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A THEORY OF HIGH ELECTRIC FIELD TRANSPORT

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The Generalized Quantum Langevin Equation (GLE) approach to the quantum transport of many electron systems is developed to study the high electric field transport. Two GLE equations, one for the center-of-mass momentum and the other for the center-of-mass energy, are obtained. The non-linear transport effects due to the presence of a high electric field are reflected directly in the memory functions of momentum and energy. By including velocity fluctuations, "collision broadening" and the "intra-collision field effect" appear in our transport equations quite naturally. In our theory, these quantum effects are illustrated by the phenomena of the level broadening, the velocity fluctuation relaxation and acceleration effects.

1. Introduction

High electric field transport has been studied for the last three decades[1–4]). In recent years it has received considerable attention due to the remarkable advance in the techniques of crystal growth and device processing[5,6]) which affords a new realm of physics on the submicrometer and subpicosecond dimensional scale. Theoretically, the high electric field transport has been discussed primarily in terms of the Boltzmann equation[1–3]). However, in the domain of ultrasmall structures (submicrometer) and ultrafast times (subpicosecond), simple arguments based on the uncertainty principle show that high field transport in semiconductors needs a more exact approach than that offered by the semiclassical Boltzmann equation[4,7]). A lot of theoretical attempts have been made toward developing a high field transport theory which would be capable of including a variety of high field effects in submicron structures, such as hot electron effects, ballistic electrons, and large electron density gradients. Among them, to name a few, there are the Monte Carlo technique[8]), the resolvent method[9,10]), the Green's function method[11,12]), the

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Wigner representation approach[13,14]), the path integral[15]), the generalized Langevin equation (GLE)[16-19]) and the balance equations[20-23]).

Through these intense theoretical studies, new phenomena of high field transport are indeed predicted. Among these, the "collisional broadening" (CB) and the "intra-collisional field effect" (ICEF)[4,9,10]) have perhaps attracted most of the attention. Originally, these two effects (CB and ICFE) were predicted by Barker in an effort to derive the steady-state Boltzmann high field transport equation by using resolvent super-operator techniques[9,10]). As the concept of a distribution is suspect under high field transport (since, to quote Thornber[15]), ". . . the electron . . . . never has a well-defined momentum . . ."), the standard approach of calculating a distribution is unsatisfactory. Also, the electron–electron interactions are hardly taken into account in the resolvent technique[9]). Many other attempts have been made toward a detailed exploration of the CB and ICFE effects[4]). Up to now a first principles account of the effects and a full understanding are still lacking. In this paper we study high field transport by the generalized Langevin equation (GLE) approach. Special attention will be given to the CB and ICFE effects.

Previously, we have used the GLE method to study the low field quantum transport, especially with high order impurity scattering[19]). Also, a brief account of the derivation of the steady state equation, as well as the transient equation by GLE have been reported[24]). In general, our method is developed by connecting the works of Ford, Lewis and O'Connell[25,26]), who have demonstrated the usefulness of the GLE approach to heat bath problems, to that of Ting et al.[18,22]), who have introduced the concept of the center of mass of the electrons to study electronic quantum transport. The basic idea of our theory is the visualization of the center of mass of the electrons as a quantum particle, while the relative electrons and phonons act as a heat bath, which is coupled to the center of mass through electron–impurity and electron–phonon interactions[19]). In the procedure of solving the second order Heisenberg equations of the fluctuating density of the relative electrons, the GLE of the center of mass of electrons is obtained directly. At the same time, the memory function, which contains all the information about the effect of the heat bath on the transport properties of the quantum particle, is also obtained without any assumed approximations for the electron–impurity, electron–phonon and electron–electron interactions. The advantages of our approach are the directness achieved in obtaining the GLE and the simplicity of including higher order approximations, which is very often difficult in other techniques[7-18]). In the present paper we apply this new approach to perform a systematic first principles study of high field transport.

Some restrictions are made for the present discussion. First, we account for the periodic lattice simply through the renormalization of the electron mass, i.e. we consider a single parabolic energy band only and neglect the interband

transition and the Stark ladder effects[27]). Secondly, the electron–electron interaction will be neglected for the sake of briefness of the discussion. The discussion of the interacting electron case under the random phase approximation (RPA) can be carried out parallel to the treatment in ref. 19, without essential difficulties. Finally, the system under investigation is assumed to be homogeneous in electron density. The inclusion of diffusion effects will be the subject of a later publication.

In section 2, the Hamiltonian of N independent electrons interacting with phonons and impurities is described in the center of mass and relative electron coordinates. The non-linear GLE equations of momentum and energy of the center of mass are obtained in their microscopic forms by eliminating the heat bath variables in the equation of motion of the momentum and energy of the center of mass, respectively. In section 3, we first present the general transport equation in our approach, which is obtained after the ensemble average over the momentum and energy GLE equations. The equations are then applied to study the steady state and transient transport. For the steady state case, we stress the velocity fluctuation effect, which induces the level broadening and mobility reduction in our treatment. In the study of transient transport, we pay special attention to the effect of acceleration due to the heat-bath–center-of-mass coupling. In the final section we discuss more about the velocity fluctuation and acceleration effects.

2. Non-linear generalized Langevin equations

Consider a set of N non-interacting electrons under the influence of a spatially uniform electric field $\boldsymbol{E}$. The electrons are coupled with phonons and an array of n_i randomly distributed impurities. In the center of mass and relative coordinates description, the total Hamiltonian is written as[19,22])

$$H = H_C + H_B + H_I , \tag{2.1}$$

$$H_C = \frac{\boldsymbol{P}^2}{2M} - NeE \cdot \boldsymbol{R} , \tag{2.2}$$

$$H_B = \sum_{k\sigma} \varepsilon_k C^\dagger_{k\sigma} C_{k\sigma} + \sum_{q\lambda} \Omega_{q\lambda} b^\dagger_{q\lambda} b_{q\lambda} , \tag{2.3}$$

$$H_I = \sum_{q,a} U(q)\, e^{iq\cdot(R-R_a)} \rho_q + \sum_{q\lambda} M(q\lambda)\, e^{iq\cdot R}(O^A_{q\lambda} + O^E_{q\lambda}) \equiv H_I^{im} + H_I^{ph} \tag{2.4}$$

Here $\boldsymbol{P}$ and $\boldsymbol{R}$, $M = Nm$ are the center-of-mass momentum, position and mass, respectively, Planck's constant $\hbar = 1$ is used throughout the calculation, and $\boldsymbol{R}_a$

is the position of the ath impurity. $U(\boldsymbol{q})$ denotes the Fourier transform of the impurity potential, $M(\boldsymbol{q}, \lambda)$ is the electron–phonon matrix element, which satisfies the Hermitian condition $M(\boldsymbol{q}, \lambda) = M^*(-\boldsymbol{q}, \lambda)$. $C^{\dagger}_{k\sigma}$ and $C_{k\sigma}$ are creation and annihilation operators for relative electrons with wave vector $\boldsymbol{k}$, spin σ and energy $\varepsilon_k = |\boldsymbol{k}|^2/2m$, and

$$\rho_q = \sum_{k\sigma} \rho_{kq\sigma} = \sum_{k\sigma} C^{\dagger}_{k-q/2,\sigma} C_{k+q/2,\sigma} \tag{2.5}$$

is the electron density operator. In addition, $b^{+}_{q\lambda}$ and $b_{q\lambda}$ are creation and annihilation operators for phonons with wave vector $\boldsymbol{q}$, energy $\Omega_{q\lambda}$ in branch λ. An electron-phonon operator is defined as

$$O_{q\lambda} = (b_{q\lambda} + b^{+}_{-q\lambda})\rho_q = O^{\mathrm{A}}_{q\lambda} + O^{\mathrm{E}}_{q\lambda} \equiv \sum_{k\lambda} O_{k\,q\lambda}\,, \tag{2.6}$$

where A and E refer to the absorption and emission, respectively, of a phonon. The superscripts "im" and "ph" refer to the impurity and phonon contributions, respectively, to the interaction Hamiltonian. Since we are here concerned with the spin-independent properties only, we suppress the spin indices σ in the following.

The Hamiltonian H may be interpreted as the Hamiltonian of a quantum particle (H_{C}) immersed in a heat bath (H_{B}), which interact through their coupling (H_{I}). This interpretation is significant because then we can relate it to the problems involving the interaction of a quantum system with a heat bath which have been studied intensively in recent years[25,26]).

The essential feature of the motion of the center of mass is that of a Brownian particle, while the phonons and relative electrons play a role as frictional and random forces via their coupling to the center of mass coordinates. We use the same basic procedure that we used in ref. 19 which, for the most part, was confined to the weak-field case. Here we concentrate on the high-field case. As in ref. 19, we eliminate the heat bath variables; in our case it is convenient to choose ρ_q and $O_{q\lambda}$, which are coupled to center of mass through H_{I} in (2.4). Then the equation of motion for the quantum particle (the center of mass) can be recast in the form of a generalized quantum Langevin equation, which is the central idea of the following derivation. In the expressions (2.2)–(2.4), we have $\boldsymbol{R}$, $\boldsymbol{P}$ as quantum particle variables, and ρ_q, $O_{q\lambda}$ the heat bath variables. Apart from the fact that ρ_q and $O_{q\lambda}$ are defined in their second quantized forms, our problem differs from that of FLO in that we have a complicated heat bath.

The equation of motion for the quantum particle is obtained via the Heisenberg equations of motion, which are (with time differentiation denoted by a dot)

$$\dot{\boldsymbol{P}} = -\mathrm{i}[\boldsymbol{P}, H] = -\mathrm{i}[\boldsymbol{P}, (H_{\mathrm{C}} + H_{\mathrm{I}})]$$
$$= NeE - \mathrm{i}\sum_{q,a} \boldsymbol{q}U(\boldsymbol{q})\,\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{R}-\boldsymbol{R}_a)}\rho_{\boldsymbol{q}} - \mathrm{i}\sum_{q\lambda} \boldsymbol{q}M(\boldsymbol{q}\lambda)\,\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}}O_{q\lambda}$$
$$\equiv Nf + \boldsymbol{g}^{\mathrm{im}} + \boldsymbol{g}^{\mathrm{ph}}\,, \tag{2.7}$$

$$\dot{\boldsymbol{R}} = \frac{\boldsymbol{P}}{M}\,. \tag{2.8}$$

Here the last two terms on the right-hand side of (2.7) are the heat bath forces due to impurities and phonons, respectively. For later use we write these heat bath forces in terms of new quantities, $U_q^{\mathrm{i}}(R)$, U_q^{i}, $M_{q\lambda}(\boldsymbol{R})$, $\boldsymbol{g}_q^{\mathrm{im}}$ and $\boldsymbol{g}_{q\lambda}^{\mathrm{ph}}$, defined by

$$\boldsymbol{g}^{\mathrm{im}} \equiv \sum_q \boldsymbol{g}_q^{\mathrm{im}} \equiv -\mathrm{i}\sum_{q,a} \boldsymbol{q}U(\boldsymbol{q})\,\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{R}-\boldsymbol{R}_a)}\rho_{\boldsymbol{q}} \equiv -\mathrm{i}\sum_q \boldsymbol{q}U_q^{\mathrm{i}}\,\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}}\rho_q$$
$$\equiv -\mathrm{i}\sum_q \boldsymbol{q}U_q^{\mathrm{i}}(\boldsymbol{R})\rho_q\,, \tag{2.9}$$

$$\boldsymbol{g}^{\mathrm{ph}} \equiv \sum_q \boldsymbol{g}_{q\lambda}^{\mathrm{ph}} \equiv -\mathrm{i}\sum_{q\lambda} \boldsymbol{q}M(\boldsymbol{q}\lambda)\,\mathrm{e}^{\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}}O_{q\lambda} \equiv -\mathrm{i}\sum_{q\lambda} \boldsymbol{q}M_{q\lambda}(\boldsymbol{R})O_{q\lambda}\,. \tag{2.10}$$

We stress that these forces on the quantum particle are of quantum mechanical nature, and the coordinates $\boldsymbol{R}$ of the quantum particle enter these forces in the form of the phase factor $\boldsymbol{q}\cdot\boldsymbol{R}$.

Following FLO, we now seek explicit expressions for the operators of the heat bath ρ_q and $O_{q\lambda}$ by solving their Heisenberg equations of motion. The details of the derivation can be found in ref. 19 and the results are listed in appendix A. Making use of (2.5) and recalling that spin indices are suppressed, we substitute the expressions ρ_{kq}, $O_{kq\lambda}^{\mathrm{A}}$ and $O_{kq\lambda}^{\mathrm{E}}$ of (A.1)–(A.3), respectively, into (2.7), and after rearranging terms we obtain the following generalized quantum Langevin equation for the motion of the center of mass:

$$M\ddot{R}_\alpha + \int_{-\infty}^{t} \mu_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t')M\dot{R}_\beta(t')\,\mathrm{d}t' = F_\alpha(t) + Nf_\alpha(t)\,. \tag{2.11}$$

Here both the forces $\boldsymbol{g}^{\mathrm{im}}$ and $\boldsymbol{g}^{\mathrm{ph}}$ defined in (2.9) and (2.10) have been split into a random force $\boldsymbol{F}(t)$ and a frictional force, the latter being the second term on the left-hand side of (2.11). The random force is

$$\boldsymbol{F}(t) = -\mathrm{i}\sum_k \sum_q \boldsymbol{q}U_q^{\mathrm{i}}(\boldsymbol{R})\,\mathrm{e}^{-\mathrm{i}\omega_{kq}t}\rho_{kq}^{\circ}$$
$$-\mathrm{i}\sum_k \sum_{q\lambda} \boldsymbol{q}M_{q\lambda}(\boldsymbol{R})[\mathrm{e}^{-\mathrm{i}(\omega_{kq}+\Omega_{q\lambda})t}O_{kq\lambda}^{\circ\mathrm{A}} + \mathrm{e}^{-\mathrm{i}(\omega_{kq}-\Omega_{q\lambda})t}O_{kq\lambda}^{\circ\mathrm{E}}]\,. \tag{2.12}$$

where ρ°_{kq} and $O^{\circ A,E}_{kq\lambda}$ are defined in (A.1)–(A.3),

$$\omega_{kq} = \varepsilon_{k+q/2} - \varepsilon_{k-q/2}\,, \tag{2.13}$$

and where ε_k and $\Omega_{q\lambda}$ refer to free electron and phonon energies, respectively. The matrix elements of the memory term $\mu(t, t')$ in (2.11) are combinations of the electron–impurity and electron–phonon scatterings, and to the lowest order of electron–impurity and electron–phonon interactions it may be written in the form

$$\begin{aligned}\mu_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t') &= \mu^{\mathrm{im}}_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t') + \mu^{\mathrm{A}}_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t') + \mu^{\mathrm{E}}_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t') \\ &= \sum_{\mathrm{s}} \mu^{\mathrm{s}}_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t')\,,\end{aligned} \tag{2.14}$$

where

$$\mu^{\mathrm{s}}_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t') \equiv \sum_{kq} \mu^{\mathrm{s}}_{kq,\alpha\beta}(\dot{\boldsymbol{R}}; t, t') = \sum_{kq} d^{\mathrm{s}}_{kq,\alpha\beta}\, \mathrm{e}^{\mathrm{i}q\cdot(\boldsymbol{R}(t)-\boldsymbol{R}(t'))-\mathrm{i}\omega^{\mathrm{s}}_{kq}(t-t')} \tag{2.15}$$

and (suppressing the phonon branch index λ)

$$d^{\mathrm{im}}_{kq,\alpha\beta} = \frac{q_\alpha q_\beta |U^1_q|^2}{M\omega^{\mathrm{im}}_{kq}}\,\phi_{kq}\,, \qquad \omega^{\mathrm{im}}_{kq} = \omega_{kq}\,, \tag{2.16}$$

$$d^{\mathrm{A}}_{kq,\alpha\beta} = \frac{q_\alpha q_\beta |M_q|^2}{M\omega^{\mathrm{A}}_{kq}}\,\phi_{kq} b_q b^{\dagger}_q\,, \qquad \omega^{\mathrm{A}}_{kq} = \omega_{kq} + \Omega_q\,, \tag{2.17}$$

$$d^{\mathrm{E}}_{kq,\alpha\beta} = \frac{q_\alpha q_\beta |M_q|^2}{M\omega^{\mathrm{E}}_{kq}}\,\phi_{kq} b^1_{-q} b_{-q}\,, \qquad \omega^{\mathrm{E}}_{kq} = \omega_{kq} - \Omega_q\,, \tag{2.18}$$

with ϕ_{kq} defined in (A.4).

We note that although the GLE in (2.11) is formally the same as that of Mori's expression[28]), they are quite different in nature. Mori's GLE is a stochastic equation obtained by projection operator techniques in which the memory term is inherently a combination of some equilibrium averaged quantities, whereas we used a microscopic method, not involving projection operator techniques, and the memory elements (2.14) are in microscopic form. Also, we note that the factor $\mathrm{i}\boldsymbol{q}\cdot(\boldsymbol{R}(t) - \boldsymbol{R}(t')) = \mathrm{i}\boldsymbol{q}\cdot\int_{t'}^{t}\dot{\boldsymbol{R}}(s)\,\mathrm{d}s$ appearing in (2.15) reflects the dependence of the memory function on the external field through the velocity of the center of mass, $\dot{\boldsymbol{R}}$. In the low field case, which we studied in ref. 19, this factor can be neglected and the memory function (2.15) is independent of the field strength. In this paper we study the high field

transport, and use $\dot{\boldsymbol{R}}$ in the argument of μ to stress the field dependent non-linear effects.

To this point we have obtained the generalized Langevin equation (GLE) (2.11) for the center-of-mass momentum. It has been shown[19]) that starting from that equation one can obtain the electric conductivity and study the transport of the many electron system very conveniently. On the other hand, in the study of the electronic quantum transport one is often interested also in energy transfer to the heat bath, which is particularly important in high field transport[1–5]). Thus we derive the GLE for the center-of-mass energy in the following.

The derivation of the GLE of the center-of-mass energy operator $\varepsilon_{\dot{R}}(t) = \frac{1}{2}M\dot{R}(t)^2$ is made quite direct by using the momentum GLE equation (2.11). Multiplying both sides of (2.11) by $\dot{\boldsymbol{R}}(t)$, and noticing that $\mathrm{d}\varepsilon_{\dot{R}}(t)/\mathrm{d}t = \dot{R}\cdot(M\ddot{\boldsymbol{R}})$, we obtain the GLE energy equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\,\varepsilon_{\dot{R}}(t) + \int_{-\infty}^{t} \mu_{\varepsilon_\alpha}(\dot{\boldsymbol{R}};\, t,\, t')\varepsilon_\alpha(t')\,\mathrm{d}t' + Q(t) = W(t) + NeE\cdot\dot{\boldsymbol{R}}(t)\,, \tag{2.19}$$

where

$$W(t) = \dot{\boldsymbol{R}}(t)\cdot\boldsymbol{F}(t) \tag{2.20}$$

is the instantaneous power supplied by the fluctuation force $\boldsymbol{F}(t)$, which is defined in (2.12). The second term on the LHS of (2.19), which acts like a frictional power, was originally in the form of $\dot{\boldsymbol{R}}\cdot\int_{\infty}^{t}\mu(t,t')M\dot{\boldsymbol{R}}(t')\,\mathrm{d}t'$. By using the expression (2.14) of $\mu(t,t')$, we performed a partial integral to obtain the second term on the LHS of (2.19), where the energy memory function

$$\mu_{\varepsilon,\alpha}(\dot{\boldsymbol{R}};\, t,\, t') = 2\sum_{\substack{\boldsymbol{kq}\\ \mathrm{s}}}\left\{\frac{1}{\mathrm{i}\omega^{\mathrm{s}}_{\boldsymbol{kq}}}\, d^{\mathrm{s}}_{\boldsymbol{kq},\alpha\alpha}\,\delta(t-t') + \frac{\boldsymbol{q}\cdot\dot{\boldsymbol{R}}(t)}{\omega^{\mathrm{s}}_{\boldsymbol{kq}}}\,\mu^{\mathrm{s}}_{\boldsymbol{kq},\alpha\alpha}(\dot{\boldsymbol{R}};\, t,\, t')\right\}, \tag{2.21}$$

and where $d^{\mathrm{s}}_{\boldsymbol{kq}}$ and $\mu^{\mathrm{s}}_{\boldsymbol{kq}}(\dot{\boldsymbol{R}};\, t,\, t')$ are the same as in (2.15). The $Q(t)$ in (2.19) represents the contribution to the frictional power due to the acceleration, which has the form

$$Q(t) = \sum_{\substack{\boldsymbol{kq}\\ \mathrm{s}}} \frac{\mathrm{i}}{\omega^{\mathrm{s}}_{\boldsymbol{kq}}}\, M\dot{R}_\alpha(t) \int_{-\infty}^{t} \mu^{\mathrm{s}}_{\boldsymbol{kq},\alpha\beta}(\dot{\boldsymbol{R}};\, t,\, t')\ddot{R}_\beta(t')\,\mathrm{d}t'\,. \tag{2.22}$$

As expected, the evolution of the center-of-mass energy, being a slow variable, obeys the GLE (2.19), which is similar to that of the GLE (2.11) of the center-of-mass momentum. The random power $W(t)$ of (2.20) obviously

preserves the features of a random term, i.e. it will disappear after averaging over the system. As for the energy memory function $\mu_\varepsilon(t, t')$ of (2.21), it is composed by two parts, namely, the Markov term with a $\delta(t)$ function and a non-Markov term. Our formula shows that in the low field limit ($\dot{\boldsymbol{R}} \to 0$), the second term on the RHS of (2.21) will be negligible, and then the evolution of the $\varepsilon_{\dot{R}}$ is basically Markovian. The non-Markov behaviour of the energy evolution represented by the second term on RHS of (2.21) will become important when the field gets higher. The interesting feature of this term is that not only does it display the memory effects through the μ^s contained in it, but also it is inhomogeneous in time due to the presence of $\dot{R}(t)$. In other words, the evolution of the center-of-mass energy, in the high field case, displays memory effects and local time behavior, and it is generally in a non-equilibrium state under the external field. So in our theory it is possible to treat the equilibrium and non-equilibrium situations consistently. Also we remark that the clean separation of the Markov and non-Markov behaviours in our energy memory function (2.21) is consistent with other studies of open systems[29]). A final note about the energy GLE (2.19) is that, as an energy evolution equation, the terms on the right-hand side are actually the sum of the rate of change of the energy of the center of mass, relative electrons and phonons. The rate of change of the energy operator of relative electrons H_e, and phonons H_{ph}, have been worked out in appendix B and appendix C. By using the expressions (B.4), (C.4) for $\dot{H}_e$ and $\dot{H}_{ph}$, respectively, we can rewrite (2.19) as an energy balance equation

$$\frac{d}{dt}\varepsilon_{\dot{R}}(t) + \dot{H}_e(t) + \dot{H}_{ph}(t) = Ne\boldsymbol{E}\cdot\dot{\boldsymbol{R}}(t)\,. \tag{2.23}$$

The derivation of the above formula is presented in appendix D. A similar expression to (2.23) has been derived by Ting et al.[22,23]) in another approach. Even though we have arrived at the same energy balance form as Ting et al. did, the $\dot{H}_e$ and $\dot{H}_{ph}$ in our expressions are different from those of Ting et al. in that they are expressed in terms of non-Markovian memory functions, as can be seen in (B.4) and (C.4), respectively. Besides, our derivation of (2.23) is direct and it is in a microscopic form, whereas Ting et al. take averages from the beginning. Furthermore, our emphasis is not on the energy balance equation per se but on the energy GLE equation (2.19), which will form the essence of our subsequent discussions.

3. High electric field quantum transport equations

We have now two GLE operator equations (2.11) and (2.19), illustrating the microscopic evolution of the momentum and energy of the electronic center of

mass, respectively. We stress that, as distinct from the low field case, the memory functions depend on $\dot{\boldsymbol{R}}$. On averaging these equations over the ensemble, we will obtain the macroscopic momentum and energy transport equations. In general, at any time t, one can denote the velocity $\dot{\boldsymbol{R}}(t)$ as the sum of the drift velocity $\boldsymbol{V}(t) = \overline{\dot{\boldsymbol{R}}(t)}$ and its fluctuation $\delta\dot{\boldsymbol{R}}(t) = \dot{\boldsymbol{R}}(t) - \overline{\dot{\boldsymbol{R}}(t)}$, i.e.

$$\dot{\boldsymbol{R}}(t) = \boldsymbol{V}(t) + \delta\dot{\boldsymbol{R}}(t)\,, \quad \overline{\delta\dot{\boldsymbol{R}}(t)} = \boldsymbol{0}\,, \tag{3.1}$$

where we use a bar to denote the ensemble average over the center-of-mass coordinates. Then after averaging over the whole system, the GLE equations (2.11) and (2.19) are replaced by

$$M\dot{V}_\alpha(t) + \int_{-\infty}^{t} \overline{\langle \mu_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t')\rangle M\dot{R}_\beta(t')}\, \mathrm{d}t' = NeE_\alpha\,, \tag{3.2}$$

$$\frac{\mathrm{d}}{\mathrm{d}t}(\varepsilon_V + \overline{\varepsilon_{\delta\dot{R}}}) + \int_{-\infty}^{t} \overline{\langle \mu_{\varepsilon,\alpha}(\dot{\boldsymbol{R}}; t, t')\rangle \varepsilon_{\dot{R},\alpha}(t')}\, \mathrm{d}t' + \overline{Q(t)} = NeE \cdot \boldsymbol{V}(t)\,, \tag{3.3}$$

where the symbol $\langle\ \ \rangle$ denotes the ensemble average over relative electrons, phonons and impurities, $\varepsilon_V = \frac{1}{2}MV(t)^2$ and $\varepsilon_{\delta\dot{R}} \equiv \frac{1}{2}M\,\delta\dot{\boldsymbol{R}}(t)^2$. The remaining complication of the present equations (3.2) and (3.3) lies in the average over the center of mass, the complication arising from the fact that the memory functions $\mu_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t')$ and $\mu_{\varepsilon,\alpha}(\dot{\boldsymbol{R}}; t, t')$ defined in (2.14) and (2.21) are all functions of $\dot{\boldsymbol{R}}$.

The momentum and energy GLE equations (3.2) and (3.3) are quite rigorous and general; while we have restricted our treatment to lowest-order in the electron–impurity and electron–phonon interactions, our results are valid for all values of the electric field. In this section we derive the transient and steady state transport equations from eqs. (3.2) and (3.3). In both cases the fluctuation parts $\delta\dot{\boldsymbol{R}}(t)$ of the velocity $\dot{\boldsymbol{R}}(t)$ in (3.1) can be treated as small quantities with respect to the drift velocity $\boldsymbol{V}(t)$.

For the purpose of solving the GLE equations (3.2) and (3.3) it is convenient to write the time dependent factor in the memory functions (2.14) and (2.21) as

$$-\mathrm{i}\omega^{\mathrm{s}}_{\boldsymbol{kq}}(t-t') + \mathrm{i}\boldsymbol{q}\cdot\int_{t'}^{t} \dot{\boldsymbol{R}}(t_1)\,\mathrm{d}t_1 = \mathrm{i}\omega_{\mathrm{s}}(V)(t-t') + \mathrm{i}\boldsymbol{q}\cdot\int_{t'}^{t}\left[-\int_{t_1}^{t}\dot{\boldsymbol{V}}(t_2)\,\mathrm{d}t_2 + \delta\dot{\boldsymbol{R}}(t_1)\right]\mathrm{d}t_1\,, \tag{3.4}$$

where

$$\omega_s(V) = \boldsymbol{q}\cdot\boldsymbol{V}(t) - \omega^s_{kq} \tag{3.5}$$

and the ω^s_{kq} are defined by (2.16)–(2.18).

3.1. *Steady state transport*

Under steady state, the transport is time independent, and we have

$$\boldsymbol{V}(t) = V_d\hat{\boldsymbol{X}}\,, \quad \dot{\boldsymbol{V}}(t) = \boldsymbol{0}\,, \quad |\delta\dot{\boldsymbol{R}}(t)| \ll V_d\,, \tag{3.6}$$

where for definiteness we have taken the electric field $\boldsymbol{E}$ along the X-direction and, as a consequence, so is the drift velocity V_d. It follows from (2.14) and (3.4) that, to the lowest non-vanishing order of $\delta\dot{\boldsymbol{R}}$,

$$\overline{\mu(\dot{\boldsymbol{R}};\, t, t')} = \sum_{\substack{kq\\ s}} d^s_{kq}\, e^{iqV_d(t-t') - i\bar{\omega}^s_{kq}(t-t')} = \mu(V_d;\, t-t')\,, \tag{3.7}$$

i.e. the dependence on time takes a simple form. In (3.7) we have used a bar over ω^s_{kq} to represent the energy level broadening effect due to $\delta\dot{\boldsymbol{R}}$ (see appendix E for detail). To lowest non-vanishing order in $\delta\dot{\boldsymbol{R}}(t)$, the evaluation of (3.2) and (3.3) is now quite straightforward. After some algebra we obtain from (3.2) the momentum transport equation

$$\frac{MV_d}{\tau} + \int_{-\infty}^{t} \langle \mu^\delta_\alpha(V_d;\, t-t')\rangle \overline{(M\,\delta\dot{R}^2_\alpha(t')/V_d)}\, dt' = NeE\,, \tag{3.8}$$

where the momentum transport time

$$\frac{1}{\tau} = \int_{-\infty}^{t} \langle \mu(V_d;\, t-t')\rangle\, dt' = \int_0^\infty \langle \mu(V_d;\, t')\rangle\, dt' = \langle \mu(V_d;\, \omega = 0)\rangle\,, \tag{3.9}$$

with $\mu(\omega)$ denoting the Fourier transform of the memory function[19]). The second term on the LHS of (3.8) is the contribution due to velocity fluctuations, which represents a new contribution to the balance equation, not considered by others, with

$$\mu^\delta_\alpha(V_d;\, t-t') \equiv \sum_{\substack{kq\\ s}} \frac{q_x V_d \omega^s_{kq}}{(q_x V_d - \omega^s_{kq})^2}\, d^s_{kq,\alpha\alpha}\, e^{i(q_x V_d - \omega^s_{kq})(t-t')}\,. \tag{3.10}$$

When $\overline{\delta\dot{R}(t')^2}$ is independent of time, we can rewrite (3.8) as

$$\frac{MV_d}{\tau} + \frac{M\,\delta V_\alpha}{\tau_\alpha^\delta} = NeE\,, \tag{3.11}$$

where $\delta V_\alpha = \overline{\delta \dot{R}_\alpha^2(t)}/V_d$, and the inverse of the momentum fluctuation relaxation time

$$\frac{1}{\tau_\alpha^\delta} = \int_{-\infty}^{t} \langle \mu_\alpha^\delta(V_d; t-t')\rangle \, dt' = \langle \mu_\alpha^\delta(V_d; \omega = 0)\rangle \tag{3.12}$$

with $\mu_\alpha^\delta(\omega)$ denoting the Fourier transform of the momentum fluctuation memory function defined in (3.10). Eq. (3.11) is the momentum balance equation in our formalism. It is new and differs from former momentum balance equations in two aspects. First, unlike the ordinary balance equation which originates from the Boltzmann equation[20,21]), where a Markovian momentum relaxation time is used, we have the momentum relaxation time τ, which depends on all time from $-\infty$ to t. Secondly, the velocity fluctuation contribution to electronic quantum transport is similar to that due to the "collisional broadening" and "intra-collision-field effect" found in the approach beyond conventional Boltzmann equation[9]). The level broadening appears very naturally in our approach when we consider the averaging effect of the velocity fluctuation (see appendix E and (3.7)). Whereas the τ_α^δ in (3.11) is similar to the "time duration of collision", the effect of the second term on the LHS of (3.11) is to reduce the overall mobility. This is easily seen, if we write (3.11) as

$$\frac{MV_d}{\tau^*} = NeE\,, \quad \frac{1}{\tau^*} = \frac{1}{\tau} + \frac{\delta V_\alpha}{V_d}\frac{1}{\tau_\alpha^\delta} > \frac{1}{\tau}\,. \tag{3.13}$$

Now we go back to the derivation of the energy balance equation. Under the steady state condition (3.6), the $Q(t)$ in (3.3) which is defined by (2.22), vanishes. It is straightforward to reduce the energy transport equation (3.3), by use of (2.23), into (to order of $\delta\dot{R}^2$)

$$\frac{d}{dt}\overline{\varepsilon_{\delta\dot{R}}} + \langle \dot{H}_e\rangle + \langle \dot{H}_{ph}\rangle = NeEV_d\,, \tag{3.14}$$

where $\varepsilon_{\delta\dot{R}} = \frac{1}{2}M\,\delta\dot{R}^2$, and

$$\langle \dot{H}_e\rangle = \frac{\frac{1}{2}MV_d^2}{\tau_\varepsilon^e} + \frac{1}{\tau_{\varepsilon,\alpha}^{\delta,e}}\overline{\varepsilon_{\delta\dot{R},\alpha}}\,, \tag{3.15}$$

$$\langle \dot{H}_{ph}\rangle = \frac{\frac{1}{2}MV_d^2}{\tau_\varepsilon^{ph}} + \frac{1}{\tau_{\varepsilon,\alpha}^{\delta,ph}}\overline{\varepsilon_{\delta\dot{R},\alpha}}\,. \tag{3.16}$$

Eq. (3.14) shows that the energy supplied by the external field is dissipated by relative electrons and phonons. In (3.15) and (3.16) we have introduced the following quantities: the inverse of the energy transport time through relative electrons in the absence of energy fluctuations is given by

$$\frac{1}{\tau_\varepsilon^{\mathrm{e}}} = \frac{2}{\tau} - \frac{1}{\rho_\varepsilon^{\mathrm{ph}}} , \tag{3.17}$$

where $1/\tau$ is defined in (3.9), and

$$\frac{1}{\tau_\varepsilon^{\mathrm{ph}}} = \int_0^\infty \langle \mu_\varepsilon^{\mathrm{ph}}(V_\mathrm{d}; t)\rangle \, \mathrm{d}t , \tag{3.18}$$

$$\mu_\varepsilon^{\mathrm{ph}}(V_\mathrm{d}; t) = \sum_{kq} \frac{2\mathrm{i}\Omega_q}{\omega_0^2}\left[\frac{d_{kq}^{\mathrm{A}}\omega_{kq}^{\mathrm{A}}}{\omega_0 - \omega_{kq}^{\mathrm{A}}} - \frac{d_{kq}^{\mathrm{E}}\omega_{kq}^{\mathrm{E}}}{\omega_0 - \omega_{kq}^{\mathrm{E}}}\right]\delta(t) . \tag{3.19}$$

The inverse of the energy fluctuation transport time due to relative electrons is given by

$$\frac{1}{\tau_\varepsilon^{\delta,\mathrm{e}}} = \frac{2}{\tau^\delta} - \frac{1}{\tau_\varepsilon^{\delta,\mathrm{ph}}} , \tag{3.20}$$

where $1/\tau^\delta$ is defined in (3.12), and the inverse of the energy fluctuation transport time due to phonons is given by

$$\frac{1}{\tau_\varepsilon^{\delta,\mathrm{ph}}} = \int_0^\infty \langle \mu_\varepsilon^{\delta,\mathrm{ph}}(V_\mathrm{d}; t)\rangle \, \mathrm{d}t , \tag{3.21}$$

where

$$\mu_\varepsilon^{\delta,\mathrm{ph}}(V_\mathrm{d}; t) = 2\mathrm{i}\sum_{kq} \Omega_q\left[\frac{d_{kq}^{\mathrm{A}}\omega_{kq}^{\mathrm{A}}}{(\omega_0 - \omega_{kq}^{\mathrm{A}})^3} - \frac{d_{kq}^{\mathrm{E}}\omega_{kq}^{\mathrm{E}}}{(\omega_0 - \omega_{kq}^{\mathrm{E}})^3}\right]\delta(t) . \tag{3.22}$$

Eq. (3.14) is the energy balance equation in our formalism, where $NeEV_\mathrm{d}$ is the power supplied by external field E. On the LHS, it shows that the energy gain of the center-of-mass electrons is dissipated through the heat bath, i.e. the relative electrons and phonons. Under steady state conditions, the energy fluctuation $\overline{\varepsilon_{\delta R}}$ is generally time independent, so the first term of the LHS of (3.14) disappears. Also, one usually assumes that the lattice is in contact with another huge heat bath which keeps the lattice temperature fixed[30]), while the energy of relative electrons do not change. Then one possible approach is to reduce the energy balance equation (3.14) into[22])

$$\langle \dot{H}_{\mathrm{ph}}\rangle = NeEV_\mathrm{d} , \tag{3.23}$$

$$\langle \dot{H}_e \rangle = 0 , \tag{3.24}$$

where $\langle \dot{H}_{ph} \rangle$ and $\langle \dot{H}_e \rangle$ are defined in (3.15) and (3.16), respectively. As $\langle H_e \rangle$ is a function of the electron temperature T_e, from (3.23) and (3.24) one can obtain T_e and then calculate V_d and other quantities via (3.8). Thus, the high field steady state transport problem is put into a numerically solvable framework.

3.2. *Transient transport*

We now investigate the time dependent behavior, the transient transport. The electric field E is assumed to be turned on at $t = 0$ and we take the initial drift velocity to be $V(0) = 0$.

In the case of transient transport, the GLE equations (2.11) and (2.19) are self-contained differential–integral equations, i.e. both sides of these equations contain $\dot{V}(t)$ as can be seen directly by substituting (2.15), (2.21) and (3.4) in these equations. Therefore, one must make an approximation with respect to the $\dot{V}(t)$ contained in (3.4). In this paper we restrict ourself to the following approximations:

$$\frac{d}{dt}\dot{V}(t) \approx 0 \quad \text{and} \quad \int_{t_1}^{t} \dot{V}(t_2)\, dt_2 \approx \int_{t-t_c}^{t} \dot{V}(t_2)\, dt_2 . \tag{3.25}$$

The first condition in (3.25) is to assume that the rate of change of the acceleration of the center of mass is very small, which is consistent with the experimental results for the transient velocity[31]). The second approximation in (3.25) is to assume that the acceleration contribution to the memory function in (3.2) and (3.3) has an effective range of time t_c, which is introduced in the present calculation phenomenologically. By using (3.4) and (3.5), the memory function (2.14) can be written as

$$\mu_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t') = \sum_{\substack{\boldsymbol{kq} \\ s}} \mu^{s}_{\boldsymbol{kq},\alpha\beta}(\dot{\boldsymbol{R}}; t, t')$$

$$= \sum_{\substack{\boldsymbol{kq} \\ s}} d^{s}_{\boldsymbol{kq},\alpha\beta} \exp\left\{ i\omega^{c}_{s}(V)(t - t') + i\boldsymbol{q} \cdot \int_{t'}^{t} \delta\dot{\boldsymbol{R}}(t_1)\, dt_1 \right\} , \tag{3.26}$$

where

$$\omega^{c}_{s}(V) = \omega_{s}(V) - \boldsymbol{q} \cdot \dot{\boldsymbol{V}}(t) t_c , \tag{3.27}$$

and $\omega_s(V)$ is defined in (3.5). In general one can also assume that the

fluctuation is relatively small, i.e. $\delta\dot{R}(t) \ll V(t)$. It follows from (2.14) that, to the lowest non-vanishing order of $\delta\dot{R}(t)$,

$$\overline{\mu_{\alpha\beta}(\dot{\boldsymbol{R}}; t, t')} = \sum_{\substack{\boldsymbol{kq} \\ \mathrm{s}}} d^{\mathrm{s}}_{\boldsymbol{kq},\alpha\beta}\, \mathrm{e}^{\mathrm{i}\bar{\omega}^{\mathrm{c}}_{\mathrm{s}}(V)(t-t')} = \mu_{\alpha\beta}(V; t-t')\,, \tag{3.28}$$

i.e. the dependence on time takes a relatively simple form. In (3.28), similar to (3.7) we have used a bar over $\omega^{\mathrm{c}}_{\mathrm{s}}$ to denote the level broadening effect due to velocity fluctuation. To lowest non-vanishing order in $\delta\dot{\boldsymbol{R}}(t)$, the evaluation of (3.2) and (3.3) is now quite straightforward. After some algebra we obtain from (3.2) the momentum transport equation

$$M\dot{V}(t) + \frac{MV(t)}{\tau(V)} + \int_{-\infty}^{t} \langle \mu^{\delta}_{\alpha}(V(t); t-t')\rangle \overline{(M\,\delta\dot{R}^2_{\alpha}(t')/V(t))}\,\mathrm{d}t'$$
$$+ A_{\mathrm{m}}(t)M\dot{V}(t) + A^{\delta}_{\mathrm{m},\alpha}(t)\,\frac{M}{V(t)}\,\frac{\mathrm{d}}{\mathrm{d}t}\,\overline{\delta\dot{R}^2_{\alpha}(t)} = NeE\,, \tag{3.29}$$

where the inverse of the instantaneous momentum transport time

$$\frac{1}{\tau(V)} = \int_{-\infty}^{t} \langle \mu_{xx}(V; t-t')\rangle\,\mathrm{d}t' = \int_{0}^{\infty} \langle \mu_{xx}(V; t')\rangle\,\mathrm{d}t' \tag{3.30}$$

and $\mu_{xx}(V; t')$ is defined in (3.28), which, we stress again, is a level broadened function. The third term on the LHS of (3.29) is the contribution due to velocity fluctuations, with

$$\mu^{\delta}_{\alpha}(V; t-t') \equiv \sum_{\substack{\boldsymbol{kq} \\ \mathrm{s}}} \frac{\omega_0(V)\omega^{\mathrm{s}}_{\boldsymbol{kq}}}{(\omega^{\mathrm{c}}_{\mathrm{s}}(V))^2}\, d^{\mathrm{s}}_{\boldsymbol{kq},\alpha\alpha}\, \mathrm{e}^{\mathrm{i}(\omega_0(V)-\omega^{\mathrm{s}}_{\boldsymbol{kq}})(t-t')}\,, \tag{3.31}$$

and it represents a new contribution to the balance equation, previously discussed by us[24]). For clarity, we emphasize that the superscript "δ" is a label, signifying contributions due to fluctuations, in contrast to the subscript "α" which denotes spatial directions. The last two terms on the LHS of (3.29) are contributions due to acceleration, not seen in the steady state equation (3.8), with

$$A_{\mathrm{m}}(t) = \sum_{\substack{\boldsymbol{kq} \\ \mathrm{s}}} \left\langle \frac{d^{\mathrm{s}}_{\boldsymbol{kq},xx}}{(\omega^{\mathrm{c}}_{\mathrm{s}}(V))^2} \left[1 + 2\left(\frac{\overline{\boldsymbol{q}\cdot\dot{\boldsymbol{R}}(t)}}{\omega^{\mathrm{c}}_{\mathrm{s}}(V)}\right)^2\right]\right\rangle, \tag{3.32}$$

$$A^{\delta}_{\mathrm{m},\alpha}(t) = \sum_{\substack{\boldsymbol{kq} \\ \mathrm{s}}} \left\langle \frac{d^{\mathrm{s}}_{\boldsymbol{kq},\alpha\alpha}\,\omega_0(V)}{2(\omega^{\mathrm{c}}_{\mathrm{s}}(V))^3} \left[\frac{\omega_0(V)}{\omega^{\mathrm{c}}_{\mathrm{s}}(V)} - \frac{3}{2}\right]\right\rangle, \tag{3.33}$$

and $\omega_0(V) = q_x V(t)$. Similarly, from (3.3) and (3.28) we obtain the energy transport equation

$$\dot{\varepsilon}_V(t) + \frac{\mathrm{d}}{\mathrm{d}t}\overline{\varepsilon_{\delta\dot{R}}(t)} + \frac{\frac{1}{2}MV^2(t)}{\tau_\varepsilon(V)} + \int_{-\infty}^{t} \langle \mu^{\delta}_{\varepsilon,\alpha}(V; t-t')\rangle \overline{\varepsilon_{\delta\dot{R}_\alpha}(t')}\,\mathrm{d}t'$$

$$+ A_\varepsilon(t)\dot{\varepsilon}_V(t) + A^{\delta}_{\varepsilon,\alpha}(t)\frac{\mathrm{d}}{\mathrm{d}t}\overline{\varepsilon_{\delta\dot{R}_\alpha}(t)} = NeEV(t)\,, \tag{3.34}$$

where the inverse of the instantaneous energy transport time

$$\frac{1}{\tau_\varepsilon(V)} = \int_{-\infty}^{t} \langle \mu_\varepsilon(V; t-t')\rangle\,\mathrm{d}t' = \int_{0}^{\infty} \langle \mu_\varepsilon(V; t')\rangle\,\mathrm{d}t'\,, \tag{3.35}$$

where $\mu_\varepsilon(V; t-t')$ is defined as in (2.21) except that the argument on the RHS is $(t-t')$, analogous to (3.28), and $\boldsymbol{q}\cdot\dot{\boldsymbol{R}}(t)$ s replaced by $q_x V(t)$. The fourth term on the LHS of (3.34) is due to the velocity fluctuations and

$$\mu^{\delta}_{\varepsilon,\alpha}(V; t-t') \equiv \sum_{\substack{\boldsymbol{kq}\\ \mathrm{s}}} \frac{2\mathrm{i}(\omega^{\mathrm{s}}_{\boldsymbol{kq}})^2}{(\omega^{\mathrm{c}}_{\mathrm{s}}(V))^3}\, d^{\mathrm{s}}_{\boldsymbol{kq},\alpha\alpha}\delta(t-t')\,. \tag{3.36}$$

Similar to the momentum transport equation (3.29), the last two terms on the LHS of the energy transport equation (3.34) are contributions due to acceleration, with

$$A_\varepsilon(t) = \sum_{\substack{\boldsymbol{kq}\\ \mathrm{s}}} \left\langle \frac{d^{\mathrm{s}}_{\boldsymbol{kq},xx}}{(\omega^{\mathrm{c}}_{\mathrm{s}}(V))^2}\left[1 + 2\left(\frac{\overline{\boldsymbol{q}\cdot\delta\dot{\boldsymbol{R}}(t)}}{\omega^{\mathrm{c}}_{\mathrm{s}}(V)}\right)\left(1 - \frac{\omega^{\mathrm{c}}_{\mathrm{s}}(V)}{\omega_0(V)}\right)\right]\right\rangle, \tag{3.37}$$

$$A^{\delta}_{\varepsilon,\alpha}(t) = \sum_{\substack{\boldsymbol{kq}\\ \mathrm{s}}} \left\langle \frac{d^{\mathrm{s}}_{\boldsymbol{kq},\alpha\alpha}}{(\omega^{\mathrm{c}}_{\mathrm{s}}(V))}\left[1 - 4\frac{\omega_0(V)}{\omega^{\mathrm{c}}_{\mathrm{s}}(V)} + \left(\frac{\omega_0(V)}{\omega^{\mathrm{c}}_{\mathrm{s}}(V)}\right)^2\right]\right\rangle. \tag{3.38}$$

In summary, eqs. (3.29) and (3.34) are the momentum and energy transient transport equations. For steady state conditions, one has $\dot{V}(t) = 0$ and $\mathrm{d}\overline{\varepsilon_{\delta\dot{R}}(t)}/\mathrm{d}t = 0$; then the terms in (3.29) and (3.34) due to acceleration vanish and one recovers the steady state transport equations (with $V(t) = V_\mathrm{d}$ and $t_\mathrm{c} = 0$) (3.11) and (3.14). Also, we have shown in the previous subsection that the total effect of the velocity fluctuations is to broaden the energy level and to reduce the overall mobility. In the following discussion we will concentrate on the effects of the acceleration on the transport properties and neglect the velocity fluctuations to write (3.29) and (3.34) as

$$(1 + A(V))M\dot{V}(t) + \frac{MV(t)}{\tau(V)} = NeE \,, \tag{3.39}$$

$$(1 + A(V))\dot{\varepsilon}_V(t) + \frac{\varepsilon_V(t)}{\tau_\varepsilon(V)} = NeEV(t) \,, \tag{3.40}$$

where $A(V)$ represents the $A_m(V)$ and $A_\varepsilon(V)$, which are defined in (3.32) and (3.37), at $\delta\dot{R} = 0$.

We note that the second term on the LHS of (3.40), according to appendix D, is

$$\frac{\varepsilon_V(t)}{\tau_\varepsilon(t)} = \langle \dot{H}_e \rangle + \langle \dot{H}_{ph} \rangle \,, \tag{3.41}$$

where $\langle \dot{H}_e \rangle$, $\langle \dot{H}_{ph} \rangle$ represents the energy changing rate of the relative electrons and phonons separately. Eq. (3.41) is equivalent to saying that the dissipation of the center-of-mass electrons is through the relative electrons and phonons. As $\langle H_e \rangle$ is a function of the electron temperature T_e, (3.39) and (3.40) are numerically solvable to obtain the time dependent $T_e(t)$ and $V(t)$. We leave the details of the numerical work for a future study and here just make a few more comments about the physics of our transient transport equations (3.39) and (3.40).

First, we remark that the $A(V)$ in (3.39) and (3.40) is related to the imaginary part of the memory function (3.28). Actually, if one performs the Fourier transform of (3.28) defined by

$$\mu(V; \omega) \equiv \mu_1(V; \omega) + i\mu_2(V; \omega) = \int_0^\infty \mu(V; t') \, e^{i\omega t'} \, dt' \,, \tag{3.42}$$

then it follows that

$$A(V) = -\sum_q \frac{d}{d\omega_s^c(t)} \mu_2(V; q, \omega_s^c(t)) \,, \tag{3.43}$$

where we used $\mu(V; \omega) = \Sigma_q \mu_q(V; \omega)$. From (3.43) one observes $A(V)$ is generally finite as $\mu_2(\omega)$ is an odd function of ω, which can be seen from (3.28) and (3.42).

Secondly, the momentum transport equation (3.39) can be rewritten into a form similar to the classical Langevin equation

$$\dot{V}(t) + \frac{V(t)}{\tau^*(V)} = \frac{eE}{m^*(V)} \,, \tag{3.44}$$

where

$$m^*(V) = m(1 + A(V)), \quad \tau^*(V) = \tau(V)(1 + A(V)). \tag{3.45}$$

To the lowest order of $V(t)$ (linear GLE case), it may be verified that (3.45) becomes time independent, which will be denoted by $m^* = m^*(V \to 0)$, $\tau^* = \tau^*(V \to 0)$. Then (3.44) has the same form as the classical Langevin equation and it is straightforward to obtain the well known Drude form of the dynamical conductivity from (3.44),

$$\sigma(\omega) = \frac{\mathrm{i}Ne^2}{m^*(\omega + \mathrm{i}/\tau^*)}, \tag{3.46}$$

which is in agreement with the result of Ting and Nee[18]), except that they have neglected the low field contribution of $A = (\delta\mu_2/\delta\omega)|_{\omega=0}$ in (3.45). On the other hand, in the high field case, the $A(V)$ of (3.43) is time dependent, and so is the $M^*(V)$ and $\tau^*(V)$ of (3.45). In consequence, the transient equations (3.39) and (3.40), where $A(V)$ plays an important role, are quite different from the ordinary transport equations. This latter conclusion is at variance with the recent work of Xing and Ting[23]), who found that the non-linear transient transport equations still have the ordinary form of the transport equation, i.e. with the $A(V)$ in (3.39) and (3.40) missing and the t_c contained in $1/\tau(V)$ and $1/\tau_\varepsilon(V)$ neglected. We feel that the absence of $A(V)$ in ref. 23 (where the non-linear GLE has not been derived) is the result of using an unjustified "classical" approximation (absence of memory effect) in the high field case (see, in particular, ref. 23, eq. (53)). This extra term gives strong support to the use of non-linear GLE equations in the analysis of non-linear transient transport problems.

In this section we have derived the high field steady state and transient quantum transport equations, starting from the GLE equations (3.2) and (3.3), which, and to first order to the electron–impurity and electron–phonon interactions, are *exact results for all values of the electric field.* In the steady state case, we obtained the momentum balance equation (3.11) and energy balance equation (3.14), where the velocity fluctuation effects have been taken into account. In the transient case, by using approximation (3.25), we get the momentum transport equation (3.29) and energy transport equation (3.34). We found that the high field transient transport equations are different from the low field corresponding equations, due to the acceleration effect.

4. Conclusion and discussion

In this paper we have presented a generalized quantum Langevin equation approach to the study of high field quantum transport for an electron–

impurity–phonon system. We have derived, rigorously, the momentum and energy nonlinear GLE for the center-of-mass electrons in their operator forms (2.11) and (2.19), respectively. After ensemble averaging, these equations are reduced to the momentum and energy transport equations (3.2), (3.3), which can be used to study steady state as well as the transient transport. Our transport equations differ from the conventional transport equations[5,20–23]), which are often deduced either from the Boltzmann equation or the motion of the center of mass without fluctuation, in three aspects. They are the energy level broadening, the transport time renormalization by velocity fluctuation and the acceleration contributions to the transient transport. These effects are basically illustrated by the diffusion constant D, the velocity fluctuation transport time τ^{δ} and the acceleration contribution $A(V)$, respectively. We discuss these three key effects in turn in the following.

4.1. *Level broadening and diffusion constant D*

It is generally recognized that the dissipation in a many-body system results from interactions between an actually observed subsystem and a heat bath into which energy flows in an irreversible way[32,33]). Nevertheless, the clear description and solutions to the ultimate transition from reversibility to irreversibility remains a difficult problem. In the study of the electron transport properties, one basic question often asked in this respect is that how can the microscopically δ-function-like electron energy level (which in many cases may cause the divergence problem) be broadened. We think that this question has got a clear answer in our GLE approach.

In our description, the fluctuation of the center of mass of the electrons (caused by the interaction with the heat bath), causes the energy level broadening in a natural way through the statistical average (see appendix E). In other words, the broadening is a statistical result, i.e. while the individual electrons are still in their quantum mechanical states, the total system displays dissipation with a decay factor proportional to Dq^2 (see (E.5)) through the average of velocity fluctuation. It is interesting to note that if we approximate the q in (E.5) as $q/\sqrt{N} \sim l^{-1}$, the inverse of the mean free path, and use $D = l^2/N\tau$, then we have $Dq^2 = 1/\tau$, and (E.4) is reduced to the result of the usual self-consistent Green's function calculation.

Also, we stress that the D which appears in (E.3) is the self-diffusion constant of the center of mass, which is known to be N times smaller than the single electron diffusion constant D_s in the low field limit[18]). In general, D is complicated and field dependent, but in the classical limit we can use the Einstein relations to write[34])

$$D(V) = \frac{k_B T_e(V)\tau}{Nm} . \tag{4.1}$$

$D(V)$ will increase dramatically following the heating of electrons in the high field (represented by $T_e(V)$). Thus, in our theory the level broadening due to velocity fluctuations will be more prominent for small systems and high fields. This latter property is analogous to that of the broadening due to the intrafield collision effect studied by Barker et al.[9]).

4.2. *Mobility reduction and velocity fluctuation transport time* τ^δ

In our study, the velocity fluctuation transport time τ^δ is introduced in the momentum transport equations (3.11) to illustrate the relaxation of the extra momentum $M\,\overline{\delta\dot{R}(t)^2}/V_d$ due to the velocity fluctuations. (Note that a similar quantity can be introduced to the transient transport eq. (3.29).) The presence of the term $M\,\overline{\delta\dot{R}(t)^2}/V_d\tau^\delta$ in (3.11) reduces the mobility of the center of mass. This can be easily observed by rewriting (3.11) as

$$\frac{MV_d}{\tau^*} = NeE\,, \tag{4.2}$$

where

$$\frac{1}{\tau^*} = \frac{1}{\tau} + \frac{\overline{\delta\dot{R}(t)^2}}{V_d^2}\,\frac{1}{\tau^\delta}\,. \tag{4.3}$$

It follows that the overall mobility $e\tau^*/m$ is smaller than the classical mobility $e\tau/m$. Furthermore, (4.3) tells us that the significance of this reduction depends on the magnitude of the dimensionless quantity

$$\frac{\overline{\delta\dot{R}(t)^2}}{V_d^2}\,\frac{\tau}{\tau^\delta} = \frac{\overline{\delta\dot{R}(t)^2}}{V_d^2}\left\{2V_d\,\frac{\partial}{\partial V_d}\ln\frac{1}{\tau} + V_d^2\left[\frac{\partial^2}{\partial V_d^2}\ln\frac{1}{\tau} + \left(\frac{\partial}{\partial V_d}\ln\frac{1}{\tau}\right)^2\right]\right\}, \tag{4.4}$$

where the equality is the result of (3.10) and (3.12). (4.4) shows that this correction term will disappear in the low field limit where $1/\tau$ is independent of V_d. Also, since in the classical limit $\overline{\delta\dot{R}^2} = k_B T_e/Nm$, this high field correction is expected to be stronger for small N (small systems) and high T_e (high field) situation. The mobility reduction and the level broadening, together with their strong dependence on the size of the system and the strength of the electric field, are the basic features of the quantum correction in the high field transport found by other theoretical methods. Our approach affords a much simpler, direct and numerical tractable way to study these effects.

4.3. *Acceleration effects and* $A(V)$

In the conventional transient equation[20,23]), the acceleration effect appears only as a Newtonian force $M\dot{V}(t)$. A contribution to the phase factor of the

memory function (3.26) due to the $-\boldsymbol{q}\cdot\dot{\boldsymbol{V}}(t)t_c$ in (3.27) and the $A(V)$ directly displayed in our transport equations (3.39) and (3.40) represent another two effects of the acceleration not considered by others. From our derivation, it is clear that both effects stem from the acceleration contributions to the frictional force due to the electron–impurity and electron–phonon interactions.

The acceleration contribution to the memory function is expected to enhance the rate of increase of the velocity when we turn on the electric field. This can be explained in the following way. The memory function (3.26) is influenced by the velocity $V(t)$ contained inside its phase factor. In the high $V(t)$ (non-linear) regime, an increase of $V(t)$ will result in a decrease of mobility, which is proportional to the inverse of the imaginary part of memory function. When we turn on the electric field ($\dot{V}>0$), the memory function (3.26) at the time t appearing in $V(t)$ should be calculated by using a smaller velocity $V(t)-\dot{V}(t)t_c$, because of the acceleration effect. So the actual instantaneous mobility (and velocity) will be larger than the value obtained by neglecting the $\boldsymbol{q}\cdot\dot{\boldsymbol{V}}(t)t_c$ term.

Finally, the terms proportional to $A(V)$ in our transient equations (3.39) and (3.40) represent another important effect of the acceleration. In particular, in the subpico-second situation when the instant transport behavior near time equal zero becomes important and the $M\dot{V}$, $\dot{\varepsilon}_V$ in (3.39) and (3.40) dominates, then the role of $A(V)$ is expected to be dramatic.

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Appendix A

The solution for the density operator ρ_{kq} and the electron–phonon operators O^{A}_{kq}, O^{E}_{kq} in the case of high electric fields and to lowest order in the electron-impurity and electron–phonon interactions

For the reader's convenience we list here the solutions without proof, and refer to ref. 19 for details. Also we suppress the phonon branch index λ.

The electron density operator is

$$\rho_{kq}(t)=\mathrm{e}^{-\mathrm{i}\omega_{kq}t}\rho^{\circ}_{kq}-\mathrm{i}\int_{-\infty}^{t}\mathrm{d}t'\,\mathrm{e}^{-\mathrm{i}\omega_{kq}(t-t')}\phi_{kq}[U^{\mathrm{i}}_{q}(\boldsymbol{R})+M_q(\boldsymbol{R})(b_q+b^{\dagger}_{-q})]\,. \tag{A.1}$$

The electron–phonon absorption operator is

$$O^{A}_{kq}(t) = e^{-i(\omega_{kq}+\Omega_q)t} O^{\circ A}_{kq} - i \int_{-\infty}^{t} dt' \, e^{-i(\omega_{kq}+\Omega_q)(t-t')} \phi_{kq} M_q(\boldsymbol{R}) b_q b^{\dagger}_q . \tag{A.2}$$

The electron–phonon emission operator is

$$O^{E}_{kq}(t) = e^{-i(\omega_{kq}-\Omega_q)t} O^{\circ E}_{kq} - i \int_{-\infty}^{t} dt' \, e^{-i(\omega_{kq}-\Omega_q)(t-t')} \phi_{kq} M_q(\boldsymbol{R}) b^{\dagger}_{-q} b_{-q} . \tag{A.3}$$

In these equations we have used the symbol "$\circ$" to denote the corresponding free operator, ω_{kq} is defined in (2.13), and

$$\phi_{kq} = [\rho_{kq}, \rho^{\dagger}_q] = C^{\dagger}_{k-q/2} C_{k-q/2} - C^{\dagger}_{k+q/2} C_{k+q/2} . \tag{A.4}$$

Appendix B

The calculation of $\dot{H}_e$, the rate of change of the Hamiltonian of the relative electrons

The equation of motion for the relative electron Hamiltonian H_e can be obained by directly using (2.1) and (2.3), from which we have

$$\begin{aligned} \dot{H}_e &= -i[H_e, H] = i \sum_{kq} [U^{i}_q(\boldsymbol{R}) + M_q(\boldsymbol{R})(b_q + b^{\dagger}_{-q})] \omega_{kq} \rho_{kq} \\ &= i \sum_{kq} [U^{i}_q(\boldsymbol{R}) \rho_{kq} + M_q(\boldsymbol{R}) O_{kq}] \omega_{kq} , \end{aligned} \tag{B.1}$$

where ω_{kq} is defined in (2.13).

Substituting the expressions listed in (A.1)–(A.3) into (B.1), we obtain

$$\begin{aligned} \dot{H}_e(t) = \dot{H}^{\circ}_e(t) + \sum_{kq} \int_{-\infty}^{t} dt' \, e^{-i\omega_{kq}(t-t') + iq\cdot(\boldsymbol{R}(t)-\boldsymbol{R}(t'))} \phi_{kq} \omega_{kq} \{ |U^{i}_q|^2 \\ + |M_q|^2 (b_q b^{\dagger}_q e^{-i\Omega_q(t-t')} + b^{\dagger}_{-q} b_{-q} e^{i\Omega_q(t-t')}) \} , \end{aligned} \tag{B.2}$$

where ϕ_{kq} is defined in (A.4) and

$$\dot{H}^{\circ}_e(t) = i \sum_{kq} \omega_{kq} e^{-i\omega_{kq}t} [U^{i}_q(\boldsymbol{R}) \rho^{\circ}_{kq} + M_q(\boldsymbol{R})(O^{\circ A}_{kq} e^{-i\Omega_q t} + O^{\circ E}_{kq} e^{+i\Omega_q t})] . \tag{B.3}$$

Performing a double partial integration with respect to the terms containing $\omega_{kq}t'$ and $(\omega_{kq} \pm \Omega_q)t'$ in (B.2), we obtain

$$\dot{H}_e(t) = \dot{H}_e^\circ(t) + \sum_{kq} \int_{-\infty}^{t} \frac{\omega_{kq}}{\boldsymbol{q}\cdot\dot{\boldsymbol{R}}} \mu^s_{\varepsilon_\alpha, kq}(\dot{\boldsymbol{R}}; t, t')\varepsilon_{\dot{R}_\alpha}(t')\,dt' + Q_e(t)\,, \tag{B.4}$$

where $\mu^s_\varepsilon = \Sigma_q\, \mu^s_\varepsilon(\boldsymbol{q})$, s = i, A, E, are defined in (2.21), $\varepsilon_{\dot{R}}(t) = \frac{1}{2}M\dot{R}(t)^2$ and

$$Q_e(t) = \sum_{\substack{kq \\ s}} \frac{\omega_{kq}}{\boldsymbol{q}\cdot\dot{\boldsymbol{R}}} \frac{M\dot{R}_\alpha(t)}{\omega^s_{kq}} \int_{-\infty}^{t} \mu^s_{\alpha\beta, kq}(\dot{\boldsymbol{R}}; t, t')\ddot{R}_\beta(t')\,dt'\,. \tag{B.5}$$

Appendix C

The calculation of $\dot{H}_{ph}$, the rate of change of the Hamiltonian for the phonons

The equation of motion for the phonon Hamiltonian H_{ph} can be obtained by directly using (2.1) and (2.3), from which we have

$$\dot{H}_{ph} = -i[H_{ph}, H] = i\sum_{kq} M_q(R)\Omega_q(O^A_{kq} - O^E_{kq})\,. \tag{C.1}$$

Substituting the expressions for $O^{A,E}_{kq}$ listed in (A.2), (A.3) into (C.1), we obtain

$$\dot{H}_{ph}(t) = \dot{H}^\circ_{ph}(t) + \sum_{kq} \int_{-\infty}^{t} dt'\, e^{-i\omega_{kq}(t-t') + i\boldsymbol{q}\cdot(\boldsymbol{R}(t)-\boldsymbol{R}(t'))}\phi_{kq}\Omega_q|M_q|^2$$
$$\times [b_q b^\dagger_q e^{i\Omega_q(t-t')} - b^\dagger_{-q} b_{-q} e^{-i\Omega_q(t-t')}]\,, \tag{C.2}$$

where

$$\dot{H}^\circ_{ph}(t) = i\sum_{kq} \Omega_q\, e^{-i\omega_{kq}t} M_q(\boldsymbol{R})(O^{\circ A}_{kq}\, e^{+i\Omega_q t} - O^{\circ E}_{kq}\, e^{-i\Omega_q t})\,. \tag{C.3}$$

Performing a double partial integration with respect to the terms containing $(\omega_{kq} \pm \Omega_q)t'$ in (C.2), (C.2) can be recast into

$$\dot{H}_{ph}(t) = \dot{H}^\circ_{ph}(t) + \int_{-\infty}^{t} \sum_q \frac{\Omega_q}{\boldsymbol{q}\cdot\dot{\boldsymbol{R}}} (\mu^A_{\varepsilon_\alpha, q}(\dot{\boldsymbol{R}}; t, t') - \mu^E_{\varepsilon_\alpha, q}(\dot{\boldsymbol{R}}; t, t'))\varepsilon_{\dot{R}_\alpha}(t')\,dt'$$
$$+ Q_{ph}(t)\,, \tag{C.4}$$

where $\mu_{\varepsilon}^{A,E} = \Sigma_q \mu_{\varepsilon}^{A,E}(q)$ are defined in (2.21), $\varepsilon_{\dot{R}_\alpha}(t) = \frac{1}{2}M\dot{R}_\alpha(t)^2$ and

$$Q_{\mathrm{ph}}(t) = \sum_{kq} \frac{\Omega_q}{q \cdot \dot{R}(t)} M\dot{R}_\alpha(t) \int_{-\infty}^{t} \left[\mu^{A}_{\alpha\beta,kq}(\dot{R}; t, t') \frac{1}{\omega^{A}_{kq}} - \mu^{E}_{\alpha\beta,kq}(\dot{R}; t, t') \frac{1}{\omega^{E}_{kq}} \right] \dot{R}(t')\, dt' . \tag{C.5}$$

Appendix D

The derivation of eq. (2.23)

Adding (B.4) and (C.4), we obtain

$$\dot{H}_{\mathrm{e}}(t) + \dot{H}_{\mathrm{ph}}(t) = \sum_{\substack{kq \\ s}} \frac{\omega^{s}_{kq}}{q \cdot \dot{R}} \left\{ \int_{-\infty}^{t} \mu^{s}_{\varepsilon_\alpha,kq}(\dot{R}; t, t') \varepsilon_{R_\alpha}(t')\, dt' + Q^{s}_{kq}(t) \right\} - W(t) , \tag{D.1}$$

where ω^{s}_{kq}, $\mu^{s}_{\varepsilon_\alpha,kq}(\dot{R}; t, t')$ and $Q^{s}_{kq}(t)$ are defined by (2.15)–(2.18), (2.21) and (2.22), respectively, and $W(t) = -\dot{H}^{\circ}_{\mathrm{e}}(t) - \dot{H}^{\circ}_{\mathrm{ph}}(t)$, $\dot{H}^{\circ}_{\mathrm{e}}(t)$ and $\dot{H}^{\circ}_{\mathrm{ph}}(t)$ are defined in (B.3) and (C.3). The Fourier transform of (D.1) is

$$\dot{H}_{\mathrm{e}}(\omega) + \dot{H}_{\mathrm{ph}}(\omega) = \sum_{\substack{kq \\ s}} \left(1 + \frac{\omega^{s}_{kq} - q \cdot \dot{R}}{q \cdot \dot{R}} \right) \{ \mu^{s}_{\varepsilon_\alpha,kq}(\dot{R}; \omega) \varepsilon_{R_\alpha}(\omega) + Q^{s}_{kq}(\omega) \} - W(\omega) . \tag{D.2}$$

It is straightforward to show that the term proportional to $(\omega^{s}_{kq} - q \cdot \dot{R})/q \cdot \dot{R}$ on the RHS of (D.2) will vanish after we take the real part of $\mu^{s}_{\varepsilon_\alpha,kq}$ and Q^{s}_{kq}. Thus (D.2) can be rewritten as

$$\dot{H}_{\mathrm{e}}(\omega) + \dot{H}_{\mathrm{ph}}(\omega) = \mu_{\varepsilon_\alpha}(\dot{R}; \omega) \varepsilon_{\dot{R}_\alpha}(\omega) + Q(\omega) - W(\omega) . \tag{D.3}$$

Next, we take the inverse Fourier transform of this equation, which we substitute into the energy GLE (2.19), to obtain (2.23).

Appendix E

The proof that level broadening is due to velocity fluctuations and the derivation of (3.7)

In this appendix, we prove the statement that in our formalism the velocity fluctuation is the cause of the level broadening in the steady state transport.

First, we recall that the momentum memory operator appearing in (3.7) before the averaging over center-of-mass coordinates is (see (2.15) and (3.1))

$$\mu(\dot{\boldsymbol{R}};\, t,\, t') = \sum_{\substack{\boldsymbol{kq} \\ \mathrm{s}}} d^{\mathrm{s}}_{\boldsymbol{kq}} \exp\left\{ \mathrm{i}(\omega_0 - \omega^{\mathrm{s}}_{\boldsymbol{kq}})(t - t') + \mathrm{i}\boldsymbol{q} \cdot \int_{t'}^{t} \delta\dot{\boldsymbol{R}}(s)\, \mathrm{d}s \right\}, \tag{E.1}$$

where $\omega_0 = q_x V_{\mathrm{d}}$ and where the velocity fluctuation $\delta\dot{\boldsymbol{R}}$ is included. Eq. (E.1) reduces to (3.7) when $\delta\dot{\boldsymbol{R}} = \boldsymbol{0}$, and in this case the time dependence of $\mu(t - t')$ is plane-wave like, and energy levels are δ functions. The situation is different once we include the $\delta\dot{\boldsymbol{R}}$ in (E.1). Now we must take average over the configurations of $\delta\dot{\boldsymbol{R}}$. Here we use the cumulant approximation[35])

$$\overline{\exp\left\{ \mathrm{i}\boldsymbol{q} \cdot \int_{t'}^{t} \delta\dot{\boldsymbol{R}}(s)\, \mathrm{d}s \right\}} = \exp\{ \mathrm{i}\boldsymbol{q} \cdot \boldsymbol{c}_1 - \tfrac{1}{2} q_\alpha^2 c_{2\alpha} \}, \tag{E.2}$$

where

$$\boldsymbol{c}_1 = \int_{t'}^{t} \overline{\delta\dot{\boldsymbol{R}}(s)\, \mathrm{d}s} = \boldsymbol{0},$$

$$c_{2\alpha} = \int_{t'}^{t} \int_{t'}^{t} \mathrm{d}s\, \mathrm{d}u\, \overline{\delta\dot{R}_\alpha(s)\, \delta\dot{R}_\alpha(u)} = \overline{(\delta R_\alpha(t) - \delta R_\alpha(t'))^2} \equiv 2D_\alpha(t - t'), \tag{E.3}$$

the last step resulting from the usual definition of the self-diffusion constant[34]) $D(t) \equiv \frac{1}{2} \lim_{t\to\infty} \overline{\delta R(t)^2}/t$. Using (E.2) and (E.3), the average of (E.1) is

$$\overline{\mu(\dot{R};\, t,\, t')} = \sum_{\substack{\boldsymbol{kq} \\ \mathrm{s}}} d^{\mathrm{s}}_{\boldsymbol{kq}}\, \mathrm{e}^{\mathrm{i}(\omega_0 - \omega^{\mathrm{s}}_{\boldsymbol{kq}})(t-t') - D_\alpha q_\alpha^2 (t-t')}. \tag{E.4}$$

Defining

$$\bar{\omega}^{\mathrm{s}}_{\boldsymbol{kq}} = \omega^{\mathrm{s}}_{\boldsymbol{kq}} - \mathrm{i}Dq^2, \tag{E.5}$$

(E.4) is reduced to (3.7).

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