Long-range proximity effect in aluminum thin films with regions of nearly identical transition temperatures

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We have investigated the resistive transition of dirty aluminum thin films whose transition temperature is spatially modulated by $\sim 2-4\%$ along the current path. At current densities as high as 600 A/cm$^2$, the system exhibits a single homogeneous transition for modulation lengths up to 50 $\mu$m, an order of magnitude longer than predicted by current theories. In the limit that the modulation length is $< 50$ $\mu$m, the $T_c$ depends sensitively and monotonically on the ratio of the lengths of the etched and the unetched regions ($d_1/d_2$), but only weakly on their absolute magnitudes. This behavior is reproduced for $T_c$ modulation parallel or perpendicular to the current flow. For samples with a controlled aperiodic (Fibonacci) modulation, the $T_c$ is compatible with that of periodically modulated films. The dependence of the $T_c$ on $d_1/d_2$ can be fitted to a simple Ginzburg-Landau theory which assumes that the effective pair coherence length is long in comparison with the modulation period. Despite the good fit, data for the full range of experimentally accessible modulation length scales are not consistent with a more detailed microscopic theory. We believe this long-range proximity effect is not the consequence of pair tunneling, but is a manifestation of the long distance over which quasiparticle phase coherence is maintained in a two-dimensional metal film.

I. INTRODUCTION

We have examined the anomalously long-range superconducting proximity effect in a two-dimensional (2D) analog of a modulated-$T_c$ superlattice. This system was prepared by surface-damaging selected regions of an otherwise uniform aluminum thin film. The technique permits us to study the proximity effect in the limit that the $T_c$'s in a "two-component" system are similar, while simultaneously constraining other transport properties of the higher- and lower-$T_c$ regions to be approximately the same. This is an unexplored regime since previous experiments$^{1,2}$ on the proximity effect typically utilize two distinct metals, which would, in general, entail rather different $T_c$'s and associated transport properties.

The accepted theory for describing the proximity effect at dirty superconducting-normal (S-N) interfaces is due to de Gennes$^{2,3}$ and Werthamer$^4$ (dGW). Numerous experiments were carried out, chiefly in the 1960s, and agreement between experiment and theory was shown to be good.$^{1,2}$ Subsequent theoretical$^5$ and experimental$^6,7$ work was carried out to encompass clean systems. In keeping with standard terminology, the "N" region may be a superconductor above its $T_c$.

In both the dirty- and clean-limit theories, the superconducting order parameter decays exponentially on the $N$ side of a S-N interface with a characteristic range, called $K_N^{-1}$. In the dGW description of a dirty system, for $T < T_c > T_{cN}$, $K_N^{-1}$ is equal to the so-called normal coherence length, $(\xi D / 2\pi k_B T)^{1/2}$. Here, $D$ is the quasiparticle diffusion constant, and $T_{cN}$ is the $T_c$ of the N region. As $T$ approaches $T_{cN}$ from above, $K_N^{-1}$ diverges,$^1$ to leading order in the reduced temperature as $[(n (T/T_{cN})]^{-1/2}$. Even at $T/T_{cN} \sim 1.01$, $K_N^{-1}$ is not expected to exceed $\sim 1$ $\mu$m in dirty aluminum thin films when $D \sim 50$ cm$^2$/s. For S-N-S systems with the $N$ region substantially longer than $1$ $\mu$m, the critical current density $J_c$ should be vanishingly small since $J_c$ is proportional to $\exp(-K_N d_N)$, where $d_N$ is the length of the $N$ region. Yet, we have found$^{8,9}$ that the observed proximity effect in our modulated-$T_c$ structures is substantially more robust, and is quantitatively incompatible with the dGW theory. Despite the apparent inconsistency, it is unclear why our dirty-limit system should be outside the regime of validity of this theory.

In this paper, we will first describe the fabrication techniques used to prepare these modulated-$T_c$ structures. In particular, the reactive-ion etching process used to locally suppress the $T_c$ of the aluminum film by a few percent will be discussed. We will present the resistive transitions of these modulated structures, emphasizing first the long length scale ($> 50$ $\mu$m) and large critical current density ($> 600$ A/cm$^2$) exhibited by the proximity effect. We then focus on the limit when the modulation period is sufficiently small ($< 50$ $\mu$m) so that the modulated structures exhibit single sharp resistive transitions. Effects of modulation direction and quasiperiodic modulation will be described. Data in the single-transition limit can be compared with different theoretical models, making precise the anomalous nature of this system. In conclusion, we will suggest a possible mechanism which may be responsible for the anomalously long range of the observed proximity effect.
II. THE SAMPLES

An aluminum thin film is first prepared by thermal evaporation and a lift-off process on a photolithographically patterned Si$_3$N$_4$/Si substrate. The nominal film thickness used throughout this study is 250 Å, while the typical width and length are 200 μm and 2 mm, respectively. The typical diffusion constant $D$ of the films is $\sim 50$ cm$^2$/s, as determined from the temperature dependence of the upper critical field close to $T_c$. This value of $D$ corresponds to a mean free path $l$ of $\sim 100$ Å. Since $l$ is substantially smaller than the intrinsic clean-limit coherence length of aluminium ($\sim 1.3$ μm), our films are always in the dirty limit as far as the properties in the superconducting state are concerned.

The substrate on which the aluminum film has been deposited is then coated with resist material. Photolithography is, once again, performed to expose selected regions along the film, with the remaining (undeveloped) resist acting as the etch mask in subsequent processing steps. The sample is then subjected to a plasma of CHF$_3$ and O$_2$ gases. This process has the effect of suppressing the $T_c$ by $\sim 2$–4 %, without appreciably affecting other transport properties. (The details of the fabrication and etching process are reported elsewhere.)

Photolithography permits any arbitrary configurations of lower-$T_c$ (etched) and higher-$T_c$ (unetched) regions to be defined on an otherwise continuous film. Two possible configurations pertinent to this work are shown schematically in Fig. 1, in which the $T_c$ is modulated periodically in a direction perpendicular [Fig. 1(a)] and parallel [Fig. 1(b)] to the current flow. For nomenclature purposes, we define the $T_c$'s of the etched and unetched section as $T_{c1}$ and $T_{c2}$, respectively. The length of the etched and unetched section along the direction of $T_c$ modulation is $d_1$ and $d_2$, respectively. The period of modulation $d_1 + d_2$ is $\Lambda$, and the $T_c$ difference $T_{c2} - T_{c1}$ is $\Delta T_c$.

Figure 2 is a schematic layout of a typical (4 mm)$^2$ sample. Each sample typically contains sixteen films together with associated current and voltage leads suitable for four-point resistance measurements in a $^3$He cryostat. This permits films with different configurations of $d_1$ and $d_2$ (e.g., $d_1/d_2$ is varied while $d_1$ is fixed) to be prepared simultaneously on a single substrate under identical conditions. On each sample, a uniformly etched and a uniformly unetched film are always available for control. Their $T_c$'s operationally define $T_{c1}$ and $T_{c2}$, respectively.

The CHF$_3$-O$_2$ plasma employed is typically used for etching Si$_3$N$_4$. As such, exposing the aluminum film to such a plasma is not formally an etch, and no appreciable amount of aluminum is expected to be removed as a result. For convenience, we will continue to refer to the region of the film exposed to the plasma as the etched region.

Due to the excellent etch resistance of photoresist, the edge roughness of the resulting etched-unetched interfaces is expected to be better than $\pm 0.2$ μm, the typical line width control of our photolithographic process. Using scanning electron microscopy and Auger electron microanalysis, we have ascertained that this process, indeed, gives rise to sharp interfaces on a length scale better than 1 μm. Further, by Auger depth profiling, we found that only surface damage results in the etched region.

The plasma apparently interacts only with the native oxide residing on the top 50 Å of the aluminum film. This oxide surface layer is always present upon exposure of the film to room air. Auger electron analysis indicates that within this top surface layer, oxygen content is reduced while the fluorine content is enhanced in the etched regions. This is clearly seen in the Auger line scans of Fig. 3. Here, the fluorine and oxygen signals are plotted as a function of position along a film with $d_1 = d_2 = 10$ μm. Although the magnitude and the sign of $\Delta T_c$ is fairly sensitive to the exact process conditions, the depression of $T_c$ in the etched region is clearly correlated with a suppression of oxygen, which is consistent with the behavior for aluminum reported in the literature.

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![FIG. 2. Layout of a typical sample. This structure contains 14 modulated films together with the uniformly etched and uniformly unetched films electrically in series. Each film constitutes a leg of this meander pattern. The associated leads and square bonding pads permit individual films to be connected to a four-terminal resistance bridge.](image)

![FIG. 1. Schematics of modulated-$T_c$ structures, illustrating etched and unetched regions whose transition temperatures are $T_{c1}$ and $T_{c2}$, respectively. We refer to the modulation in (a) as perpendicular while that in (b) as parallel.](image)
The role of the fluorine is as yet unclear, aside from its apparent propensity for replacing oxygen.

Besides the suppression of the $T_c$ by a few percent, the etching process is noninvasive to the film’s other transport properties. First, the normal-state transport properties are not substantially degraded. The resistance rises by $<20\%$, the residual resistance ratio (RRR) decreases by $<0.5\%$, and the low-temperature value of $D$ decreases by $<10\%$. More importantly, the width of the resistive transition (which we define as the temperature range for which the resistance drops from 90$\%$ to 10$\%$ of its normal-state resistance) is about the same before and after etching, both $\sim 10$ mK. Lastly, as determined from quantum transport studies, the inelastic-scattering rate is not substantially increased by the etch.

We have carried out magneto-resistance measurements$^{12,13}$ on etched films together with contiguous unetched films. These measurements were performed in the normal state for $H < 300$ G and $1.5 < T < 7$ K. By fitting these data to the theories of weak localization and superconducting fluctuations, we can obtain the inelastic-scattering (dephasing) rate, $\tau_{\text{in}}^{-1}$, as a function of temperature. For typical etching times used, we have established that both the etched and unetched films exhibit nearly identical values of $\tau_{\text{in}}^{-1}$. Also, the same temperature dependence of this rate, $\tau_{\text{in}}^{-1} \sim AT^2 BT^3$, was observed for temperatures up to $\sim 7$ K. These results further confirm the noninvasive nature of our process and are useful for future discussions.

III. EXPERIMENTAL RESULTS

In order to investigate the resistive transitions of these structures, we have patterned the films in four basic configurations: (i) perpendicular modulation [see Fig. 1(a)] with $d_1/d_2$ held fixed and the modulation period, $\Lambda = d_1 + d_2$, varied; (ii) perpendicular modulation with $d_1$, or $d_2$, held fixed and the ratio $d_1/d_2$ varied; (iii) same as (ii) except the modulation is parallel [see Fig. 1(b)], rather than perpendicular, to current path; and (iv) quasi-periodic perpendicular modulation. As the exact $T_c$'s tend to vary somewhat from sample to sample, we have found it convenient to study samples containing a multiple number of these configurations (usually two), thus allowing a systematic examination of the $T_c$ of the modulated structures for particular values of $T_{c1}$ and $T_{c2}$.

We have made measurements on a total of 11 samples, denoted by an upper-case letter. Each sample typically consists of two data sets with different configurations of $d_1$ and $d_2$, referred to as data sets “0” and “1.” (Thus, the two data sets on sample F are “F0” and “F1,” respectively.) Unless otherwise specified, all measurements were carried out at a current density of 6 A/cm², using a standard four-terminal ac resistance bridge operating at 17 Hz. Each measurement is averaged over a minimum of 30 readings.

Thermometry is achieved by monitoring the resistance of a calibrated germanium sensor. For structures with a single sharp resistive transition, $T_c$ is well defined. For these structures, we have chosen the $T_c$ as the temperature where the normalized resistance equals one-half. Since the widths of the resistive transitions are similar for all films with single sharp transitions, a different criterion for $T_c$ would not affect the conclusions of this paper. The uncertainty in $T_c$ is taken to be $\pm 0.5$ mK, the stability of our temperature control during a ramp.

A. Existence of a long length scale

The striking nature of the resistive transitions is shown in Figs. 4(a) and 4(b) (data sets F0 and D0), in which the normalized resistance of the various films is plotted against the temperature. In these samples, the ratio $d_1/d_2$ was maintained at unity, while $\Lambda$ was varied from 4 to 400 $\mu$m. In Fig. 4(a), we plot the normalized resis-

![FIG. 4. Resistive transitions with unity ratio $d_1/d_2$ for data sets F0 (a) and D0 (b). In both (a) and (b), solid and open circles are the uniformly etched and unetched films. In (a), resistive transitions shown are: $\times$, $+$, $\triangle$, and $\circ$ for $\Lambda = 4, 10, 20$, and 100 $\mu$m. In (b), resistive transitions shown are: $\times$, $+$, $\triangle$, and $\circ$ corresponding to $\Lambda = 20, 100, 200$, and 400 $\mu$m.](image-url)
of four films with Λ = 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 modulation periods ≤ 100 µm. An increase in Λ produces an increase in $T_c$ of the modulated structure, as observed in all samples measured in this study. Figure 2(b) illustrates the onset of discrete transitions in another sample for Λ ≥ 100 µm. A pronounced shoulder develops at the half-resistance point, consistent with the geometry. Taken together, these observations suggest the existence of a long length scale over which superconductivity persists.

A number of systematic features should be discussed. The shoulder observed at a normalized resistance of ~0.9 is an artifact of sample geometry and is caused by S-N interfaces between leads and the structures 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. It can be eliminated if the leads and bondings pads are uniformly etched. As expected, the resistive transitions are sensitive to $\Delta T_c$. This has the effect of shifting the onset of discrete transitions to larger Λ for samples with smaller $\Delta T_c$.

Previous experiments have propagated supercurrent across dirty S-N-S junctions of many coherence lengths. To assess the strength of the proximity effect in our system, we have measured the effect of higher excitation currents and weak applied magnetic field. In Fig. 5, we plot again the resistive transitions for the etched, unetched, and the Λ = 100 µm films of Fig. 4(a). The suppression of the temperature of the resistive transition is shown for the etched and Λ = 100 µm films at current densities up to 600 A/cm². The sharp and uniform nature of the transitions are maintained at these current densities. (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 $J_c = J_{c0}(1 - T/T_c)^{3/2}$. For the uniformly etched film, we estimate (using the data of Fig. 5) the constant $J_{c0}$ to be ~2.3 × 10⁶ A/cm², which 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.

### B. Monotonic dependence of $T_c$ on $d_1/d_2$

We now focus on the regime when $d_1 < 50$ µm. In this limit, the modulated films exhibit a single sharp resistive transition with a well-defined $T_c$. In Fig. 6, we plot the resistive transition of a sample with $\Delta T_c = 35$ mK. For this sample, $d_1$ is fixed at 5 µm, while $d_2$ is varied from 1 to 20 µm. As expected, the $T_c$'s of these modulated films are between $T_c^1$ and $T_c^2$, the $T_c$'s of the uniformly etched and unetched controls. The transitions are shifted monotonically to a higher temperature as $d_2$ is increased.

Choosing the half-resistance temperature as the criterion for $T_c$, we can examine the dependence of $T_c$ on $d_1/d_2$. For the sample of Fig. 6, the monotonic dependence of the $T_c$ on this ratio is clearly illustrated as (○) in Fig. 7. In two other samples with $\Delta T_c ~ 25$ mK, $d_1$ was fixed at 5 µm in one, while $d_2$ was fixed at 5 µm in the other. In these samples, the dependence of $T_c$ on $d_1/d_2$ is similarly monotonic. It is, in fact, identical within experimental scatter. This result is shown in the inset of Fig. 7.

The monotonic dependence of $T_c$ on $d_1/d_2$ indicates that the samples produced by the etching technique are free from artifacts. First, although our technique induces surface damage only, the undamaged section beneath the surface of the film is not electrically shunting the top, which would have resulted in all the transitions having approximately the same $T_c$. This should be obvious considering the fact that the coherence length is substantially longer than the thickness of the film, and the system is,
FIG. 7. Normalized $T_c$ shift vs $d_1/d_2$ for various samples. Data set D1 of Fig. 6 is replotted here ($\bigcirc$), illustrating the monotonic dependence of $T_c$ on $d_1/d_2$. A Fibonacci lattice ($\times$) on the same sample as D1 yields a $T_c$ compatible with the periodically-modulated films. (See Sec. III D.) Data sets E0 ($\triangle$) and E1 (+) belong to a single sample with $\Delta T_c = 24$ mK, for which $d_2 = 2 \mu m$ for E0 and $d_2 = 5 \mu m$ for E1. Note that for a given value of $\Delta T_c$, $T_c$ depends primarily on the ratio $d_1/d_2$ and only weakly on their absolute magnitudes. In the inset, data set E1 (+) is replotted along with data set C0 ($\checkmark$). C0 has $d_1 = 5 \mu m$ and $\Delta T_c = 25$ mK. Note the same dependence of $T_c$ on $d_1/d_2$, even though the etched length, rather than the unetched length, is fixed in C0. Solid lines are fits to Eq. (13) in the text.

data sets for films on a single substrate with $d_2$ fixed to be $3 \mu m$. The direction of modulation is perpendicular to current flow in one data set and parallel in another, as depicted in Figs. 1(a) and 1(b). We see that both modulation directions result in the same $T_c$ as the ratio $d_1/d_2$ is varied. Note that the film width (fixed at $200 \mu m$) is, in general, not an integral multiple of the modulation period $\Lambda$. Thus, the regions along the film edge, which are always etched, will be narrower than the designated value of $d_1$. For small values of $d_1/d_2$, the number of modulation periods across the film is large, and this effect is unimportant. However, this effect becomes important at larger values of $d_1/d_2$, consistent with the deviation between the two data sets in Fig. 8 at large $d_1/d_2$.

The independence on modulation direction means that the unetched region is not electrically shunting the etched region. Thus, the film as a whole exhibits a single-transition temperature. Also, for the current density used, our measurements are free from complication by self-heating or nonequilibrium effects associated with $N$-$S$ interfaces.\(^{14}\) Thus, the observed resistive transitions reflect transitions of the order parameter and give the thermodynamic $T_c$. Of course, these results are further evidence that the system is free from fabrication related artifacts.

D. Quasiperiodic modulation experiment

We have explored whether periodicity is an important aspect of this system. To do so, we have modulated the $T_c$ of some of the measured films in a controlled aperiodic fashion, according to the Fibonacci series:

$$u, \text{v}_e, \text{v}_e, \text{v}_e, \text{v}_e, \ldots$$

Here, $u$ and $e$ denote $5-\mu m$-long unetched and etched regions, respectively. The series is continued for a total length of 400 units to fill the 2-mm length of the film. By counting the total number of $u$ and $e$, it can be verified that the total etched length to the total unetched length is $\sim 0.6$ for this structure. This quasiperiodic modulation
scheme has been previously applied\textsuperscript{15} to study semiconductor heterostructures.

Figure 7 shows that the Fibonacci film (×) has a \( T_c \) which is compatible with the periodically modulated films prepared on the same substrate. This suggests that the periodicity of these structures is unimportant.

IV. COMPARISON WITH MODELS

A. Breakdown of the dGW description

The mean free path \( l \) of our aluminum films is \( \sim 120 \text{ Å} \), using \( v_F=1.3\times10^8 \text{ cm/s} \) and the experimentally obtained diffusion constant of \( 50 \pm 5 \text{ cm}^2/\text{s} \). This value of \( l \) is much less than the intrinsic coherence length for aluminum (\( \hbar \nu_F/2\pi k_B T_c \sim 1.3 \mu\text{m} \)). Thus, our system is dirty; a necessary condition for the dGW theory.

Within the dGW description, the order parameter (pair wave function) \( \psi \) decays exponentially in the \( N \) region with a characteristic length \( N^{-1} \) given implicitly\textsuperscript{16} by the transcendental equation:

\[
\ln \left( \frac{T}{T_{CN}} \right) = \psi \left[ \frac{1}{2} - \psi \left( \frac{1}{2} - \frac{\hbar D K_N^2}{4\pi k_B T} \right) \right].
\]  

(1)

Here, \( T_{CN} \equiv T_{c1} \), \( D \) is the diffusion constant of the \( N \) region, and \( \Psi \) is the digamma function. For the \( \Lambda = 100 \mu\text{m} \) film of Fig. 4(a), \( T_{CN} = 1.394 \text{ K} \). With this value, the decay length \( K_N^{-1} \) can be calculated numerically for \( T > T_{CN} \). The results for \( T/T_{CN} > 1 \) are plotted in Fig. 9. Note that for \( T/T_{CN} > 1 \), \( K_N^{-1} < 1 \mu\text{m} \) and that even for \( T/T_{CN} = 1.0005 \), \( K_N^{-1} < 5 \mu\text{m} \). For the \( \Lambda = 100 \mu\text{m} \) film, the resistive transition occurs at a temperature of \( 1.412 \text{ K} \). This corresponds to \( T/T_{CN} = 1.013 \) and, thus, \( K_N^{-1} \) is expected to be less than \( 1 \mu\text{m} \).

For convenience, Clarke\textsuperscript{1} has used the following closed-form expression for \( K_N^{-1} \):

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

(2)

in which the so-called normal coherence length \( \xi_N(T) = (\hbar D/2\pi k_B T)^{1/2} \) is \( \sim 650 \text{ Å} \). To within \( \pm 10\% \), the values of \( K_N^{-1} \) obtained from Eqs. (1) and (2) agree with each other for \( T/T_{CN} \) as small as 1.0005, as seen in Fig. 9. For comparison purposes only, the function \( K_N^{-1} = \xi_N(T)(T/T_{CN} - 1)^{1/2} \) is also plotted in Fig. 9 (dashed line). This is the form of the Bardeen-Cooper-Schrieffer (BCS) coherence length close to \( T_c \). Note that the divergence of \( K_N^{-1} \) in \( T/T_{CN} \) of the BCS form is somewhat slower than (1) and (2).

Previous experiments have propagated supercurrents across dirty \( S-N-S \) junctions of many coherence lengths.\textsuperscript{1,6} The critical current density \( J_c \) of these junctions has been found to scale as \( J_c = J_0(1 - T/T_{CS})^2 \exp(-K_D d_N) \), where \( d_N \equiv d_1 \). For parameters of the \( \Lambda = 100 \mu\text{m} \) film (\( d_N = 50 \mu\text{m} \)) of Fig. 4(a), the exponential factor is of order \( 10^{-27} \). The constant \( J_0 \) is typically less than \( 10^7 \text{ A/cm}^2 \) in thin films, implying that \( J_c \) would be vanishingly small. This is clearly incompatible with our measured current density of at least \( 6 \text{ A/cm}^2 \) close to \( T_c \).

The dGW theory has been successful in describing experiments in the dirty limit to date. Our work concentrates on a system where the \( S \) and the \( N \) regions have nearly identical \( T_c \)'s. This regime has not been previously explored experimentally. Provided that the gap parameters are small (always satisfied near \( T_c \)), there is no obvious reason to exclude the applicability of the theory to our system. As will be discussed further, it is likely that Josephson tunneling across a \( S-N-S \) barrier and the resulting single-exponential description of the order-parameter decay are not appropriate for our system. To elucidate the anomalous nature of this system, we now examine the data for films with \( d_1 < 50 \mu\text{m} \), which exhibit single sharp transitions. This is a regime suitable for a more detailed comparison with theories.

B. The single transition limit:

The Ginzburg-Landau regime

Jin and Ketterson\textsuperscript{17} have solved the problem of a two-component superlattice in the Ginzburg-Landau (GL) regime. In the limit \( T \rightarrow T_c \), the order parameter \( \psi \) obeys the linearized GL equation. For spatial variation only in one direction without an applied field, the equation is

\[
\frac{\hbar^2}{4m} \frac{d^2 \psi}{dx^2} = a(T) \psi.
\]

(3)

The coefficient \( a(T) \) is related to the GL coherence length \( \xi \) via

\[
a(T) = -\frac{\hbar^2}{4m \xi^2} = \alpha(T_c - T),
\]

(4)

where \( \alpha \) is a temperature-independent factor and \( m \) is the effective mass of the carriers. Note that our modulated structures satisfy the GL limit because \( T_{c1} < T < T_{c2} \) and \( T_{c2}/T_{c1} \sim 1.04 \).
Equation (3) holds separately in the etched and the unetched regions. Defining a common coordinate system (where \(0 < x < d_1\) is the etched region with \(T_c = T_{c1}\) and \(d_1 < x < d_2\) is the unetched region with \(T_c = T_{c2}\)) and observing that the order parameters for the two regions, \(\psi_1\) and \(\psi_2\), must be symmetric with respect to the center of each region, we can write

\[
\psi_1 = A \cos \left( \frac{x - d_1/2}{\xi_1} \right),
\]

\[
\psi_2 = B \cos \left( \frac{x - d_1 - d_2/2}{\xi_2} \right).
\]

The appropriate boundary condition is that the logarithmic derivative at \(x = d_1\) is continuous, i.e.,

\[
p \frac{d}{dx} (\log \psi_1) = \frac{d}{dx} (\log \psi_2).
\]

Here, \(p\) is a factor of order unity characterizing interface conditions.

The highest temperature for which a solution exists is identified as the \(T_{c}\) of the periodic structure. Applying the boundary condition (7) to (5) and (6) yields, for \(T_{c1} < T < T_{c2},\)

\[
p \tan \left( \frac{d_1}{2 \xi_1} \right) = -\frac{1}{\xi_2} \tan \left( \frac{d_2}{2 \xi_2} \right).
\]

We now assume that the effective coherence lengths of the two regions are much larger than the modulation length scales, i.e., \(\xi_1 \gg d_1\) and \(\xi_2 \gg d_2\). This assumption is used in view of the long characteristic range of the proximity effect and that the system is in the single-transition limit. Its validity will be further discussed later. Thus, (8) reduces to

\[
p \left( \frac{d_1}{\xi_1} \right) \approx -\frac{d_2}{\xi_2},
\]

which, according to (4), can be rewritten as

\[
p m_1 \alpha_d (T_c - T) = -m_2 \alpha_d (T_{c2} - T).
\]

Solving for \(T\) gives the \(T_c\) of the modulated-\(T_c\) structure,

\[
T_c = \frac{p m_1 \alpha_d T_{c1} + m_2 \alpha_d T_{c2}}{p m_1 \alpha_d + m_2 \alpha_d}.
\]

The two different regions of our film have nearly equal \(T_c\) and normal-state transport properties. Thus, the ratios \(m_1/m_2\) and \(\alpha_1/\alpha_2\) should be near unity. These ratios can be incorporated into the factor \(p\), which will be used as a fitting parameter in the following analysis of data. The transition temperature is then

\[
T_c = \frac{p d_1 T_{c1} + d_2 T_{c2}}{p d_1 + d_2}.
\]

or

\[
\frac{\Delta T_c}{T_c - T_{c1}} = 1 + \frac{d_1}{d_2}.
\]

![FIG. 10. Reduced data for data sets E1 (○), E0 (□), D1 (×), and S1 (◇), with \(\Delta T_c = 23.8, 23.8, 36.0,\) and 50.0, respectively. The inverse normalized \(T_c\) shift is plotted against the ratio \(d_1/d_2\). Solid lines are fits to Eq. (13) in the text for values of the parameter \(p = 0.75, 0.9, 1.7,\) and 2.85, respectively. Error bars for D1 are due to systematic uncertainty in the fabrication feature size, typically ±0.2 \(\mu\m).](image)

Data for all samples studied fit surprisingly well to the simple expression (13), as evident in Fig. 10. The parameter \(p\) ranges from 0.75 to 2.85 for the four data sets plotted. For larger \(\Delta T_c, p\) is shifted higher, but always of order unity. Deviation from the fit becomes pronounced for larger \(d_1/d_2\) (not shown), corresponding to the eventual breakdown of the simple law at very long modulation length scales. In Figs. 7 and 8, the solid lines fitted to the data are, in fact, Eq. (13). However, since \(\Delta T_c/(T_c - T_{c1})\) is plotted against \(d_1/d_2\) in these graphs, the linear dependence of \(\Delta T_c/(T_c - T_{c1})\) on \(d_1/d_2\) is less apparent.

C. The Cooper–de Gennes limit

When \(d_1, d_2 < \xi_1, \xi_2\), the effective electron-electron interaction strength of a nonuniform system is a weighted average of the contribution of its constituents. In this limit, called the Cooper–de Gennes limit, \(^3\)

\[
NV = \frac{N_{1}^2 V_1 d_1 + N_{2}^2 V_2 d_2}{N_1 d_1 + N_2 d_2}.
\]

Here, \(N\) is the density of states at the Fermi surface, \(V\) is the pair attraction potential, and the subscripts “1” and “2” again refer to the etched and the unetched sections, respectively. With \(NV\) specified, \(T_c\) is given by the usual BCS formula:

\[
k_B T_c = 1.14 \hbar \omega_D \exp \left( -\frac{1}{NV} \right).
\]

Since \(T_c, T_{c1}\), and \(T_{c2}\) differ by only a few percent, we can write

\[
T_{c2} - T_{c1} = (N_2 V_2 - N_1 V_1) \frac{dT_{c1}}{d(N_1 V_1)}
\]

\[
T_c - T_{c1} = (NV - N_1 V_1) \frac{dT_{c1}}{d(N_1 V_1)}.
\]

Combining with Eq. (14), an expression similar to the GL
result, Eq. (13), is readily obtained:

$$\frac{\Delta T_c}{T_c - T_{c1}} = \frac{N_2 V_2 - N_1 V_1}{N V - N_1 V_1} = 1 + \frac{N_1 d_1}{N_2 d_2}.$$  (18)

The ratio $N_1/N_2$ should be very close to unity in our system. Comparing with Eq. (13), we see that $p$ should, indeed, be of order unity. Note that no explicit account of boundary effects is included, which is equivalent to taking $p = 1$ in the GL analysis. The Cooper-de Gennes limit does not assume any inherent periodicity of the superlattice. Only the relative sizes of $d_1$ and $d_2$ matter. This is consistent with our observation that quasiperiodic (Fibonacci) modulation of the films gives rise to $T_c$'s which are compatible with periodic modulation.

Provided that the assumption $d_1, d_2 \ll \xi_1, \xi_2$ is correct, these results suggest that we have acquired an understanding of the data in the single-transition limit ($d_1 < 50 \mu m$). Yet, based on the estimates of Sec. IV A, this assumption cannot be true for our system. We believe the good fit may be fortuitous. To explore this, we now compare our data with the results of a microscopic calculation involving no adjustable parameters.

D. Self-consistent field technique

General treatments of multicomponent superconducting systems were given by de Gennes and, separately, by Takahashi and Tachiki using the self-consistent-field (SCF) method of Bogoliubov. These techniques are applicable to general nonuniform systems. The relevant equations include the Bogoliubov equations and the self-consistency equations of the single-electron potential and the pair potential, both of which may vary in space. The superlattice in question is characterized by the periodic variation of the potentials in one direction and the appropriate boundary conditions at interfaces separating regions of different $T_c$'s. Agreement between experiments and these calculations has been previously demonstrated. In particular, upper-critical-field measurements showed good agreements with the predictions of Takahashi and Tachiki.

For a two-component superlattice composed of similar materials, Jin and Ketterson showed that these complex equations reduce to a particularly simple form. This result assumes that $D$, the density of states, and the Debye frequency $\omega_D$ are constant, while the pair attraction potential $V$ is the only varying microscopic parameter in the different regions of the system. This special case is remarkably similar to our modulated-$T_c$ structures, and the result can be used directly without modification. The following paragraph states the relevant result in a form suitable for numerically evaluating the $T_c$ of our modulated structures. Details of the derivation can be found in the review article by Jin and Ketterson.

The transition temperature is identified as the highest temperature $T$ for which a solution exists for the secular equation

$$\delta_{mn} = \frac{2 \pi}{\hbar} k_B T \sum_\omega \left( \frac{m |NV| n}{2|\omega| + \Omega_m} \right) = 0,$$

(19)

where the outer vertical bars denote a matrix determinant. In our special case, the eigenvalues $\Omega_m$ are

$$\Omega_m = D \left( \frac{2 \pi m}{d_1 + d_2} \right)^2.$$

(20)

It can be shown that the summation over $\omega$ in (19) is given by

$$\delta_{mn} = \frac{2 \pi}{\hbar} k_B T \sum_\omega \left( \frac{m |NV| n}{2|\omega| + \Omega_m} \right) = \langle m |NV| n \rangle \left( \ln \frac{1.14 \hbar \omega_D}{k_B T} \right) - \chi \left[ \frac{\hbar \Omega_m}{2 \pi k_B T} \right].$$

(21)

In (21), the function $\chi$ is defined as

$$\chi(x) = \Psi \left( \frac{x}{2} + \frac{1}{2} \right) - \Psi \left( \frac{1}{2} \right),$$

(22)

where $\Psi$ is the digamma function. In (19) and (21), the matrix elements are given by

$$\langle m |NV| n \rangle = \begin{cases} \frac{N}{\pi} (V_1 - V_2) \left( \frac{1}{m- n} \sin \left( \frac{\pi (m - n) d_1}{d_1 + d_2} \right) + \frac{1}{m+n} \sin \left( \frac{\pi (m + n) d_1}{d_1 + d_2} \right) \right), & m \neq n \\ \frac{N}{d_1 + d_2} (V_1 d_1 + V_2 d_2) + \frac{N}{2 \pi n} (V_1 - V_2) \sin \left( \frac{2 \pi n d_1}{d_1 + d_2} \right), & m = n \end{cases}.$$ 

(23)

Using the experimental values of $T_{c1}$ and $T_{c2}$, the BCS equation (15) can be used to evaluate $NV_1$ and $NV_2$. With these values, $\langle m |NV| n \rangle$ can be calculated using (21) for $m, n = 1-10$. (Larger matrices do not give substantially different results.) Using (20) and (21), the $m \times n$ entries of the matrix in Eq. (19) can then be evaluated. To obtain the $T_c$ of the modulated structure, values of $T$ satisfying (19) are sought starting at $T_{c2}$, typically in decreasing order. The maximum $T$ satisfying (19) is identified as the $T_c$ of the system. Note that this pro-
procedure calls for no adjustable parameters aside from the experimental values of $T_{c1}$ and $T_{c2}$.

Figure 11 shows the results of these calculations. In Fig. 11(a), we plot $\Delta T_c/(T_{c1} - T_c)$ vs $d_1/d_2$ for a sample with $d_2 = 2 \mu m$. As was shown in Fig. 10, Eq. (13) fits the data well with $p = 0.9$. For comparison, the result of the Cooper–de Gennes limit ($p = 1.0$) is also shown (dashed line), and the result of the SCF calculation is illustrated by the lower solid line. Note that solution to (19) exists only for $d_1/d_2 < 2$, corresponding to a maximum permissible $d_1$ of $\sim 4 \mu m$. When $d_2$ is fixed at a smaller value, the range of $d_1/d_2$ over which solutions exists for (19) increases. However, the maximum $d_1$ for which such solution exists remains at $\sim 4 \mu m$. This is seen in Figs. 11(b) and 11(c), where $d_2 = 1$ and 0.5 $\mu m$, respectively. We interpret this as the maximum values of $d_1$ for which a single homogeneous transition should be observed in the modulated structures, consistent with the earlier theoretical considerations of dGW. Finally, in the limit that the modulation length scale is small ($d_2 = 0.1 \mu m$), the predictions of all three models approach each other, as seen in Fig. 11(d). This is expected since all three theories should be applicable in the limit that the modulation length scales are small in comparison with the coherence length. Since this length is $\sim 1 \mu m$ according to the estimates of Sec. IV A, the different theories should yield the same results when $d_1$ and $d_2$ are simultaneously much less than $1 \mu m$.

V. DISCUSSION

Our modulated-$T_c$ structures exhibit an anomalously long-range proximity effect which is inconsistent with the accepted microscopic description of de Gennes and Werthamer. This long length scale persists even at very high current densities, suggesting that it is not the result of a vanishingly weak order parameter in the $N$ region due to pair tunneling.

In the single-transition limit, the $T_c$ of the structures depends monotonically on the ratio $d_1/d_2$, and rather insensitively on the absolute magnitudes of $d_1$ and $d_2$. These observations are consistent with the existence of a long-range proximity effect. We have also demonstrated that the modulation direction, as well as the periodic nature of the modulation, are unimportant, indicating that our interpretation is not complicated by artifacts. These results can be fitted to a phenomenological Ginzburg-Landau model and the result in the Cooper–de Gennes limit, which are applicable, provided that the modulation length scales $d_1$ and $d_2$ are much smaller than the coherence lengths in the two regions. Such an assumption is, however, incompatible with the estimated coherence length of our dirty aluminum film. Consistent with this statement is the failure of a microscopic SCF theory to fit the experimental results. Indeed, failure to fit data is an understatement since no solution exists at all within this theory for modulation lengths >4 $\mu m$. Therefore, according to this theory, the system should not exhibit a single transition when the $N$ region exceeds this length.

We have suggested earlier that the single exponential decay description of the order parameter may simply be inadequate. However, there is no obvious reason why our system should be outside the regime of validity for the dGW theory. We believe the failure of theories is linked to small part to the similar $T_c$'s of the $S$ and $N$ regions. (This is, after all, a previously unexplored regime.) Near $T_c$, the phase coherence of the quasiparticles can be maintained over long distances in aluminum. For our films, we have determined, from quantum transport measurements, that the dephasing length close to $T_c$ is of the order of a few $\mu m$. It is possible that pairs enter a $N$ region, break up, but traverse the $N$ region without complete loss of phase memory. Once these quasiparticles reenter a $S$ region, they readily recombine to form pairs. As such, the mechanism responsible for this long-range proximity effect may not be pair tunneling, but is, instead, connected with superconducting fluctuations. Exactly how the quasiparticle coherence enhances the order parameter of the overall system awaits a detailed theoretical analysis.

In future experiments, it may be useful to systematical-
ly increase $\Delta T_c$ in a controlled fashion. The observation of a changeover from our long-range proximity effect regime to the conventional tunneling regime may be the key in understanding the present system. A large shift of the $T_c$ cannot be easily achieved using the present etching technique. Alternative approaches might include decreasing the thickness of the aluminum films, using magnetic overlayers, or applying stronger etches to damage or remove the aluminum. In addition, a different material besides aluminum should be studied. However, significant change in the transport properties of the film as a result of a more invasive processing steps is to be expected. This will substantially increase the complexity of comparing data with theories. Recall that great simplifications in the three theories examined are possible precisely because $T_{c1} \sim T_{c2}$ and parameters such as $D$, $N$, $m$, $\alpha$, and $\omega_D$ are roughly the same throughout the system.

In conclusion, we have carried out a survey of the properties of a 2D analog of a modulated transition temperature superlattice. The results are inconsistent with accepted theoretical models. It is possible that quasiparticle phase coherence plays an important role in understanding these results.

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FIG. 1. Schematics of modulated-\(T_c\) structures, illustrating etched and unetched regions whose transition temperatures are \(T_{c1}\) and \(T_{c2}\), respectively. We refer to the modulation in (a) as perpendicular while that in (b) as parallel.