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Neutron scattering study of the quasi-elastic spectral width in CeMg, CeMg₃ and NdMg₃ intermetallic compounds

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Résumé. — Des mesures de diffusion de neutrons ont été faites en vue d'étudier la partie quasi élastique du spectre magnétique dans les composés cubiques CeMg, CeMg₃ et NdMg₃ ainsi que dans des composés dilués d'yttrium ou de lanthane. La largeur de raie quasi élastique a été mesurée jusqu'à 800 K dans CeMg₃. A haute température elle suit une loi de Korringa qui intercepte l'origine à $T = 0$ K, mais dévie au-dessous de 100 K et tend vers une valeur résiduelle γ_0 pour $T \rightarrow 0$ K. La valeur de γ_0 ($\approx 0,5$ meV) est du même ordre de grandeur que dans CeMg ($\approx 0,6$ meV) et NdMg₃ ($\approx 0,8$ meV). Dans CeMg₃ et NdMg₃, la largeur résiduelle décroît lorsqu'on dilue avec le lanthane ou l'yttrium alors que dans CeMg elle est très peu affectée par la dilution avec le lanthane et même augmente significativement dans le composé avec l'yttrium. Cette différence de comportement suggère une différence dans l'origine des mécanismes de relaxation dans les deux systèmes CeMg et CeMg₃ à basse température.

Abstract. — We report neutron scattering measurements of quasi-elastic magnetic spectra in cubic compounds CeMg, CeMg₃ and NdMg₃ as well as in compounds diluted with Y and La. The quasi-elastic linewidth measured up to 800 K in CeMg₃ shows a linear Korringa law dependence at high temperature with a negligible intercept but deviates below 100 K and tends to a residual value γ_0 as $T \rightarrow 0$ K. The magnitude of γ_0 (≈ 0.5 meV) is similar to that in CeMg (≈ 0.6 meV) and NdMg₃ (≈ 0.8 meV). In CeMg₃ and NdMg₃ the residual linewidth decreases with dilution by La or Y, whereas in CeMg the linewidth is only slightly affected on dilution with La and increases significantly on addition of Y. This difference in behaviour suggests different origins of the low temperature relaxation mechanisms in the two systems CeMg and CeMg₃.

1. Introduction.

Pure cerium and some cerium intermetallic compounds are known to exhibit anomalous magnetic properties which have been classified as Kondo or intermediate valence behaviour [1, 2]. In recent years neutron scattering measurements have been reported on several of these systems, particularly measurements of the inelastic paramagnetic scattering which enable a determination of relaxation rates. In well-known intermediate valence systems CePd₃ [3] and CeSn₃ [4, 5] a broad quasi-elastic spectrum of half width ≈ 20 -30 meV was observed. More recent neutron inelastic scattering measurements on CeSn₃ reveal, at low temperature, in addition to a weakly temperature dependent quasi-elastic scattering, a broad inelastic hump centred around 40 meV which melts away into the quasi-elastic spectrum by around 130 K [6]. On the other hand the compounds like CeAl₂ and CeAl₃ [1], where the anomalies are very

much smaller and the magnetic properties close to those of the trivalent ion, are considered as Kondo systems. In these systems neutron scattering measurements reveal inelastic peaks originating from crystal field levels as well as a central quasi-elastic mode whose linewidth increases roughly as $T^{1/2}$ from a residual value $\gamma_0 = 0.5$ meV at low temperatures [7, 8]. The residual linewidth γ_0 is generally attributed to the Kondo coupling between the local moment and conduction electrons [1].

In this paper we report measurements of the spectral width of the quasi-elastic scattering performed on several cubic cerium compounds CeMg₃ [9], Ce_{0.5}La_{0.5}Mg₃, Ce_{0.5}Y_{0.5}Mg₃, CeMg [10], Ce_{0.2}La_{0.8}Mg, Ce_{0.2}Y_{0.8}Mg [11] as well as on neodymium compounds NdMg₃ [12] and Nd_{0.5}La_{0.5}Mg₃. We compare the behaviour of the quasi-elastic linewidth of compounds with different cerium or neodymium concentrations in order to understand the origin of spin relaxation in these systems.

2. Experimental.

The measurements of the quasi-elastic paramagnetic scattering were performed on time-of-flight spectrometers IN4 and IN5 at the Institut Laue-Langevin. Two incident neutron energies were used in the experiments. 12.6 meV neutrons were selected on the IN4 thermal beam spectrometer with overall elastic energy resolution 0.6 meV (f.ω.h.m). On the IN5 cold source spectrometer, neutrons of incident energy 3.5 meV were used with an overall elastic energy resolution of 0.17 meV. The low temperatures (below 300 K) were obtained by the use of ^4He cryostats and the higher temperatures (300 to 800 K) by the use of a furnace. The time-of-flight spectra were normalized for detector efficiency with the help of the measured scattering from a standard vanadium sample and corrected for background scattering and self-shielding and the spectra are converted into $S(\theta, \omega)$, for constant scattering angle θ .

3. Results.

The neutron scattering cross-section for an isotropic polycrystalline paramagnetic sample with crystal field split magnetic states can be expressed as :

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{N}{(g\mu_B)^2} \left(\frac{\gamma e^2}{m_e c^2} \right)^2 \frac{k'}{k} \left[\frac{1}{2} g f(Q) \right]^2 \times \\ \times \frac{\omega}{1 - e^{-\omega\beta}} \left[a_0 F_Q^0(\omega) + \sum_{\alpha=1}^n a_\alpha F_Q^\alpha(\omega \pm \omega_\alpha) \right]$$

where a_0, a_α ($\alpha = 1, n$) represent the normalized temperature dependent probability factors and $F_Q^0(\omega)$, $F_Q^\alpha(\omega \pm \omega_\alpha)$ ($\alpha = 1, n$) the spectral weight functions of respectively the quasi-elastic and inelastic peaks.

In figure 1, we show the spectra for $\text{Ce}_{0.2}\text{Y}_{0.8}\text{Mg}$ for one scattering angle measured with the low incident energy neutrons (3.5 meV). The solid lines in the figure are the results of fits to the quasi-elastic spectrum in the energy range $-2 \leq \omega \leq 2$ meV. In this energy range no phonon branches are cut, because of the low incident energy, hence the phonon contribution is negligible as also verified by measurements on YMg and LaMg_3 . Also the crystal field inelastic excitations occur at relatively high energy ($\Delta_{\text{CEF}} = 16$ meV in CeMg [10] and 10.6 meV in NdMg_3 [12]) and do not contribute in this energy range. For all spectra in this work we have used a Lorentzian form for $F_Q^0(\omega)$. In figure 2, we plot the temperature dependence of the quasi-elastic spectral component for CeMg , $\text{Ce}_{0.2}\text{La}_{0.8}\text{Mg}$ and $\text{Ce}_{0.2}\text{Y}_{0.8}\text{Mg}$. For all three compounds in the studied temperature range, the quasi-elastic linewidth varies linearly with the temperature and tends to a residual value γ_0 as $T \rightarrow 0$ K : $\gamma_0 \simeq 0.6$ meV for CeMg , 0.5 meV for $\text{Ce}_{0.2}\text{La}_{0.8}\text{Mg}$ and 1.1 meV for $\text{Ce}_{0.2}\text{Y}_{0.8}\text{Mg}$. For CeMg_3 (Fig. 3) the quasi-elastic linewidth γ was measured over a large temperature range (from 8 to 800 K). Above 100 K, γ increases linearly with temperature. Below 100 K, γ deviates from the linear dependence and tends to a finite value of $\gamma_0 \simeq 0.5$ meV as $T \rightarrow 0$ K. In figure 4 we present the results for CeMg_3 , $\text{Ce}_{0.5}\text{La}_{0.5}\text{Mg}_3$ and $\text{Ce}_{0.5}\text{Y}_{0.5}\text{Mg}_3$ at low temperature. The quasi-elastic linewidth of $\text{Ce}_{0.5}\text{La}_{0.5}\text{Mg}_3$ and $\text{Ce}_{0.5}\text{Y}_{0.5}\text{Mg}_3$ tends to a residual value, as $T \rightarrow 0$ K, but has a magnitude smaller than that of CeMg_3 : $\gamma_0 \simeq 0.3$ meV and 0.2 meV respectively for $\text{Ce}_{0.5}\text{La}_{0.5}\text{Mg}_3$ and $\text{Ce}_{0.5}\text{Y}_{0.5}\text{Mg}_3$. Finally, in figure 5 we show the quasi-elastic linewidth for NdMg_3 and $\text{Nd}_{0.5}\text{La}_{0.5}\text{Mg}_3$ up to 250 K and 80 K respectively. The quasi-elastic linewidth of NdMg_3 increases linearly with tempera-

Fig. 1. — $S(\theta, \omega)$ for one scattering angle for $\text{Ce}_{0.2}\text{Y}_{0.8}\text{Mg}$ at different temperatures over the energy range $-2 \leq \omega \leq 2$ meV. The continuous curves represent fits to the data using a Lorentzian as quasi-elastic spectral function.

Fig. 2. — Variation of the quasi-elastic linewidth γ (half-width at half-height) versus temperature for CeMg, Ce_{0.2}Y_{0.8}Mg and Ce_{0.2}La_{0.8}Mg. The dashed lines are guides for the eye.

Fig. 3. — Variation of the quasi-elastic linewidth γ for CeMg₃ over the temperature range 8-800 K. The dashed line represents the fit to the data using the theory of Becker *et al.* [13].

ture from a value $\gamma_0 \simeq 0.8$ meV as $T \rightarrow 0$ K. In Nd_{0.5}La_{0.5}Mg₃ the quasi-elastic linewidth is not as broad as in NdMg₃ and has the value $\gamma_0 \simeq 0.5$ meV.

4. Discussion.

The variation of the quasi-elastic linewidth in CeMg₃ was in a first approach fitted by the theory of Becker *et al.* [13] for the relaxation of crystal field excitations in metallic rare-earth systems. This calculation was performed for the classical s-f exchange Hamiltonian.

Fig. 4. — Low temperature dependence of the quasi-elastic linewidth γ in CeMg₃, Ce_{0.5}Y_{0.5}Mg₃ and Ce_{0.5}La_{0.5}Mg₃. The dashed lines are drawn to guide the eye.

Fig. 5. — Temperature variation of the quasi-elastic linewidth for the neodymium compounds NdMg₃ and Nd_{0.5}La_{0.5}Mg₃. Dashed lines are guides for the eye.

The best fit was obtained with $|Jn(E_F)| \simeq 0.3$, where J is the exchange constant. However, this value was not in agreement with the value deduced from resistivity [15] ($Jn(E_F) \simeq -0.04$) and was too high for a compound where the Kondo coupling is rather weak, as we shall see in the following.

Lopes and Coqblin [14] have, in a recent work, calculated the relaxation of crystal field excitations using the Coqblin-Schrieffer Hamiltonian. The $J=5/2$ multiplet of the cerium ion is split by the cubic symmetry into a doublet Γ_7 and a quadruplet Γ_8 . For $T \gg \Delta$ the neutron quasi-elastic spectrum is the sum of two quasi-elastic lines corresponding to the transitions within respectively Γ_7 and Γ_8 . The quasi-elastic linewidth is expressed as :

$$\gamma_{qe}(\Gamma_7) = 4 \Pi |n(E_F)|^2 [|J_{77}|^2 + 2 |J_{78}|^2] T$$

$$\gamma_{qe}(\Gamma_8) = 4 \Pi |n(E_F)|^2 [2 |J_{88}|^2 + |J_{78}|^2] T.$$

These expressions have the same form as the corresponding expressions of Becker *et al.*, except that they involve the $|J_{\text{MM}}|^2$ values instead of $|(g-1)J|^2$.

In our experiments above 300 K ($T \gg \Delta$, $\Delta = 197$ K) the transitions within the Γ_8 are the main contributions in the quasi-elastic linewidth. From the high temperature slope we deduce $|n(E_F)(2|J_{88}|^2 + |J_{78}|^2)|^{1/2} \simeq 0.08$, value which is in much better agreement with the resistivity data.

In a first analysis the residual value γ_0 of the quasi-elastic linewidth in CeMg_3 and in CeMg can be attributed to the Kondo coupling between the local moment and conduction electrons. However, the results on the dilute compounds and on NdMg_3 (Fig. 5) show that the Kondo coupling is not the only possible mechanism for the residual linewidth. In fact, since NdMg_3 is not a Kondo system, the residual linewidth in this case can only be due to the exchange coupling of the R.K.K.Y. type. In such systems the dilution by non-magnetic ions (lanthanum or yttrium) should reduce the interactions and hence the residual width. This is verified by measurements on $\text{Nd}_{0.5}\text{La}_{0.5}\text{Mg}_3$ (Fig. 5). Similarly, the reduction of the quasi-elastic linewidth observed in dilute compounds $\text{Ce}_{0.5}\text{La}_{0.5}\text{Mg}_3$ and $\text{Ce}_{0.5}\text{Y}_{0.5}\text{Mg}_3$ (Fig. 4) implies that the relaxation in CeMg_3 is mainly due to the exchange coupling. In this connection, we recall that the specific heat of both CeMg_3 and NdMg_3 has a long tail above the λ -type anomaly at the Néel temperature, at 4 and 6 K respectively, and the magnetic entropy up to T_N is lower than $R \ln 2$, which is the value for a ground state doublet Γ_7 in CeMg_3 [15] and Γ_6 in NdMg_3 [15]. This may be understood as suggesting the existence of magnetic correlations above T_N in these compounds.

The measurements on CeMg have not been made up to sufficiently high temperatures to enable any conclusions about the relaxation behaviour at high temperature to be drawn, for example the Korringa law as in CeMg_3 , or $T^{1/2}$ variation as in CeAl_2 [7] or CeAl_3 [8]. However, at low temperatures the behaviour of the quasi-elastic linewidth in CeMg , $\text{Ce}_{0.2}\text{La}_{0.8}\text{Mg}$ and $\text{Ce}_{0.2}\text{Y}_{0.8}\text{Mg}$ is different from that of CeMg_3 , $\text{Ce}_{0.5}\text{La}_{0.5}\text{Mg}_3$ and $\text{Ce}_{0.5}\text{Y}_{0.5}\text{Mg}_3$, in that the linewidth does not decrease with dilution. Previous magnetization measurements on CeMg under pressure have revealed a linear decrease of the Néel temperature with increasing pressure [16], and the relative variation of $d(\ln T_N)/dp$ is comparable to that found in CeAl_2 [17]. These results are in agreement with the theory of Doniach [18] and Lavagna *et al.* [19] for dense Kondo systems. Thus, CeMg appears similar to the Kondo system CeAl_2 . Returning to the effect of dilution, we note that dilution of CeMg by lanthanum does not significantly change the residual quasi-elastic linewidth ($\gamma_0 \simeq 0.5$ meV in $\text{Ce}_{0.2}\text{La}_{0.8}\text{Mg}$ and $\simeq 0.6$ meV in CeMg). However, dilution with yttrium enhances the residual width ($\gamma_0 \simeq 1.1$ meV in $\text{Ce}_{0.2}\text{Y}_{0.8}\text{Mg}$). This is because the Kondo coupling is enhanced by the chemical pressure due to the

addition of yttrium (YMg has a smaller lattice constant than CeMg). This is also seen from electrical resistivity measurements in $\text{Ce}_{0.2}\text{Y}_{0.8}\text{Mg}$ where a minimum is observed [11]. Similar results have been obtained on $\text{Ce}(\text{La}, \text{Y})\text{Al}_2$ systems by Steglich and Loewenhaupt [20]. All these results are also in agreement with the increase of the quasi-elastic linewidth of CeAg under external pressure [21].

It may, at first sight, appear surprising that the residual linewidth in CeMg_3 ($T_N = 4$ K) is dominated by ion-ion exchange interactions, whereas in CeMg the origin of the residual width is mainly through Kondo coupling, although in the latter compound the ordering temperature is much higher ($T_N = 19$ K). As noted above, the specific heats of CeMg_3 and NdMg_3 show a large tail above T_N suggesting persistence of magnetic correlations to relatively high temperatures. The magnetic order in CeMg on the contrary occurs as a first-order phase transition driven principally by magneto-elastic interactions and shows little or no precursory effects in susceptibility or specific heat measurements [10]. Hence, the exchange interactions do not have the same influence in CeMg as in CeMg_3 . In other words, the relative magnitudes of the ordering temperatures in CeMg and CeMg_3 is not a good indicator of the relative effect of the exchange coupling on the low temperature residual width in the two systems. Finally, the higher concentration of Ce in CeMg relative to CeMg_3 may also naively lead us to expect stronger interaction effects in the former than in the latter compound. However, such an argument clearly breaks down when carried over to the case of pure Ce.

5. Conclusions.

We have measured the quasi-elastic linewidth in cubic CeMg , CeMg_3 and NdMg_3 compounds, as well as in their diluted compounds. In CeMg_3 , measurements up to high temperature have shown a linear Korringa law behaviour with zero intercept, except below 100 K where we observe deviations giving a residual width as $T \rightarrow 0$ K. Dilution with both lanthanum and yttrium in this compound, as well as in NdMg_3 , reduces the residual linewidth. In the case of CeMg , however, dilution does not modify the residual width or increase it with yttrium. We believe the difference in behaviour between the two systems is due to the different origins of the low temperature relaxation mechanisms. It appears that the residual width in CeMg_3 is mainly due to the exchange interactions since dilution reduces the width, whereas in CeMg the origin of the residual width is the Kondo coupling between local moment and conduction electrons. This is consistent with the fact that magnetic properties of CeMg are similar to those of the Kondo compound CeAl_2 as for example in the pressure dependence of T_N .

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