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Ion-beam-induced metal-insulator transition in $YBa_2Cu_3O_{7-\delta}$: A mobility edge

J. M. Valles, Jr., A. E. White, K. T. Short, R. C. Dynes, J. P. Garno,
A. F. J. Levi, M. Anzlowar, and K. Baldwin
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

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We have studied the temperature-dependent resistivity and Hall coefficient of epitaxial $YBa_2Cu_3O_{7-\delta}$ films as a function of ion-beam-induced damage. At low ion fluences the films behave like typical metals above the superconducting transition. At higher ion fluences, the material goes continuously through the metal-insulator transition with a resistivity that varies like $\exp(T^{-1/2})$ but with a Hall coefficient that changes very little. This implies that the transition is a result of a reduction in mobility rather than a drop in carrier density.

I. INTRODUCTION

One of the puzzling characteristics of the high-T_c superconductor $YBa_2Cu_3O_{7-\delta}$ is the normal-state transport. It has been shown that in single crystals and high-quality films of $YBa_2Cu_3O_{7-\delta}$ the temperature-dependent resistivity $\rho(T)$ is remarkably linear above the transition, extrapolating to zero at $T=0$ K. Perhaps even more unusual are the results of Hall measurements which show the Hall coefficient $1/R_H$ also varying linearly with temperature and extrapolating to zero at $T=0$ K.[1,2] Using the traditional interpretation that $1/R_H$ is the carrier density for a simple single-band metal these results imply that the resistivity goes to zero as the carrier density goes to zero. This then places severe restrictions on the temperature dependence of the resultant mobility and leads one to suspect that a closer look at transport results at the metal-insulator transition may be a key to understanding the complex interactions causing this behavior.

In this work, we have studied the evolution of $\rho(T)$ and $1/R_H$ in high quality thin films of $YBa_2Cu_3O_{7-\delta}$ as defects are systematically introduced by ion-beam irradiation. As previously reported, at low fluences we see a continuous decrease in the superconducting transition temperature $T_c(R=0)$, and an increase in $\rho(T)$.[3] The ion-beam-induced defects act primarily to change the residual resistivity. At higher ion fluences, the material undergoes a continuous transition to insulating behavior. However, only very minor changes in R_H are associated with this enormous change in resistivity. If $1/R_H$ is interpreted as the carrier density, the implication is that the transition to an insulating state comes about because of a mobility edge. In Sec. II we briefly describe the experimental procedures and then present and discuss the results of the measurements. A simple model to describe the effect of the damage is introduced in Sec. III and, in Sec. IV, we summarize and draw conclusions.

II. EXPERIMENTAL PROCEDURE AND RESULTS

We studied epitaxial films of $YBa_2Cu_3O_{7-\delta}$ oriented with the c axis perpendicular to the $SrTiO_3$ substrates, which are grown using techniques described previously.[4] Three separate samples were studied extensively and the results were similar. For transport studies, 50-μm-wide bridges were photolithographically defined and etched in dilute HCl. The samples were mounted on a Cu holder which could be moved from the cryostat to the beam line for irradiation. This meant that the leads remained attached throughout the entire series of irradiations and measurements. In these studies, we chose 1 MeV Ne^+ ions for the bombardment because the range of the ions was much greater than the 1500–2000 Å film thicknesses. The nuclear-energy loss, which has been shown to be the primary damage mechanism,[5] is approximately constant throughout the film.

In Fig. 1 we show a typical series of ρ vs T curves illustrating the effects of ion irradiation on the resistivity and superconducting properties of a film of $YBa_2Cu_3O_{7-\delta}$. With increasing damage, the normal-state resistivity systematically changes from a linear dependence on temperature to insulating behavior where the resistivity is rising exponentially with decreasing temperature. Moreover, in the course of this transition from metallic to insulating behavior, the superconducting transition temperature,

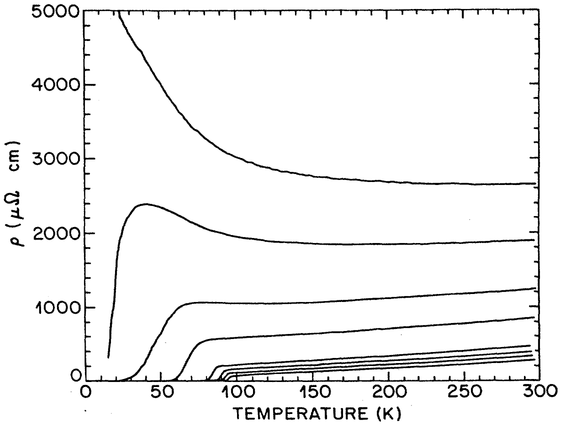

FIG. 1. Resistivity as a function of temperature for a single film after bombardment with Ne ions at fluences of (0, 0.1, 2.5, 4.0, 10.0, 15.0, 20.0, and 22.0) $\times 10^{13}$ ions/cm^2.

$T_c(R=0)$, decreases continuously and disappears in the vicinity of the metal-insulator transition. In the following, we will look more carefully at both the metallic and the insulating regimes.

In Fig. 2 we show a set of resistivity and Hall coefficient data for low Ne$^+$ fluences where the film is still metallic and superconducting. This is the region where the dependence of the superconducting critical current on fluence was previously studied.[3] The $\rho(T)$ data seem to approximately obey Matthieson's rule:

$$\rho(T)=\rho_0+\rho_1(T)\,, \tag{1}$$

where ρ_0, the zero-temperature intercept, scales linearly with ion fluence at low fluence, and $\rho_1(T)$, the resistivity intrinsic to the original starting material, remains unaffected. However, upon closer inspection of the data, it is observed that this simple description needs to be modified. Over the range shown in Fig. 2(a), the slope of $\rho(T)$ increases by 30% from 2.23 to 2.93 $\mu\Omega$ cm/K. A model which is consistent with this result is discussed in Sec. III.

The Hall coefficient measurements in Fig. 2(b) show relatively less sensitivity to fluence over the same fluence range. While the unexpected linear dependence of $1/R_H$ on temperature persists, the slopes of the $1/R_H$ vs T curves decrease quite markedly. This linear dependence of $1/R_H$ on temperature has been previously noted and an extreme two-carrier model has been proposed as an explanation.[1] At higher damage levels, this trend continues until there is almost no dependence on temperature, a result more characteristic of a normal metal.

The results of a closer look at the transport properties as a $YBa_2Cu_3O_{7-\delta}$ undergoes a transition into the insulating state are shown in Figs. 3 and 4. We find that the resistivity in the insulating regime [shown with a logarithmic scale in Fig. 3(a)] is best characterized by an exponential:

$$\rho=\rho_0 e^{(\Delta/kT)^{1/2}}\,, \tag{2}$$

where Δ can be thought of as an activation energy. At the highest damage levels studied, this dependence is observed from room temperature to 1 K and spans four decades in resistivity, as illustrated in Fig. 4. Clearly, the material is

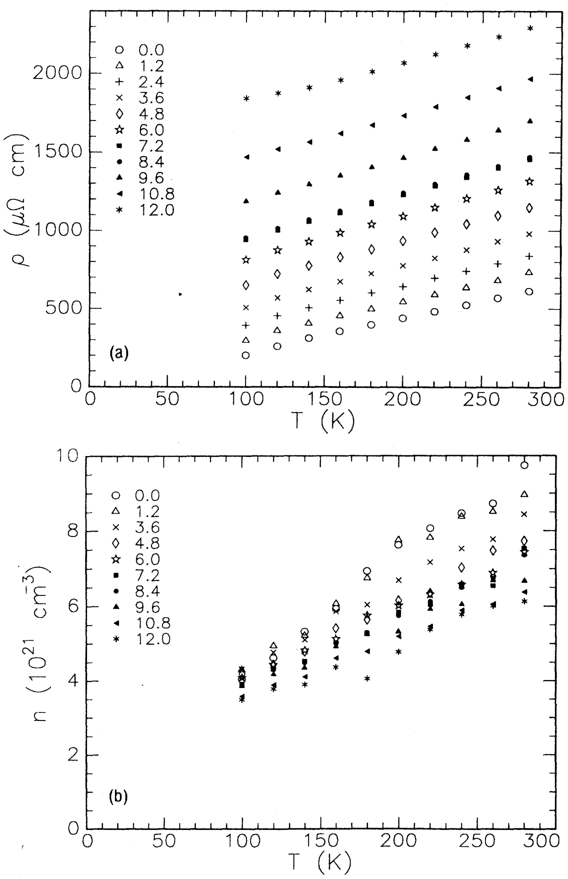

FIG. 2. Low-fluence data. (a) Resistivity vs temperature for the fluences indicated in the figure in units of 1×10^{13} ions/cm^2. (b) Carrier density (n) as a function of temperature for the same sample where n is defined as $1/(ecR_H)$.

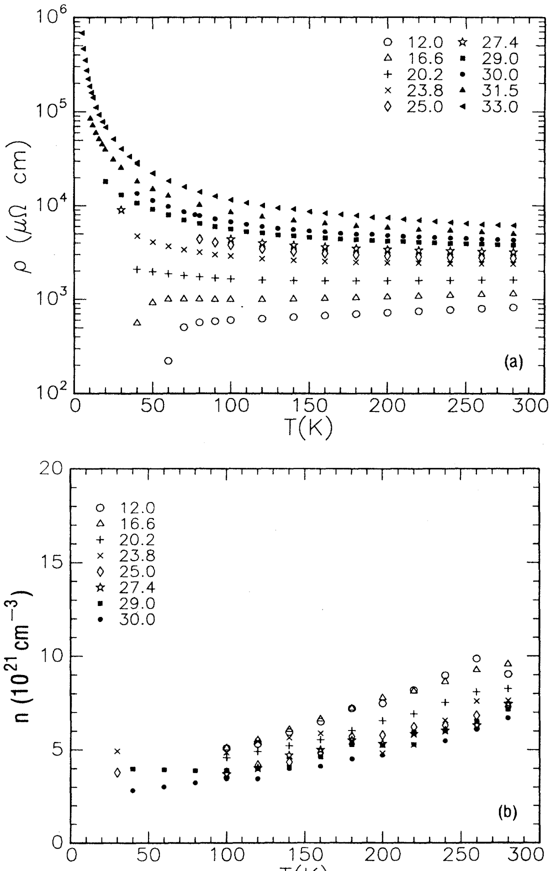

FIG. 3. High-fluence data. (a) $\log_{10}$ of the resistivity vs temperature for the fluences indicated in the figure in units of 1×10^{13} ions/cm^2. (b) Carrier density (n) vs temperature for the same sample. Note the relative insensitivity of n to damage and temperature compared to the resistivity. The slight differences in the data here compared with that in Fig. 2 are due to the fact that these are different samples. There is some variation in the values obtained for n from sample to sample (Ref. 1).

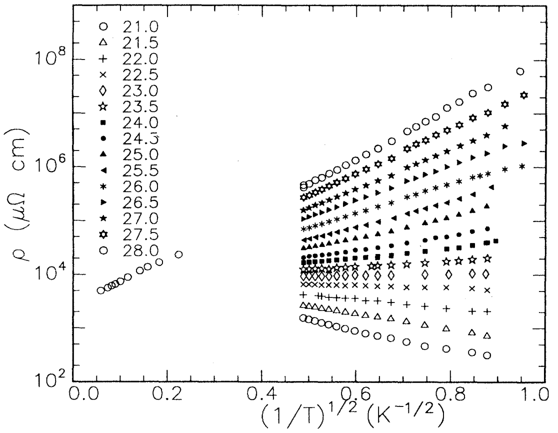

FIG. 4. $\log_{10}$ of the resistivity vs $1/T^{1/2}$ for the fluences (in units of 1×10^{13} ions/cm^2) indicated in the figure. At the highest fluence, the data fit $\exp(\Delta/T^{\alpha})$ with $\alpha=\frac{1}{2}$ much better than $\alpha=1$ or $\frac{1}{4}$.

insulating. On the other hand, $1/R_H$ [shown in Fig. 3(b)] approaches a constant value at the higher ion fluences which is not very different from the value obtained in the undamaged material just above T_c. Perhaps this constant value is the true carrier density in the material and the linear temperature dependence in the undamaged case is associated with the peculiar nature of these carriers. The slight differences between Figs. 2 and 3 are due to the fact that these data represent different samples. It is known that there is some variation in the value obtained for n from sample to sample.

It is important to reemphasize at this point that the combined data of Figs. 3(a) and 3(b) suggest that the material is insulating because of a sharply reduced carrier mobility rather than carrier freeze-out. In other words, although carrier density is almost independent of temperature, the resistivity (at 50 K, for instance) is increasing exponentially with ion fluence and decreasing temperature.

III. A MODEL AND DISCUSSION

We propose a simple model to describe the results —namely, the tracks of the incident ions create regions in the $YBa_2Cu_3O_{7-\delta}$ which are insulating. This insulating volume no longer contributes to the conductivity of the sample, so the resistance (and hence the apparent resistivity) increases. By measuring the change in slope $d\rho/dT$ in Fig. 2(a) and assuming the change is due to a reduced volume of conductor, the volume of insulating material can be estimated in the following way. If each ion leaves a cylindrical damage track of volume $\pi r_0^2 t$, where r_0 is the cylinder radius and t the film thickness, the fraction of material made insulating is given by $f=\pi r_0^2 t\times$fluence. From the data in Fig. 2(a) we find that for a fluence of 6×10^{13} ions/cm^2, $d\rho/dT$ increases by about 30% (i.e., 30% of the material is now insulating), giving a track radius of $\sim$4 Å.

This picture of an 8-Å-diam track is consistent with our earlier picture of the damage dependence of the critical-current density J_c in these materials.[3] There it was shown that at a fluence of $\sim10^{13}$ ions/cm^2 the critical current dropped markedly. By simple geometry the mean separation of the damage tracks at that fluence is $\sim$30 Å which is approximately the superconducting coherence length. At higher fluences, the insulating tracks occur at even higher densities and eventually overlap, at which point flux pinning is greatly reduced.

Applying this picture to higher fluences, we expect the damage tracks to overlap at a fluence of about (1.5–2.0) $\times10^{14}$ ions/cm^2. From Fig. 1, we see that it is just this regime where the resistivity is changing from metallic to insulatinglike behavior. Moreover, with only a small additional fluence, the film is clearly in the insulating state. It is encouraging that the same simple picture of insulating ion tracks which we used to describe the change in slope of $\rho(T)$ at low fluences is also capable of consistently explaining the crossover to insulating behavior at much higher fluences.

As an additional test of the consistency of the model, we can ask whether it is capable of describing the increase in ρ_0 with fluence. Assuming that the conduction holes are scattered from the same insulating cylinders of radius $\sim$4 Å, the mean free path λ can be determined from $\lambda=1/\phi\sigma$, where ϕ is the fluence and σ the scattering cross section (or diameter). If we assume a reasonable effective mass and Fermi velocity, we obtain a value for ρ_0 that is within a factor of 2 of the measured one. This agreement is remarkable in view of the overly simple assumptions and lends credibility to this model.

This model does *not* explain the temperature dependence of the Hall coefficient, but it can account for some aspects of the Hall results. The relative insensitivity of carrier density to damage is consistent with our model of the ions creating insulating regions in the films but not severely modifying the conducting portions. It is difficult, however, to understand the temperature dependence of $1/R_H$ and the change in that slope with increasing damage. As we mentioned, Stormer *et al.* have suggested a two-band model with light and heavy holes to account for the temperature dependence of $1/R_H$ and have argued that magnetoresistance measurements cannot rule out this picture.[1] One possibility is that disorder caused by the ion radiation could result in a broadening of the narrow band postulated in the two-band model, thereby producing the observed reduction in the temperature dependence of R_H.

We can only speculate on the microscopic nature of the damage induced defects responsible for this behavior. It is well known that the superconducting (and metallic) behavior of $YBa_2Cu_3O_{7-\delta}$ is very sensitive to the value of δ.[6] Removing oxygen from the copper-oxygen chains (δ increasing from $0\rightarrow1$) results in the destruction of superconductivity and, subsequently, the transition to insulating behavior. The oxygen in the chains is the most weakly bound species and is easily removed by thermal treatment,[7] so it seems plausible that this oxygen could be easily displaced from the chain site on, say the b axis, to the equivalent interstitial location on the a axis, creating a microtwin, and profoundly affecting the conduction properties. Such a point defect would be extremely difficult to observe directly using transmission electron microscopy or x rays and would, of course, easily anneal out. A signature of this selective defect could possibly be a progressive decrease in the orthorhombicity of the structure. We would expect eventually that when there is complete disorder in the chain oxygens that the structure would revert to tetragonal.

In the insulating regime, a temperature-dependent resistivity of the form given in Eq. (2) has been previously observed in several granular bulk and thin-film systems (cermets) and has motivated a variety of explanations.[8] One particularly appealing explanation for such behavior is based on Mott's variable-range hopping model[9] for a disordered system with an energy-dependent density of states. The generalized Mott argument[10] yields a resistivity given by $\rho=\rho_0\exp[+(2\Delta/T)^{\alpha}]$, where $\alpha=(n+1)/(n+4)$.[11] The value of n comes from the energy dependence of the density of states around the Fermi level: $N(E)\alpha(E-E_F)^n$. A constant density of states recovers the original Mott result, $\alpha=\frac{1}{4}$, while an Efros and Shklovskii "soft" gap ($n=2$) results in the observed $\alpha=\frac{1}{2}$. One might expect in the vicinity of the metal-insulator transition a region where $\alpha=\frac{1}{4}$, but careful inspection of the data indicates that the best fit indicates $\alpha=\frac{1}{2}$ over the entire range of the data presented here. However, the Hall measurements suggest that the carrier density does not change much with increasing damage. For the Coulomb-gap explanation to be valid, the majority of carriers measured in the Hall measurements must be ineffective in contributing to the conductivity. Indeed, this is quite plausible in view of the results of thermodynamic measurements (heat capacity, for instance[12]) on the insulating side of a metal-insulator transition which persist in measuring a large number of carriers. Presumably these carriers are localized and contribute to the heat capacity and Hall coefficient at finite temperatures but, because of their extremely low mobility, contribute very little to the conductivity. We caution however, that we do not treat these results as proof of this generalized variable range hopping with a Coulomb gap. In fact, the general validity of this model has been seriously questioned.[13]

Another model to explain this temperature dependence invokes the granular nature of a cermet and suggests that a distribution of charging energies due to a variation in grain sizes affects carrier transport. Such a model can indeed lead to an $\alpha=\frac{1}{2}$ behavior but again implies a density-of-states or carrier-density effect. While the $YBa_2Cu_3O_{7-\delta}$ films used in this study are not granular in the traditional sense, we have proposed a model of radiation-induced insulating regions in a metallic matrix with a spacing less than the superconducting coherence length to explain our results. It is not difficult to imagine that at some point these insulating regions will percolate.

IV. SUMMARY

We have studied the metal-insulator transition in epitaxial thin films of $YBa_2Cu_3O_{7-\delta}$ using ion-beam irradiation. Resistivity and Hall measurements have indicated that, in the course of this evolution, the carrier density does not change enough to account for the change in ρ, implying that the transition is driven by a reduction in carrier mobility. In the insulating region, the resistivity varies like $\exp(T^{-1/2})$. This dependence can be obtained within a Mott variable-range hopping model with an Efros and Shklovskii soft Coulomb gap that varies as E^2, but the Hall measurements show that the apparent carrier density remains high into the insulating region. Finally, a simple model of the ion damage in which insulating tracks result from disorder on the oxygen lattice gives a quantitatively consistent picture of the temperature-dependent resistivity, the residual resistivity, the previously observed reduction in J_c, and the crossover to insulating behavior.

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