Origin of Magnetoresistance Anomalies in Antiferromagnetic $YBa_2Cu_3O_{6+x}$

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Abstract

Specific $d$-wave angular dependence of the in-plane magnetoresistance in antiferromagnetic tetragonal $YBa_2Cu_3O_{6+x}$ ($x \sim 0.3$) on the orientation of the external magnetic field within the $(a, b)$ plane is assigned to the effective hole transport through the low-lying excited purely oxygen doublet $O2pe_u$ state in CuO$_4$ plaquette, rather than the ground $b_{1g}(d_{x^2-y^2})$ state. The external magnetic field is believed to determine the orientation of the strong exchange field for the spin-triplet $b_{1g}e_u: 3E_u$ state of the hole CuO$_4$ center and owing to the spin-orbital coupling result in a specific orbital polarization of the $E_u$ doublet. Namely this gives rise to the spatial $d$-wave like anisotropy of the hole transport. The experimental data allow to estimate the parameter of the effective spin Hamiltonian.

Key words: Cuprates, Magnetoresistance, Spin-orbital coupling

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1 Introduction

The transport properties of a single hole in a strongly correlated antiferromagnetically ordered quasi-2D cuprate have been the topic of much debate, both theoretically and experimentally. Unusual magnetoresistance anomalies in the heavily underdoped antiferromagnetic $YBa_2Cu_3O_{6+x}$ ($x = 0.30; 0.32$) crystals were reported recently by Y. Ando et al. [1]. The in-plane resistivity $\rho_{ab}$ exhibits unconventional metal-dielectric duality with the high-temperature ($T > 50K$) metal-like behavior in contrast with the low-$T$ insulating one which is not compatible both with that for a simple band insulator and for an Anderson insulator.

The crystals demonstrate an unusual behavior of the in-plane magnetoresistance, $\Delta \rho_{ab}/\rho_{ab}$ when the magnetic field $\vec{H}$ is applied along the CuO$_2$ planes.
These are a striking d-wave shaped ($\propto \cos 2\phi$) angular dependence, anomalous low-field behavior with saturation above a well-defined threshold field, and hysteretic effects at low temperatures. As the temperature decreases below $20-25$ K, the magnetic field dependence of $\rho_{ab}$ becomes essentially irreversible, and the system acquires a memory. The application of the field results in a persistent change in the resistivity. The authors consider qualitatively these features to be a manifestation of the 'charge stripe' ferromagnetic structure in this system, which could be easily rotated by a rather small external magnetic field. As the temperature is lowered, it is expected that the stripe dynamics slows down and the appropriate texture in the CuO$_2$ layers is frozen, forming somewhat like a cluster spin glass.

The antiferromagnetic domain structure in insulating YBa$_2$Cu$_3$O$_{6+\delta}$ ($x < 0.15$) with 1% Gd$^{3+}$ as a ESR probe has been recently studied in detail by A. Janossy et al. [2,3]. According to the ESR data the easy axes of the antiferromagnetic order are along [100], and [110] are hard axes in the ($a, b$) plane. In zero magnetic field the single crystal consists of the equal amounts of domains oriented along two possible easy axes. External magnetic field in the ($a, b$) plane induces a spin-flop reorientation transition, and at $h > h_{sf} = 5$ T ($T \sim 20$ K) practically all domains arrange perpendicular to the field. The authors [2,3] consider the two models of domain texture: 1) magnetic domains separated by charge domain walls within the ($a, b$) plane; and 2) perfect antiferromagnetic plane domains separated by defective ($a, b$) planes, and they conclude their ESR data qualitatively evidence in favor of the second model.

In our opinion, the experimental data [1] are unlikely to be consistent with a conventional model of the well isolated spin and orbital Zhang-Rice (ZR) singlet $^1A_{1g}$ [4] believed to be a ground state of the hole-doped CuO$_4$ center in the CuO$_2$ layers. Here, it should be noted that when speaking of a Zhang-Rice singlet as being 'well isolated', one implies that the $^1A_{1g}$ ground state for the CuO$_4$ plaquette with the two holes of the $b_{1g}(d_{x^2-y^2})$ symmetry is well separated from any other excited two-hole states. Indeed, a simple s-like spin and orbital symmetry implies the tetragonally isotropic 's-wave' transport properties of the well isolated ZR singlet in the CuO$_2$ layers. The unconventional d-wave magnetoresistance anisotropy displayed by insulating cuprates, as well as many other experimental data and theoretical model considerations evidence in favor of the more complicated structure of the valence multiplet for the hole-doped CuO$_4$ center rather than simple ZR singlet albeit namely the latter is a guideline in the overwhelming majority of current model approaches.

The nature of the valent hole states in doped cuprates is considered as being of great importance for the high-$T_c$ problem. Having solved the problem we could justify the choice of the relevant effective Hamiltonian together with the opportunities of a mapping to the single band $t - J$ or Hubbard model.
Below we show that the model of the valence $^1A_{1g} - ^3E_u$ multiplet, developed in Refs.[5–11], provides a consistent explanation of the d-wave magnetoresistance anisotropy in the CuO$_2$ layers of the doped cuprates believed to be the most important result of Ref.[1].

2 The model of the valence $^1A_{1g} - ^1E_u$ multiplet

The model implies a quasi-degeneracy in the ground state of the two-hole CuO$_4$$^-$ center with the two close in energy $^1A_{1g}$ and $^1E_u$ terms of $b_{1g}$ and $b_{1g}e_u$ configurations, respectively. In other words, one implies two near equivalent locations for the additional hole, either to the Cu3dO2p hybrid $b_{1g}(d_{x^2-y^2})$ state to form ZR singlet $^1A_{1g}$, or to purely oxygen nonbonding doublet $e_u x, y$ state with peculiar Cu$^{2+}$-Cu$^{3+}$ valence resonance. The electron density distribution for two valent hole states $b_{1g}$ and $e_u$, respectively, together with the quantitative picture of the energy spectrum for valence multiplet is shown in Fig.1. It should be noted that the symmetry of the O2pe$_ux, y$ states coincides with that of for Cu4px, y states.

In a sense, the valence ($b_{1g}^2$)$^1A_{1g} - (b_{1g}e_u)^1E_u$ multiplet for the hole CuO$_4$$^-$ center implies an unconventional state with Cu valence resonating between Cu$^{3+}$ and Cu$^{2+}$, or 'ionic-covalent' bonding [12]. In other words, the CuO$_4$ center with the valence ($b_{1g}^2$)$^1A_{1g} - (b_{1g}e_u)^1E_u$ multiplet represents a specific version of the 'correlation' polaron, introduced by Goodenough and Zhou [12].
The model is supported both by local-density-functional calculations [13], \textit{ab initio} unrestricted Hartree-Fock self-consistent field MO method (UHF-SCF) for copper-oxygen clusters [14,15], and a large variety of experimental data. To the best of our knowledge the one of the first quantitative conclusions on a competitive role of the hybrid copper-oxygen $b_{1g}(d_{x^2-y^2})$ orbital and purely oxygen O2p$_\pi$ orbitals in the formation of valent states near the Fermi level in the CuO$_2$ planes has been made by A.K. McMahan \textit{et al.} [13] and J. Tanaka \textit{et al.} [14]. Namely these orbitals, as they state, define the low-energy physics of copper oxides.

One of the most exciting experimental evidences in favor of the model with the valence $^1A_{1g} - ^1E_u$ multiplet is associated with the observation of the mid-infrared (MIR) absorption bands which polarization features are compatible with those for $^1A_{1g} - ^1E_u$ intra-multiplet dipole transitions [8]. The corresponding transition energies ($\sim \Delta_{AE}$) observed for various cuprates are of the order of a few tenths of eV, that yields a typical energy scale for the valence multiplet.

The $e_u$ hole can be coupled with the $b_{1g}$ hole both antiferro- and ferromagnetically. This simple consideration indicates clearly a necessity to incorporate in the valence multiplet both the spin singlet $(b_{1g}e_u)^1E_u$ and the spin triplet $(b_{1g}e_u)^3E_u$, which energy could be even lower due to ferromagnetic $b_{1g} - e_u$ exchange. Indeed, the low-lying spin triplet state for the two-hole CuO$_4$$^{5−}$ center was detected by $^{63,65}$Cu NQR in La$_2$Cu$_{0.5}$Li$_{0.5}$O$_4$ with a singlet-triplet separation $\Delta_{ST} = 0.13$ eV [16]. The indirect manifestations of O2p$_\pi$, or $e_u$ valent states were detected in the Knight shift measurements by NMR for 123-YBaCuO system [17]. In connection with the valence $^1A_{1g} - ^13E_u$ multiplet model for copper oxides one should note and comment the results of paper by Tjeng et al. [18], where the authors state that they "are able to unravel the different spin states in the single-particle excitation spectrum of antiferromagnetic CuO and show that the top of the valence band is of pure singlet character, which provides strong support for the existence and stability of Zhang-Rice singlets in high-$T_c$ cuprates". However, in their photoemission studies they made use of the Cu2p$_{3/2}(L_3)$ resonance condition that allows to detect unambiguously only copper photo-hole states, hence they cannot see the purely oxygen photo-hole $e_u$ states.

It should be noted that the complicated $^1A_{1g} - ^13E_u$ structure of the valence multiplet for the two-hole CuO$_4$$^{5−}$ center could be revealed in the photoemission spectra, all the more that the odd $^1E_u$ terms play here a principal role, namely these yield a nonzero contribution to the ARPES for $\vec{k} = 0$, or in other words at $\Gamma$ point. In this connection one should note the experimental measurements of the photoemission spectra in Sr$_2$CuO$_2$Cl$_2$ [19,20] and Ca$_2$CuO$_2$Cl$_2$ [21,22]. All these clearly detect a nonzero photocurrent intensity in the BZ center, thus supporting the $^1A_{1g} - ^13E_u$ structure of the ground state valence multiplet.
Overall, the model of extended $^{1}A_{1g}-^{1,3}E_u$ valence multiplet allows to explain consistently many puzzling properties both of insulating and superconducting cuprates: midinfrared absorption bands [8], (pseudo)Jahn-Teller (JT) effect and related phenomena [9–11], spin properties [23,24].

3 Anomalous magnetotransport in $b_{1g}e_u: ^{3}E_u$ hole state

The pseudo-JT polaronic nature of the spin-singlet $^{1}A_{1g}-^{1}E_u$ ground state [9–11] favors their localization. In addition, one should account for the antiferromagnetic background which leads to the crucial enhancement of the effective mass for the moving spin singlets. So, a spin-singlet small pseudo-JT polaron as a hole ground state is likely to be immobile. In such a situation the most effective channel for the hole transport could be related to the low-lying excited spin-triplet $b_{1g}e_u: ^{3}E_u$ term. This gives rise to a thermo-activated hole conductivity actually observed in most of slightly doped cuprates.

In order to obtain magnetoresistivity effect we’ll consider spin and spin-orbital interactions for the spin-triplet $b_{1g}e_u: ^{3}E_u$ state. The $e_u$ hole is strongly exchange-coupled both with the $b_{1g}$ hole on the same CuO$_4$ center and with the nearest neighboring CuO$_4$ centers. The spin state of the isolated CuO$_4$ center with the $b_{1g}e_u$ hole configuration is described by two spin operators

$$\vec{S} = \vec{s}_{b_{1g}} + \vec{s}_{e_u}, \quad \vec{V} = \vec{s}_{b_{1g}} - \vec{s}_{e_u} \quad (1)$$

($\vec{S}^2 + \vec{V}^2 = 3/2, (\vec{S} \cdot \vec{V}) = 0$), and corresponding order parameters [23,24]. For the description of the orbital $E_u$ doublet one could use the pseudospin $s = 1/2$ formalism with the Pauli matrices $\sigma_{x,y,z}$ having simple transformation properties within the $E_u x, y$ doublet:\[\footnote{We make use of standard notations for the $D_{4h}$ point group representations.}]

$$\sigma_x = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix} \propto b_{2g}, \quad \sigma_y = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \propto a_{2g}, \quad \sigma_z = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \propto b_{1g}.$$}

Then the effective spin-Hamiltonian for the spin-triplet $^{3}E_u$ state of the hole center can be represented as follows

$$\hat{H}_S = \lambda S_z \sigma_y + D S_x^2 + a(S_x^2 - S_y^2)\sigma_z + b\vec{S}_x\vec{S}_y \sigma_x - \mu_B \vec{g}_S \vec{S} - 2\mu_B \vec{H}_{ex}\vec{S}, \quad (2)$$

where $\vec{S}_x\vec{S}_y = 1/2(S_xS_y + S_yS_x)$, $\lambda$ is an effective spin-orbital coupling parameter for the $^{3}E_u$ term, $D, a, b$ the spin anisotropy constants, $\vec{g}_S$ the effective $g$-tensor, $\vec{H}_{ex}$ the internal effective spin exchange field, $\vec{h}$ the external magnetic field. Hereafter, we will assume for simplicity the ideal planar CuO$_2$
layers, isotropic spin $g$-factor and the planar direction of magnetic fields. In the frames of the strong molecular field approximation the spin operators can be replaced by appropriate averages

$$\langle S_x^2 - S_y^2 \rangle = \cos 2\Phi, \quad \langle S_x S_y \rangle = \sin 2\Phi,$$

and the spin-Hamiltonian (2) transforms into an effective Hamiltonian of the spin-induced low-symmetry crystalline field

$$\hat{H}_S = a \cos 2\Phi \sigma_z + b \sin 2\Phi \sigma_x,$$

(3)

where $\Phi$ is the azimuthal angle of the orientation for the total magnetic field $\vec{H} = \vec{H}_{ex} + \vec{h}$. Eigenvectors and eigenvalues for such a simple Hamiltonian are

$$\Psi_+ = \cos \alpha \ket{x} + \sin \alpha \ket{y}, \quad \Psi_- = \sin \alpha \ket{x} - \cos \alpha \ket{y}$$

$$\tan 2\alpha = \frac{b}{a} \tan 2\Phi, \quad E_+ = \pm \Delta(\Phi), \quad \Delta(\Phi) = \left[b^2 + (a^2 - b^2) \cos^2 2\Phi\right]^\frac{1}{2},$$

respectively, where $\ket{x, y} \equiv \ket{E_{ux, uy}}$. The quantum-mechanically and thermodynamic averages for $\sigma_{z,x}$, describing the orbital polarization (quadrupole ordering) effect, are

$$\langle \sigma_z \rangle = \rho_+ \langle \sigma_z \rangle_+ + \rho_- \langle \sigma_z \rangle_- = -\frac{a \cos 2\Phi}{\Delta(\Phi)} \tanh \beta \Delta(\Phi),$$

(5)

$$\langle \sigma_x \rangle = \pm \sin 2\alpha, \quad \langle \langle \sigma_x \rangle \rangle = -\frac{b \sin 2\Phi}{\Delta(\Phi)} \tanh \beta \Delta(\Phi),$$

respectively. Here, $\rho_\pm$ is the statistical weight of the $\Psi_\pm$ states, $\beta = 1/kT$. One should note a specific $d$-wave ($d_{x^2-y^2}$ and $d_{xy}$) angular $\Phi$-dependence for the thermodynamic averages $\langle \langle \sigma_z \rangle \rangle$ and $\langle \langle \sigma_x \rangle \rangle$, respectively (see Fig.2).

So, in frames of our approximation, the orbital state of the spin triplet $^3E_u$ is easily governed by the external magnetic field, thus providing an effective “magneto-orbital” transformation. Obviously, this spin-induced orbital polarization has considerable magnitude only at rather low temperatures. In the high-temperature limit ($|a|, |b| \ll kT$):

$$\langle \langle \sigma_z \rangle \rangle \approx -\frac{a}{kT} \cos 2\Phi.$$

(6)

It should be noted that Hamiltonian (3) possess an overall tetragonal symmetry, however, this could describe the effects of spontaneous breaking of this
assumed the only nonzero $(\text{Cu}3d\text{-O}2p)_{\sigma}$-bonding contribution to the $e_u$ hole transfer between the $E_u$ states on the nearest neighboring CuO$_4$ plaquettes, we can represent the matrix of the $e_u - e_u$ hole transfer integral $\hat{t}(e_u - e_u)$ on the $|x, y\rangle$ basis set as follows

$$\hat{t}_a(e_u - e_u) = t_\sigma \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}, \quad \hat{t}_b(e_u - e_u) = t_\sigma \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix}$$

for the transfer in the [100] and [010] directions, respectively. Here, $t_\sigma = t_a(e_u - e_u) = t_b(e_u - e_u)$. In frames of the tetragonal ansatz the $e_u$ hole energy spectrum in the tight binding approximation with the nn transfer consists of two bands $E_{x,y}$ formed by the transfer $e_u - e_u$ and $e_u - e_u$, respectively

$$E_{x, k} = 2t_\sigma \cos(k_x a), \quad E_{y, k} = 2t_\sigma \cos(k_y a). \quad (7)$$

These 1D bands are characterized by maximally anisotropic effective mass, and describe the 1D hole motion in $a$- and $b$- directions, respectively. Nevertheless, with account for rigorous $x - y$ ($a - b$) symmetry it is easy to see that the transport properties appear to be tetragonally isotropic. The spin-induced low-symmetry crystalline field (3) results in a mixing of the two bands. Indeed, instead of simple expressions for the bare hole transfer integrals we obtain matrices for the renormalized hole transfer integrals on the $|\pm\rangle$ basis set

$$\hat{t}_a(e_u - e_u) = t_\sigma \begin{pmatrix} \cos^2 \alpha & \frac{1}{2} \sin 2\alpha \\ \frac{1}{2} \sin 2\alpha & \sin^2 \alpha \end{pmatrix}.$$
\[ \hat{t}_b(e_u - e_u) = t_\sigma \left( \begin{array}{cc} \sin^2 \alpha & -\frac{1}{2} \sin 2\alpha \\ -\frac{1}{2} \sin 2\alpha & \cos^2 \alpha \end{array} \right), \] (8)

where we have simply made use of Exps.(5) for the \( \Psi_\pm \) functions.

Thus, after simple algebra we can obtain for the renormalized bands

\[ E^\pm_k = t_+ (\vec{k}) \mp \left[ t^2_+ (\vec{k}) + \Delta^2(\Phi) + 2 \Delta(\Phi) t_- (\vec{k}) \cos 2\alpha \right]^{\frac{1}{2}}, \]
\[ t_\pm (\vec{k}) = t_\sigma (\cos(k_x a) \pm \cos(k_y a)). \] (9)

The inverse in-plane effective-mass tensors for the two bands can be written as follows

\[ \left( \frac{1}{m^*} \right)_\pm = \frac{1}{m_0^*} \left[ 1 \pm \cos 2\alpha \left( \begin{array}{cc} 1 & 0 \\ 0 & -1 \end{array} \right) \right], \] (10)

where \( \frac{1}{m_0^*} = -\frac{4a^2}{\hbar^2} t_\sigma. \)

Thus, we obtain the spin-induced shift/splitting of the bare bands, which results in the breaking of the tetragonal \( x - y \) symmetry with appearance of the spin-dependent anisotropy in effective mass.

Taking into account the proportionality relation between the single-band conductivity and the inverse effective-mass tensor, one might obtain a surprisingly simple expression for the in-plane magnetoresistance

\[ \frac{\delta \rho_{a,b}}{\rho_{a,b}} = \langle \cos 2\alpha \rangle = \mp \langle \langle \sigma_z \rangle \rangle, \] (11)

where the upper (lower) sign corresponds to the [100] ([010]) directions, respectively. In other words, the in-plane magnetoresistance appears to be straightforwardly linked to the spin-induced orbital (quadrupole) polarization of the \( e_u \) states in the CuO\(_4\) centers. Rigorously speaking, the expression (11) is valid in the high-temperature region \( \Delta(\Phi) \ll kT \), when this easily (see (6)) reduces to

\[ \frac{\delta \rho_{a,b}}{\rho_{a,b}} \approx \mp \frac{a}{kT} \cos 2\phi, \] (12)

where, in contrast to (6), \( \phi \) is an in-plane azimuthal angle for the external field \( \vec{h} \), and we take into account the near orthogonality of \( \vec{H} \) and \( \vec{h} \). This extremely simple expression describes all the essential features of the magnetoresistance.
anisotropy in the CuO$_2$ layers and represents a main result of our model theory. Indeed, when comparing with experimental data [1] for YBa$_2$Cu$_3$O$_{6+x}$ ($x \sim 0.3$) one might see that the experimental angular dependence of the magnetoresistance for a rather strong external field exceeding the $h_{\text{flop}} \approx 5T$ obeys the $(\phi, T)$ dependence (12) rather well given the relatively small albeit reasonable value of the parameter $a$ of the spin anisotropy: $a \approx +0.1K$.

Observed $T$-dependent deviations from a simple $\propto \cos 2\phi$ law can be related to the JT effect within the $E_u$ doublet [9–11] which results in strong spin-vibronic effects providing the spin-induced distortions of the CuO$_4$ center. Moreover, the JT effect results in a two-well adiabatic potential with the stabilization of the hybrid structural-orbital modes of the $b_{1g}$, or $b_{2g}$ symmetry, providing the $\propto \cos 2\phi$, or $\sin 2\phi$ dependence of the magnetoresistance, respectively. These problems will be considered in details elsewhere.

We did not address important issues related to the origin of magnetic anisotropy and AF domain textures in doped insulating cuprates. In our opinion, the decisive role here is played by the doping-induced nucleation of the domains of a novel phase, which percolation at $x \geq 0.4$ results in superconductivity [25]. The effective nucleation of these domains in the CuO$_2$ layers is promoted by a strong in-plane charge inhomogeneity generated at least by three linearly arranged $nn$ chain oxygen atoms. Namely this leads to a quasi-1D stripe-like structure of the in-plane domains. The [100] and [010] oriented stripe-like domains induce the orthorhombic crystal structure distortions in the surrounding antiferromagnetic tetragonal matrix and generate an appropriate in-plane magnetic anisotropy. Thus, the CuO$_2$ layers of the underdoped tetragonal insulating YBa$_2$Cu$_3$O$_{6+x}$ could be considered as antiferromagnets with the stripe-induced fluctuating in-plane magnetic anisotropy with the competition of two easy axes [100] and [010], respectively.

For the well developed phase separation regime with the comparable volume fractions of both phases the in-plane resistivity could be qualitatively represented as a sum of three contributions: the semiconducting one, the stripe contribution, and the contact resistivity of the interface transport. One should note that the stripe domains in the hole doped cuprates could be considered as a source of the hole carriers (donors) for the semiconducting matrix.

The observed effects such as the low-temperature magnetic hysteresis and the memory could be associated with the quenching of the stripe texture below $T \approx 20K$ due to the sharp slowing down of the inter-phase boundary relaxation revealed by the $^{63,65}$Cu NQR measurements [26]. It is of interest to note, that the low-temperature ($20 \div 25K$) spin-glass like transition has been reported for heavily underdoped antiferromagnetic $Y_{1-y}Ca_yBa_2Cu_3O_6$ [27].
4 Conclusion

In conclusion, we propose a model where specific $d$-wave angular dependence of the in-plane magnetoresistance in antiferromagnetic tetragonal YBa$_2$Cu$_3$O$_{6+x}$ ($x \sim 0.3$) on the orientation of the external magnetic field in $(a, b)$ plane is assigned to the effective hole transport through the low-lying excited purely oxygen doublet O2pe$_u$ state, rather than the ground $b_{1g}(d_{x^2-y^2})$ state. Such an approach implies that the ground state of the doped CuO$_4$ plaquette in cuprates could have substantially more complicated singlet-triplet $^1A_{1g} - ^1,^3E_u$ structure instead of the well-isolated simple $^1A_{1g}$ ZR singlet. External magnetic field determines the orientation of the strong exchange field for the spin-triplet $b_{1g}e_u : ^3E_u$ state of the hole CuO$_4$ center and due to the spin-orbital coupling results in a specific orbital polarization of the $E_u$ doublet giving rise to the spatial anisotropy of the hole transport. The observed $d$-wave angular dependence of the magnetoresistance on the orientation of the external magnetic field is associated with the shift and splitting of two 1D bands, and the anisotropy of the effective mass which in turn results from the spin-induced effective low-symmetry crystalline field for the $e_u$ hole.

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