Ni are taken from Figs. 2 and 3. Similar $E(\vec{k})$ data has been obtained for Cu (Ref. 15) and is given for comparison. Theoretical values in Fig. 4 are those of Moruzzi, Williams, and Janak, who have performed *ab initio* self-consistent Kohn-Korringa-Rostoker calculations using the local-density theory of electronic exchange and correlation. In summary, Ni and Co (to a lesser extent) show significant deviations from state-of-the-art ground-state calculations.[1-4] Using the experimental exchange splittings δE_{ex} in Fig. 4 and Bohr-magneton data ($\mu_s = 2.1\mu_B$, and $0.55\mu_B$ for Fe, Co, and Ni, respectively), we observe that a phenomenological measure[19] of the intra-atomic exchange interaction U^{d-d}, namely $U^{d-d} \simeq \delta E_{ex}/\mu_s$, is approximately constant ($U^{d-d} \simeq 0.6$–0.7) for Fe, Co, and Ni. This value of U^{d-d} is smaller ($\lesssim \frac{1}{2}$) than a previous estimate based on electron-energy-loss and soft-x-ray spectra.[19]

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[1]V. L. Moruzzi, J. F. Janak, and A. R. Williams, *Calculated Electronic Properties of Metals* (Pergamon, New York, 1978).

[2]J. Callaway and C. S. Wang, Phys. Rev. B 16, 2095 (1977).

[3]Francisco Batallan, Izio Rosenman, and C. B. Sommers, Phys. Rev. B 11, 545 (1975).

[4]C. S. Wang and J. Callaway, Phys. Rev. B 16, 298 (1977).

[5]H. Capellmann, J. Phys. F 4, 1466 (1974), and to be published.

[6]V. Korenman, J. L. Murray, and R. E. Prange, Phys. Rev. B 16, 4032, 4048, 4058 (1977).

[7]D. E. Eastman, J. Phys. (Paris), Colloq. 32, C1-293 (1971).

[8]H. Hochst, S. Hufner, and A. Goldman, Phys. Lett. 57A, 265 (1976).

[9]P. Heimann, E. Marschall, H. Neddermeyer, M. Pessa, and H. F. Roloff, Phys. Rev. B 16, 2575 (1977).

[10]S. D. Kevan, P. S. Wehner, and D. A. Shirley, Solid State Commun. 28, 517 (1978).

[11]P. Heiman and H. Neddermeyer, Phys. Rev. B 18, 3537 (1978).

[12]A. Schulz, R. Courths, H. Schulz, and S. Hufner, J. Phys. F 9, L41 (1979).

[13]D. E. Eastman, F. J. Himpsel, and J. A. Knapp, Phys. Rev. Lett. 40, 1514 (1978).

[14]F. J. Himpsel, J. A. Knapp, and D. E. Eastman, Phys. Rev. B 19, 2919 (1979).

[15]J. A. Knapp, F. J. Himpsel, and D. E. Eastman, Phys. Rev. B 19, 4952 (1979).

[16]$E(k_\perp)$ dispersions for the relevant final-state bands for Fe(111) and Co(0001) were determined in a manner similar to that previously described (Refs. 13 and 14).

[17]J. Hermanson, Solid State Commun. 22, 9 (1977).

[18]Previous normal-emission data (Ref. 11) for Fe(111) and Fe(100) at $h\nu = 11.8$, 16.85, and 21.22 eV are consistent with ours (to be published), while other data (Ref. 12) exhibit somewhat lower resolution.

[19]M. B. Sterns and S. Shinozuki, Physica (Utrecht) 86-88B, 1195 (1977).

Thermodynamic and Resistive Transitions of Thin Superconducting Films

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In amorphous, nongranular, thin films of Nb_3Ge, the temperature dependence of the resistance and the current-voltage relation have been investigated at temperatures below the experimentally determined thermodynamic transition. This temperature-dependent resistance is compared to several theories which predict flux-flow resistance below the BCS transition temperature due to thermally excited vortex pairs.

In this Letter we report measurements of the temperature dependence of the electrical resistance and heat capacity of amorphous, nongranular, Nb_3Ge films deposited on thin polished-sapphire substrates. The heat capacities were measured with an ac technique.[1] We have found the samples to be resistive below the thermodynamic transition temperature T_{BCS}. A precursor to the heat-capacity jump at T_{BCS} is always observed and in some samples a bump is found on top of the BCS jump. Measurements of the magnetic field dependence of the resistance below T_{BCS} indicate that the transition broadens with field in a manner similar to that reported for granular Al and amorphous Bi films.[2,3]

The Nb_3Ge films, of thickness d in the range from 1000–1500 Å, are not two dimensional (2-D) except very close to T_{BCS}, in that $d > \xi(T)$. Here $\xi(T)$ is the temperature-dependent coherence length. On the other hand, $d \ll \lambda_T = \lambda^2(T)/d$ at all temperatures, where $\lambda(T)$ is the dirty-limit London parameter and λ_T is the transverse penetration depth. When λ_T is nearly macroscopic, which is possible in films with a high sheet resistance R_N, thermally excited vortex pairs of opposite circulation are generally believed to interact through a logarithmic force law,[4] and there is a possibility of a 2-D phase transition characterized by a condensation of these vortex excitations at a finite temperature $T_{2\text{-}D}$ which is less than T_{BCS}.[5-8] Since free vortices would be swept across a film by a transport current, a finite resistance is expected in the temperature range $T_{2\text{-}D} < T < T_{BCS}$.[5-8] The condensation of such "topological excitations" has been discussed by Berezinskii[9] and by Kosterlitz and Thouless[10] and has been the subject of extensive study in superfluid helium films.[11]

Heat capacities were measured with calorimeters suspended by their electrical leads from a thermal platform which could be cooled to 1 K. Basically the calorimeters consisted of polished 0.025–0.50-mm-thick single-crystal sapphire substrates approximately $1.2 \times 2.5\ cm^2$ in area. The films were dc sputtered onto these substrates. The heat-capacity measurements were able to resolve changes in the total heat capacity of 1–5 $\times 10^{-4}$ allowing the BCS temperature to be determined even though the Nb_3Ge films constituted only 5–8% of the total heat capacity. In this determination, for $T > T_{BCS}$ and away from the transition, the total measured heat capacity accurately fit a sum of terms only linear and cubic in the temperature. Since amorphous Nb_3Ge is a weak-coupled superconductor,[12] the data in the superconducting state, away from the transition and in the range between 1 and 2 K, were fitted to a model which included a BCS contribution, a cubic term, and a linear addendum contribution. In this fit, the only adjustable parameters were the transition temperature T_{BCS} and γ, the coefficient of the specific heat of the film in the normal state. The other parameters which were required, i.e., the total γ (film + substrate) and the coefficient of the cubic term, were provided independently from the results of the fit to $T > T_c$ data. The cryostat was suspended inside a double Mumetal shield which reduced the ambient magnetic field to less than 1 mOe.

In Fig. 1 we show the temperature dependence of the total measured heat capacity to display the quality of the data and the fit which is the solid line. In Figs. 2(a) and 2(b) we show, for two films, the temperature dependences of C_{ex}, the heat capacity in excess of the normal-state value for $T > T_{BCS}$, the BCS value for $T < T_{BCS}$, and $R(T)$. The heat capacity between 1 and 2 K, the range over which the fitting was carried out, is consistent with the observed jump at T_{BCS}, even when the resistive transition is broadened. The observation of a full BCS jump suggests that the suppression of the resistive transition is not the result of sample inhomogeneity.

If the BCS theory is fitted over the entire range of temperatures below T_{BCS} rather than between 1 and 2 K, a poorer fit results. However, the values of T_{BCS} selected by the program are only changed by about 0.010 K from the previous fit. The extra entropy associated with the peaks of Figs. 2(a) and 2(b) should be accompanied by a fall in the heat capacity below the BCS value in the range below T_{BCS} to ensure the thermodynam-

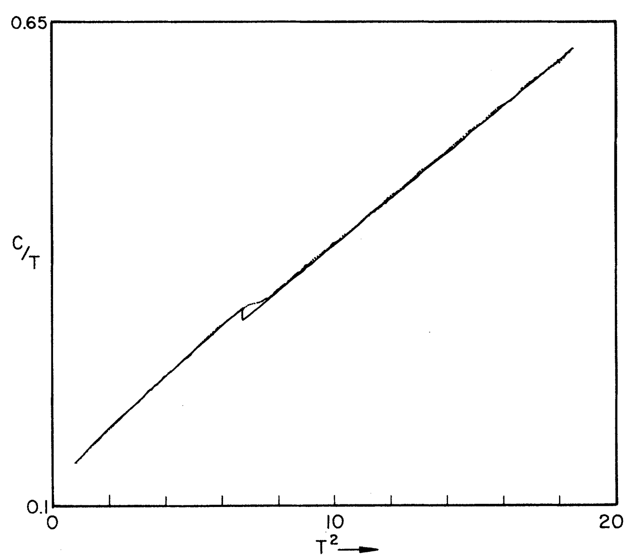

FIG. 1. Total C/T (film + substrate) vs T^2 for sample Nb_3Ge-6 from 1 to 4 K. The solid lines are the fits.

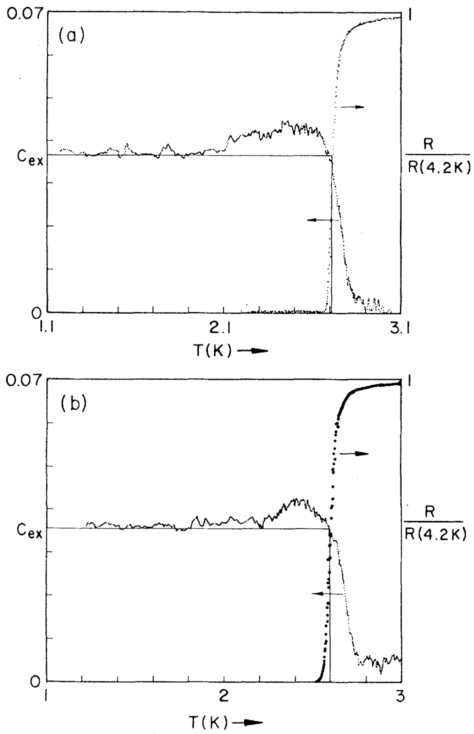

FIG. 2. C_{ex} vs T and R vs T for samples (a) Nb_3Ge-4 ($\rho = 950 \pm 150$ $\mu\Omega$ cm, $R_N = 65 \pm 10$ Ω/sq) and (b) Nb_3Ge-6 ($\rho = 800 \pm 150$ $\mu\Omega$ cm, $R_N = 63 \pm 10$ Ω/sq).

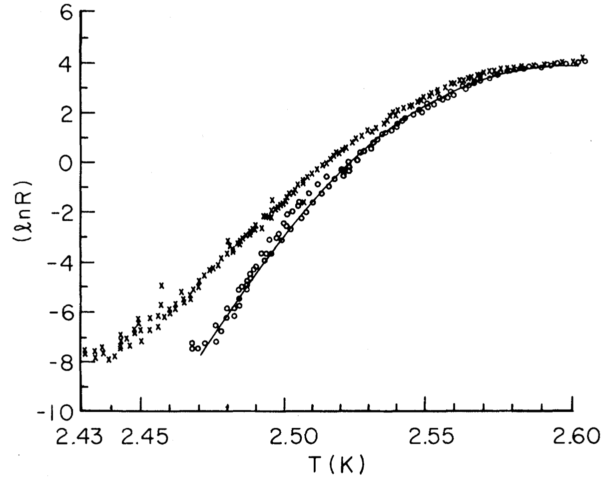

FIG. 3. Natural logarithm of the resistance vs T for the 1000-Å-thick film. The open circles are data taken in a magnetic field <0.001 G and the crosses are data in a perpendicular field of 20 G. The solid line is a fit to Turkevich's theory with modifications as outlined in the text with $\lambda(0) = 30\,000$ Å. With this value of λ, one finds $T_{2\text{-D}} = 2.54$ K. With the normal-state sheet resistance in the manner outlined in Ref. 5, $T_{2\text{-D}} = 2.59$ K.

ic consistency. As this entropy is the order of 0.1% of the total entropy at T_{BCS}, it is not surprising that the correction is lost in the scatter.

For each film, the excess heat capacity above T_{BCS} varies as $1/(T - T_{BCS})^{1/2}$, a result consistent with film thicknesses being 800–1300 Å and with the small $\xi(0)$ one expects in amorphous films. The excess conductivity above T_{BCS} also varies as $1/(T - T_{BCS})^{1/2}$, but given the complexity of the predictions for excess conductivity, extensive fitting efforts were not carried out.

In Fig. 3 we show the natural logarithm of the resistance ($\ln R$) as a function of temperature for temperatures below T_{BCS}. These data were accumulated with a current density of 1 A/cm^2 over the entire range of resistances. Also in this figure is $\ln R$ vs T with an externally applied magnetic field of ≈ 20 G perpendicular to the plane of the film.

At the present there are four theoretical papers[5-8] which discuss the possibility of thermally excited vortex-antivortex pairs giving rise to flux-flow resistance below T_{BCS}. These theories, though different in detail, rely heavily on the pioneering work of Kosterlitz and Thouless in this area. Three of the papers[6-8] predict the temperature dependence of the flux-flow resistance $R(T)$ below T_{BCS}. We have attempted to fit the theories to our zero-magnetic-field experimental results without any success for any values of the adjustable parameters. On the other hand, with a single modification, Turkevich's[7] theory could be fitted quite well (see Fig. 3). In a manner consistent with the other theories he defines $T_{2\text{-D}}$ in terms of the temperature-dependent penetration depth $\lambda(T)$. However, rather than solving self-consistently for $T_{2\text{-D}}$, he implicitly assumes $T_{2\text{-D}} \ll T_{BCS}$ in obtaining his Eq. (12) for $R(T)$. Our data were fitted by restoring the temperature dependence of λ to Eq. (12) with the only adjustable parameters being $4\pi R_N$, where R_N is the normal resistance, and $\lambda(0)$. As shown in Fig. 3, this form fitted the data over 11 to 12 "e-cades" (5 decades) of resistance. The value of $\lambda(0)$ determined in this fit was 30 000 Å which, if one assumes a-Nb_3Ge to be a weak-coupled superconductor, gives a value of $\xi(0)$ of 25 Å. The latter is within 20% of that determined from $H_{c2}(T)$.

The temperature-dependent resistance was also measured in seven different magnetic fields applied perpendicular to the sample. The magnitude of the fields ranged from 50 mG to 21 G with the data obtained in the largest field being exhibited in Fig. 3. Even though the zero-field theory of Doniach and Huberman[6] could not explain our results, the effect of externally applied fields looks qualitatively like the inset in their Fig. 1.

At temperatures below T_{BCS} the I-V characteristics were found to be non-Ohmic. The data thus far discussed were obtained with a current density of 1 A/cm^2. When the current density was decreased to 0.01 A/cm^2 there were only small changes in the measured voltages when compared with the 1-A/cm^2 data. Thus we have used the 1-A/cm^2 current density to define $R(T)$ When the current density was increased to 10 A/cm^2, the effect was to shift the resistance versus temperature curve to a lower temperature (this temperature shift was approximately 25 mK). Such behavior might be expected if the number of free vortices were dependent on the magnitude of the transport current either from the current breaking vortex-antivortex pairs or depinning additional vortices.[13]

The order of the resistive and heat capacity transitions in the present work is the reverse of that reported for granular Al.[14] Although the 800-$\mu\Omega$-cm resistivities of our films are within the resistivity range studied in Ref. 14, our film thicknesses are close to 1000 Å rather than 10 μm. The values of $R_N^\square$ in Ref. 14 range from 0.6 Ω/sq to 5.6 Ω/sq, far below those studied here. For the Kosterlitz-Thouless transition to be observable ($T_{2\text{-D}}/T_{BCS}<1$), $R_N^\square$ and not ρ_N must be large.

The high resistivities (~700 to 1000 $\mu\Omega$ cm) of the present films are less than the maximum metallic resistivity for amorphous and liquid metals as suggested by Mott.[15] X-ray diffraction analysis of the 1000-Å-thick film showed one broad weak peak. Electron diffraction analysis of the broken edge of this film showed three diffuse peaks. These results are consistent with the film being either amorphous or microcrystalline. A broad area of the film was studied with use of a JEOLCO 100-CX electron microscope in the scanning mode with a resolution of 30 Å. The surface features are consistent with those observed in other amorphous films. Although these studies were carried out on only one film, they are believed to be representative of all of the films because all of the films had similar resistivities and were deposited under identical conditions.

We have shown that the data are broadly consistent with a model of flux-flow resistance involving dissociated vortex-antivortex pairs.[7] However, although there is no feature in $R(T)$ implying a divergence in the conductivity as discussed in Refs. 6 and 8 for high-sheet resistance films at the value of $T_{2\text{-D}}=2.54$ K derived from the fitting procedure to Turkevich's formula, the data are not inconsistent with a divergence in the conductivity at a finite temperature below 2.47 K which was the limit of the data. The data measured at finite magnetic field are qualitatively consistent with the theory of Ref. 6. The apparent absence of any special feature in $R(T)$ near $T_{2\text{-D}}$ may be the result of the external magnetic field (< 0.001 Oe in this case) being greater than H_{c1} in which case the vortex-antivortex pairs would remain dissociated until a much lower temperature than reached in the present work.[16] The possibility that the disappearance of resistance below T_{BCS} is due to a transition of the type reported in thin NbN films may be ruled out by the fact that our data could not be fitted with a power law.[17] It should be noted that the value of $T_{2\text{-D}}$ determined from R_N as outlined in Ref. 5 differs from that obtained from our fit. This might be due to the use in Ref. 5 of the nearly-free-electron model to relate λ to R_N.

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[1]Paul F. Sullivan and G. Seidel, Phys. Rev. 173, 679 (1968).

[2]W. E. Masker, S. Marcelja, and R. D. Parks, Phys. Rev. 188, 745 (1969).

[3]R. E. Glover, III, Physica (Utrecht) 55, 3 (1971).

[4]J. Pearl, in *Low Temperature Physics-LT9,* edited by J. G. Daunt, D. O. Edwards, F. J. Milford, and M. Yaqub (Plenum, New York, 1965), p. 566.

[5]M. Beasley, J. Mooij, and T. Orlando, Phys. Rev. Lett. 42, 1165 (1979).

[6]S. Doniach and B. Huberman, Phys. Rev. Lett. 42, 1169 (1979).

[7]L. A. Turkevich, J. Phys. C 12, L385 (1979).

[8]B. I. Halperin and David R. Nelson, J. Low Temp. Phys. 36, 599 (1979).

[9]V. L. Berezinskii, Pis'ma Zh. Eksp. Teor. Fiz. 61, 1144 (1972) [JETP Lett. 34, 610 (1972)].

[10]J. M. Kosterlitz and D. J. Thouless, J. Phys. C 6, 1181 (1973); J. M. Kosterlitz, J. Phys. C 7, 1046 (1974).

[11]I. Rudnick, Phys. Rev. Lett. 40, 1454 (1978); D. J.

Bishop and J. D. Reppy, Phys. Rev. Lett. 40, 1727 (1978); Emilia Webster, Grant Webster, and Marvin Chester, Phys. Rev. Lett. 42, 243 (1979).
[12]C. C. Tsuei, W. L. Johnson, R. B. Laibowitz, and J. M. Viggiano, Solid State Commun. 24, 615 (1977); C. C. Tseui, J. M. Coey, and S. von Molnar, Phys. Rev. Lett. 41, 664 (1978).
[13]P. M. Horn and R. D. Parks, Phys. Rev. B 4, 2178 (1971).
[14]T. Worthington, P. Lindenfeld, and G. Deutscher, Phys. Rev. Lett. 41, 316 (1978).
[15]N. F. Mott and E. A. Davis, *Electronic Processes In Noncrystalline Materials* (Clarendon, Oxford, 1979), 2nd ed., p. 31.
[16]B. A. Huberman and S. Doniach, Phys. Rev. Lett. 43, 950 (1979).
[17]S. A. Wolf, D. U. Gubser, and Y. Imry, Phys. Rev. Lett. 42, 324 (1979).

Specific Heat of Insulating Spin-Glasses, (Eu,Sr)S, near the Onset of Ferromagnetism

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The specific heat, $C(T)$, of the insulating spin-glass $Eu_xSr_{1-x}S$ with $x = 0.40$ exhibits a linear term γT and no singularity at T_f, like metallic spin-glasses. A new term, δT^{-2}, is found which may arise from totally decoupled clusters created by frustration. The field dependence, $\gamma(B)$ and $\delta(B)$, show maxima. With $x = 0.54$ the behavior is similar. $C(T,B)$ and $\chi(T,B)$ there indicate the spin-glass phase to be supressed at $B = 1$ T.

A characteristic property of metallic spin-glass systems is the (nearly) linear thermal variation of the magnetic specific heat, $C(T)$, at low T, together with the observation of a broad maximum of C above the freezing temperature T_f but a lack of a pronounced anomaly at T_f.[1] Recent numerical calculations by Walker and Walstedt[2] confirmed the linear dependence of $C(T)$.

Recently, the insulating compounds (Eu, Sr)S were found to show the same magnetic behavior[3] as is well established for metallic spin-glasses. The oscillating Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction mediated by the conduction electrons is now replaced in (Eu, Sr)S by the well-known competing nearest- and next-nearest-neighbor exchange interactions.[4] An interesting detail of its magnetic phase diagram has been discovered by neutron diffraction experiments,[5] showing upon cooling a spin-glass state to exist below the ferromagnetic phase within a concentration regime above the "multicritical" point at $x_c = 0.51$. In this Letter we present the first calorimetric studies of $Eu_xSr_{1-x}S$ single crystals with concentrations below ($x = 0.40$) and slightly above ($x = 0.54$) x_c. One advantage of this system is the lack of an electronic contribution to the specific heat. Since, in addition, below 10 K the lattice contributes less than 0.3%, the measured specific heat is nearly identical with the magnetic part.

The specific heat was determined between 0.3 and 10 K at magnetic fields $0 \leq B \leq 1.0$ T by a quasistatic method as described in detail elsewhere.[6] A pair of carbon resistors (Allen-Bradley, 10 Ω and 180 Ω) were used as sample thermometers, which covered the ranges 0.3–1.5 K and 1.2–10 K, respectively. They were calibrated against ^{3}He and ^{4}He vapor pressure and (for $T > 4.2$ K) against a precalibrated Ge resistor. The magnetoresistance of these thermometers never exceeded $\Delta R/R(B=0) \approx 2\%$ in the present experiments. This introduced a maximum error of the specific heat, which was confined to 0.5% and could therefore be neglected in the data analysis (compare Ref. 6). The results of the specific heat were compared to the low-frequency (117

