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Electron relaxation in the conduction band of wide-band-gap oxides

Ph. Daguzan, P. Martin, S. Guizard, and G. Petite
Commissariat à l'Energie Atomique, Département de Recherche sur l'Etat Condensé, les Atomes et les Molécules,
Service de Recherche sur les Surfaces et l'Irradiation de la Matière, 91191 Gif sur Yvette, France
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Using quantum kinetic equations with memory we calculate the time-dependent distribution function of an electron gas injected suddenly in the conduction band of a wide-band-gap insulator. A comparison is made with the semiclassical Boltzmann equation approach. We show that due to the large electron-phonon coupling occurring in such materials a quantum approach is needed.

I. INTRODUCTION

Ultrashort and intense laser pulses available for a few years have motivated numerous studies on the very first events following the injection of electrons in the conduction band of wide-band-gap insulators on a subpicosecond time scale.[1-3] The way the excited material goes back to its fundamental state is of primary interest because its detailed study can give very fruitful information on the amount of energy deposited in the material, a key point for understanding optical breakdown,[4] or on the defect formation mechanism. In particular, the self-trapped exciton is considered as a precursor of permanent defects such as the well-known E' center.[5,6] This is well established in SiO_2 (Refs. 7 and 8) and also in ionic crystals.[9] In all cases, this type of defect is a manifestation of a strong electron-phonon coupling. Other evidence of this strong coupling is the direct observation of the laser heating of free electrons in the conduction band of α quartz[10,11] and the very short trapping time into deep levels lying in the band gap.[1,2] In all cases, the knowledge of the time-dependent electron distribution function is very useful because the way the electron gas thermalizes with the phonon bath is a major key in understanding such phenomena. Furthermore, the direct time-dependent observation of this distribution function is now possible with pump-probe-type experiments. Such measurements have been recently performed in semiconductors.[12-15] These experimental works provide a very sensitive test for the various theories describing the energy relaxation of the electron gas, going from Monte Carlo simulations to the most sophisticated many-body formalism. Quantum calculations, i.e., including memory effects, are now well developed for typical semiconductors.[16-23] In insulators, the situation is less advanced probably because of the lack of precise data about material parameters, such as detailed knowledge of band structure. Carrier dynamics in insulators has been mainly studied in order to understand dielectric and optical breakdown, which are problems of great importance both in fundamental physics and in technology. Various approaches have emerged[24-27] but since the beginning of the 1980s, Monte Carlo methods have had authority. Such simulations have provided relevant results about electron transport in SiO_2, and especially about hot-electron phenomena.[28-34] However, the major problem with this type of description is that it is based on the fundamental concept of scattering rates. This approach, because it is based on the Fermi golden rule, excludes any collision broadening related to the energy-time uncertainty principle, and as a consequence is justified only if the time elapsed between two collisions is (at least) longer than the time necessary for the energy conservation to take place. If not, it seems unrealistic to consider the whole process as a series of sequential elementary events. In semiconductors, it has been recently shown that such a collisional broadening can modify the electron distribution function. Therefore, because of a much higher electron-phonon coupling in materials like wide-band-gap insulators, this regime is expected to have more spectacular effects. The semiclassical Monte Carlo method has been modified in order to address this issue, including the effect of collisional broadening[32] or using a Feynman path-integral approach.[29,35] Thus a time-dependent calculation, as in semiconductors, now becomes necessary to take into account the non-Markovian behavior of the system.

In the present work, we address the problem of the relaxation of an electron gas when injected at low kinetic energy in the conduction band of a wide-band-gap insulator, for example, by an ultrashort laser pulse. The case of SiO_2 has been chosen as an example. We start with a given energy distribution of the electrons and neglect electron-electron interaction (low excitation density limit) in a one-band model context. Our goal is to focus on the kinetic aspects of the electron-phonon interaction. Two approaches are compared: in Sec. II, we present a calculation based on the direct integration of the semiclassical Boltzmann equation. We include explicitly the coupling of the electrons with two longitudinal optical phonon branches and the coupling with acoustical phonons. The interaction with acoustic phonons can be considered as elastic because the energies involved are very small. We will insist on the angular aspects of the problem showing that the acoustic phonons make the distribution isotropic almost instantaneously. Section III is devoted to a more sophisticated approach based on quantum kinetic equations. Our approach is similar to the one developed in Ref. 23. In contrast with Sec. II, the problem is simplified by the assumption (justified in II) that the distribution is isotropic, and we neglect the action of the

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electron-acoustical phonon coupling, since we are mainly interested in energy relaxation. The comparison between the two approaches will be commented in Sec. IV before the conclusion.

II. SEMICLASSICAL BOLTZMANN EQUATION APPROACH

The electron distribution function $f_{\mathbf{k}}$ is governed by the Boltzmann equation, which reads for a homogeneous system without external field,

$$\frac{\partial f_{\mathbf{k}}}{\partial t}(t)=\frac{\Omega}{(2\pi)^3}\int d\mathbf{k}'W(\mathbf{k}',\mathbf{k})f_{\mathbf{k}'}(t)[1-f_{\mathbf{k}}(t)] \\ -\frac{\Omega}{(2\pi)^3}\int d\mathbf{k}'W(\mathbf{k},\mathbf{k}')f_{\mathbf{k}}(t)[1-f_{\mathbf{k}'}(t)] \,. \qquad (1)$$

Ω is the crystal volume and $W(\mathbf{k},\mathbf{k}')$ is the probability per unit time (s^{-1}) for an electron to be scattered from state $\mathbf{k}$ to state $\mathbf{k}'$ because of its interaction with the phonons, evaluated from first-order perturbation theory with the Fermi golden rule,

$$W_{\mp}(\mathbf{k},\mathbf{k}')=\frac{2\pi}{\hbar}|\langle\mathbf{k}'|H_{e\text{-ph}}|\mathbf{k}\rangle|^2\delta(E_{\mathbf{k}'}-E_{\mathbf{k}}\mp\hbar\omega_{\mathbf{q}}) \,. \qquad (2)$$

The upper sign corresponds to the absorption of one phonon and the lower to the emission of one phonon. $H_{e\text{-ph}}$ stands for the electron-phonon interaction Hamiltonian.

SiO_2 is taken as a numerical example. We consider the main longitudinal optical (LO) phonon branches (150 and 60 meV) and the interaction with electrons is treated with the Fröhlich Hamiltonian. Only one spherical and parabolic band is considered, which is justified for the first eV's of the conduction band of SiO_2.[36] Taking the dispersion relation as constant, the transition rate can be written as[37]

$$W_{\mp}(\mathbf{k},\mathbf{k}')=\frac{2\pi}{\hbar}\frac{\alpha}{q^2}[n(\omega_{\mathrm{LO}})+\tfrac{1}{2}\mp\tfrac{1}{2}]\delta(\mathbf{k}'-\mathbf{k}\mp\mathbf{q}) \\ \times\delta(E_{k'}-E_k\mp\hbar\omega_{\mathrm{LO}}) \,, \qquad (3)$$

where $\mathbf{q}$ is the phonon momentum, $\hbar\omega_{\mathrm{LO}}$ the phonon energy, and $n(\omega_{\mathrm{LO}})$ is the phonon occupation number (Bose-Einstein statistics). The coupling constant α is given by

$$\alpha=\frac{e^2\hbar\omega_{\mathrm{LO}}}{2\Omega\varepsilon_0\varepsilon_P}, \quad \frac{1}{\varepsilon_P}=\left[\frac{1}{\varepsilon_\infty}-\frac{1}{\varepsilon_S}\right] . \qquad (4)$$

e is the electron charge, and ε_∞ and ε_S stand, respectively, for the optical and static dielectric constants. $1/\varepsilon_P=0.143$ and 0.063, respectively, for the 150- and 60-meV LO phonon modes.[24]

Electron-acoustical phonon collisions are also considered and treated as elastic and isotropic. Using a deformation potential approach, the scattering rates read[37]

$$W_{\mp}(\mathbf{k},\mathbf{k}')=\frac{\pi C^2q^2}{\rho\omega(q)\Omega}[n(q)+\tfrac{1}{2}\mp\tfrac{1}{2}] \\ \times\delta(E_{\mathbf{k}'}-E_{\mathbf{k}})\delta(\mathbf{k}'-\mathbf{k}\mp\mathbf{q}) \,. \qquad (5)$$

C is the deformation potential [$C=6$ eV (Ref. 38)] and ρ is the density, $\rho=2.6$ g/cm^3. The phonon dispersion has been approximated as[38,39]

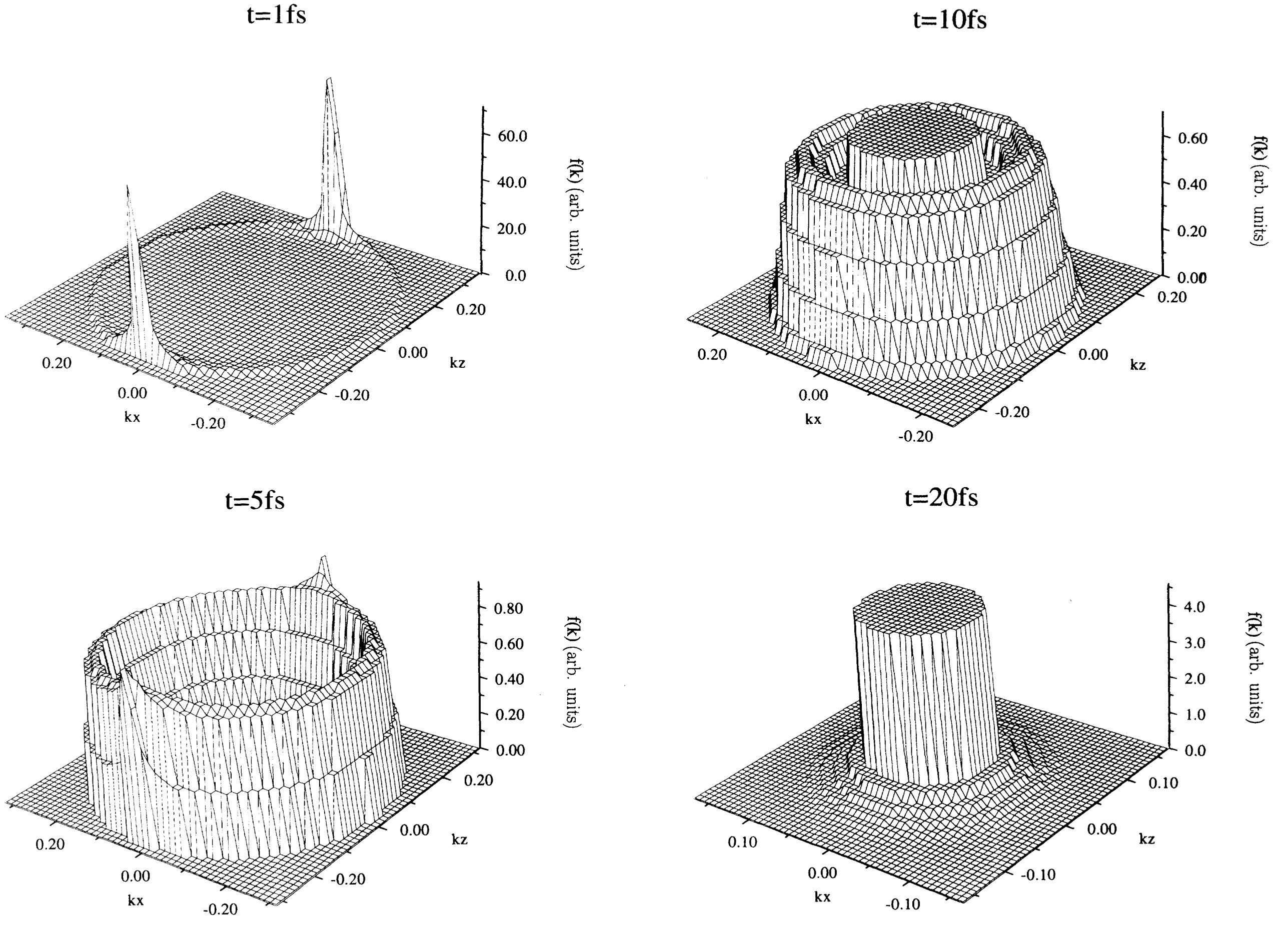
$$q<k_{\mathrm{BZ}}:\omega(q)=\frac{2}{\pi}v_sq, \quad n(q)\cong\frac{k_BT}{\hbar v_sq} \,, \\ q>k_{BZ}:\omega(q)=\frac{2}{\pi}v_sk_{\mathrm{BZ}}, \quad n(q)=n(k_{BZ}) \,, \qquad (6)$$

where k_{BZ} is the wave vector at the edge of the Brillouin zone, defined by $E_{\mathrm{BZ}}=\hbar^2k_{\mathrm{BZ}}^2/2m_e=5.5$ eV,[36] with the effective electron mass $m_e=0.5m_0$, where m_0 is the free electron mass. v_s is the sound velocity averaged over the longitudinal and transverse polarizations, $v_s=4.6\times10^5$ cm/s.[32]

Inserting the different scattering rates into Eq. (1), one obtains an integro-differential equation for each vector $\mathbf{k}$. This equation is numerically solved for each time with a fifth-order adaptive step-size Runge-Kutta procedure.

The results of this calculation are shown in Fig. 1. For design clarity, only the most energetic LO phonon branch is included. We suppose that the electrons are injected in the conduction band at $t=0$, with an energy of 1.2 eV above the bottom of the conduction band, in two opposite directions lying along kz. This condition could be a simplified representation of the initial distribution following photoexcitation with a laser polarized along kz. So the electron distribution function in the k space is highly anisotropic and is composed of two peaks. The interactions with LO phonons and acoustical phonons are turned on, and the relaxation process can take place. We observe an ultrafast (<5 fs) isotropization of the distribution function. This is mainly due to the action of the acoustical phonons. In fact, due to the Coulombic nature of the interaction with the polar LO phonons, the phonon momentum $\mathbf{q}$ appears in the denominator of (3), and thus the collisions mainly take place in the forward direction. However, in the case of acoustical phonons (5), the deformation potential approach takes the interaction as proportional to the relative displacement of ions from one unit cell to another, and thus proportional to the modulus of $\mathbf{q}$, so the large-angle collisions are favored. Therefore, collisions with acoustical phonons make the electron distribution function rapidly isotropic. From an experimental point of view, we can note that this isotropization time is of the same order, and even smaller, than the injection time in the conduction band by an ultrashort laser pulse.

The distribution function is composed of steps. This is due to the fact that electrons lose their energy-emitting LO phonons, so the energy-exchange amount is fixed (150 meV). We observe that the distribution function reaches very rapidly the bottom of the conduction band. Indeed, each electron suffers a very high number of collisions. The total rate calculated with Eq. (3) gives typically a number of the order of 1 fs^{-1}. Each collision involves an energy of 150 meV, corresponding to the optical phonon branch. In such a case, it is not surprising that the time necessary to reach the bottom of the band, starting from 1.2 eV, is of the order of a few tens of fs. We will go back to this point later.



t=1fs

t=10fs

t=5fs

t=20fs

FIG. 1. Electron distribution function in k space for various time delays after the initial conditions: 1, 5, 10, and 20 fs.

Figure 2 shows the evolution of the number of electrons $[f(E,t)\rho(E)]$ as a function of the energy and time $[\rho(E)$ being the density of states in the conduction band]. For the sake of simplicity and because the main action of the acoustical phonons is to make the distribution isotropic very rapidly, we neglect such interactions and take the distribution function as isotropic. The electron density is 10^{15} cm^{-3} and the thermostat temperature is 300 K (nondegenerate case). We have included the two optical phonon branches. The discrete structure corresponding to the phonon replicas is clearly observed. The contributions of the two different phonon branches can be easily separated, especially for short times. When the time increases, the two branch replicas can no longer be clearly identified and all the linear combinations of the two modes are observed. If now the collisions with acoustic phonons were not supposed elastic, supplementary energy exchanges would take place. They are small but continuous and they would tend to smooth the energy spectrum, but the phonon replicas would still be observable for short times.

III. QUANTUM DYNAMIC APPROACH

The quantum description of transport has been greatly improved during the last few years, especially for the semiconductor case.[40] Different models have emerged, using nonequilibrium Green's functions[22,41] and more re-

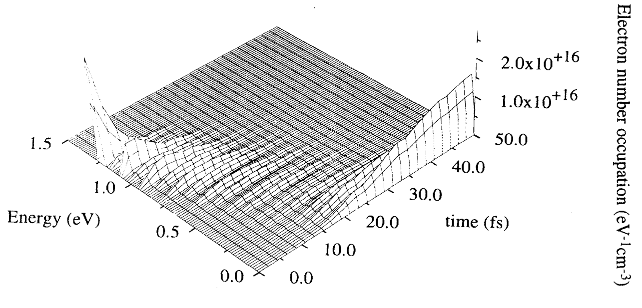


FIG. 2. Number of electrons as a function of energy for different time delays in the semiclassical approach.

cently equations of motion of single-particle density matrix.[17–20,23] The Hamiltonian describing the system of noninteracting electrons, phonons, and their coupling is written,

$$H=H_e+H_{\text{ph}}+H_{e\text{-ph}} . \tag{7}$$

We adopt the standard second quantification notation,

$$H_e+H_{\text{ph}}=\sum_{\mathbf{k}}\frac{\hbar^2k^2}{2m_e}c_{\mathbf{k}}^\dagger c_{\mathbf{k}}+\sum_{\mathbf{q}}\hbar\omega_{\mathbf{q}}b_{\mathbf{q}}^\dagger b_{\mathbf{q}} . \tag{8}$$

m_e is the electronic effective mass (we use as in Sec. II a parabolic band approximation), and $c_{\mathbf{k}}^\dagger$ ($c_{\mathbf{k}}$) and $b_{\mathbf{q}}^\dagger$ ($b_{\mathbf{q}}$) denote creation (annihilation) operators for electrons and phonons.

The electron-phonon interaction is represented by the standard Hamiltonian,

$$H_{e\text{-ph}}=\sum_{\mathbf{k},\mathbf{q}}g_{\mathbf{q}}c_{\mathbf{k}+\mathbf{q}}^\dagger c_{\mathbf{k}}(b_{\mathbf{q}}-b_{-\mathbf{q}}^\dagger) . \tag{9}$$

In the case of the Fröhlich Hamiltonian, $g_{\mathbf{q}}$ reads explicitly

$$g_{\mathbf{q}}=-\frac{i}{q}\sqrt{\alpha} . \tag{10}$$

The physical quantity of interest is the distribution function for the electrons, which is defined as $f_{\mathbf{k}}=\langle c_{\mathbf{k}}^\dagger c_{\mathbf{k}}\rangle$. In our calculation, we neglected the variation of the number of phonons $[(d/dt)n_{\mathbf{q}}=(d/dt)\langle b_{\mathbf{q}}^\dagger b_{\mathbf{q}}\rangle]$ of the heat bath which can cause a significant heating of the lattice. This is consistent with the low electronic density limit we consider here.

We start directly with a single-particle density matrix by considering the equation of motion of the distribution in the Heisenberg representation,

$$\frac{d}{dt}\langle c_{\mathbf{k}}^\dagger c_{\mathbf{k}}\rangle=\frac{1}{i\hbar}\langle[c_{\mathbf{k}}^\dagger c_{\mathbf{k}},H]\rangle . \tag{11}$$

The calculation of the right-hand side of the above equation is straightforward and gives the result

$$\frac{df_{\mathbf{k}}}{dt}=2\,\mathrm{Re}\left[\sum_{\mathbf{q}}(s_{\mathbf{k}+\mathbf{q},\mathbf{k}}-s_{\mathbf{k},\mathbf{k}-\mathbf{q}})\right] , \tag{12}$$

where $s_{\mathbf{k}+\mathbf{q},\mathbf{k}}=(i/\hbar)g_{\mathbf{q}}\langle c_{\mathbf{k}+\mathbf{q}}^\dagger b_{\mathbf{q}}c_{\mathbf{k}}\rangle$ are the so-called phonon-assisted density matrix, introduced by Zimmermann.[20,42] Equation (12) is exact. However, as explained in Ref. (23), Eq. (12) is not suitable to compute the evolution of $f_{\mathbf{k}}$ because the expectation value of such three-body operators is unknown. A first-order contribution could be obtained by factorizing the expectation values, as in the random-phase approximation.[43] But, when neglecting coherent phonon states, the single expectation value $\langle b_{\mathbf{q}}\rangle$ vanishes and, as a consequence the assisted density matrix does too. The idea is to compare the time evolution of the three-body operator $s_{\mathbf{k}+\mathbf{q},\mathbf{k}}$. Doing this, the three-body operator evolution depends on four-body operators, and an infinite set of differential equations is obtained. One needs to introduce suitable approximations in terms of a nonvanishing factorization of expectation values to truncate this process. Note that this method is the analog of the well-known Bogolivbov-Born-Green-Kirkwood-Yvon hierarchy used in the kinetic theory of gases.[44] The time derivative of $s_{\mathbf{k}+\mathbf{q},\mathbf{k}}$ is evaluated from the motion equation

$$\frac{d}{dt}s_{\mathbf{k}+\mathbf{q},\mathbf{k}}=\frac{1}{i\hbar}\left\langle\left[\frac{i}{\hbar}g_{\mathbf{q}}c_{\mathbf{k}+\mathbf{q}}^\dagger b_{\mathbf{q}}c_{\mathbf{k}},H\right]\right\rangle . \tag{13}$$

After a long but straightforward calculation one obtains, by using the approximation $\langle c_{\mathbf{k}+\mathbf{q}-\mathbf{q}'}^\dagger c_{\mathbf{k}}b_{\mathbf{q}'}^\dagger b_{\mathbf{q}}\rangle=f_{\mathbf{k}}n_{\mathbf{q}}\,\delta_{q,q'}$, the following expression for the evolution of assisted density matrix:

$$\frac{ds_{\mathbf{k}+\mathbf{q},\mathbf{k}}}{dt}=i\frac{E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega_{\text{LO}}}{\hbar}s_{\mathbf{k}+\mathbf{q},\mathbf{k}}+\frac{1}{\hbar^2}|g_{\mathbf{q}}|^2\{f_{\mathbf{k}+\mathbf{q}}(1-f_{\mathbf{k}})(n_{\mathbf{q}}+1)-f_{\mathbf{k}}(1-f_{\mathbf{k}+\mathbf{q}})n_{\mathbf{q}}\} . \tag{14}$$

At this level of approximation, (12) and (14) form a closed set of differential equations, which can be numerically solved. Without entering into details, note that the above set of equations reduce to the semiclassical Boltzmann equation if a Markov process and an adiabatic hypothesis are supposed as shown in Ref. 19. However, we found, as mentioned by other authors,[21,45] for some initial conditions, unphysical results such as distribution functions negative or larger than unity. It has been shown recently[45] that such a difficulty can be avoided by including higher-order contributions in the hierarchy mentioned above. Without repeating the explicit calculation given in Ref. 23, the above set of equations must be transformed by including a supplementary quantity in the first term on the right-hand side of (14) as follows:

$$(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega_{\text{LO}})/\hbar\mapsto(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}-\hbar\omega_{\text{LO}})/\hbar+i\Gamma_{\mathbf{k}+\mathbf{q}}+i\Gamma_{\mathbf{k}} , \tag{15}$$

where

$$\Gamma_{\mathbf{k}}=(\Gamma_{\mathbf{k}}^{\text{in}}+\Gamma_{\mathbf{k}}^{\text{out}})/2 \tag{16}$$

and

$$\Gamma_{\mathbf{k}}^{\text{in}}=\frac{2\pi}{\hbar}\sum_{\mathbf{q}}|g_{\mathbf{q}}|^2\delta(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}\pm\hbar\omega_{\text{LO}})f_{\mathbf{k}+\mathbf{q}}(n_{\mathbf{q}}+\tfrac{1}{2}\mp\tfrac{1}{2}) , \tag{17}$$

$$\Gamma_{\mathbf{k}}^{\text{out}}=\frac{2\pi}{\hbar}\sum_{\mathbf{q}}|g_{\mathbf{q}}|^2\delta(E_{\mathbf{k}+\mathbf{q}}-E_{\mathbf{k}}\pm\hbar\omega_{\text{LO}})(1-f_{\mathbf{k}+\mathbf{q}})(n_{\mathbf{q}}+\tfrac{1}{2}\mp\tfrac{1}{2}) . \tag{18}$$

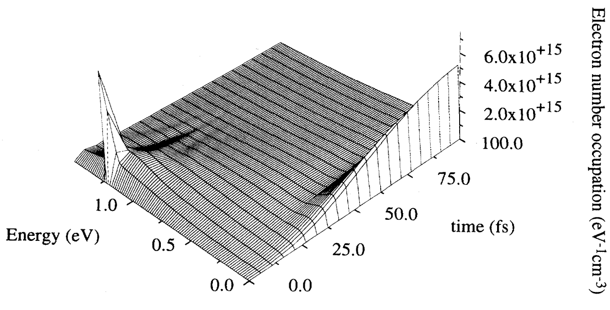


FIG. 3. Number of electrons as a function of energy for different time delays in the density matrix approach.

The above set of rate equations acts as a damping and is equivalent to assigning an intrinsic broadening to energy levels.

We present in Fig. 3 the evolution of the number of electrons as a function of the energy and time including the two LO phonon modes. In contrast with Fig. 2, the phonon replicas have disappeared. This is a consequence of the time-energy uncertainty principle. This point will be discussed in detail in the next section.

Figure 4 shows the evolutions of the average energies of the distribution functions for one (150 meV) and two (150 and 60 meV) LO phonon modes in the semiclassical case (dotted) and in the quantum case (full). Concerning the semiclassical calculation, note that the final energy in the two-phonon-mode case is lower (40 meV) than in the one-mode case (95 meV). These values must be compared with the "theoretical" result which is 39 meV ($\frac{3}{2}kT$) for a Maxwell-Boltzmann distribution. Therefore, the final distributions deviate from the unexpected equilibrium distribution, but including a supplementary scattering mechanism can give a better result. In fact, it is well known[42] that the model under consideration is too simple to reach the right final distribution (Fermi-Dirac or Maxwell-Boltzmann). One has to take into account a more realistic dispersion relation for the phonons or add other scattering processes. This is consistent with our results. The quantum calculation presents the same behavior with respect to the number of branches included in the calculation as in the semiclassical one. However, as we will discuss in the next section, we note that the time necessary for a stabilization of the energy is much longer than in the Boltzmann equation treatment case.

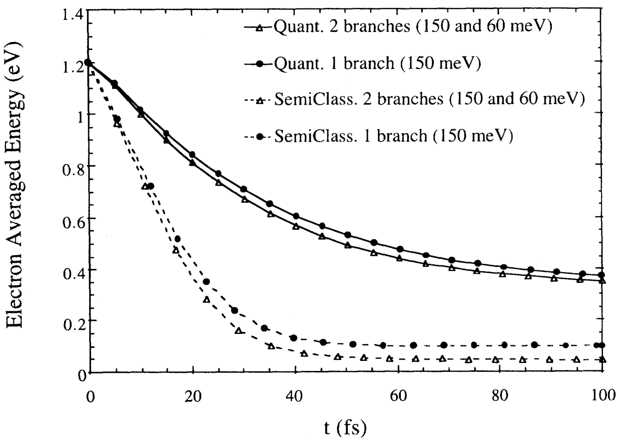


FIG. 4. Comparison between semiclassical and quantum approaches of the averaged electron energy as a function of time.

IV. DISCUSSION

The differences between the spectra shown in Figs. 2 and 3 are striking. As already mentioned, in the quantum simulation, all traces of phonon replicas are lost. Semiclassical methods are based on the concept of collision rate (s^{-1}). As is well known, these rates are obtained by using the Fermi golden rule, in which energy conservation takes place. The problem is solved by assuming that the relaxation process is a series of independent events (Markov process) excluding any memory effect. This approach is correct if the time elapsed between two collisions is much larger than the time necessary to build up the energy conservation in the golden rule which can be estimated from the time-energy uncertainty principle. For example, in the case of GaAs and by considering the case of LO phonons ($\omega_{\mathrm{LO}}=36$ meV), this time is found to be of the order of 20 fs. On the other hand, the time between two collisions calculated with Eq. (3) is of the order of 150 fs. As shown in Ref. 23 in this particular case, the electron spectrum is broadened but phonon peaks are energetically resolved, as we found too. The case of wide-band-gap insulators such as quartz is more dramatic. Considering the 150-meV LO phonon, the associated time is of 5 fs, while the time between two successive collisions for a kinetic energy of 1 eV is of the order of 2 fs. In such a case, the energy broadening is larger than the phonon energy and the discrete structure is unresolved. This shows that it is incorrect to consider the relaxation process as a series of elementary events and that a quantum calculation including memory effects is necessary to describe the evolution of the electron population.

The behavior of the mean kinetic energy of the electron is different in the semiclassical case and in the quantum case. The energy relaxation rate is slower in the latter case. This can be understood by using the general argument that an electronic system is in an equilibrium state when all the memory of its initial state is lost. In the approach based on the Boltzmann equation, the Markovian description of the temporal evolution is that the system has no memory at all. In such a case, it is easy to understand that it is able to reach its equilibrium state faster than in the approach based on the density matrix formulation which contains, at least partially, memory. From a more technical point of view, note that these contrasted behaviors can be attributed to the particular form of the Fröhlich Hamiltonian and especially to the fact that it contains a $1/q$ divergence coming from the Coulombic origin of the electron-LO-phonon interaction.

In the quantum case, transitions involving an energy exchange close to zero are favored in contrast with the semiclassical case where the exchange is fixed.[16] One must be careful about the long time behavior, because the final energy value can be strongly influenced by the choice of the model (idealized scattering mechanisms, no interaction between carriers, effects of the hierarchy truncature, initial conditions, etc.). As is well known, the passage from a kinetic regime to a thermodynamic equilibrium is a very hard physical problem. However, we observe that for times about 100 fs, there is a strong difference between the semiclassical calculation and the quantum one. The amount of deposited energy in the material differs significantly from one case to the other. Even if this difference has to be more precisely studied, it is a real fact. This can have important physical consequences in the interaction between femtosecond laser pulses and the material, especially for a precise description of the mechanism of optical breakdown,[4] or for the ultrafast carrier trapping and defect formation in wide-band-gap oxides.[1,2]

V. CONCLUSIONS

Two different approaches based, respectively, on the semiclassical Boltzmann equation and a density matrix formulation have been used in order to calculate the temporal evolution of an electron gas injected suddenly in the conduction band of a wide-band-gap insulator. We show that the strength of the electron-phonon coupling in such materials makes the spectra smooth without any phonon replicas and this is in contrast with semiconductors like GaAs. In agreement with other authors, we find that the thermalization rate is slower in the density matrix formulation case than the semiclassical case. This is certainly true. However, let us point out that our calculation neglects all effects related to carrier-carrier interactions and that the choice of the initial condition (instantaneous promotion of the electron gas in the conduction band) is, even in the case of ultrashort laser pulses, unrealistic. The role of multiphonon processes should also be investigated.[16,46] Furthermore, note that the fact that the infinite hierarchy has been truncated at second order is equivalent to a loss of information on the system we consider here. This can probably influence the relaxation time of the electronic density. More experimental works are needed to clarify the applicability range of such a model.

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