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Evidence for the Kosterlitz-Thouless Transition in Thin Superconducting Aluminum Films

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The complex impedance of an aluminum film with normal-state sheet resistance of 4130 Ω/sq has been measured as a function of temperature and frequency. The experiment provides convincing evidence that there is a vortex-antivortex dissociation transition of the type predicted by the theory of Kosterlitz and Thouless for topological long-range order in two dimensions.

It has been recently suggested that the Kosterlitz-Thouless theory of topological long-range order in two dimensions[1,2] could offer an explanation for broad transitions in the resistance of films with high normal-state sheet resistances ($R_{\square n}$).[3-6] A key assumption of the theory is the existence of thermally excited vortices which are bound pairs of opposite circulation below the phase transition temperature T_{KT} and dissociated above. The maximum separation of the bound pairs is the correlation length

$$\xi_+(T) = A\xi_{GL}(T_{KT})\exp\{[B(T/T_{KT}-1)]^{-1/2}\}, \quad (1)$$

which diverges exponentially as T_{KT} is approached from above.[2,6] Here ξ_{GL} is the Ginzburg-Landau coherence length and A and B are constants of order unity. The dynamical properties of the transition, worked out by Ambegaokar and coworkers,[7,8] explain the broadening of the superfluid transition in ^{4}He films observed by Bishop and Reppy.[9] The predicted frequency-dependent broadening should apply to thin-film superconductors as well.[3-6]

We have studied several 100-Å-thick aluminum films, prepared by evaporating aluminum onto glass substrates in an oxygen atmosphere. The films were 3-mm scribed squares, with $R_{\square n}$ ranging from 43 to 4130 Ω/sq. Complex impedances were measured at frequencies between 1 kHz and 100 MHz with the arrangement shown in the inset of Fig. 1. The film lies in a plane transverse to the axis of two displaced coaxial coils so that the mutual-coupling signal between the coils is perturbed by the induced sheet currents in the film. To increase sensitivity, we modulate the film between its superconducting and normal states with a current applied through contacts at the corners of the film. The change in the mutual-coupling signal gives the change in film-sheet admittance, essentially the admittance of the superconducting state for high-$R_{\square n}$ films.

In Fig. 1 we show the real and imaginary parts of the sheet admittance Y of a 4130-Ω/sq film as functions of temperature. For reference we also

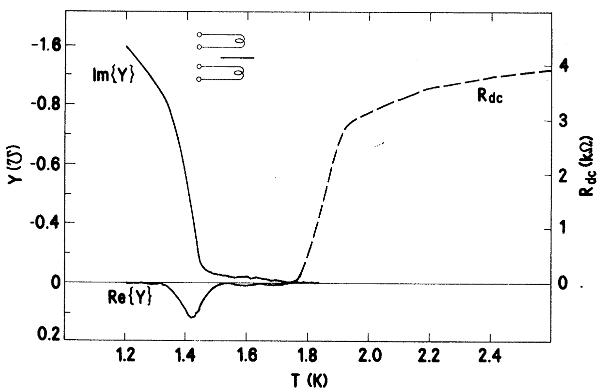

FIG. 1. Real and imaginary parts of the rf admittance Y at 17.5 MHz and dc resistance of a 4130-Ω/sq film. Inset: rf measurement configuration.

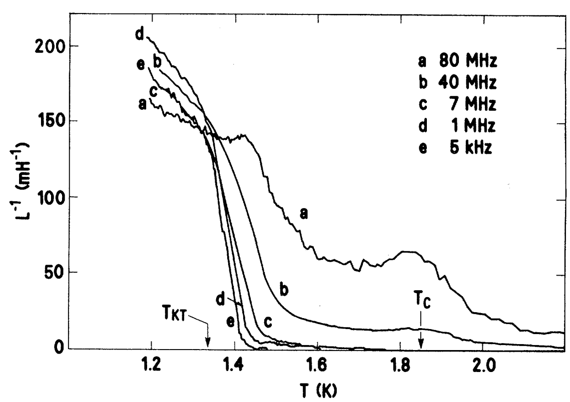

FIG. 2. Temperature dependence of the reciprocal film inductance at the frequencies indicated. The Kosterlitz-Thouless transition temperature T_{KT} and mean-field temperature T_c have been marked.

show the dc sheet resistance, measured at 1 nA and 3 Hz excitation, using van der Pauw's method.[10] The dc external magnetic field was less than 1 mG and the rf magnetic field was about 0.2 mG at the center of the film. Results independent of rf amplitude were obtained at this order of magnitude of excitation. By taking the inverse of the complex admittance, we arrive at the complex impedance, which we write as

$$Y^{-1}=R+i\omega L, \quad (2)$$

where R is the sheet resistance and L the sheet inductance. We plot the temperature dependence of L^{-1} and R in Figs. 2 and 3, respectively.

The resistance falls to zero at low temperature where the film appears to be a superconductor with purely inductive response. In this region L equals the kinetic inductance L_K, given in terms of the superelectron areal density n_s as

$$L_K=m_e/n_s e^2=2\pi\Lambda/c^2, \quad (3)$$

where Λ is the magnetic screening length in thin films that scales with $R_{\square n}$ and follows the Ginzburg-Landau $(T_c-T)^{-1}$ temperature dependence outside the critical region.[6] Except for the 80-MHz data, the linearly fitted temperature dependence of L^{-1} at low temperatures extrapolates to zero reproducibly at an effective mean-field temperature $T_c=1.85\pm0.02$ K, an average of measurements at ten frequencies. However, we find that $\Lambda(T)$ is 65% larger than the theoretical prediction for high-resistivity materials,[3-6] taking for $R_{\square n}$ the 4.2-K resistance.

Qualitatively, the data in Fig. 2 show that the transition broadens with increasing frequency. There is a change in slope of L^{-1} near 1.34 K, which we will show can be identified with T_{KT}. Another break in slope occurs at a higher temperature T_ω which increases with increasing frequency. The dissipation, as measured by R in Fig. 3, is markedly frequency and temperature dependent at low temperatures.

These results can be understood in terms of the motion of thermally excited vortices. The dissipation associated with viscous motion of vortices in superconductors has been treated by Bardeen and Stephen with the result that the vortex diffusivity D is given by[11]

$$D/kT=2\pi\xi_{GL}{}^2c^2R_{\square n}/\varphi_0{}^2, \quad (4)$$

where φ_0 is the flux quantum. We have measured D at 1.21 K by applying an external magnetic field. It is a complex quantity, because of pin-

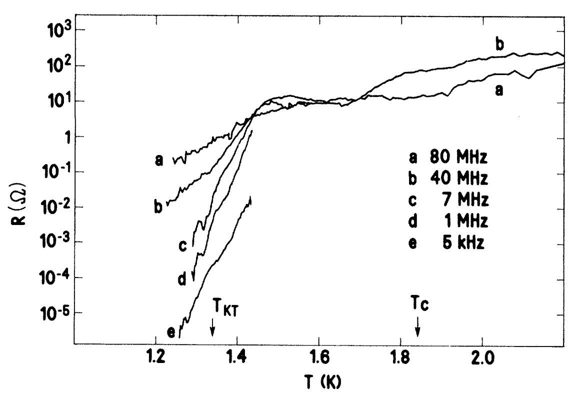

FIG. 3. Semilog plot of the temperature dependence of the rf resistance at the frequencies indicated.

ning forces, with magnitude weakly dependent on frequency and within a factor of 2 of the Bardeen and Stephen result. Previous studies[12-15] have shown that there are also nonviscous forces on vortices, such as random pinning forces, which give a contribution to L^{-1}. In the present case we expect that the forces of interaction between members of bound vortex pairs must also be included.

The contribution from the bound pairs has been treated in the dynamical theory of Ambegaokar and co-workers,[7,8] who show that the response is dominated by the reorientation (or polarization) of vortices separated by a characteristic distance set by the frequency:

$$r_\omega \approx (14D/\omega)^{1/2}. \tag{5}$$

For temperatures below T_{KT} the vortex-pair polarization picture is valid at all frequencies. However, above T_{KT} there is a crossover in behavior when

$$r_\omega = \xi_+(T_\omega). \tag{6}$$

Equation (6) implicitly defines a temperature T_ω above which the bound-vortex picture breaks down. For $T > T_\omega$ the dominant contribution is from free vortices.

Although the behavior near T_ω has not been treated accurately in the dynamical theory,[7,8] we adopt an *ad hoc* procedure for obtaining T_ω by extrapolating to zero the steep portions of the curves of L^{-1} vs T. In Fig. 4 we have plotted the quantity l_ω^{-2} against T_ω as a check of Eq. (1), where

$$l_\omega^{-2} \equiv \ln^{-2}(r_\omega/\xi_{GL}) = \ln^{-2}[(14D/\omega\xi_{GL}^2)^{1/2}] \cong B(T_\omega/T_{KT} - 1). \tag{7}$$

We have taken $A = 1$, for simplicity, since the fit is insensitive to A. The fact that there is a good fit confirms the exponential inverse square-root divergence of $\xi_+(T)$ as $T \to T_{KT}$ and gives $T_{KT} = 1.34 \pm 0.01$ K with $B = 0.24 \pm 0.02$. Further inspection of the data in Fig. 4 shows that the dissociation temperatures for frequencies below 300 kHz all lie near 1.42 K. Theoretically, the divergence of $\xi_+(T)$ cuts off at a temperature where $\xi_+(T) \sim w_{\text{film}} = 3$ mm [or at $\xi_+(T) \sim \Lambda$ if $\Lambda < w_{\text{film}}$), which corresponds to $T = 1.37$ K in the present experiment. We therefore believe that the broadening of the transition is caused by film inhomogeneity, rather than by the finite size of the film.[6]

The theory makes a fundamental prediction that

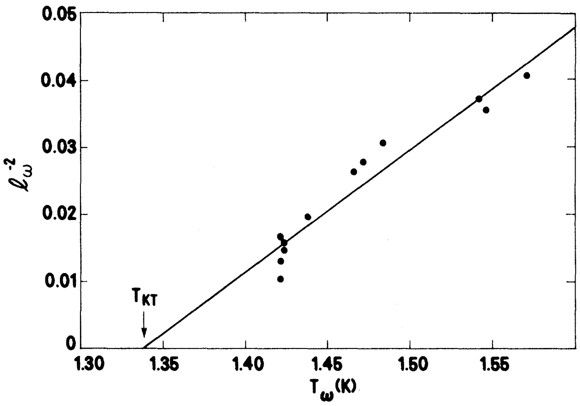

FIG. 4. Dependence of l_ω^{-2}, defined by Eq. (7), on the dissociation temperatures T_ω, with frequency of measurement increasing from left to right. The line shows the fit according to Eq. (7).

$\Lambda(T_{KT})T_{KT} = k_B^{-1}(\varphi_0/4\pi)^2 = 1.96$ cm K is a constant reflecting the universal jump in the superfluid density at the Kosterlitz-Thouless transition.[16] We find that to within experimental error $L = 7.1 \pm 0.5$ nH is frequency independent at T_{KT}, so that $\Lambda T_{KT} = 1.5 \pm 0.1$ cm K. Analysis of preliminary data on other films gives values in this same range. The theoretical value is consistent with these results, since systematic errors in the determination of the geometrical parameters of the experiment could account for the difference.

The theory predicts that dc resistance is proportional to the number of free vortices and is given by $R_{dc} \approx R_{\square n}(\xi_{GL}/\xi_+)^2$. The theory is therefore consistent with observation of negligible dc resistance for temperatures below 1.7 K, where we take values for T_{KT} and B from the rf data. The rf resistance data shown in Fig. 3 are also in qualitative accord with the theoretical prediction that R increases rapidly with frequency and temperature near T_{KT}. In the temperature region satisfying $r_\omega < \xi_+(T)$, rf resistance arises solely from bound vortex pairs. We expect that

$$R \propto \exp\left[-\frac{\varphi_0^2[\ln(r_\omega/\xi_{GL}) + \frac{1}{4}]}{4\pi^2\epsilon(r_\omega)\Lambda(T)}\right], \tag{8}$$

where the argument corresponds to the energy of a vortex pair of separation r_ω.[5-8] This expression includes the dielectric constant, $\epsilon(r_\omega)$, which describes screening of the vortex-pair interactions. We find that the temperature dependence predicted by Eq. (8) agrees with the data, except at the lowest and highest frequencies where $d\ln R/dT^{-1}$ is diminished by about a factor of 2. Deviations found at low frequencies are probably caused by film inhomogeneity and those at high frequency by pinning forces. The resistance at T_{KT} varies with frequency as ω^{α}, with $\alpha \approx 0.9$, and is smaller than the prediction of the dynamical theory[7]: $R = 2\pi^3\omega L_K l_\omega^{-2}$. This discrepancy between the theory and experiment is perhaps due to pinning effects, which were not taken into account in the theory. Apart from the deviations noted above, our results generally confirm the fundamental physical ideas of Kosterlitz and Thouless.

The authors thank D. S. Fisher for stimulating discussions, and R. P. Minnich and A. L. Bullock for technical assistance.

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New High-Field Phase in the Spin-Peierls System, Tetrathiafulvalene-$CuS_4C_4(CF_3)_4$

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New magnetization data on the spin-Peierls system tetrathiafulvalene-$CuS_4C_4(CF_3)_4$ for $h \leq 155$ kOe and $T \geq 1.5$ K provide evidence for a new phase above $h \sim 115$ kOe. Considerable hysteresis in the field-induced transitions is observed for $T \leq 5.5$ K. These results are discussed in the light of current theoretical ideas.

We report the first observation of a new phase in the spin-Peierls (SP) system tetrathiafulvalene-*bis-cis*-(1,2-perfluoromethylethylene-1,2-dithiolato)-copper [(TTF)-$CuS_4C_4(CF_3)_4$ or further abbreviated as TTF-BDT(Cu)] at high magnetic fields and low temperatures. The SP transition is a progressive spin-lattice dimerization in an assembly of quasi one-dimensional (1D) Heisenberg antiferromagnetic (AFM) chains coupled to a 3D phonon field.[1] For the class of insulating compounds (TTF)-$MS_4C_4(CF_3)_4$ [denoted as TTF-BDT(M)] the zero-field transition occurs at about 11.5–12 K for M= Cu and at 2.1 K for M = Au. More recently SP behavior has been established in (TTF)-$CuSe_4C_4(CF_3)_4$ [T_c = 6 K] (Ref. 2) and methylethylmorpholinium-(tetracyanoquinodimethanide)$_2$ [(MEM)$(TCNQ)_2$] [T_c = 18 K],[3] and perhaps in the alkali TCNQ's.[4] In the TTF-BDT(M)

