PHYSICAL REVIEW B VOLUME 45, NUMBER 10 1 MARCH 1992-II

Magnetic measurements of $CeAl_3$ to below 1 mK

O. Avenel,* J. S. Xia, B. Andraka, C. S. Jee,† M-F. Xu, Y. J. Qian,‡
T. Lang, P. L. Moyland, W. Ni, P. J. C. Signore, E. D. Adams, G. G. Ihas, M. W. Meisel,§
G. R. Stewart,** N. S. Sullivan, and Y. Takano
Department of Physics and Center for Ultra-Low Temperature Research, University of Florida, 215 Williamson Hall,
Gainesville, Florida 32611

(Received 16 September 1991; revised manuscript received 25 November 1991)

The magnetic susceptibility of a polycrystalline, single-phase sample of $CeAl_3$ has been measured from 10 K to below 800 μK. Above 40 mK, the temperature dependence of the susceptibility is consistent with the results of other groups and possesses a broad peak around 500 mK. Using standard rf superconducting-quantum-interference-device detection techniques operating at 16 Hz, the sample, which was located in a shielded environment having a residual static field of less than 2 nT, was not observed to show any magnetic anomaly from 40 mK down to the lowest achievable temperature.

In an attempt to understand heavy-fermion materials, considerable experimental and theoretical effort has been devoted to identifying the ground-state properties of these materials.[1–7] No single ground state for all the heavy-fermion systems can be identified as the materials seem to be classified as either paramagnetic, magnetic, superconducting, or both magnetic and superconducting.[1–8] For a number of years, it has been thought that $CeAl_3$, the first material to be classified as a heavy-fermion material,[1,9] possessed a paramagnetic ground state. Its magnetic susceptibility[9,10] and zero-field heat capacity at no applied pressure[10–15] show broad, low peaks at ~500 mK. These and other low-temperature results[16–20] were thought to be associated either with the development of a Fermi liquid-like state or were characteristic of a nonmagnetic Kondo lattice. Since 1988 there has been additional experimental evidence, namely, muon-spin-rotation spectroscopy[21] and transport measurements on a single-crystal sample,[22] which seem to indicate that the actual ground state of the system may be antiferromagnetic, although the precise long-range nature of this state in $CeAl_3$ remains unclear.[7] The existence of a magnetic ground state is consistent with the previous experimental results.[9–20]

The purpose of our work was to search, at significantly lower temperatures than the previous 10-mK limit,[3] for additional evidence which could assist in identification of the $CeAl_3$ ground state. Since a definitive theoretical description is lacking, the only possible guide for the experiments is a comparison of the various characteristics observed in different heavy-fermion materials.[1–7] However, such a comparison does not unambiguously eliminate the many possibilities, such as superconducting or magnetic ordering transitions, which might be anticipated. A similar discussion may also be applied to $CeCu_6$, whose properties resemble those of $CeAl_3$ (Refs. 1–7). A preliminary report of a possible phase transition, at approximately 2 mK, in $CeCu_6$ has been presented by Jin *et al.*[23]

Our polycrystalline sample was prepared by arc melting using the highest purity Ce commercially available from Ames Laboratory and was annealed at 1000 °C for 56 days. The sample was from the same batch of material that was used in zero and high magnetic field specific heat work[24] in which no anomalies were detectable at 2.5 or 4 K from the well-known second phases. The high-temperature magnetization measurements were made from 1.8 to 10 K in a magnetic field of 0.5 T using a commercial magnetometer.[25] The low-temperature low-frequency susceptibility studies were performed from 40 mK to 2.0 K using standard mutual inductance techniques operating at 317 Hz in the remnant magnetic field of the earth. For both temperature regimes, the background contribution of the entire assembly has been measured, and this small contribution has been subtracted from the results.

In a third apparatus, our investigations were performed from 100 mK down to approximately 800 μK. Susceptibility measurements were performed at 16 Hz with an ac mutual inductance bridge using a rf superconducting-quantum-interference device (SQUID) as a null detector.[26] The experimental tower was modeled after the arrangement used by Buchal *et al.*[27] and is shown in Fig. 1. The $CeAl_3$ sample was silver epoxied[28] to the end of an annealed copper finger that was bolted to the top of a copper nuclear demagnetization stage. A cylindrical tungsten sample[29] (5.84 mm long and 1.52 mm in diameter) was silver painted[30] inside a small cavity cut into the copper cold finger. The shields and the coils surrounding these samples were thermally anchored to the mixing chamber of the dilution refrigerator. Magnetic shielding is very important because the critical fields of the samples can be low, and supercooling can also suppress the transition in a small applied magnetic field. The pick-up and excitation coils were placed inside a Cryoperm[31] cylinder, and a Nb shield surrounding the whole assembly, as shown in Fig. 1. A room-temperature Mumetal shield, which was wrapped around the cryostat, was only removed after the Nb shield had been cooled to well below its superconducting transition. Prior to mounting, the Cryoperm cylinder had been carefully heat treated after welding, and degaussed at room

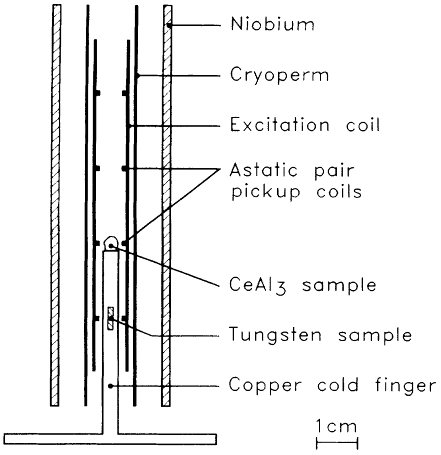

FIG. 1. A schematic of the experimental tower of the ultra-low-temperature experiment is shown.

temperature inside a Mumetal shield. Residual magnetic fields were measured by observing the shift at the SQUID output when flux was expelled from the tungsten sample at its superconducting transition,[32] and could be compensated to less than 2 nT by feeding a dc current to the excitation coil. Without any compensation, the trapped field was observed to be kept below 20 nT by the Mumetal-Nb-Cryoperm combination, and to vary by at most 2 nT from one demagnetization run to another. The magnetic field perpendicular to the axis of the detection coils could be neither detected nor compensated, but this component is believed to be small in our geometry.

Thermometry for the microkelvin experiment was provided by a strain-gauge[33] ^{3}He-melting-curve thermometer.[34] The tungsten superconducting transition provided a temperature fixed point,[34,35] and its variation with applied magnetic field[36] could also be used as a further check of the consistency of the temperature and magnetic field calibrations. The lowest temperature achieved in the present experiment was below 800 μK, a conservative estimate from the reading of the ^{3}He-melting-curve thermometer, which becomes very insensitive below 900 μK.

For the microkelvin work, the response of the ac mutual-inductance bridge operating at 16 Hz was dominated by eddy currents generated in the cold copper finger. To ensure that these currents would produce negligible heating and that the magnetic field on the sample was minimized, we only used very low excitation levels. Consequently, our sensitivity was rather low, and we were unable to make accurate quantitative susceptibility measurements. Nevertheless, at our operating excitation level of 1.7 nT rms, we could have resolved a superconductive signal corresponding to a Meissner effect involving only 1% of the sample volume. In other words, below 40 mK , any change in the $CeAl_3$ susceptibility, if present, was less than 0.01 of $(-1/4\pi)$.

The high-temperature magnetic susceptibility results are shown in Fig. 2 along with the low-temperature, low-frequency data obtained down to 40 mK, which are also given in Fig. 3. Since the mutual-inductance technique provided only relative changes in the susceptibility, the low-temperature data have been normalized to the high-temperature results in the region where the two sets of

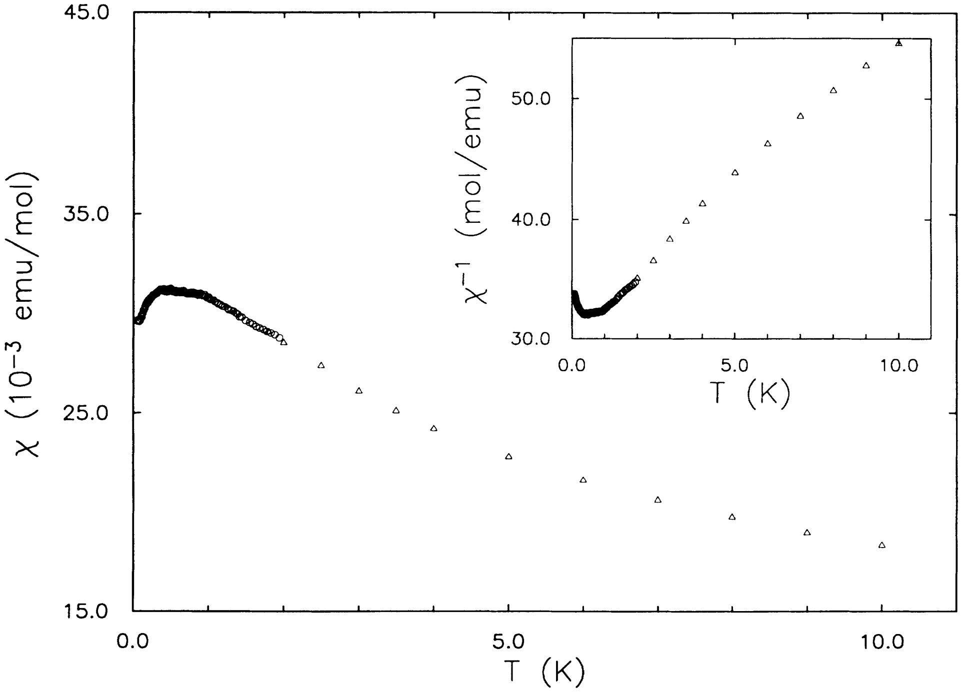

FIG. 2. The susceptibility is shown as a function of temperature. The triangles are data taken in a commercial magnetometer, and the circles are data acquired by standard low-frequency mutual-inductance techniques; see text. The inset shows the inverse susceptibility.

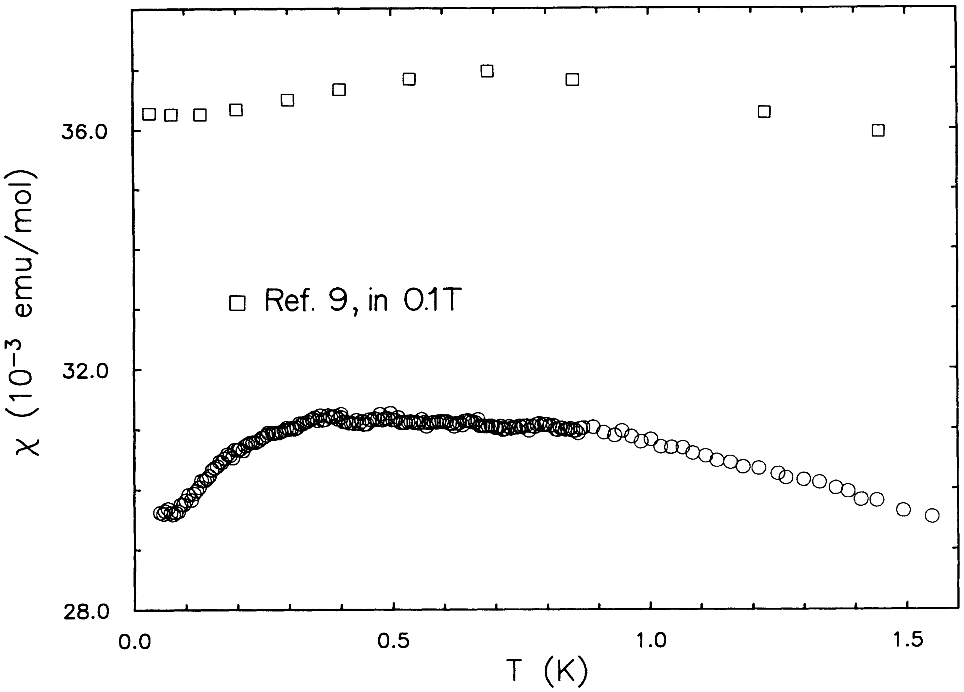

FIG. 3. The low-temperature ac susceptibility is shown from 1.5 K down to 40 mK by the circles. The results of Andres, Graebner, and Ott (Ref. 9) are shown as squares.

data overlap (see Fig. 2). Our results are in agreement with other published results,[9,10,37] including the low-temperature data of Andres *et al.*[9], which are shown in Fig. 3 for comparison.

Below 40 mK, the signal was temperature independent, and to within the aforementioned sensitivity limits, we are reasonably confident that no part of the $CeAl_3$ experienced a superconducting transition or any other kind of detectable magnetic ordering.[38] The absence of a superconducting transition is significant since $CeAl^3$ is similar to other heavy fermion superconductors which possess both antiferromagnetic interactions and superconductivity.[1-8] There is always the possibility that the remnant magnetic field suppressed any potential superconducting state. However, using BCS theory as a rough estimate for the relation between the thermodynamic critical magnetic field at T=0, i.e., $H_c(0)$, and the superconducting transition temperature T_c, we have

$$H_c(0)=T_c\ (1.0\times 10^{-6}\gamma/v_m)^{1/2}\ , \tag{1}$$

where γ is the electronic contribution to the specific heat and v_m is the molar volume. Using $\gamma=1.25$ J/(K^2 mol), $v_m=5.08\times 10^{-5}$ m^3/ mol and $T_c=800$ μK, Eq. (1) gives $H_c(0)\simeq 100$ μT, which is four to five orders of magnitude larger than the residual field present during the experiment. This result suggests that either the BCS weak-coupling theory is not applicable for an order of magnitude estimate (which is unlikely, since it is applicable for other heavy-fermion superconductors[39]), or the potential superconducting state lies at a lower temperature, or the ground state of the system is not superconducting. Finally, there are a variety of possible explanations for the observed experimental results.[7,21,22] These possibilities, which range from a magnetic glassy state to suppressed long-range ordering due to competing interactions, are too numerous to discuss in this paper. It seems clear that progress in answering the open questions will require further experimental work, at the lowest temperatures, on recently available single-crystal specimens.

We gratefully acknowledge useful conversations with S. E. Brown and engineering support from L. Frederick, A. Hingerty, G. Labbe, and the instrument makers in the Department of Physics. We wish to thank William Fogel of NIST for making the tungsten fixed-point sample available to us and Vacuumschmelze GmbH for providing the Cryoperm used in this work. The University of Florida Microkelvin Research Laboratory has been constructed and developed from funds made available by the National Science Foundation through the Instrumentation for Materials Research Program Grant No. DMR-8419267, the University of Florida Division of Sponsored Research and the State of Florida. The experiments conducted in the facility are supported in part by the State of Florida. Support for this research has been provided, in part, by the University of Florida Division of Sponsored Research (Avenel and Ihas), the National Science Foundation (DMR-8615593 and DMR-9019736 for Xia, Xu, Lang, Ni, and Adams; DMR-8902538 for Moyland and Takano; DMR-8902414 and DMR-9022733 for Signore and Meisel), and the Department of Energy (DE-FG05-86ER45268 for Andraka, Jee, and Stewart). One of us (P.J.C.S.) gratefully acknowledges the support of Scientific Instruments.

[*] Permanent address: Service de Physique de l'Etat Condensé, Centre d'Etudes Nucléaires de Saclay, 91191 Gif-sur-Yvette CEDEX, France.

[†] Present address: Department of Science Education, Kang-Weon National University, Chun-Cheon 200-701, Korea.

[‡] Present address: Department of Physics, University of Cincinnati, Cincinnati, OH 45221.

[§] Author to whom correspondence should be addressed; electronic mail address: ufmeisel@nervm.

[**] Also at Universität Augsburg.

[1] G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984).

[2] F. Steglich, Physica B **130**, 145 (1985).

[3] H. R. Ott, in *Progress in Low Temperature Physics,* edited by D. F. Brewer (North-Holland, Amsterdam, 1987), Vol. 11, p. 215.

[4] Z. Fisk, D. W. Hess, C. J. Pethick, D. Pines, J. L. Smith, J. D. Thompson, and J. O. Willis, Science **239**, 33 (1988).

[5] P. Fulde, J. Keller, and G. Zwicknagl, in *Solid State Physics,* edited by H. Ehrenreich and D. Turnbull (Academic, New York, 1988), Vol. 41, p. 1.

[6] L. Taillefer, J. Flouquet, and G. G. Lonzarich, Physica B **169**, 257 (1991).

[7] N. Grewe and F. Steglich, in *Handbook on the Physics and Chemistry of Rare Earths,* edited by K. A. Gschneider, Jr. and L. Eyring (Elsevier, Amsterdam, 1990), Vol. 14, Chap. 97.

[8] C. S. Jee, B. Andraka, J. S. Kim, H. Li, M. W. Meisel, and G. R. Stewart, Phys. Rev. B **42**, 8630 (1990).

[9] K. Andres, J. E. Graebner, and H. R. Ott, Phys. Rev. Lett. **35**, 1779 (1975).

[10] F. R. de Boer, J. Klaasse, J. Aarts, C. D. Bredl, W. Lieke, U. Rauchschwalbe, F. Steglich, R. Felten, U. Umhofer, and G. Weber, J. Magn. Magn. Mater. **47&48**, 60 (1985).

[11] A. Benoit, A. Berton, J. Chaussy, J. Flouquet, J. C. Lasjaunias, J. Odin, J. Palleau, J. Peyrard, and M. Ribault, in *Valence Fluctuations in Solids,* edited by L. M. Falicov, W. Hanke, and M. B. Maple (North-Holland, Amsterdam, 1981), p. 283.

[12] C. D. Bredl, S. Horn, F. Steglich, B. Lüthi, and R. M. Martin, Phys. Rev. Lett. **52**, 1982 (1984).

[13] G. E. Brodale, R. A. Fisher, N. E. Phillips, and J. Flouquet, Phys. Rev. Lett **56**, 390 (1986).

[14] G. E. Brodale, R. A. Fisher, C. M. Lisse, N. E. Phillips, and A. S. Edelstein, J. Magn. Magn. Mater. **54-57**, 416 (1986).

[15] G. E. Brodale, R. A. Fisher, N. E. Phillips, J. Flouquet, and C. Marcenat, J. Magn. Magn. Mater. **54-57**, 419 (1986).

[16] A. Benoit, J. Flouquet, M. Ribault, and M. Chapellier, Solid State Commun. **26**, 319 (1978).

[17] M. Ribault, A. Benoit, J. Flouquet, and J. Palleau, J. Phys. Lett. (Paris) **40**, L-413 (1979).

[18] A. P. Murani, K. Knorr, K. H. J. Buschow, A. Benoit, and J. Flouquet, Solid State Commun. **36**, 523 (1980).

[19] M. Niksch, B. Lüthi, and K. Andres, Phys. Rev. B **22**, 5774 (1980).

[20] M. J. Lysak and D. E. MacLaughlin, Phys. Rev. B **31**, 6963 (1985).

[21] S. Barth, H. R. Ott, F. N. Gygax, B. Hitti, E. Lippelt, A. Schenck, C. Baines, B. van den Brandt, T. Konter, and S. Mango, Phys. Rev. Lett. **59**, 2991 (1987).

[22] D. Jaccard, J. Sierro, J. P. Brison, and J. Flouquet, J. Phys. (Paris) Colloq. **49**, C8-741 (1988).

[23] C. Jin, L. Pollack, E. N. Smith, D. M. Lee, J. T. Markert, and M. B. Maple, Bull. Am. Phys. Soc. **36**, 717 (1991).

[24] B. Andraka, G. Fraunberger, J. S. Kim, C. Quitmann, and G. R. Stewart, Phys. Rev. B **39**, 6420 (1989).

[25] Quantum Design Inc., San Diego, CA.

[26] Biomagnetic Technologies Inc., San Diego, CA.

[27] Ch. Buchal, R. M. Mueller, F. Pobell, M. Kubota, and H. R. Folle, Solid State Commun. **42**, 43 (1982).

[28] H31LV, Epoxy Technology Inc., Billerica, MA.

[29] Sample 13b obtained from the National Institute of Standards and Technology.

[30] P3100, Johnson Matthey Materials Technology, Herts, UK.

[31] Cryoperm 10, Vacuumschmelze GmbH, Hanau, Germany.

[32] K. Kosuge, Y. Oda, and H. Nagano, Cryogenics **20**, 223 (1980).

[33] G. C. Straty and E. D. Adams, Rev. Sci. Instrum. **40**, 1393 (1969).

[34] D. S. Greywall, Phys. Rev. B **33**, 7520 (1986); Jpn. J. Appl. Phys. **26**, Suppl. 26-3, 2082 (1987).

[35] J. H. Colwell, W. E. Fogle, and R. J. Soulen, Jr., in *Proceedings of the 17th International Conference on Low Temperature Physics,* edited by U. Eckern, A. Schmid, W. Weber, and H. Wühl (North-Holland, Amsterdam, 1984), p. 395.

[36] W. C. Black, R. T. Johnson, and J. C. Wheatley, J. Low Temp. Phys. **1**, 641 (1969).

[37] K. H. Mader and W. M. Swift, J. Phys. Chem. Solids **29**, 1759 (1968).

[38] While it is true that our tungsten fixed-point sample obscures any possible other signal near 15 mK, other researchers (Refs. 9–22) have seen no anomalies in this temperature region.

[39] U. Rauchschwalbe, U. Ahlheim, F. Steglich, D. Rainer, and J. J. M. Franse, Z. Phys. B **60**, 379 (1985).