Anomalous Upper Critical Fields of Superconducting Multilayers: Verification of the Takahashi-Tachiki Effect

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We observe for the first time the anomalous upturn in the parallel upper critical field predicted for metallic multilayers composed of two materials which have different diffusion coefficients but which have the same superconducting transition temperature. We observe this upturn, which reflects the shift of the nucleation field from the clean to the dirty layers, in multilayers of $Nb/Nb_{0.6}Ti_{0.4}$ having composition wavelengths $\Lambda \geqq 420$ Å. The temperature at which the upturn occurs increases with Λ, as predicted by theory.

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Superconducting multilayers have proved to be an effective medium in which to study the effects of periodicity[1] (and even aperiodicity[2]) on the superconducting phase transition. The best known effect is a dimensional crossover in the parallel upper critical field $H_{c2\parallel}$ of periodic superconductor-insulator (or -normal-metal) multilayers. Near T_c where the coherence length is large, $H_{c2\parallel}$ is determined by the average behavior of the material, and thus the superconductor behaves three dimensionally with $H_{c2\parallel} \sim T_c - T$. As temperature is decreased, the coherence length decreases and the behavior becomes two dimensional below a characteristic temperature at which the coherence length becomes comparable to the layer periodicity. In this low-temperature regime $H_{c2\parallel} \sim (T_c - T)^{1/2}$.

Recently, Takahashi and Tachiki[3] have developed a general theory for the upper critical field of superconductive multilayers. In their theory they include spatial variations of the density of states N, the attractive interaction potential V, and the electronic diffusion constant D. The theory accounts in detail for the dimensional crossover behavior observed, for instance, in superconductor-normal-metal Nb-Cu superlattices,[4] for which the original theory by Klemm, Luther, and Beasley[5] for superconductor-insulator materials was only qualitatively applicable. More interestingly, Takahashi and Tachiki found[6] that other subtle anomalies beyond simple dimensional crossover effects can be present in the behavior of superconductive multilayers. These anomalies arise when there is a competition in the favored nucleation site between two constituent layering materials. They also found that the temperature dependence of the perpendicular critical field could exhibit anomalous upward curvature under appropriate conditions.[3] These new effects are most prominent for multilayers in which the bulk transition temperatures of the two constituent materials are equal, i. e., for materials in which NV is the same and only the diffusion constant D is different. In this paper we present an experimental confirmation of these new effects and discuss some of their consequences on the properties of superconductive multilayers.

The physical ideas underlying this new anomaly in $H_{c2\parallel}$ were discussed in Ref. 6, and so we give only a résumé here. Following Ref. 6, n will refer to the clean layers (large diffusion constant) and s will refer to the dirty layers. As for any periodic system, near T_c where the coherence length is large, the superconductivity averages over the layers and $H_{c2\parallel}$ increases as $T_c - T$. However, as the temperature decreases, nucleation of the pair potential $\Delta(r)$ will occur preferentially in the n or s layers. If nucleation occurs preferentially in the n layers, only a small fraction of $\Delta(r)$ will penetrate into the adjacent s layers because the diffusion constant of the latter is very small. Thus $\Delta(r)$ is largely confined to the n layers, and the multilayer behaves like a two-dimensional thin film with $H_{c2\parallel}$ increasing as $(T_c - T)^{1/2}$. If on the other hand, at relatively high temperatures, superconductivity nucleates preferentially in the s layers, $\Delta(r)$ will be coupled across the n layers and $H_{c2\parallel}$ will be small and linear in $T_c - T$. However, at lower temperatures when the coherence length becomes much smaller than the s-layer thickness, $\Delta(r)$ will be largely confined to these layers and the situation becomes essentially that of bulklike s layers whose H_{c2} is, by design, very high.

These ideas are illustrated schematically in Fig. 1 for a particular set of material parameters. The dashed line shows the behavior that results if nucleation occurs preferentially in the n layers, and the dash-dotted line if it occurs in the s layers. The actual $H_{c2\parallel}$ will be the larger of the two, and is shown in Fig. 1(b). The familiar 3D-to-2D crossover anomaly is evident near T_c. However, if nucleation in the s layers eventually wins out (as in the case illustrated), there is a second anomaly at a lower temperature at which nucleation centers shift from the n to the s layers. This Takahashi-Tachiki effect has not been recognized previously and is the main focus of this

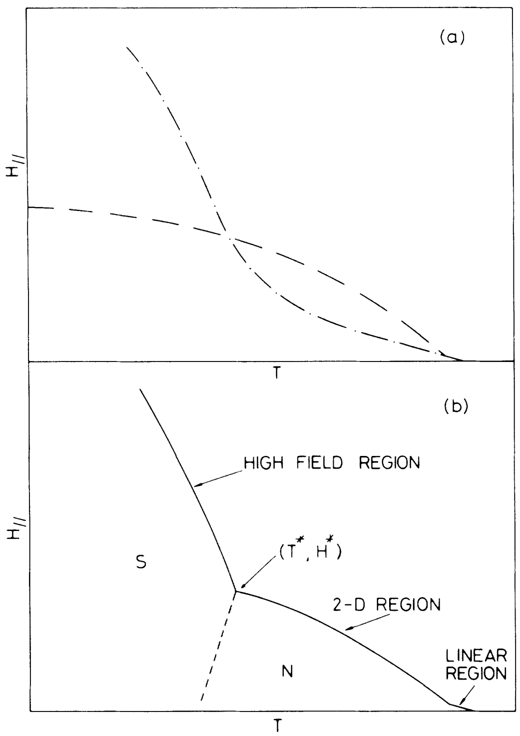

FIG. 1. $H_{c2\parallel}$ vs T after Ref. 6. In (a) the dashed line represents $H_{c2\parallel}(T)$ when $\Delta(r)$ nucleates in the n (clean) layers whereas the dash-dotted line is the result of $\Delta(r)$ nucleating in the s (dirty) layers. The solid line in (b) is the theoretically predicted result and the dashed line is the expected separation between the two regions.

paper.

The experiment was performed on Nb-$Nb_{0.6}Ti_{0.4}$ multilayers. We have made two series of samples. The first, made at the Université de Genève and measured at Stanford University, consisted of samples having wavelengths Λ of 300, 360, and 420 Å. The second series, made and measured in Geneva, had wavelengths of 300, 460, and 500 Å. The choice of Nb and $Nb_{0.6}Ti_{0.4}$ as multilayer constituents seems to be ideal for this test of the theory. Nb and Ti form a continuous solid solution over a wide composition range and the lattice constants range from 3.30 to 3.27 Å as one goes from 100% Nb to 25 at.% Nb. Thus Nb and $Nb_{0.6}Ti_{0.4}$ form a well matched lattice pair which is advantageous for the growing of multilayers. In addition the superconducting critical temperatures for the alloy in the composition range of 40 to 100 at.% Nb fall between 9 and 10 K, thus practically precluding any proximity-effect–induced anomalies in the T_c's of the multilayers themselves. Finally, NbTi alloys are well-known high-field superconductors.[7]

The multilayers were grown in a UHV system equipped with e^- beam guns for deposition, a quadrupole mass spectrometer for rate control, and a quartz thickness monitor. Substrates of (1120) Al_2O_3 heated to 575 °C were exposed to a constant flux of Nb while a shutter masking the Ti was opened and closed with a frequency determined by the evaporation rates and the desired wavelength. Each multilayer consisted of 25 layers with Nb being the first and last layer. All films were measured resistively. We also grew individual 2500-Å films of Nb and of $Nb_{0.6}Ti_{0.4}$ in order to measure the basic material parameters of the constituent materials. Their measured T_c's were 8.95 and 9.14 K, respectively, thus fulfilling the requirement that the T_c's of the constituent materials be about equal. Both films had (10–90)% transition widths $\leqq 0.1$ K wide. In order for us to see the anomalous upturn in $H_{c2\parallel}$, D_n/D_s must exceed a certain critical value which depends on the layer thicknesses d_s and d_n. For example, when $d_n = d_s = \xi_{Nb}(0)$, the critical ratio is found[6] to be 8.4. Low-field measurements yield perpendicular-field slopes near T_c of -0.75 kG/K for the Nb and -15.6 kG/K for the $Nb_{0.6}Ti_{0.4}$ (we use the perpendicular fields so as not to be affected by surface superconductivity). From these data we obtain $D_n/D_s = 20.8$ which is clearly sufficient to test the theory. The extrapolation of the perpendicular upper critical field of Nb to 0 K leads to a zero-temperature coherence length of $\xi_{Nb}(0) = 266$ Å. The T_c's of the 300-, 360-, 420-, 460-, and 500-Å multilayers are 8.81, 9.72, 9.69, 8.83, and 8.95 K, respectively,[8] and the zero-field transition widths are less than 40 mK.

The vivid experimental confirmation of the Takahashi-Tachiki prediction for the parallel-field upturn is shown in Fig. 2. This peculiar crossover, in which the maximum of $\Delta(r)$ switches from the Nb layers to the $Nb_{0.6}Ti_{0.4}$ layers, is unambiguously defined for the 420-, 460-, and 500-Å wavelength samples while it does not at all occur for the 300- and 360-Å-wavelength samples. The solid lines through the data points are a guide to the eye and also give us our definition of t^* as the intersection of the 2D-like curve with that of the high-field region curve. We find, completely consistent with the theory, that t^* $(=T^*/T)$ increases as the wavelength increases. This behavior is in accord with the physical picture of what is happening. That is, the thicker the layers, the higher the temperature at which the coherence length will be small enough to be confined entirely to the low-D_s, high-H_{c2} NbTi layers. For $\Lambda = 300$ Å, and also for the 360-Å sample, the 2D-like behavior continues down to 1.5 K, our lowest attainable temperature. This is because the layer spacing is too small ever to allow the order parameter to nucleate and be confined only in the $Nb_{0.6}Ti_{0.4}$ layers. The other regions of the curves are also consistent with the theory: Very near T_c there is the expected 3D to 2D crossover, and the magnitude of $H_{c2\parallel}$ for the three curves in the 2D region decreases with increasing wavelength for fixed t. What adds additional weight to the above is that we observe completely consistent results for the two series of samples made, and measured, at different times.

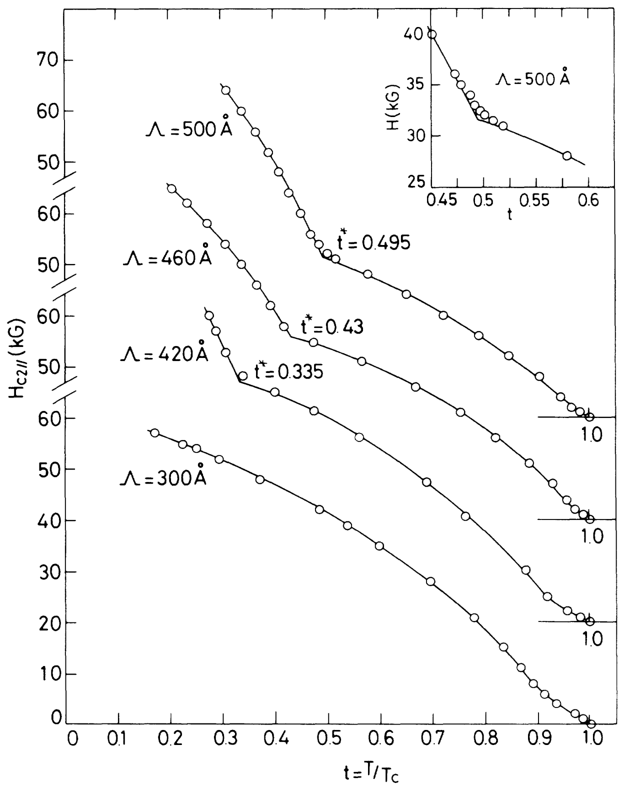

FIG. 2. Resistivity measured $H_{c2\parallel}$ vs t $(=T/T_c)$. The y axes are displaced for clarity and the solid lines are guides to the eye. Inset: Blowup of the region around t^* for the 500-Å sample.

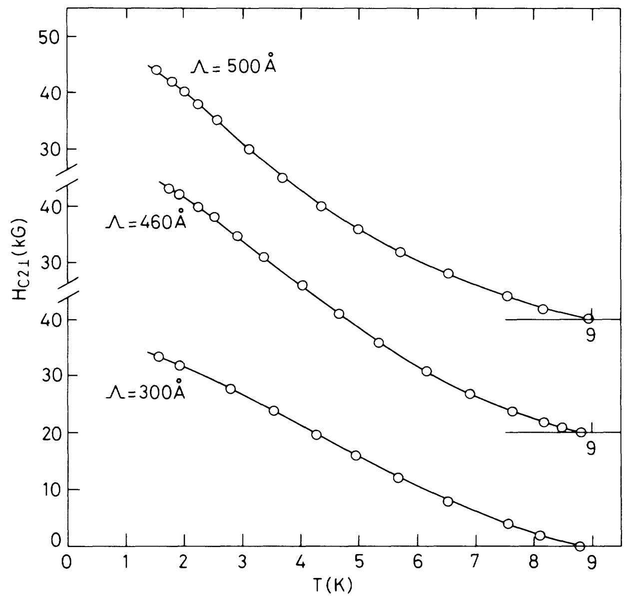

FIG. 3. $H_{c2\perp}$ vs T. The y axes are displaced for clarity and the solid lines are guides to the eye.

We have also investigated the region around T^* for the $\Lambda = 500$ Å sample and find that this crossover region is narrow (~0.25K wide) but not discontinuous as predicted theoretically. There may be several reasons for this, such as the finite thickness of the films or fluctuations, as discussed below. Another reason might be that in Ref. 6 the assumptions made about the multilayer are idealized: The interfaces are perfectly sharp and the parameters are made to change discontinuously at the layer interfaces. Low-angle x-ray diffraction measurements on these multilayers and on those with shorter wavelengths (≥ 80 Å) show that the $Nb/Nb_{0.6}Ti_{0.4}$ interfaces are about 50 Å wide. Thus the transport properties of these samples change continuously across the interfaces. Hence small deviations from the predicted discontinuous curve are not surprising.

An interesting feature of this crossover region is a bulge in the width of the transition $\Delta H = (dH/dT)\Delta T$ where ΔT is the (10–90)% resistive width and dH/dT is taken at the temperature being investigated. Outside the transition region ΔH is small. Inside it shows an increase. For example, in the case of the 500-Å-wavelength sample, $\Delta H = 3.4$ kG at $t = 0.65$ (in the 2D regime and 3.5 kG at $t = 0.34$ (in the high-field regime). However at $t = 0.51$, which is the start of the crossover region, ΔH has increased to 5.43 kG. This bulge could be indicative of enhanced fluctuations in this crossover regime where the superconductor is trying to decide in which state, 2D or high field, it should be. Put another way, this bulge could be indicative of softening in the restoring forces at t^* restraining the nucleation to occur in the s or n regions. By comparison, for the 300-Å wavelength sample $\Delta H = 2.04$ kG at $t = 0.54$, which is about $2\frac{1}{2}$ times smaller than the corresponding ΔH of the 500-Å sample. An interesting way to investigate this phenomenon would be critical-current (J_c) measurements. On general physical grounds, one would expect a minimum in the critical current in this region. This would also be a feasible way to map out the H-T phase diagram of this system. By our following the minimum in J_c, the crossover from the 2D to the high-field region could be distinguished for all H and T [this is the dashed line in Fig. 1(b)].

Finally we turn to the perpendicular-critical-field behavior. The measured perpendicular critical fields are shown in Fig. 3. The upturn seen at low temperatures in a parallel applied field vanishes when the plane of the film is perpendicular to the applied field. This comportment is again consistent with the theory. However, the Takahashi-Tachiki theory also predicts a kind of dimensional crossover in $H_{c2\perp}$ if D_n/D_s is large enough. For the perpendicular field, they found that the spatial modulation of the order parameter in a direction perpendicular to the film (and parallel to the flux lines) exponentially decreases in the n layers. When the coherence length is long, the order parameter will still be continuously coupled throughout the multilayer, but when the coherence length becomes small enough compared with the layer thickness, the damping will produce a decoupling of the s layers. Then the observed perpendicular upper critical field will be that of the s layers which is, naturally, very high. The larger the ratio D_n/D_s, the higher the temperature at which the $H_{c2\perp}$ of s will predominate for fixed $d/\xi_n(0)$. In Fig. 3 one can see the continuous transformation from the coupled-layer regime to the more or less isolated s-layer regime. Although the behavior for the perpendicular field is not as dramatic as in the parallel case (extremely high ratios of D_n/D_s are necessary in order to make the effect vivid), it still illustrates the basic consistency and correctness of the Takahashi and Tachiki theory, and the rich effects producible in metallic multilayer systems.

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