Electronic disorder, gap states, orbital depairing, and dynamics of percolative superconductors near T_c

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A theory based on local energy gaps and local gap states explains entropy transport observed by Palstra *et al.* in $YBa_2Cu_3O_7$ single crystals in crossed electric and magnetic fields above T_c. The theory also suggests that many of the reversible dissipative effects observed below T_c and conventionally ascribed solely to free vortex motion (flux flow) may have a more complex microscopic origin. The observation of entropy transport above T_c provides fundamental evidence for the microscopic separability of localized and extended states.

High-temperature superconductors (HTSC) generally contain high concentrations of defects and are strongly disordered electronically.[1–4] As a result, conventional theories of superconductivity[5–7] and normal-state fluctuations[8] must be modified to include percolative effects.[4] Here I show how percolative effects alter qualitatively several properties of HTSC near T_c. I note that much of the broadening of the resistive transition and its lack of sensitivity to the Lorentz force has been discussed in the context of a Josephson interlayer coupling model.[9] While this model explains non-Lorentz effects, there remain broadening effects of the magnetization[10] which are not described by the Abrikosov theory of the mixed state.[6,11] More striking still are the many anomalies[3] observed above T_c, including Lorentz transport, which are the focus of this paper.

In crossed electric and magnetic fields, vortices transport entropy, giving rise to heat flow with a transport line energy U_ϕ which has been measured[12] on single-crystal $YBa_2Cu_3O_{6.9}$, with the results shown in Fig. 1. In Ginzburg-Landau (GL) theory, U_ϕ is proportional to $|\mathbf{M}|$, and below T_c, where both $\mathbf{M}$ and U_ϕ are linear in T, this feature of GL theory is observed. However, although the linearity is retained as $\mathbf{M}$ increases, the slope itself decreases.[10] This result cannot be explained by GL theory or by a homogeneous model which neglects the effects of static disorder. Moreover, above T_c there should be no vortices and U_ϕ should be small if there are only dynamical two-dimensional fluctuations.[9]

From Fig. 1 we see that the main effect of increasing $\mathbf{H}$ is to broaden the transition, which explains both the reduction in linear slope below T_c and the tail above T_c. Below T_c the broadening can be explained qualitatively by inhomogeneities in Ψ combined with the repulsive vortex-vortex interaction. In terms of the GL parameter $\kappa=\lambda/\xi$, where λ is the penetration depth, ξ the coherence length, and[10] $\kappa \sim 400$, $dM/dT \propto -\kappa^{-2}$ is dominated by short-wavelength[3] fluctuations in ξ. For small $\mathbf{H}$ and low-vortex densities, the nearly normal vortex cores will occupy the regions where $\Psi^2<\Psi^2_{\rm av}$ and $\xi^2>\xi^2_{\rm av}$, giving large values of $|dM/dT|$. With increasing $\mathbf{H}$ and increasing vortex density, the repulsive vortex-vortex interaction will force the vortices into regions with larger Ψ^2, smaller ξ^2, and this reduces the magnetization slopes.

To understand the origin of the tail in U_ϕ for $T>T_c$, we compare the functional trends in the curves in Fig. 1 with those obtained in numerical simulations of the effect of thermal disorder on exciton bands.[13] The average oscillator strength per state for these bands is shown in Fig. 2 for various temperatures (analogous to $\mathbf{H}$ in Fig. 1) as a function of frequency (analogous to T in Fig. 1). The similarity for $T>T_c$ in Fig. 1 to the region $\hbar\omega > -0.5$ eV in Fig. 2 is striking and it is not accidental. The exciton oscillator strength depends on the electron $\mathbf{r}_e$, and hole $\mathbf{r}_h$ coordinates in the exciton wave function $\phi(\mathbf{r}_e-\mathbf{r}_h)$ through $|\phi(0)|^2$, so the exciton oscillator strength is a core property, just as the vortex entropy transport is associated with the normal states of the vortex core.[12] At higher T, thermal fluctuations cause increasing absorption on the high-energy side ($\hbar\omega > -0.5$ eV) of the exciton band, corresponding to exciton-phonon absorption.

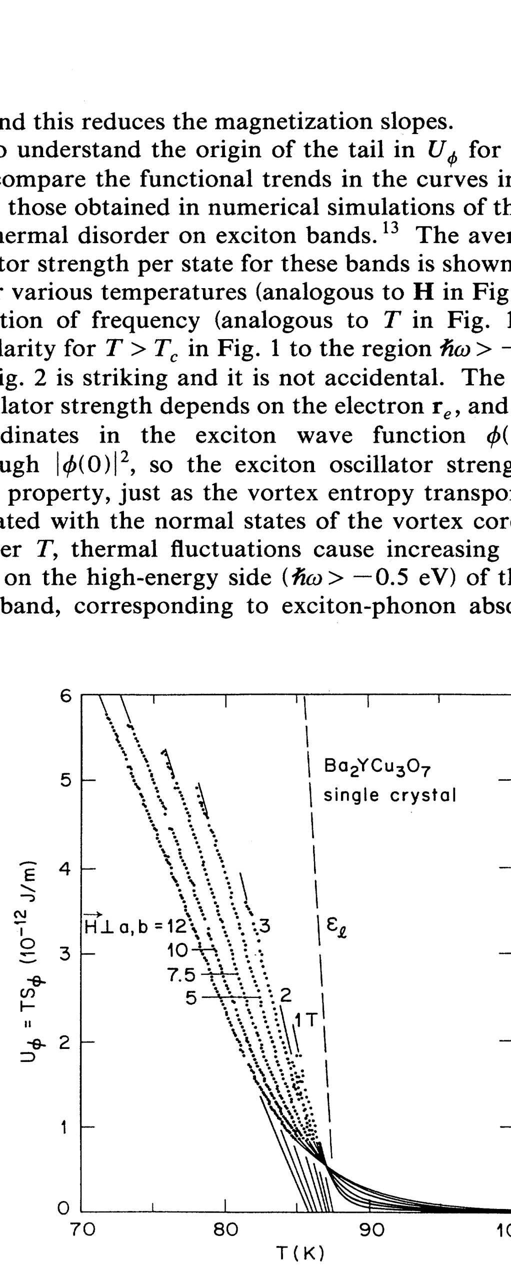

FIG. 1. Entropy transport in crossed electric and magnetic fields in single crystals of $YBa_2Cu_3O_7$, from Ref. 12.

At larger **H** due to Lorentz forces, vortices are driven into local (static) superconductive regions with $\Psi^2 > \Psi^2_{av}$ and T_c (local) $> T_c$ (bulk), giving rise to $U_\phi > 0$ for $T > T_c$ (bulk).

The picture of static local superconductivity just presented explains the results shown in Fig. 1, but it is apparently inconsistent with the fluctuation diamagnetism,[14] which is very small ($\sim 10^{-6}$) above T_c. In the local or dirty limit, where the electron mean free path $l \ll \xi$, these regions would be expected[11] to make a contribution to χ of order the fractional volume f times $[1-T/T_c$ (local)], and their contribution to U_ϕ could be no larger. Yet above T_c, $U_\phi \gtrsim 10^{-3} U_\phi$ (max). This inconsistency can be removed by assuming that we are in the nonlocal or clean limit where $l \gg \xi$. Then if the diameter of the region of interest is $d \approx \xi$, before the carriers moving under the influence of the Lorentz force can close their orbits to form a vortex, they leave the superconductive region and lose their Meissner susceptibility. This process can be called strong nonlocal orbital depairing.

The usual theory of spin and orbital depairing assumes that we are in the dirty limit where interactions are local and series expansions in Ψ [or $\Delta(\mathbf{r},\mathbf{r})$] and $\nabla\Psi$ are valid.[11] In the present case, Ψ is smoothly varying, but $\Delta = \Delta(\mathbf{r},\mathbf{r}')$ need not be local. Because it cannot be treated by perturbation theory, this case has not been solved,

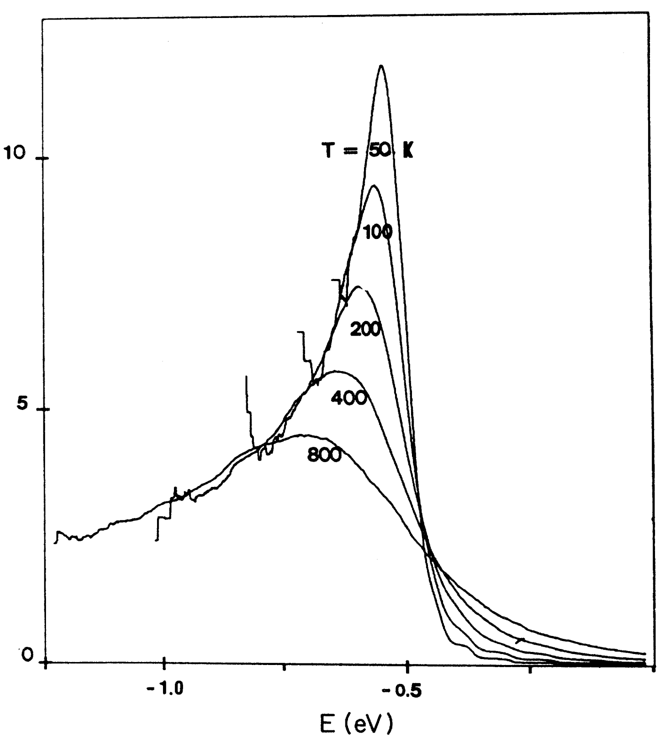

FIG. 2. Exciton oscillation strength (in arbitrary units) as a function of temperature in a one-dimensional model, from Ref. 13. Note the functional similarity of these curves near $E = -0.5$ eV to those shown in Fig. 1 near $T = 88$ K. The results for $d=2$ and 3 in Ref. 13 are similar, but the broadening with T is smaller and less well-resolved by the numerical simulation.

but for the diamagnetic susceptibility χ we can guess the answer. With $\omega_c = eH/m^*c$ and $v_F\tau = d$, Lorentz orbital closure for one electron is determined by $\omega_c\tau = eHd/p_Fc$. This parameter is the one that characteristically appears in perturbative solutions of one-electron transport equations in electromagnetic fields. However, the Meissner current is collective, and in the nonlocal case cannot be calculated perturbatively by a series expansion in powers of the coupling parameter g. This is also true for the microscopic superconductive condensation energy, which is proportional to $\exp(-g^{-1})$, and a similar relation may hold for fluctuating Meissner currents. Then for the nonlocal susceptibility one could have $\chi \approx \exp[-(\omega_c\tau)^{-1}]$. With $d \lesssim 10^{-6}$ cm and $v_F \approx 10^8$ cm/sec, $\tau \approx 10^{-14}$ sec and $\omega_c\tau \lesssim 10^{-3}$, giving a negligible diamagnetic susceptibility above T_c.

This model, however, above T_c eliminates the vortices which were supposed to contain entropy-transporting normal states in their cores. It still leaves residual local-energy gaps, and we know from Raman scattering experiments[15] that there are states in the bulk energy gap below T_c. The density of these states is linear in $\Delta E = E - E_{F'}$, so it is unlikely that they are produced by orbital pair-breaking, which would give a different energy dependence.[11] (The linear dependence is explained in quantum-percolation theory (QPT) as the result of localization.[16]) Whatever their origin, these states in the residual local-energy gaps can be occupied by normal electrons and are available for entropy transport, so that they can explain $U_\phi > 0$ for $T > T_c$ in Fig. 1 without producing a large χ. Specifically, electrons in the localized states cannot carry even a fluctuating Meissner current, because their extent is also $\lesssim \xi$, but they can transport entropy. Above T_c this transport will require energy transfer from electrons in the superconductive regions to phonons in the normal matrix in which the isolated superconductive regions are embedded. The efficiency of this conversion is unknown but it is presumably small, which is why U_ϕ is so small above T_c, even though the filling factor f associated with the percolating superconductive regions must smoothly increase with decreasing T to $f \sim \frac{1}{2}$ at $T = T_c$.

To summarize the discussion so far: in small regions above T_c we may have local-energy gaps accompanied by states in the gap. These states can transport entropy but the regions may be too small to support vortices and a comparably large Meissner current. On the other hand, the comparison between Fig. 2 and Fig. 1 is so close that it appears that states in the gap not connected with vortices could also be responsible for entropy transport and dissipation *below* but near T_c. In other words, even below T_c, where there are vortices, the vortices may be largely pinned by inhomogeneities in Ψ^2, and most of the measured value of U_ϕ may still be associated with Lorentz forces acting on electrons in the localized states in the gap associated with static disorder. Unlike normal states in vortex cores, these localized states are present[15] even when $\mathbf{H} = 0$. These remarks apply equally well to a wide variety of dissipative effects (electrical resistivity, ac magnetic susceptibility, and so on) which have been phenomenologically described in terms of flux (vortex) flow.[17] Such a description is pictorial and convenient below T_c, but as we have seen, it fails to describe U_ϕ above T_c. Because of its convenience it will probably continue to be the most popular description,[18] but it is well to remember that for distances of order ξ or less, a description in terms of localized intrinsic gap states may be preferable.

Below T_c a crossover occurs at $T=T_x$ between reversible and irreversible behavior, usually described as flux flow or creep, respectively. The behavior in the latter regime is described in terms of a thermal activation energy parameter $U_0>0$, which is found to decrease with a large negative curvature as **H** increases.[19] At $T=T_x$ the activation energy U_0 appears[19] to be zero, signaling the onset of reversibility. There are two ways to describe this onset of reversibility in the presence of inhomogeneities. First, spatially with increasing **H** the vortex cores must occupy regions where Ψ^2 is larger and approaches $\langle \Psi^2 \rangle$. In the absence of long-range vortex interactions,[11] but excluding core overlap, we eventually reach the saddle-point contours near half-filling when $U_0=0$. When long-range interactions are included in a self-consistent field, U_0 will collapse at smaller fields $H<H_{c2}$. However, the spatial renormalization procedure here in the presence of inhomogeneities is difficult to visualize. This is true of *any* model based on vortex coordinates alone, such as vortex lattice or glass melting.

The second way is simpler and more easily understood. Competition between disorder-induced gap states and vortex core states can explain the origin of the crossover temperature T_x where $U_0=0$, and its field dependence $T_x(H)$. The disorder-induced or intrinsic gap states (which are localized on a length scale[3] $d \leq 100$ Å) are little affected by fields $H<10$ T where the mean vortex spacing is larger than d. At low fields near T_c most of the dissipation occurs in the intrinsic gap states which lie outside the vortex cores. In this regime the vortices are a weakly pinned dilute solute in the electronic bath provided by the gap states, and the system is reversible for laboratory times. With increasing $T_c(H)-T$, the number of core states per vortex increases, and with increasing **H** the vortex density increases. Increasing both factors increases the fraction of the dissipation which occurs in the core states rather than the disorder-induced gap states, so that the latter no longer act as an effective solvent. Below the crossover temperature $T_x(H)$, vortex interactions dominate and irreversibility sets in. The energy spectrum of the vortex core in the nonlocal (clean) limit at high T is not known, but it seems likely that the main reason $T_x<T_c(H)$ is that a threshold $T_c(H)-T>0$ is required to bind core states well below the gap.

The foregoing discussion shows that in inhomogeneous or percolative superconductors there are three important transition temperatures, as distinguished from the one that is found in homogeneous ballistic superconductors. The highest is T_c^{on}, the onset temperature for decreasing resistance due to percolative dendrites. Next is T_c^m, the susceptibility threshold for diamagnetic vortex formation. Lowest is T_x, the crossover temperature from reversible to irreversible behavior. At T_x I suggest that dissipation in vortex core states becomes comparable to that from gap states outside vortex cores. While the vortex array may also freeze near T_x, the key point is that it is only below T_x that most of the dissipation is localized in the vortex cores. This model is consistent with the continuity of U_ϕ through T_c^m shown in Fig. 1.

The scale of lengths discussed here is microscopic and lies in the range between 10 and 10^4 Å. This is still larger than the atomic scale below 10 Å which is relevant to the electronic interactions responsible for high T_c's and large E_g/kT_c ratios. However, in strongly disordered materials it is generally assumed that whatever factor(s) is (are) responsible for qualitative material differences on one scale is probably responsible for these differences on the other scale as well. Thus the present microscopic theory is closely connected to my atomic scale theory[1] of the origin of high-temperature superconductivity which I call quantum-percolation theory (QPT). As in any two-fluid model, the separation of localized from extended states[20] reduces entropy, and the question of whether such separation is possible is the critical issue in all microscopic theories of metal-semiconductor transitions.[21] To the extent that the present microscopic theory is successful in explaining entropy transport above T_c, it provides fundamental support for this separation at the atomic level.

After submission of this paper, an extensive study of the effects of radiation damage on critical currents and magnetization irreversibility in Y-Ba-Cu-O appeared.[22] The authors find that although damage greatly enhances J_c, it has almost no effect on the irreversibility line. Their discussion shows that *all* conventional models based on vortex coordinates alone do not explain their data. As explained above, flux flow in conventional models is related to dissipation in vortex cores, but in cuprates dissipation can also occur in the intrinsic gap states which lie outside vortex cores. Thus we have *two* sets of dissipative coordinates instead of one, and quite generally the relations between J_c and the irreversibility line which apply in conventional unicoordinate models for conventional materials need no longer hold, regardless of the geometry and nature of vortex interactions alone. More specifically, radiation damage has almost no effect on the intrinsic gap states associated with native Fermi-energy pinning defects, which I now believe are associated with 6% native apical oxygen vacancies.[23] Most probably the radiation breaks CuO chains and forms local tetragonal clusters. The vortex lines are probably pinned by these clusters, which would have much lower T_c's, thus increasing J_c. In this model all the difficulties[22] are resolved.

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[1]J. C. Phillips, *Physics of High-T_c Superconductivity* (Academic, New York, 1989), p. 150.

[2]J. C. Phillips, Phys. Rev. B **39**, 7356 (1989).

[3]J. C. Phillips, Phys. Rev. Lett. **64**, 1605 (1990).

[4]J. C. Phillips, Phys. Rev. B **40**, 7348 (1989).

[5]V. L. Ginzburg and L. D. Landau, Zh. Eksp. Teor. Fiz. **20**, 1064 (1950).

[6]A. A. Abrikosov, Zh. Eksp. Teor. Fiz. **22**, 1442 (1957) [Sov. Phys.—JETP **5**, 1174 (1957)].

[7]L. P. Gorkov, Zh. Eksp. Teor. Fiz. **36**, 1918 (1959) [Sov. Phys.—JETP **9**, 1364 (1959)].

[8]H. Fukuyama, H. Ebisawa, and T. Tsuzuki, Prog. Theor. Phys. **46**, 1028 (1971); A. Schmid, Phys. Rev. **180**, 527 (1969).

[9]K. C. Woo *et al.*, Phys. Rev. Lett. **63**, 1877 (1989); W. K. Kwok *et al.*, *ibid.* **64**, 966 (1990); and (unpublished); D. H. Kim, K. E. Gray, R. T. Kampwirth, and D. M. McKay, Phys. Rev. B **42**, 6249 (1990); S. Ullah and A. T. Dorsey, Phys. Rev. Lett. **65**, 2066 (1990). Although Ullah and Dorsey claim to achieve quantitative agreement with the results of Ref. 12 with dynamical fluctuations only, their parameters include an unphysically small *c*-axis coherence length $\xi_c(0)=2$ Å, which is five times smaller than the spacing of the CuO_2 planes, which according to alloy studies [D. H. Lowndes, D. P. Norton, and J. D. Budai, Phys. Rev. Lett. **65**, 160 (1990)] are tightly coupled ($\xi_c \sim 10$ Å).

[10]U. Welp, W. K. Kwok, G. W. Crabtree, K. G. Vandervoort, and J. Z. Liu, Phys. Rev. Lett. **62**, 1908 (1989).

[11]M. Tinkham, *Introduction to Superconductivity* (Krieger, Malabar, FL, 1975).

[12]T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Wasczak, Phys. Rev. Lett. **64**, 3090 (1990).

[13]M. Schreiber and Y. Toyozawa, J. Phys. Soc. Jpn. **51**, 1537 (1982).

[14]D. C. Johnston, S. K. Sinha, A. J. Jacobson, and J. M. Newsam, Physica C **153-155**, 572 (1988).

[15]S. L. Cooper, M. V. Klein, B. G. Pazol, J. P. Rice, and D. M. Ginsberg, Phys. Rev. B **37**, 5920 (1988); D. Miller *et al.*, Bull. Am. Phys. Soc. **35**, 719 (1990).

[16]J. C. Phillips, Solid State Commun. **73**, 135 (1990).

[17]K. A. Müller, M. Takashige, and J. G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987); Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).

[18]M. Inui, P. B. Littlewood, and S. N. Coopersmith, Phys. Rev. Lett. **63**, 2423 (1989); R. Griessen, Phys. Rev. Lett. **64**, 1674 (1990).

[19]T. T. M. Palstra, B. Batlogg, R. B. van Dover, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **64**, 3090 (1990).

[20]J. C. Phillips, Solid State Commun. **47**, 191 (1983).

[21]M. H. Cohen, J. Non-Cryst. Solids **4**, 391 (1970); N. F. Mott, Philos. Mag. **31**, 217 (1975); J. C. Phillips, Philos. Mag. B **47**, 403 (1983); **58**, 361 (1988).

[22]L. Civale *et al.*, Phys. Rev. Lett. **65**, 1164 (1990); See also M. A. Kirk, Bull. Am. Phys. Soc. **36**, 833 (1991), for evidence that proton irradiation produces small (<15 Å) defects, which are mobile at room temperature and produce Y offsets.

[23]K. Brodt, H. Fuess, E. F. Paulus, W. Assmus, and J. Kowalewski, Acta Crystallogr. C **46**, 354 (1990).