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An inelastic neutron scattering study of the Kondo semiconductor CeNiSn in high magnetic field

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Abstract. We have used inelastic neutron scattering to determine the spin dynamics in the Kondo insulator CeNiSn under magnetic fields up to 10 T applied parallel to the easy a axis. The well known excitations peaked at $\hbar\omega = 2$ meV located at the Brillouin zone centre and at $\hbar\omega = 4$ meV located at the Brillouin zone border were studied comparatively. At the scattering vectors (0 1 0) and (0 0 1) the peak shifts continuously from 2 to 2.5 meV with the field. No Zeeman splitting could be detected at the accuracy of our measurements. For the '4 meV' excitation studied at the scattering vector (0 0.5 1), a field as large as 10 T begins to fill in the gap whereas there is no appreciable change in the spin dynamics between 0 and 6 T. The main feature is the broadening of the response. We compare our experimental results to recent theoretical developments.

1. Introduction

Among strongly correlated electronic systems, Ce intermetallics exhibit various behaviour classified in the literature as Kondo lattice, valence fluctuating or heavy-fermion compounds. CeNiSn is characterized by the decrease of the electronic density of states at low temperatures as established by thermodynamic and transport measurements [1]. This compound is then frequently classified as a Kondo semiconductor. The gap occurring below 10 K is in fact a pseudogap, as shown by specific heat and NMR measurements, which implies a residual density of states in the ground state [2]. The resistivity evolves surprisingly from a semiconducting behaviour for the less pure samples to a metallic-like behaviour for the best ones [3]. It is now believed that the low carrier density is localized by impurities in the lower-quality samples. By applying pressure or magnetic field or by doping, a heavy-fermion behaviour antecedent to the gap formation is recovered. The interest in this compound is thus the localization near the borderline of the magnetic–non-magnetic (M–NM) and metal–insulator (M–I) transition.

Inelastic neutron scattering (INS) experiments performed on a triple-axis spectrometer (TAS) revealed the existence of two excitations in the magnetic excitation spectrum of CeNiSn appearing below 15 K. The first one is located at the scattering vectors $\boldsymbol{Q} = (0\,0\,1)$

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[4] and $\boldsymbol{Q} = (0\,1\,0)$ [5] and is inelastic in character, with a typical energy of 2 meV. This excitation is associated with the wave vector $\boldsymbol{k}_1 = (0\,0\,0)$ at the Brillouin zone centre. It is antiferromagnetic in nature: peaks are located, in the reciprocal space, at forbidden nuclear positions. It corresponds to an antiferromagnetic arrangement of moments within the unit cell. The second excitation is observed at the scattering vectors $\boldsymbol{Q} = (Q_a n + 1/2 Q_c)$ for every Q_a and Q_c and with n integer [5] and has a typical energy of 4 meV. It is associated with a second wave vector $\boldsymbol{k}_2 = (0\,1/2\,0)$ at the border of the zone along the b axis and corresponds to fluctuations involving a doubling of the unit cell along the b axis. At present, these excitations are far from being understood. It is possible, as in many other strongly correlated Ce systems [6], to describe them as antiferromagnetic correlations associated with the wave vectors $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$. For example, such a description is well established for the heavy-fermion compound $CeRu_2Si_2$ in which the existence of incommensurate correlations peaked at $(0.31\,0\,0)$ and $(0.31\,0.31\,0)$ show the proximity to the M–NM borderline [7]. In this picture the RKKY interactions have an important contribution to the spin dynamics. On the other hand, it is possible to adopt a band picture and describe the spectrum in terms of a direct and an indirect gap. Within this approach, the excitation at 4 meV is qualitatively described as a hybridization gap [8]. This electronic gap is observed in INS because it corresponds to a transition involving quasiparticles with a high 4f spectral weight between a high density of initial and final states. Such a description was previously used for the mixed-valence semiconductor TmSe to explain the $\boldsymbol{Q}$ dependence of the signal [9]. Theoretical work is in progress to include the symmetry of CeNiSn in this picture [10, 11]. Within this approach, RKKY interactions are not taken into account; the spin dynamics is calculated from interband transitions: the occurrence of a gap is related to the complete filling of the lower band (i.e. the realization of an insulating state).

2. Experimental details

The space group reported for CeNiSn, in the orthorhombic symmetry, is either *Pnma* or $Pn2_1a$ [12]. The room-temperature lattice parameters are $a = 7.523$ Å, $b = 4.592$ Å, $c = 7.561$ Å. There are four Ce atoms per unit cell and pattern extinctions due to the glide planes n and a. We performed INS experiments on the triple-axis spectrometer IN14 installed on a cold guide at the ILL high-flux reactor. Experiments were performed at constant final wave vector $k_F = 1.55$ Å^{-1} using a horizontally focusing pyrolitic graphite (0 0 2) analyser. A beryllium filter was used in the scattered beam to reduce higher-order contamination. The full width at half maximum (FWHM) of the incoherent signal was 0.28 meV whereas it was 0.22 meV with natural collimation and a flat analyser. Single crystals of CeNiSn were grown by the Czochralski method using a tungsten crucible in a radio-frequency furnace. Their quality was the same as that of crystal No 4 in [13], which shows a metallic resistivity. Two crystals were assembled in a sample holder. They were 0.96 and 0.65 cm^3 in volume and had mosaic spreads of 0.2 and 0.5° respectively. The sample was mounted in the 12 T vertical cryomagnet of the CEA–Grenoble. The horizontal scattering plane was the (b, c) plane and the field was applied along the vertical a axis. Because of the anisotropy of the system, the dominant contribution arises from $\mathrm{Im}\chi^{aa}$, which corresponds to longitudinal fluctuations along the easy a axis. Due to the strong effects of the stray field on the metallic part of the guide shielding of IN14, the maximum energy transfer had to be reduced at the highest fields.

3. Results

The effect of a magnetic field on the magnetic excitation spectrum provides important information on both the ground state and the excited states of the system. A partial destruction of the gap for a field of about 13 T applied along the easy a axis is inferred from magnetoresistivity, magnetization and specific heat measurements [1]. In particular the linear term in the specific heat increases with the field, restoring the heavy-fermion ground state. In order to study the effect of the magnetic field on the gap in the magnetic excitation spectrum, we have determined the field dependence of both the '2 meV' and the '4 meV' modes.

3.1. The '2 meV' excitation

The field dependence at the scattering vector (0 1.025 0) is shown in figure 1 for the applied fields $H = 0$, 6 and 10 T. In all cases a single excitation is clearly observed. With increasing field, the spectrum shifts to higher energy and its intensity decreases continuously. The magnetic nature of the signal was checked by heating above 25 K. The neutron intensity was analysed from the relation $I(\boldsymbol{Q},\omega) = I_{bg} + (1+n(\omega))\chi''(\boldsymbol{Q},\omega)$, where I_{bg} is the background neutron intensity (taken as constant in the fitting procedure and determined at negative energy transfer), $n(\omega)$ is the Bose factor and χ'' the dynamical susceptibility. For this last quantity we used the functional form given in [4]:

$$\chi'' = A|\varepsilon|\omega(\mathrm{Re}(1/\sqrt{\omega^2-\varepsilon^2}))^2 \tag{1}$$

with $\varepsilon = \Delta + \mathrm{i}\Gamma$, where Δ describes the inelasticity of the lineshape and Γ its broadening, and A has the dimension of a susceptibility but its interpretation is not straightforward since the phase of the complex number ε must be taken into account in order to calculate the bulk susceptibility. This form reproduces the asymmetric shape of the spectrum, which is intermediate between a Lorentzian and a step function [4, 5]. For $\Delta = 0$ this function is equivalent to a Lorentzian of FWHM 2Γ. The fit to the expression (1) accounts well for the data at (0 1.025 0). The field dependences of the inelasticity Δ and the damping Γ are shown in the inset of figure 1 with values listed in table 1. These quantities evolve from $\Delta \approx 2$ meV ($\Gamma \approx 0.5$ meV) in zero field to $\Delta \approx 2.5$ meV ($\Gamma \approx 0.6$ meV) for a field of 10 T. The field dependence of the inelasticity Δ can be fitted using the expression

$$\Delta(H) = \sqrt{\Delta^2(0) + \delta_\Delta^2 H^2} \tag{2}$$

with $\Delta(0) \approx 2$ meV and $\delta_\Delta \approx 0.17$ meV T^{-1}, the latter value being consistent with $2\mu_B \approx 0.12$ meV T^{-1}. The use of the same expression for the damping parameter Γ gives a smaller value, $\delta_\Gamma \approx 0.045$ meV T^{-1}, indicating a weaker field effect or even no significant field dependence. This is in agreement with a visual inspection of the data which reveals a slight broadening of the signal due to the collapse of its peaked part, the high-energy tail being independent of the magnetic field. The striking point in this field dependence is that $\Delta(H)$ is characteristic of a transition between two non-degenerate levels: no linear Zeeman splitting is observed in our INS data since there is no signal appearing below 2 meV when the field is increased.

The field dependence in the vicinity of the (0 0 1) point is shown in figure 2 for the scattering vector (0 0 1.025) and fields of 0, 6 and 10 T. At the accuracy of our measurements, the magnetic responses at scattering vectors (0 1 0) and (0 0 1) appear very similar. They differ only by a 'normalization' factor of the order of 0.8 (background subtracted), qualitatively understood from the orthorhombic anisotropy observed from

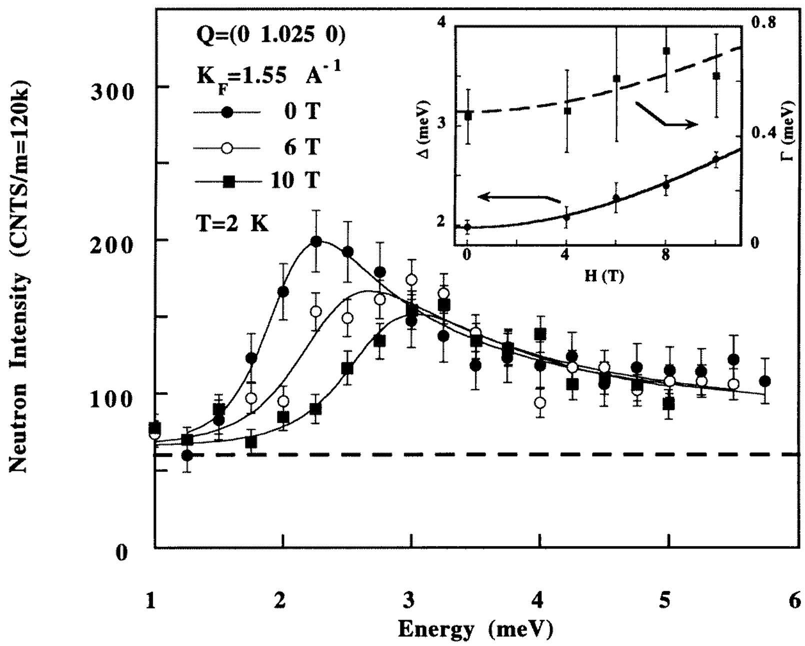

Figure 1. Energy scans at $\boldsymbol{Q} = (0\,1.025\,0)$ at $T = 2$ K for $H = 0$, 6 and 10 T. The full lines are the best fit to the function (1) defined in the text. The dashed line indicates the background. The inset shows the variation of the inelasticity Δ and the width Γ with the field with the best fit to the function (2).

Table 1. Parameters extracted from a fit of (1) to the excitation at $\boldsymbol{Q} = (0\,1.025\,0)$. A is normalized to an arbitrary monitor common for tables 1–3.

	$H = 0$ T	$H = 4$ T	$H = 6$ T	$H = 8$ T	$H = 10$ T
A	20 (2)	15 (2)	20 (3)	14 (3)	10 (2)
Δ (meV)	1.98 (07)	2.09 (11)	2.28 (15)	2.41 (10)	2.65 (08)
Γ (meV)	0.47 (10)	0.49 (15)	0.61 (23)	0.71 (15)	0.62 (15)

magnetic susceptibility measurements [1] ($\chi^a/\chi^b = 2$ and $\chi^a/\chi^c = 3$; $I(0\,0\,1) \approx \chi^a + \chi^b$ and $I(0\,1\,0) \approx \chi^a + \chi^c$). We used the functional form (1) to describe the data. The parameters extracted from the fitting procedure are given in table 2 and are, within the error bars, the same as the ones presented in table 1. This is in agreement with the three-dimensional character of this excitation put forward in [5]. This similarity permits us to improve the statistic concerning the '2 meV' excitation by a factor of $\sqrt{2}$ and reinforces our conclusion upon the absence of Zeeman splitting.

3.2. The '4 meV' excitation

Figure 3 shows the spin dynamics at the scattering vector $\boldsymbol{Q} = (0\,0.5\,1)$ for fields $H = 0$, 6 and 10 T. In zero field, the gapped lineshape characterized by a strong asymmetry is well established for this excitation. The response at 6 T turns out to be nearly identical to that in zero field. At higher fields one observes a broadening of the magnetic response, which

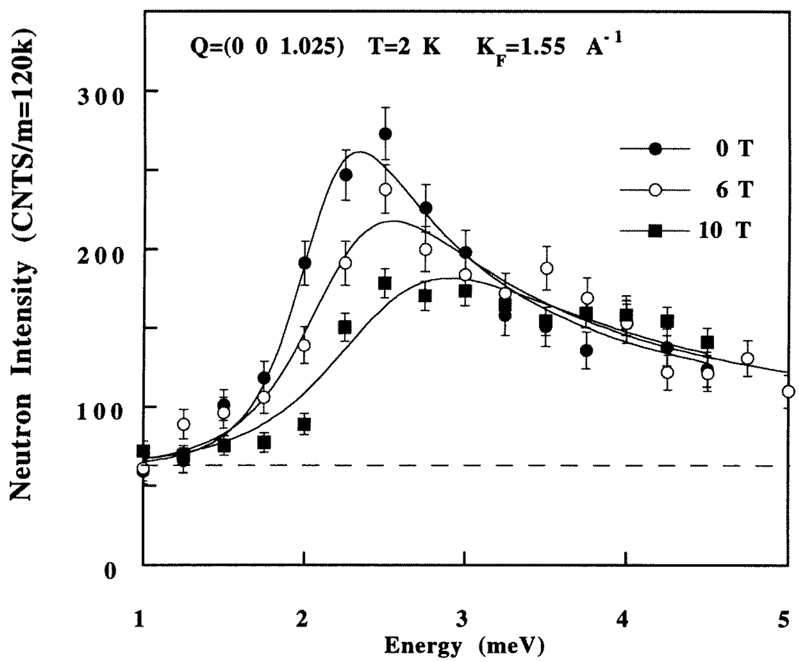

Figure 2. Energy scans performed at $Q = (0\,0\,1.025)$ at $T = 2$ K for $H = 0$, 6 and 10 T. The lines are the best fit to the function (1) defined in the text.

Table 2. Parameters extracted from a fit of (1) to the excitation at $Q = (0\,0\,0.125)$.

	$H = 0$ T	$H = 6$ T	$H = 10$ T
A	30 (4)	25 (3)	16 (3)
Δ (meV)	2.04 (10)	2.13 (11)	2.42 (10)
Γ (meV)	0.47 (10)	0.67 (16)	0.77 (15)

corresponds more probably to a transfer of intensity from the peaked part to the low-energy part of the signal. Using the same procedure as for the '2 meV' excitation, we have firstly analysed our data using the functional form (1). The best fits are obtained with parameters Δ and Γ listed in table 3 and shown in the inset of figure 3. Several facts emerge from these numbers. Firstly, only weak effects are seen on the peak position Δ, which are fully understood from the previous relation (2) with $\Delta(0) = 3.8$ meV and assuming as previously $\delta_\Delta = 0.17$ meV T^{-1}: clearly, much higher fields are required to observe a more sizeable effect on Δ. The main effect of the field is seen on the damping parameter Γ, which increases relatively rapidly from 0.6 to 1.4 meV in the field range studied.

Since the broadening of the signal is accompanied by the occurrence of a magnetic signal at energy lower than 4 meV, a Zeeman splitting cannot be ruled out unambiguously. We tentatively give an alternative analysis of the data assuming this time a response consisting of two contributions (1) centred around the shifted energies $\Delta(0) \pm \beta H^2$. The quadratic behaviour is introduced since the magnetic field has experimentally a non-linear effect. The spectrum at 6 T being almost the same as that at 0 T, a linear Zeeman splitting $\Delta(0) \pm \gamma H$

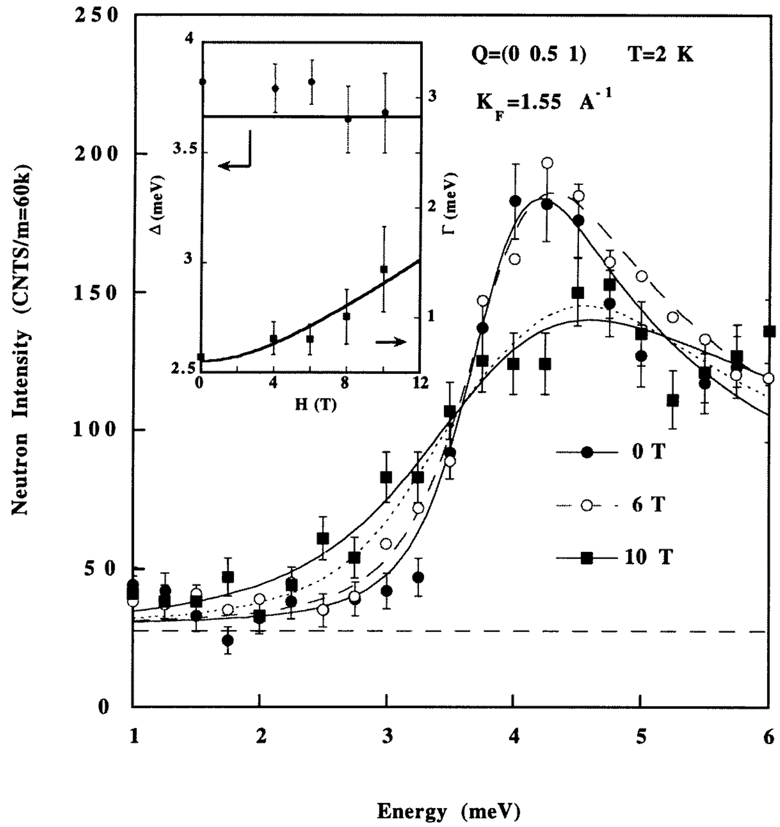

Figure 3. Energy scans performed at $\boldsymbol{Q} = (0\,0.5\,1)$ at $T = 2$ K for $H = 0$, 6 and 10 T. The lines are the best fit to the function (1) described in the text. For $H = 10$ T, the fit corresponding to a splitting described in the text is also shown (dotted line). The inset shows the variation of the inelasticity Δ and the width Γ with the field.

Table 3. Parameters extracted from a fit of (1) to the excitation at $\boldsymbol{Q} = (0\,0.5\,1)$.

	$H = 0$ T	$H = 4$ T	$H = 6$ T	$H = 8$ T	$H = 10$ T
A	28 (3)	29 (3)	26 (3)	22 (5)	27 (6)
Δ (meV)	3.82 (10)	3.79 (11)	3.82 (10)	3.65 (15)	3.68 (18)
Γ (meV)	0.64 (11)	0.81 (15)	0.80 (14)	1.01 (25)	1.44 (39)

is excluded. In this second analysis, the damping parameter Γ was kept fixed to the intermediate value $\Gamma \approx 1$ meV and β was adjusted. This procedure was chosen in order to have few parameters to fit. The best fit is achieved with a parameter $\beta \approx 0.004$ meV T^{-2}, which is consistent with the expression $\beta \approx \delta_\Delta^2/2\Delta$ expected from the expansion in H of relation (2) using $\delta_\Delta = 0.17$ meV T^{-1}. Not very surprisingly, within the experimental accuracy these two 'extreme' analyses produce indistinguishable fits for the '4 meV' peak. This is clearly shown in figure 3 for the data at 10 T. It is thus difficult to go beyond a qualitative description of the data stressing the appearance of a magnetic signal at low energy with the increase of the magnetic field. Qualitatively our data seem also consistent with the existence of a threshold field $H_T \sim 6$ T, below which the response is only weakly field dependent. We must notice that an analysis of the '2 meV' excitation in this spirit would imply use of two shifted energies $\Delta(0)+\beta_1 H^2$ and $\Delta(0)+\beta_2 H^2$ with $\beta_1 \approx 0.006$ meV T^{-2} and $\beta_2 \approx 0.01$ meV T^{-2}, but this approach is in fact similar to the one proposed previously since β_1 is close to β_2 and close to the value $\delta_\Delta^2/2\Delta = 0.007$ meV T^{-2} deduced from the analysis of subsection 3.1. As a consequence, for the '2 meV' mode, the non-degenerate picture can be kept. So, whatever the analysis procedure used, we emphasize the very different behaviour of the '2 meV' and '4 meV' modes.

In addition to changes in the energy response, important modifications are also observed in the wave vector response. This is illustrated in figure 4, which shows the evolution of two constant-energy scans at 4.25 meV for fields $H = 0$ and $H = 10$ T. The experimental lineshapes are well reproduced by a Lorentzian function $A(q) = A/(q^2 + \kappa^2)$ with the parameter κ evolving from 0.12 Å^{-1} in zero field to 0.35 Å^{-1} at 10 T. In terms of characteristic lengths, this corresponds to respectively $\xi/b = 1.8$ and $\xi/b = 0.6$ interatomic distances ($\xi = 1/\kappa$). This characteristic length reflects the correlation length (which is strictly speaking obtained by integrating a large number of constant-energy scans) and is nearly equivalent to the correlation length when the spectrum broadens. Assuming this similarity, our data point out the strong reduction of the correlations under magnetic field.

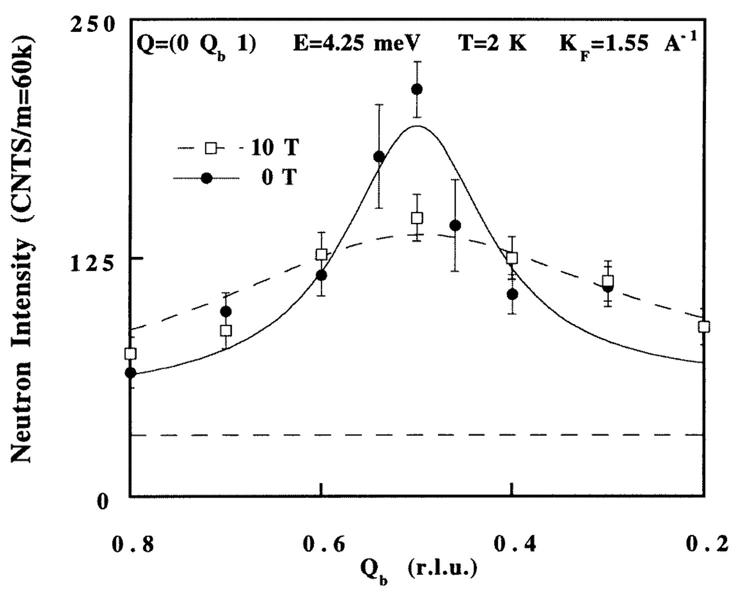

Figure 4. Q scans performed along the b axis at $T = 2$ K for $H = 0$ and 10 T at constant energy transfer equal to 4.25 meV. The lines are the best fit to a Lorentzian function.

4. Discussion

Concerning the '2 meV' excitation, the main result of this study is that the linear Zeeman splitting is not observed inasmuch as no signal appears at lower energy than the initial peak position when the field is increased. A quadratic Zeeman splitting is not relevant either at the accuracy of our measurements. From the present data, we have shown that a field as large as 10 T does not lift any degeneracy. Its effect is only to spread non-degenerate levels, whereas basically in the Kondo problem a singlet–triplet gap is expected [14]. In a naive picture, the gap corresponds to the transition between a collective non-magnetic singlet ground state formed by Kondo quenched local moments and a magnetic triplet excited state. In such a case, three modes are expected in the excitation spectrum: a field independent one and two branches with a Zeeman splitting. This situation is not observed here. It is interesting to compare our results obtained on CeNiSn with those obtained on the heavy-fermion compounds $CeCu_6$ and $CeRu_2Si_2$ [7]. Both compounds are Pauli paramagnets which exhibit a metamagnetic-like transition at 2.5 and 8 T respectively. the analysis of INS data [7] shows that two contributions must be taken into account in order to describe the magnetic excitation spectrum. The first contribution is slowly decreasing with increasing field and is associated with single site fluctuations. It is quasielastic in nature ($\Gamma_{SS} \approx 0.4$ meV for $CeCu_6$ and 2 meV for $CeRu_2Si_2$). The second reflects the antiferromagnetic-like correlations that collapses at the metamagnetic transition. It is typically inelastic, characterized by an inelasticity ω_0 and a damping parameter Γ_{IS} ($\omega_0 \approx \Gamma_{IS} \approx 0.2$ meV and $\omega_0 \approx \Gamma_{IS} \approx 1$ meV respectively). For both compounds, the two quantities ω_0 and Γ_{IS} are independent of the magnetic field. In the simple picture of a singlet–triplet gap such a dependence is expected for longitudinal fluctuations corresponding to fluctuations parallel to the field direction. The Zeeman splitting is expected only for the transverse fluctuations but has never been observed due to the strong Ising character of both compounds. It is important to note that in an earlier study of $CeCu_6$ [14] a field dependence of Γ described by relation (2) was reported, assuming a single quasielastic contribution. At present, there is no experimental evidence in CeNiSn for such an additional localized contribution.

Concerning the '4 meV' mode, the large broadening of the signal under magnetic field prevents us from being affirmative on the absence of Zeeman splitting since the magnetic response at 10 T can be interpreted with a single or a double peak. Nevertheless, this mode is the only one which clearly shows the creation of new states in the gap for a field of 10 T. This mimics the behaviour of the specific heat for which there is also a redistribution from a hump characteristic of the gap towards the low-T linear contribution when the field is increased [15]. This analogy is consistent with the fact that the '4 meV' excitation carries the dominant spectral weight compared to the '2 meV' excitation [5]. Our experimental data on the '4 meV' mode give some support to the hybridization picture. In a recent theoretical work, Ikeda and Miyake [11] considered the possibility of a hybridization potential vanishing at the zone boundary along the a axis. Such a mechanism accounts qualitatively for the pseudogap behaviour observed in specific heat, NMR and resistivity measurements. These authors have shown that it is possible to have such a node in the hybridization potential by assuming that it is the crystal field level $|5/2, \pm 3/2\rangle$ (where the quantization axis z is taken along the easy a axis) which hybridizes with the conduction band. The density of states derived has a four-peak structure with characteristic positions separated by Δ_1 and Δ_2 where $\Delta_2/\Delta_1 \sim 4.2$ (Δ_1 is the hybridization value at the zone centre $k_a = 0$, Δ_2 is the maximum hybridization value at $k_a \sim 0.7$ and this value is zero at the zone boundary $k_a = 1$). In this picture, when the field is applied, the upper and lower bands (obtained by hybridization between f electrons and the conduction band) split into up and down spin states. From this model, with increasing field, the gap in the spin susceptibility χ^{aa} remains unchanged up to a threshold value H_T, above which the bands overlap and the pseudogap vanishes. Experimentally, as shown in figure 3, there is little change of the spin dynamics for the '4 meV' excitation between 0 and 6 T, which is consistent with the idea of a threshold field of the order of 6 T. Our experimental results obtained on the '4 meV' excitation are, at least qualitatively, in agreement with the model of Ikeda and Miyake.

5. Conclusion

To summarize, our INS studies of CeNiSn under a magnetic field have brought new information concerning the ground state and the exited state of this Kondo insulator. From the field dependence of the '2 meV' excitations, we have shown that both the ground state and the first excited state are non-degenerate, as directly seen by the absence of splitting under a field. Concerning the '4 meV' excitation, models both with and without Zeeman splitting can describe the data. Our neutron measurements show quantitatively different behaviours for the '2 meV' and '4 meV' excitations. For the former, the field dependence of the magnetic response is mainly controlled by the field dependence of the mode energy, the damping being only weakly field dependent. For the latter, the magnetic response is mainly controlled by the damping, the mode energy being this time only weakly field dependent. Although the physical meanings of the parameters Δ and Γ need further theoretical justification and are without explanation so far, we believe that their very different field dependence is a strong indication of the different nature of excitations probed at wave vectors (0 1 0) and (0 1/2 0). The effect of the field is to create states in the gap, paradoxically more importantly for the '4 meV' than for the '2 meV' excitation. The results reported here stress the strong difference from the Pauli paramagnet heavy-fermion compounds previously studied in a magnetic field, e.g. $CeCu_6$ and $CeRu_2Si_2$. The specificity of CeNiSn arises certainly from the existence of a nearly insulating ground state. The understanding of the nature of this ground state requires further theoretical work. Models dealing with a hybridization picture are the most promising, owing to the similarities obtained between the calculations of $S(\boldsymbol{Q}, \omega)$ and the experimental INS data [11]. However, refinements must incorporate magnetic correlations which are missing from the original model.

Acknowledgments

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