"Normal" Tunneling and "Normal" Transport: Diagnostics for the Resonating-Valence-Bond State

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The "normal" transport properties of the high-T_c superconductors, especially tunneling and anisotropic resistivity, are extraordinarily anomalous. We show that these properties can be explained, perhaps uniquely, by a two-dimensional resonating-valence-bond state.

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The newly discovered high-T_c oxides exhibit many very strange properties. While high T_c itself has generated tremendous excitement, the normal-state behavior is also fascinating. The "normal" transport properties, among other things, are anomalous. It seems to us that much of the theoretical work on high-T_c superconductivity has ignored many of these experimental facts and concentrated only on high T_c itself, while the overwhelming probability is that all of them have the same cause.

The resonating-valence-bond (RVB) theory first proposed by Anderson and subsequently developed by other people[1] appears to be potentially able to account for most of the experimental observations. The essence of this theory is that the strong electron-electron correlation results in the separation of charge degrees of freedom from spin degrees of freedom. The low-energy excitations consist of charged boson solitons (we shall call them holons from now on) and neutral fermion solitons (spinons) with a pseudo Fermi surface. This theory based on the two kinds of excitations has many unusual experimental consequences, many of which have been confirmed. In this work we point out that the "normal" transport experiments, especially tunneling and anisotropic resistivity, provide the strongest evidence that there exist holon and spinon excitations in the RVB state. The understanding of these excitations in the normal state is crucial towards a final theory of the high-T_c superconductivity.

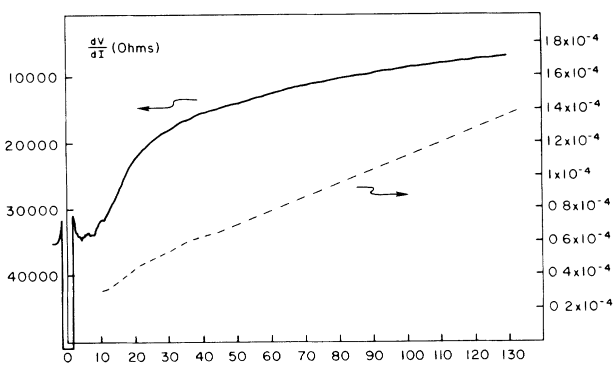

FIG. 1. Sample of Dynes's tunneling data for a single-crystal $YBa_2Cu_3O_7$/Pb junction. The upper curve is raw data of dV/dI; the straight line is the conductance; the structure at low voltage is due to Pb phonons and the Pb energy gap. The data are taken at $T=1.4$ K.

Traditionally, tunneling has been a very effective probe to understand superconductivity. However, in almost all attempts to study tunneling in the cupric superconductivity, the so-called "background" conductance at relatively high voltages is not constant but rises sharply with voltage; in fact, to a best approximation linearly, $\sigma_T \propto |V|$. Similar behavior is often found even at quite low voltages (see Fig. 1).[2-4] In neither case is there any indication of breakdown, Zeller-Giaever effects, or inelastic phonon-assisted tunneling, and the behavior is so regular and uniform as almost certainly to be intrinsic. At low voltages, it often seems that a surface layer of the material is not superconducting but normal.

Recently Tozer *et al.*[5] have measured the anisotropic conductivity of a single-crystal of $YBa_2Cu_3O_{7-x}$ with very striking results. The unexpected simplicity of the result can be brought out by replotting their data as ρT vs T^2. In both the *a-b* planes and the *c* direction the resistivity can be fitted very accurately by

$$\rho = A/T + BT \tag{1}$$

(see Fig. 2), but the coefficients are remarkably different: $A_c = 1.35$, $B_c = 3\times10^{-5}$, $A_{ab} = 0.7\times10^{-2}$, and $B_{ab} = 1.4\times10^{-6}$. (The resistivity is measured in ohm centimeters.) We note that a contact misalignment of $\frac{1}{4}°$ could account for A_{ab}, and hypothesize that

$$\rho_{ab} = 1.4\times10^{-6} T \ \Omega\ \text{cm}. \tag{2}$$

Equally, we suspect that patchiness and defects (and the difficulty of making a four-terminal measurement) account for the B_c term and—with less certainty—hypothesize that

$$\rho_c = 1.35/T \ \Omega\ \text{cm}. \tag{3}$$

We have plotted a wide variety of data on ceramic samples both of $(La\text{-}Sr)_2CuO_4$ and of "1-2-3" compounds and find that the expression (1) fits many of them very well (see note added).

We suppose that, in fact, the materials are all metals in the Cu-O planes and "semiconductors" for conduction between planes and across grain boundaries, in the sense that this conduction is invariably an inelastic process.

All of these observations are compatible with the idea that conduction in the "normal" state is mediated by holons,[6] which can be taken to be spinless bosons of charge e, and that the magnetic fluctuations in the "normal" metal are fermion solitons (spinons) with a pseudo Fermi surface, no charge, and spin $\frac{1}{2}$. Both of these excitations are solitons of the two-dimensional Cu-O layers, involving rearrangements of the entire layer wave functions, and as such cannot tunnel from one layer to another. The only three-dimensional objects are real electrons, which can tunnel between layers but must then break up into holon-spinon pairs of excitations. The charged carriers are scattered very effectively by the spinons, of which there are a number $\propto T/J$. Other scattering processes are minor if perhaps not negligible in very poor samples: Bosons of relatively long wavelength might have rather long mean free paths for impurity scattering.

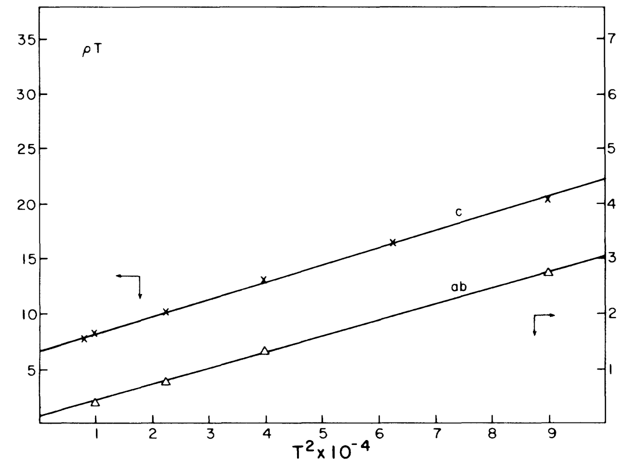

FIG. 2. Plot of the resistivity data of Tozer *et al.*: ρT vs T^2. Note that the scale for ρ_{ab} is different from that for ρ_c.

Let us derive the above results from the Zou-Anderson-Wheatley theory.[6-8] This theory leads to an effective "in-plane" Hamiltonian

$$\mathcal{H} = t_{\text{eff}} \sum_{\langle ij \rangle} e_i^\dagger e_j + t \sum_{\langle ij \rangle} (s_{i\sigma} s_{j\sigma}^\dagger - \langle s_{i\sigma} s_{j\sigma}^\dagger \rangle) e_i^\dagger e_j + \sum_k \Gamma_k s_{k\sigma}^\dagger s_{k\sigma}. \tag{4}$$

t_{eff} and $\Gamma_k \sim J$ are renormalized parameters embodying the average effects of the background of singlet pairs. Equation (4) is written in such a way as to bring out the fact that only excitations in the spinon Fermi gas scatter the holons, but that this scattering matrix element is as big as the fundamental kinetic energies and is only weakly k dependent. The spinors are twofold overcomplete and one may choose to ignore one or the other spin or simply to set $s_{i\sigma} = s_{i-\sigma}^\dagger$.[7]

First we calculate the in-plane conductivity. Roughly this is given by

$$\sigma = ne^2\tau/m_B, \tag{5}$$

and τ in turn can be estimated as the mean scattering-free time, with neglect of momentum dependence (since with the large Fermi surface umklapp processes are as common as direct ones). The elastic scattering rate τ^{-1} contains one less power of T for bosons scattered by fermions than for fermions scattering themselves, which is well known to be $\propto T^2$. This is because there is no exclusion-principle restriction on the final state of the boson.

A rough magnitude of the elastic scattering rate is estimated as follows:

$$\frac{1}{\tau} = \frac{2\pi}{\hbar} |t|^2 g_s g_b \int d\xi_q \, n_{\text{F}}(\xi_q)[1 - n_{\text{F}}(\xi_q)], \tag{6}$$

where g_s is the spinon density of states approximately given[1,7] by $g_s \approx (4\Delta J)^{-1}$, $g_b \approx (4t)^{-1}$ is the holon density of states, n_{F} is the usual Fermi function, and t is the scattering matrix element[7] between holons and spinons.

The spinon bandwidth estimated from low-temperature specific-heat measurement is of order $(1-2 \text{ eV})^{-1}$. We see that it is not surprising that the resistivity is still linear at $T \simeq 500$ K as shown by Gurvitch and Fiory.[9] Within the experimental temperature range $T \ll$ (the spinon bandwidth), the integrand in (6) is just a δ function. Thus we obtain $\tau^{-1} = (2\pi/\hbar) t^2 g_s g_b T$. This expression will reach the Mott resistivity at about $T \sim J$, which is the correct order of magnitude according to Ref. 4 and Ref. 6. Note that this scattering rate implies that the spinons are very severely scattered at all T, the mean free path $l_{\text{spinon}} \sim a/\sqrt{\delta}$, and the spinon gas is very disordered.

From the Drude formula (5), the resistivity is given by

$$\rho_{ab} = \frac{m_B^* \pi (U/t)}{32\hbar \Delta n e^2} T, \tag{7}$$

where the relation $J = 4t^2/U$ has been used, m_B^* is the holon effective mass, and n is the carrier density. A similar but slightly different result has been derived by Isawa, Maekawa, and Ebisawa.[8] Although our estimate is rather crude, the essential physics is included in (7); a more sophisticated calculation will only change the numerical factor slightly. If we take $n = 10^{21}/\text{cm}^3$ as obtained from Hall-effect measurement, $m_B^* \approx m_e$, and $\Delta = 4/\pi^2$,[1] we obtain $\rho_{ab} = 1.3 \times 10^{-6} T$ Ω cm, where the temperature T is measured in kelvins.

The process of tunneling between layers is not formally distinct. In this case a holon is scattered not within the layer but from layer to layer, again with emission or absorption of a pair of spinons, one in each layer. The only thing missing is a factor $|T_\perp/t|^2$ which gives the relative amplitude of scattering into the next layer versus scattering within the layer, $T_\perp$ being the hopping integral from layer to layer. The process in this case is responsible for the conductance rather than the resistance, and we expect the conductance to be less than the Mott conductance by a factor of order $\sigma_c/\sigma_{\rm Mott} \simeq (T_\perp/t)^2(T/J)$. This seems to give a value for $|T_\perp/t|^2$ of about 0.1 or so. The tunneling probability for holons from layer i to layer j, P_{ij} is again determined by the number of spinons available for scattering:

$$P_{ij} = (2\pi/\hbar)|T_\perp|^2 g_s g_b \int d\xi_q\, n_{\rm F}(\xi_q)[1-n_{\rm F}(\xi_q)] = (2\pi/\hbar)|T_\perp|^2 g_s g_b T. \tag{8}$$

From (8) we estimate the conductivity as

$$\sigma_c = (e^2/\hbar)2\pi|T_\perp|^2 g_s g_b^2 T(c/ab), \tag{9}$$

where ab is the area of the Cu-Cu square and c is the interlayer distance. If we set $|t/T_\perp|^2 = 10$, $t/U = 0.1$, and $J = 1000$ K, we estimate the resistivity perpendicular to the Cu-O plane as follows (measuring T in kelvins): $\rho_c = 1.7/T$ Ω cm, which is in close agreement with the experimental data. We emphasize that these numbers should not be taken literally, since we do not know the precise values of t, U, m^*, etc., and our estimate is rather crude. Nonetheless, the idea that the explanation of the anisotropic resistivity lies in this simple physical picture is hardly to be doubted. Another set of interesting transport experiments would be thermopower and thermal conductivity, for which we also expect anisotropy. The quantitative temperature dependence of the thermopower and thermal conductivity is under investigation, but seems reasonable within our picture.

Finally, we consider the normal tunneling. We have calculated this (see below) and find that the obvious phase-space argument gives the answer: The energy must be partitioned between a holon and a spinon, each with essentially constant density of states, and hence the current is proportional to V^2. Here we give a naive calculation of tunneling conductance between a normal metal and a RVB system connected through a weak junction. The electron states on the RVB side are labeled by $(\mathbf{p}\sigma)$ and on the normal size by $(\mathbf{k}\sigma)$. We assume that the junction is characterized by a tunneling matrix element T_{kp} which is taken to be independent of momentum. The tunneling Hamiltonian is thus given by $H_T = \sum_{kp\sigma}(T_{kp}c^\dagger_{p\sigma}a_{k\sigma} + \text{H.c.})$. The total Hamiltonian of the system is written as $H = H_R + H_N + H_T$, where the subscript R (N) refers to RVB (normal). All many-body effects are included in H_R. When a voltage is applied, the tunneling current is $I(t) = -e\langle d\hat{N}_R(t)/dt\rangle$. The time derivative of $\hat{N}_R$ may be obtained from its equation of motion. Following the standard procedure we can express the tunneling current in terms of the single-electron spectrum functions[10] $A_R(k,\epsilon)$ and $A_N(p,\xi_p)$:

$$I = 2e\sum_{kp}|T_{kp}|^2\int_{-\infty}^{\infty} d\epsilon\, A_N(k,\epsilon)A_R(p,\epsilon+eV)[n_{\rm F}(\epsilon) - n_{\rm F}(\epsilon+eV)]. \tag{10}$$

We shall neglect any many-body effect in the normal metal and use the free-electron spectrum function for A_N: $A_N(k,\epsilon) = 2\pi\delta(\epsilon-\xi_k)$. For the RVB state, A_R is obtained by calculation of the imaginary part of the single-particle propagator

$$G_1(p,\tau-\tau') = -\langle T_\tau c_{p\sigma}(\tau)c^\dagger_{p\sigma}(\tau')\rangle.$$

G_1 contains the complicated many-body effect which we were unable to calculate exactly. However, in the low-temperature limit ($T \ll$ spinon bandwidth), an electron tunneling into the RVB side from the normal side will have to decay into a holon and a spinon excitation. To the lowest-order approximation, we will replace them by corresponding free-particle propagators, respectively. Thus we may approximate G_1 by

$$G_1(p,i\omega_n)\frac{1}{N\beta}\sum_{q,iq_n} B(q-p,iq_n-i\omega_n)F(q,iq_n), \tag{11}$$

where B (F) is the free holon (spinon) propagator, N is the number of lattice site, and ω_n and q_n are fermionic Matsubara frequencies. The frequency summation is easily carried out and we obtain the spectrum function from (11):

$$A_R(p,\epsilon) = \frac{2\pi}{N}\sum_q [n_{\rm F}(\xi_q) + n_{\rm B}(\eta_{q-p})]\delta(\epsilon+\eta_{q-p}-\xi_q), \tag{12}$$

where $\eta_{q-p} = (\mathbf{q}-\mathbf{p})^2/2m_B^* - \mu \geq 0$ is the holon energy measured from the chemical potential, which is always nonnegative, and $n_{\rm B}$ is the usual Bose function.

Substituting A_N and A_R back into Eq. (10), one can carry out the integral at zero temperature. The final result for the linear voltage-dependent tunneling conductance is given by

$$\sigma_T = (e^2/\hbar)4\pi|T|^2 g_n g_s g_b|eV| + \sigma_0, \tag{13}$$

with the zero-bias conductance σ_0 given by

$$\sigma_0 = (4e^2/\hbar)\pi |T|^2 g_n g_s \delta, \tag{14}$$

where g_n is the density of states of the normal metal. Note that in the calculation of the integral the condition $\eta \geq 0$ imposes a limit on the integration range. The zero bias conductance (14) has been observed by Dynes,[4] and reflects the tunneling processes in which an electron of energy V tunnels into a spinon of the same energy plus a holon of approximately zero energy; namely, the electron spin is carried by the spinon and the electron charge is dumped into the "condensate." At finite temperature the integral is more complicated, but we do not expect a drastic change in $\sigma_T(V)$.

Thus the three linear phenomena—tunneling, normal-state resistivity parallel to the Cu-O plane, and normal-state conductivity perpendicular to the Cu-O plane —rather unequivocally point to the simple two-dimensional spinon-holon picture of the normal state above T_c. In this state there are no identifiable electron quasiparticles present within hundreds of degrees of Fermi energy and it would appear that "conventional" superconductivity is the last thing the system has on its mind, in agreement with simulations.[11]

We suggest that the observed superconductivity must, therefore, be a result of tunneling between the layers. In a succeeding paper[12] we will show how this tunneling can lead to electron pairing throughout the boson amplitude and to conventional superconductivity.

In the mean time, what of the electromagnetic properties of our two-dimensional layers? May[13] has proposed that all two-dimensional systems of the free bosons are superconductors, but he failed to take into account the Kosterlitz-Thouless process of vortex separation. Any reasonable estimate puts the Kosterlitz-Thouless transition T_c for this system higher than the observed T_c. We are unable as yet to resolve the question of whether the individual layers are independently topologically ordered or not.

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Note added.—Recently Hagen *et al.*[14] have tested Eq. (1) in a large number of single crystals of $YBa_2Cu_3O_7$ between room temperature and T_c and confirmed it very accurately.

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