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Existence of the transverse relaxation time in optically excited bulk semiconductors*

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Two basic types of depolarization mechanisms, carrier-carrier (CC) and carrier-phonon (CP) scattering, are investigated in optically excited bulk semiconductors (3D), in which the existence of the transverse relaxation time is proven based on the vector property of the interband transition matrix elements. The dephasing rates for both CC and CP scattering are determined to be equal to one half of the total scattering-rate-integrals weighted by the factors $(1-\cos\chi)$, where χ are the scattering angles. Analytical expressions of the polarization dephasing due to CC scattering are established by using an uncertainty broadening approach, and analytical ones due to both the polar optical-phonon and non-polar deformation potential scattering (including inter-valley scattering) are also presented by using the sharp spectral functions in the dephasing rate calculations. These formulas, which reveal the trivial role of the Coulomb screening effect in the depolarization processes, are used to explain the experimental results at hand and provide a clear physical picture that is difficult to extract from numerical treatments.

Keywords: ultrafast dephasing, carrier-carrier scattering, carrier-phonon scattering
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1. Introduction

The quantum characteristics of interactions between light and semiconductors on a short-time scale have not only been a subject of great interest in ultrafast photo-electronics, but also attracted attention to quantum cryptography and quantum computers. For example, semiconductor quantum dots are currently considered as possible candidates for solid-state-based implementations of quantum logic devices. Since the quantum operation must be completed before the quantum coherence is destroyed, the studies of dephasing or decoherence in materials are as important as the preparation of coherent states or entangled states. Indeed, since Becker *et al*[1] reported their experimental results in 1988, dephasing of the optical interband polarization in semiconductors has been a challenging subject in nonequilibrium phenomena.[1−11] The dephasing time is also called the optical transverse relaxation time T_2 which was introduced phenomenologically into nonlinear optics of atomic systems by Bloembergen and Shen.[12] The experimental results shown in Refs.[1] and [2] indicate that the optical transverse relaxation time T_2 for continuum excitation of semiconductors falls onto the femtosecond time scale. The dephasing was explained accounting for dominant CC scattering due to very strongly screened Coulomb scattering.[1,2] In comparison with the efforts dedicated to the explanation of the density-dependence of the relaxation time, the literature on such a basic problem is rather scarce. The question is whether and/or how does T_2 come into existence in the optically excited semiconductors. The definition of the dephasing rate is ambiguous in the literature and the explanation of the experimental results is usually based on empirical formulas and numerical fittings. Very recently, we have proven the existence of the transverse relaxation time in two-dimensional (2D) pulse-excited semiconductor quantum wells[13] based on the vector property of the interband transition matrix elements. By using the un-

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certainty broadening approach we have also presented an expression of density-dependent dephasing time of $T_2 \propto N^{-1/2}$, which agrees well with the experimental power-law of $T_2 \propto N^{-0.55\pm0.04}$ in 2D case,[3] where N is the density of the optically excited carriers. Naturally, one would expects that the theoretical prediction of the density-dependent dephasing rate should also hold for optically excited bulk semiconductors. The variation of the dephasing time with the 3D carrier density N was shown in Ref.[1] to follow another power-law, $T_2(\mathrm{fs}) = 4 \times 6.8[N(10^{18}\mathrm{cm}^{-3})]^{-0.3}$. One of the main goals in this paper is to explain this behaviour which is attributed to carrier-phonon scattering in bulk semiconductors.

The theoretical basis of the existence of the dephasing time in the 3D case is similar to that in the 2D case. We present a brief summary of the essential equations both for CC scattering and CP scattering in Section 2. For our purpose it is important to emphasize the dependence of the depolarization on the scattering rates. The depolarization rates turn out to be the average of the so-called momentum relaxation rates for holes (h) and electrons (e). For non-polar optical-phonon scattering, the corresponding momentum relaxation rates just equal the common scattering rates (see Section 2.2), which indicates that the momentum relaxation discussed here is somewhat different from that in transport measurements. In Section 2, we also present analytical expressions of the depolarization rates arising from CP scattering including inter-valley scattering. In Section 3, we present analytical expressions of the depolarization arising from CC scattering based on the uncertainty broadening approach. The expressions reveal the dependence of the depolarization rates on relevant parameters, such as the optical pump frequencies reflected by the Bloch wave vectors, the plasma densities, and so on. When CP scattering is taken into account in the depolarization processes, the weaker density-dependence and negligible energy dependence of the dephasing can be understood, as shown schematically in Section 4 (see Fig.1.). The comparison with the reported experimental results is also discussed in Section 4. A short summary in Section 5 concludes our paper. Some mathematical approximations for derivation of the momentum relaxation rates are presented in Appendix A.

2. Investigation of the existence of the transverse relaxation time

The interaction between light and matter, including coherent effects and relaxation phenomena, can be described by the optical Bloch equations. In optically excited semiconductors, the diagonal elements of the density matrix are often converted to the distribution functions of electrons and holes $f_{\gamma k}$, γ=e, h, where $\boldsymbol{k}$ is the Bloch wave vector marking the momentum state of the carriers. The non-diagonal elements ρ_k of the density matrix are related to the macroscopic polarization vector $\mathrm{Re}(\sum_k \rho_k \mu^*_{cv}(\boldsymbol{k}))$, where $\mu^*_{cv}(\boldsymbol{k}) = \mu_{vc}(\boldsymbol{k})$ are the matrix elements of the interband transition with band indices c and v, respectively. For our purpose it is especially important to point out that the vector property of the matrix element of the interband transition is one of the most important factors to guarantee the existence of T_2. We will neglect the renormalization effects to obtain compact expressions, though this approximation is not necessary for obtaining our conclusions. Thus, the semiconductor Bloch equations are[5,11,14]

$$\begin{aligned}\frac{\partial}{\partial t}\rho_k =& -\frac{\mathrm{i}}{\hbar}(\varepsilon_{ek}+\varepsilon_{\mathrm{h}k})\rho_k \\ &+\frac{\mathrm{i}}{\hbar}\mu_{cv}(\boldsymbol{k})\cdot\boldsymbol{E}(t)(1-f_{ek}-f_{\mathrm{h}k}) \\ &+\frac{\partial}{\partial t}\rho_k\,|_{\mathrm{scatt}}, \qquad (1a)\\ \frac{\partial}{\partial t}f_{\gamma k} =& 2\mathrm{Im}[\rho^*_k\mu_{cv}(\boldsymbol{k})\cdot\boldsymbol{E}(t)] \\ &+\frac{\partial}{\partial t}f_{\gamma k}\,|_{\mathrm{scatt}} \quad \text{with} \quad \gamma=\mathrm{e},\mathrm{h}. \qquad (1b)\end{aligned}$$

Here, $\boldsymbol{E}(t)$ is a coherent light field and $\varepsilon_{\gamma k} = \hbar^2k^2/(2m_\gamma) + (1/2)E_{\mathrm{g}}$ is the carrier energy with E_{g} the energy gap and γ=e,h.

The scattering parts of Eqs.(1) can be grouped into two classes: CP scattering processes in which the scattering rates are independent of carrier densities, and CC scattering processes in which the scattering rates depend on carrier densities. We shall first demonstrate the existence of the transverse relaxation time for CC scattering processes. And then we extend the conclusion to the carrier-density-independent processes, which consist of polar optical phonon scattering (LO), non-polar optical phonon scattering (NP), acoustic phonon scattering (AP), and inter-valley phonon scattering (IV). The expressions of the depolarization rates for CP scattering are presented in this section, while the expressions for CC scattering are discussed in Sec. 3.

2.1. Existence of T_2 in carrier-carrier scattering processes

The scattering parts of Eqs.(1) for CC scattering

are given as[5,14,16]

$$\frac{\partial}{\partial t}\rho_k \mid_{\rm scatt} = \sum_{k'} \left(\Gamma_{k,k'} \rho_{k'} - \Gamma_{k',k} \rho_k \right), \tag{2a}$$

$$\frac{\partial}{\partial t} f_{\gamma k} \mid_{\rm scatt} = \sum_{\delta,k'} (W^{\gamma\delta}_{k,k'} f_{\gamma k'} (1 - f_{\gamma k}) - W^{\gamma\delta}_{k',k} (1 - f_{\gamma k'}) f_{\gamma k}) \quad \text{with} \quad \delta = \mathrm{e}, \mathrm{h}. \tag{2b}$$

$\Gamma_{k,k'}$ and $\Gamma_{k',k}$ can be written as the sum of the transition probabilities $W^{\gamma\delta}_{k',k}$ and $W^{\gamma\delta}_{k,k'}$, namely[5,13-15]

$$\Gamma_{k',k} \equiv \sum_{\gamma,\delta} \Gamma^{\gamma\delta}_{k',k}, \tag{3a}$$

$$\Gamma_{k,k'} \equiv \sum_{\gamma,\delta} \Gamma^{\gamma\delta}_{k,k'}, \tag{3b}$$

where we have used equations as follows

$$\Gamma^{\gamma\delta}_{k',k} = \frac{1}{2}(W^{\gamma\delta}_{k,k'} f_{\gamma k'} + W^{\gamma\delta}_{k',k}(1 - f_{\gamma k'})), \tag{4a}$$

$$\Gamma^{\gamma\delta}_{k,k'} = \frac{1}{2}(W^{\gamma\delta}_{k',k} f_{\gamma k} + W^{\gamma\delta}_{k,k'}(1 - f_{\gamma k})). \tag{4b}$$

The transition probabilities are given by[5,14]

$$W^{\gamma\delta}_{k,k'} = \frac{2\pi}{\hbar} \sum_{K,K'} 2|M|^2 \delta_{k+K,k'+K'} \delta[E_\gamma(\boldsymbol{k}) + E_\delta(\boldsymbol{K}) - E_\gamma(\boldsymbol{k}') - E_\delta(\boldsymbol{K}')] \{ f_{\gamma k'} [f_{\delta \boldsymbol{K}'} (1 - f_{\delta K})] + (1 - f_{\gamma k'}) f_{\delta K} (1 - f_{\delta K'}) \}, \tag{5}$$

with the matrix elements $M = e^2 / \left[\epsilon_0 \epsilon (\boldsymbol{k} - \boldsymbol{k}')^2 V \right]$.

As we have pointed out in Ref.[13], the experimentally measured polarization is the macroscopic mechanical quantity $\mathrm{Re}(\sum_k \rho_k \mu^*_{cv}(\boldsymbol{k}))$, rather than ρ_k or $\sum_k \rho_k$. Accordingly, the relaxation of the polarization is $\frac{\partial}{\partial t} \sum_k \rho_k \mu^*_{cv}(\boldsymbol{k}) \mid_{\rm scatt}$ instead of $\frac{\partial}{\partial t} \rho_k \mid_{\rm scatt}$ or $\frac{\partial}{\partial t} \sum_k \rho_k \mid_{\rm scatt}$. From Eq.(1a), one obtains the evolution equation of the total macroscopic polarization as follows:

$$\begin{aligned} &\frac{\partial}{\partial t} \sum_k \rho_k \mu^*_{cv}(\boldsymbol{k}) \\ &= -\frac{\mathrm{i}}{\hbar} \sum_k [\rho_k \mu^*_{cv}(\boldsymbol{k})(\varepsilon_{\mathrm{e}k} + \varepsilon_{\mathrm{h}k})] \\ &\quad + \frac{\mathrm{i}}{\hbar} \sum_k [\mu^*_{cv}(\boldsymbol{k}) \mu_{cv}(\boldsymbol{k}) \cdot \boldsymbol{E}(t)(1 - f_{\mathrm{e}k} - f_{\mathrm{h}k})] \\ &\quad + \frac{\partial}{\partial t} \sum_k \rho_k \mu^*_{cv}(\boldsymbol{k}) \mid_{\rm scatt} . \end{aligned} \tag{6}$$

Here, the scattering parts due to CC scattering are

$$\begin{aligned} &\frac{\partial}{\partial t} \sum_k \rho_k \mu^*_{cv}(\boldsymbol{k}) \mid_{\rm scatt} \\ &\equiv \sum_{k,k'} (\Gamma_{k,k'} \rho_{k'} \mu^*_{cv}(\boldsymbol{k}) - \Gamma_{k',k} \rho_k \mu^*_{cv}(\boldsymbol{k})). \end{aligned} \tag{7}$$

By way of parenthesis, if the matrix element of the interband transition $\mu_{cv}(\boldsymbol{k})$ is not taken into account, it is easily to obtain $\frac{\partial}{\partial t} \sum_k \rho_k \mid_{\rm scatt} = 0$. This means that $\sum_k \rho_k$ does not decay in the scattering processes, and then we can say that $\mu_{cv}(\boldsymbol{k})$ plays an important role in the decay of the polarization. Therefore, the summation of $\sum_k \rho_k$ does not have a certain physical meaning and cannot be used to denote the polarization. However, the summations of $\sum_k f_{\gamma k}$ do represent the carrier densities. And the equations $\frac{\partial}{\partial t} \sum_k f_{\gamma k} \mid_{\rm scatt} = 0$ represent the conservation of particle number during the scattering processes.

Without the scattering term, the solution of Eq.(6) is given by

$$\sum_k \rho_k \mu^*_{cv}(\boldsymbol{k}) = \sum_k [\mu^*_{cv}(\boldsymbol{k}) \mu_{cv}(\boldsymbol{k}) \cdot \boldsymbol{\xi}(t)], \tag{8a}$$

with

$$\boldsymbol{\xi}(t) = \frac{\mathrm{i}}{\hbar} \exp\left(-\frac{\mathrm{i}}{\hbar}(\varepsilon_{\mathrm{e}k} + \varepsilon_{\mathrm{h}k}) t \right) \int_{-\infty}^{t} \{ \boldsymbol{E}(t') \exp\left[\frac{\mathrm{i}}{\hbar}(\varepsilon_{\mathrm{e}k} + \varepsilon_{\mathrm{h}k}) t' \right] (1 - f_{\mathrm{e}k} - f_{\mathrm{h}k}) \} \mathrm{d}t'. \tag{8b}$$

In the case of weak excitation ($f \ll 1$) by an optical pulse of the exponential form $\boldsymbol{E}(t) = \boldsymbol{E}_0 \exp(-\alpha|t| - \mathrm{i}\omega_0 t)$ with $\alpha > 0$, Eq.(8b) can be straightforwardly solved:

$$\boldsymbol{\xi}(t) = \begin{cases} \dfrac{\mathrm{i}\boldsymbol{E}_0 \exp(\alpha t - \mathrm{i}\omega_0 t)}{\hbar\alpha + \mathrm{i}[(\varepsilon_{\mathrm{e}k} + \varepsilon_{\mathrm{h}k}) - \hbar\omega_0]}, & t < 0; \\ \dfrac{\mathrm{i}\boldsymbol{E}_0 \exp(-\alpha t - \mathrm{i}\omega_0 t)}{-\hbar\alpha + \mathrm{i}[(\varepsilon_{\mathrm{e}k} + \varepsilon_{\mathrm{h}k}) - \hbar\omega_0]} + \dfrac{\mathrm{i}2\hbar\alpha \boldsymbol{E}_0 \exp(-\mathrm{i}(\varepsilon_{\mathrm{e}k} + \varepsilon_{\mathrm{h}k})t/\hbar)}{(\hbar\alpha)^2 + [(\varepsilon_{\mathrm{e}k} + \varepsilon_{\mathrm{h}k}) - \hbar\omega_0]^2}, & t > 0. \end{cases} \tag{8c}$$

Using the integral of motion[15]

$$4|\rho_k|^2 + (1 - f_{ek} - f_{hk})^2 = 1, \tag{9}$$

in the case of weak excitation, we can obtain the carrier distribution function after the pulse excitation (i.e. $t >> 1/\alpha$) as follows

$$\begin{aligned} f_{\gamma k} \simeq & |\rho_k|^2 \\ = & |\boldsymbol{\xi}(t) \cdot \mu_{vc}(\boldsymbol{k})|^2 \\ \simeq & \left\{ \frac{2\hbar\alpha \boldsymbol{E}_0 \cdot \mu_{vc}(\boldsymbol{k})}{(\hbar\alpha)^2 + [(\varepsilon_{ek} + \varepsilon_{hk}) - \hbar\omega_0]^2} \right\}^2 . \end{aligned} \tag{10}$$

Unlike the common Lorentzian shape that leads to the density integral, the total polarization and the mean energy over the $\boldsymbol{k}$-space do not converge, the so-called double Lorentzian shape above allows all these integrals to exist. During the pulse excitation, however, the distribution functions not only are time-dependent but considerably broader since the virtual carriers could be created in the states far away from the central frequency of the pulse. This effect is essentially related to the uncertainty principle for energy. For the long-time limit, the carrier density is time-independent. Apparently, Eqs.(8c) and (10) imply that the optically excited semiconductor is an inhomogeneously broadened system as long as the optical pulse is short enough. Then the depolarization due to the various oscillation frequencies with the corresponding sizes of wave vector is reversible.

However, when the scattering terms are considered in the evolution of the macroscopic polarization, the inherent depolarization occurs, which will be investigated right now. In this paper it is assumed that the relaxation time is long compared to the pulse duration so that we are able to solve the coherent part of the evolution equation for macroscopic polarization, and then insert the results into the relaxation part to evaluate the corresponding relaxation rates. Actually, such a procedure is always justified in experimental investigations. Consequently, decay effects can be treated separately in this situation.

Following the approach we have used for semiconductor quantum wells,[13] Eq.(7) for bulk semiconductors can also be simplified and takes approximately the same form as in the 2D case (see appendix A), i.e.,

$$\frac{\partial}{\partial t} \sum_k \rho_k \mu_{cv}^*(\boldsymbol{k}) \mid_{\text{scatt}} \equiv - \sum_k \Gamma(\boldsymbol{k}) \rho_k \mu_{cv}^*(\boldsymbol{k}), \tag{11a}$$

or in the integral formalism,

$$\frac{\partial}{\partial t} \int \rho(k) \mu_{cv}^*(\boldsymbol{k}) \mathrm{d}\boldsymbol{k} \mid_{\text{scatt}} \equiv - \int \Gamma(\boldsymbol{k}) \rho(k) \mu_{cv}^*(\boldsymbol{k}) \mathrm{d}\boldsymbol{k}, \tag{11b}$$

in which we have introduced

$$\Gamma(\boldsymbol{k}) = \frac{V}{(2\pi)^3} \int \Gamma(k', k, \cos\chi)(1 - \cos\chi) \mathrm{d}\boldsymbol{k}', \tag{11c}$$

with $\Gamma(k', k, \cos\chi) \equiv \Gamma(\boldsymbol{k}', \boldsymbol{k}) \equiv \Gamma_{k',k}$ and χ the angle between the wave vectors $\boldsymbol{k}$ and $\boldsymbol{k}'$. The approximation to obtain Eq.(11) may be called relaxation time approximation for polarization decay. Substituting Eq.(11) into Eq.(6), one obtains

$$\begin{aligned} & \frac{\partial}{\partial t} \sum_k \rho_k \mu_{cv}^*(\boldsymbol{k}) \\ = & - \frac{\mathrm{i}}{\hbar} \sum_k [\rho_k \mu_{cv}^*(\boldsymbol{k})(\varepsilon_{ek} + \varepsilon_{hk} - \mathrm{i}\hbar\Gamma(k))] \\ & + \frac{\mathrm{i}}{\hbar} \sum_k \mu_{cv}^*(\boldsymbol{k}) \mu_{cv}(\boldsymbol{k}) \cdot \boldsymbol{E}(t)(1 - f_{ek} - f_{hk}). \end{aligned} \tag{12}$$

Generally, $\Gamma(k)$ should be time-dependent because the CC scattering rate grows with increasing number of carriers during the optical excitation. Therefore, in the case of weak excitation ($f \ll 1$) and the pulse with exponential form $\boldsymbol{E}(t) = \boldsymbol{E}_0 \exp(-\alpha|t| - \mathrm{i}\omega_0 t)$, one might expect that $\Gamma(k)$ has the following time-dependent structure

$$\Gamma(k) = \begin{cases} a(k)\exp(\beta t), & \text{for } t < 0; \\ (a(k) - b(k))\exp(-\beta t) + b(k), & \text{for } t > 0, \end{cases} \tag{13}$$

where $a(k)$ and $b(k)$ are the k-dependent parameters, and $\beta > 0$ is related to α. Even in this simplified form the problem is still complicated, because one cannot solve Eq.(12) analytically. After the pulse excitation, however, the carrier density and then $\Gamma(k)$ are almost time-independent. In this oversimplified situation, we obtain the macroscopic polarization with the decay effects as follows:

$$\sum_k \rho_k \mu_{cv}^*(\boldsymbol{k}) = \sum_k \left[\mu_{cv}^*(\boldsymbol{k}) \mu_{cv}(\boldsymbol{k}) \cdot \overline{\boldsymbol{\xi}}(t) \right], \tag{14a}$$

with

$$\overline{\boldsymbol{\xi}}(t)=\begin{cases}\dfrac{\mathrm{i}\boldsymbol{E}_0\exp(\alpha t-\mathrm{i}\omega_0 t)}{\hbar[\alpha+\Gamma(k)]+\mathrm{i}[(\varepsilon_{\mathrm{e}k}+\varepsilon_{\mathrm{h}k})-\hbar\omega_0]}, & \text{for } t<0;\\[2ex] \dfrac{\mathrm{i}\boldsymbol{E}_0\exp(-\alpha t-\mathrm{i}\omega_0 t)}{\hbar[-\alpha+\Gamma(k)]+\mathrm{i}[(\varepsilon_{\mathrm{e}k}+\varepsilon_{\mathrm{h}k})-\hbar\omega_0]}+\dfrac{\mathrm{i}2\hbar\alpha\boldsymbol{E}_0\exp\left(-\mathrm{i}\left(\dfrac{\varepsilon_{\mathrm{e}k}+\varepsilon_{\mathrm{h}k}}{\hbar}-\mathrm{i}\Gamma(k)\right)t\right)}{(\hbar\alpha)^2+[(\varepsilon_{\mathrm{e}k}+\varepsilon_{\mathrm{h}k}-\mathrm{i}\hbar\Gamma(k)-\hbar\omega_0]^2}, & \text{for } t>0.\end{cases} \tag{14b}$$

$\Gamma(k)$ may then be called the damping coefficient for the quantum states with the same size of $\boldsymbol{k}$, namely, the same oscillation frequency. These quantum states that are relative to the same energy can be imagined as a homogeneously broadened system. Thus, the summation of the macroscopic polarization over $\boldsymbol{k}$-space shows that the pulse excited system can be regarded as an inhomogeneously broadened one. The dephasing (i.e. depolarization) due to the different oscillation frequencies in the inhomogeneously broadened system is reversible. However, the inherent depolarization which is indicated by $\Gamma(k)$ is irreversible. Hereafter we use the word of dephasing to refer to the irreversible depolarization which arises from the angular redistribution due to CC scattering. Therefore, the vectorial nature of the polarization plays an essential role in the depolarization process. Of course, to completely describe the evolution of the total polarization, it is appropriate to start from Eq.(6), which needs numerical calculations. Since the vectorial property of polarization is always overlooked in literature, the scattering part of Eq.(6) is then misunderstood to be zero. Consequently, the dephasing is often attributed to polarization scattering in literature. In order to emphasize the roles of vectorial nature and summation procedure in the polarization relaxation, as the first step, we shall develop the simplified model in this paper rather than adopt the numerical calculation. It should be noticed that, since $\Gamma(k)$ is obtained by summation procedure, the interplay between different $\boldsymbol{k}$-components corresponding to the Coulomb scattering is firstly averaged in our treatment. The interplay between scattering processes and the different frequencies of different $\boldsymbol{k}$-components of the polarization, however, also holds in our model, at least partially. In this treatment, we can concentrate our attention to identify how the scattering part and the coherent part work in the polarization relaxation. $\overline{\boldsymbol{\xi}}$ in Eqs.(14) is essentially different from $\boldsymbol{\xi}$ in Eqs.(8) in the absence of the Coulomb interaction. The former possesses the statistical average information over the $\boldsymbol{k}$-space. Then, unlike the case without the Coulomb interaction where the $\boldsymbol{k}$-components of polarization are $\mu^*_{cv}(\boldsymbol{k})\mu_{cv}(\boldsymbol{k})\cdot\boldsymbol{\xi}$, one cannot deduce determinately that $\boldsymbol{k}$-components of the polarization in the relaxation situation are $\mu^*_{cv}(\boldsymbol{k})\mu_{cv}(\boldsymbol{k})\cdot\overline{\boldsymbol{\xi}}$ because of the summation sign. For our purpose of only considering the total polarization, however, the summation result shown in Eq.(14a) is enough, and it is the very solution to Eq.(12). It should be emphasized that the existence of $\Gamma(k)$ depends on the vectorial nature of μ_{cv}. And besides, summation procedure is necessary to obtain the expression of $\Gamma(k)$. This is important not only for obtaining the macroscopic polarization, but also for the density of photo-excited carrier. Let us perform summation on Eq.(1b) over $\boldsymbol{k}$-space. Since $\dfrac{\partial}{\partial t}\sum_k f_{\gamma k}\,|_{\mathrm{scatt}}=0$, we have

$$\frac{\partial}{\partial t}\sum_k f_{\gamma k}=2\mathrm{Im}\left[\sum_k(\rho^*_k\mu_{cv}(\boldsymbol{k}))\cdot\boldsymbol{E}(t)\right]. \tag{15}$$

But Eq.(15) does not mean that the Coulomb interaction has no effect on the evolution of carrier density of $N=2\sum_k f_{\gamma k}$, where the factor 2 arises from the spin of electrons. The dependence of N on the Coulomb interaction is connoted through the macroscopic polarization vector of $\sum_k\rho_k\mu^*_{cv}(\boldsymbol{k})$ that will attenuate due to CC scattering. The details are not concerned in this paper. At this moment, however, we are able to further understand the essential difference between $\overline{\boldsymbol{\xi}}$ in Eqs.(14) and $\boldsymbol{\xi}$ in Eqs.(8). We gain an insight into the essential of the difference by briefly discussing the equation of motion for the density of carrier. From mathematical point of view, one can construct the solution to Eq.(15) through following steps: Firstly, let each $\boldsymbol{k}$-component of Eq.(15) equal for obtaining a fictional equation, i.e., $\dfrac{\partial}{\partial t}f_{\gamma k}=2\mathrm{Im}[(\rho^*_k\mu_{cv}(\boldsymbol{k}))\cdot\boldsymbol{E}(t)]$, which presents a striking contrast to Eq.(1b). Then, the solution to Eq.(15) equals the summation of the solution to this fictional equation over all $\boldsymbol{k}$ indices. Apparently, the symbol of $f_{\gamma k}$ in this fictional equation no longer possesses a specific physical meaning of distribution function as in Eq.(1b), but the summation of $\sum_k f_{\gamma k}$ still is the density of carrier if the

initial conditions are appropriately included.

After the excitation, the asymptotic behaviour of the optically induced polarization $\mathrm{Re}(\sum_k \rho_k \mu^*_{cv}(\boldsymbol{k}))$, which possesses a double-Lorentzian-like shape, peaks at the central frequency of light field (see Eq.(16)). Let us now denote the summation of $\sum_k \rho_k \mu^*_{cv}(\boldsymbol{k})$ by $\boldsymbol{P}(t)$ and rewrite down the asymptotic result of the total polarization of $\mathrm{Re}\boldsymbol{P}(t)$ as follows ($\alpha >> \Gamma(k)$):

$$\mathrm{Re}\boldsymbol{P}(t) = \sum_k \mathrm{Re}\left[\frac{\mathrm{i}2\hbar\alpha\mu^*_{cv}(\boldsymbol{k})\mu_{cv}(\boldsymbol{k})\cdot\boldsymbol{E}_0 \exp\left(-\mathrm{i}\left(\frac{\varepsilon_{ek}+\varepsilon_{hk}}{\hbar}\right)-\Gamma(k)\right)t}{(\hbar\alpha)^2-(\hbar\Gamma(k))^2+(\varepsilon_{ek}+\varepsilon_{hk}-\hbar\omega_0)^2-\mathrm{i}2\hbar\Gamma(k)(\varepsilon_{ek}+\varepsilon_{hk}-\hbar\omega_0)}\right]. \tag{16}$$

Noticing that the double-Lorentzian shape is greatly sharper than the common Lorentzian shape, the total polarization converges on to $\boldsymbol{k}$-space and also decays with time. From Eqs.(14), the Fourier integral transform of $\boldsymbol{P}(t)$ is given by

$$\begin{aligned} P(\omega) &= \int_{-\infty}^{\infty} P(t)\exp(\mathrm{i}\omega t)\mathrm{d}t \\ &= \sum_k \left[\begin{array}{l} \dfrac{\mathrm{i}\mu^*_{cv}(\boldsymbol{k})\mu_{cv}(\boldsymbol{k})\cdot E_0}{\hbar[\alpha+\Gamma(k)]+\mathrm{i}[(\varepsilon_{ek}+\varepsilon_{hk})-\hbar\omega_0]}\cdot\dfrac{1}{\alpha+\mathrm{i}(\omega-\omega_0)} \\ +\dfrac{\mathrm{i}\mu^*_{cv}(\boldsymbol{k})\mu_{cv}(\boldsymbol{k})\cdot E_0}{\hbar[-\alpha+\Gamma(k)]+\mathrm{i}[(\varepsilon_{ek}+\varepsilon_{hk})-\hbar\omega_0]}\cdot\dfrac{1}{\alpha-\mathrm{i}(\omega-\omega_0)} \\ +\dfrac{\mathrm{i}2\hbar\alpha\mu^*_{cv}(\boldsymbol{k})\mu_{cv}(\boldsymbol{k})\cdot\boldsymbol{E}_0}{(\hbar\alpha)^2-(\hbar\Gamma(k))^2+(\varepsilon_{ek}+\varepsilon_{hk}-\hbar\omega_0)^2-\mathrm{i}2\hbar\Gamma(k)(\varepsilon_{ek}+\varepsilon_{hk}-\hbar\omega_0)} \\ \cdot\dfrac{\hbar}{\hbar\Gamma(k)+\mathrm{i}(\varepsilon_{ek}+\varepsilon_{hk}-\hbar\omega)} \end{array}\right]. \end{aligned} \tag{17}$$

One can identify that the last term on the right hand side of Eq.(17) corresponds to the asymptotic behaviour of the polarization shown in Eq.(16). The induced polarization peaks at the central frequency of light field. Then, one can evaluate the polarization decay using $\Gamma(k_0)$, where k_0 corresponds to the light frequency ω_0. For convenience in next section (Sec. 3) and considering Eqs.(3), we rewrite the total polarization dephasing rate due to CC scattering as follows:

$$\Gamma_{\mathrm{CC}}(k_0) \equiv \sum_{\gamma,\delta} \Gamma^{\gamma\delta}_{\mathrm{CC}}(k_0), \tag{18a}$$

where

$$\Gamma^{\gamma\delta}_{\mathrm{CC}}(k_0) = \frac{V}{(2\pi)^3}\int \Gamma^{\gamma\delta}(k', k_0, \cos\chi)(1-\cos\chi)\mathrm{d}\boldsymbol{k}', \tag{18b}$$

with $\Gamma^{\gamma\delta}(k', k_0, \cos\chi) = \Gamma^{\gamma\delta}(\boldsymbol{k}', \boldsymbol{k}_0)$ and $\gamma, \delta = \mathrm{e}, \mathrm{h}$. The transverse relaxation time T_2 arising from CC scattering is then defined as $T_2 \equiv 1/\Gamma_{\mathrm{CC}}(k_0)$. Consequently, Eqs.(4) and (18) indicate that the dephasing rate due to CC scattering is equal to one half of the total scattering-rate-integrals weighted by the factors $(1-\cos\chi)$ with χ the scattering angles. A zero weight is then given to the scattering which does not change the direction of carriers, and then the weighted total scattering rate is naturally called the momentum relaxation rate.

2.2. Existence of T_2 in carrier-phonon scattering processes

The dephasing rates due to CP scattering can also be discussed with the same approach as shown in Sec. 2.1. Formally, the scattering parts in Eqs.(1) due to CP scattering are the same as in Eq.(2),

$$\frac{\partial}{\partial t}\rho_k\,|_{\mathrm{scatt}} = \sum_{k'}\left(\Gamma_{k,k'}\rho_{k'} - \Gamma_{k',k}\rho_k\right), \tag{19a}$$

$$\begin{aligned} \frac{\partial}{\partial t}f_{\gamma^k}\,|_{\mathrm{scatt}} = \sum_{p,k'}&(W^{\gamma p}_{k,k'}f_{\gamma k'}(1-f_{\gamma k}) \\ &- W^{\gamma p}_{k',k}(1-f_{\gamma k'})f_{\gamma k}). \end{aligned} \tag{19b}$$

Here p denotes phonon species, i.e., p=LO, NP, AP, IV, and $\Gamma_{k,k'}$ and $\Gamma_{k',k}$ ought to be replaced

by[11,16−18]

$$\Gamma_{k',k} = \sum_{\gamma,p} \Gamma^{\gamma p}_{k',k}, \tag{20a}$$

$$\Gamma_{k,k'} = \sum_{\gamma,p} \Gamma^{\gamma p}_{k,k'}, \tag{20b}$$

where we use equations

$$\Gamma^{\gamma p}_{k',k} = \frac{1}{2}(W^{\gamma p}_{k,k'} f_{\gamma k'} + W^{\gamma p}_{k',k}(1 - f_{\gamma k'})), \tag{21a}$$

$$\Gamma^{\gamma p}_{k,k'} = \frac{1}{2}(W^{\gamma p}_{k',k} f_{\gamma k} + W^{\gamma p}_{k,k'}(1 - f_{\gamma k})). \tag{21b}$$

Here, the transition probabilities corresponding, for example, to carrier-LO-phonon scattering are given by[11,16−19]

$$W^{\gamma \mathrm{LO}}_{k,k'} = \frac{2\pi}{\hbar} \sum_{\pm,q} |M_q|^2 \delta_{k\pm q,k'} \delta[E_\gamma(k) - E_\gamma(k') \pm \hbar\omega_{\mathrm{LO}}] \left(n(\omega_{\mathrm{LO}}) + \frac{1}{2} \pm \frac{1}{2} \right), \tag{22}$$

where $n(\omega_{\mathrm{LO}})$ is the distribution of LO phonon with frequency ω_{LO}, and the matrix element M_q is defined by

$$|M_q|^2 = \frac{e^2 \hbar \omega_{\mathrm{LO}}}{2\epsilon_0 V q^2} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_{\mathrm{s}}} \right), \tag{23}$$

with the high-frequency dielectric constant ϵ_∞, and the static dielectric constant ϵ_{s}.

Following the discussion in Sec. 2.1 (see also appendix A), we introduce $\Gamma_{\mathrm{CP}}(k_0)$ to denote the depolarization rate due to CP scattering, where k_0 corresponds to the central frequency of the light field. $\Gamma_{\mathrm{CP}}(k_0)$ reads

$$\Gamma_{\mathrm{CP}}(k_0) \equiv \sum_{\gamma,p} \Gamma^{\gamma p}_{\mathrm{CP}}(k_0), \tag{24a}$$

where

$$\Gamma^{\gamma p}_{\mathrm{CP}}(k_0) = \frac{V}{(2\pi)^3} \int \Gamma^{\gamma p}(k', k_0, \cos\chi) \times (1 - \cos\chi) \mathrm{d}\boldsymbol{k}', \tag{24b}$$

with $\Gamma^{\gamma p}(k', k_0, \cos\chi) = \Gamma^{\gamma p}(\boldsymbol{k}', \boldsymbol{k}_0) \equiv \Gamma^{\gamma p}_{k',k_0}$, γ=e,h, and p=LO, NP, AP, IV.

In the low carrier density limit ($f \ll 1$), one can obtain the dephasing rate corresponding to the carrier-LO scattering:

$$\begin{aligned}
\Gamma^{\mathrm{LO}}_{\mathrm{CP}}(k_0) &= \sum_\gamma \Gamma^{\gamma \mathrm{LO}}_{\mathrm{CP}}(k_0) \\
&= \sum_\gamma \frac{V}{(2\pi)^3} \int \Gamma^{\gamma \mathrm{LO}}(k', k_0)(1 - \cos\chi) \mathrm{d}\boldsymbol{k}' \\
&= \sum_\gamma \frac{V}{(2\pi)^3} \int \frac{1}{2} W^{\gamma \mathrm{LO}}_{k',k_0}(1 - \cos\chi) \mathrm{d}\boldsymbol{k}' \\
&= \sum_{\gamma,\pm,q} \frac{\pi V}{\hbar(2\pi)^3} \int \mathrm{d}\boldsymbol{k}' |M_q|^2 \delta_{k_0 \pm q,k'} \delta[E_\gamma(k_0) - E_\gamma(k') \pm \hbar\omega_{\mathrm{LO}}] \\
&\quad \times (n(\omega_{\mathrm{LO}}) + \frac{1}{2} \pm \frac{1}{2})(1 - \cos\chi)) \\
&= \sum_\gamma \frac{e^2 \omega_{\mathrm{LO}} m_\gamma}{16\pi\epsilon_0 \hbar^2 k_0} \left(\frac{1}{\epsilon_\infty} - \frac{1}{\epsilon_{\mathrm{s}}} \right) \left\{ 2 - \ln \left(\frac{k^\gamma_f + k_0}{k^\gamma_f - k_0} \right)^2 \right\},
\end{aligned} \tag{25}$$

in which we have introduced

$$k^\gamma_f = \left(k_0^2 \pm \frac{2 m_\gamma \omega_{\mathrm{LO}}}{\hbar} \right)^{1/2}. \tag{26}$$

The signs + and − refer to the absorption and emission respectively. This equation results from energy conservation in the carrier-LO-phonon scattering processes, and shows the relationship between the final and initial states of wave vectors of carriers. We would like to point out that the dephasing rates for GaAs calculated by Eq.(25) do not present too many differences in comparison with those when the angle correction is not taken into account.

The structures of the depolarization rates for deformation potentials scattering, such as NP, Ap and IV scattering, are the same as those in the case of the polar optical phonon scattering. However, the integrands in the integrals of the dephasing rates for these deformation potentials are almost independent of $q = |\boldsymbol{k} - \boldsymbol{k}'|$.[20,21] Consequently, the integrals of the $\cos\chi$ over $d\cos\chi$ vanish (see also Eq.(25)), leading to

the dephasing rates equal to one half of the relevant total scattering rate.

The dephasing rate due to NP scattering in the low limit of distribution of carriers is mainly determined by hole-NP scattering. Thus, the dephasing, half of the common total scattering rate,[22,23] is given by

$$\begin{aligned}\Gamma_{\rm CP}^{\rm hNP} =&\frac{1}{2}\sum_{\pm}\int W_{k',k}^{\rm hNP}\,{\rm d}\boldsymbol{k}'\\ =&\frac{3d_0^2 m_{\rm h}^{3/2}}{4\sqrt{2}\pi\hbar^3 a_0^2\rho\omega_{\rm NP}}\{(E_{\rm h}(k_0)+\hbar\omega_{\rm NP})^{1/2}n_{\rm NP}\\ &+(E_{\rm h}(k_0)-\hbar\omega_{\rm NP})^{1/2}(1+n_{\rm NP})\},\end{aligned}\tag{27}$$

where d_0 is the optical deformation potential, a_0 the lattice constant, ρ the crystal density, $E_{\rm h}(k_0)$ the initial hole energy and $n_{\rm NP}$ the optical-phonon occupation number.

Unlike the optical phonon, the energy of the acoustic phonon is small. Since the momentum of AP can be compared to that of the carriers, the AP scattering contributes significantly to depolarization. Both electron and hole should then be considered in the polarization decay. The dephasing rate, one half of the total scattering rate,[20,21] is given by

$$\begin{aligned}\Gamma_{\rm CP}^{\rm AP} =&\sum_{\gamma}\Gamma_{\rm CP}^{\gamma\,\rm AP}\\ =&\sum_{\gamma}\frac{3D_{\rm AP}^2 k_{\rm B}T\cdot(E_\gamma(k_0))^{1/2}m_\gamma}{\sqrt{2}\pi\hbar^4\rho v_L^2},\end{aligned}\tag{28}$$

where T is the lattice temperature, $D_{\rm AP}$ the deformation potential, and v_L the speed of sound.

In the IV-phonon-scattering processes an electron is scattered from the central valley to one of the outer valleys either by absorption or by emission of a phonon. The dephasing rates due to IV-phonon-scattering, which equal to one half of the common IV scattering rates,[21,24] are then given as

$$\Gamma_{\rm CP}^{{\rm e}\Gamma X}=\frac{3D_{\Gamma X}^2 m_X^{3/2}}{2\sqrt{2}\pi\hbar^3\rho\omega_{\Gamma X}}\left\{(E_{\rm e}(k_0)+\hbar\omega_{\Gamma X}-E_{\Gamma X})^{1/2}\,n_{\Gamma X}+(E_{\rm e}(k_0)-\hbar\omega_{\Gamma X}-E_{\Gamma X})^{1/2}\,(1+n_{\Gamma X})\right\},\tag{29a}$$

$$\Gamma_{\rm CP}^{{\rm e}\Gamma L}=\frac{2D_{\Gamma L}^2 m_L^{3/2}}{\sqrt{2}\pi\hbar^3\rho\omega_{\Gamma L}}\left\{(E_{\rm e}(k_0)+\hbar\omega_{\Gamma L}-E_{\Gamma L})^{1/2}\,n_{\Gamma L}+(E_{\rm e}(k_0)-\hbar\omega_{\Gamma L}-E_{\Gamma L})^{1/2}\,(1+n_{\Gamma L})\right\}.\tag{29b}$$

$E_{\rm e}(k_0)$ is the initial electron energy measured from the Γ valley edge. $D_{\Gamma X}$ ($D_{\Gamma L}$) is the deformation potential for the Γ-to-X (L) intervalley electron-phonon interaction; m_X (m_L) is the effective mass of the carriers in the X (L) valley. $E_{\Gamma X}(E_{\Gamma L})$ is the separations between the conduction-band minimum at Γ and that at $E_{\Gamma X}(E_{\Gamma L})$. $n_{\Gamma X}(n_{\Gamma L})$ is the occupation number of IV-phonon with frequency $\omega_{\Gamma X}$ ($\omega_{\Gamma L}$).

Neglecting the interference effects between Coulomb and CP interactions, the total dephasing rate Γ_t reads

$$\begin{aligned}\Gamma_t =&\Gamma_{\rm CC}(k_0)+\Gamma_{\rm CP}(k_0)\\ =&\sum_{\gamma,\delta}\Gamma_{\rm CC}^{\gamma\delta}(k_0)+\sum_{\gamma,p}\Gamma_{\rm CP}^{\gamma p}(k_0),\end{aligned}\tag{30}$$

with γ,δ=e,h, and p=LO, NP, AP, IV. The total transverse relaxation time T_2 is then defined as $T_2\equiv 1/\Gamma_t$.

3. Uncertainty broadening approach of dephasing due to carrier-carrier scattering

Scattering rate is often calculated by taking its spectral function of one-particle states as a sharp δ function to avoid overtaxing numerical capabilities[5,6] as we did for CP scattering in Sec. 2.2. However, as long as the Coulomb interaction is involved, the scattering rate for a purely Coulombic potential is effectively infinite. In equilibrium physical case, the Brooks-Herring approach is always adopted to overcome this problem, in which the screened potential drops off more rapidly with the distance between the interacting bodies. This is because electrons in the bands of semiconductors can move in response to electric fields at sufficiently long time scales. Consequently, screening of the Coulomb field may occur.[25] For the femtosecond-pulse excitation, the femtosecond echo decay time falls onto the femtosecond time scale. It is argued that on such a short time the screening is not sufficiently built up.[26] Considering this point of view, we calculate the dephasing rate using the uncertainty broadening approach proposed by Fujita *et al.*[27]

For example, the transfer rate of polarization from state $\boldsymbol{k}$ to $\boldsymbol{k}'$ in the low density limit ($f \ll 1$), corresponding to an electron scattered by holes, is given by (see also Eqs.(4) and (5))

$$\begin{aligned}\Gamma^{\mathrm{eh}}_{k',k} =&\frac{2\pi}{\hbar}\sum_{K,K'}|M|^2\,\delta_{k+K,k'+K'}\delta[E_{\mathrm{e}}(k)+E_{\mathrm{h}}(K)\\ &-E_{\mathrm{e}}(k')-E_{\mathrm{h}}(K')]f_{\mathrm{h}K},\end{aligned}\tag{31}$$

where the matrix elements $M = e^2/\left[\epsilon_0\epsilon(\boldsymbol{k}-\boldsymbol{k}')^2V\right]$. Indeed, the spectral functions entering the relaxation rates are broadened peaks rather than sharp δ functions, where the broadening is itself determined by the relaxation rates to be calculated.[16,28,29] The broadened δ function corresponding to Eq.(31) with the angle corrections taken into account can be given by[16,25,27−29]

$$D(E) = \frac{1}{\pi}\frac{\hbar\Gamma}{E^2+(\hbar\Gamma)^2}.\tag{32}$$

Here $E = E_{\mathrm{e}}(k)+E_{\mathrm{h}}(K)-E_{\mathrm{e}}(k')-E_{\mathrm{h}}(K')$ and $\Gamma = \Gamma^{\mathrm{e}}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{e}}_{\mathrm{CC}}(k')+\Gamma^{\mathrm{h}}_{\mathrm{CC}}(K)+\Gamma^{\mathrm{h}}_{\mathrm{CC}}(K')$. $\Gamma^{\mathrm{e}}_{\mathrm{CC}}(k)$ and $\Gamma^{\mathrm{h}}_{\mathrm{CC}}(K)$ are the carrier damping coefficients with the angle corrections for electrons and holes, respectively, which result from the CC scattering by different processes. For instance, the electron damping reads

$$\Gamma^{\mathrm{e}}_{\mathrm{CC}}(k) = \Gamma^{\mathrm{ee}}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k).\tag{33}$$

Here, $\Gamma^{\mathrm{ee}}_{\mathrm{CC}}(k)$ is the damping due to electron-electron collisions, $\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k)$ is the damping due to electron-hole collisions. Because of the very small value of the photon momentum, light couples valence and conduction band states with essentially the same wave vector $\boldsymbol{k}$. The wave vectors of holes are then almost equal to those of electrons, i. e., $K \simeq k$. In addition, since the transition probability for Coulomb scattering is proportional to $1/(\boldsymbol{k}-\boldsymbol{k}')^4$, the integrand of scattering rate peaks sharply at $k' = k$.[25,27] Thus, noticing Eq.(18), one deduces that

$$\begin{aligned}\Gamma \simeq&2[\Gamma^{\mathrm{ee}}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{h}h}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{he}}_{\mathrm{CC}}(k)]\\ \equiv&2\Gamma_{\mathrm{CC}}(k).\end{aligned}\tag{34}$$

This equation shows that Γ in the broadened peak function is twice as large as the dephasing rate $\Gamma_{\mathrm{CC}}(k)$ due to CC scattering. In other words, Γ equals the so-called momentum relaxation rate, which has already been pointed out by Fujita *et al.*[25,27] Consequently $\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k)$, for example, is given in the low-density limit by (see Eq.(18))

$$\begin{aligned}\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k) =&\frac{V}{(2\pi)^3}\int\Gamma^{\mathrm{eh}}(k,k',\cos\chi)(1-\cos\chi)\mathrm{d}\boldsymbol{k}'\\ =&\frac{2\pi}{\hbar}\left[\frac{V}{(2\pi)^3}\right]^3\int\mathrm{d}\boldsymbol{k}'\mathrm{d}\boldsymbol{K}\mathrm{d}\boldsymbol{K}'\\ &\times\left(\frac{e^2}{\epsilon_0\epsilon(\boldsymbol{k}-\boldsymbol{k}')^2V}\right)^2\delta_{k+K,k'+K'}\\ &\times\frac{1}{\pi}\frac{2\hbar\Gamma_{\mathrm{CC}}(k)}{E^2+(2\hbar\Gamma_{\mathrm{CC}}(k))^2}f_{\mathrm{h}K}(1-\cos\chi).\end{aligned}\tag{35}$$

According to the discussion of Fujita *et al*,[13,25,27] the term of $E = E_{\mathrm{e}}(k)+E_{\mathrm{h}}(K)-E_{\mathrm{e}}(k')-E_{\mathrm{h}}(K')$ in the denominator of Eq.(35) can be neglected against $2\hbar\Gamma_{\mathrm{CC}}(k)$ since $1/(\boldsymbol{k}-\boldsymbol{k}')^4$ has an extremely sharp maximum at $k' = k$. We therefore obtain

$$\begin{aligned}\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k) =&\frac{N}{2(2\pi)^3\hbar^2}\int\mathrm{d}\boldsymbol{k}'\left(\frac{e^2}{\epsilon_0\epsilon(\boldsymbol{k}-\boldsymbol{k}')^2}\right)^2\\ &\times\frac{1}{\Gamma_{\mathrm{CC}}(k)}(1-\cos\chi),\end{aligned}\tag{36}$$

in which $N = 2\int f_{Kh}\mathrm{d}^3K/(2\pi)^3$ is the hole density. For the optical pulse excitation, N is also the density of electrons. We rewrite Eq.(36) as follows

$$\begin{aligned}\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k)\Gamma_{\mathrm{CC}}(k) =&\frac{N}{2(2\pi)^3\hbar^2}\int\mathrm{d}\boldsymbol{k}'\left(\frac{e^2}{\epsilon_0\epsilon(\boldsymbol{k}-\boldsymbol{k}')^2}\right)^2\\ &\times(1-\cos\chi).\end{aligned}\tag{37}$$

Using the same procedures, we have

$$\begin{aligned}\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k)\Gamma_{\mathrm{CC}}(k) =&\Gamma^{\mathrm{he}}_{\mathrm{CC}}(k)\Gamma_{\mathrm{CC}}(k)\\ =&\Gamma^{\mathrm{ee}}_{\mathrm{CC}}(k)\cdot2(\Gamma^{\mathrm{ee}}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k))\\ =&\Gamma^{\mathrm{h}h}_{\mathrm{CC}}(k)\cdot2(\Gamma^{\mathrm{h}h}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{he}}_{\mathrm{CC}}(k))\\ =&\frac{N}{2(2\pi)^3\hbar^2}\int\mathrm{d}\boldsymbol{k}'\left(\frac{e^2}{\epsilon_0\epsilon(\boldsymbol{k}-\boldsymbol{k}')^2}\right)^2\\ &\times(1-\cos\chi);\end{aligned}\tag{38a}$$

thus,

$$\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k) = \Gamma^{\mathrm{he}}_{\mathrm{CC}}(k) = \Gamma^{\mathrm{ee}}_{\mathrm{CC}}(k) = \Gamma^{\mathrm{h}h}_{\mathrm{CC}}(k);\tag{38b}$$

and

$$\begin{aligned}\Gamma_{\mathrm{CC}}(k) =&\Gamma^{\mathrm{ee}}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{eh}}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{h}h}_{\mathrm{CC}}(k)+\Gamma^{\mathrm{he}}_{\mathrm{CC}}(k)\\ =&\frac{4N}{2(2\pi)^3\hbar^2}\int\mathrm{d}\boldsymbol{k}'\left(\frac{e^2}{\epsilon_0\epsilon(\boldsymbol{k}-\boldsymbol{k}')^2}\right)^2\\ &\times\frac{1}{\Gamma_{\mathrm{CC}}(k)}(1-\cos\chi).\end{aligned}\tag{38c}$$

Consequently, we have

$$\begin{aligned}(\Gamma_{\mathrm{CC}}(k))^2 =&\frac{N}{4\pi^3\hbar^2}\left(\frac{e^2}{\epsilon_0\epsilon}\right)^2\int\mathrm{d}\boldsymbol{k}'\frac{1}{(\boldsymbol{k}-\boldsymbol{k}')^4}\\ &\times(1-\cos\chi).\end{aligned}\tag{39}$$

According to Fujita *et al*,[25,27] the integral on the right hand side of Eq.(39) can be calculated exactly as $4\pi/k$. Thus

$$(\Gamma_{\rm CC}(k))^2 = \frac{N}{k}\left(\frac{e^2}{\epsilon_0\epsilon\pi\hbar}\right)^2. \qquad (40)$$

Therefore, when the dephasing due to CP scattering is not taken into account, the dephasing rate arising from CC scattering, without the effect of screening, is given by

$$\Gamma_{\rm CC}(k) = \frac{e^2}{\epsilon_0\epsilon\pi\hbar}\frac{N^{1/2}}{k^{1/2}}, \qquad (41)$$

which is a simple function of the excited carrier density and free of adjustable parameters. One also sees that Eq.(41) is free of the masses of the carriers for a certain magnitude of the Bloch vectors. This implies that all the scattering processes, such as, electron-electron (ee) scattering, electron-hole (eh) scattering, hole-hole scattering (hh) and so on, have contributions to the dephasing equivalently (see also Eq.(38b)). Furthermore, from the dimension analysis, the expression consists simply of the electron charge symbolizing the Coulomb interaction, the Planck constant symbolizing the quantum effect and the size of the Bloch wave vectors indicating a weak dependence of the excess energy of the excited carriers. It is worth pointing out that uncertainty broadening has also removed the divergence for polarization relaxation due to pure Coulomb scattering without screening.

It should be noticed that the Lorentzian function of Eq.(32) reduces to delta-function $\delta[E_{\rm e}(k)+E_{\rm h}(K)-E_{\rm e}(k')-E_{\rm h}(K')]$ in the limit of small dephasing rate $\Gamma_{\rm CC}$, if other factors in the integrand of Eq.(35) vary slowly in the neighbourhood of $E_{\rm e}(k)+E_{\rm h}(K) = E_{\rm e}(k')+E_{\rm h}(K')$, which reflects the energy conservation during the scattering processes. For the purpose of this paper, it is especially important to emphasize these comparisons among the integrands of integrals. For example, if an integral only involves a Lorentzian integrand as shown in Eq.(32), integration over the whole phase-space will give unity regardless of the value of Γ in Eq.(32). By the way, we can now realize that the replacement of $D(E)$ by a δ function for the integrations of the dephasing rates due to CP scattering is justified since the other integrand factors vary slowly under of the condition of close to energy conservation.

In the case of CC scattering however, since the square of the Fourier-transformed potential $V_q = e^2/(\epsilon_0\epsilon q^2)$ has a stronger maximum at $q = 0$ than the maximum due to the Lorentzian function of Eq.(32), the Lorentzian function can no longer be replaced by the energy conserving delta-function in the Coulomb interaction. The time required for the build-up of screening can be evaluated by the inverse plasma frequency.[26] Consequently, the screening does not play an important role in the depolarization processes at low densities, i.e. $N \leq 5\times10^{18}$ cm^{-3}, for optically excited bulk semiconductors. At very high densities, the maximum in the Lorentzian function $D(E)$ (Eq.(32)) may become steeper than the maximum in the square of the screened Coulomb potential. In this density regime the Lorentzian function $D(E)$ may then be replaced by the energy-conserving delta-function.

If we assumed that the screening could be built up at lower densities and that it were accounted for by the Debye–Hüeckel theory, the Fourier transform of the Coulomb potential, $V_q = e^2/(\epsilon_0\epsilon q^2)$, would be replaced by the screened one, namely $V_q = e^2/(\epsilon_0\epsilon(q^2+q_{\rm D}^2))$, where $q_{\rm D}$ is the Debye–Hüeckel wave vector. For Gaussian distribution of the optical excitation, $q_{\rm D}$ is given by

$$q_{\rm D}^2 = \frac{e^2N(m_{\rm e}+m_{\rm h})}{\epsilon_0\epsilon\hbar^2k_0^2}, \qquad (42)$$

where $m_{\rm e}$ ($m_{\rm h}$) is the electron (hole) mass and $k_0^2 = 2m_r(\hbar\omega_0 - E_g)/\hbar^2$ with the reduced mass m_r and photon energy $\hbar\omega_0$. At lower densities, the square of the screening potential V_q is still steeper than the Lorentzian function. Again, according to Fujita *et al*,[25,27] the screening effect might be taken into account by multiplying the square root of screening factor $g(x)$ in the right hand side term of Eq.(41), and $g(x)$ is given by

$$g(x) \equiv \frac{1}{2}(1+\frac{\pi}{2}x - (x+x^{-1})\tan^{-1}x), \qquad (43)$$

with $x \equiv k_0/q_{\rm D}$. The dephasing rate with screening effect might read

$$\Gamma_{\rm CC}(k_0) = \frac{e^2}{\epsilon_0\epsilon\pi\hbar}\frac{N^{1/2}}{k_0^{1/2}}g(x)^{1/2}. \qquad (44)$$

One can find that, even if the screening are built up immediately and effectively, this effect does not play a prominent role in the polarization relaxation except for the nearest band edge excitation (see also Fig.1).

4. Discussion and comparison with the reported experimental results

At this point it may be worth while to recapitulate what we have developed so far in this paper. We summarize two general expressions of polarization relaxation rates for continuum transitions in bulk semiconductors as follows:

$$\begin{aligned}\Gamma_t =&\Gamma_{\rm CC}(k_0)+\Gamma_{\rm CP}\\ =&\sum_{\gamma,\delta}\Gamma_{\rm CC}^{\gamma\delta}(k_0)+\sum_{\gamma,p}\Gamma_{\rm CP}^{\gamma p}\\ =&\frac{e^2}{\epsilon_0\epsilon\pi\hbar}\frac{N^{1/2}}{k_0^{1/2}}+\sum_{\gamma}\frac{e^2\omega_{\rm LO}m_\gamma}{16\pi\epsilon_0\hbar^2k_0}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_{\rm s}}\right)\\ &\times\left\{2-\ln\left(\frac{k_f^\gamma+k_0}{k_f^\gamma-k_0}\right)^2\right\}\\ &+\Gamma_{\rm CP}^{{\rm h}NP}+\Gamma_{\rm CP}^{\rm AP}+\Gamma_{\rm CP}^{{\rm e}\Gamma X}+\Gamma_{\rm CP}^{{\rm e}\Gamma L},\end{aligned}\tag{45a}$$

without screening effects; and

$$\begin{aligned}\Gamma_t =&\frac{e^2}{\epsilon_0\epsilon\pi\hbar}\frac{N^{1/2}}{k_0^{1/2}}g(x)^{1/2}\\ &+\sum_{\gamma}\frac{e^2\omega_{\rm LO}m_\gamma}{16\pi\epsilon_0\hbar^2k_0}\left(\frac{1}{\epsilon_\infty}-\frac{1}{\epsilon_{\rm s}}\right)\\ &\times\left\{2-\ln\left(\frac{k_f^\gamma+k_0}{k_f^\gamma-k_0}\right)^2\right\}\\ &+\Gamma_{\rm CP}^{{\rm h}NP}+\Gamma_{\rm CP}^{\rm AP}+\Gamma_{\rm CP}^{{\rm e}\Gamma X}+\Gamma_{\rm CP}^{{\rm e}\Gamma L},\end{aligned}\tag{45b}$$

for the screening effects to be built up immediately. Here, $\Gamma_{\rm CP}^{{\rm h}NP}, \Gamma_{\rm CP}^{\rm AP}, \Gamma_{\rm CP}^{{\rm e}\Gamma X}$, and $\Gamma_{\rm CP}^{{\rm e}\Gamma L}$ denote the dephasing rates corresponding to non-polar phonon scattering, acoustic phonon scattering, inter-valley scattering from Γ to X and from Γ to L, respectively. Each one equals one half of its common scattering rate. The total transverse relaxation time T_2 is defined as $T_2 \equiv 1/\Gamma_t$.

Figure 1(a) shows the dephasing rate versus the photon energy without screening (see Eqs.(45a)) in GaAs with two parabolic bands. The calculated depolarization rates correspond to the transitions from the heavy-hole band to the conduction band at room temperature (GaAs parameters[21,22,24,30,31] used in the model calculation : $\epsilon_\infty = 10.9$, $\epsilon_{\rm s} = 12.9$, $m_{\rm e} = 0.067m_0$, $m_{\rm h} = 0.45m_0$, $m_X = 0.58m_0$, $m_L = 0.222m_0$, $D_{\rm AP} = 7$eV, $\rho = 5.37g/cm^3$, $d_0 = 41$eV, $v_L = 5.22\times10^5$cm/s, IV-deformation potentials $D_{\Gamma L} = 1.5\times10^8$eV/cm, $D_{\Gamma X} = 10\times10^8$eV/cm, $\hbar\omega_{\rm LO} = \hbar\omega_{\rm NP} = 36$meV, IV-phonon energy, $\hbar\omega_{\Gamma X}$=30meV and $\hbar\omega_{\Gamma L}$=27.9meV, band-gap energy $E_g(T = 0)$=1.519eV, $E_{\Gamma X}$=0.46eV, $E_{\Gamma L}$=0.29eV). It can be seen that the dephasing rate increases with increasing carrier density. At very low energy states near the band edge, Fig.1(a) shows the growing trend of the unscreened dephasing rate with decreasing energy. This is pathological. Owing to the small density of states at the band edge, the weak excitation limit ($f << 1$) is no longer valid and the Pauli exclusion principle should be taken into account, which will smooth the growing trend of dephasing near the energy band-edge. Let us now suppose that the screening may be able to be built up both immediately and effectively so that the effect could be described by the Debye–Hüeckel theory. When this static screening are included, the dephasing rates with the screening factor $g(x)$ as shown by Eq.(43) are almost equal to those without the screening effect except for the nearest band edge (the related picture is shown in Fig.1(b),

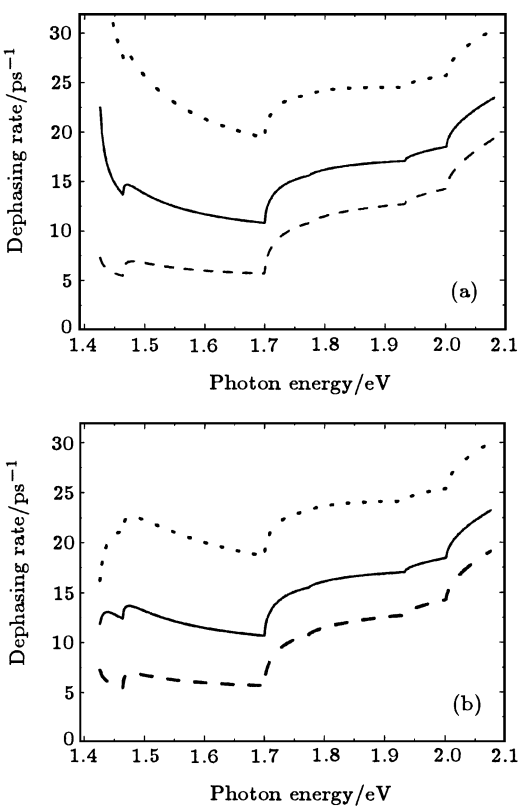


Fig.1. (a) The calculated dephasing rate without screening versus the photon energy at room temperature for various carrier densities: 1×10^{15}cm^{-3} (dashed curve), 5×10^{16}cm^{-3} (solid curve), and 3×10^{17}cm^{-3} (dotted curve). (b) The calculated dephasing rate with screening (if it is assumed to be built up) versus the photon energy at room temperature for various carrier densities: 1×10^{15}cm^{-3} (dashed curve), 5×10^{16}cm^{-3} (solid curve), and 3×10^{17}cm^{-3} (dotted curve).

where the Pauli blocking is still ignored.) Since the CP scattering rates are calculated by taking the spectral function of one-particle states as the sharp δ functions, it can also be noted that there are some dips in each curve. Two of them mark the onset of phonon-emission for electrons at $E \sim 1.46$eV and $E \sim 2$eV corresponding to the final electron states in Γ valley and X valley, respectively; one of them marks the onset of phonon-emission for heavy holes at $E \sim 1.7$eV. Another dip at $E \sim 1.94$eV marks the onset of intervalley scattering between $\Gamma \longrightarrow X$ for electrons because of phonon-absorption. Since $D_{\Gamma L}$ is smaller than $D_{\Gamma X}$,[24] the dip corresponding to the phonon-emission and absorption for $\Gamma \longrightarrow L$ appears inconspicuously. In addition, owing to the uncertainty broadening which softens the forbidden-allowed transition between the emission of phonons,[32] one should not expect that the dips should obviously occur in the experimental cases.

The uncertainty broadening rate predicts $T_2 \propto N^{-0.5}$ due to CC scattering, which deviates seriously from the experimental power-law, $T_2(\text{fs}) = 4 \times 6.8[N(10^{18}\text{cm}^{-3})]^{-0.3}$, where $T_2 = 4\tau$.[33] However, in regard of the CP scattering the calculated value is in agreement with the experimental results. In the approximation of ignoring the band-structure effects, we calculate the dephasing rates in higher energy states ($\hbar\omega_0 = 2$eV[1]). The density-independent dephasing rate Γ_{CP} due to CP scattering is estimated to be $\sim 14\text{ps}^{-1}$. The solid curve in Fig.2 describes the calculated dephasing time T_2 versus the carrier density in comparison with the measured values by Becker *et al.*[1] The dotted line is fitted using the experimental power-law,[1] i.e., $T_2(\text{fs}) = 4 \times 6.8(N(10^{18}\text{cm}^{-3}))^{-0.3}$. The dashed line is the Coulomb scattering dephasing time calculated by using Eq.(41) regardless of the CP scattering, and it deviates seriously from the measured data. One may question why the calculated results are in good agreement with the experimental data without a realistic band structure. This results from the fact that the Coulomb potential favours the exchange of small wave vectors, and then the weak k-dependence of the dephasing occurs. Besides, the contribution of the CP scattering to the depolarization is chosen to be close to the realistic case. Consequently, our model reflects the density-dependence of the dephasing and is insensitive to the nonparabolicity and anisotropy of the bands.

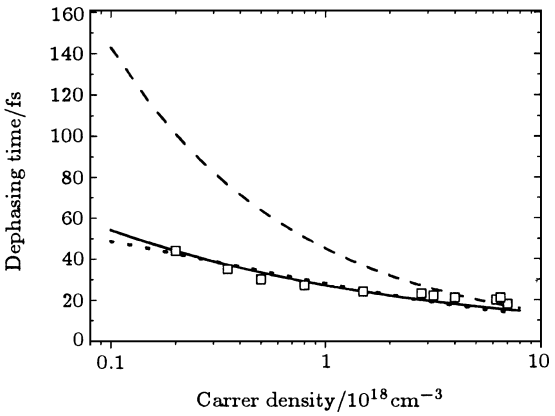


Fig.2. The calculated dephasing time (solid curve) as a function of carrier density for photon energy of 2 eV at room temperature. The measured dephasing time and the power-law fitted curve by Becker *et al*[1] are shown in open squares and the dotted curves, respectively. The dashed curve is the calculated Coulomb scattering dephasing time.

For the experimental cases where the excited carriers occupy energy states low enough to warrant the parabolic approximation, one can safely use the expressions given above to calculate the dephasing rates. In the experiments by Arlt *et al*,[2] the laser spectrum is centred at 1.54eV. Γ_{CP} is estimated to be $\sim 5\text{ps}^{-1}$. Figure 3 shows the dephasing time T_2 calculated from our model in comparison with the data points measured by Arlt *et al*[2] (the data points are obtained by using the relation $T_2 = 4\tau$ for the inhomogeneously broadened system, where τ is the measured decay time of echo signal.[33,34] The solid line is the calculated dephasing time T_2 and the dotted one is the fitted results from Ref.[2] (again $T_2 = 4\tau$). The dashed curve is the calculated Coulomb scattering dephasing time (the correction due to screening effect is neglected). Although the computed dephasing times are slightly larger than the measured values, the trend of the theoretical dephasing rate which grows at decreasing density agrees with the measurements. It is possible that additional scattering mechanisms, such as carrier-impurity scattering, play a role while they have not been taken into account in the calculations. Considering the succinctness of Eqs.(45) which is free of any adjustable parameters, the analytical expression is successful in explaining the essence of dephasing and in calculating the dephasing rate in the optically excited semiconductors.

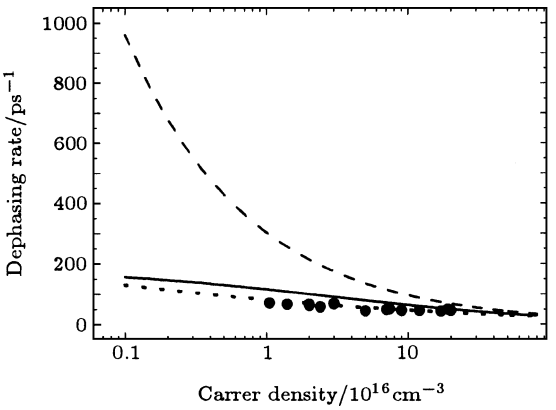


Fig.3. The calculated dephasing time of GaAs (solid curve) versus carrier density for photon energy of 1.54eV at room temperature. The measured dephasing times of GaAs sample in Ref.[2] are shown in filled circles. The dotted curve is the empirical curve obtained by using the approach of Ref.[2]. The dashed curve is the calculated Coulomb scattering dephasing time.

5. Summary

We have demonstrated the existence of the transverse relaxation time in optically excited bulk semiconductors and shown that the dephasing rates due to CC scattering and carrier-LO-phonon scattering are equal to half the so-called momentum relaxation rate. The dephasing rates arising from the deformation potentials scattering are equal to half their common scattering rates. These results arise from the vector property of the interband transition matrix elements in the total polarization. Since the Coulomb potential favours the exchange of small wave vectors in the scattering processes, the concept of transverse relaxation time is more valid in the Coulomb interaction than that in the phonon scattering regime. Based on the uncertainty broadening due to collisions, we also give the analytical expressions of the polarization dephasing due to CC scattering for bulk semiconductors, which can be used to explain the experimental results and provide a promising physical picture.

Appendix A

Now, we demonstrate Eqs.(11), i.e.

$$\frac{\partial}{\partial t}\sum_k \rho_k \mu_{cv}^*(\boldsymbol{k}) \mid_{\text{scatt}} = -\sum_k \Gamma(k)\rho_k \mu_{cv}^*(\boldsymbol{k}).$$

An exchange of the variables $\boldsymbol{k}$ and $\boldsymbol{k}'$ in the first term on the right side of Eq.(7) does not change the result of the summations, and thus Eq.(7) can be rewritten as

$$\frac{\partial}{\partial t}\sum_k \mu_{cv}^*(\boldsymbol{k})\rho_k \mid_{\text{scatt}} = -\sum_{k,k'} \Gamma_{k',k}\rho_k \left(\mu_{cv}^*(\boldsymbol{k}) - \mu_{cv}^*(\boldsymbol{k}')\right). \tag{A1}$$

Now, without loss of generality, we can concentrate our discussion on the Z component of the polarization. After switching the summation over $\boldsymbol{k}'$ to the formalism of integral over continuous coordinates, Eq.(A1) can be expressed as

$$\frac{\partial}{\partial t}\int \rho(\boldsymbol{k})\mu_z^*(\boldsymbol{k})\mathrm{d}\boldsymbol{k} \mid_{\text{scatt}} = -\frac{V}{(2\pi)^3}\int \mathrm{d}\boldsymbol{k}\left\{\rho(\boldsymbol{k})\int \mathrm{d}\boldsymbol{k}'\Gamma(k',\boldsymbol{k}) \times \left[\mu_z^*(\boldsymbol{k}) - \mu_z^*(\boldsymbol{k}')\right]\right\}, \tag{A2}$$

where V is the related volume and $\mu_z^*(\boldsymbol{k}) = \mu_{cv}^*(\boldsymbol{k})\cdot\hat{z}$, with $\hat{z}$ the unit vector of Z axis. The matrix elements $\mu_{cv}(\boldsymbol{k})$ are often replaced by $-\mathrm{i}e\boldsymbol{p}_{cv}/m\omega_0$ in the resonant approximation[35] of semiconductor physics, where $\boldsymbol{p}_{cv}$ is the momentum matrix element between the two optically coupled states with wave-vector $\boldsymbol{k}$ in the valence and conduction bands. For optical transitions in the Kane model,[36] $\boldsymbol{p}_{cv}$ (or $\mu_{cv}(\boldsymbol{k})$) is perpendicular to the wave vector $\boldsymbol{k}$ for the transition from the heavy-hole valence band (hh-c) while parallel to $\boldsymbol{k}$ for the transition from the light-hole band (lh-c).[36–38] If the direction of the wave-vector $\boldsymbol{k}$ is defined by the polar and azimuthal angles θ and ϕ, the projections of the interband matrix elements:[36–38]

$$\mu_z^*(\boldsymbol{k}) = \mu\sin\theta, \quad \text{for hh} - \text{c} \tag{A3}$$

and

$$\mu_z^*(\boldsymbol{k}) = \mathrm{i}\mu\cos\theta, \quad \text{for lh} - \text{c}, \tag{A4}$$

where $\mu = -eP/(\sqrt{2}\hbar\omega_0)$ with P the Kane momentum parameter. Therefore, for lh-c the integral in the term on the right hand side of Eq.(A2) takes the form

$$\int \mathrm{d}\boldsymbol{k}'\Gamma(k',k)\left[\mu_z^*(\boldsymbol{k}) - \mu_z^*(\boldsymbol{k}')\right] \equiv \int \mathrm{d}\boldsymbol{k}'\Gamma(k',k)\mathrm{i}\mu\left(\cos\theta - \cos\theta'\right). \tag{A5}$$

This integral can be simplified as follows. We take the direction of the wave vector $\boldsymbol{k}$ as the polar axis for the polar coordinates. Let θ and φ be the polar and azimuthal angles of Z axis relative to this polar axis, and χ, φ' be those of the wave vector $\boldsymbol{k}'$, respectively. Then, we have

$$\cos\theta' = \cos\theta\cos\chi + \sin\theta\sin\chi\cos(\varphi - \varphi'). \tag{A6}$$

The volume element $\mathrm{d}\boldsymbol{k}' = k'^2\mathrm{d}k'\sin\chi\mathrm{d}\varphi'$, where $\chi = \cos^{-1}(\boldsymbol{k}\cdot\boldsymbol{k}'/kk')$. Since $\Gamma(k',k) = \Gamma(k',k,\cos\chi)$ is independent of the angle φ', the integral of the term in $\cos(\varphi-\varphi')$ gives zero from the integration over $\mathrm{d}\varphi'$. Thus,

$$\int \mathrm{d}\boldsymbol{k}'\Gamma(\boldsymbol{k}',\boldsymbol{k})\mathrm{i}\mu(\cos\theta-\cos\theta') \equiv \int \mathrm{d}\boldsymbol{k}'\Gamma(k',k,\cos\chi)\mathrm{i}\mu\cos\theta(1-\cos\chi). \quad \text{(A7)}$$

Considering Eq.(A2), Eq.(A7) can be rewritten as

$$\int \mathrm{d}\boldsymbol{k}'\Gamma(k',k)\left[\mu_z^*(\boldsymbol{k})-\mu_z^*(\boldsymbol{k}')\right] \equiv \mu_z^*(\boldsymbol{k})\int\Gamma(k',k,\cos\chi)(1-\cos\chi)\mathrm{d}\boldsymbol{k}'. \quad \text{(A8)}$$

i.e.,

$$\frac{V}{(2\pi)^3}\int \mathrm{d}\boldsymbol{k}'\Gamma(k',k)\left[\mu_z^*(\boldsymbol{k})-\mu_z^*(\boldsymbol{k}')\right] \equiv \Gamma(k)\mu_z^*(\boldsymbol{k}), \quad \text{(A9a)}$$

where $\Gamma(k)$ is defined by Eq.(11c), i.e.,

$$\Gamma(k) = \frac{V}{(2\pi)^3}\int\Gamma(k',k,\cos\chi)(1-\cos\chi)\mathrm{d}\boldsymbol{k}'. \quad \text{(A9b)}$$

Substituting Eq.(A9) into Eq.(A2), one obtains

$$\frac{\partial}{\partial t}\int\rho(\boldsymbol{k})\mu_z^*(\boldsymbol{k})\mathrm{d}\boldsymbol{k}\mid_{\mathrm{scatt}} = -\int\Gamma(k)\rho(k)\mu_z^*(\boldsymbol{k})\mathrm{d}\boldsymbol{k}, \quad \text{(A10)}$$

or in the summation formalism

$$\frac{\partial}{\partial t}\sum_k\rho_k\mu_z^*(\boldsymbol{k})\mid_{\mathrm{scatt}} = -\sum_k\Gamma(k)\rho_k\mu_z^*(\boldsymbol{k}). \quad \text{(A11)}$$

As the choice of the Z axis is discretionary, Eq.(A11) should be valid for any coordinate projection. Thus, Eqs.(11) are obtained for lh-c. Apparently, these equations are valid for both CC scattering and CP scattering. For hh-c , however, Eq.(A5) ought to be replaced by

$$\int \mathrm{d}\boldsymbol{k}'\Gamma(k',k)\left[\mu_z^*(\boldsymbol{k})-\mu_z^*(\boldsymbol{k}')\right] \equiv \int \mathrm{d}\boldsymbol{k}'\Gamma(k',k)\mathrm{i}\mu(\sin\theta-\sin\theta'). \quad \text{(A12)}$$

Here,

$$\sin^2\theta' = 1-[\cos\theta\cos\chi+\sin\theta\sin\chi\cos(\varphi-\varphi')]^2, \quad \text{(A13)}$$

i.e.,

$$\sin\theta' = \sin\theta\cos\chi\Phi(\varphi,\varphi',\theta,\chi), \quad \text{(A14)}$$

with

$$\Phi(\varphi,\varphi',\theta,\chi) = \{1+\cot^2\theta\tan^2\chi+\tan^2\chi\sin^2(\varphi-\varphi') - 2\cot\theta\tan\chi\cos(\varphi-\varphi')\}^{1/2}. \quad \text{(A15)}$$

Then, one has

$$\int \mathrm{d}\boldsymbol{k}'\Gamma(k',k)[\mu_z^*(\boldsymbol{k})-\mu_z^*(\boldsymbol{k}')] \equiv \mu_z^*(k)\int \mathrm{d}\boldsymbol{k}'\Gamma(k',k,\cos\chi) \times(1-\cos\chi)\Phi(\varphi,\varphi',\theta,\chi), \quad \text{(A16)}$$

i.e.,

$$\frac{V}{(2\pi)^3}\int \mathrm{d}\boldsymbol{k}'\Gamma(k',k)\left[\mu_z^*(\boldsymbol{k})-\mu_z^*(\boldsymbol{k}')\right] \equiv \Gamma(k)\mu_z^*(\boldsymbol{k}), \quad \text{(A17a)}$$

where $\Gamma(k)$ is defined by

$$\Gamma(k) = \frac{V}{(2\pi)^3}\int \mathrm{d}\boldsymbol{k}'\Gamma(k',k,\cos\chi) \times\{(1-\cos\chi)\cdot\Phi(\varphi,\varphi',\theta,\chi). \quad \text{(A17b)}$$

Because Eq.(A17b) for hh-c is more complicated than Eq.(A9b) for lh-c, the transverse relaxation rate here is a little difficult to be discussed. Even so, one can easily see that zero weight is also given to scattering which dose not change the direction of carriers. Furthermore, for the Coulomb interaction and a linearly polarized optical excitation (the latter assumption is used here for convenience but it is not necessary), if Z axis is chosen to be parallel to the electric field of the optical pulses, one obtains that $\Phi(\varphi,\varphi',\theta,\chi)$ approaches to 1, and then Eq.(A17b) for hh-c takes the form of Eq.(A9b) for lh-c. This is explained as follows.

First, because the nondiagonal elements of the density matrix $\rho_k \propto \mu_{cv}(\boldsymbol{k})\cdot\boldsymbol{E}(t) \propto \sin\theta$ for hh-c, the linearly polarized optical excitations favour the transitions in which the wave vectors $\boldsymbol{k}$ are perpendicular to the direction of the electric field of the pulses, and then $\cot\theta \ll 1$. Second, since the transition probability for the Coulomb interaction is proportional to $1/(\boldsymbol{k}-\boldsymbol{k}')^4$, it strongly favours small angle scattering, namely, $\sin\chi \ll 1$. This conclusion is also weakly valid for the Fröhlich interaction, in which the transition probability is proportional to $1/(\boldsymbol{k}-\boldsymbol{k}')^2$ (see also Eq.(23)). Consequently, $\Phi(\varphi,\varphi',\theta,\chi) \longrightarrow 1$ (see Eq.(A15)).

For deformation potentials scattering, the large wave vectors are exchanged during the scattering processes. For example, the momenta of AP phonons can be compared to those of the carriers though the energy of AP is so small that is makes the scattering

approximately elastic in nature. Consequently, the "in scattering" marked by the term of $\mu^*_{cv}(\boldsymbol{k}')$ in the right hand side of Eq.(A1) can be neglected on the basis of a random direction consideration: Since $\mu^*_{cv}(\boldsymbol{k}')$ is a vector quantity, different $\boldsymbol{k}'$ contributions in the second term cancel each other, leading to the depolarization rates equal to one half of their total scattering rate. Indeed, even if the weighting factor $(1-\cos\chi)$ is taken into account for the calculation of their dephasing rates, the contribution of "in scattering" to the depolarization also vanish (see also the discussion in Sec. (2.2)). All in all, we especially emphasize in this paper the angle correction for the Coulomb scattering, this is because small angle scattering dominates the scattering processes. For deformation potentials scattering however, the small angle scattering does not play the main role in the scattering processes, and then it is not necessary to take into account the angle correction in the depolarization scattering processes.

We now discuss elliptically including circularly polarized light excitation for hh-c. Since the nondiagonal elements of the density matrix $\rho_k \propto \mu_{cv}(\boldsymbol{k}) \cdot \boldsymbol{E}(t)$, the elliptically polarized light excitations also favour the transitions in which the wave vectors $\boldsymbol{k}$ are perpendicular to the direction of the electric field of the pulses. Besides, the polarized light can be expanded in the linearly polarized basis. Consequently, the conclusions based on the linearly polarized pulse excitation should be still valid for the case of the elliptically polarized light excitation , i.e., the replacement of $(1-\cos\chi)\Phi(\varphi,\varphi',\theta,\chi)$ by $(1-\cos\chi)$ is reasonable for the Coulomb interaction in the hh-c situation.

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