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Abstract

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MATTHEY, Daniel, GARIGLIO, Stefano, TRISCONE, Jean-Marc. Field-effect experiments in $NdBa_2Cu_3O_{7-\delta}$ ultrathin films using a $SrTiO_3$ single-crystal gate insulator. *Applied Physics Letters*, 2003, vol. 83, no. 18, p. 3758-3760

DOI : 10.1063/1.1624635

APPLIED PHYSICS LETTERS VOLUME 83, NUMBER 18 3 NOVEMBER 2003

Field-effect experiments in $NdBa_2Cu_3O_{7-\delta}$ ultrathin films using a $SrTiO_3$ single-crystal gate insulator

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(Received 23 June 2003; accepted 12 September 2003)

We report on the electrostatic modulation of superconductivity in very thin films of cuprate superconductors using a field-effect device based on a $SrTiO_3$ single-crystal gate insulator. A T_c modulation of 3.5 K and a 37% change of the normal state resistance have been observed in an epitaxial bilayer composed of an insulating $PrBa_2Cu_3O_{7-\delta}$ layer deposited on top of a superconducting $NdBa_2Cu_3O_{7-\delta}$ film, two unit cells thick. To achieve large electric fields, the thickness of the commercial dielectric single-crystal $SrTiO_3$ substrate (also used as the gate insulator) was reduced to 110 μm. The dielectric properties of the gate insulator were characterized as a function of temperature and electric field and the magnitude of the field effect was quantified. A T_c enhancement of 2.8 K was obtained for an applied field of -1.8×10^6 V/m, corresponding to a polarization of -4 μC/cm^2. [DOI: 10.1063/1.1624635]

Field-effect experiments in high-temperature superconductors (HTS) have been intensively studied in past years.[1,2] These doped Mott insulators are particularly attractive for field-effect experiments because of their relatively low carrier density (1–2 orders of magnitude smaller than metals), and also because their superconducting properties depend markedly on the carrier density. The electric field effect allows the carrier density to be modulated without modification of the chemical composition or structure. In the simplest geometry, an electric field applied between two electrodes (one of them being the superconducting film of interest), across a dielectric, will induce a charge redistribution necessary to screen the external electric field inside the electrodes. Field-effect devices (FEDs) using a $SrTiO_3$ (STO) thin film as the dielectric gate insulator and ferroelectric FEDs have been extensively studied with reports of substantial modulations of T_c and J_c.[1–12] STO is indeed an interesting candidate as a dielectric since it has a large dielectric constant ϵ[13–15] at low temperatures and because of its good lattice match to HTS such as $YBa_2Cu_3O_{7-\delta}$. The polarization for an ideal parallel plate capacitor is $\epsilon\epsilon_0 V/d$, where d is the thickness of the dielectric, V the applied voltage, and ϵ_0 the vacuum permittivity. For a given voltage, substantial polarizations will be obtained if large dielectric constant materials and small thicknesses are used. The maximum charge density that can be transferred for a given dielectric is proportional to ϵE_{BD}, where E_{BD} is the breakdown field. The latter has been shown, for some materials, to scale with the inverse of ϵ at room temperature.[16] In this letter, we report on experiments using a FED based on a thinned $SrTiO_3$ single-crystal substrate as a gate dielectric, and show that this approach can lead to substantial modulation of T_c and resistivity in a bilayer made of an insulating $PrBa_2Cu_3O_{7-\delta}$ (PBCO) and a superconducting $NdBa_2Cu_3O_{7-\delta}$ (NBCO) thin films. The idea to thin down the thickness of a single-crystal substrate was explored by Nakajima *et al.* in STO bicrystals, leading to a 5% change in the resistance of a $YBa_2Cu_3O_{7-\delta}$ grain boundary junction.[9] Applied here to a conventional FED this approach seems very promising, since it can be used to perform field-effect experiments on the many different interesting oxide compounds that can be epitaxially grown on STO.

The device structure we used is shown in the inset of Fig. 1. The NBCO and PBCO thin films were first grown by off-axis magnetron sputtering onto (100) STO substrates. The growth temperature was typically 700 °C and the Ar $+O_2$ sputtering pressure was 0.15 Torr (O_2/Ar$=$0.1). In the experiments reported here, the thickness of the NBCO layer was 24 Å and the thickness of the PBCO layer 550 Å. The bilayer was protected by an amorphous insulating PBCO layer deposited *in situ*. After deposition, the sample was photolithographically patterned using ion milling. The superconducting path was located in the middle of the sample. Finally, the thickness of the STO single crystal was reduced down to 110 μm using a dimpling grinder (model 150) from Fischione instruments. The sample was first glued on a rotating plate and thinned down by using a 10-mm-radius rotating wheel with diamond paste. The superconducting path was located in the region of minimal thickness. We estimated the

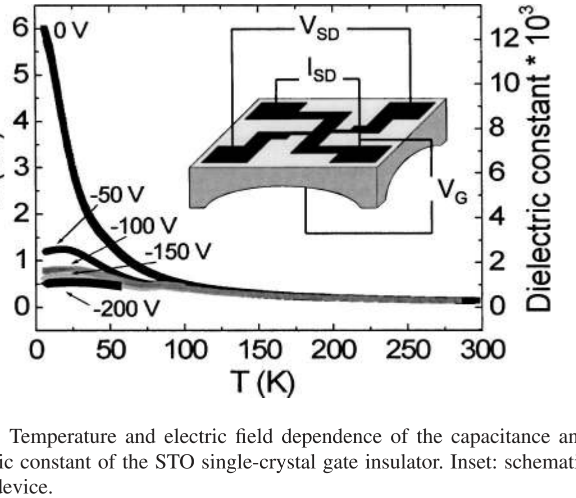

FIG. 1. Temperature and electric field dependence of the capacitance and dielectric constant of the STO single-crystal gate insulator. Inset: schematic of the device.

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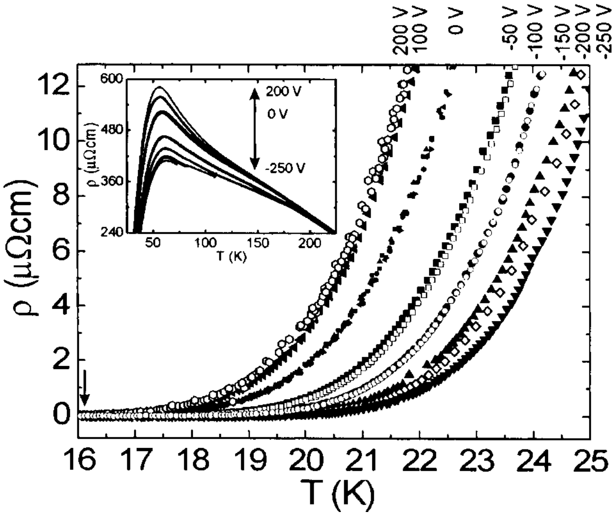

FIG. 2. Resistivity as a function of temperature of the PBCO/NBCO heterostructure close to the foot of the transition and over the whole temperature range (inset) for different applied fields across the gate dielectric. The resistivity was calculated using the thickness of the NBCO layer. The arrow indicates the value of T_{KT} for zero applied field.

maximal thickness difference over a 1 mm^2 region to be of the order of 12 μm, a value confirmed by direct optical measurements. Fluctuations of the electric field over the measured superconducting path due to thickness variations thus did not exceed 10%. The last step was the deposition of a gold electrode on the back of the sample, in the ground-down region (not shown in the inset of Fig. 1).

Figure 1 shows the typical temperature and electric field dependence of the dielectric constant of the STO single-crystal gate insulator. The capacitance of the thinned STO single crystal was directly measured using an Agilent 4284A LCR meter as a function of temperature and (static) electric field. The frequency of the ac signal used for the capacitance measurements was 1 kHz. The static electric field was applied using a Kepco voltage source. The dielectric constant versus temperature curve was obtained using the temperature dependence of the capacitance.[17] The zero-field value of the dielectric constant is found to be about 12 500 at 4.2 K. The temperature dependence of the capacitance of the reduced STO single crystal shows a Curie–Weiss dependence $C = C_0/(T-T_0)^{\gamma}$, with $T_0=40$ K and $\gamma=1$ down to 109 K as previously observed.[13] This temperature is associated with the cubic–tetragonal structural phase transition of STO. Applying an electric field across the STO single crystal hardens the optical phonon mode and thus drastically reduces the dielectric constant. The dielectric properties of the device were measured up to -200 V, the limit of the LCR meter. The polarization P was calculated using the measured value of the capacitance and the geometry of the device. The "breakdown field" E_{BD} at a given temperature was estimated by measuring the field at which an abrupt rise in the leakage current through the dielectric was observed.[18] We found that ϵE_{BD} varies between -2.7×10^{8} V/m at 292 K and -1.7×10^{9} V/m at 82 K. Between each measurement at a given voltage, a measurement without any applied electric field was performed to check for reproducibility. We note that at low temperatures, the maximum measured polarization is -4 μC/cm^2 for an applied voltage of -200 V.

In Fig. 2, the resistivity of the PBCO/NBCO bilayer, measured between the source and drain contacts, is shown as a function of temperature for different voltages applied across the 110-μm-thick gate dielectric. The resistivity measurements were performed using a standard four-point technique with a source–drain current of typically 10 μA, and gate leakage currents below 1 nA. Negative voltages correspond to hole doping of the sample, resulting, as expected, in a decrease of the resistivity and an increase of the critical temperature. At high temperatures, one notices that the resistivity increases while decreasing the temperature. This is a signature of the thick PBCO layer whose semiconductor-like resistivity is dominant. At high temperatures, the change in the resistivity as a function of the applied voltage is small because of the low STO dielectric constant and the relatively high conductivity of PBCO. At lower temperatures, the NBCO layer becomes superconducting and the resistivity of the bilayer goes to zero. The maximum change in the resistivity is about 37% and occurs just above the superconducting transition. Between each measurement at a given voltage, a measurement at zero voltage was carried out. All the curves at zero voltage collapse, as can be seen in Fig. 2. To demonstrate the reproducibility of the data, two different measurements at -50 and at -100 V are shown on the graph. At low temperatures, the reproducibility is excellent. At higher temperatures, in the normal state, drifts in the resistivity for a given voltage of up to 1.5% have been observed (not shown on the figure). By defining T_{c0} as the temperature at which $R=0.1$ Ω, the maximum change in T_{c0} is 3.5 K and the maximum T_{c0} enhancement is 2.8 K. We note that such large shifts have been reported in the literature, but for applied fields reducing the hole density, thus resulting in a T_c reduction.[8,19]

Because STO is piezoelectric, a concern is that the deformation of the cell could lead to modifications of T_c. We estimated the maximum change in the critical temperature of the NBCO layer which could be induced by a change of the lattice parameters due to the applied fields used in this study. Applying an electric field along the c axis of the STO crystal will induce an in-plane mechanical strain. The critical temperature of $\mathrm{Re\,Ba_2Cu_3O_7}$ has been shown to depend upon uniaxial pressures.[20–22] Strain ε can be related to the piezoelectricity of the STO using $\varepsilon_{ij}=d_{kij}E_k$, where d_{kij} is the piezoelectric coefficient and E_k the applied electric field. Strain can then be converted to a pressure σ using $\sigma_{ik}=C_{iklm}\varepsilon_{lm}$ where C_{iklm} is the elastic coefficient of the NBCO layer. The piezoelectric constant d_{caa} was found to be as large as 100×10^{-12} m/V for $T\sim20$ K.[23] For an applied field of -1.8×10^{6} V/m the maximum T_c change is $\Delta T_c=0.05$ K using the values of the elastic coefficients of $\mathrm{YBa_2Cu_3O_7}$ (Refs. 21 and 24) and the T_c dependence on pressure for $\mathrm{GdBa_2Cu_3O_7}$ (see Ref. 20) and $\mathrm{YBa_2Cu_3O_7}$,[22] suggesting that a piezoelectric effect cannot explain the T_c changes observed.

To further analyze the data, we consider the phase fluctuation scenario proposed by Emery and Kivelson.[25] In their model, classical fluctuations give an upper limit for the critical temperature (the temperature at which long-range phase coherence is established) in the underdoped region of the phase diagram. This upper limit is related to the physics of

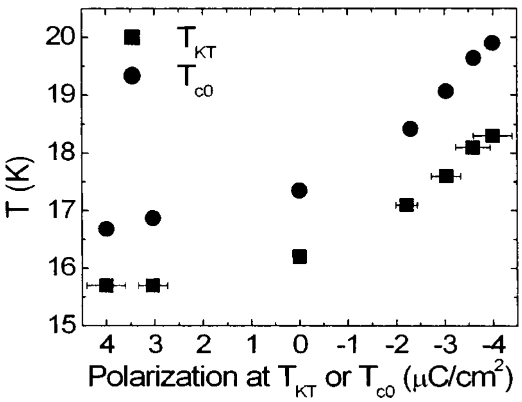

FIG. 3. Critical temperature T_{KT} (the Kosterlitz–Thouless temperature), and T_{c0}, the temperature at which $R=0.1\ \Omega$ of the NBCO layer as a function of the measured polarization at T_{KT} or T_{c0}.

the XY model and is proportional to the superfluid density. We thus used a Kosterlitz–Thouless analysis of the tail of the resistive transitions to extract $T_c(=T_{\mathrm{KT}})$ and compared our results with this theoretical prediction.[26] If we assume that the change in the superfluid density is proportional to the change in the number of carriers, one would expect a linear relation between T_{KT} and Δn_s in the underdoped region, and thus, between T_{KT} and the polarization. Figure 3 shows the critical temperature T_{KT} as a function of the measured polarization at T_{KT}. An essentially linear increase of T_{KT} as a function of polarization is observed, a result in agreement with the fluctuation scenario discussed earlier. T_{c0}, as defined earlier, is also shown in Fig. 3 as a function of the polarization at T_{c0}, and displays a behavior very similar to that of T_{KT}; however, with a larger slope. An asymmetry between the + and − polarity of the applied field is also observed. Our data suggest that it is easier to increase T_c than the opposite. A possible explanation is related to the short Thomas–Fermi screening length. Applying a negative voltage increases the hole density at the interface, eventually leading to an increased T_c. Applying a positive voltage reduces the number of carriers at the interface. However, since T_c here is determined resistively, a more favorable conducting path across the sample, away from the interface, will not be affected by the electric field, potentially reducing the observed effect.

The authors would like to thank J. Mannhart and C. Berthod for stimulating discussions, P. Stadelmann and D. Laub for their help in the early stage of the project, R. Cartoni and D. Chablaix for valuable technical help, and P. Paruch for a careful reading of the manuscript. This work was supported by Swiss National Science Foundation through the National Center of Competence in Research, "Materials with Novel Electronic Properties, MaNEP" and division II, NEDO, and ESF (Thiox).

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