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Cu-O Bilayers in a Highly-Anisotropic Superconductor***

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Direct Evidence for, and the Nature of, Josephson Coupling between Cu-O Bilayers in a Highly-Anisotropic Superconductor

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ABSTRACT

The angular-dependent, *c*-axis resistivity for oxygen-deficient $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals is shown to be a maximum for fields parallel to the *c*-axis, i.e., for zero macroscopic Lorentz force, and agrees with a series stack of Josephson tunnel junctions. The *c*-axis component of field dominates the *c*-axis dissipation in most cases. The results indicate the possibility of an unusual normal-state *c*-axis conductance and that the *c*-axis junctions may be extremely underdamped. [word count=80]

Recent experiments on^{1,2} $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and² $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ crystals related to the c-axis conduction in highly-anisotropic, high-temperature superconductors (HTS) and a subsequent interpretation³ of the field dependence found in Ref. 1 have strongly suggested that these HTS materials behave as a stack of Josephson junctions between close, well-coupled, bi- or tri-layers of Cu-O. The strong, $1/B$ dependence of Josephson coupling energy, E_j , on the field, B , parallel to the c-axis, used in Ref. 3, was first proposed⁴ to understand the irreversibility behavior of the ab-plane resistivity for fields parallel to the c-axis. While such a field dependence was in quantitative agreement with measurements⁵ in discrete, thin-film Nb-based junctions, a more convincing test of this aspect of the model for HTS requires direct c-axis resistivity measurements. This also eliminates the uncertain absolute magnitude of E_j inherent in the analysis of Ref. 4.

Thus we report measurements of the c-axis resistance, as a function of magnetic field and its orientation with respect to the c-axis, using oxygen-deficient, and thus reasonably anisotropic⁶, $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals ($T_c=35-75$ K). The measured angular-dependent c-axis resistance, at temperatures somewhat below the transition temperature, T_c , exhibits a maximum in dissipation for fields parallel to the c-axis, i.e., for zero macroscopic Lorentz force: this tends to rule out⁷ the motion of vortices from the external field. The dependences of the dissipation on applied field strength and angle with respect to the c-axis are in excellent agreement with the field dependence predicted by Refs. 3-5 for a series stack of Josephson tunnel junctions, assuming that only the c-axis component of the field plays a role⁸. The data define a Josephson coupling energy^{9,10} which shows the same field dependence as found⁵ in the discrete, thin-film Nb-based junctions.

Single crystals, grown using a self-flux method¹¹, had their oxygen composition modified (reduced) by annealing for 200 hours at 500 °C in appropriate mixtures¹² of O_2 in N_2 such that 0.095, 3.7 and 8.3% O_2/N_2 yielded $T_c=35, 65$ and 75 K as measured by electrical transport. Silver epoxy was placed on the plate-like crystal surfaces ($\sim 1\text{mm} \times 1\text{mm} \times 50\mu\text{m}$) before annealing. The Ag diffuses into the crystal during the oxygen anneal to form an electrical contact pad to which Au leads were attached, two on each face. Large current pads decrease heating effects and improve current uniformity. For very large resistivity anisotropies, currents redistribute rapidly in the ab-planes creating an effectively uniform c-axis current. This uniformity was confirmed by exchanging current and voltage leads on the bottom (only) of the crystal and getting identical results. More isotropic samples than used here (i.e., higher T_c) failed this test because of the (relatively) smaller c-axis resistivity.

Measurements of dc electrical transport properties were carried out in a 7 T split-coil radial-access magnet in which the sample probe is rigidly mounted on a rotational feedthrough which was stepper-motor controlled with a reproducibility of better than 0.01 degrees. Voltage-current measurements confirmed that the data were always collected in the linear response regime (using $J \sim 1 \text{ A/cm}^2$).

The c-axis resistance, R_c , of an oxygen-deficient $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystal with $T_c \sim 65 \text{ K}$ is shown in Fig. 1 as a function of the angle, ϕ , between the field direction and the c-axis for a temperature of 60 K and a field, B , of 1 T. The maximum R_c occurs for $B \parallel c$, which corresponds to a *minimum* macroscopic Lorentz force on the Abrikosov vortices and it drops over three orders-of-magnitude into the noise for the *maximum* Lorentz force, i.e., $B \parallel ab$. This tends to rule out⁷ the motion of vortices from the external field as the cause of the dissipation, but it is shown below to be consistent with dissipation across inter-bilayer Josephson junctions.

At reduced temperatures, $t \equiv T/T_c$, closer to one, dissipation due to flux motion¹³ can be relatively more important because the pinning energy is $\sim (1-t)^2$, while the Josephson coupling is $\sim (1-t)$. Such additional dissipation is clearly seen nearer to T_c but only for fields close to the minimum-pinning, maximum-Lorentz-force, ab-plane direction. We do not discuss this result further, but it emphasizes the clear distinction between dissipation across Josephson junctions and that due to motion of vortices from the external field.

We also measured $R_c(T)$ at $\phi=0$ as a function of B and determined the activation energy, U , from Arrhenius plots. These results are shown in Fig. 2 for two $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystals ($T_c=65$ and 75 K), together with identical determinations made on two discrete, thin-film Nb junctions⁵. The qualitative similarity is striking, and thus the present results seem to imply consistency with the model of Ref. 5 in which the effective coherent Josephson area, A_{eff} , of the Nb junctions is experimentally shown to be Φ_0/B at high fields. Note that at low fields, A_{eff} is limited by junction dimensions or defects to some fixed A_0 , which we incorporate into the model by:

$$A_{\text{eff}} = \Phi_0 / (B + B_0), \quad (1)$$

where $B_0 \equiv \Phi_0 / A_0$: then the fits shown in Fig. 2 indicate $B_0 \sim 0.05 \text{ T}$, or $A_0 \sim 0.04 \mu\text{m}^2$, for the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystals.

In Fig. 3, we show *experimental* evidence of the extent to which it is the c-axis component of B which primarily determines R_c (as first described in Ref. 8 for *ab-plane transport*). The solid lines represent data for field sweeps with $B \parallel c$ from three

YBa₂Cu₃O_{7-δ} crystals ($T_c=35, 65$ and 75 K), while the symbols represent rotations at fixed fields (of 4, 2 and 1.6 T) for the same crystals. Note that for the 65 K crystal the rotation data for 4 and 2 T were taken at 51 and 49.5 K respectively, while the field sweep was done at 50 K, leading to the slight systematic offsets shown. This new result is consistent with Josephson dissipation of Refs. 3-5 but not flux motion.

Encouraged by this excellent qualitative agreement with Josephson dissipation, we seek a more quantitative fit of the data using simple extensions of conventional Josephson models. To fit the data in Fig. 1, we use¹⁰

$$R_c(T) = R_{Nc}(T) \{ [L_0(E_j/2k_B T)^2]^{-1}, \quad (2)$$

which at low temperatures reduces to

$$R_c(\phi) = R_{Nc} \exp\left(-\frac{e_j(T)\Phi_0}{(B\cos\phi+B_0)k_B T}\right), \quad (3)$$

and is thus consistent with the Arrhenius activation leading to Fig. 2. Here the fitting constants are R_{Nc} and $e_j \equiv E_j/A_{eff}$, since $B_0=0.05$ T (see above). The excellent agreement with Eq. 3, shown in Fig. 1, is strong supporting evidence for the overall Josephson model of Refs. 3-5 including further confirmations of the $1/B$ dependence of E_j and the significance of the c -axis component of B . We now express e_j within the Anderson-Ambegaokar-Baratoff⁹ (AAB) model as:

$$e_j(T) = \frac{\pi \hbar \Delta(T)}{2e^2 s \rho_{Nc}} \tanh\left(\frac{\Delta(T)}{2k_B T}\right), \quad (4)$$

where s is the repeat distance of the Cu-O bilayers, and ask whether values for the c -axis resistivity, ρ_{Nc} , and energy gap, $\Delta(T)$, can be found which are compatible with R_{Nc} and a suitable $\Delta(T)$. We expect ρ_{Nc} to be given by $R_{Nc}A_{xtal}/t_{xtal}$, where A_{xtal} and t_{xtal} are the area and c -axis thickness of the crystal, but in that case there is a significant disagreement with a reasonable $\Delta(T)$ for rotational data taken at lower temperatures, and conversely, using $\Delta(T)/\Delta(0)$ from the BCS theory with $\Delta(0)=1$ to 1.4 times the BCS value¹⁴, $\Delta_{BCS}(0)$, of 10 meV, there is a significant disagreement at lower temperatures with the expected ρ_{Nc} .

In order to systematically address the discrepancy with the standard Josephson model^{9,10} we have measured R_c for the crystal with $T_c=65$ K over a wide range of B and T and include a parallel quasiparticle conductance in fits, as in Ref. 3. This

procedure is more clearly presented in terms of the *c*-axis conductances, $Y_c \equiv 1/R_c$, which are plotted in Fig. 4, and fit by^{3,10}

$$Y_c(T) = Y_{Nc}(T) \{ I_0(E_j/2k_B T)^2 - 1 + \sigma_{qp}(T) \}, \quad (5)$$

where the first two terms in brackets represent the pair conductance³, I_0 is the modified Bessel function¹⁰ and $\sigma_{qp}(T)$ is the quasiparticle conductance, normalized to its normal-state value, which is given analytically in Taylor's thesis¹⁵. For the 60 K data, the dotted curve represents *only* the pair conductance, while the solid curve includes σ_{qp} , with $\Delta(0) = \Delta_{BCS}(0)$. It is clear that σ_{qp} has little effect on the vast majority of the data *which thus accurately determine* E_j and Y_{Nc} from Eq. 5.

As above, we cannot find values for ρ_{Nc} which are compatible with Y_{Nc} , E_j and a suitable $\Delta(T)$. To show this we plot in Fig. 5 the *c*-axis resistance of the $YBa_2Cu_3O_{7-\delta}$ crystal ($T_c=65$) in zero field as a function of T , together with values determined from the fits to Fig. 4 of $1/Y_{Nc}$ and $\rho_{Nc}t_{xtal}/A_{xtal}$. Only the latter parameter is affected by $\Delta(0)$, through Eq. 4, so they are shown for both $\Delta(0) = \Delta_{BCS}(0)$ and¹⁴ $1.4\Delta_{BCS}(0)$. A similar problem occurs in fits³ to the *c*-axis resistivity of $Bi_2Sr_2CaCu_2O_8$: using a temperature-independent Y_{Nc} , the *low*-conductance fit to the Josephson model was consistent with the measured Y_{Nc} above T_c , but $Y(T)$ at low-temperatures needed a larger E_j .

A difficulty with this analysis is our inability to unambiguously determine the normal-state resistance, even above T_c , since expected fluctuation effects³ on $R_{Nc}(T > T_c)$ cannot be easily estimated because the strictly two-dimensional nature of the Cu-O bilayers is less certain in $YBa_2Cu_3O_{7-\delta}$, with its partial chains, than for $Bi_2Sr_2CaCu_2O_8$. Nevertheless, we can speculate. One possibility is that the *c*-axis transport may be more coherent at low temperatures, either: below T_c , since the superconductivity will diminish in-plane fluctuations that would otherwise dephase adjacent bilayers¹⁶; or at low temperature such that the direct, metallic-like *c*-axis hopping rate exceeds that for boson assisted processes¹⁷. The smaller values and temperature dependence of $\rho_{Nc}t_{xtal}/A_{xtal}$ compared to $R_{Nc}(T \geq T_c)$ could result from this. For the opposite dependence of $1/Y_{Nc}$, it is possible that these *c*-axis Josephson junctions are not overdamped, and thus the prefactors for the pair conductance in Eqns. 2, 3 and 5 may not be simply the normal-state one. Data on extremely underdamped, discrete Nb-Pb junctions find¹⁸ a prefactor resistance which is inversely proportional to the quasiparticle density, increasing dramatically

as T decreases and equal to R_N below T_c . The fit values of the prefactor Y_{Nc} could certainly be consistent with such a picture.

In conclusion, while our data undeniably shows that Josephson coupling and dissipation dominates for transport along the c -axis, the details indicate the possibility of an unusual normal-state conductance and that the c -axis Josephson junctions may be extremely underdamped. We also show for the first time that the c -axis component of magnetic field dominates the c -axis dissipation in most cases (except for fields nearly parallel to the ab planes).

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[text word count=1950; characters=11382]

REFERENCES

1. G. Briceno, M.F. Crommie and A. Zettl, *Phys. Rev. Lett.* **66**, 2164 (1991).
2. R. Kleiner, F. Steinmeyer, G. Kunkel and P. Müller, *Phys. Rev. Lett.* **68**, 2394 (1992); R. Kleiner and P. Müller, preprint.
3. K.E. Gray and D.H. Kim, *Phys. Rev. Lett.* **70**, 1693 (1993).
4. D.H. Kim, K.E. Gray, R.T. Kampwirth, J.C. Smith, D.S. Richeson, T.J. Marks, J.H. Kang, J. Talvacchio and M. Eddy, *Physica C177*, 431 (1991).
5. D.H. Kim, K.E. Gray and J.H. Kang, *Phys. Rev.* **B45**, 7563 (1992).
6. K.E. Gray, D.H. Kim, B.W. Veal, G.T. Seidler, T.F. Rosenbaum and D.E. Farrell, *Phys. Rev.* **B45**, 10071 (1992).
7. K.E. Gray, R.T. Kampwirth, D.J. Miller, J.M. Murduck, D. Hampshire, R. Herzog and H.W. Weber, *Physica C174*, 340 (1991).
8. P.H. Kes, J. Aarts, J. van den Berg, C.J. van der Beek and J.A. Mydosh, *Supercond. Sci. Technol.* **1**, 242 (1989).
9. P.W. Anderson, Lectures at Ravello Spring School, 1963; V. Ambegaokar and A. Baratoff, *Phys. Rev. Lett.* **10**, 486 (1963).
10. V. Ambegaokar and B.I. Halperin, *Phys. Rev. Lett.* **22**, 1364 (1969).
11. D. L. Kaiser, F. Holtzberg, M. F. Chisholm and T. K. Worthington, *J. Crystal Growth* **85**, 593(1987).
12. J. D. Jorgensen, B. W. Veal, A. P. Paulikas, L. J. Nowicki, G. W. Crabtree, H. Claus and W. K. Kwok, *Phys. Rev.* **B41**, 1863 (1990).
13. A.F. Volkov and Y.I. Latyshev, *JETP Lett.* **53**, 213 (1991).
14. Tunneling measurements consistently see features near 20 meV for $\text{YBa}_2\text{Cu}_3\text{O}_7$ with $T_c \sim 90$ K, i.e., roughly 1.4 times the BCS value, e.g., see M. Kawasaki, et al, *Jpn. J. Appl. Phys.* **32**, 1612 (1993). Such a large value is consistent with tunneling into many other HTS cuprates which range up to twice BCS, see J.F. Zasadzinski, et al, *J. Phys. Chem. Solids* **53**, 1635 (1992).
15. B.N. Taylor, Thesis, University of Pennsylvania, 1963, unpublished, p. 50.
16. A.J. Leggett, private communication.
17. R.J. Radtke, C.N. Lau and K. Levin, preprint.
18. P. Silvestrini, O. Liengme and K.E. Gray, *Phys. Rev.* **B37**, 1525 (1988).

Figure Captions

- Fig. 1. The *c*-axis resistance of a $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystal ($T_c=65$ K) as a function of the angle between the field direction and the *c*-axis taken at 60 K and 1 T. The solid line is a fit to Eq. 3 for purely Josephson dissipation, using $R_{Nc}=0.1$ ohms and $e_j(60 \text{ K})=2.6 \times 10^{-6} \text{ J/m}^2$.
- Fig. 2. The activation energy determined from Arrhenius plots of $R_c(T)$ as a function of B for $\phi=0$. Two $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystals ($T_c=65$ and 75 K) and two discrete, thin-film Nb junctions⁵ are shown (open symbols) to be strikingly similar with a $1/B$ dependence at high fields.
- Fig. 3. The *c*-axis resistance of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystals ($T_c \sim 35, 65$ and 75 K) as a function of the *c*-axis component of B . The solid lines are data for field sweeps (with $\phi=0$), while the symbols represent rotations at fixed fields (of 4, 2 and 1.6 T) for the same crystals. Note that for the 65 K crystal the data were taken at slightly different temperatures, which are consistent with the slight systematic offsets.
- Fig. 4. The *c*-axis conductance of a $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystal ($T_c=65$ K) for various temperatures, as a function of B for $\phi=0$. Fits are shown to the pair conductance alone (dotted line for 60 K data only) and a sum of the pair and quasiparticle conductances (Eq. 5) for $\Delta(0)$ equal to the BCS value.
- Fig. 5. The *c*-axis resistances of a $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ crystal ($T_c=65$) in zero field as a function of T (open circles), together with values determined from the fits to Fig. 5 of $1/Y_{Nc}$ (triangles) and $\rho_{Nc}t_{\text{xtal}}/A_{\text{xtal}}$. The latter are shown for $\Delta(0)$ equal to $\Delta_{\text{BCS}}(0)$ (solid squares) and $1.4\Delta_{\text{BCS}}(0)$ (dashed line) since the values of ρ_{Nc} , but not E_j , obtained from the fits in Fig. 5 depend on the actual (but unknown) magnitude of $\Delta(0)$.

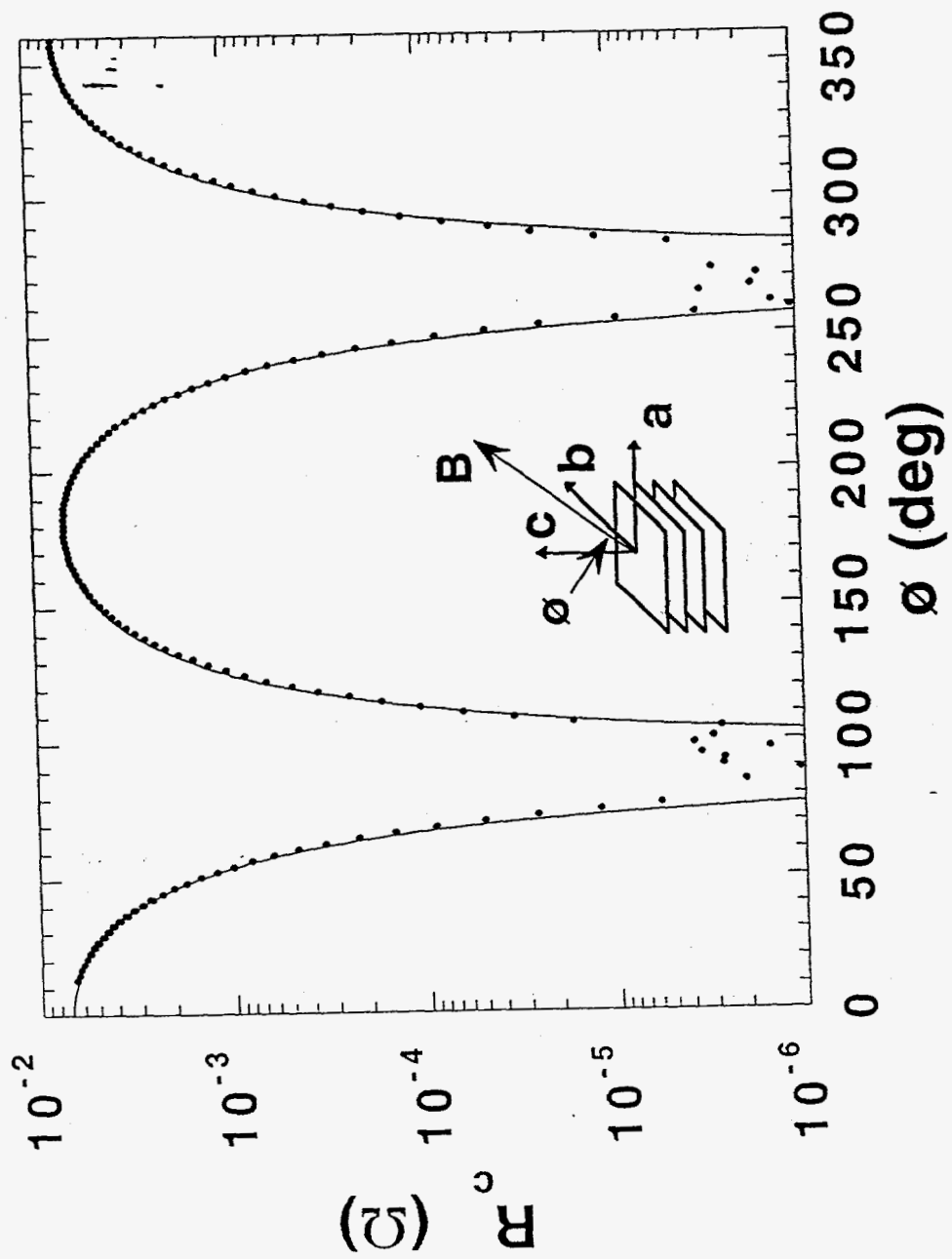


Fig 1

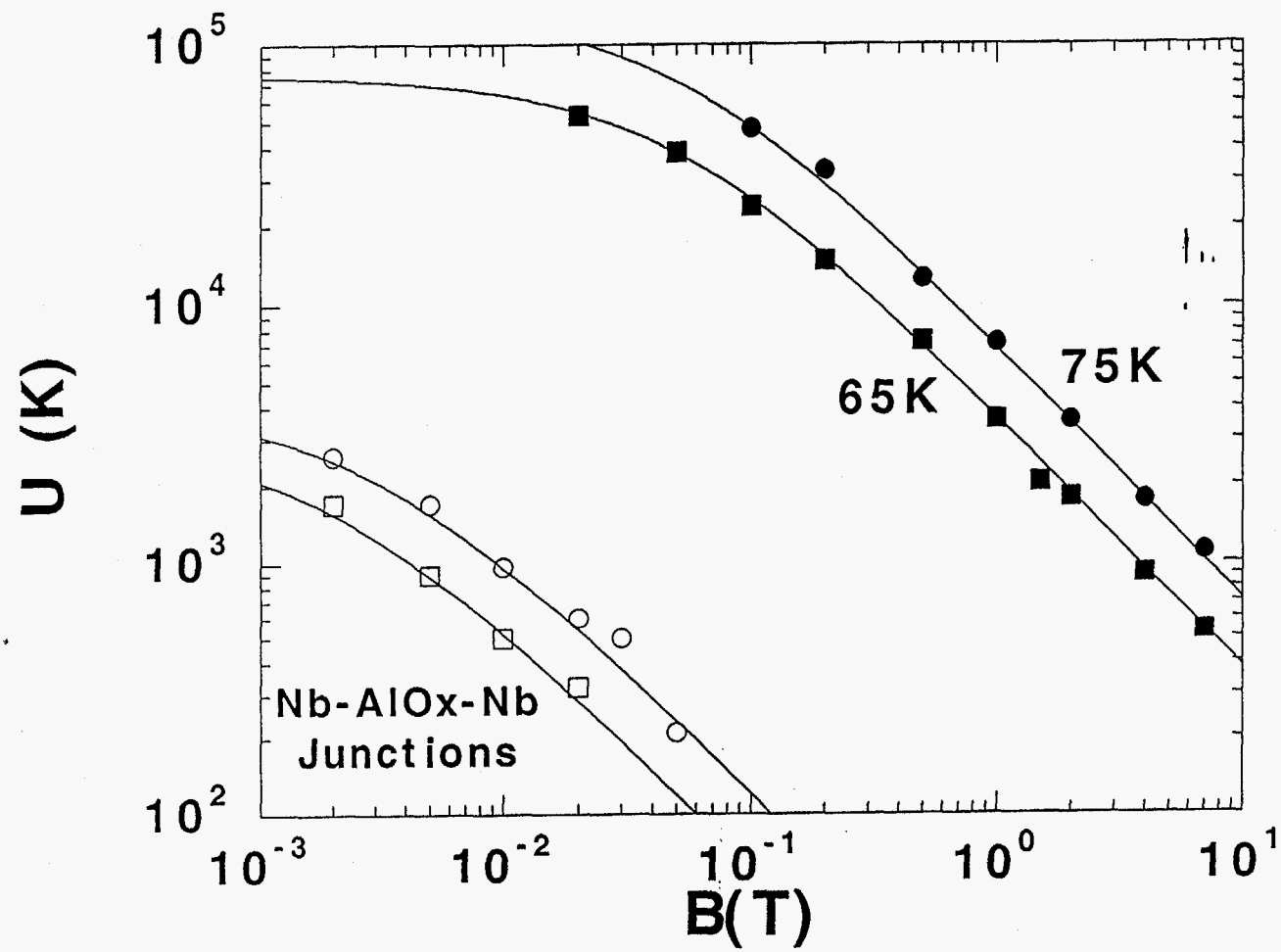


Fig 2

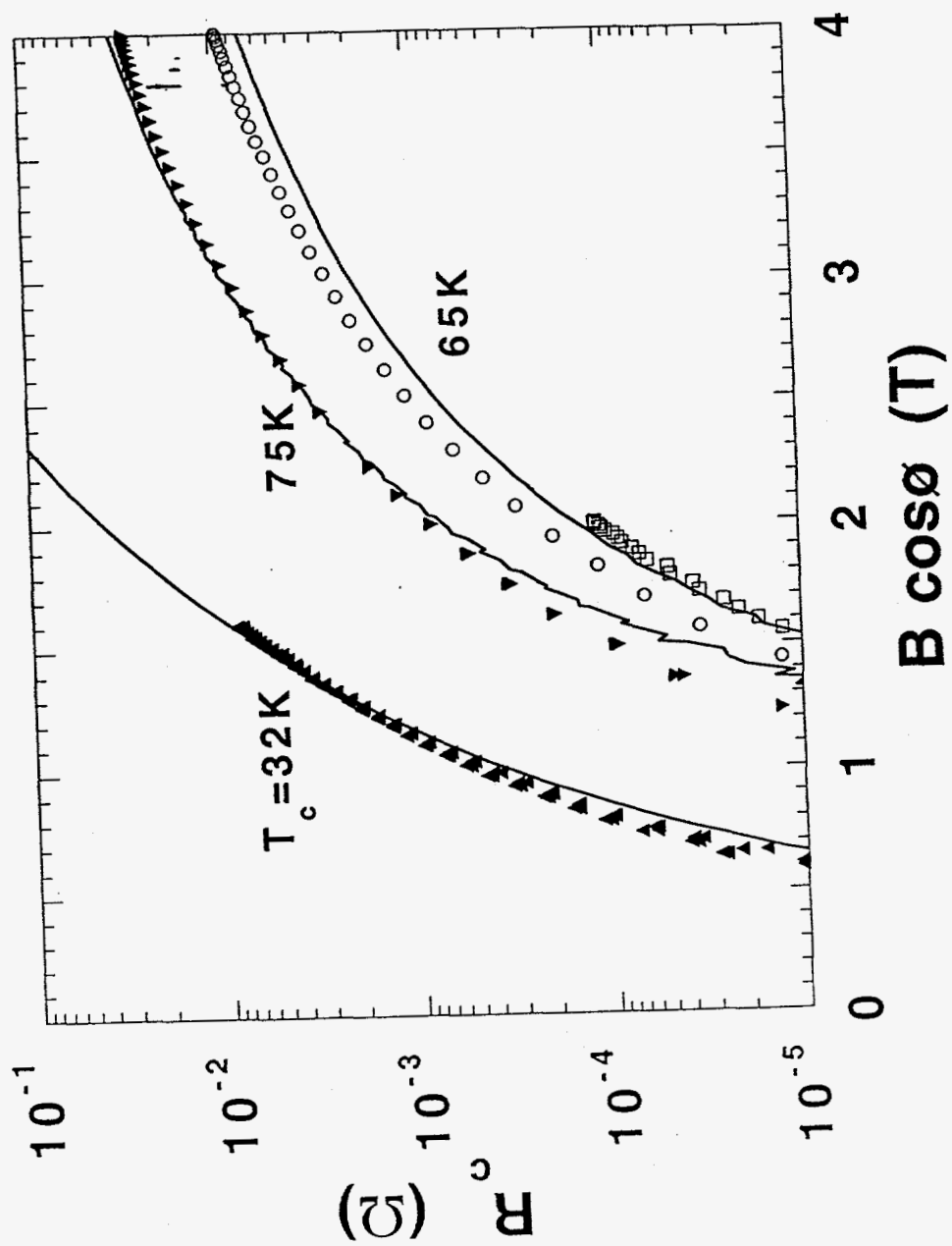


Fig 3

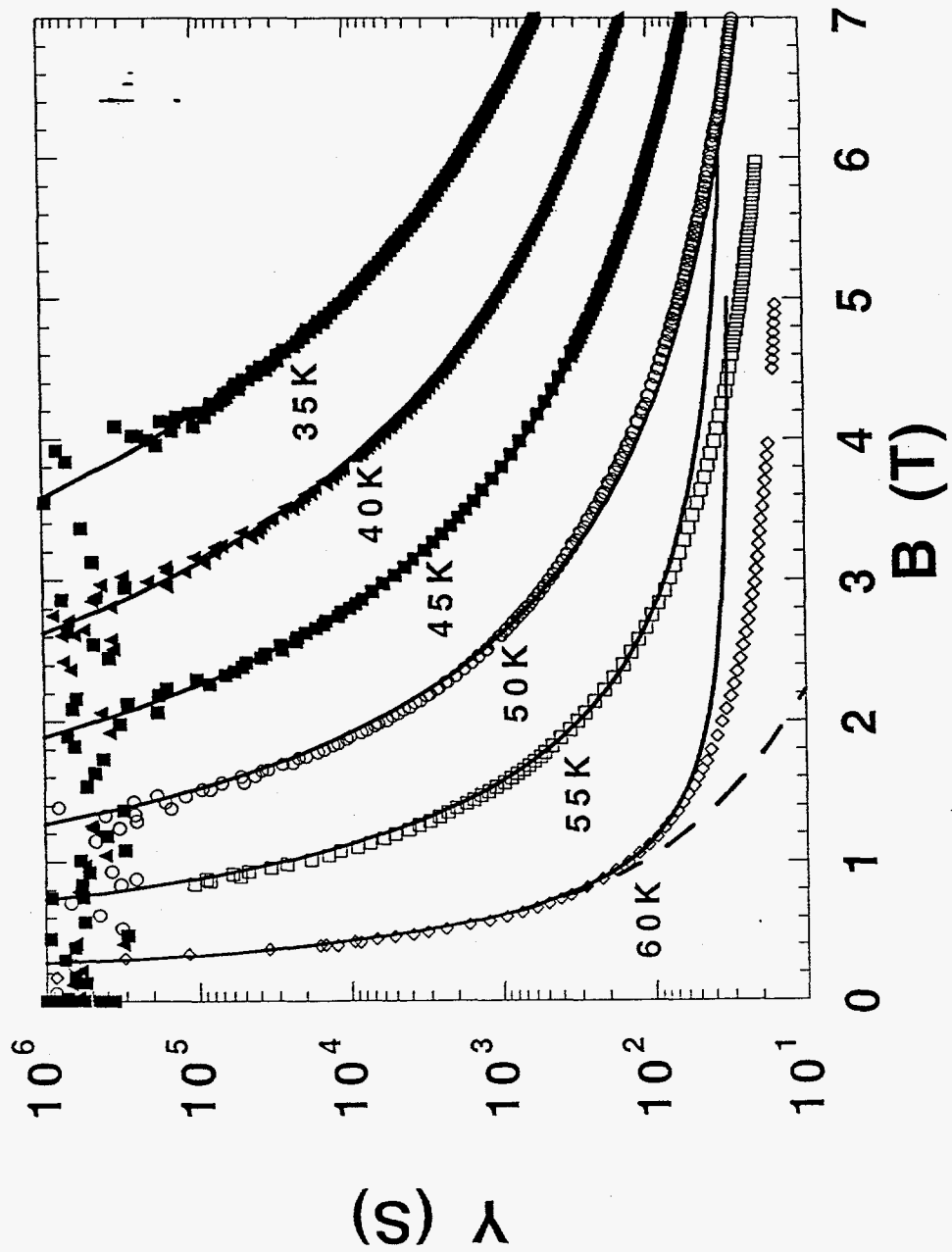


Fig 4

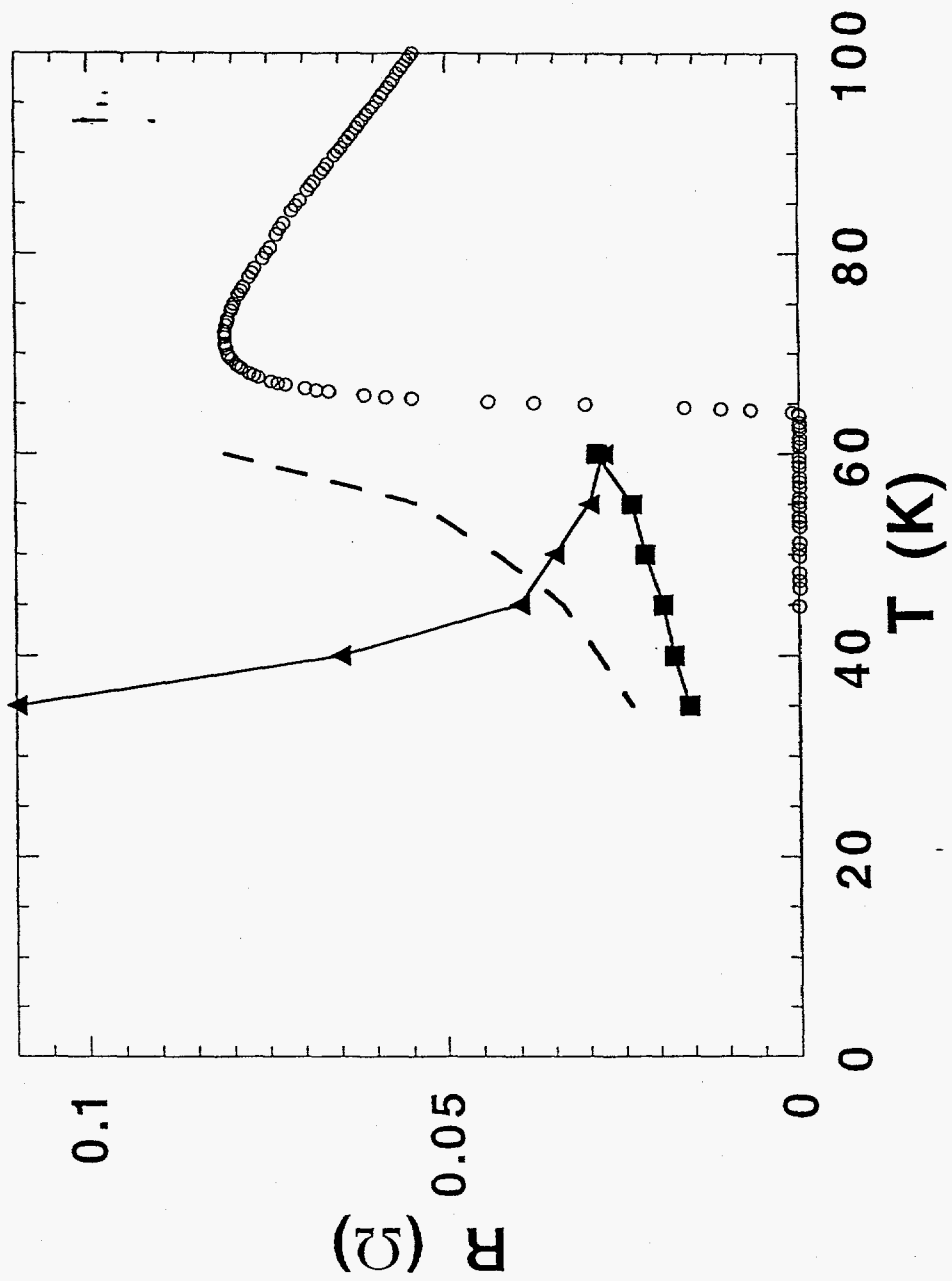


Fig 5