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Authors
Flouquet, J.
Haen, P.
Lejay, P.
et al.

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MAGNETIC INSTABILITY IN Ce HEAVY FERMION COMPOUNDS

J. Flouquet°, P. Haen°, P. Lejay°, P. Morin+, D. Jaccard[x], J. Schweizer≠, C. Vettier[xx], R.A. Fisher* and N.E. Phillips*

°CRTBT[□], +Laboratoire Louis-Néel[□], C.N.R.S., BP 166 X, 38042 Grenoble-Cédex, France
[x]Département de la Matiére Condensée, 1211 Genéve, Switzerland
≠DRF/SPhMDN, C.E.N.G., BP 85 X, 38041 Grenoble-Cédex, France
[xx]ILL, 156 X, 38042 Grenoble-Cédex, France
*MMRD Lawrence Berkeley Laboratory, University of California, Berkeley, CA 94720, USA.

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J. Flouquet°, P. Haen°, P. Lejay°, P. Morin+, D. Jaccard[x], J. Schweizer‡, C. Vettier[xx], R.A. Fisher*, and N.E. Phillips*

°CRTBT[□], +Laboratoire Louis-Néel[□], C.N.R.S., BP 166 X, 38042 Grenoble-Cédex, France

[x]Département de la Matière Condensée, 1211 Genève, Switzerland

‡DRF/SPhMDN, C.E.N.G., BP 85 X, 38041 Grenoble-Cédex, France

[xx]ILL, 156 X, 38042 Grenoble-Cédex, France

*MMRD Lawrence Berkeley Laboratory, University of California, Berkeley, CA 94720, USA.

Abstract

Different types of cerium heavy fermion compounds are discussed : cubic antiferromagnetic ground states ($CeAl_2$, $CePb_3$, $CeIn_3$) and Pauli paramagnetic ground states ($CeSn_3$, $CeRu_2Si_2$). Applying external variables such as pressure or magnetic field restore a localized Fermi liquid state. Special attention is paid to $CeRu_2Si_2$ which is approaching a singular magnetic instability for $T \rightarrow 0$. Comparison between pressure and alloying effects emphasize the important role of the itineracy of the 4f electrons at the magnetic instability.

Keywords : Ce compounds, Antiferromagnetism, Neutron diffraction, Pressure effect, Specific heat, Thermal expansion, magnetostriction.

Running title : Magnetic Instability in Ce Heavy Fermion Compds.

[□]Laboratoires associés à l'Université Joseph Fourier, Grenoble.

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The large variety of situations in heavy fermion systems arises from the interplay between different couplings generally referred to as a single site local fluctuation reminiscent of the Kondo effect (energy $k_B T_K$), the crystal field splitting (Δ) governing the degeneracy of the level and the intersite magnetic coupling J_{ij}. Another important ingredient is the crystal symmetry which may lead to strong magnetic anisotropy and more generally may determine the shape of the Fermi surface, allowing nesting when the itinerant character of the f electrons play a role. Here, an attempt is made to classify different situations according to the hierarchy of these terms. We shall focus only on cerium compounds and on the discussion of magnetism in normal phases. The approach of the magnetic to non-magnetic (M-NM) transition from a long range antiferromagnetic (AF) ground state to a Pauli paramagnet (PP) ground state has been studied by applying pressure (P) on AF cubic materials : $CeAl_2$, $CePb_3$, $CeIn_3$ [1]. The situation of the Pauli paramagnet $CeSn_3$ is interesting since it reproduces the state of above mentioned materials for P heigher than the critical pressure (P_c) for the M-NM transition. Emphasis is given to the appearance of a large d contribution to the magnetic form factor [2,3] and its rapid collapse with the defect content. An illuminating example is the reappearance of AF ordering in Ce_2Sn_5, a superstructure of $CeSn_3$ [4]. Near a M-NM transition, the sensibility of defects is certainly high ; high purity materials are needed to study the low energy excitation of a Ce lattice.

Experimentally, the compound $CeRu_2Si_2$ is an ideal case since it is located just at the vicinity of a M-NM transition as revealed by : i) the strong increase of the magnetization which appears for a field H_M ($\approx$ 8.3 T at 4.2 K [5,6]) applied along the c-axis of the tetragonal structure in single crystals,
ii) the existence of intersite AF correlations which vanish at H_M [7], and
iii) the occurrence of a M-NM transition at zero field in the system $Ce_{1-x}La_xRu_2Si_2$ for a small critical concentration $x_c \geq 0.08$ [8]. This compound is also characterized by an unusual value of the electronic Grüneisen parameter $\Omega_f \sim 180$ (the same value is derived from magnetic measurements) [6]. This large value of Ω_f leads to unusual magnetic field effects on the elastic properties, i.e. on the compressibility and on the volume itself [6,9]. We shall discuss the enhancement at H_M of the coefficient γ in the low temperature specific heat. It is strongly suggested that a singular point occurs at H_M for $T \rightarrow 0$, i.e. that a magnetic instability (referred to as a metamagnetic transition) can be induced just at H_M at 0 K. Comparison with

$Ce_xLa_{1-x}Ru_2Si_2$ alloys shows that the breakdown of the lattice invariance restores rapidly a localized picture [10,11].

Finally, the situation of $CeAl_3$ which has been considered for more than a decade as the archetype of PP heavy fermion compound is still a puzzle. The strong non-linearity of its properties on applying P and H may be characteristic of the crossing of M-NM transition and of the sensibility to defects [12].

Pressure induced instability in cubic materials

$P < P_c$: Antiferromagnetic ground state : $CeAl_2$, $CePb_3$, $CeIn_3$:

At P = 0, these three compounds order antiferromagnetically at T_N = 3.8, 1.2 and 10 K, respectively [1]. Static measurements such as specific heat, susceptibility or magnetic form factor show that the degeneracy of the 4f level is a doublet at low temperature. In dynamic measurements such as resistivity or inelastic neutron scattering, the observation of the crystal field splitting is unambiguous for $CeAl_2$ and $CePb_3$ but extremely difficult in $CeIn_3$. That suggests at P = 0, for $CeAl_2$ and $CePb_3$, $k_BT_K < \Delta$ while, for $CeIn_3$, $k_BT_K \sim \Delta$.

The evolution of the magnetic structure and the variations of T_N vs. P as determined by neutron diffraction experiments (and for T_N in some cases by transport properties measurements) are shown in Fig. 1. For the two well localized cases ($CeAl_2$ and $CePb_3$) the main feature is the initial rapid decrease of T_N under pressure before a change at $P_{I \to II}$ from a modulated structure (I) to a $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ structure (II) analoguous to that of $CeIn_3$ at P = 0. The transition at $P_{I \to II}$ is interpreted as a signature of the weakness of the magnetic anisotropy correlated to the entrance in the P regime where $k_BT_K \sim \Delta$.

Thus $CeIn_3$ is an excellent candidate to study a magnetic instability under pressure. In this compound, T_N decreases first weakly on applying P. Its initial magnetic Grüneisen parameter $\Omega_M = -\partial \ln T_N / \partial \ln V$ (where V is the volume) equals -2, which is opposite to the electronic Grüneisen constant Ω_f = +7 as derived from the resistivity maximum [13]. This suggests competition between intersite (J_{ij}) and local coupling k_BT_K. For P > 18 kbar,

$T_N(P)$ decreases rapidly and almost linearly. Ω_M (20 kbar) ~ -48. It would be intersting to determine a critical exposant near P_c.

$P > P_c$: Emergence of a d contribution in pure $CeSn_3$

$CeSn_3$ is often chosen as an archetype of either high temperature Kondo lattice ($k_BT_K > \Delta$) or even intermediate valence compound, with a PP ground state. The case of $CeSn_3$ is equivalent to that of $CeIn_3$ where a M-NM transition has been driven, either by pressure ($P > P_c$) or by alloying (such as in $CeIn_{3-x}Sn_x$ for $x > 2.5$ [14]). Like other cubic materials ($CePd_3$, $CeBe_{13}$), $CeSn_3$ is characterized by a quasi-constant value of its electronic Grüneisen parameter, $\Omega_f \sim +10$, rather analoguous to the value of ~ +7 mentioned above for $CeIn_3$. One of the major result in $CeSn_3$ is that the Ce magnetic form factor exhibits below 40 K, an extracontribution of 5d character reaching for the best sample 49 % of the 4f amplitude [2] (Fig. 2). Systematic studies on different crystals of $CeSn_3$ [3] but also of the compound Ce_2Sn_5 [4] (which will appear in $CeSn_3$ as a parasitic phase induced by stacking faults) demonstrate that the 5d component is gradually lowered when the number of defects is intentionally increased. This component disappears for 1.2 % of defects, corresponding to a critical mean free path between 20 and 50 Å. The periodicity of the lattice is required for the development of the delocalized part of the magnetization. This strongly suggests that defects or doping may wipe out the critical regime at P_c. Another illustrating point is that Ce_2Sn_5 which exhibits two Ce sites in its structure (Fig. 3) becomes AF with $T_N = 2.5$ K. Its particularity is that the Ce_I site equivalent to Ce in $CeSn_3$ remains non magnetic while the Ce_{II} site with two Ce nearest neighbors carries a magnetic moment. Low temperature specific heat measurements lead below T_N to $\gamma = 380$ mJ mole^{-1} K^{-2}, i.e. 190 mJ K^{-2} per Ce_{II} atom instead of $\gamma = 35$ mJ mole^{-1} K^{-2} for $CeSn_3$ [4].

Singular behavior of $CeRu_2Si_2$ at $H = H_M$ for $T \to 0$ K

The series $Ce_{1-x}La_xRu_2Si_2$ has been extensively studied [5, 7-11, 15-17]. In PP ground state alloys ($x < 0.08$) the differential susceptibility ($\chi_H = \partial M/\partial H$) exhibits a huge maximum at a metamagnetic-like field H_M [5,15]. At 1.5 K the value of H_M is reduced from 7.9 T for x = 0 to 5.6 T for x = 0.05 [15]. At $T \to 0$, the latter values become, 7.7 T for x = 0 and ≈ 5.3 T for x = 0.05, as measured on new samples [11,17]. For x = 0, a rapid decrease of the residual initial susceptibility $\chi_0 = \chi(T \rightarrow 0)$ is observed on applying pressure [6], leading to the large value $\Omega_f = 180$ mentioned above. The remarkable feature is that the metamagnetic transition appears for a critical value M_c of the magnetization, i.e. the product $\chi_0 H_M$ is pressure invariant. That leads to relate different quantities by a scaling law. The approach to an instability at $H = H_M$ as $T \rightarrow 0$ K, is due to the field expansion of the lattice : $\frac{\Delta V}{V}$ $(H > H_M) \sim 10^{-3}$. The latter value is almost the same as the difference (2.2×10^{-3}) between the volumes for $x = 0$ and $x = x_c$: in a simple picture, a negative pressure of 2 kbar could induce AF order in $CeRu_2Si_2$.

Fig. 4a shows the specific heat of $Ce_{1-x}La_xRu_2Si_2$ single crystals at $H = 0$ in term of C/T vs. T. These data confirm the absence of order for $x = 0$ and $x = 0.05$, while, according to magnetization experiments [15b], long range order occurs for $x = 0.10$ and $x = 0.13$, at $T_N \approx 2.7$ and ~ 4.1 K, respectively. The magnetization of the AF ground state alloys exhibits below T_N a metamagnetic step at a field H_c inducing a ferromagnetically polarized state. For $x = 0.10$ and $x = 0.13$, $H_c \approx 3.5$ T at 1.5 K [15]. Fig. 4b represents C/T of the same crystals for a magnetic field (still applied along the c-axis) close to H_M in the PP cases or close to H_c in the AF cases. Interesting features can be observed in Fig. 4b : i) a weak maximum of C/T appears for $CeRu_2Si_2$ while only a continuous increase of C/T is observed for $x = 0.05$; ii) In the ordered compounds, a peak or a maximum of C/T emerges more clearly than at $H = 0$. It corresponds to the value of H_c obtained by magnetization [15b].

A plot of γ ($\equiv$ C/T extrapolated to 0 K) shows an enhancement of γ of 28 % at H_M for $x = 0.05$, while none is found for the AF alloys [10]. For $x = 0$ and $x = 0.05$, these experimental values are in good agreement (Fig. 5) with the variation of γ derived (through the Maxwell relation) from magnetization data gathered below 1 K which show a T^2 variation of M at constant field [11,17]. For $x = 0$, an enhancement of γ of 62 % at H_M is derived from these data when taken in the $0.1 \text{ K} < T < 0.4$ K range, while data collected only above 1.3 K [16] led to a lower effect. Similarly, an enhancement of only 30 % could be predicted [5] by comparing the values at $H = 0$ and $H \approx H_M$ of the coefficient B in the linear term (BT) in the resistivity (ρ) between 1.5 and 4.2 K (*) , while if a scaling, $A \propto \gamma^2$, is applied to these data, assuming $\rho \propto AT^2$ below 1 K, the correct enhancement of γ is predicted.

(*) For $x = 0.05$, magnetoresistance data [R. Djerbi, thesis Grenoble (1988) ; see also R. Djerbi *et al.*, J. Mag. Mag. Mat. **76&77** (1988) 265] show an enhancement of B of only 15 %.

Thus, the mass enhancement appears to decrease on warming. This point has been clarified recently [11] by a measure of the thermal dilatation $\alpha(T)$ in different applied fields. These data show that the extremum T_m^α of $\alpha(T)$ goes from 9 K at H = 0 to ~ 350 mK at H_M and increase again above H_M (inset Fig. 5). This variation of T_m^α vs. H delimit a high temperature region, where $CeRu_2Si_2$ appears as a disordered paramagnet, from two low temperature regions : a strongly correlated Fermi liquid below H_M and a purely field localized polarized Fermi liquid above H_M. In other words, the characteristic temperature where Fermi liquid is recovered (i.e. T law in C and α, T^2 law in M and ρ) reaches a deep minimum at H_M. This shift of the characteristic temperature agrees with the strong temperature increase of χ_{H_M} on cooling, in contrast with the achievement of a T^2 susceptibility at $H \to 0$ below 4 K [5,10b]. The picture of $CeRu_2Si_2$ is that of a heavy fermion lattice approaching a Kondo lattice collapse driven by a magnetic instability.

The values predicted for $\gamma(H_M)$: 560 and 630 $mJ.mole^{-1}K^{-2}$ for x = 0 and x = 0.05 respectively, appear close to the value $\gamma(0)$ = 600 $mJ.mole^{-1}K^{-2}$ measured at H = 0 for x = 0.13 [10]. This might be a critical value for the M-NM transition.

Furthermore, for x = 0 the enhancement of γ appears as a sharp feature in the reduced scale H/H_M by contrast with the data for x = 0.05. The already prepared samples of $CeRu_2Si_2$ have a residual resistivity close to 2 $\mu\Omega$cm [5], suggesting that their quantity of defects might be important. An open question is as to whether these defects play any role in the variation of γ, by comparison with alloying which smears out drastically all anomalies at H_M. The itineracy of the quasi-particles is primordial. A coherent picture seems to be recovered on increasing x (> x_c) when a static magnetic order is realized. However, no continuity occurs between x = 0 and x > 0.08. The common feature is the local character of the magnetism (dominant factor for x > x_c) but the nesting condition of the Fermi surface may appear only when the lattice invariance is respected.

$CeAl_3$: Non-linear pressure and field effects

During more than a decade $CeAl_3$ has been considered as the archetype PP heavy fermion compound. The observations of a spontaneous Larmor precession at H = 0 below 0.7 K [19], of a T^3 term in the resistivity of single crystals below 1.6 K [20] and of NMR Al broadening at 1.2 K [21] seem to classify now $CeAl_3$ as an AF ground state with a value $m_o \sim 0.3\ \mu_B$ for the average staggered magnetization. However muon spin experiments show a gradual onset of magnetic correlation on cooling. A broad distribution of local magnetic fields centered around zero appear below 2 K on only part of the muon sites. Neutron diffraction experiments have failed to reveal long range magnetic order. The specific heat of $CeAl_3$ shows a large difference between the temperature $T_M^{C/T} \sim 350$ mK of the maximum in C/T and its inflection at T" ~ 1.6 K [22]. (At the opposite, for the AF alloy $Ce_{0.87}La_{0.13}Ru_2Si_2$, the values $T_M^{C/T} \sim 3.3$ K and T" ~ 4.2 K shown by Fig. 4a are rather near $T_N \sim 4.5$ K measured directly by neutron [8]). In $CeAl_3$, the large temperature range where C/T increases on cooling is certainly not governed by imperfections in the crystal since samples prepared in different laboratories have quite similar specific heats [22]. It was suggested that the particular behavior of $CeAl_3$ is due to frustrating interactions [23].

Let us consider the low temperature resistivity measured at different pressures and magnetic fields on a polycrystal (Fig. 6) .When the resistivity is analyzed in terms of residual ρ_o and AT^2 contributions, the main features are i) the occurrence of a maximum in A at $P^* \sim 1.2$ kbar, ii) the strong correlation between the pressure variation of A and ρ_o (i.e. the strong coupling between defects and lattice), iii) the occurrence of a metamagnetic transition as observed in $CeRu_2Si_2$. The initial P increase of A is in excellent agreement with the negative sign of the Grüneisen parameter $\Omega(P=0) = -200$ [22]. Above 1.2 kbar, a positive Grüneisen parameter is recovered. Another evidence of the crossing through P_c with pressure is that usual scaling laws, applied in ordinary heavy fermion compounds ($P >> P_c$)

$$A \sim 1/\sqrt{T_M^{\ell}} \text{ or } \frac{1}{\sqrt{T^*}},$$

(T^*, temperature up to which a T^2 law is obeyed) cannot be applied for $P \gtrsim P^*$ [12]. It is tempting to identify P^* and P_c. However, the metamagnetic field H_M seems to increase drastically only above 3 kbar. By analogy with the results of the serie $Ce_xLa_{1-x}Ru_2Si_2$ [15], P_c of $CeAl_3$ may be chosen as 3 kbar.

Conclusion

Up to now enhancement of γ at H_M has been found in systems ($CeRu_2Si_2$, UPt_3) where the itineracy of the quasi particules is well established, in agreement with dHvH experiments [24]. UPt_3 is considered as a weak antiferromagnet [25] while up to now no evidence of AF ordering has been found in $CeRu_2Si_2$. By contrast for $CeAl_2$ where the local nature of the Ce atoms drives the magnetic ordering ($T_N \sim T_K$), no enhancement of γ is observed at H_C. The same behaviour is found in $Ce_xLa_{1-x}Ru_2Si_2$ when by alloying the AF ordering is induced. The disappearance of the electronic lattice anomaly in systems containing defects is shown for the metamagnetic transition but also for a system like $CeSn_3$ known as far from any magnetic instability. The specific situation of $CeAl_3$ ($P_c \sim 3$ kbar) shows also the strong interplay between defects and lattice. Finally, we wish to stress the importance exhibited by quantitative studies of the heavy fermion magnetic instability on pure materials by varying continuously pressure and field through their critical points (P_c, H_M).

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FIGURE CAPTIONS

Fig. 1 : Temperature-Pressure magnetic phase diagrams of $CeAl_2$, $CePb_3$ and $CeIn_3$ (from ref. 1). Region III corresponds to the paramagnetic state, I to the incommensurate structures of $CeAl_2$ and $CePb_3$ at low pressure, II to the $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ commensurate structures of the three compounds. For $CePb_3$ the open triangles were obrained by transport properties measurements. For $CeIn_3$, $T_N(P)$ was deduced from (●) neutron, (○) specific heat, and (■) resistivity measurements.

Fig. 2 : Ce magnetic form factor in $CeSn_3$ at 4.2 K in an applied field of 4.6 T [3]. The arrows show the value obtained in reference.

Fig. 3 : Substitution zone in $CeSn_3$ and structure of Ce_2Sn_5 [4]. The latter is a surstructure of $CeSn_3$ obtained by removing one plane of Sn atoms every four $CeSn_3$ cells and by a a/2 glide of next cells.

Fig. 4 : Plots of C/T as a function of T for $Ce_xLa_{1-x}Ru_2Si_2$: a) for H = 0, b) for H close to H_M or to H_c, i.e. for H = 7.5, 5.5, 3.5 and 3.5 T, for x = 0, 0.05, 0.10 and 0.13, respectively.

Fig. 5 : Dependence of γ for $CeRu_2Si_2$ and $Ce_{0.95}La_{0.05}Ru_2Si_2$ as a function of H/H_M ; comparison with experimental data (■). The insert shows the variation of T_m^{α} as a function of H in $CeRu_2Si_2$.

Fig. 6 : For $CeAl_3$: pressure and field dependence of the A coefficient of the AT^2 resistivity term and of the residual resistivity ρ_o. The open circles in the basal plane represent H_c or H_M as observed in magnetoresistivity experiments.

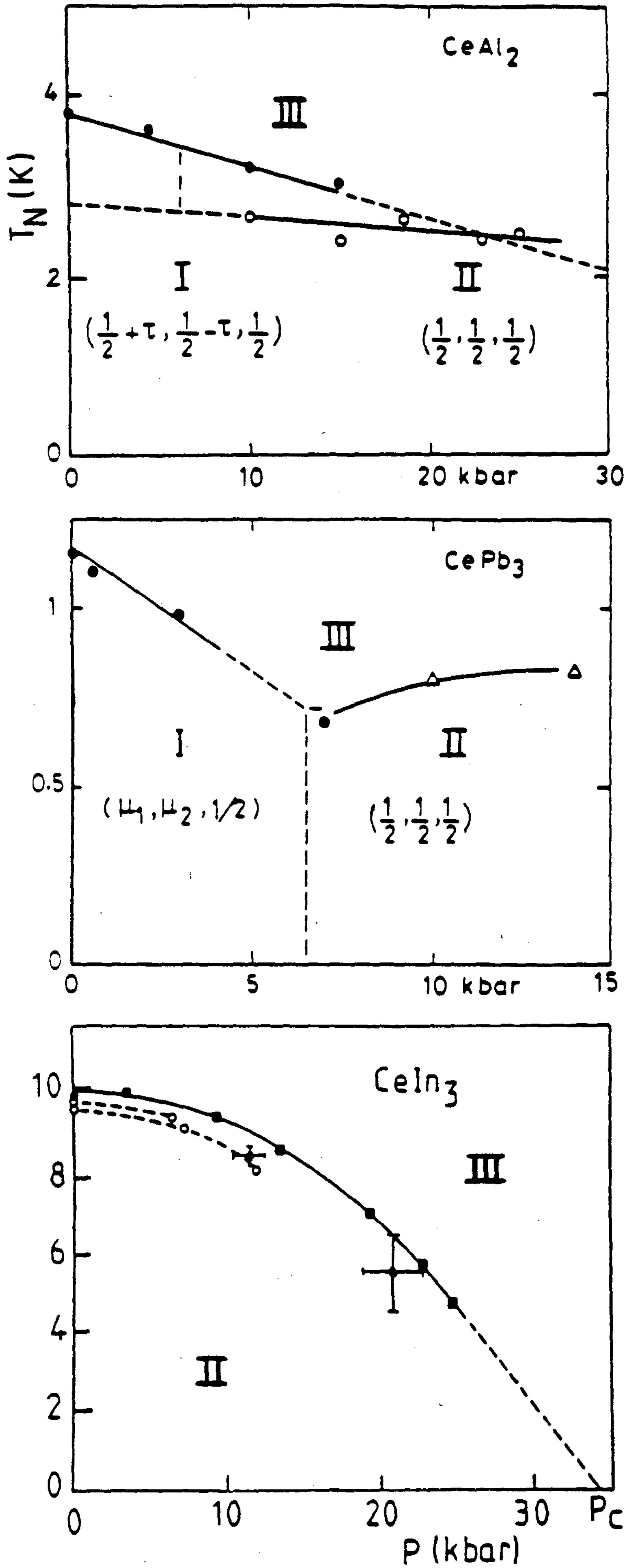

CeAl2
III
I
II
(1/2+τ, 1/2−τ, 1/2)
(1/2, 1/2, 1/2)
TN (K)
20 kbar
CePb3
III
I
II
(μ1, μ2, 1/2)
(1/2, 1/2, 1/2)
10 kbar
CeIn3
III
II
Pc
P (kbar)

FIG. 1

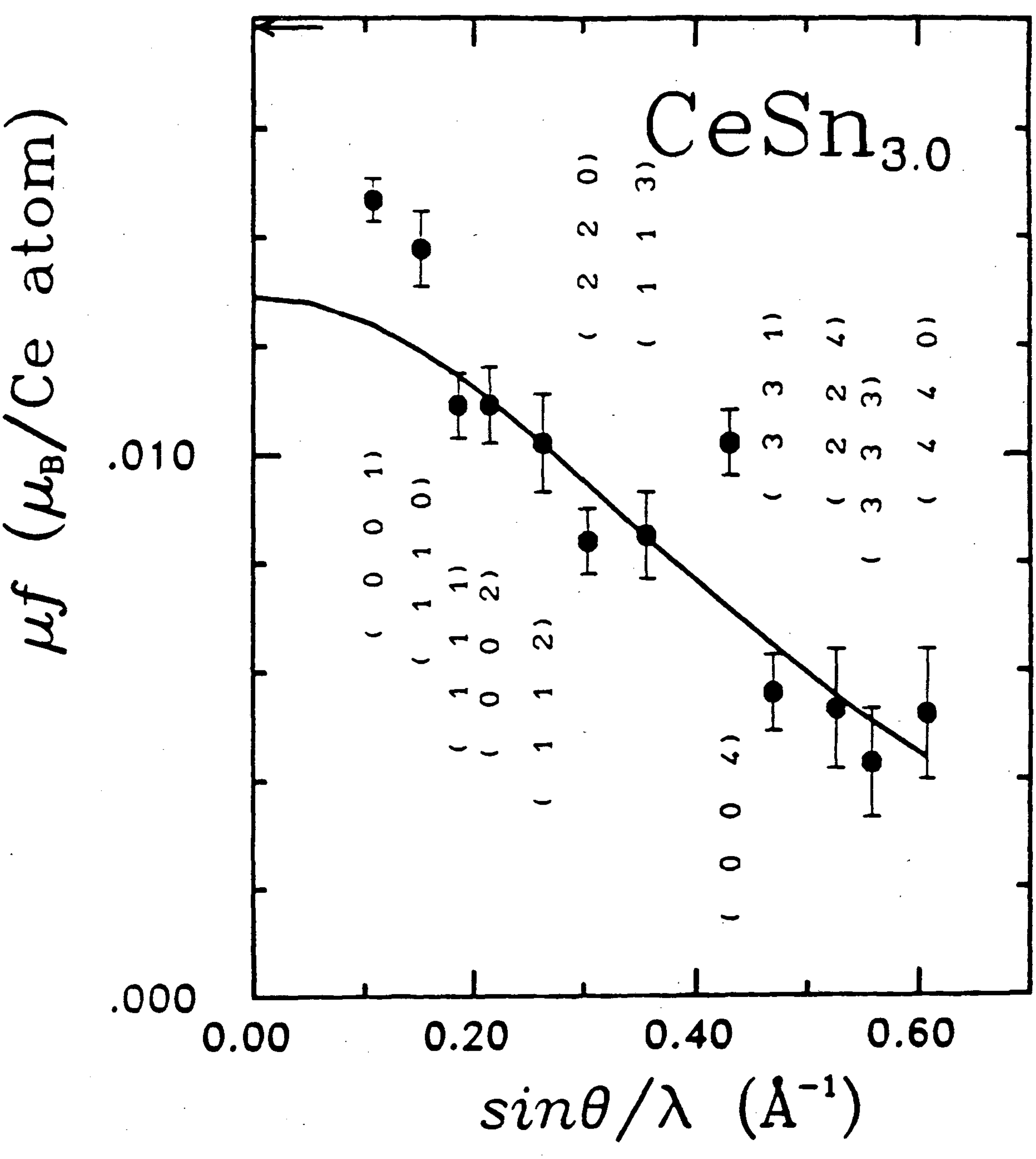
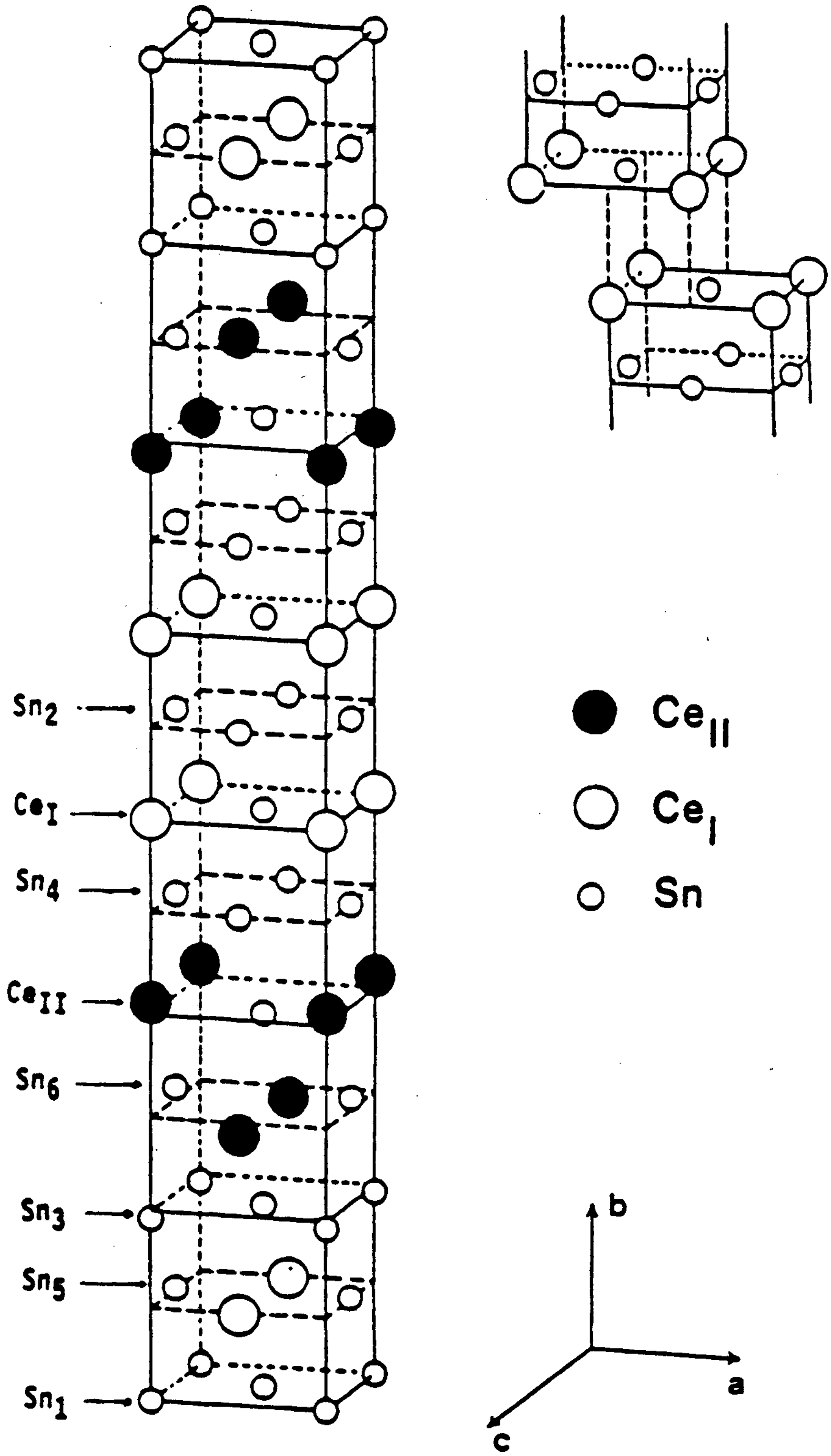
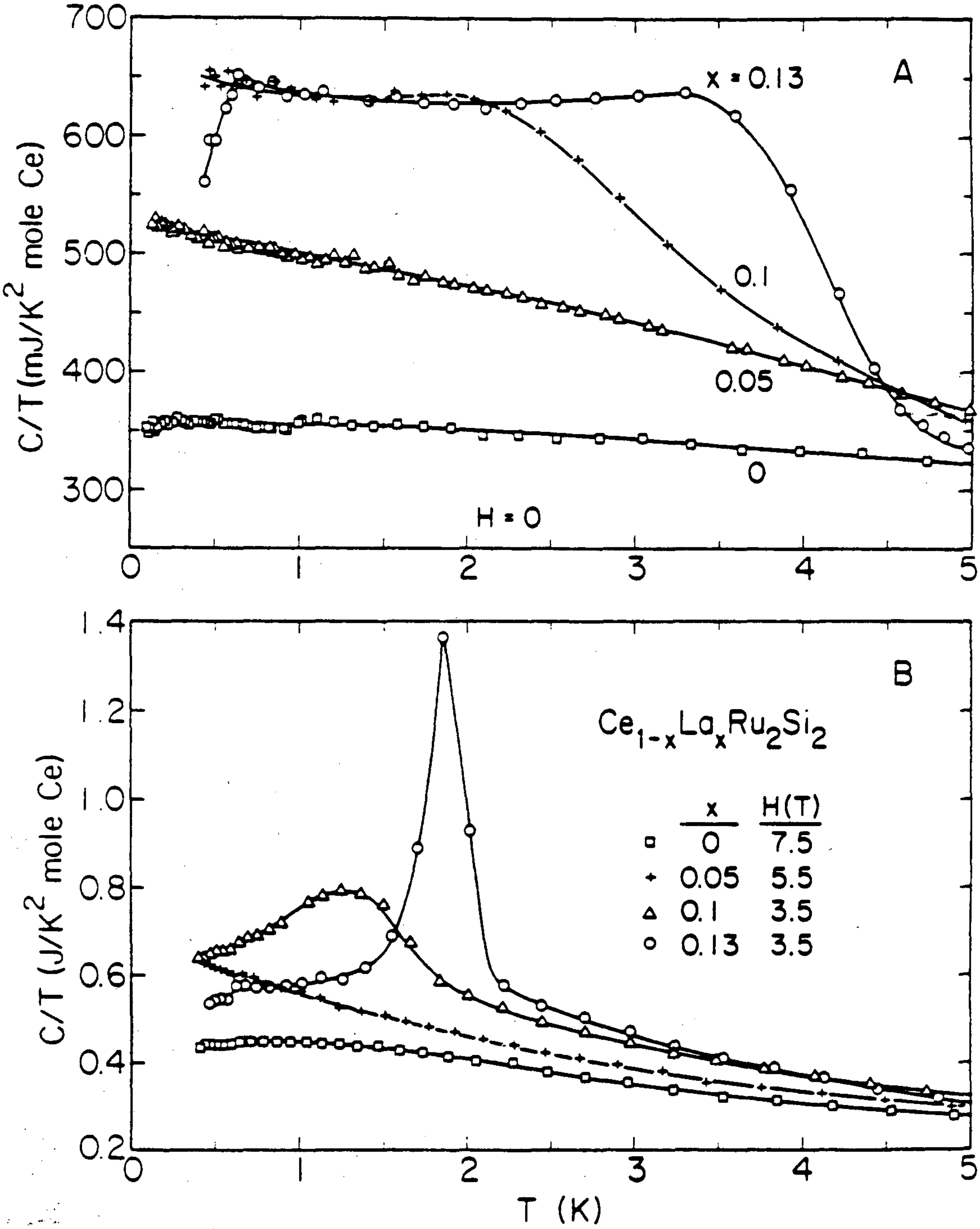

CeSn3.0
μf (μB/Ce atom)
.010
.000
0.00
0.20
0.40
0.60
sinθ/λ (Å-1)
(0 0 1)
(1 1 0)
(1 1 1)
(0 0 2)
(1 1 2)
(2 2 0)
(1 1 3)
(0 0 4)
(3 3 1)
(2 2 4)
(3 3 3)
(4 4 0)

FIG. 2



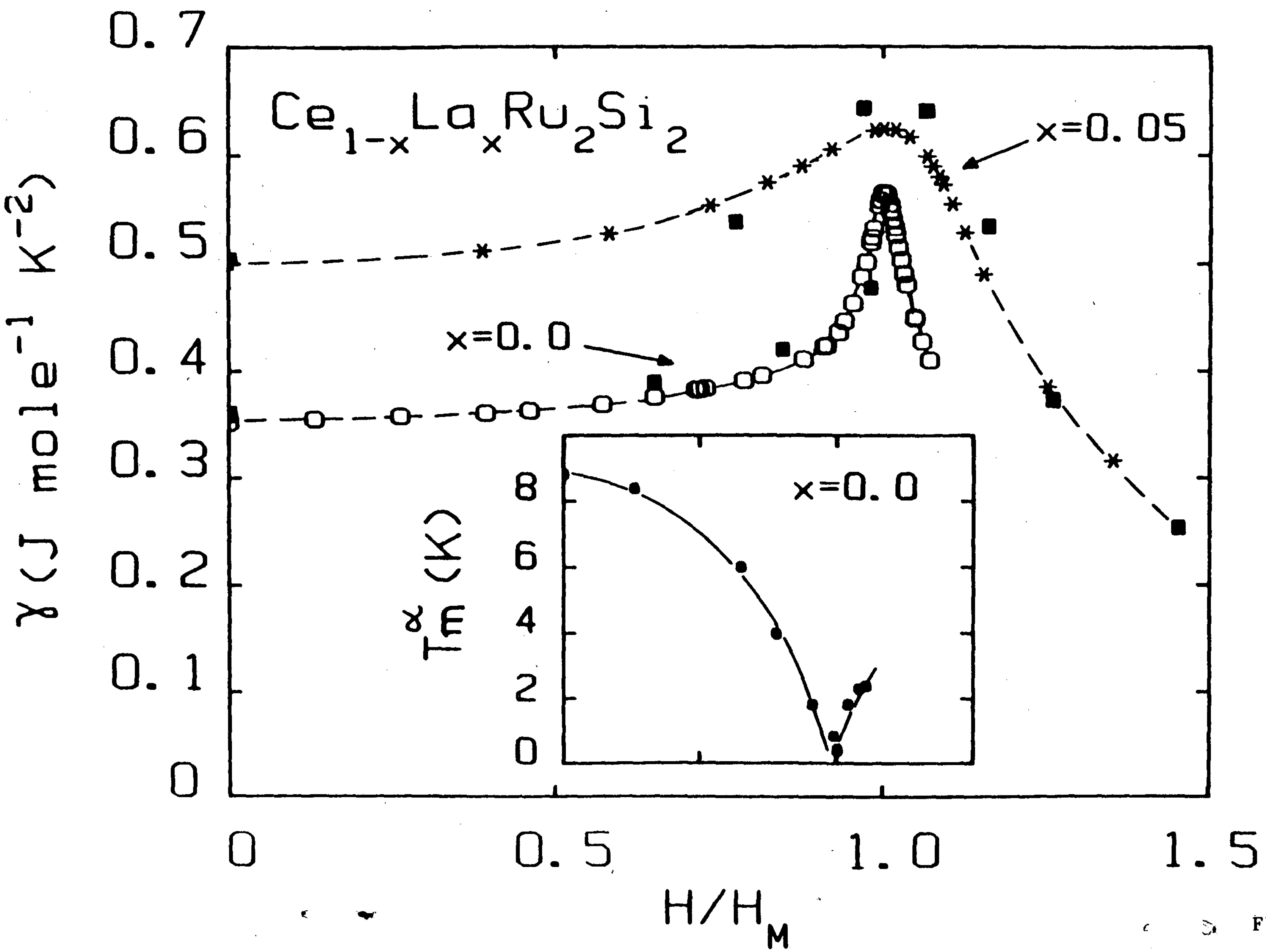
Ce_2Sn_5

FIG. 3



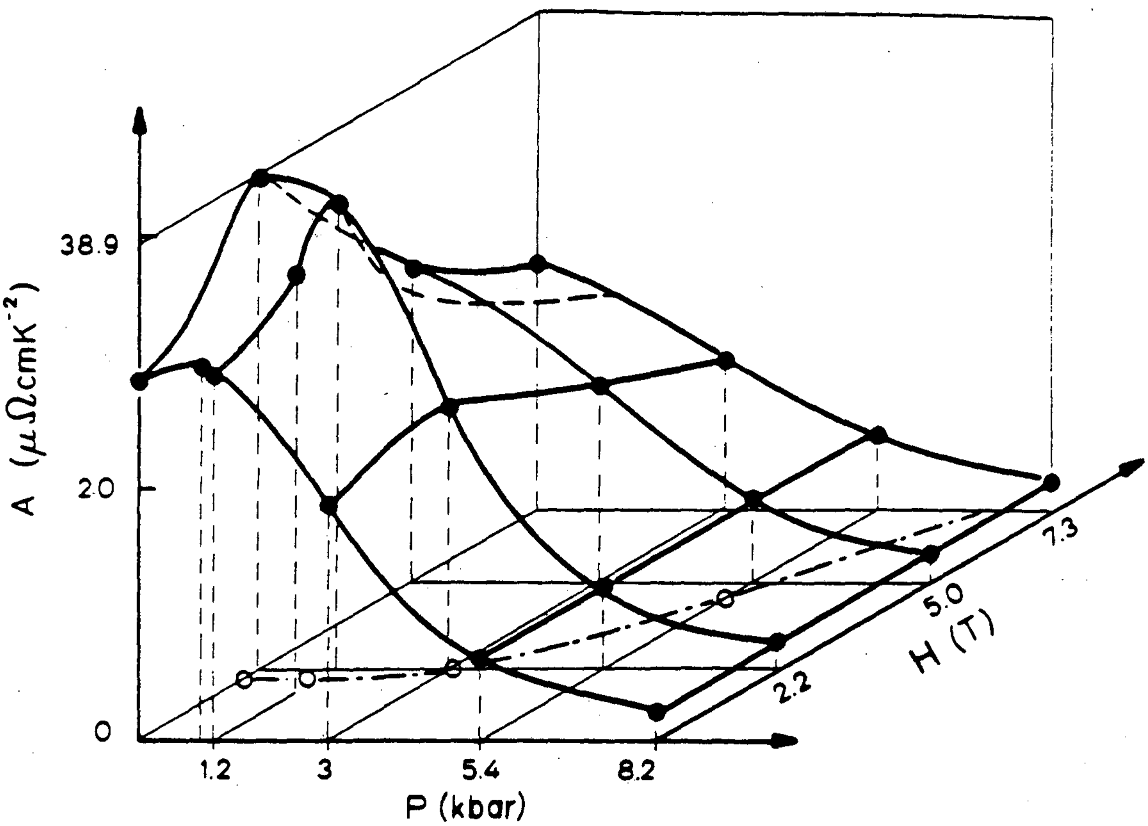
700
600
500
400
300
C/T (mJ/K^2 mole Ce)
x = 0.13
A
0.1
0.05
0
H = 0
0
1
2
3
4
5
1.4
1.2
1.0
0.8
0.6
0.4
0.2
C/T (J/K^2 mole Ce)
B
$Ce_{1-x}La_xRu_2Si_2$
x
H(T)
0
7.5
0.05
5.5
0.1
3.5
0.13
3.5
T (K)

FIG. 4

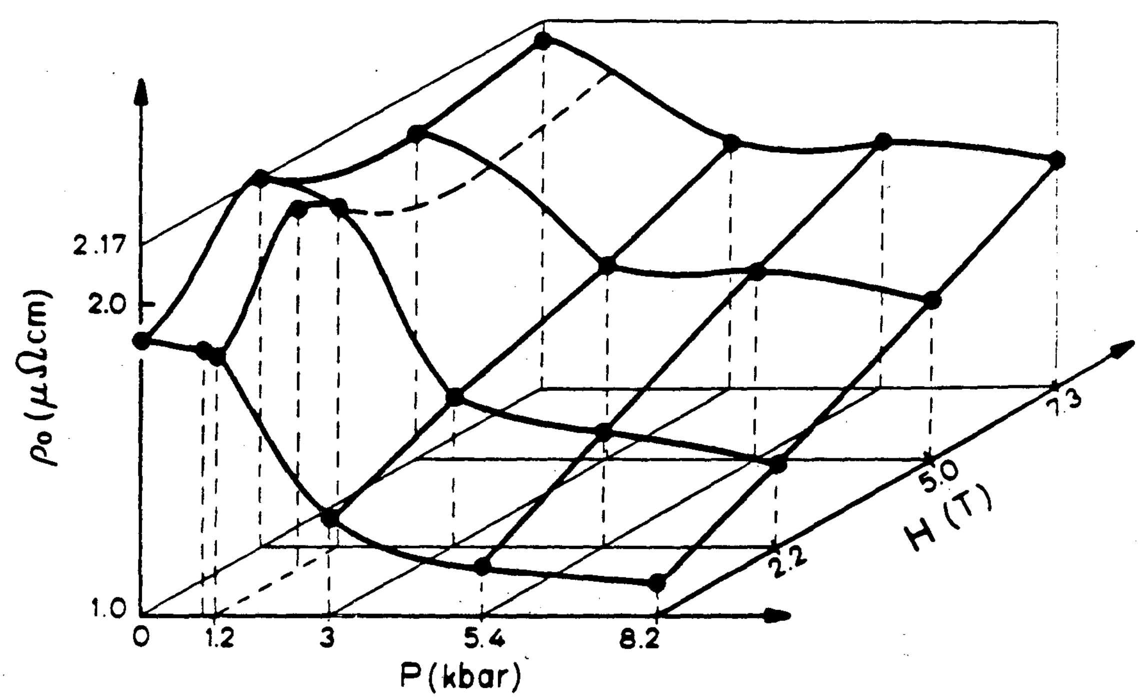


0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
γ (J mole⁻¹ K⁻²)
Ce1-xLaxRu2Si2
x=0.05
x=0.0
8
6
4
2
0
Tmα (K)
x=0.0
0
0.5
1.0
1.5
H/HM

FIG. 5



38.9
A (μΩcmK⁻²)
2.0
0
1.2
3
5.4
8.2
P (kbar)
2.2
5.0
7.3
H (T)



2.17
2.0
ρ₀ (μΩcm)
1.0
0
1.2
3
5.4
8.2
P (kbar)
2.2
5.0
7.3
H (T)

FIG. 6