PHYSICAL REVIEW B VOLUME 33, NUMBER 1 1 JANUARY 1986

## Brief Reports

*Brief Reports are short papers which report on completed research which, while meeting the usual* **Physical Review** *standards of scientific quality, does not warrant a regular article. (Addenda to papers previously published in the* **Physical Review** *by the same authors are included in Brief Reports.) A Brief Report may be no longer than 3½ printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

# Low-temperature specific heat of uranium monosulfide

J. E. Gordon
*Department of Physics, Amherst College, Amherst, Massachusetts 01002*

R. Troc
*Institute for Low Temperature and Structure Research, Polish Academy of Sciences,*
*P.O.Box 937, PL-50-950 Wroclaw, Poland*

(Received 22 July 1985)

The specific heat of a single crystal of US has been measured between 1.25 and 4.55 K. A $(C/T)$-vs-$T^2$ analysis of the data yields a good straight-line fit which corresponds to values for $\gamma$ and $\Theta_D$ of 25.3 mJ/(mole K$^2$) and 181 K. The data show no evidence of a $T^{3/2}$ spin-wave contribution, but do support the view that the linear term in the specific heat is not solely a conduction-electron contribution.

## INTRODUCTION

The specific heat of polycrystalline US was measured between 1.6 and 350 K some years ago.[1,2] The authors reported that the low-temperature specific heat included a $T^{3/2}$ spin-wave contribution and further that $\gamma$, the coefficient of $T$ in the linear contribution to the specific heat, appeared to be considerably smaller at high temperatures than at low. This latter characteristic has been noted in other of the uranium monopnictides and chalcogenides.[3-5]

Recently, Rudigier, Fierz, Ott, and Vogt[6] have measured the low-temperature specific heat of single crystals of USb and UTe. These were the first such measurements on single crystals of the cubic binary uranium compounds, and they yielded $\gamma$ values for these two substances which are significantly smaller than the low-temperature values reported for other of these compounds.[7] Blaise *et al.*[5] have speculated that the large low-temperature $\gamma$'s may include a magnetic term which could be sample dependent, and which might well be larger for polycrystalline than for single-crystal samples. Because of this possible difference between single and polycrystalline samples, and because the $T^{3/2}$ term reported to be present in the earlier low-temperature specific-heat measurements on US is surprising in view of the magnetic anisotropy present in that material, we have made specific-heat measurements on a single crystal of US in the temperature region 1.25–4.55 K.

## EXPERIMENT

The sample used was a 3.933-g single crystal of uranium monosulfide which came from the same batch as the single crystals used in the magnetic measurements made by Alfred and Troc.[8] The sample preparation is described in Ref. 8. The specific-heat data were obtained using the transient method discussed by Gordon, Hall, Lee, and Mortimer.[9] In the present experimental arrangement the sample was sandwiched between two copper plates, one of which was thermally connected to a 1.1-K pumped-$^4$He pot by a small stainless-steel wire. A calibrated germanium thermometer was connected to this plate and a one kilohm film heater was attached to the plate on the opposite side of the sample. The heat capacity of the addenda in this experimental arrangement was measured. It was found to be about 16% as large as the sample heat capacity at 1.25 K and about 38% as large at 4.5 K. In order to check the experimental method, we replaced the single crystal of US by a 2.661-g sample of high-purity aluminum. Specific-heat measurements between 1.25 and 4.55 K yielded values for $\gamma$ and $\Theta_D$ of 1.34 mJ/(mole K$^2$) and 423 K, respectively. These values lie within ~1% of the values of 1.35 and 427.7 reported for Al by Phillips.[10]

## RESULTS AND DISCUSSION

Our specific-heat results for US are plotted in Fig. 1, along with the low-temperature data of Westrum, Walters, Flotow, and Osborne.[1] The agreement between the two sets of data is reasonably good, although the latter data fall slightly lower on the graph. The difference is about 3% at the low-temperature end and about 1% at the higher temperatures. This disparity doubtless reflects both experimental uncertainty in the two sets of measurements and actual differences in sample composition. Nevertheless, it is small compared to the disparities which exist in the case of specific-heat measurements on other cubic uranium compounds.

Figure 2 is a conventional $(C/T)$-vs-$T^2$ graph of the present data. It is evident that the data are well fitted by a straight line, and show no anomalies below 4 K of the kind

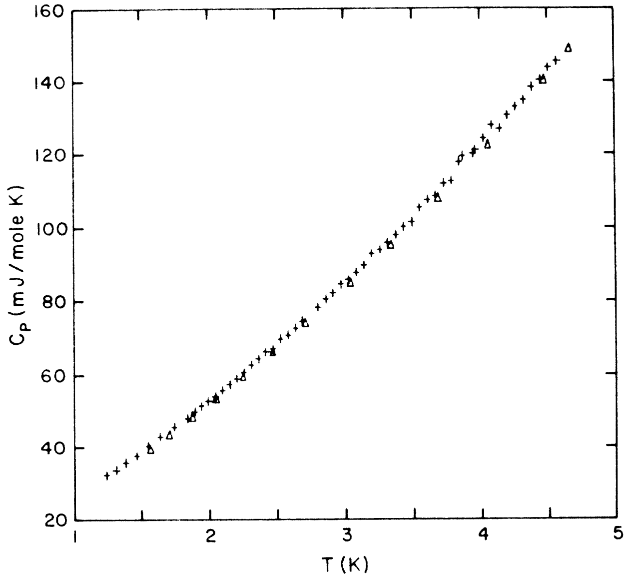

FIG. 1. Molar specific heat of uranium monosulfide. The crosses represent the present measurements and the triangles those of Westrum *et al.* (Ref. 1).

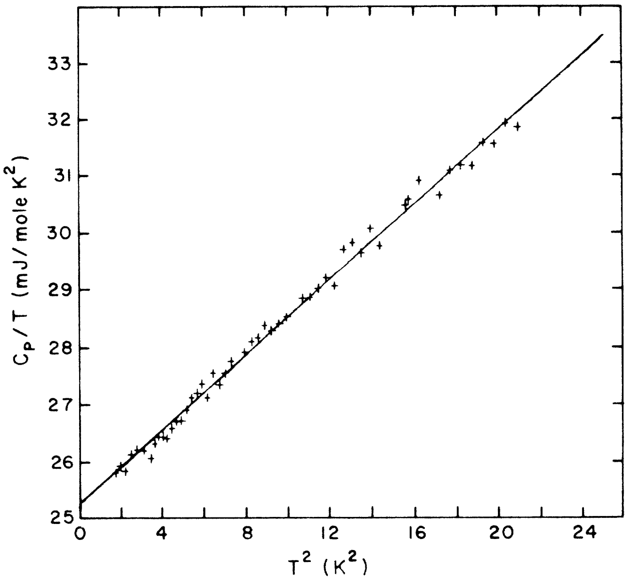

FIG. 2. $(C/T)$-vs-$T^2$ graph of the molar specific heat of US. The straight line is determined by a least-squares fit to the data and corresponds to the equation $C/T=25.3+0.330T^2$ mJ/(mole K$^2$).

reported by Rudigier *et al.*[6] A least-squares fit to the data yields an expression for the low-temperature specific heat of US which is given by Eq. (1):

$$C=25.3T+0.330T^3\ \text{mJ/(mole K)}\ . \tag{1}$$

Equation (1) implies values for $\gamma$ and $\Theta_D$ of 25.3 mJ/(mole K$^2$) and 181 K (assuming $3N$ Debye modes), respectively.

It is possible, of course, to include a $T^{3/2}$ term in a least-squares fitting of our experimental results for the specific heat. If we do so, we obtain

$$C=24.2T+0.856T^{3/2}+0.287T^3\ \text{mJ/(mole K)}\ . \tag{2}$$

This expression is to be compared with that used by Westrum *et al.*[1] and Flotow, Osborne, and Walters[2] to represent their data:

$$C=23.3T+1.099T^{3/2}+0.283T^3\ \text{mJ/(mole K)}\ . \tag{3}$$

The coefficients in the two expressions for $C$ are quite similar. In each the second term contributes relatively little to the value of $C$ over the temperature range 1.25–4.55 K. The coefficient of the $T^{3/2}$ term in Eq. (2) is obtained from the least-squares fit, while that in Eq. (3) was evaluated from the formula given by Dyson[11] for an ideal Heisenberg ferromagnet. However, US is a highly anisotropic ferromagnet.[12] One would therefore not expect a $T^{3/2}$ contribution to the low-temperature specific heat.[13,14] While we have included such a term in arriving at Eq. (2) in order to allow comparison with the earlier work, statistical criteria for goodness of fit to our experimental results provide no justification for inclusion of this term. In our view, then, neither spin-wave theory nor the present experimental results lead to the conclusion that the low-temperature specific heat of single-crystal US includes a $T^{3/2}$ spin-wave contribution.

On the other hand, it is likely that the linear term in Eq. (1) contains more than just a conduction-electron contribution. Whether this enhancement is due to magnetic effects of the kind postulated in Ref. 5, to spin-fluctuation effects, or to electron-phonon or other many-body effects is not clear. However, it does seem evident that this enhancement, whatever its origin, is roughly the same for single and polycrystalline samples. Therefore, the apparent difference in high- and low-temperature $\gamma$'s seen in the polycrystalline US (Ref. 2) is likely to be present in the single crystal as well. We hope to make specific-heat measurements on this single crystal from helium to room temperature to determine if this is, in fact, the case.

## SUMMARY

We find that the low-temperature specific heat of a single crystal of US is well represented by a simple sum of a linear and a cubic term. Our data provide no evidence for a $T^{3/2}$ spin-wave contribution. The $\gamma$ value obtained from our measurements is in reasonably good agreement with that obtained earlier for a polycrystalline sample of US,[1] and is therefore considerably larger than the high-temperature value of $\gamma$ reported in Ref. 2. It is also larger than would be predicted on the basis of band-structure calculations.[15] The origin of this apparent enhancement of the low-temperature $\gamma$ of US, as well as of the low-temperature $\gamma$'s of other binary uranium compounds, appears to us to merit further study, for the effect can now be regarded as an intrinsic property of these materials and not merely as an anomaly due to impurity or polycrystalline effects.

*Note added in proof.* H. Rudigier, H. R. Ott, and O. Vogt [Physica B **130**, 538 (1985)] have recently reported specific-heat results on single crystals of a number of cubic uranium compounds other than US. The results reported here are consistent with their data and conclusions.

## ACKNOWLEDGMENT

We wish to thank Digital Equipment Corporation for donating the data-acquisition system used in these experiments.

[1] E. F. Westrum, Jr., R. R. Walters, H. E. Flotow, and D. W. Osborne, J. Chem. Phys. **48**, 155 (1968).

[2] H. E. Flotow, D. W. Osborne, and R. R. Walters, J. Chem. Phys. **55**, 880 (1971).

[3] H. Yokokawa and Y. Takahashi, J. Phys. Chem. Solids **40**, 603 (1979).

[4] A. Blaise, R. Troc, R. Lagnier, and M. J. Mortimer, J. Low Temp. Phys. **38** 79 (1980).

[5] A. Blaise, J. E. Gordon, R. Lagnier, M. J. Mortimer, and R. Troc, J. Magn. Magn. Mater. **38**, 109 (1983).

[6] H. Rudigier, Ch. Fierz, H. R. Ott, and O. Vogt, Solid State Commun. **47**, 803 (1983).

[7] A. Blaise, J. Phys. (Paris) Colloq. **40**, C4-49 (1979), and references therein.

[8] A. T. Aldred and R. Troc, J. Magn. Magn. Mater. **36**, 217 (1983).

[9] J. E. Gordon, R. O. A. Hall, J. A. Lee, and M. J. Mortimer, Proc. R. Soc. London, Ser. A **351**, 179 (1976).

[10] N. E. Phillips, Phys. Rev. **114**, 676 (1959).

[11] F. J. Dyson, Phys. Rev. **102**, 1230 (1956).

[12] D. L. Tillwick and P. de V. du Plessis, J. Magn. Magn. Mater. **3**, 319 (1976).

[13] A. R. Mackintosh, Phys. Lett. **4** 140 (1963).

[14] B. R. Cooper, Proc. Phys. Soc. London **80**, 1225 (1962).

[15] H. L. Davis, in *The Actinides: Electronic Structure and Related Properties,* edited by A. J. Freeman and J. B. Darby (Academic, New York, 1974), Vol. II, p. 1.