The Role of Nonequilibrium Dynamical Screening in Carrier Thermalization

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We investigate the role played by nonequilibrium dynamical screening in the thermalization of carriers in a simplified two-component two-band model of a semiconductor. The main feature of our approach is the theoretically sound treatment of collisions. We abandon Fermi’s Golden rule in favor of the Schwinger-Bakshi-Mahantappa-Keldysh (nonequilibrium field theoretic) formalism as the former is applicable only in the long-time regime. We also introduce the concept of nonequilibrium dynamical screening. The dephasing of excitonic quantum beats as a result of carrier-carrier scattering is brought out. At low densities it is found that the dephasing times due to carrier-carrier scattering is in picoseconds and not femtoseconds, in agreement with experiments. The polarization dephasing rates are computed as a function of the excited carrier density and it is found that the dephasing rate for carrier-carrier scattering is proportional to the carrier density at ultralow densities. The scaling relation is sublinear at higher densities, which enables a comparison with experiment.

I. INTRODUCTION

The relaxation of hot electron distributions via emission of LO optical phonons has been studied by a number of authors. However, the role of carrier-carrier scattering has received less attention for two main reasons. The theory of screening involved in the proper description of carrier-carrier scattering is poorly understood. Any reasonable attempt to include carrier-carrier scattering is computationally intensive. Secondly, the perception that carrier-carrier scattering is significant only for large carrier densities has resulted in few attempts at accurately modeling the phenomenon. In particular, authors in Refs. 1–4 have conducted theoretical studies of the relaxation of hot electrons when the emission of LO optical phonons is the dominant mechanism. The authors in Ref. 5–7 have attempted to investigate the role of carrier-carrier scattering in momentum and energy relaxation. On the experimental side, authors in Refs. 8–15 have investigated the relaxation of hot carriers in bulk GaAs as well as quantum well structures such as GaAs/AlGaAs. A number of experiments have been performed in the recent years on the density dependence of the scattering rate of free carriers which provide information about the role of carrier-carrier scattering. Prominent among them are the experiments by authors in Refs. 16 and 20. The use of nonequilibrium Green function techniques in the study of relaxation phenomena is not widespread. However, attempts have been made in the recent past. Notable among those are the study of memory effects due to relaxation by phonons by authors in Refs. 5–7 and an attempt by authors in Ref. 30 to model the nonequilibrium aspects of screening of the Coulomb interaction, similar to the approach outlined here.

It has been pointed out in Ref. 6 that changes in the scattering rates with carrier density are solely due to coulomb scattering of carriers. Therefore an investigation of the density dependence of carrier-carrier scattering should provide a good testing ground for theories of coulomb scattering. The authors in Ref. 5–7 have developed a theory of carrier-carrier scattering using the quasi-classical Boltzmann equation coupled with Fermi’s Golden rule for evaluating the collision terms. Objections to the use of Fermi’s Golden rule may be raised, however. Firstly, Fermi’s Golden rule is derived from time-dependent perturbation theory as an asymptotic time approximation which translates into a strict energy conservation rule. Quantitatively, the time resolution of a typical photon echo measurement is of the order of $\Delta t \sim 10\text{fs}$. This translates to an energy uncertainty of an order of $\Delta E \sim 80\text{meV}$.

This is substantial when compared to the plasmon energy at densities of the order of $\rho_0 \sim 2 \times 10^{19}/\text{cm}^3$ or the LO phonon energy which is $\omega_{LO} = 36\text{meV}$. At these densities the plasmon energy is $\omega_p \sim 6.2\text{meV}$. We therefore abandon the familiar rule in favor of a more elaborate explicitly nonequilibrium field-theoretic formulation which we shall discuss subsequently. There is at least one other significant conceptual drawback in the present theories of Coulomb scattering in carrier thermalization. We are primarily interested in the dynamics that occur within 10 ps. of the switching on of the external field. Within this time scale, the system is in a highly nonequilibrium state. Therefore it
does not, in particular, make sense to use the naive generalisation of the equilibrium dynamically screened coulomb interaction which essentially involves replacing in the formula for the RPA dielectric function, the equilibrium carrier densities by nonequilibrium ones. This is because when time translation invariance is broken as is the case here, the RPA (random phase phase approximation) as it is commonly understood, is no longer valid as it is not possible to transform to the frequency domain. Our field-theoretic formulation involves the use of Schwinger-Bakshi-Mahantappa-Keldysh-Kadanoff-Baym approach in deriving the collision terms (henceforth the phrase ‘collision terms’ will refer to the right side of the Boltzmann equation. A good introduction to this approach may be found in Ref. 27) We assume that the collision terms in the Boltzmann equation can be expanded in a power series of the external (time-dependent) vector potential. This approach is similar to linear (or quadratic) response theory. The main difference being that we solve the full Boltzmann equation numerically after the collision terms have been obtained so that the densities and currents no longer depend only on finite powers of the external fields. We also obtain explicit formulas for the screened coulomb interaction in terms of the external fields. These formulas clearly indicate the nonequilibrium nature of the dynamical screening. This screened potential also produces a finite collision cross-section in both two and three space dimensions.

II. SEMICONDUCTOR BLOCH EQUATIONS

We consider a three dimensional, two component, undoped, electron-hole system interacting with a transverse classical electromagnetic field along with a source for the electromagnetic field in addition to the usual coulombic carrier-carrier interaction. It is important to include other terms such as electron-phonon interaction as this is the dominant mechanism of energy relaxation. Impurity scattering is relatively less important for the ultra-pure sample that we consider. Further, it has been argued in the literature that only carrier-carrier scattering contributes significantly to the density dependence of the scattering rates. Intuitively, this is understandable as the electrons can independently emit phonons at a fixed rate irrespective of how many carriers there are to scatter off from. We shall verify these expectations rigorously later on.

We assume that the system is in thermodynamic equilibrium at temperature $T=0$ before time $t=0$ (we use natural units throughout, $\hbar =1$). The fields are assumed to have been turned on at $t=0$ and last for a duration of $\tau_X = 250 fs$. The excitation energy coincides with the band gap $\omega_X = E_g - E_h$, where $E_g$ and $E_h$ are the band gap and exciton binding energy, respectively. Fig.1 is a schematic illustration of the conditions under which the simulations are performed. The semiconductor Bloch equations (SBE for short) (see for example Ref. 33) are nothing but the equations of motion for the equal time component of the conditions under which the simulations are performed. The semiconductor Bloch equations (SBE for short) (see for example Ref. 33) are nothing but the equations of motion for the equal time component of the Green functions of the system. The equations in the form given below were first derived in Refs. 28–31 (Our notation is slightly different though). The Green functions in the equation below are proportional to the inter-band polarization and the electron and hole density distributions.

$$g_{hh}(kt) = i\langle \psi^+_k(kt)\psi_h(kt)\rangle = i\, f(k, t)$$

$$g_{he}(kt) = i\langle \psi_h(kt)\psi_e(kt)\rangle$$

$$i\frac{\partial}{\partial t}g_{hh}(kt) = 2\text{Re}(\Omega(kt)g_{he}(kt)) + R_{hh}(kt)$$

$$i\frac{\partial}{\partial t}g_{he}(kt) = -\Omega(kt)(i - 2g_{hh}(kt)) + (\epsilon_h(k) + \epsilon_e(k) - 2\Sigma(kt))g_{he}(kt) + R_{he}(kt)$$

$$\Omega(kt) = \frac{e}{mc}A_{ext}(t).p_{ve} - i\sum_{k'}v_{k-k'}g_{he}(kt')$$

$$\Sigma(kt) = -i\sum_{k'}v_{k-k'}g_{hh}(kt')$$

(1)
constant relaxation rates: $R$. In our case the Hartree Green function lacks time translation invariance and therefore it is not possible to solve in the shielded potential approximation as discussed in Ref. 27. This is exactly what we do, except that in field. Alternatively, it may be obtained by replacing the full Green function by the Hartree Green function setting the full Green-functions of the system. (Collisionless or Hartree-Fock(HF) approximation therefore means collisionless or Hartree-Fock(HF) approximation).

Relaxation through recombination occurs on a time scale of nanoseconds. Therefore the approximation for $R$ for most purposes unless one is interested exclusively in studying coherent phenomena. The approximation for $R$ in terms of a constant population relaxation rate suggests that the total kinetic energy of the holes decays with the same rate as the total number of holes. We know that this is not true, because when phonons are present the kinetic energy relaxation takes place on a time scale of picoseconds whereas population relaxation through recombination occurs on a time scale of nanoseconds. Therefore the approximation for $R$ in terms of a constant relaxation rate is particularly bad. A more sophisticated approach would involve the use of Fermi’s Golden rule to evaluate the collision terms. Here, the relaxation terms have a complicated dependence on the polarization and densities. However, the rates $R$ and $R$ still depend on the polarization and densities at time $t$ and not on all previous times. Higher in the hierarchy of theoretical sophistication is the nonequilibrium Green function approach. Here, the collision terms depend on the full Green function of the system and therefore effectively, on the polarization and densities at all times previous to time $t$ at which the collision rates are evaluated. Such a dependence of the collision terms on the history of the system is known as the memory effect. Each approach has its pros and cons. While the method of using constant relaxation rates has in its favor, simplicity, it contains phenomenological parameters that have to be fitted from experiment. In fact the phenomenon of the photon echo has been succesfully used to measure the polarization dephasing ($\gamma_{hh}$). The use of Fermi’s Golden rule works well for studying relaxation via phonons but is ambiguous when applied to carrier-carrier scattering (for reasons mentioned in the introduction and for reasons that will become clearer later.). In light of these arguments we take an alternative route. The collision terms in the Boltzmann equation occur as the product of the collision self-energy and the full Green function. The collision self-energy in turn depends on the full Green function. When the external fields are absent the collision terms are identically zero. Therefore, it is sensible to expect a power series expansion for the collision terms in powers of the external fields. In order to simplify matters further, we assume that for the purpose of evaluating these terms, it is sufficient to retain only the coupling of the carriers to the electromagnetic field, as the latter is responsible for generating all the dynamics. Therefore, the zeroth term in this expansion is just the collision self-energy evaluated using the free Green function multiplied by the free Green function which turns out to be identically zero. The next term would involve the first power of the total vector potential multiplied by a linear response term which we evaluate. A more precise quantitative meaning of this approximation will be given in the next section. The screened fock self-energy ( the GW approximation as it is sometimes called) involves the evaluation of the screened coulomb interaction. To this end, we introduce the concept of nonequilibrium dynamical screening.

### A. Nonequilibrium Dynamical Screening

The RPA is obtained by linearizing the Hartree equations of motion for the Green function in the external field. Alternatively, it may be obtained by replacing the full Green function by the Hartree Green function in the shielded potential approximation as discussed in Ref. 27. This is exactly what we do, except that in our case the Hartree Green function lacks time translation invariance and therefore it is not possible to solve the resulting integral equation for the shielded potential by transforming to the frequency domain. This will force us to seek an expansion in terms of a small parameter.

In order to derive the relevant formulas for the screened coulomb interaction, we proceed as follows. The equation of motion for the mean-field (Hartree) Green function of the system, in the presence of an external classical electromagnetic field may be written as:

$$
(i\frac{\partial}{\partial t_1} - H_0(1))G(1,1') = \delta(1-1') + \frac{e}{mc}A(1)\cdot p^T M G(1,1') + \frac{e^2}{2mc^2}|A(1)|^2 G(1,1').
$$

Here and henceforth, we use the abbreviation $1 = (x_{11},t_1)$. This Green function will be used as the starting point for derivation of the formulas for the screened coulomb interaction. Note here that the Hartree-Coulomb terms are absent because of charge neutrality. The Green function in the above equation is defined as...
The derivatives are computed in the usual manner by first inverting Eq. (2) to obtain an equation for the k of the same using the equation of motion for the mean-field Green function. The dimensionless quantity expansion in powers of the dimensionless quantity defined below.

\[ G(1, 1') = \begin{pmatrix} G_{ee}(1, 1') & G^*_e(1, 1') \\ G_{he}(1, 1') & G_{hh}(1, 1') \end{pmatrix}, \]  

(3)

and

\[ M = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \]  

(4)

Here the contour-ordered Green functions are defined as,

\[ G_{hh}(1, 1') = \frac{-i(T_c S \psi_h(1)\psi_h^*(1'))}{T_c S}, \]

\[ G_{ee}(1, 1') = \frac{-i(T_c S \psi_e(1)\psi_e^*(1'))}{T_c S}, \]

\[ G^*_e(1, 1') = \frac{-i(T_c S \psi_e^*(1)\psi_e(1'))}{T_c S}, \]

\[ G_{he}(1, 1') = \frac{-i(T_c S \psi_h(1)\psi_e^*(1'))}{T_c S}. \]  

(5)

Here S is the S-matrix that involves the part of the hamiltonian that generates the external field. Using the same methods outlined in Ref. [27] the screened coulomb interaction \( v_S(1, 3) \) satisfies:

\[ v_S(1, 3) = v(1 - 3) - i(2S + 1) \int d2 \int d4 \ v_S(1, 2) F(2, 4)v(4 - 3), \]

\[ F(2, 4) = G_{he}(4, 2)G^*_e(2, 4^+) + G_{hh}(4, 2)G_{hh}(2, 4^+) + G_{ee}(4^+, 2)G_{ee}(2, 4) + G^*_e(4^+, 2)G_{he}(2, 4). \]  

(6)

We can now expand \( v_S(1, 3) \) in powers of the vector potential by expanding the Green-function in powers of the same using the equation of motion for the mean-field Green function. The dimensionless quantity that appears in this expansion is denoted by \( \Lambda(kt) \). The expansion in powers of the vector potential is an expansion in powers of the dimensionless quantity defined below:

\[ \Lambda(kt) = \int_0^t dt' \frac{e}{mc} A_{ext}(t') \cdot p_{cv} \exp(-ie(kt')), \]

\[ \Lambda_0 = \frac{e}{mc} A_{ext} \cdot p_{cv} \tau_X. \]  

(7)

Here t is some arbitrary time after \( t = 0 \) and \( A_{ext} \) is the amplitude of \( A_{ext}(t) \) and \( \tau_X = 250 fs \) is the pump duration. We require the magnitude of \( \Lambda(kt) \) quantity to be small compared with unity for all values of \( \epsilon(k) = \frac{k^2}{2m} + E_g \) which is the interband transition energy at wavevector \( k \). The expansion in powers of \( \Lambda \) is synonymous with working at low-densities. The leading terms in the expansion for the Green functions can be derived by expanding the Green function in powers of the vector potential.

\[ G(1, 1') = G_0(1, 1') + \int d3 \ (\frac{\delta G(1, 1')}{\delta A(3)})_0 A(3) + \frac{1}{2} \int d3 \int d4 \ (\frac{\delta^2 G(1, 1')}{\delta A(3)\delta A(3)})_0 A(3)A(4). \]  

(8)

The derivatives are computed in the usual manner by first inverting Eq. (2) to obtain an equation for the inverse \( G^{-1}(1, 1') \).

\[ G^{-1}(1, 1') = (i \frac{\partial}{\partial t_1} - H_0(1))\delta(1 - 1') - \frac{e}{mc} A(1) p_{cT} M \delta(1 - 1') - \frac{e^2}{2mc^2} [A(1)]^2 \delta(1 - 1') \]  

(9)
and then by using relations such as,

\[ \delta G(1,1') = - \int d3 \int d4 \, G(1,3)(\delta G^{-1}(3,4))G(4,1') \]  

(10)

Such an approach leads us to expressions for the leading terms in the expansion for the Green functions of the system.

\[ G_{ee}^{\geq}(1,1') = i \sum_k \phi_{ck}(x_1) \phi^*_{ck}(x_1') \exp(i\epsilon_c(k)(t_1 - t_1')) \]

\[ G_{eh}^{\geq}(1,1') = -i \sum_k \phi_{ck}(x_1) \phi^*_{ck}(x_1') \exp(i\epsilon_e(k)(k t_1 - t_1')) \Lambda(k t_1) \Lambda^*(k t_1') \]

\[ G_{eh}^{\leq}(1,1') = - \sum_k \phi_{ck}(x_1) \phi^*_{ck}(x_1') \exp(i\epsilon_e(k)(k t_1)) \Lambda(k t_1) \Lambda^*(k t_1') \]

\[ G_{he}^{\leq}(1,1') = \sum_k \phi_{ck}(x_1) \phi^*_{ck}(x_1') \exp(-i\epsilon_h(k)(k t_1)) \exp(-i\epsilon_e(k)(k t_1')) \Lambda^*(k t_1) \]

\[ G_{hh}^{\leq}(1,1') = i \sum_k \phi_{ck}(x_1) \phi^*_{ck}(x_1') \exp(-i\epsilon_h(k)(k t_1 - t_1')) \Lambda(k t_1') \Lambda^*(k t_1) \]

\[ G_{hh}^{\geq}(1,1') = -i \sum_k \phi_{ck}(x_1) \phi^*_{ck}(x_1') \exp(-i\epsilon_h(k)(k t_1 - t_1')) \]  

(11)

From the above formulas it is obvious that the leading contribution to the collision part of the screened coulomb interaction depends quadratically on the vector potential.

\[ v_S(2,1) = \int d3 \, d4 \, \frac{1}{2} \frac{\delta^2 v_S(1,2)}{\delta A(3) \delta A(4)} A(3) A(4) \]  

(12)

Assuming space translational invariance (in the one conduction band and one valence band model), the (fourier transform of the collision part of the screened coulomb interaction is

\[ v_q^2(t_1, t_2) = -\frac{1}{\Omega} (2S + 1) \sum_k -F_1(k, q; t_1, t_2) \Lambda(k + q; t_1) \Lambda^*(k; t_2) \]

\[ -F_1^+(k, q; t_2, t_1) \Lambda(k; t_1) \Lambda^*(k + q; t_2) \]

\[ +F_h(k, q; t_2 - t_1) \Lambda(k; t_1) \Lambda^*(k; t_2) \]

\[ +F_1^-(k, q; t_2 - t_1) \Lambda(k; t_1) \Lambda^*(k; t_2) \]

\[ + F_1(k, q; t_2 - t_1) \Lambda(k; t_1) \Lambda^*(k; t_2), \]  

(13)
\[ v_\mathbf{q}^< (t_1, t_2) = -(v_\mathbf{q}^> (t_1, t_2))^*, \]

\[ F_1(\mathbf{k}, \mathbf{q}; t_1, t_2) = \exp(i(\epsilon_h(\mathbf{k} + \mathbf{q}) - \epsilon_h(\mathbf{k}))t_2)\exp(i(\epsilon_c(\mathbf{k} + \mathbf{q}) - \epsilon_c(\mathbf{k}))t_1), \]

\[ F_h(\mathbf{k}, \mathbf{q}; t_2 - t_1) = \exp(i(\epsilon_h(\mathbf{k} + \mathbf{q}) - \epsilon_h(\mathbf{k}))(t_2 - t_1)), \]

\[ F_c(\mathbf{k}, \mathbf{q}; t_2 - t_1) = \exp(i(\epsilon_c(\mathbf{k} - \mathbf{q}) - \epsilon_c(\mathbf{k}))(t_2 - t_1)), \]

For the screened Coulomb interaction it is clear that for small \( q \) the screened Coulomb interaction \( v_\mathbf{q}^>(t_1, t_2) \) is proportional to \( \frac{1}{q^2} \) in two dimensions, and \( v_\mathbf{q} = \frac{2\pi e^2}{\epsilon_0 q^2} \) in three dimensions. The spin \( S = 1/2 \) for fermions. \( \epsilon_c(\mathbf{k}) = \frac{k^2}{2m_c} + E_g \) and \( \epsilon_h(\mathbf{k}) = \frac{k^2}{2m_h} \) and the effective masses are \( m_c^* = 0.067 m_e \) and \( m_h^* = 0.5 m_e \) and \( e_0 = 12.4 \).

From the above formulas for the screened Coulomb interaction in the steady state limit becomes clear when one realises that in the steady state limit the Green functions possess time translation invariance and therefore Eq. (6) can be fourier transformed and inverted algebraically to yield an expression for the screened Coulomb interaction. This screened interaction is expressed in terms of the full (steady state) Green function of the system. (Physically, the steady state limit can be realised by applying an infinitely weak field for an infinitely long time.) When the full Green function is replaced with the Hartree Green function, we recover the familiar RPA. If we assume further that the densities involved are small, it is easy to see that the collision part of the screened Coulomb interaction is quadratic in the unscreened potential. This is equivalent to iterating the integral equation (Eq. (6)) once. The steady state limits of Eqs. (13) and (14) are therefore identical to the low density limit of RPA. However, the RPA (i.e., the replacement of the full Green function by the Hartree Green function) is known to be exact only in the ultra high density limit and its low density limit is not likely to capture all the physics. In light of the complexity of the problem we are dealing with here, and all the other approximations that we have made, it is reasonable to proceed further, after pointing out these pitfalls to the reader.

Quasistatic screening is the popular choice in the literature but it is well known that this type of screening grossly overestimates the importance of screening yielding an exponentially decaying Yukawa type of potential in real space. The naive generalisation of the RPA involves replacing in the formula for the equilibrium RPA, equilibrium densities (and polarizations) by nonequilibrium ones. When this is done, it is not clear what the meaning of the frequency that appears in the dielectric function is. Nor is it clear how that screened Coulomb interaction should be coupled with Fermi’s Golden Rule. Our approach is more systematic and perhaps more appealing. The approach by authors in Ref. [36] comes closest to the one just discussed above. The authors however do not solve the SBE including the detailed nonequilibrium screening that they introduce.

B. Collision Terms due to Screening of the Coulomb Interaction

The collision terms are evaluated using the same scheme that was used in the evaluation of the screened Coulomb interaction. \( R_{hx}(\mathbf{k} t) \) is expanded in powers of \( \Lambda(\mathbf{k} t) \). The collision self-energy is \( \Sigma_v(1, 1') = iv_\mathbf{q}(1, 1')G(1, 1') \) Defining
\[
R(1,1') = \begin{pmatrix} R_{ee}(1,1') & R_{eh}(1,1') \\ R_{he}(1,1') & R_{hh}(1,1') \end{pmatrix} = \int d2 \Sigma_c(1,2)G(2,1') - \int d2 G(1,2)\Sigma_c(2,1'),
\]

the relevant component of \( R(1,1') \) namely,
\[
R_{he}(x_1t_1, x_2t_1') = \sum_k \phi_{ek}(x_1)\phi^\ast_{ek}(x_1')R_{he}(kt_1)
\]
yields the necessary collision term. An expansion of \( R(1,1') \) in powers of \( \Lambda \) implies that the collision term \( R_{he}(kt) \) is proportional to \( \Lambda^3 \).
\[
R_{he}(kt) = \frac{1}{V} \sum_q \int_0^t dt_2 \left[ U_1(k,q;t,t_2)\Lambda^\ast(k - q,t_2) - U_2(k,q;t,t_2)\Lambda^\ast(k,t_2) \right],
\]
\[
U_1(k,q;t,t_2) = v_q^\ast(t,t_2)\exp(-i\epsilon_h(k - q) + \epsilon_c(k))t\exp(-i\epsilon_c(k - q) - \epsilon_c(k))t_2
\]
\[
+ v_q^\ast(t_2,t)\exp(-i\epsilon_h(k - q) - \epsilon_h(k))t_2\exp(-i\epsilon_c(k - q) + \epsilon_h(k))t,
\]
\[
U_2(k,q;t,t_2) = v_q^\ast(t,t_2)\exp(i\epsilon_c(k - q) - \epsilon_h(k))t_2\exp(-i\epsilon_h(k - q) + \epsilon_c(k))t
\]
\[
+ v_q^\ast(t_2,t)\exp(-i\epsilon_c(k) - \epsilon_c(k) - \epsilon_c(k))t_2\exp(-i\epsilon_h(k) + \epsilon_c(k))t,
\]
and \( R_{hh}(k,t) \) and \( R_{ee}(k,t) \) are of the order of \( \Lambda^4 \) and are therefore neglected as far as coulomb scattering is concerend.

C. Collision Terms due to Phonons

For treating the electron-phonon collisions we must expand both \( R_{hh}(kt) \), \( R_{ee}(kt) \) and \( R_{he}(kt) \) in powers of \( \Lambda(kt) \). The expansion yields the following for the collision terms.
\[
R_{he}(kt) = -\int_0^t dt_2 \frac{2}{V} \sum_q |M_q|^2 \sin(\omega_{LO}(t_2 - t))\left[ \exp(i\epsilon_c(k - q) - \epsilon_c(k))t_2 \right]
\]
\[
\exp(-i\epsilon_c(k - q) + \epsilon_h(k))t\Lambda^\ast(kt_2) - \exp(i\epsilon_h(k) - \epsilon_h(k))t_2 \exp(-i\epsilon_c(k) + \epsilon_h(k))t_2 \Lambda^\ast(k - q,t_2)
\]
\[
- \exp(-i\epsilon_c(k - q) - \epsilon_h(k))t_2 \Lambda^\ast(k - q,t_2)
\]
\[
- \exp(-i\epsilon_h(k - q) - \epsilon_h(k))t_2 \Lambda^\ast(k - q,t_2)
\]
\[
\exp(-i\epsilon_h(k - q))t\Lambda(k - q,t)
\]
\[
\exp(-i\epsilon_c(k) - \epsilon_c(k))t_2 \Lambda^\ast(k - qt)\Lambda^\ast(kt)
\]

7
and similarly for $R_{hh}(kt)$.

$$R_{hh}(kt) = -\frac{2i}{V} \sum_{q} |M_q|^2 \sin(\omega_{LO}(t - t_2)) \left[ -\exp(i(\epsilon_e(k) - \epsilon_e(k - q)t_2) \right.$$

$$\exp(i(\epsilon_h(k) - \epsilon_h(k - q)t_2)) \Lambda(k t_2) \Lambda^*(k - q t)$$

$$+ \exp(i(\epsilon_h(k - q) - \epsilon_h(k)t_2))$$

$$\exp(i(\epsilon_h(k) - \epsilon_h(k - q)t_2)) \Lambda(k - q t_2) \Lambda^*(k - q t) \right]$$

$+ c.c.$

It can be seen that the leading contribution $R_{he}$ due to phonons is proportional to $\Lambda$ and the next term is proportional to $\Lambda^3$. In the term proportional to $\Lambda$ it appears that we have to use the total vector potential and not the external vector potential. This is because corrections to $\Lambda$ resulting from such a replacement is proportional to $\Lambda^2$ which is larger in magnitude than the next leading term in $R_{he}$ which is proportional to $\Lambda^3$. But in fact, this apparent inconsistency is not so severe, as we have found that for times when the exciting fields are present, the two $\Lambda$’s are within 1% of each other. However, for times when the external fields are absent, it is important as a matter of principle to include this correction. However, we neglect this correction because of a more basic reason, namely when the external fields are off the fluctuations in the vector potential are going to be comparable to the expectation value of the same. Since we are anyway going to treat the fields classically, we are obligated to ignore the difference between the two $\Lambda$’s as well, after the external fields are turned off.

The fact that the electron and hole effective masses are different means that $R_{hh}(kt)$ and $R_{ee}(kt)$ are not exactly equal. This is inconsistent with charge conservation. A remedy is to use the reduced mass instead of the effective masses thereby forcing charge conservation. This approximation will not affect the qualitative results and has the added appeal of obeying a conservation law.

D. Scaling Relation of the Dephasing Rate with Density at Ultra-Low Densities

The theory of the semiconductor photon echo is fairly well developed. The central feature of the theory is that the energy of the echo signal is known to vary exponentially with the time delay $\tau$ between the two pulses involved in the echo. Quantitatively, $E(\tau) \sim \exp(-\tau/T_{echo})$ and the echo time is related to the polarization dephasing rate $\gamma$ through $T_{echo} = \frac{1}{4\gamma}$. The echo time can be measured by measuring the echo energy as a function of the temporal separation of the pulses. The polarization dephasing rate $\gamma$ is a phenomenological constant that accounts for the decay of the polarization due to various relaxation processes. From our analysis it is possible to predict with accuracy the scaling relation of this phenomenological dephasing rate with the total carrier density at ultra-low densities. We have argued that the density dependence of the relaxation rate is solely due to carrier-carrier scattering. Our analysis shows that $R_{hc}(kt)$ is proportional to $\Lambda^3$ for coulomb scattering and proportional to $\Lambda$ for phonon scattering. Further, the total density which is related to $g_{hh}(kt)$ scales as $\Lambda^2$ and $g_{he}(kt)$ is proportional to $\Lambda$ for small $\Lambda$. Therefore the polarization dephasing rate at ultra-low densities is proportional to the density (from carrier-carrier scattering) and also has a added part independent of the density due to phonons. A critical evaluation (and a measure of the smallness of the density) of this conclusion as well as a comparison with the results of other authors will be given at the end.

III. COMPUTATIONAL PROCEDURE

Having obtained the collision terms, we are now ready to solve the semiconductor Bloch equations numerically. We follow closely the approach outlined in Ref. 32. We assume an isotropic distribution and
perform all angular integrations analytically. The singular coulomb terms in the HF part are particularly troublesome and need special care in their evaluation. This is a well-known approach outlined in Ref. 33.

For example, a sum of the type shown below can be rewritten as described:

$$\sum_{k \neq k'} v_{k-k'} f(k') = \sum_{k \neq k'} v_{k-k'}(f(k') - f(k)) + f(k) \sum_{k \neq k'} v_{k-k'}.$$

The last part can be evaluated analytically and in the first part the angular integrations performed analytically. We also assume a cut-off for the wavevector $k_{\text{max}}$. The above sum may then be written as

$$\sum_{k \neq k'} v_{k-k'} f(k') = \frac{(4\pi e^2)}{(2\pi)^2} \int_0^{k_{\text{max}}} dk' \left( \frac{k'}{k} \right) \ln \left( \frac{(k' + k)}{(k' - k)} \right) (f(k') - f(k))$$

$$\quad + \left( \frac{(4\pi e^2)}{(2\pi)^2} f(k) \left( \frac{1}{k} \right) \frac{1}{2} (k_{\text{max}}^2 - k^2) \ln \left( \frac{(k_{\text{max}} + k)}{(k_{\text{max}} - k)} \right) \right) + k k_{\text{max}}.$$

The cutoff $k_{\text{max}}$ is chosen arbitrarily and its validity is justified a posteriori. For example, for the case of excitation at the excitonic absorption edge we choose $k_{\text{max}} = \frac{12.0 \pi}{X}$. This choice is justified on the grounds that the tail of the occupation probability distribution becomes close to zero as the wavevector approaches $k_{\text{max}}$. We choose $A_{\text{ext}}(t) \cdot P_{\text{vec}} = A_{\text{pump}} P_{\text{vec}} \cos(\omega_{\text{pump}} t) \theta(t_X - t)$ and $A_{\text{pump}} = \frac{A_0}{(P_{\text{vec}} \omega_{\text{trans}})}$. Here, $P_{\text{vec}}$ is the inter-band momentum matrix element, $\tau_X = 250 f.s.$ is the pump duration. We can vary the intensity by tuning the dimensionless parameter $A_0$. For the numerical solution of the semiconductor Bloch equations we use the standard Runge-Kutta procedure. The screened coulomb interaction is first numerically evaluated and tabulated in an array before proceeding to the Runge-Kutta algorithm. It is important to use the rotating wave approximation described in detail in the seminal work of R. Binder et al. [23] in order to get sensible results. The collision terms are partly evaluated analytically, tabulated and then used in the computations.

**IV. RESULTS AND DISCUSSIONS**

The main results of our formalism are shown in Figs.2 and 3. The phenomenological dephasing rate $\gamma = 1/T_2$ is plotted as a function of the total excited carrier density. The simulations as depicted in Fig.2a confirm our claim that at ultra-low density, the dephasing rate scales linearly with the density. The scale for the smallness of the density is set by the value of $A_0$. In fact, our simulation shows (Fig.2b) that the total excited carrier density at ultra-low densities is given by $\rho_0 = 7.8A_0^3$ (in units of $10^{13} cm^{-3}$). Therefore, for a choice of $A_0 = 0.1$ which is small compared to unity, we find that the upper limit on the densities that we can work with is $7.8 \times 10^{13} cm^{-3}$. At these low densities, unfortunately, experimental data are hard to come by, because the signal becomes very weak at low densities. It is worthwhile to stress the following point. The fact that we are able to extract the density dependence of the dephasing rate does not mean that the actual relaxation via carrier-carrier scattering is simply an exponential relaxation. The quantity $T_2$ is merely a useful phenomenological parameter that gives us an estimate of the rate of relaxation. It also happens to be directly measurable experimentally, via four-wave mixing. Strictly speaking, we should have simulated the four-wave mixing experiment including the realistic Coulomb scattering, plotted the energy of the echo signal as a function of the time delay between the pulses, and then extracted the dephasing rate from the plot. This is much too difficult to carry out and therefore we have chosen the simpler route outlined here.

It is possible to go beyond these densities by making a slight modification to our formalism. The phenomenological collision term $R_{\text{he}}$ contains terms proportional to the powers of the polarization via the self-energy and the Green function. The polarization in turn decays with a time constant of $T_2$. So, $R_{\text{he}}$ itself should decay at the same rate or faster. In our nonequilibrium formalism however, this does not happen. The decay has to come from the polarization which is proportional to $A$. This $A$ as we have defined it, does not decay with time. However, we know that the polarization does. So we define a new $A$ as follows.

$$A'(kt) = A(kt) \exp(-t/T_2)$$
with a $T_2$ to be determined later. Therefore, we replace all the $\Lambda(k\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!t)$'s in the collision terms by $\Lambda'(k\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!t)$'s. This means that the actual polarization (for small $\Lambda_0$) is proportional not to $\Lambda(k\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!t)$ but to $\Lambda'(k\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!\!t)$. This ensures that the collision terms decay with time. The time constant $T_2$ has to be determined self-consistently. We vary the value of $T_2$ and plot the total density as a function of time(not shown). For time scales that we consider, (less than 1 ps.) no recombination occurs and so we should see a constant total density when the external fields are switched off. For $\Lambda_0$ close to unity and large enough $T_2$, we see that the theory is asymptotically unstable. In other words, the total density increases without saturation. This unphysical result comes about because of a lack of self-consistency in the formalism. In order to derive expressions for the screened coulomb interaction and the collision terms we have ignored the effects of collisions. This inconsistency manifests itself as asymptotic instability. In other words, the theory is unreliable at long time scales. The device of introducing $T_2$ is precisely to partially restore this self-consistency. The value of $T_2$ is tuned from a large value to smaller and smaller values till we see that the total excited carrier density remains constant after the exciting field has been switched off. Any smaller value of the $T_2$ than this critical value will also yield the same result, of course. However, too small a value of $T_2$ will spoil the small time behaviour of our formalism which is modeled exactly. Therefore, the critical value for $T_2$ provides the dephasing rate. The Fig.3a shows the result of the simulations. The experimental data from Ref.14 shows that at densities larger than $10^{15}cm^{-3}$ the polarization dephasing rate scales as $1/\tau \propto \rho_0^{31}$. This sublinearity in the scaling is confirmed by our simulations. The difference between the predicted exponent and the observed is probably attributable to lack of full self-consistency in the formalism and the use of a simple two-band model. The sublinearity originates because of a saturation effect on the dephasing rate brought about by imposing partial self-consistency to the formalism. We have found that the dephasing rate at relatively high densities ($>1 \times 10^{15}cm^{-3}$) is given by

$$\frac{1}{\tau_2} \text{(in ps}^{-1}) = 4.0(\rho_0)^{0.48}$$

where $\rho_0$ is in $10^{15}cm^{-3}$. In addition, there is a additive contribution of 5 ps$^{-1}$ to $1/\tau_2$, independent of the density (for densities in the range $10^{12}$ to $5 \times 10^{15}cm^{-3}$) coming from phonons (not shown in the figs.). Therefore, the carrier-carrier dephasing rate becomes equal to the carrier-phonon dephasing rate at a density of $2 \times 10^{15}cm^{-3}$. These results are also confirmed by experiments in Refs.5 and 21. The conventional wisdom that carrier-carrier scattering is much faster than carrier-phonon scattering is true at densities much higher than those considered here. The dependence of the density on the parameter $\Lambda_0$ is shown in Fig.3b. The dependence is non-parabolic as $\Lambda_0$ approaches and exceeds unity. The dephasing rate for phonons is computed in a manner analogous to the method used for carrier-carrier scattering. The fact that the dephasing rate is independent of the density in the density range considered here suggests that the term that is proportional to $\Lambda^3$ contributes insignificantly. We have confirmed this by plotting various quantities with and without this term and registering only a marginal change. The explanation is, a possible accidental cancellation of the various components of this term. In fig.4 we see that for a pulse $\Lambda_0 = \pi$, in the absence of collisions (shown in dotted lines) the phenomenon of quantum beats which is the oscillation of the hole occupation probability in time after the external field has been turned off ($\tau_X = 250fs$). This is due to the superposition of discrete excitonic frequencies resulting in the beating behaviour. The presence of carrier-carrier scattering (in solid) results in the decay of the quantum beats with a rate given by $1/\tau_2 = 21.9/ps$. The value of $\Lambda_0 = \pi$ is much too large compared to unity and significantly beyond the range of applicability of our formalism, and therefore the value of $T_2$ quoted above should not be taken too literally. The large value of $\Lambda_0$ ensures a large value of $1/\tau_2$ and therefore emphatically illustrates the phenomenon of dephasing.

V. CONCLUSIONS

We have been able to construct an elaborate theory of screening in the presence of external classical time varying electromagnetic fields. We have obtained explicit formulas for the screened coulomb interaction, and demonstrated that they produce finite collision cross-sections in both two as well as three dimensions. We have also explained the range of validity of these formulas. Using the theory we have demonstrated the following: The density dependence of the polarization dephasing rate is due to carrier-carrier scattering alone, in the range of densities considered, and the energy relaxation is due mainly to phonons. Phonons contribute to the dephasing rate as an additive term independent of the density. We have also derived scaling
relations of the polarization dephasing rates with density. We have also provided quantitative estimates of the dephasing rates and demonstrated that they agree qualitatively with experiments. In particular, we have demonstrated that the dephasing times at low densities due to carrier-carrier scattering is in picoseconds and not femtoseconds as is sometimes believed. We have also studied the time evolution of the probability distribution and demonstrated the phenomenon of dephasing of excitonic quantum beats as a result of carrier-carrier scattering. Further improvements should involve the use of more realistic band structures such as the inclusion of valence subbands.

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Fig. 1 Figure demonstrating the conditions under which the simulations are performed.
Fig. 2a Linear scaling of the dephasing rate with density at ultra-low density.
Fig. 2b Parabolic dependence of the excited carrier density on $\Lambda_0$.
Fig. 3a Sublinear scaling of the dephasing rate with density at higher densities.
Fig. 3b Non-parabolic dependence of the excited carrier density on $\Lambda_0$ for higher densities.
Fig. 4 Dephasing of the excitonic quantum beats as a result of carrier-carrier scattering.
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