Electrostatic Control of the Evolution from Superconductor to Insulator in Ultrathin Films of Yttrium Barium Copper Oxide

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The electrical transport properties of ultrathin YBa$_2$Cu$_3$O$_{7-x}$ films have been modified using an electric double layer transistor configuration employing an ionic liquid. The films were grown on SrTiO$_3$ substrates using high pressure oxygen sputtering. 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 6K to 22K were found to collapse onto a single scaling function, which suggests the presence of a quantum critical point. However the scaling failed at the lowest temperatures suggesting the presence of an additional phase between the superconducting and insulating regimes.

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

For high-$T_c$ cuprates, such as YBa$_2$Cu$_3$O$_{7-x}$ (YBCO), since thickness cannot be varied continuously and the upper critical field $H_{c2}$ is huge 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.

On the other hand, electrostatic charging is promising since it keeps the structure fixed and it’s continuous and reversible. 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 $T_c$, suggesting that the situation is more complicated. Work using SrTiO$_3$ (STO) as the dielectric indicates that although the carrier concentration is changed by the order of $10^{13}$ cm$^{-2}$, this change is not large enough to traverse from insulator to superconductor, as the induced carriers may reside on the CuO$_2$ chains while the CuO$_2$ planes are only indirectly affected.

Recently, ionic liquids have been used to make FET-like devices to induce large carrier concentration changes in inorganic materials such as ZrNCl, STO, ZnO and YBCO. The devices are electric double layer transistors (EDLTs), since in response to the electric field, ions are driven to sample surfaces to form an interfacial double layer with the induced carriers. The gap between these two layers is of the order of nanometers. The capacitance is huge and the induced sheet carrier density can be as high as $8 \times 10^{14}$ cm$^{-2}$. With such high carrier concentration, superconductivity was induced in ZrNCl and STO samples and a $T_c$ shift of about 50K was observed in YBCO thin films. However, no continuous transition from insulator to superconductor 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 unit cell (UC) will be affected by gating. In a thick sample, the proximity effect will dominate and no transition will be seen. Recently, Bollinger et al. reported superconductor-insulator transition in a 214 compound La$_{2-x}$Sr$_x$CuO$_4$ using an EDLT configuration. Here we report a clear electrostatically...
induced transition between superconducting and insulating behavior in YBCO, which is a 123 compound. The 214 and 123 compounds have different structures.

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. A sketch of the device is shown in Fig. 1. Amorphous Al$_2$O$_3$ 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 hours 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 minutes. 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 were 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 didn’t carry out any processing after film growth. Films were covered with ionic liquid (IL) and were quickly cooled down to 240K. The IL used was N,N-diethly-N-(2-methoxyethyl)-N-(methylammonium bis(trifluoromethylsulphonyl)-imide, (DEME-TFSI). This condenses into a rubber-like state at 240K, 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 to 240K and being kept below this temperature, no changes are seen over several days. For measurements, the gate voltage was changed at 240K where it is held for 1 hour 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. 2. The sample starts as a superconductor with $T_c^{onset}$ ~ 77K. Here $T_c^{onset}$ 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 1.52V. We notice that $T_c^{onset}$ is a nonlinear function of $V_G$. There is a $V_G$ threshold of about 0.3V below which almost no change can be seen. Then, $T_c^{onset}$ drops by about 30K as $V_G$ increases from 0.5V to 1.1V, which is about 5K/0.1V. However, as $V_G$ increases from 1.1V to 1.25V, a change of 0.2V to 0.3V can induce a $T_c^{onset}$ shift of about 5K. This nonlinearity suggests that the gate voltage cannot be used as a tuning parameter for quantitative analysis.

As $V_G$ increases to 1.37V, $R(T)$ curves flatten out at the lowest temperature then undergo a small upturn at $V_G$ =1.40V 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. 2. This low temperature behavior is similar to that observed in amorphous MoGe films and granular superconductors and will dramatically affect any quantitative scaling analysis. We suggest that this is evidence of a mixed phase separating the superconducting and insulating regimes.

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 can’t be used because of the complicated electronic properties of YBCO. We assume that Drude behavior is found at high temperatures and take $1/R_{\text{sheet}}(T=200K)$ to be proportional to the carrier concentration 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, we find that they match with a constant coefficient 0.165 as plotted in Fig. 8. This is especially true 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 quantum critical point 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 5 shows isotherms from 2K to 22K. A clean crossing point can be seen if we neglect the lowest temperature isotherms. The critical resistance per square is about 6kΩ, which is very close to the quantum resistance given by $R_Q = h/(2e)^2 = 6.45kΩ$ 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 general 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 Fig. 6. All the sheet resistance data from 6K to 22K collapse onto a single function $R_{\text{sheet}} = R_c f(|x - x_c|/\nu z)$ if $\nu z = 2.2$. Assuming $z=1$, the value $\nu = 2.2$ is close to the universality class of metal-insulator transition in anisotropic 2D system $(7/3)$ or quantum percolation model (2.43). Scaling breaks down for data below 6K, which is not surprising due to the nonmonotonic behavior of $R(T)$ at low temperature at gate voltages in the transition regime.

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. 4) and bulk phase diagram. This surprising considering the fact that the active layer

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**FIG. 4:** (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 $T_{\text{onset}}$. The white area in the bottom left hand corner is a regime of with nonlinear I-V curves.

**FIG. 5:** Isotherms of $R(x)$ at temperatures ranging from 2K to 22K. The carrier concentration $x$ is calculated using the phenomenological relation $x=0.165/R_{\text{sheet}}(200K)$. The arrow indicates the crossing point where $R_c=6.0kΩ$ and $x_c=0.048$ carriers/Cu.

**FIG. 6:** Finite size scaling analysis of $R(T)$ using the calculated carrier concentration $x=0.165/R_{\text{sheet}}(T = 200K)$ as a tuning parameter. Only data in the range from 6K to 22K are shown here and the best collapse is found when $\nu z = 2.2$. 

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 develops at the lowest temperatures, resulting in curves similar to those found in granular superconductors. The resultant SI transition is not direct.

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