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Temperature dependence of anisotropic lower critical fields in $(La_{1-x}Sr_x)_2CuO_4$

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The anisotropic lower critical fields H_{c1} are determined from a novel procedure based on the Bean's critical-state model to pick up the onset of flux penetration in broad zero-field-cooled magnetization curves in $(La_{1-x}Sr_x)_2CuO_4$ ($x \approx 0.07$) single crystals. The values of H_{c1} at $T=0$ K are estimated to be 70 G for $H \parallel$layer and 300 G for $H \perp$layer. The temperature dependence of H_{c1} is such that it tends to saturate towards the lowest temperature measured (2 K) in contrast to the other recent results which report the abnormal upturn of the H_{c1}-T curve at low temperatures. Instead the results indicate that the critical current density in plane has a strong temperature dependence even in the temperature range well below T_c and becomes $\sim 10^6$ A/cm^2 at 2 K.

Many experimental studies have been reported on the lower critical field (H_{c1}) of the new high-T_c oxide superconductors.[1-6] However, none of the magnitude, the anisotropy, or the temperature dependence of H_{c1} have been established yet. The reported values for H_{c1} have ranged from a few to thousands of gauss. Especially the reported results on the temperature dependence have been controversial. Some results have shown unusual behavior,[5] such as no saturation or upturn at low temperatures, which has even invoked the question whether the superconducting mechanism of the new high-T_c oxide superconductors is a conventional BCS type or not. The discrepancies in the reported results on H_{c1} come partly from the following problems in determining the lower critical fields of the high-T_c oxide superconductors from the magnetization curves.

The first is a well-known *old* problem due to strong irreversible behavior in *M-H* curves for type-II superconductors.[7] In ideal type-II superconductors, the magnetization ($-4\pi M$) drops sharply at H_{c1}. However, in actual superconductors in which flux pinning is present, flux is reluctant to enter the specimen in an increasing field, and the anomaly at H_{c1} in the *M-H* curve becomes increasingly weaker as the critical current density (J_c) of the superconductor becomes larger. In any case, in principle, the lower critical field H_{c1} is determined by an onset of the deviation from perfect diamagnetism. However, the magnetization deviates very gradually from perfect diamagnetism for large-J_c specimens, and hence, the accurate location of H_{c1} is often hard to pick up.

The second is a material problem involved with the high-T_c oxide superconductors. In the early stages of the research on high-T_c superconductors, most of the measurements were performed using ceramic specimens or low-quality single crystals which have nonsuperconducting second phases in the grain and/or along grain boundaries, leading to weak-link behavior in many aspects of the superconducting properties. For example, magnetic measurements by Senoussi, Oussena, and Ribault[3] using La-Sr-Cu-O ceramic specimens showed very complicated behavior in *M-H* curves, apparently affected by weak links in grain boundaries. Senoussi *et al.* suggested two threshold fields characterizing flux entry: H_{c1}^w, at which the first vortex penetrates the weak links, and H_{c1}^g, at which the first vortex enters the grains themselves. If one strictly takes the threshold field of flux entry as H_{c1} according to the above-mentioned rule for *hard* superconductors, such weak-link behavior causes a large error in estimating H_{c1}. This problem can be avoided by using good single-crystal specimens for measurements, although some people suggest that inherent weak links may exist even in single crystals.[8]

The third problem is due to demagnetization corrections. Single crystals of the high-T_c oxide superconductors are often disk shaped with the **a** and **b** axes in plane. Therefore the measurements for a magnetic field perpendicular to the layer must be corrected by a very large demagnetization factor. In addition, if the crystals have imperfect demagnetization geometries (i.e., nonellipsoidal shape), the conventional demagnetization correction itself may not be quantitatively accurate. Hence it is difficult to obtain the accurate value of H_{c1} for $H \perp$layer.

This paper reports the first extensive magnetic measurements on high-quality $(La_{1-x}Sr_x)_2CuO_4$ single crystals. We believe these measurements overcome the above-mentioned difficulties and provide convincing data on the anisotropic values and temperature dependences of H_{c1} of $(La_{1-x}Sr_x)_2CuO_4$.

Single crystals of $(La_{1-x}Sr_x)_2CuO_4$ ($x \approx 0.07$) used in this work were grown by the traveling solvent floating zone method. The details of the growth and the characterization of crystals are described in Ref. 9, and the transport properties and the upper critical fields are discussed in Refs. 10 and 11. The grown crystals have a cylindrical shape of the dimensions ~6-mm diam×~30 mm, with the crystalline **a** axis nearly along the cylinder axis. The specimens prepared to investigate the anisotropy had the following three different orientations as shown in the insets of Fig. 1: (a) $H \parallel$layer, $J_c \perp$layer, (b) $H \parallel$layer, $J_c \parallel$layer, and (c) $H \perp$layer, $J_c \parallel$layer. To minimize the demagnetization effect, the specimens were cut into disk-shaped slices (2-4 mm×1-2 mm×0.2-0.3 mm). Magnetic measurements were made by a commercial superconducting quantum interference device magnetometer (Quantum Design, Inc.) in the magnetic field range from 0-55 kOe at temperatures from 1.8-30 K. Isothermal *M-H* curves were taken after zero-field cooling (ZFC). The initial slopes of the *M-H* curves slightly exceeded 100% of perfect diamagnetism due to the small but not negligible demagnetization effect: 110% for (a), 110% for (b), and 120% for (c). Here H refers to the applied field which is smaller than the effective field on the sample.

Figure 1 compares magnetization curves for the above three orientations taken at temperatures between 2-30 K. Figures 1(a) and 1(b) have their magnetic fields parallel to the layer. The circulating current perpendicular to the field axis is induced to sustain the gradient of flux density $[\mathrm{rot}\,\mathbf{h}(\mathbf{r}) = 4\pi\mathbf{J}/c]$. The direction of the induced current flow, except along the edge side, is perpendicular to the layer in (a) and parallel to the layer in (b). The configuration in Fig. 1(c) has the magnetic field perpendicular to the layer, and consequently the current is parallel to the layer. The marked difference between the two magnetization curves of Figs. 1(a) and 1(b) comes solely from the difference in the directions of the flowing currents. Keeping in mind that the *M-H* curve becomes broader as J_c becomes larger, this difference clearly shows that the critical current density ($J_c^{\perp}$) perpendicular to the layer is considerably lower than the critical current density ($J_c^{\parallel}$) parallel to the layer. This observation is just expected from the layered structure of La-Sr-Cu-O. In any configuration, as the temperature lowers, the increase of J_c causes the *M-H* curve to broaden. This is especially true for the configurations in Figs. 1(b) and 1(c). For these configurations, except at the temperatures close to T_c, the very gradual deviation of the *M-H* curve from the perfect diamagnetic line makes it difficult to determine the accurate location of H_{c1}. Hence the following analysis was adopted to avoid the ambiguity in picking up H_{c1} from these broadened *M-H* curves.

According to the Bean's critical-state model[12] for the flux entry to superconductors containing pinning centers, the magnetization for a slab of thickness D is expressed in practical units, i.e., gauss (or oersteds) and amperes/cm^2 by[13]

$$-4\pi M_{\mathrm{obs}} = H \quad (H \le H_{c1}), \tag{1}$$

$$-4\pi M_{\mathrm{obs}} = H - \frac{10}{4\pi J_c D} B_{\mathrm{eq}}^2(H) \quad (H_{c1} < H \le H^*), \tag{2}$$

$$-4\pi M_{\mathrm{obs}} = H - B_{\mathrm{eq}}(H) + \frac{\pi J_c D}{10} \quad (H > H^*), \tag{3}$$

where H is an external magnetic field, $B_{\mathrm{eq}}(H)$ is the equilibrium flux density, J_c is the critical current density, and H^* is the field defined as $B_{\mathrm{eq}}(H^*) = \pi J_c D/5$. In these equations it is assumed that the current density is field independent, which may be a good approximation for low fields. Equation (2) can be modified as

$$B_{\mathrm{eq}}(H) = \left[\frac{4\pi J_c D}{10}\right]^{1/2} (H + 4\pi M_{\mathrm{obs}})^{1/2} \quad (H_{c1} < H < H^*), \tag{4}$$

where $H + 4\pi M_{\mathrm{obs}} (\equiv 4\pi\Delta M)$ is an amount corresponding to the deviation of the observed magnetization from perfect diamagnetism. Equation (4) indicates that, in principle, the $(\Delta M)^{1/2}$ vs H plot should give the equilibrium *B-H* curve. Hence the lower critical field is given by the threshold field of this plot, and the critical current density is also given by the slope of this plot at $H \gg H_{c1}$.[14] Figure 2 shows a typical example of such a plot. Figure 2(a) shows raw data and a straight line representing perfect diamagnetism which is a least-squares fit to the low-field

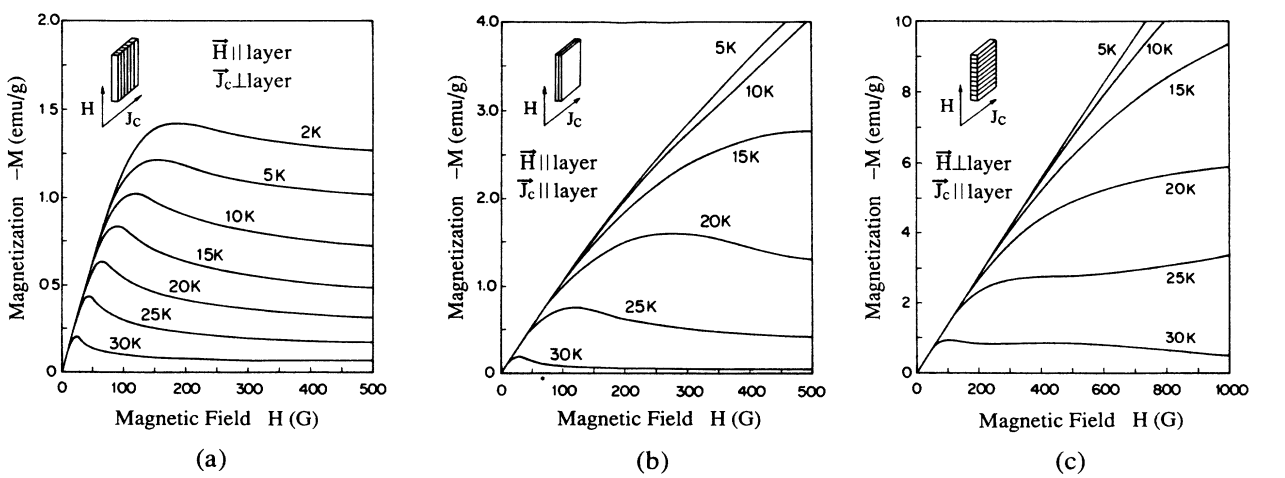

FIG. 1. *M-H* curves of the $(La_{1-x}Sr_x)_2CuO_4$ single crystals: (a) $H \parallel$layer and $J_c \perp$layer, (b) $H \parallel$layer and $J_c \parallel$layer, (c) $H \perp$layer and $J_c \parallel$layer.

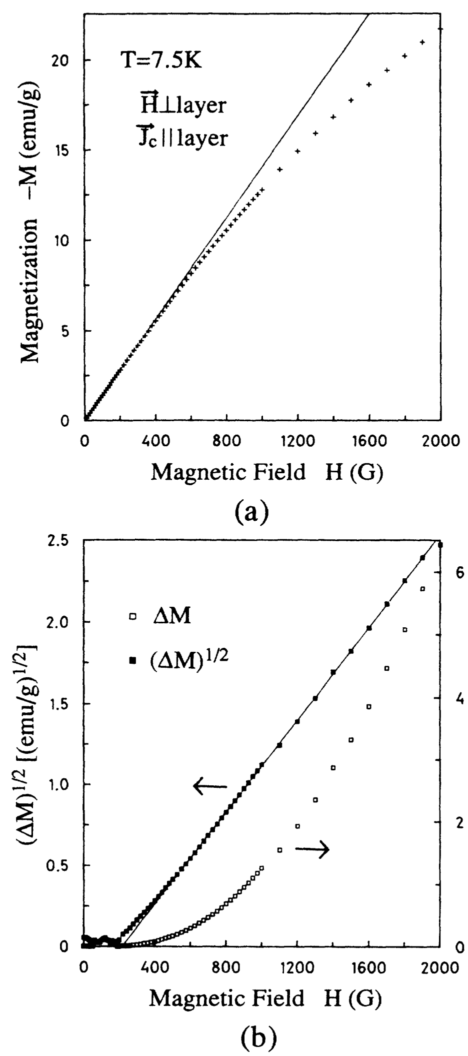

FIG. 2. (a) M-H curve at 7.5 K for $H\perp$layer and $J_c\|$layer and a straight line representing perfect diamagnetism which is a least-squares fit to the data points between 0–220 G. (b) ΔM vs H and $(\Delta M)^{1/2}$ vs H plots of the upper data (a), where ΔM is the difference between the straight line and the observed magnetization.

part of the M-H curve. Figure 2(b) shows ΔM vs H and $(\Delta M)^{1/2}$ vs H plots. The threshold of the $(\Delta M)^{1/2}$ vs H plot is slightly smeared probably due to the demagnetization field at the sharp corners of the specimen. Therefore, H_{c1} is obtained by extrapolating the linear part of the $(\Delta M)^{1/2}$ vs H curve to the horizontal axis and multiplying the intercept ($H_{\text{intercept}}$) by the numerical factor f.[15]

The lower critical fields ($H_{c1}^{\|}$ and $H_{c1}^{\perp}$) parallel[16] and perpendicular to the layer thus obtained for each temperature are plotted against the temperature in Fig. 3. The values of $H_{c1}^{\|}$ can be obtained from either of the data of Figs. 1(a) and 1(b). The results essentially agree with each other, except that the values estimated from Fig. 1(a) are less scattered, which are only plotted in Fig. 3. The temperature dependences of the critical current densities for three configurations estimated from the slope of the linear part of the $(\Delta M)^{1/2}$ vs H curve are summarized

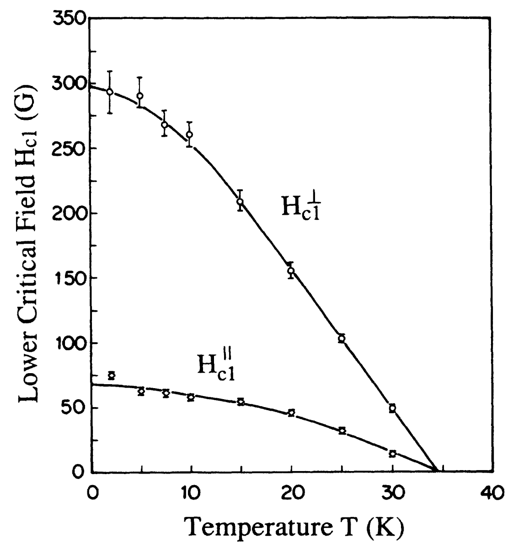

FIG. 3. Temperature dependences of the lower critical fields of $(La_{1-x}Sr_x)_2CuO_4$ for $H\|$layer and $H\perp$layer. The solid curves are drawn as a guide for the eye.

in Fig. 4. It should be noted here that the temperature dependence of $J_c^{\|}$ for $H\|$layer and that for $H\perp$layer appear to be different. In principle, $J_c^{\|}(H\|\text{layer})$ and $J_c^{\|}(H\perp\text{layer})$ need not be the same. However, it should also be noted that in the configuration in Fig. 1(b), flux penetration from the edge sides becomes dominant at low temperature, where $J_c^{\|}(H\|\text{layer}) \gg J_c^{\perp}(H\|\text{layer})$. Therefore the saturation of $J_c^{\|}(H\|\text{layer})$ at low temperatures may not be real.

As mentioned at the beginning of this paper, it has sometimes been reported that the temperature dependence

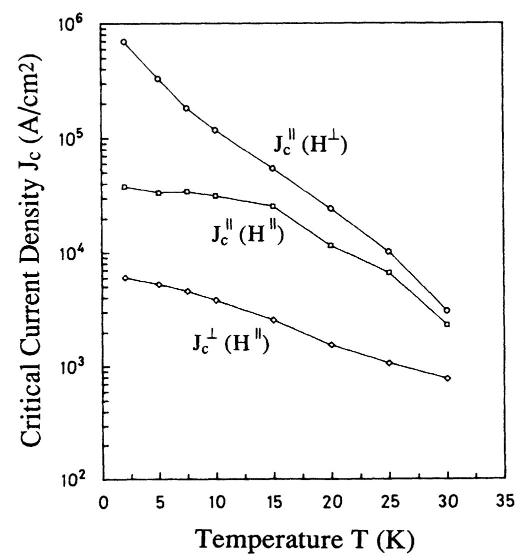

FIG. 4. Temperature dependences of the critical current densities of $(La_{1-x}Sr_x)_2CuO_4$ for the following three configurations: ($H\|$layer, $J_c\perp$layer), ($H\|$layer, $J_c\|$layer), and ($H\perp$layer, $J_c\|$layer).

of H_{c1} in high-T_c oxide superconductors might be unusual. For this issue, it can be concluded from the results of this study that the unusual temperature dependence of H_{c1} reported previously was incorrect due to insufficient analysis that did not account for the strong temperature dependence of the critical current density ($J_c^{\parallel}$) parallel to the layer. As apparent in Figs. 1(b) and 1(c), the magnetization curves grow markedly at low temperatures. However, this growth is not caused by the steep rise of H_{c1}, but by the steep rise of $J_c^{\parallel}$. This can be directly understood from the comparison between Figs. 1(a) and 1(b). A careful analysis based on the Bean's model also supports this conclusion. The temperature dependence of H_{c1} is normal as seen in Fig. 3, although it may be too soon to discuss it in terms of conventional theories such as the strong- or weak-coupling theory, or the dirty- or clean-limit theory. The following summarizes the anisotropic lower critical field values: $-(dH_{c1}^{\parallel}/dT)_{T_c}=18$ G/K, $-(dH_{c1}^{\perp}/dT)_{T_c}=55$ G/K, $H_{c1}^{\parallel}(T=0)=70$ G, $H_{c1}^{\perp}(T=0)=300$ G, $H_{c1}^{\perp}/H_{c1}^{\parallel}=\sim 3.0$ $(T\sim T_c)$, and ~ 4.2 $(T\sim 0$ K).

In contrast, the critical current density does have an unusual temperature dependence. The flux-creep theory[16] predicts that $J_c(T)$ should depend linearly on the temperature: $J_c(T)=J_c(0)(1-k_BT\ln t/U)$, where U is the magnitude of the pinning potential and t is the time scale for magnetic measurements (typically several tens of minutes to an hour). The observed temperature dependence is much stronger than this prediction and nearly follows $\exp(-T/T_0)$, where $T_0=5.3$ K and 14 K for J_c parallel and perpendicular to the layer, respectively. A nearly exponential dependence of J_c on the temperature as observed here has been reported from the magnetization hysteresis measurements on $YBa_2Cu_3O_7$ at much higher fields as well.[17] Generally speaking the critical current density is not an intrinsic parameter of superconducting materials, but a structure-sensitive parameter depending on the metallurgical processing of materials. However, the pinning mechanisms for high-T_c oxide superconductors seem to be dominated more by the microstructure of a finer length scale than do those of conventional superconductors because of the short coherence lengths. This may suggest that the critical current density of high-T_c oxide superconductors is a *quasi-intrinsic* parameter, and hence, the temperature dependence of J_c should be examined more closely from a microscopic viewpoint.

In summary, the anisotropic lower critical fields of single crystal $(La_{1-x}Sr_x)_2CuO_4$ were determined by using a novel procedure to accurately pick up the onset of flux penetration from the broad M-H curves. It was concluded from this evaluation that the temperature dependence of H_{c1} is essentially "normal" and that, instead, the critical current density has a strong temperature dependence, even at low temperatures.

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