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Semiconductor Kadanoff-Baym Equation Results for Optically Excited Electron–Hole Plasmas in Quantum Wells

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We present results from solutions of the semiconductor Kadanoff-Baym equations (full two-time semiconductor Bloch equations) with self-energies in quasistatic Born approximation, for GaAs single quantum wells. We concentrate on memory and correlation effects under fs-pulse excitation conditions. A remarkable feature is the observed kinetic energy increase which is due to the build-up of correlations among the generated carriers. We demonstrate that the two-time approach is (i) very well suited to study correlation phenomena both on short and long times, thereby avoiding well-known problems of one-time kinetic equations, and (ii) that it is becoming practical with the use of efficient integration techniques.

1. Introduction

Electron-electron correlations have been recognized to be important for a quantitative description of short time-scale optical response of semiconductors. Recently, important aspects, such as the consistent treatment of polarization scattering (including off-diagonal terms in the dephasing matrix) [1 to 4], dynamical screening (including vertex corrections) [2,3], and strong coupling effects [5] have been investigated. A second group of carrier correlation effects are related to non-Markovian phenomena such as incomplete collisions, retardation and the build-up of screening [6 to 13].

While the first group of effects are well understood, the practical relevance of the second is still an open question. Previous calculations [11 to 13] have shown rather weak signatures of memory effects in Coulomb scattering, despite the (theoretically) obvious failure of Markovian equations on a femtosecond time scale. Unfortunately, conclusive experiments on this scale are very difficult, in particular due to the unavoidable large spectral width of the optical pulse. There is, however, another aspect of non-Markovian behavior which predicts these effects to persist also on a long time scale. It has been pointed out that memory effects are due to the build-up of correlations in a system of interacting particles [12]. As a result of these correlations, the system does not relax towards a Fermi function (as in the ideal gas case) but towards a correlated (momentum) distribution which may be easier to verify experimentally.

For a systematic theoretical analysis of these problems in optically excited semiconductors, the interband Kadanoff-Baym equations (KBE) for the two-time correlation

functions [14] or, equivalently, the Keldysh nonequilibrium Green function formalism [15], appear to be the most promising tool. We will motivate this choice in Section 2. In this paper, we directly solve the Kadanoff-Baym equations for the (interband and intraband) two-time correlation functions for a two-band quantum well system, including all Coulomb scattering effects on the level of the quasi-static Born approximation, Section 3. The most striking result is the build up of Coulomb correlations, leading to an additional heating of the carriers. In Section 4 we provide some details of our numerical scheme.

2. The Interband Kadanoff-Baym Equations

Memory effects are mostly studied in the frame of non-Markovian quantum kinetic equations for the (one-time) Wigner functions. The derivation of these equations from the KBE involves a number of approximations, such as the generalized Kadanoff-Baym ansatz (GKBA) [16], assumptions for the spectral function and its damping, quasiparticle approximations etc., which are difficult to control and which lead to a significant amount of inconsistency. Among the problems are violation of total energy conservation, wrong equilibrium solutions, phenomenological ingredients, e.g. for the damping coefficients. These problems are well recognized and would certainly be avoided if the KBE could be solved efficiently. We show below that this is in fact possible not only on supercomputers [11]. The main advantages of the KB approach are that (i) the full (two-time) dynamical information in the nonequilibrium spectral function is retained without any approximation, (ii) total energy (kinetic + potential) is conserved during the whole evolution, (iii) the relaxation leads to the correct correlated equilibrium state, (iv) the model is intrinsically consistent, because the level of approximation is determined by a single quantity, the self-energy, and (v) two-particle properties (e.g. correlation energy) are very conveniently computed from one-particle (but two-time) quantities. Of course, the price we have to pay is the limitation to rather simple situations.

The KBE [14] govern the evolution of two-time, nonequilibrium correlation functions

$$G^{<}_{\mu_1\mu_2}(\mathbf{k}t_1t_2) = \frac{i}{\hbar}\,\langle \psi^{\dagger}_{\mu_2\mathbf{k}}(t_2)\,\psi_{\mu_1\mathbf{k}}(t_1)\rangle\,, \qquad G^{>}_{\mu_1\mu_2}(\mathbf{k}t_1t_2) = -\frac{i}{\hbar}\langle \psi_{\mu_1\mathbf{k}}(t_1)\psi^{\dagger}_{\mu_2\mathbf{k}}(t_2)\rangle\,,$$

where $\psi(\psi^{\dagger})$ are Heisenberg-picture electron annihilation(creation) operators and the bracket $\langle\ldots\rangle$ denotes averaging with the initial ($t_1 = t_2 = -\infty$) density operator. In general nonequilibrium situations, these correlation functions depend on both time arguments, not just the time difference, as would be the case in equilibrium situations. The correlation functions also carry a pair of band indices μ_1,μ_2, each of which runs through all the energy bands under consideration. Kinetic quantities such as carrier distributions and interband polarizations are given by the time-diagonal part: for example, in a two-band model with one conduction band (μ = c) and one valence band (μ = v),

$$f_{e\mathbf{k}}(t) = -i\hbar G^{<}_{\mathrm{cc}}(\mathbf{k}tt)\,, \qquad f_{h\mathbf{k}}(t) = i\hbar G^{>}_{\mathrm{vv}}(-\mathbf{k}tt)\,, \qquad P_{\mathbf{k}}(t) = -i\hbar G^{<}_{\mathrm{cv}}(\mathbf{k}tt)\,. \tag{1}$$

Correlations are contained in the time off-diagonal components of $G^{<}$. Each correlation function evolves with respect to both time arguments according to a pair of Kadanoff-

Baym equations, which are Hermitian conjugates of each other,

$$\left[i\hbar\,\frac{\partial}{\partial t_1}-\varepsilon_{\mu_1}(\mathbf{k})\right]G^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}t_1t_2)=\sum_{\bar{\mu}}\hbar\Omega_{\mu_1\bar{\mu}}(\mathbf{k}t_1)\,G^{\gtrless}_{\bar{\mu}\mu_2}(\mathbf{k}t_1t_2)+I^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}t_1t_2)\,,$$
$$\left[-i\hbar\,\frac{\partial}{\partial t_2}-\varepsilon_{\mu_2}(\mathbf{k})\right]G^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}t_1t_2)=\sum_{\bar{\mu}}G^{\gtrless}_{\mu_1\bar{\mu}}(\mathbf{k}t_1t_2)\,\hbar\Omega_{\bar{\mu}\mu_2}(\mathbf{k}t_2)-I^{\gtrless *}_{\mu_2\mu_1}(\mathbf{k}t_2t_1)\,. \tag{2}$$

Here, $\varepsilon_\mu(k)$ are the one-particle energies which, in the following, are assumed to be parabolic with effective masses m_e and m_h and band gap E_g. The effective Rabi energy $\hbar\Omega$ contains the dipole energy $d_{\mu_1\mu_2}E(t)$ due to the external light field $E(t)$ and the Coulomb (Hartree-Fock) renormalization

$$\hbar\Omega_{\mu_1\mu_2}(\mathbf{k}t)=-d_{\mu_1\mu_2}E(t)\,(1-\delta_{\mu_1\mu_2})+i\hbar\sum_{\mathbf{k}'}G^{<}_{\mu_1\mu_2}(\mathbf{k}'tt)\,V(\mathbf{k}-\mathbf{k}')\,. \tag{3}$$

The Coulomb potential $V(k-k')$ is, in the following, assumed to be a statically screened 2D potential. The collision integrals are given by

$$I^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}t_1t_2)=\sum_{\bar{\mu}}\int_{-\infty}^{t_1}\mathrm{d}\bar{t}[\boldsymbol{\Sigma}^{>}_{\mu_1\bar{\mu}}(\mathbf{k}t_1\bar{t})-\boldsymbol{\Sigma}^{<}_{\mu_1\bar{\mu}}(\mathbf{k}t_1\bar{t})]\,G^{\gtrless}_{\bar{\mu}\mu_2}(\mathbf{k}\bar{t}t_2)$$
$$-\sum_{\bar{\mu}}\int_{-\infty}^{t_2}\mathrm{d}\bar{t}\boldsymbol{\Sigma}^{\gtrless}_{\mu_1\bar{\mu}}(\mathbf{k}t_1\bar{t})\,[G^{>}_{\bar{\mu}\mu_2}(\mathbf{k}\bar{t}t_2)-G^{<}_{\bar{\mu}\mu_2}(\mathbf{k}\bar{t}t_2)]\,. \tag{4}$$

Here, $\boldsymbol{\Sigma}^{\gtrless}$ are the self-energies of the electrons in the medium, which, in the exact theory, are related to the two-electron Green functions, whose evolution, in turn, is driven by higher order correlation functions. An important advantage of the KB approach is that there exist systematic diagrammatic tools to derive approximations for $\boldsymbol{\Sigma}^{\gtrless}$. Here, we use the quasi-statically screened direct Born approximation,

$$\boldsymbol{\Sigma}^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}t_1t_2)=i\hbar\sum_{\mathbf{k}'}V(\mathbf{k}'-\mathbf{k},t_1)\,V(\mathbf{k}'-\mathbf{k},t_2)\,\Pi^{\gtrless}(\mathbf{k}-\mathbf{k}',t_1t_2)\,G^{\gtrless}_{\mu_1\mu_2}(\mathbf{k}'t_1t_2)\,, \tag{5}$$

where the polarization propagators $\Pi^{\gtrless}$ are

$$\Pi^{\gtrless}(\mathbf{q},t_1t_2)=-2i\hbar\sum_{\mathbf{k}'\lambda\mu}G^{\gtrless}_{\mu\lambda}(\mathbf{k}'+\mathbf{q},t_1t_2)\;G^{\lessgtr}_{\lambda\mu}(\mathbf{k}',t_2t_1)\,, \tag{6}$$

containing all carrier–carrier and carrier–interband polarization (diagonal and off-diagonal with respect to k) contributions. Equations (2) to (4) are solved for the correlation functions $G^{\gtrless}_{\mu_1\mu_2}$ without further approximations.

3. Results and Discussions

In order to investigate the effect of correlations, we have studied a GaAs single quantum well (two-band model) with moderate excitation density $n\approx 2\times10^{11}$ cm^{-2}. The optical pulse is chosen to have a duration of 50 fs, a peak amplitude at time $t=0$ corresponding to $d_\mathrm{cv}E(t=0)=10$ meV and a center frequency ω_0 corresponding to $\hbar\omega_0$ being 10 meV (case I) or 25 meV (case II) above E_g, respectively. In the calculation, $E_\mathrm{g}=1.5$ eV, and

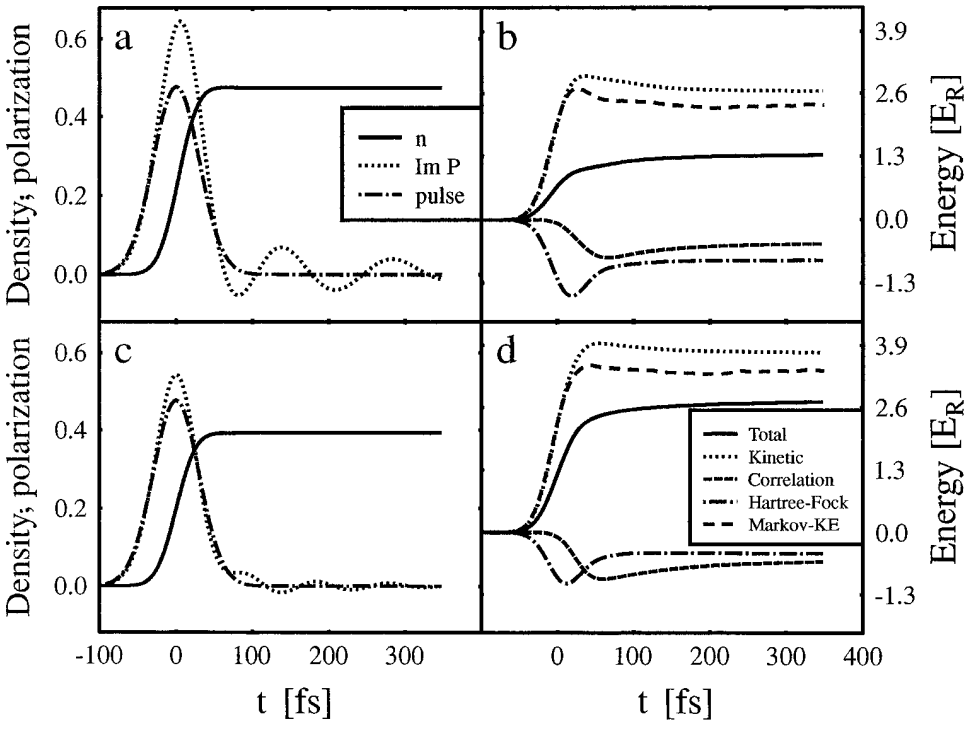

Fig. 1. a), c) Time evolution of density and polarization $P(t)$ and b), d) various energy contribution (summed over both bands). The calculations are for a GaAs single quantum well with an excess laser energy of 10 meV (parts a, b) and 25 meV (parts c, d) above E_g, respectively. Density and polarization are in units of a_B^{-2}, where a_B is the 3D exciton Bohr radius, and $E_R = 4.2$ meV

the background dielectric constant ε_0 entering the Coulomb potential is 13. The interband KBE were solved numerically using a time step of 2.5 fs and a momentum space grid of 0.4 a_B^{-1} ($a_B = 132$ Å), with $k_{max} = 9.2 a_B^{-1}$. These parameters allow for an accurate solution (density and energy conservation within 1% [1])) if the mean-field (i.e., coherent excitonic contributions) is not included. With those included, a bigger momentum range is required to achieve comparable accuracy. However, test calculations have shown that the results related to the correlation dynamics do not change qualitatively with increasing k_{max}.

In Fig. 1 we trace the time development of the density, the imaginary part of the interband polarization $P(t) = 2\sum_k P_k(t)$ and various components of the total energy (case I: Fig. 1a and b; case II: Fig. 1c and d). With a lower excess energy, the polarization (envelope) in case I is more dominated by the exciton. The oscillation frequency in

[1]) We mention that the Born approximation (6) with a time dependent screening parameter κ is not strictly energy conserving (even when the computation is numerically exact). In the calculations in this paper, we have used the static 2D Lindhard form $\kappa = 2\frac{e^2}{\varepsilon_0}\sum_{\mu=e,h}\frac{m^*_\mu}{\hbar^2} f_\mu(k=0)$, which leads to an energy violation of approximately 10 %. The violation is more pronounced in 2D than in 3D.

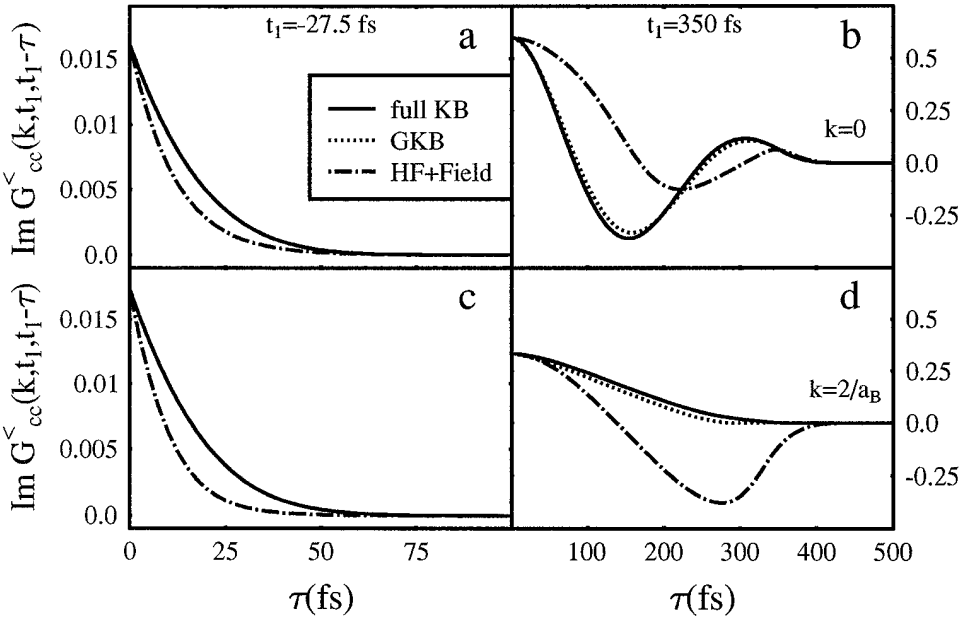

Fig. 2. Imaginary part of $G^<_{cc}(k, t_1, t_2 = t_1 - \tau)$ beginning at the diagonal ($\tau = 0$) and going back in time t_2. The curves correspond to the full two-time result, the GKB ansatz with exact G^r and GKB with G^r in Hartree-Fock approximation. a), c) correspond to $t_1 = -27.5$ fs and b), d) to $t_1 = 350$ fs and to the momenta $k = 0$ (parts a, b) and $k = 2/a_B$ (parts c, d), respectively. The parameters are for case II. At early times, the curves "full KB" and "GKB" are indistinguishable

each case approximately corresponds to the difference between $\hbar\omega_0$ and the exciton frequency. The polarization shows only weak deviations from the corresponding Markovian result. [2]) The potential energy consists of a Hartree-Fock (HF) part and a correlation part, the latter coming from the collision integral Eq. (4). It reflects the build-up of Coulomb correlations among the carriers and, due to total energy conservation, translates into an additional increase of kinetic energy. Therefore, the final equilibrium state is at a higher temperature than predicted by Markovian calculations (curves "Markov KE"). Moreover, due to the correlation energy stored in the system, the equilibrium momentum distribution may deviate from a simple Fermi function. On the other hand, the relative significance of correlations varies with momentum: it is most prominent at low momenta. The KB approach allows to study these features in detail by considering the two-time quantities away from the time diagonal. In Fig. 2, we show the imaginary part of $G^<_{cc}(t_1, t_1 - \tau)$ for two fixed values of t_1 as a function of the distance τ from the diagonal (moving back in time along the t_2 direction). Along with the exact curves from our solution ("full KB") we show two approximations which are based on the GKBA [16]. Applying it to semiconductors [17], one replaces $G^<_{cc}(t_1, t_2)$ by $\sum_\mu G^r_{c\mu}(t_1, t_2)\, G^<_{\mu c}(t_2, t_2)$, with $t_2 = t_1 - \tau, \tau \geq 0$ here. The most accurate approximation is

[2]) Deviations occur only in the tail and are difficult to resolve in the figure. More refined comparisons of Markovian versus KB results over a wider parameter range, including spectra, will be given in a forthcoming paper.

obtained if for the (unknown) Green's function G^{r} the exact one from the solution of the KBE is taken (curves labeled "GKB''). The good agreement shows that, at least for low momenta, the exact GKB reproduces the full two-time evolution very well. In practice, however, the exact G^{r} are not known and approximations are used. Their quality strongly depends on the treatment of correlations, as can be seen in Fig. 2, where we have also plotted the result for G^{r} with correlations neglected (i.e. in Hartree-Fock approximation [17]). Improving on the simple Hartree-Fock ansatz is a topic of ongoing interest, see e.g. [18], and our KBE results can serve as benchmarks for the various approximations.

4. Numerical Scheme and Capabilities

The computation of the self-energies is the limiting factor for the numerical solution of the KBE. In a naive estimate, the numerical effort needed to evaluate them in the statically screened Born approximation Eq. (5) for one carrier component, would scale with the time duration of the run T (in practice, T may be replaced by the smaller memory depth) and the number of grid points per dimension N as T^2N^{3d}, where d is the number of dimensions of the system. In an M-band system the number of collision terms, including polarization scattering, increases like M^4 (however, the number of operations increases only like M^2, cf. Eq. (6). This scaling looks quite prohibitive. However, for many situations of interest, there exists a way around this problem. Consider the Born approximation (5). It involves two (three-dimensional) momentum integrals which are both of *convolution type*: one convolution in $\boldsymbol{\Pi}^{\gtrless}$, Eq. (6) and the second in $\boldsymbol{\Sigma}^{\gtrless}$ between $\boldsymbol{\Pi}\boldsymbol{W}$ and G, which can be evaluated very efficiently by Fast Fourier Transforms (FFT). This technique was first used in solving KBE in [19] with a Fourier-Bessel Transform in cylindrical coordinates, whereas we use a Cartesian coordinates FFT [20], which has the advantage of being able to treat arbitrary anisotropic situations. We summarize the main features of the FFT approach: 1. drastically improved scaling with the number of k-points $(NlogN)^d$; 2. surprisingly high stability (very good conservation of carrier number and total energy) even with a large time step. 3. In contrast to other techniques, one always computes the full six-dimensional momentum integral, allowing for arbitrary anisotropic situations (e.g. warping, anisotropic distributions [13]). 4. 2D calculations are becoming essentially simpler than 3D ones. 5. The scheme works for all non-Markovian scattering integrals, including those for one-time functions (Wigner distribution). 6. The Markov limit is readily obtained from the same code by modifying the time integration [20]. Of course the applicability of this scheme is limited to scattering integrals of convolution type. It is not possible to treat directly exchange scattering integrals, strongly inhomogeneous systems (where momentum conservation does not hold) or strong scattering (beyond the Born approximation). Further details of the computational scheme are discussed in [21].

5. Summary

We have demonstrated that the solution of the two-time Kadanoff-Baym equations for two-band quasi-2D semiconductors is now computationally feasible with acceptable effort. It allows us to study, apart from carrier distributions and polarizations, the dynamics of correlations in detail. It is shown that Coulomb correlations lead to an additional heating of the carriers and that their effect is particularly strong at low momenta.

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References

[1] F. Rossi, S. Haas, and T. Kuhn, Phys. Rev. Lett. **72**, 152 (1994).
[2] F. Jahnke et al., Phys. Rev. Lett. **77**, 5257 (1996).
[3] A. Knorr, S. Hughes, T. Stroucken, and S.W. Koch, Chem. Phys. **210**, 27 (1996).
[4] G. Manzke, Q. Y. Peng, U. Moldzio, and K. Henneberger, phys. stat. sol. (b) **206**, 37 (1998).
[5] D. O. Gericke, S. Kosse, M. Schlanges, and M. Bonitz, phys. stat. sol. (b) **206**, 257 (1998).
[6] D.B. Tran Thoai and H. Haug, Z. Phys. B **91**, 199 (1993).
[7] K. El Sayed, L. Banyai, and H. Haug, Phys. Rev. B **50**, 1541 (1994).
[8] K. El Sayed, S. Schuster, H. Haug, F. Herzel, and K. Henneberger, Phys. Rev. B **49**, 7337 (1994).
[9] G. Manzke, K. Henneberger, J. Heeg, K. El Sayed, S. Schuster, and H. Haug, phys. stat. sol. (b) **188**, 395 (1995).
[10] F.X. Camescasse, A. Alexandrou, D. Hulin, L. Banyai, D.B. Tran Thoai, and H. Haug, Phys. Rev. Lett. **77**, 5429 (1996).
[11] W. Schäfer, J. Opt. Soc. Amer. **B13**, 1291 (1996).
[12] M. Bonitz, D. Kremp, D.C. Scott, R. Binder, W.D. Kraeft, and H.S. Köhler, J. Phys.: Condensed Matter **8**, 6057 (1996).
[13] R. Binder, S.H. Köhler, M. Bonitz, and N. Kwong, Phys. Rev. B **55**, 5110 (1997).
[14] L. Kadanoff and G. Baym, Quantum Statistical Mechanics, Benjamin Co., New York 1962.
[15] L.V. Keldysh, Soviet Phys. – J. Exper. Theor. Phys. **20**, 1018 (1965).
[16] P. Lipavsky, V. Spicka, and B. Velicky, Phys. Rev. B **34**, 6933 (1986).
[17] D. B. Tran Thoai, L. Banyai, E. Reitsamer, and H. Haug, phys. stat. sol. (b) **188**, 387 (1995).
[18] H. Haug and L. Banyai, Solid State Commun. **100**, 303 (1996).
[19] P. Danielewicz, Ann. Phys. (USA) **152**, 239 (1984).
[20] H.S. Köhler, Phys. Rev. C **51**, 3232 (1995).
[21] H. S. Köhler, N. H. Kwong, and H. Yousif, in preparation.