Hall Resistance in the Hopping Regime: A "Hall Insulator"?

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The Hall conductivity and resistivity of strongly localized electrons at low temperatures and small magnetic fields are obtained. The results depend on whether one uses the conductivity or resistivity tensor to obtain the macroscopic Hall resistivity. In the second case the Hall resistivity always *diverges* exponentially as $T \to 0$. However, when the Hall resistivity is derived from the conductivity, the resulting temperature dependence is sensitive to the disorder configuration, and the Hall resistivity may approach a constant value as $T \to 0$. This is the Hall insulating behavior. It is argued that for strictly dc conditions the transport quantity that should be averaged is the resistivity, and this shows no Hall insulating behavior.

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Recently it has been stated [1–3] that the zero-temperature Hall resistivity $\rho_{xy}(\omega)$ of noninteracting electrons in the insulating regime remains finite as the frequency $\omega \to 0$. This puzzling result was derived from the Kubo formula for the frequency-dependent conductivity. It was found that at zero temperature the disorder-averaged $\sigma_{xy}(\omega)$ of the Anderson insulator vanishes at low frequencies proportionally to ω^2. Since to leading order in ω the longitudinal conductivity $\sigma_{xx} \sim i\omega\varepsilon_0$, where ε_0 is the dielectric constant, this yielded that the Hall resistivity $\rho_{xy} \sim \sigma_{xy}/(\sigma_{xx}^2 + \sigma_{xy}^2)$ approaches a constant in the small-frequency, zero-temperature limit.

In the present Letter we show that the above prediction of a "Hall insulator" came about because of an averaging procedure inappropriate for dc conditions. We examine the Hall resistance of strongly localized electrons at finite temperatures, and find that the correct dc result diverges. As in Ref. [3], we consider the problem using the Holstein model [4] for the Hall effect in a system with localized states. The smallest cluster that yields a Hall resistivity contains a triangle of three sites. The correct way to obtain the macroscopic Hall resistivity from the single-triangle conductivity tensor is different for ac or dc measurements. Under ac conditions, one has to average the conductivity tensor, and a Hall insulator behavior is possible. This procedure, at zero temperature, has been followed in Refs. [1–3]. On the other hand, as discussed below, under dc conditions one has to first invert the conductivity tensor and then to average over different orientations of the triangle. In this case the macroscopic Hall resistivity diverges as the temperature tends to zero, similar to the findings of Friedman and Pollak [5]. Thus our Letter presents a simple explanation for the nonexistence of the dc Hall insulator.

The previous [1–3] discussions of the zero-temperature Hall resistivity were all based on the ensemble averaged Kubo formula, which yields that $\bar{\sigma}_{xx} = \bar{\sigma}_{yy}$ and that $\bar{\sigma}_{xy}$ vanishes proportionally to the magnetic field (the overbar above the quantity indicates ensemble averaging). However, before averaging, σ_{xy} includes a field-independent term. We show that in the strongly localized regime this term is comparable in magnitude to σ_{xx} and σ_{yy}. This leads to delicate cancellations when the local (unaveraged) conductivity tensor is inverted to obtain the resistivity, and consequently to the very different temperature dependences described above.

It is convenient to investigate the transport properties of electrons in the hopping regime by constructing the rate equations for the electron distributions, utilizing the electronic transition probabilities between localized states. One has

$$\frac{dn_i}{dt} = \sum_j (P_{ji} - P_{ij}), \tag{1}$$

in which n_i is the electronic population of site i (the term "site" is used for a localized state) and P_{ij} is the rate of the population decay by phonon-assisted hopping into site j. A delicate point is the dependence of the rate on the magnetic field, H. As was shown by Holstein [4], this is due to interference of the "direct" amplitude to hop from i to j, with the indirect amplitude via a third site ℓ, $i \to \ell \to j$. The magnetic field dependence of P_{ij} then necessitates the consideration of at least three sites. To write this rate explicitly, we employ the Holstein model [6] for the electron-phonon interaction, in which the ion displacements are coupled to the local (site) density of the electrons, and denote by ϵ_i the single-particle energies of the localized states, which are assumed to be randomly distributed, and by J_{ij} the overlap of two wave functions localized at sites i and j. The strong localization regime is characterized by [4,7] $|J_{ij}| \ll |\epsilon_{ij}|$, $\epsilon_{ij} = \epsilon_i - \epsilon_j$.

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One finds

$$P_{12} = P_{12}^{\rm dir} + P_{12}^{\rm indir}. \quad (2)$$

Here $P_{12}^{\rm dir}$ arises from the direct amplitude alone and is independent of the magnetic field,

$$P_{12}^{\rm dir} = n_1(1 - n_2)\,|J_{12}|^2 \int_{-\infty}^{\infty} dt\, e^{i\epsilon_{12}t} e^{2[g(t)-g(0)]},$$

$$g(t) = \sum_q \frac{|v_q|^2}{\omega_q^2}\,[(1 + N_q)e^{-i\omega_q t} + N_q e^{i\omega_q t}], \quad (3)$$

where ω_q is the phonon frequency, v_q is the interaction matrix element, and $N_q = 1/(e^{\beta\omega_q} - 1)$ ($\hbar = 1$). $P_{12}^{\rm indir}$ comes from the interference between the direct ($1 \to 2$) and the indirect ($1 \to 3 \to 2$) amplitudes

$$P_{12}^{\rm indir} = n_1(1 - n_2)\,|J_{12}J_{23}J_{31}|2\Re e^{i\phi_{132}} \times \int_0^{\infty} dt_2 \int_{-\infty}^{\infty} dt_1\, e^{g(t_1)+g(t_2)+g(t_1+t_2)-3g(0)} \times [(1 - n_3)e^{i\epsilon_{12}t_1 + i\epsilon_{13}t_2} - n_3 e^{i\epsilon_{12}t_1 + i\epsilon_{32}t_2}], \quad (4)$$

and contains the occupation of site 3, n_3, and the magnetic phase, ϕ_{132},

$$\phi_{132} = \frac{e}{c}\vec{H}\cdot\vec{S},$$

$$\vec{S} = (\vec{R}_1 \times \vec{R}_3 + \vec{R}_3 \times \vec{R}_2 + \vec{R}_2 \times \vec{R}_1)/2. \quad (5)$$

Here $\vec{R}_i$ is the radius vector of site i, and $\vec{S}$ is the vectorial area of the triangle. The field-dependent part of P_{12} includes a term even in the field (proportional to $\cos\phi_{132}$) and a term odd in it (proportional to $\sin\phi_{132}$). The first gives a correction to the direct rate and will be discarded henceforth. Obviously it is the term odd in the field that is capable of producing the Hall resistance.

We now apply the rate equation (1) to a group of three sites, 1, 2, and 3, to obtain the current driven by an external ac electric field $\vec{E}$ of small frequency ω. In the presence of an electric field the occupations n_i will be modified in a way that can be expressed by changes $\delta\mu_i$ in the local chemical potentials [7]

$$n_i = n_i^0 - \beta n_i^0(1 - n_i^0)\delta\mu_i, \quad (6)$$

where n_i^0 is the Fermi distribution. Also, the rate P_{ij} that depends on $\epsilon_i - \epsilon_j$ is changed to depend on $\epsilon_i - \epsilon_j + e\vec{E}\cdot(\vec{R}_i - \vec{R}_j)$. This way one obtains the response of the system to the local electrochemical potential differences [7]

$$\zeta_{ij} = \frac{\delta\mu_i - \delta\mu_j}{e} - \vec{E}\cdot\vec{R}_{ij}, \quad \vec{R}_{ij} = \vec{R}_i - \vec{R}_j. \quad (7)$$

Explicit calculations of Eqs. (3) and (4) yield

$$e\frac{dn_1}{dt} = e(P_{21} - P_{12} + P_{31} - P_{13}) = G_{12}\zeta_{12} + G_{31}\zeta_{13} + \sin\phi\,\Gamma\zeta_{23}, \quad (8)$$

with analogous expressions for the other two sites, where ϕ is the magnetic flux enclosed in the triangle. These equations determine the electrochemical potential differences, ζ_{ij}. However, in practice one does not have to solve for the ζ_{ij}s since Eq. (8) gives the current [7] [see Eq. (13) below]. In (8), G_{ij} is the conductance of the bond ij, arising from the direct rate,

$$G_{ij} = G_{ji} = e^2\beta n_i^0(1 - n_j^0)\,|J_{ij}|^2 \int_{-\infty}^{\infty} dt\, e^{i\epsilon_{ij}t} e^{2[g(t)-g(0)]}. \quad (9)$$

At low temperatures [7] and for weak electron-phonon coupling, the bond conductance becomes

$$G_{ij} = e^2\beta|J_{ij}|^2 \frac{v^2\mathcal{N}(|\epsilon_{ij}|)}{|\epsilon_{ij}|^2}\, e^{-\beta(|\epsilon_i|+|\epsilon_j|+|\epsilon_i-\epsilon_j|)/2}, \quad (10)$$

where $\mathcal{N}$ is the phonon density of states. (Energies are measured from the Fermi energy.) The interference process leads to the phase-dependent term, with

$$\Gamma = e^2\beta n_1^0(1 - n_2^0)\,|J_{12}J_{23}J_{31}| \times \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2\, e^{g(t_1)+g(t_2)+g(t_1+t_2)-3g(0)} \times e^{i\epsilon_{12}t_1}[(1 - n_3^0)e^{i\epsilon_{13}t_2} + n_3^0 e^{i\epsilon_{32}t_2}]. \quad (11)$$

Expanding this expression for weak electron-phonon interaction and using (10) we find that Γ can be written in the form

$$\Gamma = \frac{|J_{12}J_{23}J_{31}|}{4e^2\beta}\left(\frac{G_{31}G_{12}}{|J_{31}J_{12}|^2 n_1^0(1 - n_1^0)} + \frac{G_{12}G_{23}}{|J_{12}J_{23}|^2 n_2^0(1 - n_2^0)} + \frac{G_{23}G_{31}}{|J_{23}J_{31}|^2 n_3^0(1 - n_3^0)}\right). \quad (12)$$

This expresses the fact that the indirect rate involves *two* scattering events by the phonons [4,8]. The temperature dependence of Γ, at low temperatures, can be obtained from (10), using $n_i^0(1 - n_i^0) \sim \exp[-\beta|\epsilon_i|]$.

The current density is given by

$$\vec{j} = e\sum_i \frac{dn_i}{dt}\vec{R}_i. \quad (13)$$

If one now introduces the effective field $\vec{E}_{\rm eff}$, which produces the electrochemical potential,

$$\zeta_{ij} = -\vec{E}_{\rm eff}\cdot\vec{R}_{ij},$$

$$\vec{E}_{\rm eff} = \frac{1}{2S_z}[\zeta_{23}\vec{R}_1 + \zeta_{31}\vec{R}_2 + \zeta_{12}\vec{R}_3]\times\hat{z}, \quad (14)$$

the current density becomes

$$\vec{j} = \overleftrightarrow{\sigma}\vec{E}_{\rm eff},$$

$$\overleftrightarrow{\sigma} = \vec{R}_{12}\vec{R}_{12}G_{12} + \vec{R}_{23}\vec{R}_{23}G_{23} + \vec{R}_{31}\vec{R}_{31}G_{31} + \Gamma\sin\phi(\vec{R}_{23}\vec{R}_1 + \vec{R}_{31}\vec{R}_2 + \vec{R}_{12}\vec{R}_3). \quad (15)$$

A remarkable observation is that the part of $\vec{j}$ that is proportional to the magnetic field is perpendicular to the

effective electric field. In deriving the expression for $\overleftrightarrow{\sigma}$ it was assumed that the triangle lies in the x-y plane, perpendicular to $\hat{z}$. Note that $\vec{E}_{\rm eff}$, Eq. (14), is invariant to the choice of the coordinate origin.

The current response to the electrochemical potential difference is our central result. From (15) one finds for the conductivity tensor

$$\sigma_{xx} = (R^x_{12})^2 G_{12} + (R^x_{23})^2 G_{23} + (R^x_{31})^2 G_{31} \qquad (16)$$

(σ_{yy} is given upon replacing R^x_{ij} by R^y_{ij}), and

$$\sigma^{xy}_{yx} = R^x_{12}R^y_{12}G_{12} + R^x_{23}R^y_{23}G_{23} + R^x_{31}R^y_{31}G_{31} \pm 2S_z\Gamma\sin\phi\,. \qquad (17)$$

It is straightforward to check that these results follow very simply also from the Kubo formulation, with the understanding that it yields the response to the *effective field* $\vec{E}_{\rm eff}$:

$$\sigma_{ij}(\omega) = \frac{ie^2\omega}{\rm Vol}\sum_{m,n}\left[\frac{\langle m|x_i|n\rangle\langle n|x_j|m\rangle}{\omega - \epsilon_{nm} + i\eta} - \frac{\langle m|x_j|n\rangle\langle n|x_i|m\rangle}{\omega + \epsilon_{nm} + i\eta}\right](f_m - f_n)\,, \qquad (18)$$

where η is a positive infinitesimal, $|m\rangle$ and $|n\rangle$ are eigenstates, and f_n, f_m their populations. We generalize the derivation of [3] to include electron-phonon coupling and consider the $\omega > 0$, $\omega \to 0$ limit. For example, to get the leading, (1,2)-type terms, in (16) and in the first line of (17), the two relevant states are $|1\rangle$: electron in site 1 with a phonon bath in equilibrium and $|2,q\rangle$: electron in site 2, with the same minus one phonon in state q, where $\omega_q \cong \epsilon_{21}$. Thus, the approximate eigenstates are

$$|m\rangle = |1\rangle + \sum_q \frac{J_{12}\nu_q}{\epsilon_{12} + \omega_q}|2,q\rangle\,, \qquad (19)$$

$$|n\rangle = |2,q\rangle + \frac{J_{21}\nu^*_q}{\epsilon_{21} - \omega_q}|1\rangle\,, \qquad (20)$$

where $\nu_q = v_q\sqrt{N_q}/\omega_q$, and

$$\langle n|x|m\rangle \cong \frac{J_{12}|\nu_q|^2 R^x_{12}}{\omega}\,. \qquad (21)$$

Using Eq. (18), this produces the first term in (16),

$$\frac{1}{\rm Vol}(R^x_{12})^2 e^2\beta J^2_{12}\sum_q \frac{|v_q|^2}{\omega_q^2}N_q\pi\delta(\omega - \epsilon_{21} - \omega_q) = \pi\beta e^2 J^2_{12}(R^x_{12})^2\frac{|v_q|^2}{\omega_q^2}N_q\mathcal{N}(\epsilon_{21})|_{\omega_q=\epsilon_{21}}\,. \qquad (22)$$

Using the expression similar to (20) for the matrix elements of y yields the first term in (17). The (2,3) and (1,3) terms in Eqs. (16) and (17) are similarly obtained. To get the Γ term in (17), one has to mix in Eq. (19) also the state $|3,q,q'\rangle$ (i.e., $|3\rangle$ minus the phonons q and q') in two ways.

(a) A straightforward correction $\sum_{q'}[J_{13}\nu_q\nu_{q'}/(\epsilon_{13} + \omega_q + \omega_{q'})]|3,q,q'\rangle$, which is first order in J but second order in the electron-phonon interaction. It will be dominated by the "resonant" contribution (q' such that $\omega_q + \omega_{q'} = \epsilon_{31}$) and by the nonresonant contribution given by $q' = q''$ such that $\omega_{q''} = \epsilon_{32}$.

(b) The Holstein contribution, the mixing of $|2,q\rangle$ via the intermediate state $|3,q,q'\rangle$: $\sum_{q'}[J_{13}J_{32}\nu_q|\nu_{q'}|^2/(\epsilon_{13} + \omega_q + \omega_{q'})(\epsilon_{12} + \omega_q)]|2,q\rangle$. Here, as found by Holstein, the resonant contribution with $\epsilon_{31} = \omega_q + \omega_{q'}$ will yield the needed phase to have a term odd in the magnetic field (the Γ term) in (17). Putting all the above together we find within the required accuracy

$$\langle n|x|m\rangle \cong \frac{J_{12}\nu_q}{\omega}R^x_{12} - 2i\pi\delta(\epsilon_{31} - \omega_q - \omega_{q'}) \times J_{13}J_{32}\nu_q|\nu_{q'}|^2\left[R^x_3 - \frac{R^x_1 + R^x_2}{2}\right]. \qquad (23)$$

Using this in Eq. (18) produces the additional Γ term in Eq. (17).

The result of the above calculations is the conductivity tensor $\overleftrightarrow{\sigma}$ of a single triangle. From that, one would like to obtain *macroscopic* quantities, such as the Hall resistivity ρ_{xy}. This is quite difficult. One often tries to do that by some averaging. Two different properties to be averaged over are the orientations and sizes of the triangles and then the (widely distributed) on-site energies. *A priori* at least two averaging procedures exist. One can average $\overleftrightarrow{\sigma}$, resulting in $\bar{\sigma}$, and then calculate $\rho_{xy} \equiv (\bar{\sigma}^{-1})_{xy}$. One can also average the resistivity tensor of a triangle $\overleftrightarrow{\sigma}^{-1}$ to obtain $\rho_{xy} \equiv \overline{(\sigma^{-1})}_{xy}$. As we will see, these two procedures lead to qualitatively different results. This fact, which has not been noticed before, demonstrates the trickiness of the averaging procedure.

We note that σ_{xy} includes a term independent of the magnetic field, which is of the same order of magnitude as σ_{xx}. Were we to average $\overleftrightarrow{\sigma}$ over directions *before* inverting it, then this term would have disappeared. However, if $\overleftrightarrow{\rho}$ is to be averaged, then this term remains, and leads to delicate cancellations in the denominator of $\overleftrightarrow{\rho}$. This, in turn, is the cause of the two different temperature dependences of the Hall resistivity.

We first average the conductivity tensor over directions and then invert it. In that case,

$$\rho^{(1)}_{xy} = \frac{2S_z\Gamma\sin\phi}{(R^x_{12})^4 G^2_{12}}\,, \qquad (24)$$

where we have assumed for simplicity that G_{12} is the largest conductance. To obtain the temperature dependence we consider the situation in which the magnetic field-free hopping conduction takes place along the bond 12 and site 3 supplies the interference path. Thus we imagine ϵ_1 and ϵ_2 to be below and above the Fermi level, but close to it, while ϵ_3 is away from the Fermi energy. Then [cf. Eqs. (10) and (12)]

$$\rho^{(1)}_{xy} \sim \frac{\Gamma}{G^2_{12}} \sim \exp[-\beta(\epsilon_3 - 2\epsilon_2 + \epsilon_1)], \qquad (25)$$

and is sensitive to the averaging procedure over the on-site energy distribution. One may imagine that the energy ϵ_2 is mostly in between the energies ϵ_1 and ϵ_3, in which case the log average of $\rho_{xy}^{(1)}$ will lead to a constant value for the Hall resistivity at very low temperatures—i.e., a "Hall-insulating behavior."

We next consider the transverse resistivity obtained by inverting the full conductivity tensor and then averaging over directions to restore rotational invariance. The result is

$$\rho_{xy}^{(2)} = \frac{1}{2S_z} \frac{\Gamma \sin\phi}{G_{12}G_{23} + G_{23}G_{31} + G_{31}G_{12}}. \quad (26)$$

Because of the cancellations occurring when $\overleftrightarrow{\sigma}$ of Eqs. (16) and (17) is inverted, the denominator here includes $G_{12}G_{23}$, etc., but not the (larger) term G_{12}^2, as in (24). Consequently, $\rho_{xy}^{(2)}$ increases exponentially as the temperature tends to zero, independent of the specific configuration of the single-particle energies. This is because of the factors $n_i^0(1 - n_i^0)$ in Eq. (12). Consider, for example, the energy configuration specified above. In that case the leading term in Γ is of order $\exp[-\beta(\epsilon_3 - \epsilon_1)]$ while $G_{12}G_{23}$ ($G_{12} \sim \exp[-\beta(\epsilon_2 - \epsilon_1)]$, $G_{23} \sim \exp[-\beta\epsilon_3]$) dominates the denominator in Eq. (26), leading to $\rho_{xy}^{(2)} \sim \exp[\beta\epsilon_2]$.

Both $\rho_{xy}^{(1)}$ and $\rho_{xy}^{(2)}$ are *independent* of the strength of the coupling to the phonons. This is in analogy with the "classical" (Boltzmann equation) result for the Hall coefficient, which does not contain the mean free path.

The physically correct way of averaging may depend on whether the experiment is a dc one or an ac one. In the ac case the current contacts are irrelevant, the current is inside the sample, and the macroscopic current density is obtained by summing the contributions from all triangles within a unit volume. This is equivalent to the averaging *of the conductivity tensor* of a single triangle.

In the dc case, the current is flowing from one current contact to the other through a percolation chain of bonds. Thus, the direction of the current in each bond is defined. To find the elementary Hall voltage created at this bond we can use the resistivity tensor of a single triangle. The total Hall voltage is obtained in this case by summing over the bonds along the percolation chain. This is equivalent to the averaging of the resistivity tensor of a single triangle. Thus, the dc Hall resistivity should be calculated by averaging over ρ. This is in accordance with the discussion given in Ref. [9]. We do not have a definitive idea as to the crossover frequency between the ac and dc regimes. One might *speculate* that it is the inverse of the time it takes the electron to traverse the sample from one current contact to the other. However, a different length scale may be involved in this.

To summarize, two independent derivations of $\overleftrightarrow{\sigma}$ at zero frequency but finite temperatures, were given for the Holstein model. Here the Hall conductivity too has a finite dc value when real, phonon-mediated, transitions are allowed. Surprisingly enough, we find that the answer depends on which transport quantity is averaged over directions. It was shown that the ensemble averaging needed to get the macroscopic Hall resistivity is subtle, and the result depends on whether $\overleftrightarrow{\sigma}$ is averaged before or after having been inverted. The former procedure leads to a possible "Hall insulating" behavior. The latter leads to a ρ_{xy}, which grows exponentially when the temperature is lowered. Using the percolating path picture (see, e.g., Ref. [7]), we argued that the latter is the proper averaging procedure for the dc limit. Finally, we would like to remark that we have not specifically considered here the deep quantum, large field, limit [10], which we hope to address in future work.

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