Letter

Limitations and improvements of the relaxation time approximation in the quantum master equation: Linear conductivity in insulating systems

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The nonequilibrium steady states of quantum materials have many challenges. Here, we highlight issues with the relaxation time approximation (RTA) for the dc conductivity in insulating systems. The RTA to the quantum master equation (QME) is frequently employed as a simple method, yet this phenomenological approach is exposed as containing a fatal flaw, displaying non-negligible dc conductivity in the linear response regime. We find that the puzzling behavior is caused by the fact that the density matrix in the RTA incompletely incorporates the first order of the external field. To solve this problem, we have derived a calculation scheme based on the QME that ensures correct behavior in low electric fields. Our method reproduces well the overall features of the exact electric currents in the whole field region. It is not time consuming, and its application to lattice systems is straightforward. This method will encourage progress in this research area as a simple way to more accurately describe nonequilibrium steady states.

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Introduction. Field-induced phenomena in quantum systems under strong electric fields have attracted much attention with the recent developments in laser technology [1–6]. The electromagnetic responses of condensed-matter systems beyond the linear response regime lead to a variety of intriguing phenomena, including bulk photovoltaic effects [7–12] and nonreciprocal transport of quantum geometric origin [13–17]. Laser light with stronger intensities triggers photoinduced phase transitions as a result of nonperturbative quantum effects, which is experimentally demonstrated in the ultrafast timescale and investigated theoretically [18–26].

When the driving field becomes strong, dissipation to environment plays a vital role in determining the distribution function far from equilibrium, which is generally difficult and has a long history of research [27–35]. The theoretical description of open dissipative systems necessitates the nonequilibrium Green's function [1,36–40] or the density matrix [41–45] as a fundamental quantity, rather than the wave function. The nonequilibrium Green's function can be calculated using the diagrammatic approach formulated on the Schwinger-Keldysh contour, while the density matrix is calculated on the basis of the quantum master equation (QME) [46,47]. Recently, nonperturbative modulation of the distribution function due to the Landau-Zener tunneling [48–50] has been formulated by combining the nonequilibrium Green's functions approach with analytic methods [14]. There, it was discussed that systems with broken inversion symmetry lead to interesting nonperturbative phenomena such as tunnel spin current and nonreciprocal current. The Green's function method is a powerful tool, but it is time consuming and its application to lattice systems requires the cumbersome treatment of dynamical phases. The development of QME methods as a complementary method is therefore considered important.

The relaxation time approximation (RTA) is the simplest approximation to describe the nonequilibrium steady state in the QME approach [51–53]. It has been frequently used in the context of the semiclassical Boltzmann equation and the semiconductor Bloch equation. Quite recently, however, it has been pointed out that the RTA has a problem in its application to nonlinear optics [44,45]. We show here that, in addition to this problem, the RTA treatment in insulating systems involves a fatal flaw that the dc conductivity exhibits conducting behavior in the linear response regime, despite the absence of Fermi surfaces. The reason why this has been overlooked until now is that the detailed studies of transport properties in multiband systems have only recently progressed, and that for metallic systems, the problem is only quantitative.

In this Letter, we demonstrate the fatal flaw inherent in the RTA by applying it to the Landau-Zener model which is a minimal model for real insulators. We find that it originates from the fact that the density matrix in the RTA incorporates the first order of the external field E in an incomplete form. On the basis of the QME, we derive a calculation scheme to fix this issue in low electric fields by sequentially incorporating the perturbation correction of the external field. Our method reproduces well the overall features of electric currents in the whole field region. It is less time consuming and comparable to RTA calculations. Its application to lattice systems is straightforward.

Relaxation time approximation. To demonstrate a fatal flaw contained in the standard RTA, let us consider a two-band system with a finite gap. With two eigenenergies $\varepsilon_{k\pm}$, the band gap at each k point is given by $\Delta_k = \varepsilon_{k+} - \varepsilon_{k-} > 0$. We introduce the dc electric field E via the Peierls substitution, $k \to k - eEt$ (hereafter $e = 1$). Following Ref. [14], we introduce the snapshot basis $|\Phi_{k\alpha}(t)\rangle = |u_{\alpha,k-Et}\rangle e^{-i\Theta_\alpha(t)}$ ($\alpha = \pm$) with $|u_{\alpha,k}\rangle$ being the eigenstate of the snapshot Hamiltonian $H(k)$. The phase factor $\Theta_\alpha(t)$ consists of the dynamical and Berry phases [54]. In this basis, the QME for the density matrix $[\rho_k(t)]_{\alpha\beta}$ is written in the 2×2 matrix form as

$$\frac{d\rho_k(t)}{dt} = -i[\mathcal{W}_k(t), \rho_k(t)] + \mathcal{D}(\rho_k(t)), \tag{1}$$

where $[\mathcal{W}_k(t)]_{\alpha\alpha} = 0$, $[\mathcal{W}_k(t)]_{+-} = [\mathcal{W}_k(t)]^*_{-+} \equiv W_k(t) = E\langle u_{+,k-Et}|i\partial_k|u_{-,k-Et}\rangle e^{i\Theta_+(t)-i\Theta_-(t)}$ denotes the transition dipole matrix elements. Here, the dissipation to environment is described by $\mathcal{D}(\rho_k(t))$, whose form in the RTA is given by

$$[\mathcal{D}(\rho_k(t))]^{\mathrm{RTA}}_{\alpha\beta} = \frac{f_D(\varepsilon_{k\alpha}(t))\delta_{\alpha\beta} - [\rho_k(t)]_{\alpha\beta}}{\tau_{\alpha\beta}}, \tag{2}$$

where f_D is the Fermi-Dirac distribution function, and $\varepsilon_{k\alpha}(t) = \varepsilon_{k-Et,\alpha}$. $\tau_{++} = \tau_{--} = \tau_1$ and $\tau_{+-} = \tau_{-+} = \tau_2$ denote the longitudinal and transverse relaxation time, respectively.

By solving Eq. (1), we obtain the density matrix of the nonequilibrium steady state. Then the electric current $J(t)$ is calculated as $J(t) = -e\int \frac{dk}{2\pi}\mathrm{Tr}[\partial_k H(k)\rho_k(t)]$, which can be decomposed into intra- and interband contributions as

$$J(t) = J_{\mathrm{intra}}(t) + J_{\mathrm{inter}}(t), \tag{3}$$

$$J_{\mathrm{intra}}(t) = -\sum_{\alpha=\pm}\int \frac{dk}{2\pi}\frac{\partial\varepsilon_{k\alpha}(t)}{\partial k}[\rho_k(t)]_{\alpha\alpha}, \tag{4}$$

$$J_{\mathrm{inter}}(t) = 2\,\mathrm{Im}\left[\int \frac{dk}{2\pi}\frac{W_k(t)\Delta_k(t)}{E}[\rho_k(t)]_{-+}\right]. \tag{5}$$

The electric current in the nonequilibrium steady states is obtained as the long-time limit of $J(t)$. The presence of J_{inter} was pointed out in the early stage [55], and recently it was rederived and its importance was discussed in detail in the context of the Landau-Zener tunneling [14].

Hereafter, let us consider the Landau-Zener model for simplicity,

$$H(k) = \begin{pmatrix} vk & \delta \\ \delta & -vk \end{pmatrix}, \tag{6}$$

where 2δ corresponds to the band gap. In Fig. 1(a), we show the field dependence of the electric current J^{RTA} in the RTA for several τ with $\tau_1 = \tau_2 = \tau$ at the temperature $T = 0.01\delta$. In the inset, one can see a remarkable increase of the current J^{RTA} at around $E = 0.5E_{\mathrm{th}}$, consistent with the generation of tunneling carriers. Such an increase of tunneling carriers is implied by the rapid increase of the tunneling probability $P_{\mathrm{LZ}} = \exp(-\pi E_{\mathrm{th}}/E)$ with $E_{\mathrm{th}} = \delta^2/v$, as shown in the dashed line of the main panel of Fig. 1(a) [48,56].

Surprisingly, an unexpected linear E dependence with the slope increasing with τ^{-1} is observed in the low-E regime, indicating conducting behavior even though the system is actually an insulator. What is responsible for this finite linear dependence? To clarify the origin, in Fig. 1(b), we separately show the contributions from $J^{\mathrm{RTA}}_{\mathrm{intra}}$ (dashed line) and $J^{\mathrm{RTA}}_{\mathrm{inter}}$ (dotted line) for $\tau^{-1} = 0.02\delta$. One can see that the linear E behavior comes from the interband contribution $J^{\mathrm{RTA}}_{\mathrm{inter}}$.

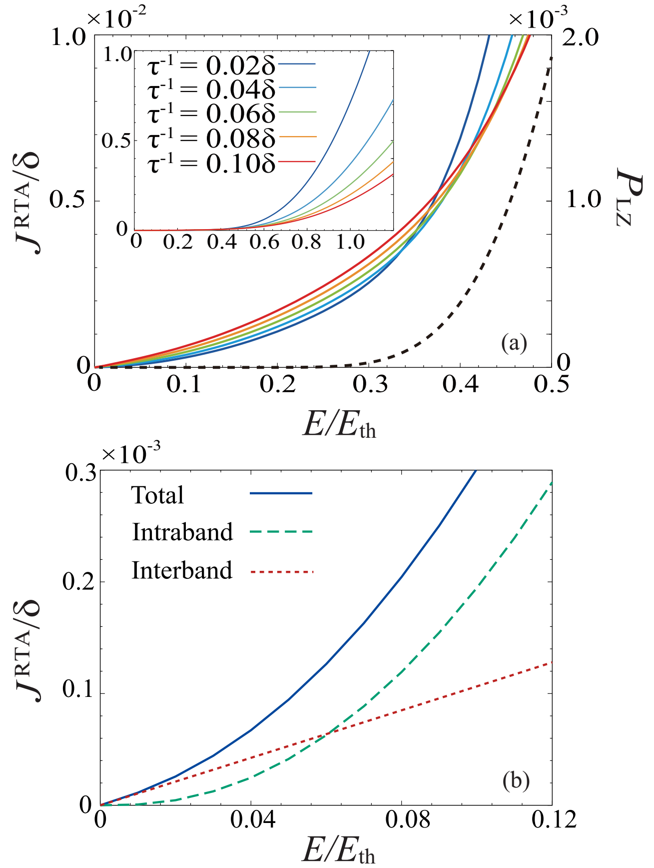

FIG. 1. (a) Field dependence of the dc current J^{RTA} in the RTA at $T = 0.01\delta$. The inset depicts J^{RTA} in a wide range. Solid lines denote the RTA currents for several τ^{-1}. The dotted line represents the tunneling probability P_{LZ}. One can see the exponential behavior in the inset, but encounter an unexpected linear behavior at a low E limit in the main panel. (b) Intraband $J^{\mathrm{RTA}}_{\mathrm{intra}}$ (dashed line) and interband $J^{\mathrm{RTA}}_{\mathrm{inter}}$ (dotted line) contributions of J^{RTA} at $\tau^{-1} = 0.02\delta$ as a function of E/E_{th}. $J^{\mathrm{RTA}}_{\mathrm{inter}}$ shows unphysical linear E behavior.

In fact, the density matrix in the low-E limit can be calculated as

$$[\rho_k(t)]^{\mathrm{RTA}}_{\alpha\alpha} \sim f_D(\varepsilon_{k\alpha}(t)) + E\tau_1\frac{\partial f_D(\varepsilon_{k\alpha}(t))}{\partial k}, \tag{7}$$

$$[\rho_k(t)]^{\mathrm{RTA}}_{+-} \sim -\frac{W_k(t)}{\Delta_k(t) - i\tau_2^{-1}}\delta f_k(t), \tag{8}$$

with $\delta f_k(t) = f_D(\varepsilon_{k-}(t)) - f_D(\varepsilon_{k+}(t))$, by solving Eq. (1) in a perturbative manner [57]. Then, within the linear response regime, the electric conductivities $\sigma^{\mathrm{RTA}} = \lim_{E\to 0} J^{\mathrm{RTA}}/E$ are given by

$$\sigma^{\mathrm{RTA}}_{\mathrm{intra}} = \tau_1\sum_{\alpha=\pm}\int \frac{dk}{2\pi}\left(\frac{\partial\varepsilon_{k\alpha}}{\partial k}\right)^2\left(-\frac{\partial f_D}{\partial\varepsilon_{k\alpha}}\right), \tag{9}$$

$$\sigma^{\mathrm{RTA}}_{\mathrm{inter}} = 2\tau_2^{-1}\int \frac{dk}{2\pi}\frac{|\langle u_{+,k}|i\partial_k|u_{-,k}\rangle|^2\Delta_k}{\Delta_k^2 + \tau_2^{-2}}\delta f_k. \tag{10}$$

In the insulating case, the intraband conductivity $\sigma^{\mathrm{RTA}}_{\mathrm{intra}}$ vanishes at $T = 0$, which is consistent with the Boltzmann theory. On the other hand, the interband conductivity $\sigma^{\mathrm{RTA}}_{\mathrm{inter}}$ does not vanish even in the insulating case, which gives the slope of $J_{\mathrm{inter}}^{\mathrm{RTA}}$ at $E \to 0$. This term increases roughly in proportion to damping τ_2^{-1}. The presence of the small but finite conductivity is only a quantitative problem in metallic systems, but critical in insulating systems. It is a fatal flaw of the RTA in the QME [58]. It has also been recently argued that the RTA is problematic for the optical response [44,45], which is another problem different from the unphysical behavior observed here for the dc current. As we will show later, the current problem comes from the presence of $i\tau_2^{-1}$ in the denominator of Eq. (8). This is due to the fact that the RTA does not properly incorporate the first-order contribution of the electric field E. In our formalism beyond the RTA, this term in the denominator vanishes [cf. Eq. (19)]. This $i\tau_2^{-1}$ term also affects the Hall current, with which the Hall conductivity does not quantize within the RTA [51].

Beyond RTA. To fix the issue in the RTA, here we consider a two-band insulating system coupled to a fermionic reservoir [1,59] within the QME formalism, and then derive the dissipation term $\mathcal{D}(\rho_k(t))$ microscopically. We start with the Born-Markov master equation [46,60]

$$\frac{d\tilde{\rho}_k(t)}{dt} = -\int_{-\infty}^{t} \mathrm{Tr}_{\mathrm{B}}[\tilde{H}_{I,k}(t), [\tilde{H}_{I,k}(s), \tilde{\rho}_k(t) \otimes \tilde{\rho}_B]]ds, \tag{11}$$

where $\tilde{\rho}_k(t)$ is the reduced density operator and $\tilde{\rho}_B$ is the thermal density operator of the fermionic reservoir (bath), respectively. Here, the tildes on operators denote the interaction picture. $\tilde{H}_{I,k}(t)$ represents the interaction term between the system and the bath, $\tilde{H}_{I,k}(t) = \sum_{\sigma p} V_p \tilde{b}_{k\sigma p}^{\dagger}(t)\tilde{c}_{k\sigma}(t) + \mathrm{H.c.}$, where $\tilde{c}_{k\sigma}(t)$ and $\tilde{b}_{k\sigma p}(t) = \tilde{b}_{k\sigma p}(t_0)e^{-i\omega_p(t-t_0)}$ are respectively the annihilation operators of an electron in the system and the bath with momentum k and pseudospin σ. $\mathrm{Tr}_{\mathrm{B}}[\cdots]$ means tracing out the bath degrees of freedom. We impose the broadband condition for the spectral density of the fermionic reservoir as

$$\sum_p \pi |V_p|^2\, \delta(\omega - \omega_p) = \Gamma \text{ (const)}. \tag{12}$$

The key point of our formalism is to express Eq. (11) in the snapshot basis by introducing the transformed field operator

$$\tilde{\psi}_{k\alpha}(t) = \sum_{\sigma} \langle \Phi_{k\alpha}(t)|\sigma\rangle \tilde{c}_{k\sigma}(t), \tag{13}$$

and evaluate the integral in Eq. (11) with employing the adiabatic perturbation theory [13,14]. The key part of time evolution is contained in the dynamical phase as

$$W_k(t-s) \sim e^{-i\Delta_k(t)s} W_k(t), \tag{14}$$

$$|\Phi_{k\alpha}(t-s)\rangle \sim e^{i\varepsilon_{k\alpha}(t)s}|\Phi_{k\alpha}(t)\rangle. \tag{15}$$

Specifically in the adiabatic limit $E \to 0$, these are the exact expressions. We further evaluate the dissipation term in a perturbative manner, such that the resultant QME is exact up to $O(E)$ while maintaining the unitarity of the time evolution operator up to $O(E^2)$. We call this approximation the dynamical phase approximation (DPA).

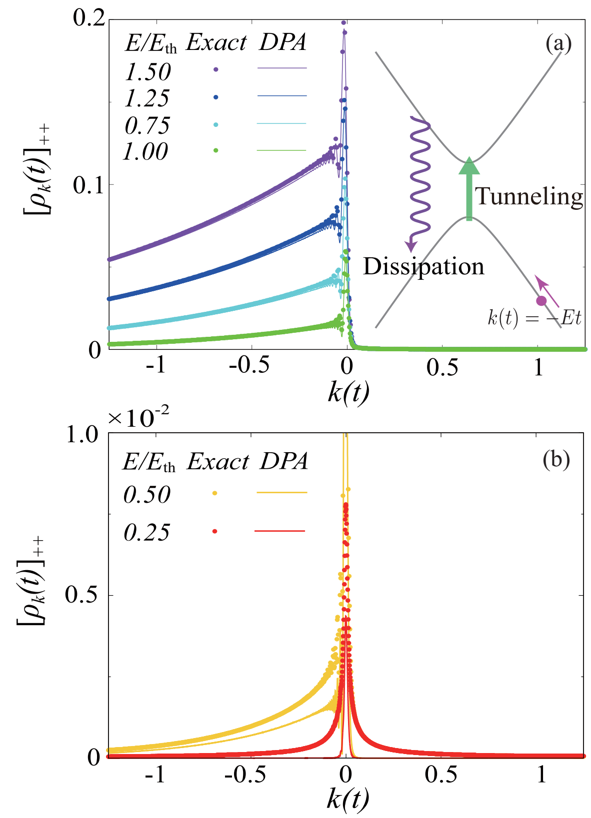

FIG. 2. Occupation number of a single electron in the upper band. Solid lines and solid circles denote the results in DPA and the numerically exact results [14], respectively. The schematic figure represents the electron dynamics in the Landau-Zener model. The DPA results are almost consistent with the exact results. The small deviation is due to the damping-induced excitation inherent in open systems.

Finally, we obtain the dissipation term in the DPA as $\mathcal{D}(\rho_k(t)) = \mathcal{D}_0 + \mathcal{D}_1 + \mathcal{D}_2$, where

$$[\mathcal{D}_0]_{\alpha\beta} = -2\Gamma([\rho_k(t)]_{\alpha\beta} - f_D(\varepsilon_{k\alpha}(t))\delta_{\alpha\beta}), \tag{16}$$

$$[\mathcal{D}_1]_{\alpha\beta} = -2\Gamma\frac{[\mathcal{W}_k(t)]_{\alpha\beta}}{\Delta_k(t)}\delta f_k(t), \tag{17}$$

$$[\mathcal{D}_2]_{\alpha\beta} = 2\Gamma\alpha\frac{|W_k(t)|^2}{\Delta_k^2(t)}\big(\delta f_k(t) + \Delta_k(t) f_D'(\varepsilon_{k,-\alpha}(t))\big)\delta_{\alpha\beta}. \tag{18}$$

The subscript n of $\mathcal{D}_n$ denotes the perturbation order of W_k [57]. Note that the expression here is a simplified one under the particle-hole symmetry, $\varepsilon_{k+}(t) = -\varepsilon_{k-}(t)$. Here, the zeroth-order term $\mathcal{D}_0$ corresponds to the RTA with $\tau_1 = \tau_2 = 1/2\Gamma$, while $\mathcal{D}_1, \mathcal{D}_2$ describe the field-induced correction terms. Specifically, in the presence of Eq. (17), the low-E expression for $[\rho_k(t)]_{+-}$ is replaced from Eq. (8) into

$$[\rho_k(t)]_{+-}^{\mathrm{DPA}} \sim -\frac{W_k(t)}{\Delta_k(t)}\delta f_k(t), \tag{19}$$

with which the unphysical linear term in $J_{\mathrm{inter}}^{\mathrm{RTA}}$ [Eq. (10)] is completely canceled out.

Now, let us compare the DPA calculation with the numerically exact calculation [14]. Figures 2(a) and 2(b) show the

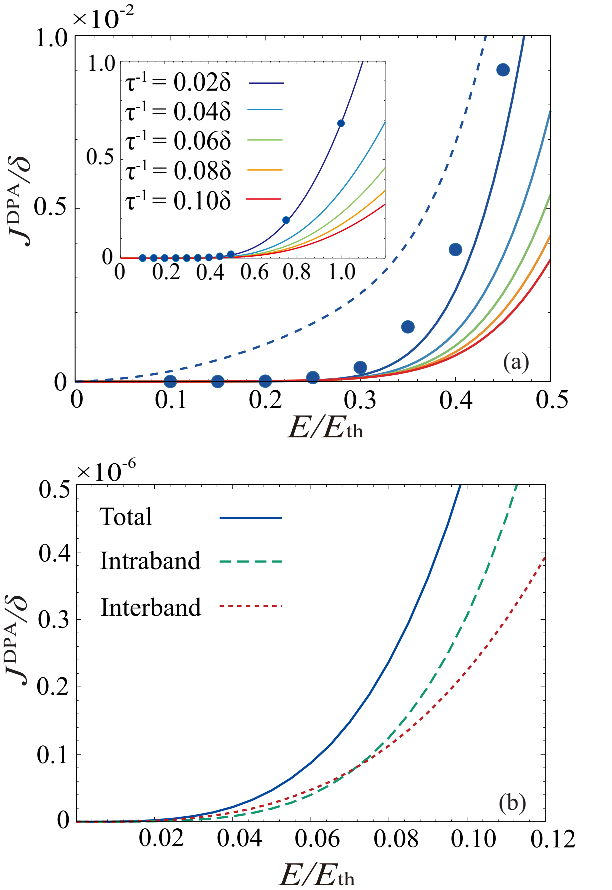

FIG. 3. (a) Field dependence of the dc current J^{DPA} in the DPA. The inset depicts J^{DPA} in the wide range. In the main panel, solid lines denote the DPA currents for several τ^{-1}, the dashed line represents the RTA current at $\tau^{-1} = 0.02\delta$, and the solid circles the result of the numerically exact result. In the inset, one can see that the DPA result reproduces the exact result well. J^{DPA} almost vanishes at $E < 0.2E_{\mathrm{th}}$. We can verify in (b) that J^{DPA} has no linear E terms with an accuracy of less than $10^{-7}\delta$ at $E < 0.06E_{\mathrm{th}}$. At the intermediate region $E \sim 0.4E_{\mathrm{th}}$, J^{DPA} deviates from the exact result by $\sim 10^{-3}\delta$ but roughly increases with the tunneling probability P, implying that the excited carriers are carrying the electric current.

occupation numbers of the upper band at $\tau^{-1} = 2\Gamma = 0.02\delta$ and $T = 0.01\delta$ as a function of $k(t) = -Et$. Electrons starting from $t = t_0 < 0$ ($k > 0$) are excited by Landau-Zener tunneling as they pass through the gap minimum at $t = 0$ ($k = 0$). Then, at $t > 0$ ($k < 0$), the excited electrons decay with a relaxation time τ. As shown in Fig. 2(a), the DPA results are consistent with the exact result at higher temperatures and stronger electric fields. The small deviation in the low-E regime in Fig. 2(b) is due to the damping-induced excitation inherent to the present fermionic reservoir, which is only partially included in the DPA. While this is incorrect from the viewpoint of the rigorous treatment of the fermionic reservoir, it could be considered an advantage since this particular excitation should be absent for the ideal environment.

Finally, we illustrate the field dependence of the electric current in the DPA in Fig. 3. Solid lines in Fig. 3(a) denote the DPA currents for several damping parameters, the dotted line represents the RTA current at $\tau^{-1} = 0.02\delta$, and the solid circles are the numerically exact results. It is clear that the RTA fails to capture qualitative features of the exact result. On the other hand, as shown in the inset of Fig. 3(a), the DPA result well describes the exact one. The most significant improvement is the disappearance of the linear E dependence in the interband contribution. This is due to the fact that the $i\tau^{-1}$ term in the off-diagonal term of the density matrix in the RTA is completely canceled by correctly treating the electric field up to the first order. This also improves the behavior of the intraband contribution, where the exponential behavior is more pronounced. As a result, the total current in the low-field region is also greatly improved and shows exponential behavior. Indeed, J^{DPA} almost vanishes at $E < 0.2E_{\mathrm{th}}$. It can be verified in Fig. 3(b) that J^{DPA} has no linear E terms with an accuracy of less than $10^{-7}\delta$ at $E < 0.06E_{\mathrm{th}}$. At the intermediate region $E \sim 0.4E_{\mathrm{th}}$, J^{DPA} has an error of $\sim 10^{-3}\delta$. This is thought to be due to the damping-induced excitation and/or the fact that the DPA calculation was terminated up to $\mathcal{D}_2$. Although further refinements to incorporate these effects are available in principle, the overall features of the dc current are described well enough qualitatively and semiquantitatively in our DPA. In addition, it has a great advantage that the computational time required to obtain these results is almost the same as for the RTA.

Conclusion. This Letter highlights issues with the RTA for the dc current in insulating systems and proposes an improvement based on the QME. The RTA is frequently employed as a simple method, yet this phenomenological approach is exposed as containing a fatal flaw, displaying non-negligible dc conductivity at the low-E limit in generic insulating systems. This puzzling behavior is because of the incomplete inclusion of the first-order contribution of the electric field in the density matrix. We reevaluate the QME and incorporate the dynamical phase of the transition matrix, thereby correctly capturing the first-order terms of the electric field. We obtained a scheme that accurately describes the insulating behavior. It was demonstrated that it accurately predicts the correct off-diagonal terms in the density matrix and is in semiquantitative agreement with the numerically exact result. This also improves the calculation of quantum Hall effects. While numerically exact calculations can be performed for the present fundamental model, it is often time consuming. Our method is not rigorous but correctly captures the overall features in the dc current. Furthermore, it does not require much computation time and is straightforward to apply to lattice systems. The DPA is an alternative to the RTA, which describes nonequilibrium steady states more correctly. We believe that this method will encourage progress in this research field.

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[58] In the diagrammatic viewpoint, Eqs. (9) and (10) correspond to the contribution of bubble diagrams in linear response theory. It contains only self-energy corrections. The simplest vertex correction vanishes in one-band systems but has a non-negligible contribution in multiband systems. This has the same order contribution of electric field E as the bubble diagram. We believe that our formalism properly includes such key contributions, and then reproduces the insulating behavior.

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