Magnetic field, frequency and temperature dependence of complex conductance of ultrathin $\text{La}_{1.65}\text{Sr}_{0.45}\text{CuO}_4/\text{La}_2\text{CuO}_4$ films and the organic superconductors κ - $(\text{BEDT-TTF})_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$

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We used atomic-layer molecular beam epitaxy to synthesize bilayer films of a cuprate metal ($\text{La}_{1.65}\text{Sr}_{0.45}\text{CuO}_4$) and a cuprate insulator (La_2CuO_4), in which interface superconductivity occurs in a layer that is just one-half unit cell thick. We have studied the magnetic field and temperature dependence of the complex sheet conductance, $\sigma(\omega)$, of these films, and compared them to κ - $(\text{BEDT-TTF})_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ single crystals. The magnetic field H was applied both parallel and perpendicular to the 2D conducting layers. Experiments have been carried out at frequencies between 23 kHz and 50 MHz using either two-coil mutual inductance technique, or the LC resonators with spiral or rectangular coils. The real and the imaginary parts of the mutual-inductance $M(T, \omega)$ between the coil and the sample were measured and converted to complex conductivity. For H perpendicular to the conducting layers, we observed almost identical behavior in both films and κ - Br single crystals: (i) the transition onset in the inductive response, $L_k^{-1}(T)$ occurs at a temperature lower by 2 K than in $\text{Re } \sigma(T)$, which shows a peak close to the transition, (ii) this shift is almost constant with magnetic field up to 8 T; (iii) the vortex diffusion constant $D(T)$ is not linear with T at low temperatures as in the case of free vortices, but is rather exponential due to pinning of vortex cores. These features are absent for parallel orientation of the dc magnetic or the high-frequency (RF) field. These results can be described by the extended dynamic theory of the Berezinski-Kosterlitz-Thouless (BKT) transition and dynamics of bound vortex-antivortex pairs with short separation lengths.

1. Background

Ultrathin films of high-temperature superconductors have been studied intensely, with numerous attempts to observe the Berezinski-Kosterlitz-Thouless (BKT) transition (see references in [1]). This has been a matter of substantial debate, though [1]. It was noted that a precondition [2] for the BKT transition to occur in a superconductor, i.e., that the sample size $L_s < \lambda_{eff}$, where $\lambda_{eff} = 2\lambda^2/d$ is the effective (Pearl) penetration depth and d is the film thickness, is not satisfied even in the thinnest possible (one unit cells, 1 UC thick) $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) films. Hence, it is

still controversial not only whether the true BKT transition has ever been really observed in cuprates in the dc transport data, but even whether it can occur at all.

On the other side, Minnhagen [2] argued that although the usual BKT transition is not present when $L_s > \lambda_{eff}$, it is still possible to observe at high frequencies the BKT-like response from the bound pairs such that the vortex separation length $r < \lambda_{eff}$. According to the *BKT* theory extended to finite frequencies [2-6], higher-frequency currents sense vortex-antivortex pairs of smaller separations. At high frequency, the electromagnetic response of a 2D superconductor is dominated by those bound pairs that have $r \sim l_w$, where $l_w = (14D/\omega)^{1/2}$ is the vortex diffusion length and D is the vortex diffusion constant. We estimate that $l_w < 1 \mu\text{m}$, which is much less than $\lambda_{eff} \sim 40 \mu\text{m}$ as found in *1 UC* thick YBCO films. This implies that it should be possible to detect the response of vortex-antivortex pairs with short separation lengths at radio (*RF*) and microwave frequencies in ultrathin cuprate films, even though the usual *BKT* transition cannot be seen in DC and low-frequency measurements.

Recently, we observed large shift ($\Delta T_c = 4\text{K}$) between the onsets of the superconducting transition observed in the real part of complex sheet conductance, $Re\sigma(\omega, T)$, and in the imaginary part, $Im\sigma(\omega, T)$, in ultrathin (1 UC to 3 UC thick) YBCO films sandwiched between semiconducting $\text{Pr}_{0.6}\text{Y}_{0.4}\text{Ba}_2\text{Cu}_3\text{O}_{7-x}$ layers, at radio frequencies [8], and in bilayers of a cuprate metal ($\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4$) and a cuprate insulator (La_2CuO_4) film, ($\Delta T_c = 2\text{K}$) [1]. Here, we report new data on the temperature dependence of $\sigma(\omega)$ in $\text{La}_{1.55}\text{Sr}_{0.45}\text{CuO}_4/\text{La}_2\text{CuO}_4$ heterostructures (*LSCO/LCO* in what follows) with $T_c = 36 \text{ K}$, significantly improved compared to the earlier study ($T_c = 21 \text{ K}$) [1]. While neither of the two constituent materials is superconducting per se, the *LSCO/LCO* bilayers show interface superconductivity [9], which has been proven to reside in a single CuO_2 layer, about 0.2 nm thick [10]. Our new results presented below indicate that such bilayers are a very good 2D model system for studying the physics of BKT transition. We also report the temperature dependence of $Re\sigma(\omega, T)$ and $Im\sigma(\omega, T)$ for 2D organic single crystals $\kappa\text{-(BEDT-TTF)}_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$, for comparison.

2. Experimental

We have grown bilayer *LSCO/LCO* films in a unique atomic-layer-by-layer molecular beam epitaxy (ALL-MBE) system [11] that incorporates *in situ* surface science tools such as time-of-flight ion scattering and recoil spectroscopy (TOF-ISARS) and reflection high-energy electron diffraction (RHEED). ALL-MBE system enables synthesis of atomically smooth films as well as multilayers with perfect interfaces [9,10]. Typical surface roughness determined from atomic force microscopy (AFM) data is $0.2 - 0.5 \text{ nm}$, less than *1 UC* height which in *LSCO* is 1.3 nm . The films were grown on single-crystal LaSrAlO_4 substrates polished perpendicular to the [001] crystallographic direction, so that the *c*-axis of *LSCO* was normal to the film surface. The microstructure, growth mechanism of *LSCO/LCO* heterostructures, and their superconducting properties have been reported before [9,10].

We have used a contactless, single-coil inductance technique [1,12,13] to measure the absolute value of the magnetic field penetration depth, $\lambda(T)$, and the real part of high-frequency conductivity, $Re\sigma(\omega, T)$, in superconducting ultrathin film samples. This technique has the same advantages as the two-coil mutual inductance method [14], while it has much higher sensitivity to the variations of $\lambda(T)$.

Also, we used a two-stage Gifford-McMahon cryocooler from ULVAC technologies Inc., with a UR4K03 cold head combined with a C10 compressor, running on 60 Hz power. For temperature control, a heater driven by a commercial temperature controller (Lake Shore 340) is mounted on the sample stage. The sample with sapphire holder was mounted either horizontally or vertically. A copper coil placed on the top of the cryocooler head allowed us to apply a small magnetic field (~ 100 Gauss) either perpendicular or parallel to the film surface. Additional measurements have been done in a superconducting magnet with fields up to 9 T.

In this radio-frequency technique, we measure the change of mutual inductance, ΔM , between a film and a one-layer pancake coil with diameter l mm. This coil has 15 turns of 46 awg Cu wire; it is located in the proximity of the film and connected in parallel to a capacitor C . The LC circuit is driven by a TE 1000 RF Vector Impedance Analyzer operating at 0.5-150 MHz, with a high frequency stability of 1 Hz. The film is placed in vacuum, at a small distance (~ 0.1 mm) above the coil; this allows the sample temperature to vary from room temperature down to 3.5 K. The instrument measures the impedance as a function of frequency, which is scanned near the resonant one. Thus we determine the resonance frequency and the impedance (at resonance) as a function of the temperature. Since in our bilayer films the superconducting layer thickness $d \sim 0.2$ nm $\ll \lambda$, the change in the complex mutual inductance of the coil, $M(T)$, that occurs because of the superconducting transition in the film, may be calculated (see details in [1]). Actually, the mutual inductance is a complex quantity because the coil impedance Z is complex [1]. The change of the imaginary part of M with temperature can be determined from the real part of the LC circuit impedance, $ReZ(T)$, as well [1].

We have also used two-coil mutual inductance technique. In these experiments, the film is clamped between two axially symmetric coils of the average radius l mm. The in-phase and quadrature components of the voltage at the receiving coil, in response to an ac current in the drive coil, are detected by conventional lock-in techniques [14]. For our films with $d \ll \lambda$, the in-phase signal is roughly proportional to mutual inductance between drive and pick-up coils: $M(T) \approx M_0(2\lambda^2/Rd)$, while the quadrature signal is proportional to the imaginary part of $M(T)$, where M_0 is the mutual inductance between the coils without the film, d is the film thickness, and R is the effective radius of the coils.

We used radio frequency LC technique [15] to measure $\lambda(T)$ for organic single crystals κ -(BEDT-TTF) $_2$ Cu[N(CN) $_2$]Br. This technique employs a circular solenoid coil into which the sample (0.5x0.5mm) is placed with a face parallel or perpendicular to the high-frequency field. The coil is a part of the LC circuit driven by the impedance meter (VM-508 TESLA). Changes in the properties of the sample lead to the change of the coil's inductance that in turn results in the change of the resonance frequency of the LC circuit, similar to the one-spiral-coil technique.

3. Results

The inversion from $M(T)$ in single coil technique to $\lambda(T)$ and $\sigma(T)$ is simple compared to the more familiar two-coil mutual inductance method [14]. Calculating the integrals in inversion equations (see [1]) one gets the values of $ReM(T)$ and $ImM(T)$ at a fixed temperature. From this, we obtain $\lambda(T)$, the kinetic inductance of the film, $L_k^{-1}(T) = \mu_0 \lambda^2 / d$, and the high-frequency conductivity, $\sigma(\omega, T)$.

In Fig. 1 we show the values of $L_k^{-1}(T)$ and $\omega \text{Re}\sigma(T)$ of an M - I bilayer film measured at different frequencies, $8 \text{ MHz} < \nu < 51 \text{ MHz}$, in single-coil inductance experiments, together with the two-coil mutual inductance data taken at 23 kHz . One can notice a large difference between the high-frequency and low-frequency data; in the later, the change in the inductive part of the coil impedance, $\text{Re}M(T)$, is much more abrupt. Also the temperature at which the superconducting transition onsets becomes apparent, T_c , is lower in low-frequency data; it increases with ω up to 51 MHz , in both $L_k^{-1}(T)$ and $\omega \text{Re}\sigma(T)$. Apart from this, there is also a small difference, $\Delta T = 0.22 \text{ K}$, in the onsets observed in $\text{Re}M(T)$ and $\text{Im}M(T)$, in the low-frequency data.

Fig. 1. (Color online) Temperature dependence of $L_k^{-1}(T)$ and $\omega \text{Re}\sigma(T)$ for a $LSCO/LCO$ bilayer film, measured by the single-coil inductance technique at $12.5 - 50.8 \text{ MHz}$, together with $\text{Re}M/M(0)$ and $\text{Im}M/M(0)$ at 23 kHz measured by the two-coil mutual inductance method.

In Fig. 2, we show the derivative functions $d\text{Re}M(T)/dT$ and $d\text{Im}M(T)/dT$ measured in another bilayer $LSCO/LCO$ film exposed to a weak magnetic field B , oriented either parallel or perpendicular to the film surface. As one can see, the onset point for $d\text{Re}M(T)/dT$ is shifted downwards with respect to the onset of $d\text{Im}M(T)/dT$ by a nearly constant value of about $\Delta T_c = 1.2 \text{ K}$. This shift is almost independent on whether the weak magnetic field ($H = 100 \text{ Gauss}$) is on or off. However, it rises from $\Delta T_c = 0.22 \text{ K}$ in low-frequency data at 23 kHz , to $\Delta T_c = 1.2 \text{ K}$ at 50 MHz . Since the observed high-frequency response is dominated by the vortex-antivortex pairs with the vortex separation length $r \sim l_\omega \sim \omega^{-1/2}$, one indeed expects T_{BKT} to increase with the frequency [1,2]. Thus the observed frequency dependence may be considered as a proof that a dynamic BKT transition is being seen. Similar two-step behavior of $H_{c2}(T)$ was observed in ultrathin YBCO films and bilayers with lower T_c as well [1,8].

Another test for T_{BKT}^* can be made by analysing the onset points of strong dependence of $L_k^{-1}(T)$ and $\omega Re\sigma(T)$ on the frequency. A central quantity in the dynamic description of BKT transition is the frequency-dependent complex dielectric function $\varepsilon(\omega)$ which describes the response of a 2D superconductor to an external time-dependent field. The measured $L_k^{-1}(T)$ is renormalized from the BCS value $L_{k0}^{-1}(T)$ in the absence of vortices: $L_{k0}(T)/L_k(T) = n_s/n_{0s} = Re[1/\varepsilon(\omega)]$.

Figure 2. (Color online) Temperature dependence of the derivatives $dReM(T)/dT$ and $dImM(T)/dT$ of a bilayer LSCO/LCO film at different frequencies, with or without external magnetic field ($H = 100$ Gauss) perpendicular and parallel to the film surface.

After Refs. 1 and 3, one can derive the following relation in the high-frequency limit:

$$\frac{L_k^{-1}(T)}{\omega Re\sigma} = \frac{\pi(Y-1)}{2Y \ln Y}, \quad (1)$$

where: $Y = (l_\omega / \xi^+)^2$, $l_\omega = 14D/\omega^{1/2}$ is the vortex diffusion length and D is the vortex diffusion constant. Both real and imaginary part of the $1/\varepsilon(\omega)$ are directly related to $Y(T)$ [3]. Using the $L_k^{-1}(T)$ and $\omega Re\sigma(T)$ data from Fig. 1, we solved Eq. (1) for $Y(T)$ and in Fig. 3 plotted Y versus $(1/T)$ curves.

The qualitative explanation of the $Y(T)$ dependence is as follows. By probing the system at finite frequencies, the observed bound-pair response is dominated by those pairs with $r \sim l_\omega$. At temperatures below T_{BKT}^{dc} , the dissipation is proportional to the number of such vortex-antivortex pairs [2,3]. This number grows gradually with temperature up to T_{BKT}^* . On the high-temperature side, $Re\sigma$ decreases with increasing temperature since $Re\sigma \propto 1/n_f\mu$, where n_f is the density of free vortices and μ is the vortex mobility [3]. Dissipation is the largest when the correlation length $\xi^+(T)$, i.e. the average distance between thermally induced free vortices above T_{BKT}^* , becomes equal to l_ω , which determines the BKT transition temperature at a given frequency, T_{BKT}^* . This transition temperature is determined as the point at which $Y=1$ (corresponding to the maximum of $\omega Re\sigma(T)$ curve) and is frequency-dependent due to $r \sim l_\omega$ relation.

Figure 3. (Color online) Temperature dependence of $Y = (L/\xi^+)^2$ at two different frequencies: 8 MHz and 50 MHz.

Other candidates for exhibiting the BKT state are the quasi-two-dimensional (2D) organic superconductors, which show 2D electronic structure with extremely weak interlayer coupling [16]. Fig. 4 shows the temperature dependence of the $ReM(T)$ and $ImM(T)$ in κ -Br single-crystal samples. A clear upturn of the $ImM(T)$ is observed at T_{c1} , while $ReM(T)$ stays at zero when the high-frequency magnetic field, H_{rf} , is aligned precisely perpendicular to the conducting BEDT-TTF layer.

Figure 4. Temperature dependence of ReM and ImM of κ -Br samples close to T_c with H_{rf} parallel and perpendicular to the conducting layer.

This upturn disappears when the RF field is oriented parallel to 2D conducting layer. Based on the comparison with the *LSCO/LCO* film data presented above, we infer that T_{c1} is the BCS transition temperature, while T_{c2} is the BKT transition temperature. In Figure 5, we present the magnetic field dependence of these curves, showing that T_c shift is almost independent from the magnetic field.

Figure 5. Temperature dependence of ReM and ImM of k -Br samples at different magnetic fields perpendicular to the conducting layer: $B = 0, 0.5, 1.0, 1.9, 2.8$, and 3.8 T.

In conclusion, we have studied the temperature dependence of the real and the imaginary parts of the complex sheet conductance, $\sigma(T, \omega)$ in *LSCO/LCO* bilayer films and κ -(BEDT-TTF)₂Cu[N(CN)₂]Br single crystals. Our RF measurements showed three key features: (i) a steep jump in $L^{-1}_k(T)$, (ii) a maximum in $\omega Re\sigma(T)$, and (iii) a systematic downward shift of the T_c onset point of $L^{-1}_k(T)$ curves compared to the transition onset of $\omega Re\sigma(T)$ curves. Weak magnetic field does not change this temperature shift. Although the first two features are in agreement with the BKT model, independence of the shift on magnetic field is rather surprising.

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