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HYPERFINE SPECIFIC HEAT OF URANIUM METAL*

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We have measured the specific heats of U^{235} and U^{238} metals below 0.75° K. Our results show a large T^{-2} contribution to the specific heat of U^{235} , whereas the corresponding term in the U^{238} data is almost negligible. This indicates the presence of a large hyperfine interaction in U^{235} . To our knowledge this is the first such measurement on an actinide metal.

Conventional heat capacity measuring techniques could not be employed in these experiments because of alpha-particle heating. For example, the heating which arises from the U^{234} impurity in U^{235} (approximately 1%) is sufficiently large to warm a sample from 0.4 to 1°K in a minute if the sample is isolated from the cooling paramagnetic salt. Therefore a new method, similar to that devised by Miedema and Haseda,¹ was employed.

Figure 1 shows the essential features of the apparatus. A strip of brass (70% Cu, 30% Zn; approximately 8 mm long, 6 mm wide, and 0.3 mm thick) provided a weak thermal link between the sample and the cold salt. Two carbon resistance thermometers (Speer 470 ohm, 1/2 watt) were mounted at the two ends of the brass link, and a manganin heater was attached to the side of the sample opposite the brass. Provided the power input to the resistors is kept low (less than 10^{-12} watt), the temperature-resistance relationship is given by $T = A \log R / (\log R - B)^2$, where the constants A and B are determined by calibration in the helium region. After cooling the sample we observed the response of the resistors as the input from the heater was increased in small steps from zero to 350 ergs/sec. If the thermal resistance between sample and salt is known, the transient response of the sample thermometer, after the heat input is abruptly changed, gives the sample's heat capacity. The steady-state response for a given heat input gives information

about the thermal resistance between sample and salt.

Assuming (1) the principal thermal resistance is that of the brass link, and (2) the brass' thermal conductivity varies linearly with temperature, we can say for any heater input that

$$C(dT_{U}/dt) = P_{H} + P_{\alpha} - b(T_{U}^{2} - T_{S}^{2}), \qquad (1)$$

where C is the heat capacity of the sample and addenda, P_H is the heater power, P_{α} is the heat input/sec due to α heating and stray heat leaks to the sample, T_U is the temperature of the upper resistor (assumed to be the sample temperature), T_S is the temperature of the lower resistor (T_S is essentially constant for a given P_H), and b is a constant whose value is dependent upon the thermal conductance of the brass strip.



FIG. 1. Schematic diagram of apparatus for measuring specific heat. The various parts of the apparatus are thermally bonded to one another with soft solder or Indalloy solder.

If these assumptions are valid, a graph of P_H versus $(T_U^2 - T_S^2)$ for the steady state $(dT_U/dt = 0)$ should yield a straight line of slope *b* and intercept P_{α} . Further, the value of *B* should be that predicted by the Wiedemann-Franz Law. The steady-state experimental results did, in fact, yield a good straight line. The constant *b* was found to have a value of 800 ergs/sec-deg² in good agreement with the theoretical prediction. In the case of U^{235} , P_{α} was determined as (90 ± 8) ergs/sec, consistent with an expected 87-erg/sec heat input from the α particles.

If P_H is suddenly changed to a new value, Eq. (1) predicts that the transient response of T_{II} will be an approximate exponential rise with a time constant $\tau = C/2bT_U$, if the change in P_H is small. Having obtained b from the analysis of the steady-state conditions, we can infer the value of C by measuring the time constants (6 to 100 seconds in these experiments) associated with changes from one steady-state condition to another. This procedure makes it possible to measure low-temperature specific heats even in the presence of extremely large alpha-particle heating. The method has the disadvantage that it does not yield results of high accuracy and that at sufficiently low temperatures the assumptions which underlie Eq. (1) are no longer valid. To test the method, we used it to determine the specific heat of a 160-mg holmium sample between 0.3° and 0.7° K. The results obtained agree $(\pm 3\%)$ with the predictions of Bleaney.² The results are consistently larger than those of Lounasmaa³ (1% at 0.7° K, 7% at 0.38° K), possibly indicating a smaller quadrupole interaction than that reported by Lounasmaa.

The results for a 4.87-g sample of U^{235} and a 14.03-g sample of U^{238} are presented in Fig. 2. The U^{238} sample was spectroscopically analyzed at Brookhaven National Laboratory. The following impurities were reported: Cu, 0.0001%; Mg, 0.0001%; Al, 0.001%; Si, 0.001%; Mn, 0.005%, Ca, 0.01%. Measurements on U^{238} were feasible down to 0.17°K because of the relatively small amount of α -particle heating. The U^{238} results can be represented by the equation

$$C_{238} = \gamma T + \beta T^{-2}, \qquad (2)$$

where $\gamma = (12.1 \pm 0.3) \text{ mJ/mole-deg}^2$, $\beta \approx 0.15$ mJ-deg/mole. Although magnetic measurements have shown some samples of uranium to be superconducting,⁴⁻⁶ it was not possible to fit our U²³⁸ results with an electronic term consist-



FIG. 2. Specific heats of U^{235} and U^{238} , in mJ/moledeg, corrected for addenda. {The major addenda contributions are a linear term from 5 g of copper, and both a linear and a T^{-2} term [J. C. Ho, H. R. O'Neal, and N. E. Phillips, Rev. Sci. Instr. <u>34</u>, 782 (1963)] from the 0.5-g manganin heater.} Circles represent data obtained in a 600-gauss field; triangles represent zero-field data.

ent with the BCS⁷ predictions for a superconductor. Moreover, our value for γ is in good agreement with that obtained by Smith and Wolcott⁸ in the helium region, and our results were unaffected by the application of a 600-gauss magnetic field. We conclude that our U²³⁸ sample was not superconducting. This is not surprising in view of the variety of results which have been reported for uranium. The hyperfine term in Eq. (2) is larger (by a factor of 2, see below) than can be accounted for by the 0.7% U²³⁵ normally present in U²³⁸. This term is, however, very small, and the discrepancy could be caused by a failure of our technique below 0.3°K.

As can be seen from Fig. 2, the molar specific heat of U^{235} is far larger than that of U^{236} over the entire range of measurement. If we represent the U^{235} results by the expression

$$C_{235} = f(T) + \beta T^{-2}, \qquad (3)$$

where the first term is the electronic contribution and the second the hyperfine contribution, then we find that $\beta = (10.9 \pm 0.7)$ mJ-deg/mole. This uncertainty in β results partly from the scatter in the data and partly from uncertainty as to the form of f(T). The two curves, A and B, correspond to the two choices f(T) = (12.1T)and $f(T) = 70 \exp(-1/T)$ mJ/mole-deg, respectively. The latter expression is the approximate BCS expression for a superconductor with γ = 12.1 mJ/mole-deg² and a critical temperature of 0.7° K.⁶ Either curve represents an approximate fit to all the data. It is therefore impossible to tell whether or not the U^{235} sample was superconducting. However, any reasonable choice for f(T) leads to approximately the same value for the hyperfine contribution.

It is not possible at present to give an unambiguous interpretation of the hyperfine specific heat of U^{235} . Both nuclear magnetic and nuclear electric quadrupole interactions give rise to heat capacities proportional to T^{-2} in the high-temperature approximation. The hyperfine term in the Hamiltonian appropriate to a metal in a magnetically ordered state may be written³

$$H = a'I_{2} + P\{I_{2}^{2} - \frac{1}{3}I(I+1)\}, \qquad (4)$$

where the first term is the magnetic interaction and the second the electric quadrupole interaction, and I = 7/2 for U²³⁵. If we assume the hyperfine interaction to be entirely magnetic, our value of β gives $a' \approx 12 \times 10^{-3}$ deg, a value close to that which one would $expect^2$ if the ions in the metal had three 5f electrons, and if the metal were perfectly ordered. However, while there are some indications that uranium metal may be ordered at low temperatures,⁹ present experimental evidence indicates it is unlikely that uranium ions in the metal have three 5f electrons.¹⁰ Therefore it is possible that the major, and perhaps the total, contribution to the specific heat of U^{235} metal arises from the electric quadrupole interaction. If we assume that the interaction is entirely electric, we obtain $P \approx 8 \times 10^{-3}$ °K. Such a value of P is comparable to those obtained for U²³⁵ in various salts.^{11,12}

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HELICON-PHONON INTERACTION IN SOLIDS

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The existence of the type of electromagnetic wave propagation in a conducting medium known as the helicon wave is now well established both in theory and by experiments on a number of metals and semiconductors.¹ The phase velocity of a helicon wave can be very much less than the velocity of light (velocities of the order of 100 cm \sec^{-1} have been observed) and, in particular,

may be equal to an acoustic velocity in a solid. If a mechanism for interaction exists, the two types of wave will be coupled and their behavior may be expected to deviate significantly from the usual. The purpose of this Letter is to present the results of a calculation of the interaction of acoustic waves and helicon waves in a solid. Several effects are suggested which should be ex-