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Angular dependence of the upper critical field in $Bi_2Sr_2CuO_{6+\delta}$

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The angular dependence of the upper critical magnetic field has been investigated in a wide range of temperatures in very high-quality $Bi_2Sr_2CuO_{6+\delta}$ single crystals with critical temperature T_c (midpoint) $\simeq 9$ K in magnetic fields up to 28 T. Although the typical value of the normal state resistivity ratio $\rho_c/\rho_{ab} \approx 10^4$, the anisotropy ratio $H_{c2\|ab}$ / $H_{c2\perp ab}$ of the upper critical fields is much smaller and shows an unexpected temperature dependence. A model is proposed based on a strong anisotropy and a small transparency between superconducting layers.

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One of the puzzling phenomena of high-T_c superconductors (HTSC) is the anomalous positive curvature of the temperature dependence of the upper critical field H_{c2}, which has been observed in some superconducting oxides [1 – 4]. The magnitude of the critical field at zero temperature was far in excess of the Werthamer-Helfand-Hohenberg extrapolation [5] and any quadratic saturation of H_{c2} was not found down to temperatures in the mK range for low-critical-temperature $Tl_2Ba_2CuO_6$ single crystals [1] and $Bi_2Sr_2CuO_6$ films [2]. In addition, it was deduced from measurements in 61-T pulsed magnetic fields applied parallel to the c-axis that the shape of $H_{c2}(T)$ in the high-T_c cuprates depends on the anisotropy of the materials, becoming more conventional as the normal state anisotropy gets smaller [6]. The authors argued that the normal state anisotropy plays a key role in determining the curvature of $H_{c2}(T)$ [6]. However, in the different models [7 – 9] which have been proposed to account for the upward curvature in $H_{c2}(T)$, the effect of anisotropy is not so important. This problem of anisotropy in the superconducting properties is related to the more general question for the understanding of high-T_c superconductivity on the basis of the normal-state properties.

In transport measurements on the low-T_c phase $Bi_2Sr_2CuO_6$ (Bi2201) we obtained a much smaller (nearly two orders of magnitude) anisotropy for the superconducting critical field than for the normal-state resistivity. To explain this contrasting behavior with respect to the situation for $Bi_2Sr_2CaCu_2O_8$ (Bi2212) where both measured anisotropies are comparable [10], we analysed the angular dependence of the upper critical field in a model based on weakly-coupled superconducting layers.

The anisotropy is usually expressed by the ratio $\gamma = \sqrt{m_c/m_{ab}}$ between the effective masses of the quasiparticles along the c-axis and the ab-plane, which can be related to the transport anisotropy with $\gamma \simeq \simeq \sqrt{\rho_c/\rho_{ab}}$ using $\rho = m/ne^2\tau$ for the out-of-plane resistivity ρ_c and in-plane resistivity ρ_{ab} [8, 10]. For the three-dimensional (3D) limit with a superconducting coherence length ξ_c larger than an interlayer distance s, the anisotropic Ginzburg-Landau (AGL) relation $H_{c2}(\theta) = H_{c2\|ab}(\cos^2\theta + \gamma^2\sin^2\theta)^{-0.5}$ describes the angular dependence of the upper critical field with $\gamma = H_{c2\|ab}/H_{c2\perp ab} = \sqrt{m_c/m_{ab}}$ for the applied field $H \parallel ab$-plane and $H \perp ab$-plane [11]. Here, θ is the angle between the magnetic field and the ab-plane. For layered superconductors with a high degree of anisotropy, such that $\xi_c(T) < s$, a 2D situation with decoupled layers arises. For such a thin-film superconductor in the vicinity of a critical temperature T_c (in the GL approximation), Tinkham has proposed a qualitative model with the angluar dependence $|H_{c2}(\theta)\sin\theta/H_{c2\perp ab}| + + [H_{c2}(\theta)\cos\theta/H_{c2\|ab}]^2 = 1$ [12]. The thin-film model results in a cusp at $\theta = 0°$ with $dH_{c2}/d\theta \neq 0$, which behaviour has been observed in superconducting multilayers [13]. An important feature is that the both

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models predict a temperature-independent critic-field anisotropy $H_{c2\|ab}/H_{c2\perp ab}$.

For $YBa_2Cu_3O_7$ with a not too strong anisotropy (typically $\gamma \simeq 10-30$), there are indications for a dimensional crossover from 3D to 2D with decreasing temperature [14, 15]. For Bi2212 with γ values up to 1000, deviations from the AGL theory have been observed in resistivity experiments in the vicinity of T_c by Palstra et al. [16], but without a clear indication of the dimensional crossover. In similar experiments with a better angular resolution, Marcon et al. [17] and Naughton et al. [18] have found the 2D behaviour in Bi2212 crystals. Because of the high H_{c2} values in these systems, a direct determination of the H_{c2} anisotropy can only been done close to T_c. To extend the temperature region further below T_c, the anisotropy of irreversibility has been investigated.

The low T_c of Bi2201 does not restrict the critical field studies to temperatures close to T_c while its structure and properties are closely related to HTSC. Single crystals of the pure Bi2201-phase are difficult to grow, because the crystals are nonstoichiometric and, as a rule, not perfect. For this reason, most of the measurements were carried out on single-phase La-doped Bi2201 single crystals. The three slightly overdoped crystals investigated in this study were grown without doping by a KCL-solution-melt method in the stoichiometry $Bi_{2+x}Sr_{2-(x+y)}Cu_{1+y}O_{6+\delta}$ with Bi excess in order to have good quality single crystals [19]. The zero-field critical temperatures defined by the 10% and 90% points of the resistive transition equal 8.1 – 9.8, 8.7 – 9.5, and 8.1 – 8.9 K for samples No. 1, 2, and 3, respectively. In the inset of Fig.1 we have plotted the temperature dependence of the resistive transition for sample No. 1. In the four-probe resistance measurements the transport current was in the ab-plane of the crystals and orthogonal to the field in all cases. The angular resolution was better than half a degree with the $\theta = 0°$ orientation obtained from the highest value of $H_{c2}(0)_{c2\|ab}$.

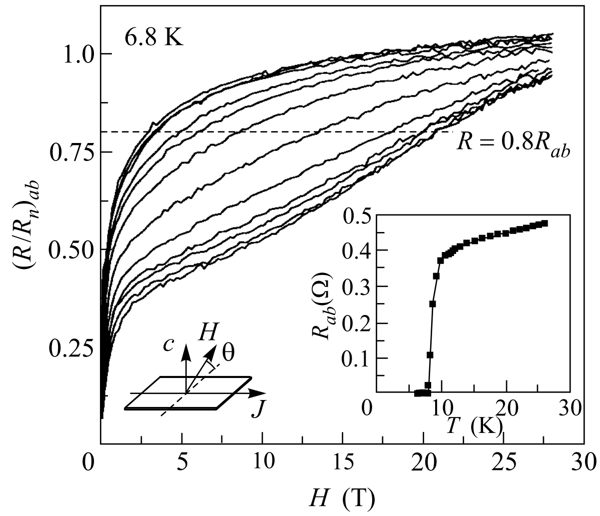

Fig.1. In-plane resistive transitions of sample No. 1 as a function of applied field for various field orientations relative to the ab-plane of the crystal at 6.8 K. Angle θ from above is 90°, 72°, 54°, 36°, 27°, 18°, 9°, 4.5°, 2.7°, 1.8°, 0.9°, 0°. The resistance has been normalized to extrapolated normal-state resistance R_n at the highest fields. The inset shows the temperature-dependent superconducting transition of the same sample

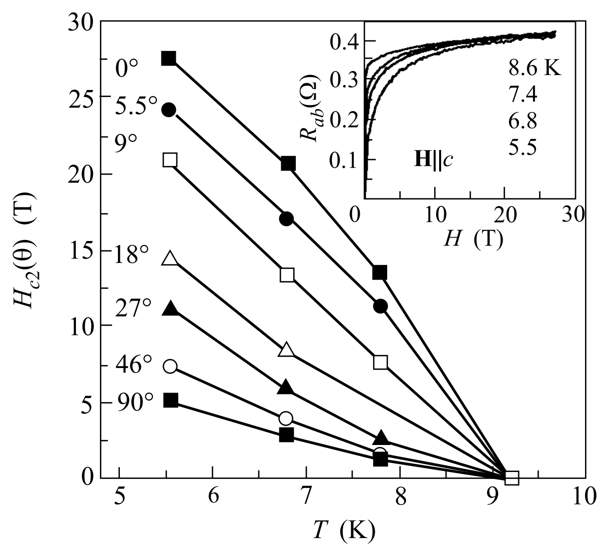

Fig.2. Temperature dependencies of the upper critical field $H^*_{c2}(T)$ for sample No. 1 at various angles θ extracted from the 80 %-resistive transitions of the crystals. The inset shows the magnetoresistance curves for the same sample at various temperatures with the field direction perpendicular to the ab-plane of the crystal

Fig.1 shows the in-plane resistive transitions for sample No. 1 at 6.8 K as a function of applied field for various field orientations relative to the ab-plane of the crystal. The resistance has been normalized to the extrapolated high-field normal-state resistance R_n. The inset in Fig.2 shows the magnetoresistance curves for the same sample at various temperatures with the field direction perpendicular to the ab-plane of the crystal. In spite of the strong broadening of the magnetic transitions at high temperatures one can see in the inset that the resistive transitions in the normal state are completed at $H > 13$ T, even at $T = 5.5$ K (the weak increase of the normal-state resistance is due to a magnetoresistance contribution in high magnetic fields).

The field induced resistive transitions for samples No. 2 and 3 were similar to those shown in Fig.1. The influence of the flux-flow dissipation on the broadening of the superconducting transition, becomes less noticeable for a critical field determination close to R_n [16, 20, 6]. In support of this, we display in Fig.2 the $H^*_{c2}(T)$ phase diagram for sample No. 1 at various angles θ obtained from a 80% criterion of its normal-state value R_n. For the lowest θ values these temperature dependencies are analogous to the dependence for conventional type II superconductors. The phase diagram obtained from the $R = 0.5R_n$ criterion exhibited for all field orientations a strong anomalous concave upturn of $H^*_{c2}(T)$.

In Fig.3 we show $H^*_{c2}(\theta)$ for sample No. 1 at 6.8 and 5.55 K. The inset shows similar data for sample No. 2 at 1.42 and 0.82 K and for sample No. 3 at 4.2 K (data points). For the angular dependence $H^*_{c2}(\theta)$ a good agreement is found with the 3D AGL model with anisotropy parameters $\gamma = H_{c2\|ab}/H_{c2\perp ab}$ equal to 9.8, 7.15 and 5.3 at 7.8, 6.8 and 5.55 K, respectively, (crystal No. 1). This anisotropy is much lower than $\gamma = \sqrt{\rho_c/\rho_{ab}}$ found from resistivity which equals 140 at 6 K. Moreover, the obtained anisotropy parameter γ varies with temperature which is unexpected for a similar temperature dependence of the critical fields in the two field orientations. We conclude therefore that the anisotropy parameter γ of our Bi2201 single crystals cannot be deduced from the angular dependence of $H_{c2}(\theta)$ using the available models as has been done before for Bi2212.

A temperature-dependent critical-field anisotropy was observed in layered low-T_c 2H-TaS_2 [21] and MoS_2 [22] single crystals intercalated with a variety of organic molecules and alkali metals. The data have been analyzed using the theory of dimensional crossover developed by Klemm et al. [23]. Klemm et al. have extended the Lawrence-Doniach model [24] and found the conditions necessary for observing crossover to a 2D behavior characterized by the temperature-dependent critical-field anisotropy and a strong upward curvature in $H_{c2\|ab}$ and vs T. In our case the $H^*_{c2\|ab}(T)$ phase diagram obtained from a 80% criterion of its normal-state value R_n does not show the upward curvature in $H^*_{c2\|ab}$ and is analogous to the dependence for conventional type II superconductors (Fig. 2 and Ref. [20]).

In the following we propose a model for the observed critical-field anisotropy based on a superconductor with a high degree of anisotropy consisting of stacked two-dimensional superconducting planes with an effective thickness d coupled by weak Josephson coupling [24]. The upper critical field $H_{c2\|ab}$ is determined by the depairing currents in the ab-plane, and will be finite even for $\rho_c/\rho_{ab} \to \infty$. We neglect spin effects such that $H_{c2\|ab}$ remains smaller than the paramagnetic limit. In this model, the upper critical field at an arbitrary orientation with respect to the crystal and an arbitrary temperature is determined by lowest eigenvalues of the operator $\widehat{L} = -(\hbar\frac{\partial}{\partial \mathbf{r}} - \frac{1}{c}2ie\mathbf{A})^2$, where $\mathbf{A}$ is the vector potential. For the magnetic field H oriented at an angle θ to the ab-plane (xy-plane), the vector potential $\mathbf{A}$ can be written as

$$\mathbf{A} = (-Hy\sin\theta + Hz\cos\theta, 0, 0), \qquad (1)$$

where z lies in the range of $-d/2 < z < d/2$. Under the assumption that the effective thickness d is less than the correlation length in the ab-plane of crystal, the order parameter will be independent of x and z coordinates. In a strongly layered anisotropic superconductor for a magnetic field directed along one of general axes of the crystal, the vortex cross-section has the shape of an ellipse. At low transparency between layers, the small half-axis of the vortex can be less than d. The effective mass approximation is no longer valid. In this case currents between the superconducting layers may be neglected and we can consider one single isolated layer with effective thickness d. As a result, the operator $\widehat{L}$ is given by

$$\widehat{L} = -\hbar^2\frac{\partial^2}{\partial y^2} + \frac{4e^2}{c^2}H^2y^2\sin^2\theta + \frac{4e^2}{c^2}H^2\cos^2\theta < z^2 >, \qquad (2)$$

where $< z^2 >= d^2/12$. From Eq. (2) we obtain the relationship between the angular dependent critical field $H(\theta)$ and the upper critical field $H_{c2\perp ab}$:

$$H(\theta)\sin\theta + \gamma' H^2(\theta)\cos^2\theta = H_{c2\perp ab}, \qquad (3)$$

where $\gamma' \simeq ed^2/6$. This expression is similar to the one given above for thin-film superconductors as proposed by Tinkham [12] and Harper and Tinkham [25], but the coefficient γ' is now a material constant and hence temperature-independent. This equation is valid over a wide temperature region except for temperatures in the vicinity of T_c (see below). From a rough estimation from the experimental data in Fig.3, we obtain the temperature-independent value $\gamma' = H_{c2\perp ab}/H^2_{c2\|ab} \simeq$ $\simeq 0.0068\ T^{-1}$.

For small θ, we do not observe the expected cusp-like structure for the 2D model descibed by Eq. (3). In practice, the CuO_2 layers in the crystal are slightly misoriented with respect to the distribution of c-axis orientations. Also defects may cause an enhancement of a link between layers and thus increase the effective thickness of the superconducting layers. Such crystal imperfections have no influence for high angles θ. For small θ, the expression for the angular dependence of the upper critical field $H_{c2}(\theta)$ can be rewritten with a Gaussian distribution of the c-axis orientations on a sphere

$$H_{c2}(\theta) = \frac{\beta}{\gamma'} \int_0^\infty du\; u e^{-\beta u^2} \frac{\sin\sqrt{\theta^2+u^2}}{\cos^2\theta} \times$$

$$\times \left[\sqrt{1 + \frac{4H_{c2\perp ab}\gamma' \cos^2\theta}{\sin^2\sqrt{\theta^2+u^2}}} - 1 \right], \qquad (4)$$

where $\beta >> 1$ determines the angular width in the misalignment of the CuO_2 layers and the possible existence of shorted layers in the crystal. The solid curves shown in Fig.3 are fits of Eq. (4) to our experimental data for sample No. 1 using β as a temperature independent parameter. The inset shows the same data for samples No. 2 and 3. With the dashed lines we have shown the cusp-like structure for $\beta \to \infty$.

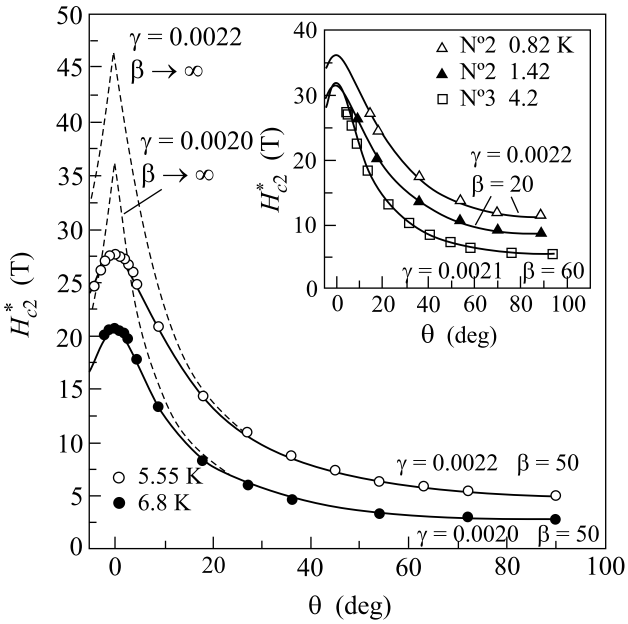

Fig.3. Angular dependence of the upper critical field H^*_{c2} for sample No. 1 at 6.8 and 5.55 K. The inset shows similar data for the samples No. 2 and 3 at lower temperatures. Full lines are fits of Eq. (4) to the experimental data for sample No. 1, 2 and 3 (see also inset) with the indicated β and γ' parameters. The long dashed lines show the result without angular broadening ($\beta \to \infty$)

Using this analysis (at $\beta \to \infty$) of the critical-field data, we obtained the same parameter $\gamma' = 0.0021 \pm \pm 0.0001$ T^{-1} for samples No. 1, 2 and 3. Because the angular with in the misalignment of the CuO_2 layers in the investigated crystals is much less than we obtained from the parameter γ' (these crystals showed X-ray rocking curves with a width of about $0.1° - 0.3°$), this means that even a small proportion of the shorted layers is of first importance. From this value of γ' we evaluate an effective thickness of the superconducting layers $d \simeq \sqrt{6\hbar c\gamma'/e} = 2.7 \cdot 10^{-7}$ cm. This value is close to the lattice parameter along c-axis ($2.46 \cdot 10^{-7}$ cm) which looks reasonable. The critical-field anisotropy $\gamma = H_{c2\|ab}/H_{c2\perp ab}$ depends now on temperature. Because the temperature dependence in Eq. (3) is only determined by the temperture dependence of $H_{c2\perp ab}$ ($\sim 1 - T/T_c$ for $T_c - T << T_c$), one gets $\gamma \sim \sim 1/\sqrt{1-T/T_c}$. From measured values of $H^*_{c2\perp ab}(T)$ data and the value of γ', we determined the magnitude of $H^*_{c2\|ab}(T)$. Using a linear extrapolation of $H^*_{c2\|ab}(T)$ and $H^*_{c2\perp ab}(T)$ from T_c to zero temperature, we found $H^*_{c2\|ab}(0) \simeq 90$ T and $H^*_{c2\perp ab}(0) \simeq 16$ T for sample No. 1 and 2 yielding $H^*_{c2\|ab}/H^*_{c2\perp ab} \simeq 5.6$ at $T = 0$ K.

The thin-film approximation ($\xi_c < s$) holds for $H_{c2}(\theta) m_c/m_{ab} > 1/\gamma'$. Near T_c with $H_{c2} \sim 1 - T/T_c$ and at sufficiently large mass ratio m_c/m_{ab}, there is only a narrow region near T_c where this inequality does not hold and an effective mass approach with a diffusion tensor becomes more adequate. In the studied crystals at $T = 6.8$ K, $H^*_{c2\perp ab} = 2.9$ T, $\rho_c/\rho_{ab} = 2 \cdot 10^4$, and $\gamma' = 0.0021$ this condition breaks only down at $1 - T/T_c \simeq 0.002$ which limit may not be reached in view of the broadening of the superconducting transitions.

In conclusion, the effective-mass approximation is not suitable for layered superconductors with a very low transparency between the superconducting layers. In a magnetic field parallel to layer, the vortex cross-section has the shape of ellipse with the small half-axis much less than the lattice parameter along c-axis of the crystal. For this high degree of anisotropy, the proposed model of weakly-coupled superconducting layers allows one to explain the observed angular dependence of the upper critical field.

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1. A. P. Mackenzie, S. R. Julian, G. G. Lonzarich et al., Phys. Rev. Lett. **71**, 1238 (1993).

2. M. S. Osofsky, R. J. Soulen, Jr., S. A. Wolf et al., Phys. Rev. Lett. **71**, 2315 (1993).
3. D. J. C. Walker, O. Laborde, A. P. Mackenzie et al., Phys. Rev. **B51**, 9375 (1995).
4. K. Karpinska, A. Malinowski, Marta Z. Cieplak et al., Phys. Rev. Lett. **77**, 3033 (1997).
5. N. R. Werthamer, E. Helfand, and P. C. Hohenberg, Phys. Rev. **147**, 295 (1966).
6. Y. Ando, G. S. Boebinger, A. Passner et al., Phys. Rev. **B60**, 12 475 (1999).
7. Yu. N. Ovchinnikov and V. Z. Kresin, Phys. Rev. **B52**, 3075 (1995).
8. G. Kotliar and C. M. Varma, Phys. Rev. Lett. **77**, 2296 (1996).
9. T. Koyama and M. Tachiki, Physica **C263**, 25 (1996).
10. A. Schilling, R. Jin, J. D. Guo, and H. R. Ott, Phys. Rev. Lett. **71**, 1899 (1993).
11. V. G. Kogan, Phys. Rev. **B24**, 1572 (1981).
12. M. Tinkham, Phys. Rev. **129**, 2413 (1963).
13. B. Y. Jin, J. B. Ketterson, E. J. McNiff et al., J. Low Temp. Phys. **69**, 39 (1987).
14. D. E. Farrell, J. P. Rice, D. M. Ginsberg, and J. Z. Liu, Phys. Rev. Lett. **64**, 1573 (1990).
15. B. Lundqvist, O. Rapp, and M. Andersson, Phys. Rev. **B62**, 3542 (2000), and references cited herein.
16. T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer et al., Phys. Rev. **B38**, R5102 (1988).
17. R. Marcon, E. Silva, R. Fastampa, and M. Giura, Phys. Rev. **B46**, 3612 (1992).
18. M. J. Naughton, R. C. Yu, P. K. Davies et al., Phys. Rev. **B38**, R9280 (1988).
19. V. P. Martovitsky, J. I. Gorina, and G. A. Kaljushnaia, Solid State Commun. **96**, 893 (1995); J. I. Gorina, G. A. Kaljushnaia, V. P. Martovitsky et al., Solid State Commun. **108**, 275 (1998).
20. S. I. Vedeneev, A. G. M. Jansen, E. Haanappel, and P. Wyder, Phys. Rev. **B60**, 12 467 (1999).
21. R. V. Coleman, G. K. Eiserman, S. J. Hillenius et al., Phys. Rev. **B27**, 125 (1983).
22. J. A. Woollam and R. B. Somoano, Phys. Rev. **B13**, 3843 (1976).
23. R. A. Klemm, A. Luther, and M. R. Beasley, Phys. Rev. **B12**, 877 (1975).
24. W. Lawrence and S. Doniach, in *Proc. 12th Int. Conf. Low Temp. Phys.*, Ed. E. Kanda, Keigaku, Tokyo, 1971, p. 361.
25. F. E. Harper and M. Tinkham, Phys. Rev. **B172**, 441 (1968).