Nernst effect of Dirac fermions in graphene under a weak magnetic field

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The derivation for the transport coefficients of an electron system in the presence of temperature gradient and the electric and magnetic fields are presented. The Nernst conductivity and the transverse thermoelectric power of the Dirac fermions in graphene under charged impurity scatterings and weak magnetic field are calculated on basis of the self-consistent Born approximation. The result is compared with so far the available experimental data.

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I. INTRODUCTION

The Nernst effect as well as the thermoelectric power is a sensitive probe of the impurity scatterings in an electron system. Recently, the Hall and the Nernst effects in graphene have been studied experimentally at relatively strong[1–3] and moderate[2] magnetic fields. Graphene in most of the experiment devices is absorbed on the surface of SiO_2. There are strong evidences that the charged impurities in the substrate are responsible for the carrier density dependences of the electric conductivity[4–10] and the Hall coefficient[11] as measured in the experiments by Novoselov *et al.*[12] At strong magnetic field, the carriers are in the Landau quantized states. In the interior of the system under the strong magnetic field, the carriers are mostly localized around the charged impurities. Though, the Hall effect seems weakly dependent of the impurity scatterings because the current is most likely conducted by the edge states that are not localized.[13] The standard Green's function theory of many body system has difficulty to treat the charge transport under scatterings of the charged impurities in a strong magnetic field since it deals with the bulk states of the electrons. On the other hand, at weak magnetic field when the effect of Landau quantization is negligible, the standard Green's function theory should be applicable for investigating the magnetothermoelectric transports of the electron system.

Based on the self-consistent Born approximation (SCBA) for Dirac fermions under the charged impurity scatterings, we have recently developed an electronic transport formalism for graphene.[9,11,14] It has been shown that the experimentally measured electric conductivity, the inverse Hall coefficient[12] and the thermoelectric power[1–3] are successfully explained by our approach. In this work, along the same approach, we study the Nernst effect of Dirac fermions as a function of the carrier density under a weak magnetic field. Though there exists no experimental measurements of the Nernst effect in a weak field so far, we show that our obtained results for the transverse thermoelectric power could qualitatively compare with the experimental measurements[2] in magnetic fields of moderate strength. We intend to examine to what extend the theory is valid in dealing with the transport properties of graphene.

Meanwhile in doing this work, we present a derivation of the transport coefficients of an electron system under the temperature gradient and the electric and magnetic fields being applied.

The model of electrons in graphene is established from its energy band structure in the first Brillouin zone corresponding to a honeycomb lattice. At low carrier concentration, the low energy excitations of electrons in graphene can be viewed as massless Dirac fermions.[12,15–20] That is, the energy linearly depends on the momentum around the two Dirac points in the first Brillouin zone. Using the Pauli matrices σ's and τ's to coordinate the electrons in the two sublattices (a and b) of the honeycomb lattice and two valleys (around the two Dirac points 1 and 2), respectively, and suppressing the spin indices for briefness, the Hamiltonian of the system is given by

$$H = \sum_k \psi_k^\dagger (v\vec{k}\cdot\vec{\sigma}\tau_z - \mu)\psi_k + \frac{1}{V}\sum_{kq} V_i(q)\psi_{k-q}^\dagger \psi_k, \qquad (1)$$

where $\psi_k^\dagger = (c_{ka1}^\dagger, c_{kb1}^\dagger, c_{kb2}^\dagger, c_{ka2}^\dagger)$ is the fermion operator, the momentum k is measured from the center of each valley, v ($\sim$5.86 eV Å) is the velocity of electrons, μ is the chemical potential, V is the volume of system, and $V_i(q) = n_i(-q)v_0(q)$ is the charged impurity potential.[11,14] Here, $n_i(-q)$ is the impurity density and $v_0(q)$ is given by the Thomas-Fermi (TF) type

$$v_0(q) = 2\pi e^2/(q + q_{\mathrm{TF}})\epsilon \qquad (2)$$

where $q_{\mathrm{TF}} = 4k_F e^2/v\epsilon$ is the TF wave number, $k_F = \sqrt{\pi n}$ (with n as the carrier density) is the Fermi wave number, and $\epsilon \sim 3$ is the effective dielectric constant. For briefness, we hereafter use units of $v = \hbar = k_B$(the Boltzmann constant) $= 1$.

With SCBA,[21,22] the Green's function

$$G(k,\omega) = [\omega + \mu - \vec{k}\cdot\vec{\sigma}\tau_z - \Sigma(k,\omega)]^{-1}$$

$$\equiv g_0(k,\omega) + g_c(k,\omega)\hat{k}\cdot\vec{\sigma}\tau_z$$

and the self-energy $\Sigma(k,\omega)$ of the single particles are determined by coupled integral equations.[9] The diagonal and off-diagonal parts, g_0 and g_c, respectively, of the Green's function can be expressed as

$$g_{0,c}(k,\omega) = [g_+(k,\omega) \pm g_-(k,\omega)]/2$$

with $g_\pm$ as the upper and lower-band Green's functions. Corresponding to the SCBA to the self-energy, the current vertex

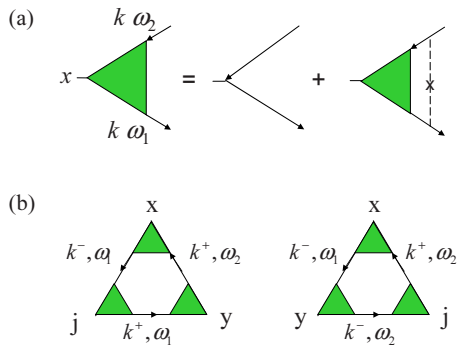

FIG. 1. (Color online) (a) Self-consistent Born approximation for the Current vertex correction. (b) Diagrams for calculating the function $P_{xy}(\omega_1,\omega_2)$. The vertex j is associated with the vector potential A_j. $k^{\pm}=k\pm q/2$.

correction $\Gamma_x(k,\omega_1,\omega_2)$ is given by the ladder-diagrams approximation as shown in Fig. 1(a). $\Gamma_x(k,\omega_1,\omega_2)$ is expanded as

$$\Gamma_x(k,\omega_1,\omega_2)=\sum_{j=0}^{3} y_j(k,\omega_1,\omega_2)A_j^x(\hat{k}), \tag{3}$$

where $A_0^x(\hat{k})=\tau_z\sigma_x$, $A_1^x(\hat{k})=\sigma_x\vec{\sigma}\cdot\hat{k}$, $A_2^x(\hat{k})=\vec{\sigma}\cdot\hat{k}\sigma_x$, $A_3^x(\hat{k})=\tau_z\vec{\sigma}\cdot\hat{k}\sigma_x\vec{\sigma}\cdot\hat{k}$, and $y_j(k,\omega_1,\omega_2)$ are determined by four-coupled integral equations.[9] The functions y_j describe how the current vertex is renormalized by the impurity scatterings from the bare one $A_0^x(\hat{k})$.

II. FORMALISM

A. General formula of the transport coefficients

To study the Nernst effect of graphene, we consider the electronic transport of Dirac fermions under weak in-plane temperature gradient ∇T, electric potential ϕ, and weak magnetic filed $\vec{B}=\nabla\times\vec{A}$ perpendicular to the graphene plane. Here, $\vec{A}$ is the vector potential. Since the temperature gradient ∇T is not a dynamic quantity, we cannot directly apply the linear response theory (LRT) to treat the current response to ∇T. To use LRT, one usually introduces a fictitious gravitational potential (that couples with the Hamiltonian) following the work of Luttinger and obtains the transport coefficients using the Einstein argument relating the currents response to the external perturbations.[23] Here, we present a derivation from a microscopic point of view.

First, following the idea of Luttinger,[23] suppose the system with a variable temperature $T(r)$ in locally equilibrium everywhere in space. Specifically, consider the system is divided into small cells but microscopically large enough. The Hamiltonian of the cell at r_j with chemical potential $\mu(r_j)$ and temperature $T(r_j)$ are given by $H_j=\int_{\text{cell}-j}d\vec{r}\psi^{\dagger}(r)[h(r)-\mu(r_j)]\psi(r)$ with $h(r)=\vec{\sigma}\tau_z\cdot(-i\nabla+\vec{A})+V_i(r)$ and $\psi(r)$ as the operator of Dirac fermions in real space. The distribution function of the system is then given by

$$\rho_1=Z^{-1}\exp\left[-\sum_j H_j/T(r_j)\right], \tag{4}$$

where Z is the normalization constant. Instead of considering $T(r)$, we intend to find out an equivalent system determined by an effective Hamiltonian H_{eff} at constant temperature T_0. Its distribution function is

$$\rho_2=Z^{-1}\exp(-H_{\text{eff}}/T_0). \tag{5}$$

From $\rho_1=\rho_2$, we have $H_{\text{eff}}=\Sigma_j H_j T_0/T(r_j)$. Suppose each cell is macroscopically so small enough that the summation can be replaced with integral over space. H_{eff} reads

$$\begin{aligned}H_{\text{eff}}&=\int d\vec{r}\psi^{\dagger}(r)[h(r)-\mu(r)]\circ\frac{T_0}{T(r)}\psi(r)\\&=\int d\vec{r}\psi^{\dagger}(r)[h(r)+h(r)\circ\Psi(r)+\Phi(r)-\mu_0]\psi(r)\\&\equiv H[\Phi,\Psi]\end{aligned}$$

with $h(r)\circ\Psi(r)=\{h(r),\Psi(r)\}/2$ (here $\{A,B\}$ is the anticommutation relation between A and B) and

$$\Phi(r)=\mu_0-T_0\mu(r)/T(r), \tag{6}$$

$$\Psi(r)=T_0/T(r)-1. \tag{7}$$

Here, T_0 and μ_0 are the average temperature and chemical potential, respectively. In the limit $T(r)\to T_0$, we have $[\mu(r),\Phi(r),\Psi(r)]\to[\mu_0,0,0]$. By so doing, the system with variable temperature $T(r)$ in the local equilibrium state is now described by an equivalent one under the potentials $[\Phi(r),\Psi(r)]$ at constant T_0 and μ_0.

Now we go back to the original problem: how do the currents respond to the temperature gradient ∇T? Initially the system is in the equilibrium state of $H[0,0]$. With gradually turning on ∇T, the system $H[0,0]$ becomes unstable because the equilibrium state shifts to $H[\Phi(r),\Psi(r)]$ and thereby currents are produced. {In the shifting process $[0,0]\to[\Phi(r),\Psi(r)]$, T_0 and μ_0 are kept as constants.} The perturbation here is the difference $H[0,0]-H[\Phi(r),\Psi(r)]$. This is different from the usual case that the perturbations are due to applying the external dynamic potentials and the initial equilibrium state given by $H[0,0]$ keeps unchanged.

Generally, in addition to ∇T, with the external scalar potential ϕ being applied, the system under consideration is $H[\phi,0]$. With respect to the equilibrium system $H[\Phi,\Psi]$, the perturbation is $H[\phi,0]-H[\Phi(r),\Psi(r)]$. Mathematically, we have

$$H[\phi,0]=H[\Phi,\Psi]+H[\phi,0]-H[\Phi,\Psi]\equiv H[\Phi,\Psi]+H'$$

with H' given by

$$\begin{aligned}H'&=\int d\vec{r}\psi^{\dagger}(r)[-h(r)\circ\Psi(r)+\phi(r)-\Phi(r)]\psi(r)\\&\equiv\int d\vec{r}\psi^{\dagger}(r)[\Phi_1(r)+\xi(r)\circ\Phi_2(r)]\psi(r),\end{aligned}$$

where $\Phi_1(r)=\phi(r)-\Phi(r)-\mu_0\Psi(r)$, $\Phi_2(r)=-\Psi(r)$, and $\xi(r)=h(r)-\mu_0$. Here $\Phi_1(r)$ and $\Phi_2(r)$ take the role as the pertur-

bation potentials. In the limit $\nabla T \to 0$, the negative forces $\nabla\Phi_1(r)$ and $\nabla\Psi_1(r)$ read

$$\nabla\Phi_1(r) = \nabla[\phi(r) - \mu(r)] = e\vec{E}, \tag{8}$$

$$\nabla\Phi_2(r) = -T_0\nabla[1/T(r)]. \tag{9}$$

Hereafter we denote μ_0 and T_0 simply as μ and T, respectively, for briefness. According to LRT, we need to find out the corresponding currents determined from the equations of continuity. We here consider the relevant currents.

(i) For the potentials Φ_1 and Φ_2 (actually the corresponding vector potentials), the coupling currents to be determined are $J_1(r)$ and $J_2(r)$, respectively. One may consider to convert the facts $\psi^\dagger(r)\psi(r)$ (coupled to Φ_1 in H') and $\psi^\dagger(r)\xi(r)\psi(r)$ (coupled to Φ_2) into the respective currents in the picture of $H[\Phi,\Psi]$ (from which the perturbed system evolves). The resulted currents then contain the terms of Φ_1 and Φ_2. However, since H' is already linear in Φ_1 and Φ_2, we just only need to consider them in the picture of $H[0,0]$. For the unperturbed system $H[0,0]$, they are the particle current and the heat current,

$$\vec{J}_1(r) \equiv \vec{J}(r) = \psi^\dagger(r)\vec{j}\psi(r), \tag{10}$$

$$\vec{J}_2(r) \equiv \vec{J}^Q(r) = \psi^\dagger(r)\xi(r)\circ\vec{j}\psi(r). \tag{11}$$

with $\vec{j}=\vec{\sigma}\tau_z$.

(ii) The currents of the reference (using subscript r) system $H[\Phi,\Psi]$ itself can be obtained from the known results of the particle current $\vec{J}_{1r}$ and energy current $\vec{J}_r^E$ in Ref. 23. The results are

$$\vec{J}_{1r}(r) = \vec{J}(r)\circ[1+\Psi(r)],$$

$$\vec{J}_{2r}(r) = \vec{J}_r^E(r) - \mu\vec{J}_{1r}(r)$$
$$= \vec{J}^Q(r)\circ[1+2\Psi(r)] + [\Phi(r)+\mu\Psi(r)]\circ\vec{J}(r).$$

Their averages under $H[\Phi,\Psi]$ vanish.

(iii) The currents of the system $H[\phi,0]$ under physical (using subscript p) observation are

$$\vec{J}_{1p}(r) = \vec{J}(r), \tag{12}$$

$$\vec{J}_{2p}(r) = \vec{J}^Q(r) + \phi(r)\circ\vec{J}(r). \tag{13}$$

In terms of $\vec{J}_{1r}(r)$ and $\vec{J}_{2r}(r)$, and Φ_1 and Φ_2, they read

$$\vec{J}_{1p}(r) = \vec{J}_{1r}(r) + \vec{J}_1(r)\circ\Phi_2(r),$$

$$\vec{J}_{2p}(r) = \vec{J}_{2r}(r) + \vec{J}_1(r)\circ\Phi_1(r) + 2\vec{J}_2(r)\circ\Phi_2(r).$$

The observed current densities are their averages in the system $H[\phi,0]$.

By the linear response theory, we have

$$\langle\vec{J}_{1p}(r)\rangle = \langle\vec{J}_1(r)\circ\Phi_2(r)\rangle_0 - \hat{K}^{11}\cdot\nabla\Phi_1 - \hat{K}^{12}\cdot\nabla\Phi_2, \tag{14}$$

$$\langle\vec{J}_{2p}(r)\rangle = \langle\vec{J}_1(r)\circ\Phi_1(r)\rangle_0 + 2\langle\vec{J}_2(r)\circ\Phi_2(r)\rangle_0 - \hat{K}^{21}\cdot\nabla\Phi_1 - \hat{K}^{22}\cdot\nabla\Phi_2, \tag{15}$$

where $\langle\cdots\rangle_0$ is the average under $H[\Phi,\Psi]$, $\hat{K}^{ij}$'s are 2×2 constant tensors [with respect to the directions of the coordinates hereafter denoted by subscripts (x,y) or (μ,ν), the superscripts 1 corresponding to particle current and 2 to heat current] determined by the Kubo formula

$$\hat{K}^{ij} = -\lim_{\Omega\to0}\mathrm{Im}\hat{\chi}^{ij}(\Omega+i0)/\Omega \tag{16}$$

with $\hat{\chi}^{ij}(\Omega+i0)$ as the retarded response function. In the bosonic Matsubara frequency Ω_m, $\hat{\chi}^{ij}$ reads

$$\hat{\chi}^{ij}(i\Omega_m) = -\frac{1}{V}\int_0^\beta d\tau e^{i\Omega_m\tau}\langle T_\tau\vec{J}_i(\tau)\vec{J}_j(0)\rangle_0 \tag{17}$$

where $\vec{J}_i=\int d\vec{r}\vec{J}_i(r)$. Now that $\langle\vec{J}_{ip}(r)\rangle$ given by Eqs. (14) and (15) are already explicitly linear in the perturbations Φ_1 and Φ_2, in calculating the averages for the constants in Eqs. (14) and (15), the equilibrium point $[\Phi,\Psi]$ can be shifted to [0,0]. To the first order of $\nabla\Phi_i$, the term $-\langle\vec{J}_i(r)\circ\Phi_j(r)\rangle_0$'s in Eqs. (14) and (15) are calculated as

$$\langle\vec{J}_i(r)\circ\Phi^j(r)\rangle_0 = \langle\vec{J}_i(r)\circ(\vec{r}\cdot\nabla\Phi_j)\rangle_0.$$

For the diagonal elements $\langle J_{ix}(r)\circ x\rangle_0 \equiv M^i_{xx}$, we have

$$M_{xx} = \langle J_x(r)\circ x\rangle_0 = i\langle\psi^\dagger(r)[\xi(r),x^2]\psi(r)\rangle_0/2 = 0,$$

$$M^Q_{xx} = \langle J^Q_x(r)\circ x\rangle_0 = i\langle\psi^\dagger(r)[\xi^2(r),x^2]\psi(r)\rangle_0/4 = 0,$$

which can be shown by expanding $\psi(r)$ in terms of the eigenstates of $\xi(r)$. For the off-diagonal elements, $\langle J_{ix}(r)\circ y\rangle_0 \equiv M^i_{xy}$, we get

$$M^i_{xy} = \langle J_{ix}(r)y\rangle_0,$$
$$= \langle[J_{ix}(r)y - J_{iy}(r)x]\rangle_0/2,$$
$$= -M^i_{yx}, \tag{18}$$

because the system is invariant under rotation around the z axis perpendicular to the plane. The element M^i_{xy} is the magnetization of the electrons. Substituting the results into Eqs. (14) and (15), we get

$$\langle\vec{J}_{1p}(r)\rangle = -\hat{N}^{11}\cdot\nabla\Phi_1 - \hat{N}^{12}\cdot\nabla\Phi_2, \tag{19}$$

$$\langle\vec{J}_{2p}(r)\rangle = -\hat{N}^{21}\cdot\nabla\Phi_1 - \hat{N}^{22}\cdot\nabla\Phi_2, \tag{20}$$

with

$$\hat{N}^{11} = \hat{K}^{11}, \tag{21}$$

$$\hat{N}^{12} = \hat{K}^{12} - \hat{M}, \tag{22}$$

$$\hat{N}^{21} = \hat{K}^{21} - \hat{M}, \tag{23}$$

$$\hat{N}^{22} = \hat{K}^{22} - 2\hat{M}^{Q}. \tag{24}$$

These forms have been obtained in Ref. 24 with a phenomenological approach using the Einstein argument and the consideration classifying the transport components in each kind of the local currents as the observable currents.

Though we consider the Dirac fermions here, the derivation above is valid for general electron systems.

For a noninteracting system such as the one considered here, the average $\langle\cdots\rangle_0$ under $H[0,0]$ reduces, in principle, to the independent single-particle problem. Using the bases of single-particle states $\{|n\rangle\}$ for a given impurity configuration, one can obtain formally the expressions for $\hat{N}^{11}$ and $\hat{N}^{12}=\hat{N}^{21}$ as

$$\hat{N}^{11} = -\int \frac{d\omega}{2\pi} f'(\omega)\hat{C}(\omega), \tag{25}$$

$$\hat{N}^{12} = \hat{N}^{21} = -\int \frac{d\omega}{2\pi} f'(\omega)\omega\hat{C}(\omega), \tag{26}$$

with

$$\hat{C}(\omega) = \frac{2\pi^2}{V}\left\langle \sum_{n\neq m} \mathrm{Re}\, \vec{j}_{nm}\vec{j}_{mn}\delta(\omega-\xi_n)\delta(\omega-\xi_m)\right\rangle_i, \tag{27}$$

where $f'(\omega)=df(\omega)/d\omega$ with $f(\omega)$ the Fermi distribution function, $\vec{j}_{nm}=\langle n|\vec{j}|m\rangle$, ξ_n is the eigenvalue of $\xi(r)$ of the state $|n\rangle$ and $\langle\cdots\rangle_i$ in Eq. (27) means the average over the impurity configurations. These forms have been obtained in Ref. 25. For the readers' convenience, we give a derivation in the Appendix. The compact form given by Eqs. (26) is so obtained because of large cancellations between $\hat{K}^{12}=\hat{K}^{21}$ and $\hat{M}$. The tensors $\hat{N}^{11}$ and $\hat{N}^{12}=\hat{N}^{21}$ are thus related via the function $\hat{C}(\omega)$.

However, Eq. (27) is not a convenient formula to start with. To proceed, one needs to derive $\hat{C}(\omega)$ in terms of the Green's function. From Eq. (17), carrying out the τ integral, we have

$$\begin{aligned}\chi^{11}_{\mu\nu}(i\Omega_m) &= \frac{1}{\beta V}\sum_n \int d\vec{r}\int d\vec{r}'\ \mathrm{Tr}\langle G(r,r',i\omega_n)J_{1\mu}\\ &\quad\times G(r',r,i\omega_n+i\Omega_m)J_{1\nu}\rangle_i\\ &\equiv \frac{1}{\beta}\sum_n P_{\mu\nu}(\omega_n,\omega_n+\Omega_m),\end{aligned} \tag{28}$$

where $G(r,r',i\omega_n)$ is the Green's function for a given impurity distribution, ω_n is the fermionic Matsubara frequency, $\langle\cdots\rangle_i$ again the average over the impurity configurations, and the function $P_{\mu\nu}(\omega_1,\omega_2)$ is so defined by the equation. Taking the analytical continuation $\Omega_m\to\Omega+i0$, we have for $\hat{N}^{11}$,

$$\begin{aligned}\hat{N}^{11} &= -\int \frac{d\omega}{2\pi} f'(\omega)\mathrm{Re}[\hat{P}(\omega^-,\omega^+)-\hat{P}(\omega^-,\omega^-)]\\ &\quad+\int \frac{d\omega}{2\pi} f(\omega)\mathrm{Re}\frac{\partial}{\partial\omega'}[\hat{P}(\omega',\omega^+)-\hat{P}(\omega^+,\omega')]_{\omega'=\omega^+}\\ &\equiv \int \frac{d\omega}{2\pi} f'(\omega)\mathrm{Re}[\hat{P}(\omega^-,\omega^-)-\hat{P}(\omega^-,\omega^+)-\hat{Y}(\omega^+)],\end{aligned}$$

where $\omega^{\pm}=\omega\pm i0$, and

$$\hat{Y}(\omega) = \int_{-\infty}^{\omega} dz\frac{\partial}{\partial z'}[\hat{P}(z',z)-\hat{P}(z,z')]_{z'=z}.$$

Therefore, we get

$$\hat{C}(\omega) = \mathrm{Re}[\hat{P}(\omega^-,\omega^+)-\hat{P}(\omega^-,\omega^-)+\hat{Y}(\omega^+)].$$

B. Hall and Nernst conductivities of Dirac fermions

The Nernst effect describes the response of the transverse current to a temperature gradient ∇T in the presence of a perpendicular magnetic field B but $\vec{E}=0$. It is reflected by the Nernst conductivity N^{12}_{xy}. Here, we study it in the limit of $B\to 0$.

In the limit of $B\to 0$, the elements N^{11}_{xx} and N^{12}_{xx} are independent of the magnetic field. They are related to the electric conductivity $\sigma=e^2N^{11}_{xx}$ and thermoelectric power $S=-N^{12}_{xx}/eTN^{11}_{xx}$ which we have given in previous works.[11,14] Though the element N^{11}_{xy} was calculated previously for studying the Hall coefficient, the function $C_{xy}(\omega)$ was not given explicitly.[11] To calculate N^{12}_{xy}, we here need to find out the explicit expression for $C_{xy}(\omega)$.

As in the calculation of the Hall conductivity in the limit of $B\to 0$,[11] by introducing the vector potential via $\vec{A}(r)=\vec{A}(q)\exp(i\vec{q}\cdot\vec{r})$ with $\vec{B}=i\vec{q}\times\vec{A}(q)$ and taking the limit $q\to 0$, one obtains $\Phi^{12}_{xy}(i\Omega_m)$ in terms of the average of the multiplication of three current operators.[26,27] As shown in Fig. 1(b), $P_{xy}(\omega_1,\omega_2)$ is obtained as

$$\begin{aligned}P_{xy}(\omega_1,\omega_2) &= \lim_{q\to 0}\frac{2e}{V}\sum_k \mathrm{Tr}\{\Gamma_x(k^-,k^+,\omega_1,\omega_2)\\ &\quad\times[G(k^+,\omega_2)\Gamma_y(k^+,\omega_2,\omega_1)V(k^+,k^-,\omega_1)\\ &\quad+V(k^+,k^-,\omega_2)\Gamma_y(k^-,\omega_2,\omega_1)G(k^-,\omega_1)]\},\end{aligned}$$

where the factor 2 stems from the spin degeneracy, $k^{\pm}=k\pm q/2$, $V(k^+,k^-,\omega)=G(k^+,\omega)\vec{\Gamma}(k^+,k^-,\omega,\omega)\cdot\vec{A}G(k^-,\omega)$, and $\Gamma_x(k,\omega_1,\omega_2)\equiv\Gamma_x(k,k,\omega_1,\omega_2)$ as given by Eq. (3). The vertex $\Gamma_\mu(k^-,k^+,\omega_1,\omega_2)$ satisfies the 4×4 matrix equation

$$\begin{aligned}\Gamma_\mu(k^-,k^+,\omega_1,\omega_2) &= \tau_3\sigma_\mu+\frac{1}{V}\sum_{k_1} n_i v_0^2(k-k_1)G(k_1^-,\omega_1)\\ &\quad\times\Gamma_\mu(k_1^-,k_1^+,\omega_1,\omega_2)G(k_1^+,\omega_2).\end{aligned} \tag{29}$$

To find out the limit of $q\to 0$, we need to expand the right hand side of Eq. (28) to the first order in q and then use $\vec{B}=i\vec{q}\times\vec{A}$. The manipulation is tedious but elementary. We only outline the key points in the derivation below.

(i) The expansions of the Green's function $G(k^{\pm},\omega)$ and the vertex functions $\Gamma_\mu(k^{\pm},\omega_1,\omega_2)$ can be easily obtained by definition. The most involved expansion is for the vertex function $\Gamma_x(k^-,k^+,\omega_1,\omega_2)$. By expanding both sides of Eq. (29) to the first order in q, one gets $\Gamma_\mu(k^-,k^+,\omega_1,\omega_2)=\Gamma_\mu(k,\omega_1,\omega_2)+\vec{\gamma}_\mu(k,\omega_1,\omega_2)\cdot\vec{q}/2$ with $\vec{\gamma}_\mu(k,\omega_1,\omega_2)$ determined by

$$\begin{aligned}\vec{\gamma}_\mu(k,\omega_1,\omega_2)=&\frac{1}{V}\sum_{k'} n_i v_0^2(k-k')G(k',\omega_1)\vec{\gamma}_\mu(k',\omega_1,\omega_2)\\&\times G(k',\omega_2)-\frac{1}{V}\sum_{k'} n_i v_0^2(k-k')\\&\times[\nabla G(k',\omega_1)\Gamma_\mu(k',\omega_1,\omega_2)G(k',\omega_2)\\&-G(k',\omega_1)\Gamma_\mu(k',\omega_1,\omega_2)\nabla G(k',\omega_2)],\end{aligned}\tag{30}$$

where ∇ means the gradient with respect to k'.

(ii) From the identity

$$\begin{aligned}&\frac{1}{V}\sum_{kk'} n_i[(\nabla_k+\nabla_{k'})v_0^2(k-k')]\mathrm{Tr}[G(k,\omega_1)\vec{\gamma}_\mu(k,\omega_1,\omega_2)\\&\quad\times G(k,\omega_2)G(k',\omega_2)\Gamma_\nu(k',\omega_2,\omega_1)G(k',\omega_1)]=0,\end{aligned}\tag{31}$$

performing the integral by part in the left hand side of Eq. (31) and using Eq. (30) and the equation for $\Gamma_\nu(k,\omega_1,\omega_2)$, we obtain

$$\begin{aligned}&\sum_k \mathrm{Tr}\{\vec{\gamma}_\mu(k,\omega_1,\omega_2)[\nabla_k G(k,\omega_2)\Gamma_\nu(k,\omega_2,\omega_1)G(k,\omega_1)\\&\quad+G(k,\omega_2)\Gamma_\nu(k,\omega_2,\omega_1)\nabla_k G(k,\omega_1)]\}\\&=-\sum_k \mathrm{Tr}\{[\nabla G(k,\omega_1)\Gamma_\mu(k,\omega_1,\omega_2)G(k,\omega_2)\\&\quad-G(k,\omega_1)\Gamma_\mu(k,\omega_1,\omega_2)\nabla G(k,\omega_2)]\nabla_k\Gamma_\nu(k,\omega_2,\omega_1)\}.\end{aligned}$$

(iii) For $V(k^+,k^-,\omega)$, using Eq. (29), we get the expansion

$$\begin{aligned}&G(k^+,\omega)\Gamma_\alpha(k^+,k^-,\omega,\omega)G(k^-,\omega)\\&=\frac{\partial}{\partial k_\alpha}G(k,\omega)+i\sigma_z\hat{\alpha}\cdot[b(k,\omega)\hat{k}\hat{\phi}-a(k,\omega)\hat{\phi}\hat{k}]\cdot\vec{q}.\end{aligned}\tag{32}$$

where ϕ is the angle of $\vec{k}$, $\hat{\phi}$ is the unit vector in ϕ direction, and the first term in the right hand of Eq. (32) comes from the Ward identity $G(k,\omega)\Gamma_\alpha(k,\omega,\omega)G(k,\omega)=\partial G(k,\omega)/\partial k_\alpha$. The coefficients $a(k,\omega)$ and $b(k,\omega)$ are determined by solving Eq. (30). Since the final result depends on their combination $a(k,\omega)+b(k,\omega)\equiv c(k,\omega)$, the function $c(k,\omega)\equiv z(k,\omega)+[g_0^2(k,\omega)-g_c^2(k,\omega)]X(k,\omega)$ is determined by following equations:

$$\begin{aligned}z(k,\omega)=&[g_0'(k,\omega)g_c(k,\omega)-g_0(k,\omega)g_c'(k,\omega)][y_0(k,\omega,\omega)\\&-y_3(k,\omega,\omega)]-g_c(k,\omega)\{g_0(k,\omega)[y_0(k,\omega,\omega)\\&+y_3(k,\omega,\omega)]+2g_c(k,\omega)y_1(k,\omega,\omega)\}/k,\end{aligned}$$

$$\begin{aligned}X(k,\omega)=&\frac{1}{V}\sum_{k'} n_i v_0^2(k-k')\{z(k',\omega)\\&+g_+(k',\omega)g_-(k',\omega)X(k',\omega)\},\end{aligned}$$

with $g'(k,\omega)=\partial g(k,\omega)/\partial k$.

Using the results given above, one gets a final expression $P_{xy}(\omega_1,\omega_2)=[Z(\omega_1,\omega_2)-Z(\omega_2,\omega_1)]/2i$ with

$$\begin{aligned}Z(\omega_1,\omega_2)=&\frac{2Be}{V}\sum_k \mathrm{Tr}\Bigg[G_1\Gamma_{x,12}\left(\frac{\partial G_2}{\partial k_x}\frac{\partial\Gamma_{y,21}}{\partial k_y}-\frac{\partial G_2}{\partial k_y}\frac{\partial\Gamma_{y,21}}{\partial k_x}\right)\\&-ic(k,\omega_1)\Gamma_{x,12}G_2\Gamma_{y,21}\sigma_z\Bigg],\end{aligned}$$

where $\Gamma_{x,ij}=\Gamma_x(k,\omega_i,\omega_j)$ (and the same meaning for $\Gamma_{y,ij}$), $G_j=G(k,\omega_j)$. Since $P_{xy}(\omega^-,\omega^-)=0$, we have $C_{xy}(\omega)=\mathrm{Im}\,Z(\omega^-,\omega^+)+\mathrm{Re}\,Y_{xy}(\omega^+)$, and

$$\mathrm{Re}\,Y_{xy}(\omega)=\int_{-\infty}^{\omega} dz\,\mathrm{Im}\,R(z)$$

with $R(\omega)=\frac{\partial}{\partial\omega'}[Z(\omega',\omega)-Z(\omega,\omega')]_{\omega'=\omega}$.

Knowing the function $C_{xy}(\omega)$, we can calculate N_{xy}^{11} and the Nernst conductivity N_{xy}^{12} according to Eqs. (25) and (26), respectively. Since we are interested in the low temperature cases, we here give their expressions in the limit of $T\to 0$. They are

$$N_{xy}^{11}=\frac{1}{2\pi}\mathrm{Im}\,Z(0^-,0^+)-\int_0^\infty\frac{dz}{2\pi}\mathrm{Re}\,R(iz),\tag{33}$$

$$N_{xy}^{12}=\frac{\pi T^2}{6}\mathrm{Im}\left[\frac{\partial}{\partial\omega}Z(\omega^-,\omega^+)\big|_{\omega=0}+R(0^+)\right],\tag{34}$$

In obtaining N_{xy}^{12}, the use of the expanding $C_{xy}(\omega)\approx C_{xy}(0)+\omega C_{xy}'(0)$ has been made. In addition, the z integral in $\mathrm{Re}\,Y_{xy}(0)$ reduces to the path-integral around the negative axis $(-\infty,0)$. Because $R(z)$ is an analytical function (by definition) in the upper and lower z plane, respectively, the integral path has been deformed to the imaginary axis, giving rise to the last term in Eq. (33). The integral along the imaginary axis is simple to handle for the numerical calculation since there is no singularity in the Green's function. If this term is neglected, the expression for the Hall conductivity $\sigma_{xy}=e^2N_{xy}^{11}$ will reduce to the form as in the previous work.[11] The fact that the contribution from this term is negligible will be checked later. In analogous to the thermoelectric power $S=E_x/(\nabla T)_x$, we define $S_{xy}=E_x/(\nabla T)_y$ which describes the production of the transverse electric field due to a temperature gradient in the absence of current flow. By setting $\langle J_{1p}(r)\rangle=0$ in Eq. (19), S_{xy} is obtained as

$$S_{xy}=-(S\sigma_{xy}+eN_{xy}^{12}/T)/\sigma.\tag{35}$$

Clearly, S_{xy} includes two parts. The quantity $S\sigma_{xy}$ describe the process for the response of the transverse electric field to ∇T through the way: because there exists a longitudinal electric field (with the magnitude proportional to the thermoelectric power S) due to the temperature gradient, the current could flow transversely by the Hall process. In different to

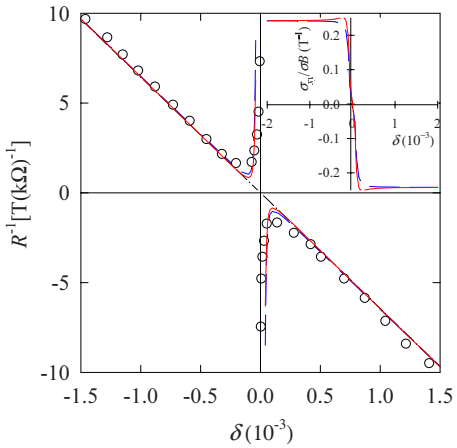

FIG. 2. (Color online) The inverse Hall coefficient R^{-1} (in unit of 10^{-3} Tesla/Ohm) as a function of δ. The solid (red) and dashed (blue) lines correspond, respectively, to the calculations with and without the contribution from the last term in Eq. (33) [Inset, the corresponding results for σ_{xy} normalized by σB with B is in unit of Tesla (T)]. Dot-dashed line: classical result. Symbols: experimental data (Ref. 12).

this, eN_{xy}^{12}/T implies an additional transverse field due to a transverse current response directly to ∇T.

III. RESULTS

The functions $g_{0,c}(k,\omega)$ and $y_j(k,\omega_1,\omega_2)$ and their derivatives with respect to k and ω are involved in the calculation. In a recent work,[14] we have described how to numerically solve the corresponding integral equations to determine these functions. In our numerical calculation, we take the impurity density as $n_i=1.15\times10^{-3}a^{-2}$ (with a as the lattice constant of graphene) the same as in our previous works for reproducing the experimental measurements of the electric conductivity, the Hall coefficient and the thermoelectric power.[9,11,14]

The numerical results for the Hall conductivity σ_{xy} and the inverse Hall coefficient $R^{-1}=B\sigma^2/\sigma_{xy}$ as functions of the carrier concentration δ (doped carrier per carbon atom) are shown in Fig. 2. The calculations with and without the last term in Eq. (33) for both results of R^{-1} and σ_{xy} (normalized by σB in inset of Fig. 2) are almost indistinguishable. This fact means that the contribution from the last term in Eq. (33) is negligible small. The experimental data[12] (symbols) and the classical prediction $R^{-1}=-nec$ are also plotted in Fig. 2 for comparison. As we have stated previously, the divergence of R^{-1} at $\delta=0$ stems from the vanishing of σ_{xy} while the conductivity σ remains finite.

At low T, the Nernst conductivity N_{xy}^{12} is proportional to T^2. In Fig. 3, we depict eN_{xy}^{12}/T^2B as a function of the electron doping concentration δ. Because of the electron-hole symmetry, N_{xy}^{12} is even for $\delta\to-\delta$. N_{xy}^{12} comes mainly from the first term in the square bracket in Eq. (34). This is similar to the case as in N_{xy}^{11}. The derivation $\partial Z(\omega^-,\omega^+)/\partial\omega|_{\omega=0}$ is very delicate. Here, it cannot be considered simply as $\propto\partial\sigma_{xy}(\mu)/\partial\mu$ because the scattering potential here depends strongly on the electron doping concentration. For comparison, we also depict the result for $S\sigma_{xy}/TB$ in Fig. 3. In the regime of δ studied here, the magnitudes of both quantities are overall the same. But at low δ, eN_{xy}^{12}/T^2B is bigger than $S\sigma_{xy}/TB$. In the absence of the electric field, the transverse current density is solely determined by $J_x=-N_{xy}^{12}(\nabla T)_y/T$.

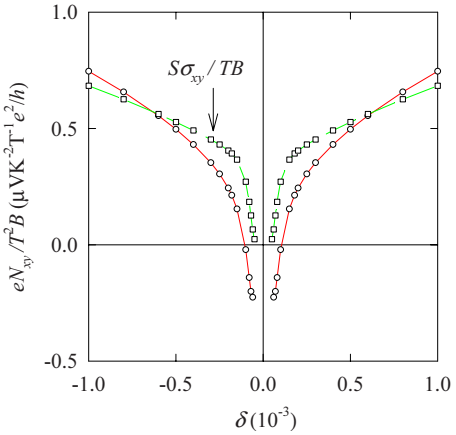

FIG. 3. (Color online) The coefficient eN_{xy}^{12}/T^2B (red line with circles)(in unit of $\mu VK^{-2}T^{-1}e^2/h$ with h the Planck constant) as a function of δ. The dashed line (green with squares) is $S\sigma_{xy}/TB$ for comparison.

In Fig. 4, we show the transverse thermoelectric power S_{xy} divided by TB as functions of δ at the average impurity densities $n_i=1.15\times10^{-3}a^{-2}$ and $n_i=1.6\times10^{-3}a^{-2}$. S_{xy} is a superposition of $S\sigma_{xy}$ and eN_{xy}^{12}/T. As a result, S_{xy} is linear in T and B. At large carrier doping, both of $S\sigma_{xy}$ and eN_{xy}^{12}/T have about the same contribution to S_{xy}. While at low doping, eN_{xy}^{12}/T is predominant. S_{xy} changes sign at low doping because eN_{xy}^{12}/T does. The factor $S\sigma_{xy}+eN_{xy}^{12}/T$ of S_{xy} decreases quickly as δ decreasing at low δ regime. Its slop normalized by a negative constant ($\sim-\sigma/\delta$, $\sigma\propto\delta$ at large δ and is flat at very low doping) is almost the behavior of S_{xy}. The dip in S_{xy} corresponding to the maximum of the slop. So far there exist no measurements of the Nernst effect of Dirac

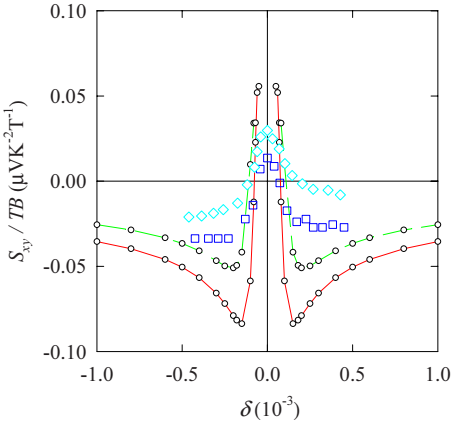

FIG. 4. (Color online) The coefficient S_{xy}/TB of transverse thermoelectric power (in unit of $\mu VK^{-2}T^{-1}$) as a function of δ. The solid (red with small symbols) and dashed (green) lines are the calculations at impurity densities $n_i=1.15\times10^{-3}a^{-2}$ and $1.6\times10^{-3}a^{-2}$, respectively. The squares (blue at $B=1$ Tesla) and diamonds (cyan at $B=3$ Tesla) are the experimental results of Ref. 2.

fermions in a weak magnetic field. But for the purpose to make a qualitative comparison, the experimental results for the transverse thermoelectric power of Ref. 2 with the magnetic fields $B=1$ Tesla (T) (squares) and 3 T (diamonds) are plotted. To judge whether the magnetic field is weak or not, we consider the ratio between the overall magnitude of spacing $\epsilon_0=v\sqrt{2eB/\hbar c}$ of the Landau level $E_n=\pm\epsilon_0\sqrt{|n|}$ and the Fermi energy $E_F=(4\pi\delta)^{1/2}v/3^{1/4}a$. The weak magnetic field means that the ratio ϵ_0/E_F is much less than unity. Clearly, for a magnetic field, this ratio is smaller at larger carrier doping δ. Here the ratio is about 0.35 for a doping level at 1.0×10^{-4} and $B=1$ T. Though the strengths of these magnetic fields could not be regarded as weak, the data indicate a tendency toward to the theoretical prediction as the magnetic field is decreasing.

We note that the present calculations fit nicely to the Hall data (shown in Fig. 2) but not to the Nernst effect (the transverse thermoelectric power shown in Fig. 4). As is known, the Nernst effect is a more sensitive probe of the impurity scatterings than the electric conductivity and the Hall conductivity since it detects not only the current-current correlation function but also its derivative. One of the reasons for the discrepancy between the present calculation and the experiment measurement might be due to other possible sources of impurity scatterings besides the charged scatters. Because of the sensitive dependency of the impurity scatterings, the measurements of the Nernst effect as well as the longitudinal thermoelectric power may vary from sample to sample. As we noted from the data for the latter one, there are obvious differences between the experimental results (see Fig. 8 of Ref. 14) due to other impurity scatterings. For the present case besides the charged impurities, the short range scatters may be also relevant for the physics close to the Dirac point. However, it is not the predominant component because it is not able to produce the linear carrier density dependency of the electric conductivity.

The present calculation is based on the assumption that the impurities are randomly distributed. Actually, at low carrier doping, graphene is an inhomogeneous system due to the impurity correlations in the substrate as observed by experiment.[28–31] It seems there exist the electron and hole puddles. Nonetheless, in each puddle, the average number of the carrier is less than unity (or in the order of unity). Moreover, the mean free path of the electrons is much longer than the length scale (~tens nanometers) of the puddles. Within a mean free path, a carrier can transfer through many such puddles. The puddles can be thus regarded as the microscopic wrinkles. In addition, at low carrier concentration, there exists significant quantum coherence between the upper- and lower-band states,[32] resulting in the minimum electric conductivity,[9,33–35] the unconventional behaviors of the inverse Hall conductivity[11,12] and the thermoelectric power.[1–3,14] Therefore, the carrier must be treated quantum mechanically and the physics by the present approach is qualitatively correct. However, at very low carrier doping, because of weak screening, the charged impurity scatterings are very strong and the SCBA thereby becomes quantitatively incorrect. The validity of SCBA can be judged by the ratio of the Fermi level broadening γ and the Fermi energy E_F. The SCBA is good if $\gamma/E_F<1$. In the present study for the impurity density $n_i=1.15\times10^{-3}a^{-2}$, $\gamma/E_F<1$ when $\delta>1.0\times10^{-4}$.[36] Below this carrier doping, the results given here have only the qualitative meanings.

Finally, we compare our calculation with the semiclassical Boltzmann theory. By the Boltzmann theory within the relaxation time-τ approximation, the function g_k describing the difference between the disturbed distribution function and the Fermi distribution function f is determined by[37]

$$g_k=\tau\frac{\partial f}{\partial\xi_k}\vec{v}\cdot\left(\frac{\xi_k}{T}\nabla T+e\vec{E}\right)+\tau e(\vec{v}\times\vec{B})\cdot\nabla_k g_k \qquad (36)$$

using units of $c=\hbar=1$ again. Here $\xi_k=vk-\mu$ for electrons in graphene. From Eq. (36), one obtains

$$\sigma=\tau v e^2 k_F/\pi,$$

$$\sigma_{xy}=B\tau^2 v^2 e^3/\pi,$$

$$N_{xx}^{12}=T^2\tau\pi/3.$$

The inverse Hall coefficient is $B\sigma^2/\sigma_{xy}=ne$. The Mott relation[38] is given by $N_{xx}^{12}=T^2\pi^2\sigma'/3e^2$ with $\sigma'=\tau e^2/\pi$. However, using the Mott relation, one obtains zero Nernst conductivity $N_{xy}^{12}=0$ because σ_{xy} is constant independent of the chemical potential μ. This is different from the present result that the Nernst conductivity is in the order of $S\sigma_{xy}T/e$ as shown in Fig. 3.

IV. SUMMARY

In summary, we have derived the formula for the transport coefficients for the Dirac fermions in graphene in the presence of the temperature gradient, the electric field and the magnetic field. The derivation is valid for general electron systems. It is different from the usual perturbation process with the external dynamic potentials applied (in that case the original equilibrium state is unchanged) since the perturbation due to turning on the temperature gradient shifts the equilibrium state. The physical observed system is in the perturbed state with respect to this equilibrium state.

On the basis of self-consistent Born approximation, we have studied the Nernst effect of the Dirac fermions under the charged impurity scatterings and weak magnetic field in graphene. The transverse thermoelectric power is closely related with the Hall conductivity and the longitudinal thermoelectric power for which the theory has been shown to be in good agreement with the experiment. The Nernst conductivity is dealt with the similar approach as for the Hall conductivity. The present calculation is a prediction to the Nernst conductivity of the Dirac fermions in graphene under a weak magnetic field.

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APPENDIX: DERIVATION OF EQS. (25) AND (26)

We here give a derivation of Eqs. (25) and (26). Consider the response function $\chi_{AB}(\tau)$ defined by

$$\chi^{AB}(\tau) = -\frac{1}{V}\langle T_\tau A(\tau)B(0)\rangle_0, \tag{A1}$$

where A and B (of Hermitians) can be either the current or the heat current operator. Using the basis of the single-particle states $\{|n\rangle\}$, we have

$$\begin{aligned} A(\tau) &= \int d\vec{r} e^{H[0,0]\tau}\psi^\dagger(r)a(r)\psi(r)e^{-H[0,0]\tau} \\ &= \sum_{nm} e^{(\xi_n-\xi_m)\tau}a_{nm}c_n^\dagger c_m, \end{aligned} \tag{A2}$$

where $a_{nm}=\langle n|a|m\rangle$ and $c_n^\dagger$ (c_n) creates (annihilates) a particle in state $|n\rangle$. The function $\chi_{AB}(\tau)$ is written as

$$\chi^{AB}(\tau) = -\frac{1}{V}\left\langle \sum_n f_n a_{nn}b_{nn} + \sum_{n\neq m} f_n(1-f_m)e^{(\xi_n-\xi_m)\tau}a_{nm}b_{mn}\right\rangle_i,$$

where $f_n=\langle c_n^\dagger c_n\rangle$ is the Fermi distribution function. Taking the Fourier transform, we get

$$\begin{aligned} \chi^{AB}(i\Omega_\ell) = -\frac{1}{V}\Bigg\langle & \sum_n f_n a_{nn}b_{nn}\beta\delta_{\ell,0} \\ & - \sum_{n\neq m}\frac{f_n-f_m}{i\Omega_\ell+\xi_n-\xi_m}a_{nm}b_{mn}\Bigg\rangle_i, \end{aligned} \tag{A3}$$

where $\beta=1/T$. Taking the analytical continuation $i\Omega_\ell\to\Omega+i0$ [for which the first term in the braces in Eq. (A3) can be disregarded because of $\Omega_\ell>0$], we have

$$\begin{aligned} \mathrm{Im}\,\chi^{AB,r}(\Omega) &= \frac{1}{V}\mathrm{Im}\left\langle \sum_{n\neq m}\frac{f_n-f_m}{\Omega+\xi_n-\xi_m+i0}a_{nm}b_{mn}\right\rangle_i \\ &= \frac{1}{V}\Bigg\langle \sum_{n\neq m}(f_n-f_m)\Bigg[\frac{\mathrm{Im}(a_{nm}b_{mn})P}{\Omega+\xi_n-\xi_m} \\ &\quad - \pi\,\mathrm{Re}(a_{nm}b_{mn})\delta(\Omega+\xi_n-\xi_m)\Bigg]\Bigg\rangle_i \\ &= \frac{1}{V}\Bigg\langle \sum_{n\neq m}(f_n-f_m)\Bigg[\frac{\mathrm{Im}(a_{nm}b_{mn})\Omega P}{\Omega^2-(\xi_n-\xi_m)^2} \\ &\quad - \pi\,\mathrm{Re}(a_{nm}b_{mn})\delta(\Omega+\xi_n-\xi_m)\Bigg]\Bigg\rangle_i, \end{aligned} \tag{A4}$$

where P means taking the principle value in the summation, and in the P term the exchange $n\leftrightarrow m$ and the use of $\mathrm{Im}(a_{mn}b_{nm})=-\mathrm{Im}(a_{nm}^\dagger b_{mn}^\dagger)=-\mathrm{Im}(a_{nm}b_{mn})$ have been made in the last equality. Substituting the result of Eq. (A4) into Eq. (16), we obtain

$$\begin{aligned} K^{AB} = \frac{1}{V}\Bigg\langle \sum_{n\neq m}\Bigg[& \mathrm{Im}(a_{nm}b_{mn})\frac{(f_n-f_m)P}{(\xi_n-\xi_m)^2} \\ & - \pi\,\mathrm{Re}(a_{nm}b_{mn})f_n'\delta(\xi_n-\xi_m)\Bigg]\Bigg\rangle_i, \end{aligned} \tag{A5}$$

where $f_n'=df_n/d\xi_n$.

Note first, the desired forms given by Eqs. (25) and (26) are obtained from the contribution from the last term in the square brackets in Eq. (A5).

Second, for the diagonal elements, the first term in the square brackets in Eq. (A5) vanishes. In the present case, A and B are vectors. For the diagonal elements, $a_{x,nm}\propto b_{x,nm}$ and $a_{x,nm}b_{x,mn}$ is real.

Therefore, in following, we will consider only the contribution from the first term in the square brackets in Eq. (A5) for the case of off-diagonal elements. Denote it as

$$R(A,B) = \frac{1}{V}\sum_{n\neq m}\frac{(f_n-f_m)P}{(\xi_n-\xi_m)^2}\mathrm{Im}(a_{nm}b_{mn}), \tag{A6}$$

dropping the symbol of the average $\langle\cdots\rangle_i$ for briefness. We only need to prove that the off-diagonal element of $R(A,B)$ is cancelled by the corresponding matrix element of M_{xy}^i given by Eq. (18).

(i) $A=B=\vec{J}$. Using $\vec{j}=i[\xi(r),\vec{r}]$, $R(\vec{J},\vec{J})$ reads

$$\begin{aligned} R(J_x,J_y) &= \frac{1}{V}\sum_{n\neq m}(f_n-f_m)\mathrm{Im}(x_{nm}y_{mn}) \\ &= \frac{1}{V}\sum_n f_n\,\mathrm{Im}\langle n|[x,y]|n\rangle = 0. \end{aligned}$$

(ii) $A=\vec{J}$ and $B=\vec{J}^Q$ with $\vec{J}^Q(r)=\{\vec{j}(r),\xi(r)\}/2$. Using $\vec{j}_{nm}^Q=\vec{j}_{nm}(\xi_n+\xi_m)/2=i\vec{r}_{nm}(\xi_n^2-\xi_m^2)/2$, one gets

$$R(J_x,J_y^Q) = \frac{1}{2V}\sum_{n\neq m}(\xi_n+\xi_m)(f_n-f_m)\mathrm{Im}(x_{nm}y_{mn}).$$

On the other hand, we note

$$\begin{aligned} -M_{xy} &= -\frac{1}{2V}\sum_{nm} f_n[j_{x,nm}y_{mn}-j_{y,nm}x_{mn}] \\ &= \frac{1}{V}\sum_{nm} f_n(\xi_n-\xi_m)\mathrm{Im}(x_{nm}y_{mn}) \\ &= \frac{1}{2V}\sum_{nm}(f_n+f_m)(\xi_n-\xi_m)\mathrm{Im}(x_{nm}y_{mn}), \end{aligned}$$

where we have made use of the exchange $n\leftrightarrow m$ in the last equality. Therefore, we have

$$\begin{aligned} R(J_x,J_y^Q)-M_{xy} &= \frac{1}{V}\sum_{nm}(f_n\xi_n-f_m\xi_m)\mathrm{Im}(x_{nm}y_{mn}) \\ &= \frac{1}{V}\sum_n f_n\xi_n\,\mathrm{Im}\langle n|[x,y]|n\rangle = 0. \end{aligned} \tag{A7}$$

(iii) The case of $A=\vec{J}^Q$ and $B=\vec{J}$ is the same as (ii) and $N^{12}=N^{21}$.

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