

Diffusion of photoinjected carriers in plasma in nonequilibrium semiconductors

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We consider effects of diffusion in the photogenerated carrier system in highly photoexcited polar semiconductors. We develop a quantum quasihydrodynamic description of the system based on the nonequilibrium statistical operator formalism. We derive a generalized Fick's diffusion equation for the charge density of the carriers, with the ambipolar diffusion coefficient obtained at the microscopic level and depending on the evolving macroscopic (nonequilibrium thermodynamic) state of the sample. A detailed numerical calculation for the case of GaAs is done, obtaining good agreement with experimental data.

I. INTRODUCTION

Ultrafast responses and functioning under far-from-equilibrium conditions in semiconductor systems pose new and interesting problems in the physics of condensed matter. These systems, as we have emphasized on other occasions, become an interesting testing ground for theoretical ideas in nonequilibrium statistical thermodynamics and many-body physics. Besides the interest in the comprehension of the basic physical principles underlying these important situations, there is a parallel technological interest because of their multiple practical applications.

The study of optical and transport properties is of large relevance in the physics of highly excited semiconductors. Pump-probe experiments have been extensively performed in recent years. In these, electron-hole pairs (carriers) are photogenerated through illumination by laser light; for sufficiently large carrier concentration (typically larger than 10^{16} cm^{-3}) the pairs are on the metallic side of Mott transition and can be regarded as a two-component Landau's Fermi fluid. The carriers and the lattice background form what is termed a highly excited plasma in semiconductors (HEPS). The investigation of the relaxation and transport processes in HEPS is of scientific relevance and technological interest. It is presently acknowledged—as a result of a number of experimental and theoretical studies—that the relaxation processes can be broken down into three stages:¹ In the first stage, the energy and momentum are redistributed among the nonequilibrium carriers owing to carrier-carrier scattering; as a result the electrons and holes attain an internal thermalization characterized by a quasitemperature $T_c^*(t)$, and quasichemical potentials, $\mu_e(t)$ and $\mu_h(t)$. Quasitemperature T_c^* is, in general, much larger than the lattice temperature, but keeps decreasing due to the rapid process in which the carriers lose large part of their excess energy due to optical-phonon emission. In the second stage, T_c^* decreases until optical phonons are no longer efficient at removing the excess energy. In the first and second stages the optical phonons

have populations in excess of equilibrium, mainly on a restricted off-center portion of the Brillouin zone; in this region an overshoot of the quasitemperature of some modes occurs (i.e., the quasitemperature adjudicated to the excited mode is larger than T_c^*),¹⁻³ and finally (in the tenfold picosecond scale) there follows internal thermalization of all optical-phonon modes with the carriers. Finally, in the third stage the cooling of both carriers and optical phonons through emission of acoustic phonons occurs, and heat diffusion from the latter to the thermal reservoir. In the nanosecond time scale, recombination of electron-hole pairs begins to play a role.

In most pump-probe experiments, where the time evolution of $T_c^*(t)$, was determined, a delay in the energy relaxation of the carriers, was observed, in conditions of high carrier concentration, with the presence of a long plateau in T_c^* vs. t , of extension in the hundredfold picosecond range, and values of T_c^* well above the thermal reservoir temperature. The phenomenon was tentatively ascribed to screening by the carrier charge density of the Fröhlich carrier-LO-phonon interaction, or due to the optical-phonon quasitemperature overshoot referred to above. It can be proved that both effects contribute to a delay of the carrier's relaxation, but only on a very short time scale, namely, during the action of the laser pulse and a few picoseconds thereafter. However, there exists experimental evidence of a process that significantly reduces the carrier density in the hundredfold picosecond time scale.⁴ This is the result of diffusion of carriers out of the volume of laser illumination, that occurs with a time of diffusion of, typically, 50 ps: this diffusion process contributes to the observed warmup of the carriers.⁵

Considering then the relevance of carrier diffusion on the time evolution of the nonequilibrium macrostate in HEPS, we study in this paper such transport phenomenon in those systems. It is well known that for systems slightly deviated from thermodynamic equilibrium exact closed expressions for response functions and transport coefficients can be obtained in the form of correlation functions in equilibrium. This is the fluctuation-dissipation theorem,⁶ and calculations can be

performed in a quite practical way using the double-time thermodynamic Green-function algorithm devised by Bogoliubov and Tyablikov and described in a paper by Zubarev.⁷ The actual calculations may be difficult for the case of interacting many-body systems but it is formally closed at this level. But the question we are considering is one of systems strongly departed from equilibrium where one faces a situation with a much higher level of complexity, with response functions and transport coefficients depending on the instantaneous nonequilibrium macroscopic state of the system. In this regime, nonlinearities are present and, in many cases, nonlocal and memory effects may become relevant. There exist several methods used to determine the form of nonlinear transport equations based along different approaches.⁸ A seemingly powerful one is the nonequilibrium statistical operator method (NSOM) consisting of an appropriate extension of Gibbs ensemble algorithm to nonequilibrium situations,⁹ and modeled upon a far-reaching generalization of the Chapman-Enskog approach in kinetic theory.¹⁰

We resort here to NSOM, in Zubarev's approach,¹¹ to deal with the proposed study of carrier diffusion in HEPS. As noted, the carrier system is treated as a Fermi liquid in Landau's quasiparticle approach, with Coulomb interaction dealt with in the random-phase approximation (RPA). We build the quasihydrodynamic NSOM equations of evolution, in a Markovian approximation, that describe the material movement of the carriers. From the resulting generalized Fick's constitutive equations (the so-called Maxwell-Cattaneo equations of extended irreversible thermodynamics^{12,13}) we derive an equation of motion for the carrier density, and, from it, we characterize the diffusion coefficient $D(t)$ in the far-from-equilibrium conditions in HEPS. We evidence the contributions to $D(t)$ coming from carrier-optical-phonon interactions and the interactions of the carriers with the laser and recombination radiation fields, and their relative contributions are discussed. The diffusion coefficient is related to the average kinetic energy and the lifetime of the momentum density of the carriers. The dependence of D on the nonequilibrium macrostate of HEPS is discussed, and its time evolution calculated in the conditions of the experiment of Ref. 4, a result briefly reported elsewhere.¹⁴

II. QUASIHYDRODYNAMICS OF CARRIERS IN HEPS

We consider an intrinsic direct-gap polar semiconductor sample illuminated by a laser pulse of duration t_L , with power flux I_L and photon frequency ω_L . The photo-generated electron-hole pairs are assumed to behave as free carriers, i.e., to be on the metallic side of Mott transition (typically for concentrations higher than 10^{16} cm⁻³). These carriers relax energy to the lattice while their total number varies as it grows due to photon absorption but diminishes in recombination and diffusion processes. We assume a constant laser light intensity throughout the volume of laser light focalization, and stimulated emission and self-absorption are neglected in comparison with spontaneous recombination. The sys-

tem Hamiltonian consists of the carrier energy operator (electrons and holes in band energy states taken in the effective-mass approximation plus Coulomb interaction dealt with in RPA); the Hamiltonian of the phonon field; and the energy operators describing the interaction of carriers with lattice vibrations and with the laser and recombination radiation fields. The carrier-radiation interaction is treated in the dipole approximation with the photon field described by a classical field incorporated in the carrier's Hamiltonian. Optical phonons are described in an Einstein (dispersionless) model. The acoustic phonons are taken as a thermal bath at fixed temperature T_0 . These and the laser source are taken as ideal reservoirs, i.e., they are assumed to remain in stationary unaltered conditions while constantly coupled with the open semiconductor sample.

We have the formation of HEPS in nonequilibrium conditions, which we analyze; as anticipated in the Introduction, we resort to NSOM (Ref. 9) and Zubarev's approach¹¹ in the calculations. We briefly recall that NSOM requires the choice of a basic set of variables to describe the nonequilibrium macroscopic state of the system, say, $Q_j(t)$, $j=1,2,\dots,n$, which are the average values with the nonequilibrium statistical operator (NSO) of a corresponding set of dynamical quantities P_j , the NSO being a functional of these and only these variables. Such a choice is based on the separation of the total Hamiltonian into two parts, namely,

$$H = H_0 + H', \quad (1)$$

where H_0 contains the kinetic energies and the part of the interactions that produce very rapid relaxation processes (the Coulomb interaction in HEPS), and H' is related to the slow relaxation processes. Furthermore, the quantities P_j and the relevant part H_0 of the Hamiltonian are connected by Peletminskii-Zubarev symmetry condition, namely,

$$[P_j, H_0] = \sum_{k=1}^n \Omega_{jk} P_k, \quad (2)$$

in an appropriate quantum representation, and where Ω are c numbers. In this way it may be said that it has been eliminated from the description the fast relaxing variables, and the macrostate of the system is characterized in terms of the contracted description generated by the set of slow relaxing variables.

As described elsewhere⁹ the construction of the NSO, originally done resorting to either heuristic or projection operator techniques approaches, can be put under a unifying approach resorting to a variational principal encompassed in Jaynes's predictive statistical mechanics¹⁵ with memory effects and *ad hoc* hypothesis. In Zubarev's approach the NSO is given by

$$\rho_\varepsilon(t) = \exp \left\{ \varepsilon \int_{-\infty}^t dt' e^{\varepsilon(t'-t)} \ln \bar{\rho}(t', t'-t) \right\}, \quad (3)$$

and $\bar{\rho}$ is an auxiliary NSO given by

$$\bar{\rho}(t_1, t_2) = \exp \left\{ -\phi(t_1) - \sum_{j=1}^n F_j(t_1) P_j(t_2) \right\}, \quad (4)$$

where t_1 stands for the time dependence of the thermodynamic variables ϕ (which ensures normalization of $\bar{\rho}$) and F_j , and t_2 refers to the time evolution of operators P_j in a Heisenberg representation. ε is a positive infinitesimal (which ensures irreversible evolution from the initial state) that goes to zero after the trace operation in the calculations of average values have been performed. The nonequilibrium thermodynamic variables F_j are related to the Lagrange multipliers in the variational method, and are thermodynamically conjugated to the variables Q_j in the sense that

$$Q_j(t) = -\delta\phi(t)/\delta F_j(t), \quad (5)$$

where δ stands for the functional derivative (for the connection of NSOM with phenomenological irreversible thermodynamics, see Refs. 9 and also 13).

Finally, a fundamental step in the theory is the construction of the equation of motion for the basic variables, i.e., those that describe the irreversible evolution of the macroscopic state of the system. They are the average over the nonequilibrium macrostate of Heisenberg equations of motion, i.e.,

$$\frac{d}{dt} Q_j(t) = \text{Tr} \left\{ \frac{1}{i\hbar} [P_j, H] \rho_\varepsilon(t) \right\}. \quad (6)$$

This is an extremely complicated set of coupled equations, in general, which are nonlinear and nonlocal in space and time. However, the separation of the Hamil-

tonian as given by Eq. (1), and the use of the symmetry condition of Eq. (2), allow us to write Eq. (6) in ways that are easier to handle.¹⁰ After very lengthy and elaborate mathematical manipulations the right-hand side of Eq. (6) can be written in terms of an infinite series of collision operators which are instantaneous in time (given as averages over the auxiliary NSO at the time of measurement) and organized in increasing powers of n of the interaction strengths contained in H' , namely,

$$\frac{d}{dt} Q_j(t) = \sum_{n=0}^{\infty} J_j^{(n)}(t) \quad (7)$$

with the construction of quantities $J_j^{(n)}$ described in detail in Ref. 10. The form of the collision operators given by Eq. (7) allows for the introduction of approximations by means of a truncation of the series of partial collision operators in a given order of interaction. The lowest order that introduces relaxation effects ($n \leq 2$) is a truncation in second order in the interaction strengths: It renders the equations Markovian in character and we have called it the second-order approximation in relaxation theory (SOART).⁹ It is usually referred to in the literature as the quasilinear theory of relaxation,¹⁶ a name we avoid because of the misleading term "linear" that refers to a certain approximation in the description of relaxation effects; in SOART the equations of evolution are a set of coupled highly nonlinear integrodifferential equations, namely,

$$\frac{d}{dt} Q_j(t) \simeq J_j^{(0)}(t) + J_j^{(1)}(t) + J_j^{(2)}(t), \quad (8)$$

where

$$J_j^{(0)}(t) = \frac{1}{i\hbar} \text{Tr} \{ [H_o, P_j] \bar{\rho}(t, 0) \}, \quad (9a)$$

$$J_j^{(1)}(t) = \frac{1}{i\hbar} \text{Tr} \{ [H', P_j] \bar{\rho}(t, 0) \}, \quad (9b)$$

$$J_j^{(2)}(t) = \left[\frac{1}{i\hbar} \right]^2 \int_{-\infty}^0 dt' e^{\varepsilon t'} \text{Tr} \{ [H'(t')_o, [H', P_j]] \bar{\rho}(t, 0) \} + \frac{1}{i\hbar} \sum_k \int_{-\infty}^0 dt' e^{\varepsilon t'} \text{Tr} \{ [H'(t')_o, P_k] \bar{\rho}(t, 0) \} \frac{\delta J_j^{(1)}(t)}{\delta Q_k(t)}, \quad (9c)$$

with $H'(t')_o$ meaning that the operator is given in the Heisenberg representation with the Hamiltonian H_o , and $\bar{\rho}$ is given by Eq. (4). We next apply NSOM which is just very briefly reviewed to the study of the HEPS described at the beginning of this section.

In HEPS the system of carriers attains a very rapid internal thermalization (subpicosecond time scale) as a result of Coulomb interaction,¹⁷ and then can be described in terms of a quasitemperature, $kT_c^*(t) = \beta^{-1}(t)$, and quasichemical potentials, $\mu_e(t)$ and $\mu_h(t)$.^{1,18} These β , μ_e , and μ_h 's are F -type thermodynamic variables in the NSOM formalism above; the conjugated Q -type variables are the carrier band energies, and the number (or concentration) of electrons and of holes, the same for both since they are produced in pairs. On the other hand, the optical phonons received part of the excess energy pumped by the laser source on the carrier system and are warmed

up. This occurs in a differentiated way with modes in a restricted region of the Brillouin zone being preferentially excited.³ Hence, the macroscopic description of the optical phonons should be done in terms of the mode occupation function $v_q(t)$.

The carrier concentration and the laser flux power are related through the NSOM-SOART equation¹

$$\begin{aligned} \frac{d}{dt} n(t) &= \alpha_1(\omega_L) (1 - f_L^e - f_L^h) I_L \\ &\quad - \frac{2\pi}{\hbar} \sum_{\mathbf{k}\mathbf{q}} |U^R(\mathbf{k}, \mathbf{q})|^2 f_{\mathbf{k}}^e f_{\mathbf{k}}^h \delta(\epsilon_{\mathbf{k}}^x + E_G - \hbar\omega_q) \\ &\quad - n(t)/\tau_D, \end{aligned} \quad (10)$$

where U^R is the matrix element of the recombination processes; $\epsilon_{\mathbf{k}}^x = \hbar^2 k^2 / 2m_x$, with $m_x^{-1} = m_e^{-1} + m_h^{-1}$; ω_q is

the frequency of the photons produced in recombination processes, i.e., $cq/\epsilon_\infty^{1/2}$, with ϵ_∞ being the high-frequency dielectric constant; $f_{\mathbf{k}}^{e(h)}$ are the Fermi-Dirac distribution functions for electrons (e) and for holes (h); E_G is the energy gap; $f_L^{e(h)}$ are Fermi-Dirac distribution functions for carrier energies ($m_x/m_{e(h)}$) ($\hbar\omega_1 - E_G$); and $\alpha_1(\omega_L)$ is the one-photon absorption coefficient at the laser photon frequency. Evidently, on the right-hand side of Eq. (10), the first term is the one associated with pair production by photon absorption, the second accounts for recombination processes, and the last is a phenomenological term associated with diffusion of pairs out of the volume of laser illumination. Whereas the first two terms arise in the NSOM-SOART calculation, the last one is obtained by solving Fick's diffusion equation, assuming a constant concentration n in the active volume of the sample and null outside; τ_D is a diffusion time given by $\tau_D = \bar{l}^2/D$, where D is the ambipolar diffusion coefficient and \bar{l} an average linear dimension of the active volume; and we take for it the penetration length at the laser frequency. The concentration and quasichemical potentials are related, in the internally thermalized carrier subsystem, by¹⁹

$$n = n_e^0 F_{1/2}(\beta\mu_e) = n_h^0 F_{1/2}(\beta\mu_h), \quad (11)$$

where $F_{1/2}$ is the Fermi function of the index one-half, and

$$n_{e(h)}^0 = 2[2\pi m_{e(h)}/\hbar^2\beta]^{3/2}. \quad (12)$$

The equation of evolution for the carrier energy is given elsewhere;¹ this equation, coupled to Eq. (10) for the carrier density, and Eq. (11), once solved, produce the evolution in time of the quasitemperature and the quasichemical potentials (see, for example, applications in Refs. 1, 5, 18, and 20).

Insofar as we have introduced in the basic set of macrovariables the homogeneous ones for the hydrodynamic-like analysis we want to perform, such a set of variables must be enlarged with local dependent variables whose evolution, imposed over the homogeneous reference state, we are going to analyze. We begin with the carrier density $n(\mathbf{r}, t)$, which we better—for practical reasons—replace by the equivalent Fourier amplitudes $n(\mathbf{Q}, t)$. In a plane-wave representation we have for the corresponding dynamical operators

$$\hat{n}^e(\mathbf{Q}) = \sum_{\mathbf{k}} \hat{n}_{\mathbf{k}\mathbf{Q}}^e = \sum_{\mathbf{k}} C_{\mathbf{k}+\mathbf{Q}}^\dagger C_{\mathbf{k}}, \quad (13a)$$

$$\hat{n}^h(\mathbf{Q}) = \sum_{\mathbf{k}} \hat{n}_{\mathbf{k}\mathbf{Q}}^h = \sum_{\mathbf{k}} h_{-\mathbf{k}} h_{-\mathbf{k}-\mathbf{Q}}^\dagger, \quad (13b)$$

where C (C^\dagger) and h (h^\dagger) are annihilation (creation) operators in the electron (e) and hole (h) states, respectively. Following NSOM [Cf. Eq. (2)] we calculate

$$[\hat{n}^{e(h)}(\mathbf{Q}), H_0] = \hbar \frac{\mathbf{Q}}{m_{e(h)}} \cdot \hat{\mathbf{p}}^{e(h)}(\mathbf{Q}), \quad (14)$$

where

$$\hat{\mathbf{p}}^{e(h)}(\mathbf{Q}) = \sum_{\mathbf{k}} \hbar(\mathbf{k} + \frac{1}{2}\mathbf{Q}) \hat{n}_{\mathbf{k}\mathbf{Q}}^{e(h)} \quad (15)$$

are the Fourier amplitudes for the density of the linear momentum of electrons and of holes, respectively. Hence, both quantities should be incorporated to the basic set of variables, and next we calculate

$$[\hat{\mathbf{p}}^{e(h)}(\mathbf{Q}), H_0] = \hbar \hat{\varphi}^{e(h)}(\mathbf{Q}) \mathbf{Q}, \quad (16)$$

where

$$\hat{\varphi}^{e(h)}(\mathbf{Q}) = \sum_{\mathbf{k}} \frac{\hbar^2}{m_{e(h)}} [(\mathbf{k} + \frac{1}{2}\mathbf{Q}) : (\mathbf{k} + \frac{1}{2}\mathbf{Q})] \hat{n}_{\mathbf{k}\mathbf{Q}} \quad (17)$$

is the flux of linear momentum and the double dots stand for the tensorial product of the vectors. According to NSOM we ought to incorporate the tensor $\hat{\varphi}$ to the basic variables and continue the procedure which introduces tensorial fluxes of higher and higher rank.¹³ Then, a practical solution requires us to introduce a truncation procedure. For that purpose we retain only $n(\mathbf{Q})$ and $\mathbf{p}(\mathbf{Q})$, and, therefore, to close the coupled set of equations of evolution for the averages we need to express the average of quantity $\hat{\varphi}$ (or, equivalently, the average of $n_{\mathbf{k}\mathbf{Q}}$ on which it depends) in terms of the truncated set of basic variables. Of course, this is an approximation for which a price needs to be paid. In terms of the criterion established elsewhere¹³ it will produce inaccurate results in the dependence of energy dispersion relations on the quadratic and higher-order contributions in the wave number Q . This is unimportant in the hydrodynamic regime (limit of small Q) that we introduce in the calculation. It should be noted that a complete hydrodynamic description of the carrier system would require us to consider the density of energy and its flux and higher rank tensorial fluxes associated to them. We omit this part, which is associated to the propagation of thermal perturbation,²¹ and we concentrate only on the propagation of density perturbations. Both groups of variables (material and thermal) are linked by cross-kinetic terms in the equations of evolution; hence, our final expressions simply omit them.

In the conditions stated above, the auxiliary NSO is for the present case

$$\bar{\rho}(t, 0) = \exp \left\{ -\phi(t) - \beta(t) [H_c - \mu_e(t) N_e - \mu_h(t) N_h] + \beta(t) \mathbf{v}(t) \cdot \mathbf{p} - \sum_{\mathbf{q}} F_{\mathbf{q}}(t) \hat{\nu}_{\mathbf{q}} \right. \\ \left. - \sum_{\mathbf{Q}} ' [\beta(t) \mu^e(\mathbf{Q}, t) \hat{n}^e(\mathbf{Q}) + \beta(t) \mu^h(\mathbf{Q}, t) \hat{n}^h(\mathbf{Q})] + \sum_{\mathbf{Q}} ' [\beta(t) \mathbf{v}^e(\mathbf{Q}, t) \cdot \hat{\mathbf{p}}^e(\mathbf{Q}) + \beta(t) \mathbf{v}^h(\mathbf{Q}, t) \cdot \hat{\mathbf{p}}^h(\mathbf{Q})] \right\}, \quad (18)$$

where the upper prime in the sum sign indicates that $Q=0$ (the homogeneous terms) is excluded. In Eq. (18) we have two sets of basic variables, one for the homogeneous variables, namely the carrier Hamiltonian H_c , the number operators for electrons N_e and holes N_h , and the total linear momentum \mathbf{p} . They are accompanied by the intensive nonequili-

brum thermodynamic variables corresponding, respectively, to the reciprocal quasitemperature $\beta(t)$, the quasichemical potentials $\mu_e(t)$ and $\mu_h(t)$, and the drift velocity $\mathbf{v}(t)$. The other set is the one associated to the inhomogeneous variables $n^{e(h)}(\mathbf{Q}, t)$, $\mathbf{p}^{e(h)}(\mathbf{Q}, t)$ and \mathbf{v}_q , accompanied by the nonequilibrium thermodynamically conjugated intensive variables $\mu^{e(h)}(\mathbf{Q}, t)$, $\mathbf{v}^{e(h)}(\mathbf{Q}, t)$, and $F_q(t)$.

Next, we derive the equations of evolution for the basic nonhomogeneous variables in NSOM-SOART [cf. Eqs. (8) and (9)]. The relevant equations for the analysis we want to perform in this paper are those for the local density and local linear momentum of the carriers, namely,

$$\frac{\partial}{\partial t} n^e(\mathbf{Q}, t) = -i \frac{Q}{m_e} \cdot \mathbf{p}^e(\mathbf{Q}, t) + \sum_{\mathbf{k}} B_{\mathbf{kQ}}^{eh}(t) [n_{\mathbf{kQ}}^h(t) + n_{\mathbf{k}, -\mathbf{Q}}^{h*}(t)] - \sum_{\mathbf{k}} B_{\mathbf{kQ}}^{he}(t) [n_{\mathbf{kQ}}^e(t) + n_{\mathbf{k}, -\mathbf{Q}}^{e*}(t)] , \quad (19a)$$

$$\begin{aligned} \frac{\partial}{\partial t} \mathbf{p}^e(\mathbf{Q}, t) = & i \varphi^e(\mathbf{Q}, t) \mathbf{Q} - i Q V(\mathbf{Q}) n_e [n^e(\mathbf{Q}, t) + n^h(\mathbf{Q}, t)] + \sum_{\mathbf{k}} \hbar \mathbf{k} B_{\mathbf{kQ}}^{eh}(t) [n_{\mathbf{kQ}}^h(t) + n_{\mathbf{k}, -\mathbf{Q}}^{h*}(t)] \\ & - \sum_{\mathbf{k}} \hbar \mathbf{k} B_{\mathbf{kQ}}^{he}(t) [n_{\mathbf{kQ}}^e(t) + n_{\mathbf{k}, -\mathbf{Q}}^{e*}(t)] - \sum_{\mathbf{kq}} \hbar \mathbf{q} A_{\mathbf{kq}}^e(t) [n_{\mathbf{kQ}}^e(t) + n_{\mathbf{k}, -\mathbf{Q}}^{e*}(t)] , \end{aligned} \quad (19b)$$

$$\frac{\partial}{\partial t} n^h(\mathbf{Q}, t) = \text{same as Eq. (19a) with exchange } e \leftrightarrow h , \quad (19c)$$

$$\frac{\partial}{\partial t} \mathbf{p}^h(\mathbf{Q}, t) = \text{same as Eq. (19b) with exchange } e \leftrightarrow h . \quad (19d)$$

In Eqs. (19), φ is defined by the average of Eq. (17),

$$\varphi^{e(h)}(\mathbf{Q}, t) = \text{Tr} \{ \hat{\psi}^{e(h)}(\mathbf{Q}) \bar{\rho}(t, 0) \} . \quad (20a)$$

Furthermore,

$$V(\mathbf{Q}) = 4\pi e^2 / V \epsilon_0 Q^2 \quad (20b)$$

is the matrix element of the Coulomb interaction dealt with in the RPA (ϵ_0 is the static dielectric constant),

$$B_{\mathbf{kQ}}^{eh}(t) = A_L \delta(\epsilon_{\mathbf{k}}^x + E_G - \hbar \omega_L) + A_R (\epsilon_{\mathbf{k}}^x + E_G) f_{\mathbf{k}}^e(t) + \text{same term with exchange } \mathbf{k} \rightarrow \mathbf{k} + \mathbf{Q} , \quad (20c)$$

$$B_{\mathbf{kQ}}^{he}(t) = \text{same as Eq. (20c) with exchange } e \leftrightarrow h \quad (20d)$$

with

$$A_R = e^2 E_G / \epsilon_\infty \hbar^2 c^3 m_x , \quad (20e)$$

$$A_L = (2\pi^2 e^2 E_G / \epsilon_\infty \hbar c \omega_L^2 m_x) I_L , \quad (20f)$$

i.e., the B 's are terms associated with the interaction of carriers with the radiation (laser and recombination) fields. Finally,

$$\begin{aligned} A_{\mathbf{kq}\gamma}^{e(h)}(t) = & \frac{1}{\hbar} |V_{\mathbf{q}\gamma}^{e(h)}|^2 \{ [(v_{\mathbf{q}\gamma} + 1) [1 - f_{\mathbf{k}+\mathbf{q}}^{e(h)}(t)] + v_{\mathbf{q}\gamma} f_{\mathbf{k}+\mathbf{q}}^{e(h)}(t)] \delta(\epsilon_{\mathbf{k}+\mathbf{q}}^{e(h)} - \epsilon_{\mathbf{k}}^{e(h)} + \hbar \omega_{\mathbf{q}\gamma}) \\ & + \{ (v_{\mathbf{q}\gamma} + 1) f_{\mathbf{k}+\mathbf{q}}^{e(h)}(t) + v_{\mathbf{q}\gamma} [1 - f_{\mathbf{k}}^{e(h)}(t)] \} \delta(\epsilon_{\mathbf{k}+\mathbf{q}}^{e(h)} - \epsilon_{\mathbf{k}}^{e(h)} - \hbar \omega_{\mathbf{q}\gamma}) \} \end{aligned} \quad (20g)$$

comes from the interaction between carriers and phonons, with $V_q^{e(h)}$ being the matrix element of this interaction, and γ stands for the type of phonons. Furthermore,

$$f_{\mathbf{k}}(t) = \text{Tr} \{ C_{\mathbf{k}}^\dagger C_{\mathbf{k}} \bar{\rho}(t, 0) \} . \quad (21)$$

As already noted we need next to close the system of equations, i.e., to write that the $n_{\mathbf{kQ}}$ that appears in φ and in the terms with the coefficients B 's and A 's, in terms of the basic variables $n(\mathbf{Q}, t)$ and $\mathbf{v}(\mathbf{Q}, t)$. For that purpose we resort to the Heims-Jaynes perturbation expansion for averages²² specified for the case of the auxiliary NSO of Eq. (18); we find in first order (linear approximation) in the local deviations from homogeneity,

$$n_{\mathbf{kQ}}^{e(h)}(t) = a_1^{e(h)}(\mathbf{k}, \mathbf{Q}; t) \beta(t) \mu_{e(h)}(\mathbf{Q}, t) + a_2^{e(h)}(\mathbf{k}, \mathbf{Q}; t) \beta(t) \mathbf{v}^{e(h)}(\mathbf{Q}, t) , \quad (22)$$

where

$$a_1^{e(h)}(\mathbf{k}, \mathbf{Q}; t) = \frac{1 - e^{-\beta(t) \Delta E_{\mathbf{kQ}}^{e(h)}}}{\beta(t) \Delta E_{\mathbf{kQ}}^{e(h)}} f_{\mathbf{k}}^{e(h)}(t) [1 - f_{\mathbf{k}+\mathbf{Q}}^{e(h)}(t)] , \quad (23a)$$

$$a_2^{e(h)}(\mathbf{k}, \mathbf{Q}; t) = \hbar (\mathbf{k} + \frac{1}{2} \mathbf{Q}) a_1^{e(h)}(\mathbf{k}, \mathbf{Q}; t) , \quad (23b)$$

$$\Delta E_{\mathbf{kQ}}^{e(h)} = \epsilon_{\mathbf{k}+\mathbf{Q}}^{e(h)} - \epsilon_{\mathbf{k}}^{e(h)} , \quad (23c)$$

where $f_k^{e(h)}(t)$ are the distribution functions of carriers in state \mathbf{k} in the homogeneous state, namely,

$$f_k^{e(h)}(t) = [1 + \exp\{-\beta(t)[\epsilon_k^{e(h)} - \mu_{e(h)}(t)] + \hbar\beta(t)\mathbf{v}(t) \cdot \mathbf{k}\}]^{-1}, \quad (24)$$

which from now on replaces the population function of Eq. (21) in Eqs. (20c), (20d), and (20g), i.e., we neglect in them the correction due to local inhomogeneities.

With the help of Eq. (22) we can write

$$n^{e(h)}(\mathbf{Q}, t) = a_{11}^{e(h)}(\mathbf{Q}, t)\beta(t)\mu_{e(h)}(\mathbf{Q}, t) + \mathbf{a}_{12}^{e(h)}(\mathbf{Q}, t)\beta(t)\mathbf{v}^{e(h)}(\mathbf{Q}, t), \quad (25a)$$

$$\mathbf{p}^{e(h)}(\mathbf{Q}, t) = \mathbf{a}_{21}^{e(h)}(\mathbf{Q}, t)\beta(t)\mu_{e(h)}(\mathbf{Q}, t) + \mathbf{a}_{22}^{e(h)}(\mathbf{Q}, t)\beta(t)\mathbf{v}^{e(h)}(\mathbf{Q}, t), \quad (25b)$$

where

$$a_{11}^{e(h)}(\mathbf{Q}, t) = \sum_{\mathbf{k}} a_1^{e(h)}(\mathbf{k}, \mathbf{Q}; t), \quad (26a)$$

$$\mathbf{a}_{12}^{e(h)}(\mathbf{Q}, t) = \mathbf{a}_{21}^{e(h)}(\mathbf{Q}, t) = \sum_{\mathbf{k}} \mathbf{a}_2^{e(h)}(\mathbf{k}, \mathbf{Q}; t), \quad (26b)$$

$$\mathbf{a}_{22}^{e(h)}(\mathbf{Q}, t) = \sum_{\mathbf{k}} \hbar^2 [(\mathbf{k} + \frac{1}{2}\mathbf{Q}) : (\mathbf{k} + \frac{1}{2}\mathbf{Q})] a_1^{e(h)}(\mathbf{k}, \mathbf{Q}; t). \quad (26c)$$

Next, Eqs. (25) allow us to express μ in terms of n and \mathbf{v} , to obtain, after replacement in Eq. (22),

$$n_{\mathbf{kQ}}^{e(h)}(t) = g_1^{e(h)}(\mathbf{k}, \mathbf{Q}; t)n^{e(h)}(\mathbf{Q}, t) + \mathbf{g}_2(\mathbf{k}, \mathbf{Q}; t) \cdot \beta(t)\mathbf{v}^{e(h)}(\mathbf{Q}, t), \quad (27)$$

where

$$g_1^{e(h)}(\mathbf{k}, \mathbf{Q}; t) = a_1^{e(h)}(\mathbf{k}, \mathbf{Q}; t) / a_{11}^{e(h)}(\mathbf{Q}; t), \quad (28a)$$

$$\mathbf{g}_2^{e(h)}(\mathbf{k}, \mathbf{Q}; t) = \mathbf{a}_2^{e(h)}(\mathbf{k}, \mathbf{Q}; t) = \frac{a_1^{e(h)}(\mathbf{k}, \mathbf{Q}; t)}{a_{11}^{e(h)}(\mathbf{Q}; t)} \mathbf{a}_{12}^{e(h)}(\mathbf{Q}, t). \quad (28b)$$

The result of Eq. (27) can now be replaced in Eqs. (19) to obtain the generalized hydrodynamic equations

$$\frac{\partial}{\partial t} n^e(\mathbf{Q}, t) = i \frac{\mathbf{Q}}{m_e} \cdot \mathbf{p}^e(\mathbf{Q}, t) + \gamma_1^{eh}(\mathbf{Q}, t)n^h(\mathbf{Q}, t) + \gamma_2^{e(h)}(\mathbf{Q}, t) \cdot \beta(t)\mathbf{v}^h(\mathbf{Q}, t) - \gamma_1^{he}(\mathbf{Q}, t)n^e(\mathbf{Q}, t) + \gamma_2^{he}(\mathbf{Q}, t)\beta(t)\mathbf{v}^e(\mathbf{Q}, t), \quad (29a)$$

$$\begin{aligned} \frac{\partial}{\partial t} \mathbf{p}^e(\mathbf{Q}, t) &= \underline{A}^e(\mathbf{Q}, t)i\mathbf{Q}n^e(\mathbf{Q}, t) + \underline{B}^e(\mathbf{Q}, t)i\mathbf{Q}[\mathbf{Q} \cdot \mathbf{v}^e(\mathbf{Q}, t)] \\ &+ \underline{C}^e(\mathbf{Q}, t)[i\mathbf{Q} \times \beta(t)\mathbf{v}^e(\mathbf{Q}, t)] - i\mathbf{Q}N_e V(\mathbf{Q})[n^e(\mathbf{Q}, t) + n^h(\mathbf{Q}, t)] \\ &+ \Gamma_1^{eh}(\mathbf{Q}, t)n^h(\mathbf{Q}, t) + \Gamma_2^{eh}(\mathbf{Q}, t)\beta(t)\mathbf{v}^h(\mathbf{Q}, t) - \Gamma_1^{he}(\mathbf{Q}, t)n^e(\mathbf{Q}, t) - \Gamma_2^{he}(\mathbf{Q}, t)\beta(t)\mathbf{v}^e(\mathbf{Q}, t) \\ &+ \frac{1}{2}\hbar\mathbf{Q}\lambda_1^{he}(\mathbf{Q}, t)n^e(\mathbf{Q}, t) + \frac{1}{2}\hbar\mathbf{Q}\lambda_2^{he}(\mathbf{Q}, t) \cdot \beta(t)\mathbf{v}^e(\mathbf{Q}, t) \\ &- \frac{1}{2}\hbar\mathbf{Q}\lambda_1^{eh}(\mathbf{Q}, t)n^h(\mathbf{Q}, t) - \frac{1}{2}\hbar\mathbf{Q}\lambda_2^{eh}(\mathbf{Q}, t)\beta(t)\mathbf{v}^h(\mathbf{Q}, t) - \Lambda_1^e(\mathbf{Q}, t)n^e(\mathbf{Q}, t) - \Lambda_2^e(\mathbf{Q}, t)\beta(t)\mathbf{v}^e(\mathbf{Q}, t), \end{aligned} \quad (29b)$$

$$\frac{\partial}{\partial t} n^h(\mathbf{Q}, t) = \text{same as Eq. (27a) with exchange } e \leftrightarrow h, \quad (29c)$$

$$\frac{\partial}{\partial t} \mathbf{p}^h(\mathbf{Q}, t) = \text{same as Eq. (27b) with exchange } e \leftrightarrow h, \quad (29d)$$

where

$$\gamma_1^{e(h)}(\mathbf{Q}, t) = \sum_{\mathbf{k}} B_{\mathbf{kQ}}^{eh}(t)[g_1^h(\mathbf{k}, \mathbf{Q}, t) + g_1^h(\mathbf{k}, -\mathbf{Q}, t)], \quad (30a)$$

$$\gamma_2^{eh}(\mathbf{Q}, t) = \sum_{\mathbf{k}} B_{\mathbf{kQ}}^{eh}(t)[\mathbf{g}_2^h(\mathbf{k}, \mathbf{Q}, t) + \mathbf{g}_2^h(\mathbf{k}, -\mathbf{Q}, t)], \quad (30b)$$

$$\gamma_1^{he}(\mathbf{Q}, t) = \text{same as Eq. (30a) with exchange } e \leftrightarrow h, \quad (30c)$$

$$\gamma_1^{he}(\mathbf{Q}, t) = \text{same as Eq. (30b) with exchange } e \leftrightarrow h, \quad (30d)$$

$$\underline{A}^e(\mathbf{Q}, t) = \frac{\hbar^2}{m_e} \sum_{\mathbf{k}} [(\mathbf{k} + \frac{1}{2}\mathbf{Q}) : (\mathbf{k} + \frac{1}{2}\mathbf{Q})] g_1^e(\mathbf{k}, \mathbf{Q}, t), \quad (30e)$$

$$\underline{B}^e(\mathbf{Q}, t) = \frac{\hbar^2}{m_e} \sum_{\mathbf{k}} \frac{1}{Q^2} (\mathbf{k} \cdot \mathbf{Q} + \frac{1}{2}Q^2) [(\mathbf{k} + \frac{1}{2}\mathbf{Q}) : \mathbf{g}_2^e(\mathbf{k}, \mathbf{Q}, t)], \quad (30f)$$

$$\underline{\Gamma}^e(\mathbf{Q}, t) = \frac{\hbar^2}{m_e} \sum_{\mathbf{k}} \frac{1}{Q} (\mathbf{k} \cdot \mathbf{Q} + \frac{1}{2} Q^2) [(\mathbf{k} + \frac{1}{2} \mathbf{Q}) : \mathbf{g}_2^e(\mathbf{k}, \mathbf{Q}, t)], \quad (30g)$$

$$\underline{\Gamma}_1^{eh}(\mathbf{Q}, t) = \sum_{\mathbf{k}} \hbar \mathbf{k} B_{\mathbf{k}\mathbf{Q}}^{eh} [g_1^h(\mathbf{k}, \mathbf{Q}, t) + g_1^h(\mathbf{k}, -\mathbf{Q}, t)], \quad (30h)$$

$$\underline{\Gamma}_2^{eh}(\mathbf{Q}, t) = \sum_{\mathbf{k}} B_{\mathbf{k}\mathbf{Q}}^{eh} \hbar \mathbf{k} : [g_2^h(\mathbf{k}, \mathbf{Q}, t) + g_2^h(\mathbf{k}, -\mathbf{Q}, t)], \quad (30i)$$

$$\underline{\Gamma}_1^{he}(\mathbf{Q}, t) = \text{same as Eq. (30h) with exchange } e \leftrightarrow h, \quad (30j)$$

$$\underline{\Gamma}_2^{he}(\mathbf{Q}, t) = \text{same as Eq. (30i) with exchange } e \leftrightarrow h, \quad (30k)$$

$$\lambda_1^{he}(\mathbf{Q}, t) = \sum_{\mathbf{k}} B_{\mathbf{k}\mathbf{Q}}^{eh} [g_1^h(\mathbf{k}, \mathbf{Q}, t) - g_1^h(\mathbf{k}, -\mathbf{Q}, t)], \quad (30l)$$

$$\lambda_2^{he}(\mathbf{Q}, t) = \sum_{\mathbf{k}} B_{\mathbf{k}\mathbf{Q}}^{eh} [g_2^h(\mathbf{k}, \mathbf{Q}, t) - g_2^h(\mathbf{k}, -\mathbf{Q}, t)], \quad (30m)$$

$$\lambda_1^{eh}(\mathbf{Q}, t) = \text{same as Eq. (30l) with exchange } e \leftrightarrow h, \quad (30n)$$

$$\lambda_2^{eh}(\mathbf{Q}, t) = \text{same as Eq. (30m) with exchange } e \leftrightarrow h, \quad (30o)$$

$$\Lambda_1^e(\mathbf{Q}, t) = \sum_{\mathbf{k}\mathbf{q}} \hbar \mathbf{q} A_{\mathbf{k}\mathbf{q}}^e(t) [g_1^e(\mathbf{k}, \mathbf{Q}, t) + g_1^e(\mathbf{k}, -\mathbf{Q}, t)], \quad (30p)$$

$$\Lambda_2^e(\mathbf{Q}, t) = \sum_{\mathbf{k}\mathbf{q}} A_{\mathbf{k}\mathbf{q}}^e(t) \hbar \mathbf{q} : [g_2^e(\mathbf{k}, \mathbf{Q}, t) + g_2^e(\mathbf{k}, -\mathbf{Q}, t)], \quad (30q)$$

$$\Lambda_1^h(\mathbf{Q}, t) = \text{same as Eq. (30o) with exchange } e \leftrightarrow h, \quad (30r)$$

$$\Lambda_2^h(\mathbf{Q}, t) = \text{same as Eq. (30q) with exchange } e \leftrightarrow h. \quad (30s)$$

Next, we introduce a simplifying assumption which consists of neglecting the dependence on \mathbf{Q} of the kinetic coefficients of Eqs. (30); this means resorting to a local theory or, in other words, neglecting nonlocal effects. In fact, e.g., the term $\underline{A}^e(\mathbf{Q}, t) i \mathbf{Q} n^e(\mathbf{Q}, t)$ becomes

$$\int d^3 r' \underline{A}^e(\mathbf{r} - \mathbf{r}'; t) \nabla' n^e(\mathbf{r}, t), \quad (31a)$$

when going over to the direct space. The local approximation implies

$$\underline{A}^e(\mathbf{r} - \mathbf{r}', t) = \underline{A}^e(\mathbf{Q} = 0, t) \delta(\mathbf{r} - \mathbf{r}'), \quad (31b)$$

and then we have in direct space the simple term

$$A^e(t) \nabla n^e(\mathbf{r}, t). \quad (31c)$$

In this local approximation, and, furthermore, neglecting in the population of Eq. (24) the drift term, i.e., we take the limit of low drift velocities [or, more specifically, when it is verified the inequality $m_{e(h)} v_{e(h)}^2 / 2 \ll \beta^{-1}(t)$], and because the resulting isotropy tensors become scalars and several coefficients cancel out because they involve odd powers of \mathbf{k} in sums over this variable, one has that in Eqs. (29), $\underline{B}^{e(h)}$, $\underline{C}^{e(h)}$, Γ^{eh} , Γ_1^{he} , λ_1^{he} , λ_2^{he} , λ_1^{eh} , λ_2^{eh} , and $\lambda_1^{e(h)}$ are null. Finally, we obtain

$$\frac{\partial}{\partial t} n^e(\mathbf{Q}, t) = i \frac{\mathbf{Q}}{m_e} \cdot \mathbf{p}^e(\mathbf{Q}, t) + \gamma_1^{eh}(t) n^h(\mathbf{Q}, t) - \gamma_1^{he}(t) n^e(\mathbf{Q}, t), \quad (32a)$$

$$\begin{aligned} \frac{\partial}{\partial t} \mathbf{p}^e(\mathbf{Q}, t) = & A^e(t) i \mathbf{Q} n^e(\mathbf{Q}, t) - i \mathbf{Q} N_e(t) V(\mathbf{Q}) [n^e(\mathbf{Q}, t) + n^h(\mathbf{Q}, t)] \\ & + \Gamma_2^{eh}(t) \beta(t) \mathbf{v}^h(\mathbf{Q}, t) - \Gamma_2^{he}(t) \beta(t) \mathbf{v}^e(\mathbf{Q}, t) - \Lambda_2^e(t) \beta(t) \mathbf{v}^e(\mathbf{Q}, t), \end{aligned} \quad (32b)$$

$$\frac{\partial}{\partial t} n^h(\mathbf{Q}, t) = \text{same as Eq. (32a) with exchange } e \leftrightarrow h, \quad (32c)$$

$$\frac{\partial}{\partial t} \mathbf{p}^h(\mathbf{Q}, t) = \text{same as Eq. (32b) with exchange } e \leftrightarrow h. \quad (32d)$$

Because of the presence of the local drift velocities $v^{e(h)}$ in Eqs. (32), to close this system of equations we take into account that, from Eq. (25b), with $\mathbf{a}_{21} = 0$ in the limit of small \mathbf{Q} it follows that

$$\mathbf{p}^{e(h)}(\mathbf{Q}, t) = a_{22}^{e(h)}(t) \beta(t) \mathbf{v}^{e(h)}(\mathbf{Q}, t), \quad (33)$$

where

$$a_{22}^{e(h)}(t) = \frac{1}{3} \sum_{\mathbf{k}} \hbar^2 k^2 f_{k(t)}^{e(h)} [1 - f_{k(t)}^{e(h)}]; \quad (34)$$

hence, we are left with a system of four coupled equations of evolution, i.e., those for n^e , n^h , \mathbf{p}^e , \mathbf{p}^h .

We consider next the diffusive movement of the carriers, which is assumed to be a uniform movement, i.e., without

acceleration due to the fact that, after a transient time, the drift force resulting from the gradient of concentration is compensated by the viscous force; this implies that in taking the near steady-state condition, $\partial \mathbf{p}^{e(h)}(\mathbf{Q}, t)/\partial t \simeq 0$. Hence, Eqs. (32b) and (32d), after multiplication by $i\mathbf{Q}$, becomes

$$-A^e(t)Q^2 n^e(\mathbf{Q}, t) + m_{e(h)} \omega_{pe}^2 [n^e(\mathbf{Q}, t) + n^h(\mathbf{Q}, t)] + \Gamma^{eh}(t) i\mathbf{Q} \cdot \mathbf{p}^h(\mathbf{Q}, t) - \Gamma^{he}(t) i\mathbf{Q} \cdot \mathbf{p}^e(\mathbf{Q}, t) - \Lambda^e(t) i\mathbf{Q} \cdot \mathbf{p}^e(\mathbf{Q}, t) = 0, \quad (35a)$$

same as Eq. (35a) with exchange $e \leftrightarrow h$,

$$(35b)$$

where we wrote $\omega_{pe(h)}^2(t) = 4\pi e^2 n(t)/m_{e(h)}$,

$$\begin{aligned} \Gamma^{eh} &= \Gamma_2^{eh}/a_{22}^h, \\ \Gamma^{h(e)} &= \Gamma_2^{he}/a_{22}^e, \\ \Lambda^{e(h)} &= \Lambda_2^{e(h)}/a_{22}^{e(h)}, \end{aligned} \quad (36)$$

and $A^{e(h)}$ is given by Eq. (30e), which for $Q=0$ becomes a scalar because of isotropy.

Rearranging Eqs. (35) we find that

$$\begin{aligned} \frac{i\mathbf{Q}}{m_e} \cdot \mathbf{p}^e(\mathbf{Q}, t) &= -D_e(t) Q^2 n^e(\mathbf{Q}, t) \\ &+ \alpha^{eh}(t) [n^e(\mathbf{Q}, t) + n^h(\mathbf{Q}, t)] \\ &+ \bar{\Gamma}^{eh}(t) i\mathbf{Q} \cdot \mathbf{p}^h(\mathbf{Q}, t). \end{aligned} \quad (37a)$$

same as Eq. (37a) with exchange $e \leftrightarrow h$,

$$(37b)$$

where

$$D_e(t) = A^e(t)/[\Gamma^{he}(t) + \Lambda^e(t)] m_e, \quad (38a)$$

$$D_h(t) = A^h(t)/[\Gamma^{eh}(t) + \Lambda^h(t)] m_h, \quad (38b)$$

$$\alpha^{eh}(t) = \omega_{pe}^2(t)/[\Gamma^{he}(t) + \Lambda^e(t)], \quad (38c)$$

$$\alpha^{he}(t) = \omega_{ph}^2(t)/[\Gamma^{eh}(t) + \Lambda^h(t)], \quad (38d)$$

$$\bar{\Gamma}^{eh}(t) = \Gamma^{eh}(t)/[\Gamma^{he}(t) + \Lambda^e(t)] m_e, \quad (38e)$$

$$\bar{\Gamma}^{he}(t) = \Gamma^{he}(t)/[\Gamma^{eh}(t) + \Lambda^h(t)] m_h. \quad (38f)$$

Neglecting the coupling of both linear momenta, after replacing Eqs. (37) into Eqs. (32a) and (32c), we find in direct space

$$\begin{aligned} \left[\frac{\partial}{\partial t} - D_e(t) \nabla^2 \right] n^e(\mathbf{r}, t) &= \alpha^{eh}(t) n(\mathbf{r}, t) - \gamma^{he}(t) n^e(\mathbf{r}, t) \\ &+ \gamma^{eh}(t) n^h(\mathbf{r}, t), \end{aligned} \quad (39a)$$

$$\begin{aligned} \left[\frac{\partial}{\partial t} - D_h(t) \nabla^2 \right] n^h(\mathbf{r}, t) &= \alpha^{he}(t) n(\mathbf{r}, t) - \gamma^{eh}(t) n^h(\mathbf{r}, t) \\ &+ \gamma^{he}(t) n^e(\mathbf{r}, t), \end{aligned} \quad (39b)$$

where α^{eh} and α^{he} are given by Eqs. (38c) and (38d), and γ^{eh} are those of Eqs. (30a) and (30c) in the limit of $Q=0$, and

$$n(\mathbf{r}, t) = n^e(\mathbf{r}, t) + n^h(\mathbf{r}, t). \quad (40)$$

We can see that Eqs. (39) are of the type of equations of diffusion for electrons and for holes (D_e and D_h playing the role of diffusion coefficients) with driving forces arising from Coulomb interaction and radiation fields, which couple both equations. Both types of carriers do not diffuse independently, and because of the local charge balance (neutrality of charge) in the movement we have $n^e(\mathbf{r}, t) = n^h(\mathbf{r}, t) = n(\mathbf{r}, t)/2$. To describe the ambipolar diffusion, i.e., the diffusive movement of the total charge $n(\mathbf{r}, t)$, we multiply Eq. (37a) by D_h , Eq. (37b) by D_e , and adding up both we find

$$\left[\frac{\partial}{\partial t} - D(t) \nabla^2 \right] n(\mathbf{r}, t) = \frac{1}{2} D(t) \left[\frac{\omega_{pe}^2(t)}{A^e(t)} + \frac{\omega_{ph}^2(t)}{A^h(t)} \right] n(\mathbf{r}, t) + \frac{D_e(t) - D_h(t)}{D_e(t) + D_h(t)} [\gamma^{he}(t) - \gamma^{eh}(t)] n(\mathbf{r}, t), \quad (41)$$

which is a diffusion-type equation with driving forces (Coulomb and radiation fields), where we have introduced the ambipolar diffusion coefficient

$$D^{-1}(t) = \frac{1}{2} [D_e^{-1}(t) + D_h^{-1}(t)], \quad (42)$$

defined in terms of the individual diffusion coefficients of electrons and of holes. If we write

$$\begin{aligned} 3A^{e(h)}/m_{e(h)} &= c_{e(h)}^2, \\ [\Gamma^{he} + \Lambda^e]^{-1} &= \tau_e, \\ [\Gamma^{eh} + \Lambda^h]^{-1} &= \tau_h, \end{aligned} \quad (43)$$

where c has the dimension of velocity and τ of time, we have

$$D_{e(h)}(t) = \frac{1}{3} c_{e(h)}^2(t) \tau_{e(h)}(t), \quad (44)$$

defining diffusion coefficients with a form typically of classical kinetic theory,²³ given at time t in terms of the macroscopic state of the system. In the nondegenerate limit [when $f_k(t)$ becomes an instantaneous Maxwell distribution] we find for the velocity c the thermal velocity, and $\tau_{e(h)}$ is clearly [cf. Eqs. (32b) and (32d) together with Eq. (33)] the relaxation time of each momentum, composed of two contributions [cf. Eqs. (38a), and (38b)], one arising from collisions with phonons (part Λ) and another as a result of the interaction with the radiation fields (part Γ). Therefore, we have an expression for the ambipolar diffusion coefficient at the microscopic level (i.e., in terms of the dynamics generated by the interactions present in

the Hamiltonian) and given at each instant of time in terms of the nonequilibrium macroscopic state that is evolving under the action of the pumping and dissipative effects that develop in HEPS. We next proceed to perform analytical and numerical calculations.

III. ALGEBRAIC AND NUMERICAL RESULTS FOR GaAs

We are now in condition to obtain expressions and to evaluate computationally the diffusion coefficients $D_{e(h)}(t)$. First, we restrict the calculations to the nondegenerate limit, taking for $f_k(t)$ the instantaneous Maxwell distributions characterized by $\beta(t)$ and $n(t)$. In that case, as already noted, $c_{e(h)}$ are the thermal velocities of electrons (holes), and we need then to evaluate the momentum relaxation times $\tau_{e(h)}(t)$ of Eqs. (44) and (43). We notice that they are composed of two contributions, one coming from the interaction with the radiation fields, and the other arising from collisions with phonons. We next consider the situation after the laser pulse has been switched off, and the subsequent delay times in the hundredfold picosecond time scale. In this time interval, contribution from recombination to the relaxation times is negligible when compared with the one arising from interaction with the phonons. The latter has contributions from, in polar semiconductors. Fröhlich [involving longitudinal optical (LO) phonons] and deformation-potential interactions (involving phonons in all branches). The Fröhlich interaction predominates over the others and we will only consider it in the calculations.

It should be noted that the calculation of τ is coupled to the solution of the time evolution of the homogeneous part of the macrostate of the system, i.e., $\tau(t)$ depends on $\beta(t)$, $n(t)$, and $v_{qLO}(t)$. The dependence on the latter makes it not possible to obtain algebraic expressions, and also makes the computational calculations very difficult. To overcome this difficulty we introduce an approximation, namely, while keeping in the calculations of β and n the explicit dependence on $v_{q\gamma}$, in the expression for τ we take the LO phonons as having a unique reciprocal quasitemperature $\beta_{LO}(t) = 1/kT_{LO}^*(t)$. The latter is obtained from the expression for the energy of the LO phonons given by

$$E_{LO}(t) = \sum_q \hbar\omega_0 [\exp\{\beta_{LO}(t)\hbar\omega_0\} - 1]^{-1} \quad (45)$$

where we use an Einstein model for the LO phonons, ω_0 being the dispersionless frequency.

In such conditions, and for the nondegenerate carrier system we find that

$$\begin{aligned} \tau_{e(h)}^{-1}(t) = & \frac{1}{\tau_{e(h)}^0} x_{(t)}^{3/2} e^{-x(t)/2} \\ & \times \left\{ \left[1 + 2v_{LO}(t) + \frac{v_{LO}(t)}{\bar{v}(t)} \right] K_1 \left[\frac{x(t)}{2} \right] \right. \\ & \left. + \left[1 - \frac{v_{LO}(t)}{\bar{v}(t)} \right] K_0 \left[\frac{x(t)}{2} \right] \right\}, \quad (46) \end{aligned}$$

$$1/\tau_{e(h)}^0 = \frac{2}{3} e E_{e(h)} (2\pi m_{e(h)} \hbar\omega_0)^{-1/2}, \quad (47a)$$

$$x(t) = \beta(t) \hbar\omega_0, \quad (47b)$$

$$v_{LO}(t) = [e^{\beta_{LO}(t)\hbar\omega_0} - 1]^{-1}, \quad (47c)$$

$$\bar{v}(t) = [e^{x(t)} - 1]^{-1}, \quad (47d)$$

where $E_{e(h)}$ is the Fröhlich coupling strength²⁴ and K_n are modified Bessel functions of the second kind.

We proceed next to present a complete calculation that describes the time evolution of the macroscopic state of HEPS and the ambipolar diffusion coefficient, in the conditions of the experiment of Ref. 4. To obtain the evolution of the quasitemperature and quasicchemical potentials of carriers we solve the coupled system of equations of evolution for the energy,¹ relating it to $\beta_c(t)$, $\mu_e(t)$, and $\mu_h(t)$ through the equation

$$E(t) = \sum_k [\epsilon_k^e f_k^e(t) + \epsilon_k^h f_k^h(t)], \quad (48)$$

and for the density $n(t)$, Eq. (10), together with Eqs. (11). The equations of evolution for the LO-phonon populations are

$$\frac{d}{dt} v_{qLO}(t) = -\tau_{qLO}^{-1}(t) [v_{qLO}(t) - \bar{v}_{qLO}(t)], \quad (49)$$

where

$$\begin{aligned} \tau_{qLO}^{-1}(t) = & x^{1/2} [1 - e^{-x}] \\ & \times \sum_{\alpha} \frac{\Omega_{\alpha}^{LO}}{y_{\alpha}^3} \exp\{-\frac{1}{4}x(y_{\alpha} - y_{\alpha}^{-1})^2\}, \quad (50a) \end{aligned}$$

$$x(t) = \beta(t) \hbar\omega_0, \quad (50b)$$

$$y_{\alpha} = \Lambda_{\alpha} q, \quad (50c)$$

$$\Lambda_{\alpha} = (2m_{\alpha}\omega_0/\hbar)^{-1/2}, \quad (50d)$$

$$\Omega_{\alpha} = n(t)\pi^{3/2}\hbar\Lambda_{\alpha}\gamma_{\alpha}/m_{\alpha}, \quad (50e)$$

$$\begin{aligned} \gamma_{\alpha} = & (e^2/\hbar)(\epsilon_{\infty}^{-1} - \epsilon_0^{-1})(m_{\alpha}/2\hbar\omega_0)^{1/2} \\ = & eE_{\alpha}/\hbar\omega_0(2m_{\alpha}\omega_0/\hbar)^{1/2}, \quad (50f) \end{aligned}$$

and

$$\bar{v}_{qLO}(t) = [e^{x(t)} - 1]^{-1}. \quad (50g)$$

In Eqs. (50), γ_{α} is Fröhlich coupling constant, \bar{v} has the form of the Planckian population function of phonons LO at the quasitemperature of the carriers, and the carriers have been considered as a nondegenerate gas. As already noticed the form for the equation of evolution of the populations of the LO phonons leads to the production of LO phonons in excess of equilibrium in a privileged off-center region of reciprocal space, and phonon quasitemperature overshoot,^{1,2} and results in a slowing down of the carrier relaxation.¹ We recall that Eq. (49) has been used in the calculation of the evolution of the macroscopic thermodynamic state of the carriers, Eq. (46) was obtained under the assumption of taking the LO-phonon populations in the form of Eq. (47c), with the LO-phonon quasitemperature obtained from Eq. (45).

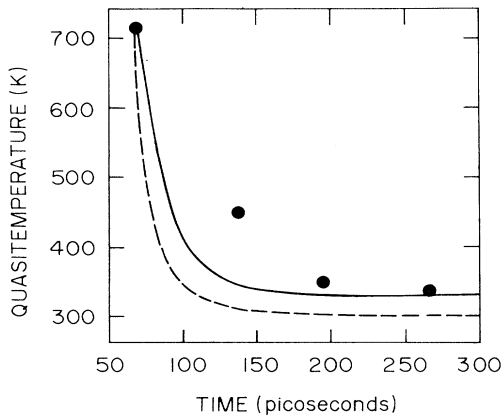


FIG. 1. Evolution of the carrier quasitemperature; dots are experimental points from Ref. 4. Full line is the case when diffusion, in the way described in the text, is taken into account, and the dashed line corresponds to a calculation neglecting diffusion effects.

Furthermore, we also recall that in Eq. (10)

$$\frac{1}{\tau_D(t)} = \frac{D(t)}{\bar{l}^2} = \frac{1}{\bar{l}^2} \frac{2D_e(t)D_h(t)}{D_e(t)+D_h(t)}, \quad (51)$$

where $D_{e(h)}$ are related to $\tau_{e(h)}$ through Eq. (44), and we take for the average diffusion length \bar{l} the value 7×10^{-5} cm, i.e., of the order of the penetration depth. We take as initial values those corresponding to the first experimental point in Ref. 4.

In Fig. 1 we show the evolution of the carrier quasitemperature, where a good agreement can be observed with the experimental data. The dashed line shows the

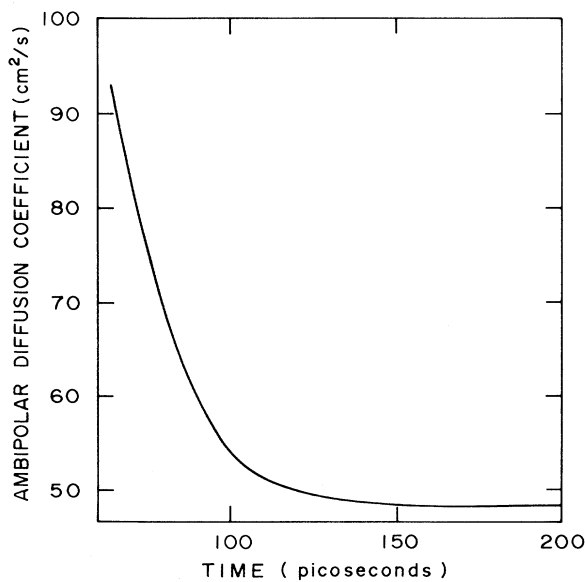


FIG. 2. Evolution of the ambipolar diffusion coefficient accompanying that of the macroscopic state of the sample.

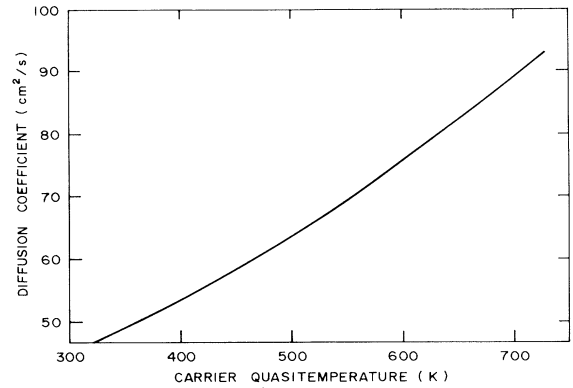


FIG. 3. Dependence of the ambipolar diffusion coefficient with the instantaneous carrier quasitemperature.

evolution of the quasitemperature in the absence of diffusion effects, which makes explicit the role of these in the slowing down of the carrier relaxation.

Figure 2 displays the curve corresponding to the calculation of the evolution of the ambipolar diffusion coefficient, while in Fig. 3 it is shown the dependence of D with the instantaneous carrier quasitemperature. On approaching the equilibrium temperature of 300 K the value of D approaches that calculated by Young and van Driel,²⁵ as well as measured experimental values.²⁶

It can be observed that D increases with temperature in a nonlinear way, and to be noticed that in the nondegenerate regime in which we performed the calculations, the individual diffusion coefficients of Eq. (44) take the form of an Einstein law,²⁷ namely,

$$D_{e(h)}(t) = kT_c^*(t)\tau_{e(h)}(t)/m_{e(h)}. \quad (52)$$

Figure 4 shows the kinetic equation for the carrier density in reasonably good agreement with experiment.

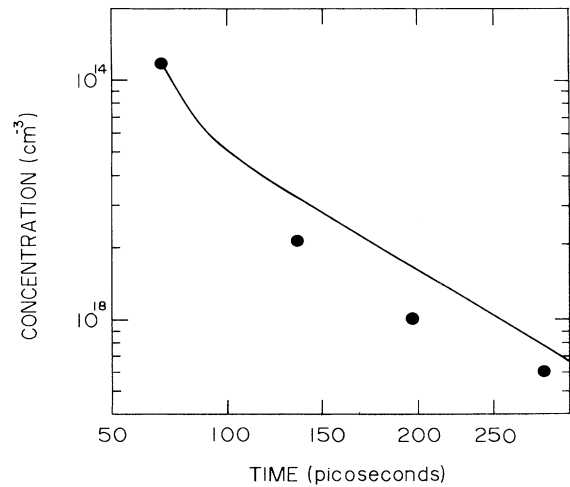


FIG. 4. Kinetics of the carrier concentration governed by the ambipolar diffusion coefficient of Fig. 2; dots are experimental points from Ref. 4.

IV. CONCLUDING REMARKS

We have presented a study of diffusion effects in the carrier system in HEPS. Usually this is done in terms of phenomenological quasihydrodynamic approaches in near-equilibrium conditions. But HEPS evolve under, in many cases, far-from-equilibrium conditions. For that reason we have resorted first to a microscopic description—in the sense that we introduce a dynamic description governed by quantum-mechanical equations of movement determined by the knowledge of the system Hamiltonian—and, second, for the description of the instantaneous macroscopic (nonequilibrium thermodynamic) state of HEPS and its evolution to a powerful mechanostatistical formalism, namely the NSOM (Ref. 9) in Zubarev's approach.¹¹ However, it should be stressed that we did not use the whole power of the method^{9,10} but restricted ourselves to linear calculations in the space deviation [cf. Eq. (22)], local, and Markovian approximations, i.e., we obtained a space-independent and instantaneous diffusion coefficient in a generalized diffusion equation. The expression for the diffusion coefficient of each type of carriers resembles the form of the one given in classical kinetic theory [cf. Eq. (44)], i.e., one-third the square of a mean velocity (in fact, proportional to the mean kinetic energy) times the linear momentum relaxation time, all quantities given in terms of the nonequilibrium macroscopic state of HEPS at each time t . This relaxation time depends on the interaction of the carriers with phonons and photons. Usually, in polar semiconductors the effect of the Fröhlich interaction predominates; this $\tau_{e(h)}$ depends on the population of the nonequilibrium phonon modes, but in the unique quasitemperature approximation we used for them in the calculation of τ an algebraic expression can be derived as given by Eq. (46). In this case the dependence of $\tau_{e(h)}$ on the carrier quasitemperature can be obtained, with numerical results for the case of GaAs displayed in Fig. 3 and dis-

cussed in the main text.

The ambipolar diffusion coefficient is related in a simple form with the individual diffusion coefficients for both types of carriers [cf. Eq. (42)], an expression, on the one hand, in coincidence with the one in the phenomenological descriptions,²⁸ and, furthermore, reminiscent of Mathiessen's rule.²⁹ The calculations performed in the conditions of the experiment of Ref. 4 allow us to show the explicit time evolution of the ambipolar diffusion coefficient, and with it the kinetics of the carrier density, with results that are in good agreement with experiment (cf. Fig. 4).

Hence, we have been able to offer a study of the diffusion of carriers in HEPS. The question of including nonlocal and memory effects, as well as the comparison of the diffusion coefficient with the mobility coefficient, will be reported elsewhere;³⁰ we anticipate that the Einstein relation between these two transport coefficients does not hold, except, approximately, in the limit of very weak drift velocity. Young and van Driel²⁶ have shown that at high levels of photoinjection many-body effects affect the ambipolar diffusion coefficient producing a diminution of its values as compared with those obtained in independent-carrier transport theories. Such effects were not considered in our formulation and, thus, it would be desirable to enlarge the study of ambipolar diffusion in HEPS bringing together both treatments, i.e., including many-body effects (the band-gap gradient in the shrinking effect that the presence of the carrier density produces) in the context of the nonequilibrium statistical thermodynamic approach we used in this paper.

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