Hall resistance in the hopping regime, a “Hall Insulator”?

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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 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 \( \omega^2 \). Since to leading order in \( \omega \) the longitudinal conductivity \( \sigma_{xx} \sim i\omega\varepsilon_0 \), where \( \varepsilon_0 \) is the dielectric constant, this yielded that the Hall resistivity \( \rho_{xy} \sim \sigma_{xy}/(\sigma_{xx}^2 + \sigma_{yy}^2) \) approaches a constant in the small-frequency, zero-temperature limit.

Obviously one would like to examine the Hall resistance at finite temperatures. Here we address this question for strongly localized electrons. We consider the problem using the Holstein model [4] for the Hall effect in a system with localized states. The conductivity tensor for a triangle of three sites is obtained first. To get the macroscopic Hall resistivity one may attempt to apply two averaging procedures. We find that the result strongly depends on whether the conductivity tensor is averaged before inverting it or vice versa. In the first case a Hall insulator behavior is possible, while in the second the macroscopic Hall resistivity increases with decreasing temperature. The latter is similar to the finding of Friedman and Pollak [5]. The two behaviors are argued to be related with “ac” and “dc” conditions.

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 bar above the quantity indicates ensemble averaging). However, before averaging, \( \sigma_{xy} \) includes a field-independent term. We show that in the strongly localized regime this term is comparable in magnitude to \( \sigma_{xx} \) and \( \sigma_{yy} \). 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

\[
\frac{dn_i}{dt} = \sum_j (P_{ji} - P_{ij}),
\]

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 \( \ell \), \( i \to \ell \to j \). 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 [4] for the electron-phonon interaction, in which the ion displacements are coupled to the local (site) density of the electrons, and denote by \( \epsilon_i \) 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 \(|J_{ij}| \ll |\epsilon_{ij}|, \epsilon_{ij} = \epsilon_i - \epsilon_j \). One finds

\[
P_{12} = P_{12}^{\text{dir}} + P_{12}^{\text{indir}}.
\]
Here $P_{12}^{\text{dir}}$ arises from the direct amplitude alone and is independent of the magnetic field,

$$P_{12}^{\text{dir}} = n_1 (1 - n_2) \left| J_{12} \right|^2 \int_{-\infty}^{\infty} dt \, e^{i \xi_{12} t} \, e^{2(g(t)-g(0))},$$

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

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

$$P_{12}^{\text{indir}} = n_1 (1 - n_2) \left| J_{12} J_{23} J_{31} \right| 2 \Im \, e^{i \phi_{132}} \int_0^\infty dt_2 \int_0^\infty dt_1 \, e^{g(t_1) + g(t_2) + g(t_1 + t_2) - 3g(0)} \left[ (1 - n_3) e^{i \xi_{12} t_1 + i \xi_{13} t_2} - n_3 e^{i \xi_{12} t_1 + i \xi_{32} t_2} \right],$$

and contains the occupation of site 3, $n_3$, and the magnetic phase, $\phi_{132}$,

$$\phi_{132} = \frac{e}{c} \vec{H} \cdot \vec{S}, \quad \vec{S} = (\vec{R}_1 \times \vec{R}_3 + \vec{R}_3 \times \vec{R}_2 + \vec{R}_2 \times \vec{R}_1)/2.$$  

(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 which 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 $\omega$. In the presence of an external 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 $\mu_i$

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

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

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

(7)

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

$$e^2 \frac{dn_1}{dt} = e(P_{21} - P_{12} + P_{31} - P_{13}) = G_{12} \zeta_{12} + G_{31} \zeta_{13} + \sin \phi \Gamma \zeta_{23},$$

(8)

with analogous expressions for the other two sites, where $\phi$ is the magnetic flux enclosed in the triangle. These equations determine the electrochemical potential differences, $\zeta_{ij}$. However, in practice one does not have to solve for the $\zeta_{ij}$’s since Eq. (8) gives the current (9) (see Eq. (13) below.) In (9), $G_{ij}$ is the conductance of the bond $ij$, arising from the direct rate,

$$G_{ij} = G_{ji} =$$

$$e^2 \beta n_{ij}^0 (1 - n_{ij}^0) \left| J_{ij} \right|^2 \int_{-\infty}^{\infty} dt \, e^{i \xi_{ij} t} \, e^{2(g(t)-g(0))}.$$  

(9)

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

$$G_{ij} = e^2 \beta \left| J_{ij} \right|^2 \frac{v^2 N(|\varepsilon_{ij}|)}{|\varepsilon_{ij}|^2} e^{-2(|\varepsilon_i| + |\varepsilon_j| + |\varepsilon_i - \varepsilon_j|)},$$

(10)
where $N$ is the phonon density of states. (Energies are measured from the Fermi energy.) The interference process leads to the phase-dependent term, with

$$\Gamma = e^2 \beta n_i^0 (1 - n_i^0) | J_{12} J_{23} J_{31} | \int_{-\infty}^{\infty} dt_1 \int_{-\infty}^{\infty} dt_2 \ e^{g(t_1) + g(t_2) + g(t_1 + t_2) - 3g(0)} e^{i \epsilon_{12} t_1} \left( (1 - n_i^0) e^{i \epsilon_{13} t_2} + n_i^0 e^{i \epsilon_{32} t_2} \right).$$

(11)

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

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

(12)

This expresses the fact that the indirect rate involves two scattering events by the phonons. The temperature dependence of $\Gamma$, 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

$$\vec{J} = e \sum_i \frac{d n_i}{d t} \vec{R}_i.$$  

(13)

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

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

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

(14)

the current density becomes

$$\vec{J} = \partial \vec{E}_{\text{eff}}$$

$$\vec{E}_{\text{eff}} = \vec{R}_{12} \vec{R}_{12} G_{12} + \vec{R}_{23} \vec{R}_{23} G_{23} + \vec{R}_{31} \vec{R}_{31} G_{31} + \Gamma \sin \phi (\vec{R}_{23} \vec{R}_1 + \vec{R}_{31} \vec{R}_2 + \vec{R}_{12} \vec{R}_3).$$

(15)

A remarkable observation is that the part of $\vec{J}$ which is proportional to the magnetic field is perpendicular to the effective electric field. In deriving the expression for $\vec{E}_{\text{eff}}$ it was assumed that the triangle lies in the $x - y$ plane, perpendicular to $\hat{z}$. Note that $\vec{E}_{\text{eff}}$, 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

$$\sigma_{xx} = (R_{12}^x)^2 G_{12} + (R_{23}^x)^2 G_{23} + (R_{31}^x)^2 G_{31},$$

(16)

($\sigma_{yy}$ is given upon replacing $R_{ij}^x$ by $R_{ij}^y$, and

$$\sigma_{yx} = R_{12}^x R_{23}^y G_{12} + R_{23}^x R_{31}^y G_{23} + R_{31}^x R_{12}^y G_{31}$$

$$\pm 2 S_z \Gamma \sin \phi.$$  

(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, $\vec{E}_{\text{eff}}$:

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

(18)
where η is a positive infinitesimal, |m> and |n> are eigenstates and f_n, f_m their populations. We generalize the derivation of (18) to include electron-phonon coupling and consider the ω > 0, ω → 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>: electron in site 1 with a phonon bath in equilibrium and |2, q >: electron in site 2, with the same minus one phonon in state q, where ω_q ≡ ϵ_{21}. Thus, the approximate eigenstates are:

|m> = |1> + \sum q J_{12}ν_q \epsilon_{12} + ω_q |2, q >,

|n> = |2, q > + \frac{J_{21}ν_q^*}{ε_{21} - ω_q} |1 >, (19)

where ν_q = ν_q \sqrt{N_q}/ω_q, and

<n|m> ≡ \frac{J_{12}|ν_q|^2 R_{12}^x}{ω}.

(20)

Using Eq. (18), this produces the first term in (14):

\begin{align*}
\frac{1}{Vol}(R_{12}^x)^2 e^2 βJ^2_{12} \sum \frac{|ν_q|^2}{ω_q^2} N_q \delta(ω - ϵ_{21} - ω_q) = \\
4\pi βe^2 J^2_{12} ω^2 |N_q N(ϵ_{21})| |ω_q - ϵ_{21}|.
\end{align*}

(22)

Using the expression similar to (24) for the matrix elements of y, yields the first term in (17). The (2,3) and (1,3) terms in Eqs. (10) 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’> (i.e. |3> > minus the phonons q and q’) in two ways:

(a) A straightforward correction \sum q’ |J_{13}ν_q ν_q’/(ε_{13} + ω_q + ω_q’)|3, q, q’>, 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 ω_q + ω_q’ = ϵ_{31} and by the nonresonant contribution given by q’ = q” such that ω_q” = ϵ_{32}.

(b) The Holstein contribution, the mixing of |2, q > via the intermediate state |3, q, q’>: \sum q’ |J_{13}J_{32}ν_q | ν_q’ |2 /((ε_{13} + ω_q + ω_q’)(ε_{12} + ω_q)) | 2, q >. Here, as found by Holstein, the resonant contribution with ϵ_{31} = ω_q + ω_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

\begin{align*}
(n|x|m) ≡ \frac{J_{12}ν_q}{ω} R_{12}^x - 2iπδ(ε_{31} - ω_q - ω_q’)
\end{align*}

\begin{align*}
J_{13}J_{32}ν_q | ν_q’ |2 \left[ R_{3} - \frac{R_{1}^2 + R_{2}^2}{2} \right].
\end{align*}

(23)

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

The result of the above calculations is the conductivity tensor \sigma of a single triangle. We would like to have macroscopic quantities like the Hall resistivity \rho_{xy}. For that purpose one might average \sigma over the orientations and sizes of the triangles and also over the on-site energies. A priori two averaging procedures exist. One can average \sigma, resulting in \bar{σ}, and then calculate \rho_{xy} ≡ (σ^{-1})_{xy}. One can also average the resistivity tensor of a triangle \tilde{σ}^{-1} resulting in \overline{σ}^{-1} and then calculate \rho_{xy} ≡ (\overline{σ}^{-1})_{xy}. As we will see these two procedures lead to qualitatively different results.

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 \sigma over directions before inverting it, then this term would have disappeared. However, if \tilde{σ} is to be averaged, then this term remains, and leads to delicate cancellations in the denominator of \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,

\begin{align*}
ρ_{xy}^{(1)} = \frac{2S_z Γ \sin φ}{(R_{12}^z)^2 G_{12}^2},
\end{align*}

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 $\epsilon_1$ and $\epsilon_2$ to be below and above the Fermi level, but close to it, while $\epsilon_3$ is away from the Fermi energy. Then [cf. Eqs. (10) and (13)]

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

and is sensitive to the averaging procedure over the on-site energy distribution. One may imagine that the energy $\epsilon_2$ is mostly in-between the energies $\epsilon_1$ and $\epsilon_3$, in which case the log-average of $\rho^{(1)}_{xy}$ 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

$$\rho^{(2)}_{xy} = \frac{1}{2S_2 G_{12}G_{23} + G_{23}G_{31} + G_{31}G_{12}} \frac{\Gamma \sin \phi}{\beta \epsilon_2}$$

Due to the cancellations occurring when $\tilde{\sigma}$ of Eqs. (10) and (17) is inverted, the denominator here includes $G_{12}G_{23}$, etc., but not the (larger) term $G^2_{12}$, as in (24). Consequently, $\rho^{(2)}_{xy}$ increases exponentially as the temperature tends to zero, independent of the specific configuration of the single-particle energies. This is because of the factors $n^0_i(1 - n^0_i)$ in Eq. (12). Consider for example the energy configuration specified above. In that case the leading term in $\Gamma$ 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 $\rho^{(2)}_{xy} \sim \exp[\beta \epsilon_2]$.

Both $\rho^{(1)}_{xy}$ and $\rho^{(2)}_{xy}$ 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. We consider a measurement to be an ac one if the frequency is such that the electron cannot traverse the sample from one current contact to the other during one field period. In this 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 summation is equivalent to the averaging of the conductivity tensor of a single triangle.

We define a measurement as a dc one when the electron traverses the sample during a time short compared to the period of the field. The current is therefore flowing from one current contact to the other through a percolation chain of bonds. This means that the direction of the current in each bond is defined. To find the elementary Hall voltage created at this bond we have to invert the conductivity tensor of the triangle attached to this bond and express this elementary Hall voltage in terms of 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 summation is equivalent to the averaging of the resistivity tensor of a single triangle.

To summarize: Two independent derivations of $\tilde{\sigma}$ 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 before or after having been inverted. The former procedure leads to a possible “Hall insulating” behavior. The latter leads to a $\rho_{xy}$ which grows exponentially when the temperature is lowered. Using the percolating path picture of [7], one might speculate that the latter is the proper averaging procedure for the dc limit.

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