Quenching of spin fluctuations by high magnetic fields in the heat capacity of $CeSn_3$

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The low-temperature (1.3–20.0-K) heat capacity of the mixed-valent compound $CeSn_3$ was measured in magnetic fields up to ~10 T. The electronic specific heat constant γ decreases with increasing fields and at 9.98 T reaches $\sim\frac{3}{4}$ of the zero-field value. The nearly constant value of γ for $H > 7.5$ T suggests that the spin fluctuations are probably completely quenched by 10 T. The $T^3 \ln T$ term of the heat capacity observed in zero field disappears in fields > 2.5 T. Furthermore, the applied magnetic field induces a magnetic moment on the Ce atoms, and this leads to a magnetic contribution to the heat capacity which has a T^3 temperature dependence. Analysis of the heat-capacity data of $CeSn_3$ yields the many-body enhancement factor due to paramagnons (λ_{spin}) as 0.65, the Stoner enhancement factor (S) as 4.23, the spin-fluctuation temperature (T_s) as 5.8 K, and the effective field sufficient to quench spin fluctuations (H_{eff}) as 8.6 T. The field dependence of the electronic contribution to the heat capacity is compared with that predicted by two theoretical models and the agreement is fair in one case and poor in the second case.

I. INTRODUCTION

The effect of spin fluctuation in weakly and nearly ferromagnetic metals has been of considerable interest theoretically for over a decade because it is connected with the origin of ferromagnetism in metals.[1] It was pointed out by Brinkman and Engelsberg,[2] by Béal-Monod *et al.*,[3] and more recently by Hertel *et al.*[4] that the application of high magnetic fields offers one way of testing the spin fluctuation theory. Brinkman and Engelsberg[2] explained that high magnetic fields of the order of characteristic spin fluctuation temperature, $T_s \equiv T_F/S$, are required to quench the spin fluctuation enhancements. Here, T_F and S are, respectively, the Fermi temperature and the Stoner exchange-enhancement factor. On the other hand, Béal-Monod *et al.*[3] showed that the decrease in the heat capacity at 0 K of a nearly ferromagnetic Fermi liquid is proportional to H^2.

If the magnetic field is sufficiently large so that the Zeeman splitting energy of opposite spin states is comparable to or larger than the characteristic spin fluctuation energy, then the paramagnons no longer have enough energy to flip the spins and, therefore, the inelastic spin-flip scattering is quenched. Hence a decrease of the specific-heat enhancement with increasing magnetic field is to be expected. Hertel *et al.*[4] who have made a more detailed mathematical analysis, suggest that if the Stoner enhancement and the mass enhancement due to spin fluctuation are large (~4 and ~1.5, respectively) and if the spin fluctuation temperature T_s is small (~15 K) a depression of the electronic contribution to the heat capacity of a few percent might be expected at 10 T. They have suggested an f-band metal such as UAl_2 might have the three parameters of the appropriate magnitude to see such an effect. Trainor *et al.*[5] have measured the low-temperature heat capacity for the nearly ferromagnetic UAl_2 compound at 0, 1.0, and 4.3 T. Their data indicate a slight decrease (1.5%) in the heat capacity at the highest field, but considering the experimental error one cannot conclusively conclude that there was such a shift.

Recently, the quenching of spin fluctuations, which was theoretically predicted,[2–4] has been found in the low-temperature heat-capacity measurements at high magnetic fields up to ~10 T for the strongly Pauli paramagnetic $LuCo_2$ (Ref. 6) and weakly ferromagnetic Sc_3In (Ref. 7) compounds. The results for $LuCo_2$ show that the electronic specific-heat constant decreases with increasing magnetic fields (11% at 9.98 T), while the Debye temperature does not change.[6] On the other hand, the heat-capacity peak observed around $T_c = 6.0$ K in zero field for Sc_3In becomes smaller with increasing fields and at 9.98 T its magnetic entropy is ~18% of the zero-field value.[7] These anomalous behaviors in $LuCo_2$ and Sc_3In are thought to be due to the depression of spin fluctuation contribution to the heat capacity by the high magnetic fields.

Because of the observed quenching of spin fluctuations observed in a strongly Pauli paramagnet and a weak ferromagnet it was felt that a similar effect might be observed in a material which exhibits valence fluctuations. The intermetallic compound $CeSn_3$ was chosen for such a study because valence

(or spin) fluctuations are thought to account for the enhanced magnetic susceptibility and electronic specific-heat constant, and a T^2 term in the resistivity.[8,9] $CeSn_3$ has the fcc Cu_3Au-type structure and its magnetic susceptibility exhibits a Curie-Weiss behavior between 250 and 800 K, a broad maximum at ~ 135 K and a relatively rapid rise below 30 K.[8,10–14] In order to explain the deviation from Curie-Weiss behavior below 250 K, Tsuchida and Wallace[11] assumed a gradual transition of cerium from the magnetic trivalent state to the nonmagnetic tetravalent state at low temperatures.

This interpretation of the temperature dependence of susceptibility has obtained further support in the NMR measurements by Borsa *et al.*[15] and by Malik *et al.*,[8,16,17] and the anomalous thermal expansion measurements reported by Harris and Raynor.[18] The ^{119}Sn Knight shift has a broad maximum around 115 K in agreement with the susceptibility but the NMR does not show a rapid rise below 30 K. The ^{119}Sn Knight-shift measurements down to 1.2 K[8,16,17] suggest that $CeSn_3$ does not order ferromagnetically down to this temperature. In a polarized neutron scattering study for $CeSn_3$ at a field of 4.23 T between 4.2 and 300 K, Stassis *et al.*[19] have found that below 40 K a magnetic moment in the vicinity of the Ce sites contains both a $5d$ and $4f$ component. They also note that the latter is not connected with the rapid rise of the bulk susceptibility below this temperature and this probably accounts for the lack of proportionality between the magnetic susceptibility and the Knight shift.

Recently, detailed measurements of magnetic susceptibility for $CeSn_3$ were made by Lawrence[13] and by Dijkman *et al.*[14] Lawrence and Murphy[13,20] have measured the susceptibility for $CeIn_{3-x}Sn_x$ $(0 \leq x < 3)$ and found in the intermediate-valence regime of $0.8 \leq x \leq 3.0$, $T_s(x)$ oscillates with x in proportion to the conduction electron density of states. On the other hand, Dijkman *et al.*[14] have measured the susceptibility between 1.6 and 1400 K and the magnetization at 4.2 K in fields up to 34 T for $Ce_{1-x}La_xSn_3 (0 \leq x \leq 1)$ and found that the intermediate valence state of Ce is preserved throughout the series, with the characteristic temperature unaffected by La substitution. Both authors[13,14] believe that the sharp increase below 30 K in the susceptibility of $CeSn_3$ is due to impurities because it can be suppressed to a large extent by preparing samples with an excess amount of tin.[14] Therefore, they[13,14] conclude that the anomaly of the χ-T curve at low temperatures for $CeSn_3$ is not intrinsic and may be due to the presence of magnetic Ce atoms which are stabilized in the Ce^{3+} state by neighboring lattice defects or local atomic disorder. This observation may also account for the differences found in the magnetic form factors for the two $CeSn_3$ samples studied by Stassis *et al.*[19]

Cooper *et al.*[12] performed measurements of the low-temperature heat-capacity, electrical resistivity and thermoelectric power. They,[12] and also Malik *et al.*[8] found an increase in the C/T vs T^2 plot with decreasing temperatures below $T \simeq 3$ K ($T^2 \simeq 10$ K^2) and that the electronic specific-heat constant γ is large ($\gamma \simeq 13.2$ mJ/g at. K^2). According to Malik *et al.*[17] and Stalinski *et al.*[21] the electrical resistivity goes as T^2 at low temperatures and increases continuously up to 300 K with a negative curvature above 150 K. In contrast Maury *et al.*[22] found a resistance minimum at ~ 40 K, which the authors thought could be a Kondo minimum, but they also stated that their results do not necessarily rule out that $CeSn_3$ is an intermediate valence system. Moreover, the thermoelectric power is giant and positive with a maximum around 220 K.[12] These experimental results show that $CeSn_3$ exhibits mixed valent properties, and that a change in the nature of the f states occurs from the bandlike states ($T = 4.2$ K) to the localized states ($T \geq 40$ K).[23,24] At low temperatures of $T < 40$ K, $CeSn_3$ is the typical strongly Pauli paramagnetic material, where the ground-state properties are successfully described by the exchange enhanced Fermi liquid model.[23,25–29] Therefore, this compound seems to be one of the better materials to see if the persistent spin fluctuations in strong Pauli paramagnets can be quenched by large magnetic fields.

II. EXPERIMENTAL

The measurements were performed on a large grain polycrystalline sample (~ 8 g) prepared in this laboratory. The sample was first prepared by arc melting 99.9 at.% pure Ce and 99.99 at.% pure Sn. The arc-melted button was placed in a tungsten crucible in an attempt to grow a single crystal by the Bridgman technique. Although this method was successful in growing single crystals for neutron scattering[30,31] and de Haas–van Alphen[32] studies, this particular run only yielded several large grains of $CeSn_3$, but this sample was suitable for the heat-capacity studies. After cooling no evidence was found for any appreciable oxidation or reaction of the sample with the tungsten crucible.

The low-temperature calorimeter used in this work is of the usual isolation heat-pulse type with a mechanical heat switch. The temperature was measured by using a germanium resistance thermometer which had been calibrated at magnetic fields of 0, 2.50, 5.39, 7.62, and 9.98 T.[33] The heat capacity of the 1965 Calorimetry Conference standard[34] copper sample was measured to serve as a check of the apparatus and experimental technique. Accordingly, the low-temperature heat-capacity results obtained on our copper standard sample were in agreement with

the previous zero-field data[34] within ±1% at 0 T and ±2% at the four nonzero magnetic fields.

III. RESULTS

The heat-capacity measurements on $CeSn_3$ were made between 1.3 and 20.0 K at magnetic fields of 0, 2.50, 5.39, 7.62, and 9.98 T. Figure 1 shows the experimental results between 1.3 and 7.7 K. The zero-field heat capacity is nearly the same (within a few percent) as the previous results[8,12] up to about 3 K. Above this temperature our data are systematically lower than the other published results, by ~14% from Malik *et al.*[8] and by ~20% from Cooper *et al.*[12] at ~7.5 K (56 K^2) which was near the upper end of their measurements. Below ~4 K (16 K^2) the zero-field heat capacity exhibits a gentle upturn which becomes nearly flat. With increasing magnetic fields this gentle upturn in the C/T vs T^2 curves becomes smaller, and above $H=2.50$ T the C/T vs T^2 curve becomes linear with the exception of a sharp upturn below ~1.8 K.

This sharp upturn below ~1.8 K becomes more pronounced with increasing magnetic fields and is probably due to the superparamagnetic behavior of magnetic impurities, such as iron. However, the possibility of cerium atom clusters (e.g., due to disordered cerium atoms occupying tin sites) or local lattice defects which stabilize the Ce^{3+} ion (as suggested by Lawrence[13] and Dijkman *et al.*[14] to explain the low-temperature susceptibility anomaly) might also account for such a behavior. Because of this we analyzed our $CeSn_3$ sample for iron impurities, and found that it contained between 160 and 186 at. ppm Fe. The iron was probably introduced into the sample during arc melting and it most likely accounts for the sharp upturn below 1.8 K. Based on our experience with other materials doped with iron, we believe the iron impurities have no effect on the heat capacity above 1.8 K.

The electronic specific-heat constant γ and the Debye temperature at 0 K, Θ_D, were calculated from a least-squares fit of the heat-capacity data for each field between 2 and 5 K to the equation

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

The results of a least-square fit to Eq. (1) are given in Table I. For the heat-capacity data at 5.39, 7.62, and 9.98 T, this equation is probably of the correct form, but it may not be for the 0 and 2.50 T field data. We will return to this point shortly. The solid line in Fig. 1 is the result of a least-square fit for the data at 9.98 T.

Figure 2 shows the results of heat-capacity measurements up to 20.0 K at 0 and 9.98 T. As seen in this figure, the largest difference between the two C/T vs T^2 curves, besides that observed at the lowest temperature, occurs at ~15 K(225 K^2), and above ~18 K the curves merge. This suggests that the paramagnon contribution to the heat capacity of strongly Pauli-paramagnetic compound $CeSn_3$ predominates at low temperatures and the enhancement due to the spin fluctuations is suppressed be-

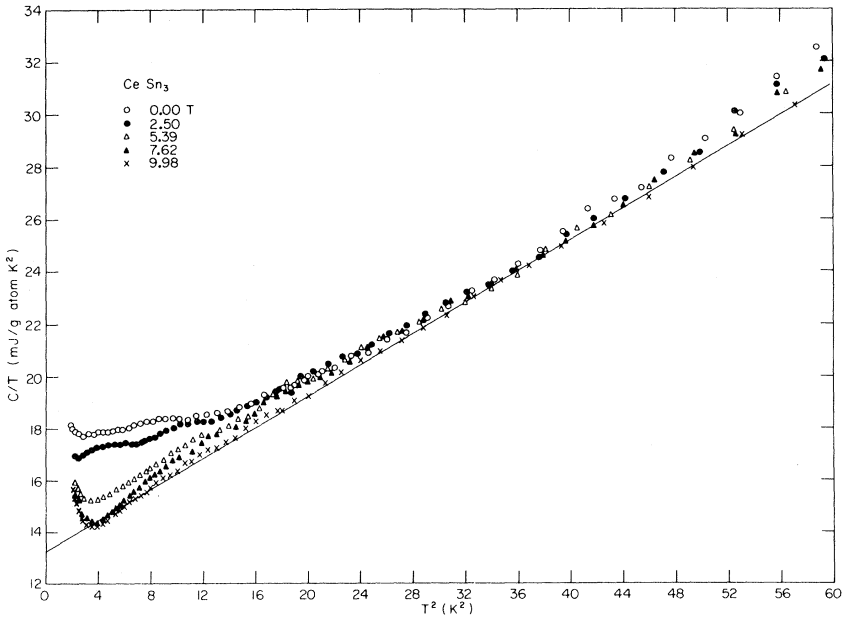

FIG. 1. The heat capacity of $CeSn_3$ from 1.3 to 7.7 K at five magnetic fields: 0, 2.50, 5.39, 7.62, and 9.98 T. The solid line is the result of a least-square fit of the 9.98-T data to Eq. (1).

TABLE I. The results of a least-squares fitting of the heat-capacity data of $CeSn_3$ at five magnetic fields to the equation $C/T=\gamma+\beta T^2$ and a least-square fitting of the 0 and 2.50 T results to the equation $C/T=A+BT^2+DT^2\ln T$.

H (T)	γ (mJ/g at. K^2)	β (mJ/g at. K^4)	Apparent Θ_D (K)
0.00	17.35±0.04	0.122±0.003	252±2
2.50	16.40±0.06	0.168±0.004	226±2
5.39	14.16±0.03	0.283±0.001	190±1
7.62	13.58±0.06	0.302±0.002	186±1
9.98	13.26±0.03	0.300±0.001	186±1

H (T)	A (mJ/g at. K^2)	B (mJ/g at. K^4)	D (mJ/g at. K^4 lnK)
0.00	18.15±0.04	−0.230±0.014	0.211±0.008
2.50	17.33±0.09	−0.200±0.030	0.222±0.017

cause of the renormalization of spin fluctuation as the temperature increases.[35,36]

For a nearly ferromagnetic metal, theory[2,25,26,35] predicts that the low-temperature heat capacity well below T_s is given by

$$C=\gamma_0 T[m^*/m+\alpha(T/T_s)^2\ln(T/T_s)]+\beta T^3 \quad . \qquad (2)$$

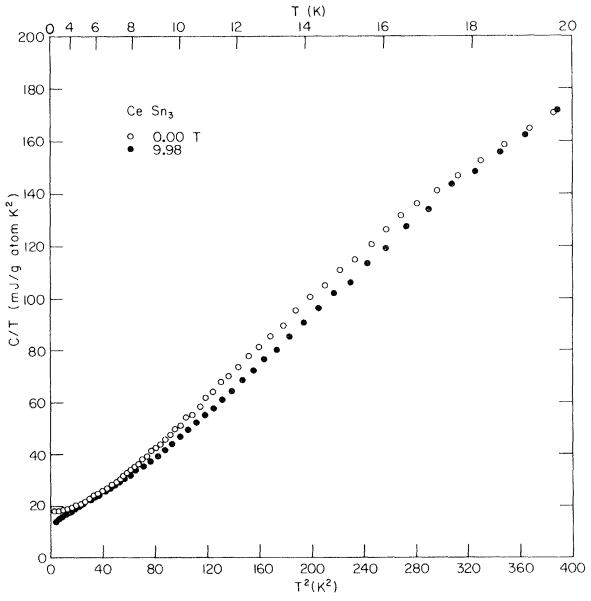

FIG. 2. The heat capacity of $CeSn_3$ from 1.3 to 20.0 K at 0 and 9.98 T.

Here, m^*/m is the zero temperature many-body mass enhancement, which includes spin-fluctuation and electron-phonon contributions, γ_0 is the electronic specific-heat constant determined from the band-structure density of states, α is proportional to $S(1-S^{-1})^2$ (where S is the Stoner enhancement factor) and βT^3 is the usual lattice contribution. As shown by Lederer and Mills[37] and by Engelsberg *et al.*[38] Eq. (2) is valid only for the case of a uniform enhancement throughout the lattice, but if one takes into account local enhancement on one atom (the magnetically dilute solute atom) then the $T^3\ln T$ term is negligible except for $T<T_s/100$ and the next leading term is small and has a T^3 dependence.

Because of the unusual shape of the C/T vs T^2 curves at 0 and 2.50 T below ~5 K, the heat-capacity data for these two fields were analyzed by using the following relationship:

$$C/T=A+BT^2+DT^2\ln T \quad . \qquad (3)$$

Comparing Eqs. (2) and (3) we find that $A=(m^*/m)\gamma_0=\gamma$, $B=\beta-(\alpha\gamma_0/T_s^2)\ln T_s$, and $D=\alpha\gamma_0/T_s^2$. Table I also shows the results of a least-square fit of the heat-capacity data at 0 and 2.50 T between 1.3 and 5.0 K to Eq. (3). Figure 3 shows the measured heat capacity of $CeSn_3$ below 6 K, where the solid lines for the data at 0 and 2.50 T, and at 5.39, 7.62, and 9.98 T are the results of a least-square fit to Eq. (3) and to Eq. (1), respectively. As seen in this figure, the $T^3\ln(T/T_s)$ correction term is not needed to describe the heat capacity of $CeSn_3$ at magnetic fields greater than 2.50 T.

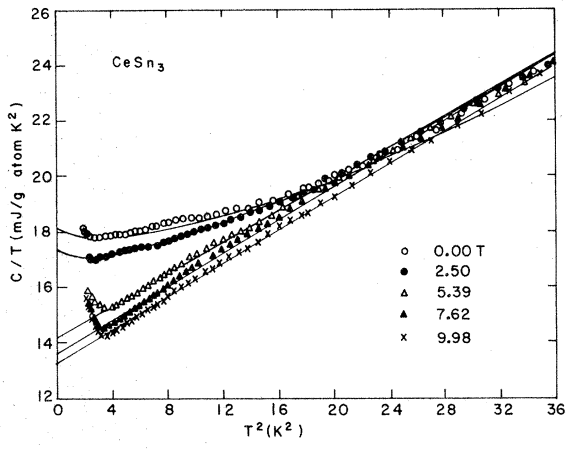

FIG. 3. The heat capacity of $CeSn_3$ at five magnetic fields. The solid lines for the data at 0 and 2.50 T, and at 5.39, 7.62, and 9.98 T are the results of a least-squares fit to Eq. (3) and to Eq. (1), respectively.

The resultant γ values from the fit of the experimental data to the two equations [(1) and (3)] are nearly the same, they differ by $\sim 5\%$ for both $H=0$ and 2.50 T measurements, see Table I and Fig. 4. This suggests that the resultant parameters γ are not too sensitive to the choice of the fitting equation. It is our belief, however, that the γ values obtained from Eq. (3) are the more reliable ones. On the other hand the rapid drop in the "Debye temperature" with increasing field especially between 0 and 2.5 T might suggest that Eq. (1) probably is not valid. But

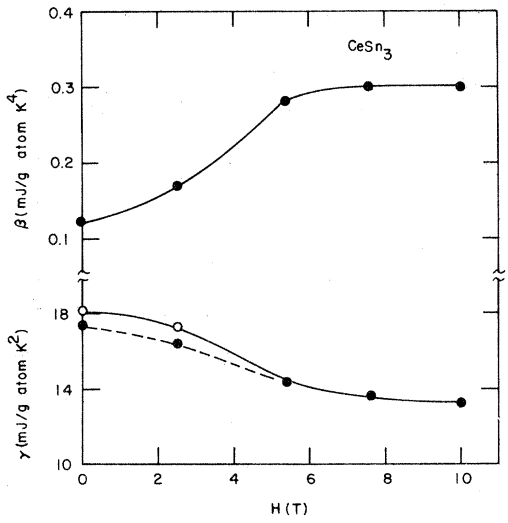

FIG. 4. The electronic specific-heat constant γ and the coefficient of the T^3 term β for $CeSn_3$ as a function of magnetic field from zero to 9.98 T. The solid and open circles were obtained from a least-square fit of the heat-capacity data at each field to Eq. (1) and to Eq. (3), respectively.

the Debye temperature calculated from the phonon spectrum as obtained from inelastic neutron scattering experiments (240 K at $T=0$ K and $H=0$ T)[31] is slightly less than (within 5% of) our heat-capacity value at zero field. This good agreement gives us confidence that the Θ_D value at $H=0$ T listed in Table I and that the listed β values, which we also show in Fig. 4, are close to the "true value," within $\sim 5\%$. More will be said shortly about the variation of β with the applied field.

As shown in Fig. 4 both the γ and β values rapidly change at low applied fields, then more slowly with increasing magnetic field for $H \geq 5$ T, and appear to be leveling off at ~ 9 and ~ 6 T, respectively. At 9.98 T the γ value dropped by 27% (the open circle at $H=0$ T) and β value increased by 146% from the zero-field values.

A comparison of our zero-field data with the previous results obtained on $CeSn_3$ and $LaSn_3$ is shown in Table II. At first glance it would appear that there is a wide discrepancy between the previously reported heat-capacity results for $CeSn_3$ and our recent measurements. Most of this discrepancy is due to the fact that there is a gentle curvature in the C/T vs T^2 plot and the resultant γ and Θ_D values greatly depend upon the temperature range over which the data are considered to have a linear dependence. The reason for this is obvious upon examination of the temperature dependence of Θ_D as given by Stassis *et al.*[31] who show that Θ_D drops from 240 K at $T=0$ K to ~ 200 K at 5 K and 150 K at ~ 14 K. Such a temperature variation is most unusual. For example, if the data of Malik *et al.*[8] are used over the same range used to fit our results, then the γ and Θ_D values would be about 5% smaller than ours. The data of Cooper *et al.*[12] cannot be analyzed in this range because there are only four points and considerable scatter, but as noted earlier there is good agreement between the three heat-capacity measurements below 3 K.

Furthermore, the analysis of Stassis *et al.*[31] adds credence to the above conclusion. These authors estimated a γ value, which is less than 4% smaller than our value, by subtracting off the lattice contribution to the heat capacity (as calculated from the Θ_D value obtained from their inelastic neutron scattering results) from the heat-capacity results of Cooper *et al.* and Malik *et al.*

The low-temperature heat capacity of the isostructural compound $LaSn_3$ has been measured by Umlauf *et al.*[39] in magnetic fields at 0, 0.5, and 5 T. The 0.5 T results are listed in Table II, where one notes the nearly identical values for the Debye temperatures of $LaSn_3$ and $CeSn_3$ (compared Stassis *et al.*'s value, which is the most reliable Θ_D for $CeSn_3$) and the sixfold difference in γ, which is not unusual when Ce is in a mixed valence state. Furthermore, Umlauf *et al.* found that the normal-state heat capacities at

TABLE II. Comparison of electronic specific-heat constants and Debye temperature of $CeSn_3$ at $H=0$ T and $LaSn_3$ at 0.5 T.

Compd.	Authors	γ (mJ/g at. K^2)	Θ_D (K)	Temp. range of fit (K)
$CeSn_3$	Cooper *et al.* (Ref. 12)	13.2±2.5	161[a]	2 –9.5
	Malik *et al.* (Ref. 8)	14.8[a]	179[a]	2.7 –6.5
	This study	18.2	252	2 –4.5
	Stassis *et al.* (Ref. 31)	17.5[b]	240[c]	. . .
$LaSn_3$	Umlauf *et al.* (Ref. 39)	3.0±0.1	235±4[d]	0.5 –10

[a]Data estimated from a C/T vs T^2 plot given by the respective authors using the straight line *they* had drawn through their data points.
[b]Estimated by subtracting the lattice contribution (based on the Θ_D value obtained from neutron scattering results) from the heat-capacity results of Malik *et al.* and Cooper *et al.*
[c]Calculated from phonon spectrum which was obtained from inelastic neutron scattering measurements.
[d]Erroneously reported as 214 K.

0.5 and 5 T are nearly the same from 0.5 to 10 K, except that the latter becomes slightly larger below ~3 K, which the authors thought was due to the presence of magnetic lanthanide impurities. The absence of any change in γ (or possibly a slight increase) for $LaSn_3$, which contrasts with the 27% decrease in $CeSn_3$, supports our views that the decrement of the low-temperature heat capacity with increasing field (Fig. 3 and Table I) is probably due to the depression of the spin fluctuation enhancement to the heat capacity by magnetic fields.

IV. DISCUSSION

In this section of our paper the field dependence of β is examined, and then the heat-capacity results are analyzed to derive several useful parameters, i.e., the spin fluctuation temperature and the paramagnon enhancement factor. These latter quantities will be used to compare our results with two theoretical models which deal with the variation of the electronic heat capacity as a function of applied field.

A. Field dependence of β

As seen in Fig. 4, the coefficient of the "lattice" or T^3 contribution to the heat capacity β increases with increasing applied field. This is unexpected, since intuitively a magnetic field would not expect to soften a lattice (which an increase in β implies, i.e., a decrease in Θ_D). Indeed our previous measurements on $LuCo_2$ show that β or Θ_D remains constant to ±0.5% as the field varied from 0 to 10 T.[6]

The neutron scattering results of Stassis *et al.*[19] suggest an explanation of this unusual phenomena. These authors have found that an applied magnetic field of 4.23 T induces a magnetic moment on the Ce sites in $CeSn_3$. Thus this gives rise to a magnetic contribution to the heat capacity at $H>0$ T, C_M, much like a material which orders magnetically. The only difference in the two cases is that in the intrinsic material the moments are aligned by an internal field and in $CeSn_3$ the moments are aligned by an external field. However, if the external field were extremely large >100 T the moments would be frozen at $T<20$ K and there would be no contribution to the heat capacity. From Fig. 3 it is seen that the β term saturates at $H\simeq 6$ T and thus a temperature of the order of 4 K will overcome the magnetic energy causing thermal motion of the spin waves. By using the following equation the value of C_M can be obtained:

$$C_M = C_T - C_E - C_L \quad \text{for } H \geq 5.0 \text{ T} , \tag{4}$$

where C_T is the measured heat capacity and C_E and C_L are the electronic and lattice contributions to the heat capacity. The C_E term (which is equal to γ T) is obtained from the measured γ value as determined at each field. The lattice contribution was calculated from the Debye temperature as given by Stassis *et al.*[19] who found it (Θ_D) to vary considerably with temperature.

The temperature variation of C_M at two fields is shown in Fig. 5, where it is seen to rise quickly, peak at 4 K ($H=5.35$ T) or 5 K ($H=9.98$ T), and fall even more sharply on the high-temperature side. From a plot of $\ln C_M$ vs $\ln T$ the low-temperature values of C_M (from 2 to 3.6 K) were found to lie on a straight line with a slope of 3. The T^3 dependence

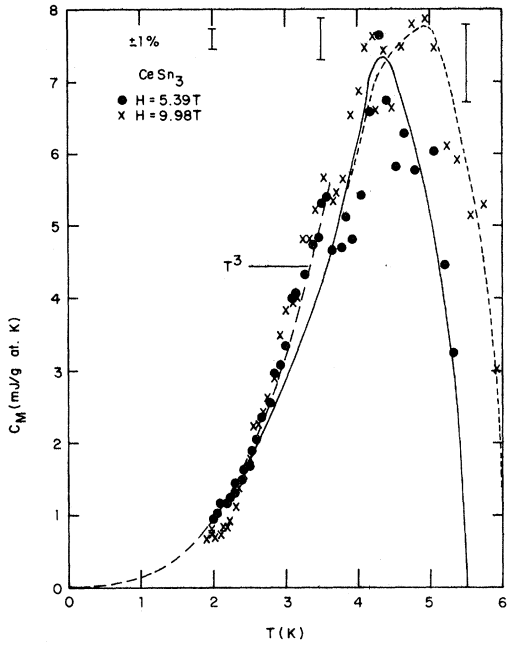

FIG. 5. The temperature dependence of the magnetic contribution to the heat capacity for two applied fields. The error bars at the top of the figure indicate a 1% error of the total heat capacity at 2.0, 3.5, and 5.5 K.

of C_M is clearly seen in Fig. 6. If we assume that the coefficient β of the T^3 term to the heat capacity is composed of two contributions, the lattice (β_L) and magnetic (β_M), then one can check to see if the slopes of the data presented in Fig. 6 are correct. The β_L value (0.173) was determined from the Debye temperature (224 K) at 3 K (the midpoint range of the T^3 fit) as given by Stassis *et al.*[31] By subtracting β_L from the measured β values listed in Table I, values of 0.110 and 0.127 were obtained for β_M at magnetic fields of 5.39 and 9.98 T, respectively. The lines corresponding to these β_L values (slopes) are shown in Fig. 6.

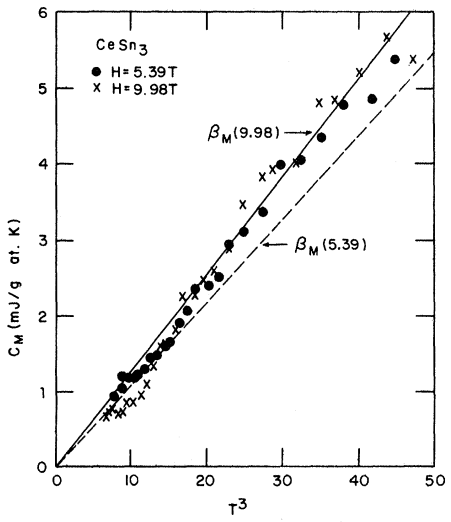

FIG. 6. A plot of the magnetic contribution to the heat capacity (C_M) vs T^3 at two applied magnetic fields. The magnetic contribution to the T^3 term of the heat capacity (β_M), i.e., the slope, was calculated from the experimental β value and Debye temperature at applied fields of 5.39 and 9.98 T (see text).

TABLE III. The magnetic entropy due to the induced magnetic moments in $CeSn_3$ at high magnetic fields.

H (T)	S_M (mJ/g at. K)
5.39	4.4
7.62	5.3
9.98	5.1
Theory ($s=\frac{1}{2}$)	5.76×10^3

The magnetic entropy was calculated for three fields and the results are tabulated in Table III. The constant entropy value at the two highest fields is consistent with the observation that the β value is constant for $H > 7.5$ T (Fig. 4). The magnitude of the entropy is quite small $\sim\frac{1}{1000}$ of the theoretical value for $s=\frac{1}{2}$ (i.e., $S_M = R\ln 2$). This low value is not unexpected, since the measured magnetic entropies for itinerent magnet systems are much smaller than the theoretically expected value [e.g., $\sim 2\%$ for Sc_3In (Ref. 40)]. However, for $CeSn_3$ where the moment is induced by an applied field, S_M is apparently even an order of magnitude smaller. This small entropy indicates that only a small fraction of the electrons are involved in the ordering process.

B. Spin fluctuation temperature

As shown above, after Eq. (3), the fit parameters B and D may be used to determine the spin fluctuation temperature, i.e.,

$$B = \beta - D\ln T_s \tag{5}$$

provided the Debye temperature is known, since

$$\beta = (1.9437\times10^6)/\Theta_D^3 \ . \tag{6}$$

By using the Θ_D value at 0 K given by Stassis *et al.*[31] and the zero field B and D fit parameters a value of 5.8 K is obtained for T_s.

The effective field required to quench spin fluctuations, $H_{\rm eff}$, is given by

$$H_{\rm eff} = k_B T_s/\mu_B = 1.488\,T_s \ , \tag{7}$$

where k_B is the Boltzmann constant and μ_B is the

Bohr magneton. By using $T_s = 5.8$ K, an $H_{\text{eff}} = 8.6$ T is obtained. This effective field is consistent with our observation that γ is essentially constant above 7.6 T, see Fig. 4.

C. Paramagnon (spin fluctuation) enhancement factor

The enhancement of the electronic contribution to the heat capacity due to many-body effects is given by

$$\frac{N(E_F)}{N_0(E_F)} = \frac{\gamma}{\gamma_0} = 1 + \lambda_{e-\text{ph}} + \lambda_{\text{spin}} \ , \tag{8}$$

where $N(E_F)$ is the experimental density of states (DOS), $N_0(E_F)$ is the calculated DOS, $\lambda_{e-\text{ph}}$ is the electron-phonon enhancement factor, λ_{spin} is the paramagnon enhancement factor, and γ_0 is the electronic specific-heat constant calculated from $N_0(E_F)$. As noted above, at $H = 10$ T the spin fluctuations are quenched and Eq. (8) can be written as

$$\frac{\gamma(H=10)}{\gamma_0} = 1 + \lambda_{e-\text{ph}} \ , \tag{9}$$

where $\gamma(H=10)$ is the electronic specific-heat constant measured at a field of 10 T. If $\lambda_{e-\text{ph}}$ is known then γ_0 can be evaluated without calculating the theoretical DOS. Fortunately a reasonable estimate of $\lambda_{e-\text{ph}}$ can be obtained by assuming that it is the same for $CeSn_3$ as it is for $LaSn_3$. This is a good assumption since the experimental phonon spectrum for $CeSn_3$ (Ref. 31) is quite similar to that of $LaSn_3$.[41] The $\lambda_{e-\text{ph}}$ value for $LaSn_3$ was estimated by Stassis *et al.*[41] from the McMillian equation to be 0.763, and we have calculated it to be 0.787 by using Eq. (8), assuming $\lambda_{\text{spin}} = 0$ and a value of $\gamma_0 = 1.65$.[42] Using the average value ($\lambda_{e-\text{ph}} = 0.775$) for $CeSn_3$ a value of 7.49 is obtained for γ_0 from Eq. (9). Substituting $\lambda_{e-\text{ph}}$ and γ_0 into Eq. (8) and using the γ value measured at $H = 0$ (Table I) a value of 0.65 is found for λ_{spin}. This number is quite reasonable in view of the experimental data, and is consistent with Liu's estimate for λ_{spin}[24] and the de Hass–van Alphen results reported by Johanson *et al.*[32]

The value of γ_0 obtained above (7.49) is considerably larger than the calculated band-structure value of 2.65.[42] The calculated value was obtained by using a linearized augmented plane wave (APW) self-consistent relativistic model which included spin-orbit coupling. The discrepancy may be due to the extreme local nature of the $4f$ charge density, which makes the local density approximation for the exchange-correlation potential questionable.

Another possible explanation for the factor of three discrepancy between our estimated value of γ_0 and that deduced from band-structure calculations, is that the Fermi surface is anisotropic and that we have only quenched part of the spin fluctuations. That is, applied fields up to 100 T or higher would be required to completely quench the spin fluctuations associated with other parts of the Fermi surface, i.e., a plot of γ vs H would show a series of steps in which γ decreases and then remains constant before decreasing again. If this were the case our value for γ_0 would be reduced.

The most likely situation is a combination of both explanations. Heat-capacity studies in magnetic fields up to 50 or 100 T would be helpful in solving this discrepancy.

D. Comparison to theoretical models

Béal-Monod *et al.*[3] have shown that the shift of the electronic specific-heat constant at 0 K by an applied field H,

$$[C(0) - C(H)]/C(0) \approx [\gamma(0) - \gamma(H)]/\gamma(0) = \delta\gamma/\gamma(0) \ ,$$

should be

$$\frac{\delta\gamma}{\gamma(0)} \approx 0.1 \times \left(\frac{S}{\ln S}\right) h^2 \ , \tag{10}$$

where S is the Stoner enhancement factor and $h = \mu_B H / k_B T_s$. The Stoner enhancement factor, $S \equiv \chi(\text{obs})/\chi_0$, can be estimated from the observed magnetic susceptibility, extrapolated to 0 K [χ(obs)] and the Pauli-paramagnetic susceptibility at 0 K as determined from the density of states at the Fermi surface (χ_0). We have adopted a value of 0.43×10^{-3} emu/g atom, which is given by Misawa[27] as χ (obs), whereas the χ_0 value is estimated from the γ_0 value obtained above (7.49). Thus, a value of $S = 4.23$ is obtained and Eq. (10) for $CeSn_3$ becomes $\delta\gamma/\gamma(0) \approx 29.33h^2$ (%). The predicted variation of $\delta\gamma/\gamma(0)$ as a function of h is shown in Fig. 7 as curve c along with the observed results.

On the other hand, Hertel *et al.*[4] have recently made a more detailed mathematical analysis for the quenching effect of the spin fluctuation enhancement of the electronic specific heat caused by magnetic fields. They have shown the effective mass enhancement $\lambda_{\text{spin}}(H)/\lambda_{\text{spin}}(0)$ as a function of the reduced field, $h \equiv \mu_B H / k_B T_s$, for the metals with $S = 10$ and $\lambda_{\text{spin}} = 0.37$ (case a) and with $S = 4$ and $\lambda_{\text{spin}} = 0.345$ (case b). These results are also shown in Fig. 7.

The experimental results at low h appear to be in reasonable agreement with the b curve of Hertel *et al.* and in fair agreement with their a curve. This would be expected since the parameters used to generate curve b are closer to those found in $CeSn_3$ than the parameters used to generate curve a. The Stoner enhancement value for the model of Hertel *et al.* (4) is nearly the same as the experimental value (4.23),

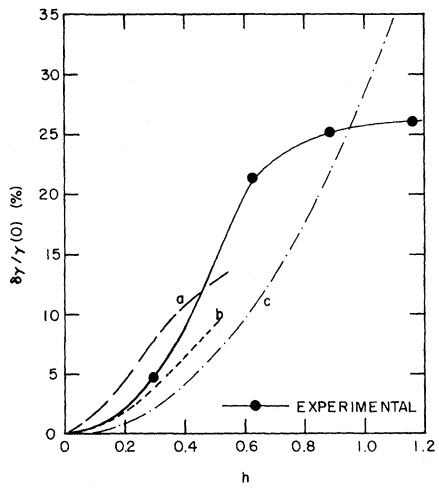

FIG. 7. The shift of the electronic specific-heat constant at 0 K, $\delta\gamma/\gamma(0) = [\gamma(0) - \gamma(H)]/\gamma(0)$, of $CeSn_3$ caused by an applied field as a function of the reduced field, $h \equiv \mu_B H/k_B T_s$. The theoretical curves a and b are taken from Hertel *et al.*[4] (case a: $S = 10$ and $\lambda_{spin} = 0.37$, and case b: $S = 4$ and $\lambda_{spin} = 0.345$) and from Béal-Monod *et al.* (Ref. 3) (curve c: $S = 4.23$ and $T_s = 5.8$ K).

but the λ_{spin} value used to calculate curve b is about half of the experimental value (0.345 vs 0.65).

For the model of Béal-Monod and co-workers the agreement is fair to poor, although the experimentally derived parameters S and T_s are used to calculate curve c. A change of the constant in Eq. (10) from 0.1 to 0.2 would shift curve c right on top of the experimental curve up to $h \simeq 0.6$. At high values of $h > 0.6$ the model of Béal-Monod *et al.* would not be expected to hold since the applied fields are approaching $H_{eff}(h \simeq 1.0)$ and the spin fluctuations are nearly quenched. This is evident in Fig. 7 by the leveling of the experimental curve for $H > 0.7$.

E. Other considerations

The anomalous temperature dependence of the magnetic susceptibility in $CeSn_3$, which has a broad maximum around 150 K, has been discussed from two points of view. Misawa[27] and Beal-Monod and Lawrence[28] have reanalyzed the susceptibility of $CeSn_3$ following the Fermi liquid model and they have attributed the broad maximum to the $T^2 \ln T$ and T^2 terms of the susceptibility inherent in this model. On the other hand, Lethuiller and Lacroix-Lyon-Caen[43] have described this compound as a Kondo metal with the Kondo temperature $T_K = 120$ K. However, Anderson[44] suggested that this problem should be discussed from the mixed valence nature of this compound because the magnetic moments appear with increasing temperature. Our experimental heat-capacity measurements on $CeSn_3$ below 20 K in magnetic fields up to 10 T, give additional support to the Fermi liquid model rather than the Kondo model.

The discussion based on the magnetic susceptibility measurements has suggested a high value of T_s for $CeSn_3$ ($T_s \approx 200$ K), which is over 30 times larger than our results ($T_s = 5.8$ K). A similar inconsistency in T_s has been found for $LuCo_2$ (Ref. 6) and Pd.[45] Usually, the characteristic spin fluctuation temperature, T_s, has been obtained from the analyses of the χ vs T curve by using the definition, $T_s = T_F/S$. Thus values of $T_s \sim 630$ K was estimated for $LuCo_2$ and $T_s \sim 400$ K for Pd. However, a remarkable decrease of the electronic specific-heat constant for these materials (11% at 9.98 T for $LuCo_2$ and 8% at 11 T for Pd) was observed in the low-temperature-high magnetic field heat-capacity measurements. These facts strongly suggest that the heretofore estimated values for T_s are unreasonably large by at least one order of magnitude, since magnetic fields of the order of T_s are required to quench the spin fluctuation enhancements. This inconsistency may require a new definition of T_s, which contains the more detailed characteristics of the band structure besides T_F and S, or perhaps two different T_S values are necessary to describe the material, one associated with the heat capacity and the other with the magnetic susceptibility.

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