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Kinetics of Screening in Optically Excited Semiconductors

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Kinetic equations for the density correlation and density–density response function are derived. Solving these equations numerically and comparing its solution with the adiabatic approximation the change in the transmitted spectrum of a single electronic transition is calculated where a red-shift can be observed.

Es werden kinetische Gleichungen für die Dichtekorrelationsfunktion und die Dichte–Dichte-Responsefunktion abgeleitet. Die numerische Lösung dieser Gleichungen wird mit der adiabatischen Approximation verglichen. Die Berechnung der Änderung im Transmissionspektrum für einen einzelnen elektronischen Übergang ergibt eine Rotverschiebung.

1. Introduction

In the Boltzmann equation screening is usually calculated with the actual one-particle distribution function. Under short-pulse excitation in semiconductors this adiabatic concept is expected to fail, and so we here present first steps towards a kinetics of screening in non-equilibrium systems. We focus on a two-band situation in a semiconductor with optically excited carriers. In this system the kinetics of screening is connected with the question: How fast does the gap shrinkage react to the build-up of charge density? Clearly it means to ask for the relevant time scale for building up longitudinal excitations like plasmons or LO phonons and their distribution if density is excited in a semiconductor by an external field and furthermore to ask for the value of the gap shift which causes changes in the transmission spectrum due to the non-equilibrium excitations.

The layout of the paper is as follows: After deriving kinetic equations for the density correlation function and for the density–density response function within the Keldysh formalism in Section 2 we summarize in Section 3 the equilibrium properties. In Section 4 we compare in a simple case the numerically exact solution of these equations with the adiabatic approximation and calculate the change in the transmitted intensity for a single electronic transition.

2. The Longitudinal Propagator for Non-Equilibrium Systems

The definition of the longitudinal propagator within the Keldysh framework and variational derivative technique is [1]

$$D(I, I') = \frac{\delta \langle U(I) \rangle}{\delta \varrho^{\text{ext}}(I')}, \quad (1)$$

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where $I = q_1, t_1, \eta_1$ and $\eta_1 = \pm 1$ denotes the double time contour [2]. $U(I)$ is the longitudinal field which obeys Poisson's equation

$$U(I) = v(q_1) [\varrho(I) + \varrho^{\text{ext}}(I)] \quad (2)$$

with the Coulomb potential $v(q_1)$ and the induced charge density $\varrho(I)$. Introducing the polarization function

$$P(I, I') = \frac{\delta \langle \varrho(I) \rangle}{\delta \langle U(I') \rangle}, \quad (3)$$

we obtain from (2) the equation for the longitudinal propagator for a homogeneous and isotropic situation ($q = |q|$)

$$D(qt_1t'_1) - v(q) \int dt_2 P(qt_1t_2) D(qt_2t'_1) = v(q) \delta(t_1 - t'_1). \quad (4)$$

To make the definition (1) of the longitudinal propagator more transparent we decompose the Keldysh matrix according to [2] (q is dropped in the following)

$$D(t_1t_2) = \begin{bmatrix} D^+(t_1t_2) + D^<(t_1t_2) & -D^<(t_1t_2) \\ D^>(t_1t_2) & D^+(t_1t_2) - D^>(t_1t_2) \end{bmatrix}, \quad (5)$$

where the density correlation functions

$$\begin{aligned} D^<(t_1t'_1) &= -\frac{i}{\hbar} [\langle U(t_1) U(t'_1) \rangle - \langle U(t_1) \rangle \langle U(t'_1) \rangle], \\ D^>(t_1t'_1) &= -\frac{i}{\hbar} [\langle U(t'_1) U(t_1) \rangle - \langle U(t_1) \rangle \langle U(t'_1) \rangle] \end{aligned} \quad (6)$$

and the retarded response function

$$D^+(t_1t'_1) = \theta(t_1 - t'_1) [D^>(t_1t'_1) - D^<(t_1t'_1)] + v(q) \delta(t_1 - t'_1) \quad (7)$$

were introduced. Sometimes it is convenient to work with the advanced function

$$D^-(t_1t'_1) = -\theta(t'_1 - t_1) [D^>(t_1t'_1) - D^<(t_1t'_1)] + v(q) \delta(t_1 - t'_1). \quad (8)$$

The four fundamental functions in (6) to (7) are connected by the Keldysh relation

$$D^>(t_1t'_1) - D^<(t_1t'_1) = D^+(t_1t'_1) - D^-(t_1t'_1) \quad (9)$$

and fulfil the ϵ -symmetry relations

$$\begin{aligned} D^<(t_1t'_1) &= D^>(t'_1t_1), \\ D^<(t_1t'_1) &= -D^{<*}(t'_1t_1), \\ D^{+*}(t_1t'_1) &= D^-(t'_1t_1) = D^+(t_1t'_1). \end{aligned} \quad (10)$$

As we will see $D^<$ describes the distribution of the longitudinal excitations, whereas the imaginary part of the Fourier transform of D^+ accounts for the spectral behaviour. For D^+ and $D^<$ we obtain from (4) the following equations:

$$D^+(qt_1t'_1) - v(q) \int_{t'_1}^{t_1} dt_2 P^+(qt_1t_2) D^+(qt_2t'_1) = v(q) \delta(t_1 - t'_1), \quad (11)$$

$$D^<(qt_1t'_1) = \int_{-\infty}^{t_1} dt_2 \int_{-\infty}^{t'_1} dt_3 D^+(qt_1t_2) P^<(qt_2t_3) D^-(qt_3t'_1), \quad (12)$$

where the polarization functions P^+ and $P^<$ are introduced according to (5). Restricting ourselves to the electron–electron interaction in the random phase approximation and with the one-particle Green's function [3],

$$\begin{aligned} G^<(kt_1t'_1) &= \frac{i}{\hbar} \langle \psi^+(kt'_1) \psi(kt_1) \rangle, \\ G^>(kt_1t'_1) &= -\frac{i}{\hbar} \langle \psi(kt_1) \psi^+(kt'_1) \rangle, \\ G^+(kt_1t'_1) &= \theta(t_1 - t'_1) [G^>(kt_1t'_1) - G^<(kt_1t'_1)], \end{aligned} \quad (13)$$

we obtain from (3)

$$\begin{aligned} P^{\leq}(qt_1t'_1) &= -i\hbar \sum_k G^{\leq}(q+k, t_1t'_1) G^{\geq}(kt'_1t_1), \\ P^+(qt_1t'_1) &= \theta(t_1 - t'_1) [P^>(qt_1t'_1) - P^<(qt_1t'_1)]. \end{aligned} \quad (14)$$

The polarization function (14) exhibits the same symmetries as the longitudinal propagator (10).

3. Equilibrium Situation

Before solving the kinetic equations (11) and (12) in the general non-equilibrium case we will summarize some results for equilibrium. In the latter all quantities depend on the time difference only and we can perform the Fourier transformation with respect to it ending up with

$$D^+(q\omega) = \frac{v(q)}{1 - v(q)P^+(q\omega)} = v(q)\varepsilon^{+1}(q\omega) = V_s^+(q\omega), \quad (15)$$

$$D^{\leq}(q\omega) = D^+(q\omega)P^{\leq}(q\omega)D^-(q\omega) = P^{\leq}(q\omega) \frac{\text{Im } D^+(q\omega)}{\text{Im } P^+(q\omega)}. \quad (16)$$

The last expression follows from (9). So D^+ is the usual dynamically screened Coulomb potential where ε^+ is the dielectric function. For pure electron–electron interaction $\text{Im } D^+$ has a peak at the plasma frequency $\pm\omega_p(q)$.

For the interpretation of $D^<(q\omega)$ it is useful to introduce a distribution function $N(q\omega)$ for the longitudinal excitations according to [2]

$$D^<(q\omega) = 2iN(q\omega) \text{Im } D^+(q\omega). \quad (17)$$

Then from (16) follows

$$N(q\omega) = \frac{P^<(q\omega)}{P^>(q\omega) - P^<(q\omega)}; \quad P^<(q\omega) = 2iN(q\omega) \text{Im } P^+(q\omega). \quad (18)$$

In the equilibrium situation the one-particle Green's function obeys the Kubo–Martin–Schwinger relation [4]

$$G^<(k\omega) = -e^{-\beta(\hbar\omega - \mu)} G^>(k\omega), \quad (19)$$

where μ is the chemical potential and $\beta = 1/kT$. So we recover from (14) the well-known relation

$$P^>(q\omega) = e^{\beta h\omega} P^<(q\omega) \quad (20)$$

which holds, however, quite generally.

Then for the distribution one yields from (18) the well-known Bose function to a given temperature

$$N(\omega) = \frac{1}{e^{\beta h\omega} - 1}. \quad (21)$$

4. Non-Equilibrium Excitations

In the general case of non-equilibrium we introduce as customary central and relative coordinates according to

$$T = \frac{t_1 + t'_1}{2}, \quad t = t_1 - t'_1, \quad (22)$$

$$D(qt_1 t'_1) \Rightarrow D(qtT). \quad (23)$$

From (11) and (12) then the kinetic equations follow,

$$D^+(qtT) - v(q) \int_0^t dt' P^+ \left(q, t - t', T + \frac{t'}{2} \right) D^+ \left(q, t', T - \frac{t - t'}{2} \right) = v(q) \delta(t), \quad (24)$$

$$D^<(qtT) = \int_0^\infty d\bar{t} \int_0^\infty dt' D^+ \left(q, \bar{t}, T + \frac{t - \bar{t}}{2} \right) P^< \left(q, t + t' - \bar{t}, T - \frac{t' + \bar{t}}{2} \right) \\ \times D^+ \left(q, t', T - \frac{t + t'}{2} \right). \quad (25)$$

Now in the non-equilibrium theory it is usual to make the fundamental assumption that all functions in (24) and (25) vary slowly in their central time T on a microscopic scale which is determined by the relative time t . Then one can linearize (24) and (25) with respect to the central time derivative. This concept is used e.g. in plasma physics [2], but it works well only at the plasma dispersion $\omega = \omega_p(q)$ if damping is not too large. Additionally, this assumption is expected to fail for short pulse excitation. To avoid this problem we shall strictly solve the integral equation numerically in the time domain. For this we have specified first the polarization function in (24) and (25). Corresponding to (17) we make for the Fourier transform of the one-particle Green's function $G^<(k\omega)$ in (14) the ansatz

$$G^<(k\omega) = 2if(k\omega) A(k\omega), \quad (26) \\ G^>(k\omega) = -2i(1 - f(k\omega)) A(k\omega)$$

with the spectral function A and the particle distribution f which is the solution of a general Boltzmann equation. At the moment we are not able to solve the one-particle kinetics and the screening kinetics self-consistently and therefore we use for f a Boltzmann distribution with total density following from integrating a Gaussian excitation light pulse. For the spectral function $A(k\omega)$ we assume a Lorentzian with constant damping γ which converts

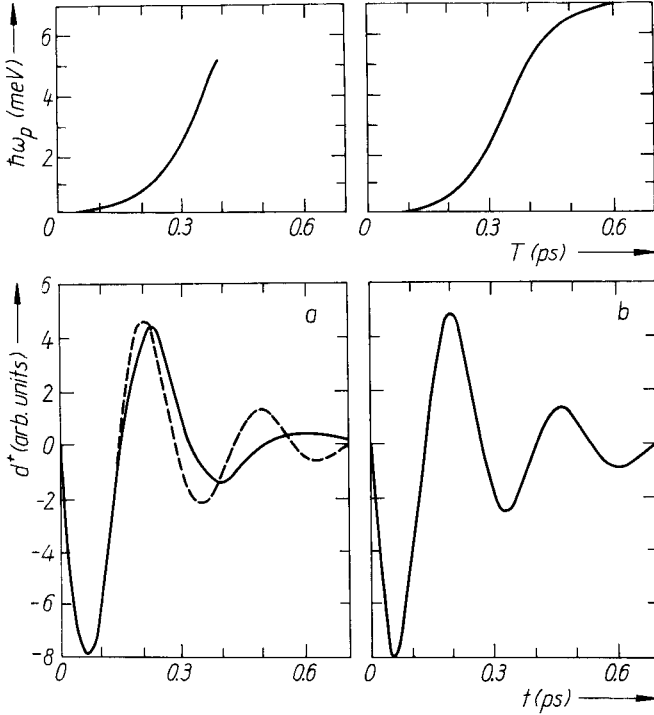


Fig. 1. Density response function d^+ in dependence on relative time t for zero wave vector in the adiabatic approximation (dashed curve) and exact solution (full curve). The parameters are: one-particle damping $\gamma = 0.75 \text{ ps}^{-1}$, $T = 77 \text{ K}$, density rise time 0.5 ps . a) Non-stationary situation with an actual plasma frequency $\hbar\omega_p = 5 \text{ meV}$ (see upper figure), b) stationary situation with $\hbar\omega_p = 7 \text{ meV}$ (final density)

in the time domain into

$$A(kt) = \frac{1}{2} e^{-iE_c(k)t - \gamma|t|}. \quad (27)$$

This gives for the polarization function, in the low density limit,

$$P^<(\eta t T) = \frac{1}{i\hbar} N(T) \exp\left(i\eta t - 2\gamma|t| - \frac{\eta}{\hbar\beta} t^2\right), \quad (28)$$

$$P^+(\eta t T) = -\frac{2}{\hbar} N(T) \theta(t) \sin \eta t \exp\left(-2\gamma t - \frac{\eta}{\hbar\beta} t^2\right), \quad (29)$$

where η is

$$\eta = \frac{\hbar}{2m} q^2 \quad (30)$$

and $N(T)$ is the excited density.

Due to the time convolution in (24) and (25) we expect memory effects which means e.g. that the plasma frequency ω_p does not correspond to the actual excited density in the system. In order to illustrate this fact we will compare the numerically exact solution of the kinetic equations with the adiabatic approximation which means that the system feels only the actual excited density independent of the density history. Mathematically in the

adiabatic limit from (24) and (25) follows

$$D_{\text{ad}}^+(qtT) - v(q) \int_0^t dt' P^+(q, t - t', T) D_{\text{ad}}^+(qt'T) = v(q) \delta(t), \quad (31)$$

$$D_{\text{ad}}^-(qtT) = \int_0^\infty dt' \int_0^\infty dt'' D_{\text{ad}}^+(qt'T) P^-(q, t + t' - t'', T) D_{\text{ad}}^+(qt''T). \quad (32)$$

In the case of zero wave vector ($q = 0$) we can solve the adiabatic equations (31) and (32) analytically ending up with

$$d_{\text{ad}}^+(\eta = 0, tT) = -\theta(t) \omega_p(T) \sin(\omega_p(T) t) e^{-2\eta t}, \quad (33)$$

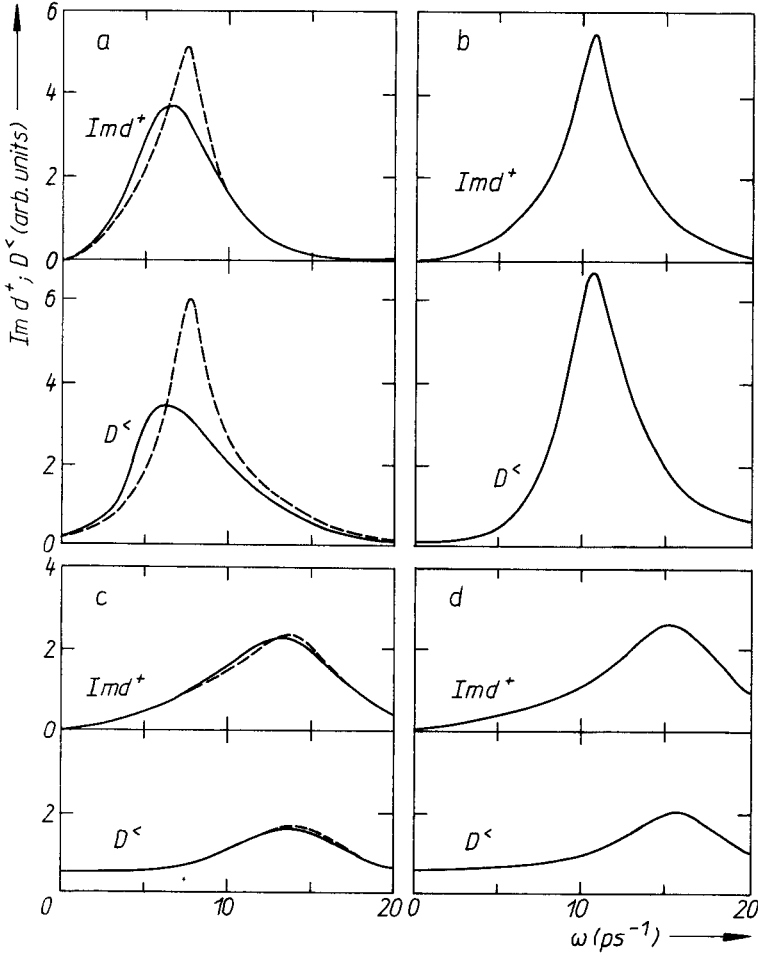


Fig. 2. The imaginary part of the response function $d^+(\omega)$ and the correlation function $D^<(\omega)$ for different wave vectors in the non-stationary (left-hand side) and in the stationary situation (right-hand side) (dashed curves adiabatic approximation). The parameters are the same as in Fig. 1. a) $\eta = 0, \hbar\omega_p = 5 \text{ meV}$; b) $\eta = 0, \hbar\omega_p = 7 \text{ meV}$ (final density); c) $\eta = 2 \text{ ps}^{-1}, \hbar\omega_p = 5 \text{ meV}$; d) $\eta = 2 \text{ ps}^{-1}, \hbar\omega_p = 7 \text{ meV}$ (final density)

where $D^+ = \nu\delta + \nu\delta^+$ and $\omega_p^2(T) = e^2N(T)/m$. So the response function performs damped oscillations with a period depending on the plasma frequency. For two "snapshots" T in Fig. 1 $d^+(\eta = 0, tT)$ is depicted in the adiabatic limit and for the numerically exact solution following from (24) and (25). There is a definite difference between the oscillation periods for the exact solution and for the adiabatic approximation, respectively.

In Fig. 1b stationarity is already reached and no deviation between both curves can be seen. This will become more clear if we consider the Fourier transform with respect to the relative time, shown in Fig. 2. In the general case the plasma frequency lags behind the adiabatic approximation until the stationary situation is reached. This is due to the smaller density values at earlier stages. For larger wave vectors there is not much difference between both curves due to the fact that plasmons are strongly damped in the electron excitation continuum.

However, can these tiny deviations from the adiabatic approximation be detected? To have a rough impression we consider the self-energy shift of a simple electronic transition. For this we will calculate the transmitted intensity [5, 6] according to

$$I(\omega T_D) = \text{Im} \int dt' e^{-i\omega(T_D-t')} P_{cv}(t'), \quad (34)$$

where the spectrum was probed by an extreme broad-band test beam

$$E_t = 2\pi E_0 \delta(t - T_D) \quad (35)$$

with a delay time T_D with respect to the exciting pulse. P_{cv} is the test-beam induced polarization. The change in the one-particle Green's function linear in the excited density is $\delta G_c = G_c \circ DG_c \circ G_c$, which follows from integrating Dyson's equation (\circ means time convolution).

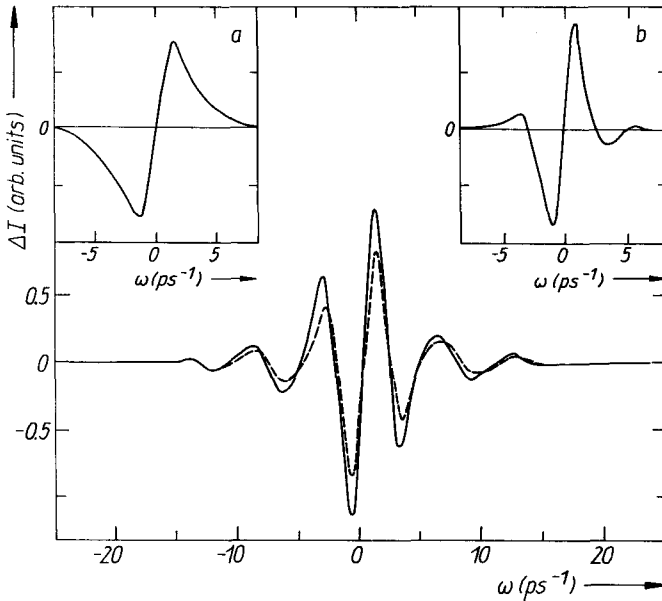


Fig. 3. The change of the transmitted intensity with the same parameters as in Fig. 1 for zero delay time (dashed curve adiabatic approximation); zero of frequency scale is $E_c - E_v$. Inset a: stationary situation, delay $T_D = 1$ ps, inset b: long-pulse excitation (density rise time 1.0 ps) for zero delay

The differential transmission spectrum between pump-on and pump-off considering only a single electronic transition is then given by

$$\Delta I(\omega T_D) = \text{Im } i \sum_q \int_0^\infty dt e^{i(\omega - E_c + E_v)t - 2\gamma t} \int_0^t dt' \int_0^t dt'' D^<(q, t' - t'', T_D + \frac{t' + t''}{2}).$$

Numerical results are shown in Fig. 3. In the stationary situation (inset a) one can observe a red shift due to the negative value of the self-energy. For medium rise time of the density (inset b) we find side structures, so-called coherent oscillations, which are found in any transmission experiment with rapidly changing optical constants. To see the non-adiabatic behaviour we have to use shorter light pulses. Then we obtain in the adiabatic approximation a smaller gap shift as for the exact solution and so the adiabatic approximation fails. However, there might be problems to detect the rapidly changing self-energy shift because the coherent oscillations tend to blur the observation [7].

5. Conclusion and Outlook

We have derived kinetic equations for the density correlation function and for the density-density response function within the Keldysh framework. Via the polarization function there is a coupling to the one-particle kinetics, but for numerical simplicity we have assumed an equilibrium one-particle distribution function. The treatment of one-particle and screening kinetics self-consistently has to be done in future. In contrast to the concepts used previously where kinetic equations were solved only linear in the central time derivative we have calculated the equations in the whole time domain numerically. The comparison with the adiabatic approximation has shown that memory effects will become important if the rise time of the one-particle density is shorter than a few hundred fs. In the differential transmission spectrum one can observe a stronger red-shift as in the adiabatic approximation but coherent oscillations make the observation difficult.

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