Anisotropic spin fluctuations in cubic $CeSn_3$

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(Received 13 December 1983)

The low-temperature high-field heat capacity of a $CeSn_3$ single crystal was found to develop an anisotropy for $H > 2.50$ T with the magnetic field parallel to the [100] and [110] directions. The magnetic susceptibility of $CeSn_3$ below 5 K was also anisotropic with $\chi_{110} > \chi_{100}$ by $\sim 5\%$ at 1.8 K. For the first time ever an anisotropic quenching of spin fluctuations in any highly enhanced paramagnet has been observed.

Recently the low-temperature heat capacity (1–20 K) of polycrystalline $CeSn_3$ was measured in magnetic fields up to 10 T.[1] The authors found that the electronic specific-heat constant γ was depressed with increasing magnetic field while β, the coefficient of the T^3 term in the temperature dependence of the heat capacity, was raised. The former was attributed to the quenching of spin fluctuations, while the latter was thought to be due to the development of an induced magnetic moment on the Ce atoms by the applied field (metamagnetism). An earlier investigation of $CeSn_3$ by polarized neutrons[2] indicated the development of an anisotropy in the form factor of $CeSn_3$ at a field of 4.3 T.

As a result of these two studies, we raised the question whether or not the quenching of spin fluctuations by large magnetic fields and the development of an anisotropic form factor in $CeSn_3$ were related. For this reason we undertook a study of the heat capacity as a function of magnetic field and the magnetic susceptibility of a $CeSn_3$ single crystal aligned in two orientations with the magnetic field parallel to the [100] and [110] directions.

The sample was first prepared by arc melting 99.9 at.% pure Ce and 99.99 at.% pure Sn. The arc-melted button was then placed in a tungsten crucible to grow a single crystal by the Bridgeman technique. The sample used in the magnetic-susceptibility measurement was cut from a section of the $CeSn_3$ single crystal adjacent to the sample used in the heat-capacity measurement. The single crystals were cut such that after the alignment of either a [100] or a [110] direction parallel to the magnetic field the second orientation is obtained by a simple 90° rotation. *Thus the same large single crystal (8.73 g) was used for the heat-capacity measurements in the two directions, and a second small single crystal (0.26 g) was used for the magnetic-susceptibility measurements for the two orientations.* This procedure was used to ensure that if any difference was observed in one measurement it was not due to the use of two different crystals (one of each orientation), even though the two crystals may have been cut from the same larger single crystal. The experimental details regarding the low-temperature calorimetry have been described previously.[3] The magnetic-susceptibility measurements were made from 1.7 to 300 K by using the Faraday technique at 1.83 T except for the field-dependence measurements.[4]

The low-temperature heat capacity for the two orientations of the single crystal at all five fields exhibited the same general behavior as we had observed for our polycrystalline sample,[1] except that the rapid upswing observed in the C/T vs T^2 curves for the polycrystalline sample below 1.9 K was absent in the single crystals. As indicated in our earlier paper the upswing was attributed to iron impurities (160 to 185 ppm atomic) in the polycrystalline sample. Since the single crystal had a low iron content (6 ppm atomic) the absence of the upswing is easily accounted for. As observed earlier the zero-field and 2.50-T heat-capacity curves clearly exhibited a $T^3 \ln T$ term in addition to the linear electronic and cubic lattice terms. But at higher fields the $T^3 \ln T$ term no longer was observed, and these data could be fitted to the usual linear C/T vs T^2 curve, i.e.,

$$C/T = \gamma + \beta T^2 \ . \tag{1}$$

The low-field data ($H \leq 2.50$ T) were fitted to the equation

$$C/T = A + BT^2 + DT^2 \ln T \ , \tag{2}$$

where $A = \gamma$, $B = \beta - (\alpha\gamma_0/T_s^2)\ln T_s$, $D = \alpha\gamma_0/T_s^2$, T_s is the spin-fluctuation temperature, α is proportional to $S(1-S^{-1})^2$ with S being the Stoner enhancement factor, and γ_0 is the electronic specific-heat constant determined from band-structure density of states. Equation (1) gives the electronic and lattice contribution to heat capacity, while Eq. (2) gives the low-temperature heat capacity for a nearly ferromagnetic metal[5] well below T_s.

The field dependence of γ and β are shown in Fig. 1, where it is seen that below 5 T the γ and β values are identical when the applied fields are oriented parallel to the [100] and [110] directions. At ~ 5 T an anisotropy begins to develop and becomes quite evident above 7 T. Thus the quenching of spin fluctuations (i.e., the lowering of γ)[1] is 6.2% less in the [100] direction than in the [110] direction, while the strength of the induced magnetic moment (i.e., the increase in β)[1] is 36% larger in the [110] direction than in the [100] direction. The average change in β_{100} and β_{110} due to an applied field of 10 T is about the same as observed in the polycrystalline sample, but the lowering of γ in the polycrystalline material (-26.9%) is significantly larger than the change observed in γ_{100} (-15.4%) and in γ_{110} (-21.1%) when the applied field is increased from 0 to 10 T (see Table I).

The field dependence of the magnetic susceptibility is shown in Fig. 2. For temperatures ≥ 4.2 K, the susceptibility shows no field dependence. But at 1.77 K the susceptibility increases as the field is lowered from 1.8 to 0.9 T, indicating the presence of magnetic impurities. We believe the magnetic impurities are disordered Ce atoms, i.e., face-centered Ce atoms which have interchanged positions with the corner Sn atoms of the cubic $AuCu_3$-type structure giving rise to a tetrad of Ce atoms in which the face-centered Ce atom is in contact with the three remaining Ce corner

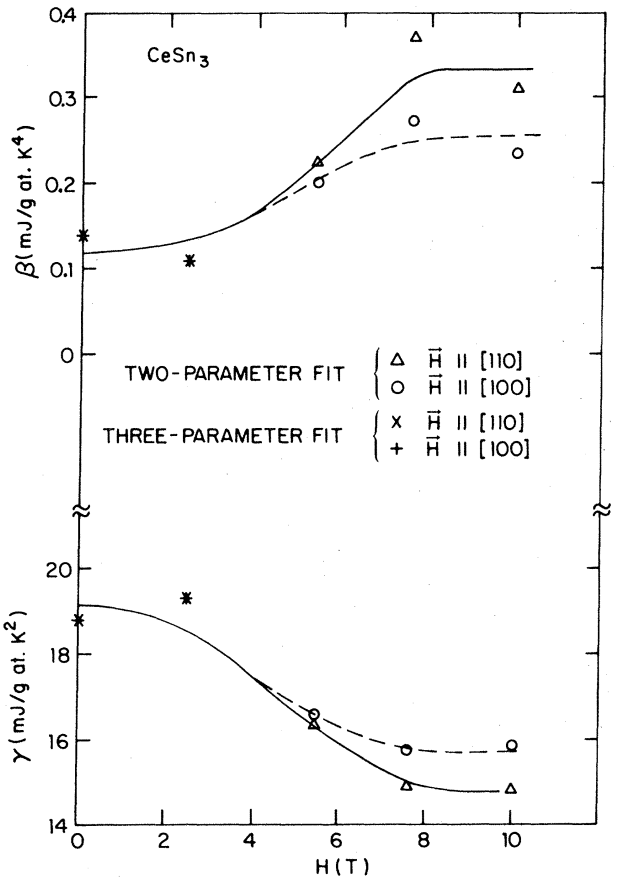

FIG. 1. Magnetic field dependence of the electronic specific-heat constant γ and the slope of the standard C/T vs T^2 plot, β.

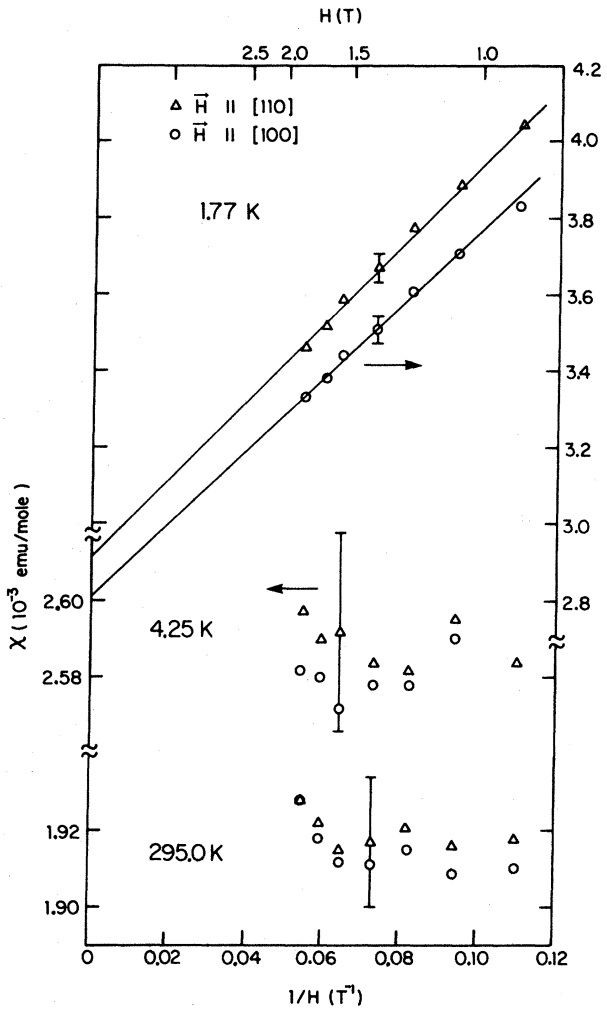

FIG. 2. Field dependence of susceptibility at 1.77, 4.2, and 295 K. Since $\chi_{obs}(H)=\chi_{CeSn_3}+\Delta\chi_{imp}(H)$, the slope is proportional to the contribution to χ from impurity. Note the change in scale for the 1.77-K data relative to that for the 4.25- and 295.0-K results. The error bars indicate a 1% uncertainty.

atoms (see Fig. 3). Following the method of Honda and Owen,[7] the true susceptibility and the susceptibility due to magnetic impurities are extracted. The impurity susceptibility is given by $\Delta\chi_{imp}=c\sigma_{sat}/H$, where σ_{sat} is the saturation magnetization of the impurity at the temperature of the susceptibility measurement and c is the impurity concentration.

TABLE I. Summary of some of the important heat-capacity and magnetic-susceptibility parameters.

Property	Single crystals H_{100}	Single crystals H_{110}	Polycrystalline[a]
γ $(H=0)$, mJ/g-at. K^2	18.75 ±0.03	18.75 ±0.03	18.15 ±0.04
β $(H=0)$, mJ/g-at. K^4	0.141[b]	0.141[b]	0.141[b]
Θ_D $(H=0)$, K	240[b]	240[b]	240[b]
T_s, K	5.6	5.6	5.8
γ $(H=10$ T), mJ/g-at. K^2	15.86 ±0.04	14.79 ±0.04	13.16 ±0.03
β $(H=10$ T), mJ/g-at. K^4	0.232 ±0.008	0.310 ±0.005	0.300 ±0.001
$\Delta\chi_{imp}$ at 1 T, emu/mole	0.96×10^{-3}	1.02×10^{-3}	· · ·

[a]Reference 1.
[b]The Debye temperature was assumed to be the same as that given by Stassis *et al.* from neutron scattering experiments (Ref. 6), and the β value was calculated from this Θ_D value.

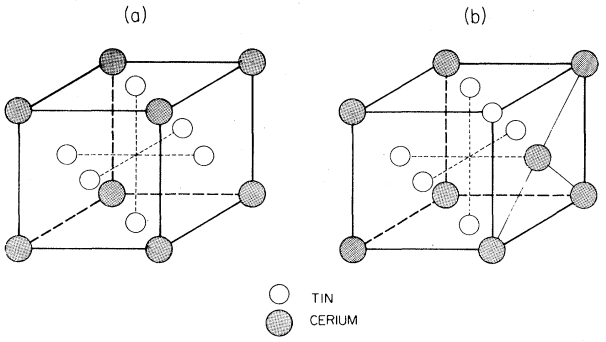

FIG. 3. Perfectly ordered $CeSn_3$ structure (a) and the $CeSn_3$ structure in which a corner Ce atom has interchanged positions with a face-centered Sn atom giving rise to a Ce tetrad (b).

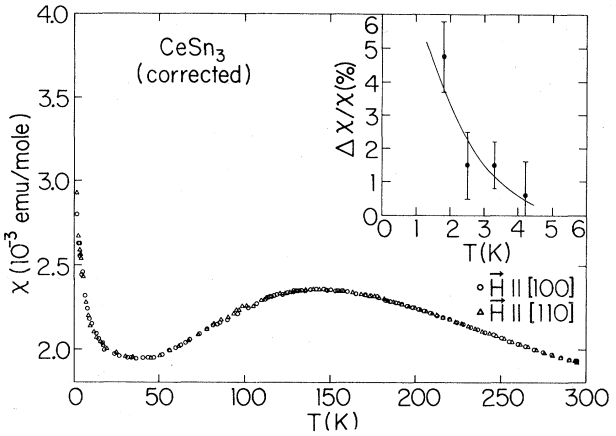

FIG. 4. Magnetic susceptibility of $CeSn_3$ from 1.7 to 295 K. Insert shows the magnetic anisotropy.

Taking the impurity to be Ce with an effective moment of $2.54\mu_B$, the concentration of impurity is estimated to be 7.12×10^{-4} Ce atoms/mole, which is equivalent to about 60 Ce-Sn interchanges per 10^5 atoms.[8] As is evident in Table I the correction to the magnetic susceptibility is $\sim 6\%$ larger in the [110] direction than in the [100] direction. The average value of $\Delta\chi_{imp}$ was used to calculate the number of Ce-Sn interchanges.

The magnetic susceptibility of a $CeSn_3$ single crystal from 1 to 300 K is shown in Fig. 4, where the contribution due to magnetic impurities has been corrected for, and is similar to that reported previously. The magnetic susceptibility is isotropic within $\pm 0.5\%$ down to ~ 5 K. Below 5 K, where spin fluctuations are important, an anisotropy developed with the susceptibility $\sim 5\%$ larger in the [110] direction than in the [100] at 1.7 K.

The origin of the anisotropy in the quenching of spin fluctuations and magnetic susceptibility is not well understood. It could be due to the spin-orbit interaction which causes the band structure to shift as the spins try to follow the field direction and "drag" along some of the orbital moment. Such an effect was used to explain field-induced changes observed in the band structure and Fermi surface of nickel.[9] A second possibility is that the Fermi energy lies in a region in which the density of states varies extremely rapidly with energy, and when a magnetic field is applied the Zeeman splitting of the spin-up and spin-down bands gives rise to different density-of-states values for the up and down bands. In both of these cases the magnetization is nonlinear, so that $M=\chi_1 H+\chi_2 H^3+\cdots$, and the higher-order terms can give rise to an anisotropy through a directional dependence of χ_2.

There is also a third possibility, which is due to the disordered Ce atoms. As is seen in Fig. 3, the only possible direct Ce-Ce contacts are along the face diagonals, i.e., the [110] directions. The Ce-Ce distances in the [110] directions are $0.707a$, while those in the [100] direction are a, where a is the lattice parameter. Thus one would expect to see an enhancement in the magnetic susceptibility in the [110] direction, and likewise a larger depression of the spin fluctuations and a stronger-induced moment in the same direction, which is observed. This, however, could not explain the anisotropy observed in the neutron form-factor measurements, since these are made using coherent scattering, while the tetrads would be expected to occur randomly in the crystals. Furthermore, if the tetrad developed an ordered arrangement the concentration of these would be much too small to be detected by neutron scattering. It is, however, possible that the Ce tetrads could account for the anisotropy in γ and β because the concentration levels are only ~ 1.7 times lower than the 1 per 1000 calculated in our earlier paper from the magnetic entropy.[1] Also, the fact that the impurity correction for the magnetic susceptibility, $\Delta\chi_{imp}$, is larger in the [110] direction (see Table I) would also tend to favor the tetrad explanation rather than the first two arguments based on the band structure.

Regardless of which model should eventually prove correct, we have shown that the quenching of spin fluctuations and related metamagnetic behavior in a highly enhanced, nearly ferromagnetic paramagnet exhibits an anisotropy even in a cubic material. Other experiments, such as high-field magnetic-susceptibility measurements and additional measurements on samples which have been heat treated to introduce different amounts of disorder or perfection in them, should help elucidate our understanding of this material and the nature of spin fluctuations.

ACKNOWLEDGMENTS

The authors wish to thank B. N. Harmon, A. R. Mackintosh, and C. Stassis for their helpful comments and discussions. The Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-ENG-82. This work was supported by the Office of Basic Energy Sciences.

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[8]If one assumes that $\Delta\chi_{imp}$ is due to Fe impurities, then one calculates an impurity content of about 200 ppm atomic Fe, which is ~ 30 times larger than that measured for our sample. Furthermore, the absence of an upswing below 1.9 K in the heat capacity also indicated that the Fe content must be significantly below this concentration; see results published in Ref. 1 for a $CeSn_3$ sample containing 160 to 180 ppm Fe.
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