Negative differential transmission in graphene

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By using the Kubo linear response theory with the Keldysh Green function approach, we investigate the mechanism leading to the negative differential transmission in system with the equilibrium electron density much smaller than the photon-excited one. It is shown that the negative differential transmission can appear at low probe-photon energy (in the order of the scattering rate) or at high energy (much larger than the scattering rate). For the low probe-photon energy case, the negative differential transmission is found to come from the increase of the intra-band conductivity due to the large variation of electron distribution after the pumping. For the high probe-photon energy case, the negative differential transmission is shown to tend to appear with the hot-electron temperature being closer to the equilibrium one and the chemical potential higher than the equilibrium one but considerably smaller than half of the probe-photon energy. We also show that this negative differential transmission can come from both the inter- and the intra-band components of the conductivity. Especially, for the inter-band component, its contribution to the negative differential transmission is shown to come from both the Hartree-Fock self-energy and the scattering. Furthermore, the influence of the Coulomb-hole self-energy is also addressed.

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I. INTRODUCTION

Graphene is an easily accessible, truly two-dimensional system which is attractive from the point of view of both the basic physics and possible applications. In the past decade, it has attracted immense investigations. Among these works, the time-resolved optical pump-probe measurement is widely used to investigate the dynamics of electrons in graphene. With this method, a pump pulse is used to excite electrons from the valence band to the conduction one and then a time-delayed probe pulse with the probe-photon energy \( \omega \) is applied to detect the differential transmission (DT). The experimentally obtained DT under a probe-photon energy much higher than twice of the Fermi energy is often positive with its fast relaxation of several hundred femtoseconds followed by a slower picosecond relaxation. From the DT, the conductivity \( \sigma(\omega) \) is extracted. Moreover, if \( \omega \) is also much larger than the scattering rate and the electrons are considered to be free, the conductivity is given by \( (\hbar = 1) \):

\[
\sigma(\omega) \approx \sigma_{\text{free}}(\omega) = -\sigma_0 [f_{k_1} - f_{k_1}^{-}],
\]

(1)

in which \( \sigma_0 = e^2/4 \), \( f_{k,\eta} \) is the electron distribution with \( \eta = 1 (-1) \) representing the conduction (valence) band and \( k_\omega = \omega/(2\hbar \nu_F) \) stands for the resonant absorption state. With this equation and the temporal evolution of \( \sigma_{\text{free}}(\omega) \), the evolution of the electron distribution difference at the resonant absorption state \( f_{k_1} - f_{k_1}^{-} \) is directly detected.

In addition to the positive DT, the negative DT, which indicates the increase of the conductivity after the pumping (positive differential conductivity), has also been observed in different experiments. Soon after that, the negative DT was also observed by George et al. in the case where the probe-photon energy is as low as tens of meV. These two kinds of negative DT can be well understood theoretically. By considering the inter-band conductivity of free electrons [Eq. (1)], the negative DT reported by Sun et al. is shown to come from the weakening of the Pauli blocking due to the heating of electrons by the pump pulse. As for the case with low probe-photon energy, the heating of the electrons can still contribute to the negative DT. Nevertheless, with the scattering taken into consideration, the intra-band conductivity was found to be also important for the negative DT.

In contrast to the previous two cases with moderate or small probe-photon energies, the negative DT has also been observed with the probe-photon energy being very high (e.g., > 1.5 eV). The mechanisms lead to this kind of negative DT is still unclear. At such high probe-photon energy, the Fermi surface is less possible to be as high as half of the probe-photon energy and hence the weakening of the Pauli blocking is less important. As for the intra-band conductivity, its strength is largely suppressed and hence whether it is strong enough to lead to the negative DT is unclear. Moreover, in previous works investigating problems of the negative DT, the scattering strength used was assumed to be constant. With constant scattering strength, the inter-band conductivity can not lead to the negative DT. Nevertheless, with the variation of the scattering strength taken into consideration, whether it can lead to negative DT is still unknown. In addition, the variation of the Coulomb self-energy after the pumping was also suggested to be the
mechanism leading to the increase of the inter-band conductivity and hence the negative DT.\textsuperscript{18,24,27} However, whether it is strong enough to lead to a negative DT has not been fully theoretically demonstrated and it is even claimed that the negative DT itself may come from external factors.\textsuperscript{30}

In this work, we calculate the optical conductivity based on the linear response theory with the Keldysh Green function approach\textsuperscript{37,38} by taking into account the contribution of the electron-phonon and the electron-impurity scatterings as well as the Coulomb self-energy explicitly. We show that for graphene with low equilibrium electron density without the Coulomb-hole (CH) self-energy,\textsuperscript{39,40} the negative DT can appear when the probe-photon energy $\omega$ is low (in the order of the scattering rate) or when $\omega$ is high (much larger than the scattering rate). For the low $\omega$ case, the negative DT mainly comes from the large increase of the intra-band conductivity after the pumping due to the large variation of the electron distribution. As for the high $\omega$ case, the negative DT is shown to tend to appear when the hot-electron temperature $T_e$ is close to the equilibrium one and the chemical potential is higher than the equilibrium chemical potential but considerably lower than $\omega/2$. This negative DT is found to come from the variation of both the intra- and the inter-band components of the conductivity. Especially, for the inter-band conductivity, we find that both the scattering and the Hartree-Fock (HF) self-energy can cause the negative DT.\textsuperscript{31,32} In addition, the influence of CH self-energy is also addressed.

This paper is organized as follows. In Sec. II, we set up the model and lay out the formalism. In Sec. III the results obtained numerically are presented. We summarize and discuss in Sec. IV.

II. MODEL AND FORMALISM

The effective Hamiltonian of graphene near the $K$ and $K'$ points can be described by\textsuperscript{32}

$$H_0 = \sum_{k\sigma\mu\eta\eta'} \eta \bar{c}_{k\sigma\mu\eta} c_{k\sigma\eta'}.$$

(2)

Here, $\mu = 1 \ (-1)$ stands for $K \ (K')$ valley, $\eta = 1 \ (-1)$ represents conduction (valence) band, $s$ denotes spin and $\bar{\varepsilon}_k = v_F k$ with $v_F$ being the Fermi velocity. In this work, we only investigate the linearly polarized normal incident light with the electric field along $\hat{x}$ direction. Then, based on the effective Hamiltonian, the current operator reads (the in-plane photon momentum $q = 0$)\textsuperscript{34}

$$\hat{j} = \sum_{k\sigma\mu\eta\eta'} d_{\sigma\mu\eta\eta'}^{\mu} c_{k\sigma\mu\eta}^\dagger c_{k\sigma\eta'}.$$

(3)

with the dipole matrices being

$$d_{\sigma\mu\eta\eta'}^{\mu} = -ie[v_F'(\eta' e^{i\theta_k} + \eta e^{-i\theta_k})/2].$$

(4)

It has been shown that the hot-electron Fermi distribution is quickly established after the pumping with the hot electron temperatures in the conduction and valence bands being the same while the chemical potentials being different.\textsuperscript{18,43} Based on this result, the electron density matrix without interactions is set to be $\rho_0 = \exp[-\sum_{k\mu\eta\eta'} |\beta_\eta(\eta|\varepsilon_k - \mu_\eta)|^2 k_\mu\eta\eta']$ with $\mu_\eta$ being the chemical potential in band $\eta$, $\beta_\eta = k_B T_e$ and $T_e$ standing for the electron temperature. Then, from the Kubo formula with the Keldysh Green function approach, the optical conductivity is expressed as\textsuperscript{33,37,38,44}

$$\sigma(\omega) = \int dk \int d\omega_d \sum_{\eta,\eta'} d_{\eta\mu\eta'}^{\mu} d_{\eta'\eta''\eta''}^{\eta'} A(k, \eta', \omega_1 + \omega)$$

$$\times A(k, \eta, \omega_1)[F(\omega_1, \eta) - F(\omega_1 + \omega, \eta')]/(16\pi^3 \omega).$$

(5)

Here, $g_d = 4$ comes from the valley and spin degeneracies and $F(\omega, \eta) = \{\exp[(\omega - \mu_\eta)/(k_B T_e)] + 1\}^{-1}$. In this equation, the two band indices $\eta$ and $\eta'$ in $[F(\omega_1, \eta) - F(\omega_1 + \omega, \eta')]$ can be used to distinguish the contributions of the intra- and inter-band conductivities. The intra-band conductivity is defined as terms with $\eta = \eta'$ and the inter-band one with $\eta \neq \eta'$. With this definition of the inter- and intra-band conductivities, their differential conductivities are defined as the difference of the corresponding conductivities after and before the pumping. The electron spectral function is given by

$$A(k, \eta, \omega) = -2i \text{Im} \{[\omega - \eta \varepsilon_k - \Sigma_R(k, \eta, \omega)]^{-1}\}.$$  

(6)

Here, $\Sigma_R(k, \eta, \omega)$ represents the retarded self-energy from the electron-electron, electron-phonon and electron-impurity interactions. The self-energy from the electron-phonon interaction is calculated from\textsuperscript{34}

$$\Sigma_i(k) = -i\pi n_i/2 \int \frac{d^2 k'/(2\pi)^2}{\epsilon_q} \delta(\varepsilon_k - \varepsilon_{k'})$$

$$\times [1 - \cos(\theta_k - \theta_{k'})][1 + \cos(\theta_k - \theta_{k'})].$$

(7)

Here, $\epsilon_q$ is the dielectric function calculated as Scharf et al.\textsuperscript{34} and $V_q = 2\pi \epsilon_r^2/(\kappa q)$ represents the two-dimensional bare Coulomb potentials with $\kappa$ standing for the background dielectric constant.\textsuperscript{34} For the self-energy from the electron-phonon scattering, although the temperatures of electrons and phonons can be different, the Feynman rules and diagrammatic technique for the Keldysh Green function approach are still valid.\textsuperscript{34} With this approach, the retarded self-energy is obtained as

$$\Sigma_{ph}^{R}(k, \eta, \omega) = \sum_{\eta', \lambda} M_{\eta, \eta', \lambda} M(k - q, \omega)$$

$$\times M_{\eta', \lambda} \left[ \frac{n_{q\lambda} + 1 - F(\epsilon_{|k-q|}, \eta')}{\omega + i0^+ - \eta'\epsilon_{|k-q|} - \omega\lambda(q)} \right]$$

$$+ \frac{n_{q\lambda} + F(\epsilon_{|k-q|}, \eta')}{\omega + i0^+ - \eta'\epsilon_{|k-q|} + \omega\lambda(q)}. $$

(8)
Here, $M_{\eta',\lambda}(k,-q)$ are the electron-phonon scattering matrices for phonons in branch $\lambda$ with their detailed forms given by Scharf et al.\textsuperscript{34} $n_{q\lambda} = \{\exp[\omega_{q\lambda}/(k_B T_{ph})] - 1\}^{-1}$ is the distribution for phonons with energy $\omega_{q\lambda}$ and temperature $T_{ph}$. In the following, we ignore the effect of polaronic shifts, i.e., the real part of the electron-phonon self-energy and compensate such approximation by renormalizing the spectral function to fulfill $\int dq A(k,\eta,\omega) = 2\pi$ as done by Scharf et al.\textsuperscript{34}

As for the self-energy from the Coulomb interaction, we take into account the HF self-energy, which is expressed as

$$\Sigma_{HF}^{\eta,\eta'}(k,\omega) = \frac{1}{2} \sum_{k',\eta''} (1 + \eta'' \cos(\theta_k - \theta_{k'})) \times F(\eta'' v_k' k', \eta'') \frac{V_{k'k}}{\epsilon_{k'k} - \epsilon_{k-k}}.$$

with the exchange term of the full valence band subtracted as it is already included in the original single-particle energy.\textsuperscript{35} However, it is known in semiconductor optics that there is a Debye shift or the CH self-energy in addition to the HF self-energy, which is expressed as

$$\Sigma_{CH}^{\eta,\eta'}(k,\omega) = \sum_q \eta \cos(\theta_{k} - \theta_{k+q}) V_q (1/\epsilon_q - 1/\epsilon_q^0)/2,$$

with $\epsilon_q^0$ representing the dielectric function at $T_e = 0$ K and $\mu_1 = \mu_{-1} = 0$ meV. The physics of this CH self-energy is as follows. After the pumping, the pump-excited electrons change the screening strength. As a result, the subtracted exchange term of the full valence band also varies [Eq. (11)]. It has been shown in semiconductor optics that the CH self-energy leads to a strong renormalization of the band structure (band-gap shrinkage)\textsuperscript{36,46}. Due to the similar renormalization of the band structure in graphene, it was speculated that this variation of the Coulomb self-energy is responsible for the negative DT.\textsuperscript{22} In this work, we also calculate the influence of the Coulomb self-energy to check this speculation.

III. RESULTS

From the relation between the transmission and conductivity\textsuperscript{33,47-49}

$$T_{\omega} = \left|1 + N_{lay} \sigma(\omega) \sqrt{\mu_0/\epsilon_0}/(1 + n_{ref})\right|^{-2},$$

one finds that the negative DT emerges when the optical conductivity increases after the pumping. Therefore, instead of calculating the DT, we investigate the variation of the conductivity before and after the pulse (differential conductivity) $\Delta \sigma(T_e, \mu_1, \mu_{-1}, \omega) = \sigma(T_e, \mu_1, \mu_{-1}, \omega) - \sigma(T_0, \mu_0^1, \mu_0^{1-1}, \omega)$. Here, for electrons before the pumping, we choose their temperature $T_0 = 300$ K. We concentrate on the case with the photon-excited electron density much larger than the equilibrium one and hence choose the chemical potential $\mu_1^0 = \mu_0^{1-1} = 0$ for convenience. Then, due to the symmetry between electrons and holes, one always has $\mu_1 = -\mu_{-1}$ after the establishment of the Fermi distribution. In our calculation, the material parameters are the same as in Ref.\textsuperscript{34} and the impurity density $n_i = 5 \times 10^{11}$ cm$^{-2}$. The temperature of phonons $T_{ph}$ is chosen to be the same as electrons and the substrate is chosen to be SiO$_2$ unless otherwise specified.

A. Contributions of intra- and inter-band differential conductivities

In this section, we first give an analytical analysis on the intra- and inter-band components of the conductivity and then show their contributions to the total differential conductivity numerically.

1. Analytical analysis on the intra- and inter-band conductivities

The increase of the total conductivity after pumping may come from both the intra- and the inter-band components of the conductivity. For the intra-band component in Eq. (13), it can be simplified by assuming the self-energy $\Sigma(k, \eta, \omega) = Re\Sigma_{HF} + i/(2\pi)$. Further by using the condition $\mu_1 = \mu_{-1}$ and taking the limit $\omega \to 0$, one has

$$\sigma_{intra}(\omega) = e^2\pi D(T_e, \mu_1, \mu_{-1})/[\pi(1 + \omega^2\tau^2)].$$

Here, $Re\Sigma_{HF}$ is the real part of the self-energy which comes from the HF and CH self-energies and

$$D(T_e, \mu_1, \mu_{-1}) = 2 \int_0^\infty d\epsilon \epsilon\left[-\partial_\tau F(\epsilon + Re\Sigma_{HF}, 1)\right]$$

$$= 2 \int_0^\infty dx (k_B T_e x + \mu') \frac{e^x}{(e^x + 1)^2},$$

with $\mu' = \mu_1 - Re\Sigma_{HF,1}$. From Eq. (13), one finds that the increase of the scattering strength, i.e., the decrease of $\tau$, leads to the increase (decrease) of the conductivity when the probe-photon energy $\omega$ is larger (smaller) than $\tau^{-1}$. Moreover, from Eqs. (13) and (14), it is found that the increase of the electron temperature $T_e$ and chemical potential $\mu_1$ as well as the decrease of the self-energy Re$\Sigma_{HF,1}$ lead to the increase of $D(T_e, \mu_1, \mu_{-1})$ and hence $\sigma_{intra}$.

As for the inter-band conductivity, its increase may come from the variations of the scattering (the imaginary part of the self-energy), the HF self-energy and the CH
self-energy (the real part of the self-energy). For the scattering, it can be shown that the inter-band conductivity increases with the decrease of the scattering strength. Therefore, the positive differential conductivity may appear when the scattering strength decreases. On the other hand, for the HF and CH self-energies, their influences are understood from the simplified Eq. (13) by neglecting the electron-impurity and electron-phonon scatterings:

\[
\sigma(\omega) = \frac{g_d\sigma_0^2\omega}{\omega} \int_0^\infty dk k \delta(\omega - 2\epsilon_k + \Sigma_{k_{-1}}^{ee} - \Sigma_{k_{-1}}^{ee,1}) \\
\times \left\{ F(-\epsilon_k + \Sigma_{k_{-1}}^{ee},-1) - F(\omega - \epsilon_k + \Sigma_{k_{-1}}^{ee,1},1) \right\} \\
= \frac{g_d\sigma_0^2v_F^2k_\omega}{\omega} \left\{ F(-\epsilon_k + \Sigma_{k_{-1}}^{ee,1},-1) - F(\omega - \epsilon_k + \Sigma_{k_{-1}}^{ee},1) \right\} \\
\times \left\{ 2\omega - \partial_\omega \phi_k(\Sigma_{k_{-1}}^{ee} - \Sigma_{k_{-1}}^{ee,1}) \right\}. 
\]

(15)

Here, \(k_\omega\) satisfies \(\omega - 2\epsilon_{k_{-1}} + \Sigma_{k_{-1}}^{ee} - \Sigma_{k_{-1}}^{ee,1} = 0\) and \(\Sigma_{k_{-1}}^{ee,1}\) stands for the corresponding self-energy from the Coulomb interaction (HF and CH self-energies). After the pumping, the difference of the HF self-energy \(\Sigma_{k_{-1}}^{ee} - \Sigma_{k_{-1}}^{ee,1}\) may increase due to the increase of the carrier density and that of the CH self-energy \(\Sigma_{k_{-1}}^{ee} - \Sigma_{k_{-1}}^{ee,1}\) can also increase due to the enhancement of the screening strength. As a result, \(k_\omega\) may increase, which tends to enhance the inter-band conductivity.

2. Numerical investigation on the contributions of intra- and inter-band differential conductivities

We then numerically calculate the probe-photon energy dependence of the total differential conductivity. The case without the CH self-energy is first investigated. It is noted that the chemical potential \(\mu_1\) and temperature \(T_e\) of hot-electrons vary with the temporal evolution after the pumping. Here, we take \(T_e = 500\) K and \(\mu_1 = -0.1 = 300\) meV as an example to investigate the possible mechanisms discussed in Sec. IIIA1. The total differential conductivity as function of probe-photon energy is plotted in Fig. 1(a) (red solid curve). To identify the individual contributions of inter- and intra-band components, we also plot the differential conductivity with only the inter-band component in the figure (open squares). One finds that, at very low probe-photon energy \(\omega < 50\) meV, the total differential conductivity is positive and far away from zero while the inter-band differential conductivity is negative. This indicates that the large positive differential conductivity at low \(\omega\) comes from the intra-band component.

As discussed in Sec. IIIA1, this large positive intra-band differential conductivity at low probe-photon energy \(\omega\) comes from the variation of \(D(T_e,\mu_1,\mu_{-1})\) and \(\tau\) after the pumping. The variation of \(D(T_e,\mu_1,\mu_{-1})\) can be obtained from Eq. (14) if the electron distribution (i.e., \(T_e\) and \(\mu_1\)) and the HF self-energy \(\text{Re}\Sigma_{k_{-1}}^{ee}\) are known. It is noted that \(\text{Re}\Sigma_{k_{-1}}^{ee}\) for the temperatures and chemical potentials calculated here is always less than \(3\) meV, much smaller than the chemical potential after the pumping (300 meV). Hence, the influence of the HF self-energy on \(D(T_e,\mu_1,\mu_{-1})\) can be neglected and one obtains \(D(T_e,\mu_1,\mu_{-1}) = 35.8\) and 600 meV for the cases before and after the pumping, respectively. To further identify the contribution of \(\tau\), we fit the total conductivity in the region \(\omega < 50\) meV with

\[
\Delta\sigma/\sigma_0 = \frac{\delta(\omega)}{\sigma_0} = \sigma_{\text{intra}}(\omega) + \sigma_{\text{free}}(\omega). 
\]

(16)

Here, \(\sigma_{\text{free}}(\omega)\) is included to take into account the influence of the inter-band conductivity. \(\tau\) is fitted to be

![FIG. 1: (Color online) Inter-band as well as total differential conductivities with and without the CH self-energy as function of the probe-photon energy \(\omega\). The states after the pumping are chosen to be (a) \(T_e = 500\) K and \(\mu_1 = -0.1 = 300\) meV, and (b) \(T_e = 1000\) K and \(\mu_1 = -0.1 = 300\) meV. The total conductivity without the CH self-energy in (a) is fitted for \(\omega \leq 50\) meV by Eq. (16) with the fitted results plotted as the green dotted curve. The inset zooms the range at high \(\omega\) with the inter-band differential conductivity without the CH and HF self-energies plotted as brown solid triangles. The black chain line in the figure marks \(\Delta\sigma/\sigma_0 = 0\).](image)
0.07 meV$^{-1}$ and 0.17 meV$^{-1}$ for the cases before and after pumping, respectively [the obtained differential conductivity is plotted in Fig. (a) as the green dotted curve]. One finds that the relative change of $\tau$ is much smaller than that of $D(T_e, \mu_1, \mu_{-1})$. Hence the variation of the intra-band conductivity mainly comes from the variation of the electron distribution, which leads to the huge increase of $D(T_e, \mu_1, \mu_{-1})$. Moreover, since the influence of the HF self-energy on $D(T_e, \mu_1, \mu_{-1})$ can be neglected, its influence on the intra-band component of the differential conductivity is also marginal. It is further noted from the figure that, with the increase of the probe-photon energy $\omega$, $\Delta \sigma$ decreases fast and becomes negative around $\omega = 55$ meV. This quick decrease of $\Delta \sigma$ can be understood from Eq. (13), which infers that the influence of the intra-band component of the differential conductivity is significant only when $\omega$ is in the order of $\tau^{-1}$.

In addition to the large positive differential conductivity shown in the case of low probe-photon energy $\omega$, another small positive total differential conductivity also appears when $\omega$ is high (larger than 1200 meV) as shown by the red solid curve in the inset. One also finds from the inset that the inter-band differential conductivity (open squares) is also positive. Moreover, it is smaller than the total one with their difference being in the same order ($\sim 10^{-5} \sigma_0$) as the inter-band differential conductivity. This means that the positive total differential conductivity comes from both the inter- and the intra-band components and both contributions are important at such high $\omega$.

For the positive contribution from the intra-band component at such high $\omega$, it still mainly comes from the large variation of the electron distribution after the pumping as the case with low probe-photon energy $\omega$. This is confirmed by calculating the intra-band component in Eq. (5) with the distribution $[F(\omega_1, \eta) - F(\omega_1 + \omega, \eta)]$ fixed with the pumping. We find that the obtained intra-band conductivity decreases more than 75% for all $\omega$ shown in the inset (not plotted in the figure).

As for the positive contribution from the inter-band component, it may come from the variations of both the HF self-energy and the scattering strength after pumping as discussed in Sec. IIIA1. Although the influence of the HF self-energy on the intra-band component is marginal, it may have non-negligible influence on the inter-band component. To further identify its contribution, we also plot the inter-band differential contribution without the HF self-energy in the inset (brown solid triangles). One finds that the one with the HF self-energy is about 2~3 times of that without the HF self-energy. This means that the variations of both the HF self-energy and the scattering strength have important contributions to the positive inter-band differential conductivity at such high probe-photon energy.

We further investigate the influence of the CH self-energy on the differential conductivity. With the CH self-energy, the total differential conductivity and the one with only inter-band component are plotted as the blue dashed curve and the blue solid dots in the figure, respectively. One finds that their qualitative behaviors are similar to those without the CH self-energy, i.e., the large positive differential conductivity from the intra-band component at low probe-photon energy $\omega$ as well as the emergence of positive differential conductivity at high $\omega$. Nevertheless, it is seen that the differential conductivity with the CH self-energy included varies markedly compared with the one without the CH self-energy. This means that the CH self-energy has pronounced influence on the differential conductivity. Moreover, it is also seen that with the CH self-energy, the total and the inter-band differential conductivities are very close at high $\omega$ as shown in the inset. This indicates that the large influence of the CH self-energy at high $\omega$ is mainly on the inter-band component.

To further show the influence of $T_e$, we plot the probe-photon energy $\omega$ dependence of the differential conductivities at the electron temperature $T_e = 1000$ K and chemical potential $\mu_1 = 300$ meV in Fig. (b). For the case without the CH self-energy, the total differential conductivity (red solid curve) at high $\omega$ (> 1200 meV as shown in the inset) becomes negative, which is different from the case at $T_e = 500$ K. To show the origin of this negative differential conductivity, we plot the the inter-band one with (red open squares) and without (brown solid triangles) the HF self-energy in the inset. It is shown that they are also negative and the total differential conductivity is close to the inter-band one without the HF self-energy. This indicates that the negative total differential conductivity comes from the negative inter-band differential conductivity without the CH and HF self-energies under high temperature at high $\omega$. To see the influence of the CH self-energy, we plot in the inset both the total (blue dashed curve) and the inter-band (blue solid dots) differential conductivities. One finds both become positive for the high $\omega$ case. This again shows that the CH self-energy has a large influence on the differential conductivity.

B. Condition for negative DT

In the previous section, the mechanisms and their contributions to the positive differential conductivity are investigated under specific electron temperatures $T_e$ and chemical potential $\mu_1$. However, the hot-electron temperature and the chemical potential after the pumping vary with time. Here, we investigate the conditions of $T_e$ and $\mu_1$, as well as the probe-photon energy $\omega$ for the emergence of the positive differential conductivity (i.e., negative DT). Since the positive differential conductivity in the low $\omega$ case has been shown to come from the intra-band component of the conductivity, in this section, we only investigate the case with high $\omega$, where the negative DT was speculated to come from the scattering and the Coulomb self-energy. The cases without
the CH self-energy are first studied with the total differential conductivity in the \( \omega - \mu_1 \) parameter space plotted in Fig. 2(a). Here, the electron temperature is chosen to be 700 K, which is among those estimated in the experiment and the phonon temperature is chosen to be the same as that of electrons. We only plot the region with positive differential conductivity for clarity. It is found that if the probe-photon energy is larger than 1250 meV, the positive differential conductivity emerges with the corresponding range of \( \mu_1 \) depending on \( \omega \). To further show the contributions of the inter- and intra-band components, we plot the chemical potential dependence of the total and the inter-band differential conductivities in Fig. 2(b) with \( \omega = 1500 \) and 1700 meV. From the figure, one finds that the inter-band differential conductivities (open squares and dashed curve) are negative but the total conductivity is positive when \( 160 \text{ meV} < \mu_1 < 420 \text{ meV} \) for \( \omega = 1700 \text{ meV} \) (solid curve) and 140 meV < \( \mu_1 < 340 \text{ meV} \) for \( \omega = 1500 \text{ meV} \) (dashed curve). This indicates that the positive differential conductivity emerges from the inter-band component. The result is consistent with previous ones that the intra-band differential conductivity increases with \( T_e \) and \( \mu_1 \) [Eq. (14)] while the inter-band one is negative if the hot-electron temperature is much larger than the equilibrium one. Moreover, one also finds that peaks appear in the chemical potential dependences of both the total and the inter-band differential conductivities with the corresponding \( \mu_1 \) considerably smaller than \( \omega/2 \). Considering that the intra-band differential conductivity increases with \( \mu_1 \) monotonously, the peak shown in the total differential conductivity is understood to come from the inter-band one. In addition, it is also shown in the figure that with the increase of \( \omega \), the peak of the inter-band differential conductivity appears at higher \( \mu_1 \) (pointed by the arrows), this explains that the positive total differential conductivity tends to appear at higher \( \mu_1 \) with the increases of \( \omega \) as shown in Fig. 2(a). To further show the influence of the HF self-energy on the inter-band differential conductivity, we plot the inter-band differential conductivity at \( \omega = 1700 \text{ meV} \) without the HF self-energy (green dashed curve) in Fig. 2(b). It is shown that the inter-band differential conductivities with and without the HF self-energy are close at small \( \mu_1 \) (< 200 meV). This is understood since the HF self-energy is small when the excited electron density is small (i.e., \( \mu_1 \) is small). On the other hand, when the excited electron density is much larger than the equilibrium one (i.e., \( \mu_1 \) is large), the HF self-energy also becomes large and it leads to an obvious increase of the inter-band differential conductivity as shown in the figure.

One also finds from Fig. 2(a) that the positive differential conductivity (negative DT) emerges only when the probe-photon energy \( \omega \) is high enough. This is supported by the experimental data by Shang et al. 24 They showed that the negative DT appears when the probe-photon energy \( \omega \) is larger than 2000 meV but disappears when \( \omega \) is smaller than 1835 meV under very strong pumping strength. From the numerical results shown in Fig. 2(a) (where \( T_e = 700 \text{ K} \)), this property can be shown by electrons with the chemical potential \( \mu_1 \) around 490 meV. One finds that the corresponding differential conductivity (DT) changes from negative (positive) to positive (negative) when \( \omega > 1900 \text{ meV} \). It is noted that the ele-
electron density under these $T_e$ and $\mu_1$ is $1.9 \times 10^{13}$ cm$^{-2}$. This electron density is also reasonable since the pumping strength used in the experiment is very strong.

It is noted that in our calculation, all modes of phonons are assumed to be at the same temperature as electrons. Nevertheless, in genuine situation, the phonon-phonon scattering is not strong enough to build up such phonon distribution. To further investigate the influence of the phonon temperature, we also show the results with the phonon temperature being 200 K lower than the electron temperature $T_e$ in Fig. 2(c). It is shown that with the decrease of the phonon temperature, the region of the positive differential conductivity is enlarged. This is understood since the electron-phonon scattering strength decreases with the decrease of the phonon temperature, which increases the inter-band differential conductivity.

To confirm this, we also plot the chemical potential dependence of the total and the inter-band differential conductivities with $T_{ph} = 500$ K and $\omega = 1700$ meV in Fig. 2(d). Compared to the one with $T_{ph} = 700$ K [Fig. 2(b)], it is found that the inter-band differential conductivity without the HF self-energy (green dashed curve) increases markedly and it can even lead to positive differential conductivity at 45 meV $< \omega < 350$ meV.

We further give a detailed investigation on the total differential conductivity in the electron-temperature–chemical-potential ($T_e$–$\mu_1$) space at a fixed probe-photon energy $\omega = 1500$ meV. The phonon temperature is chosen to be the same as the electrons and the results are plotted in Fig. 2(e). One finds that the positive differential conductivity can appear when $T_e < 730$ K. To further identify the contributions of the inter- and intra-band components, we plot the temperature dependence of the differential conductivity in Fig. 2(f) with $\mu_1 = 320$ meV. One finds that the total differential conductivity (solid curve) is obviously larger than the inter-band one (dotted curve). This indicates the important positive contribution of the intra-band component. Moreover, it is also shown that the difference between the inter-band differential conductivities with (red dotted curve) and without (green dashed curve) the HF self-energy varies slowly with the temperature. This means that in the parameter range investigated here, the influence of temperature on the HF self-energy is small. In addition, it is also found that the inter-band differential conductivity with only scattering (i.e., without the HF self-energy) can also be positive when $T_e$ is close to the equilibrium one (300 K). However, it decreases with the increase of the temperature quickly and becomes negative when $T_e > 550$ K.

Now, we can lay out the conditions for the emergence of the negative DT (or equivalently, positive differential conductivity) in graphene with low equilibrium electron density. The negative DT can appear both in the low probe-photon energy $\omega$ (in the order of the scattering rate) case and in the high $\omega$ case (much larger than the scattering rate). For the low $\omega$ case, this negative DT mainly comes from the increase of the intra-band conductivity. This increase is mainly due to the large variation of the electron distribution after the pumping and is almost not influenced by the HF self-energy in the temperature range investigated here. As for the high $\omega$ case, although the DT for free electrons [i.e., the one with the conductivity calculated from Eq. (1)] can never be negative, the negative DT can appear if the scattering is taken into consideration. With the scattering, the inter-band conductivity tends to lead to a negative DT when the hot-electron temperature $T_e$ is close to the equilibrium one and the chemical potential $\mu_1$ is higher than the equilibrium chemical potential but considerably smaller than $\omega/2$. Moreover, this region of the negative DT determined by the inter-band conductivity can be further expanded by the intra-band one, which always tends to decrease the DT if $T_e$ and $\mu_1$ increase after the pumping. Furthermore, the HF self-energy can also contribute to the negative DT by increasing the inter-band differential conductivity when $\mu_1$ is much larger than the equilibrium chemical potential (about several hundred milli-electron volts larger) in the temperature range usually estimated in the experiment (around 1000 K). This further expands the region of the negative DT at high $\omega$. We further note that if the phonon temperature is considered to be smaller than that of electrons, the inter-band differential conductivity increases, which also expands the region of the negative DT.

![Diagram](image_url)

**FIG. 3:** (Color online) Total differential conductivity with the CH self-energy for graphene on SiO$_2$ (a) and SiC (b) in $T_e$–$\mu_1$ parameter space. For clarity, only the positive differential conductivity is plotted.

C. Influence of CH self-energy

Finally, we address the influence of the CH self-energy on the emergence of the negative DT (positive differential conductivity). The total differential conductivity with the CH self-energy for graphene on SiO$_2$ substrate in the electron-temperature–chemical-potential ($T_e$–$\mu_1$) parameter spaces is plotted in Fig. 3(a). Here, the probe-photon energy $\omega$ is chosen to be 1500 meV. Comparing with the results without the CH self-energy [Fig. 2(e)], one finds that the CH self-energy enhances both the region and the magnitude of the positive differential conductivity markedly. This means that the variation of the CH self-energy after the pumping is the dominant mechanism.
leading to the positive differential conductivity at such high $\omega$ if it is taken into consideration. It is worth noting that, the shape for the region of the positive differential conductivity with the CH self-energy is similar to the one without it [Fig. 2(e)] (this is also true for $\omega - \mu_1$ parameter space). This means that the tendency for the emergence of the total differential conductivity summarized in Sec. IIIB is still valid, although the variation of the CH self-energy after the pumping becomes the dominant mechanism leading to the positive differential conductivity.

It is noted that since the CH self-energy mainly comes from the variation of the screening, its influence should be smaller if the graphene layer is on a substrate with higher background dielectric constant. To confirm this, we show the results for graphene on SiC [Fig. 3(b)], whose background dielectric constant is 2.2 times of that for SiO$_2$. One finds that both the magnitude and the region of the positive differential conductivity decrease obviously compared with the one on SiO$_2$ substrate. Nevertheless, the positive differential conductivity (negative DT) region shown on SiC substrate is still very large.

Actually, the negative DT region shown on SiO$_2$ and SiC substrates with the CH self-energy is so large that it disagrees with some of the existing experiments.\textsuperscript{13,30,31} Dawlaty \textit{et al.}\textsuperscript{24} have shown experimentally for graphene on SiC substrate that the DT is positive when the optical excited electron density is about 10$^{12}$ cm$^{-2}$ with the corresponding $T_e$ estimated to be around 900 K and $\mu_1$ around zero under the probe-photon energy close to 1500 meV.\textsuperscript{20} However, the numerically obtained DT with these parameters is negative as shown in Fig. 3(b). Moreover, Ruzicka \textit{et al.}\textsuperscript{39} and Brida \textit{et al.}\textsuperscript{31} also showed in graphene on SiO$_2$ substrate that the DT is positive when the excited electron density is about 10$^{13}$ cm$^{-2}$ with the corresponding $T_e$ estimated to be around 1000 K and $\mu_1$ around 350 meV.\textsuperscript{20} However, the numerically obtained DT with the CH self-energy is also negative as shown in Fig. 3(a). Nevertheless, for the case without the CH self-energy, the DTs under these parameters are all positive [Fig. 2(e)] for the SiO$_2$ substrate and that for the SiC substrate is not shown\textsuperscript{1}. Further considering the fact that the result obtained without the CH self-energy is consistent with the experiment with the negative DT (Sec. IIIB)\textsuperscript{24} one concludes that although the calculation of the CH self-energy used here is very successful in semiconductors\textsuperscript{39,40} it overestimates the Coulomb self-energy in the gapless graphene system. This may come from the approximations used to obtain Eq. (11). For example, the background dielectric constant $\kappa$ is taken to be constant although it is shown to increase with $q$ [Ref. 52] and the vertex correction\textsuperscript{38} for the Coulomb screening is neglected. A better description on the influence of the Coulomb self-energy is beyond the scope of this investigation.

### IV. SUMMARY

In summary, we have investigated the emergence of the negative DT by calculating the optical conductivity based on the linear response theory with the Keldysh Green function approach. In our calculation, the influences of the electron-phonon and electron-impurity scatterings as well as the Coulomb self-energy are explicitly included. We investigate the system with the photoexcited electron density much larger than the equilibrium one and show that the negative DT (or equivalently, the positive differential conductivity) can appear at low probe-photon energy $\omega$ (in the order of the scattering rate) or at high $\omega$ (much larger than the scattering rate).

For the low $\omega$ case, the negative DT is shown to mainly come from the large increase of the intra-band conductivity due to the large variation of the electron distribution after the pumping. As for the high $\omega$ case, it is shown that the inter-band conductivity with only scattering can lead to the negative DT when the hot-electron temperature $T_e$ is close to the equilibrium one and the chemical potential $\mu_1$ is higher than the equilibrium chemical potential but considerably lower than $\omega/2$. Moreover, the intra-band conductivity can further expand this region of the negative DT because it always tends to decrease DT if $T_e$ and $\mu_1$ increase after the pumping. Furthermore, in the temperature range usually estimated in the experiments (around 1000 K), the HF self-energy is also shown to be able to contribute to the negative DT by increasing the inter-band differential conductivity considerably when $\mu_1$ is much higher than the equilibrium one. This further expands the region of the negative DT at high $\omega$. Nevertheless, the HF self-energy has little influence on the intra-band conductivity and hence its effect on the negative DT at low $\omega$ can be neglected. In addition, we also show that if the phonon temperature is smaller than that of electrons, the inter-band differential conductivity is increased which also expands the region of the negative DT. Our numerical results are consistent with the $\omega$ dependence of the negative DT obtained experimentally by Shang \textit{et al.}\textsuperscript{24}

The influence of the CH self-energy is also investigated. We find that it markedly expands the region of the negative DT. However, this expansion is too large which makes the obtained region of the negative DT become incompatible with the existing experiments.\textsuperscript{13,30,31} This means that the CH self-energy calculated with the well established approach in semiconductors\textsuperscript{39,40} overestimates the Coulomb self-energy in the gapless graphene system. More investigation to take into account the high order correlation is needed.

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