Spin mechanism for hopping magnetoresistance in La_2CuO_4

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A spin mechanism is proposed for a hopping magnetoresistance. The mechanism is based on a splitting of impurity states by spin in a molecular field and on a difference between the probabilities for hops with and without spin flip. As a result, the resistance depends on the magnitude and relative orientation of the molecular fields near impurities. This orientation is in turn controlled by an external magnetic field.

Insulating La_2CuO_4 crystals exhibit[1] a three-dimensional hopping conductivity with a variable hop length $R = R_0 \exp[(T_0/T)^{1/4}]$, where $T_0 = 10^5$–10^6 K, at least at low temperatures.[2] The magnetoresistance of these samples is extremely unusual.[3,4] As a field $\mathbf{H}\|\mathbf{b}$ ($\mathbf{b}$ is the orthorhombic axis, perpendicular to the CuO_2 planes) is increased, $R(H)$ decreases abruptly at $H = H_c$ (Fig. 1). The field H_c is a critical field, at which a weak ferromagnetic transition occurs from an antiferromagnetic ordering to a ferromagnetic ordering of the moments of planes.[3,5] The latter arise because of a noncollinearity of the Cu^{2+} spins ($\mathbf{S}_n$), itself a consequence of the Dzyaloshinskiĭ-Moriya interaction ($\boldsymbol{\omega}_2[\mathbf{S}_n \times \mathbf{S}_m]$), where $\boldsymbol{\omega}_0$ is proportional to the orthorhombic rotation vector. The corresponding spin rotation angle is[3] $\theta \approx 3\times 10^{-3}$.

The abrupt change in the magnetoresistance results from a reorientation of localized spins [Fig. 1(a)], which is evidence of a strong interaction of charge carriers with

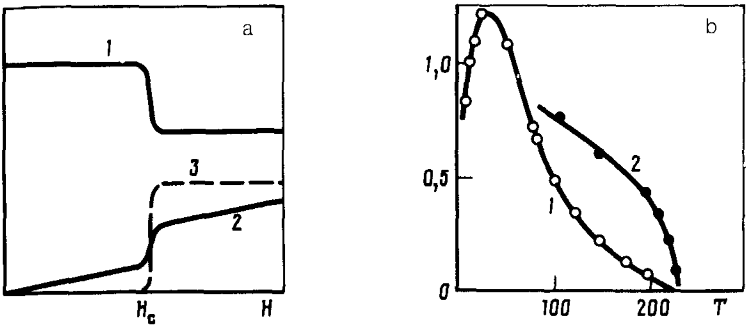

FIG. 1. a: Qualitative field dependence. 1—Of the resistance R; 2—of the macroscopic magnetization M^F; 3—of the quantity $|\mathbf{M}^+| = |\mathbf{M}_1 + \mathbf{M}_2|$ (the "staggered magnetization"[3]), where $\mathbf{M}_\alpha$ is the average antiferromagnetism vector in neighboring planes ($\mathbf{H}\|\mathbf{b}$). b: Temperature dependence. 1—Of the relative jump in the resistance, $\Delta R/R_H$: 2—of the jump in the ferromagnetic moment, ΔM^F (from Ref. 3). Here $\Delta M^F \sim \theta M$.

spins. If we assume that the degree of orthorhombic nature is uniform throughout the sample, we would have $\Delta R/R \propto \theta^2 \sim 10^{-5}$, but this result is completely at odds with the experimental results. In an effort to resolve this contradiction, let us assume that the degree of orthorhombic nature intensifies near impurities at which holes are localized. A possibility of this sort was studied in Ref. 6 for certain types of acceptors. While we have $\omega_0 \approx 8$ K in the volume (Refs. 3 and 4), we have $\omega = \omega_0 \varphi_{\text{loc}}/\varphi_0$ locally, near an impurity, and this quantity can reach a value ~ 200 K (with $\varphi_{\text{loc}} \sim 1$ and $\varphi_0 \approx 0.05$ as the orthorhombic rotation angles near the impurity and in the volume).

Calculations based on very simple models[6,7] show that the spin of a neutral acceptor is $S = 1/2$. Acting on each spin $\mathbf{S}_i$ is a molecular field

$$\mathbf{h}_i = [\mathbf{n}_i \times \vec{\omega}], \tag{1}$$

where $\mathbf{n}_i$ is the unit antiferromagnetism vector at the position of acceptor i; here $\langle \mathbf{n}_i \rangle = \mathbf{M}_\alpha$, where $\alpha = 1,2$ for neighboring layers. We wish to stress that $\langle \mathbf{h}_i \rangle$ depends on only the layer index α, not on the position of the impurity within the layer. The energy of an impurity associated with the orientation of its spin in $h_i \sigma_i$, where $\sigma_i = \pm 1/2$ is the projection of $\mathbf{S}_i$ onto $\mathbf{h}_i$.

The assumption that there is a pronounced local orthorhombic deformation near an impurity leads to hops of a polaron nature. We will restrict the present letter to temperatures which are not too low: $T > T^* = \hbar\bar{\omega}/2 \ln(W/\Delta) \sim 10$ K, where $\hbar\bar{\omega} \sim 100$ K is the frequency of a local orthorhombic mode,[6] and W/Δ is the ratio of the polaron shift to the width of the Mott band.[8] The dependence of the hopping probability on $\Delta_{ij} = \epsilon_j + h_j \sigma_j - \epsilon_i - h_i \sigma_i$ (the difference between the energies of the final and initial states) then reduces[9] to a factor $\exp\{-\Delta_{ij}/2T\}$, regardless of the sign of Δ_{ij}, where ϵ_i is the nonmagnetic part of the energy of the impurity state.

Estimates show that the following inequalities hold:

$$\tau_S \ll \tau_n \ll \tau_0, \tag{2}$$

where τ_0 is the lifetime of a hole at one impurity, τ_n is the time scale of the fluctuations of $\mathbf{n}$, and τ_S is the relaxation time of an impurity spin. From inequality (2) we can find the probability for the filling of impurity sublevels σ_i: $g(h_i\sigma_i) = \exp(-h_i\sigma_i/T)/2\mathrm{ch}(h_i/2T)$. The quantization axis $\mathbf{S}_i$ tracks the instantaneous direction of $\mathbf{h}_i$. The probability for a transition $i \to j$ over times smaller than τ_n but larger than τ_S [i.e., for the instantaneous values of $\mathbf{n}$ and $\mathbf{h}$; see (1)] is

$$W_{ij}^{\mathbf{n}_i,\mathbf{n}_j} = w_0 \exp\{-\frac{2\tilde{r}_{ij}}{a}\} \sum_{\sigma_i\sigma_j} \exp\{-\frac{\Delta_{ij}}{2T}\} g(h_i\sigma_i)[\frac{1+\eta}{2} + 2(1-\eta)\sigma_i\sigma_j \frac{(\mathbf{h}_i\mathbf{h}_j)}{h_i h_j}]$$

$$= w_0 \exp\{\frac{\epsilon_i - \epsilon_j}{2T} - \frac{2\tilde{r}_{ij}}{a}\}[\cosh(h_i/2T)]^{-1}$$

$$\times \{(1+\eta)\cosh(\frac{h_i}{4T})\cosh(\frac{h_j}{4T}) + (1-\eta)\sinh(\frac{h_i}{4T})\sinh(\frac{h_j}{4T})\frac{(\mathbf{h}_i\mathbf{h}_j)}{h_i h_j}\}, \tag{3}$$

Here $\tilde{r}_{ij}/a = [\Sigma_\mu (r_{ij}^\mu/a_\mu)^2]$,$^{1/2}$ a_μ is the anisotropic radius of the impurity state ($\mu = a, b, c$), and η is the ratio of the probabilities for transitions with and without spin flip. It can be shown that the spin-flip process requires the involvement of an additional long-wavelength magnon, so we would expect $\eta < 1$. Calculations for η for a specific model will be published separately.

The average number of transitions is

$$\Gamma_{ij} = f(\epsilon_i - \mu_i)[1 - f(\epsilon_j - \mu_j)]\langle W_{ij}^{\mathbf{n}_i,\mathbf{n}_j}\rangle_{\mathbf{n}}, \tag{4}$$

where $f(\epsilon)$ is the Fermi function, and the chemical potentials μ_i and μ_j are found from the principle of detailed balance for Γ_{ij}. Here we have $\mu_i = \mu_j = \mu$ if $|\langle \mathbf{h}_i \rangle| = |\langle \mathbf{h}_j \rangle|$. Figure 2 shows a scheme of the transitions between impurity sublevels. At $T < h_i$, transitions between low-lying sublevels are predominant; these transitions are accompanied by a spin flip if $\mathbf{h}_i = -\mathbf{h}_j$ but not if $\mathbf{h}_i = \mathbf{h}_j$.

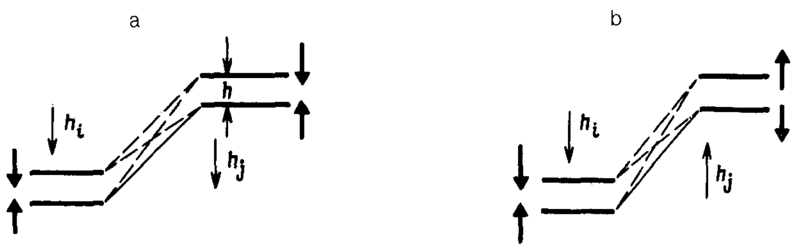

FIG. 2. Scheme of transitions between spin sublevels of impurities i and j. a—The case $\mathbf{h}_i = -\mathbf{h}_j$; b—the case $\mathbf{h}_i = -\mathbf{h}_j$. A solid line shows a transition which is predominant at $T < h$.

Each coupling ij corresponds to a resistance[10] $R_{ij} = T/e^2\Gamma_{ij}$. The three-dimensional nature of the Mott conductivity shows that the hops usually occur between different layers. For layers whose indices are of the same parity we would have $\langle \mathbf{h}_i \rangle = \langle \mathbf{h}_j \rangle$, and the coupling resistance would be $R_{ij}^+ = B\exp(\xi_{ij})$. For layers of different parity we would have $\langle \mathbf{h}_i \rangle = -\langle \mathbf{h}_j \rangle$ and $R_{ij}^- = B\exp(\xi_{ik} + \delta)$, and δ (which does not depend on ij!) is calculated from (3) and (4). We thus have a percolation problem, in which the nodes are divided into two equal groups. The function describing the coupling between nodes within each group is ξ_{ij}, while the function describing the coupling between nodes from different groups is $\xi_{ij} + \delta$. The percolation threshold in this problem is $\xi_c(\delta) \approx \xi_c(0) + \delta/2$ for $\delta \ll \xi_c(0)$. The latter conduction allows $\delta \gtrsim 1$, since we have $\xi_c(0) \sim 10$ at $T = 100$ K. Our original problem [with $\delta \ll \xi_c(0)$] is thus equivalent to a network of random resistances with nodes of a single type, but with $R_{ij} = \sqrt{R_{ij}^+ R_{ij}^-}$ (the geometric-mean rule).

In a field $\mathbf{H} \| \mathbf{b}$ with $H < H_c$, the directions of $\langle \mathbf{h}_i \rangle$ in the layers alternate ($|\langle \mathbf{h}_i \rangle|$ does not depend on H), and the resistance is $R = R_0 = B\exp[\xi_c(0) + \delta/2]$; at $H > H_c$ the direction of $\langle \mathbf{h}_i \rangle$ is the same in all layers, so we have $R = R_H = B\exp[\xi_c(0)]$. For the relative jump in the resistance we have $\Delta R/R_H = \exp(\delta/2) - 1$.

Evaluating δ in the mean-field approximation, i.e., using the substitution $\mathbf{h} \to \langle \mathbf{h}_i \rangle = [\mathbf{M}_\alpha \times \boldsymbol{\omega}]$ in (3), we find

$$\Delta R/R_H = \{[\cosh(\frac{\omega M}{2T}) + \eta]/[1 + \eta\cosh(\frac{\omega M}{2T})]\}^{1/2} - 1, \tag{5}$$

where $M = |\mathbf{M}_\alpha|$. In the case $M \ll 1$ (i.e., near the Néel point), we have

$$\Delta R/R_H \approx \frac{1-\eta}{1+\eta}\frac{\omega^2}{16T^2}M^2. \tag{6}$$

Expression (5) agrees qualitatively with the experimental behavior at $\omega \sim 100$ K. The behavior of the curves in Fig. 1(b) confirms the proportionality of $\Delta R/R_H$ and M^2. It should be kept in mind that a quantitative comparison with experiment will require eliminating the band component of the conductivity which arises at $T > 50$ K (Refs. 2 and 4).

Incorporating the fluctuations of $\mathbf{n}$ near the Néel temperature leads to only a renormalization of the coefficient in (6). At low temperatures the fluctuations are more important; they suppress the magnetoresistance. Neutron experiments[11] and also theoretical considerations[12] indicate an intensification of the fluctuations in $\mathbf{n}$ at low temperatures. It is possible that the low-temperature dip in $\Delta R/R_N$ [Fig. 1(b)] is a consequence of specifically this factor.

We will discuss the effect of fluctuations in $\mathbf{n}$ and the more complex situation which arises in the case[4] $\mathbf{H} \perp \mathbf{b}$ in a separate publication.

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