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# Enhancement of $H_{c1}$ and its influence on the irreversibility lines in multilayer YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>/PrBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> thin films

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Abstract. The irreversibility line  $H_{irr}$  cannot cross  $H_{c1}$ , and the temperature dependence of  $H_{irr}(T)$  differs from  $H_{c1}(T)$ . Therefore there exists an intersection between them, and it is expected that the simple power-law behaviour of the irreversibility line  $H_{irr}(T) \sim (1 - T/T_c)^{\alpha}$  will break down over a wide range of temperature. We have observed this interesting phenomenon in our three YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>/PrBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> (YBCO/PrBCO) multilayer thin films with different YBCO superconducting layers for the first time due to the enhancement of  $H_{c1}$  in a parallel (|| CuO plane) magnetic field. The temperature dependence of  $H_{c1}(T)$  in these three films is in good agreement with calculations of the enhanced  $H_{c1}$  derived from the London approximation for the case of a slab of thickness  $\tau$  which is thinner than the London penetration depth  $\lambda$  when the applied field is parallel to the sample surface, and they mimic the results observed by Civale and co-workers in thinner pure YBCO ultra-thin films (~25 nm).

### 1. Introduction

According to the conventional definition, the lower critical field can be understood as the field at which a single flux line occurs in a sample. The lower critical field  $H_{c1}$  in bulk YBCO (both single crystal as well as thin film) has been measured by either r.f. surface resistance or magnetization techniques [1–3]. In these peculiar type-II high- $T_c$  superconductors,  $H_{c1}$ is usually very small. Even in pure YBCO thin films it is only about several hundred gauss.  $H_{cl}$  is usually very small for thin films, so the intersection between  $H_{cl}(T)$  and  $H_{irr}(T)$ occurs in a very narrow temperature range  $\Delta(T) = T_e - T_c < 10^{-3}$  K, and is therefore experimentally difficult to observe. So in high- $T_c$  superconducting materials,  $H_{irr}$  normally obeys a simple power-law behaviour, i.e.  $H_{\rm irr}(T) \propto (1 - T/T_{\rm c})^{\alpha}$ , with  $\alpha$  in the range between 4/3 and 2. However, Civale and co-workers [4] have very recently found that in thinner YBCO thin films (~250 Å, less than the London penetration depth  $\lambda$ ), the lower critical field parallel to the CuO plane is greatly enhanced up to several tesla; the simple power-law behaviour of  $H_{irr}(T)$  breaks down over a wide range of temperature, since the irreversibility line cannot cross  $H_{c1}(T)$ . Considering the special structure of YBCO/PrBCO multilayers, in which the flux line can be strongly pinned in PrBCO when the magnetic field is applied parallel to the a-b plane, the magnetic field penetrates through the PrBCO layer; thus the superconducting layer can be treated as similar to the thinner YBCO film so that  $H_{c1}(T)$  should also be enhanced. Moreover, the thickness of the superconducting YBCO layer can be adjusted conveniently, so we can investigate further the YBCO thickness dependence of the enhanced  $H_{c1}$  in YBCO/PrBCO superlattices.

# 2. Experiment

The following four samples, (YBCO)<sub>5</sub>/(PrBCO)<sub>2</sub> (YP143), (YBCO)<sub>8</sub>/(PrBCO)<sub>2</sub> (YP150), (YBCO)<sub>18</sub>/(PrBCO)<sub>2</sub> (YP176) and pure YBCO (t114) thin films, were selected as candidates to be investigated. The thickness of these films was about 250 nm. They were prepared by continuous sequential high-pressure d.c. sputtering onto a heated SrTiO<sub>3</sub>(100) substrate in a pure oxygen atmosphere ( $P_{O_2} \approx 3 \text{ mbar}$ ) from planar stoichiometric YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> and PrBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> targets ( $\phi \approx 50 \text{ mm}$ ) as reported elsewhere [5]. The film was characterized by x-ray diffraction (XRD), which showed clearly the presence of satellite peaks reflected from the superlattice structures with higher intensity. The resistance measurements were carried out on a 100  $\mu$ m long by 10  $\mu$ m wide strip (patterned by ion milling) by the standard four-probe method. The sample was held on a sample holder which could be rotated from -180° to 180°, and the resolution of the angle was 0.04°. The measurements were performed in a liquid helium cooled cryostat. A magnetic field up to 12 T was generated by a superconducting solenoid.

## 3. Results and discussion

The irreversibility line of a type-II superconductor separates the H-T plane into two regions: a region above the line where the critical current is zero and the magnetic properties are reversible and a region below the line where a critical current is observed and the magnetic properties are irreversible. In our experiments zero resistance (in fact, R = 0 is restricted by the resolution of the instruments; in our experiments  $1.5 \times 10^{-3} \mu\Omega$  cm was used as a criterion to determine the irreversibility line) was taken as a criterion to determine the H-Tphase diagram through measuring magnetoresistance at a fixed temperature. For a fixed temperature  $T_i$ , there is present a field  $H_i$  above which the resistance of the sample appears; thus we can get a point  $(T_i, H_i)$  of the H-T phase diagram, and successively we can obtain the whole H-T phase diagram. The measuring current density J is about 500 A cm<sup>-2</sup>, much less than the critical current density of the sample.

Figure 1(a) shows the relation of H to temperature for the sample YP176, both in a magnetic field parallel to the a-b plane (o) and perpendicular to the a-b plane ( $\Delta$ ) determined by the method mentioned above. Comparing these two situations, we can see clearly that the irreversibility line for H parallel to the a-b plane shifts to a higher temperature range. Further, it is obvious that there exists a knee at  $H \approx 2$  T which can be understood as the irreversibility line run into  $H_{c1}(T)$ . In order to investigate the temperature dependence of  $H_{irr}$ , we use the reduced temperature  $t = 1 - T/T_c$  instead of T, then plot  $H_{irr}$  as a function of t on a double logarithmic scale, as shown in figure 1(b). It is clear that, for H perpendicular to the a-b plane, almost all of the experimental data fall onto one line, the slope of which is about 1.37. That means the temperature dependence of  $H_{irr}$ obeys a simple power-law behaviour, i.e.  $H_{irr} \sim (1 - T/T_c)^{1.37}$ . In the case of H parallel to the a-b plane, when H exceeds  $H_c$  (about 2T), the experimental data fall onto a line, the slope of which is about 1.45; however, when H is below 2T, the data clearly deviate from the line, indicating that in this region  $H_{irr}$  does not obey the simple power-law behaviour.

Similar behaviour has also been observed in other samples, YP150 and YP143, whose H-T diagrams are shown in figures 2 and 3, respectively. Their  $H_c$  take place at about 5.5 T and 7.5 T, respectively. One interesting point is that for YP143, even in the case of H perpendicular to the a-b plane, there is also deviation from a simple power-law behaviour of the irreversibility line below H = 0.8 T, as shown clearly in the inset of figure 3. We



Figure 1. (a) H-T phase diagram for YP176(18:2) with the magnetic field both parallel to the a-b plane (o) and perpendicular to the a-b plane ( $\Delta$ ) determined by the zero-magnetoresistance method. (b) Double logarithmic relation between H and the reduced temperature  $t = 1 - T/T_c$ .

suggest that it is also caused by enhancement of  $H_{c1}^{\perp}$  due to the strong pinning in this superlattice. We propose that when the modulation ratio of the superlattices is small, there is a larger dislocation between the superconducting layer and the insulating layer, which means that in some regions there exist six u.c. YBCO layers and in other regions there exist five u.c. YBCO layers (on average there are about 5.5 u.c. YBCO layers, i.e. the modulation ration in YP143 is about 5.5:2; this is consistent with the result measured by XRD and also with the fitting results of  $H_{c1}(T)$  for YP143 to be mentioned later). So there are many kinks present at the boundary of these two kinds of region. These kinks form the pinning centres in the superlattices. Finally, they lead to the enhancement of  $H_{c1}^{\perp}(T)$ , and also in the case of H perpendicular to the a-b plane.

In order to rule out the effectiveness of the criteria of the H-T by measuring the magnetoresistance on the characteristics of the irreversibility line, we use the same criteria to check the irreversibility line characteristics in pure YBCO thin films. Figure 4 shows such an H-T diagram for a pure YBCO film obtained by the method mentioned above. In magnetic fields parallel and perpendicular to the a-b plane,  $H_{irr}^{\perp}(T)$  and  $H_{irr}^{\parallel}(T)$  obey the



Figure 2. H-T phase diagram for YP150(8:2) with the magnetic field both parallel to the a-b plane ( $\phi$ ) and perpendicular to the a-b plane ( $\Delta$ ).



Figure 3. H-T phase diagram for YP143(5:2) with the magnetic field both parallel to the a-b plane (o) and perpendicular to the a-b plane ( $\Delta$ ). The inset shows  $\log(H)$  against  $\log(1-T/T_c)$  for YP143 in a magnetic field perpendicular to the a-b plane; it shows clearly the deviation from a simple power-law behaviour of the irreversibility line below H = 0.8 T.

same power-law relation, with a different exponent. For H perpendicular to the a-b plane,  $\alpha = 1.68$ ; the exponent is about 1.90 for H parallel to the a-b plane. So we suggest that the enhancement of  $H_{c1}$  and its influence on the irreversibility line is an intrinsic characteristic of the sample caused by this special kind of multilayer structure.

For these quasi-two-dimensional structural superlattices [6], since the superconducting layer is less than the London penetration depth, when the external magnetic field is applied parallel to the a-b plane,  $H_{c1}$  will be enhanced [7]. Although Somin [8] and Clem and co-workers [9] have discussed the problem of  $H_{c1}$  for ultra-thin-film and Josephson-coupled high- $T_c$  superconductors, and obtained their individual calculating formulae, respectively, neither of them can describe our experimental data quantitatively. Considering the special structural characteristics of the sample, we think that the flux line will be strongly pinned in the PrBCO layer. In other words, when  $H < H_c$ , the flux line (repelled from the superconducting YBCO layer) penetrates only through the PrBCO layer; however, when  $H > H_c$ , the flux line starts to penetrate the YBCO layer. Here,  $H_c$  is defined as the lower



Figure 4. H-T phase diagram for a pure YBCO thin film determined by the same method mentioned in the text.

critical field  $H_{cl}$ . In fact, the enhancement of  $H_{cl}$  in three multilayer structural samples is actually similar to that in thin films observed by Ciavel and co-workers [10]. They have derived a temperature dependence of the enhanced lower critical field on the basis of the London approximation, expressed as follows:

$$H_{c1}^{\tau} = H_{c1}^{\infty} \frac{1 + \left[2/(K_0(\frac{1}{\kappa})\right] \sum_{n=1}^{\infty} (-1)^n K_0(\frac{n\tau}{\lambda})}{1 - 1/\cosh(\tau/2\lambda)}$$
(1)

where  $H_{c1}^{\infty}$  represents the lower critical field for bulk YBCO,  $\lambda$  is the London penetration depth,  $\tau$  is the thickness of the YBCO thin film,  $K_0$  is the Hankel function of zero order and imaginary argument and  $\kappa = \lambda/\xi$  is the Ginzburg-London parameter. The temperature dependence of  $H_{c1}^{\tau}$  is displayed through the temperature dependence of  $\lambda$  and  $H_{c1}^{\infty}$ . Assuming the G-L temperature dependence of  $\lambda$  and  $H_{c1}^{\infty}$  and using the bulk value for  $\lambda = 1300$  Å [11] and  $\kappa = 300$ ,  $H_{c1}^{\infty} = 180$  Oe etc, they have calculated  $H_{c1}^{\tau}$ . They found that the calculation for a YBCO thin film with a thickness of about 250 Å can well describe their experimental data. Taking into account the similar structural characteristics of our samples to the thinner film mentioned above, we expect our experimental data can also be described by (1). Assuming a G-L temperature dependence of  $\lambda$  and  $H_{c1}^{\infty}$ , i.e.

$$\lambda = \lambda_0 \left( 1 - \frac{T}{T_c} \right)^{-1/2} \qquad H_{c1}^{\infty} = H_{c1}^{\infty}(0) \left( 1 - \frac{T}{T_c} \right)$$

using the same parameter, we have calculated  $H_{c1}^{\tau}$  for three samples. The  $\tau$  in  $H_{c1}^{\tau}$  is now replaced by the thickness of the YBCO layer for YP143,  $\tau = 6.48$  nm (in fact, the modulation ratio of YP143 is about 5.5:2, this is consistent with the result measured by XRD), and 9.34 nm and 21 nm for YP150 and YP176, respectively. The calculated results are shown in figure 5. The simulated results are in good agreement with the experimental data in the range of experimental accuracy, because there are no other fitting parameters except the thickness of the YBCO layer during the process of calculation. We must state, however, that the experimental data which are mentioned here are not really the lower critical fields. In fact, the values we obtained by the method of zero magnetoresistance are larger than the real  $H_{c1}$ , i.e. the experimental data shift to a slightly higher temperature range compared with the calculated results. However, we believe that the experimental data have the same temperature and thickness dependence.



Figure 5. H-T phase diagram for superlattices YP143, YP150 and YP176 with YBCO thickness 6.48 nm, 9.34 nm and 21 nm, respectively. The full curves are calculated in terms of (1) for corresponding YBCO thicknesses. The inset shows that of YP143.

One point should be noted: when T is very near  $T_c$  (e.g.  $T > 0.995T_c$ ) the lower critical field calculated by (1) becomes negative—it is physically meaningless. This is caused by the divergence of  $\sum_{n=1}^{\infty} (-1)^n K_0 \left(n \frac{\tau}{\lambda_0} (1-t)^{1/2}\right) \approx \frac{1}{2} \ln \left(\frac{\tau}{\lambda_0} (1-t)^{1/2}\right) - 0.289$ ; however,  $K_0(1/\kappa)$  has a finite value, therefore leading to the numerator in (1) becoming negative and diverging at the point  $T = T_c$ . For this situation, equation (1) should be treated in the following way. When T tends to  $T_c$ ,  $\kappa = \lambda/\xi$  will not be constant, since  $\lambda = \lambda_0 (1-t)^{-1/2}$  tends to infinity, but the coherence length  $\xi$  tends to a limiting value of the thickness of the YBCO layer because of the restriction of the sample geometry when the magnetic field

is parallel to the a-b plane; so  $\kappa = \lambda/\xi$  will be also infinite when T is very near  $T_c$ , and therefore  $K_0(1/\kappa)$  is also divergent at  $T = T_c$ . Finally, the numerator of (1) tends to a finite value, i.e.  $H_{c1}(T \to T_c)$  becomes very small and almost tends to zero; this is in good agreement with the experimental data.

## 4. Conclusion

In summary, we have observed the enhancement of  $H_{c1}$  in YBCO/PrBCO high- $T_c$  superconducting superlattices with the thinner YBCO layer separated by two unit cells from the PrBCO layers. Due to the enhancement of  $H_{c1}$ , the irreversibility line in the case of a magnetic field parallel to the a-b plane no longer obeys a simple power-law behaviour. The temperature dependence of  $H_{c1}$  can be well described by the London approximation, which indicates that in these multilayer structural samples it can be treated as a slab of thickness  $\tau$  which is thinner than the London penetration depth  $\lambda$ , when the applied field is parallel to the sample surface.

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