High-field specific heat of the spin-fluctuation system UAl_2

G. R. Stewart and A. L. Giorgi
Materials Science and Technology Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545

B. L. Brandt and S. Foner
Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

A. J. Arko
Materials Science and Technology Division, Argonne National Laboratory, Argonne, Illinois 60439
(Received 8 February 1983)

Measurements of the low-temperature specific heat, both in 0- and 7-T applied field, and magnetic susceptibility on polycrystalline, unannealed single-crystal and annealed single-crystal samples of UAl_2 show no sample dependence for these properties to ±5%, in contrast to recent results for $TiBe_2$. Measurements of the specific heat of the annealed single crystal of UAl_2 from 2 to 23 K in fields to 12.5 T and from 4 to 16 K in fields to 17 T indicate a partial (~40%) suppression of the spin-fluctuation contribution, which occurs at 2 K in 12.5 T, and the possible existence of a small competing effect which increases the specific heat with increasing field above 4 K. These results are compared with recent magnetization and magnetoresistivity results which indicate suppression of spin fluctuations in UAl_2 between 15 and 20 T.

I. INTRODUCTION

Only two metallic systems are known which exhibit what has been called[1] the only "hard proof" of spin fluctuations, that is, a $T^3\ln T$ term in the low-temperature specific heat (LTSH). Such a divergent LTSH term was discovered[2] in UAl_2 in 1975 and in $TiBe_2$ in 1982.[3] Many other systems exist for which spin fluctuations have been suggested, e.g., Pd,[4] Sc,[5] $LuCo_2$,[6] and $CeSn_3$.[7] Measurements made on these systems include resistivity, magnetic susceptibility, and LTSH. Such measurements as a function of applied magnetic field are of particular interest due to the expectation of suppression of spin fluctuations and therefore changing physical properties at some applied field.

Unfortunately, many of these measurements seem to be contradictory. For example, the LTSH of Pd in an applied magnetic field of 11 T shows[4] an 8% decrease in the coefficient γ of the term linear in temperature T,

$$C=\gamma T+\beta T^3 . \tag{1}$$

This was interpreted[4] as a partial suppression of the spin-fluctuation mass enhancement of the electrons

$$0.141\gamma=N(0)(1+\gamma_{\rm SF}+\gamma_{e\text{-ph}}) \tag{2}$$

by the applied field, where $N(0)$, the bare electronic density of states, is "dressed" by both an electron-phonon interaction, $\gamma_{e\text{-ph}}$, and the spin-fluctuation interaction $\gamma_{\rm SF}$. However, magnetization measurements[8] on Pd to 30 T show no apparent field-dependent susceptibility to 2%. A further problem is that the observed magnitudes of field effects, i.e. $[\gamma(H)-\gamma(0)]/\gamma(0)$, in the LTSH for Pd,[4] Sc,[5] $LuCo_2$,[6] and $CeSn_3$ (Ref. 7) are much larger than would be predicted from estimates based on the spin-fluctuation temperature $T_{\rm SF}$ and the Stoner enhancement factor S derived from resistivity or susceptibility data. In Pd, for example, $T_{\rm SF}$ is estimated to be about 250 K and S is about 10. Hertel, Appel, and Fay predict[9] a 1% depression of $1+\gamma_{\rm SF}$ for a 10-T applied field versus the reported 7% effect.[4] One possibility is that estimates of $T_{\rm SF}$ based on the temperature where the susceptibility has a peak may be incorrect. It has been suggested[6] that this peak for $LuCo_2$ may be due to a band-structure effect, rather than due to spin fluctuations.

Firstly, one advantage in measuring either $TiBe_2$ or UAl_2 is that, with some approximations discussed below, one may derive $T_{\rm SF}$ from the $T^3\ln T$ term in the LTSH with the knowledge that at least $T_{\rm SF}$ is derived from an effect generally believed[1] due to spin fluctuations. Secondly, one can measure $C(H)$ in the temperature range where the $T^3\ln T$ term is quite sizable and therefore once again be reasonably certain that one is measuring the response of the spin fluctuations to field.

We have previously reported[10] such LTSH measurements to 17 T for $TiBe_2$. The present work is a similar study for UAl_2, and provides a comparison to a recent study[11] of the magnetization and magnetoresistance of UAl_2.

II. EXPERIMENTAL

Owing to the sample dependence of ρ, χ, C, and $[\gamma(H)-\gamma(0)]/\gamma(0)$ observed[3] for the $TiBe_2$ system, the present work used three different samples of UAl_2: an unannealed, polycrystalline, arc-melted sample prepared at Los Alamos, plus an unannealed and an annealed single-crystal sample prepared at Argonne National Laboratory. The latter two samples came from a large single crystal grown by the induction-melting floating-zone technique in an argon atmosphere. The residual resistivity ratio (between 300 and 4 K) for the unannealed single crystal was about 12. The annealed single crystal was kept at 1000 °C for 2.5 d under argon at twice atmospheric pressure and had a resistivity ratio (between 300 and 4 K) of 23. The lattice parameters measured by a Debye-Scherrer powder camera for the polyscrystalline, unannealed, and annealed

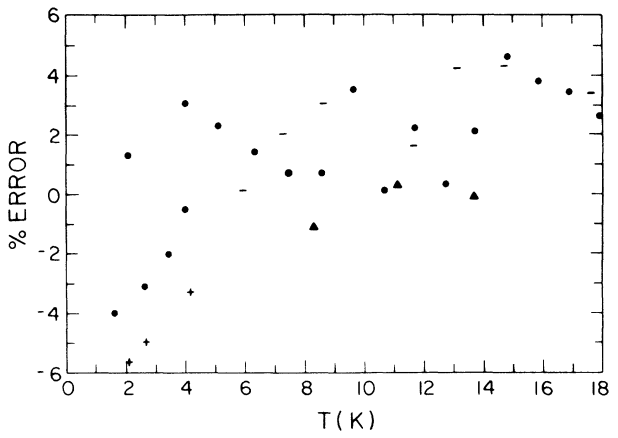

FIG. 1. Shown is the percentage error, $100\times(C^{\text{measured}}-C)/C$ (where C is derived from Ref. 26), found in measuring the LTSH of a 212-mg sample of high-purity Ge at zero (closed circles), 10- (dashes), 12.5- (pluses), and 17-T (closed triangles) applied field. For a detailed discussion of measurement technique in applied field, see Refs. 10, 13, and 15.

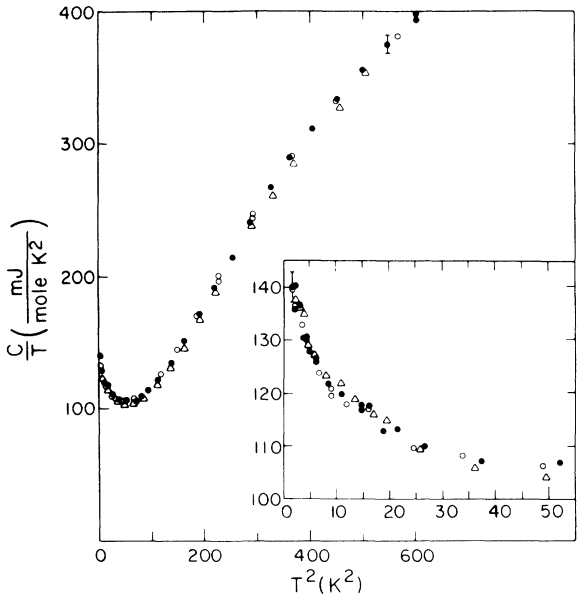

FIG. 2. LTSH data for polycrystalline (open circles), unannealed single-crystal (closed circles), and annealed single-crystal UAl_2 (open triangles) from 1.4 to 24.5 K. These data, plus the data of Ref. 2, all agree to within ±2%. Not shown for the sake of clarity in this figure are data in 7 T for all three samples for $T>4$ K which show very little change (1—2% increase) from the zero-field values.

single-crystal samples were 7.7639, 7.7640, and 7.7656±0.0003 Å, respectively.

The susceptibility of the three UAl_2 samples was measured at 1.2 T in a vibrating sample magnetometer[12] adapted to a superconducting magnet. The low-temperature specific-heat apparatus has been described elsewhere.[10,13–15] Two sets of LTSH data were taken in overlapping field ranges. The first set of LTSH data was obtained at the Francis Bitter National Magnet Laboratory in a water-cooled, high-power Bitter magnet in fields of 0, 6.5, 11.4, 14.2, and 17.0 T. The second set of field data was measured at Los Alamos in a superconducting state magnet in fields of 0, 7, 11, and 12.5 T. Agreement between the two sets of data at similar fields was better than ±2%. LTSH measurements in the normal-state magnet were limited to $T>4$ K due to the large amount of electrical and vibrational noise present in the Bitter magnet (see Ref. 13 for a complete discussion). Measurements in the superconducting magnet extended down to 2 K. Temperatures in zero field were accurate to 0.050 K; temperatures in field were determined by using both a capacitance thermometer and the known[16] magnetoresistence of a carbon-glass thermometer. The uncertainty of this temperature determination in a field was less than ±0.1 K. Additionally, the LTSH of a piece of high-purity Ge was measured in field in both the Bitter and the superconducting magnets as a check on the accuracy of the $C(H)$ data. These results are shown in Fig. 1. Clearly, the accuracy of the measurements in field is close to the accuracy of the zero-field results.

III. RESULTS AND DISCUSSION

The susceptibility of all three different samples was measured to be $(15.1\pm0.7)\times10^{-6}$ emu/g G at 4.2 K. These data were taken on small pieces ($\leq$200 mg) of the samples to avoid the problem of a non-zero-field gradient over the volume of the sample. This number is in agreement with the results of Trainor *et al.*,[2] obtained by using a Faraday balance, and of Franse *et al.*,[11] using an integration method, of $\chi=14.7\times10^{-6}$ emu/g G. Using yet another method, that of extraction, Fournier[17] obtained $\chi(4.2\text{ K})=13.8\times10^{-6}$ emu/g G for UAl_2.

The main result of the susceptibility measurements in the present work that we wish to focus on is the lack of significant sample dependence. The specific-heat data on the same three samples are shown in Fig. 2 and also show no sample dependence (to ±2%). In addition, these data agree to within approximately 2% of the original published data of Ref. 2 over the whole temperature range of measurement. This indicates that the UAl_2 system does not have the side issue of sample-dependent properties which has hampered the understanding of $TiBe_2$.[3]

A. LTSH at zero field

The zero-field data of Fig. 2 may be expressed as

$$C/T=\gamma_0(1+\lambda_{\text{SF}}+\lambda_{e\text{-ph}})+\epsilon T^2+\alpha T^4+\delta T^2\ln T\ . \quad (3)$$

A least-squares computer fit of this expression to the zero-field (closed circles) LTSH data for the annealed UAl_2 crystal over the temperature range 2—23 K is shown in Fig. 3. The accuracy of the fit to these 32 data points is as good as the scatter in the data, ±2%. In order to compare these results to the earlier analysis of the LTSH of UAl_2 by Trainor *et al.*,[18] a fit only to the low-temperature data (shown in the expanded inset in Fig. 3) has also been done using Eq. (3) without the αT^4 term. The results of both these fits are shown in Table I. The αT^4 term is needed to fit the negative curvature of the data shown in

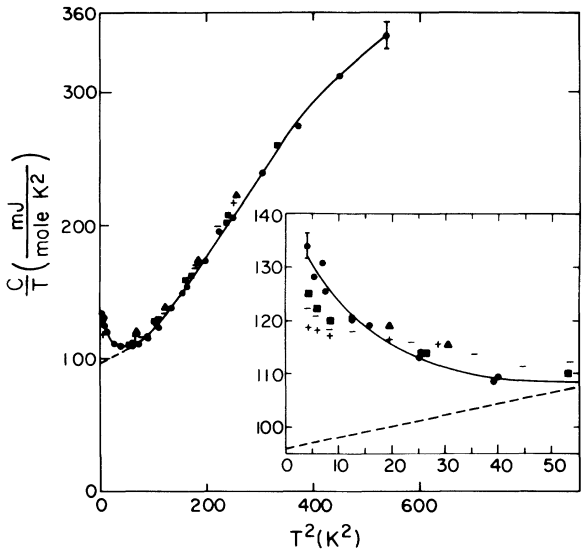

FIG. 3. LTSH data in 0–17 T for the annealed single crystal of UAl_2. The solid line through all the zero-field data (closed circles) is a least-squares computer fit of the data to Eq. (3). The solid line through the low-temperature zero-field data shown in the inset is a similar fit, with the T^4 term omitted. The closed squares are data in 8 T for $T^2<25$ K^2 and in 7 T otherwise, the dashes are data in 11 T, the pluses are data in 12.5 T for $T^2<20$ K^2 and 14.2 T otherwise, and the closed triangles are data in 17 T. A data point ($C/T=127.1$ mJ/mole K^2; $T^2=4.248$ K^2) at 4 T is omitted for clarity. The dotted lines are drawn to follow Eq. (7) using $\Theta_D=304$ K to give approximate values for full suppression of the spin fluctuations as discussed in the text.

Fig. 3 above $T^2=300$ K^2 and clearly makes very little difference in the other parameters. Also shown in Table I are the results[18] of a three-term fit [Eq. (3) with no αT^4 term] to the data of Ref. 18. Agreement is again fairly close.

The authors of Ref. 18 extracted the spin-fluctuation temperature T_{SF} from their fit to the data by making an approximation to $\gamma_0(1+\lambda_{e\text{-ph}})$ from their LTSH data from 10 to 20 K and presuming that spin fluctuations (λ_{SF}) were unimportant in that temperature range. Visually, this is a compelling argument. Figure 3 shows the telltale upturn due to the $T^2\ln T/T_{SF}$ term [the negative part of which $-T^2\ln T_{SF}$ is part of T^2 in Eq. (3) above] only at low temperatures. However, the supposition that λ_{SF} is negligible in UAl_2 already at 10 K seems inconsistent with the spin-fluctuation temperature derived in the very same work of 23–25 K.

Nevertheless, without a calculation for γ_0 (mJ/mole K^2), or equivalently the bare electronic density of states $N(0)$ for UAl_2, some approximation must be made to obtain the Stoner enhancement factor S in order to calculate T_{SF},

$$S=\chi^{\text{measured}}/\mu_B^2 N(0)\ , \tag{4}$$

where

$$N(0)=0.1415\gamma_0\ . \tag{5}$$

Once S is known along with γ_0, then one uses δ, the coefficient of the $T^2\ln T$ term in Eq. (3), and the theory of Ref. 19 to calculate T_{SF}:

$$T_{SF}=\frac{3\pi^2}{4}\left[\frac{\gamma_0 S}{5\delta}\right]^{1/2}\left[1-\frac{1}{S}\right]^2\ . \tag{6}$$

How shall γ_0 be approximated? In a normal metal

$$\frac{C}{T}=\gamma_0(1+\lambda_{e\text{-ph}})+\beta T^2\ , \tag{7}$$

where β (mJ/mole K^4) is related to the Debye temperature Θ_D via

$$\Theta_D=10\left[\frac{1944\times 3}{\beta}\right]^{1/3}\ . \tag{8}$$

Thus, on a C/T vs T^2 plot such as in Fig. 3, the intercept at $T=0$ is $\gamma_0(1+\lambda_{e\text{-ph}})$ and the slope of the data is just β. From knowing[20] Θ_D for $LaAl_2$ ($=374$ K), $\beta^{UAl_2}=0.208$,

$$\Theta_D^{UAl_2}=\left[\frac{m^{LaAl_2}{}_2}{m^{UAl_2}}\right]^{1/2}\Theta_D^{LaAl_2}\ , \tag{9}$$

where m is the mass of the respective compound. A dotted line fitting a relationship as in Eq. (7) and having this slope is drawn in Fig. 3, giving $\gamma_0(1+\lambda_{e\text{-ph}})=96$. Using $\lambda_{e\text{-ph}}\simeq 0.3$, Eq. (5), and $\chi^{\text{measured}}=15.1\times 10^{-6}$ emu/g G, Eq. (4) gives $S\simeq 4.35$. T_{SF} is then 30 K if δ is derived only from the low-temperature data (Table I), and 28 K if δ is derived from the high-temperature data.

TABLE I. Analysis of the LTSH data (mJ/mole K) in Fig. 3 for annealed, single-crystal UAl_2.

$\gamma_0(1+\lambda_{SF}+\lambda_{e\text{-ph}})$	ϵ	α	δ	
142.3	−3.644	−0.00169	1.566	(four-term fit to data over entire temperature range)
142.1	−3.437		1.413	(three-term fit to the low-temperature data only)
142	−3.48		1.35	(three-term fit from Ref. 17 of their data from 0.9 to 3 K)

A way to bypass the uncertainty of approximating $\gamma_0(1+\lambda_{e\text{-ph}})$ is to use the coefficient of the T^2 term in Eq. (3)

$$\ln T_{SF}=\frac{\epsilon\beta}{\delta}+B_0+\ln(0.79)\left[1-\frac{1}{S}\right] \tag{10}$$

to calculate T_{SF} if $N(0)$ and therefore S is known, Eq. (4). However, there is disagreement[19,21] over what to use for B_0. This uncertainty leads to a factor of 2.11 ($\simeq 8$) between results from the two theories for T_{SF}. Therefore Eq. (6) should be used to obtain T_{SF}.

B. LTSH in fields to 17 T

The field data are shown by different symbols in Fig. 3. The highest field data (14.2 and 17.0 T) exist only for $T>4$ K. The dotted line in Fig. 3 and in the expanded inset serve as an ansatz for what the specific heat would approximately be in the absence of spin fluctuations. Reference 18 chose an intercept $\gamma_0(1+\lambda_{e\text{-ph}})$ slightly lower (90 vs 96) than the one chosen in the present work, i.e., the percentage of the LTSH assigned to spin fluctuations in Fig. 3 is a conservative, lower-limit estimate.

What is immediately striking about the $C(H)$ data in Fig. 3 is that there is *no* apparent suppression of the spin-fluctuation-caused upturn in the specific heat up to fields of 17 T for $T\geq 4$ K. In fact, the field appears to increase the specific heat slightly (4–6% at 17 T) for $T>5$ K. [LTSH data in 7 T for all three samples (not shown in Fig. 2 for sake of clarity) for $T\geq 4$ K shows that the field effect on the specific heat, at least to 7 T, is sample independent to $\pm 1\%$.]

This decrease in field of the specific heat only at the very lowest temperatures is in contrast with the results[10] for $TiBe_2$, where the magnitude of the decrease grew smaller as temperature increased but did not vanish until about 15 K. However, since $TiBe_2$ has a significantly higher S (65) than UAl_2 and a T_{SF} which is approximately the same,[3,22] one would expect qualitatively a field effect smaller in UAl_2 than in $TiBe_2$, since[23] at low fields

$$\frac{C(H)-C(0)}{C(0)}\propto\frac{H^2}{T_{SF}^2}\frac{S}{\ln S}\,.$$

The right-hand side of this relation, assuming T_{SF} is identical for UAl_2 and $TiBe_2$, is a factor of 5 larger for $S=65$ than for $S=3.2$. $[C(12.5\,\mathrm{T})-C(0)]/C(0)$ at 2 K is -10.5% for UAl_2 and approximately[10] -20% for $TiBe_2$ for 12.5 T. The slight increase in the LTSH for $T>4$ K in field appears to be outside the error limits graphically shown in Fig. 1 where the error in $C(H)$ for a standard is shown.

It is possible that this 4–6% increase of the data for $T>4$ K in 17 T is due to a competing effect, i.e., some part of the specific heat which, as temperature increases, increases slightly with field. At $T\leq 4.5$ K, an increase in β [Eq. (7)] for $CeSn_3$ has been seen[7] which saturated at about 6 T at a value twice the zero-field value. This increase was ascribed[7] in a qualitative way to an induced magnetic moment on the Ce atoms which had been seen by neutron scattering. Although a similar induced magnetic moment on the U atoms in UAl_2 has also been observed by neutron scattering,[24,25] what is observed in the field data for UAl_2 is quite different from the results for $CeSn_3$. Not only does the increase appear not to saturate up to 17 T, the increase is found at higher temperatures.

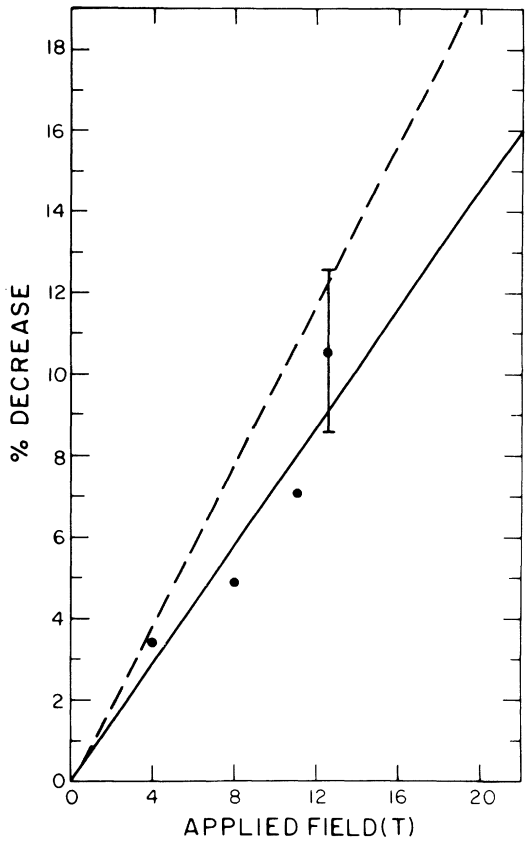

FIG. 4. This figure shows the percent decrease in the LTSH at 2.1 K vs field. The dotted line is the corrected decrease vs field assuming a competing effect of + 3% at 11 T, as discussed in the text. It should be noted that the percent decrease at 4 T (3.4%) is somewhat larger than that reported in Ref. 2 at the same temperature and field.

The percentage decrease in C with field is plotted in Fig. 4 at the lowest temperature of measurement. If the 11-T point at $T^2=53.5$ K^2 [i.e., at a temperature where spin fluctuations are unimportant (see Fig. 3)] is used to infer the percentage change to be assigned to the competing effect, the percentage decrease shown in Fig. 4 for 11 and 12.5 T would be increased by 3%. These two possibilities for $\Delta C(H)$ at 2.1 K are shown as solid and dotted lines, respectively, in Fig. 4. The dotted line shown in the inset in Fig. 3 may be used as discussed above for an indication of how far the LTSH would need to be decreased to correspond to full suppression of spin fluctuations. At 2.1 K, full suppression of the spin fluctuations in UAl_2 would be accompanied by approximately a 26% decrease in the specific heat. With the use of the steeper of the two lines in Fig. 4, this amount of decrease is extrapolated to occur at 26 T. Of course, this is really only a very rough approximation due to (1) the scatter in the data in Fig. 4; (2) the uncertainty in assigning a contribution to the supposed competing effect; (3) the possibility that the correct behavior of $\partial C/\partial H$ is nonlinear.

Therefore, while the 17-T LTSH data for UAl_2 shown

in Fig. 3 appears at first glance in contradiction to the recent statement[11] that spin fluctuations in UAl_2 are fully suppressed between 15 and 20 T, the lower-field LTSH data below 4 K implies the presence of an effect which partially counteracts the expected[11] decrease in the LTSH due to suppression of spin fluctuations.

IV. CONCLUSIONS

UAl_2, in contrast to $TiBe_2$, exhibits very little sample dependence in both its low-temperature susceptibility and specific heat. Partial ($\sim$40%) suppression of the spin-fluctuation-caused upturn in the low-temperature specific heat of UAl_2 is observed at 2.1 K in 12.5 T, consistent with both the rather low spin-fluctuation temperature calculated in the present work and recent magnetization and resistivity experiments[11] in high field. An increase in the specific heat with increasing field at temperatures above 4 K remains unexplained.

(A recent, more thorough measurement of a Ge standard in high field, with over 20 data points, indicates that about half of this unexplained increase above 4 K in field is due to a slight decrease in the thermal conductivity of the platform support wires. The remaining 3% anomalous increase may be said to be within our error bars.)

ACKNOWLEDGMENTS

The authors would like to thank L. G. Rubin for technical assistance and helpful discussion. Part of this work was performed at the Francis Bitter National Magnet Laboratory which is supported at Massachusetts Institute of Technology by the National Science Foundation. Work at Los Alamos and Argonne was completed under the auspices of the U.S. Department of Energy.

[1]M. B. Brodsky, Rep. Prog. Phys. 41, 1547 (1978). It should be pointed out that the electron-phonon interaction has also been predicted to give a $T^3 \ln T$ term in the LTSH. See G. M. Eliashberg, Zh. Eksp. Teor. Fiz. 43, 1005 (1962) [Sov. Phys.—JETP 16, 780 (1963)].

[2]R. J. Trainor, M. B. Brodsky, and H. V. Culbert, Phys. Rev. Lett. 34, 1019 (1975).

[3]G. R. Stewart, J. L. Smith, A. L. Giorgi, and Z. Fisk, Phys. Rev. B 25, 5907 (1982).

[4]T. Y. Hsiang, J. W. Reister, H. Weinstock, G. W. Crabtree, and J. J. Vuillemin, Phys. Rev. Lett. 47, 523 (1981).

[5]K. Ikeda, K. A. Gschneidner, Jr., T.-W. E. Tsang, and F. A. Schmidt, Solid State Commun. 41, 889 (1982).

[6]K. Ikeda and K. A. Gschneidner, Jr., Phys. Rev. Lett. 45, 1341 (1980).

[7]K. Ikeda and K. A. Gschneidner, Jr., Phys. Rev. B 25, 4623 (1982).

[8]F. A. Muller, R. Gersdorf, and L. W. Roeland, Phys. Lett. 31A, 424 (1970). In agreement with the 30-T data is a lower-field (15-T) magnetization measurement on Pd, see S. Foner and E. J. McNiff, Jr., Phys. Rev. Lett. 19, 1438 (1967).

[9]P. Hertel, J. Appel, and D. Fay, Phys. Rev. B 22, 534 (1980).

[10]G. R. Stewart, J. L. Smith, and B. L. Brandt, Phys. Rev. B 26, 3783 (1982).

[11]J. J. M. Franse, P. H. Frings, F. R. de Boer, A. Menovsky, C. J. Beers, A. P. J. van Deursen, H. W. Myron, and A. J. Arko, Phys. Rev. Lett. 48, 1749 (1982).

[12]S. Foner, Rev. Sci. Instrum. 30, 548 (1959). See also, A. Zieba and S. Foner, *ibid.* 53, 1344 (1982), and references cited therein.

[13]G. R. Stewart, B. Cort, and G. W. Webb, Phys. Rev. B 24, 3841 (1981).

[14]G. R. Stewart, Cryogenics 18, 120 (1978); G. R. Stewart and A. L. Giorgi, Phys. Rev. B 17, 3534 (1978).

[15]G. R. Stewart, Rev. Sci. Instrum. 54, 1 (1983).

[16]H. H. Sample, B. L. Brandt, and L. G. Rubin, Rev. Sci. Instrum. 53, 1129 (1982).

[17]J. M. Fournier, Solid State Commun. 29, 111 (1979).

[18]R. J. Trainor, M. B. Brodsky, and G. S. Knapp, in *Plutonium 1975,* edited by H. Blank and R. Lindner (North-Holland, Amsterdam, 1976), p. 475.

[19]W. F. Brinkman and S. Engelsberg, Phys. Rev. 169, 417 (1968).

[20]R. J. Schiltz and J. F. Smith, J. Appl. Phys. 45, 4681 (1974).

[21]S. Doniach and S. Engelsberg, Phys. Rev. Lett. 17, 750 (1966).

[22]A. L. Giorgi and G. R. Stewart, Solid State Commun. 44, 1465 (1982).

[23]M. T. Beal-Monod, S.-K. Ma, and D. R. Fredkin, Phys. Rev. Lett. 20, 929 (1968).

[24]V. C. Rakhecha, G. H. Lander, A. J. Arko, and R. M. Moon, J. Appl. Phys. 52, 1636 (1981).

[25]Another fact that may have bearing on the cause of the observed positive $\partial C/\partial H$ is the existence of a field-dependent susceptibility in UAl_2. This field dependence saturates by 2 T, is thought to be due to magnetic impurities, and is found in all uranium compounds: M. B. Brodsky (private communication). Although this field-dependent susceptibility is only a few percent in magnitude, the presence of magnetic impurities in UAl_2 will undoubtedly add a (unknown) contribution to $\partial C/\partial H$.

[26]P. Flubacher, A. J. Leadbetter, and J. A. Morrison, Philos. Mag. 4, 273 (1959).