addition to these phenomena, with the pump–probe method, optical gain has also been reported in optically pumped graphene very recently [28].

In addition to these experimental works, many theoretical studies have been devoted to obtain a clear comprehension on the dynamics of photoexcited carriers and phonons [20, 25, 29, 31–40]. Among these works, the simple rate equation approach [20, 25, 32, 40] assumes the establishment of the Fermi distribution as a starting point and, therefore, fails to incorporate the pump process and the buildup of the Fermi distribution. The widely used time-dependent Boltzmann equation approach [31, 35, 39] can fulfill the above aims but neglects interband coherence [36], which should be very important during the pump process. More elaborate microscopic investigations with the interband coherence included have been carried out based on many-particle density-matrix approach [34, 36–38]. However, these theoretical investigations are still far from completion, as stated in the following.

Among papers with the interband coherence included [34, 36–38], the scattering terms under the Markovian approximation with Coulomb screening in the static limit are applied. Nevertheless, with static screening, the electron–electron Coulomb scattering diverges when the initial and final states of the scattered electrons satisfy the condition \( \hbar v_F |q| = |\hbar \omega| \) [41, 42], where \( q \) and \( \hbar \omega \) are the differences in momentum and energy between the initial and final states, respectively. To avoid this divergence, in these papers, the delta function originating from the Markovian approximation and reflecting the energy conservation in the Coulomb scattering term, was replaced by a Lorentzian function with the broadening attributed to the non-Markovian effect [43]. Nevertheless, under the more natural dynamic screening [31, 44–46], this broadening is not needed. This is because the dynamic screening also diverges in a higher order when \( \hbar v_F |q| = |\hbar \omega| \) [47, 48], and therefore reduces the original divergent scattering amplitudes to zero.

Another important issue frequently investigated is the influence of the Auger process. In previous works, the interband coherence was neglected and the Auger process was defined as the process where one of the two scattered electrons is transferred from one band to the other while the other electron remains in the same band [34, 35, 37]. In the works by Winzer and co-workers [34, 37], such a process was particularly stressed. However, under the dynamic screening, this Auger process is totally forbidden because it also satisfies the strong restriction \( \hbar v_F |q| = |\hbar \omega| \) due to the energy and momentum conservation under linear energy dispersion approximation. However, in the case with the optical transition, interband coherence is generated. With the interband coherence, there are Coulomb scattering processes with three out of the four band indices in the Coulomb interaction Hamiltonian being the same and the left one being different. This is also a kind of Auger process. However, the influence of such a process has not been addressed in the literature.
Finally, in the microscopic equations obtained based on the many-particle density-matrix framework by Knorr and co-workers [34, 36–38], the pump term was obtained under the vector potential gauge. However, the drift term, which describes the acceleration of the electrons by the electric field, is not included. The drift term may be unimportant when the pump-photon energy is very high as investigated in these works. Nevertheless, it can be expected to be important when the pump-photon energy is low. On the other hand, in the time-dependent Boltzmann equation approach, the drift term is included but the pump term in the equation has to be introduced based on a Fermi-golden-rule-like approximation and hence is suitable only for a weak pump intensity [35]. Moreover, Rabi flopping [49] is absent in this approach. Therefore, a general equation based on the microscopic many-particle density-matrix approach that includes both the drift and the pump terms naturally is needed.

In this paper, we perform a microscopic investigation of the dynamics of photoexcited carriers and phonons in graphene under the linear energy dispersion approximation by setting up and solving the kinetic Bloch equations with the electron–phonon, electron–impurity and electron–electron Coulomb scatterings explicitly included [49, 50]. In our study, dynamic screening [47, 51–53] is adopted in Coulomb scattering. Moreover, with the gauge invariant approach [49], both the drift and pump terms are obtained naturally. We look into the dynamics of carriers photoexcited by the linearly polarized laser pulses with the pump-photon energy in both near-infrared and THz regimes under medium pump intensity. When the pump-photon energy is high enough (with the corresponding oscillation period much smaller than the scattering time and the pulse width), the influence of the drift term is shown to be negligible compared to that from the pump process. Moreover, the anisotropic photoexcited electrons tend to be isotropic under the scattering and an isotropic hot-electron Fermi distribution with separate conduction- and valence-band chemical potentials is established before the end of the pulse investigated here. In contrast, when the pump-photon energy is low (with the corresponding oscillation period larger than the pulse width), the drift term causes a net momentum transfer from the electric field to the electrons. Together with the dominant Coulomb scattering, a drifted Fermi distribution is established in several hundred femtoseconds. Moreover, the form of the drifted Fermi distribution differs from the one obtained under a static electric field [46]. Besides, we also show that the temporal evolution of the DT measured by Hale et al [25] can be well fitted with our microscopic calculation. In addition, under the linear energy spectrum approximation, although the Auger process involving only the diagonal terms of the density matrices is forbidden by dynamic screening, we show that the Auger process involving the interband coherence still contributes to the dynamics of carriers. However, its contribution is important only when the pump-photon energy is low (comparable to the variation speed of the distribution). Similarly, it is shown that the terms neglected in the rotating-wave approximation [49], which is widely accepted in semiconductor optics, also become important in the case of low pump energy. We also investigate the dynamics of phonons. Owing to the linearly polarized pump pulse, the q-resolved hot-phonon temperatures are anisotropic. Moreover, the anisotropic hot-phonon temperatures under high pump energy are very different from those under low pump energy. Finally, we investigate the negative DT by fitting the temporal evolution of DT in the experimental work by Sun et al [14]. Our results support their suggestion that the negative DT comes from the weakening of Pauli blocking due to the heating of the electrons by the pump pulse.

This paper is organized as follows. In section 2, we set up the model, lay out the kinetic Bloch equations and then give a simple analysis of the kinetic Bloch equations. In section 3 the
results obtained numerically from the kinetic Bloch equations are presented. We summarize in section 4.

2. Model and formalism

We start our investigation from graphene on SiO$_2$ and SiC substrates. In the absence of an external field, the energy dispersion of graphene near $K$ and $K'$ points can be described by the effective Hamiltonian [54] ($\hbar = 1$)

$$H_0^{\mu,AB} (\mathbf{k}) = v_F (\mu \tau_x k_x + \tau_y k_y).$$

(1)

Here, $v_F$ represents the Fermi velocity, $\mu = 1$ ($-1$) for $K$ ($K'$) valley and $\tau$ are the Pauli matrices in the pseudospin space formed by the A and B sublattices of the honeycomb lattice. The eigenvalues of $H_0^{\mu,AB}$ are $\varepsilon_{\mathbf{k} \mu} = \eta v_F k$ and the corresponding eigenstates are $\psi_{\mathbf{k} \mu} = (\mu \eta e^{-i\mathbf{k} \cdot \mathbf{r}}, 1)^T / \sqrt{2}$ with $\eta = 1$ ($-1$) for conduction (valence) band and $\theta_k$ representing the polar angle of $\mathbf{k}$. The additional electron–optical-field interaction Hamiltonian under the vector potential is given by [55]

$$H_{\text{photon}}^{\mu,AB} (\mathbf{k}, t) = \frac{|e| v_F [\mu \tau_x A_x (t) + \tau_y A_y (t)]}{c}.$$  

(2)

Note that this Hamiltonian is consistent with the one obtained from the tight-binding approach [56, 57]. Here, we only investigate the linearly polarized light with the electric field $\mathbf{E}(t) = -\mathbf{E}_0 \cos(\omega_L t) \exp[-t^2/(2\sigma^2)]$ along the $x$ direction. For simplicity, the vector potential is taken to be $\mathbf{A}(t) = \frac{1}{c} \mathbf{E}_0 \sin(\omega_L t) \exp[-t^2/(2\sigma^2)]$. Since the effective Hamiltonian equation (1) used is under the linear approximation, we restrict our investigation with the pump-photon energy $\omega_L$ lower than 2000 meV, for which the linear approximation is good for electrons with their energy under the resonant absorption energy $\omega_L/2$ [58]. Our investigation is performed in the base set of the eigenstates of the Hamiltonian (equation (1)). Under this base set, the Hamiltonians are given by

$$H_0^\mu (\mathbf{k}) = v_F k \sigma_z,$$

$$H_{\text{photon}}^\mu (\mathbf{k}, t) = \frac{|e| v_F [\mu \tau_x A_x (t) + \tau_y A_y (t)]}{c}.$$  

(4)  

(5)

with $\sigma$ representing the Pauli matrices in the conduction and valence band space, $\hat{\mathbf{k}}$ indicating the direction of $\mathbf{k}$ and $\hat{\mathbf{z}}$ standing for the unit vector along the $z$ direction.

Exploiting the non-equilibrium Green’s function approach with the gradient expansion as well as the generalized Kadanoff–Baym ansatz, the gauge invariant kinetic equations [49] of the carriers read [46, 50, 59, 60] (see appendix A for a detailed derivation of the coherent and drift terms)

$$\partial_t \rho_{\mu \mathbf{k}} = \partial_t \rho_{\mu \mathbf{k}} \big|_{\text{coh}} + \partial_t \rho_{\mu \mathbf{k}} \big|_{\text{drift}} + \partial_t \rho_{\mu \mathbf{k}} \big|_{\text{scat}}$$

(6)

in which $\rho_{\mu \mathbf{k}}$ stand for the density matrices with their diagonal terms $\rho_{\mu \mathbf{k}, \eta \eta} = f_{\mu \mathbf{k}, \eta}$ representing the electron distribution functions and the off-diagonal terms $\rho_{\mu \mathbf{k}, 1, -1} = \rho_{\mu \mathbf{k}, -1, 1}$ standing for the interband coherence. The drift terms

$$\partial_t \rho_{\mu \mathbf{k}} \big|_{\text{drift}} = |e| \mathbf{E} \cdot \nabla_k \rho_{\mu \mathbf{k}}$$

(7)
describe the acceleration of electrons by the laser field $E$ which is related to the Drude absorption mechanism. The coherent terms are given by

$$\partial_t \rho_{\mu k} \big|_{\text{coh}} = -i [v_F k \sigma_z + \Sigma_{\mu k}^{\text{HF}} + H_{\text{Pump}}^{\mu}, \rho_{\mu k}].$$

(8)

Here $[A, B] \equiv AB - BA$ is the commutator and $H_{\text{Pump}}^{\mu} = -|e|v_F \mu A, \sin \theta_k \sigma_z / c$ comes from the laser field describing the pump process which describes the interband absorption. $\Sigma_{\mu k}^{\text{HF}} = - \sum_q \hat{S}^\mu_{k,k-q} \rho_{\mu k-q} \hat{S}^\mu_{k-q,k} V^r(q, 0)$ stand for the Hartree–Fock self-energies, in which $\hat{S}^\mu_{k,k-q}$ are the form factors with their elements being $S^\mu_{k,k-q,\eta'\eta} = \psi^\mu_{kq} \psi^{\mu*}_{kq}$, and $V^r(q, \omega) = V^r_0 / \epsilon(q, \omega)$ represent the screened two-dimensional Coulomb potentials with $V^r_0 = 2\pi v_F r_s / q$ being the bare Coulomb potential. Here $r_s$ stands for the dimensionless Wigner–Seitz radius [52, 61–64]. $\epsilon(q, \omega)$ is the dielectric function under the dynamic screening [49, 52, 61], which is given by [47, 51–53]

$$\epsilon(q, \omega) = 1 - \sum_{\mu
u q \eta} |S^\mu_{k,k+q,\eta q}|^2 2(f_{\mu \eta, k} - f_{\mu \eta, k+q} V^r_0 \epsilon_{\eta k} - \epsilon_{\eta k+q} + \omega + io)^{-1}$$

(9)

It is noted that the laser field $E$ not only pumps electrons but also accelerates them, as revealed in our gauge invariant kinetic Bloch equations (equation (6)) [49] by the pump and the drift terms, respectively. However, in previous works by Knorr and co-workers [34, 36–38], the drift term is not included and hence the case of low pump energy, for which the drift term is expected to be important, cannot be investigated with their equations. In contrast, our kinetic Bloch equations can deal with both the high and low pump energies completely.

Under the laser field with pump-photon energy $\omega_L$, the off-diagonal terms of the density matrices are generated around the resonant absorption point (where $2v_F k \approx \omega_L$). They oscillate with the frequency around $\omega_L$. Following the approach adopted in semiconductor optics [49], we introduce slowly varying interband-polarization component $P_{\mu k} = \rho_{\mu k,1,-1} e^{i\omega_L t}$. Then, the kinetic equations can be rewritten as

$$\partial_t f_{\mu k, \eta} = \eta \text{Im}(\Omega_{\mu k}^*) P_{\mu k} + |e| E \cdot \nabla_k f_{\mu k, \eta} + \partial_t f_{\mu k, \eta} \big|_{\text{scat}},$$

$$\partial_t P_{\mu k} = -i \Omega_{\mu k} (f_{\mu k,1} - f_{\mu k,1})/2 - iv^\mu_k P_{\mu k} + |e| E \cdot \nabla_k P_{\mu k} + \partial_t P_{\mu k} \big|_{\text{scat}}$$

(10)

with the detuning $v^\mu_k = 2v_F k_1 - \omega_L + \text{Tr}[S^{\text{HF}}_{\mu k} + H^{\mu}_{\text{Pump}}] \sigma_z$ and the Rabi frequency [49]

$$\Omega_{\mu k} = -2 e^{i\omega_L t} \left( \Sigma_{\mu k,1,-1}^{\text{HF}} + H^{\mu}_{\text{Pump},1,-1} \right)$$

$$= \mu |e| v_F E_0 \sin \theta_k e^{-i\pi/2(1 - e^{2i\omega_L t})/\omega_L} - 2 \Sigma_{\mu k,1,-1}^{\text{HF}} e^{i\omega_L t}.$$  

(11)

When the pump-photon energy is much higher than the variation speeds of $P_{\mu k}$ and $f_{\mu k, \eta}$, these equations can be further simplified by neglecting the high-frequency oscillating terms (rotating-wave approximation) [65] and the Rabi frequency becomes

$$\Omega_{\mu k} = \mu |e| v_F E_0 \sin \theta_k e^{-i\pi/2(1 - e^{2i\omega_L t})/\omega_L} + 2 \sum_q \hat{S}^\mu_{k,k-q,1,-1} \hat{S}^\mu_{k-q,k,1,-1} V^r(q, 0) P_{\mu k-q}.$$  

(12)

Then, one finds that except for the drift and scattering terms, equation (10) is of the same form as that in the semiconductor optics [49]. However, when the pump energy is low, the rotating-wave approximation is no longer valid and the Rabi frequency must be calculated with equation (11).

2 The influence of the full valence band is subtracted in our calculation following [49, chapter 16].
We further define a new set of matrices $\hat{\rho}_{\mu k}$ with $\hat{\rho}_{\mu k, \eta \eta'} = f_{\mu k, \eta}$ and $\hat{\rho}_{\mu k, 1-1} = \hat{p}_{\mu k}^* = P_{\mu k}$. With such a definition, $\hat{\rho}_{\mu k}$ vary slowly with time. We perform the Markovian approximation in the interaction picture [49, 65, 66]. Then, the scattering terms in equation (10), which include the electron–electron Coulomb, electron–impurity, electron–optical phonon and electron–remote-interfacial (RI) phonon scatterings, are given as

$$\partial_t \hat{\rho}_{\mu k, \eta \eta'}|_{\text{ee}} = \left\{ -\pi \sum_{q \eta \eta'} S^\mu_{k,k-q,\eta \eta'} \hat{\rho}_{\mu k-q,\eta \eta'} S^\mu_{k,k,q,\eta \eta'} \hat{\rho}_{\mu k,\eta \eta'} \sum_{k_1, k_2, \eta_1 \eta_2} V^\pi(q, \epsilon_{\eta_1 k}\pm q - \epsilon_{\eta_2 k}) \right.$$  

$$\times V^\pi(q, \epsilon_{\eta_1 k}\pm q - \epsilon_{\eta_2 k}) S^\mu_{k_1,k_1+q,\eta_1 \eta_1'} \hat{\rho}_{\mu k_1+q, \eta_1 \eta_1'} S^\mu_{k_2,k_2+q,\eta_2 \eta_2'} \hat{\rho}_{\mu k_2, \eta_2 \eta_2'} \times e^{i(\eta_1 - \eta_2 + \eta_1' - \eta_2')} \delta(\epsilon_{\eta_1 k} - \epsilon_{\eta_2 k} + \epsilon_{\eta_1' k_1+q} - \epsilon_{\eta_2' k_1}) \left\{ \begin{array}{c} \{ \eta \leftrightarrow \eta' \} \\ \text{[} \leftrightarrow \text{]} \end{array} \right\} + \{ \eta \leftrightarrow \eta' \}^* \right\}, \quad (13)$$

$$\partial_t \hat{\rho}_{\mu k, \eta \eta'}|_{\text{ei}} = \left\{ -\pi \sum_{q \eta \eta'} S^\mu_{k,k-q,\eta \eta'} \hat{\rho}_{\mu k-q,\eta \eta'} S^\mu_{k,k,q,\eta \eta'} \hat{\rho}_{\mu k,\eta \eta'} |U(\epsilon_{\eta_2 k} - \epsilon_{\eta_2 k} q)|^2 e^{i(\eta_1 - \eta_2) \omega q t / 2} \right.$$  

$$\times \delta(\epsilon_{\eta_2 k} - \epsilon_{\eta_2 k}) \left\{ \begin{array}{c} \{ \eta \leftrightarrow \eta' \} \\ \text{[} \leftrightarrow \text{]} \end{array} \right\} + \{ \eta \leftrightarrow \eta' \}^* \right\}, \quad (14)$$

$$\partial_t \hat{\rho}_{\mu k, \eta \eta'}|_{\text{ep}} = \left\{ -\pi \sum_{q \eta \eta'} M^\mu_{k,k-q,\eta \eta'} M^\mu_{k,k,q,\eta \eta'} \right.$$  

$$\times (N_{q, \eta}^\pm \hat{\rho}_{\mu k-q,\eta,\eta'} \hat{\rho}_{\mu k,\eta,\eta'} - N_{-q, \eta}^\pm \hat{\rho}_{\mu k-q,\eta,\eta'} \hat{\rho}_{\mu k,\eta,\eta'}) \times e^{i(\eta_1 - \eta_2 + \eta_1' - \eta_2')} \delta(\epsilon_{\eta_2 k} - \epsilon_{\eta_2 k} \pm \omega q \lambda) \right\} + \{ \eta \leftrightarrow \eta' \}^* \right\}, \quad (15)$$

in which $[\leftrightarrow]$ stands for the same term in the previous [] but interchanging $\leftrightarrow$, and $\{ \eta \leftrightarrow \eta' \}^*$ represents the conjugate of the same term in the previous [] except the exchange of $\eta$ and $\eta'$. In equation (15), $N_{q, \eta}^\pm = n_{q, \eta}^\pm + 1/2 \pm 1/2$ with $n_{q, \eta}^\pm$ standing for the distribution function of the phonon in branch $\lambda$; $V^\pi(q, \epsilon_{\eta k+q} - \epsilon_{\eta k}) = [V^\pi(q, \epsilon_{\eta k+q} - \epsilon_{\eta k})]^*; \hat{\rho}_{\mu k} = 1 - \hat{p}_{\mu k}$ and $\hat{p}_{\mu k} = \hat{p}_{\mu k}$. The optical phonons include the transverse optical phonons (KO) near the $K$ ($K'$) point and the longitudinal (LO) as well as transverse optical (TO) phonons near the $\Gamma$ point. The detailed forms of the scattering matrix elements $U(\epsilon_{\eta_2 k} - \epsilon_{\eta_2 k} q)$ and $M^\mu_{k,k-q,\eta \eta'}$ are given in appendix B.

In this work, the $q$-resolved dynamics of the RI and optical phonons is also studied, while the acoustic phonons are set to be in equilibrium with the environment at temperature $T_0$. The kinetic equations of the hot phonons are

$$\partial_t n_{q, \eta}^\lambda = \partial_t n_{q, \eta}^\lambda + \partial_t n_{q, \eta}^\lambda |_{\text{pp}} \quad \text{(16)}$$
in which $\partial n^\lambda_{q | \epsilon p}$ come from the carrier–phonon scattering and $\partial n^0_{q | pp}$ describe the anharmonic decay of hot phonons in relaxation time approximation. They are given by

$$\partial n^\lambda_{q | \epsilon p} = \text{Re} \left[ 2\pi \sum_{k(\xi), q(\xi), \lambda} M^{\mu \nu}_{\lambda | k, q, \lambda, \eta \xi} M^{\mu \nu}_{\lambda | k, k + q, \lambda, \eta \xi} \right. \times \left( \hat{\rho}^\xi_{\mu | k, q, \xi, \eta \xi} \hat{\rho}^\xi_{\mu | k, k + q, \xi, \eta \xi} N^\lambda_{q} - \hat{\rho}^\xi_{\mu | k, q, \xi, \eta \xi} \hat{\rho}^\xi_{\mu | k, k + q, \xi, \eta \xi} N^\lambda_{q} \right) \times e^{i(\eta \xi q + \eta \xi q) n_{\lambda, q}} / 2 \delta (\epsilon_{\eta \xi k} - \epsilon_{\eta \xi k} - \omega q), \right]$$

(17)

$$\partial n^0_{q | pp} = -(n^0_{q} - n^0_{\lambda q}) / \tau_{pp}, \quad \text{(18)}$$

where $\tau_{pp}$ is the phenomenological relaxation time from the phonon–phonon scattering and $n^0_{\lambda, q}$ is the number of the $\lambda$ branch phonons at environmental temperature $T_0$.

Before we show our numerical results, we first give a simple analysis of the Auger process. If only the diagonal terms of the density matrices (i.e. $\eta = \eta'$, $\eta_1 = \eta_2$, $\eta_3 = \eta'$, $\eta_1' = \eta_2'$ and $\eta_3' = \eta_4'$) are involved, the Coulomb scattering terms become

$$\partial f_{\mu k, \eta} = \begin{bmatrix} -2\pi \sum_{q \eta} S^\mu_{k, k - q, \eta \eta_1} f_{\mu k - q, \eta \eta_1} S^\mu_{k, k - q, \eta \eta_1} f_{\mu k - q, \eta \eta_1} \sum_{k \eta_1' \eta_2' \eta_1''} V^\eta (q, \epsilon_{\eta_1' k_1 + q} - \epsilon_{\eta_1'' k_1}) \\
\times V^\eta (q, \epsilon_{\eta_1' k_1 + q} - \epsilon_{\eta_1'' k_1}) S^\mu_{k_1, k_1 + q, \eta_1' \eta_1''} f_{\mu k_1 + q, \eta_1''} f_{\mu k_1 + q, \eta_1'} S^\mu_{k_1, k_1 + q, \eta_1' \eta_1''} f_{\mu k_1 + q, \eta_1''} \delta (\epsilon_{\eta_1 k - q} - \epsilon_{\eta k} + \epsilon_{\eta_1 k + q} - \epsilon_{\eta_1' k_1}) \end{bmatrix} - [> \leftrightarrow <].$$

(19)

From this equation, one finds that it describes the scattering process with one electron being scattered between bands $\eta$ and $\eta_1$ while the other electron between bands $\eta_2$ and $\eta_1'$ under the energy conservation condition $\delta (\epsilon_{\eta_1 k - q} - \epsilon_{\eta k} + \epsilon_{\eta_1 k + q} - \epsilon_{\eta_1' k_1})$. Moreover, the vertices of the two involved scattering processes, $V^\eta (q, \epsilon_{\eta_1' k_1 + q} - \epsilon_{\eta_1'' k_1}) S^\mu_{k_1, k_1 + q, \eta_1' \eta_1''} S^\mu_{k_1, k_1 + q, \eta_1' \eta_1''}$ and $V^\eta (q, \epsilon_{\eta_1' k_1 + q} - \epsilon_{\eta_1'' k_1}) S^\mu_{k, k - q, \eta, \eta_1} S^\mu_{k, k - q, \eta, \eta_1}$, are conjugated with each other. For the Auger-type process here, it means that three out of the four involved band indices ($\eta$, $\eta_1$, $\eta_1'$ and $\eta_2'$) are the same and the left one is different ($\eta \eta_1 \eta_1' \eta_2'$ = $\eta_1$), corresponding to the case where one of the two scattered electrons is transferred from one band to the other while the other electron remains in the same band. Then, taking $\eta_1 = \eta_1' = \eta_2' \neq \eta$ for example and substituting the energy dispersion, the requirement of energy conservation becomes $V_{\eta}(|k - q| + |k| + |k_1 + q| - |k_1| = 0$. Since $|k - q| + |k| \geq |k - q - k| = q = |k_1 + q - k_1| \geq |k_1| - |k_1 + q|$, one finds that the condition

$$V_{\eta} = |\epsilon_{\eta_1' k_1 + q} - \epsilon_{\eta_1'' k_1}| \quad \text{(20)}$$

must be satisfied. Although this condition is obtained in the special case $\eta_1 = \eta_1' = \eta_2' \neq \eta$, one can prove that it is valid as long as the band indices in the delta function are Auger type. On the other hand, it has been shown that, under this condition, the dielectric function under the dynamic screening $\epsilon(q, \epsilon_{\eta_1' k_1 + q} - \epsilon_{\eta_1 k_1})$ also diverges [47, 48, 67, 68]. Therefore, both $V^\eta (q, \epsilon_{\eta_1' k_1 + q} - \epsilon_{\eta_1'' k_1})$ and $V^\eta (q, \epsilon_{\eta_1' k_1 + q} - \epsilon_{\eta_1'' k_1})$ are reduced to zero and the corresponding
processes are forbidden. Nevertheless, if the scatterings involving the off-diagonal terms are taken into consideration, terms like

\[
\pi \sum_{q_1, q_2} \sum_{\eta_1, \eta_2} S^\mu_{k-k, q_1, q_2, \eta_1, \eta_2} \hat{\rho}_{k-k, q_1, q_2, \eta_1, \eta_2} \hat{\rho}_{k-k, q_1, q_2, \eta_1, \eta_2} V^\tau(q, \epsilon, k_1 + q_1 - \epsilon, k_1) \\
\times V^\alpha(q, \epsilon, k_1 + q - \epsilon, k_1) S^\mu_{k-k, q_1, q_2, \eta_1, \eta_2} \hat{\rho}_{k-k, q_1, q_2, \eta_1, \eta_2} \\
\times \hat{S}^\alpha_{k-k, q_1, q_2, \eta_1, \eta_2} \hat{\rho}_{k-k, q_1, q_2, \eta_1, \eta_2} e^{i(\eta - \eta_1 + \eta - \eta_1' - 1/2)} \delta(\eta_1 - \eta_1') \\
\times \delta(\epsilon, k - \epsilon, k + q - \epsilon, k) \delta(\eta_1, \eta_2, \eta_1' \eta_2')
\]  

(21)

also exist in \( \partial \hat{\rho}_{k, \eta} \big|_{\text{ee}} \). Here, \( \delta(\eta_1 - \eta_1') \) indicates an Auger-type scattering process for scattering with the vertex \( V^\tau(q, \epsilon, k_1 + q_1 - \epsilon, k_1) S^\mu_{k-k, q_1, q_2, \eta_1, \eta_2} \hat{\rho}_{k-k, q_1, q_2, \eta_1, \eta_2} \). However, \( \delta(\eta_1 - \eta_1') \) guarantees that the scattering process restricted by the energy conversion (whose vertex is \( V^\alpha(q, \epsilon, k_1 + q - \epsilon, k_1) S^\mu_{k-k, q_1, q_2, \eta_1, \eta_2} \hat{\rho}_{k-k, q_1, q_2, \eta_1, \eta_2} \)) is non-Auger type, which means that the required equation (20) is no longer necessary. Therefore, these scattering terms are permitted. It is noted that these terms exist naturally when the optical dipoles are taken into consideration. In semiconductor optics these terms are absent because the Auger-type Coulomb vertex in the electron–electron Coulomb interaction Hamiltonian is too small and is neglected [65]. Moreover, from the corresponding scattering term (equation (21)), one finds that there is a prefactor \( \exp[i(\eta - \eta_1 + \eta - \eta_1') - 2/2] \) with \( \eta - \eta_1 + \eta - \eta_1' \neq 0 \). Since the other factors in the scattering term all vary slowly with time, the contribution of such a scattering term oscillates very fast when \( \omega_1 \) is large. Then, when the \( \omega_1 \) is much larger than the variation speed of \( \hat{\rho}_{k, \eta} \), this term will have little influence on the dynamics of carriers. However, when the \( \omega_1 \) is small enough (in the order of the variation speed of \( \hat{\rho}_{k, \eta} \)), this kind of Auger process is expected to have a marked influence on the evolution of the electron distribution function. It is noted that the influence of the energy-spectrum correction from the electron–electron interaction on the Auger process involving only the diagonal terms of the density matrices was also investigated in the literature [42, 69, 70]. It was shown that with this energy spectrum corrected from the linear one, the equations \( \eta \eta_1 \eta_1' = -1 \) and \( \epsilon, k - \epsilon, k_1 + q_1 - \epsilon, k_1 = 0 \) are incompatible with each other, indicating that the energy-conservation condition \( \delta(\epsilon, k - \epsilon, k_1 + q - \epsilon, k_1) \) has no Auger-type solutions in equation (19). This means that such an Auger process is forbidden, which is in agreement with our conclusion obtained by using dynamic screening. In contrast, since the delta-function in equation (21) is non-Auger type, the new Auger process proposed in this work, i.e. the Auger process involving the interband coherence, is still valid. We further point out that the investigation performed here is under the linear spectrum approximation. However, if the electron energy is so high that the linear approximation is no longer valid, the Auger process can exist even under dynamic screening [71].

3. Results

The kinetic Bloch equations, (6) and (16), with all of the terms included are numerically solved following the method detailed in [72], except the drift term. In this paper, the drift term is treated up to the third order to suppress the noise (see appendix C). Then, the temporal evolutions of the carrier and phonon distribution functions are obtained. From equation (10), one finds that
The equilibrium chemical potential is taken to be 18.6 meV. According to the fitting of the experimental data here (section 4), when the distribution is isotropic, equation (23) can be simplified to the one given in [13] \( \sigma_{\text{opt}}(t) = -e^2 \left[ f_{k_{\alpha,1}}(t) - f_{k_{\alpha,-1}}(t) \right]/4 \).

\[ f_{\mu k, \eta} = f_{-\mu k, \eta} \text{ and } P_{\mu k} = -P_{-\mu k}. \] Therefore, we only show the results in \( K \) valley (\( \mu = 1 \)) with the valley index \( \mu \) no longer appearing for simplicity. With the evolution of carrier distribution, the temporal evolution of the optical transmission at the probe-photon energy \( \omega_{pr} \) is calculated from [13, 73, 74]

\[ T_{\text{opt}}(t) = \left| 1 + N_{\text{lay}} \sigma_{\text{opt}}(t) \sqrt{\mu_0/\epsilon_0}/(1 + n_{\text{ref}}) \right|^{-2}, \tag{22} \]

where \( n_{\text{ref}} \) is the refractive index of the substrate and \( N_{\text{lay}} \) is the number of graphene layers. Here, the corresponding interband optical conductivity, which is determined by carrier distributions, is given by [13, 55, 75]\(^3\)

\[ \sigma_{\text{opt}}(t) = -\frac{e^2}{4\pi} \int_0^{2\pi} d\theta_{k_{\alpha}} \sin^2(\theta_{k_{\alpha}}) \left[ f_{k_{\alpha,1}}(t) - f_{k_{\alpha,-1}}(t) \right] \tag{23} \]

in which \( |k_{\alpha}| = \omega_{pr}/(2v_F) \) is the resonant absorption point. DT is calculated from \( \Delta T_{\text{opt}}(t)/T^0_{\text{opt}} = [T_{\text{opt}}(t) - T^0_{\text{opt}}]/T^0_{\text{opt}} \), with \( T^0_{\text{opt}} \) representing the transmission before the pump. It is noted that we only calculate the real part of the conductivity here. This is because in the case investigated here, \( N_{\text{lay}} \sigma_{\text{opt}}(t) \sqrt{\mu_0/\epsilon_0}/(1 + n_{\text{ref}}) \) is small and the contribution of the imaginary part of the conductivity to the transmission is much smaller than that from the real part [76]. We present our numerical results with the material parameters given in table 1. The full-width at half-maximum (FWHM) of the pump pulse is chosen to be 100 fs (corresponding to \( \sigma_\mathrm{f} = 42.5 \text{ fs} \)) and \( \tau_{\text{pp}} = 2.8 \text{ ps} \).\(^4\) The equilibrium chemical potential is taken to be 18.6 meV (the corresponding electron density is \( 1.43 \times 10^{11} \text{ cm}^{-2} \)), the environmental temperature is set to be 300 K and the impurities are absent, unless otherwise specified.

\(^3\) When the distribution is isotropic, equation (23) can be simplified to the one given in [13] \( \sigma_{\text{opt}}(t) = -e^2 \left[ f_{k_{\alpha,1}}(t) - f_{k_{\alpha,-1}}(t) \right]/4 \).

\(^4\) According to the fitting of the experimental data here (section 3.1.2), \( \tau_{\text{pp}} = 2.8 \text{ ps} \).
3.1. High pump-photon energy

We first investigate the dynamics of carriers and phonons with high pump-photon energy. The substrate is chosen to be SiO₂. In the calculation, \( \omega_L = 1500 \text{ meV} \) (corresponding to near-infrared light), unless otherwise specified.

3.1.1. Influence of drift term. We first investigate the influence of the drift term on the dynamics of electrons. To make it clearer, the pump process is switched off by excluding \( \mathcal{H}_{\text{Pump}} \) in equation (8). In the calculation, the maximal electric field in equation (3) is taken as \( E_0 = 150 \text{ kV cm}^{-1} \) and the pump-photon energy \( \omega_L = 150 \text{ meV} \). The \( k \) dependences of \( \ln(f_{k,\eta}^{-1} - 1) \) along the \( \mathbf{k}_x \) and \( \mathbf{k}_y \) directions at different times are plotted in figures 1(a) and (b), respectively. Under the small equilibrium chemical potential used in our calculation (18.6 meV, which is even smaller than \( k_B T_0 \)), the dynamics of holes is similar and is not shown here. From the figures, one finds that before the pulse (e.g. \( t = -100 \text{ fs} \)), the electron distribution is the Fermi distribution

\[
\ln(f_{k,\eta}^{-1} - 1) = (\eta v_F |\mathbf{k}| - \mu_\eta)/(k_B T_\eta)
\]  

(24)
as shown in the figure. Here, \( T_\eta \) and \( \mu_\eta \) are the electron temperature and chemical potential in corresponding band \( \eta \), respectively. After \( t = -100 \text{ fs} \), the electrons drift under the influence of the electric field (equation (3)) along the \( \mathbf{x} \) direction. From this oscillating electric field, a net oscillating momentum \( \mathbf{k}_{\text{net}}(t) = \sum_k \mathbf{k}_k f_{k,\eta}(t) / \sum_k f_{k,\eta}(t) \) along the \( \mathbf{x} \) direction is gained by the electrons. As a result, the location of the minimum of \( \ln(f_{k,\eta}^{-1} - 1) \) along the \( \mathbf{k}_x \) directions oscillates around \( k = 0 \), as shown in figure 1(a). However, the minimum along the \( \mathbf{k}_y \) directions remains fixed at \( k = 0 \) but its magnitude oscillates, as revealed in figure 1(b). To further illustrate the influence of the electric field on the net momentum \( \mathbf{k}_{\text{net}}(t) \), we plot the temporal evolution of \( k_{\text{net}} \) in figure 1(d). The electric field \(-E(t)\) is also plotted in the same figure for reference. One finds that its phase is always \( \pi/2 \) in advance of \( k_{\text{net}} \). Moreover, after the pulse (\( t > 100 \text{ fs} \), when \( |E| < 0.1|E_0| \)), the net momentum gained from the electric field tends to zero. In addition, it is found that for curves at \( t = -100, 0 \) and 100 fs in figure 1(a), the magnitudes of the slopes of the curves away from the minima decrease with temporal evolution, indicating that the electrons are heated by the drift pulse (see equation (24)). Then, with negligible net momentum, the isotropic hot-electron Fermi distribution is expected to be established under Coulomb scattering after the pulse. To show this, we plot the \( k \) dependences of \( \ln(f_{k,\eta}^{-1} - 1) \) along the \( \mathbf{k}_x \) and \( \mathbf{k}_y \) directions at \( t = 100 \) and 120 fs in figure 1(c). One finds that at \( t = 100 \text{ fs} \), the curve is almost linear with \( k \) for each direction, which indicates the establishment of the Fermi distribution along that direction. To further show the buildup of the isotropic Fermi distribution, one fits the distribution with equation (24) along the direction \( \theta_k \) and obtains the corresponding hot-electron temperature \( T_{\eta \theta_k} \) and chemical potential \( \mu_{\eta \theta_k} \). Then, the establishment of the isotropic Fermi distribution is identified when \( T_{\eta \theta_k} \) along different directions are very close. Here, it is found that at \( t = 120 \text{ fs} \) the relative difference of the temperature increase \( T_{1\theta_k} - T_0 \) along different directions is less than 15%.

Then, we investigate the influences of the pump-photon energy and impurity density on the thermalization of electrons by the drift term. We plot the pump-photon energy and impurity density dependences of the hot-electron temperature \( T_1 \) in figure 2. With different \( \omega_L \) and \( N_i \), the isotropic Fermi distributions are established at different times. Here, \( T_1 \) is fitted along the \( \mathbf{k}_x \) direction at \( t = 230 \text{ fs} \), when the isotropic Fermi distribution has just been established (relative
Figure 1. (a)–(c) $\ln (f_{k_1}^{-1} - 1)$ as a function of $k$ with $\omega_L = 150$ meV along the $x$ and $y$ directions at different times. Here, the pump process is switched off. The black dotted curve in (c) is $\left( v_F |k| - \mu_1 \right) / (k_B T_1)$ with $T_1 = 333$ K and $\mu_1 = 11.4$ meV. To make the results clearer, the curve for $t = 100$ fs plotted in (c) is $\ln (f_{k_1}^{-1} - 1) + 1$. (d) Temporal evolution of the total momentum $k_{net}(t)$ with $\omega_L = 150$ meV (solid curve with the scale on the left-hand side of the frame). The temporal evolution of the electric field $-E(t)$ is also plotted for reference (dotted curve with the scale on the right-hand side of the frame).

The difference between temperature increase $T_{1|k} - T_0$ along the $k_x$ and $k_y$ directions is less than 10% for the slowest case in our calculation. The impurity density dependence of $T_1$ is obtained with $\omega_L = 300$ and 500 meV. It is shown that $T_1$ decreases monotonically with the increase of the pump-photon energy. Moreover, a peak appears in the impurity density dependence as shown in figure 2(b). These phenomena can be well understood from the Boltzmann equation under the relaxation time approximation: $\frac{\partial}{\partial t} f_{k_{n},\eta} = e|E| \cdot \nabla_k f_{k_{n},\eta} - (f_{k_{n},\eta} - f_{k_{n},\eta}^0) / \tau_p$, with $f_{k_{n},\eta}^0$ being the equilibrium distribution and $\tau_p$ standing for the relaxation time. From this equation, one obtains...
the conductivity under the ac electric field $\mathbf{E} = E_0 \exp (-i\omega_L t)\hat{x}$ as
\[
\sigma(\omega_L) = \frac{e^2 v_F^2}{\pi} \int_0^\infty dk k^2 \frac{\tau_p}{1 + (\omega_L \tau_p)^2} \sum_\eta \left( -\frac{\partial f_{k\eta}}{\partial \epsilon_{k\eta}} \right)
\]
which is the same as the intraband conductivity (which corresponds to the intraband absorption) given under the limit $\omega_L \to 0$ in the literature [40, 55, 74, 84]. From this equation, it is found that the conductivity and hence the energy absorbed from the electric field decreases monotonically with the increase of $\omega_L$ and a peak exists in its $N_i$ dependence. These phenomena can also be understood more physically. One finds from equation (25) that the conductivity tends to zero in the limit of no scattering or infinitely large scattering. This can be comprehended as the effect of the electric field on the electron is totally cancelled in one oscillation period in the case without scattering and the electrons cannot be drifted by the electric field in the limit of infinitely large scattering strength. Then, in the case with finite scattering strength, a peak must appear as shown in figure 2(b). As for the decrease of $T_1$ with increasing $\omega_L$, it is understood as the electron finding it more and more difficult to follow the laser field with the increase of the oscillation frequency.

To further compare the influence from the drift and the pump terms, we also perform the calculation with the pump process included but the drift term excluded. In the calculation, $\omega_L = 500$ meV, $N_i = 0$ and the other parameters remain the same. At the time in figure 2, i.e. $t = 230$ fs, the isotropic Fermi distribution is checked to have already been established and the obtained hot-electron temperature is $T_1 = 792$ K, much larger than 7 K established by the drift term only. In the sample usually reported in the experiments [85, 86], the mobility is larger than $1000$ cm$^2$ V$^{-1}$s$^{-1}$ ($N_i < 2.5 \times 10^{12}$ cm$^{-2}$). Within this impurity density range, the temperature increase due to the drift term is less than 40 K. This is much smaller than that from the pump process (the influence of impurity on the pump process is small with such pump intensity and impurity density range). Moreover, since the thermalization due to the drift effect decreases with increasing $\omega_L$, the drift term is negligible in the case with pump-photon energy higher than 500 meV. This is consistent with the investigation in semiconductors where the energy of the

![Figure 2](http://www.njp.org/)
pump photon is very high due to the large band gap [49]. However, in the case with very low pump-photon energy as we will investigate in the next subsection, the drift term is expected to be very important, especially for the case where the electric field does not change direction during the pulse and a large net momentum is transferred to the electrons via the drift term.

3.1.2. Dynamics of electrons. Before the investigation of electron dynamics, we first calculate the DT and compare it with the experimental data in single-layer graphene by Hale et al [25] (figure 3(a)). It is noted that this experiment has been fitted in our previous work [31]. However, in that paper, the Fermi distribution was assumed to be established all of the time and the hot phonons are averaged with a fitting parameter $E_{\text{max}}$, which describes the upper energy limit of the hot carriers being able to emit phonons. In this paper, with the inclusion of the pump term and the $q$-resolved hot phonons, the buildup of the Fermi distribution can be studied elaborately and the fitting parameter $E_{\text{max}}$ is no longer needed. Then, the influence of the approximations in our previous paper can be checked. In the calculation here, the probe-photon energy $\omega_{\text{pr}} = 1100 \text{ meV}$, the FWHM is taken to be 180 fs and the pump-photon energy $\omega_L = 1500 \text{ meV}$, as indicated in the experiment. It is noted that with $N_i = 0$ and such high pump-photon energy, the drift effect of the laser field is negligible as shown in the previous subsection. In addition to these parameters, the fitting parameters are the phonon relaxation time $\tau_{\text{pp}} = 2.8 \text{ ps}$, the substrate-dependent dimensionless Wigner–Seitz radius $r_s = 1.5$ and the maximal electric field $E_0 = 400 \text{ kV cm}^{-1}$ which leads to the maximal electron density being $3.6 \times 10^{12} \text{ cm}^{-2}$. This electron density is comparable with the experimental estimation and that in our previous paper ($4.6 \times 10^{12} \text{ cm}^{-2}$). It is noted that the photon-excited electron density is so large that the small initial equilibrium electron density has a marginal influence on the results. For the relaxation of DT which begins from $t = 133 \text{ fs}$, as shown in figure 3(a), one finds that our numerical results agree very well with the experimental data with a fast relaxation followed by a slow one with the relaxation times being 0.28 and 1.33 ps, respectively. The discrepancy between our numerical results with the experimental data at the initial increase of the DT is understood to come from the approximation in our model where the interference between the pump and probe pulses is neglected and the Markovian approximation [49] is applied. It is well known that the dynamics of the electrons can be well described under the Markovian approximation when the related timescale is long enough. However, during the ultrafast pump process, the use of the Markovian approximation leads to inaccuracy of the results. Nevertheless, under this approximation, the dynamics of the electrons during the pulse can still be described qualitatively [49]. Based on this consideration, this approximation is widely used in the optical problems in both semiconductor [49, 65, 88, 89] and graphene [34, 36–38]. A more accurate investigation requires non-Markovian kinetics which is beyond the scope of this investigation.

Then, we investigate the dynamics of electrons, especially the buildup of the Fermi distribution under linearly polarized light. We plot $\ln(f_{k,1}^{-1} - 1)$ as a function of $k$ along different directions at different times in figures 3(b)–(d). With the strong scattering and the negligible drift effect, the distribution is center symmetric and hence the distribution functions along the $-x$ and $-y$ directions are not shown. Besides, the dynamics of the holes is almost the same [31] (due to the low equilibrium chemical potential) and is not plotted here. From figure 3(b), one finds that the distribution of the pumped electrons is anisotropic and mainly around $k = 1.14 \text{ nm}^{-1} \approx \omega_L/(2\hbar v_F)$ along the $y$-axis as shown by the valley there. This comes

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5 We have checked that in the parameter range investigated in this paper, the multi-photon process is irrelevant.
Figure 3. (a) DT from the numerical results compared with the experimental data in single-layer graphene [25]. The results are normalized as Hale et al [25]. The arrow in the figure indicates $t = 133$ fs, which is the beginning of the relaxation. (b)–(d) $\ln(f_{k,1}^{-1} - 1)$ as a function of $k$ along the x and y directions at $t = -90$, 0, 100 and 131 fs. The black dotted curves in (b) and (c) are the case before the pump pulse. The curve for $t = 100$ fs plotted in (d) is $\ln(f_{k,1}^{-1} - 1) + 1$. The cases without the Auger process are also plotted in (b)–(d). The comparison of the electron distributions $f_{k,1}$ at $t = -100$ (black dotted curve) and 131 fs (red solid curve) are plotted in the inset.

from the linearly polarized pump light as shown by $\sin \theta_k$ in equation (11). During the pump process, the scattering also plays an important role by smearing out the valleys and making the distribution tend to be isotropic as shown in figures 3(b) and (c). Moreover, it is found that the slopes of the curves around the Dirac point decrease with the temporal evolution, indicating their thermalization under the scattering. At $t = 131$ fs shown in figure 3(d), one finds that the
curves become almost linear with \( k \) for each direction. By fitting the curves with equation (24), one obtains the hot-electron temperature \( T_{\text{h}} \) and chemical potential \( \mu_{\text{h}} \) along the \( x \) direction as well as \( T_{\text{h}} \) and \( \mu_{\text{h}} \) along the \( y \) direction. Further fitting the distribution of electrons in the valence band (not shown), one obtains \( T_{\text{h}} = 1738.5 \text{ K} \) and chemical potential \( \mu_{\text{h}} = 49 \text{ meV} \) along the \( x \) direction as well as \( T_{\text{h}} = 1773.4 \text{ K} \) and chemical potential \( \mu_{\text{h}} = 47.5 \text{ meV} \) along the \( y \) direction. From these results, one finds that the relative differences along the \( x \) and \( y \) directions for both \( T_{\text{h}} \) and \( \mu_{\text{h}} \) in band \( \eta \) are very small (less than 4.2\%). Moreover, the chemical potentials in the valence band are almost opposite to those in the conduction band. These results indicate that the distribution established at \( t = 131 \text{ fs} \) is an isotropic one with separate chemical potentials for electrons in conduction and valence bands. This is understood due to the fact that the recombination and generation of electrons and holes is much weaker than the intraband thermalization. Furthermore, noting that the pulse (equation (3)) ends after \( t = 165 \text{ fs} \) (when \( |E| < 0.1|E_0| \)), one concludes that in the case with such a long pump pulse width, such a hot-electron Fermi distribution is established during the pulse. Considering that the relaxation of the DT begins at 133 fs which is after the establishment of the Fermi distribution, the approximation of the buildup of the Fermi distribution in our previous work is acceptable. However, this approximation leads to the inaccuracy of the hot-electron temperature (3200 K at the beginning of the relaxation in our previous work [31]).

To further show the influence of the Auger process involving the interband coherence, we also plot the results without the Auger process in figures 3(b)–(d). One finds that the exclusion of the Auger process has little influence on the evolution of the distribution function. This is in agreement with our argument in section 2 that the Auger process that survives dynamic screening in the presence of the optical polarization does not contribute due to the high pump-photon energy. Similarly, also due to the high pump energy, the rotating-wave approximation is checked to be valid (not shown in the figure).

3.1.3. Dynamics of phonons. With the inclusion of the \( q \)-resolved hot phonons, we are able to give a more detailed investigation on the dynamics of phonons than our previous work [31]. We calculate the temperature \( T_{\text{ph}}(q) \) for phonons with momentum \( q \) at branch \( \lambda \) by \( T_{\text{ph}}(q) = \omega_{\lambda}/|k_{\lambda}\ln(1 + 1/n^2_{\lambda})| \) and then, plot them in momentum space at \( t = 131 \text{ fs} \) for the KO and RI phonons in figures 4(a) and (b). The \( q \)-resolved temperatures of the other phonons are similar to these two phonon branches. It is shown that due to the anisotropic distribution of electrons, the thermalization of phonons under the electron–phonon scattering is also anisotropic. Moreover, it is found that the thermalization of phonons with small \( q \) (\( q < \omega_{q\lambda}/v_F \), which are the phonons within the black dotted circle) is weaker than those with \( q > \omega_{q\lambda}/v_F \).

This can be understood from the energy conservation during the electron–phonon scattering process (equation (15)), \( \delta(\varepsilon_{\eta_2,q-k-q} - \varepsilon_{\eta_1,k} \pm \omega_{q\lambda}) \). By using the relation \( v_F|k - q - k| = v_F|k - q - k| = v_F q \), one finds that the phonons with \( q < \omega_{q\lambda}/v_F \) and \( q > \omega_{q\lambda}/v_F \) are involved in the interband (\( \eta_2 \neq \eta_3 \)) and intraband (\( \eta_2 = \eta_3 \)) electron–phonon scattering, separately. Therefore, the weaker thermalization of the phonons with \( q < \omega_{q\lambda}/v_F \) means that before the establishment of the hot-electron Fermi distribution (\( t \leq 131 \text{ fs} \)), the recombination of electrons and holes due to the interband electron–phonon scattering is weaker. In order to show this clearly, we plot the \( q \) dependence of the angle average phonon temperature \( T_{\text{ph}}^q = \int_0^{2\pi} dq q T_{\text{ph}}(q)/(2\pi) \) at different times in figures 4(c)–(f), as done by Knorr and co-workers [33, 36]. The locations \( v_F q = \omega_{q\lambda} \) for each branch of phonons are indicated by arrows in figures 4(c)–(f). It is found that in the first hundred femtoseconds, only the thermalization
Figure 4. (a) and (b) Typical $\mathbf{q}$-resolved hot-phonon temperatures of KO and RI$_1$ phonons at $t = 131$ fs, respectively. The black dotted circles indicate $q = \omega_{q\lambda}/v_F$. (c)–(f) The angle averaged hot-phonon temperatures for different phonon branches as a function of $v_F q$ at $t = -90, 25, 131$ and 300 fs, respectively. The arrows indicate the location of the corresponding phonon energy $v_F q = \omega_{q\lambda}$. It is noted that the phonon energies of TO and LO phonons are degenerate and we only plot the arrow for the TO phonons here. The region close to $v_F q = 0$ is enlarged in the inset for KO and RI$_2$ phonons at $t = 300$ fs.

of the phonons with $v_F q > \omega_{q\lambda}$ is efficient. After that, the temperatures of the phonons with $v_F q < \omega_{q\lambda}$ increase markedly. In particular, for the KO and RI$_2$ branches, the hottest phonons appear among the phonons with $v_F q < \omega_{q\lambda}$, as shown in the inset of figure 4(f). This is consistent with the results by Knorr and co-workers [33, 36] and can be understood as the interband electron–phonon scattering happens only among the low-energy electrons with its energy $|\epsilon_{q\lambda}| < \omega_{q\lambda}$. However, the energies of the photoexcited electrons are very high ($\omega_L/2 > 3.9\omega_{q\lambda}$) and before the low-energy electrons are heated under the scattering, the energy gained by the electrons first relaxes through the intraband electron–phonon scattering process. Then, after the thermalization of the low-energy electrons, the heating of the phonons with $v_F q < \omega_{q\lambda}$ becomes important.

One also observes from figures 4(a) and (b) that the hottest phonons are along the $\mathbf{q}_y$ direction. This comes from the angular dependence of the electron–phonon scattering matrices. Since the hottest phonon appears with $q > \omega_{q\lambda}/v_F$, we only consider the intraband scattering process. Then, with only the diagonal terms of the density matrices taken into consideration, the scattering matrices in equation (15) $|M_{\mu\nu;\lambda}^{k,k-q}\eta\eta}|^2$ are proportional to $1 - \cos(\theta_k - \theta_{k-q})$ for the KO phonons and $1 + \cos(\theta_k - \theta_{k-q})$ for the RI$_1$ phonons [64, 79, 80]. One immediately understands that the parallel or antiparallel scattering is the strongest. Then, since the
photoexcited electrons are mainly along the $\mathbf{k}$ direction, the hottest phonons, which come from the electron–phonon scattering, also have to be along the $\mathbf{y}$ direction as shown in the figures. In addition, figures 4(c)–(f) show that the thermalized phonons in KO and LO branches occupy a larger momentum range than the other phonons. This also comes from their different angular dependences of the scattering matrices as revealed by Malic et al [36].

3.2. Low pump-photon energy

We then investigate the dynamics of electrons and phonons with the pump-photon energy $\omega_L$ down to the THz regime. The maximal pump field in equation (3) is taken as $E_0 = 5 \text{kV cm}^{-1}$. The substrate is again chosen to be SiO$_2$.

3.2.1. Dynamics of electrons. In this subsection, we investigate the dynamics of electrons with $\omega_L = 15 \text{ meV} \ (3.6 \text{ THz})$. With such a low pump-photon energy, the corresponding oscillation period of the electric field is 276 fs, which is even larger than twice the FWHM. Therefore, during the pump process, the laser field $E$ almost keeps along one direction and a large net momentum is transferred to the electrons. In this case, the drift term is very important since it describes the momentum transfer. Furthermore, due to the transferred net momentum, a drifted Fermi distribution after the pulse is expected. In previous works with static electric field [46, 90], it has been shown that in the case with strong Coulomb scattering, the drifted Fermi distribution is expressed as

$$f_{k\eta} = \{\exp[(\eta v_F|\mathbf{k} - \mathbf{u}_\eta| - \mu_\eta)/(k_B T_\eta)] + 1\}^{-1} \quad (26)$$

with $\mathbf{u}_\eta$ being the drift velocity. However, in graphene, it has been shown by Zhou and Wu [46] that the Coulomb scattering is not strong enough compared with the drift effect of the static electric field and the drifted Fermi distribution is instead described by

$$f_{k\eta} = \{\exp[(\eta v_F|\mathbf{k} - \mathbf{u}_\eta| - \mu_\eta)/(k_B T_\eta)] + 1\}^{-1}. \quad (27)$$

Although both distributions are the same in semiconductors with parabolic spectrum, they have different properties in graphene due to the linear dispersion. For equation (27), one finds that $\mathbf{u}_\eta = \sum_k \mathbf{k} f_{k\eta}(t)/\sum_k f_{k\eta}(t)$ is nothing but the center-of-mass drift velocity. However, in equation (26), $\mathbf{u}_\eta$ is only an effective parameter. Moreover, in the $k$ dependence of $\ln(f_{k1}^{-1} - 1)$ along the direction of $\mathbf{u}_\eta$, equation (26) indicates a minimum at $k = 0$ and the magnitudes of the slopes for $\theta_k$ and $\theta_k + \pi$ directions are different. However, equation (27) shows that the minimum deviates from $k = 0$ and the magnitudes of the slopes are identical. To reveal the distribution established here and the dynamics of electrons, we plot $\ln(f_{k1}^{-1} - 1)$ as functions of $k$ along the direction of the electric field $E$ (i.e. $\theta_k = 0$ and $\pi$) at different times in figure 5(a). One finds that during the pump process, the minimum of the curve first drifts away from the Dirac point as shown by the curves at $t = -50$ and 25 fs. However, before the end of the pulse ($t > 100 \text{ fs}$, when $|E| < 0.1|E_0|$), the minimum has already started to tend to the Dirac point and the magnitudes of the slopes for the curves away from Dirac point become different. Then, at $t = 300 \text{ fs}$, the minimum is around the Dirac point and the curves along $\theta_k = 0$ and $\pi$ directions are almost linear with $k$, but with different magnitudes of slopes. With such different magnitudes of slopes, the corresponding drift of the center-of-mass $k_{\text{net}}$ is $0.012 \text{ nm}^{-1}$. This indicates the establishment of the distribution equation (26), as shown by the fitting plotted as the black dotted lines in the figure. It is understood that the drifted Fermi distribution is established after
Figure 5. (a) $\eta \ln (f_{k,n}^{-1} - 1)$ along $\theta_k = 0$ and $\pi$ directions for conduction band electrons ($\eta = 1$) at $t = -50, 25, 100, 300$ and $500$ fs, as well as that for valence band electrons ($\eta = -1$) at $t = 300$ fs. Here, $\omega_L = 15$ meV and the impurity density $N_i = 0$. The black dotted curve is calculated from equation (26) with $T_1 = 302.97$ K, $u_1 = 0.093v_F$ and $\mu_1 = 18.5$ meV. (b) Temporal evolution of $T_{\eta\theta_k}$ and $u_{\eta\theta_k}$ (with the scale on the right-hand side of the frame) for electrons in conduction (solid curves) and valence (dashed curves) bands obtained by fitting equation (26) along directions $\theta_k = 0$ and $\pi$ (red squares), as well as $\theta_k = \pi/4$ and $5\pi/4$ (blue dots).

the pulse in our present investigation. Then, with the Coulomb scattering being the dominant scattering, the distribution as equation (26) is established.

It is also shown in figure 5(a) that at $t = 300$ fs, the similar drifted hot-electron Fermi distribution in the valence band ($\eta = -1$) has also been established since the minimum of the corresponding curve is around $k = 0$ and the curves along $\theta_k = 0$ and $\pi$ directions are almost linear with $k$ with different magnitudes of slopes. Moreover, the distributions along the other directions $\theta_k$ are also similar. To further investigate the buildup of the unitary drifted Fermi distribution along all directions, we fit the distributions of electrons along different directions in both the conduction and the valence bands with equation (26). The obtained $T_{\eta\theta_k}$ and $u_{\eta\theta_k}$ are plotted in figure 5(b). One finds that after $t = 350$ (420) fs, the relative difference between the drift velocities along different directions for electrons in conduction (valence) band is less than 10% and the relative difference of the temperature increases $T_{\theta_k} - T_0$ ($T_{\theta_k} - T_0$) is less than 10%. This indicates the buildup of the unitary drifted Fermi distribution as equation (26) for all directions in conduction (valence) band. It is noted that the case studied here is n-doped, which leads to different buildup times, hot-electron temperatures and drift velocities of the unitary drifted Fermi distributions for electrons in conduction and valence bands. However, under the interband Coulomb scattering, the temperatures and drift velocities in conduction and valence bands tend to be identical. From our calculation, the relative difference of the drift velocities is less than 10% after $t = 550$ fs and that of the temperature increase is less than 10% after $t = 350$ fs, as shown in the figure.
We also investigate the influence of impurities and pump-photon energy on the drift velocity of electrons. $u_{1\theta k}$ obtained from the distributions along $\theta_k = 0$ and $\pi$ directions under different pump-photon energies and impurity densities are plotted in figure 6(a). It is noted that after $t = 350$ fs, the relative differences of the drift velocities along different directions are less than 7% for all of the cases investigated here. One finds that in the case without impurity, the drift velocity $u_{1\theta k}$ and hence the total momentum relax slowly (with the relaxation time being about 1 ps). However, when the impurities with $N_i = 5 \times 10^{11}$ cm$^{-2}$ (the corresponding mobility is about 5000 cm$^2$ V$^{-1}$ s$^{-1}$) are introduced into the system, the relaxation of the total momentum becomes very fast (with the relaxation time being 27 fs) and at $t = 350$ fs, the corresponding $u_{1\theta k} = 0.0005v_F$ is negligible. This is because the relaxation of the drift velocity comes from the electron–impurity and electron–phonon scatterings. However, the electron–phonon scattering strength is weak and only when the impurity density is large (e.g. $5 \times 10^{11}$ cm$^{-2}$) can the relaxation of the net momentum become fast. Besides, it is also shown in the figure that when $\omega_L$ increases from 15 to 30 meV, the value of $u_{1\theta k}$ decreases markedly. This is because with $\omega_L = 30$ meV, the laser field $E$ changes direction during the pulse and the net momentum gained by the electrons is partially cancelled.

We further investigate the influence of the pump and Auger processes on the evolution of the electron density by switching them off separately. The results are plotted in figure 6(b). In the calculation, $\omega_L = 15$ meV and $N_i = 0$. It is noted that without the pump process, the electrons can still be excited from the valence band to the conduction band as indicated by Tani et al [39]. This is because the electrons are heated by the drift effect of the laser field, which leads to the interband electron–phonon scattering. Moreover, it is also found that the Auger process indeed affects the dynamics of electrons and it leads to the change of the excited electron density at the end of the pulse ($t = 100$ fs) as large as 70%, which is consistent with our analysis in section 2. Furthermore, with such a low pump-photon energy, the widely accepted rotating-wave approximation in semiconductors optics is no longer valid. The temporal evolution of the photoexcited electron density under the rotating-wave approximation is also plotted in

Figure 6. (a) Temporal evolution of $u_{1\theta k}$ for the cases with $\omega_L = 15$ and 30 meV without impurity, as well as $\omega_L = 15$ meV with impurity density $N_i = 5 \times 10^{11}$ cm$^{-2}$. (b) Temporal evolution of the electron densities for the cases with all the processes included, without Auger process, without pump process and under the rotating-wave approximation, separately. Here $\omega_L = 15$ meV and $N_i = 0$. 

We further investigate the influence of the pump and Auger processes on the evolution of the electron density by switching them off separately. The results are plotted in figure 6(b). In the calculation, $\omega_L = 15$ meV and $N_i = 0$. It is noted that without the pump process, the electrons can still be excited from the valence band to the conduction band as indicated by Tani et al [39]. This is because the electrons are heated by the drift effect of the laser field, which leads to the interband electron–phonon scattering. Moreover, it is also found that the Auger process indeed affects the dynamics of electrons and it leads to the change of the excited electron density at the end of the pulse ($t = 100$ fs) as large as 70%, which is consistent with our analysis in section 2. Furthermore, with such a low pump-photon energy, the widely accepted rotating-wave approximation in semiconductors optics is no longer valid. The temporal evolution of the photoexcited electron density under the rotating-wave approximation is also plotted in...
3.2.2. Dynamics of phonons. We further investigate the dynamics of phonons. We plot the \( \mathbf{q} \)-resolved temperatures of the hot RI\(_1\) and KO phonons with \( \omega_L = 15 \) meV under the impurity densities \( N_i = 0 \) and \( 5 \times 10^{11} \) cm\(^{-2}\) in figure 7. The other phonons are similar and not shown. The temperatures of these two branches of phonons are obtained at \( t = 350 \) fs when the unitary distribution of electrons for all directions has been established. From the figures, one finds that in the case without impurity, the \( \mathbf{q} \)-resolved temperatures of the hot phonons are highly anisotropic. Moreover, by considering that \( \mathbf{u}_p \) in both conduction and valence bands are along the \( \theta_k = 0 \) direction, it is shown that the hottest phonons appear in the direction parallel to \( \mathbf{u}_p \) and the coldest phonons antiparallel to \( \mathbf{u}_p \). This phenomenon is also shown in semiconductors under static electric field [91]. It can be understood from the generation rate of phonons due to the electron–phonon scattering (equation (17)) after the establishment of the drifted Fermi distribution of electrons. With only the diagonal terms of the density matrices taken into

![Figure 7](http://www.njp.org/)

**Figure 7.** (a) and (b) ((c) and (d)) \( \mathbf{q} \)-resolved hot-phonon temperatures with \( N_i = 0 \) (5 \( \times 10^{11} \)) cm\(^{-2}\) at \( t = 350 \) fs for RI\(_1\) and KO phonons, respectively.

figure 6(b). One finds that the fast oscillation of the electron density during the pump process disappears and the excited electron density at the end of the pulse changes about 2.6 times.
consideration, equation (17) is given by
\[
\partial_t n_q^{\pm} = 2\pi \sum_{\mu_1, \mu_2} |M^{\mu_1 \mu_2}_{k+q, k, \eta_1, \eta_2}|^2 \delta(\epsilon_{\eta_1 k+q} - \epsilon_{\eta_2 k} - \omega_{q, \lambda}) (f^{\lambda}_{\mu_1 k, \eta_1} f^{\lambda}_{\mu_2 k+q, \eta_2} N_{q}^{\pm} - f_{\mu_1 k, \eta_1} f_{\mu_2 k+q, \eta_2} N_{q}^{\mp}).
\]

Substituting the drifted Fermi distribution equation (26) and the Bose distribution of phonons \( n_q^{\pm} = 1/[\exp[\omega_{q, \lambda}/(k_B T)] - 1] \) into it, one finds that whether the phonons are emitted or absorbed is determined by the sign of the term
\[
\exp[q \cdot \mu (k+q, \eta) - q \cdot \mu (k, \eta) - \mu_u (k+q, \eta) + \mu_u (k, \eta)]/(k_B T) \leq 1 - \exp[q \cdot \mu (k+q, \eta) - q \cdot \mu (k, \eta) - \mu_u (k+q, \eta) + \mu_u (k, \eta)]/(k_B T).
\]

Here, to simplify the analysis, the temperatures of phonons and electrons are taken to be the same \( T \) and the chemical potentials and drift velocities for electrons in different bands are also set to be unified \( \mu \) and \( u \), respectively. Then, due to the energy conservation \( \delta(\epsilon_{\eta_1 k+q} - \epsilon_{\eta_2 k} - \omega_{q, \lambda}) \), equation (29) is simplified to \( \exp[q \cdot \mu (k+q, \eta) - q \cdot \mu (k, \eta) - \mu_u (k+q, \eta) + \mu_u (k, \eta)]/(k_B T) \leq 1 - \exp[q \cdot \mu (k+q, \eta) - q \cdot \mu (k, \eta) - \mu_u (k+q, \eta) + \mu_u (k, \eta)]/(k_B T) \). One immediately understands that the hottest phonons appear among phonons with \( \mu \) parallel to \( q \) and the coldest phonons appear among phonons with \( \mu \) antiparallel to \( q \) as shown in figures 7(a) and (b). This phenomenon can also be understood physically from the relaxation of the drift velocity of electrons. During the relaxation, the net momentum gained from the laser field is transferred from the electrons to the phonons, which implies the tendency of the absorption of the phonons with \( q \cdot u < 0 \) and the emission of the phonons with \( q \cdot u > 0 \). On the other hand, in the system where the net momentum relaxes very quickly through the electron–impurity scattering, the distributions of phonons become more isotropic as shown in figures 7(c) and (d).

3.3. Negative differential transmission in system with high Fermi energy

In this subsection, we further investigate negative DT due to the weakening of Pauli blocking. Sun et al [14] measured the DT with high probe-photon energy (> 500 meV) and showed that a peak exists in the temporal evolution of DT. Moreover, it is also found that there is a range of the probe-photon energy where negative DT appears during the evolution. To understand this, they suggested that their epitaxial sample consists of a stack of graphene layers [13, 92, 93] with some layers heavily doped and some layers undoped. Moreover, the Fermi energy of the heavily doped layers is assumed to be around half of the upper boundary of the probe energy range for the appearance of negative DT. Then, with the energies of the resonant absorption states of the probe pulse smaller than the Fermi energy in the heavily doped layers, the corresponding distribution difference \( f_{k^{\pm} - 1} - f_{k^{\pm} + 1} \) in equation (23) increases in the undoped layers but decreases in the heavily doped layers after the pulse. This leads to the decrease of the conductivity in undoped layers but increase in heavily doped ones. These two opposite tendencies compete in the evolution and are responsible for the peak and the negative DT. Moreover, with the decrease of the probe-photon energy, the variation of the conductivity from the undoped layers is strengthened whereas that from the heavily doped layers is weakened. Furthermore, if the probe-photon energy is higher than twice the Fermi energy in the heavily doped layers, the conductivities in both the heavily and undoped layers decrease. These properties lead to the fact that the negative DT appears only in a probe-energy range smaller than twice the Fermi energy.

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To check the above conjecture, we fit their experimental data with our microscopic approach. The substrate is chosen to be SiC, the environmental temperature is 50 K, the FWHM of the pulse is 100 fs and the pump-photon energy $\omega_L = 1550$ meV as indicated by Sun et al [14]. The material-dependent Wigner–Seitz radius $r_s = 0.9$ and the maximal electric field in equation (3) is taken to be $E_0 = 175$ kV cm$^{-1}$. With such a pump intensity, the photoexcited electron density is about $2.5 \times 10^{11}$ cm$^{-2}$ per layer. The probe-photon energy $\omega_{pr} = 550$ meV is the same as that in the experiment. It is noted that with such a high probe energy, the intraband absorption is suppressed [75]. To model the multilayer structure in their sample, $N_{lay} \sigma_{ap}(t)$ in equation (22) is replaced by $N_h \sigma_h(t) + N_u \sigma_u(t)$ with $\sigma_h(t)$ and $\sigma_u(t)$ being the conductivity in heavily and undoped graphene, respectively and $N_h$ and $N_u$ being the corresponding layer numbers. In our computation, $\sigma_h(t)$ and $\sigma_u(t)$ are calculated separately with the corresponding equilibrium Fermi energy being $\mu_0 = 350$ and 0 meV, respectively, as estimated in the experiment. $N_h = 1$ and $N_u = 29$ are taken as fitting parameters which are very close to those estimated in the experiment. The results are plotted in figure 8(a). One finds that

Figure 8. (a) DT from the numerical results compared with the experimental data extracted from figure 3(a) in [14]. The inset shows the electron distributions before ($t = -100$ fs) and after ($t = 200$ fs) the pulse in the heavily doped and undoped layers. (b) Temporal evolution of DT under different probe-photon energies.
our numerical results repeat the peak and the negative DT shown by the experimental data. Especially, when \( t > 500 \text{ fs} \), the fitting is quite good. The discrepancy between our numerical results and the experimental data for the magnitude of the peak is again from the approximation mentioned in section 3.1.2 as well as the simplification of the complex layer configuration to two kinds of doped layers. The electron distributions before and after the pulse are plotted in the inset of figure 8(a), which is consistent with the arguments of Sun et al [14]. Moreover, when the probe-photon energy is small enough or larger than twice the Fermi energy, the DT remains positive, as shown by the curves with \( \omega_{pr} = 520 \) and 710 meV in figure 8(b). Based on these results, our microscopic investigation supports their argument that the negative DT comes from the weakening of the Pauli blocking in the heavily doped layers.

4. Summary

In summary, we have investigated the non-equilibrium dynamics of carriers and phonons in graphene under linearly polarized near-infrared and THz laser pulses via the microscopic kinetic Bloch equation approach [49, 50]. The energy dispersion is described under the linear approximation. Moreover, the carrier–impurity, carrier–phonon and carrier–carrier Coulomb scatterings are explicitly included and the dynamic screening for Coulomb scattering is utilized. In addition, based on the vector potential gauge and the gauge invariant approach [49], both the drift and pump terms are naturally included in our approach.

We first investigate the thermalization of electrons due to the drift term. It is shown that the thermalization is weakened with the increase of the pump-photon energy and it is even negligible when the pump-photon energy is higher than 500 meV (with the corresponding oscillation period being much smaller than the pulse width and the scattering time). Moreover, a peak appears in the impurity density dependence of the thermalization. Then, the dynamics of carriers and phonons with the pump-photon energy up to the near-infrared regime is studied. We show that the temporal evolution of the DT reported by Hale et al [25] can be well fitted. Moreover, under the linearly polarized laser pulse, the electrons are photoexcited anisotropically. However, their distribution tends to be isotropic under the scattering. Then, the isotropic hot-electron Fermi distribution with separate conduction- and valence-band chemical potentials is found to be established within \( t = 131 \text{ fs} \), which is before the end of the pulse (the FWHM of the pulse is 180 fs). This is consistent with the previous works [26, 31, 36]. In contrast to the high pump-photon energy case, in the case with the pump-photon energy down to the terahertz regime (with the corresponding period larger than the pulse width investigated here), we show that a large net momentum is transferred to the electrons through the drift term and a drifted Fermi distribution different from the one under the static field is established in several hundred femtoseconds [46]. We further show under the linear energy spectrum approximation that the Auger process investigated in the previous works [34, 35, 37], which only involves the diagonal terms of density matrices, is forbidden by the dynamic screening. However, the Auger process involving the interband coherence contributes to the dynamics of carriers. Nevertheless, its influence is important only when the pump-photon energy is low (comparable to the variation speed of the distribution). In contrast, for the rotating-wave approximation which is widely accepted in semiconductor optics, we show that it fails when the pump energy is low. We also apply our microscopic theory to investigate the negative DT in the case where the equilibrium Fermi energy is high and the probe-photon energy is around twice the Fermi energy. We show
that the negative DT appears due to the weakening of Pauli blocking under the pump pulse, which supports the suggestion by Sun et al [14] in their experimental work.

In addition, the momentum-resolved hot-phonon temperatures are also studied. When the pump-photon energy is high, the hot phonon temperatures are anisotropic due to the linearly polarized laser field and the hottest phonons appear with their momentum in the direction perpendicular to the laser field $E$. Moreover, we also show that the electron–hole recombination due to the interband electron–phonon scattering is weak in the first hundred femtoseconds. On the other hand, when the pump-photon energy is low and the relaxation of the net momentum is slow, the hot-phonon temperatures are also highly anisotropic with the hottest and the coldest phonons appearing in the directions parallel and antiparallel to the drift velocity, respectively, due to the drifted Fermi distribution of electrons. However, if the relaxation of the net momentum is fast as in the case with high impurity density, the hot-phonon temperatures become more isotropic.

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**Appendix A. Gauge invariant kinetic Bloch equations**

We use the non-equilibrium Green function approach to derive the gauge invariant kinetic Bloch equations under the optical field [49, 50]. The effective Hamiltonian without external field in the real space is given by [55]

$$H^\mu_{0,t}(-i\nabla) = v_F(-i\mu \tau_x \nabla_x - i\tau_y \nabla_y)$$  \hspace{1cm} (A.1)

and the electron–optical-field interaction Hamiltonian is described by [55]

$$H^\mu_{\text{photon},t}(t) = |e|v_F[\mu \tau_x A_x(t) + \tau_y A_y(t)]/c.$$  \hspace{1cm} (A.2)

Then, in the base set of the eigenstates of $H^\mu_{0,t}(-i\nabla)$, the above Hamiltonians read

$$H^\mu_{0,t}(\mathbf{p}) = v_F p \sigma_z,$$  \hspace{1cm} (A.3)

$$H^\mu_{\text{photon},t}(\mathbf{p}, t) = |e|v_F A(t) \cdot (p \sigma_x + \mu \hat{\mathbf{z}} \times p \sigma_y)/(cp)$$  \hspace{1cm} (A.4)

with $p_x = -i\partial_x$, $p_y = -i\partial_y$ and $p = \sqrt{p_x^2 + p_y^2}$. It is noted that this electron–optical-field interaction Hamiltonian is consistent with the previous works based on the tight-binding approach [56, 57].

The contour-ordered Green function $G^\mu(1, 2)$ are matrices with the matrix elements defined as $G^\mu_{\eta \eta'}(1, 2) = -i\langle T_c [\psi^\dagger_{\eta'}(1) \psi^\mu(2)] \rangle$. Here, $\psi^\mu_{\eta'}$ and $\psi^\mu_{\eta'}^\dagger$ are the Fermion field operators in the Heisenberg picture for electrons in band $\eta$ and valley $\mu$; $(1) = (\mathbf{r}_1, t_1)$ and $(2) = (\mathbf{r}_2, t_2)$ stand for the space–time points on the contour and $G^\mu_0$ represent the free-particle Green function. By multiplying $i \partial_n - H^\mu_{0,t}(\mathbf{p}_{1})$ and $-i \partial_n - H^\mu_{0,t}(\mathbf{p}_{2})$ to the Dyson equation of $G^\mu(1, 2)$ and subtracting the two obtained equations from each other, one obtains the equation with

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\( \mathbf{r}_1, \mathbf{r}_2 \) and \( \mathbf{t}_1, \mathbf{t}_2 \). Then, transforming the equation to the center-of-mass and relative variables \( \mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2, \mathbf{T} = (\mathbf{t}_1 + \mathbf{t}_2)/2, \mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2 \) and \( \tau = t_1 - t_2 \) and omitting \( \mathbf{R} \) due to the uniformity of the system studied here, one obtains [49]

\[
\begin{align*}
& i\partial_\tau G^\mu(\tau, T, \mathbf{r}) = [H^\mu_{0,\tau}(\mathbf{p}_r) + H^\mu_{\text{photon,}r}(\mathbf{p}_r, T + \tau/2)]G^\mu(\tau, T, \mathbf{r}) - G^\mu(\tau, T, \mathbf{r})[H^\mu_{0,\tau}(\mathbf{p}_r) + H^\mu_{\text{photon,}r}(\mathbf{p}_r, T - \tau/2)].
& \quad \text{(A.5)}
\end{align*}
\]

Here \( \mathbf{p}_r \) represents the momentum operator with respect to the relative variable. The gauge invariant functions are constructed as [49]

\[
G^\mu(\mathbf{k}, \tau, T) = \int d^2r \exp(-i\mathbf{k} \cdot \mathbf{r})G^\mu(\tau, T, \mathbf{r})
\]

with \( \mathbf{k} = \mathbf{k} - \int_{-1/2}^{1/2} d\lambda|e|A(T + \lambda \tau)/c. \) Applying this transformation to equation (A.5) and setting \( \tau = 0^+ \), one has

\[
\partial_\tau \rho_{\mu k}(T) + |e|\partial_\tau A(T) \cdot \nabla_k \rho_{\mu k}(T)/c + i[H^\mu_{0,}\mathbf{k} + H^\mu_{\text{photon,}r}(\mathbf{k}, T), \rho_{\mu k}(T)] = 0
\]

(A.7)

with \( \rho_{\mu k}(T) = -iG^\mu(\mathbf{k}, 0^+, T) \) being the local density matrix of electrons with momentum \( \mathbf{k} \) and \( H^\mu_{\text{photon,}r}(\mathbf{k}, T) \) coming from the electron–optical-field interaction given by equation (5). \( |e|\partial_\tau A(T) \cdot \nabla_k \rho_{\mu k}(T)/c \) comes from the transformation of \( \partial_\tau G^\mu(\tau, T, \mathbf{r}) \) and is just the drift term since \( \mathbf{E}(T) = -\partial_\tau A(T)/c \) under the vector potential gauge. Moreover, the derivation here is based on the assumption that the field \( \mathbf{A} \) is small and can be treated perturbatively. Then, expanding \( |\mathbf{k}| \) around \( |\mathbf{k}| \) and keeping only the first order term of \( \mathbf{A}(t) \), one obtains equation (6) except for the scattering term and Hartree–Fock term.

**Appendix B. Expression of the scattering terms**

In this section, we give the scattering matrices used in equations (14) and (15). \( U(\varepsilon_{q,k} - \varepsilon_{q,k-q}) = \sqrt{N}Z_1V_r(\varepsilon_{q,k} - \varepsilon_{q,k-q})\exp(-qd) \) with \( Z_1 = 1 \) being the charge number of the impurity, \( N_I \) standing for the impurity density and \( d \) representing the effective distance of the impurity layer from the graphene sheet. \( M^\mu_{\kappa,\kappa',\mu',\mu} \) are scattering matrices for electron–phonon scattering with phonons in branch \( \lambda \). For the acoustic phonon, \( \omega_{qac} = v_{ph}q \) with \( v_{ph} \) being the sound velocity and the corresponding scattering matrices are

\[
M^\mu_{\kappa,\kappa',\mu',\mu} = D\sqrt{q}S_{\kappa',\kappa}Q_{\delta_{\mu',\mu}}/\sqrt{2\rho_m v_{ph}}
\]

in which \( D \) is the deformation potential and \( \rho_m \) denotes the graphene mass density [77, 78]. For the RI phonons,

\[
M^\mu_{\kappa,\kappa',\mu',\mu} = \sqrt{2\pi}\omega_{RI}\varepsilon_{RI}^{q}\varepsilon_{q}\delta_{\mu',\mu}/\varepsilon_{RI}^{q}
\]

where \( \varepsilon_{RI}^{q} \) is the substrate-dependent dielectric function. For the two branches of phonons RI \( \text{I} \) and RI \( \text{II} \) in the sample on SiO\(_2\), the corresponding dielectric functions are [94]

\[
1/\varepsilon_{RI}^{q} = 1/[\varepsilon_{1} + \varepsilon(q, 0)] - 1/[\varepsilon_{0} + \varepsilon(q, 0)],
\]

\[
1/\varepsilon_{RI}^{q} = 1/[\varepsilon_{\infty} + \varepsilon(q, 0)] - 1/[\varepsilon_{1} + \varepsilon(q, 0)]
\]

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with $\epsilon_i = 3.36$, $\epsilon_0 = 3.90$ and $\epsilon_{\infty} = 2.40$ [82]. For the only branch of RI phonons in graphene on SiC substrate, it is [94]

$$1/\epsilon_{q_{RI}}^{q} = 1/[(\epsilon_{\infty} + \epsilon(q, 0))] - 1/[\epsilon_0 + \epsilon(q, 0)]$$  \hspace{1cm} (B.5)

with $\epsilon_0 = 9.7$ and $\epsilon_{\infty} = 6.5$ [82, 95].

For the optical phonons, the scattering matrices are obtained according to the work by Ishikawa and Ando [96] as

$$M^{\text{OP}_{\mu'n'}}_{\mu'k,k_0} = A^{\text{OP}_{\mu'n'}}_{\mu'k,k_0} / \sqrt{\rho_m \epsilon_{\text{OP}}}$$  \hspace{1cm} (B.6)

with the corresponding parameters of the optical phonons given by

$$A^{\text{LO}_{\mu'n'}}_{\mu'k,k_0} = i \delta_{\mu'\mu} \sqrt{\langle D_1^2 \rangle} [\eta e^{i\mu(\theta_k - \theta_n)} - \eta e^{i\mu(\theta_k - \theta_n)}] / \sqrt{2},$$ \hspace{1cm} (B.7)

$$A^{\text{TO}_{\mu'n'}}_{\mu'k,k_0} = -\mu \delta_{\mu'\mu} \sqrt{\langle D_1^2 \rangle} [\eta e^{i\mu(\theta_k - \theta_n)} + \eta e^{i\mu(\theta_k - \theta_n)}] / \sqrt{2},$$ \hspace{1cm} (B.8)

$$A^{\text{KO}_{\mu'n'}}_{\mu'k,k_0} = \sqrt{\langle D_1^2 \rangle} [\mu \eta e^{-i\mu(\theta_k + \theta_n)} + \mu' \eta e^{i\mu(\theta_k + \theta_n)}] \delta_{\mu,-\mu'} / \sqrt{2}.$$ \hspace{1cm} (B.9)

Appendix C. Numerical scheme for drift term

In this section, we present the numerical scheme for the drift term. The truncated two-dimensional momentum space is divided into $N \times M$ control regions in polar coordinate system [72], each with equal energy and angle intervals. The grid point $k_{n,m}$ is located at the center of the control region $(n, m)$ with its radial coordinate $k_n = (n + 0.5) \Delta k$ and angular coordinate $\theta_m = m \Delta \theta$. Here, $\Delta k$ is the momentum interval and $\Delta \theta = 2\pi / M$ is the angular interval.

The drift term is dealt with in the discrete conservation principle:

$$|e|^2 E(t) \cdot \nabla_k \rho_{\mu k,n'n'} |_{k=k_{n,m}} \simeq \int_{\Omega_{n,m}} d^2 k \frac{|e|^2 E(t) \partial_k \rho_{\mu k,n'n'} }{\Delta \theta \Delta k}$$

$$= \frac{|e|^2 E(t)}{\Delta \theta \Delta k} \int_{\Omega_{n,m}} d\theta_k \frac{\partial_e (k \rho_{\mu k,n'n'} \cos \theta_k) - \partial_{\theta_k} (\rho_{\mu k,n'n'} \sin \theta_k)}{\Delta k}$$

$$\simeq \frac{|e|^2 E(t)}{\Delta \theta \Delta k} \left[ \Delta \theta (F^{r,n,m';}_{\mu,n,m,n'} - F^{r,n,m';}_{\mu,n-1,m,n'}) 
- \Delta k (\sin \theta_{m+0.5} F^{\theta,n,m';}_{\mu,n,m,n'} - \sin \theta_{m-0.5} F^{\theta,n,m';}_{\mu,n-1,m,n'}) \right].$$  \hspace{1cm} (C.1)

Here $\Omega_{n,m}$ and $\partial \Omega_{n,m}$ are the control regions which contain the grid point $k_{n,m}$ and the corresponding boundary, respectively, and the electric field is set to be along the x direction. In the last step of the above equation, the integration of the boundary is replaced by the summation over the first order quadrature on the four (or three if the control region is the neighbor of $k = 0$) sides of the boundary $\partial \Omega_{n,m}$ and the fluxes treated up to third order [97] in the polar coordinate are given by

$$F^{r,n,m'}_{\mu,n,m,n'} = -k_{n-1} \cos \theta_m \rho_{\mu k_{n-1,m},n-n'} / 6 + \cos \theta_m (5k_{n} \rho_{\mu k_{n},n-n'} + 2k_{n+1} \rho_{\mu k_{n+1},n-n'}) / 6;$$  \hspace{1cm} (C.2)

$$F^{\theta,n,m'}_{\mu,n,m,n'} = 5 \rho_{\mu k_{n},n-n'} / 6 + \rho_{\mu k_{n+1},n-n'} / 3 - \rho_{\mu k_{n+1},n-n'} / 6.$$  \hspace{1cm} (C.3)

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In order to satisfy the requirement of numerical stability, \( m' \) in \( F_{\mu, m', n}^{\theta, m, n} \) is taken to be \( m \) if 
\[ -|e| E(t) \cdot \hat{x} \sin \theta_{m+0.5} > 0 \] and \( m + 1 \) otherwise. Moreover, \( n' \) in \( F_{\mu, n', m}^{\theta, n, m} \) is chosen to be \( n \) if 
\[ -|e| E(t) \cdot \hat{x} \cos \theta_m > 0 \] and \( n + 1 \) otherwise. Besides, if \( n' - 1 \geq 0 \), \( m' \) is taken to be \( m \). On the other hand, if \( n' - 1 < 0 \), the term \( n' - 1 \) in the equation is replaced by 0 and \( m' = m + M/2 \). It is noted that with this numerical scheme, the temperature variation due to the noise is within 3 K for the pulse investigated in figure 2.

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