Dimensional crossover in superlattice superconductors

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We have observed dimensional crossover from anisotropic three-dimensional behavior to two-dimensional behavior in both the temperature dependence and the angular dependence of the upper critical fields of Nb/Cu superlattice superconductors. The crossover occurs when the coherence length $\xi_\perp$ perpendicular to the layer planes is of the order of the copper layer thickness D_{Cu}. We have observed the crossover (1) by varying the temperature, thereby varying $\xi_\perp$ relative to D_{Cu} and (2) by varying D_{Cu} relative to $\xi_\perp$. The complicating effects due to surface superconductivity have been eliminated by placing thick copper films as the outside layers of the samples. The measurements are in qualitative agreement with theoretical models of layered superconductors.

I. INTRODUCTION

Dimensional crossover from three-dimensional (3D) to two-dimensional (2D) behavior in layered superconductors has been the subject of detailed studies over the years. This crossover should be observable in both the temperature dependence of the parallel (to the layers) upper critical field $H_{c2\|}(T)$ and the angular dependence of the upper critical field $H_{c2}(\theta)$ (θ is the angle between the applied field and the plane of the layers).[1–4] Among the layered superconductors previously investigated are naturally occuring layered compounds, e.g., intercalated transition-metal dichalcogenides,[5,6] and artificially grown layered materials, e.g., Nb/Ge and Nb/Cu.[7–12] Many of these experiments strongly suggest the existence of dimensional crossover, especially in the dependence of $H_{c2\|}$ on temperature. However, to our knowledge, dimensional crossover in both the temperature dependence $H_{c2\|}(T)$ and the angular dependence $H_{c2}(\theta)$ has not yet been reported.

In this paper we report the observation of dimensional crossover in *both* the temperature dependence of $H_{c2\|}$ and the angular dependence of H_{c2} for Nb/Cu superlattices. This crossover occurs when a simple criterion is met: The perpendicular coherence length $\xi_\perp$ is approximately equal to the separation of the superconductor layers (the copper layer thickness D_{Cu} in our case). When $\xi_\perp \gg D_{Cu}$, many superconducting layers are coupled and 3D behavior is expected and observed. When $\xi_\perp \ll D_{Cu}$, the superconducting layers are decoupled. If, in addition, the niobium layer thickness D_{Nb} is less than the Ginzburg-Landau coherence length of bulk niobium, the Nb/Cu superlattice exhibits 2D behavior. We have observed this 3D-to-2D crossover in both the temperature dependence $H_{c2\|}(T)$ and the angular dependence $H_{c2}(\theta)$.

Previous experiments (e.g., intercalated transition-metal dichalcogenides[5,6] and Nb/Ge superlattices[7,9]) were performed on superconducting layers coupled by tunneling through insulating layers. This relatively weak coupling required very careful control of a very small separation (~ 10 Å) between the superconducting layers. In contrast, for the present experiment with Nb/Cu superlattices, the superconducting Nb layers are coupled through the Cu layers by the proximity effect.[10,11] This relatively strong coupling allows a larger separation (~ 100 Å) between superconducting layers which can be controlled more accurately. In addition, the results of previous experiments was complicated by the possible existence of surface superconductivity.[9–12] This interference by surface superconductivity in the measurement of H_{c2} was reported earlier for Nb/Cu samples.[10–12] In this experiment, surface superconductivity has been eliminated by adding thick (1500-Å) films of copper as the first and last layers. The method of preparation of the Nb/Cu-superlattice samples, the crystallographic analysis of their structure,[13] and the experimental details of the H_{c2} measurement have been described earlier.[10–12]

This paper is organized as follows. Section II will briefly review the theory of anisotropic critical fields and dimensional crossover for layered superconductors. The results of our measurements on the temperature dependence of upper critical fields in Nb/Cu and the observation of dimensional crossover will be described in Sec. III. The corresponding dimensional crossover behavior in the angular dependence will be discussed in Sec. IV. Table I provides a detailed summary of the samples included in this paper.

II. THEORY

Lawrence and Doniach developed a model for layered superconductors based on an anisotropic Ginzburg-Landau theory having different coherence lengths parallel ($\xi_\|$) and perpendicular ($\xi_\perp$) to the plane of the layers.[1] The upper critical fields in this model are given by

$$H_{c2\|}(T)=\frac{\Phi_0}{2\pi\xi_\|(T)\xi_\perp(T)} \tag{1}$$

and

$$H_{c2\perp}(T)=\frac{\Phi_0}{2\pi\xi_\|(T)^2}\ . \tag{2}$$

The angular dependence of the upper critical field is

TABLE I. Nb/Cu-superlattice sample parameters.

D_{Nb} (Å)	D_{Cu} (Å)	T_c (K)	T (K)	$H_{c2\parallel}$ (kG)	$H_{c2\perp}$ (kG)	$\xi_\perp$ (Å)
23	23	3.34	1.17	12.5	8.75	136
47	22	6.05	1.17	24.0	17.6	100
44	44	5.07	1.17	13.3	9.16	131
47	87	3.05	1.17	2.75	2.08	301
168	147	7.00	1.37	26.9	12.2	74.5
168	147	7.00	3.33	12.2	8.03	134
168	147	7.00	4.86	5.83	4.64	212
168	147	7.00	5.28	4.74	3.78	235
168	147	7.00	6.04	2.78	2.07	297
168	147	7.00	6.62	1.07	.749	463
171	376	5.85	1.17	23.4	7.47	67.1
171	376	5.85	3.04	11.4	4.04	101
171	376	5.85	3.63	5.63	2.91	174
171	376	5.85	4.08	3.56	2.16	237
171	376	5.85	4.47	2.75	1.51	256
171	376	5.85	4.86	1.90	.97	298
171	376	5.85	5.28	.992	.485	403
171	376	5.85	5.41	.747	.404	489
171	376	5.85	5.62	.383	.189	651
170	90	7.70	1.17	38.7	16.4	60.1
168	168	6.75	1.17	27.1	10.6	68.9
168	168	6.75	1.96	21.7	9.27	80.4
168	168	6.75	5.28	4.15	2.59	222
172	255	6.50	1.56	21.4	8.67	79.0
172	333	6.05	1.10	26.5	9.86	68.0
172	333	6.05	1.17	26.9	9.54	65.8
176	585	4.84	1.10	35.3	8.68	48.0
176	585	4.84	3.33	9.52	2.13	88.1
176	585	4.84	3.77	2.50	1.19	250
172	1240	3.55	1.17	29.6	7.03	51.4

$$H_{c2}(\theta,T)=\frac{\Phi_0}{2\pi\xi_{\parallel}^2(T)[\sin^2\theta+(m/M)\cos^2\theta]^{1/2}} . \quad (3)$$

Here θ is the angle between the direction of the applied field and the plane of the layers, and

$$M/m=[H_{c2\parallel}(T)/H_{c2\perp}(T)]^2$$

is the anisotropy in the effective Ginzburg-Landau mass.

The upper critical field for a 2D superconducting film $[\xi(T)<D]$ is given by[14]

$$H_{c2\parallel}(T)=\frac{\Phi_0}{2\pi\xi(T)D/\sqrt{12}} \quad (4)$$

and

$$H_{c2\perp}(T)=\frac{\Phi_0}{2\pi\xi(T)^2} . \quad (5)$$

In a parallel magnetic field the vortex has its radius in the direction perpendicular to the film restricted to a length $\sim D/2$ which accounts for a factor $D/\sqrt{12}$ in the denominator of Eq. (4). The angular dependence is given implicity by the equation

$$\left|\frac{H_{c2}(\theta)\sin\theta}{H_{c2\perp}}\right|+\left[\frac{H_{c2}(\theta)\cos\theta}{H_{c2\parallel}}\right]^2=1 . \quad (6)$$

Near $\theta=0°$ there is a qualitative difference between anisotropic 3D and 2D behavior in the slope of $H_{c2}(\theta)$. For 3D behavior [Eq. (3)] the slope is zero, $dH_{c2}/d\theta\,|_{\theta=0°}=0$. For 2D behavior, Eq. (6) leads to

$$\left[\frac{1}{H_{c2}}\frac{dH_{c2}}{d\theta}\right|_{\theta=0°}=-\frac{H_{c2\parallel}}{H_{c2\perp}} . \quad (7)$$

It should be possible to observe a transition between anisotropic 3D behavior and 2D behavior in a stack of coupled thin superconducting layers. Each layer if isolated (weakly coupled) acts as a 2D superconductor and obeys Eq. (6). With strong coupling, the material will act as an anisotropic 3D superconductor with an upper critical field described by Eq. (3). Several theories have been developed with the coupling between superconducting layers being due to Josephson tunneling.[1–4] These theories predict that the crossover from 3D to 2D behavior occurs when the perpendicular coherence length $\xi_\perp$ is approximately equal to the separation between superconducting layers. Although no theory exists to date for proximity-effect-coupled layered superconductors, it seems reasonable to assume that the same condition defines the crossover region for Nb/Cu, i.e., $\xi_\perp \approx D_{Cu}$.

Therefore, the crossover from 3D to 2D behavior can be observed by varying the temperature, thereby changing

$\xi_\perp$ with respect to D_{Cu}, or by varying the separation D_{Cu} between the superconducting layers at fixed temperature.

III. TEMPERATURE DEPENDENCE

Figure 1 shows the critical fields as a function of temperature for a thick niobium layer [Cu(1500 Å)/Nb(8500 Å)/Cu(1500 Å)]. The $T=0$ Ginzburg-Landau coherence length ξ_0 estimated from this data is 125 Å. Since $D_{Nb} \gg \xi_0$, there is little anisotropy ($H_{c2\parallel} \approx H_{c2\perp}$), and $H_{c2} \propto 1-T/T_c$, as expected for an isotropic 3D superconductor. The addition of Cu(1500 Å) as the outermost layers supresses the anisotropy due to surface superconductivity. (The existence of surface superconductivity would lead to $H_{c2\parallel}/H_{c2\perp}=1.7$.[15]) The critical field for a thin ($D_{Nb} \sim \xi_0$) niobium film [Cu(1500 Å)/Nb(191 Å)/Cu(1500 Å)] is shown in Fig. 2. In agreement with Eqs. (4) and (5), the temperature dependences for this film is 2D-like: $H_{c2\parallel}(T) \propto (1-T/T_c)^{1/2}$ and $H_{c2\perp}(T) \propto 1-T/T_c$. The behavior shown in Figs. 1 and 2 has been observed for a large number of samples with essentially identical results.

Figure 3(a) shows H_{c2} values for a superlattice with $D_{Nb}=172$ Å and $D_{Cu}=333$ Å. At high temperatures near T_c, the sample behaves like a 3D superconductor ($H_{c2\parallel}$ is linear in T), and at lower temperatures, $T \lesssim 3$ K, it behaves like a 2D superconductor ($H_{c2\parallel}$ is square root in T [Eq. (4)]).[16] The perpendicular coherence length obtained using Eqs. (1) and (2) is given by

$$\xi_\perp(T) = \left[\frac{\Phi_0}{2\pi H_{c2\perp}(T)} \right]^{1/2} \frac{H_{c2\perp}(T)}{H_{c2\parallel}(T)} . \quad (8)$$

$\xi_\perp$ calculated in this fashion from the data of Fig. 3(a) is plotted in Fig. 3(b). In agreement with the theoretical expectations discussed in Sec. II, dimensional crossover occurs when $\xi_\perp \sim D_{Cu}$.

Dimensional crossover can also be observed by varying the copper layer thickness. The 3D-to-2D transition is clearly shown in a plot of the anisotropy $H_{c2\parallel}/H_{c2\perp}$ versus the ratio $\xi_\perp/D_{Cu}$ (Fig. 4). The anisotropy is constant to about $\xi_\perp/D_{Cu} \approx 0.6$, and then it suddenly increases in a universal fashion for a variety of different copper thicknesses ($90 < D_{Cu} < 1240$ Å) and temperatures (≈ 1.2 K $\leq T < T_c$). This is in agreement with a theoretical model[3] of layered superconductors that suggests that the increase in $H_{c2\parallel}/H_{c2\perp}$ should occur at a value of $\xi_\perp/D_{Cu} = 1/\sqrt{2} \approx 0.7$.

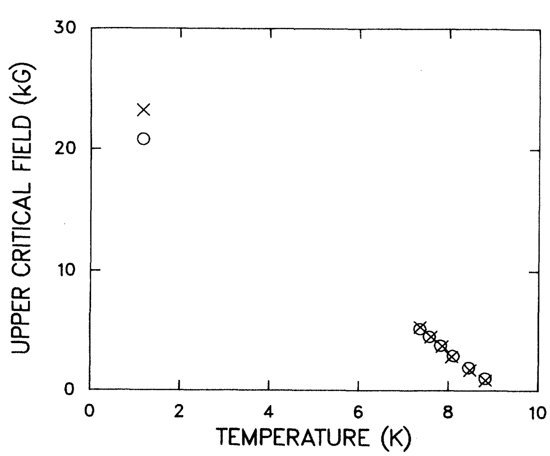

FIG. 1. Critical fields for a 3D, thick niobium film [Cu(1500 Å)/Nb(8500 Å)/Cu(1500 Å)]; ×, $H_{c2\parallel}$; ○, $H_{c2\perp}$.

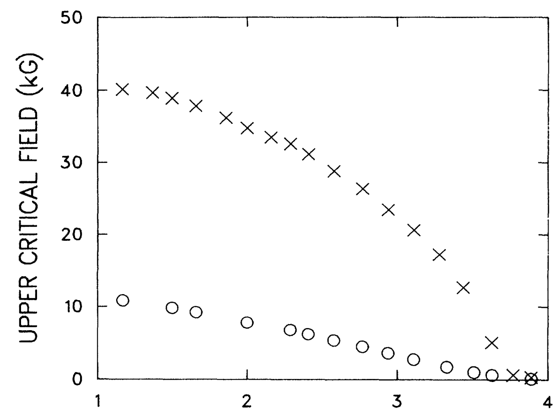

FIG. 2. Critical fields for a 2D, thin niobium film [Cu(1500 Å)/Nb(191 Å)/Cu(1500 Å)]; ×, $H_{c2\parallel}$; ○, $H_{c2\perp}$.

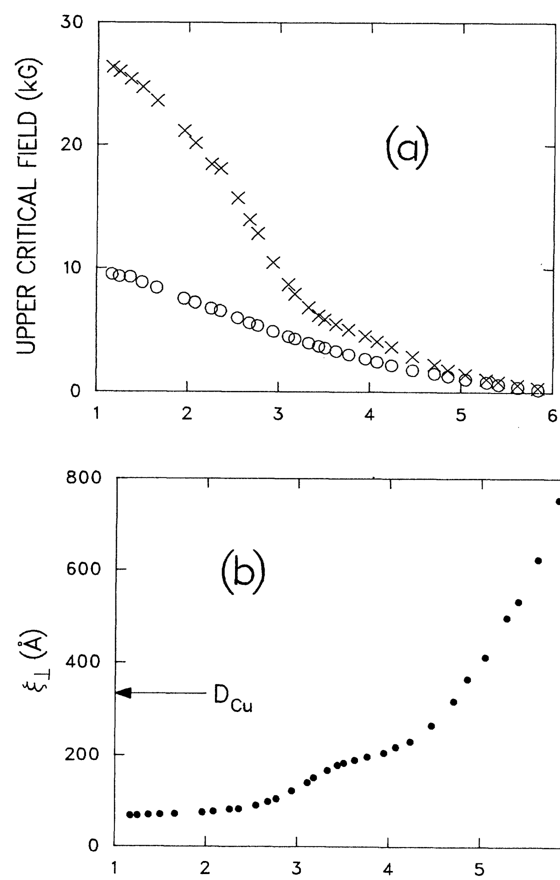

FIG. 3. (a) Upper critical fields for a Nb/Cu superlattice. $D_{Nb}=172$ Å and $D_{Cu}=333$ Å (1500 Å of Cu as the outside layers); ×, $H_{c2\parallel}$; ○, $H_{c2\perp}$. (b) Perpendicular coherence lengths calculated using Eq. (8) for the same sample.

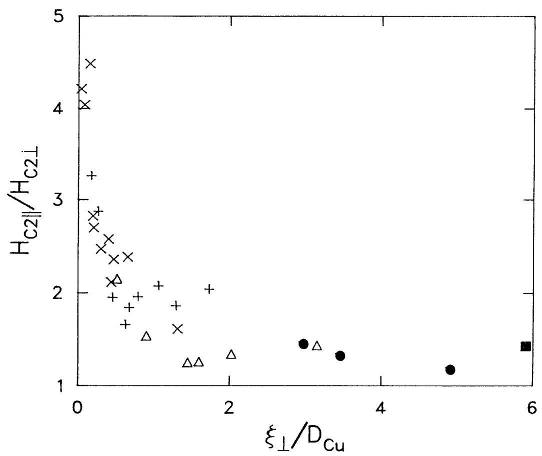

FIG. 4. Critical-field anisotropy vs $\xi_\perp/D_{Cu}$ at various temperatures. $+$, $D_{Nb}=171$ Å and $D_{Cu}=376$ Å; $\triangle$, $D_{Nb}=168$ Å and $D_{Cu}=147$ Å; $\times$, $168 \leq D_{Nb} \leq 176$ Å and $90 \leq D_{Cu} \leq 1240$ Å; $\bullet$, $44 \leq D_{Nb} \leq 47$ Å and $22 \leq D_{Cu} \leq 87$ Å; $\blacksquare$, $D_{Nb}=23$ Å and $D_{Cu}=23$ Å.

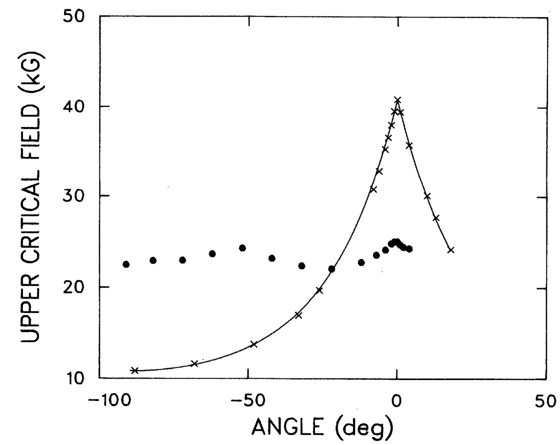

FIG. 5. Angular dependence of critical fields for samples with a single niobium layer (same samples as shown in Figs. 1 and 2). $\bullet$, thick niobium [Cu(1500 Å)/Nb(8500 Å)/Cu(1500 Å) at 1.17 K]; $\times$, thin niobium [Cu(1500 Å)/Nb(191 Å)/Cu(1500 Å) at 1.17 K]. The solid curve is a fit to the Tinkham curve [Eq. (6)] for the sample with a thin niobium layer.

IV. ANGULAR DEPENDENCE

The angular dependence of the upper critical field is shown in Fig. 5 for a thick ($D_{Nb} \gg \xi$) and a thin ($D_{Nb} \lesssim \xi$) niobium layer whose temperature dependences were shown in Figs. 1 and 2. The thick niobium layer, which showed 3D-type temperature dependence, shows a very small anisotropy with negligible surface superconductivity effects. The thin niobium layer, which shows 2D-type temperature dependence, has an angular dependence which matches very well with the Tinkham curve [Eq. (6)] expected for a 2D superconductor. Therefore, a single niobium layer shows the proper behavior in the 3D and 2D limits, in temperature as well as angular dependences.

Figure 6(a) shows the upper-critical-field curve for a sample with $D_{Nb}=D_{Cu}=23$ Å. The angular dependence is in qualitative agreement with the expected behavior for an anisotropic 3D superconductor [Eq. (3)]. In particular $dH_{c2}/d\theta\,|_{\theta=0^\circ}=0$.

The angular dependence of H_{c2} for a Nb/Cu sample with 2D behavior is shown in Fig. 6(b). The angular dependences were taken at a low temperature (1.17 K) where $\xi_\perp=70$ Å $\ll D_{Cu}=333$ Å. The general characteristic which distinguishes 2D behavior (a cusp at parallel field [Eq. (6)]) from anisotropic 3D behavior (a rounded curve at parallel field [Eq. (3)]) is present in Fig. 6(b). At intermediate angles, however, the data falls consistently below the Tinkham prediction. This tendency to fall below the Tinkham interpolation curve at intermediate angles is common to the Nb/Cu samples in the 2D region.

The upper-critical-field curves evolve continuously from a cusp-type behavior when $\xi_\perp=D_{Cu}$ to a rounded behavior when $\xi_\perp \simeq 6D_{Cu}$. This evolution is illustrated in Fig. 7. The sharpness of the cusp near parallel field is characterized by the logarithmic derivative of $H_{c2}(\theta)$ be-

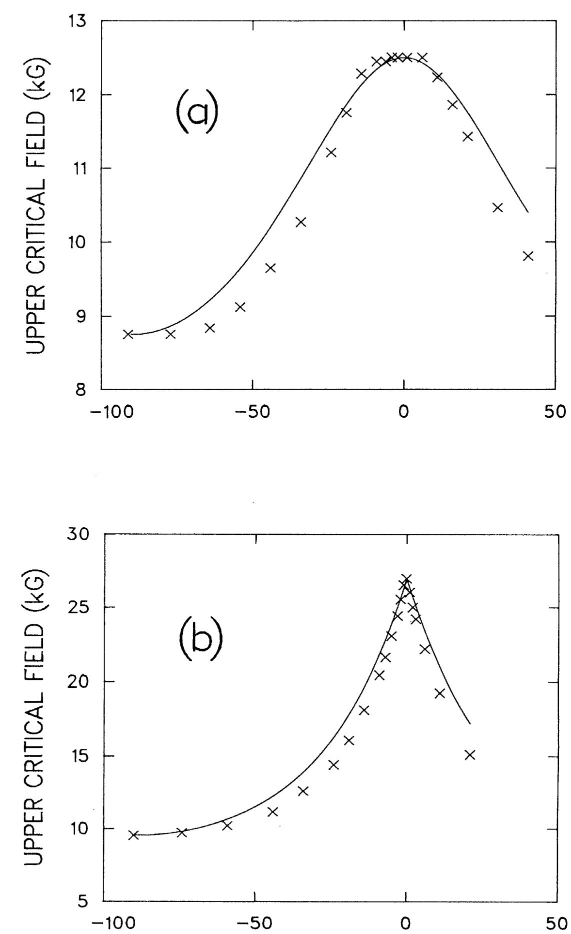

FIG. 6. Angular dependence of critical fields for a Nb/Cu superlattice showing (a) anisotropic 3D behavior ($D_{Nb}=23$ Å, $D_{Cu}=23$ Å, and $T=1.17$ K), and (b) 2D behavior ($D_{Nb}=172$ Å, $D_{Cu}=333$ Å, and $T=1.17$ K).

tween 0° and 10°. This logarithmic derivative is plotted, as is done traditionally, against the critical-field anisotropy $H_{c2\parallel}/H_{c2\perp}$.[17] In the discussion of Fig. 4 we concluded that the abrupt increase in $H_{c2\parallel}/H_{c2\perp}$ at $\xi_\perp/D_{Cu}\sim 0.6$ corresponds to the crossover from anisotropic 3D to 2D behavior. The angular dependences as shown in Fig. 7 quite clearly reflects this. If $H_{c2\parallel}/H_{c2\perp}\lesssim 1.5$, the logarithmic derivative is zero or near zero, as is expected for anisotropic 3D behavior. If $H_{c2\parallel}/H_{c2\perp}>2$, the logarithmic derivative is large, indicating a sharp cusp as expected for 2D behavior. The behavior of the intermediate samples is a smooth interpolation between these two regimes with no abrupt distinction. The solid line in Fig. 7 corresponds to the Tinkham angular dependence [Eq. (6)] for a single thin-film superconductor. As noted earlier, the cusp in $H_{c2}(\theta)$ in the 2D region is sharper than that predicted by Tinkman, and therefore the experimental data lies below the theoretical curve in the region $H_{c2\parallel}/H_{c2\perp}>2$.

In Fig. 7 we have combined data as a function of temperature for fixed D_{Cu} and data for several values of D_{Cu} at a variety of fixed temperatures. It is quite remarkable that all the data falls into a universal curve, showing again that the relevant physical parameter characterizing the dimensional behavior is $\xi_\perp/D_{Cu}$.

V. SUMMARY

We have observed dimensional crossover in Nb/Cu superlattices in the temperature and angular dependences of the superconducting critical fields. This crossover occurs when the coherence length $\xi_\perp$ perpendicular to the layer planes is of the order of the Cu layer thickness D_{Cu}. This criterion is expected intuitively and predicted by detailed theoretical models of layered superconductors. We have observed the crossover (1) by varying the temperature, and thereby varying $\xi_\perp$ relative to D_{Cu}, and (2) by varying D_{Cu} relative to $\xi_\perp$. The complicating effects of surface superconductivity have been eliminated by using thick copper layers as the outside layers of the samples.

This is the first report of the observation of dimensional crossover in the critical fields for layered superconductors where complicating effects due to surface superconductivity have been eliminated and the crossover is observed consistently in *both* the temperature and angular dependences.

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APPENDIX

It was shown in Sec. III that anisotropic 3D behavior implies a linear temperature dependence of $H_{c2\parallel}$ [Eq. (1)], whereas 2D behavior is typically square-root-like [Eq. (4)]. Naively, one might identify the crossover temperature as the temperature T^* at which the $H_{c2\parallel}$ curve turns sharply upward [3.2 K in Fig. 3(a)].

It is clear that as the copper thickness becomes smaller, the coupling between adjacent superconducting layers becomes stronger, and the dimensional crossover then should occur at a lower reduced temperature, T/T_c. The values T^*/T_c (solid circles in Fig. 8) decrease down to a thickness of $\approx$200 Å. However, below this thickness, T^* increases, approaching T_c, contrary to expectations. Clearly, T^* is not the crossover temperature.

We have shown that crossover occurs when the coher-

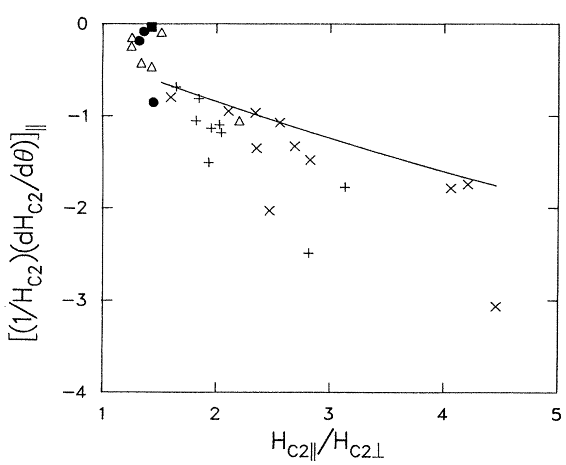

FIG. 7. Logarithmic derivative of $H_{c2}(\theta)$ near $\theta=0°$ vs critical-field anisotropy. +, $D_{Nb}=171$ Å and $D_{Cu}=376$ Å; △, $D_{Nb}=168$ Å and $D_{Cu}=147$ Å; ×, $168\leq D_{Nb}\leq 176$ Å and $90\leq D_{Cu}\leq 1240$ Å; ●, $44\leq D_{Nb}\leq 47$ Å and $22\leq D_{Cu}\leq 87$ Å; ■, $D_{Nb}=23$ Å and $D_{Cu}=23$ Å. The solid line is the logarithmic derivative of the Tinkham curve [Eq. (6)].

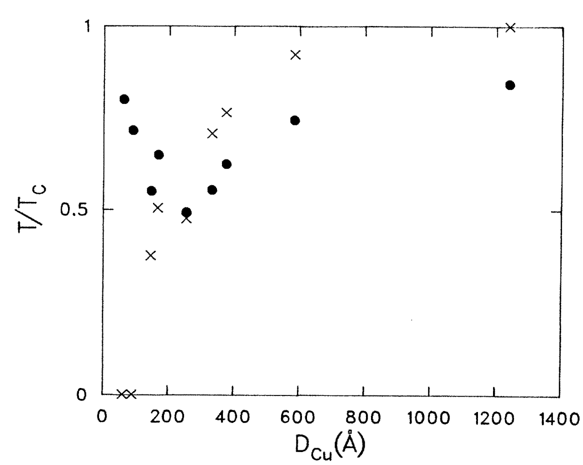

FIG. 8. ●, T^*/T_c—the reduced temperture at the upturn in the plot of $H_{c2\parallel}$ vs temperature; ×, $\hat{T}/T_c$—the reduced temperature at dimensional crossover, i.e., $\xi_\perp(\hat{T})=D_{Cu}/\sqrt{2}$.

ence length $\xi_\perp$ is of the order of D_{Cu}. Klemm, Beasley, and Luther suggest that the crossover temperature is defined by the condition $\xi_\perp(\hat{T})=D_{Cu}/\sqrt{2}$.[2] In agreement with our expectations, $\hat{T}/T_c$ (crosses in Fig. 8) decreases as a function of D_{Cu}. This leads us to conclude that the proper crossover temperature is $\hat{T}$ not T^*. The behavior of T^* [the temperature at which $H_{c2\parallel}(T)$ turns sharply upward] is not addressed by the Klemm-Beasley-Luther theory, and therefore its unusual copper thickness dependence is not unexpected.

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