Coupling Behavior of Cu-O₂ Layers in Oxygen-Deficient YBCO Films and Ultrathin YBCO Films

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We have measured the resistivity as a function of temperature in a magnetic field at various field angles θ for oxygen-deficient YBCO films and ultrathin YBCO films; θ is the angle between the crystal c axis and the direction of the magnetic field. The angular dependence of the critical field, H_{c2} , shows a cusplike behaviour at $\theta = 90^\circ$ for the oxygen-deficient 60 K YBCO films indicating that the Cu-O₂ layers are two-dimensional in nature. Both T_c versus θ in a constant field and H_{c2} as a function of θ show a plateau at $\theta = 90^\circ$ for 83 K (zero resistance) ultrathin YBCO films with a thickness of 20 units of the YBCO cell; these results indicate that there is a coupling between the Cu-O₂ layers. The dimensionality of the Cu-O₂ layers in oxygen-deficient YBCO films and ultrathin YBCO films is discussed in terms of the anisotropic Ginzburg-Landau model and the Tinkham thin-film model.

High- T_c oxide compounds are characterized by layer structures (Cu-O₂ layers). Oxide compounds of this class are of intrinsic interest due to the coupling between the Cu-O₂ layers. Earlier work on the layered superconductors has shown that in the isotropic but three-dimensional system, H_{c2} is a continuous, differentiable function of orientation in field as derived from the anisotropic Ginzburg-Landau (GL) theory.¹ In contrast Tinkham² pointed out the discontinuous derivative $dH_{c2}/d\theta$ at $\theta = 90^\circ$ for the two-dimensional thin film, θ is the angle between the normal of the film and the field direction. Recent efforts³⁻⁵ in the coupling of the Cu-O₂ layers using 90 K YBCO single crystals have shown that YBCO compounds show a three-dimensional behavior indicating that there is a coupling behavior between the Cu-O₂ layers. We report here the angular dependence of the

critical field, H_{c2} , in the oxygen-deficient YBCO films ($T_{c,zero} \approx 60$ K) and ultrathin YBCO films (20 unit cells of YBCO). The data are analyzed with Tinkham's two-dimensional (2D) thin film model and the three-dimensional (3D) anisotropic Ginzburg-Landau model.

Tinkham has derived the angular dependence of the critical field in 2D thin film. The result is

$$|H_{c2}(\theta) \cos \theta / H_{c2,c}| + [H_{c2}(\theta) \sin \theta / H_{c2,ab}]^2 = 1 \quad (1)$$

where $H_{c2,c}$ is the critical field with the applied field parallel to the c axis and $H_{c2,ab}$ is the critical field with the applied field parallel to the ab plane, and θ is the angle the magnetic field makes with the crystal c axis. As Eq. (1) predicts a discontinuity in $dH_{c2}(\theta)/d\theta$ at $\theta = 90^\circ$, one expects to observe a cusp-like behavior in the angular dependence of H_{c2} at $\theta = 90^\circ$.

The angular dependence of H_{c2} in the anisotropic GL theory is

$$H_{c2}(\theta) = H_{c2,c}(\theta = 0^\circ) [\cos^2 \theta + [(m_{ab}/m_c) \sin^2 \theta]^{1/2}] \quad (2)$$

where m_{ab}/m_c is the effective mass ratio which is related to the critical fields by the relation: $m_{ab}/m_c = (H_{c2,c}/H_{c2,ab})^2$. Equation (2) predicts a continuous change of $dH_{c2}/d\theta$ at $\theta = 90^\circ$. So one should observe a continuous derivative of $dH_{c2}/d\theta$ with $dH_{c2}/d\theta = 0$ at $\theta = 90^\circ$.

The resistivity as a function of temperature in a magnetic field for the samples was measured by the four-probe method. A superconducting cryostat which can generate a horizontal magnetic field up to 7 T was used to measure the resistivity of the film under various field directions. The orientation of the sample relative to the magnetic field is shown in the inset of Fig. 1. The angle θ which the c axis makes with respect to the direction of the magnetic field can be changed by rotating the sample about the z axis.

The oxygen deficient YBCO films with $T_{c,zero}$ at about 60 K and ultrathin YBCO films were grown in situ by the rf magnetron sputtering technique. The sputtering target was the stoichiometric $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ oxide compound. The substrates were MgO(001). The sputtering pressure was a mixture of Ar and O_2 (7:3). We made the oxygen deficient in the samples by controlling the oxygen content in the sample preparation processes. A detailed description of the sample preparation is reported elsewhere.⁶ The as-grown films were shiny and smooth. The X-ray diffraction pattern shows that the films have a preferred orientation with the crystal c axis perpendicular to the surface of MgO(001).

Figure 1 shows the temperature dependence of the resistance in a magnetic field parallel to the crystal c axis and the ab plane for the oxygen-deficient YBCO film. The zero resistance of the film is about 60 K. The increment field is 1 T in each curve. The film shows $dH_{c2,ab}/dT = -10.5$ T/K, $dH_{c2,c}/dT = -1.6$ T/K at the 10% resistive transition.

Fig. 1. Temperature dependence of the resistance in fields (a) parallel to the crystal c axis and (b) to the ab plane. The inset of (b) shows the orientation of the film with respect to the direction of the magnetic field; θ is the angle between the crystal c axis and the magnetic field.

Figure 2 shows the angular dependence of the critical field at 71.1 K. The theoretical calculation based on the Tinkham thin film model and the anisotropic GL model are also shown. We have used the experimental values of $H_{c2,c}(\theta=0)$ and $H_{c2,ab}(\theta=90^\circ)$ in the theoretical calculation. A cusplike behavior was found and the data fitted much better the Tinkham thin-film model. The data indicate that the Cu-O₂ layer in the oxygen-deficient films are 2D in nature. There is a depletion of oxygen in the Cu-O chain as one varies the sample from the 90 K YBCO oxide compounds to the 60 K YBCO oxide compounds. A 3D behavior has been observed for the 90 K YBCO single crystal.³⁻⁵ Our present data show a 2D behavior in the Cu-O₂ plane in oxygen-deficient YBCO films. The data tend to indicate that the Cu-O chain plays a role in the coupling of the Cu-O₂ layers.

The coupling behavior in the Cu-O₂ layers is related to the anisotropic effective mass ratio m_c/m_{ab} . The YBCO films show a coupling behavior in the Cu-O₂ layers and an effective mass ratio $m_c/m_{ab} = 13.1$.⁷ Our oxygen-deficient YBCO films show $m_c/m_{ab} = 33$, which is more anisotropic than that of YBCO films, and indicate a 2D behavior in the Cu-O₂ layers. The anisotropic effective mass ratio for BSCCO film is $m_c/m_{ab} = 207$,⁸ which is even more anisotropic than that of oxygen-deficient YBCO films. Therefore BSCCO films show a 2D behavior in the reported data.⁵

Figure 3 shows the resistance as a function of temperature in a field parallel to the *ab* plane and the crystal *c* axis respectively for the ultrathin YBCO film (sample *a*) grown onto SrTiO₃(001) with 96 nm thick PrBa₂Cu₃O_{7-y} (PBCO) Buffer layer. The as-grown ultrathin YBCO film is

Fig. 2. Critical fields as a function of θ at $T = 71.1$ K derived from the 10% resistive transition. Also shown are the theoretical fittings based on the Tinkham 2D thin-film model and the anisotropic GL 3D model.

Fig. 3. Temperature dependence of resistance in magnetic fields (a) perpendicular and (b) parallel to the crystal c axis for ultrathin YBCO films with a thickness 24 nm. The increment of the magnetic field in each curve is 2 T.

24 nm thick (20 units of YBCO cell) and has T_c onset at 88 K and zero resistance at 83 K. The X-ray diffraction pattern shows a preferred orientation with the crystal c axis perpendicular to the plane of SrTiO₃. The increment of the magnetic field in each curve is 2 T. The angular dependence of T_c derived from the 10% resistive transition at $H = 6$ T is shown in Fig. 4. We found a plateau in the angular dependence of T_c at $\theta = 90^\circ$. Bauhofer *et al.*³ reported a plateau in the angular dependence of T_c in YBCO single crystals and suggested a coupling behavior in the CuO₂ plane. Our present data in ultrathin YBCO films show a similar feature (a plateau) in the angular dependence of T_c at $\theta = 90^\circ$. These results indicate that the coupling between Cu-O₂ layers exists in ultrathin YBCO films with a thickness of 20 units YBCO cell.

Fig. 4. Angular dependence of T_c obtained from the 10% resistive transition at $H = 6$ T for ultrathin YBCO films with a thickness 24 nm.

Fig. 5. Angular dependence of H_{c2} at $T = 86.4$ K for ultrathin YBCO films with a thickness 24 nm. H_{c2} is obtained from the 10% resistive transition. The data fit better to the anisotropic GL 3D model.

Figure 5 shows the angular dependence of H_{c2} derived from the 10% resistive transition for sample *a* at $T = 86.4$ K. Also shown in Fig. 5 is the theoretical calculation based on Tinkham's 2D thin film model and the anisotropic 3D GL model. The data fit better to the 3D GL model. The data suggest that there is a coupling between the Cu-O₂ layers in ultrathin YBCO film with $T_{c,zero} = 83$ K.

The critical fields for sample *a* obtained from the 10% resistive transition are $H_{c2,ab} = 4.53$ T and $H_{c2,c} = 0.55$ T at 86.4 K, where $H_{c2,ab}$ is the critical field with the field applied along the *ab* plane and $H_{c2,c}$ the critical field with the applied field along the crystal *c* direction. If we put $H_{c2,ab} = 4.53$ T and $H_{c2,c} = 0.55$ T into the GL relations; $\xi_{ab}^2 = \Phi_0 / (2\pi H_{c2,c})$ and $\xi_c = \Phi_0 / (2\pi H_{c2,ab} \xi_{ab})$, we obtain $\xi_{ab} = 24.5$ nm and $\xi_c = 2.97$ nm, where ξ_{ab} is the coherence length along the *ab* plane and ξ_c the coherence length along the crystal *c* direction. We note that ρ_c is larger than the distance between the Cu-O₂ layer which is 0.8 nm⁹. Therefore there can be a coupling behavior between the Cu-O₂ layers in ultrathin YBCO films.

In conclusion we have studied anisotropic critical fields of oxygen-deficient 60 K superconducting YBCO films and the ultrathin 83 K YBCO film. The angular dependence of H_{c2} shows a cusplike behavior indicating that the Cu-O₂ layer in the 60 K YBCO films is 2D in nature. The angular dependence of T_c in a constant field for ultrathin YBCO films shows a plateau at $\theta = 90^\circ$ and the angular dependence of H_{c2} at T near T_c fits well to the GL 3D model. These data indicate that the coupling behavior of the Cu-O₂ layers exists for ultrathin YBCO films at T near T_c .

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