Quantum kinetic theory of confined electrons in a strong time-dependent field

M. Bonitz *

Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität Kiel Leibnizstrasse 15, D-24098 Kiel, Germany

J.W. Dufty

Department of Physics, University of Florida, Gainesville, FL 32611-8440

Abstract

A gauge-invariant Green's function approach to quantum transport of spatially confined electrons in strong electromagnetic fields is presented. The theory inlcudes mean-field and exchange effects, as well as collisions and initial correlations. It allows for a self-consistent treatment of spectral properties and collective effects (plasmons), on one hand, and nonlinear field phenomena, such as harmonic generation and multiphoton absorption, on the other. It is equally applicable to electrons in quantum dots, ultracold ions in traps or valence electrons of metal clusters.

Key words: Quantum dots, quantum kinetic theory, strong field effects

There is a wide range of interesting new phenomena being observed for driven systems of confined electrons, such as those in quantum dots and metallic

* Corresponding author. Fax: +49.431.880.4094
Email address: bonitz@theo-physik.uni-kiel.de (M. Bonitz).

clusters. Their theoretical description in the full generality of confinement potential and time dependent external field is still evolving. The objective here is to describe a formal kinetic theory based on a gauge-invariant nonequilibrium Green's function approach which extends previous work [1,2] to inhomogeneous systems.

We consider a system of N electrons (charge $e = -e_0$) in a confinement potential V_I and an external electromagnetic field described by the hamiltonian

$$\hat{H}_e = \sum_{i=1}^{N} \left\{ \frac{\left(p_i + \frac{e_0}{c}\mathbf{A}(\mathbf{r}_i,t)\right)^2}{2m_e} + \phi(\mathbf{r}_i,t) + V_I(\mathbf{r}_i,t) \right\} + \sum_{j<i}^{N} \frac{e_0^2}{|\mathbf{r}_i - \mathbf{r}_j|}, \qquad (1)$$

where the field is represented by the vector and scalar potentials $\mathbf{A}$ and ϕ which are solutions of Maxwell's equations. The nature of the confinement potential will not be specified, it can be given by an external field (e.g. open quantum dot or trap), by an internal interface potential or by an ensemble of ions (in case of metal clusters), it is assumed to be externally given. The electrons are represented in standard way by fermion field operators $\psi, \psi^\dagger$, and their nonequilibrium properties are described by the two–time correlation functions $g^>(1,1') = \frac{1}{i\hbar}\langle\psi(1)\psi^\dagger(1')\rangle\,, \qquad g^<(1,1') = -\frac{1}{i\hbar}\langle\psi^\dagger(1')\psi(1)\rangle$, where $1 \equiv \mathbf{r}_1, t_1$. The two-time functions $g^>$ $g^<$ contain the complete dynamical and statistical information yielding, on the time-diagonal, the Wigner distribution $f(\mathbf{p},\mathbf{R},t) = -i\hbar g^<(\mathbf{p},\mathbf{R};t_1,t_1')|_{t_1=t_1'=t}$, whereas the dynamical information (e.g. the single–particle spectrum and the correlations) follows from the function values *across the diagonal* in the $t_1 - t_1'$–plane, in particular, from the *spectral function* $a(1,1') \equiv i\hbar\left\{g^>(1,1') - g^<(1,1')\right\}$.

The time evolution of the correlation functions in an electromagnetic field is determined by the Kadanoff–Baym/Keldysh equations reading, in the vector

potential gauge,

$$\left[i\hbar\frac{\partial}{\partial t_1}-\frac{1}{2m_e}\left(\frac{\hbar}{i}\nabla_1+\frac{e_0}{c}\mathbf{A}(1)\right)^2-V_I(1)\right]g^{\gtrless}(1,1')=$$
$$\int d\bar{\mathbf{r}}_1\,\Sigma^{\rm HF}(1,\bar{\mathbf{r}}_1t_1)g^{\gtrless}(\bar{\mathbf{r}}_1t_1,1')+I^{IC}(1,1')+ \qquad (2)$$
$$\int_{t_0}^{t_1}d\bar{1}\;\left[\Sigma^{>}(1,\bar{1})-\Sigma^{<}(1,\bar{1})\right]\,g^{\gtrless}(\bar{1},1')-\int_{t_0}^{t_1'}d\bar{1}\,\Sigma^{\gtrless}(1,\bar{1})\;\left[g^{>}(\bar{1},1')-g^{<}(\bar{1},1')\right],$$

which have to be fulfilled together with the adjoint equations. The l.h.s. of this equation contains all single-particle terms, whereas the r.h.s. contains all corrections due to mean field, exchange and correlations. Here, $\Sigma^{\rm HF}$ is the Hartree-Fock selfenergy (mean field plus exchange), while $\Sigma^{\gtrless}$ are the correlation selfenergies describing collisions in the presence of the confinement and electromagnetic fields. Further, t_0 denotes the (arbitrary finite) initial time, and $I^{IC}(1,1')$ is an additional collision integral due to initial correlations existing in the system at $t=t_0$ [3].

We now derive gauge-invariant kinetic equations for the correlation functions in Wigner representation, $g^{\gtrless}(\mathbf{R},\mathbf{k},t,t')$, which are defined by the gauge-invariant Fourier transform,

$$\tilde{g}(\mathbf{R},\mathbf{k},t_1,t_1')=\int d\mathbf{r}\,\exp\left[-\frac{i}{\hbar}\mathbf{r}\cdot\left(\mathbf{k}-\frac{e_0}{c}\int_{t_1'}^{t_1}\frac{dt'}{\tau}\,\mathbf{A}(t')\right)\right]g(\mathbf{R},\mathbf{r},t_1,t_1'), \quad (3)$$

Here, R and r are the center of mass and relative coordinates, $2R=r_1+r_1'$, $r=r_1-r_1'$ and T and τ are the respective macroscopic and relative times. A lengthy but straightforward calculation yields the following pair of adjoint equations

$$\left\{\left[i\hbar\frac{\partial}{\partial t_1}+i\hbar\,\frac{\mathbf{K}^A(t_1,t_1')}{t_1-t_1'}\nabla_{\mathbf{k}}\right]-\frac{1}{2m_e}\left[\frac{1}{2}\frac{\hbar}{i}\nabla_R+\mathbf{k}-\mathbf{K}^A(t_1,t_1')\right]^2\right\}$$

$$\times\ \tilde{g}^{\gtrless}(\mathbf{R},\mathbf{k},t_1,t_1')$$
$$-\int d^3r \int \frac{d^3k_1}{(2\pi\hbar)^3}\ e^{-\frac{i}{\hbar}\mathbf{r}(\mathbf{k}-\mathbf{k}_1)}\, V_I^{\rm eff}\left(\mathbf{R}+\frac{\mathbf{r}}{2},t\right)\tilde{g}^{\gtrless}(\mathbf{R},\mathbf{k}_1,t_1,t_1')$$
$$=\tilde{I}^{\gtrless}(\mathbf{R},\mathbf{k},t_1,t_1'), \tag{4}$$

$$\left\{\left[-i\hbar\frac{\partial}{\partial t_1'}+i\hbar\,\frac{\mathbf{K}^A(t_1',t_1)}{t_1-t_1'}\nabla_{\mathbf{k}}\right]-\frac{1}{2m_e}\left[-\frac{1}{2}\frac{\hbar}{i}\nabla_R+\mathbf{k}-\mathbf{K}^A(t_1',t_1)\right]^2\right\}$$
$$\times\ \tilde{g}^{\gtrless}(\mathbf{R},\mathbf{k},t_1,t_1')$$
$$-\int d^3r \int \frac{d^3k_1}{(2\pi\hbar)^3}\ e^{+\frac{i}{\hbar}\mathbf{r}(\mathbf{k}-\mathbf{k}_1)}\, V_I^{\rm eff}\left(\mathbf{R}+\frac{\mathbf{r}}{2},t\right)\tilde{g}^{\gtrless}(\mathbf{R},\mathbf{k}_1,t_1,t_1')$$
$$=-\left[\tilde{I}^{\gtrless}(\mathbf{R},\mathbf{k},t_1',t_1)\right]^*, \tag{5}$$

where the effective potential includes the Hartree mean field, $V_I^{\rm eff}(\mathbf{r},t) = V_I + \Sigma^H$ which is the solution of Poisson's equation $\Sigma^{\rm H}(\mathbf{r},t) = \int d^3r_1 V(\mathbf{r}-\mathbf{r}_1)n(\mathbf{r}_1,t)$ with n denoting the nonequilibrium local electron density $n(\mathbf{r},t) = \int \frac{d^3p}{(2\pi\hbar)^3} f(\mathbf{p},\mathbf{r},t)$. The integral $\tilde{I}^{\gtrless}$ on the r.h.s. contains all terms of the r.h.s. of Eq. (3) except the one containing Σ^H. Further, we defined

$$\mathbf{K}^A(t,t') \equiv -\frac{e_0}{c}\int_{t'}^{t} dt''\,\frac{\mathbf{A}(t)-\mathbf{A}(t'')}{t-t'}, \tag{6}$$

These equations are still exact and contain the full field and confinement potential dependence in the mean field and collision terms. They are, in principle, well suited for direct numerical solution generalizing the field-free spatially homogeneous results, e.g. [4,5].

Alternatively, these equations can be used as a rigorous starting point for analytical approximations. In particular, we can obtain the equation for the single-time distribution function, which follows from the difference of equations (4) and (5) for the function $g^<$ at equal times $t_1 = t_1' = t$,

$$\left\{\frac{\partial}{\partial t}+\frac{\mathbf{k}}{m_e}\nabla_R - e_0\mathbf{E}(t)\nabla_{\mathbf{k}}\right\} f(\mathbf{R},\mathbf{k},t)$$

$$+\frac{2}{\hbar}\int d^3r \int \frac{d^3k_1}{(2\pi\hbar)^3}\ \sin\frac{\mathbf{r}\,(\mathbf{k}-\mathbf{k}_1)}{\hbar}\, V_I^{\rm eff}\left(\mathbf{R}+\frac{\mathbf{r}}{2},t\right) f(\mathbf{R},\mathbf{k}_1,t) = I(\mathbf{R},\mathbf{k},t),$$

where the electric field $\mathbf{E}(t) = -c\int_{t_0}^{t} dt'\mathbf{A}(t')$ is assumed to be homogeneous over the range of the confiment volume and $I(\mathbf{R},\mathbf{k},t) \equiv -2\mathrm{Re}[\tilde{I}^{<}(\mathbf{R},\mathbf{k},t,t)]$.

In the *local approximation* the integral on the left side reduces to the classical gradient expression, $-\nabla_R V_I^{\rm eff}(\mathbf{R},t)$, while the exchange contribution vanishes, and the collision integrals coincide with those of a homogeneous system. To account for weak spatial inhomogeneities arising from the confinement potential, we consider the first order gradient approximation to equations (4, 5). The resulting quantum kinetic equation for the Wigner distribution reads, allowing for an arbitrary band structure $\epsilon(\mathbf{k})$,

$$\left\{\frac{\partial}{\partial t} + \nabla_{\mathbf{k}}\bar{\epsilon}\nabla_R - \left[\nabla_{\mathbf{R}}\bar{\epsilon} + e_0\mathbf{E}(t)\right]\nabla_{\mathbf{k}}\right\} f(\mathbf{R},\mathbf{k},t) = I_{\rm coll}(\mathbf{R},\mathbf{k},t) + I^{IC}(\mathbf{R},\mathbf{k},t), \tag{7}$$

$$\text{with} \qquad \bar{\epsilon}(\mathbf{R},\mathbf{k},t) = \epsilon(\mathbf{k}) + \Sigma^H(\mathbf{R},t) + \Sigma^F(\mathbf{R},\mathbf{k},t) + V_I(\mathbf{R},t), \tag{8}$$

$$\Sigma^F(\mathbf{R},\mathbf{k},t) = -\int \frac{d^3k_2}{(2\pi\hbar)^3} V(k_2) f(\mathbf{R},\mathbf{k}-\mathbf{k}_2,t). \tag{9}$$

Consistency and conservation laws require the collision integrals to be used in first order gradient approximation as well:

$$\begin{aligned} I_{\rm coll}(\mathbf{R},\mathbf{k},t) = -2\mathrm{Re}\int_{t_0}^{t} d\bar{t}\Big\{&\Sigma^{>}[\mathbf{R},\mathbf{k}+\mathbf{K}^A(t,\bar{t}),t,\bar{t}]\,g^{<}[\mathbf{R},\mathbf{k}+\mathbf{K}^A(t,\bar{t}),\bar{t},t] \\ &-\Sigma^{<}[\mathbf{R},\mathbf{k}+\mathbf{K}^A(t,\bar{t}),t,\bar{t}]\,g^{>}[\mathbf{R},\mathbf{k}+\mathbf{K}^A(t,\bar{t}),\bar{t},t]\Big\}. \\ +\hbar\,\mathrm{Im}\int_{t_0}^{t} d\bar{t}\,&\left(\nabla_{\mathbf{R}_1}\nabla_{\mathbf{k}_2} - \nabla_{\mathbf{R}_2}\nabla_{\mathbf{k}_1}\right) \\ &\Big\{\Sigma^{>}[\mathbf{R}_1,\mathbf{k}_1+\mathbf{K}^A(t,\bar{t}),t,\bar{t}]\,g^{<}[\mathbf{R}_2,\mathbf{k}_2+\mathbf{K}^A(t,\bar{t}),\bar{t},t] \\ &-\Sigma^{<}[\mathbf{R}_1,\mathbf{k}_1+\mathbf{K}^A(t,\bar{t}),t,\bar{t}]\,g^{>}[\mathbf{R}_2,\mathbf{k}_2+\mathbf{K}^A(t,\bar{t}),\bar{t},t]\Big\}_{\mathbf{R}_1=\mathbf{R}_2=\mathbf{R};\,\mathbf{k}_1=\mathbf{k}_2=\mathbf{k}} \end{aligned} \tag{10}$$

and similarly for I^{IC}. Note that the momentum arguments in all functions are

shifted by the field-dependent momentum K^A which reflects the explicit field-dependence of the scattering process giving rise to the intracollisional field effect and to nonlinear phenomena including collisional harmonics generation and (inverse) bremsstrahlung [1].

To obtain a closed equation for f, the two-time correlation functions in Eq. (10) have to be expressed by the Wigner distributions. The appropriate solution in nonequilibrium is the generalized Kadanoff-Baym ansatz [6] which was generalized to the presence of external fields in [2]. Here, the spatial inhomogeneity due to the confinement potential requires a further generalization by including gradient corrections:

$$\begin{aligned}
\pm g^{\gtrless}(\mathbf{R},\mathbf{k},t,t') = {} & g^R(\mathbf{R},\mathbf{k},t,t') f^{\gtrless}[\mathbf{R},\mathbf{k}-\mathbf{K}^A(t',t),t'] \\
& - f^{\gtrless}[\mathbf{R},\mathbf{k}-\mathbf{K}^A(t,t'),t] g^A(\mathbf{R},\mathbf{k},t,t'), \\
& + \frac{i\hbar}{2}(\nabla_{\mathbf{R}_1}\nabla_{\mathbf{k}_2} - \nabla_{\mathbf{R}_2}\nabla_{\mathbf{k}_1}) \\
& \quad \Big\{ g^R(\mathbf{R}_1,\mathbf{k}_1,t,t') f^{\gtrless}[\mathbf{R}_2,\mathbf{k}_2-\mathbf{K}^A(t',t),t'] \\
& \quad - f^{\gtrless}[\mathbf{R}_1,\mathbf{k}_1-\mathbf{K}^A(t,t'),t] g^A(\mathbf{R}_2,\mathbf{k}_2,t,t') \Big\}_{\mathbf{R}_1=\mathbf{R}_2=\mathbf{R};\, \mathbf{k}_1=\mathbf{k}_2=\mathbf{k}}
\end{aligned} \tag{11}$$

where $f^{>} \equiv 1 - f$ and $f^{<} \equiv f$. Finally, the retarded and advanced Green's functions $g^{R/A}$ describe the correlated single-particle spectrum. Their equation of motion follows from the sum of the two-time equations (4, 5) which has to be treated with the same approximations as the kinetic equation for f [7].

Equations (7 – 11) provide a closed system for the electron Wigner distribution function in a confinement potential $V_I(R)$ and a homogeneous time-dependent electromagnetic field $\mathbf{E}(t)$ of arbitrary strength. It is completely general as far as the treatment of correlations is considered, arbitrary scattering processes can be included by specifying the selfenergies $\Sigma^{\gtrless}$. The first order gradient approximation generally restricts the model to weak inhomogeneities, whereas

for parabolic potentials $V_I(R)$ it is exact. Our result is of importance for electrons in quantum dots, ions in traps, metal clusters etc. It essentially improves previous treatments, most of which incorrectly treat the exchange [by neglecting it completely or in the ∇_k term in Eq. (7)] or neglect the field dependence of the scattering terms. Numerical solutions of Eq. (7) seem feasible [8] and are subject of ongoing research.

This work has been supported by the Deutsche Forschungsgemeinschaft (via SFB 198/B10) and the Department of Energy (Grant DE-FG02ER54677). M.B. acknowledges hospitality of the University of Florida where this work was performed.

References

[1] D. Kremp, Th. Bornath, M. Bonitz and M. Schlanges, Phys. Rev. E **60**, 4725 (1999)

[2] M. Bonitz, Th. Bornath, D. Kremp, M. Schlanges, and W.D. Kraeft, Contrib. Plasma Phys. **39**, 329 (1999)

[3] D. Semkat, D. Kremp and M. Bonitz, Phys. Rev. E **59**, 1557 (1999); J. Math. Phys. **41**, 7458 (2000)

[4] M. Bonitz et al., J. Phys. Cond. Matt. **8**, 6075 (1996)

[5] *Progress in Nonequilibrium Green's Functions II*, M. Bonitz and D. Semkat (Eds.), World Scientific Publ., Singapore 2002

[6] P. Lipavsky, V. Spicka, and B. Velicky, Phys. Rev. B **34**, 6933 (1986)

[7] M.Bonitz and J.W. Dufty, to be published

[8] Solutions of the field-dependent spatially homogeneous equation have been reported by H. Haberland, M.Bonitz, and D. Kremp, Phys. Rev. E **64**, 026405 (2001) where also earlier references can be found.