

Nonlinear transport in photoexcited plasma in semiconductors: Non-Ohmic mobility and a generalized Einstein relation

A. R. Vasconcellos, A. C. S. Algarte, and R. Luzzi

Instituto de Física "Gleb Wataghin," Universidade Estadual de Campinas, Unicamp, 13083-970 Campinas, São Paulo, Brazil

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Resorting to a theory of responses to thermal and mechanical perturbations, based on statistical irreversible thermodynamics for systems arbitrarily away from equilibrium, we obtain the diffusion and mobility coefficients in a highly photoexcited plasma in semiconductors in the presence of an electric field. They are dependent on the evolution of the nonequilibrium thermodynamic state of the system. From these transport coefficients we derived a generalized Einstein relation for ultrafast transient regimes and for non-Ohmic conditions. In all cases this generalized Einstein law acquires values that are field dependent and larger than those in its original form only valid in steady-state conditions and the limit of weak fields. Numerical results appropriate for the case of a GaAs sample are presented.

I. INTRODUCTION

In a previous paper,¹ hereafter referred to as I, we considered the question of ambipolar diffusion in highly photoexcited plasma in semiconductors, in the context of a quasihydrodynamic description in terms of informational statistical thermodynamics based on the nonequilibrium statistical operator method.² We derived 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 was performed, obtaining good agreement with experimental data. In I we commented that, even in a linear transport regime, the Einstein relation does not hold in the ultrafast transient regime, but becomes valid in the linear transport regime (very weak drift velocity) once the system attains a steady state well after the transient time has elapsed. Here we return to this question to consider the transient nonlinear regime.

We recall that the photoinjected plasma in semiconductors consists of the presence of a double fluid of charged particles (electrons and holes produced in pairs by strong photoexcitation at densities high enough to be on the metallic side of the Mott transition) moving in the lattice background. The carriers interact with the latter via carrier-phonon interaction. Study of the electrical conductivity has been extensively performed in the case of the usual gaseous plasmas, which of course have different physical characteristics than the photoinjected plasma in semiconductors.³ On the other hand, electron transport in solids has also received much attention for a long time: kinetic theories were applied, as described in the classical textbooks by Conwell and Ziman,⁴ as well as Kubo's formula for the current-current correlation function and quantum Boltzmann-like equations.⁵ In general, these approaches did not allow us to tackle situations where the system is far away from equilibrium satisfactorily, as is the case of the highly excited plasma in semi-

conductors, and when submitted to high-field intensities. More recently Monte Carlo-like methods have also been introduced⁶ with relative success. As already noted, here we consider the question of diffusion and conductivity in the highly excited plasma in semiconductors in far-from-equilibrium conditions produced by high levels of photoexcitation and under the action of an electric field, and therefore in the presence of highly nonlinear dissipative processes. For such a purpose we resort to a seemingly powerful, concise, and practical formalism, which is encompassed in a broad generalization of Gibbs theory to arbitrary nonequilibrium conditions, namely the nonequilibrium statistical operator method² already referred to, and its accompanying nonlinear, nonlocal, memory-dependent nonequilibrium quantum transport theory,⁷ a far-reaching generalization of Boltzmann's and Mori's approaches.^{2,8} This method provides appropriate microscopic foundations to irreversible thermodynamics,⁹ and, consequently, to a generalized hydrodynamics which provides the diffusion equation for the photoinjected carrier system in the presence of the electric field, the generalized nonlinear Ohm's law, and, finally, the Einstein relation for nonlinear charge transport, as described below.

The Einstein relation, sometimes also called the Nernst-Townsend-Einstein relation¹⁰ (NTER for short from now on), establishes that the ratio between the diffusion coefficient and the mobility multiplied by the absolute temperature is a universal constant (k_B/e). We stress in I that since the plasma is in far-from-equilibrium conditions, the absolute temperature is replaced by the carrier quasitemperature. (For the question of the concepts of temperature and entropy in systems in conditions far away from equilibrium, see Ref. 11.) It has been noted that NTER fails for large fields in the case of gaseous plasma,¹² and we will stress this fact here in relation to the highly excited plasma in semiconductors. The case of a gas of rigid spheres was considered by Garcia Colin and Uribe,¹³ within the framework of extended irreversible thermodynamics,¹⁴ which in a sense is a method also used by us here within its informational statistical approach in the context of the nonequilibrium sta-

tistical operator method.⁹

The paper is organized as follows: In Sec. II we briefly review the main results of I, and add the calculation of the mobility. Of the contributions to it from the different carrier-phonon scattering mechanisms, namely the deformation potential, piezoelectric interaction and Fröhlich interaction, the latter is the relevant one to be considered. In Sec. III we discuss the definition of a generalized NTER in the linear (in the transient and stationary conditions) and nonlinear (non-Ohmic systems) transport regimes. Numerical calculations for the particular case of a GaAs sample are presented, though restricted to field strengths below those corresponding to enter into the runaway-in-mobility regime. In Sec. IV we summarize and comment upon the results.

II. DIFFUSION AND MOBILITY IN HEPS

Let us consider the photoinjected highly excited plasma in semiconductors (HEPS) described in I: an intrinsic direct-gap polar semiconductor illuminated by a laser pulse of duration t_L , power flux I_L , and photon frequency ω_L . The process produces photogenerated electron-hole pairs (carriers) that are assumed to behave as free carriers, i.e., to be on the metallic side of Mott's transition. These carriers relax their excess energy to the lattice, while their total number (concentration) varies as it grows due to (laser) photon absorption but diminishes in recombination and diffusion processes. The dynamics of relaxation of HEPS is reviewed in Ref. 15. We simply recall that the macroscopic nonequilibrium thermodynamic state of the carrier system is described in terms of a quasitemperature $k_B T_c^*(t) = \beta^{-1}(t)$, and quasichemical potentials $\mu_e(t)$ and $\mu_h(t)$ for electrons and holes, respectively. We stress that this contracted description of the carrier system, in terms of only these few nonequilibrium thermodynamic parameters, is possible because of the very rapid (subpicosecond time scale) internal thermalization (in nonequilibrium conditions) that follows as a consequence of the ultrafast redistribution in energy-momentum space of the excess energy pumped by the external sources (the laser pulse and the electric field). This is because the carrier fluid is dense—in units of the exciton radius the intercarrier spacing r_s is smaller than $2(r_s < 2)$ —and therefore is in the so-called metallic regime.¹⁶ This condition is verified theoretically,^{17,18} and the quasitemperature and quasichemical potentials are experimentally characterized and measured.^{15,18,19} The nonequilibrium macrostate of the phonons is described by quasitemperatures for the different branches, $k_B T_\gamma^*(t) = \beta_\gamma^{-1}(t)$. This is appropriate for the case of the acoustic phonons, but the optical phonons require, for short delay times after application of the laser pulse, a description in terms of populations per mode,^{20,21} and thus is verified the phenomenon of quasitemperature overshoot;²¹ hence we are introducing an approximation acceptable for delay times that are not too short. The carrier Hamiltonian is taken to be composed of electron and hole bands in the effective-mass approximation, and the Coulomb interaction is dealt with in the random-phase approximation. The acoustic phonons are taken in the

Debye approximation, and the Einstein (dispersionless) model is used in the case of the optical phonons. The electron-phonon interaction is composed of the deformation potential and piezoelectric interactions in the case of A phonons, the deformation potential in the case of TO phonons, and the deformation potential and Fröhlich interaction in the case of LO phonons. Next we consider the phenomena of carrier diffusion and mobility.

A. Diffusion coefficient

As described in I, to proceed with a description of the nonequilibrium thermodynamic state of the photoinjected plasma in semiconductors, we resort to the nonequilibrium statistical operator method, introduced as basic variables to characterize the carrier state, the energy and concentration of which are homogeneous variables, and inhomogeneous variables consisting of the local density and momentum density, or better, their Fourier amplitudes $n^{e(h)}(\mathbf{Q}, t)$ and $\mathbf{p}^{e(h)}(\mathbf{Q}, t)$, where $\mathbf{Q} \neq 0$ ($\mathbf{Q} = 0$ is the homogeneous state). For the phonon system we take the energies of the different branches. (This implies, as noted above, the internal thermalization of the carrier and phonon systems, and, in the case of the former, we have added local variations of density and momentum.) The homogeneous variables are related in the nonequilibrium statistical operator method to a set of intensive variables, namely T_c^* , μ_e , and μ_h , as already described.¹⁵

The equations of evolution for the local density and momentum density were derived in I using, (1) the second-order approximation in relaxation theory⁷ (the instantaneous-in-time approximation and the second order in the interactions), (2) linearization in the inhomogeneous variables (weak deviations from the homogeneous state), and (3) a local approximation (space correlations are neglected). We obtained Eqs. (32a) and (32b) in I, complemented by Eq. (33). Furthermore, assuming a near-mechanical equilibrium of driving and viscous forces (that is, taking $\partial p^{e(h)}/\partial t \simeq 0$), we arrived at the equations of diffusion [with sources arising from random-phase approximation (RPA) polarization effects and the presence of the laser and recombination electromagnetic fields] given by Eqs. (39a) and (39b) in I, for electrons and holes, respectively. In those equations what is of relevance to us here is the expression for the diffusion coefficients, namely [cf. Eq. (44) in I]

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

which has a form closely resembling that of the kinetic theory,²² where c has dimensions of velocity and τ has dimensions of time, both defined in Eqs. (43) in I. Equation (1) provides the diffusion coefficients for electrons and holes, given at time t , while dissipative processes develop in the nonequilibrium system. Moreover, since the particles are charged, once the neutrality of the charge is imposed (brought about by the Coulomb interaction) it is possible, as shown in I, to define the ambipolar diffusion coefficient $D(t)$ given by the expression

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

Consider now the presence of an electric current generated by an electric field.

B. Mobility coefficient

Let the system be in the presence of a homogeneous and constant electric field of intensity \mathcal{E} . The interaction between the carriers and field adds to the Hamiltonian the energy interaction

$$H_{\mathcal{E}} = -e\mathcal{E} \sum_j [x_j^{(e)} - x_j^{(h)}], \quad (3)$$

where x_j is the coordinate (in the direction of the field) of the j th carrier. As basic variables in the framework of the nonequilibrium statistical operator method, we consider the carrier's energy, linear momentum (in the direction of the field), and density, and the energies of the phonons. The thermodynamically conjugated variables (Lagrange multipliers that the method introduces) are the carrier's reciprocal temperature $\beta(t)$, also $-\beta(t)\mu_e(t)$ and $-\beta(t)\mu_h(t)$, involving the quasichemical potentials, as in Sec. II A, and, further, $-\beta(t)v_e(t)$ and $-\beta(t)v_h(t)$ where $v_{e(h)}$ play the role of drift velocities (in the direction of the electric field), and finally the reciprocal temperatures of the phonons in the different branches. Moreover, we

take a constant temperature (that of the thermal bath) for the phonons, since its increase has been proved to be small,²³ and we neglect the change in time of the carrier concentration, which can be done when one considers delay times below the nanosecond characteristic time for recombination. It has been proven that transient transport occurs on the picosecond scale, and so the stationary state is covered in that way.²³ Consequently we only need to carry the equations of evolution for the energy and linear momentum. In the second-order approximation in relaxation theory,⁷ we obtain that

$$\frac{d}{dt} E(t) = -(e/m_e)\mathcal{E}P_e(t) + (e/m_h)\mathcal{E}P_h(t) - J_E^{(2)}(t), \quad (4a)$$

$$\frac{d}{dt} n(t) \simeq 0, \quad (4b)$$

$$\frac{d}{dt} P_{\alpha}(t) = ne_{\alpha}\mathcal{E} - J_{\alpha}^{(2)}(t), \quad (4c)$$

where $\alpha = e$ or h , $e_e = -e$, $e_h = e$, E and P are the energy and momentum per unit volume, n is the carrier density (we recall that it is taken to be constant), and $J_E^{(2)}$ and $J_{\alpha}^{(2)}$ are the terms of relaxation due to interaction with the phonon field; they are given by

$$J_E^{(2)}(t) = \frac{2\pi}{\hbar} \sum_{k\alpha q\gamma, i} |V_{q\gamma}^{\alpha, i}|^2 \hbar\omega_{q\gamma} \{v_{q\gamma} f_k^{\alpha}(t) [1 - f_{k+q}^{\alpha}(t)] - [1 + v_{q\gamma}] f_{k+q}^{\alpha}(t) [1 - f_k^{\alpha}(t)]\} \delta(\epsilon_{k+q}^{\alpha} - \epsilon_k^{\alpha} - \hbar\omega_{q\gamma}), \quad (5a)$$

$$J_{\alpha}^{(2)}(t) = \frac{2\pi}{\hbar} \sum_{kq\gamma, i} |V_{q\gamma}^{\alpha, i}|^2 \hbar q_{\parallel} \{v_{q\gamma} f_k^{\alpha}(t) [1 - f_{k+q}^{\alpha}(t)] - [1 + v_{q\gamma}] f_{k+q}^{\alpha}(t) [1 - f_k^{\alpha}(t)]\} \delta(\epsilon_{k+q}^{\alpha} - \epsilon_k^{\alpha} - \hbar\omega_{q\gamma}) + \{[1 + v_{q\gamma}] f_k^{\alpha}(t) [1 - f_{k-q}^{\alpha}(t)] - v_{q\gamma} f_{k-q}^{\alpha}(t) [1 - f_k^{\alpha}(t)]\} \delta(\epsilon_{k-q}^{\alpha} - \epsilon_k^{\alpha} + \hbar\omega_{q\gamma}), \quad (5b)$$

where q_{\parallel} is the phonon wave vector in the direction of the electric field, and index i , in matrix elements V , refers to the different possible types of carrier-phonon interactions. Moreover, calculation of the corresponding average values lead to the results that

$$P_{\alpha}(t) = nm_{\alpha} v_{\alpha}(t), \quad (6)$$

and that f and v are carrier and phonon distribution functions given by

$$f_k^{\alpha}(t) = [\exp\{\beta(t)[\epsilon_k^{\alpha} - \mu_{\alpha}(t)] - \beta(t)\hbar\mathbf{k} \cdot \mathbf{v}_{\alpha}(t)\} + 1]^{-1} \quad (7)$$

and

$$v_{q\gamma} = [\exp\{\beta_{\gamma}\hbar\omega_{q\gamma}\} - 1]^{-1}. \quad (8)$$

We stress that the calculated expression for the carrier distribution function takes the form of an instantaneous time-shifted Fermi-Dirac distribution function: as already discussed at the beginning of this section, this is a

result of the ultrafast internal thermalization of the carrier system in this dense plasma in semiconductors. Moreover, let us consider the system under conditions such that it follows a *nondegeneratelike regime*, when Eq. (7) becomes

$$f_k^{\alpha}(t) = \left[\frac{4\pi^3 \hbar^3 n}{(2\pi m_{\alpha})^{3/2}} \right] \beta^{3/2}(t) \times \exp \left\{ \frac{-\beta(t)[\hbar\mathbf{k} - m_{\alpha}\mathbf{v}_{\alpha}(t)]^2}{2m_{\alpha}} \right\}. \quad (9)$$

Equation (9) has the shape of an instantaneous shifted-Maxwell-Boltzmann distribution, which is a very good approximation of the distribution of Eq. (7) in most general situations, except for very large carrier concentrations—typically of the order of 10^{19} cm^{-3} and up, namely near optical saturation—and low quasitemperature. This is not the case since we are considering a highly excited system. Moreover, for LO phonons we

write

$$v_0 = [\exp\{\beta_0 \hbar \omega_0\} - 1]^{-1} \quad (10)$$

where ω_0 is the LO-phonon dispersionless frequency (Einstein model), and β_0 is the LO-phonon reciprocal quasitemperature. Using Eqs. (9) and (10) in Eqs. (5) we can obtain analytic expressions for the relaxation terms which depend on the electric-field intensity. It should be noted that the relaxation terms of energy and momentum [cf. Eqs. (5)] are a simple superposition of the scattering mechanism due to different types of carrier-phonon interactions, in other words a manifestation of Mathiessen's rule.⁴ For simplicity, in the case of these polar semiconductors we concentrate our attention on Fröhlich interaction only, since it is the predominant one. One point needs to be stressed here: In classical plasma the so-called Debye-Onsager-Falkenhagen effect is present, which is a consequence of the modification of the collision integral as a result of screening effects by the mobile charges, which is dependent on the field strength, and leads to intracollisional field effects and a collisionally broadening effect.²⁴ In the present case of a highly excited plasma in semiconductors, it is accounted for in the screening of the Fröhlich interaction through the presence of a dielectric function calculated in the random-phase approximation (Lindhart dielectric function).⁴ However, under the conditions that lead to the use of Eq. (9), the RPA screening factor reduces to the Debye-Hückel expression, with a squared screening wave number given by $q_{\text{DH}}^2 = 8\pi n e^2 / k_B T_c^*(\mathcal{E})$, where the dependence on the electric-field strength is contained in the quasitemperature. However, under strong excitation conditions (high quasitemperature, higher and higher with increasing field strength) and not too large densities, as is the situation under consideration, the screening effect is very weak and shall be neglected.

In the conditions thus stated, we find that

$$J_E^{(2)}(t)|_{\alpha, \text{LO}}^F = n \exp\{-x_\alpha(t)\} \sum_{l=0}^{\infty} x_\alpha^l(t) \Psi_{\alpha l}(z(t)), \quad (11a)$$

$$\begin{aligned} J_\alpha^{(2)}(t)|_{\alpha, \text{LO}}^F &= n x_\alpha^{1/2}(t) \exp\{-x_\alpha(t)\} \\ &\quad \times \sum_{l=1}^{\infty} x_\alpha^{l-1}(t) \Phi_{\alpha l}(z(t)) \\ &= x_\alpha^{1/2}(t) \varphi_\alpha(x_\alpha(t), z(t)), \end{aligned} \quad (11b)$$

where we have defined the quantities

$$x_\alpha(t) = \beta(t) \frac{1}{2} m v_\alpha^2(t), \quad (12a)$$

$$z(t) = \beta(t) \hbar \omega_0, \quad (12b)$$

and the different quantities that appear in Eqs. (11) are listed in the Appendix.

We now look for the mobility coefficient. First we note that the density current \mathcal{J} (in the direction of the field) is

$$\begin{aligned} \mathcal{J}(t) &= -en v_e(t) + ev_h(t) \\ &= - \left[\frac{e}{m_e} \right] P_e(t) + \left[\frac{e}{m_h} \right] P_h(t), \end{aligned} \quad (13)$$

and that we can write Eq. (4c) as

$$\frac{d}{dt} P_\alpha(t) = ne_\alpha \mathcal{E} - \frac{P_\alpha(t)}{\tau_{p\alpha}(t)}, \quad (14)$$

where

$$\tau_{p\alpha}^{-1}(t) = \left[\frac{\beta(t)}{2m_\alpha} \right]^{1/2} \varphi_\alpha(x_\alpha(t), z(t)), \quad (15)$$

with φ defined in Eq. (11b). The initial condition for Eq. (14) is $P_\alpha(0) = 0$. Equation (14) is a highly nonlinear differential equation since, we recall, the relaxation time of Eq. (15) is a highly nonlinear functional of x , and then of P (or v) and, therefore, of \mathcal{E} . But Eq. (14), with the given initial condition, can be transformed in an equivalent highly nonlinear integral equation, namely,

$$P_\alpha(t) = ne \mathcal{E} \tau_{c\alpha}(t), \quad (16)$$

with $\tau_{c\alpha}$ being a characteristic time for conduction given by

$$\tau_{c\alpha}(t) = \frac{\int_0^t dt' \exp\left\{ \int_0^{t'} dt'' \tau_{p\alpha}^{-1}(t'') \right\}}{\exp\left\{ \int_0^t dt' \tau_{p\alpha}^{-1}(t') \right\}}, \quad (17)$$

where, we stress, the momentum relaxation time $\tau_{p\alpha}(t)$ depends on the instantaneous nonequilibrium macroscopic state of the system, i.e., on $\beta(t)$, $\mu_0(t)$, and also on $v_\alpha(t)$. Introducing Eq. (16) into Eq. (13), we find for each type of carrier that

$$\mathcal{J}_\alpha(t) = \sigma_\alpha(t) \mathcal{E}, \quad (18)$$

with

$$\sigma_\alpha(t) \equiv \sigma_\alpha\{\beta(t), r_\alpha(t)\} = n \left[\frac{e^2}{m_\alpha} \right] \tau_{c\alpha}(t), \quad (19)$$

and then in Eq. (18) $\sigma(t)$ plays the role of an instantaneous conductivity and Eq. (19) is reminiscent of Drude's result, but is a highly nonlinear expression in the drift velocity, and then on the field strength. Equation (18) stands for a *generalized Ohm's law for the nonlinear charge transport regime in the nonequilibrium highly photoexcited and field-dependent state of the system*.

Let us consider, for the sake of a simplified illustration, the particular limit of Eq. (18) corresponding to the linear transport regime, defined by the condition that

$$x_\alpha(t) = \beta(t) \frac{1}{2} m_\alpha v_\alpha^2(t) \ll 1, \quad (20)$$

namely, a drift kinetic energy much smaller than the thermal energy, and then up to the lowest order in x , when only the contribution $l=1$ in Eq. (11b) is taken and introduced in Eq. (15), we find that

$$\tau_{p\alpha}^{-1}(t) \simeq \left[\frac{\beta(t)}{2m_\alpha} \right]^{1/2} \Phi_{\alpha 1}(z(t)), \quad (21)$$

with Φ_1 given by Eq. (A10). Also, it ought to be noted that in the stationary state (ss), when in Eq. (17) $\tau_{p\alpha}$ is taken as time independent, we find that $\tau_{c\alpha}^{(\text{ss})} = \tau_{p\alpha}^{(\text{ss})}$, meaning that the characteristic time for conduction coincides

with the momentum relaxation time.

Finally the mobility⁴ η_0 for each type of carrier, once σ_α is given by Eq. (19), takes the form

$$\eta_\alpha(t) = \left[\frac{e}{m_\alpha} \right] \tau_{c\alpha}(t). \quad (22)$$

Consequently, given the diffusion coefficient in Sec. II A, and the mobility in Sec. II B, we can proceed to consider the Einstein relation.

III. GENERALIZED EINSTEIN RELATION

Let us now consider a NTER in general nonequilibrium and nonlinear conditions. First of all, care must be taken of the fact that the presence of an electric field breaks the otherwise spherical symmetry of the model, and so we must define a diffusion tensor. We will express this in a reference frame consisting of one axis in the direction of the field (let us call it the parallel axis), and two in the perpendicular plane (let us call them the transverse axes). In it the diffusion tensor is diagonal with one component D_{\parallel} and two identical D_{\perp} . We next consider two separate situations, namely (1) the limit of weak fields (linear regime) when for the diffusion coefficient we can use the expression of Eq. (1), and then

$$\mathcal{N}(t)_\alpha = \frac{eD_\alpha(t)}{k_B T^*(t)\eta_\alpha(t)} = \frac{m_\alpha c_\alpha^2(t)}{3k_B T^*(t)} \frac{\tau_{p\alpha}(t)}{\tau_{c\alpha}(t)}, \quad (23)$$

with τ_c given by Eqs. (17), in which is introduced $\tau_{p\alpha}$ given by Eq. (21), and (2) in the case of intermediate to high fields (nonlinear regime) when for the parallel component of the diffusion tensor we find that

$$\mathcal{N}(t)_{\alpha\parallel} = \frac{eD_{\alpha\parallel}(t)}{k_B T^*(t)\eta_\alpha(t)} = \frac{m_\alpha c_{\alpha\parallel}^2(t)}{k_B T^*(t)} \frac{\tau_{\alpha\parallel}(t)}{\tau_{c\alpha}(t)}. \quad (24)$$

In Eq. (24) we have introduced

$$\frac{1}{2} m_\alpha c_{\alpha\parallel}^2(t) = \frac{b_{\alpha\parallel}(t)}{m_\alpha a_{\alpha}(t)}, \quad (25a)$$

$$\tau_{\alpha\parallel}(t) = \frac{b_{\alpha\parallel}(t)}{A_{\alpha\parallel}(t)}, \quad (25b)$$

a_α is the same as in Eq. (43) in I, and

$$b_{\alpha\parallel}(t) = \sum_{\mathbf{k}} h^2 k_{\parallel}^2 f_{\mathbf{k}}^\alpha(t) [1 - f_{\mathbf{k}}^\alpha(t)], \quad (26a)$$

$$A_{\alpha\parallel}(t) = \sum_{\mathbf{k}q_{\parallel}} A_{\mathbf{k}q_{\parallel}}^\alpha \hbar^2 q_{\parallel} k_{\parallel} f_{\mathbf{k}}^\alpha(t) [1 - f_{\mathbf{k}}^\alpha(t)], \quad (26b)$$

where A is given by Eq. (20g) in I, and k_{\parallel} and q_{\parallel} are the corresponding wave vectors in the direction of the electric field. Moreover, $f_{\mathbf{k}}$ is given by Eq. (9), and the characteristic time for diffusion τ_{\parallel} takes the form

$$\tau_{\alpha\parallel}^{-1}(t) = \left[\frac{\beta(t)}{2m} \right]^{1/2} \xi(x_\alpha(t), z(t)), \quad (27)$$

where

$$\begin{aligned} \xi(x_\alpha(t), z(t)) &= 2e^{-x_\alpha(t)} \\ &\times \sum_{l=1}^{\infty} x_\alpha^{l-1}(t) \left[l - \frac{1}{2} - x_\alpha(t) \right] \Phi_{\alpha l}(z(t)). \end{aligned} \quad (28)$$

Let us next consider these two limiting cases (1) and (2) in some detail.

A. Linear regime (transient and stationary conditions)

This regime is characterized by the condition imposed by Eq. (20), in which, after using Eq. (9), the velocity c is given by

$$\frac{1}{2} m_\alpha c_\alpha^2(t) = \frac{3}{2} k_B T_\alpha^*(t), \quad (29)$$

and Eq. (23) becomes

$$\mathcal{N}(t)_\alpha = \frac{\tau_{p\alpha}(t)}{\tau_{c\alpha}(t)}. \quad (30)$$

We recall that in this regime and the nondegenerate limit we are considering, the characteristic time for diffusion becomes the momentum relaxation time.

In the *linear transient regime* and in *stationary conditions*, i.e., after the transient time following application of the field has elapsed, we have shown that $\tau_{p\alpha}$ and $\tau_{c\alpha}$ coincide, and then we recover the original Einstein relation, valid in the domain of the linear theory of relaxation, and the nondegenerate regime

$$\mathcal{N}|_{\text{linear}}^{(\text{ss})} = 1. \quad (31)$$

This is not the case, we emphasize, during the ultrafast transient time when Eq. (30) remains valid. To illustrate the case we proceed to perform numerical calculations involving a photoexcited GaAs sample, by solving Eqs. (4)

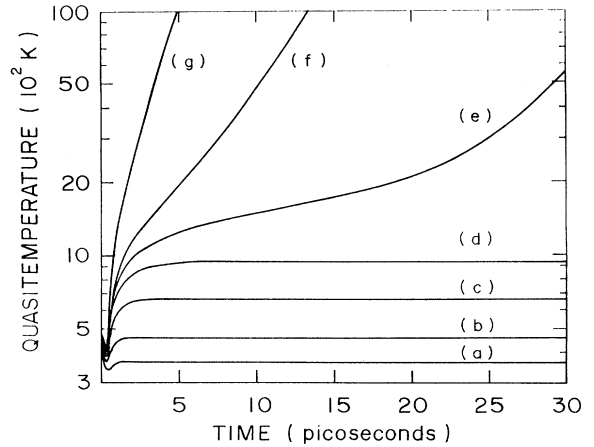


FIG. 1. Evolution in time of the carrier quasitemperature for several values of the electric-field strength: (a) 4 kV/cm, (b) 6 kV/cm, (c) 8 kV/cm, (d) 9 kV/cm, (e) 9.5 kV/cm, (f) 10 kV/cm, and (g) 12 kV/cm. (The runaway effect arises for a field strength of roughly 9.3 kV/cm.)

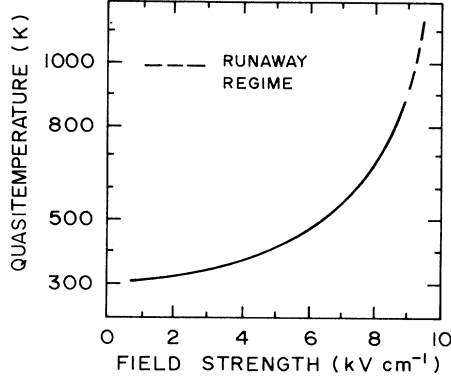


FIG. 2. The carrier quasitemperature in the stationary state as a function of the electric-field strength.

in the linear regime, using only the contributions up to order $l=0$ in Eq. (11a) and $l=1$ in Eq. (11b) and taking the exponential as equal to 1. We consider an ultrashort laser pulse with a photon energy of 120 meV above the gap value: the concentration (fixed by the pulse intensity) is taken as $n=10^{17} \text{ cm}^{-3}$, and several values of the electric-field intensity are considered. We concentrate our attention on the contributions due to the electrons, the relevant ones (the drift velocity of the electrons is nearly one order of magnitude larger than that of the holes). In Fig. 1 we show the evolution of the carrier quasitemperature, while in Fig. 2 are indicated its stationary values as a function of the electric-field intensity. Figure 3 shows the evolution of both characteristic times for electrons, and in Fig. 4 we can see the evolution of the NTER. Inspection of this figure tells us that, as predicted, the Einstein relation holds in the steady state, while during the transient the relation is higher than 1. At $t=0$, since $\tau_c(t)$ is proportional to $v(t)$, one has $\tau_c(0)=0$ (that is, the mobility is null previous to the application of

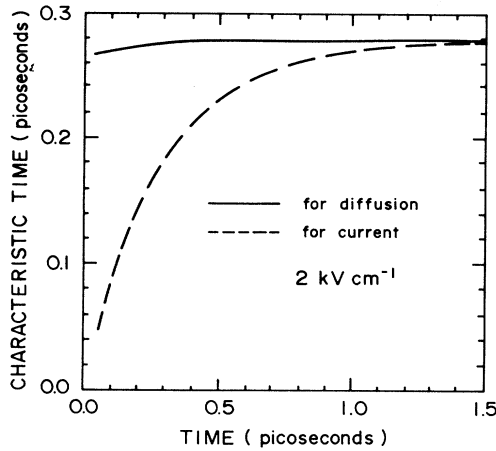


FIG. 3. Evolution in the ultrafast transient of the electron characteristic times for the diffusion and for the current.

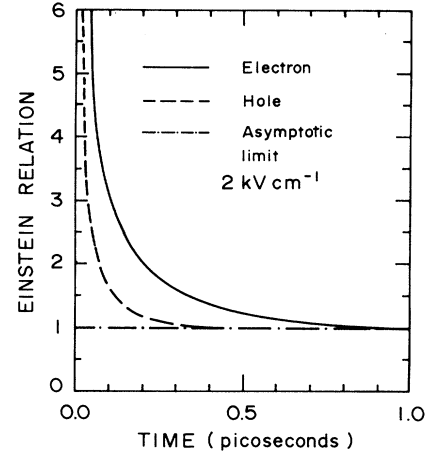


FIG. 4. Evolution in the ultrafast transient of the Einstein relation for electrons and for holes: in each case all the curves are in near coincidence in the range of fields between 1 and 3 kV/cm.

the electric field). Hence the characteristic time for conduction is initially much smaller than the characteristic time for diffusion, but keeps increasing rapidly until both coincide in the steady state, when they become equal to the stationary relaxation time for the momentum. Hence

$$\mathcal{N}(t)_{\text{linear}} \geq 1, \quad (32)$$

the equal sign following, as indicated by Eq. (31), in the steady state.

B. Nonlinear regime

Consider now the NTER outside the linear domain. In this case and in the regime characterized by Eq. (9), it follows that

$$\frac{1}{2}m_\alpha c_{\alpha\parallel}^2(t) = \frac{1}{2}k_B T^*(t) + \frac{1}{2}m_\alpha v_\alpha^2, \quad (33)$$

where v_α is the drift velocity of Sec. II B. Therefore, the NTER as given by Eq. (24) in the nonlinear transport regime (intermediate to high fields) is given by

$$\mathcal{N}(t)_{\alpha\parallel} = [1 + 2x_\alpha(t)] \times \frac{[\beta(t)/2m_\alpha]^{-1/2} \xi^{-1}(x_\alpha(t), z(t))}{\tau_{c\alpha}(t)}. \quad (34)$$

Let us consider the steady state, when, we recall, $\tau_{c\alpha} = \tau_{p\alpha}$, and then after using Eqs. (15) and (16) we obtain that

$$\begin{aligned} \mathcal{N}_{\alpha\parallel}^{(ss)} &= [1 + 2x_\alpha] \frac{\varphi(x_\alpha, z)}{\xi(x_\alpha, z)} \\ &= [1 + 2x_\alpha] \frac{\sum_{l=1}^{\infty} x_\alpha^{l-1} \Phi_{l\alpha}(z)}{2 \sum_{l=1}^{\infty} (1 - \frac{1}{2} - x_\alpha) x_\alpha^{l-1} \Phi_{l\alpha}(z)}. \end{aligned} \quad (35)$$

Equation (35), although exact to any power in x , and so in \mathcal{E} (it ought to be recalled that the relation depends on the macroscopic state of the nonequilibrium system, i.e., can only be computed after the equations of evolution [Eqs. (4)] have been solved), has a very cumbersome expression. Evidently, for a quite general case one needs to resort to computational methods to solve the set of highly nonlinear equations of evolution for the basic set of nonequilibrium thermodynamic variables, Eqs. (4), together with the response functions and relations of Eqs. (17), (19), and (35).

Before closing this section we shall consider a particular case as a matter of illustration. For that purpose let us simply consider in Eq. (35) the contribution up to first order in x (numerical calculations for the case of GaAs already described show the approximation to be a good one for $x \leq 1$, which is satisfied below the runaway in the mobility domain, meaning electric fields smaller than 8 kV cm^{-1}). After some algebra, we find that

$$\mathcal{N}_{\alpha||}^{(ss)} \simeq 1 + 2[2 - \lambda_{\alpha}(z)]x_{\alpha}, \quad (36)$$

where

$$\lambda_{\alpha}(z) = \frac{\Phi_{\alpha 2}(z)}{\Phi_{\alpha 1}(z)}, \quad (37)$$

and Φ_1 and Φ_2 given by Eqs. (A10) and (A11). We note that x is proportional to the squared drift velocity and so to the square of the current. Moreover, using Eq. (18), we can write

$$x_{\alpha} = \left[\frac{m_{\alpha} \beta^0}{2e^2 n^2} \right] \sigma_{\alpha 0}^2 \mathcal{E}^2, \quad (38)$$

where the index zero stands for β and σ in the linear regime, to be consistent with the approximation used to keep terms only up to first order in x in Eq. (36). Hence

$$\mathcal{N}_{\alpha||}^{(ss)} \simeq 1 + C_{\alpha}(z_0) \mathcal{E}^2, \quad (39)$$

where

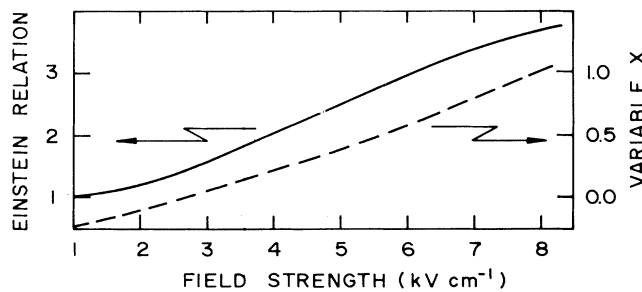


FIG. 5. Dependence of the Einstein relation on the electric-field strength (full line and left ordinate). The dashed line (right ordinate) shows the values of the variable x of Eq. (12a), which characterizes the nonlinearity in the transport theory [cf. Eqs. (11), (19), (32), and (39)].

$$C(z_0) = 2[2 - \alpha_{21}(z_0)]z_0 \left[\frac{m_{\alpha}}{2e^2 n^2 \hbar \omega_0} \right] \sigma_{\alpha}^2(z_0), \quad (40)$$

and coefficient C is dependent on the electric field through the variable z_0 (i.e., through β^0). We can then resort to results already derived for the case of the GaAs sample of Sec. III A, to obtain the dependence of the NTER on the electric-field intensity, in the given approximation shown in Fig. 5. Inspection of this curve tells us that

$$\mathcal{N}_{\alpha||}^{(ss)} \geq 1, \quad (41)$$

with the equal sign standing for low-field intensities, i.e., when $x \ll 1$. In Fig. 5 we have shown the case of electrons, since in the case of the more massive holes the NTER remains near the value 1 (at most 5% larger for 8 kV cm^{-1}).

IV. CONCLUDING REMARKS

We have considered a highly photoexcited plasma in polar semiconductors, and derived a generalized Einstein relation, that is, an Einstein relation in the nonequilibrium conditions of the system whose macroscopic thermodynamic state is evolving under the pumping and dissipative processes to which it is submitted. The theoretical method was based on a quasihydrodynamic approach associated with the informational statistical thermodynamics that the method provides.^{2,8,9} The generalized Einstein relation we considered is the ratio of the diffusion coefficient to the mobility multiplied by the carrier quasitemperature. They are dependent on the nonequilibrium thermodynamic variables and given at each time once they are dependent on the instantaneous macrostate of the system away from equilibrium. We have resorted to Zubarev's approach to the statistical method we used,² and have introduced approximations that are local in space and instantaneous in time; i.e., memory effects and space correlations are neglected which, as opposed to the case of gaseous systems, are unimportant in HEPS as a result of the characteristics of the carrier-phonon interactions.

Sections II A and II B were devoted to the derivation of the diffusion and mobility coefficients, respectively, which were used to obtain the generalized NTER of Eqs. (23) and (24). The characteristic time for diffusion τ and the characteristic time for conduction τ_c are functionals of the basic thermodynamic variables, namely, the carrier quasitemperature, drift velocity, and also the concentration, the latter taken as a constant, and the phonon temperatures; the evolution of these are accounted for in Eqs. (4). All type of phonons contribute to these characteristic times, but we have considered LO phonons exclusively, since the Fröhlich interaction between carriers and this type of phonons is the relevant one. It should be noted that Mathiessen's rule is satisfied in the sense that the inverse of these characteristic times is a simple superposition of the inverse relaxation times associated with each kind of carrier-phonon interaction [cf. Eq. (5)].

Considering the nondegeneratelike limit characterized

by Eq. (9) (a very good approximation in typical situations), we were able to obtain compact analytic expressions for these characteristic times. In the regime of weak electric-field intensities the characteristic time for diffusion was shown to coincide with the momentum relaxation time. As a consequence, in the steady-state regime, when the characteristic time for conduction also coincides with the momentum relaxation time, we recover the NTER in its usual form [cf. Eq. (31)]. This is not the case during the ultrafast transient, when the instantaneous characteristic time for conduction is smaller than the instantaneous momentum relaxation time. This was shown for the case of photoexcited GaAs in Figs. 1–3.

In Sec. III B we considered the case of intermediate to high fields, to obtain what can be termed the generalized NTER for nonlinear charge transport. We looked for the diagonal component of the diffusion tensor in the direction of the applied electric field. Again in the nondegeneratelike regime characterized by Eq. (9), we obtain the characteristic time for diffusion for arbitrary field intensity [cf. Eq. (27)], while that for the momentum relaxation is given in Sec. II B [cf. Eq. (15)]. The NTER in these general conditions is given by Eq. (34), an extremely cumbersome expression: in it $\tau_c(t)$ is given by Eq. (17), and we stress once again that the calculation of the NTER is coupled to the solution of the equations of evolution for the basic variables, viz., Eq. (4).

In the steady state, as noted, the characteristic time for conduction coincides with the momentum relaxation time, and so we can write Eq. (35). It has a formidable structure, which makes its general analysis quite difficult. For illustrative purposes we resorted to calculating the first nonlinear correction, which is quadratic in the drift velocity (current), implying that in the case of field strengths the quantity x is smaller than 1. The result is given in Eqs. (38) and (40), and the dependence on the field is displayed in Fig. 5. We can see that the NTER increases with the electric-field strength, essentially as a result of the increasing electric force added to the driving force associated with diffusion in inhomogeneous media, though restricted to weak concentration gradients. The procedure can be extended to include higher-order electric effects so as to cover the cases of very strong electric fields. Those cases would need appropriate computational

methods to solve the set of Eqs. (34) together with (4), (11), and those of the Appendix.

Formal derivations of a higher-order Einstein relation for nonlinear charge transport, including corrections to second order in the field intensity and the concentration gradient, were given by Hope, Feat, and Landsberg.²⁵ A phenomenological treatment for the case of ion gases based in extended irreversible thermodynamics is due to Gonzalez and Jou,²⁶ and a kinetic one is given by Garcia-Colin and Uribe.¹³

In conclusion, resorting to an appropriate statistical irreversible thermodynamic approach we have been able to derive a generalized Einstein relation for ultrafast transient regimes and for non-Ohmic conditions in a plasma in polar semiconductors with inverted parabolic valence and conduction bands. Since the relation involves the diffusion and mobility coefficients, their expressions need be coupled to the equations of evolution for the macrovariables that the method requires for the characterization of the macroscopic nonequilibrium thermodynamic state of the system.^{2,9} As a final word we stress that the numerical results presented above were performed in a nondegeneratelike limit for the carrier system. As stated, this is a good approximation in usual experimental conditions in highly excited semiconductor plasma. It should begin to fail at high concentrations (typically larger than 10^{19} cm^{-3} and carrier quasitemperatures that are not too high), and then—in this degeneratelike regime—the Einstein relation in the conditions that led to Eqs. (23) and (24) does not hold. The characteristic time for diffusion in Eq. (1) does not coincide with the momentum relaxation time of Eq. (15): the coincidence of both [and so, it follows, Eq. (35)] occurs only in the nondegeneratelike regime.

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APPENDIX: DETAILS OF EQS. (11)

In Eqs. (11) we have introduced the quantities

$$\Psi_{al}(z(t)) = A_{\alpha, LO}^F z^{1/2}(t) 2^{2l} \left[(v_0 + 1) \int_0^\infty \frac{dq}{q} \Gamma[l + 1, \beta \epsilon_\alpha^{(+)}(q)] - v_0 \int_{q_{||\alpha}}^\infty \frac{dq}{q} \Gamma[l + 1, \beta \epsilon_\alpha^{(-)}(q)] \right], \quad (\text{A1})$$

where

$$e_\alpha^{(\pm)}(q) = \left[\frac{m_\alpha \omega_0^2}{q^2} \right] \left[1 \pm \left[\frac{\hbar q^2}{2m_\alpha \omega_0} \right] \right], \quad (\text{A2})$$

$$q_\alpha = \left[\frac{2m_\alpha \omega_0}{\hbar} \right]^{1/2}, \quad (\text{A3})$$

$$A_{\alpha, LO}^F = eF_{0\alpha} \left[\frac{2\hbar \omega_0}{\pi m_\alpha} \right]^{1/2}, \quad (\text{A4})$$

and $\Gamma(a, b)$ is an incomplete gamma function,²⁷ and

$$\Phi_{\alpha l}[z(t)] = \beta_{\alpha, \text{LO}}^F 2^{2l} \frac{2l}{(2l+1)!} z^{l+1}(t) \{ \nu_0 [I_l^{(1)}[z(t)] - I_l^{(2)}[z(t)] - (\nu_0 + 1) e^{-z(t)} [I_l^{(1)}[z(t)] + I_l^{(2)}[z(t)]] \} . \quad (\text{A5})$$

where

$$\begin{aligned} I_l^{(1)}[z(t)] &= \frac{\Gamma(l - \frac{1}{2})}{1} z^{-(l+2)/2}(t) e^{z(t)/2} \mathcal{W}_{1/2(l-1), 1/2(l-1)}[z(t)] \\ &\quad + \frac{1}{2}(l-1) \Gamma(l - \frac{3}{2}) z^{-(l+5)/2}(t) e^{z(t)/2} \mathcal{W}_{1/2(l-2), -1/2(l-2)}[z(t)] \\ &\quad - (l-1)(l-2) z^{-2}(t) \int_0^\infty du u^{2(l-3)} \exp\{-u^2 z(t)\} \text{arcSh}(u) , \end{aligned} \quad (\text{A6})$$

$$I_l^{(2)}[z(t)] = \frac{1}{2} \Gamma(l + \frac{1}{2}) z^{-(l+2)/2}(t) e^{z(t)/2} \mathcal{W}_{1/2(l-1), 1/2(l+1)}[z(t)] , \quad (\text{A7})$$

$$B_{\alpha, \text{LO}}^F = \frac{2eE_{0\alpha}}{\sqrt{\pi}} , \quad (\text{A8})$$

with $E_{0\alpha}$ being Fröhlich's field-coupling strength, and $\mathcal{W}_{a,b}(\xi)$ are Whittaker special functions.²⁷ In particular (results explicitly used in the numerical calculations),

$$\Psi_{\alpha 0}[z(t)] = eE_{0\alpha} \left[\frac{2\hbar\omega_0}{\pi m} \right]^{1/2} \left[1 - \frac{\nu_0}{\bar{\nu}(t)} \right] z^{1/2}(t) \exp\{-\frac{1}{2}z(t)\} K_0\left[\frac{1}{2}z(t)\right] , \quad (\text{A9})$$

$$\begin{aligned} \Phi_{\alpha 1}[z(t)] &= \left[\frac{2eE_{0\alpha}}{3\sqrt{\pi}} \right] z(t) e^{z(t)/2} \left\{ -[\nu_0 - e^{-z(t)}(\nu_0 + 1)] K_0\left[\frac{z(t)}{2}\right] \right. \\ &\quad \left. + [\nu_0 + e^{-z(t)}(\nu_0 + 1)] K_1\left[\frac{z(t)}{2}\right] \right\} , \end{aligned} \quad (\text{A10})$$

$$\begin{aligned} \Phi_{\alpha 2}[z(t)] &= \left[\frac{2eE_{0\alpha}}{3\sqrt{\pi}} \right] z(t) e^{z(t)/2} \frac{1}{3} \left\{ -[\nu_0 - e^{-z(t)}(\nu_0 + 1)] [\Gamma(\frac{3}{2}) \mathcal{W}_{-1/2, -1/2}[z(t)] + 2\Gamma(\frac{1}{2}) z^{-3/2}(t) \mathcal{W}_{0,0}[z(t)]] \right. \\ &\quad \left. + [\nu_0 + e^{-z(t)}(\nu_0 + 1)] 2\Gamma(\frac{5}{2}) \mathcal{W}_{1/2, 3/2}[z(t)] \right\} . \end{aligned} \quad (\text{A11})$$

In Eqs. (A9)–(A11) we introduced the auxiliary function

$$\bar{\nu}(t) = [\exp\{\beta(t)\hbar\omega_0\} - 1]^{-1} , \quad (\text{A12})$$

that is, the distribution of the phonons at the carrier quasitemperature: then, from Eqs. (14a) and (18a), the exchange of energy between both subsystems ceases when mutual thermalization is attained, as it should. Furthermore, K_n is the n th modified Bessel function of the second kind.²⁷

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