

HEAT CAPACITY OF α URANIUM AT A PRESSURE OF 10 kbar, BETWEEN 0.3 AND 6°K*

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We have measured the heat capacity of α uranium at a pressure of 10 kbar between 0.3 and 6°K both in zero magnetic field and at 2000 Oe. A heat-capacity anomaly typical of a bulk superconducting transition was observed at 2°K; the anomaly coincided with a superconducting transition observed magnetically on the same sample. Since zero-pressure heat-capacity measurements to temperatures as low as 0.1°K have consistently failed to detect a bulk superconducting transition,¹⁻³ these measurements constitute the first observation of bulk superconductivity in α uranium and show that 10 kbar increases T_c by a factor of at least 20—an even more striking effect than that suggested by magnetic measurements.⁴ It seems possible that α uranium may even be the first example of a metal that becomes superconducting only under pressure without undergoing a crystallographic transition. The application of 10-kbar pressure also increased the normal-state density of states by 18% and eliminated a low-temperature (<0.7°K) heat-capacity anomaly that was observed at zero pressure.

Superconducting transitions in α uranium at zero pressure have been observed⁵⁻⁸ either magnetically or resistively, with values of T_c ranging from 0.2°K to above 1°K, but there is no indication of an anomaly of the type associated with a superconducting transition in heat-capacity measurements on one sample¹ to 0.15°K or in measurements on two other samples² to 0.65°K. More recent heat-capacity measurements on two samples to 0.1°K and on two other samples to 0.3°K also failed to show evidence of bulk superconductivity, although three of the samples had been studied magnetically and in each case these measurements showed superconducting transitions within the range of the calorimetric measurements.³ Thus, the heat-capacity measurements show that at zero pressure, α uranium is not a bulk superconductor above 0.1°K, and that all previously observed transitions were the consequence of

multiply connected superconducting filaments. Since the superconductivity of α uranium is of particular interest in connection with the role of the $5f$ states, these measurements were undertaken to determine whether or not the transition observed magnetically at high pressure⁴ is a bulk transition, and to look for any associated change in the normal-state density of states.

The sample was contained in a small Be-Cu piston and cylinder arrangement in which a pressure applied in a press at room temperature could be retained when the assembly was cooled to low temperatures. The pressure at the low temperature was determined from the magnetically measured T_c by use of the known pressure dependence.⁴ Although the heat capacity of the cell was 20 to 40 times the normal-state heat capacity of the sample, reasonable accuracy was obtained by measuring the heat capacities of the filled and empty cell to a precision of a few tenths of 1%, using a germanium thermometer that retained its calibration between the two measurements.

Figure 1 shows the results of the high-pressure measurements for temperatures below 3°K, and, for comparison, a dashed curve which represents the zero-pressure heat capacity of a larger sample from which the high-pressure sample was cut. The crosses represent the total sample heat capacity; the triangles and the dashed curve have been corrected for the hyperfine heat capacity of the U^{235} by subtraction of a T^{-2} term that was derived from lower-temperature measurements at zero pressure.³ The zero-pressure heat capacity is independent of magnetic field to at least 5000 Oe, and the anomaly below 1°K was also observed in three other samples.³ Above 1°K the dashed curve is represented by $C = 10.3T + 0.323T^3$ mJ/mole deg, and, therefore, the coefficient of the electronic heat capacity is $\gamma = 10.3$ mJ/mole deg². At 10 kbar, the zero-field measurements show an anomaly typical of a superconducting

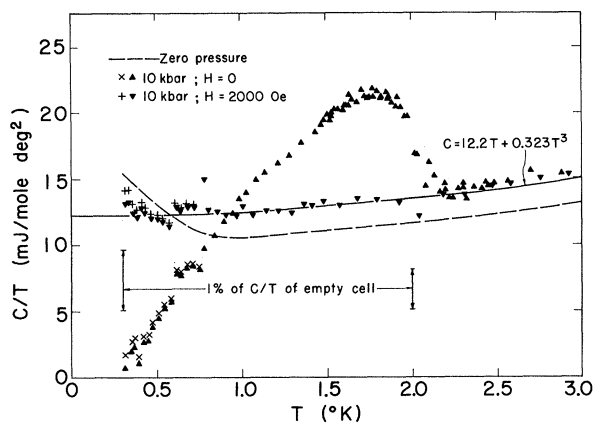


FIG. 1. The heat capacity of α uranium at zero pressure and at 10 kbar. Points represented by triangles and the dashed curve have been corrected for the hyperfine heat capacity by subtraction of a T^{-2} term. The crosses represent the total sample heat capacity. For $T > 1^\circ\text{K}$ the dashed curve corresponds to $C = 10.3T + 0.323T^3$.

transition at $T_C \approx 2^\circ\text{K}$, in good agreement with the magnetically determined T_C . The 2000-Oe 10-kbar measurements show no anomaly at any temperature in the range of measurement and give $\gamma = 12.2$ mJ/mole deg². The accuracy with which the T^3 term can be determined is insufficient to permit any conclusion about its pressure dependence, but the 18% change in γ , which is proportional to the normal-state density of states $N(0)$, is well outside the experimental error. Thus, the application of a pressure of 10 kbar to α uranium raises T_C from below 0.1 to 2°K , increases $N(0)$ by 18%, and eliminates the low-temperature zero-pressure anomaly in the heat capacity.

The BCS expression⁹ for T_C is

$$T_C \approx 0.85\Theta_D \exp[-1/N(0)V],$$

where V measures the strength of the phonon-mediated electron-electron interaction and Θ_D is the Debye temperature. To explain an increase of T_C from 0.1 to 2°K , this relation would require a 65% increase in $N(0)V$. (Θ_D is expected to change by no more than 2% and, therefore, makes a negligible contribution to the change of T_C .) Thus, the enhancement of T_C cannot be ascribed to the change in $N(0)$ alone, and a marked increase in V is implied.

Geballe *et al.*⁸ have recently suggested that the pressure enhancement of the superconducting interaction and the anomalous physical properties of α uranium at 43°K might both be re-

lated to a populating of localized $5f$ states below 43°K . They suggest that the electrons in these $5f$ states inhibit superconductivity, just as $4f$ electrons in lanthanum do,¹⁰ but that application of pressure raises the $5f$ states above the Fermi surface. Such a pressure dependence of the $5f$ states would be similar to that of the $4f$ states in cerium.¹¹ We may employ this model and analogies with the lanthanide metals to explain the effect of pressure on γ and on the zero-pressure low-temperature heat-capacity anomaly. The heat-capacity anomalies in cerium^{12,13} associated with the ordering of $4f$ electrons also disappear¹⁴ on application of 10 kbar. Furthermore, the available γ values¹⁵ for the lanthanide metals suggest that $N(0)$ increases with increasing number of electrons in the $6s5d$ conduction band, but that the localized $4f$ states do not contribute to $N(0)$. Thus, if the zero-pressure anomaly in α uranium is associated with the ordering of $5f$ electrons, its disappearance at 10 kbar and the accompanying increase in γ could both be understood if the $5f$ and $7s6d$ states behaved in the same way as the $4f$ and $6s5d$ states in the lanthanides. (Since the $5f$ states in the actinides are generally believed to be less highly localized than the $4f$ states in the lanthanides, these analogies, although suggestive, should be applied with a certain amount of caution.) Although there is no evidence of a temperature-dependence magnetic susceptibility for α uranium below 43°K ,¹⁶ the number of electrons involved may be very small (that part of the entropy of the zero-pressure anomaly actually observed above 0.1°K amounts to only $3 \times 10^{-4}R$) or, alternatively, the ordering may be antiferromagnetic with an anomaly in χ which is small compared with the relatively large Pauli paramagnetism.¹⁷

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MAXIMUM LOSSLESS CURRENT IN A SUPERCONDUCTING FOIL WITH A SURFACE SHEATH*

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The maximum lossless transport current density of the superconducting surface sheath of a long foil of rectangular cross section is controlled by the larger dimension $2a$ of the cross section when the applied magnetic field is parallel to $2a$ and perpendicular to the current. When $2a$ becomes very large the maximum current density becomes very small.

The maximum current density of the superconducting surface sheath has attracted a great deal of theoretical attention recently.¹⁻⁴ In particular, it was predicted from energy considerations³ that the magnetization per unit volume of a long cylinder due to persistent currents which flow around the axis of a cylinder should be size dependent. This is in quantitative agreement with recent experiments on good surfaces.^{5,6} It was also predicted that the maximum current density of a surface sheath which is infinite in two dimensions is finite,^{1,2} and that for a foil which is infinite in two dimensions with one or both surfaces superconducting the maximum current density depends⁴ on the thickness $2b$ of the foil.

We shall show that for an infinite surface sheath or a foil in two dimensions the maximum lossless current density is zero, contrary to the earlier prediction.^{1,2,4} However, a foil of thickness $2b$, of width $2a$, and of infinite length can carry a finite lossless surface current density whose magnitude is controlled by the width $2a$ and not by the thickness $2b$ when $(b/a)^2 \ll 1$ and $b > \Delta$ (Δ is the thickness of the surface sheath).

We employ the same physical principles as in Ref. 3, namely, that the Gibbs free-energy

difference $\Delta G_{SN}(H_0)$ between the superconducting state in a magnetic field with a current and the normal state (assumed nonmagnetic) without a current is zero for maximum lossless current in the surface sheath. Park⁴ states that he uses the same criterion whereas, in fact, he equates the free energy of the superconducting and normal states with a current flowing in each. $\Delta G_{SN}(H_0)$ may be written³

$$\oint dV \{ (\vec{H} - \vec{H}_0)^2 - \frac{1}{2} |\Psi|^4 \} = \Delta G_{SN}(H_0), \quad (1)$$

where the order parameter $\Psi(x, y, z)$ and the magnetic field $\vec{H}(x, y, z) = \text{curl } \vec{A}(x, y, z)$ have to be determined from the Ginzburg-Landau⁷ equations and Eq. (1). $\vec{H}(x, y, z)$ is the local magnetic field at the applied magnetic field H_0 . Equation (1) is written in the usual Ginzburg-Landau normalization⁷ and the integral is to be extended over all space. When one neglects the more sophisticated details of the internal current distribution of the surface sheath, Eq. (1) means roughly the following: The first term on the left-hand side is the total energy which arises from a total current I in the specimen ($J = I/2a$). The consequence of this current at the applied field H_0 is the magnetic field $(H - H_0)$