The Mixed State in '1212' Superconductors

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INTRODUCTION

Since the discovery of high-temperature superconductors (HTSs), many attempts have been made to explain the vortex state in these materials (Zhu et al., 1999; Kolacek & Vasek, 2000; Kim et al., 2000) but to date no consensus has yet been reached. One particular vortex-state phenomenon that remains controversial is the sign reversal of the Hall resistivity, which is seen in practically all HTSs. A thorough understanding of the mixed state would allow for the understanding of flux pinning in these materials. Aside from solving an old mystery, this would also make it possible to optimize the pinning capability of HTSs. Such optimization would make it possible to greatly improve the current-carrying ability of these materials, thus making them even more viable for industrial applications, especially in power transmission.

This study offers to shed light on these issues by examining and comparing the mixed state of two isomorphic superconductors, $TlBa_2CaCu_2O_{7-\delta}$ (Tl-1212, $T_c \sim 90$ K) and $HgBa_2CaCu_2O_{6+\delta}$ (Hg-1212, $T_c \sim 120$ K). Since the structural anisotropy of these two are practically identical, any similar properties in their mixed state could be ascribed mainly to the *physical* structure of an HTS and any dissimilar properties could be ascribed mainly to the *electronic* structure of an HTS. The 30-K difference in the T_c of these species has been ascribed to differences in electronic structure, primarily the redistribution of oxygen in the unit cell as a result of exchanging Tl^{3+} cations with Hg^{2+} cations (Gapud et al., 1999). This study is also expected to shed more light on this possibility. It is also possible for such oxygen redistribution to alter mixed-state behavior.

The following phenomena were studied in particular: (1) transitions in the H-T phase diagram, (2) the behavior of the mixed-state component of the Hall angle, $\tan \theta_H$, and (3) the sign reversal of Hall resistivity.

EXPERIMENTAL DETAILS

High-quality, thin-film samples (thickness of around 0.7 μm) were used in this study. The Hg-1212 films were fabricated using a recently patented cation exchange process (Wu et al., 1999; Yan et al., 1998) in which Tl-1212 precursor films are converted into Hg-1212 films. Because of this, the superconducting '1212' layered structure is preserved under the cation exchange. The Tl-1212 films were fabricated using a modified crucible process (Siegal et al., 1997; Siegal et al., 1998) in which DC-sputtered superconducting films were further improved by annealing in Tl vapor.

Basically, two measurement programs were conducted: (1) Magnetoresistivity, both as a function of temperature and field, and (2) Hall resistivity. In both, thin (38 μm) Pt-wire leads were fused to DC-sputtered silver contacts using silver adhesive, making for negligible contact resistance signal-to-noise ratio of at least $\sim 10^6$.

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Thermovoltaic effects were eliminated by taking data while reversing the current at about 1 Hz. Resolutions are as follows: temperature, ~0.01 K; voltage, ~10 nV; current, ~10 μA. Magnetic fields of 0 to 5.5 T were provided at the central core of a 6-T superconducting magnet.

Transition lines in the H-T phase diagram were determined from measurements of magnetoresistivity and from current-voltage (I-V) data. In all these measurements, a four-point configuration was used. Hall measurements utilized a five-point contact configuration, and any misalignment effects were eliminated by taking data for reversed magnetic field.

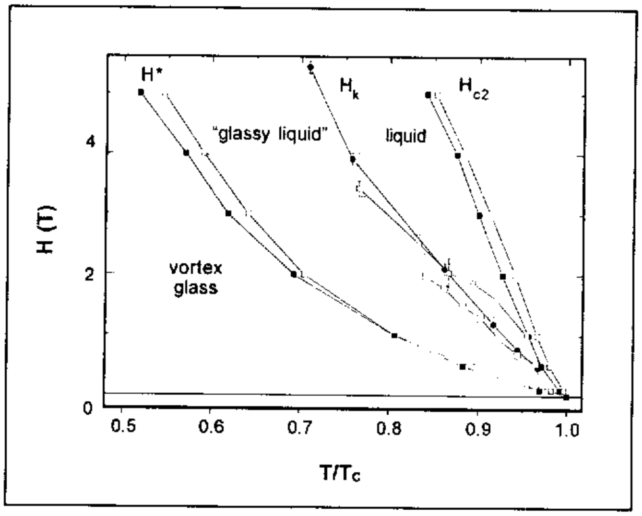

Fig. 1. H-T phase diagram of Hg-1212 (solid) and Tl-1212 (open)

DISCUSSION OF RESULTS

The irreversibility lines of 1212 species were already shown to *overlap* when H is plotted against reduced temperature, T/T_c, which supported the assertion that the 30-K difference in T_c is largely independent of the physical structure (Gapud et al., 1999). Fig. 1 shows that the other transition lines overlap as well. $H_{c2}(T)$ was obtained from constant-field resistivity curves by defining the temperature T at which the resistivity in field $H = H_{c2}$ is halfway into the transition to superconductivity. $H^*(T)$ line was obtained in a similar way, but with the temperature T being the critical temperature for the field $H = H^*$. Between H^* and H_{c2} another transition had been found, the H_k line, defined as the field above which magnetoresistivity has $H^{-1/2}$ dependence, thus marking a transition to thermally activated flux flow or a "real" vortex liquid from a "glassy" vortex liquid (Kang et al., 1998). Since these transitions are determined mainly by intrinsic pinning mechanisms, which in turn are largely dictated by physical anisotropy, such overlap for an isomorphic pair is not surprising.

The mixed-state behavior of $\tan \theta_H$ is obtained by subtracting the normal-state portion, AH/T^2, which leaves $\tan \theta_H - AH/T^2 = \tan \theta_M$. A is the slope obtained from plotting $\tan \theta_H/H$ versus T^2 for each constant-field curve. $\tan \theta_M$ is plotted for both species in Fig. 2. Above a certain temperature, the curves of each species overlap, signifying field independence – and

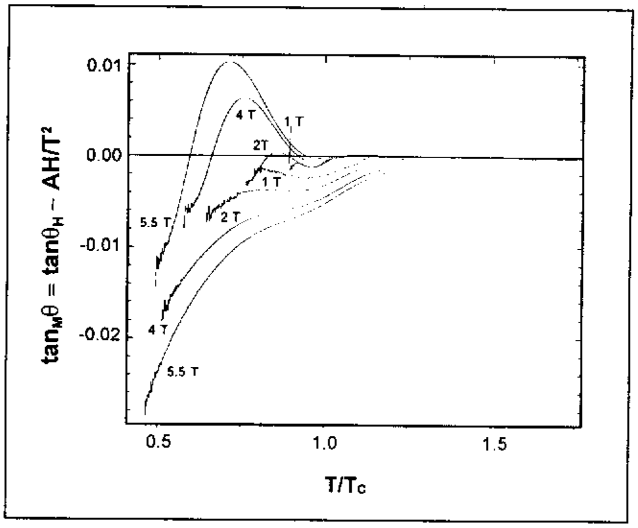

Fig. 2. Tan θ_M at different fields for Hg-1212 – top four curves – and Tl-1212 – bottom four curves

therefore independence of pinning effects. Below this temperature, one would expect to see effects due to pinning. But here one sees divergent behavior between the two species which is too subtle to be detected in the H-T phase diagram.

This is consistent with differences in the Hall sign reversal for both species. Fig. 3, also plotted in reduced temperature, T/T_c. At all fields, Hg-1212 has a much greater tendency towards double sign reversal than Tl-1212, with a subtle double reversal at 4 T. At first

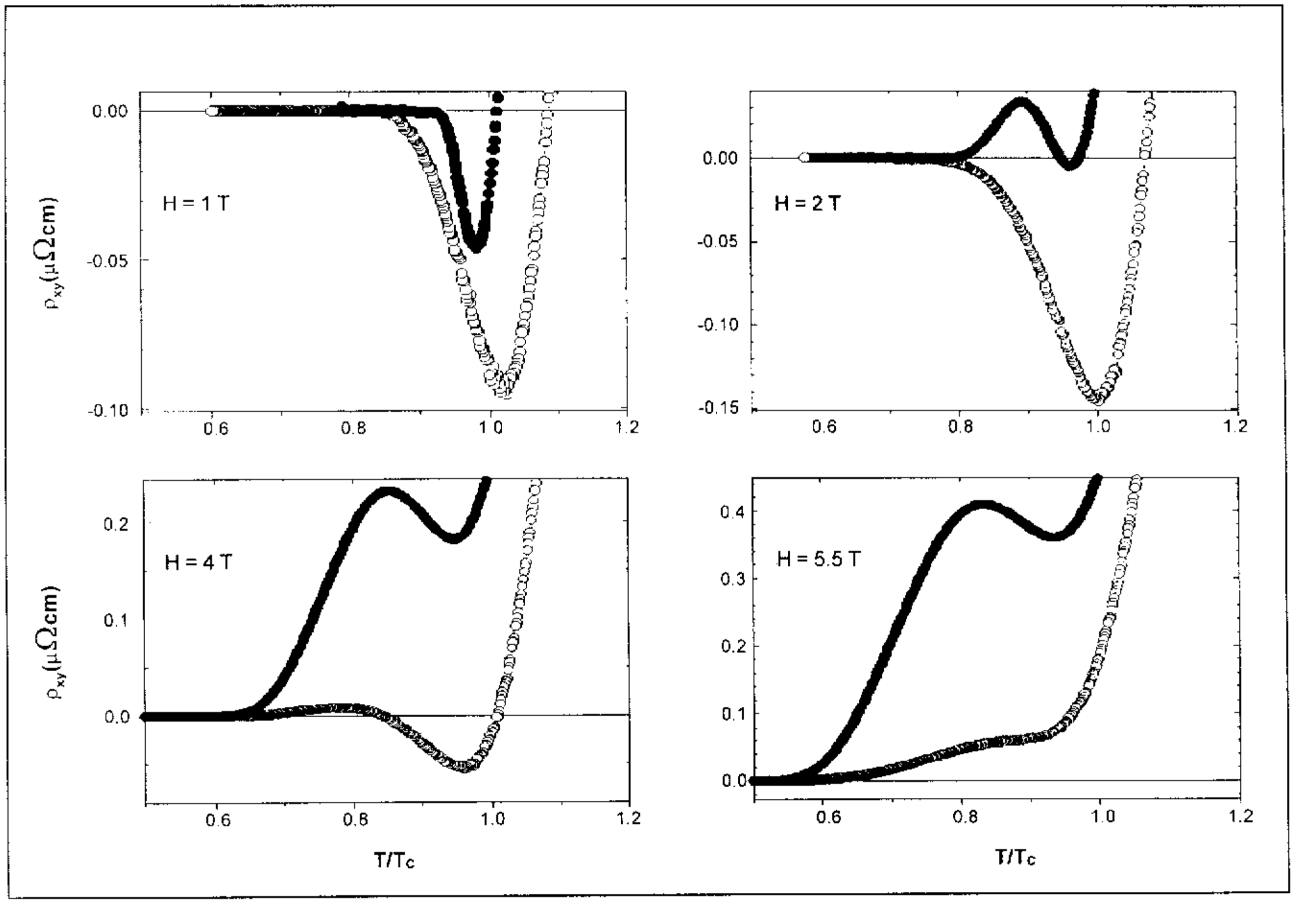

Fig. 3. Sign reversal of Hall resistivity for Hg-1212 (solid) and Tl-1212 (open) at different fields. T_c refers to the zero-field value.

glance, the implications of Fig. 2 and Fig. 3 seem to contradict those of the H-T phase diagram, Fig. 1, which implies that the intrinsic pinning is similar for both species. However the magnetically divergent behavior in tan θ_M and in the Hall sign reversal is not necessarily due to differences in pinning. There is already evidence that Hall sign reversal is not affected by artificial pinning sites but is dependent on the *doping level*, which is also consistent with the time-dependent Ginzburg-Landau model (Kopnin et al., 1993), one of the popular models to date. Sign reversal has been shown to occur mainly for underdoped HTSs (Nagaoka, 1998), and the *second* sign reversal occurs for the lowest doping levels (Kang et al., 1999).

The results therefore imply that the electronic structure of Hg-1212 and Tl-1212 differ such that Hg-1212 is actually at a *lower* doping level than Tl-1212. It would seem, then, that the divergent magnetic behavior in the mixed state as illustrated by tan θ_M is more an effect of differing doping levels and less an effect of pinning. Since there is a magnetic dependence in this regime, this further suggests the existence of differing *spin* structure between the species, either as a result of the different properties of the Hg^{2+} cation and the Tl^{3+} cation, or the different distribution of oxygen, or both, and this could provide an important mechanism behind the 30-K difference in their T_c.

In summary, similar pinning continues to be observed in '1212' superconductors Hg-1212 and Tl-1212, but observations in the mixed state have uncovered and magnified subtle differences in the electronic structure behind the 30-K difference in T_c, implying that Tl-1212 is actually less underdoped than Hg-1212. The exchange of cations and/or the resultant redistribution of oxygen could also produce a difference in magnetic spin structure, causing magnetically divergent behaviors only seen in the mixed state. *All this strongly suggests that the vortex state in HTSs is governed more by doping level, electronic structure, and spin structure, than by intrinsic pinning.*

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REFERENCES

Gapud, A.A., J. Z. Wu, B. W. Kang, S. L. Yan, Y. Y. Xie, & M. P. Siegal, 1999. Giant T_c shift in $HgBa_2CaCu_2O_{6+\delta}$ and $TlBa_2CaCu_2O_{7-\delta}$ super-conductors due to Hg-Tl exchange. *Phys. Rev. B* Vol. 59, No. 1, pp. 203-206.

Kang, B. W., J. Z. Wu, A. A. Gapud, W. N. Kang, D. K. Christen, & R. Kerchner, 1998. Unpublished data, private communication.

Kang, B. W., J. Z. Wu, W. N. Kang, Q. Y. Chen, W. K. Chu, & Z. Ren, 1999. Effect of anion doping on Hall sign anomaly of Tl-2201 films. *Philos. Mag. Lett.*

Kim, Wan-Seon, W. N. Kang, Mun-Seog Kim, & Sung-Ik Lee, 2000. Vortex phase diagram of Hg-1223 thin films from magnetoresistance measurements. *Phys. Rev. B* Vol. 61, No. 17, pp. 11317-11320.

Kolacek, Jan & Petr Vasek, 2000. Hall voltage sign reversal in type II superconductors. *Physica C* Vol. 336, pp. 199-204.

Kopnin, N. B., B. I. Ivlev, & A. V. Kalatsky, 1993. *J. Low Temp. Phys.* Vol. 90, p. 1.

Nagaoka, T., Y. Matsuda, H. Obara, A. Sawa, T. Terashima, I. Chong, M. Takano, & M. Suzuki, 1998. Hall anomaly in the superconducting state of high-T_c cuprates: universality in doping dependence. *Phys. Rev. Lett.* Vol. 80, No. 16, pp. 3594-3597.

Siegal, M. P., E. L. Venturini, D. L. Overmyer, & P. P. Newcomer, 1998. Growth and characterization of TlBaCaCuO thin films. *J. of Superconductivity* Vol. 11, p. 135.

Siegal, M. P., E. L. Venturini, B. Morosin, & T. L. Aselage, 1997. Synthesis and properties of Tl-Ba-Ca-Cu-O superconductors. *J. of Materials Research* Vol. 12, p. 2825.

Wu, J. Z., S.L. Yan, & Y. Y. Xie, 1999. Cation exchange: A scheme for synthesis of mercury-based high-temperature superconducting epitaxial thin films. *Appl. Phys. Lett.* Vol. 74, No. 10, pp. 1469-1471.

Yan, S. L., Y. Y. Xie, J. Z. Wu, T. Aytug, A. A. Gapud, B. W. Kang, L. Fang, M. He, S. C. Tidrow, K. W. Kirchner, J. R. Liu, & W. K. Chu, 1998. High critical current density in epitaxial $HgBa_2CaCu_2O_x$ thin films. *Appl. Phys. Lett.* Vol. 73, No. 20, pp. 2989-2991.

Zhu, B.Y., D. Y. Xing, Z. D. Wang, B. R. Zhao, & Z. X. Zhao, 1999. Sign reversal of the mixed-state Hall resistivity in type-II superconductors. *Phys. Rev. B* Vol. 60, No. 5, pp. 3080-3083.