PHYSICAL REVIEW B VOLUME 36, NUMBER 10 1 OCTOBER 1987

Fermi-liquid scaling in the paramagnetic spectral response of the compounds $CeSn_{3-x}In_x$

A. P. Murani
Institut Laue-Langevin, 156X, 38042 Grenoble Cedex, France
(Received 5 May 1987)

The low-temperature paramagnetic spectra of the compounds $CeSn_{3-x}In_x$ $(0<x<1)$ show broad inelastic peaks at energies ω_i which scale with T_{max}, the temperature of the maxima in the bulk susceptibility, and with χ $(T\rightarrow 0)$ and γ $(=C/T)$, in quantitative agreement with Fermi-liquid relations. The spectra are well described by an analytic function $\chi''(\omega)$ for the degenerate Anderson model. Fits to the data suggest that, if $n_f\simeq 1$, the ground-state degeneracy of Ce is close to 6 $(=2J+1)$.

A number of valence-fluctuation (intermediate valence), Kondo-lattice (heavy-fermion) systems have been investigated by neutron scattering techniques in recent years. Valence-fluctuation systems like Ce-Th,[1] $CeSn_3$, and $CePd_3$ (Ref. 2) show broad quasielastic spectral distributions of half-width typically 25–30 meV at high temperatures (above >100 K) while at lower temperatures one observes inelastic humps in the magnetic response, as, for example, at ~ 40 meV and ~ 55 meV in $CeSn_3$ (Ref. 3) and $CePd_3$,[4] respectively. In another IV system, namely $YbAl_3$, the form of the magnetic response at low temperature shows a steplike rise around ~ 32 meV, with the paramagnetic spectrum extending to >100 meV.[5] In Kondo-lattice or heavy-fermion systems, i.e., those with large values of γ, such as $CeAl_3$, crystal-field excitations are observed in addition to a relatively narrow ($\Gamma\sim 0.5$ meV) residual quasielastic distribution (as $T\rightarrow 0$ K).[6] The temperature dependence of the latter follows a $T^{1/2}$ law as found also in other heavy-fermion systems $CeCu_2Si_2$ and $CeCu_6$.[7,8] For the latter compound, i.e., $CeCu_6$ measurements on a single-crystal sample show strong Q dependence of the magnetic response due to antiferromagnetic correlations, although the compound does not order magnetically down to very low temperatures.[9,10]

While neutron scattering data are available for a variety of valence-fluctuation and Kondo-lattice systems, there have been relatively fewer neutron studies of a given system as it evolves from a valence fluctuation towards a Kondo-lattice or heavy-fermion state as a function of a parameter such as the average conduction electron per atom ratio, as, for example, in systems like $CePd_{3-x}Ag_x$ (Ref. 11) and $CeSn_{3-x}In_x$.[12,13] The latter system is particularly interesting as it shows a homogeneous evolution from the IV state of the Sn-rich end towards Kondo-lattice behavior for $x\leq 1.9$. In the region $x\leq 1$, the susceptibility and electronic specific-heat coefficient γ (Ref. 14) as well as T_{max}, the temperature of the maximum in the bulk susceptibility vary roughly linearly with x. Also $\chi(0)$, the magnitude of χ as $T\rightarrow 0$ K, and γ, the coefficient of the linear (electronic) term in the specific heat, attain their maximum values around $x=1.1$ [γ varies from 60 mJ mole^{-1}K^{-2} at $x=0$ to ~ 270 mJ mole^{-1}K^{-2} at $x=1.1$].[14,15] The present study is restricted to this concentration range $(0<x<1)$ where the magnetic properties evolve linearly with x.

The normalized spectra measured at 5 K with the time-of-flight technique using neutrons of incident energy 50 meV were converted to $S(\theta,\omega)$ after correction of empty cell scattering in the usual manner.[3] The results are shown in Fig. 1 for three $CeSn_{3-x}In_x$ compounds with $x=0.5$, 0.75, and 1.0. Also included in the figure is the spectrum $S(\theta,\omega)$ for a $LaSn_2In$ sample measured in an identical manner, which shows the typical phonon contribution. It is clear that the magnetic scattering in the cerium-based samples is largely dominant. Phonon corrections were, however, applied to extract the purely magnetic contribution using the scaling procedure adopted previously.[3] The spectra corrected for phonons as well

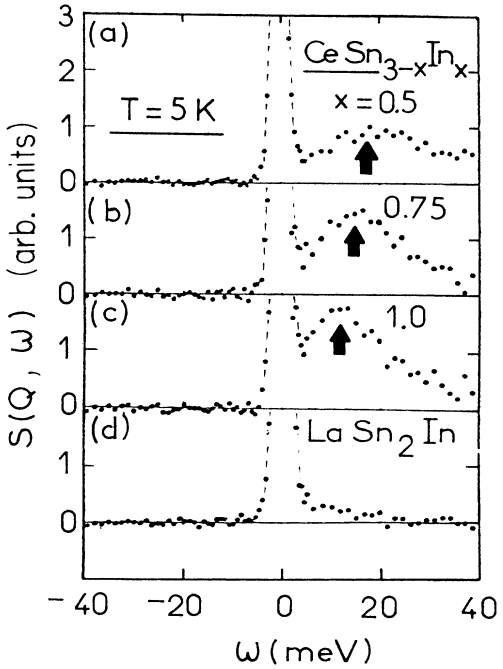

FIG. 1. The observed spectra $S(\theta,\omega)$ vs $\omega(2\theta=10°)$ at 5 K for the $CeSn_{3-x}In_x$ compounds with $x=0.5$, 0.75, and 1.0 as well as a $LaSn_2In$ sample. The arrows indicate the position ω_i of the peaks in the spectral distributions.

as variation of intensity with energy transfer ω for fixed scattering angles 2θ using the Ce^{3+} form-factor dependence[16,17] are shown in Fig. 2.

An important question regarding the present results concerns the origin of the inelastic peaks in these compounds. As pointed out some years ago, the magnetic resistivity of both $CeSn_3$ and $CeSn_2In$ (Ref. 18) bear strong similarity to the behavior expected for a Kondo system in the presence of crystalline electric field (CEF) splitting hence, naturally, the question arises whether the observed inelastic peaks represent crystal-field excitations. However, as discussed previously for $CeSn_3$,[3] it is rather difficult to reconcile the observed neutron spectra with a crystal-field description, for in a cubic crystal field the $2J+1$ $(=6)$ fold degenerate Ce^{3+} state should split into a Γ_7 doublet and a Γ_8 quartet states so that we should expect to observe both an inelastic as well as a quasielastic (or elastic) spectral components with relative weights in the ratio of the appropriate transition probabilities. If we analyze the observed spectra in terms of the usual function consisting of Lorentzian quasielastic and inelastic spectral components, namely,

$$S(Q,\omega)=\left[\frac{\omega}{1-\exp(-\omega/T)}\right]\left\{\frac{A}{\pi}\left[\frac{\Gamma_0}{\Gamma_0^2+\omega^2}\right]+\frac{B}{2\pi}\left[\frac{\Gamma_i}{\Gamma_i^2+(\omega+\omega_i)^2}+\frac{\Gamma_i}{\Gamma_i^2+(\omega-\omega_i)^2}\right]\right\}, \tag{1}$$

where the denominator inside the square brackets is the detailed balance factor, ω_i the position of the inelastic peak, Γ_0 and Γ_i the half-widths of the quasielastic and inelastic components, and A and B their respective Kramers-Kronig weights; we find that the ratio A/B for the $x=0.5$ and the $x=0.75$ samples is equal to 5/8 and 3/26, respectively, while for the $CeSn_2In$ sample $(x=1)$ the best fit is obtained with $A=0$, i.e., there is negligible quasielastic scattering. We note that there is no resolution limited elastic magnetic scattering either, as cross checked by comparison of the elastic cross sections of the various samples including $LaSn_2In$, taking account of the appropriate incoherent cross sections of Ce, La, In, and Sn. The absence of the quasielastic (or elastic) magnetic scattering in the $CeSn_2In$ sample clearly rules out crystal-field interpretation. Indeed, as discussed below, this conclusion is also consistent with the results of form-factor measurements on $CeSn_3$ (Ref. 16) and particularly $CeSn_2In$,[17] which show no anisotropy of crystal-field origin similar to that found, for example, in $CeIn_3$,[19] which has the Γ_7 doublet ground state.

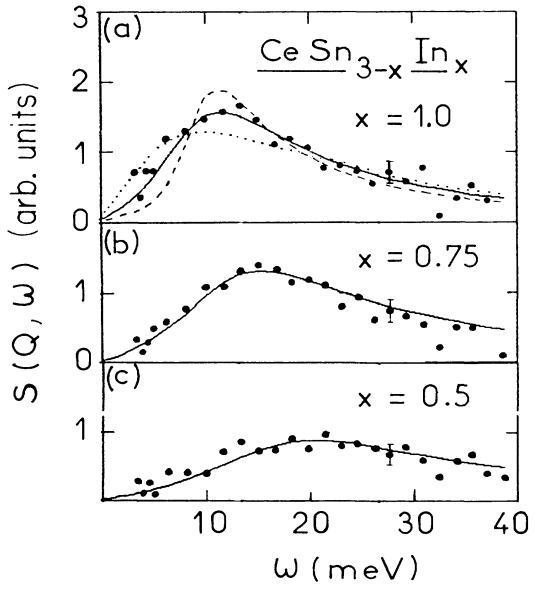

FIG. 2. $S(\theta,\omega)$ vs $\omega(2\theta=10°)$ at 5 K for $CeSn_{3-x}In_x$ samples corrected for phonons and Ce^{3+} form factor dependence. The solid curves represent the best fits to the data with $n_f=1$, $N=6$, and $E_f=9\pm1$, 12 ± 1, and 16 ± 2 for the $x=0.5$, 0.75, and 1.0 samples, respectively. In (a) the dotted and dashed curves represent the best fits with $N=2$ and $N=14$, respectively.

In Table I we list the values of the energies ω_i determined by fits to Eq. (1) and the temperature T_{max} for the series of $CeSn_{3-x}In_x$ compounds which suggest that $\omega_i \sim 4T_{max}$, in reasonable accord with Schlottmann's calculations.[24] In the latter model the energy ω_i of the peak (hump) corresponds to the energy of excitation of the f electron (hole) to the Fermi sea. Kuramoto and Müller-Hartmann[25] give a similar interpretation for ω_i which, except for a logarithmic term, is equal to the position ε_f^* of the renormalized f level relative to the Fermi energy, i.e., $\varepsilon_f^* = \omega_i + \ln(\omega_i)$. Alternatively, according to the authors, the energy ω_i represents the Kondo temperature T_K, as suggested also by Gunnarson and Schönhammer.[26] Thus, the results in Table I indicate that the Kondo temperature T_K is roughly equal to 4 times T_{max}, the temperature of the maximum in the susceptibility.

Fermi-liquid relations between $\chi(0)$, γ, and an energy parameter Δ, or T_K (or ω_i) have been derived by several authors. In terms of the Kondo temperature $T_K(=\omega_i)$

TABLE I. Positions ω_i of the inelastic peaks and the temperatures T_{max} of the maxima in the bulk susceptibility for some Ce-based systems.

Compound	ω_i (meV)	ω_i (K)	T_{max} (K)	ω_i/T_{max} ±0.5
$CePd_3$	55 ± 5[a]	640 ± 60[a]	130 ± 10[b,c]	4.9
$CeSn_3$	40 ± 5[d]	460 ± 60[d]	130 ± 10[e,f]	3.6
$CeSn_{2.5}In_{0.5}$	18 ± 2[g]	210 ± 20[g]	60 ± 5[h,i]	3.5
$CeSn_{2.25}In_{0.75}$	13 ± 1[g]	150 ± 10[g]	38 ± 5[h,i]	3.9
$CeSn_2In$	9 ± 1[g]	104 ± 10[g]	25 ± 3[h,i]	4.0

[a]Reference 4.
[b]Reference 20.
[c]Reference 21.
[d]Reference 3.
[e]Reference 22.
[f]Reference 23.
[g]This work.
[h]Reference 12.
[i]Reference 13.

TABLE II. Predicted and observed values of $\Delta = |\tilde{\varepsilon}_0 - \tilde{\varepsilon}_f| = \omega_i$; Δ_χ from susceptibility; Δ_γ from specific heat.

Compound	$\chi(0)$ (10^3 emu mole^{-1})	γ (mJ mole^{-1}K^{-2})	Δ_χ (meV)	Δ_γ (meV)	Δ_{obs} (meV)
$CePd_3$	1.2[a]–1.4[b]	39[c]	58–50	60	55 ± 5[d]
$CeSn_3$	1.7[e]–2.0[f]	57[g]–68[h]	41–35	41–35	40 ± 5[i]
$CeSn_{2.5}In_{0.5}$	3.3[j]	. . .	20.0	. . .	18 ± 2[k]
$CeSn_{2.25}In_{0.75}$	5.4[j]	190[h]	12.8	12.5	13 ± 1[k]
$CeSn_{1.9}In_{1.1}$	8.0[j]	270[h]	8.6	8.8	
$CeSn_2In$	8.0[j]	. . .	8.6	. . .	9 ± 1[k]

[a]Reference 20.
[b]Reference 21.
[c]Reference 29.
[d]Reference 4.
[e]Reference 22.
[f]Reference 23.
[g]Reference 15.
[h]Reference 14.
[i]Reference 3.
[j]References 12 and 13.
[k]This work.

Kuramoto and Müller-Hartmann[25] obtain

$$\chi(0) = \frac{N\mu^2}{3\pi k_B T_K} \sin\left(\frac{\pi n_f}{N}\right), \tag{2}$$

where $\mu^2 = (g_J \mu_B) J(J+1)$, n_f is the $4f$ occupancy, and N the degeneracy of the $4f$ level. An identical relationship has been derived by Newns, Hewson, Rasul, and Read[27] for a resonant f level of bandwidth Δ, with T_K replaced by Δ in Eq. (2). For large $N(\geq 6)$ or small n_f, these reduce to the Fermi-liquid relations of Ramakrishnan and Sur,[28] namely,

$$\chi(0) = \frac{\mu^2}{3k_B |\tilde{\varepsilon}_0 - \tilde{\varepsilon}_f|} n_f \text{ and } \gamma = \frac{\pi^2}{3} \frac{k_B}{|\tilde{\varepsilon}_0 - \tilde{\varepsilon}_f|} n_f, \tag{3}$$

where $|\tilde{\varepsilon}_0 - \varepsilon_f|$ is the energy difference between the renormalized f levels f^0 and f^1. If we identify this energy parameter with the observed ω_i's we obtain an excellent quantitative agreement with the values of $\chi(0)$ and γ assuming $n_f = 1$. The results are shown in Table II.

The form of the paramagnetic response for the $CeSn_{3-x}In_x$ compounds $(0 < x < 1)$ and particularly that for $CeSn_3$ (Ref. 3) is qualitatively similar to that calculated numerically by Schlottmann[24] for an Anderson impurity in the $U \rightarrow \infty$ limit. Also, single-ion model calculation of the Anderson Hamiltonian by Cox, Bickers, Wilkens,[30] and Kuramoto and Kojima[31] as well as Gunnarson and Schönhammer[26] predict a spectral shape similar to that observed for these compounds. We have attempted to fit the spectra to the analytic form of $\chi''(\omega)$ given by Kuramoto and Müller-Hartmann,[25] namely,

$$\chi''(\omega) = C \frac{N\omega}{n\tilde{E}_f^2} \frac{\sin\alpha}{u^2(u^2 + 4\sin^2\alpha)} \left\{ \sin\alpha \ln[(1-u^2)^2 + 4u^2\sin^2\alpha] + |u| \left[\frac{\pi}{2} - \tan^{-1}\left(\frac{1-u^2}{2|u|\sin\alpha} \right) \right] \right\} \tag{4}$$

where $\tilde{E}_f$ is a characteristic energy closely related to the Kondo temperature T_K, $u = \omega/\tilde{E}_f$, $\alpha = \pi n_f/N$, and $C = \frac{1}{3}(g_J\mu_B)^2 J(J+1)$.

We have proceeded to fit the data for the $CeSn_{3-x}In_x$ samples keeping $\alpha(=\pi n_f/N)$ constant at various values corresponding to $N = 2$, 4, 6, or 14 and $n_f = 1$. Overall best fits resulted for α values corresponding to 4 and 6, while for both $N = 2$ and 14 the resultant spectral shapes deviate strongly from the observed form, as shown by the dotted and dashed curves in Fig. 2. Of the two possible values of N (i.e., 4 or 6) we found that for the energy region $\omega > E_f$ the fits were distinctly better for $N = 6$ although, in the low-energy region the fitted curves deviate slightly below the data points. This, however, is to be expected since, as pointed out by Kuramoto and Müller-Hartmann,[25] their analytical spectral function is accurate for $\omega > E_f$, but gives an underestimate relative to the exact Bethe-ansatz result as $\omega \rightarrow 0$. Good fits with $N = 6$ are obtained for all three samples except for small discrepancies in the low-energy region yielding values of E_f of 9 ± 1, 12 ± 1, and 16 ± 2, meV, respectively. We conclude from the results that the most appropriate value of the ground-state degeneracy N in this compound is close to 6, in good agreement with the form-factor studies of $CeSn_3$ and $CeSn_2In$[16,17] where no evidence of anisotropy of crystal-field origin is found, in contrast to the case of $CeIn_3$,[19] for example, where the amplitudes of the $(h00)$ reflections are higher than for the $(0kk)$ reflections consistent with the Γ_7 doublet ground state in this compound. The opposite would hold for the case of Γ_8 quartet.

In conclusion, the present results on the series of $CeSn_{3-x}In_x$ compounds reveal inelastic peaks (humps) whose energies ω_i scale quantitatively with $\chi(0)$ and γ (as well as T_{max}) via the Fermi-liquid relations.[25,27,28] This, to our knowledge, is the first demonstration of such a systematic variation. If the energies ω_i are identified with

the Kondo temperature T_K the results suggest that $T_K \sim 4T_{max}$. Finally, the form of the observed spectra can be reasonably well accounted for by the analytic formula obtained by Kuramoto and Müller-Hartmann[25] which leads to the conclusion that, assuming the $4f$ occupancy $n_f \simeq 1$, the degeneracy N is close to 6, i.e., the ground state is fully degenerate.

It is a pleasure to acknowledge helpful discussions and correspondence with G. W. Crabtree, S. Doniach, D. M. Edwards, E. Müller-Hartmann, J. Pierre, and S. M. Shapiro. Assistance of R. Raphel and M. Maeder with sample preparation and bulk susceptibility measurements, respectively, is also gratefully acknowledged.

[1]S. M. Shapiro, J. D. Axe, R. J. Birgeneau, J. M. Lawrence, and R. D. Parks, Phys. Rev. B **16**, 2225 (1977).
[2]E. Holland-Moritz, M. Loewenhaupt, W. Schmatz, and D. K. Wohlleben, Phys. Rev. Lett. **38**, 983 (1977).
[3]A. P. Murani, Phys. Rev. B **28**, 2308 (1983); J. Phys. C **33**, 6359 (1983).
[4]R. M. Galera, A. P. Murani, J. Pierre, and K. R. A. Ziebeck, J. Magn. Magn. Mater **63–64**, 594 (1987).
[5]A. P. Murani, Phys. Rev. Lett. **54**, 1447 (1985).
[6]A. P. Murani, K. Knorr, K. H. J. Buschow, A. Benoit, and J. Flouquet, Solid State Commun. **36**, 523 (1980).
[7]S. Horn, E. Holland-Moritz, M. Loewenhaupt, F. Steglich, H. Scheuer, A. Benoit, and J. Flouquet, Phys. Rev. B **23**, 3171 (1981).
[8]S. M. Shaprio, Physica B **136**, 365 (1986), and references therein.
[9]G. Aeppli, M. Yoshikawa, Y. Endoh, E. Bucher, J. Hutnagl, Y. Onuki, and T. Komatsubara, Phys. Rev. Lett. **57**, 122 (1986).
[10]L. P. Regnault, W. A. C. Erkelens, J. Rossat-Mignot, J. Flouquet, E. Walker, D. Jaccard, A. Amato, and B. Hennion, J. Magn. Magn. Mater. **63–64**, 289 (1987).
[11]T. Mihalisin, P. Scoboria, and J. A. Ward, Phys. Rev. Lett. **46**, 862 (1981).
[12]J. Lawrence, Phys. Rev. B **20**, 3770 (1979).
[13]W. H. Dijkman, F. R. de Boer, P. F. de Chatel, and J. Aarts, J. Magn. Magn. Mater. **15–18**, 970 (1980).
[14]R. A. Elenbaas, C. J. Schinkel, and C. J. M. Van Deudekom, J. Magn. Magn. Mater. **15–18**, 979 (1980).
[15]J. R. Cooper and C. Rizzuto, J. Phys. (Paris) Colloq. **32**, C1-1136 (1971).
[16]C. Stassis, C. K. Loong, B. N. Harmon, and S. H. Liu, J. Appl. Phys. **50**, 7567 (1979).
[17]A. Benoit, J. X. Boucherle, J. Flouquet, J. Sakurai, and J. Schweizer, J. Magn. Magn. Mater. **47–48**, 149 (1985).
[18]A. Maury, P. Scoboria, J. E. Crow, and T. Mihalisin, J. Appl. Phys. **50**, 7572 (1979).
[19]J. X. Boucherle, J. Flouquet, Y. Lassailly, J. Palleau, and J. Schweizer, J. Magn. Magn. Mater. **31–34**, 409 (1983).
[20]W. E. Gardner, J. Penfold, T. F. Smith, and I. R. Harris, J. Phys. F **2**, 133 (1972).
[21]J. P. Kappler, G. Krill, M. J. Besnus, M. F. Ravet, N. Handaoui, and A. Meyer, J. Appl. Phys. **53**, 2152 (1982).
[22]T. Tscuchida and W. E. Wallace, J. Chem. Phys. **43**, 3811 (1965).
[23]J. G. Sereni, J. Phys. F **10**, 2831 (1980).
[24]P. Schlottmann, Phys. Rev. B **25**, 2371 (1982); **29**, 4468 (1984).
[25]Y. Kuramoto and E. Müller-Hartmann, J. Magn. Magn. Mater. **52**, 122 (1985).
[26]O. Gunnarson and K. Schönhammer, J. Magn. Magn. Mater. **52**, 227 (1985).
[27]D. M. Newns, A. C. Hewson, J. W. Rasul, and N. Read, J. Appl. Phys. **53**, 7877 (1982).
[28]T. V. Ramakrishnan and K. Sur, Phys. Rev. B **26**, 1798 (1982).
[29]R. D. Hutchins, V. V. S. Rao, J. E. Greedan, and R. S. Craig, J. Phys. Soc. Jpn. **32**, 451 (1972).
[30]D. L. Cox, N. E. Bickers, and J. W. Wilkins, J. Magn. Magn. Mater. **54–57**, 333 (1983).
[31]Y. Kuramoto and H. Kojima, Z. Phys. F **10**, 2429 (1980).