Magnetotransport in manganites and the role of quantal phases I: Theory

Y. Lyanda-Geller, P. M. Goldbart, S. H. Chun and M. B. Salamon

Department of Physics and Materials Research Laboratory,
University of Illinois at Urbana-Champaign, Urbana, Illinois 61801

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A microscopic picture of charge transport in manganites is developed, with particular attention being paid to the neighborhood of the ferromagnet-to-paramagnet phase transition. The basic transport mechanism invoked is inelastically-assisted carrier hopping between states localized by magnetic disorder. In the context of the anomalous Hall effect, central roles are played by the Pancharatnam and spin-orbit quantal phases.

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Introduction: The double-exchange interaction (DEI) has long been understood to play a major role in the ferromagnet to paramagnet transition (FPT) in the manganite systems La$_{1-x}$A$_x$MnO$_3$ (where A stands for Ca, Sr or Pb), the transition being accompanied by a metal-insulator transition (MIT). In this DEI picture, proposed by Zener and elaborated by Anderson and Hasegawa [1], intra-atomic Hund’s Rule coupling leads to a modulation of the amplitude for the hopping of outer-shell carriers between neighboring Mn ions. It is now recognized, however, that the physics of the DEI is insufficient to fully explain the observed phenomenon of colossal magnetoresistance (CMR) (i.e., the strong magnetic-field induced suppression of the resistivity, and the shift to higher temperatures of the peak in its temperature-dependence [2]). Moreover, interest in CMR has led to a re-examination of the nature of the FPT and the MIT in manganites and related compounds. In these contexts, Millis, Shraiman and co-workers [3] have proposed that the DEI is accompanied by a large Jahn-Teller lattice distortion that would cause the polaronic collapse of any conduction band. Varma [4] and Sheng et al. [5] have argued, in contrast, that the MIT in manganites is an Anderson localization transition, resulting from magnetic and non-magnetic disorder.

The purpose of the present Letter is to address charge transport in manganites in the vicinity of the FPT and MIT from the vantage point afforded by the Hall effect. In a companion Letter [6], we present and analyze experimental data on the Hall effect and CMR in La$_{2/3}$(Ca,Pb)$_{1/3}$MnO$_3$. We shall argue that, near the FPT, owing to charge-carrier localization, transport is via hopping between localized states.

The central part of our analysis is the discussion of the microscopic mechanism of the Hall effect (HE) in manganites. In ferromagnetic metals HE’s include an ordinary Hall effect (an OHE, which arises from the Lorentz force acting on the current carriers), as well as an anomalous Hall effect (AHE), i.e., a Hall current proportional to the average magnetization and independent of the demagnetization effects. For metallic states, microscopic mechanisms yielding the AHE have been discussed, e.g., in Ref. [7], the essential ingredient being the spin-orbit interaction (SOI), which leads to an AH current in the presence of magnetization (of any origin) [8]. If charge transport near the FPT and MIT in manganites does indeed occur via hopping, then we are led to the general issue of the microscopic mechanism of the AHE in hopping conductors. This AHE cannot be captured by a picture based solely on the Anderson-Hasegawa analysis [1] of the DEI within a pair of Mn ions. Such a picture includes only the modulation of the magnitude of the hopping between the pair determined by the relative alignment of the core spins on the ions [via a factor cos($\theta/2$), where $\theta$ is the angle between (semiclassical) directions of the core spins]. This insufficiency of a pair-based picture is an analog of Holstein’s observation [9] that to capture the OHE in hopping conductors requires the analysis of at least triads of atoms, and of the attendant Aharonov-Bohm (AB) fluxes through the polygons whose vertices are the atomic sites. Therefore, we shall examine a mechanism for the AHE involving hopping within triads of sites, in which fundamental roles are played by two quantal phases: (i) the SOI phase, acquired by electrons propagating in the presence of SOI; and (ii) the (quantal) Pancharatnam phase (an electronic analog of the (optical) Pancharatnam phase accrued by classical light under a sequence of polarization changes [10,11]). In this electronic analog, outer-shell carriers, hopping from ion to ion, acquire a phase determined by the solid angle subtended by the spherical polygon whose vertices are the orientations of the core-spins of the ions visited.

Recently, Kim et al. [12] revisited the theory of the AHE, in the context of a model that includes DE, SOI, and gauge fluxes arising from interactions. In work done in parallel with the present work, Ye et al. [13], focusing on the metallic regime, address the relationship between the AHE, Berry phases [14] and the SOI, and like the present work, incorporate the effect of topological spin excitations.

Localization of carrier states in manganites: Several general ideas support the notion that the carrier states are localized at temperatures near to the (zero magnetic field) FPT, as well as at higher temperatures. Approaching the FPT from the ferromagnetic side, there is a net magnetization of the core spins, but strong thermal fluctuations...
render typical instantaneous configurations of the spins rather inhomogeneous. Among these fluctuations there are “hedgehog” excitations which, owing to their topological stability, are long-lived, and become more numerous as the FPT is approached \([17\).\] Due to the resulting inhomogeneity, the carrier-transfer matrix elements are reduced \([17\). In the quasi-static approach, the (fast) carrier motion takes place through a slowly (time-)varying background core-spin configuration. In generic instantaneous random backgrounds, the carriers are expected to be localized. Support for this notion comes from the close similarity between manganites and a system of randomly located identical impurities (i.e., off-diagonal disorder) for which localization has been established by Lifshitz \([13\].\) Although spin-induced randomness in manganites \(\text{arising from the random } \cos(\theta/2) \text{ factors}\) is weaker than the randomness considered in \([13\], the two systems are expected to exhibit similar localization behavior. Furthermore, the condition for localization (viz., that the characteristic spatial scale of the outer-shell wavefunctions be much smaller than distance between sites) is well obeyed in manganites. Therefore, provided that there is appreciable randomness in the core-spins orientations, the transport properties should be determined by the short-distance physics of clusters of ions and magnetic correlations between such clusters. Moreover, nonmagnetic disorder and possible states bound to the A-ions are capable of amplifying the trend towards localization \([16\].\)

Thus the following picture of transport in manganites emerges. (i) In the paramagnetic insulating state, the percolative motion of strongly localized carriers is suppressed by magnetic randomness. (ii) With decreasing temperature, the carrier hopping (which is assisted by phonons) becomes less frequent, so that the resistivity grows, and (iii) reaches a maximum when the core spins become sufficiently correlated that a tenous but infinite conducting network emerges. (iv) With further reduction in temperature, the resistivity decreases abruptly, in line with the traditional percolation picture \([14\], as more and more hopping paths become available to carriers, owing to further alignment of core spins. This abrupt decrease terminates when the newly available hopping paths are effectively shunted by the existing network. (v) Further decrease in temperature leads to further core-spin alignment and, ultimately, to the metallic state.

Anomalous Hall effect and the Pancharatnam phase: In order to discuss the AHE in conditions of charge-carrier localization, we begin by considering a triad of magnetic sites formed by neighboring Mn ions, as shown in Fig. 1. Within such triads, there is an elementary AHE, which arises from interference between hopping processes connecting two sites: e.g., between the direct process (having complex amplitude \(A\)) and the indirect process of hopping via the third site (with amplitude \(A'\)). Ignoring any AB flux (as we shall not be concerned with the OHE), we observe that any phase difference between the amplitudes \(A\) and \(A'\) stems from spin quantal phases and transfer-assisting mechanisms (e.g., electron-phonon processes); we call the latter transfer phases \(\phi_T\).

To understand the nature of the spin quantal phases we examine the single-particle quantum mechanics of a carrier hole added to a triad of Mn\(^{3+}\) ions. We regard the spin-3/2 core spins of the Mn ions as large enough to be treated classically, so that one can assign a definite direction to each. Thus, a generic configuration is characterized by the unit vectors \(\{\mathbf{n}_1, \mathbf{n}_2, \mathbf{n}_3\}\) located at the triad of sites \(\{\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3\}\) (see Fig. 1). Due to Hund’s Rules there is, at each site, a single state available to the added hole, its spin opposing the core-spin direction. We treat the remaining spin (and orbital) states as simply being inaccessible. Postponing to below the effects of SOI, we assume that the transfer of holes (being effected by either the kinetic energy or the electron-phonon interaction) has no effect on the spin of the carriers. However, such transfer in the presence of the constraints set by the core-spin orientations has a striking effect on the quantal dynamics of the carriers: in the quantal amplitude for a hole in the presence of the constraints set by the core-spin, \(\Omega/2 = \tan^{-1} \left[ \mathbf{n}_j(\mathbf{n}_2 \times \mathbf{n}_3)/(\mathbf{n}_1 \cdot \mathbf{n}_2 + \mathbf{n}_2 \cdot \mathbf{n}_3 + \mathbf{n}_3 \cdot \mathbf{n}_1) \right] \), which modulates the interference between direct and indirect hopping between sites of a triad. \(\Omega\) is the (oriented) solid angle of the geodesic triangle on the unit sphere having vertices at \(\{\mathbf{n}_1, \mathbf{n}_2, \mathbf{n}_3\}\). It is the quantal analogue of the classical optical phase discovered in the context of polarized light by Pancharatnam \([11,12\].\) What Pancharatnam showed is that a cyclic change of the polarization state of light is accompanied by a phase shift (i.e., a phase anholonomy) determined by the geometry of the cycle, as represented on the Poincaré sphere of light polarizations, via the area \(\Omega\) of the geodesic polygon whose vertices are these polarizations.

In the DE electronic analog, the transporting of a d-shell carrier to an ion with a differently oriented core spin in a spin-independent process amounts to a connection, which determines the phase of the spin state in terms of the sequence of sites visited. A hole returning to a site returns to the same spin-state, except that its phase is augmented by a quantal Pancharatnam phase, determined by the geometry of the cycle, as represented on the sphere of core-spin orientations, via \(\text{half the area of the geodesic polygon whose vertices are these orientations. In contrast to Berry’s adiabatic phase} [13\], the phenomenon
described here is associated with sudden changes in the carrier-spin state, and need not be slow.

In the hopping regime, this Pancharatnam phase leads to an AHE in an elementary triad in much the same way that an AB flux leads to the OHE in Holstein’s spinless model \(10\). In Holstein’s model, carrier hopping between sites of a triad occurs due to carrier-phonon interaction; the Hall current arises due to the interference of direct and indirect hopping. The transfer phase is nontrivial \((\phi_T = \pi/2)\) when this interference involves processes assisted by two phonons \(10\). (For the longitudinal conductivity, a single phonon-assisted transfer is sufficient.) In a uniform magnetic field \(B\), processes associated with a nontrivial \(\phi_T\) lead to an OH conductivity \(10\),

\[
\sigma_{\text{OH}} = G\{\epsilon_j\} \sin \phi_T \sin (B \cdot Q/\phi_0), \tag{1}
\]

where \(\phi_0\) is the (electromagnetic) flux quantum, \(Q\) is the (oriented, real space) area of the triad, and \(\{\epsilon_j\}_{j=1}^3\) are the energies of the three single-particle eigenstates, which are invariant under reversal of the AB flux. The explicit expression for \(G\) can be found in Ref. \(10\). \((G\) also depends on the populations of these states, which themselves may depend on particle-particle correlations.)

We now turn from the OHE in a spinless triad to the elementary AHE in a triad of magnetic sites. Like the OHE, this AHE results from two-phonon processes, but is due to the Pancharatnam phase instead of the AB phase. (We do not yet included the effects of the SOI.) \textit{Mutatis mutandis}, we arrive at the AH conductivity,

\[
\sigma_{\text{AH}} = G\{\epsilon_j\} \sin \phi_T \cos \frac{\theta_{13}}{2} \cos \frac{\theta_{32}}{2} \cos \frac{\theta_{21}}{2} \sin \frac{\Omega}{2}, \tag{2}
\]

where \(\cos \theta_{jk} \equiv \mathbf{n}_j \cdot \mathbf{n}_k, \cos (\theta_{jk}/2)\) are Anderson-Hasegawa factors, and \(\{\epsilon_j\}_{j=1}^3\) are the energies of the three single-particle eigenstates consistent with Hund’s Rules, these energies depending on \(\mathbf{n}_j \cdot \mathbf{n}_k\) and \(\cos (\Omega/2)\). Note that \(G\) is invariant under Pancharatnam flux reversal \(\Omega \rightarrow -\Omega\), and \(\sigma_{\text{AH}}\) is odd under it.

We have shown that, for a triad with given set of core-spin orientations, an AHE arises from the quantal Pancharatnam flux. However, there is a significant difference between this AHE and the OHE. In the former (nonmagnetic) case, a uniform applied magnetic field leads to a net macroscopic OHE, even though contributions of triads may cancel one another \(23\). In the latter case (magnetic sites, Pancharatnam flux, and no SOI), even the presence of macroscopic magnetization of the core spins is insufficient to cause a macroscopic Hall current. The reason for this is that in obtaining the macroscopic AH current from Eq. \(2\) we must average over the configurations of the core spins. In the absence of SOI, the distribution of these configurations, although favoring a preferred direction (i.e., the magnetization direction \(\mathbf{m} \equiv \mathbf{M}/|\mathbf{M}|\)), is invariant under a reflection of all core-spin vectors in any plane containing the magnetization. This fact, coupled with the fact that \(\{\epsilon_j\}\) are also invariant under such reflections, guarantees that the macroscopic AH current will average to zero. (We do, however, expect significant AH current noise, in the FTP regime, owing to the fluctuations of the Pontryagin charge \(22\) of the triads of core spins and, hence, elementary Pancharatnam fluxes.)

In order to capture the AHE in materials such as manganese, we must consider not only the Pancharatnam phase but also some agent capable of lifting the reflection invariance of the energies \(\{\epsilon_j\}\) and the distribution of core-spin configurations, and hence of inducing sensitivity to the sign of the Pancharatnam flux. Such an agent is provided by the SOI, \(H_{\text{so}} = \alpha \mathbf{p} \cdot (\mathbf{r} \times \nabla U)\), where \(U\) includes ionic and impurity potentials, \(\alpha\) is the SOI constant, \(\mathbf{p}\) is the electron momentum, and \(\sigma\) are the Pauli operators. The SOI leads to an effective SU(2) gauge potential \(A_{\text{so}} = \alpha m (\mathbf{r} \times \nabla U)\), providing an additional source of quantal phase. For a given core-spin configuration, SOI favors one sense of carrier-circulation around the triad over the other, and thus favors one sign of Pancharatnam phase over the other.

There are two resulting contributions to the AHE. The first, \(j^{(1)}_{\text{AH}}\), arises from the SOI-generated dependence of \(\{\epsilon_j\}\) on the three vector-products \(N_{jk} \equiv \mathbf{n}_j \times \mathbf{n}_k\) which, together with the magnetization direction \(\mathbf{m}\), yield a preferred value for the triad Pontryagin charge \(q_p = [\mathbf{n}_1 \cdot (\mathbf{n}_2 \times \mathbf{n}_3)]\) and, hence, a preferred Pancharatnam flux. To see the origin of this dependence on \(N_{jk}\), let us analyze corrections, due to the SOI, of hole eigenenergies. If the on-site energies of the holes are nondegenerate, it is straightforward to determine that phase sensitivity first enters at third order (in the transfer matrix elements): \(\delta \epsilon_j = \sum_h \frac{k_p}{2} (\frac{\partial}{\partial \mathbf{k}} \langle \epsilon_j^2\rangle - \langle \epsilon_j \rangle^2)\), where \(T_{jk} \equiv P_j V_{jk} P_k\) are the hopping amplitudes, \(V_{jk}\) are the hopping matrix elements, and \(P_j\) denotes a trace in spin space. (For degenerate \(\epsilon\’s\) one should obtain the splitting of the \(\epsilon\’s\) due to transfer in the absence of SOI, and then include SOI at the final step, arriving at the result to be given below.) The hopping matrix elements are sensitive to the SOI quantum phase, and can be written in the form \(V_{jk} = V_{jk}^{\text{orb}} L_{jk}\), where \(L_{jk} \equiv (1 + i \mathbf{g}_{jk} \cdot \mathbf{r}_{jk} / h)\), \(V_{jk}^{\text{orb}}\) is an orbital factor, and \(\mathbf{g}_{jk}\) (\(\propto \alpha_{\text{so}}\) is an appropriate vector that describes the average SOI for the transition \(j \rightarrow k\) \(23\). Then, e.g., the first-order (in \(\alpha\)) shifts in the \(\epsilon\’s\) are given by

\[
\delta \epsilon_j \propto \text{Tr} T_{13} T_{32} T_{21} = 4 \text{Re} \text{Tr} P_1 L_{13} P_3 L_{32} P_2 L_{21} = -N \cdot \mathbf{g} + 2 (N_{13} \cdot \mathbf{g}_{13} + N_{32} \cdot \mathbf{g}_{32} + N_{21} \cdot \mathbf{g}_{21}), \tag{3}
\]

where \(N \equiv N_{13} + N_{32} + N_{21}\), and \(g \equiv g_{13} + g_{32} + g_{21}\). When \(U\) in the SOI is a superposition of spherically-symmetric ionic potentials, the vectors \(g_{jk}\) have a transparent geometrical meaning, and are proportional to the triangle area \(Q\). In this case, \(g_{jk} = a_{jk} (R_j - R_k) \times (R_k - R_h)\). Then the SOI-generated shift in the carrier eigenenergies has the Dzyaloshinski-Moriya form \(23\). By incorporating the shifts \(3\), together with the Pancharatnam phase, we arrive at the elementary AH conductivity
\[ \sigma^{(1)}_{\text{AH}} = \mathbf{n}_1 \cdot (\mathbf{n}_2 \times \mathbf{n}_3) \sum_j \delta \varepsilon_j \frac{\partial G}{\partial \varepsilon_j}, \]

As discussed above, Eq. (3) has a nonzero macroscopic average, owing to the presence of a characteristic Pontryagin charge constructible from the \( N_{jk} \), that feature in the energy shifts, and the magnetization direction. A second consequence of the SOI-generated carrier-energy shift (3) leads to the second contribution, \( \sigma^{(2)}_{\text{AH}} \). Due to the feedback of the (fast) carrier freedoms, which provide an effective potential for the (slow) spin system, determined by Eq. (3), the equilibrium probabilities of spin configurations having opposing Pancharatnam fluxes will no longer be equal. (For this contribution, which is related not to \( \partial G/\partial \varepsilon_j \) but to \( G \) itself, there is no need to account for SOI-induced carrier-energy shifts in the current now being averaged over a nonsymmetric spin-configuration distribution.) A contribution with this origin has also been considered in Ref. [3]. \( \sigma^{(1)}_{\text{AH}} \) and \( \sigma^{(2)}_{\text{AH}} \) are of the same order of magnitude.

We now consider the question of how the physics of elementary triads relates to the macroscopic properties of manganites. For hopping conductivity, the pathways taken by the current depend sensitively on the details of the core-spin configuration, and regions having certain local spin configurations will tend to be avoided by the current. This fact renders rather subtle the spin-configuration averaging procedure, which must also account for effects such as local spin correlations and excitations of various types. Let us try to identify which triads the AH current tends to favor. To favor their participation in the conducting network, the triads should at least have positive components along the magnetization direction. For magnetic compatibility with its neighbors, the net magnetization of the triad should be roughly that of the bulk. Furthermore, to contribute appreciably to the AH current, the triad should be as splayed as possible, given the above constraints. This favors symmetrical configurations of the triad spins; we call these triads optimal triads. As we shall see in the companion Letter [1], these observations allow us to explain the striking experimental finding that the Hall resistivity depends on the magnetic field and temperature only through the magnetization and, moreover, to predict the explicit form of this dependence.

**Spin-orbit quantal phase and AHE in nonmagnetic triads:** We conclude with a remark concerning the hopping AHE in systems with nonmagnetic ions (in which case no Pancharatnam phases arise). In this case, the SOI quantal phase itself leads to an AHE. In the presence of the SU(2) gauge potential \( A_{\text{so}} \), electrons moving around a nonmagnetic triad acquire a full SU(2) phase, not projected due to Hund’s Rules. Due to carrier-spin polarization, this phase leads to an AH current in the same way that the AB flux leads to the OHE [10]. We emphasize that that the OH and AH effects in such systems should be experimentally distinguishable from one another. For example, the AHE in the hopping regime should be observable in inversion layers of doped semiconductors in the absence of magnetic field, when electron spin-polarization is induced by circularly polarized light.

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