

On nonequilibrium many-body systems V: ultrafast transport phenomena

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Received 25 May 1989

Abstract The Nonequilibrium Statistical Operator Method and its accompanying non-linear quantum transport theory, described in Rev. Brasil. **Fis.** **15**, 106 (1985); *ibid* **16**, 495 (1986), are used to **perform** an analytical study of the ultrafast mobility transient of central-valley photoinjected carriers in direct-gap polar semiconductors. **Expressions** for the time-resolved mobility of the hot **carriers** submitted to general conditions of **photoexcitation** and electric field intensity are derived. Numerical results for the case of low to **moderately high** fields are shown, and a **qualitative comparison** with available experiments is done. It is shown that the mobility transient displays a structure composed of a **maximum** and a minimum before attaining the steady state. The characteristics of **this** structure depend on the degree of photoexcitation and electric field intensity. A **brief** discussion of the carriers' diffusion **coefficient** is done.

1. Introduction

Studies of the optical and transport properties of semiconductors under high levels of excitation have shown **novel** and quite interesting features, evidenced in ultrafast **laser** spectroscopy experiments. Notable improvements in time resolved laser spectroscopy have **made** it a very **useful tool** to be used with **confidence** for the investigation of very rapid microscopic mechanisms in the biological and physical **realms**¹. These kind of studies are of great interest because of the **variety** of phenomena observed, most of them of **relevance** in the functioning of some

semiconductor devices, **and** also because they **provide** an excellent **testing** ground for theoretical **ideas** in the field of many-body systems far from equilibrium. The question of very **fast** relaxation processes in highly photoexcited **plasma** in semiconductors (HEPS) **was** recently the object of experimental **and** theoretical study (see list of **references 1**). We **analyzed** the question of relaxation of photoinjected carriers in direct-gap polar semiconductors resorting to the **nonequilibrium statistical** operator method (NSOM)². The NSOM is a powerful formalism that **seems** to offer an **elegant and concise valuable** analytical treatment in the theory of irreversible processes, **adequate** to deal with a large **class** of experimental situations. It is considered a far reaching generalization of the Chapman-Enskog approach in the kinetic theory of **gases**³ or of the **Mori-Langevin** formalism⁴, **and** we use it to calculate the mobility of far-from-equilibrium carriers in HEPS⁵.

Several approaches to hot-carriers quantum transport are presently available, and numerical methods, such as the Monte Carlo computational approach, have shown recent remarkable improvements⁶. However, **analytical** methods for studying the nonlinear transport in HEPS under the action of intense electric fields are highly desirable in order to obtain a better physical insight and for the interpretation of new phenomena. In particular, the NSOM **allows** us to study nonlinear ultrafast transient transport **in** HEPS arbitrarily away from equilibrium **and** for any value of the electric field intensity. The generalized nonlinear quantum transport theory derived from the NSOM is reviewed in the work of ref.7, where we applied it to construct an extensive treatment of mobility in HEPS. As shown in that work we can demonstrate the **existence** of a **novel** feature in the evolution curves of the drift velocity which we termed structured ultrafast transport, viz. maxima - relative or absolute (overshoot) - and minima that may appear before a steady-state is reached. A criterion for the occurrence of this structure, and **also** overshoot effects, was derived and **several** general properties stated.

In this **article** we **present** a complete study of the ultrafast mobility transient of photoinjected carriers in HEPS in the so-called linear NSOM-linear theory of relaxation, and numerical calculations appropriate for the case of **GaAs** are done.

We confirm numerically the general qualitative results reported in ref. 7, in particular, the criterion for the **existence** of structured transient transport is verified, and further we show that there exist three different regimes in the transient transport depending on the range of values of the electric field intensity.

2. Nonlinear quantum transport in HEPS

We consider the case of a polar semiconductor described by a two inverted-parabolic bands model, where a concentration n of electron-hole pairs has been created by an intense pulse of laser light. These carriers are in a state strongly departed from equilibrium but in a condition of internal equilibrium (hot carriers) as a result of the Coulomb interaction^{2,8}. A constant electric field of intensity \mathcal{E} in, say, that x -direction is applied, accelerating these carriers, which, at the same time, transfer energy and momentum to the phonon field. The **sample** is in contact with a thermal reservoir at temperature T_0 , and the phonons are warmed up in scattering events involving Frölich, deformation potential, and piezoelectric interactions with the carriers⁹.

To deal with the irreversible thermodynamic evolution and transport properties of this system we resort, as indicated in the Introduction, to the NSOM. We **recall** that the NSOM requires, as a first step, the choice of a basic set of variables to describe the macrostate of the nonequilibrium **system**^{4,5}. For the **present** case we select the eight dynamical quantities P_j , $j = 1, 2, \dots, 8$, consisting of: the carriers, longitudinal optical (L), transverse optical (TO), and **acoustic** (A) phonon Hamiltonians (H ., H_{LO} , H_{TO} , H_A , respectively), the number operators for electrons N_e , and for holes, N_h , and the linear momenta (its component in the direction of the electric field) of electrons, $P_{e\cdot}$, and of holes, P_h . The nonequilibrium macroscopic variables, i.e. the average values of these eight dynamical quantities over the nonequilibrium ensemble. $Q_j(t) = \text{Tr}(P_j \rho(t))$, are - after dividing by the volume of the system - the densities of the corresponding energies, $E_e(t)$, $E_{LO}(t)$, $E_{TO}(t)$, $E_A(t)$, the density of pairs, $\mathbf{n}(t)$ (equal to the density of electrons and of holes), and the density of linear momenta, $\pi_e(t)$, and $\pi_h(t)$. Further, there are eight intensive nonequilibrium variables, $F_j(t)$, thermodynamically

conjugate to the $Q_j(t)$, that define the four reciprocal quasi-temperatura $\beta_e(t) = 1/k T_e^*(t)$, $\beta_{Lo}(t) = 1/k T_{Lo}^*(t)$, $\beta_{To}(t) = 1/k T_{To}^*(t)$, $\beta_A(t) = 1/k T_A^*(t)$, two are associated to quasi-chemical potentials, $-\beta_e(t) \mu_e(t)$, $-\beta_e(t) \mu_h(t)$, and, finally, two are associated do drift velocities, $-\beta_e(t) v_e(t)$, $-\beta_e(t) v_h(t)$.

Among the different approaches in the **NSOM-theory**^{4,7} we resort here to the use of Zubarev's **method**¹⁰. Zubarev's NSO for the isolated system composed of the sample, the thermal reservoir, and the source of the electric field is approximated by

$$\rho_{tot.}(t) = \rho_r \otimes \rho_e(t) , \tag{1}$$

where ρ_r is the statistical operator of the reservoir and source, which is assumed to remain in a stationary state, i.e. practically unchanged by the interaction with the open system defined by the nonequilibrium semiconductor sample, whose NSO is

$$\rho_e(t) \exp \left\{ -\epsilon \int_{-\infty}^0 dt' e^{\epsilon t'} \left[\phi(t+t') + \sum_{j=1}^8 F_j(t+t') P_j(t') \right] \right\} \tag{2}$$

with ϵ going to zero after the trace operation in the calculation of averages has been performed. It can be rewritten^{4,7}

$$\rho_e(t) = \bar{\rho}(t, 0) + \rho'(t) , \tag{3}$$

where

$$\bar{\rho}(t_1, t_2) = \exp \left\{ -\phi(t_1) - \sum_{j=1}^8 F_j(t_1) P_j(t_2) \right\} \tag{4}$$

is an auxiliary statistical operator that defines the instantaneous average values

$$Q_j(t) \text{Tr} \{ P_j \rho_e(t) \} = \text{Tr} \{ P_j \rho_{eq}(t, 0) \} , \tag{5}$$

and $\rho'(t)$ contains the information on the dynamics relevant to the description of the irreversible evolution of the system. Eq.(5) determines the nonequilibrium thermodynamic parameters $F_j(t)$, making them conjugate to **macrovariables** $Q_j(t)$ in the sense of generalized irreversible **thermodynamics**^{4,10}.

Eq. (2) defines the operation of selecting the subgroup of retarded solutions of the Liouville equation corresponding to the initial value problem $\rho_\epsilon(t_0) = \rho_{e_g}(t_0)$ ($t_0 \rightarrow -\infty$). This is guaranteed by the presence of an infinitesimal source in the equation for $\rho_\epsilon(t)$

$$\frac{p}{pt} \ln \rho_\epsilon(t) + \frac{1}{i\hbar} [\ln \rho_\epsilon(t), H] = -\epsilon [\ln \rho_\epsilon(t) - \ln \rho_{e_g}(t, 0)] \quad (6)$$

where H is the total Hamiltonian of the system^{4,10}. Hence, irreversibility is associated to this symmetry breaking and the average of any physical quantity A is defined by a quasi-average in Bogoliubov's sense¹¹

$$\langle A|t \rangle = \lim_{t \rightarrow 0} \text{Tr}\{A\rho_\epsilon(t)\} \quad (7)$$

Invariance under time-reversal transformations is not satisfied for these quasi-averages because of the removal of the corresponding degeneracy in eq. (6); this is equivalent to introducing Prigogine's dynamic condition for dissipativity¹².

Further, the total Hamiltonian H is separated in the form $H = H_0 + H' + H_\epsilon$, where H_0 contains the energy operators of each individual subsystem, i.e. $H_0 = H_L + H_{LO} + H_{TO} + H_A$. The interaction between them and with the external reservoirs are included in H' , i.e. it is composed of the interaction energies of carriers with the phonon field, anharmonic interaction between phonons, and the interaction with the thermal reservoir (responsible for heat diffusion out of the sample). Finally, the interaction of the carriers with the electric field is

$$H_\epsilon = -e\mathcal{E} \sum_j (x_{e_j} - x_{h_j}) \quad (8)$$

where $x_{e(h)_j}$ is the coordinate of the j -th electron (hole).

For the given choice of the basic set of dynamical variables we find $[P_j, H_0] = 0$ and $[P_j, P_k] = 0$, and the Coulomb interaction between carriers, contained in H_0 , is treated in the random phase approximation¹³, i.e. the carriers are considered as a two-component Landau's Fermi fluid. Coulomb interaction is only called forth indirectly to ensure the internal thermalization of carriers at any time⁸. We recall

that the conduction and (heavy hole) valence Bloch bands are taken in the effective mass approximation.

Using these results, the evolution equations for macrovariables $Q_j(t)$, $j = 1, 2, \dots, 8$, i.e.,

$$\frac{d}{dt} Q_j(t) = \frac{1}{i\hbar} \text{Tr}\{[P_j, H_0 + H' + H_\varepsilon] \rho_c(t)\} , \quad (9)$$

can be written in the NSOM-linear theory of relaxation (LTR)^{7,14} in the form

$$\frac{d}{dt} Q_j(t) = J_j^{(1)}(t) + J_j^{(2)}(t) , \quad (10)$$

with the collision operator \mathbf{J} given for this case by⁷

$$J_j^{(1)}(t) = \frac{1}{i\hbar} \text{Tr}\{[P_j, H_\varepsilon] \rho_{cg}(t, 0)\} , \quad (11a)$$

$$J_j^{(2)}(t) = \left(\frac{1}{i\hbar}\right)^2 \int_{-\infty}^0 dt' \exp(\varepsilon t') \text{Tr}\{[H'(t'), [H', P_j]] \rho_{cg}(t, 0)\} . \quad (11b)$$

Although the LTR truncates, to second order in H' , the series of collision operators containing averages over the coarse-grained ensemble of terms of increasing order in the interaction strengths, eqs. (9) are highly nonlinear differential equations. The right-hand sides of them are functionals of the intensive variables F_j , and then it is convenient to rewrite the left-hand side in terms of the latter, which can be accomplished noting that

$$\frac{d}{dt} Q_j(t) = \sum_k \frac{\delta Q_j(t)}{\delta F_k(t)} \frac{d}{dt} F_k(t) = - \sum_k C_{jk}(t) \frac{d}{dt} F_k(t) , \quad (12)$$

where $C_{jk}(t)$ are the elements of the correlation matrix,

$$C_{jk}(t) = \text{Tr}\{P_j(t) \Delta P_k(t) \rho_{cg}(t)\} \equiv (P_j; P_k|t) , \quad (13)$$

with $\Delta P(t) = P - \langle P|t \rangle$.

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We are now in a condition to calculate the generalized transport equations for the **basic** set of macrovariables, which are

$$\frac{1}{V} \cdot \frac{d \langle H_C | t \rangle}{dt} = \sum_{\alpha} \frac{e \mathcal{E}}{m_{\alpha} V} \cdot \langle P_{\alpha} | t \rangle - \sum_{\alpha, \eta, i} \dot{E}_{\alpha, \eta}^i(t) \quad , \quad (14a)$$

$$\frac{1}{V} \cdot \frac{d \langle H_{LO} | t \rangle}{dt} = \sum_{\alpha, i} \dot{E}_{\alpha, LO}^i(t) - \dot{E}_{LO, AN}(t) \quad , \quad (14b)$$

$$\frac{1}{V} \cdot \frac{d \langle H_{TO} | t \rangle}{dt} = \sum_{\alpha, i} \dot{E}_{\alpha, TO}^i(t) - \dot{E}_{TO, AN}(t) \quad , \quad (14c)$$

$$\begin{aligned} \frac{1}{V} \cdot \frac{d \langle H_{AC} | t \rangle}{dt} &= \sum_{\alpha, i} \dot{E}_{\alpha, AC}^i(t) + \dot{E}_{LO, AN}(t) \\ &+ \dot{E}_{TO, AN}(t) - \dot{E}_{AC, DIF}(t) \end{aligned} \quad (14d)$$

$$\frac{1}{V} \cdot \frac{d \langle P_{\alpha} | t \rangle}{dt} = n e \mathcal{E} - \sum_{\eta, i} \dot{\pi}_{\alpha, \eta}^i(t) \quad . \quad (14e, f)$$

where we took the modulus of the linear momentum in the direction of the electric field. The two equations for the average number of electrons and **holes** are not considered since, for our purposes here, they are constant because recombination effects are relevant in a near nonosecond time scale while our interest is in the few picoseconds' time scale.

In eq.(14a) the first term on the right-hand side accounts for the **energy transfer** from the electric field, the second is the rate of variation due to the **carrier-phonon** interaction having the contributions

$$\begin{aligned} \dot{E}_{\alpha, \eta}^i(t) &= \frac{2\pi}{\hbar} \cdot \sum_{\vec{k}, \vec{q}} \hbar \omega_{\vec{q}, \eta} \cdot |M_{\alpha \eta}^i(\vec{q})|^2 \cdot \{ \nu_{\vec{q}, \eta}(t) \cdot f_{\vec{k}, \alpha}(t) \cdot [1 - f_{\vec{k}+\vec{q}, \alpha}(t)] - \\ &- [1 + \nu_{\vec{q}, \eta}(t)] \cdot f_{\vec{k}+\vec{q}, \alpha}(t) \cdot [1 - f_{\vec{k}, \alpha}(t)] \} \cdot \delta(\epsilon_{\vec{k}+\vec{q}, \alpha} - \epsilon_{\vec{k}, \alpha} - \hbar \omega_{\vec{q}, \eta}) \quad (15) \end{aligned}$$

where the index n is LO, TO, or A, and the upper index i refers to the different types of interactions, PD, **PZ** and FR for deformation potential, **piezoelectric**, and Fröhlich interactions respectively. Further,

$$\nu_{\vec{q},n}(t) = 1/\{\exp [\beta_n(t)\hbar\omega_{\vec{q},n}] - 1\} \quad (16)$$

are the distribution functions for n-type phonons, and $\omega_{\vec{q},n}$ the corresponding frequency dispersion relations, and

$$f_{\vec{k},\alpha}(t) = [4\pi^3 \hbar^3 n / (2\pi m_\alpha)^{3/2}] \beta_c^{3/2}(t) \exp \{ -\beta_c(t) [\hbar\vec{k} - m_\alpha \vec{v}_\alpha(t)]^2 / 2m_\alpha \} \quad (17)$$

are the carriers' distribution functions; at the high excitation levels being considered we can use the above **instantaneous Maxwell-Boltzmann** distribution, which contains a shift term in the exponential due to the **presence** of the electric field.

In eqs. (14 **b,c**, and d) the first term on the right-hand **side** accounts for the energy transfer from the carriers to the phonon (it is equal to the corresponding term in eq.(14**a**) with a change of sign). The second term in eqs.(14**b** and c), $\dot{E}_{LO(TO),AN}$, is the rate of energy transfer to the acoustic phonons via **anharmonic** interactions, and is written in the form

$$\dot{E}_{LO(TO),AN}(t) = \sum_{\vec{q}} \hbar\omega_{\vec{q},LO(TO)} \frac{\nu_{\vec{q},LO(TO)}(t) - \nu_{\vec{q},LO(TO)}(t, \beta_{AC})}{\tau_{LO(TO)}} \quad (18)$$

where

$$\nu_{\vec{q},LO(TO)}(t, \beta_{AC}) = 1/\{\exp [\beta_{AC}(t)\hbar\omega_{\vec{q},LO(TO)}] - 1\} \quad (19)$$

and $\tau_{LO(TO)}$ is a phenomenological relaxation time, to be evaluated from **Raman** scattering linewidths. The same term with a change of sign **appears** in eq. (14**d**). The **last** term in eq. (14**d**) is the rate of change of the A-phonons' energy due to heat diffusion to the thermal reservoir, and we write for it

$$\dot{E}_{AC, DIF}(t) = \sum_{\vec{q}} \hbar\omega_{\vec{q},AC} \tau_{AC}^{-1} [\nu_{\vec{q},AC}(t) - \nu_{\vec{q},AC}(\beta_B)] \quad (20)$$

where

$$\nu_{\vec{q},AC}(\beta_B) = 1/\{\exp[\beta_B \cdot \hbar\omega_{\vec{q},AC}] - 1\} \quad (21)$$

β_B is the reciprocal temperature of the reservoir, and τ_{AC} is a phenomenological relaxation time which depends on the diffusion coefficient and the dimensions of the surface of the active volume of the crystal¹⁵.

Finally, in eqs. (14e and f) the first term on the right-hand side is a drift force due to the action of the electric field. The second term contributes to the rate of variation of the momentum as a result of collisions with phonons, and is given by

$$\begin{aligned} \dot{\pi}_{\alpha,\eta}^i(t) = & \frac{2\pi}{\hbar} \cdot \sum_{\vec{k},\vec{q}} \hbar q_{\mathcal{E}} \cdot |M_{\alpha,\eta}^i(\vec{q})|^2 \cdot \left\{ \nu_{\vec{q},\eta}(t) \cdot f_{\vec{q},\eta}(t) \cdot [1 - f_{\vec{k}+\vec{q},\alpha}(t)] - \right. \\ & - [1 + \nu_{\vec{q},\eta}(t)] \cdot f_{\vec{k}+\vec{q},\alpha}(t) \cdot [1 - f_{\vec{k},\alpha}(t)] \left. \right\} \cdot \delta(\epsilon_{\vec{k}+\vec{q},\alpha} - \epsilon_{\vec{k},\alpha} - \hbar\omega_{\vec{q},\eta}) + \\ & + \frac{2\pi}{\hbar} \cdot \sum_{\vec{k},\vec{q}} \hbar q_{\mathcal{E}} \cdot |M_{\alpha,\eta}^i(\vec{q})|^2 \cdot \left\{ [\nu_{\vec{q},\eta}(t) + 1] \cdot f_{\vec{k},\alpha}(t) \cdot [1 - f_{\vec{k}-\vec{q},\alpha}(t)] - \right. \\ & \left. - \nu_{\vec{q},\eta}(t) \cdot f_{\vec{k}-\vec{q},\alpha}(t) \cdot [1 - f_{\vec{k},\alpha}(t)] \right\} \cdot \delta(\epsilon_{\vec{k}-\vec{q},\alpha} - \epsilon_{\vec{k},\alpha} + \hbar\omega_{\vec{q},\eta}) \quad (22) \end{aligned}$$

where $q_{\mathcal{E}}$ is the modulus of the component of \vec{q} in the direction of the electric field.

Next we take Einstein's model for the optical phonons (with dispersionless frequencies ω_{LO} and ω_{TO}) and a Debye model for acoustic phonons ($\omega_{AC}(\mathbf{q}) = sq$, with triple degeneracy; \mathbf{s} is the **velocity** of sound). Next, performing the integrations in reciprocal space, i.e. the **summation** over \vec{k} and \vec{q} , we obtain

$$\begin{aligned} \dot{E}_{\alpha,AC}^{PD}(t) = & A_{\alpha,AC}^{PD}(t) \exp[-x_{\alpha}(t)] \left\{ M(3,3/2, x_{\alpha}(t)) + \right. \\ & + \left[3y_{\alpha}(t) - \frac{\beta_C(t)}{\beta_{AC}(t)} \right] \cdot M(2,3/2, x_{\alpha}(t)) - \\ & - \frac{3\pi^{1/2}}{2} y_{\alpha}^{1/2}(t) M(5/2,3/2, x_{\alpha}(t)) - \pi^{1/2} \cdot y_{\alpha}^{3/2}(t) \cdot \exp(x_{\alpha}(t)) - \\ & \left. - \frac{1}{3} y_{\alpha}(t) [1 - 3\sqrt{2}y_{\alpha}(t)] \cdot M(1,3/2, x_{\alpha}(t)) \right\}, \quad (23) \end{aligned}$$

$$\begin{aligned} \dot{E}_{\alpha,AC}^{PZ}(t) = & A_{\alpha,AC}^{PZ}(t) \exp[-x_{\alpha}(t)] \left\{ M(2, 3/2, x_{\alpha}(t)) + \right. \\ & + \left[y_{\alpha}(t) - \frac{\beta_C(t)}{\beta_{AC}(t)} \right] \cdot M(1, 3/2, x_{\alpha}(t)) - \\ & \left. - \pi^{1/2} y_{\alpha}^{1/2}(t) \exp[x_{\alpha}(t)] \right\} \end{aligned} \quad (24)$$

where

$$A_{\alpha,AC}^{PD}(t) = \theta_{AC} n \left(\frac{2^{7/2} m_{\alpha}^{5/2} E_{1\alpha}^2}{\pi^{3/2} \hbar^4 \rho} \right) \beta_C^{-3/2}(t), \quad (25)$$

$$A_{\alpha,AC}^{PZ}(t) = \theta_{AC} n \left(\frac{2^{1/2} m_{\alpha}^{3/2} e^2 H_{PZ}^2}{\pi^{3/2} \hbar^2 \rho \epsilon_0^2} \right) \beta_C^{-1/2}(t), \quad (26)$$

$$x_{\alpha}(t) = \beta_C(t) \frac{1}{2} m_{\alpha} v_{\alpha}^2(t), \quad (27a)$$

$$y_{\alpha}(t) = \beta_C(t) \frac{1}{2} m_{\alpha} s^2. \quad (27b)$$

$E_{1\alpha}$ is the deformation potential coupling constant, H_{pz} the piezo-electric potential coupling constant, ϵ_0 the static dielectric constant, ρ the density of the material, $\theta_{AC} = 3$ is the degeneracy of the acoustic modes, and $M(a, b, x)$ are Kummer functions¹⁶; also

$$\begin{aligned} \dot{E}_{\alpha,LO}^{FR}(t) = & A_{\alpha,LO}^{FR}(t) \exp[-z_{LO}(t) - x_{\alpha}(t)] \cdot \\ & \cdot \sum_{\ell=1}^{\infty} \sum_{\ell'=0}^{\infty} \frac{1}{(2\ell+1)!} \cdot \frac{[2^3 \cdot z_{LO}(t) \cdot x_{\alpha}(t)]^{\ell'}}{2\ell-1} \left\{ [1 + \nu_{LO}(t)] \cdot \right. \\ & \exp[-z_{LO}(t)] \Gamma(\ell + 1/2) \cdot \\ & U(\ell + 1/2, \ell' + 2; 2z_{LO}(t)) - \nu_{LO}(t) \exp[z_{LO}(t)] \Gamma(\ell' + L + 1/2) \cdot \\ & \left. \cdot U(\ell' + \ell + 1/2, \ell + 2; 2z_{LO}(t)) \right\} \end{aligned} \quad (28)$$

$$\begin{aligned}
 \dot{E}_{\alpha,LO}^{PD}(t) &= A_{\alpha,LO}^{PD}(t) \exp[-z_{LO}(t) - x_{\alpha}(t)] \\
 &\cdot \sum_{\ell=0}^{\infty} \frac{[2^3 \cdot z_{LO}(t) x_{\alpha}(t)]^{\ell}}{(2\ell+1)!} \cdot \left\{ [1 + \nu_{LO}(t)] \exp[-z_{LO}(t)] \cdot \Gamma(3/2) \cdot \right. \\
 &U(3/2, \ell+3; 2z_{LO}(t) - \nu_{LO}(t) \exp[z_{LO}(t)] \Gamma(\ell+3/2) \cdot \\
 &\left. \cdot U(\ell+3/2, \ell+3; 2z_{LO}(t)) \right\} \quad (29)
 \end{aligned}$$

where

$$\nu_{LO}(t) = 1 / \exp[\beta_{LO}(t) \hbar \omega_{LO}] - 1 \quad , \quad (30)$$

$$A_{\alpha,LO}^{FR}(t) = \theta_{LO} n \left(\frac{2^{3/2} e E_{0\alpha}}{\pi^{1/2} m^{1/2}} \right) (\hbar \omega_{LO})^2 \beta_C^{3/2}(t) \quad , \quad (31)$$

$$A_{\alpha,LO}^{PD}(t) = \theta_{LO} n \left(\frac{m_{\alpha}^{3/2} D_{LO\alpha}^2}{\pi^{3/2} \hbar^2} \right) (\hbar \omega_{LO})^2 \beta_C^{3/2}(t) \quad , \quad (32)$$

$$L_O(t) = \beta_C(t) \hbar \omega_{LO} / 2 \quad , \quad (33)$$

$E_{0\alpha}$ is Fröhlich field, $D_{LO\alpha}$ is LO-phonon deformation potential interaction, $\theta_{LO} = 1$ is the degeneracy of LO phonons, $\Gamma(a)$ are gamma functions, and $U(a, b, z)$ are confluent hypergeometric functions¹⁶, and

$$\dot{E}_{\alpha,TO}^{PD} = \text{same as } \dot{E}_{\alpha,LO}^{PD} \text{ with exchange } LO \rightarrow TO \quad , \quad (34)$$

and $\theta_{TO} = 2$.

Further,

$$\dot{E}_{LO(TO),AN}(t) = \frac{1}{V_{\text{cell}}} \hbar \omega_{LO} \tau_{LO}^{-1} [\nu_{LO(TO)}(t) - \nu_{LO(TO)}(t, \beta_{AC})] \quad , \quad (35)$$

$$\dot{E}_{AC,AN}(t) \simeq \frac{1}{V_{\text{cell}}} \tau_{AC}^{-1} \left[\frac{1}{\beta_{AC}(t)} - \frac{1}{\beta_B} \right] \quad , \quad (36)$$

where V_{cell} is the unit cell volume.

The different contributions to the equations of evolution for the linear momentum are

$$\begin{aligned} \dot{\pi}_{\alpha,AC}^{PD}(t) = & B_{\alpha,AC}^{PD}(t)v_{\alpha}(t)\exp[-x_{\alpha}(t)]\{[1-6y_{AC,\alpha}(t)]M(3,5/2,x_{\alpha}(t))+ \\ & + 2\pi^{1/2}y_{AC,\alpha}^{1/2}(t)[\beta_{AC}(t)/\beta_C(t)]^{1/2}M(7/2,5/2,x_{\alpha}(t))+\dots\} \end{aligned} \quad (37)$$

$$\begin{aligned} \dot{\pi}_{\alpha,AC}^{PZ}(t) = & B_{\alpha,AC}^{PZ}(t)v_{\alpha}(t)\exp[-x_C(t)]\{[1-2y_{AC,\alpha}(t)]M(2,5/2,x_{\alpha}(t))+ \\ & y_{AC,\alpha}^{1/2}(t)[\beta_{AC}(t)/\beta_C(t)]\exp[x_C(t)]\} \end{aligned} \quad (38)$$

where

$$B_{\alpha,AC}^{PD}(t) = \theta_{AC}n\left(\frac{2^{3/2}m_{\alpha}^{5/2}E_{1\alpha}^2}{\pi^{3/2}\hbar^4s^2\rho}\right)\frac{\beta_C^{-1/2}(t)}{\beta_{AC}(t)}, \quad (39)$$

$$B_{\alpha,AC}^{PZ}(t) = \theta_{AC}n\left(\frac{2^{1/2}m_{\alpha}^{3/2}e^2H_{PZ}^2}{3\pi\hbar^2s^2e^2\rho}\right)\frac{\beta^{-1/2}(t)}{\beta_{AC}(t)}, \quad (40)$$

$$y_{AC,\alpha}(t) = \beta_{AC}(t)\frac{1}{2}m_{\alpha}\cdot s^2 \quad (41)$$

Also,

$$\begin{aligned} \dot{\pi}_{\alpha,LO}^{FR}(t) = & B_{\alpha,LO}^{FR}(t)x_{\alpha}^{-1/2}(t)\exp[-2z_{LO}(t)-x_{\alpha}(t)] \\ & \cdot \sum_{\ell=0}^{\infty} \left\{ \left[\frac{2^{3\ell}}{(2\ell+1)!} - \frac{2^{3\ell}}{(2\ell)!} \right] [x_{\alpha}(t)z_{LO}(t)]^{\ell} \cdot \right. \\ & \{ \Gamma(\ell+1/2)[(1+\nu_{LO}(t))U(3/2,\ell+2;2z_{LO}(t))+ \\ & + \nu_{LO}(t)U(\ell+1/2,\ell+2;2z_{LO}(t))] + \\ & + \sum_{\ell'=1}^{\infty} \frac{1}{(2\ell'-1)!} [(1+\nu_{LO}(t))\Gamma(\ell'+1/2)U(\ell'+1/2,\ell+1;2z_{LO}(t))- \\ & \left. - \nu_{LO}(t)\Gamma(\ell+\ell'-1/2)U(\ell+\ell'-1/2,\ell+1;2z_{LO}(t))] \right\}, \end{aligned} \quad (42)$$

$$\begin{aligned}
 \dot{\pi}_{\alpha,LO}^{PD}(t) &= B_{\alpha,LO}^{PD}(t) x_{\alpha}^{-1/2}(t) \exp[-x_{\alpha}(t)] \\
 &\quad \left\{ \left[\left[1 + \frac{\nu(t)}{\nu_{LO}(t)} \right] + \exp[-2z_{LO}(t)] \left[1 - \frac{\nu(t)}{\nu_{LO}(t)} \right] \right\} \right. \\
 &\quad \left\{ \sum_{\ell=0}^{\infty} \left[\frac{2^{3\ell}}{(2\ell+1)!} - \frac{2^{3\ell}}{(2\ell)!} \right] [x_{\alpha}(t) z_{LO}(t)]^{\ell} \frac{6 \cdot z_{LO}(t)}{2\ell+1} U(5/2, \ell+4; 2z_{LO}(t)) \right\} + \\
 &\quad + \left\{ \left[1 - \frac{\nu(t)}{\nu_{LO}(t)} \right] + \exp[-2z_{LO}(t)] \left[1 - \frac{\nu(t)}{\nu_{LO}(t)} \right] \right\} \\
 &\quad \left\{ \sum_{\ell=0}^{\infty} \left[\frac{2^{3\ell}}{(2\ell+1)!} - \frac{2^{3\ell}}{(2\ell)!} \right] [x_{\alpha}(t) z_{LO}(t)]^{\ell} \left[U(3/2, \ell+2; 2z_{LO}(t)) + \right. \right. \\
 &\quad \left. \left. + \left(1 - \frac{3}{2\ell+1} \right) U(3/2, \ell+3; 2z_{LO}(t)) \right] \right\} \quad (43)
 \end{aligned}$$

where

$$B_{\alpha,LO}^{FR}(t) = \theta_{LO} n \left(\frac{eE_{0\alpha}}{\pi^{1/2}} \right) \hbar\omega_{LO} \beta_C(t) , \quad (44)$$

$$B_{\alpha,LO}^{PD}(t) = \theta_{LO} n \left(\frac{m_{\alpha}^2 E_{10\alpha}^2}{2^2 \pi \hbar^4 s^2 \rho} \right) (\hbar\omega_{LO})^{5/2} \nu_{LO}(t) \beta_C(t) , \quad (45)$$

$$\nu(t) = 1 / \{ \exp[\beta_C(t) \cdot \hbar\omega_{LO}] - 1 \} . \quad (46)$$

and finally,

$$\dot{\pi}_{\alpha,TO}^{FR} = \text{same as } \dot{\pi}_{\alpha,LO}^{FR} \text{ with the exchange } LO \leftrightarrow TO , \quad (47)$$

$$\dot{\pi}_{\alpha,TO}^{PD} = \text{same as } \dot{\pi}_{\alpha,LO}^{PD} \text{ with the exchange } LO \leftrightarrow TO , \quad (48)$$

The left-hand sides of eqs.(14) are expressed in terms of the intensive variables, i.e. quasi-temperatures and drift velocities, using the relations

$$E_c(t) = \sum_{\vec{k}_{\alpha}} \epsilon_{\vec{k},\alpha} f_{\vec{k},\alpha}(t) , \quad (49a)$$

$$E_n(t) = \sum_{\vec{q}} \theta_n \hbar\omega_{\vec{q},n} \nu_{\vec{q},n}(t) , \quad (49b)$$

$$\frac{1}{V} \langle P, |t\rangle = n m_\alpha v, (t) . \quad (49c)$$

So far we have obtained a complete derivation of the equations that govern the evolution of the basic set of variables that describe the macrostate of the photoinjected HEPS in a constant electric field. They are valid for any intensity of the electric field strength but relaxation effects due to collision with phonons have been treated in the NSOM-linear theory of relaxation. The collision operators are expressed in terms of series of Kummer and confluent hypergeometric functions, and thus are of a rather difficult numerical manipulation. To simplify these expressions we restrict the calculations to the case of low to moderately high electric fields assuming that the carrier drift energy is smaller or at most comparable with the thermal energy, i.e.

$$\beta_c(t) m_\alpha v_\alpha^2(t)/2 \lesssim 1 . \quad (50)$$

In these conditions the series in eqs.(21, 23, 28, 29, and 34) can be rearranged in the form of a dominant term plus corrections. We retain only the main terms to obtain

$$\dot{E}_{\alpha,AC}^{PD}(t) = A_{\alpha,AC}^{PD}(t) \left[1 - \frac{\beta_C(t)}{\beta_{AC}(t)} \right] , \quad (51)$$

$$\dot{E}_{\alpha,AC}^{PZ}(t) = A_{\alpha,AC}^{PZ}(t) \left[1 - \frac{\beta_C(t)}{\beta_{AC}(t)} \right] , \quad (52)$$

$$\begin{aligned} \dot{E}_{\alpha,LO}^{FR}(t) &= n \theta_{LO} e E_{0\alpha} \left(\frac{4\hbar\omega_{LO}}{\pi m_\alpha} \right)^{1/2} \left[1 - \frac{\nu_{LO}(t)}{\nu(t)} \right] z_{LO}^{1/2}(t) \\ &\exp \left[-z_{LO}(t) \right] K_0(z_{LO}(t)) , \end{aligned} \quad (53)$$

$$\begin{aligned} \dot{E}_{\alpha,LO}^{PD}(t) &= n \theta_{LO} D_{\alpha,LO}^2 \left(\frac{2m_\alpha^3 \hbar\omega_{LO}}{\pi^3 \hbar^4 \rho^2} \right) \left[1 - \frac{\nu_{LO}(t)}{\nu(t)} \right] z_{LO}^{1/2}(t) \\ &\exp \left[-z_{LO}(t) \right] K_1(z_{LO}(t)) , \end{aligned} \quad (54)$$

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$$\dot{E}_{\alpha,TO}^{PD}(t) = \text{same as } \dot{E}_{\alpha,LO}^{PD}(t) \text{ exchanging } LO \leftrightarrow TO , \quad (55)$$

for the terms associated with energy relaxation (we used also that $\beta_{AC} \hbar s q \ll 1$), and for the terms associated with momentum relaxation we find.

$$\dot{\pi}_{\alpha,AC}^{PD}(t) = B_{\alpha,AC}^{PD}(t) v_{\alpha}(t) , \quad (56)$$

$$\dot{\pi}_{\alpha,AC}^{PZ}(t) = B_{\alpha,AC}^{PZ}(t) v_{\alpha}(t) , \quad (57)$$

$$\dot{\pi}_{\alpha,AC}^{FR}(t) = \theta_{LO} n m_{\alpha} \gamma_{\alpha}(t) v_{\alpha}(t) , \quad (58)$$

$$\begin{aligned} \dot{\pi}_{\alpha,LO}^{PD}(t) &= \theta_{LO} n \left(\frac{2^{-1/2} m_{\alpha}^{3/2} E_{10\alpha}^2}{3\pi^{3/2} \hbar^4 s^2 \rho} \right) (\hbar \omega_{LO})^3 \nu_{LO}(t) \beta_C^{3/2}(t) \exp[z_{LO}(t)] v_{\alpha}(t) \cdot \\ &\left\{ \left[\left(i + \frac{\nu(t)}{\nu_{LO}(t)} \right) + \exp(-2z_{LO}(t)) \left(1 - \frac{\nu(t)}{\nu_{LO}(t)} \right) \right] K_2(z_{LO}(t)) - \right. \\ &\left. \left[\left(1 - \frac{\nu(t)}{\nu_{LO}(t)} \right) - \exp(-2z_{LO}(t)) \left(1 - \frac{\nu(t)}{\nu_{LO}(t)} \right) \right] K_1(z_{LO}(t)) \right\} , \quad (59) \end{aligned}$$

$$\dot{\pi}_{\alpha,TO}^{PD}(t) = \text{same as } \dot{\pi}_{\alpha,LO}^{PD}(t) \text{ exchanging } LO \leftrightarrow TO , \quad (60)$$

where

$$\begin{aligned} \gamma_{\alpha}(t) &= \gamma_{\alpha 0} x_{LO}^{3/2}(t) \exp[z_{LO}(t)] \nu_{LO}(t) \\ &\left\{ \left[\left(i + \frac{\nu(t)}{\nu_{LO}(t)} \right) + \exp(-2z_{LO}(t)) \left(1 - \frac{\nu(t)}{\nu_{LO}(t)} \right) \right] K_1(z_{LO}(t)) - \right. \\ &\left. \left[\left(1 - \frac{\nu(t)}{\nu_{LO}(t)} \right) - \exp(-2z_{LO}(t)) \left(1 - \frac{\nu(t)}{\nu_{LO}(t)} \right) \right] K_0(z_{LO}(t)) \right\} , \quad (61) \end{aligned}$$

with

$$\nu(t) = 1 / \left[\exp(\beta_C(t) \hbar \omega_{LO}) - 1 \right] \quad (62)$$

$$\gamma_{\alpha 0} = (2^{5/2} e E_{0\alpha} / 3) (1/2\pi m_{\alpha} \hbar \omega_{LO})^{1/2} , \quad (63)$$

and $K_n(\mathbf{z})$ are Bessel functions of the second kind¹⁶.

Simple mathematical manipulations allow us to put eq. (61) in the form given by Conwell⁵ but with the instantaneous values of $T_c(t)$ and $T_{LO}(t)$.

Next, we assume that Frölich interaction predominates over all other carrier-phonon interactions, and, since in the very early stages of relaxation after finalization of the laser pulse there is practically no heating of A phonons, we take T_{AC} equal to the reservoir temperature. Hence, the original set of six generalized transport equations reduce to four equations once those for the A- and TO-phonons' rate of energy variation are dropped. Using eqs. (49) they take the form

$$\frac{d\beta_C(t)}{dt} = -\frac{\beta^2(t)}{3} \sum_{\alpha} \frac{v_{\alpha}}{n} \frac{\dot{E}_{\alpha,LO}^{FR}}{\pi_{\alpha,LO}}(t) - \dot{E}_{\alpha,AC}^{PD}(t) - \dot{E}_{\alpha,LO}^{FR}(t) , \quad (64a)$$

$$\frac{d\beta_{LO}(t)}{dt} = \frac{2V_{cell}}{(\hbar\omega_{LO})^2} [1 - \cosh(\beta_{LO}(t)\hbar\omega_{LO})] \left\{ \sum_{\alpha} \dot{E}_{\alpha,LO}^{FR}(t) - \dot{E}_{LO,AN}(t) \right\} \quad (64b)$$

$$\frac{dv_{\alpha}}{dt} = (e/m_{\alpha})E - \gamma_{\alpha}(t)v_{\alpha}(t) . \quad (64c)$$

Eq. (64c) is a Newton-Langevin-type equation with $\gamma_{\alpha}(t)$ playing the role of the reciprocal of an instantaneous momentum relaxation time. Formally, eqs. (64c,d) are of the same form of eqs.(42) in ref. 7, but $\gamma_{\alpha}(t)$ in the latter depends on a supercorrelation function (eq.(43) in 7), a functional, with a highly complicated dependence, on all the nonequilibrium variables, including the drift velocities. Differently, $\gamma_{\alpha}(t)$ of eqs.(64c,d), calculated in the NSOM-linear theory of relaxation, depends only on $T_c(t)$ and $T_{LO}(t)$, but it is independent of $v_{\alpha}(t)$. Thus, eqs. (64c,d) are first order linear differential equations for each drift velocity, possessing the solutions

$$v_{\alpha}(t) = (e/m_{\alpha})\mathcal{E}\tau_{\alpha}(t) , \quad (65)$$

where

$$\tau_\alpha(t) = \exp(-\psi(t)) \int_0^t dt' \exp(\psi(t')) , \quad (66)$$

$$\psi_\alpha(t) = \int_0^t dt' \gamma_\alpha(t') , \quad (67)$$

and we have taken the initial condition $\mathbf{v}_\alpha(0) = 0$.

Defining the currents $\mathbf{I}_\alpha(t) = n\mathcal{E}\mathbf{v}_\alpha(t)$ and using eq. (65) we obtain a Drude-type conductivity

$$\sigma_\alpha(t) = (ne^2/m_\alpha)\tau_\alpha(t) , \quad (68)$$

with an instantaneous transport relaxation time depending on time through the quasi-temperatures $T_c(t)$ and $T_{LO}(t)$, and then varying in time with the irreversible evolution of the macrostate of the system.

The results obtained in the work of ref. 7 are **easily reverified**: at extremal points of $\mathbf{v}_\alpha(t)$, at, say, $t = t_x$, we obtain from eqs.(65) and (66) that

$$\left. \frac{d\mathbf{v}_\alpha}{dt} \right|_{t_x} = (e/m_\alpha)\mathcal{E}[1 - \gamma(t_x)\tau(t_x)] = 0 , \quad (69)$$

and so extremal values of the drift velocities should appear whenever, during their transient, there occurs a crossover of the evolution curves for the momentum relaxation time and transport relaxation time. Such extremum is a maximum or a minimum if the second time derivative at t_x ,

$$\left. \frac{d^2\mathbf{v}_\alpha}{dt^2} \right|_{t_x} = -(e/m_\alpha)\mathcal{E}\tau_\alpha(t_x) \left. \frac{d\gamma_\alpha}{dt} \right|_{t_x} = v_\alpha(t_x)\gamma_\alpha^2(t_x) \cdot \left. \frac{d\gamma_\alpha^{-1}}{dt} \right|_{t_x} , \quad (70)$$

is **negative** or positive respectively. Further,

$$\left. \frac{d\gamma_\alpha^{-1}}{dt} \right|_{t_x} = \frac{\partial\gamma_\alpha^{-1}}{\partial T_c} \Big|_{T_c} + \frac{\partial\gamma_\alpha^{-1}}{\partial T_{LO}} \cdot \left. \frac{dT_{LO}}{dt} \right|_{t_x} , \quad (71)$$

and, since the last term is expected to be much smaller than the first² neglecting it we find that on cooling ($dT_c/dt < 0$) a maximum occurs if $\partial\gamma^{-1}/\partial T_c > 0$, and a minimum for $\partial\gamma^{-1}/\partial T_c < 0$. Once $\tau(t)$ begins at zero and increases, the

first extremum, if it occurs, is a maximum, and thus a transient with structure (a maximum and a minimum) should follow if on cooling γ^{-1} passes through a minimum. It ought to be emphasized that this characteristic of the mobility transient remains valid for the quite general case of any intensity of the electric field strength and large relaxation effects, as shown in ref. 7. We recall that the expression for $\gamma(t)$ given by eq. (61) is valid for low to moderately high fields and the NSOM-linear theory of relaxation. Other properties of this structured mobility are given in ref. 7; we call the reader's attention to the fact that the maximum is an overshoot if the momentum relaxation time at t_x is larger than γ^{-1} in the steady state.

We have drawn in fig. 1 the curves for the reciprocal of the momentum relaxation time dependent on the carriers' quasi-temperature for several values of the LO-phonon quasi temperature. There, $\theta_0 = \hbar\omega_0/k$ is Einstein's temperature, and γ_α is normalized in terms of the $\gamma_{\alpha 0}$ of eq. (63). The existence can be seen of a maximum of γ (minimum of the momentum relaxation time γ^{-1}) for a quasi-temperature $T_c^{ex} \simeq 2\theta_0$, with a negative slope (or alternatively $d\gamma^{-1}/dT_c > 0$) for $T_c > T_c^{ex}$ and a positive slope (or $d\gamma^{-1}/dT_c < 0$) for $T_c < T_c^{ex}$; also $d\gamma/dT_{LO}$ is smooth.

Next, we apply these results to a specific case to obtain numerical solutions.

3. Structured mobility in GaAs

Consider a sample of GaAs illuminated by an intense pulse of laser light. To fix initial conditions we take as an example the case of the experiment of Shank et al.¹⁷ which are already used to study relaxation phenomena in HEPS². A very short laser pulse of 0.25 psec produces a density of photoinjected carriers $n = 2 \times 10^{18} \text{ cm}^{-3}$, having an excess kinetic energy of roughly 2.4 eV, and in contact with a thermal reservoir at 300 K. Immediately after the pulse the initial quasi-temperature of carriers is, roughly, 6700 K, and $T_{LO} \sim 303 \text{ K}$, $T_A \sim T_0 = 300 \text{ K}^2$. We have also used relaxation times for anharmonic processes of 10 psec and a heat diffusion relaxation time of 1 nsec.

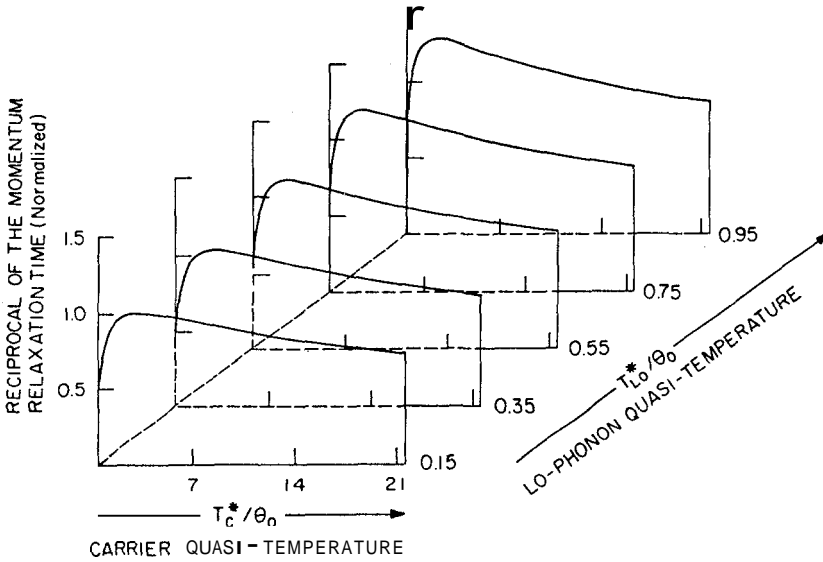


Fig.1 - Dependence of the reciprocal of the momentum relaxation time with the carrier quasi-temperature for several values of the LO-phonons quasi-temperature. The normalization factors are indicated in the main text.

The coupled set of integro-differential equations, eqs.(64), is solved using standard computational techniques. Our results are displayed in the accompanying figures. Fig. 2 shows the time evolution of the carriers' quasi-temperature for several values of the electric field. It can be noted that for $\mathcal{E} \lesssim 4 \text{ kV/cm}$ the carriers cool down to a steady state in times shorter than 5 psec. For $4 \lesssim \mathcal{E} \lesssim 9.4 \text{ kV/cm}$ the carriers cool down at a slower pace, and the steady state follows in times from 5 psec to more than 30 psec. For $\mathcal{E} \gtrsim 9.4 \text{ kV/cm}$ the steady state follows after times of more than 30 psec, and the steady-state quasi-temperature is larger than the initial one. This is a result of the fact that Joule heating effects overcome the energy relaxation of the phonon field. It is expected to occur starting at a value of the field intensity such that (cf. eq. (142)),

$$ne^2 \mathcal{E}^2 \left(\frac{\tau_e}{m_e} + \frac{\tau_h}{m_h} \right) \approx \dot{E}_{e,LO}^{FR} + \dot{E}_{h,LO}^{FR} \quad (72)$$

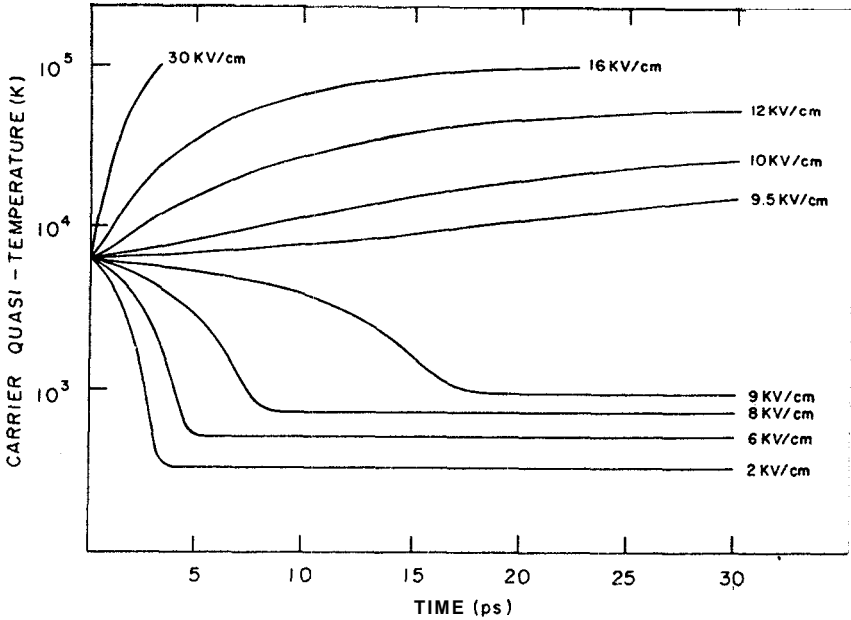


Fig.2 - Evolution of the carriers quasi-temperature for several values of the electric field intensity.

Fig. 3 shows the evolution of the LO-phonons quasi-temperature. One can note an increase in T_{LO} of less than 10% of the reservoir temperature (300 K), and the fact that for fields larger than, roughly, 9.4 kV/cm, one finds smaller values of T_{LO} with increasing fields.

Fig. 4 shows the evolution of the electron-drift velocity (almost identical curves are obtained for holes except that the ordinate scale must be reduced by a factor of, roughly, 15). Fig. 5 provides the evolution of the momentum relaxation time and transport relaxation time, and in fig. 6 we find the dependence of the stationary drift velocity with the electric field intensity.

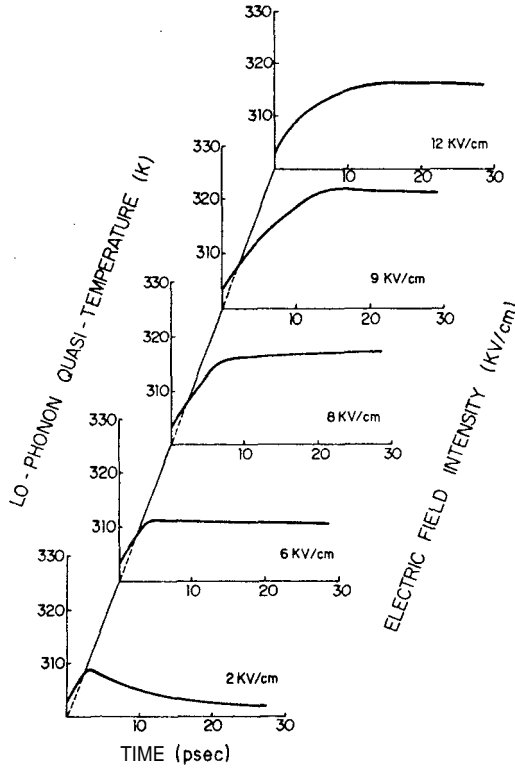


Fig.3 - Evolution of the LO-phonon quasi-temperature for several values of the electric field intensity.

Inspection of these curves tells us that the ultrafast mobility of hot carriers in the central valley of **GaAs** (and expected to be valid for HEPS in general) shows three well defined regimes.

- i) A structured mobility with a relative maximum (no overshoot) and a minimum at low electric field intensities (in our case $\mathcal{E} \lesssim 4 \text{ kV/cm}$), with the mobility in the steady state following a near Ohmic law,
- ii) A structured mobility with an absolute maximum (overshoot) and a minimum, at low to intermediate electric field ($4 \lesssim \mathcal{E} \lesssim 9.4 \text{ kV/cm}$), and non-Ohmic behavior in the stationary state,

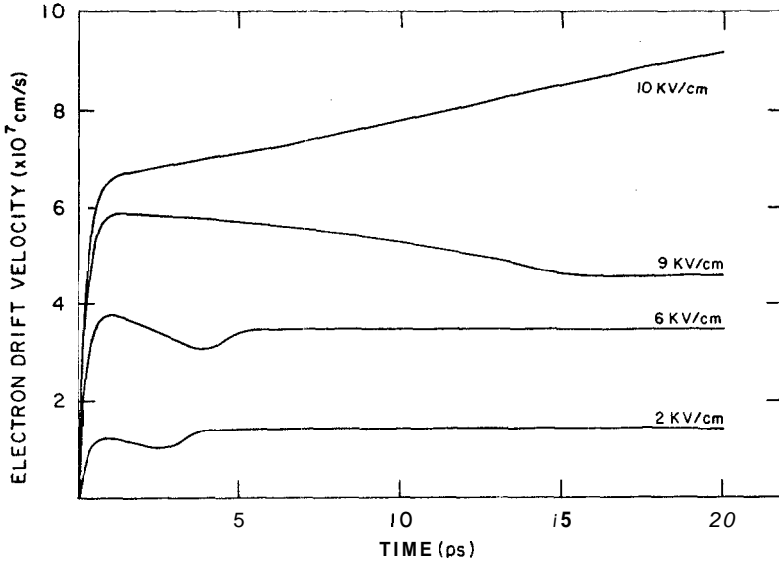


Fig.4 - Evolution of the electron drift velocity for for several values of the electric field intensity.

iii) Normal behavior, i.e. a monotonic increase of the mobility towards its stationary value, and a near Ohmic dependence of the latter.

Fig. 5 confirms the stated criterion that maxima (minima) appears when there occurs a crossover of the evolution curves of $\tau(t)$ and $\gamma^{-1}(t)$, and the latter is decreasing (increasing) at that point.

Fig. 7 shows the dependence with the electric field of the carriers' quasi-temperature in the steady state, and fig. 8 that of the electron transport relaxation time (observe that it coincides with the momentum relaxation time in the steady-state).

The differential stationary conductivity is,

$$\sigma_{diff}(E) = ne^2 \frac{d}{d\mathcal{E}} \left[E \left(\frac{\tau_e}{m_e} + \frac{\tau_h}{m_h} \right) \right] =$$

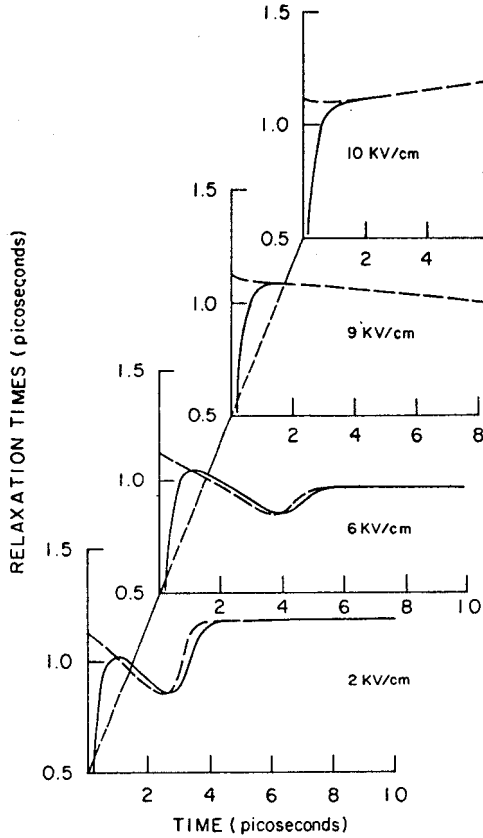


Fig.5 - Evolution of the momentum and transport relaxation times for for several values of the electric field intensity.

$$\begin{aligned}
 &= ne^2 \left(\frac{\tau_e}{m_e} + \frac{\tau_h}{m_h} \right) + (ne^2 \mathcal{E}) \frac{d}{d\mathcal{E}} \left(\frac{\tau_e}{m_e} + \frac{\tau_h}{m_h} \right) \simeq \\
 &\simeq \sigma_{\text{Drude}} + (ne^2 \mathcal{E}) \cdot \left(\frac{1}{m_e} \frac{\partial \gamma_e^{-1}}{\partial T_e} + \frac{1}{m_h} \frac{\partial \gamma_h^{-1}}{\partial T_e} \right) \frac{dT_e}{d\mathcal{E}} \quad (73)
 \end{aligned}$$

Hence, eq. (73) together with fig. 8 indicates that at fields $\mathcal{E} \lesssim 9 \text{ kV/cm}$, σ_{dif} is smaller than Drude conductivity, a very steep increase follows in the interval $9 \lesssim \mathcal{E} \lesssim 9.4 \text{ kV/cm}$, and σ_{dif} becomes larger than Drude's value.

Fig. 9 shows the dependence of the height of the maximum and the depth of

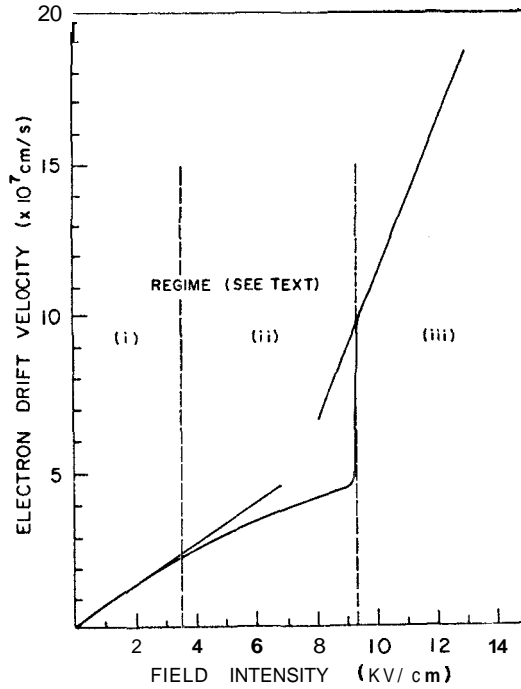


Fig.6 - Dependence of the drift velocity in the steady state with the the electric field intensity.

the minimum, relative to the stationary value (positive values of the first correspond to overshoot). Fig. 10 shows the temporal length of both kind of extrema, at half height, and fig. 11 shows the temporal localization of both maximum and minimum. One observes an (almost linear) increase of the overshoot with electric field intensity for $4 \lesssim \mathcal{E} \lesssim 9.4$ kV/cm, and the disappearance of it at this last value when the system is entering regime (iii). For $E \simeq 8$ kV/cm the overshoot is, roughly, 25% of the velocity in the stationary state. The temporal length of the overshoot also increase with the electric field intensity, but its peak value is

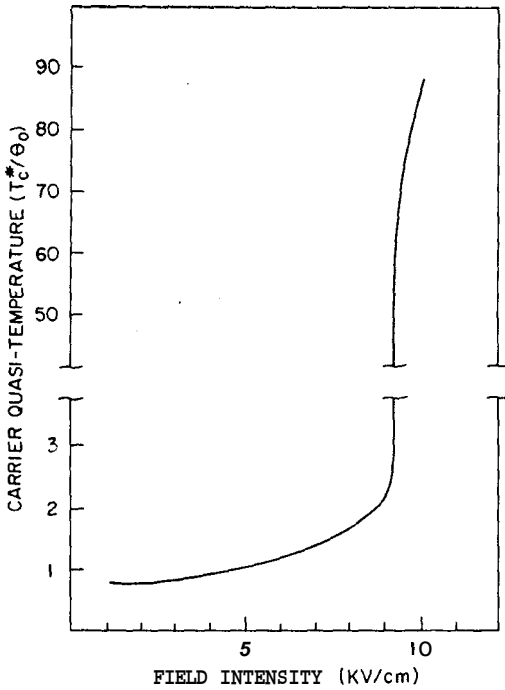


Fig.7- Dependence of the carrier quasi-temperature in the steady state with the electric field intensity.

present with almost the same delay time of, roughly, 1 psec (cf. fig. 11).

We have analyzed the effect of the initial conditions on the structured mobility transient. Fig. 12 shows the evolution of the electron drift velocity for $\mathcal{E} = 6$ kV/cm, and different values of the initial carrier quasi-temperature (i.e. increasing values of the laser frequency). At low energy transfer it follows normal behavior; with increasing energy transfer a structured mobility begins to appear, becoming more and more evident leading, at high energy transfer, to the appearance of an increasingly more pronounced overshoot. This is the result of the fact that the minimum of γ^{-1} is at near $2\theta_0$, i.e. ~ 920 K in GaAs; then, for $T_c(0) = 700$ K the carrier system evolves without γ^{-1} passing through such minimum and no

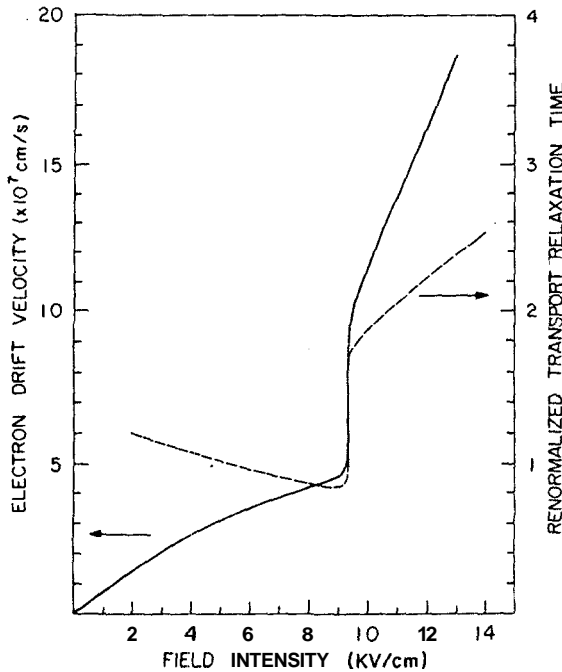


Fig.8 - Dependence of the electron drift velocity in the steady state with the electric field intensity.

structure can be produced. The subsequent values of $T_c(0)$ correspond to initial values of γ^{-1} that allow it to pass through its minimum while the carriers cool down and structure appears. With increasing values of $T_c(0)$, the initial value of γ^{-1} is up and up on the positive-slope side of the curve γ^{-1} vs T_c . Since the stationary value of the drift velocity is the same in all cases (independes of the initial conditions and being fixed only by the value of \mathcal{E} , for a certain value of $T_c(0)$ the maximum becomes an overshoot, with increasing heigth with increasing $T_c(0)$.

Finally, we note that the existence of an instantaneous transport relaxation time allows to write an instantaneous Einstein relation linking it to an instantaneous diffusion coefficient,

$$D_\alpha(t) = (kT_c(t)/m_\alpha)\tau_\alpha(t) \quad (74)$$

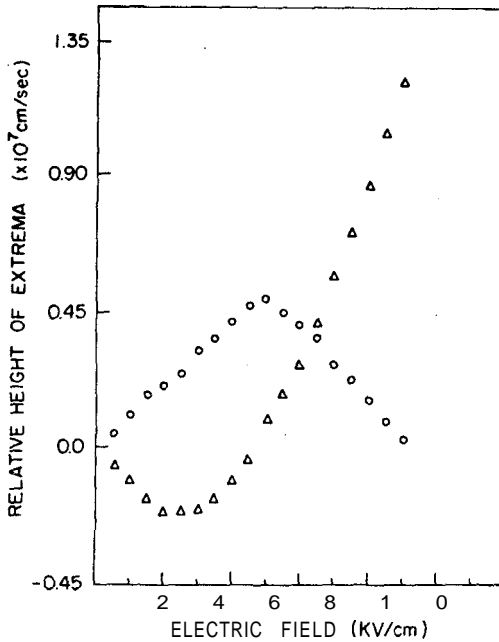


Fig. 9 - Height of extrema relative to the velocity in the steady for several values of the electric field intensity. The dip of the mininum (dot) is given in absolute value.

Because of the rapid decay of the carrier quasi-temperature in the early stages of relaxation, the structure in the transient of $D(t)$ is washed out, as shown in fig. 13, but a very pronounced diffusion overshoot is present.

4. Conclusions

We have presented a detailed analytical study of the ultrafast mobility transient of initially far-from-equilibrium carriers in highly photoexcited plasma in polar semiconductors (HEPS). For that purpose we resorted to the powerful non-linear quantum transport theory derived from the nonequilibrium statistical operator method (NSOM) in Zubarev's approach. A coupled set of nonlinear integro-

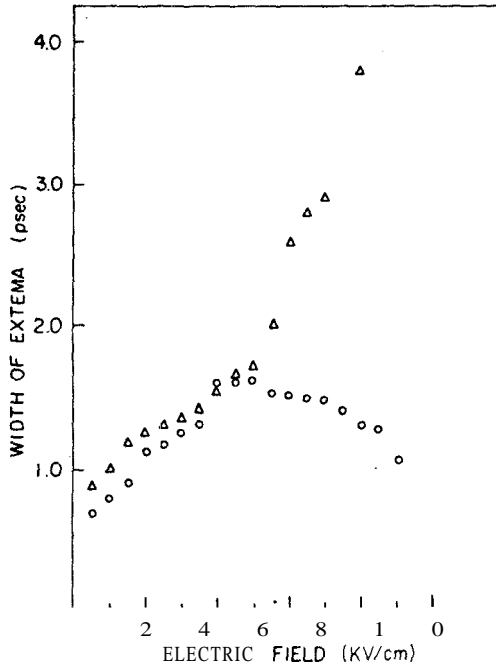


Fig. 10 - Width of the extrema (dots for the minimum, triangles for the maximum) taken at the value of the drift velocity in the steady state.

differential generalized transport equations for a basic set of nonequilibrium thermodynamic variables, deemed appropriate for the description of the macroscopic state of the HEPS, was derived. Two of these equations are those for the carriers' drift velocity (or mobility). The generalized collision operators were calculated in the NSOM-linear theory of relaxation.

We recover in this approximation the prediction that⁷, depending on how it proceeds the irreversible evolution of the nonequilibrium macrostate of the system, maxima and minima may be observed during the ultrafast mobility transient of the photoinjected carriers in the central valley of polar semiconductors. We called this previously unreported effect *structured mobility of hot carriers*. A criterion for this effect to be found is given in Section 2 which, as shown, basically depends

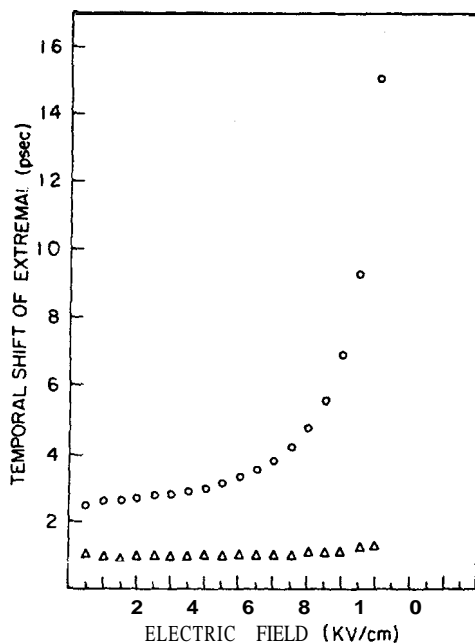


Fig. 11 - Delay time for occurrence of the minimum (dots) or maximum (triangles) after application of the electric field.

on the fact that the momentum relaxation time passes through a **minimum value** while the HEPS evolves from its initial nonequilibrium state to a stationary state for **fixed** electric field intensity.

The expressions for the collision operators we obtained, however valid for any intensity of the electric field, are quite complicated and difficult to handle. To make possible rather accessible mathematical manipulations, we restricted the numerical analysis to the case of up to moderately high field intensities, when the kinetic energy of drift of the carriers is smaller than **their** thermal energy. The coupled set of basic nonlinear generalized transport equations **was** solved for a given initial

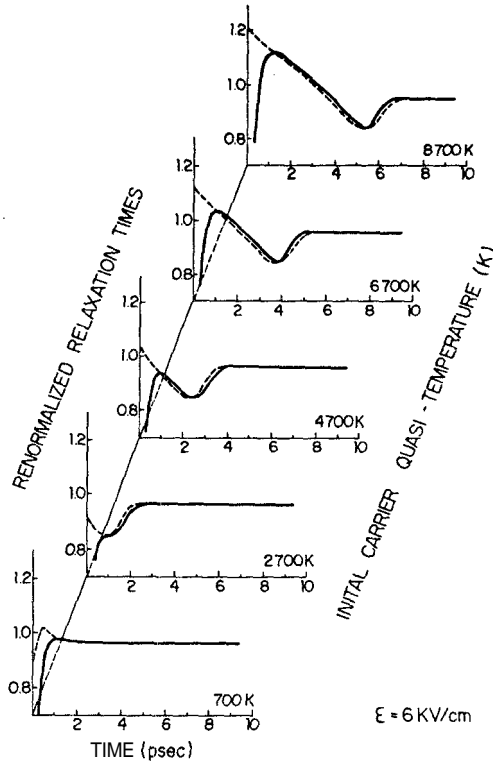


Fig. 12 - Evolution of the electron drift velocity (normalized to its value in the steady state) for an electric field of 6 kV/cm, and different values of the level of photoexcitation, characterized by the initial carrier quasi-temperatures.

condition, and the results presented in Section 2. We showed that there exist three differentiated regimes in the ultrafast mobility transient dependent on the field strength.

For a sufficiently intense electric field the carriers' system keeps heating up (or starts to cool down and next heats up) so that the momentum relaxation time does not attain its minimum and a structured mobility is excluded. Hence, there exist a maximum value of the field above which the mobility only presents normal evolution. This regime follows, on increasing field strength, from another

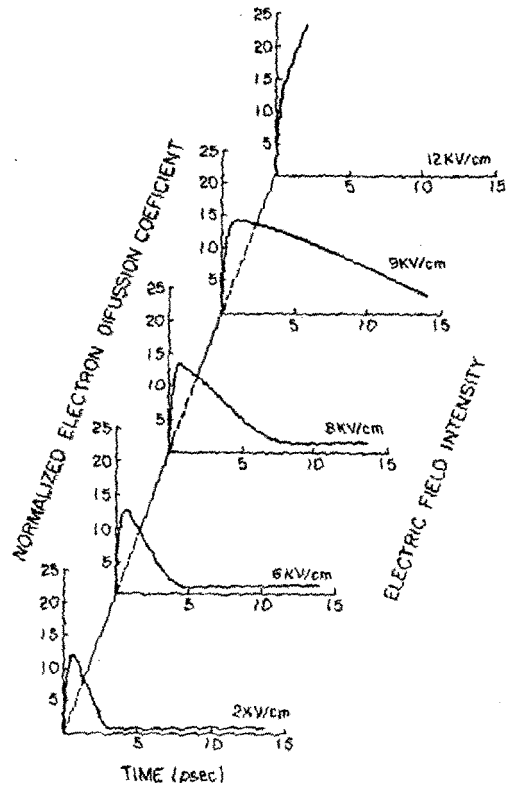


Fig. 13 - Evolution of the electron diffusion coefficient for several values of the electric field intensity.

one where structured mobility is present and the maximum is an overshoot. The height of the overshoot diminishes with decreasing field intensity and there is a lower limit of this intensity below which this maximum is no longer an overshoot. (Cf. fig. 3). The regime with overshoot is strongly correlated with the region of values of electric field intensity for which the stationary mobility is non-Ohmic (Cf. fig. 6).

Further, as shown by fig. 12, and the ensuing discussion in Section 2, there is a lower level of photon laser energy for the phenomenon to occur, viz., the

one that allows **excess** kinetic energy of the carriers to be high enough for the macroscopic state of the system to allow, at the start, the momentum relaxation time to decrease with decreasing temperature.

Experimental observation of ultrafast mobility in HEPS is scarce, and the existing few reports are not detailed **enough**^{18,19}. It may be mentioned that there is a certain qualitative and semi-quantitative agreement with **Hammond's measurements**¹⁹ in that he reports a lower and upper limit for the field intensity for overshoot to be observed. Also, it must be stressed that we have studied the **dependence** of the mobility of nonequilibrium carriers in HEPS in a single valley. However, the band structure of direct-gap polar semiconductors displays multiple valleys, and therefore intervalley scattering of carriers needs be considered. It could lead to additional structure at sufficiently high levels of excitation as a result of the transfer of carriers to higher energy valleys where they have larger effective masses, but this effect seems to be smoothed out by carrier **collision**²⁰. Quite recently, Liu et al.²¹ have applied **Zubarev's NSOM** to the study of the steady state of high-field electron transport in multi-valley semiconductors. The present paper provides an extended discussion of the results reported in ref. **22**.

This **article** is based on the **Ph.D.** Thesis of V.N. Freire, who at the time was a Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP) pre-doctoral fellow. The other two **authors** (ARV and RL) are Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq) research fellows.

References

1. R.R. Alfano, in (a) *New Techniques and Ideas in Quantum Measurement Theory*, edited by D.M. Greenberger, pp. 118-126 (The New York Academy of Sciences, New York, 1986); (b) *Biological Events Probed by Ultrafast Laser Spectroscopy*, and (c) *Semiconductor Processes Probed by Ultrafast Laser Spectroscopy*, Vols. I and II, edited by R.R. Alfano (Academic, New York, **1983** and 1984 respectively).

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2. R. Luzzi and A.R. Vasconcellos, in Ref. **1c**, Vol.I. pp. 135-169; A.C. Algarte and R. Luzzi, Phys. Rev. B27, 7563 (1983); R. Luzzi, in: *High* Excitation and Short Pulse Phenomena, edited by M.H. Pilkuhn, pp. 318-332 (North Holland, Amsterdam, 1985) and reference therein.
3. R. Zwanzig, in: Perspectives in Statistical Physics, edited by H.J. Ravechè, pp. 123-134 (North Holland, Amsterdam, 1981) and Kinam (Mexico) 3, 5 (1981).
4. A.C. Algarte, A.R. Vasconcellos, R. Luzzi, and A.J. Sampaio, Rev. Brasil. Fis. 15, 106 (1985).
5. A brief summary of the principal **results** were communicated in, V.N. Freire, A.R. Vasconcellos, and R. Luzzi, Solid State Commun. (in press), and Proc. Fifth Int. Conf. on Hot Carriers in Semiconductors, edited by J. Shah (Boston, MA, 1987) in: Solid State Electronics. Vol. 31 (Pergamon, Oxford, 1988).
6. M. Rieger, P. Kocivar, P. Bordone, P. Luigli, and L. Reggiani, Proc. Fifth Int. Conf. on Hot Carriers, edited by J. Shah, (Boston, MA, 1987) in: Solid State Electronics, Vol 31 (Pergamon, Oxford, 1988). C. Jacoboni and L. Reggiani, Rev. Mod. Phys. 55, 3 (1983); L. Reggiani, in Proc. *Fourth* Int. Conf. on Hot Carriers, edited by E. Gornik, G. Bauer, and E. Vass (Innsbruck, 1985): Physica **134B**, 123 (1985).
7. V.N. Freire, A.R. Vasconcellos, and R. Luzzi, submitted *Nuovo Cim.D*.
8. A. Elci, M.D. Scully, A.L. Smirl, and J.C. Matter, Phys. Rev. **B16**, 191 (1977); J. Collet, T. Amand, and M. Pugno, Phys. Lett. **96A**, 368 (1983).
9. E. Conwell, Solid State Phys., Suppl. 9, 1 (1967).
10. D.N. Zubarev, Fortsch. Phys. **18**, 125 (1970); Neravnoovesnaia *Statisticheskai*a Termodinamika, Ch. IV (Idz. (Nauka, Moskwa, 1971) [English Trans: Nonequilibrium Statistical *Thermodynamics*(Plenum, New York, 1974)]).
11. N.N. Bogoliubov, Lectures in Statistical Mechanics, Vol. II (Gordon and Breach, New York, 1970).
12. I. Prigogine, Nature (London) 246, 67 (1973).
13. D. Pines, Elementary Excitations in *Solids*(Benjamin, Readings, MA, 1964).

14. In Ref. **10b**, Ch. IV, Section 22.3; also **R. Luzzi**, A.R. Vasconcellos, Rev. Brasil. Fis. **16**, 495 (1986).
15. J.M. Ziman, *Electrons and Phonons* (Oxford University Press, Clarendon, **1956**).
16. M. Abramovitz and I. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1970).
17. C.V. Shank, D.H. Auston, E.P. Ippen and O. Teschke, *Solid State Commun.* **26**, 567 (1978).
18. C.V. Shank, R.L. Fork, **B.T.** Greene, F.K. Reinhardt, and R.A. Logan, *Appl. Phys. Lett.* **38**, 104 (1981).
19. **R.B.** Hammond, *Physica* **134B**, 475 (1985).
20. M.A. Osman, U. Ravaioli, and D.K. Ferry, in *High Speed Electronics*, edited by **B. Kallback** and **H. Beneking** (Springer, Berlin-Heidelberg, 1986).
21. M. Liu, D.Y. Xing, C.S. Ting, and W.T. Xu, *Phys. Rev.* **37**, 21997 (1988).
22. V.N. Freire, **A.R.** Vasconcellos, and R. Luzzi, *Phys. Rev.* **B39**, 13264 (1989).

Resumo

São usados o Método do Operador Estatístico de Não Equilíbrio e a teoria não-linear **quântica** de transporte que tem associado, como descrito em Rev. Bras. Fis. **15**, 106 (1985); *ibid* **16**, 495 (1986), para realizar um estudo analítico da mobilidade transiente ultra-rápida de portadores fotoinjectados no vale central de um semiconductor polar de gap direto. Obtemos expressões para a mobilidade resolvida temporalmente destes portadores quentes quando submetidos a condições gerais de fotoexcitação e intensidade de campo elétrico. Resultados numéricos são apresentados no caso de campos fracos até moderadamente fortes, e uma **comparação** qualitativa com experimentos disponíveis é feita. **É** mostrado que a mobilidade transiente apresenta uma estrutura composta de um máximo e um **mínimo** antes de ser atingido o estado estacionário. As características desta estrutura dependem do nível de fotoexcitação **é** da intensidade do campo elétrico. **É** apresentada uma breve discussão sobre o coeficiente de difusão dos portadores.