Two-carrier percolation model of normal-state transport properties of single-crystal $YBa_2Cu_3O_{7-x}$

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Linear temperature dependences of the planar resistivity and nominal planar Hall carrier concentration which extrapolate almost to zero at $T=0$ have been observed in this high-T_c superconductor. A simple model containing no exotic constructs explains the experimental data.

Perhaps the most characteristic feature of cuprate high-T_c superconductors is the linearity of their normal-state resistivity $\rho(T)=a(T_0'+T)$ from T_c to temperatures $T\gtrsim 300$ K where oxygen evolution begins. This behavior was first observed on powders of these materials.[1] Single-crystal measurements have shown that $\rho_{\alpha\beta}(T)$ is highly anisotropic,[2] corresponding to metallic resistivities $\rho_{\parallel}(T)$ in the a-b plane and semiconductive hopping conduction along the c axis normal to these planes. Single-crystal[3] and epitaxial film[4] studies of the planar Hall resistance $R_{\parallel}^{H}$ of $YBa_2Cu_3O_{7-x}$ have shown an even more remarkable linearity. The nominal carrier concentration $n_{\parallel}(T)=(R_{\parallel}^{H}e)^{-1}=bV_0(T_0''+T)$ appears to freeze out as T is extrapolated to zero. Moreover, with sample quality characterized by small transition temperature widths ΔT_c and large critical currents j_c, the parameters T_0' and T_0'' both become small ($\lesssim 10$ K) for the best samples.[4]

Together with the high value of T_c, the small value of T_0' has led to a number of exotic theories which are widely cited[3] in public. So far no exotic theory has been advanced to explain $n_{\parallel}(T)$, but one could be expected soon. Here I advance a very simple theory of both $\rho_{\parallel}(T)$ and $n_{\parallel}(T)$, which is intended to be as ordinary as possible. This means that I assume that the currents are carried by quasiparticles, that the scattering is due to phonons, and that the temperature dependence is derived from the Boltzmann equation in the usual relaxation-time $\tau(T)$ approximation.

My theory contains three additional elements, each of which is justified by other experiments. The first element is carrier localization at low T in the $Cu(2)O_2$ planes and the Cu(1)O chains, as evidenced by nuclear quadrupole relaxation experiments.[5] The second element is soft optic-mode phonons with frequencies in the range 25–50 cm^{-1}, as observed by Raman spectroscopy on powders[6] and by neutron scattering from a large single crystal[7] of the analogue compound La_2NiO_4. The third element is anisotropic percolative conductivity in a planar network consisting of twinned metallic domains and a minority fraction of defective (nonmetallic) inclusions.[8] With this geometry each separate metallic domain orientation is also a minority fraction which by itself may not percolate in two dimensions, but both orientations may percolate together. This observation is pivotal to what follows.

The general properties of the chain (1) and plane (2) carriers are the result of the local environments of the Cu and O atoms in these structural subunits $i=(1,2)$. With the y or b axis along the chains, the Fermi velocities v_{ai} can be described approximately as $v_{x2}=v_{y2}=v_{y1}=v_F$ and $v_{x1}=0$. There are two $Cu(2)O_2$ planes and one Cu(1)O chain per unit orthorhombic cell, and the $Cu(2)O_2$ planes are therefore buckled (fourfold but no mirror local symmetry with respect to z) while the local environment of the Cu(1)O atoms has at least the orthorhombic symmetry of the unit cell. The low-frequency phonons[9] with which these charged carriers interact most strongly are probably the infrared-active transverse acoustic modes of the Cu(1)O chain with **x** polarization (B_{3u}). Although those modes are localized on the Cu(1)O chains, because chain (1) carriers must be backscattered [that is, $(0,k_y)\rightarrow(0,-k_y)$], these modes do not contribute to $\tau_{y1}(T)$, which is probably dominated by scattering from oxygen vacancies $O^{\square}$ on the Cu(1)O chains which makes $\tau_{y1}(T)=\tau_{y1}^{0}$ a constant. The planar (2) carriers, however, can be scattered from $(0,k_y)$ to any point on or near the Fermi line (k_x,k_y) by partially screened dipolar interactions with these modes, which are expected to dominate $\tau_{x2}(T)$ and $\tau_{y2}(T)$ in the temperature range 100 K $\lesssim T\lesssim 300$ K.

The **x**-polarized soft optic modes (frequencies 25–50 cm^{-1}, equivalent temperatures $T_0\sim 35$–70 K) are much softer than those found in most intermetallic compounds, but such soft modes are often associated with ferroelectric transitions in perovskites.[6] (An even better analogy, in view of the very high oxygen diffusivity in these materials, may be with ternary solid electrolytes.[10] These materials have scaffolded structures which are closely parallel to the tubular structure of the electronic charge density in cuprates.[11]) These modes are stabilized by a nearly constant density of optic modes with frequencies up to ~ 600 cm^{-1} (800 K). Thus the linearities which are observed in the range 100 K $\lesssim T\lesssim 800$ K can easily be explained in terms of carrier interactions dominated by fully excited soft phonons with occupation numbers $n_0(T)$ proportional to T/T_0.

The planar resistivity $\rho_{\parallel}(T)$ is described by

$$\rho_{\parallel}(T)=a(T_0'+T) \tag{1}$$

and we include a residual resistivity ρ_d due to defect or impurity scattering as well as a thermal scattering term

proportional to

$$n_0(T) = [\exp(T_0/T) - 1]^{-1} \tag{2}$$

so that

$$\rho_{\parallel}(T) = \rho_d + a n_0(T) T_0 \tag{3}$$

which corresponds to (1) with

$$T_0' = -T_0/2 + \rho_d/a\,, \tag{4}$$

showing that $|T_0'|$ can be made even smaller than $T_0/2 \lesssim 20$ K by compensation between thermal and defect scattering.

For a single orthorhombic domain, the Hall resistivity R^H_{xyz} is given[12] by

$$R^H_{xyz} = E_y/j_x B_z = \sigma^d_{xyz}/\sigma^d_{xx}\sigma^d_{yy}\,. \tag{5}$$

Suppose we have only one carrier but $\tau_a(T)$ is anisotropic. Then σ_{aa} is proportional to τ_a and $\sigma_{a\beta\sigma}$ is proportional to $\tau_a\tau_\beta$, leaving R^H_{xyz} temperature-independent. This is no longer the case for the present anisotropic two-carrier model. Because $v_{x1} = 0$, $\sigma^d_{xyz} \propto \tau_{x2}\tau_{y2}$ (only the planes carry a Hall current). Similarly $\sigma^d_{xx} \propto \tau_{x2}$. However, *for a single domain*,

$$\sigma^d_{yy} \propto \tau^d_{y1} + b_2\tau^d_{y2}\,, \tag{6}$$

where b is a constant of order unity. The chain current j_{y1} is not scattered by the soft phonons, so that

$$\sigma^d_{yy} \propto b_1\Theta_D + b_2(T_0' + T) \tag{7}$$

with $b_1 \gg b_2$. (Note that the term b_1 in (7) is absent from $\sigma_{yy} \propto [\rho_{\parallel}(T)]^{-1}$ because the chains alone do not percolate.) What Eq. (7) says, in effect, is that for a single domain τ_{y2} and T do not appear in σ^d_{yy} because the plane is shorted by the chain. With this effect

$$R^{Hd}_{xyz} \propto \tau^d_{x2}\tau^d_{y2}/\tau^d_{x2}\tau^d_{y1}\,, \tag{8}$$

$$(eR^{Hd}_{xyz})^{-1} \propto n_{\parallel}(T) \propto (T_0'' + T)\,, \tag{9}$$

where T_0'' differs from T_0' because of the second term in (7). Because the planar Cu(2)O_2 currents percolate, the bulk Hall resistivity R^H_{xyz} will have the same temperature dependence as R^{Hd}_{xyz}.

The present percolative model agrees very well not only with the experimental[1-4] relations (1) and (9) but also with the qualitative trends observed[4] in epitaxial films of varying quality and composition. In particular, the "freeze-out" of the nominal carrier density $n_{\parallel}(T)$ occurs quite differently in the best film compared to the worst (deliberately enriched with Cu, two-phase) films. This can be explained in terms of interlayer scattering which for the best film sets in gradually near $T_b \sim 200$ K, as one would expect for a dynamical crossover. However, in the two-phase films the freeze-out is much more abrupt near $T_b \sim 120$ K, just as one would expect from a genuine spatial or geometrical percolation threshold. It would be interesting to extend the theory of the Hall effect in a percolative anisotropic medium[13] to the present macroscopically isotropic but microscopically twinned geometry. It would also be interesting to know (for instance, from an interatomic force field model[9]) whether the assumption made here of planar but not chain continuity across twin boundaries is reasonable: The former involves strains of order $|b-a|/(b+a) \sim 10^{-2}$, while the latter involves bond-angle twists of order $\pi/2$.

Note added in proof. A recent paper [K. Char *et al.*, Phys. Rev. B **38**, 834 (1988)] contains Hall resistivity data $R_H(T)$ on epitaxial $Y_2Ba_4Cu_8O_{20-x}$ films which are similar in structure to $YBa_2Cu_3O_{7-x}$ films but which contain an extra Cu-O chain in each unit cell. Their $R_H(T)$ closely resembles that described in Ref. 4 for the Cu-rich samples, indicating that the Cu-O chains can make an important contribution to the Hall anomalies, as assumed in the present model.

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