Role of the Exchange Interaction in the Short-Time Relaxation of a High-Density Electron Plasma

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We incorporate the exchange interaction into a simulation in which all electron motion, including that involved in screening, is treated explicitly with a molecular-dynamics simulation run concurrently within an ensemble Monte Carlo treatment of scattering. Exchange is treated for the first time in such an approach by a semiclassical modification of the molecular dynamics which takes full account of the Fermi statistics, and in particular does not violate the exclusion principle. A comparison is made with the short-time relaxation data of Becker *et al.*

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The development of laser-pulse compression techniques has provided a technique to examine nonequilibrium electron distributions on time scales of a few tens of femtoseconds. In the standard pump-and-probe experiments used to study this regime, a short initial laser pulse creates a nonthermal electron-hole plasma; this "pump" pulse bleaches the absorption. By using a second, "probe" pulse to monitor the absorption, one observes the subsequent thermalization of the plasma. Recently, Becker *et al.*[1] used femtosecond photon-echo techniques to probe the electron-electron scattering relaxation process in dense electron-hole plasmas in semiconductors. Most recently, such fast laser excitation studies have focused on the high-density regime.[2] In this high-density regime, one cannot ignore the uncertainty principle. However, this is not the case in the short-time, low-density regime, which could be modeled effectively by using a single-particle picture of electrons which ignored the Fermi character of their statistics.[3] That is, one could choose a position uncertainty small compared to the interelectron length r_s, so the single-particle potential was well defined, and simultaneously leave the momentum well defined. These facts justified an ensemble Monte Carlo (EMC) approach based on classical electron motion interupted by phonon interaction events.[4] These EMC simulations could be coupled effectively with exact treatment of the Coulomb interaction between carriers using a molecular-dynamics (MD) approach.[5] Numerical values of the energy- and density-dependent thermalization times for the various carrier-carrier and carrier-phonon processes have been worked out theoretically, and agree with the experimental data at low density.[6-8]

Because of the high electron excitation energies occurring in these systems, it is difficult to find analytical treatments of screening which properly describe the nontrivial temporal and wave-vector dependences of the screening. To overcome this difficulty, joint molecular-dynamics and ensemble Monte Carlo simulations provide a real-space treatment of the Coulomb interaction and avoid such problems.[5] In this approach, individual electrons interact through a Coulomb interaction which is screened only by the high-frequency (valence-electron) dielectric constant. The remaining part of the screening, due to the conduction electrons, arises explicitly from the combined motion of the individual simulated electrons.

At densities near 10^{19} cm^{-3}, which can now be achieved routinely, there are strong theoretical and experimental[9] grounds to expect that many-electron effects will be important. In principle, these higher densities may be modeled by fully quantum-mechanical descriptions which are well understood. In practice, however, the far-from-equilibrium distributions are prohibitively expensive to study numerically, and accurate analytical approaches are not yet available even in the semiclassical limit. A desirable alternative, therefore, is to extend the existing Monte Carlo approaches into the slightly degenerate regime. This has the added advantage of retaining a picture which is accessible to (classical) intuition.

The incorporation of the exchange-energy interaction into such a nearly classical approximation is the subject of this Letter. The method we describe below incorporates the exchange interaction among electrons, and describes an electron distribution which satisfies both the exclusion principle and the uncertainty principle while obeying a set of equations of motion which resemble those of a classical electron gas. The need to satisfy the uncertainty principle gives rise to corrections to the usual Coulomb interaction. Finally, we test the new approach by simulating the bulk GaAs system studied experimentally by Becker *et al.*,[1] finding that the new terms tend to decrease the discrepancy between the experimental results and the classical Monte Carlo results that were previously found at high density.

In practice, semiclassical (EMC) approaches treat the electrons as having both a well-defined plane-wave

momentum and a well-defined position. To go beyond this approximation, we make an explicit choice of wave packets, which serve as the basis for a more rigorous quantum description. To preserve as much as possible the classical picture, we choose minimum-uncertainty wave packets

$$\phi_i(\mathbf{r}) = \phi(\mathbf{r}-\mathbf{x}_i,\mathbf{p}_i) = (2\pi\sigma^2)^{-3/4}\exp\left[-\frac{(\mathbf{r}-\mathbf{x}_i)^2}{4\sigma^2}+\frac{i}{\hbar}\mathbf{p}_i\cdot(\mathbf{r}-\mathbf{x}_i)\right]. \quad (1)$$

This packet describes an electron localized about position $\mathbf{x}_i$ with uncertainty σ and about momentum $\mathbf{p}_i$ with uncertainty $\hbar/2\sigma$. When the interactions among electrons can be ignored, and when the external potentials vary slowly, the value of σ does not enter explicitly into the equations of motion. One then recovers the classical equations of motion. This is roughly equivalent to making the approximation $\langle\nabla V(\mathbf{r})\rangle = \langle\nabla V\rangle(\mathbf{r})$ in Ehrenfest's theorem. At the high densities we study, however, exchange gives rise to momentum-dependent forces which make a large value of σ preferable. In this situation, σ will appear explicitly, and we shall need to determine a particular value for it.

We can use packets of the form (1) as the basis for an expansion of the many-electron system:

$$a_i = \int d^3r\,\phi^*(\mathbf{r}-\mathbf{x}_i,\mathbf{p}_i)\psi(\sigma_i,\mathbf{r}), \quad (2)$$

where $\psi(\sigma_i,\mathbf{r})$ is the field operator which annihilates a particle of spin σ_i at the point $\mathbf{r}$. For any single fixed value of σ_i, if $(\mathbf{x}_i,\mathbf{p}_i)$ is allowed to vary over all phase-space points, then a_i generates an overcomplete basis for describing the electron system. In the spirit of single-particle semiclassical approximations, we represent the many-electron system by a combination of wave packets: $|\Psi\rangle = a_1^\dagger a_2^\dagger\cdots a_N^\dagger|\,\rangle$, where $|\,\rangle$ is the "vacuum" of the unexcited semiconductor conduction band, so $\langle\mathbf{x}_1,\mathbf{x}_2,\ldots,\mathbf{x}_N|\Psi\rangle$ is a Slater determinant wave function. In the independent (or single-) electron approach, $\mathbf{x}$ and $\mathbf{p}\equiv\hbar\mathbf{k}$ represent the position and momentum of a particular electron. In the present approach, on the other hand, the parameters $\mathbf{x}_i$ and $\mathbf{p}_i$ *do not* represent the position and momentum of the ith electron. Instead, they represent the ith phase-space region near which *an* electron may be found. In this way, there is no conflict between the quantum-mechanical identity of the electrons and the existence of distinct positions and momenta. Classically, the nonidentity of the electrons is based simply on the possibility of following their smooth trajectories exactly.

For simplicity, we will derive the equations of motion for a single parabolic band of mass m. We use a Coulomb potential $V(\mathbf{r}-\mathbf{r}') = e^2/4\pi\varepsilon|\mathbf{r}-\mathbf{r}'|$, with ε taken to be the high-frequency dielectric constant. Using this potential, the second-quantized Hamiltonian $\mathcal{H}$ yields an energy $H(\{\sigma_i,\mathbf{x}_i,\mathbf{p}_i\}_{i=1}^N) \equiv \langle\Psi|\mathcal{H}|\Psi\rangle$. This energy is composed of kinetic-energy, direct Coulomb, and exchange-interaction parts:

$$H = E_K + E_D + E_{XC}, \quad (3)$$

$$E_K = \frac{1}{2m}(\mathbf{p}_1^2+\mathbf{p}_2^2+\cdots+\mathbf{p}_N^2)+\frac{N}{8m\sigma^2}, \quad (4a)$$

$$E_D = \tfrac{1}{2}\sum_{i\neq j} V_\sigma(\mathbf{x}_i-\mathbf{x}_j), \quad (4b)$$

$$E_{XC} = -\tfrac{1}{2}\sum_{i\neq j}\delta(\sigma_i,\sigma_j)\hat{V}_\sigma(|\mathbf{p}_j-\mathbf{p}_i|/\hbar)\Delta\sigma(\mathbf{x}_j-\mathbf{x}_i), \quad (4c)$$

where $\Delta_\sigma(\mathbf{x}) = (4\pi\sigma^2)^{-3/2}\exp(-\mathbf{x}^2/4\sigma^2)$, and $\delta(\sigma_i,\sigma_j)$ is a Kronecker delta. The potential

$$V_\sigma(\mathbf{x}) = \int d^3r\,V(\mathbf{r}+\mathbf{x})\Delta_\sigma(\mathbf{r}) \quad (5)$$

is a coarse graining over a length scale σ of the Coulomb potential, while

$$\hat{V}_\sigma(\mathbf{k}) = \int d^3r\,V(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}-\mathbf{r}^2/4\sigma^2}. \quad (6)$$

We can incorporate exchange effects in the joint MD and EMC simulation described earlier by deriving new semiclassical equations of motion from the Hamiltonian (3):

$$\frac{d\mathbf{p}_i}{dt} = -\frac{\partial H}{\partial\mathbf{x}_i} = \sum_{\substack{j=1\\(i\neq j)}}^N \mathbf{F}_{ij}^D + \sum_{\substack{j=1\\(i\neq j)}}^N \delta(\sigma_i,\sigma_j)\mathbf{F}_{ij}^{XC}, \quad (7a)$$

$$\frac{d\mathbf{x}_i}{dt} = \frac{\partial H}{\partial\mathbf{p}_i} = \frac{\mathbf{p}_i}{m} - \sum_{\substack{j=1\\(i\neq j)}}^N \delta(\sigma_i,\sigma_j)\mathbf{G}_{ij}, \quad (7b)$$

where we have written out the form of the terms that arise from (4).

As is clear from (4), the equations of motion involve the packet size σ explicitly. For a parabolic band, the σ-dependent term in the kinetic energy clearly does not affect the equation of motion. This term is the energy cost of confining electrons in finite-size packets. It is necessary in order to satisfy the exclusion principle in a finite-density system. The gradient of the direct Coulomb interaction (4b) gives rise to the $\mathbf{F}^D$ force term in (7a), which depends only on the position parameters. For interparticle separations much greater than σ, $\mathbf{F}^D$ approaches the classical Coulomb potential. In order to maintain a picture as close as possible to the classical one, we would like the direct Coulomb force to act between essentially pointlike particles, and so we will want $\sigma \ll r_s$.

The exchange interaction is more complex than either the direct interaction or the kinetic energy; it depends on every parameter in the problem and contributes terms to

both (7a) and (7b):

$$\mathbf{F}_{ij}^{XC}=\hat{V}_\sigma(\mathbf{k}_j-\mathbf{k}_i)\left[\frac{\partial}{\partial \mathbf{x}_i}\Delta_\sigma(\mathbf{x}_j-\mathbf{x}_i)\right], \tag{8a}$$

$$\mathbf{G}_{ij}=-\mathbf{G}_{ji}\equiv\frac{1}{\hbar}\left[\frac{\partial}{\partial \mathbf{k}_i}\hat{V}_\sigma(\mathbf{k}_j-\mathbf{k}_i)\right]\Delta_\sigma(\mathbf{x}_j-\mathbf{x}_i). \tag{8b}$$

The second quantity plays a role in the position time evolution (7b) which is essentially that of a density-dependent effective-mass term. In precise contrast with the direct interaction, the appropriate limit for computing the exchange energy is $\sigma\rightarrow\infty$. This can be seen most clearly for spatially homogeneous systems (which include the one we treat here). Confining the electrons in a large box of volume V, we ignore correlation by assuming that the spatial and momentum factors in (4c) can be averaged independently. The homogeneity assumption then leads to

$$E_{XC}=\frac{1}{V}\sum_{\substack{i,j=1\\(i\neq j)}}^{N}\delta(\sigma_i,\sigma_j)\frac{e^2}{2\varepsilon|\mathbf{k}_i-\mathbf{k}_j|^2}, \tag{9}$$

which is the exact exchange energy (ignoring correlation). In the limit $\sigma=0$, the exchange energy given by (4c) is zero.

It is clear from the foregoing discussion that no single value of σ can yield an accurate value for both the direct Coulomb and the exchange forces. That is, the value of σ selects a class of basis states, in which either the exchange or direct energy is well defined, and thus susceptible to a classical approximation. One seeks a compromise σ value which treats both interactions with acceptable accuracy. There is no unique or best way to do this. The approach we have adopted is to compute the exact exchange energy for the initial distribution using (9), and choose σ to minimize the difference between this "exact" result and our σ-dependent approximation. Our initial distribution is uncorrelated by definition, so (9) is exact, and our approximation (4c) approaches this exact value. The differences, and justification for this approach, will be discussed in a subsequent paper. The resulting σ value has to be computed separately for each density. This σ is small at low densities, rising to a constant value of ≈ 18 Å above densities of 10^{18} cm^{-3}; for thermal rather than photoexcited electrons, these are the densities at which the semiconductor simulated (GaAs) becomes strongly degenerate at room temperature.

We have performed EMC and MD simulations to model the femtosecond photon-echo experiments of Becker *et al.*,[1] performed in bulk GaAs. The simulations evolved an ensemble of 2000 electrons initially excited from three valence bands (heavy- and light-hole bands, split-off band) into the Γ valley of the conduction band. The initial distribution of carriers was determined explicitly from an excitation pulse energy of 2 eV, with thermal broadening at a lattice temperature of 300 K taken into account. Parameters such as valley masses and deformation potentials were taken from other measurements where possible,[3,6,10] and consistency with *ab initio* calculations has also been found.[7] Holes were neglected, an approximation that is justified *at short times* by the holes' higher mass.[11]

In these earlier studies, we have found that the time constants determined from the photon-echo experiments agreed well with the results of our joint MD and EMC simulations *at low densities*. With increasing density, the experiments (crosses in Fig. 1) exhibited an approximate power-law decrease of the time constant. In contrast, the simulations (triangles in Fig. 1), showed a knee between 10^{17} and 10^{18} cm^{-3}, with the time constant varying slowly at the lower densities, and falling off rapidly at the higher densities. The low-density behavior simply reflects the dominance of phonon-scattering processes, while strong density dependence in the complementary regime reflects the dominance of electron-electron scattering at high density. In the range of densities below $\sim 2\times 10^{18}$ cm^{-3}, the experimental and numerical-simulation data were consistent. At higher densities, however, the experimental time constant was systematically larger than the simulation.

Using the formalism described above, we have included exchange corrections (circles in Fig. 1). It is clear that inclusion of exchange signficantly improves the agreement between experiment and simulation at high densities. At the low densities, where both the electron-electron interaction and the exchange corrections to it are small, the previous simulations remain valid.

We can understand qualitatively why exchange increases the time constant at high density by considering the momentum time-evolution equation (7a). The dominant term on the right-hand side of (7a) is the Coulomb force. The main correction to this force is a cutoff of the force at small ($|\Delta\mathbf{r}|\lesssim\sigma$) interelectron spacings. This

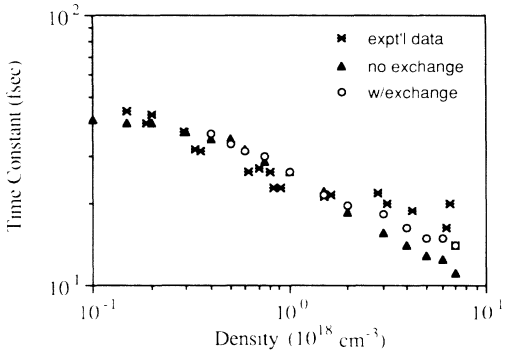

FIG. 1. The relaxation time constant for scattering of carriers out of the initial excitation volume in momentum space. The crosses refer to the data of Becker *et al.* (Ref. 1); triangles are calculations ignoring the role of exchange, and circles include this effect through the method described in the text.

corresponds to the well-known "exchange hole" which arises from the Puali exclusion principle. A second effect arises directly from the exchange energy: In equilibrium, the exchange energy of a homogeneous electron gas is an integral of the density distribution with a strictly negative kernel, so the exchange interaction is attractive. That is confirmed by the form of the exchange force (8a). The potential $\hat{V}_\sigma(\mathbf{k})$ given by (6) is strictly positive, so the exchange force acts in a direction opposite to the direct Coulomb force, partially canceling it in the short-range region where it is strongest.

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