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INELASTIC SCATTERING OF THERMAL NEUTRONS ON HEAVY FERMION SYSTEM $CeAl_3$

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Intermetallic compound $CeAl_3$ is one of the most representative members of the fast enriching class of heavy fermion systems. These systems are characterized by a large effective mass of conduction electrons near the Fermi level[1].

In this work we present the results of measurements of inelastic scattering of thermal neutrons (INS) on the $CeAl_3$, $Ce_{0.97}Pr_{0.03}Al_3$ and $LaAl_3$ compounds, using the time-of-flight spectrometer in inverted geometry with beryllium filter and pyrollitic graphite (as monochromator) in front of the detector at the IBR-2 pulsed reactor[2]. The resolution on the elastic line was 0.6 meV; and the regime of measurements, energy loss due to the scattering process.

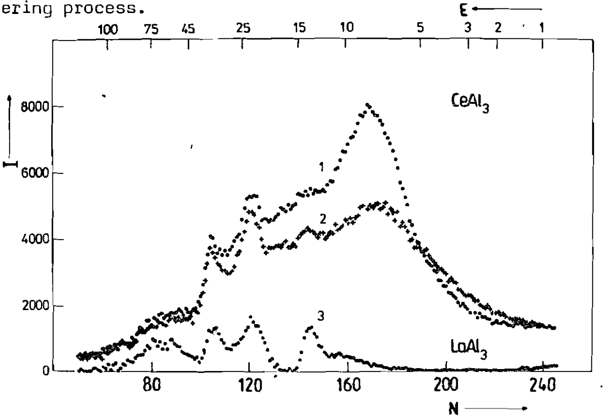

Fig. 1. Inelastic scattering spectra on $CeAl_3$ sample at 10 K (1), 77 K (2) and on LaAl3 sample at 77 K (3). The three scattering angles were summed up: φ = 30°, 50°, 70°. The background is subtracted. $\mathcal{E}$ -energy transfer in meV; N - channel number of 128 μs width. On the ordinate (I) - the number of counts per channel.

The neutron scattering spectra for $CeAl_3$, at 10 K, 77 K, together with those for $LaAl_3$ at 77 K are shown in fig. 1. The measurements on $LaAl_3$ have been performed to estimate the phonon contribution to the $CeAl_3$ spectrum. Two isostructural compounds have similar values of lattice parameters, but La has no magnetoactive 4f electrons. The lanthanum phonon cross section is three times larger than that of cerium, so that the $LaAl_3$ spectra (measured under the same conditions as those of $CeAl_3$) gave us an upper limit estimation of the phonon contribution in $CeAl_3$. Comparing $CeAl_3$ and $LaAl_3$ spectra which are shown in fig. 1 and also considering the observed temperature behaviour of intensity in $CeAl_3$ (it is increasing with decreasing temperature for an energy transfer greater than 5 meV), we may confirm the existence of a strong magnetic scattering up to 60 meV energy transfer. As is seen in the same figure, the $CeAl_3$ spectrum at 10 K contains a well-defined inelastic peak at the energy transfer $\approx$ 8 meV.

The double differential cross section (DDCS) for the paramagnetic scattering of unpolarized neutrons envolving the 4f-electron-crystalline electric field (CEF) interaction has the form[3]:

$$\frac{d^2\sigma}{d\Omega d\varepsilon} \sim \frac{k_1}{k_0} F^2(Q) \frac{\varepsilon}{1 - e^{-\varepsilon/k_B T}} \left\{ \sum_m \chi_c P^m_{mm}(\varepsilon, \Gamma_{mm}) + \frac{1}{2} \sum_{m \neq n} \chi^{mn}_W \left(1 - e^{-\frac{\Delta_{mn}}{k_B T}}\right) P_{mn}(\varepsilon - \Delta_{mn}, \Gamma_{mn}) \right\}, \quad (1)$$

where $\vec{k}_0$ and $\vec{k}_1$ are the wave vectors of incident and scattered neutrons, respectively; F(Q) - the magnetic form factor; χ_C^m and χ_W^{mn} the Curie and Van Vleck susceptibility; $P_{mn}(\mathcal{E}-\Delta_{mn}, \Gamma_{mn})$ the normalized to unity Lorentzian with the half-width at half-maximum Γ_{mn} ; Δ_{mn} the energy distance between the levels of the splitted ground multiplet in CEF.

The 4f-electrons-CEF interaction of Ce^{3+} ion in the hexagonal point symmetry case, splits the $^2F_{5/2}$ ground multiplet in three doublets between which the two transitions are possible observable at low temperature ($T < \Delta_{mn}$) by INS in the down scattering regime, if the ground level is $\Gamma_9\,|\pm 3/2\rangle$. In the above mentioned way the neutron scattering measurements were interpreted in /4/. There the data were prelucrated by decomposition of the spectrum into two Lorentzians and were established in addition to the inelastic peak at 7.6 meV the presence of a magnetic peak at 5.2 meV corresponding to the $\Gamma_9|\pm 3/2\rangle \to \Gamma_8|\pm 5/2\rangle$ transition. In our INS spectra we did not find such a line neither visually, nor by decomposition of the spectra in separated components, although the energy resolution was about two times better in this energy transfer range.

The solid line in fig. 2 was obtained by means of the least square fit of the experimental spectra of $CeAl_3$ at 10 K and 77 K by using the relation (1) and by taking into account the resolution function of the spectrometer /5/. For this there was assumed the existence of two quasielastic lines (dotted line 1,2 in fig.2) and one inelastic (dotted line 3 in fig.2) since only in this way we could obtain a satisfactory description of measured spectra. In order to get a quantitative estimation of the observed spectral component intensity (χ_C^m and χ_W^{mn} in relation (1)) of magnetic response function of $CeAl_3$ we have performed the INS measurement on

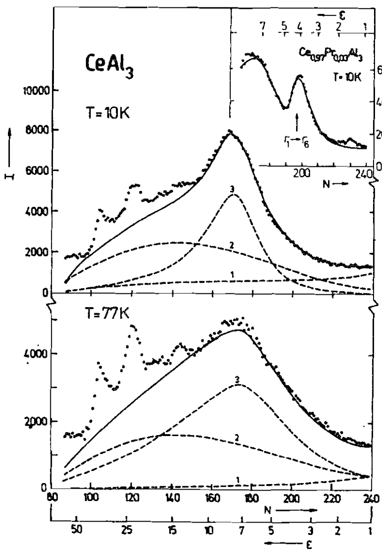

Fig. 2.

INS spectra of $CeAl_3$(points). Solid curves were obtained by convolution of expression (1) with the instrumental resolution using intensities, positions and widths of separate spectral components given in the table. Dotted 1,2 -- quasielastic, 3 -- inelastic component of the spectrum. The notations are the same as in fig. 1. The insert shows a fragment of inelastic spectrum on $Ce_{0.97}Pr_{0.03}Al_3$ at T=10 K. The solid line was calculated with the same spectral characteristics as for $CeAl_3$ and inelastic line due to scattering on $\Gamma_1 \to \Gamma_6$ transition of Pr^{+3} ion in CEF.

$Ce_{0.97}Pr_{0.03}Al_3$ compound at T=10 K. A fragment of the INS spectra on $Ce_{0.97}Pr_{0.03}Al_3$ sample is displayed in the insert of fig.2. There is the inelastic peak at the energy transfer $\Delta_{\Gamma_1 \to \Gamma_6}$ = 4.31 meV, which corresponds to the $\Gamma_1 \to \Gamma_6$ transition of the ground multiplet $^3H_4$ of Pr^{+3} ion splitted by CEF /6/. The intensity of this inelastic line is proportional to Van Vleck susceptibility of the system of $\Gamma_1 - \Gamma_6$ levels and yields : $\chi_W^{\Gamma_1-\Gamma_6} = 6.4\times10^{-2}\ \frac{EME}{mol}$. If all the factors entering the expression for DDCS (1) are taken into account, then one may use the Van Vleck susceptibility of the $\Gamma_1 - \Gamma_6$ transition and its corresponding intensity in the $Ce_{0.97}Pr_{0.03}Al_3$ spectrum to normalize the spectral component intensity of $CeAl_3$.

The magnetic response function characteristics of the $CeAl_3$ compound determined by the above mentioned procedure are summarized in the table. In the same table there

Table

	Δ,meV	Γ,meV	$\chi^N \times 10^{-2}\ \frac{emu}{mol}$	
T=10K	0	1,2 ± 0,3	1,1	$\chi^N = 2{,}1\times10^{-2}\ \frac{emu}{mol}$
	0	13 ± 2	0,6	
	7,6 ± 0,3	5,0 ± 0,5	0,4	$\chi^{bulk} = 2{,}2\times10^{-2}\ \frac{emu}{mol}$
T=77K	0	2,2 ± 0,5	0,1	$\chi^N = 0{,}85\times10^{-2}\ \frac{emu}{mol}$
	0	19 ± 3	0,3	
	5,4 ± 0,5	10 ± 1	0,45	$\chi^{bulk} = 0{,}75\times10^{-2}\ \frac{emu}{mol}$

are also listed the measured bulk susceptibility /7,8/. The magnitudes of the total static susceptibility determined by INS on $CeAl_3$(χ^N) are in good agreement with those obtained by magnetic measurements (χ^{bulk}). The fact indicates that the used analysis of data is adequate.

Unexpected for the magnetic response function parameter values in $CeAl_3$ are the two facts. Firstly : the existence of a single inelastic peak only. This contradicts with the widespread assertion that the ground state of $^2F_{5/2}$ multiplet in CEF of $CeAl_3$ is a doublet $\Gamma_9\,|\pm 3/2\rangle$ /4,8-11/, because there becomes possible one transition to the level $\Gamma_9\,|\pm 3/2\rangle$ only if the ground level is $\Gamma_7\,|\pm 1/2\rangle$. In the framework of the standard CEF-theory, the Van Vleck susceptibility for the $\Gamma_7 - \Gamma_9$ transition yields: $\chi_W^{\Gamma_7-\Gamma_9} = 0.83\times10^{-2}\ \frac{EME}{mol}$ which is two times larger than those measured by the INS at T=10 K.This divergence suggests possible that strong interaction in $CeAl_3$ of 4f localized electrons with the conduction electrons modifies the wave functions of 4f electrons in CEF and the usual CEF theory cannot be applied for to calculate the observed intensity of inelastic magnetic scattering.

The second peculiarity of the magnetic response function characteristics in $CeAl_3$ is the existence of two quasielastic components with a difference between their linewidths by an order of magnitude. Heavy fermion systems are characterized by anomalously large values of the electronic specific heat coefficient γ and of the magnetic susceptibility χ at $T\to0$ (for $CeAl_3$ $\gamma(0) = 1.62\ \frac{J}{mol\cdot K^2}$ /12/ $\chi(0) = 3.6\times10^{-2}\ \frac{EME}{mol}$ /12/) what corresponds to the resonance width near Fermi level of about 1 meV. The INS measurements on $CeAl_3$ /13/ and $CeCu_6$ /14/ samples, performed with a high resolution, but limited to a short energy transfer range (±2.5meV), pointed out that the quasielastic peak width had the value of 0.5 meV for $T\to 0$ which was in agreement with the estimation of the scale width on the basis of specific heat data ($\Gamma(T\to 0)\approx \gamma(0)^{-1} \approx$ 1 meV).

In our experiments the magnetic quasielastic scattering with a width of about 1 meV at T = 10 K was also observed. At the same time in the INS experiments on heavy fermion systems based on uranium (UBe_{13} /15/, UPt_3 /16/) there is not found a correlation between the $\gamma(0)$ magnitude and the quasielastic response width, the latter being by an order greater than expected and coincides with the width of the second quasielastic peak found in this work for $CeAl_3$.

The presence of the two quasielastic components in magnetic response function in $CeAl_3$, permits to assume the existence of the two types of magnetic fluctuations (slow and fast) with characteristic relaxation times differeing by an order of magnitude.

The possible explanation for the presence of two quasielastic peaks in INS spectra of $CeAl_3$ could be the neutron scattering on 4f localized moments of the Ce^{+3} ions and on"compensating conduction electron spin clouds". Of course such a supposition needs further experimental study (INS experiments with other heavy fermion systems in a larger energy transfer range) and detailed theoretical consideration.

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Горемычкин Е.А. и др. E14-87-278
Неупругое магнитное рассеяние тепловых нейтронов на системе с тяжелыми фермионами $CeAl_3$

В экспериментах по неупругому рассеянию тепловых нейтронов на системе с тяжелыми фермионами $CeAl_3$ наблюдалась необычная форма магнитной функции отклика. Наряду с особенностью при передаче энергии ≈ 8 мэВ, обусловленной переходом между уровнями расщепленного кристаллическим электрическим полем основного мультиплета иона Ce^{+3}, обнаружено интесновное магнитное рассеяние до передач энергий ≈60 мэВ.

Работа выполнена в Лаборатории нейтронной физики ОИЯИ.

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Goremychkin E.A. et al. E14-87-278
Inelastic Scattering of Thermal Neutrons on Heavy Fermion System $CeAl_3$

Inelastic scattering of thermal neutrons on heavy fermion system $CeAl_3$ in a large energy transfer range (up to ≈ 100 meV) has revealed an unusual aspect of the magnetic response function.Besides the peculiarity at the energy transfer of ≈8 meV due to crystal level transition within the ground multiplet of the Ce^{+3} ion in the crystalline electric field, strong magnetic scattering up to 60 meV energy was observed.

The investigation has been performed at the Laboratory of Neutron Physics, JINR.

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