Superconductivity in Quasi-Two-Dimensional Layered Composites

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Finely layered superconducting Nb/Ge composites have been fabricated with use of a novel deposition scheme and studied as model systems for quasi-two-dimensional superconductivity. Systematic variation of superconducting properties has been observed along with two- to three-dimensional crossover behavior in both the parallel critical fields below T_c and the fluctuation conductivity above T_c.

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Systems that exhibit a high degree of anisotropy have been attracting considerable attention recently because of their unique physical properties. Superconducting examples include naturally occurring quasi-two-dimensional (2D) systems such as the layered transition-metal dichalcogenides[1] and artificial layered composites.[2,3] In addition theoretical studies[4-6] of the superconductivity of such systems based on Josephson-coupled models have led to a variety of unusual predictions. Interest remains high in both the material and physical properties of such systems.[7,8] In this Letter we describe the results of a study of the critical magnetic fields of a novel system of artificially layered superconductors produced by advanced sequential-sputtering techniques previously developed by Barbee and Keith.[9] Our results for the parallel upper critical fields $H_{c2\parallel}(T)$ unambiguously demonstrate dimensional crossover behavior in quantitative agreement with the Josephson-coupled model of quasi-2D superconductors. The expected crossover in the fluctuation conductivity above T_c is also observed. Thus our results not only provide the first quantitative confirmation of the Josephson-coupled model, they also demonstrate the utility of sequential-sputtering techniques in producing model systems for the study of quasi-2D superconductors.

Our multilayered composites consist of very thin alternating layers of Nb and Ge sequentially deposited by sputtering onto sapphire substrates by means of a turntable arrangement that periodically moves the sample into two carefully isolated sputtering regions. Samples with individual layer thickness ranging from 5 to 100 Å have been produced. Typically fifty layer pairs are deposited yielding total thicknesses ranging from 0.1 to 1 μm. X-ray-diffraction analysis shows a distinct superlattice structure at low angles associated with the layer periodicity and a broadened Nb ⟨110⟩ line at high angles. From these data the layer periodicity and Nb-layer thickness can be accurately determined. The Ge layers are found to be amorphous. These x-ray data in conjunction with sputter Auger analysis demonstrate the high degree of regularity in the layering of these samples. In general it is found that for large Ge thicknesses (~ 100 Å) the critical temperatures T_c of these multilayered sputtered samples are very close to those reported for single-layer Nb films, although somewhat higher critical temperatures are observed for samples with thinner Ge layers. These results will be discussed elsewhere. A list of the material parameters for our samples is given in Table I.

For the critical-field studies the samples were etched into 300-μm-wide bridges and H_{c2} was determined from the midpoint of the ac resistive transition. Other choices for the definition of H_{c2} made no qualitative changes in the $H_{c2}(T)$ phase boundary. Representative behavior is presented in Fig. 1. For the samples shown D_{Nb} is essentially constant and D_{Ge} is varied through the range where Josephson decoupling takes place. The strong anisotropy of all the samples is evident from the vastly different critical fields in the par-

TABLE I. Parameters for Nb-Ge composites of Figs. 1 and 2. $D_{Nb}+D_{Ge}\equiv s$. All samples have fifty Nb layers except "45/7" with 84.

D_{Nb}/D_{Ge} (Å/Å)	T_c (K)	$\xi_z(0)$ (Å)	R (Ω-layer)	δ
45/7	5.79	38.6	80.8	...
65/35	6.88	9.64	25.0	0.28
37/32	4.95	8.13	91.6	0.34
45/50	4.69	0	83.3	0.15

allel and perpendicular directions. Also, except for temperatures very near T_c (see below), $H_{c2\perp}$ is to first order independent of D_{Ge}. However, the parallel critical fields are very different and clearly show dimensional crossover both as a function of normal layer thickness and temperature. More specifically for the $D_{Ge}=7$ Å sample $H_{c2\parallel}(T)$ exhibits strongly coupled, bulklike, 3D behavior $[H_{c2}(T)\propto T_c - T]$, whereas for $D_{Ge}=50$ Å decoupled, thin-film-like, 2D behavior $[H_{c2}(T) \propto (T_c-T)^{1/2}]$ is observed. This is perhaps not so surprising. However for $D_{Ge}=35$ Å the novel temperature-dependent dimensional crossover expected from theory is clearly seen. The critical-field behavior goes from being 3D near T_c to 2D well below T_c. The solid lines are theoretical fits obtained from the Josephson-coupling theory.

These results confirm quantitatively one of the more striking predictions of the Josephson-coupled model of quasi-2D superconductors[5,6]: namely, that interlayer orbital pair breaking in a parallel magnetic field can become ineffective as the temperature is reduced, leading (in the absence of subsidiary pair breaking) to a formally infinite critical field at a temperature $T^* < T_c$ defined by the relation $\xi_z(T^*) = s/\sqrt{2}$. Here $\xi_z(T)$ is the Ginzburg-Landau coherence length perpendicular to the layers and s is the layer repeat distance. In the presence of subsidiary pair breaking (e.g., due to Pauli limiting and finite superconductor thickness), T^* corresponds to a dimensional crossover temperature for the critical fields. Prober, Schwall, and Beasley[10] and Deutscher and Entin-Wholman[11] have reported likely observations of this effect in layered compounds and granular superconductors, respectively. Further evidence has been reported recently by Vincent, Hillenius, and Coleman.[7] However, to date no really definitive test of the Josephson model or satisfactory model system for studying quasi-2D superconductivity has been available.

In making the theoretical fits shown in Fig. 1,

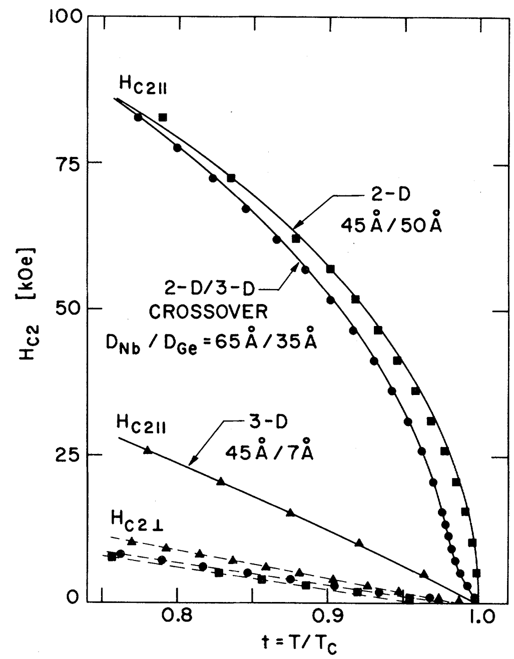

FIG. 1. Upper critical fields of the layered composites. Decreasing the Ge thickness effects a progression from anisotropic 3D, to "crossover", to "decoupled" or 2D behavior. The solid lines are from the Josephson-coupling theory.

we have used the calculations of Klemm, Luther, and Beasley[5] extended to include the effects of finite superconducting layer thickness.[11] The parameters that enter the theory are T_c, $\xi_\parallel(0)$, $\xi_\perp(0)$, D_{Nb}, s, and the spin-orbit scattering rate $\lambda_{s.o.}$. With the exception of $\lambda_{s.o.}$ all the parameters are determined directly from experiment, $\xi_\parallel(0)$ and $\xi_\perp(0)$ being determined from the parallel- and perpendicular-critical-field slopes near T_c. The only adjustable parameter then is $\lambda_{s.o.}$ which is determined by the high-field data. Note, however, that in the region of the crossover the fits are insensitive to the choice of $\lambda_{s.o.}$.

We have also measured the zero-field fluctuation conductivity $\sigma'(T)$ of our samples. Data are shown in Fig. 2 for the strong (3D), intermediate, and fully decoupled (2D) cases. As seen in Fig. 2, the data for the 2D samples are well fitted by the standard Aslamazov-Larkin (AL) fluctuation theory including the Maki-Thompson (MT) corrections.[12] The fitted pair-breaking parameter δ is in satisfactory accord with the results obtained for single 2D Nb films.[13] The 3D sample was found to exhibit a $(T-T_c)^{1/2}$ temperature dependence of σ' in accord with previous experimental work on amorphous materials[14] and the AL theo-

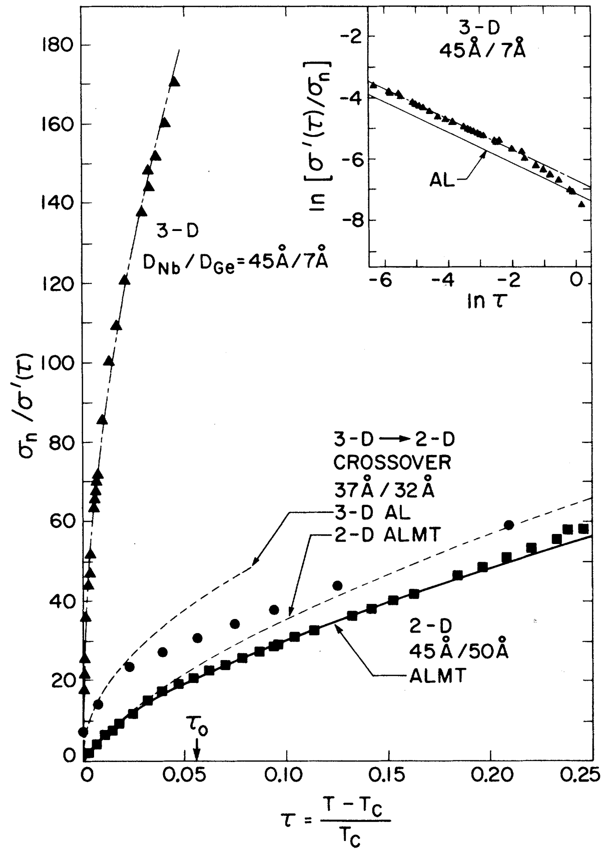

FIG. 2. Temperature dependence of the zero-field fluctuation conductivity above T_c. Shown are samples exhibiting 2D (squares), crossover (circles), and 3D (triangles) behavior. τ_0 is the predicted crossover temperature of the crossover sample. Inset shows ln-ln replotting of the 3D data comparing the square-root fit with the AL result.

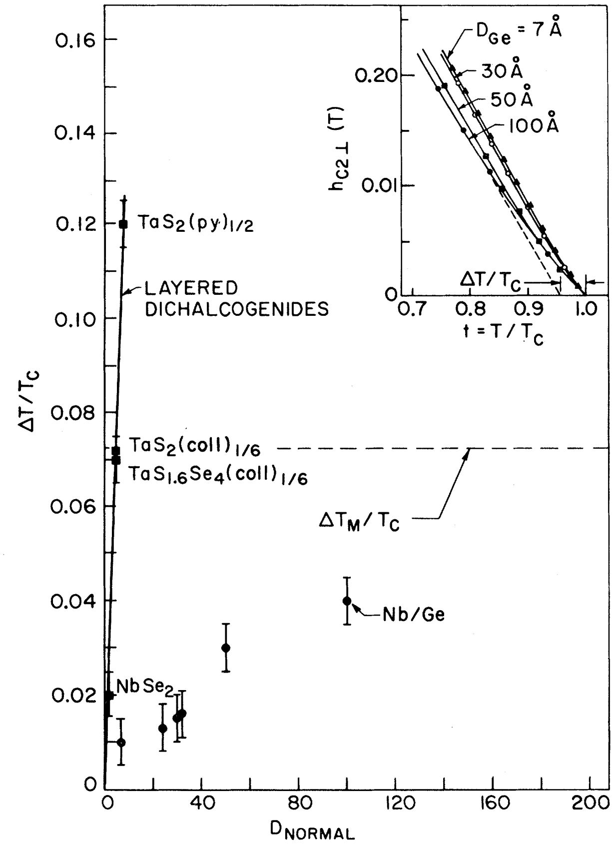

FIG. 3. Inset: $h_\perp(T) \equiv H_{c2\perp}(T)/T_c[dH_{c2\perp}(T)/dT]_{T_c}$ near T_c for roughly constant D_{Nb}. The extrapolation of the linear portion of $H_{c2}(T)$ to zero for each sample is quantified by the parameter ΔT. D_{normal} is the separation distance between superconducting layers. The dotted line is the average value of $\Delta T_m/T_c$ for the Nb/Ge samples (see Ref. 19). The layered dichalcogenides are shown for comparison.

ry. However, as shown in the inset, the magnitude of σ'^{-1} is larger than the AL prediction by a factor of ~ 1.5. Inclusion of the MT corrections could not be satisfactorily employed to remove this discrepancy.[15] In any event, the sample with intermediate coupling [which also showed an upturn in $H_{c2\parallel}(T)$ in accord with theory] does evidently exhibit a crossover in its fluctuation behavior from 3D near T_c to 2D well above T_c. We have plotted the appropriate 3D AL and 2D ALMT results for this sample along with the expected dimensional crossover temperature τ_0.[4] Detailed curve fitting was not possible because the MT corrections have not been included concomitantly with finite-thickness effects in the crossover fluctuation[4,16,17] theories. Hence, except for a possible detailed quantitative discrepancy in the 3D limit, these data reflect nicely the dimensional crossover behavior expected for coupled 2D systems. We note that the superconducting parameters used in fitting σ' and calculating τ_0 were determined solely from the critical-field measurements below T_c.

Finally let us turn to the perpendicular-critical-field data. As first emphasized by Woollam *et al.*,[18] layered-compound superconductors consistently show an anomalous positive curvature in the temperature dependence of their perpendicular critical fields. To date there has been neither a satisfactory explanation of this effect nor entirely convincing evidence that it is an intrinsic phenomenon. Our artificially layered composites also show such anomalies, as do the similar composites studied by Haywood and Ast[2] in which no apparent Josephson coupling was present. As shown in Fig. 3, we find that the magnitude of the anomaly increases with increasing layer separation [at roughly fixed $R_\square$ (≈ 85 Ω-layer)]. To quantify the magnitude of the anomaly, we have used the extrapolated T_c shift normalized to the actual T_c as illustrated in the inset of Fig. 3. Data on the layered-compound superconductors analyzed in a similar way are also shown for comparison. In both cases, a systematic dependence on D_N is evident, which suggests an intrinsic origin for this phenomenon.[19]

In conclusion, then, we see that these artificial, layered composites appear to provide a model system for studying quasi-two-dimensional superconductivity. Moreover, the results obtained with these systems strengthen the case that novel superconducting behavior exists in the *intra*layer as well as the *inter*layer properties of these systems.

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[1]F. R. Gamble, F. J. Di Salvo, R. A. Klemm, and T. H. Geballe, Science 168, 568 (1970).

[2]T. W. Haywood and D. G. Ast, Phys. Rev. B 18, 2225 (1978).

[3]J. M. Dupart and J. Baixeras, Appl. Phys. Lett. 30, 123 (1977); C. R. Spencer, P. Martinoli, E. D. Gibson, J. D. Verhoeven, and D. K. Finnemore, Phys. Rev. B 18, 1216 (1978).

[4]W. Lawrence and S. Doniach, in *Proceedings of the Twelfth International Conference on Low Temperature Physics*, edited by Eizo Kanda (Academic Press of Japan, Kyoto, 1971), p. 361.

[5]R. A. Klemm, A. Luther, and M. R. Beasley, Phys. Rev. B 12, 877 (1975).

[6]L. N. Bulaevskii, Usp. Fiz. Nauk 116, 449 (1975) [Sov. Phys. Usp. 18, 514 (1976)].

[7]J. L. Vincent, S. J. Hillenius, and R. V. Coleman, Phys. Rev. Lett. 44, 892 (1980).

[8]I. K. Schuller, Phys. Rev. Lett. 44, 1597 (1980).

[9]T. W. Barbee, Jr. and D. Keith, in Proceedings of the Workshop on X-ray Instrumentation for Synchrotron Radiation Research, edited by H. Winick and G. Brown, Stanford University Synchrotron Radiation Laboratory Report No. 78-04, 1978 (unpublished).

[10]D. E. Prober, R. E. Schwall, and M. R. Beasley, Phys. Rev. B 21, 2717 (1980).

[11]G. Deutscher and O. Entin-Wohlman, Phys. Rev. B 17, 1249 (1978).

[12]W. J. Skocpol and M. Tinkham, Rep. Prog. Phys. 38, 1049 (1975).

[13]T. Kwaguti and Y. Shibuya, Phys. Lett. 45A, 395 (1973).

[14]W. L. Johnson, C. C. Tsuei, and P. Chaudhari, Phys. Rev. B 17, 2884 (1978); P. J. Silverman, Phys. Rev. B 16, 2066 (1977).

[15]K. Maki, Prog. Theor. Phys. 45, 1016 (1971).

[16]R. A. Klemm, J. Low Temp. Phys. 16, 381 (1974); K. Yamaji, Phys. Lett. 38A, 43 (1972).

[17]L. G. Aslamosov and A. A. Varlamov, J. Low Temp. Phys. 38, 223 (1980).

[18]J. A. Woollam, R. B. Somoano, and P. O'Connor, Phys. Rev. Lett. 32, 712 (1974).

[19]While no specific mechanism can be definitively identified for this anomaly, we note that recent theories of two-dimensional superconductors [B. A. Huberman and S. Doniach, Phys. Rev. Lett. 43, 950 (1979), and in Proceedings of the Conference on Inhomogeneous Superconductors, Berkley Springs, West Virginia, 1979 (unpublished)] predict melting of the vortex lattice in thin superconducting films at reduced temperatures in the range of the observed anomalies. The predicted melting temperature $\Delta T_m/T_c$ for independent films with $R_\square = 85$ Ω is shown by the dashed line in Fig. 3.