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Nodeless pairing in superconducting copper-oxide monolayer films on $Bi_2Sr_2CaCu_2O_{8+\delta}$

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Abstract The pairing mechanism of high-temperature superconductivity in cuprates remains the biggest unresolved mystery in condensed matter physics. To solve the problem, one of the most effective approaches is to investigate directly the superconducting CuO_2 layers. Here, by growing CuO_2 monolayer films on $Bi_2Sr_2CaCu_2O_{8+\delta}$ substrates, we identify two distinct and spatially separated energy gaps centered at the Fermi energy, a smaller U-like gap and a larger V-like gap on the films, and study their interactions with alien atoms by low-temperature scanning tunneling microscopy. The newly discovered U-like gap exhibits strong phase coherence and is immune to scattering by K, Cs and Ag atoms, suggesting its nature as a nodeless superconducting gap in the CuO_2 layers, whereas the V-like gap agrees with the well-known pseudogap state in the underdoped regime. Our results support an *s*-wave superconductivity in $Bi_2Sr_2CaCu_2O_{8+\delta}$, which, we propose, originates from the modulation-doping resultant two-dimensional hole liquid confined in the CuO_2 layers.

Keywords Copper oxides · Molecular beam epitaxy · Nodeless pairing · Modulation doping

1 Introduction

The discovery of high temperature (T_c) superconductivity (HTS) in cuprates [1] has triggered tremendous efforts to elucidate as to why they superconduct at high T_c. However, this enigma remains unresolved, apparently owing to the layered structure in which the superconducting CuO_2 layers are sandwiched between non-superconducting charge reservoir layers, for example, BiO/SrO in the case of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212). Upon chemical doping in the reservoir layers, rather than direct chemical doping in the CuO_2 layers, the resulting carriers transfer into the CuO_2 layers and boost superconductivity therein, resembling the HTS in single-unit-cell FeSe films on $SrTiO_3$ [2]. By layer-by-layer removal with an Ar^+ bombardment technique, we could reveal respective quasiparticle (QP) excitations of the constituent planes of Bi-2212 [3] and Bi-2201 [4]. However, the CuO_2 surface obtained is too small to systematically investigate its superconducting property.

Here we grow ultrathin CuO_2 films on the BiO surfaces of the cleaved Bi-2212 crystals by a state-of-the-art ozone molecular beam epitaxy (MBE) technique. This approach allows direct measurements of the electronic structure of

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the CuO_2 layers by in situ scanning tunneling microscopy (STM). We demonstrate that the superconducting gap in the CuO_2 layers is nodeless, contradictory to the nodal *d*-wave pairing scenario that is often thought to be the most important result in the 30-year study of the HTS mechanism of cuprates.

2 Experimental

Our experiments were conducted in an ultrahigh vacuum low temperature STM system equipped with ozone-assisted MBE chamber (Unisoku), with a base pressure of $\sim 1 \times 10^{-10}$ Torr. The copper-oxide films were prepared by evaporating high-purity Cu (99.9999%) sources from a standard Knudsen cell under ozone flux beam of $1.0–5.0 \times 10^{-5}$ Torr. The flux beam of ozone from a home-built ozone gas delivery system was injected into the MBE using a 1/2 in. stainless tube (Swagelok), $\sim$50 mm distant from the sample. The K, Cs and Ag atoms were respectively evaporated from alkali-metal dispensers (SAES Getters) and home-made Ta boat, with the samples kept at approximately 50 K. Polycrystalline PtIr tips were cleaned by *e*-beam heating in UHV and calibrated on the MBE-grown Ag/Si(111) films before STM measurements. All STM images were acquired at 4.2 K in constant-current mode, and the differential tunneling d*I*/d*V* spectra were measured by using a standard lock-in technique with a small bias modulation of 1 mV at 966 Hz, unless otherwise specified.

3 Results and discussion

MBE growth of copper-oxide films on Bi-2212 with $T_c = 91$ K (Fig. 1a–d) proceeds in layer-by-layer mode, and epitaxial crystalline films down to a single monolayer (ML) could be prepared. The STM topographic images (Fig. 1a, d) reveal atomically flat and nearly defect-free surfaces on the cleaved Bi-2212 crystal. The characteristic *b*-axis supermodulation (Fig. 1c) is clearly seen in the uncovered BiO regions in Fig. 1a. The atomically resolved image in Fig. 1d reveals the nearest-neighboring Cu ions in the ML films to be 3.8 Å apart. The square structure and the in-plane lattice constant are consistent with those of the CuO_2 plane in bulk Bi-2212. For rocksalt-structured tetragonal CuO (T-CuO) [5–7], a $\sqrt{2} \times \sqrt{2}$ surface reconstruction might blur half Cu ions and lead to the same lattice constant. However, the identification of the films as CuO_2 is sustained by a large separation of 2.35 eV between the Fermi level (E_F) and the valence band of T-CuO [6], while the corresponding value here is only 1.40 eV (Fig. 1e).

In scanning tunneling spectroscopy (STS), the differential conductance d*I*/d*V* measures the local density of states (DOS) as a function of bias voltage (*V*). Our d*I*/d*V* measurements reveal several distinctive features of the CuO_2 films from the cleaved BiO surfaces that have been extensively studied by STM/STS [8]. First, the overall electronic spectrum of the CuO_2 films is characterized by a Mott–Hubbard-like gap of 2.21 eV without prominent states in it (Fig. 1e). The result is expected since the ground state of CuO_2 is a Mott insulator as previously observed by STM in $Ca_2CuO_2Cl_2$ [9]. The pronounced DOS at 1.1 eV can be assigned to the upper Hubbard band, while the shoulders around -1.82 eV (see the arrow in Fig. 1e) the charge transfer band (CTB) [10] or Zhang–Rice–Singlet [11] because of strong on-site Coulomb repulsion. The CTB gap magnitude of 2.21 eV agrees with 2.2 eV as measured in $Ca_2CuO_2Cl_2$ [9] and 2.0 eV for $La_{2-x}Sr_xCu_2O_4$ [12].

Second, and the most surprisingly, in some regions the low-energy QP excitations of the CuO_2 films disclose a well-defined U-gap (Fig. 1f) (Δ_s). Its two pronounced conductance peaks at gap edges ($\pm$18.6 meV) and the vanishing DOS in between, suggest that Δ_s is an *s*-wave-like gap. To avoid possible artificial effects in our measurements, we collected more than 3,000 spectra on different locations of the CuO_2 films from more than forty samples. The persistence of the U-gap in these regions is illustrated in Fig. 1h: despite changes in the coherence peak intensity and the gap size ranging from 16 to 22 meV, the essential U shape can clearly be seen. The observation invalidates the general assumption that the V-shaped STS spectra on the cleaved BiO surfaces are due to tunneling from the underlying CuO_2 planes in Bi-2212 [8]. Other regions are characterized by a larger V-shaped gap (Δ_p) (the black curve in Fig. 1g). At boundaries between the two types of regions, a double-gap feature with mixed U- and V-gaps (violet curve in Fig. 1g) is present and expected. Within our instrumental resolution, we find no difference in the detailed features of these V-gaps and the pseudogap (PG) from the BiO surfaces in Fig. 1c [8]. The two-gap feature, together with the systematic spatial evolution of d*I*/d*V* spectra observed throughout, suggests two types of QP gaps at different energy scales in the CuO_2 films on Bi-2212, although their spectral weights (*W*) are spatially varied. Such spatial inhomogeneity might originate from different amount of charge transfer from the substrate to CuO_2, e.g. the U-gap regions have more hole carriers as compared to the V-gap regions. This charge transfer seems not induce apparent large-energy-scale states in the CTB gap (Fig. 1e). The result is very significant and implies that external doping of the CuO_2 Mott insulator doesn't alter its fundamental electronic structure, an issue fiercely debated