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Upper critical field in anisotropic superconductors

Ctirad Uher and Joshua L. Cohn

Physics Department, The University of Michigan, Ann Arbor, Michigan 48109

Ivan K. Schuller

Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439 (Received 18 April 1986; revised manuscript received 14 July 1986)

The upper critical field of anisotropic superconductors (Mo/Ni superlattices) exhibits large nonlinearities in temperature dependence. This behavior is unexpected in light of current theories of upper critical field including surface superconductivity effects.

Anisotropic superconductivity has been studied in a variety of one-dimensional,¹ intercalated,²⁻⁶ and artificially prepared⁷⁻¹¹ systems. Much of the experimental work in these systems is related to measurements of the temperature and angular dependence of the upper critical fields H_{c2} . In many cases, the experiments determine the critical field at which surface superconductivity nucleates, i.e., H_{c3} .¹² The relationship between H_{c3} and H_{c2} is well established both theoretically and experimentally in isotro-pic superconductors.¹³ In anisotropic superconductors, very little work has been done either experimentally or theoretically regarding the angular and temperature dependence of H_{c3} .^{14,15} We present here the experimental temperature dependence of H_{c3} for an anisotropic superconductor. The results show that the temperature dependence of H_{c3} is $(T_c - T)^{1/2}$ unlike in homogeneous superconductors where it is linear. Moreover, at low temperature, the ratio $H_{c3}/H_{c2} \sim 1.7 \pm 0.1$, as found and predicted in dirty isotropic superconductors at all temperatures.^{13,16}

The anisotropic superconductors studied in this work were prepared by sequential magnetron sputtering of Mo and Ni on sapphire substrates using a technique described earlier.¹⁷ The sputtering system was calibrated to prepare equal layer thicknesses of Mo(6.9 Å)/Ni(6.9 Å) ($\lambda \simeq 13.8$ Å) and Mo(8.3 Å)/Ni(8.3 Å) ($\lambda \simeq 16.6$ Å) samples. The total thickness of the samples was kept around 1 μ m. Detailed x-ray measurements^{18,19} show that the $\lambda = 13.8$ Å sample is quite disordered, akin to an amorphous mixture of Mo and Ni, whereas the $\lambda = 16.6$ Å sample still preserves the layered structure. Cooling was achieved with the aid of a dilution refrigerator provided with a custommade reduced tail section to incorporate an 8-T superconducting solenoid. Samples were attached to an oxygenfree high-conductivity copper sample holder with a thin coating of Apiezon M grease and fastened with dental floss. Two separate runs were made, one for the parallel orientation of magnetic field with respect to the film surface, and the other for the perpendicular configuration. The width of superconducting transitions is between 10-40 mK (increasing with the field) and we have monitored the 50% resistive transition point. Temperature was monitored by a combination of a calibrated Ge sensor and a Speer carbon resistance thermometer. The latter is known to possess a small and reproducible magnetoresistance, while the Ge sensor serves to provide its zero-field

calibration curve. Current densities used in the measurements were very small, $J = 0.1 \text{ A cm}^{-2}$, to avoid any heating of the structure at sub-Kelvin temperatures.

The length scale that determines the behavior of layered superconductors is the anisotropic coherence length (parallel ξ_{\parallel} and perpendicular ξ_{\perp}). From the anisotropic Ginzburg-Landau theory the coherence lengths relevant for anisotropic superconductors can be obtained using

$$H_{c\parallel}(T) = \frac{\phi_0}{2\pi} \frac{1}{\xi_{\parallel}(T)} \frac{1}{\xi_{\perp}(T)} , \qquad (1)$$

$$H_{c\perp}(T) = \frac{\phi_0}{2\pi} \frac{1}{\xi_{\parallel}^2(T)} , \qquad (2)$$

where $H_{c\parallel}$ and $H_{c\perp}$ are the experimentally measured *upper* critical fields, and ϕ_0 is the flux quantum.

Table I shows the "zero" (extrapolated from 90 mK down) temperature critical fields and coherence lengths for the two samples measured in these experiments. It is clear from this table that both coherence lengths (ξ_{\parallel}) and (ξ_{\perp}) are much larger than the layer thicknesses of the samples, and therefore the material should behave as a bulk superconductor, without any dimensional effects. One therefore would expect H_{\perp} to be linear at high temperature and then saturate at low temperature, and $H_{\parallel}/H_{\perp} = 1.69$ independent of temperature as predicted and observed for surface superconductivity of bulk isotropic superconductors.^{13,16}

However, a measurement of the temperature dependence of $H_{c\parallel}$ and $H_{c\perp}$ gave a rather surprising result, as shown in Figs. 1 and 2. Although $H_{c\perp}$ is linear as expected, $H_{c\parallel}$ deviates considerably from theoretical expectations for a bulk homogeneous superconductor (i.e., $H_{c\parallel}/H_{c\perp}$ = 1.69 independent¹⁶ of temperature). In fact, the ratio

TABLE I. Low-temperature critical fields and coherence lengths for Mo/Ni superlattices.

Parameter	$\lambda = 13.8$ Å	$\lambda = 16.6$ Å
$H_{c\perp}(0)$	0.98 T	3.5 T
$H_{c\parallel}(0)$	1.62 T	6.2 T
$\xi_{\parallel}(0)$	200 Å	110 Å
$\xi_{\perp}(0)$	130 Å	60 Å

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FIG. 1. Temperature dependence of the parallel $H_{c\parallel}$ and perpendicular $H_{c\perp}$ critical fields for Mo-Ni heterostructures with $\lambda = 16.6$ Å. Inset shows critical field anisotropy. X-ray structural data indicate that layering is preserved in this structure.



FIG. 2. Temperature dependence of the parallel $H_{c\parallel}$ and perpendicular $H_{c\perp}$ critical fields for structure with $\lambda = 13.8$ Å. Inset shows critical field anisotropy. X-ray structural data reveal a typical glassylike diffraction pattern, signaling a structural collapse.

deviates greatly from a constant as shown by the insets in both figures.

A plot of $H_{c\parallel}$ vs $(T_c - T)^{1/2}$ (Fig. 3) indicates that this behavior is obeyed throughout the temperature range studied. This square-root dependence is completely unexpected for the behavior of either H_{c2} or H_{c3} in a bulk homogeneous superconductor. In general, nonlinearities in the upper parallel critical field may arise from the following physical phenomena: dimensional crossover, spin orbit scattering, proximity effect, localization, or disordered structure.

Dimensional crossover. Nonlinear temperature dependences have been predicted and observed for $H_{c2\parallel}$ for a set of two-dimensional (2D) superconducting layers weakly coupled through the Josephson effect^{7,8} and for 2D layers coupled by the proximity effect in the absence of surface superconductivity.¹² In these cases, the nonlinearities occur when the ratio of $\xi_{\perp}/D_N \leq 1$, where D_N is the thickness of the separator between the 2D superconducting films. Table I shows that in the present experiments the opposite is true, i.e., $\xi_{\perp}/D_N \gg 1$, and therefore the Mo layers are strongly coupled together. Consequently, the materials should behave as 3D superconductors with a linear temperature dependence of $H_{c2\parallel}$. It is clear therefore that dimensionality effects are not operational here.

Spin-orbit scattering. Another possible explanation using the spin-orbit scattering time $\tau_{s.o.}$ as an adjustable parameter is not believed to operate here either. Although a reasonable fit using $\tau_{s.o.}$ as an adjustable parameter can be obtained,²⁰ the nonlinearity should be the same in both $H_{c\parallel}$ and $H_{c\perp}$. The reason for this is that, as pointed out above, the critical-field ratio $(H_{c\parallel}/H_{c\perp})$ should be independent of temperature, as predicted¹³ and found experimentally¹⁶ in homogeneous superconductors.

Proximity effect. A long awaited and never observed effect relates to the surface superconductivity of a superconductor in contact with a normal metal.²¹ In this case, it



FIG. 3. Parallel critical field exhibiting a $(T_c - T)^{1/2}$ temperature dependence.

was shown that the parallel critical field can exhibit strong nonlinearities if the normal-state conductivity σ_s of the superconductor is much larger than the conductivity of the normal metal σ_N . The low-temperature $H_{c\parallel}/H_{c\perp}$ ratio is also predicted to be close to 1.7 if $\sigma_s/\sigma_N \approx 100$. The experimental data observed in the present study strongly resemble the predictions of Hurault. However, it is not clear at present what the ratio of the conductivities between the two constituents (Mo and Ni) is, because for these small thicknesses it is very hard to unravel the contribution from each one of the metals. Therefore, it is not clear whether the condition $\sigma_s/\sigma_N \gg 1$ is satisfied in this case. Moreover, since $\xi_{\perp} \gg D$ it is not obvious that Hurault's predictions are operating here.

Localization. One intriguing possibility is that surface superconductivity is strongly affected by electron localization. We have shown earlier that the transport properties of the superlattices exhibit a negative temperature coefficient of resistivity at low temperatures, characteristic of electron localization-interaction effects.¹⁹ It would be quite remarkable if the interaction of superconductivity and localization would have such a drastic effect on the critical fields. To our knowledge, no theory has been developed to study the effect of localization on surface superconductivity.

In the absence of further theories that address the dependence of surface superconductivity in anisotropic superconductors or superconductors incorporating strongly scattering, randomly distributed inclusions, it is not possible to identify the exact mechanism that is responsible for the observed anisotropies. However, the fact that such a system gives rise to a well-defined temperature dependence implies that complicating effects due to surface superconductivity should be understood and accounted for before theories of H_{c2} are applied to complex superconducting systems, such as metallic heterostructures.

In summary, we have observed a strong nonlinearity in the temperature dependence of the parallel critical field of anisotropic Mo/Ni superconductors. The origin of the nonlinearity has not been unambiguously established at the present time. It should be stressed that the present results show that surface superconductivity can exhibit nonlinear behavior quite similar to dimensional behavior, and this has to be taken into account when critical-field measurements are interpreted in inhomogeneous superconductors.

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- ¹L. J. Azevedo, W. G. Clark, G. Deutscher, R. L. Greene, G. B. Street, and L. J. Sutter, Solid State Commun. **19**, 197 (1976).
- ²R. C. Morris and R. V. Coleman, Phys. Rev. B 7, 991 (1973).
- ³S. Foner and E. J. McNiff, Phys. Lett. **45A**, 429 (1973).
- ⁴D. E. Prober, R. E. Schwall, and M. R. Beasley, Phys. Rev. B 21, 2117 (1980).
- ⁵J. A. Woolam, R. B. Somoano, and P. O'Connor, Phys. Rev. Lett. **32**, 712 (1974).
- ⁶P. deTrey, S. Gygax, and J. P. Jan, J. Low Temp. Phys. **11**, 421 (1973).
- ⁷T. W. Haywood and D. G. Ast, Phys. Rev. B 18, 2225 (1978).
- ⁸S. T. Ruggiero, T. W. Barbee, and M. R. Beasley, Phys. Rev. Lett. 45, 1229 (1980).
- ⁹H. K. Wong, B. Y. Jin, H. Q. Yang, J. E. Hilliard, and J. B. Ketterson, Superlattices Microstr. 1, 259 (1985).
- ¹⁰I. Banerjee, Q. S. Yang, C. M. Falco, and I. K. Schuller, Phys. Rev. B 28, 5037 (1980).
- ¹¹For a recent review, see S. T. Ruggiero and M. R. Beasley, in Synthetic Modulated Structures, edited by L. L. Chang and B. C. Giessen (Academic, New York, 1985), Chap. 10.

- ¹²C. S. L. Chun, G.-G. Zheng, J. L. Vicent, and I. K. Schuller, Phys. Rev. B 29, 4915 (1984).
- ¹³D. Saint-James and G. Sarma, *Type II Superconductors* (Pergamon, Oxford, 1969).
- ¹⁴E. V. Minenko, Fiz. Nizk. Temp. 9, 1036 (1983) [Sov. J. Low Temp. Phys. 9, 535 (1983)].
- ¹⁵B. Ya. Shapiro (private communication).
- ¹⁶G. Ebneth and L. Tewordt, Z. Phys. 185, 421 (1965).
- ¹⁷M. R. Khan, C. S. L. Chun, G. P. Felcher, M. Grimsditch, A. Kueny, C. M. Falco, and I. K. Schuller, Phys. Rev. B 27, 7186 (1983).
- ¹⁸I. K. Schuller, Phys. Rev. Lett. 44, 1597 (1980).
- ¹⁹C. Uher, R. Clarke, G.-G. Zheng, and I. K. Schuller, Phys. Rev. B 30, 453 (1984).
- ²⁰C. Uher, W. J. Watson, J. L. Cohn, and I. K. Schuller, in *Lay-ered Structures and Epitaxy*, edited by J. M. Gibson, G. C. Osbourne, and R. M. Tromp (North-Holland, Amsterdam, 1986).
- ²¹J. P. Hurault, Phys. Lett. 20, 587 (1966).