Electrostatic Control of the Evolution from a Superconducting Phase to an Insulating Phase in Ultrathin $YBa_2Cu_3O_{7-x}$ Films

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The electrical transport properties of ultrathin $YBa_2Cu_3O_{7-x}$ films have been modified using an electric double layer transistor configuration employing an ionic liquid. A clear evolution from superconductor to insulator was observed in nominally 7 unit-cell-thick films. Using a finite size scaling analysis, curves of resistance versus temperature, $R(T)$, over the temperature range from 6 to 22 K were found to collapse onto a single scaling function, which suggests the presence of a quantum critical point. However, the scaling fails at the lowest temperatures indicating the possible presence of an additional phase between the superconducting and insulating regimes.

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Superconductor-insulator transitions at zero temperature induced by varying external parameters such as thickness, magnetic field, and carrier concentration are examples of quantum phase transitions (QPTs) [1,2]. A signature of a QPT at nonzero temperature is the success of finite size scaling in describing the data [3–5]. Basically, the resistance of a 2D system near a quantum critical point (QCP) collapses onto a single scaling function $R = R_c f(\delta T^{-1/\nu z})$, where R_c is the critical resistance, δ is tuning parameter, T is the temperature, ν is the correlation length critical exponent, and z is the dynamic critical exponent [6–9].

For high-T_c cuprates, such as $YBa_2Cu_3O_{7-x}$ (YBCO), since thickness cannot be varied continuously [10–12], and the upper critical field H_{c2} is huge [13–15] tuning carrier concentration would be the most practical approach. There are two ways to do this, by chemical doping or by electrostatically charging. The disadvantage of the former is that it can introduce structural or chemical changes and cannot be tuned continuously [16]. On the other hand, electrostatic charging is promising since it keeps the structure fixed and it is continuous and reversible [17–20]. Moreover, a large effect is expected in high-T_c cuprates due to their much lower carrier densities compared with those of conventional metals. However, most electrostatic charging experiments with cuprates show only small changes in T_c [21–23] suggesting that the situation is more complicated.

Recently, ionic liquids have been used to make field-effect-transistor-like devices to induce carrier concentration changes up to 8×10^{14} cm^2 in materials such as ZrNCl, $SrTiO_3$ (STO), ZnO, and YBCO [24–27]. With such high carrier concentration, superconductivity was induced in ZrNCl and STO samples and a T_c shift of 50 K was observed in YBCO thin films. However, no continuous transition from superconductor to insulator has thus far been found in YBCO. The reason is that films as thin as 10 unit cells (UC) are still too thick. The Thomas-Fermi screening length in YBCO is short, typically several angstroms. Thus the order of only one UC will be affected by gating. In a thick sample, the proximity effect will dominate and no transition can be seen. In a recent paper, Bollinger *et al.* reported superconductor-insulator transition in $La_{2-x}Sr_xCuO_4$ (LSCO) using an electric double-layer transistor (EDLT) configuration [28]. Compared to LSCO, YBCO has a different crystal structure and chemical disorder which has important consequences. First, YBCO has CuO_x chains and previous work using STO as a gate dielectric indicates that most of the induced carriers may reside on the CuO_x chains while the CuO_2 planes are only indirectly affected [29]. Second, disorder in YBCO is limited to oxygen defects and there is no presence of the characteristic A type disorder of LSCO, resulting in a much higher spatial homogeneity for YBCO [30,31]. Thus, the properties of the superconductor-insulator transition in these two systems may be expected to be very different since the nature of the disorder may affect the character of electron localization. Here we report a clear electrostatically induced transition between superconducting and insulating behavior in YBCO. By means of finite size scaling analysis of different $R(T)$ curves, the presence of a QCP is suggested. Nonetheless, we noticed that there might be another phase between the superconducting and insulating regimes since the scaling fails at temperature below 6 K. Moreover, the phase diagram derived from our data shows striking similarity with bulk phase diagram.

Films were grown on (001)-oriented $SrTiO_3$ substrates using a high pressure oxygen sputtering system. Previous work has shown that this technique provides high quality epitaxial ultrathin YBCO films [12,32]. The device we are using is similar to the one shown in Fig. 1 of Ref. [25]. Amorphous Al_2O_3 films were deposited as masks, and the substrates were etched in buffered (10:1) HF solution and annealed in an O_2/O_3 mixture for 6 h at 750 °C. Atomic force microscopy (AFM) was used to make sure that the substrate surfaces were clean and were TiO_2 terminated.

The oxygen pressure during deposition was 2.0 mbar and the substrate temperature was 900 °C. After deposition, films were cooled in an 800 mbar O_2 atmosphere and annealed at 500 °C for 30 min. Films were characterized *ex situ* by AFM and x-ray diffraction. Thickness was measured using x-ray reflectivity. A series of samples with thicknesses ranging from 5 to 10 UC was fabricated and measured using standard four-probe techniques. Films thinner than or equal to 6 UC were insulating whereas thicker ones were superconducting. This suggests that the first 5 to 6 UC are insulating and that the 7 UC thick film has a superconducting layer that is actually only 1 to 2 UC thick.

Ultrathin YBCO films are sensitive and react with most chemicals so we did not carry out any processing after film growth. Films were covered with ionic liquid (DEME-TFSI) [25] and were quickly cooled down to 240 K. The ionic liquid condenses into a rubberlike state at 240 K, a temperature at which most chemical reactions are suppressed. If the device is kept at room temperature for several hours with ionic liquid on it, we see an increase in resistance and a drop in T_c, possibly due to chemical reaction. However, after cooling down and being kept below 240 K, no changes are seen over several days. For measurements, the gate voltage was changed at 240 K where it is held for 1 h before cooling down. The gate voltage was kept constant during measurements.

Resistance vs temperature, $R(T)$, curves at various gate voltages of a 7 UC thick YBCO sample are shown in Fig. 1. The sample starts as a superconductor with $T_c^{\mathrm{on}} \sim 77$ K. Here T_c^{on} is taken to be the temperature at which the sheet resistance falls to 90% of its normal value. We realize an insulator with a gate voltage (V_G) of only 1.52 V. We notice that T_c^{on} is a nonlinear function of V_G. There is a V_G threshold of about 0.3 V below which almost no change can be seen. Then, T_c^{on} drops by about 30 K as V_G increases from 0.5 to 1.1 V, which is about 5 K/0.1 V. However, as V_G increases from 1.1 to 1.25 V, a change of 0.2 to 0.3 V can induce a T_c^{on} shift of about 5 K. This nonlinearity suggests that the gate voltage cannot be used as a tuning parameter for quantitative analysis.

As V_G increases to 1.37 V, $R(T)$ curves flatten out at the lowest temperature then undergo a small upturn at $V_G = 1.40$ V, although they initially decrease as temperature decreases. Finally, $R(T)$ evolves to an insulating state as V_G increases further, as can be seen clearly in the inset of Fig. 1. Interestingly, we have found that the resistance in the insulating side up to $V_G = 1.48$ V is thermally activated with an Arrhenius type $R(T)$ relation. Above this voltage the logarithm of sheet resistance is fit by a $T^{-1/3}$ dependence consistent with 2D Mott variable range hopping. More detailed study is in progress.

The low temperature upturn behavior is similar to that observed in amorphous MoGe films and granular superconductors [4,33–36] and will dramatically affect any quantitative scaling analysis. We suggest that this is evidence of a mixed phase separating the superconducting and insulating regimes. Nevertheless, the thickness fluctuations in the single unit cell active layer could also contribute to this upturn. We also cannot rule out the possibility that we have not completely depleted the superconductivity of the underlayers.

For further quantitative analysis of the data, we need the induced carrier concentration. As discussed above, gate voltage is not simply related to the carrier concentration and the Hall resistance cannot be used because of the complicated electronic properties of YBCO. We assume that Drude behavior is found at high temperatures and take $1/R_s$ (200 K) to be proportional to the carrier concentration

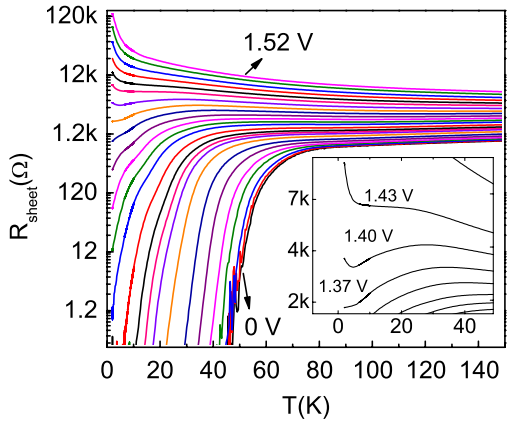

FIG. 1 (color online). Logarithm of the sheet resistance vs temperature with gate voltage varying from 0 to 1.52 V. From bottom to top: 0, 0.3, 0.5, 0.7, 0.8, 0.9, 1, 1.1, 1.12, 1.14, 1.17, 1.20, 1.25, 1.27, 1.30, 1.33, 1.35, 1.37, 1.40, 1.43, 1.44, 1.46, 1.48, 1.50, 1.52 (V). Inset: enlarged low temperature part near the transition between the superconducting and insulating regimes.

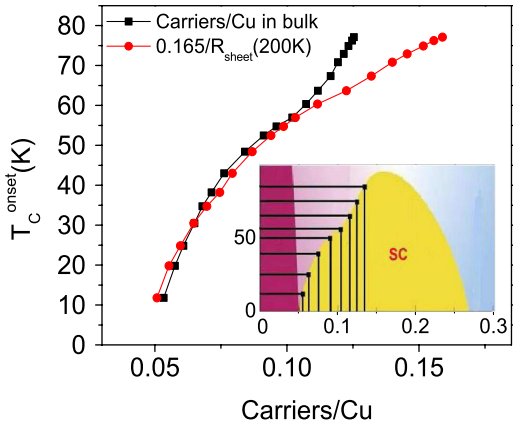

FIG. 2 (color online). Carrier concentration calculated using $0.165/R_s$ (200 K) (red dots) compared to that derived from the phase diagram of bulk high-T_c cuprates (black squares). The inset shows how the carrier concentration was determined from the bulk phase diagram using measured values of T_c^{on} at different gate voltages.

measured as carriers per in-plane Cu atom. Comparing this with the carriers per in-plane Cu atom derived from the phase diagram of bulk YBCO [37], we find that they match with a constant coefficient 0.165 as plotted in Fig. 2. This is especially true in the transition regime which is the focus of quantitative analysis.

Using measured $R(T)$ curves and calculated carrier concentrations, a phase diagram can be constructed and is plotted as Fig. 3. It is similar to the phase diagram of bulk high-T_c cuprates derived from chemical doping. Here it is measured continuously and perhaps more accurately, especially in the transition and insulating regimes. The data plotted here are in the linear regime of the I-V curves. There are nonlinear I-V curves deep in the insulating regime which are denoted by white in the figure.

When a QCP is approached, resistance isotherms as a function of the carrier concentration x should cross at the critical resistance R_c and all resistance data should collapse onto a single scaling function. Figure 4 shows isotherms from 2 to 22 K. A clean crossing point can be seen if we neglect the lowest temperature isotherms. The critical resistance per square is about 6.0 kΩ, which is very close to the quantum resistance given by $R_Q = h/(2e)^2 = 6.45$ kΩ considering that imprecise shadow masks were used to define the four-terminal configuration. The critical carrier concentration is $x_c = 0.048$ carriers/Cu, close to that derived from the bulk phase diagram (0.05). But this is not surprising given that the bulk phase diagram was used to calibrate the carrier concentration.

The results of the finite size scaling analysis are shown in the inset of Fig. 4. All the sheet resistance data from 6 to 22 K collapse onto a single function $R_s = R_c f(|x - x_c|T^{-1/\nu z})$ if $\nu z = 2.2$. Assuming $z = 1$ [38], the value $\nu = 2.2$ is close to that of the universality class of a metal-insulator transition in an anisotropic 2D system (7/3) [39] or that of the quantum percolation model (2.43) [40].

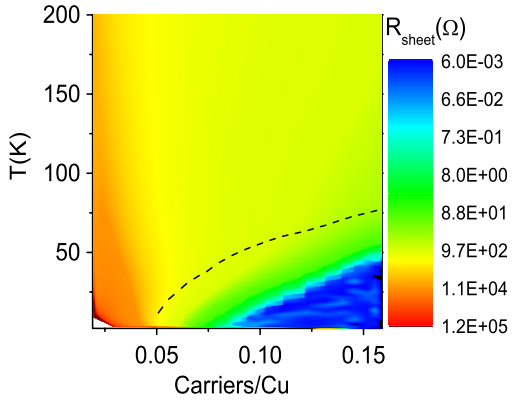

FIG. 3 (color online). Color plot of the resistance vs temperature and calculated carrier concentration. This can be interpreted as a phase diagram. Different colors represent different sheet resistances. The dash line is T_c^{on}. The white area in the bottom left-hand corner is a regime of with nonlinear I-V curves.

Previous studies of QPTs in amorphous bismuth [5] and STO [41] found $\nu z \approx 0.7$, which is compatible with the (2 + 1)D XY model. In amorphous MoGe films [3,4] $\nu z \approx 1.3$, which is consistent with classical percolation theory. The value of $\nu z = 2.2$ for YBCO is different from $\nu z = 1.5$ found for LSCO [28]. This could be a consequence of the difference in the disorder of the two systems. However, the scaling of the YBCO data breaks down below 6 K, which is a consequence of the nonmonotonic behavior of $R(T)$ at low temperature at gate voltages in the transition regime. These results suggest that the transition from superconductor to insulator is indirect and may involve an intermediate phase and possibly multiple QCPs. We would like to emphasize that this transition between superconductor and insulator would be different from the QPT inside the superconducting dome suggested to give rise to the anomalous behavior of underdoped cuprates [42].

In summary, a transition from a superconductor to an insulator has been induced in ultrathin superconducting YBCO films using an EDLT device configuration. A striking feature of the data is the similarity between the phase diagram (Fig. 3) and bulk phase diagram. This is surprising considering the fact that the active layer in our sample is only one to two UCs and that there are high electric fields in the double layer. All resistance data collapse onto a single scaling function with critical exponent $\nu z = 2.2$ except the lowest temperature part. This suggests an approach to a quantum critical point. However, new physics appears to develop at the lowest temperatures, resulting in curves similar to those found in granular superconductors. The resultant superconductor-insulator transition may not be direct.

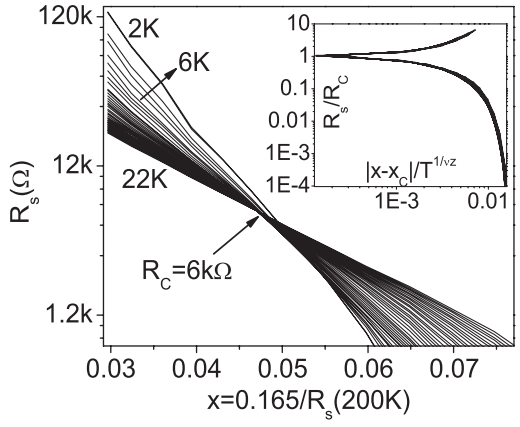

FIG. 4. Isotherms of $R(x)$ at temperatures ranging from 2 to 22 K. The carrier concentration x is calculated using the phenomenological relation $x = 0.165/R_s$ (200 K). The arrow indicates the crossing point where $R_c = 6.0$ kΩ and $x_c = 0.048$ carriers/Cu. Inset: Finite size scaling analysis of $R(T)$ for data in the range from 6 to 22 K. The calculated carrier concentration $x = 0.165/R_s$ (200 K) is used as the tuning parameter. The best collapse is found when $\nu z = 2.2$.

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