

ac hopping magnetotransport across the spin-flop transition in lightly doped La_2CuO_4 Oleg P. Sushkov¹ and Valeri N. Kotov²¹*School of Physics, University of New South Wales, Sydney 2052, Australia*²*Department of Physics, Boston University, 590 Commonwealth Avenue, Boston, Massachusetts 02215, USA*

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The weak ferromagnetism present in insulating La_2CuO_4 at low doping leads to a spin flop transition, and to transverse (interplane) hopping of holes in a strong external magnetic field. This results in a dimensional crossover $2\text{D} \rightarrow 3\text{D}$ for the in-plane transport, which in turn leads to an increase of the hole's localization length and increased conduction. We demonstrate theoretically that as a consequence of this mechanism, a frequency-dependent jump of the in-plane ac hopping conductivity occurs at the spin flop transition. We predict the value and the frequency dependence of the jump. Experimental studies of this effect would provide important confirmation of the emerging understanding of lightly doped insulating $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$.

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I. INTRODUCTION AND PROBLEM OVERVIEW

The interplay between the charge and spin degrees of freedom is crucial for our understanding of the high-temperature superconductivity. A lot can be learned about this interplay by studying the insulating phases of the copper oxides. Lightly doped La_2CuO_4 is especially interesting because of the weak ferromagnetism which exists in the Néel phase. The weak ferromagnetism provides a “handle” that allows one to influence the spins by application of a moderate magnetic field and hence to study how the charge dynamics is influenced by the spins. This issue has been recently addressed both experimentally^{1–6} and theoretically.^{7–9}

The present work is relevant to $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$, and $\text{La}_2\text{CuO}_{4+y}$ at very low doping. Generally these compounds have very different properties. $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ has an insulator-superconductor transition at $x \approx 0.055$,¹⁰ while $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$ remains an insulator at all dopings.¹¹ Elastic and inelastic neutron scattering in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ reveals incommensurate magnetic peaks^{12–15} while neutron scattering in $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$ demonstrates only an inelastic peak that is always commensurate.^{16–18} There are, however, similarities between these compounds: the long-range Néel order is destroyed at rather close values of doping, $x=0.02$ in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, and $x=0.03$ in $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$.^{10,19} The most important similarity is that at low doping all compounds exhibit variable-range hopping (VRH) conductivity^{2,10} that unambiguously indicates localization of holes. Localized holes in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ can lead to a diagonal spiral distortion of the spin background, and the corresponding theory^{20–25} explains quantitatively the whole variety of magnetic data. The transport theory based on localization^{26,27} explains quantitatively the in-plane anisotropy of the dc and ac conductivities in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ as well as the negative in-plane dc magnetoresistance in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{La}_2\text{CuO}_{4+y}$.

In the low-doping region there exist anisotropies in the spin-spin interactions, such as Dzyaloshinsky-Moriya (DM) and XY terms, in addition to the Heisenberg exchange. In the Néel phase the anisotropies confine the spins to the (*ab*) plane and fix the direction of the Néel vector to the \hat{b} -orthorhombic axis. Moreover, the DM interaction induces

a small out-of-plane spin component that is ferromagnetic in the plane (weak ferromagnetism) but staggered in the out-of-plane \hat{c} -direction. This component can be easily influenced by an external magnetic field applied in different directions.

Magnetic field directed along the \hat{c} axis can cause an alignment of the out-of-plane moments via a spin flop transition at a critical field H_f , determined by the competition between the DM and interlayer Heisenberg exchange. Typically $H_f \approx 5–7$ T.^{3,5,6} Theoretically a more complicated behavior than just a spin flop is also possible.⁷ Which particular regime is realized depends on the values of the anisotropy parameters. The experimental data are not 100% conclusive, but still they mostly indicate a simple spin flop and this is the picture that we accept in the present work. Magnetic field directed along the orthorhombic \hat{b} axis causes a continuous rotation of the spins towards the \hat{c} axis.^{1,4,5,8,9,28} The spins align completely along \hat{c} at a field $H_{c2} \sim 20$ T. An intermediate in-plane spin flop is also possible at $H_{c1} < H_{c2}$.^{8,9} The intermediate in-plane spin flop is very sensitive to doping and the situation is different for Sr, Li, and zero doping.²⁸ However the intermediate in-plane spin flop practically does not influence the transport properties.

In the present work we study the in-plane ac magnetoresistance (MR) across the spin flop transition, and our results are applicable to $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$, and $\text{La}_2\text{CuO}_{4+y}$. Doping is assumed to be so small that the compounds are in the Néel phase (the weak ferromagnetism regime). For definitiveness and simplicity we concentrate on the case of a field along the \hat{c} direction where the spin flop transition is sharp, although an extension for a field in the \hat{b} direction is straightforward. It was pointed out first in the experimental papers^{1,2} that the large negative MR, for both the dc and ac case, is due to an increase of the hole's localization length. A specific mechanism, based on a three-dimensional (3D) VRH transport scenario, was first proposed and explored in Ref. 29. We have recently revisited the problem within a different, quasi-two-dimensional formulation,²⁷ which we have argued to be relevant to any reasonable temperature range where experiments are performed. Our theory provides a detailed knowledge of the evolution of the hole's localized wave function across the spin flop transition,²⁷ lacking in all previous studies. This evolution takes into ac-

count the opening up of an interplane hopping channel at the spin flop and was used to explain the negative dc MR, in excellent agreement with experiment.²⁷ Here we calculate the ac MR within this theoretical framework, and show that a large negative MR is also expected in this case. The MR value depends on the mechanism of ac hopping conduction, determined by the value of the ac frequency ω compared to the temperature T . The variation of the ac MR with ω is found to be slow, due to the logarithmic dependence of the VRH distance on ω . Moreover, at low frequency a saturation of the MR is predicted. These features distinguish the ac regime from the dc case which has been studied more extensively in experiment.

The rest of the paper is organized as follows. In Sec. II we discuss the high-frequency quantum case, and in Sec. III the phonon-dominated regime. Section IV contains our conclusions.

II. IN-PLANE ac CONDUCTIVITY JUMP AT THE SPIN-FLOP TRANSITION FOR FIELD $H \parallel \hat{c}$: ZERO TEMPERATURE PHONONLESS CASE

First we consider the quantum, phononless regime that corresponds to very low temperatures ($T \ll \omega$). In this case the ac absorption is due to resonant electromagnetic transitions between hybridized symmetric and antisymmetric states formed by a hole bound to two impurities separated by distance r . It was demonstrated in Ref. 27 that at $H < H_f$ the hole hopping matrix element between the CuO_2 planes is extremely small, and therefore we have a pure two-dimensional (2D) situation. According to Shklovskii and Efros,³⁰ the conductivity in the quantum regime (which also takes into account the Coulomb interaction e^2/r within the resonant pair), appropriately modified for our 2D case, is

$$\sigma(\omega) \propto \int_{r_\omega}^{\infty} \left(\omega + \frac{e^2}{r} \right) \frac{r^3 I(r) dr}{\sqrt{\omega^2 - 4I(r)}}, \quad (1)$$

where

$$I(r) = I_0^2 P(r). \quad (2)$$

Here $I(r)$ is the overlap integral of the two localized states, and $I_0 \sim \epsilon_0 \approx 10$ meV, where ϵ_0 is the hole binding energy.²⁷ The tunneling probability $P(r)$ is

$$P(r) = e^{-2\kappa r}, \quad (3)$$

where κ is the inverse localization length. From now on we omit the exact prefactors in formulas like Eq. (1) since we will be interested only in conductivity ratios. The lower limit of integration r_ω in Eq. (1) is determined by the zero of the denominator of the integrand. Performing the integration in Eq. (1) with logarithmic accuracy, one obtains the well known answer³⁰

$$\sigma(\omega) \propto \frac{\omega}{\kappa} \left(\omega + \frac{e^2}{r_\omega} \right) r_\omega^3, \quad (4)$$

where

$$r_\omega = \frac{1}{\kappa} \ln \left(\frac{2\epsilon_0}{\omega} \right). \quad (5)$$

Now we turn to the case $H > H_f$. As discussed in Ref. 27, the spin flop gives rise to hopping in the \hat{c} direction and this effectively changes the dimensionality of the bound state, increasing the localization length. The hopping in the \hat{c} direction is described by the parameter $Zt_\perp \sim 0.5$ meV, where t_\perp is the effective hopping matrix element, and $Z \approx 0.3$ is the hole quasiparticle residue determined by spin quantum fluctuations. We estimate the hopping relative to the binding energy

$$\frac{2Zt_\perp}{\epsilon_0} \sim 0.1 - 0.2. \quad (6)$$

Due to the dimensional crossover the wave function of the hole bound state is changed in a peculiar way. The general integral representation for the probability to find the hole at a distance r from the trapping center is derived in Ref. 27. The result is simplified greatly in two limiting regimes: (1) Very large distances, $\kappa r \gg \epsilon_0/(2Zt_\perp)$, and (2) intermediate large distances, $\epsilon_0/(2Zt_\perp) \gg \kappa r \gg 1$. The typical hopping distance is given by Eq. (5), and therefore the very large distance regime is realized for $\ln(2\epsilon_0/\omega) \gg \epsilon_0/(2Zt_\perp)$, and the intermediate large distance regime is valid for $\epsilon_0/(2Zt_\perp) \gg \ln(2\epsilon_0/\omega) \gg 1$.

In the very large distance regime, the main result of Ref. 27 is that the localization length increases to

$$\bar{\kappa}^{-1} = \kappa^{-1} [1 - (4Zt_\perp/\epsilon_0)]^{-1/2}, \quad H > H_f. \quad (7)$$

The tunneling probability is basically given by the same Eq. (3) with the replacement $\kappa \rightarrow \bar{\kappa}$. Therefore, Eq. (4) is also valid and hence the ratio of conductivities after and before the flop is

$$\frac{\sigma_{H>H_f}}{\sigma_{H<H_f}} = \left(1 - \frac{4Zt_\perp}{\epsilon_0} \right)^{-2} \left[\frac{1 + V_\omega \sqrt{1 - \frac{4Zt_\perp}{\epsilon_0}}}{1 + V_\omega} \right], \quad (8)$$

$$\kappa r_\omega \gg \epsilon_0/(2Zt_\perp).$$

We have introduced here the relative strength of the Coulomb interaction

$$V_\omega \equiv \frac{e^2/r_\omega}{\omega} = \frac{\epsilon_0}{\omega \ln(2\epsilon_0/\omega)}, \quad (9)$$

and taken into account that $\epsilon_0 = e^2 \kappa_0$. For typical frequencies the Coulomb interaction always dominates, $V_\omega \gg 1$. It is clear that the conductivity increases after the flop, $\sigma_{H>H_f}/\sigma_{H<H_f} > 1$, i.e., the magnetoresistance is negative.

In the intermediate large distance regime the propagation probability is²⁷

$$P(r) = e^{-2\kappa r} \left(1 + 4(\kappa r)^2 \frac{(Zt_\perp)^2}{\epsilon_0^2} \right). \quad (10)$$

One has to substitute this expression into Eq. (1) and perform the integration with logarithmic accuracy. Note that the lower limit of integration is changed, $r_\omega \rightarrow r'_\omega = r_\omega [1 + 2(Zt_\perp/\epsilon_0)^2 \ln(2\epsilon_0/\omega)]$. After simple calculations, we obtain

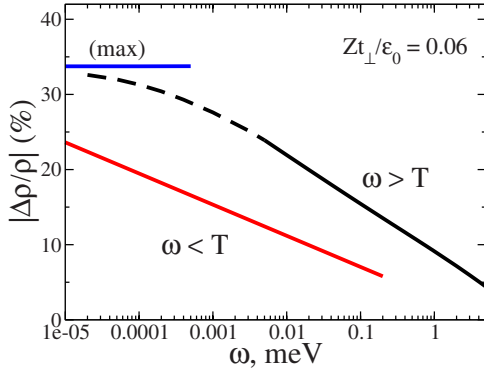


FIG. 1. (Color online) Relative resistivity jump at the spin flop transition vs frequency. In the quantum limit $\omega > T$, the black solid line corresponds to the Intermediate large distance regime described by Eq. (11), and is justified at $\omega \gtrsim 10^{-2}$ meV. The horizontal line, labeled “max,” corresponds to the very large distance regime (ultralow frequency) described by Eq. (8). The smooth crossover between these two limiting cases is shown schematically by the dashed line. At high temperatures $\omega < T$, the relaxation absorption formula Eq. (17) is plotted in red within its region of applicability; it is expected to saturate at $\omega \sim 10^{-8}$ meV (not shown).

$$\frac{\sigma_{H>H_f}}{\sigma_{H<H_f}} = 1 + \left(\frac{Zt_{\perp}}{\epsilon_0}\right)^2 \ln\left(\frac{2\epsilon_0}{\omega}\right) \frac{10 + 8V_{\omega}}{1 + V_{\omega}},$$

$$\kappa r_{\omega} \ll \epsilon_0 / (2Zt_{\perp}). \quad (11)$$

From now on we use the value $Zt_{\perp} / \epsilon_0 = 0.06$, which fits well the dc MR of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$,²⁷ and we take $\epsilon_0 = 10$ meV.

As the frequency decreases, the ratio $(\sigma_{H>H_f} / \sigma_{H<H_f})$ slowly increases, following Eq. (11). This behavior is illustrated in Fig. 1, where we have plotted the magnitude of the MR jump, i.e., the quantity $|\Delta\rho/\rho \equiv (\rho_{H>H_f} / \rho_{H<H_f}) - 1|$. The intermediate large distance formula (11) fails somewhere below $\omega \sim 10^{-2}$ meV. At lower frequencies one needs to perform the integration in Eq. (1) using the exact numerical expression for $P(r)$ derived in Ref. 27. This is expected to provide a smooth crossover to the ultralow frequency asymptotic value given by Eq. (8) and shown in Fig. 1 by the horizontal line.

III. IN-PLANE ac CONDUCTIVITY JUMP AT THE SPIN-FLOP TRANSITION FOR FIELD $H \parallel \hat{c}$: PHONON-DOMINATED REGIME, $\omega < T$

The regime $\omega > T$ considered in the previous section is not very realistic, since experimentally it is easier to achieve $\omega < T$. So, the regime $\omega < T$ that we consider in the present section is the most interesting one. To calculate the ac conductivity in this limit we use the Austin-Mott approach based on the relaxation mechanism.³¹ On the technical side we follow Ref. 32. In this regime there is a local quasistatic equilibrium established in an external ac electric field between two hole bound states separated by distance r . The relaxation comes from the phonon assisted tunneling, and the characteristic relaxation time τ is

$$\frac{1}{\tau(r)} = \nu P(r), \quad (12)$$

where $\nu \sim 1-10$ meV is a characteristic phonon frequency, and $P(r)$ is the tunneling probability considered previously. According to Eq. (4.7) of Ref. 32 the conductivity is, adapted to our 2D case,

$$\sigma(\omega) \propto \omega^2 \int_0^{\infty} \frac{r^3 \tau dr}{1 + \omega^2 \tau^2} G(r, T), \quad (13)$$

where the function $G(r, T)$ is expected to be independent of r at large distances, and $G(r, T) \propto T$. We have then

$$\sigma(\omega) \propto (\omega^2 / \nu) T \int_0^{\infty} \frac{r^3 P(r) dr}{P^2(r) + (\omega / \nu)^2}. \quad (14)$$

Define

$$\tilde{r}_{\omega} = \frac{1}{2\kappa} \ln(\nu / \omega). \quad (15)$$

Similar to the previous section we have the very large distance regime, $\kappa \tilde{r}_{\omega} \gg \epsilon_0 / (2Zt_{\perp})$, and the intermediate large distance regime, $\epsilon_0 / (2Zt_{\perp}) \gg \kappa \tilde{r}_{\omega} \gg 1$. The typical hopping distance in this case, given by Eq. (15), contains an extra factor 1/2, compared to Eq. (5). We take $\nu = 5$ meV. Therefore the very large distance regime sets in round 10^{-8} meV, i.e., in the KHz range. The intermediate large distance regime on the other hand is realized at 10^{-5} meV (i.e., MHz range) and higher.

Using Eq. (10) and performing the integration in Eq. (14), we obtain the conductivity after the spin flop in the intermediate large distances regime

$$\sigma_{H>H_f}(\omega) \propto \omega T \left(\frac{1}{2\kappa}\right) \tilde{r}_{\omega}^3 \left[1 + 10 \left(\frac{Zt_{\perp}}{\epsilon_0}\right)^2 (\kappa \tilde{r}_{\omega})\right]. \quad (16)$$

Therefore, the conductivity ratio is, for $\kappa \tilde{r}_{\omega} \ll \epsilon_0 / (2Zt_{\perp})$,

$$\frac{\sigma_{H>H_f}}{\sigma_{H<H_f}} = 1 + 5 \left(\frac{Zt_{\perp}}{\epsilon_0}\right)^2 \ln(\nu / \omega). \quad (17)$$

To make the consideration complete we also present the result for the very large distance regime, $\kappa \tilde{r}_{\omega} \gg \epsilon_0 / (2Zt_{\perp})$,

$$\frac{\sigma_{H>H_f}}{\sigma_{H<H_f}} = \left(1 - \frac{4Zt_{\perp}}{\epsilon_0}\right)^{-2}. \quad (18)$$

Our discussion so far did not account for the Coulomb interaction. For strong interaction $e^2 / \tilde{r}_{\omega} \gg T$, easily achievable for not too small ω and not too high T , the conductivity (16) is expected to cross over to a temperature-independent behavior, as discussed in Ref. 32. In this case the function G that appears in Eq. (13) is $G(\tilde{r}_{\omega}, T) \propto e^2 / \tilde{r}_{\omega}$, $e^2 / \tilde{r}_{\omega} \gg T$. Consequently we find that Eq. (16) becomes $\sigma_{H>H_f} \propto \omega \kappa^{-1} \tilde{r}_{\omega}^2 [1 + 8(Zt_{\perp} / \epsilon_0)^2 (\kappa \tilde{r}_{\omega})]$, within the limits of its applicability, while $\sigma_{H<H_f} \propto \omega \kappa^{-1} \tilde{r}_{\omega}^2$. From here one can deduce that the coefficient 5 is replaced by 4 in Eq. (17).

Within the accuracy of our calculations (in particular the value of ν) such a small difference can be ignored. We also mention that the exponent in Eq. (18) changes from -2 to $-3/2$ in the strong interaction case, which can be easily obtained by combining the dependence $\sigma(\omega) \propto \omega \kappa^{-1} \tilde{r}_\omega^2$ and Eq. (7), suitable for the regime of very large distances.

IV. DISCUSSION AND CONCLUSIONS

We are only aware of old work² that has studied the problem experimentally in $\text{La}_2\text{CuO}_{4+y}$ (and not theoretically explained until now) in the high-temperature regime, $\omega \ll T$; our results in the relaxation regime are completely consistent with those measurements. The weak ferromagnetism of La_2CuO_4 provides a unique opportunity to study the interplay between the charge and spin dynamics, and the jump of the in-plane ac conductivity at the spin flop transition is the most direct manifestation of this interplay. Therefore further experimental studies of the jump could provide important confirmation of our emerging understanding of lightly doped insulating $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$.

In conclusion, we have developed a description of the ac magnetotransport across a spin flop transition in the strongly

localized regime (low T and ω compared to the hole's binding energy ~ 10 meV). Figure 1, which roughly spans the MHz to THz frequency range, summarizes the typical main features of our results, namely (1) slow, logarithmic variation over a wide frequency range [Eqs. (11) and (17)], (2) existence of an upper limit for the MR at low ω [Eq. (8)], and (3) different magnitudes of the MR and saturation frequencies, depending on the nature of the ac VRH mechanism (resonant absorption or relaxational). We hope that the rich ac behavior found in this work also stimulates additional experiments, as both our theoretical understanding and sample quality have improved dramatically during the last few years. Magnetotransport in the strongly localized regime is a case where the presence of disorder actually simplifies considerably the problem of spin-charge interplay, and consequently our theory applies mostly to the La family of cuprates.

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