Hall effect in the perovskite manganites

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We compute the zero temperature phase diagram and the Hall response of the doped perovskite manganites within the model of double exchange and Jahn-Teller coupling, employing the $d \rightarrow \infty$ approximation proposed by Millis et al.. We find that in this scenario a "hole-like" $R_H$ for the hole doped manganites, as observed at low temperature by Mati et al., can be obtained only when lattice distortions persist in the metallic state as $T \rightarrow 0$. The calculated temperature dependence of $R_H$ seems to be consistent with the measured "normal" Hall coefficient in thin films.

I. INTRODUCTION

With the current interest in "colossal magnetoresistance" the ferromagnetic manganese oxides have been intensely studied over the last few years. These compounds, of the form $La_{1-x}(Sr,Ca)_xMnO_3$, are antiferromagnetic insulators in the undoped phase ($x=0$), and have a ferromagnetic ground state beyond a critical doping. The ferromagnetism is conventionally understood in terms of the double exchange model. The transition from the ferromagnetic to the paramagnetic phase is accompanied by a simultaneous metal-insulator 'transition'. The transport is typically activated in the paramagnetic phase.

The large increase in resistivity near $T_c$ and the rather low transition temperature prompted the suggestion that, in addition to the double exchange mechanism, carrier localisation due to electron-phonon coupling effects could be relevant in these materials. Detailed calculations incorporating both double exchange and the Jahn-Teller coupling have been successful in reproducing most of the qualitative features of the data. So, although there are possibly complications due to electron correlation effects as well as disorder due to cation substitution, the basic interplay between charge, spin and lattice degrees of freedom in the manganites seems to be adequately described by the double exchange/Jahn-Teller scenario.

Experimentally there is overwhelming evidence for dynamical lattice distortions in these compounds, including isotope effects on $T_c$ and direct measurement of the Debye-Waller factor. There is also a systematic and large change in lattice constant with temperature. The activated transport in the paramagnetic phase has been interpreted in detail in terms of small polaron hopping. While it is obvious and acknowledged that the electron phonon coupling is central to understanding the "high temperature", $T \gtrsim T_c$, physics, its effect on the low temperature metallic phase is much less studied. This spin polarised phase is believed to admit an almost "band theoretic" description, with electron-magnon interactions describing the transport. The measurement of the Hall effect in $La_{0.67}Ca_{0.33}MnO_3$ at low temperature, however, suggests that the electron-phonon coupling may be crucial in understanding this phase too, as we discuss next.

II. HALL MEASUREMENTS ON THE MANGANITES.

The Hall response of the manganites has signatures of various interaction effects. There is strong field dependence to the Hall resistivity, as is usual in ferromagnets, and the data can be parametrised as $\rho_{xy} = R_H(T)B + R_S(T)M$, where $R_H$ and $R_S$ are respectively the "normal" and "anomalous" Hall coefficients, $B$ the induction and $M$ the spontaneous or induced magnetisation. Measurements indicate that $R_H > 0$, while the anomalous term $R_S < 0$ (in the Ca doped system) and $|R_S| \gg R_H$ for $T \sim T_c$. In fact, at all but the lowest temperatures, $\rho_{xy}$ is dominated by the anomalous term. The sign and temperature dependence of $R_S$ is intriguing and it has been suggested that its origin may lie in a combination of spin-orbit coupling effects and strong electronic correlations. Here we concentrate on the normal Hall coefficient which, although much smaller than $R_S$, also has several mysterious features, not necessarily linked to the ferromagnetism; (a), $R_H$ is "hole-like" although a simple electron with degenerate Mn orbitals would predict an "electron-like" Hall coefficient. (b). The carrier density, inferred from $R_H^{-1}$, at $x=0.33$ seems to be five times the nominal doping. So, either the Drude form $R_H^{-1} \sim x$, naively expected for the doped insulator, is inapplicable, or there is a serious discrepancy between the nominal and actual carrier density. Therefore, beyond the anomalies attributable to spin orbit coupling effects, the doping and temperature dependence of the normal Hall coefficient itself in the manganites is not understood. We have calculated $R_H$ within the model of double-exchange and Jahn-Teller coupling introduced by Millis et al. as a function of electron-phonon coupling, $g$, electron density, $n$, and temperature, $T$. 


III. MODEL

We use the standard Hamiltonian,

$$\hat{H} = \hat{H}_{el} + \hat{H}_{d-ex} + \hat{H}_{ph} + \hat{H}_{el-ph}$$

The electronic band is specified by \(\hat{H}_{el} = \sum_{ij} \epsilon_{ij}^{\alpha\beta} c_i^{\dagger} c_j^{\beta} - \sum_i \mu n_i\), where \(\alpha\) and \(\beta\) refer to the two \(e_g\) orbitals. The double-exchange term \(\hat{H}_{d-ex} = -J_H \sum_{i} \delta_i \sigma_i\) couples the conduction electrons to the core ‘spin’ \(3/2\), which we treat classically, assuming \(J_H S_{c}/t \rightarrow \infty\). The classical phonons are described by a harmonic distortion term \(\hat{H}_{ph} = \sum_i k_i^2 / 2\), and the Jahn-Teller coupling \(\hat{H}_{el-ph} = \theta \sum_i \hat{r}_i \hat{c}_i^{\dagger \sigma} \hat{c}_i^{\sigma} \hat{c}_i^{\dagger} \hat{c}_i^{\sigma}\) where \(\theta\) is the lattice distortion beyond which the system, with electron density \(n\), becomes a polaronic insulator, and \(\langle r \rangle\). \(\langle r \rangle \geq r_c(n)\) corresponding to the insulating state. Obviously, since phononic effects completely disappear in the first case, the Hall effect would simply correspond to that of a spin polarised band, i.e. the band theoretic result. In the third case, since the conductivity vanishes as \(T \rightarrow 0\), the Hall coefficient is ill-defined. The most interesting regime corresponds to \(b\), where lattice distortions persist in the \(T \rightarrow 0\) metallic state and \(R_H\) can significantly differ, in sign and magnitude, from the band-theoretic value. The metallic or insulating character of the ground state is determined from the density of states, \(N(\omega)\), at \(\omega = 0\). For the Fermi liquid this corresponds to the band DOS, for the ‘rigid band’ phase it is finite but suppressed from the band value, and for the polaronic insulator \(N(0) = 0\).

Our result for the “phase boundaries” separating the Fermi liquid, the distorted metallic phase and the polaronic insulator at \(T = 0\) are shown in Fig.1. Before discussing the behaviour of \(R_H\) in the various phases let us establish the band theoretic result. Consider half-filling first, ignoring the antiferromagnetism for the moment. If the two \(e_g\) orbitals were equally occupied, and the spin states were degenerate, then the “half-filled” state would correspond to quarter filling of spin degenerate bands, and the Hall effect would be “electron-like”. However, if the \(T \rightarrow 0\) state were spin polarised the majority carriers would inhabit two orbitally degenerate half-filled bands. This state, for a particle-hole symmetric bandstructure would correspond to \(R_H = 0\). Hole doping on this state, i.e a reduction of the electron count, would lead to an electron-like Hall effect, not hole-like as has been measured.

The numerically obtained \(T \rightarrow 0\) result for \(R_H\) confirms that for \(g < g_c(n = 1) \sim 1.14\), where the half-filled state becomes insulating, the Hall coefficient, at all doping, correspond to the band prediction. These states all have vanishing distortion as \(T \rightarrow 0\). The results on \(R_H\), for several interaction strengths, \(g = 0.9, 1.0, 1.3\), are superposed in Fig.2. Between \(g_c(1)\) and \(g'_c(1)\) the half-filled state remains insulating while states away from half-filling are metallic, but with finite \(\langle r \rangle\). However, for a given \(g\), with \(g_c(1) < g < g'_c(1)\), beyond a critical deviation away from half-filling the distortion seems to disappear. In this region, bounded by \(g_c(n)\) separating the Fermi liquid and distorted phase, \(R_H\) exhibits interesting deviation from the band value. Notice that while \(R_H = 0\) would occur only at the particle-hole symmetric point, \(n = 1\), in the Fermi liquid phase, it occurs away from half-filling in the distorted phase. This is the key to the anomalous behaviour in the doped insulator.

We have shown a typical plot, for \(g = 1.3\), in Fig.2.

This permits a simple classification of the various regimes as \(T \rightarrow 0\). Following [12] these are \(a\), Fermi liquid; \(\langle r \rangle \rightarrow 0\) as \(T \rightarrow 0\), electron-phonon interaction effects vanish and there are no “quenched” distortions. \(b\), “Rigid band” phase, \(0 < \langle r \rangle < r_c(n)\), where \(r_c(n)\) denotes the critical distortion beyond which the system, with electron density \(n\), becomes a polaronic insulator, and \(c\). \(\langle r \rangle \geq r_c(n)\) corresponding to the insulating state. Obviously, since phononic effects completely disappear in the first case, the Hall effect would simply correspond to that of a spin polarised band, i.e. the band theoretic result. In the third case, since the conductivity vanishes as \(T \rightarrow 0\), the Hall coefficient is ill-defined. The most interesting regime corresponds to \(b\), where lattice distortions persist in the \(T \rightarrow 0\) metallic state and \(R_H\) can significantly differ, in sign and magnitude, from the band-theoretic value. The metallic or insulating character of the ground state is determined from the density of states, \(N(\omega)\), at \(\omega = 0\). For the Fermi liquid this corresponds to the band DOS, for the ‘rigid band’ phase it is finite but suppressed from the band value, and for the polaronic insulator \(N(0) = 0\).

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IV. RESULTS

A. \(T = 0\), phase diagram, and doping dependence of \(R_H\)

At low temperature, \(T \ll T_c\), the spin background is almost completely polarised. In the absence of any orbital ordering, the phonon distribution depends only on the scalar distortion, \(r = |r|\), and, for \(T \rightarrow 0\), the probability distribution, \(P(r)\), for the (classical) phonons is sharply peaked, since the variance \(\langle \Delta r^2 \rangle \sim T\). Therefore, in this limit, the electronic spectrum is completely determined by the average onsite distortion \(\langle r \rangle = r_0\).
to illustrate this behaviour. Notice that with approach to half-filling, where the interaction effects are strongest due to the lowest kinetic energy, $R_H$ moves off the band result, changes sign somewhere, and tends to diverge as \( n \to 1 \). It is numerically difficult to push the imaginary frequency calculation below $T \sim 0.01$, but an asymptotic analytic solution (see Appendix) at $T = 0$ indicates that for $x \to 0$, $R_H \sim x^{-1/3}$. Increasing $g$ beyond $g_c'(1)$ states away from half-filling rapidly turn insulating. The boundary $g_c(n)$ separating the polaronic insulator from the Fermi liquid and distorted metallic phases is shown in Fig.1.

**B. Temperature dependence of $R_H$**

Electron-phonon coupling leads to a suppression of electron kinetic energy, as does magnetic disorder within the double exchange model. So, qualitatively, the effect of increasing temperature, reducing magnetisation, and suppressing electron motion would look like increasing $g$ at $T = 0$. The finite $T$ results bear this out, as shown in Fig. 3. For the chosen coupling constant, $g = 1.5$, for $x = 0.2$ the hole-like $R_H$ at $T = 0$ becomes larger as $T \to T_c$, for $x = 0.3$ the low temperature negative Hall coefficient changes sign to hole-like at some intermediate temperature, while for $x = 0.4$ it becomes less negative with increasing $T$ but never changes sign. A complementary situation would hold for ‘electron-doped’ systems, if they could be experimentally realised. Overall, $R_H$ deviates even more from its band theoretic value with increasing temperature ($T \leq T_c$).

Within our approximation the magnetic disorder is maximal at $T = T_c$ (the spin distribution becomes isotropic) and there is no further suppression of kinetic energy for $T > T_c$. In this regime the behaviour of $R_H$ is probably determined by the activation effects in $\sigma_{xx}$.

**V. DISCUSSION**

A ‘hole-doped’ insulator is intuitively expected to have a hole-like $R_H$, so our approach might seem to be an elaborate way of arriving at an obvious conclusion. The state we describe is, however, rather non-trivial, and below we contrast it both to (a) the doping of a simple band insulator and (b) the mean field picture of the “distorted” metallic phase (where every site has the same distortion $\vec{r}$). Next, (c) we contrast the properties of this mean-field state to that with randomly oriented distortions and, finally, (d), comment on quantum effects which we have neglected in our calculation.

(a) Usually “electron” or “hole” doping refers to the doped carriers occupying a specific part of the Brillouin zone. For a band insulator, the doped holes, by definition, occupy the hole like parts of the zone, and lead to a positive $R_H$. When the insulating state arises out of interaction effects, at an electron density where band theory would have predicted a metal, it is not clear at all that a reduction in electron density is equivalent to “hole doping” in the earlier sense. Also, if $R_H$ as $T \to 0$, away from the insulating state, is required to deviate from the simple band value, some signature of interaction effects must persist in the metallic ground state. We have stated this earlier in the context of our results, here we would like to emphasize that, quite independent of specific models, a change in the sign of the $R_H$ at $T = 0$ requires “quenched” distortions of some sort. Proximity to an insulator is necessary, but not sufficient, to obtain the behaviour observed.

(b). Since the Jahn-Teller coupling can lead to a lattice distortion and splitting of the electronic band, naive band theory predictions based on degenerate $\epsilon_g$ levels are incorrect, as we have argued before. One could, however, construct a mean-field picture incorporating Jahn-Teller distortions, which might well provide an adequate description of the half-filled insulating state. By continuity one might argue that a similar mean-field picture for Jahn-Teller distortions away from half-filling, with a self-consistently computed $\vec{r}$ would provide a reasonable description of the metallic phase. In that case one would indeed dope into the ‘lower band’, and the quasiparticles, in this orbitally ordered phase, will show a hole-like $R_H$. That situation, though conceptually appealing, does not seem to be experimentally relevant. There is no signature for orbital long range order in the metallic state of the manganites and one is required to deal with spatially random distortions, as we have done.

(c). The phase we consider has some distinct, non mean-field like, properties. Within the scheme we have adopted for the transport calculation, the single particle spectral function completely determines the transport coefficients. Since the spectral function $A(k, \omega) = -(1/\pi)\text{Im}(\omega + \mu - \epsilon_k - \Sigma(\omega))^{-1}$, the low frequency behaviour $\lim_{\omega \to 0}(\mu - \Sigma(\omega)) \sim \mu - i\pi \Gamma$, determines the low temperature transport. Here $\mu = \mu - \Sigma_R(0)$ and $\Gamma = -(1/\pi)\Sigma_i(0)$ These parameters, $\mu$ and $\Gamma$ have a physical interpretation. $\Gamma$ is the single particle scattering rate, and determines the resistivity within this model, while $\epsilon_k = \mu$ determines the location of the Fermi surface and the Hall coefficient. This distinction between the intuitive mean-field picture and the orbitally disordered phase shows up in the behaviour of these parameters, and have direct physical consequences. The implications are (i) When the electrons propagate in a background of random distortions, they will experience a large scattering rate, in contrast to the orbitally ordered phase where $\Gamma$ and $\rho(T) \to 0$ as $T \to 0$. (ii) The orbitally ordered metallic phase will show a Fermi surface in photoemission, while the state computed by us does not have any discontinuity in its momentum distribution function since, near half-filling, since one finds $\mu > 2D$ the bandwidth (see Appendix). It is somewhat difficult to cleanly verify these effects in the electronic spectrum because
there is already large substitutional disorder, leading to a finite $\Gamma$ as $T \to 0$, and a smearing of the Fermi surface.

$(d)$ We have made a classical approximation for the phonons and, even in the absence of orbital ordering, below some coherence scale the quantum effects could become important and anneal out the random lattice distortions. In this regard we can gain some insight from what is known about doped Mott insulators. In the Hubbard model at large $U$, over an intermediate temperature range, one can view the system as electrons interacting with static random local moments. This doped Mott insulator has a doping dependence of $R_H$ similar to what we have found here. However, below $T \sim xD$, where $x$ is the doping, a Fermi liquid ground state is recovered and $R_H$ becomes band like again. A similar situation could arise in this model also, as we outline below. Suppose the relevant optical phonon in our case has a frequency $\omega_0 = \sqrt{k/m}$. For $T \ll \hbar\omega_0/R_H$ we could integrate out the phonons and generate a term in the Hamiltonian of the form $\hbar^2 \sum_\alpha \langle \dot{c}_\alpha^+ \dot{c}_\alpha \rangle / 2$. At $T = 0$ the model reduces to a “Hubbard” model involving two orbital species $c_{\uparrow\alpha}$ with an effective onsite repulsion $U_{eff} = U_{coupl} + \hbar^2 / \omega_0$ (at finite $T$ we need to consider the other spin species too since the ferromagnet is not fully polarised). This suggests that there could be a temperature scale below which our results, involving static distortions, would not hold. Apart from the presence of substitutional disorder, it is not clear why such a quantum scale is not visible in the manganites.

We can now discuss how the measured $R_H$ fits within this framework. Since experimentally the half-filled state is insulating the relevant regime for us would be $g > g_0(1)$. Let us choose $g = 1.3$, as shown in Fig.2, for illustration. There $R_H$ is indeed large and positive near half-filling. However, the behaviour changes for $x \sim 0.3$ where $R_H \sim 0$ and changes sign at larger doping. This is effectively the interpolation between a doped insulator (strong coupling, low kinetic energy) and ‘band metal’ (weak coupling, large kinetic energy). So the enhancement seen in $R_H^0$ at $x = 0.33$ could be simply due to the proximity of $R_H$ to a zero crossing. For our choice of $g, x = 0.33$ itself is electron like, but that depends on the exact value of the coupling, which is anyway unknown. The two predictions one can make within this scheme are: (a) With increasing doping, within a given chemical family, $La_{1-x}Ca_xMnO_3$ say, $R_H$ should change sign from hole-like to electron like, unless some other ordering phenomena intervenes (b). With increasing coupling, $Pb$ substitution on $Cu$, keeping the doping level fixed, $R_H$ should deviate further from its band-theoretic value (should become larger if it is already hole-like, etc).

On the $T$ dependence of $R_H$ itself, there does not seem to be any published data. Unpublished results from Bryan Lin et al. [13] for $La_{0.67}Ca_{0.33}MnO_3$ indicates that the hole-like $R_H$ at $T = 0$ increases with $T$ and has a peak near $T_c$, beyond which it seems to fall off. This seems to be consistent with our calculated $T$ dependence ($x = 0.2$, Fig.3). However, $\rho_H$ measurements on the manganite are rather difficult, due to the large magnetoresistance, and it would be useful to have independent confirmation of this trend.

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VI. APPENDIX; THE $T \to 0$ EQUATIONS

As we have seen, much of the physics of $R_H(x, T)$ is controlled by the coupling constant and doping dependence at $T = 0$, so it may be helpful to consider the $T = 0$ equations in some detail. We essentially follow the derivation in [12]. Since the spins are completely polarised, it is sufficient to consider only the Greens function for electron spin parallel to the magnetisation, $G_{\uparrow\uparrow}$.

Within the $d \to \infty$ framework one solves a local problem, of a site hybridising with a self-consistently computed bath, with the local Green’s function $G_{\text{loc}}(\omega)$ being the local projection, $G_{\text{loc}}(\omega)$, of the lattice Green’s function. Broadly, for our electron-phonon problem, there are three steps: (a) Compute the local Green’s function, $G_{\uparrow\uparrow}^{\text{loc}}(r, \omega)$ for a given value of local distortion, $r$. This is elementary since the local fermion problem is quadratic. (b) Compute the full local Greens function by averaging over coordinates, with the appropriate distribution function.

$$G_{\uparrow\uparrow}^{\text{loc}}(\omega) = \int d\bar{r} P(r) G_{\uparrow\uparrow}^{\text{loc}}(r, \omega)$$

This is possible since the phonons are classical. (c) Calculate the distribution function, $P(r)$, as the electronic free energy for distortion $r$ as $P \sim e^{-T\Gamma lnG(r)}$. This closes the self consistent loop.

More specifically, for the semicircular band DOS that we have assumed, the following equations need to be solved for $T \to 0$.

$$a(i\omega_n) = i\omega_n + \mu - \frac{1}{2} \int d\bar{r} P(r) \left\{ \frac{1}{a - gr} + \frac{1}{a + gr} \right\}$$

which combines steps (a) and (b) above. $a(\omega)$ is related to $G_{\uparrow\uparrow}^{\text{loc}}(\omega)$ by

$$G_{\uparrow\uparrow}^{\text{loc}}(\omega) = \frac{1}{2} \int d\bar{r} P(r) \left\{ \frac{1}{a - gr} + \frac{1}{a + gr} \right\}$$

The phonon distribution

$$P(r) = \frac{1}{Z_{\text{loc}}} e^{\exp\left[-\frac{r^2}{2T} + \sum_n \ln\left(\frac{q_n^2 - q_{r}^2}{(i\omega_n)^2}\right)\right]}$$

and $Z_{\text{loc}} = \int d\bar{r} P(r)$. A more complicated version of these equations hold at finite $T$, where the distribution of local spin orientations also has to be taken into account.

To understand the damping dependence of transport coefficients we need to solve the equations above in the case
where \( P(r) \), in the limit \( T \to 0 \), reduces to a delta function at one non zero value, i.e.,

\[
a(\omega_n) = i\omega_n + \mu - \frac{1}{2} \left( \frac{1}{a} - gr \right) + \frac{1}{a + gr} \tag{3}
\]

and \( r \) is determined by maximising \( P(r) \). We can view this as a cubic equation for \( a(\omega) \), for a specified \( gr \). It is possible to solve this equation near the band edge, parametrised in terms of the deviation of the chemical potential \( \mu \) from \( \mu^* \) (where one begins to dope into the lower band), and the DOS looks like,

\[
\sim \frac{1}{\pi \omega - (\mu^* - \mu)}
\]

From this the number of doped holes can be estimated to be \( x = 1 - n \sim (\mu^* - \mu)^{3/2} \).

To obtain \( \sigma_{xx} \) etc., we need to use the relation

\[
\mu - \Sigma(\omega) = G_{ret}(\omega) + G_{ret}^{-1}(\omega)
\]

which, for \( ReG(0) \neq 1 \), leads to \( Im\Sigma(0) \sim ImG(0) \sim (\mu^* - \mu)^{1/2} \). Combining this with the result for \( n \) before, we obtain \( \Gamma \sim x^{1/3} \). Also \( \mu - Re\Sigma(0) \sim ReG(0) + ReG^{-1}(0) \geq 2 \) since \( ImG(0) \to 0 \).

Since \( \Gamma \to 0 \) while \( \mu - Re\Sigma(0) - \epsilon_x \neq 0 \), the spectral functions, which enter the transport calculation can be roughly approximated as \( A(\tilde{k}, \omega) \sim \Gamma/(\epsilon_x - \mu)^2 \). From which, employing the formulae cited previously in the text, one obtains \( \sigma_{xx} \to \Gamma^2 \), i.e. \( \rho \sim x^{-2/3} \), and \( \sigma_{xy} \sim \Gamma^3 \).

This leads to \( R_H \sim \sigma_{xy}/\sigma_{xx}^2 \sim x^{-1/3} \).

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