Anisotropic critical fields in superconducting superlattices

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(Received 25 July 1983)

The temperature and angular dependence of critical fields (H_c) have been studied as a function of layer thickness for superconducting Nb/Cu superlattices. For layer thicknesses between 100 and 300 Å, dimensional crossover has been observed in the temperature dependence of H_c. Associated with the crossover we find a change in the angular dependence of H_c to that given by the effective-mass theory. This is the first time that a relationship has been found between dimensional crossover observed in the temperature dependence and that in the angular dependence of critical fields.

Layered superconductors, artificially prepared superlattices, and intercalated transition-metal dichalcogenides have received considerable attention recently because of the high degree of anisotropy in their physical properties.[1-3] For example, they show unusually high superconducting critical fields when the magnetic field is applied parallel to the layers. The superconductive coupling between the layers changes with layer spacing, and so it is possible to tune the character of the system in a continuous fashion from where the layers are three dimensional (3D) and weakly coupled to where the layers are two dimensional (2D) but strongly coupled (Fig. 1). In the intermediate region, which we designated as a "2D-coupled" system, dimensional crossover can take place as a function of the temperature because the superconducting coherence length is strongly temperature dependent. The crossover phenomenon has been a subject of several theoretical and experimental investigations.[4,5] Lawrence and Doniach[6] (LD) have proposed a theory of critical-field anisotropy for a set of 2D superconductors *coupled by the Josephson effect.* In this study we show that such a crossover has indeed been observed in a purely metallic *proximity-effect—coupled* system. Furthermore, we have observed for the first time, the predicted LD effective-mass angular dependence of the critical fields in a system showing dimensional crossover.

Samples of equal layer thickness were prepared by a sputtering technique, and their critical temperature and structure have been described in earlier papers.[7] The same set of samples as used for those studies[7] was used for the critical-field studies. A 0.3-mm-wide strip was photolitographically etched into each sample, and standard four-probe techniques were used to measure the transition field in a radial-access magnet up to 50 kG. The critical field was defined as the midpoint of the ac resistive transition, which did not show appreciable (less than 3%) broadening as a function of magnetic field or temperature. The critical field was independent of the measuring frequency (200 Hz) and current density (0.3 A/cm^2).

Figure 1 shows the ratio of $H_{\parallel}$ to $H_{\perp}$ at 1.17 K (lowest temperature) as a function of layer thickness d. Between 100- and 300-Å layer thickness there is a sharp peak in the $H_{\parallel}/H_{\perp}$-vs-d curve, which we designated as the 2D-coupled region. It is interesting to note that the peak occurs at a thickness equal to the parallel coherence length

$$\xi_{\parallel}=\left[\frac{\phi_0}{2\pi}\frac{1}{H_{c\perp}}\right]^{1/2}=161\pm17 ,$$

in Å, which is the same for *all* samples below $d=400$ Å. For a layer thickness $d \ll \xi_{\rm Nb}$ (the coherence length of Nb), the individual layers are 2D. However, since the coherence length extends over a few layer spacings, the measured properties are characteristic of some average behavior of the entire sample. In our terminology, the layers in this case are "strongly coupled." For the very thick layers ($d > \xi_{\rm Nb}$), on the other hand, the individual layers are bulklike. The measured properties are representative of the properties of the individual layers, the copper having a minor effect. The most interesting region is the

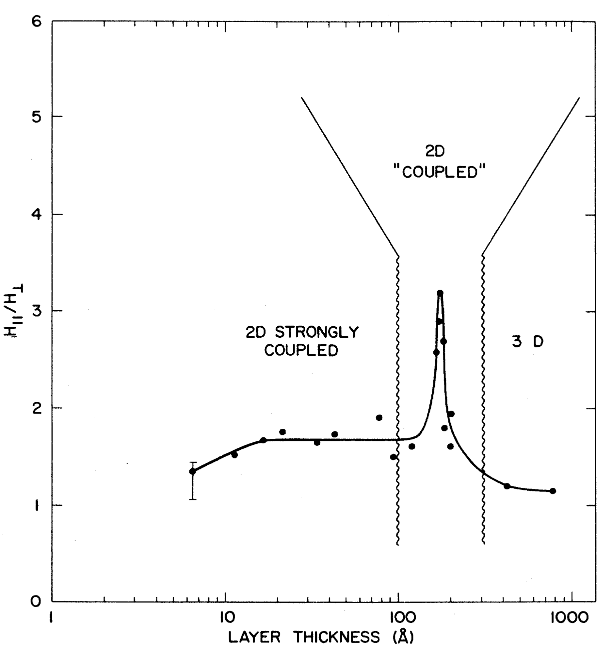

FIG. 1. Ratio of parallel ($H_{\parallel}$) perpendicular ($H_{\perp}$) critical field vs layer thickness. Boundaries of the 2D-coupled region are not precisely defined and are only to be taken as an indication of the region where the 2D Nb films become coupled.

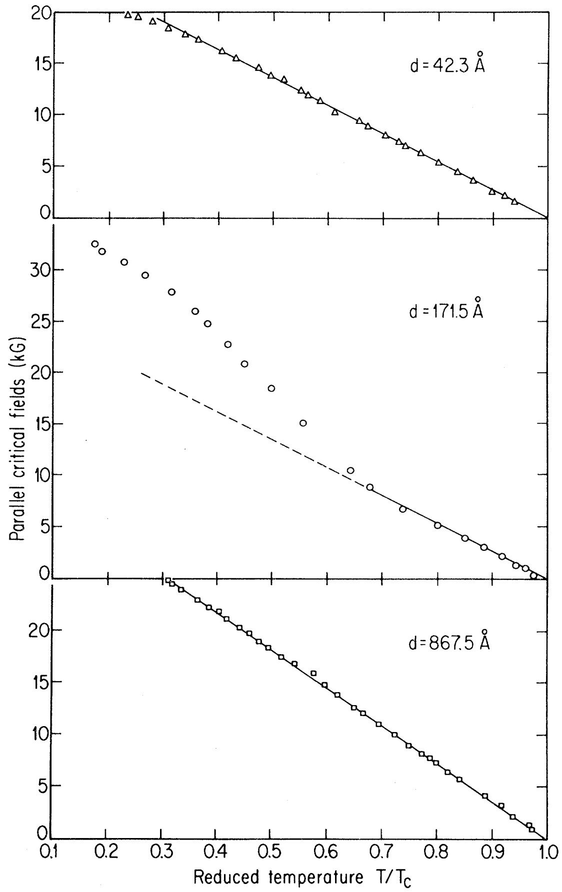

FIG. 2. Temperature dependence of $H_{\parallel}$ for representative samples of each of the regions described in the text.

"peak" region in Fig. 1, i.e., the 2D-coupled region where $d \sim \xi_{\mathrm{Nb}}$. In this region a dimensional crossover takes place and the angular dependence of critical fields changes with temperature.

We should point out that at first sight one might expect these effects to occur for a much larger thickness of Cu. This is because Josephson coupling comparable to that across an oxide layer occurs for considerably thicker Cu layers. However, Josephson-coupling experiments are zero-field experiments, whereas our experiments are in a strong magnetic field close to the critical field. In addition, the present effects are due to the proximity effect and not the Josephson effect across the layers. Clearly, Fig. 1 shows this to be true.

Figure 2 shows the parallel critical field ($H_{\parallel}$) versus temperature for a few samples, representative of the different regions discussed above. The sample with $d=42.3$ Å has a critical field which varies linearly close to T_c and shows a tendency to saturate at low temperatures, just as an ordinary bulk superconductor.[8] In this regime, (i.e., 2D strongly coupled) the coherence length extends over a few layer spacings, hence the superconducting properties are representative of an average over many layers. In fact, the ratio of 1.7 of $H_{\parallel}$ to $H_{\perp}$ suggests strongly that the $H_{\parallel}$ is H_{c3} due to surface superconductivity ($H_{c3}/H_{c2}=1.69$).[9] The same behavior is observed in samples in which the individual Nb layers are 3D, although $H_{\parallel}/H_{\perp}<1.68$ due to the proximity with the Cu.[10] However, for the 171.5-Å sample, the behavior is entirely different. The temperature dependence is almost linear close to T_c, but at $T \simeq 0.65\ T_c$ there is a dramatic upturn in the critical field, and subsequently a tendency to saturation at low temperatures. This upturn is representative of a 3D-to-2D transition discussed previously by Klemm, Luther, and Beasley,[4] and Ruggiero, Barbee, and Beasley.[5]

It should be stressed at this point that Nb/Cu consists of proximity-effect—coupled Nb layers as opposed to systems studied previously[5] where the coupling was due to the Josephson effect across a semiconductor (Ge). In fact, the present results prove that the qualitative ideas forwarded in theories of Josephson-coupled superconductors[4,6] are valid also for proximity-coupled superconductors. However, a detailed comparison of experiment to theory would require an extensive modification in the theory. Although the Hamiltonian used in these theories is probably applicable to proximity-coupled systems, the boundary conditions are inappropriate. We will now address the question of angular dependence in such a system.

Tinkham,[10] based on the argument of fluxoid quantization, determined that the angular dependence of a 2D thin film ($D<\xi$) is given by

$$\left|\frac{H_c(\theta)\sin\theta}{H_{\perp}}\right| + \left[\frac{H_c(\theta)\cos\theta}{H_{\parallel}}\right]^2 = 1 , \qquad (1)$$

where the coordinate system is such that $\theta=0$ parallel to the film. As opposed to this, for thick films ($D>\xi$) superconductivity first nucleates at the surface (H_{c3} effect), and the angular dependence close to $\theta=0$ has been worked out by Yamafuji, Kusayanagi, and Irie[11] and Saint-James.[12] In practice there is little difference between the Yamafuji *et al.*, Saint-James, and Tinkham values for $H_c(\theta)$, given $H_{\parallel}$ and $H_{\perp}$. In fact, for a single 8500-Å Nb film we find very good agreement with Tinkham's theory although, we should stress, the angular dependence of H_{c3} should not necessarily be described by the Tinkham result. We therefore will use Eq. (1) *only* for descriptive purposes. On the other hand, for the LD effective-mass model[6] (developed for Josephson coupled superconductors), $H_c(\theta)$ can be written as[4]

$$\left[\frac{H_c(\theta)\sin\theta}{H_{\perp}}\right]^2 + \left[\frac{H_c(\theta)\cos\theta}{H_{\parallel}}\right]^2 = 1 . \qquad (2)$$

From Eq. (1),[9]

$$[dH_c(\theta)/d\theta]_{\theta=0} = H_{\parallel}^2/2H_{\perp} > 0 .$$

In other words, there is a cusp at $\theta=0$, for either an isolated 2D film or an isolated 3D film. Eq. (2), however, shows $[dH_c(\theta)/d\theta]_{\theta=0}=0$. Thus for Josephson-coupled 2D films the curve is smooth at $\theta=0$ and "bell" shaped.

The angular dependence for the 42.3-Å sample (Fig. 3) shows a cusp in the experimental data and falls below the values calculated from Eq. (1). This suggests that in addition to the H_{c3} anisotropy, there is some added anisotropy induced by the layering process. The two anisotropies, be-

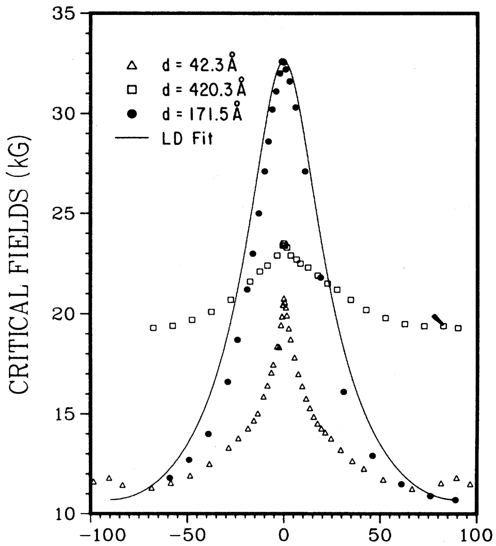

FIG. 3. Angular dependence of parallel critical fields for representative samples of each of the regions described in the text.

ing effective at the same time, induced a larger anisotropy in the system. This extra anisotropy can be qualitatively explained by an anisotropic effective mass: The anisotropic effective mass in conjunction with the H_{c3} anisotropy will give a sharper cusp than that predicted by the Tinkham theory.

In contrast, the 420.5-Å sample, while also having a cusp, shows an anisotropy less than that predicted from simple surface superconductivity (Fig. 3). Here the thick copper layers backing the 3D niobium depress the effect of surface superconductivity, and hence the anisotropy is less than predicted theoretically.

Finally, the most interesting feature is the anisotropy shown by the samples in the 2D-coupled region. For instance, for the sample with $D = 171.5$ Å at 1.17 K, $H_{\parallel}/H_{\perp}$ is 3.2, and the angular dependence does not show a cusp, but instead agrees closely with the LD effective-mass theory (solid line in Fig. 3). Here the coupling between the layers is "perfect" in the sense that the effective mass dominates the surface-superconductivity effect. If the above interpretation is correct, we expect to see a change in the angular dependence at higher temperatures. Since $\xi(T) \rightarrow \infty$ as $T \rightarrow T_c$, the layers should become strongly coupled again close to T_c. Interestingly, as expected, at high temperature the angular dependence does

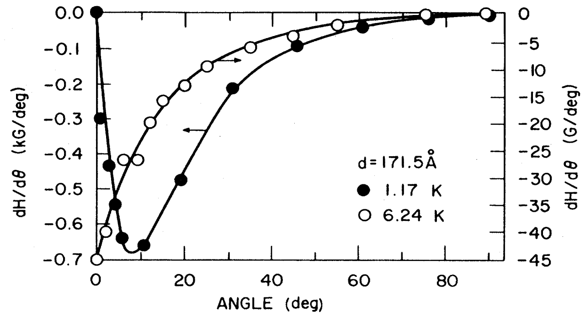

FIG. 4. Derivative of the critical field vs angle for one of the samples in the 2D-coupled region at two temperatures.

show a cusp, the data points falling slightly below the Tinkham line. To emphasize our conclusions, Fig. 4 shows $dH/d\theta$ vs θ for the $d = 171.5$-Å sample. Notice that at $\theta = 0$, the 1.17-K slope is zero, whereas the 6.24-K slope is nonzero, clearly showing the features of a cusp.

At this point, we should stress that these experiments clearly show that for all thicknesses *both* effects are present: Angular-dependent surface superconductivity and anisotropic critical fields as predicted by effective-mass theories. By properly adjusting the layer thicknesses it is possible to observe one or the other effect. For a quantitative comparison, theoretical work is needed to combine both effects. We hope that the present results and future measurements will motivate further theoretical and experimental work on the subject.

In conclusion, the proximity-effect—coupled Nb/Cu layered system clearly shows a dimensional crossover. Associated with the crossover there are changes in the angular dependence of the critical fields, and for the first time the LD dependence associated with the crossover has been observed.

We would like to thank M. R. Beasley, G. Deutscher, R. C. Dynes, J. B. Ketterson, and W. McMillan for helpful comments and discussion, and G. W. Crabtree and P. Roach for advice on cryogenics. One of us (I.K.S.) would like to thank P.L.A.M. Shapiro for suggesting the idea for a scattering experiment. We would especially like to thank J. Vicent and G. G. Zheng for extending some of the experiments showing dimensional crossover. We would particularly like to thank R. A. Klemm for many helpful discussions and insight into the theoretical aspects of the field. This work was supported by the U.S. Department of Energy.

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