Kronig-Penney-Ising picture of colossal magnetoresistance

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From general arguments, it is shown that a magnetic Kronig-Penney model based on the thermodynamics of an Ising model can be used for describing the colossal magnetoresistance (CMR) phenomenon. The model considers a tunnelinglike transmission process of hopping electrons through a dynamic lattice characterized by evolving magnetic clusters. In this model, correlations between the magnetic states are considered to be more relevant than the lattice strain effects for obtaining the CMR features. Physical arguments lead to the theoretical description of the intrinsic temperature and field dependences of the CMR observed in typical manganite materials. [S0163-1829(99)13713-5]

It is often argued that the colossal magnetoresistance (CMR) is due to a complicated interplay between electronic and lattice degrees of freedom (polaron). In the following, we show that the magnetic degrees of freedom contain the basic contribution to the field and temperature CMR behavior.

Let us recall the intriguing CMR phenomenon. The resistivity of perovskite materials has an ''enormous'' peak at some temperature $T_{\text{M-I}}$ considered as related to a metal-insulator (M-I) transition. This transition is often accompanied by a second-order ferromagnetic transition at a temperature T_c. Under a magnetic field, the $\rho(T,H)$ appreciably decreases near the temperature where $\rho(T,0)$ has its maximum. Many theories have been proposed for CMR, but the exact temperature and field behaviors are not yet fully satisfactory understood.

The CMR phenomenon in perovskite based materials is here below explained through critical phenomenon behaviors and analytical laws for describing the main observations. The model seems much more simple than previously presented models. At this stage, it should not be requested that it solves all known puzzles for all compounds with CMR. Simplifications are made here below for presenting the approach. In fact, we present an alternative view with respect to the most popular theories based on the magnetic polaron idea[1] including the double exchange scenario (DES).[2] Various features seem hard indeed to be put into this polaronic-DES framework, e.g., at high temperature the small polaron picture with a few meV activation energy due to lattice distortion[3] does not directly match to the large polaron picture of the metallic state at low temperature, where a band picture should emerge and hold below the magnetic transition.[4] In fact, the question can be raised whether there is a change in the mobility of carriers or in their number (or both) or whether there is even a change in the band structure necessarily implying some conductivity transition. For instance, it can be accepted that the mean-free path is small at high temperature, but at low temperature the zero-field resistivity dependence seems to be that of metallic ferromagnets.[5,6] Also, transport features at fixed magnetization m indicate that for large or small magnetization values the exponentially stretched dependence is markedly different.[7,9–11]

The role of lattice strain as shown on films grown on different substrates does not seem to explain the findings, and should be supplemented by taking into account magnetic domainlike effects.[8] This implies that magnetic features should be reemphasized. The DES has been recently also much criticized because it was thought that it could not explain the qualitative difference in electrical conduction for the whole range of x in one of the most often studied CMR materials, i.e., $La_{1-x}Ca_xMnO_3$. However, it has been shown that a more correct treatment of the DES leads to an acceptable view in the interesting doping range $[x=0.16,0.40]$, i.e., where the magnetic and conducting transitions are high (near room temperature). Outside this interval, further work should be envisaged since the magnetic structure has quite another periodicity, but the following concepts should still hold as it will be easily seen.

In fact, three simple ingredients can be combined in a realistic way in order to emphasize the magnetic degrees of freedom role, i.e., the Ising model for the magnetic spins, the Drude formula for the electrical conductivity, and some type of scattering for hopping electrons. By combining these basic ingredients, we essentially take into account magnetic cluster effects, but the more so their correlated fluctuations. The former scattering strength and magnetic state life time are the only microscopic physical parameters.

Notice that (i) grains are usually pretty small, whence there is much grain boundary scattering. We neglect such a (extrinsic) background term here, though it can be easily inserted in the scattering strength if necessary. (ii) The number of carriers is also kept field and temperature independent. To take into account a density of states temperature (and even field) dependence is a rather trivial generalization to be made, within a self-consistent picture taking into account band and localized state carriers (with possible spin states). (iii) The temperature dependent lattice distortion[3] role for hopping charge carriers is also neglected here, but can be included at a later stage again in the definition of a

temperature-dependent lattice parameter and mapped into an effective carrier mass or into an effective localized spin-coupling strength.

In so doing, we do not claim that the present model should give precise quantitative values at this time because of the extremely limited number of parameters that we are using. Nevertheless, the theory will be in good agreement with experimental data. Moreover, extensions seem easy in light of the *paraphernalia* of solid state physics ideas and techniques. This model is surely not the unique alternative to the magnetic polaron model. However, this paper shows that the correlation between spins is the key ingredient to be integrated in the understanding of the CMR phenomenon, seen as a transport property in presence of magnetic states rather than a set of such near equilibrium specific states controlled by some unknown exchange interaction.

For the following developments, let us reduce the problem to a two-dimensional $(d=2)$ case, allowing as a first approximation an in-plane conduction like in thin films.[8]

The Ising model[13] on a $d=2$ square lattice is used for the spins on the manganese sites assumed to represent the local magnetization of the system. This simplified picture allows for a faster way of obtaining the following results, but the spin-spin exchange interactions could be as well of indirect origin as in DES without loss of generality. Simply, we let each lattice site i contains a two-state spin $\sigma_i=\pm 1$. The dimensionless Hamiltonian reads

$$E=-K\sum_{i,j}\ \sigma_i\sigma_j-h\sum_{i}\ \sigma_i\,, \tag{1}$$

where $K=J/kT$ is the dimensionless interaction between nearest-neighboring i,j spins, and h is the dimensionless magnetic field H/kT orienting the spins. The $(h=0,\ K>0)$ case is a classical problem taught in classrooms because it has a nontrivial phase transition as demonstrated by Onsager, i.e., a logarithmic divergence of the specific heat near the reduced critical temperature $K_c=-\frac{1}{2}\ln(\sqrt{2}-1)$. The Ising model implies that there are droplets (clusters) of, e.g., $+1$ or -1 spins that nucleate, grow, coalesce, and disappear as a function of temperature.[12] It is well known[13,14] that a ferro-paramagnetic transition takes place exactly at K_c for $h=0$ on such a lattice. In each grain or if the intergrain coupling is adequate, clusters of respectively up and down spins coexist and the average size ξ of these clusters diverges at K_c following $\xi\sim|K-K_c|^{-\nu}$ with $\nu=1$.[15] The other properties like $m(K,h)$ are not known exactly because the Ising model in a field has not yet been solved.

In view of the partially covalent-ionic bonding in the plane, the quasilocalized carriers are supposed to be (spinless) electrons having a linear hopping motion along the electrical field imposed across the lattice. The Lorentz force is neglected here because of the rather short mean-free path.

In the computer experiments, we launch electrons toward the right at random from the left side of the ''sample.'' Each electron jumps to the right from a site to the next nearest-neighbor site at each time step as follows. In a so-called ''magnetic cluster,'' the electron hopping is free. When an electron reaches a magnetic cluster wall, the electron is stopped with a probability $(1-p)$ or transmitted with a probability p. By analogy with tunneling effect, p is assumed to be the exponential of some measure of the cluster size s ahead of the electron, i.e.,

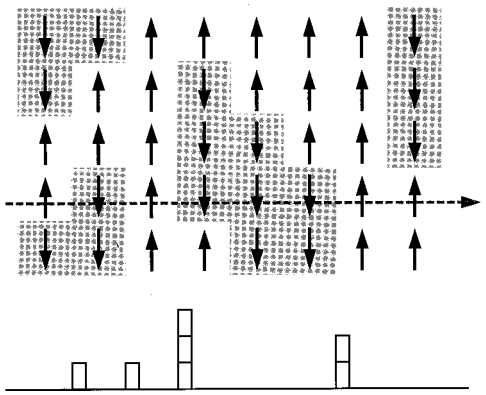

FIG. 1. Schematic illustration of the magnetotransport process as discussed in the text: (a) The square lattice on which $+1$ and -1 magnetic domains are distributed. On this lattice, one electron follows the linear motion illustrated by the dashed arrow; (b) The barrier landscape viewed by this electron at the time corresponding to the snapshot of (a).

$$p=\exp(-\gamma s), \tag{2}$$

where γ is a dimensionless parameter that is like a potential barrier strength of the cluster. There is no retention time upon a site nor phonon nor magnetic drag, nor other type of scattering. At each time step the magnetic structure is recalculated according to a Monte Carlo procedure for the Ising Hamiltonian. We count the carrier arrival time τ on the right-hand side of the ''sample.'' This time obviously depends on the sign distribution fluctuations of the spins for a given K and h on the line during the electron hopping.

Following the Drude formula, the resistivity is directly obtained from

$$\rho=\frac{\tau}{L}, \tag{3}$$

where L is the size of the lattice. At high temperature, when the spins are completely disordered the resistivity is of course large; it is smaller but not negligible at lower temperature; near the critical point K_c for $h=0$, the resistivity should become enormous: indeed the electron is a little bit ''at a loss'' because the spin fluctuations are huge and much hamper the electron motion. A magnetic field stiffens the clusters (or reduces the fluctuations). Therefore, the resistivity should be reduced because the electron has a greater chance to find its way through. Thus, the qualitative features of CMR are immediately found in this simple model.

Notice that this CMR version is somewhat like a temperature-dependent ''magnetic Kronig-Penney model'' in an electric field since each wall is a potential barrier of which strength γ is controlled by magnetic and thermal conditions [Fig. 1(b)], just like in disordered thin films.[16,17] The nontrivial (new) ingredient is that the ''barriers'' are correlated and controlled by the thermodynamics of the Ising model, in space and time.

In order to obtain a good numerical convergence, we have left the magnetic system to reach a pseudosteady state before

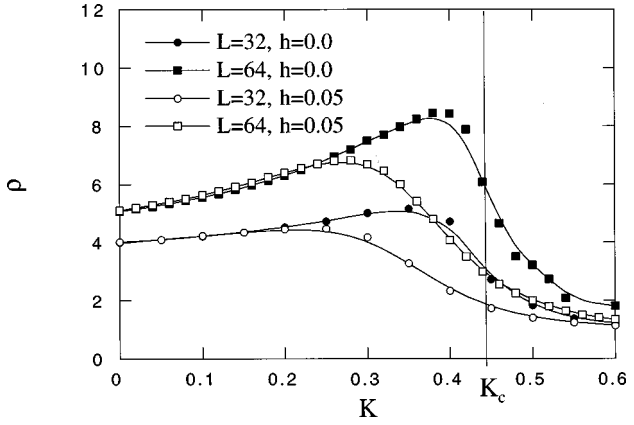

FIG. 2. Theoretical resistivity ρ as a function of K. Two cases are shown: $h=0$ and $h\neq 0$. Different lattice sizes are illustrated: $L=32$, $L=64$.

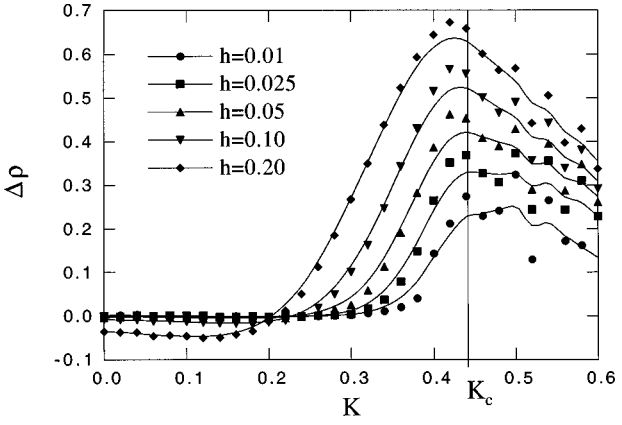

FIG. 3. Magnetoresistance $\Delta\rho$ as a function of K.

launching the ''electrons.'' Lattices up to 256×256 were used. For conciseness, we fixed arbitrary herein $\gamma=1$. The results do not change drastically with γ. Clearly, at a later stage of investigations, the γs term can be itself temperature and/or magnetization dependent for taking into account the lattice strain. More complicated schemes taking into account different spin channels can be also imagined within an effective medium approximation. Here we consider that the spin density corresponds to the case where the majority of polarized (up or down) spins is much larger than the minority. The spinless approximation of the charge carrier is not even a strong approximation. Indeed, it is clear that the true electron hopping is depending on the availability of a neighboring state of similar nature. Thus, the electron will be rather stopped in front of a wall after which the spins states have the opposite sign to that of the incoming electron. This is also in the DES spirit in fact.

Figure 2 presents the resistivity ρ as a function of K. Both zero and nonzero magnetic field cases are illustrated. The ferroparamagnetic transition at K_c is indicated by the vertical line. As expected, a bump is observed in ρ below K_c and an inflexion at K_c. Moreover, the bump height decreases as h increases. Such a bump can be viewed as the signature of a percolation transition[18,19] rather than a strict metal-insulator transition. On both sides of K_c, the resistivity ρ decreases exponentially. This shows that the experimentally observed decreasing behavior in the high-temperature phase might have nothing to do with a semiconducting phase or a ''metal-insulator'' transition as often claimed, but rather to the number of available final state in the scattering.

The dimensionless excess resistivity defined by

$$\Delta\rho=\frac{\rho(0)-\rho(h)}{\rho(0)} \tag{4}$$

is shown as a function of K in Fig. 3. Data due to different values of the magnetic field are shown. This quantity $\Delta\rho$ is found to be independent of the length L of the lattice. The qualitative features of the CMR are well observed, i.e., (i) a peak at some intermediate temperature, (ii) an increase of $\Delta\rho$ with h, (iii) a shift of the peak towards high temperatures and (iv) a wide transition region.

A nontrivial test of the model and theory is in order in Fig. 4, which presents the evolution of the maximum of $\Delta\rho$ as a function of h in a semilog plot. The value of $\Delta\rho$ for $K=K_c$ is also given. A logarithmic increase of $\Delta\rho$ with h is observed, i.e.,

$$\Delta\rho\sim\ln h. \tag{5}$$

This logarithmic behavior can be found in data on $La_{1-x}Ca_xMnO_3$, $La_{1-x}Mg_xMnO_3$, and $Pr_{1-x}Sr_xMnO_3$ compounds[8,20] (Fig. 5). The predicted logarithmic behavior (straight lines) seems to hold quite well for moderate to high-magnetic fields. It is true that one should distinguish between low-field, moderate, and high-field CMR.[21] At lower field values (e.g., $H<0.2$ T) inhomogeneities play a relevant role,[8,21] an effect outside the present investigation.

In addition, since CMR can be seen as related to a second-order critical phenomenon, it should be possible to describe CMR with the help of scaling arguments.

For $h=0$, the time for an electron to cross the system of size L is approximately given by

$$\tau=L+\frac{L}{\xi}\sum_{i=1}^{+\infty}[1-\exp(-\gamma\xi)]^i, \quad \text{for} \quad \xi<L, \tag{6}$$

where ξ is the characteristic size of the clusters and $[1-\exp(-\gamma s)]^i$ is the probability that the electron remains

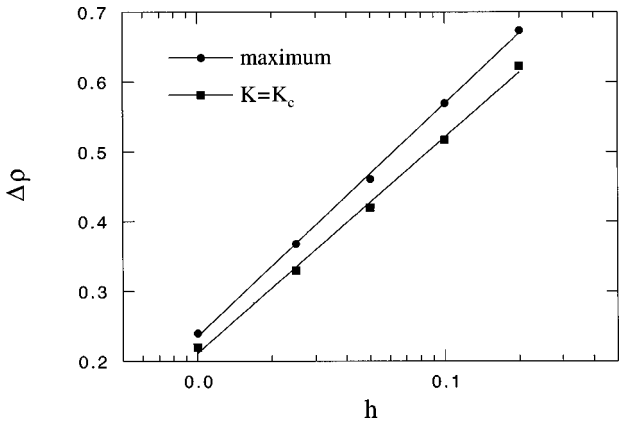

FIG. 4. Semilog plot of the evolution of the maximum of the magnetoresistance $\Delta\rho$ as well as $\Delta\rho$ for $K=K_c$ as a function of the magnetic field h.

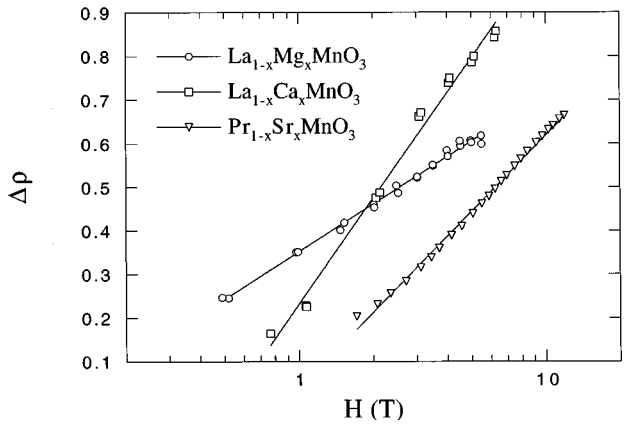

FIG. 5. Semilog plot of the evolution of the magnetoresistance $\Delta\rho$ as a function of the magnetic field for various compounds: $La_{1-x}Ca_xMnO_3$ from Ref. 8, $La_{1-x}Mg_xMnO_3$ from Ref. 8, and $Pr_{1-x}Sr_xMnO_3$ from Ref. 20. The magnetoresistance is taken near T_c. In all cases, the logarithmic behavior (straight line) predicted by the model [Eq. (6)] is clearly observed to hold over more than one decade of magnetic-field values.

blocked i successive times on a magnetic wall before being transmitted to the next site. Using the Drude formula, one has

$$\rho = 1 + \frac{1}{\xi \ln[1-\exp(-\gamma\xi)]}. \tag{7}$$

The above relationship neglects the correlations between wall fluctuations. Nevertheless, the form of Eq. (8) explains the finite size effects (Fig. 2), to be seen when $\xi \approx L$ and for small L values. Close to the ferroparamagnetic transition, one can develop Eq. (8) and obtain

$$\rho \sim \exp(\xi) \approx \exp\left(\frac{1}{|K-K_c|}\right). \tag{8}$$

The wall fluctuation correlations being in fact negligible far away from K_c, the above exponential scaling behavior seems to be correct on both sides of K_c. A quantitative comparison to available data is not immediate at low field ($H < 300$ mT) since grain boundary background is very much field dependent there. In such a regime because of inherent inhomogeneity of (usually small) grains, the $\Delta\rho(h)$ dependence is indeed to be known as nonuniversal.[22]

In the case of large h values, it has been argued elsewhere[23] that the in-plane correlation length ξ near K_c scales as

$$\xi \sim h^{-\nu/(b\delta)}, \tag{9}$$

where $b = 1/8$ and $\delta = 15$ for the $d = 2$ Ising model whence the $\Delta\rho \sim \ln h$ behavior should not be expected. Introducing the latter scaling relation in Eq. (9) leads to a stretched exponential, a law that qualitatively implies some apparent parallelism of $\Delta\rho(h)$ decaying curves with an amplitude being temperature dependent. This feature can be understood as resulting from the non-negligible far away from K_c cluster fluctuations and from some drift due to the magnetic field h.

It may be recalled that an analogous treatment to ours occurred in the pioneer theoretical work of Fisher and Langer[24] using a $d = 2$ Ising model instead of a true $d = 3$ model for describing the experimental results on the resistivity at ferromagnetic transitions. A divergence was predicted at T_c while experimentally an inflexion (only) was seen. This paradoxical situation was found to be due to using a two-dimensional rather than a three-dimensional Ising model. Extensions towards a better agreement to $d = 3$ cases should follow the same generalizing lines[5] in the future for our CMR model.

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