Long-Range Proximity Effect in Aluminum Thin Films with Spatially Modulated T_c

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We report resistive measurements on a thin-film analog of a modulated-transition-temperature superlattice. The aluminum film was reactive-ion etched in strips perpendicular to the current path, decreasing T_c of the etched sections by $\approx 2\%$, thus spatially modulating the transition temperature. At current densities ≤ 600 A/cm^2, the resulting structures exhibit a single homogeneous transition for etched lengths up to 50 μm, substantially longer than expected from the order-parameter decay length of 0.81 μm for the proximity effect in the dirty limit.

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Superconducting junctions[1,2] and superlattices[3] have been extensively studied before. For a superconductor-normal-metal-superconductor (S-N-S) junction, the order parameter in the normal region decays exponentially over a characteristic length K_N^{-1}. Providing that the critical current is not exceeded, the junction should display a sharp resistive transition. For larger currents, or longer lengths of the normal material, this homogeneity disappears, giving rise to discrete resistive transitions.

In this Letter, we report results on a thin-film analog of a modulated-T_c superlattice. This system exhibits strong proximity effects over a length scale quantitatively incompatible with the standard picture outlined above (originally formulated by de Gennes[4] and Werthamer[5]). Our samples consistently exhibit sharp resistive transitions for normal[6] regions up to 50 μm and for current densities up to 600 A/cm^2. This length is substantially longer than K_N^{-1} for thin-film aluminum in the dirty limit. Thus, superconductivity in this system persists over an unexpectedly long length scale.

Our samples are patterned aluminum thin films (thickness 250 Å, width 200 μm, and length 2 mm), simultaneously vapor deposited onto a silicon-nitride/silicon substrate. To modulate the T_c, a sample is coated with photoresist material and openings defined by photolithography. A reactive-ion etching process (described elsewhere[7]) is then performed, in which the undeveloped photoresist acts as an etch mask.[8] This process changes the T_c of the etched regions by $\approx 2\%$ with respect to the T_c of the unetched region. The mechanism for the change of T_c in the etched films is not understood. In our previous study,[7] the T_c of the etched region was enhanced by ~ 40 mK relative to the unetched sample. The etch used in the present study had a higher energy density than in the previous study and the sample stage was cooled during processing, resulting in a suppression of T_c by ~ 30 mK. We speculate that surface and substrate conditions are likely to be affected by these changes in the processing conditions and may be the controlling factors in the absolute T_c shifts.

A sample typically has fourteen modulated films, fabricated simultaneously on the same substrate under identical conditions. Each modulated film has different etched and unetched lengths, d_1 and d_2, respectively, patterned in series with one another. (One film is shown schematically in Fig. 1.) For control, uniformly etched and unetched films are always available on each sample. The T_c's of these uniform regions are referred to as T_{c1} and T_{c2}, respectively. A notable difference between this work and previous studies of others is that the constituents of our "superlattice" have nearly identical T_c's.

Our etching process is fairly noninvasive. The residual-resistance ratios (RRR) of the etched and unetched regions differ by no more than 0.5% and the sheet resistance by no more than 10%. The diffusion constants, as determined by the slope of the upper critical field as a function of temperature,[9] differ by no more than 20%. The similar resistive transition widths are further evidence of the consistent quality of the etched and unetched films.

The nature (whether discrete or homogeneous) of the resistive transitions was investigated by varying d_1 and d_2. Since the patterns were defined lithographically, these dimensions can be conveniently controlled with great accuracy. We have elected to fabricate the films in two configurations: (i) d_1/d_2 was held fixed while the modulation period, d_1+d_2 (denoted as Λ), was varied; (ii) d_1, or d_2, was held fixed while the ratio d_1/d_2 was

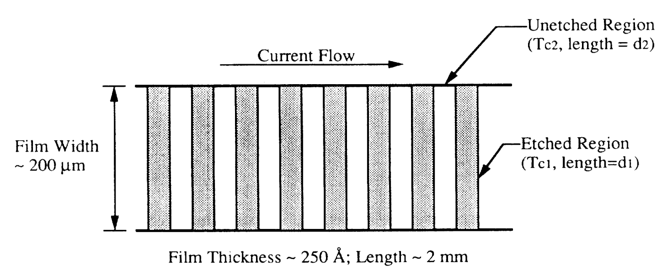

FIG. 1. Schematic of a modulated T_c structure, illustrating etched and unetched regions (d_1, etched length; d_2, unetched length).

varied. As T_c's tend to vary from sample to sample, we have found it convenient to study samples containing both configurations, thus allowing a systematic examination of the T_c of the modulated structures for particular values of T_{c1} and T_{c2}.

Unless otherwise specified, all measurements were carried out at a current density of 6 A/cm^2, using a standard four-terminal ac resistance bridge operating at 17 Hz. The resistance was measured by applying a constant current through a common pair of leads, while pairs of voltage leads corresponding to particular modulation periods Λ were monitored. In this Letter, T_c was chosen as the temperature where the normalized resistance equals one-half.[10] The uncertainty in T_c is taken to be ± 0.5 mK, the resolution of our thermometry and temperature control. Our quoted values of d_1 and d_2 have a systematic uncertainty of ± 0.2 μm, the typical linewidth control achieved in our lithographic process. Because of the excellent etch resistance of the photoresist, the resulting interface "roughness" is less than this value.

The striking nature of the resistive transitions is clearly seen in samples with a unity ratio of d_1/d_2, while Λ was varied from 4 to 400 μm. The resulting transitions are shown in Figs. 2(a) and 2(b) (data sets[11] F and D) in which the normalized resistance of the various films is plotted against the temperature. In Fig. 2(a), we plot the transitions of four films with $\Lambda = 4$, 10, 20, and 100 μm, along with the normalized resistance of a uniformly etched film on the left and a uniformly unetched film on the right. This sample exhibits sharp resistive transitions for these modulation periods, demonstrating that the proximity effect extends over a length scale of at least 50 μm (length of the etched section). This length scale is in quantitative disagreement with the de Gennes-Werthamer theory. We will elaborate on this point later in this Letter. As Λ is increased, the single transition should break up into two discrete transitions. This trend is borne out in our experiment and is illustrated in Fig. 2(b) for another sample with $\Lambda \gtrsim 100$ μm. A pronounced shoulder develops at the half resistance point, consistent with the geometry. We note that the system does not yet display two discrete transitions for Λ as large as 400 μm.

There are other noteworthy features in the data shown in Fig. 2. The shoulder observed at a normalized resistance of ≈ 0.9 is likely an artifact of sample geometry, or possibly associated with S-N interfaces between voltage probes and the films under study. It should be emphasized that this feature is present even in the uniformly etched film and is unrelated to the onset of the resistive transition in the modulated structures. Figure 2(a) also shows that an increase in Λ produces an *increase* in T_c of the modulated structure. This unexpected characteristic was observed in all samples measured in this study. Last, the resistive transitions are sensitive to the T_c difference $\Delta T_c \equiv T_{c2} - T_{c1}$. This has the consequence that the onset of discrete transitions is shifted to larger Λ for samples with smaller ΔT_c.

To unambiguously demonstrate that these observations are not the result of processing artifacts (which would presumably extend over a fixed length), we have examined structures in which the unetched length (d_2) was held fixed at 2 or 5 μm, while the etched lengths were varied so as to span a ratio of d_1/d_2 from 0.5 to 10. All transitions were sharp, consistent with the picture of a single homogeneous transition. The results for data set $E1$ with $d_2 = 5$ μm are shown in Fig. 3. Similar experiments were carried out on samples with a fixed d_1 of 5 μm while the ratio d_1/d_2 was varied over the same range of values. Again, the normalized T_c decreased smoothly and monotonically with increasing d_1/d_2, and, in fact,

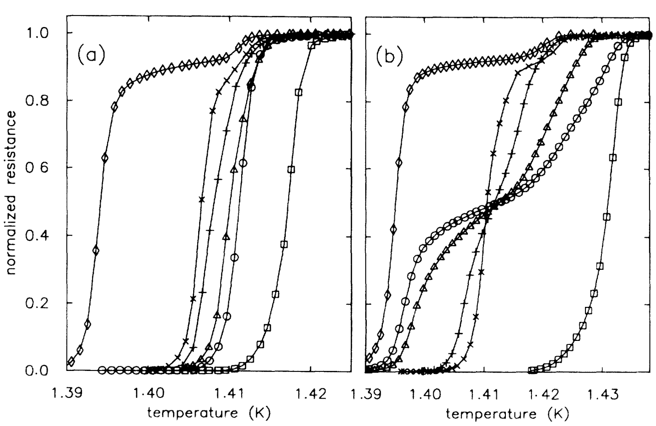

FIG. 2. Resistive transitions for data sets (a) F and (b) D with unity ratio d_1/d_2. In both (a) and (b), ◇ and □ are the uniformly etched and unetched films. In (a), resistive transitions shown are ×, +, △, and ○ for $\Lambda = 4$, 10, 20, and 100 μm. In (b), resistive transitions shown are ×, +, △, and ○ corresponding to $\Lambda = 20$, 100, 200, and 400 μm.

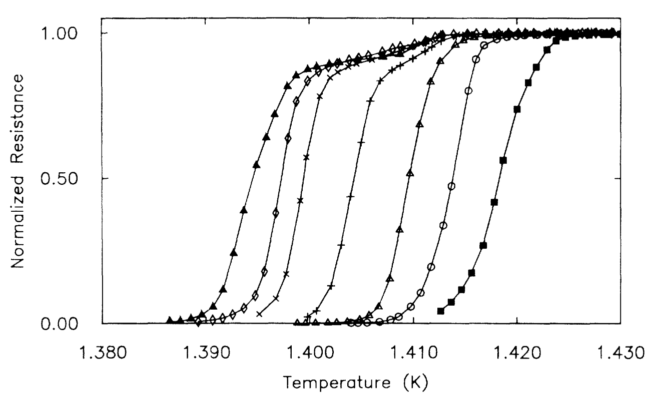

FIG. 3. Resistive transitions for data set $E1$ in which the ratio of etched to unetched lengths (d_1/d_2) is varied from 0.5 to 10 while d_2 is fixed and 5 μm. ■ and ▲ are the uniformly unetched and uniformly etched films. The symbols ○, △, +, ×, and ◇, correspond to etched lengths of 2.5, 5, 10, 25, and 50 μm, respectively.

the data of Fig. 3 were reproduced. This evidence confirms the absence of etching-induced artifacts on the masked regions.

Previous experiments have propagated supercurrents across dirty S-N-S junctions of many coherence lengths.[12,13] To assess the strength of the proximity effect in our system, we have measured the effects of higher excitation currents and weak applied magnetic fields. In Fig. 4, we plot the resistive transitions for the etched, unetched, and the $\Lambda = 100$ μm films of Fig. 2(a). The suppression of the transition temperatures at current densities up to 600 A/cm^2 are shown for the etched and $\Lambda = 100$ μm films. The sharp and uniform nature of the transitions is maintained at these current densities, supporting our identification of a strong, long-range proximity effect. (Higher current densities could not be explored using these samples due to heating effects.) For homogeneous thin films close to T_c, the current density varies as[9] $J_c = J_{c0}(1 - T/T_c)^{3/2}$. For the uniformly etched film, we estimate (using the data of Fig. 3) the constant J_{c0} to be $\approx 2.3 \times 10^6$ A/cm^2. This is a reasonable value for thin films.

A magnetic field of 1 mT produced noticeably broader transitions and suppressed T_c by ≈ 50 mK. However, the homogeneous signature of the resistive transition was preserved in these fields. Typical thin-film behavior was observed down to ≈ 0.4 K, in fields up to 30 mT, but substantial broadening of the transitions is present.

To examine the dependence of the transition temperature on the ratio d_1/d_2, we present the results of three representative data sets ($D1$, E, and $E1$) in Fig. 5. To facilitate the comparison between data sets, we have normalized the transition-temperature shift, $T_c - T_{c1}$, to ΔT_c. The data sets fall on similar curves and can be fitted by the expression

$$\frac{\Delta T_c}{T_c - T_{c1}} = 1 + p\frac{d_1}{d_2}. \qquad (1)$$

This is Eq. 6-66 in Ref. 3 (the result of a linearized Ginzburg-Landau treatment of a superlattice), in the limit that the effective coherence length is much longer than the modulation lengths. The factor p, which is of order unity, is a parameter in the theory related to the logarithmic derivative of the order parameter across an interface. Here, we treat p as a fitting parameter. We have selected this equation since we observed that the quantity $T_c - T_{c1}$ was strongly affected by the ratio d_1/d_2 and only weakly dependent on their absolute magnitudes. The ΔT_c of our samples was typically between 22 and 46 mK. The fitting parameter p was found to be of order 1. We note the apparent size dependence of p (data sets E and $E1$ of Fig. 5). These data were taken from films fabricated simultaneously on a single substrate and should have identical values of ΔT_c. The two data sets have the same range of the ratios d_1/d_2 but of different fixed unetched lengths ($d_2 = 2$ μm for E and $d_2 = 5$ μm for $E1$). The values of p which best fit the data are 0.9 and 0.75, respectively.

The results presented in this Letter are summarized as follows: (a) The proximity effect extends over lengths up to 50 μm, corresponding to a periodicity of 100 μm. (b) The normalized transition temperatures for $\Lambda \leq 50$ μm are determined by the ratio d_1/d_2. (c) As ΔT_c decreases, the normalized transition temperature at which a structure having a fixed value of d_1/d_2 undergoes its superconducting transition increases. (d) There is a weak dependence of the normalized transition temperature on Λ. (Smaller Λ results in a lower T_c for a fixed ratio of

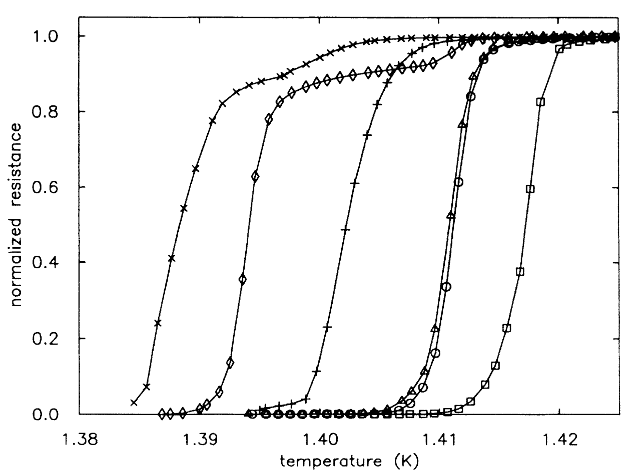

FIG. 4. Resistive transitions of uniformly etched (◇), unetched (□), and $\Lambda = 100$ μm (○) films at a current density of 6 A/cm^2. Also shown are results at 60 A/cm^2 for the $\Lambda = 100$ μm (△) film; and at 600 A/cm^2 for the etched (×) and $\Lambda = 100$ μm (+) films. Note the homogeneous transitions for all current densities used, confirming the existence of a long length scale.

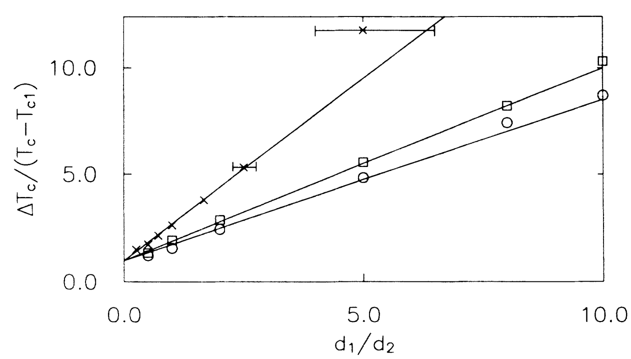

FIG. 5. Reduced data for data sets $E1$ (○), E (□), and $D1$ (×), with $\Delta T_c = 23.8$, 23.8, and 36.0 mK, respectively. The inverse normalized T_c shift is plotted against the ratio d_1/d_2. Solid lines are fits by Eq. (1) in text for values of the parameter $p = 0.75$, 0.9, and 1.7, respectively. Note the dependence of p on modulation length in the data sets E and $E1$.

d_1/d_2.)

Within the theory of de Gennes and Werthamer, which is applicable[14] to systems in the dirty limit ($l \ll \hbar v_F/2\pi k_B T$), the order parameter decays exponentially in the normal region with a characteristic length[15]

$$K_N^{-1} = \xi_N(T) \left[1 + \frac{2}{\ln(T/T_{cN})} \right]^{1/2}, \qquad (2)$$

where $\xi_N(T) \equiv (\hbar D/2\pi k_B T)^{1/2}$, $T_{cN} \equiv T_{c1}$, and D is the diffusion constant of the normal region. The mean free path l is estimated to be 120 Å using $v_F = 1.3 \times 10^8$ cm/s and the experimental value of the diffusion constant $D = 50 \pm 5$ cm^2/s. For the $\Lambda = 100$ μm film of Fig. 2(a), $\xi_N(T) = 650$ Å, and thus K_N^{-1} is estimated to be 0.81 μm for $T = 1.412$ K and $T_{cN} = 1.394$ K. We note that the theory of de Gennes and Werthamer explicitly accounts for the possibility that the normal region is a superconductor above its transition temperature.

The critical current density J_c of dirty *S-N-S* junctions has been found to scale[12,13] as $J_c = J_0(1 - T/T_{cs})^2 \times \exp(-K_N d_N)$, where d_N is the length of the normal region (d_1). For parameters of the $\Lambda = 100$ μm film ($d_N = 50$ μm), the exponential factor is of order 10^{-27}. The constant J_0 is typically less than 10^7 A/cm^2 in thin films, implying that J_c would be vanishingly small. This is clearly incompatible with our measured current density of *at least* 6 A/cm^2 close to T_c.

The de Gennes–Werthamer theory has been successful in describing experiments in the dirty limit to date. The theory assumes that the order parameter decays as a single exponential with characteristic length given by Eq. (2). Our system is always in the regime where the calculated K_N^{-1} is small compared with the length of the normal (etched) region. This is the requirement for the validity of the single-exponential description. However, the superconducting and normal regions of our system have nearly identical transition temperatures. This regime has not been explored by any previous experimenters. Provided the gap parameters are small (always satisfied near T_c), there is no obvious reason to exclude the applicability of the theory in this regime.[16] We speculate that our system, with its similar T_c's, may not be adequately described by a single-exponential decay of the order parameter.

To observe the limitations of and departure from existing theory, it would be useful to systematically increase ΔT_c in a controlled fashion without significantly changing other transport properties. Unfortunately, a large shift of the T_c cannot be easily achieved using the present technique. Possible alternative approaches might include varying the thickness of the aluminum films, using magnetic/paramagnetic overlayers, and stronger etches to damage or remove the aluminum.

In conclusion, we have carried out a survey of the properties of a novel 2D analog of a modulated-transition-temperature superlattice. In the different samples studied, homogeneous transitions are observed for samples having 1:1 modulation periods as large as 100 μm. The magnitude of the critical current, as well as the reasonable agreement with Eq. (1), strongly suggests that the order parameter decays over a length substantially *longer* than expected. We believe that a satisfactory explanation of this effect should also account for the observed trends in Λ and ΔT_c.

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[4]G. Deutscher and P. G. de Gennes, in *Superconductivity,* edited by R. D. Parks (Marcel Dekker, New York, 1969).

[5]N. R. Werthamer, Phys. Rev. **132**, 2440 (1963).

[6]Here, normal refers to a superconductor at a temperature above its T_c.

[7]Y. K. Kwong *et al.*, J. Vac. Sci. Technol. B **7**, 2020 (1989).

[8]The Freon etch used in the process is not expected to react with the aluminum or the resist. The use of the word "etch" refers to the processing procedure and not the chemistry. We expect the process results in negligible removal of the aluminum.

[9]M. Tinkham, *Introduction to Superconductivity* (Krieger, Malabar, FL, 1980).

[10]Since the widths of the resistive transitions are similar for all films with homogeneous transitions, a different criterion for T_c would not affect the conclusions presented in this Letter.

[11]We have taken data on seven samples denoted *A* through *G*. A sample typically consists of two data sets with different configurations of d_1 and d_2.

[12]J. Clarke, Proc. Roy. Soc. London A **308**, 447 (1969).

[13]For experiments performed in the clean limit, see, for example, J. G. Shepherd, Proc. Roy. Soc. London A **326**, 421 (1972). These and other experiments confirmed the clean-limit theory.

[14]For a discussion of the regime of validity for the de Gennes–Werthamer theory, see Clarke (Ref. 12), p. 462.

[15]K_N is given by the roots of a transcendental equation. See, for example, Eq. (3) in P. G. de Gennes and J. P. Hurault, Phys. Lett. **17**, 181 (1965), and Eq. (18) of Ref. 4. Our numerical solution for K_N using this result agrees within 15% with the simpler Eq. (2) in the text for T/T_{cN} ranging from 1.004 to 2. Thus, Eq. (2) is adequate for estimating K_N^{-1}.

[16]It has been suggested to us that this system may be viewed as an array of thin-film *S-N-S* junctions in series, which may introduce different boundary conditions than those in the de Gennes–Werthamer theory.

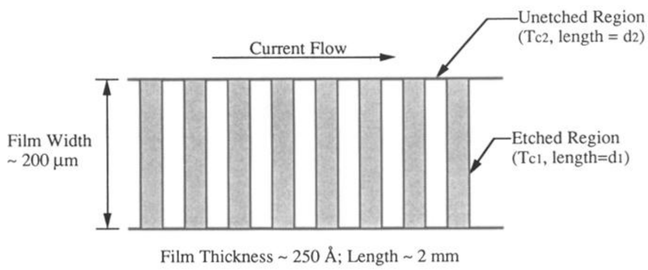

FIG. 1. Schematic of a modulated T_c structure, illustrating etched and unetched regions (d_1, etched length; d_2, unetched length).