Pergamon

Solid State Communications, Vol. 101, No. 11, pp. 825–829, 1997

0038–1098/97 $17.00+.00

PII: S0038–1098(96)00231-4

MAGNETIC SPECTRAL RESPONSE OF CERIUM BASED KONDO SYSTEMS

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(*Received* 14 *September* 1996; *in revised form* 11 *November* 1996 *by C.N.R. Rao*)

Inelastic neutron scattering studies on two cerium Kondo systems, CeSiGa and $CeSn_2In$ are reported. In CeSiGa the thermal evolution of the quasielastic line width indicates a strong competition between Kondo and RKKY interaction and the spectral response shows only one inelastic peak, the possible reasons for this behaviour are discussed. In the case of $CeSn_2In$ our results agree well with those of previous workers.

1. INTRODUCTION

Inelastic neutron scattering is one of the most powerful tools to investigate crystal field and magnetic excitations in solids. A large number of valence fluctuating, heavy fermion or Kondo lattice compounds have been studied by neutron scattering technique. In case of valence fluctuating systems, the quasielastic line-width is independent of temperature whereas for Kondo lattice compounds it follows a $\sqrt{T}$ dependence for non-magnetic state and a linear temperature dependence for magnetically ordered state. In this paper we report our measurements on two cerium based Kondo systems, $CeSn_2In$ and CeSiGa, using the triple axis spectrometer (TAS) at Dhruva reactor.

CeSiGa is a ferromagnetic dense Kondo system belonging to $CeSi_{2-x}Ga_x$ series which is reported to show a interesting interplay between the Kondo and the RKKY type interactions. The magnetization, specific heat and resistivity measurements suggest that the ground state is spit by the crystalline electric field (CEF) in three doublets [1, 2]. Moschalkov *et al.* [2] have analysed the resistivity curve by assuming 160 K and 330 K splitting between the CEF split ground state and the two excited states. This assumption implies two Kondo temperatures for the system, $T_K^h = 106$ K and $T_K^l = 13.8$ K. Very close values of T_K^l and the Curie temperature, T_C ($\approx$13 K) suggest that there is a strong competition between the magnetic interaction and the Kondo spin fluctuations in this system.

For comparison, the spectral response of $CeSn_2In$ is also presented in this paper. $CeSn_2In$ is a well characterized non-magnetic Kondo system belonging to the series $CeSn_{3-x}In_x$ [3]. Neutron inelastic measurements have been carried out [4, 5] on $CeSn_2In$, both on a polycrystalline sample and a single crystal. These show a 7 meV broad inelastic peak at about 9 meV energy transfer.

2. EXPERIMENTAL

The two systems studied here along with their La based isostructural compounds, viz $LaSn_2In$ and LaSiGa were prepared by arc melting, under argon atmosphere, appropriate amounts of rare earths, Sn, In, Si and Ga to obtain about 15–20 g of sample. The alloy buttons were remelted several times to homogenise the samples. All the samples were checked by neutron diffraction to confirm their phase purity. Inelastic neutron scattering measurements were carried out on triple axis spectrometer installed on T1007, tangential thermal neutron beam hole. The spectrometer, designed to operate at a medium resolution, employs a Cu(1 1 1) plane as monochromator and Si(1 1 1) plane as an analyser. The collimations used are open inpile, 60′ between monochromator and sample, 60′ between sample and analyser and open between analyser and detector [6, 7]. The spectrometer was operated at a constant final energy, $E_f = 25$ meV. The measurements were made in neutron energy loss mode at constant scattering angle. The data were recorded at various temperatures between 10 and 300 K using a closed cycle refrigerator. The recorded data were corrected for background signal and phonon scattering which was estimated using the La compounds as per the standard procedure [8]. The pure magnetic response thus obtained can be related to the imaginary part of the dynamical susceptibility, Im $\chi(E)$, $\chi''(Q,E)$,

$$S(Q,E) = \left[\frac{1}{1-\exp(-\hbar\omega/k_BT)}\right] f^2(Q)\chi''(Q,E). \quad (1)$$

The quantity in the square bracket is the detailed balance factor and $f(Q)$ represents the magnetic form factor of Ce^{3+} ion.

3. RESULTS AND DISCUSSION

3.1. *$CeSn_2In$*

The pure magnetic response of $CeSn_2In$ at 12 K is shown in Fig. 1. The experimental data (circles) shows a single peak centered at about 8 meV. This data was fitted to an analytical expression given by Kuramoto and Müller-Hartmann [KMH] [9] for the dynamical susceptibility of a degenerate Anderson lattice.

$$\chi''(Q,E) = \frac{CNE}{\pi\tilde{E}_F^2}\frac{\alpha}{u^2(u^2+4\alpha^2)} \times\left\{\alpha\ln[(1-u^2)^2+4u^2\alpha^2]+|u|\left[\frac{\pi}{2}-a\tan\left(\frac{1-u^2}{2|u|\alpha}\right)\right]\right\}, \quad (2)$$

where $u = E/\tilde{E}_F$ and $\alpha = \sin(\pi n_f/N)$ with $\tilde{E}_F$ is a characteristic energy closely related to Kondo temperature, n_f is the occupancy and N is the degeneracy of the $4f$ level.

According to this expression, the presence of the inelastic peak in Im $\chi(\omega)$ represents a quasi-threshold energy $\tilde{E}_F$ of magnetic excitations and the $4f$ electrons with their energy $E > \tilde{E}_F$ get an additional decay channel into the conduction band coupled through magnetic excitations. The fitting was done assuming $n_f = 1$ and

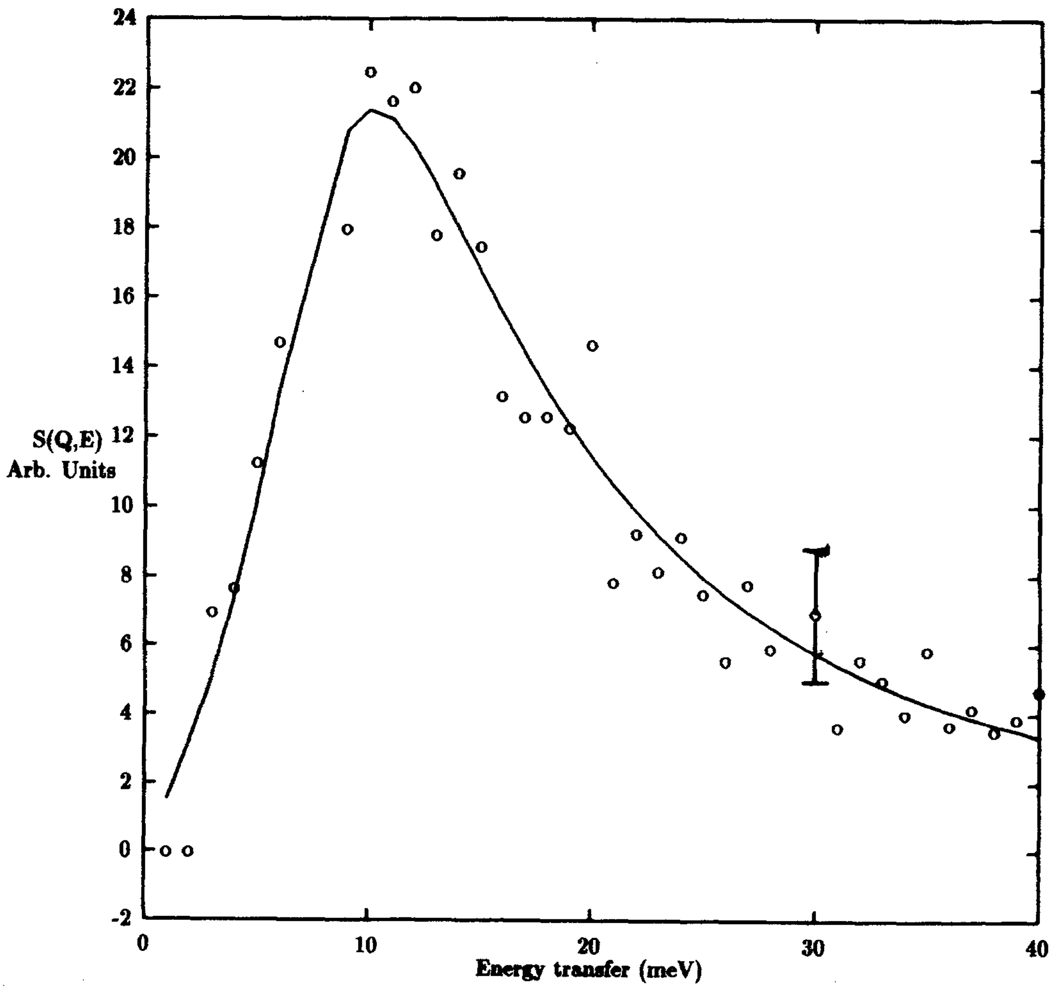

Fig. 1. Magnetic spectral response of $CeSn_2In$. Circles are experimental data points and solid line represents KMH fit.

by keeping the value of α constant at a value corresponding to $N = 6$. Thus there is only one free parameter namely $\tilde{E}_F$ and its value was obtained as 8.2 ± 1.3 meV. The fit, in the low energy region, slightly deviates from the experimental data. This is explained by KMH themselves, that their analytical expression is accurate for $E > \tilde{E}_F$ but under estimates with respect to Bethe Ansatz results as $E \rightarrow 0$. The values of n, N and $\tilde{E}_F$ are in good agreement with those reported in literature [3].

3.2. *CeSiGa*

In the case of CeSiGa the neutron spectra were recorded at various temperatures (10 K, 25 K, 50 K and 100 K). The spectrum shows a quasielastic peak and an inelastic peak centered around 15 meV energy transfer. It was difficult to estimate the width of the quasielastic peak at $T = 12$ K with $E_f = 25$ meV as it was just within the energy resolution of the spectrometer. We have therefore repeated the experiment in quasielastic region with $E_f = 20$ meV. Figure 2 shows the total magnetic response of CeSiGa at 12 K. Solid line represents the least-squares fit to the imaginary part of the susceptibility which can be expressed via the Krammers Kronig relation as

$$\chi''(Q,E) = \hbar E \chi(Q) F(Q,E), \tag{3}$$

where $\chi(Q)$ is the static susceptibility and $F(Q,E)$ is the normalized spectral response given by

$$\chi(Q)F(Q,E) = \frac{A_0(T)\Gamma_0(T)}{\Gamma_0^2(T) + E^2} + \sum_1^n \frac{A_i(T)\Gamma_i(T)}{\Gamma_i^2(T) + (E \pm E_i)^2}, \tag{4}$$

where A_0, A_1 are the amplitudes and Γ_0, Γ_i the half width of the quasielastic and inelastic lines respectively.

In Table 1 we give the fitted parameters at all the temperatures. The data in all the cases have been fitted to two lorentzians, one centered at zero energy (Quasielastic peak) and the other at 15 meV energy transfer, the inelastic peak. A typical fitted profile is shown in

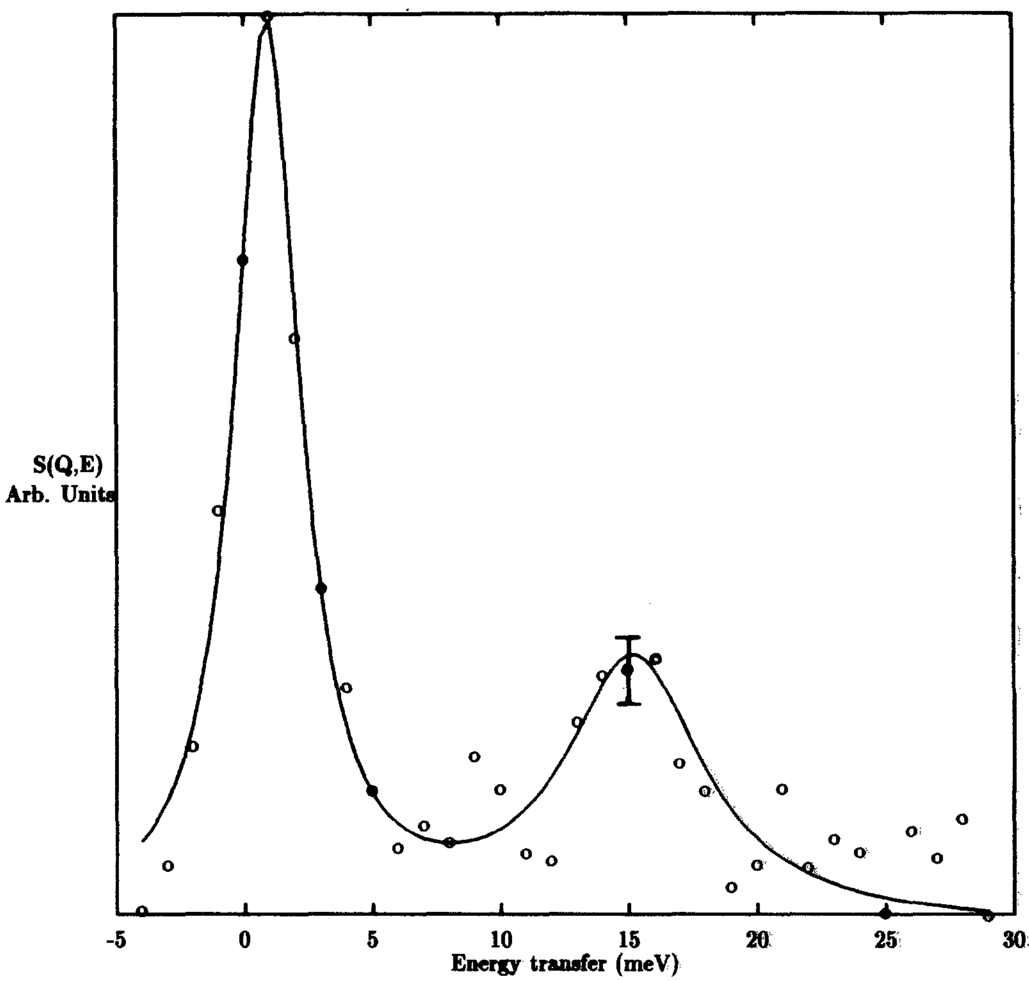

Fig. 2. Magnetic spectral response of GeSiGa. The solid line represents the least squares fit to the experimental data points (circles).

Table 1. Values of positions (E_i), amplitudes (A_i) and widths (Γ_i) of the Lorentzians in the fits to the magnetic response of CeSiGa

T (K)	E_i (meV)	A_i (arb. unit)	Γ_i (meV)
12	0 ± 1	155.24	3.1 ± 1
	15.3 ± 2	48.00	11.6 ± 3
25	0 ± 1	109.30	3.8 ± 1
	14.8 ± 2	31.42	13.6 ± 3
50	0 ± 1	86.4	4.6 ± 1
	15.2 ± 2	20.60	16.4 ± 3
100	0 ± 1	80.00	5.8 ± 1
	15.4 ± 2	12.00	19.5 ± 3

Fig. 2. Both the quasielastic and inelastic peak broaden out with reduction in their amplitudes as the temperature is increased. In Fig. 3 we have shown the thermal evolution of the quasielastic linewidth. The data can be fitted to both curves, $\Gamma = \Gamma_0 + AT$ (dashed line) and $\Gamma = \Gamma_0 + AT^{1/2}$ (solid line). Both the fits are found to be almost equally good with the $\sqrt{T}$ being slightly better and therefore it is difficult to say about the exact nature of thermal evolution of the quasielastic peak. This may perhaps be due to the competition between the single site

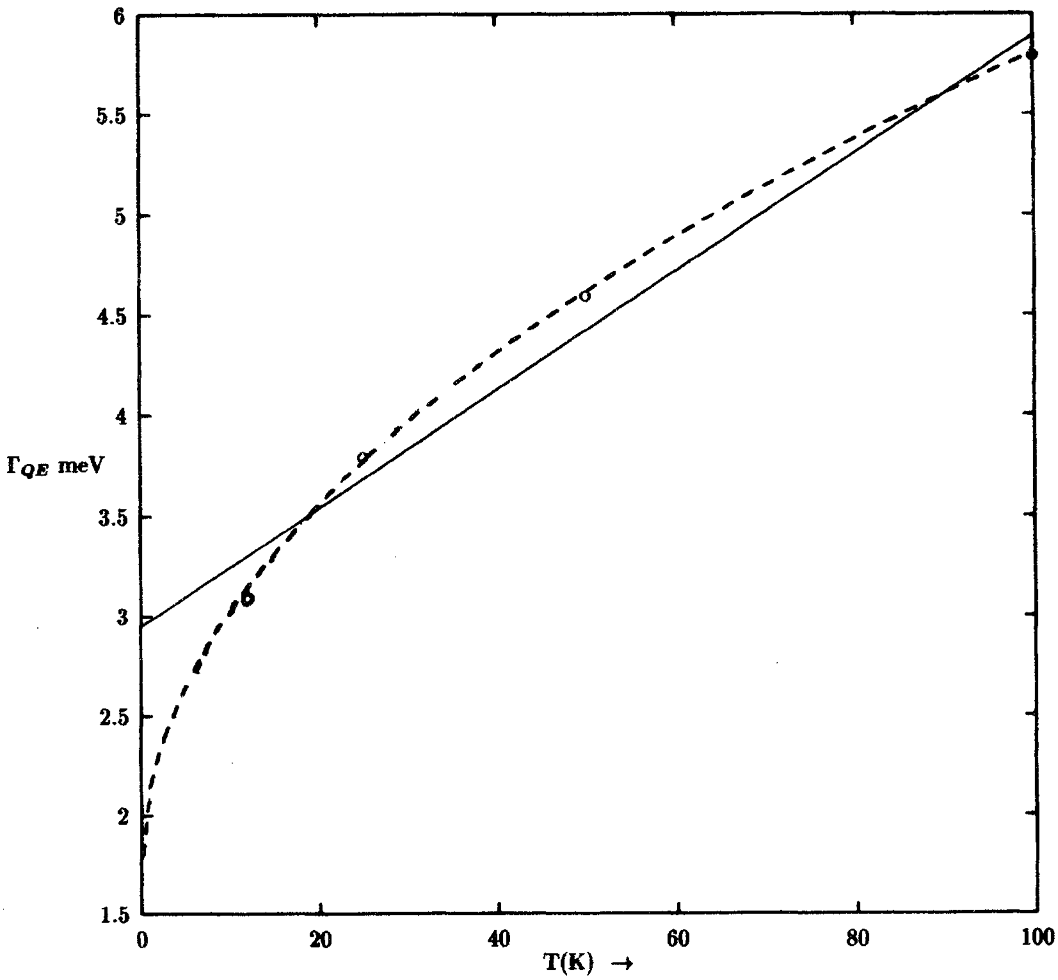

Fig. 3. Plot of quasielastic line width with temperature. Solid line represents the $T^{1/2}$ fit and the dashed line represents the T fit.

Kondo interactions and the intersite RKKY interactions. Using the $T^{1/2}$ curve the Kondo temperature estimated was $T_K = 17$ K which is quite close to the value of T_K^l deduced by Moschalkov *et al.* [2].

We believe that the origin of the inelastic peak is due to CEF splitting of the Ce^{3+} ground state. In the case of tetragonal symmetry the CEF splits the $J = 5/2$ multiplet into three CEF doublets. The inelastic peak at 15 meV is very close to 160 K, the first CEF split level as proposed by Moschalkov *et al.* [2], however, we do not see any excitation at 330 K. This could be due to two possibilities. Firstly, as in the case of $CeCu_2Si_2$ and $CeCu_2Ge_2$ [10, 11], the ground state even in tetragonal point symmetry can split into a doublet and a quasi-quartet with the CEF transition having a large width. This can be related to the strong interaction between the *f*-electrons with conduction band which is related to the increase in number of hybridization channels with high degeneracy of the excited level [12]. Secondly, it could be due to strong competition between the Kondo and the RKKY interactions wherein the intensity of the CEF excitations get reduced and due to which it is difficult for us to see the second CEF excitation. Such a behaviour is observed in ferromagnetic $CeNi_{0.8}Pt_{0.2}$ [13].

We are now extending our study to other members of the series, $CeSi_{2-x}Ga_x$ and this is expected to make it possible to propose the CEF model for this series.

Acknowledgements—We [R.B.P., P.R.S. and K.R.P.] are greatly indebted to IUC-DAEF, Indore for the financial assistance under the projects IUC/BOM/02 and IUC/BOM/48. We would also like to thank Drs B.A. Dasannacharya, Prof. B. Srinivasan and Dr K.R. Rao for their constant encouragement throughout the work.

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