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The evolution of the crystal-field states upon increasing hybridization in $Ce(Cu_{1-x}Ni_x)_2Ge_2$

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Abstract. Inelastic neutron scattering (INS) experiments have been performed on polycrystalline $Ce(Cu_{1-x}Ni_x)_2Ge_2$ for the concentrations $x = 0.1$, 0.28, 0.5, 0.65 to study the effect of the increasing hybridization between the f electrons and the band states on the crystal-field (CF) states. For both $x = 0.1$ and $x = 0.28$, CF level schemes very similar to that of pure $CeCu_2Ge_2$ have been found. In contrast, no well defined CF excitations could be detected for $x = 0.5$ and $x = 0.65$. The absence of properly developed CF states in high-T^* Kondo and/or valence fluctuation compounds is a well known fact. Remarkably, in $Ce(Cu_{1-x}Ni_x)_2Ge_2$ for $x = 0.5$ and $x = 0.65$, the Kondo temperatures as determined by quasielastic neutron scattering are at least one order of magnitude below the typical CF splitting energies observed for $x = 0.1$ and $x = 0.28$. The evolution of the CF splitting in $Ce(Cu_{1-x}Ni_x)_2Ge_2$ and its distinctive behaviour is discussed in comparison with other heavy-fermion compounds.

1. Introduction

$CeCu_2Ge_2$ is a magnetically ordered heavy-fermion system (HFS) where the RKKY intersite and the Kondo-type on-site interactions are almost equal in strength [1]. Within its tetragonal crystalline environment, the $4f^1$ multiplet of the $^2F_5/2$ Hund's rule ground state is split into three doublets. Inelastic neutron scattering (INS) studies revealed a ground-state characterized by a wave function $0.9|\pm 5/2\rangle - 0.435|\mp 3/2\rangle$ separated from the first and accidentally degenerate excited state corresponding to $|\pm 1/2\rangle$ and $0.9|\mp 3/2\rangle + 0.435|\pm 5/2\rangle$ [2]. The degeneracy of the excited state has to be understood in terms of the two wave functions being closer in energy than the experimental resolution and hence forming a quasiquartet. On alloying nickel with $CeCu_2Ge_2$ the unit cell is compressed and the hybridization between the f electrons and the band states increases. Indeed, $CeNi_2Ge_2$ shows no magnetic order, but a typical Fermi-liquid behaviour. For intermediate concentrations, experimental evidence for the existence of heavy-fermion band magnetism (HFBM) [3, 4] has been provided. In the regime of HFBM the spin degrees of freedom are assumed to be transferred from the 4f sites to the band states, while the 4f charges reside at the cerium sites. Quasielastic neutron scattering, as well as macroscopic measurements revealed a slight increase of the Kondo-lattice temperature T^* with increasing Ni content x up to $x \leqslant 0.7$. For $x \geqslant 0.7$, T^* increases strongly, reaching values of 30–40 K for $CeNi_2Ge_2$ [3]. No well defined CF states could be detected for $CeNi_2Ge_2$ by means of INS [5]. For

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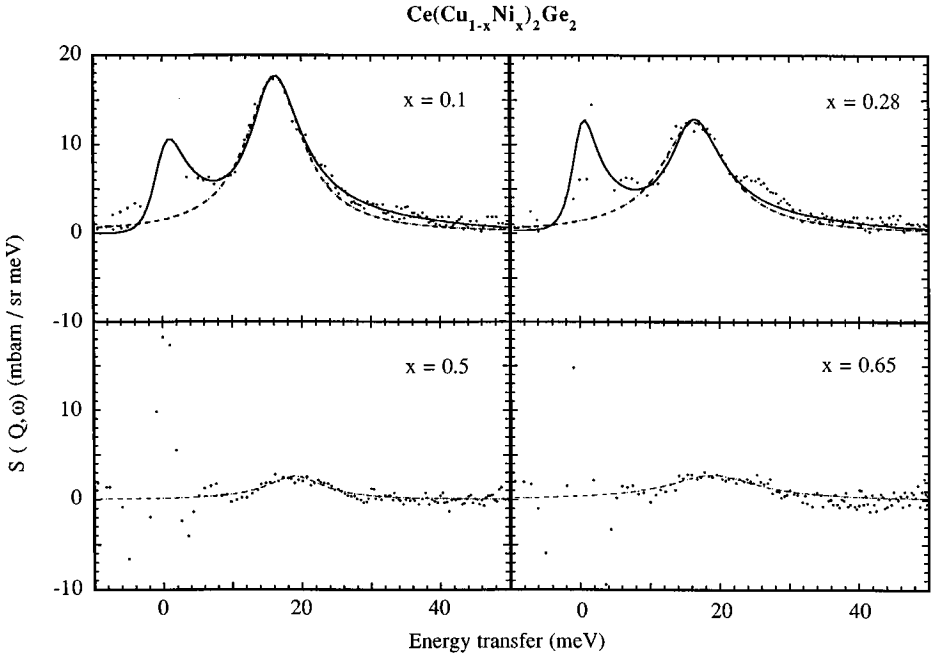

Figure 1. Magnetic excitation spectra of $Ce(Cu_{1-x}Ni_x)_2Ge_2$ at $T = 20$ K with an incident neutron energy of $E_i = 69$ meV. Full and dashed lines for are fits as described in the text. For $x = 0.5$ and $x = 0.65$, the magnetic intensities were too weak for any analysis in terms of CF wave functions.

details of the phase diagram, the reader is referred to [3] and references therein. The CF potential is essential in determining the thermodynamic and magnetic properties of rare-earth compounds. It has been proposed that the details of the CF potential itself are mainly based on the hybridization interaction between f-electron and band states that is responsible for the heavy-fermion behaviour [6].

Direct access to the CF potential can be obtained by means of INS, since the magnetic scattering cross section $S(\boldsymbol{Q}, \omega, T)$ is proportional to the generalized magnetic susceptibility $\chi(\boldsymbol{Q}, \omega, T)$. We have performed INS experiments to study the evolution of the CF splitting in $Ce(Cu_{1-x}Ni_x)_2Ge_2$ upon increasing hybridization. The absence of well defined CF excitations in high-T^* Kondo and/or valence fluctuation compounds is a well known fact. So far, a progressive evolution from a CF-split heavy fermion to a fully degenerate valence fluctuation state has been demonstrated experimentally only for $CeIn_{3-x}Sn_x$ by Murani *et al* [7]. They also found a concomitant broadening and shift of the $^2F_{5/2} \rightarrow {}^2F_{7/2}$ spin–orbit excitation. As will be shown in the following, the present results on $Ce(Cu_{1-x}Ni_x)_2Ge_2$ display a rather different behaviour compared to that of $CeIn_{3-x}Sn_x$.

2. Experimental details

The samples have been synthesized by arc melting the pure elements in an argon atmosphere. X-ray diffraction and microprobe analysis indicated single-phase material over the whole composition range. The INS experiments have been performed on the thermal time-of-flight (TOF) spectrometer IN4 at the Institute Laue–Langevin, Grenoble. Incident neutron

energies of $E_i = 17$ meV and $E_i = 69$ meV have been chosen, respectively. For the determination of the CF level schemes, samples with concentrations $x = 0.1$, 0.28, 0.5 and 0.65 were measured in the paramagnetic state at $T = 20$ K [3]. The results showed that the CF excitation energies exceeded the 17 meV window. Consequently, the low-energy data could only confirm the results of our quasielastic neutron scattering study [3] and, in the following, we will concentrate on results obtained with an incident neutron energy of $E_i = 69$ meV. Additionally, the nonmagnetic reference compounds $LaCu_2Ge_2$ and $LaNi_2Ge_2$, a vanadium standard, an empty sample holder and a cadmium plate have been measured to account for the phonon part of the scattering, detector efficiency, background and absorption in a standard way.

The phonon spectra of $LaCu_2Ge_2$ and $LaNi_2Ge_2$ differed only slightly. The corresponding phonon spectra of $Ce(Cu_{1-x}Ni_x)_2Ge_2$ were obtained by a linear interpolation from the two reference compounds. First, the phonon correction was performed by subtracting the interpolated phonon intensities of the nonmagnetic compounds normalized to the mean scattering length. Secondly, the phonon correction can be based on the different Q-dependences of the phononic and magnetic scattering, respectively. Here, the nonmagnetic reference compounds are used to determine the ratio of the high-angle and low-angle scattering intensities. This ratio is used to scale the corresponding magnetic intensities [8]. The correction procedure has been performed in a mutually consistent way for the two methods. The corrected data, which show the magnetic intensities of $Ce(Cu_{1-x}Ni_x)_2Ge_2$ only, are displayed in figure 1. For $x = 0.1$ and $x = 0.28$ a well defined peak is observed close to $\hbar\omega = 18$ meV. For higher concentrations only weak humps are located at around 20 meV. The magnetic transition within the ground state is hidden by the incoherent elastic scattering. For $x = 0.28$, 0.5 and 0.65 small peaks appear close to 6 meV. We believe that these additional intensities are artificially introduced by the subtraction of the quasielastic intensities. A shoulder appears on the high-frequency wing in $S(\boldsymbol{Q}, \omega)$ for $x = 0.28$. This contribution is possibly of magnetic origin and we did not take it into account in the subsequent analysis.

Table 1. The parameters intensity, resonance frequency Δ and damping Γ resulting from fitting the INS spectra of $Ce(Cu_{1-x}Ni_x)_2Ge_2$ by a single Lorentzian line.

$Ce(Cu_{1-x}\ Ni_x)_2Ge_2$			
x	I (au)	Δ (meV)	Γ (meV)
0.10	8.8	15.9	5.0
0.28	6.9	16.2	5.4
0.50	1.3	19.2	5.4
0.65	2.0	19.2	7.9

To analyse the data we proceeded as follows. In a first step we fitted the excitations close to 20 meV simply by a single Lorentzian line. In a second step we analysed $S(\boldsymbol{Q}, \omega)$ for $x = 0.1$ and 0.28 using a complete CF Hamiltonian in order to test whether a consistent description of the CF can be achieved. On the basis of a detailed analysis of the CF level scheme in $CeCu_2Ge_2$ using INS, susceptibility and magnetization data [2] as well as on the basis of heat capacity experiments [9] we analysed the data using a single Lorentzian, assuming that the peak at an energy transfer of approximately 20 meV is due to a pseudoquartet [2]. Furthermore, we assumed that any residual linewidth comes from an intrinsic line broadening of the CF levels due to hybridization effects. The results of the

fits using, in addition to the background, the three parameters of a Lorentzian line, namely the intensity I, the resonance frequency Δ and the damping Γ, are indicated as dashed lines for all concentrations in figure 1. The resulting parameters are listed in table 1.

Table 2. CF parameters and corresponding level schemes of $Ce(Cu_{1-x}Ni_x)_2Ge_2$ for $x = 0$, 0.1, 0.28. The data for pure $CeCu_2Ge_2$ are included for comparison and are taken from reference [2].

$CeCu_2Ge_2$		
CF	$W = 5.436$ meV	$B_2^0 = -0.757$ meV
parameters	$x_{40} = -0.05$	$B_4^0 = -0.005$ meV
	$x_{44} = 0.531$	$B_4^4 = 0.24$ meV
Level	16.46 meV	$0.435\|\pm 5/2\rangle + 0.9\|\mp 3/2\rangle$
scheme		$0.23\|\pm 1/2\rangle$
	0 meV	$0.9\|\pm 5/2\rangle - 0.435\|\mp 3/2\rangle$
$Ce(Cu_{1-x}Ni_x)_2Ge_2$, $x = 0.1$		
CF	$W = 5.26 \pm 1.45$ meV	$B_2^0 = -0.028 \pm 0.007$ meV
parameters	$x_{40} = -0.502 \pm 0.22$	$B_4^0 = -0.044 \pm 0.02$ meV
	$x_{44} = 0.482 \pm 0.302$	$B_4^4 = 0.211 \pm 0.13$ meV
Level	15.74 meV	$0.39\|\pm 5/2\rangle + 0.92\|\mp 3/2\rangle$
scheme	15.46 meV	$\|\pm 1/2\rangle$
	0 meV	$0.92\|\pm 5/2\rangle - 0.39\|\mp 3/2\rangle$
$Ce(Cu_{1-x}Ni_x)_2Ge_2$, $x = 0.28$		
CF	$W = 5.14 \pm 0.55$ meV	$B_2^0 = -0.125 \pm 0.02$ meV
parameters	$x_{40} = -0.628 \pm 0.08$	$B_4^0 = -0.054 \pm 0.007$ meV
	$x_{44} = 0.305 \pm 0.274$	$B_4^4 = 0.131 \pm 0.1$ meV
Level	16.11 meV	$\|\mp 1/2\rangle$
scheme	15.75 meV	$0.23\|\pm 5/2\rangle - 0.97\|\mp 3/2\rangle$
	0 meV	$0.97\|\pm 5/2\rangle + 0.23\|\mp 3/2\rangle$

To get a more detailed description of the CF parameters, next we tried to fit the results for $x = 0.1$ and 0.28 using the complete CF Hamiltonian. Of course, the analysis is hampered by the fact that only a single line has been observed, and that the intensities close to or even below the elastic line are ill defined. The CF Hamiltonian operator of tetragonal symmetry, following the parametrization proposed by Hutchings, according to [10], is given by

$$H_{CF} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4. \tag{1}$$

Generalizing the formalism of Lea, Leask and Wolf [11], this Hamiltonian can be rewritten for cases of lower than cubic symmetry [12]:

$$H_{CF} = W(x_{20}\mathcal{O}_2^0 + x_{40}\mathcal{O}_4^0 + x_{44}\mathcal{O}_4^4). \tag{2}$$

As $\sum |x_{nm}| = 1$ and $\mathrm{sgn}(x_{20}) = \mathrm{sgn}(W)$, it is sufficient to determine the three parameters W, x_{40} and x_{44}. Since only one peak could be detected in our INS measurements within the experimental resolution, additional information is needed to perform an analysis in terms

of CF wave functions. Within the fitting procedure, a Lorentzian line shape has been assumed for the CF transitions. It has been shown theoretically to be a good approximation, particularly at high temperatures [13]. The quasielastic linewidths, as determined on IN6 before [3], were taken as a measure of the width of the CF excitations and kept fixed. Otherwise, the series did not converge and many solutions are permitted to describe the data. This is the crucial assumption, and it is justified by the fact that we could not find any significant variation of the scattering law in its dependence on Q. The widths are 0.69 meV for $x = 0.1$ and 0.83 meV for $x = 0.28$, respectively [3]. In combination with the Lorentzian line shape we conclude that any broadening of a transition is, at least chiefly, dominated by dynamical relaxation processes and not due to dispersion or coupling to other modes. In HFS, this intrinsic relaxational broadening is determined by the residual quasielastic linewidth as a measure of the Kondo-lattice temperature. The Lorentzian lines have been convoluted with the instrumental resolution. In accord with symmetry considerations, three Kramers doublets have been assumed. Based on the three plausible assumptions of a constant (quasielastic) linewidth, a Lorentzian line shape and a level scheme consisting of three doublets, the CF parameters for $x = 0.1$ and $x = 0.28$ could be determined. The starting values for pure $CeCu_2Ge_2$ were taken from reference [2] and then the CF parameters were fitted to the data for $CeCu_{1.8}Ni_{0.2}Ge_2$. The parameters obtained were then taken as new starting values for $CeCu_{1.44}Ni_{0.56}Ge_2$. The fits describe the data satisfactorily and are displayed in figure 1 as full lines. According to our assumptions, these curves each correspond to a sum of three Lorentzians, multiplied by the detailed-balance factor. The positions of the two Lorentzians describing the first and second excited states as well as the intensity ratios are determined by the CF level scheme, whereas the widths were kept fixed. The CF parameters and the corresponding level schemes are summarized in table 2. For comparison, the values of $CeCu_2Ge_2$ are included too. The CF splitting and the corresponding wave functions are very similar for all three compounds. This provides further support for the validity of our assumptions, since for low Ni concentrations, no drastic changes of the CF parameters of $CeCu_2Ge_2$ are expected. This is also evident from resistivity and static susceptibility measurements [3]. As is evident from figure 1, the assumption of three doublets cannot be extracted from the data alone. Indeed, the level schemes in table 2 indicate an energy separation of the second from the first excited state less than the intrinsic width of each level. As in the case of pure $CeCu_2Ge_2$ [2] and $CeCu_2Si_2$ [14], the CF splitting of $Ce(Cu_{1-x}Ni_x)_2Ge_2$ for $x = 0.1$ and $x = 0.28$ may well be interpreted as a doublet–quasiquartet. A totally different behaviour is observed for $x = 0.5$ and $x = 0.65$, as displayed in the lower part of figure 1. In these cases, the (inelastic) magnetic signal was too weak to provide a reasonable fit, and no CF level schemes could be determined.

3. Discussion and conclusions

The typical heavy-fermion phenomena take place on a low-energy scale determined by the Kondo-lattice temperature, and are thus limited to a low-temperature range. At higher temperatures, HFS usually behave like ordinary magnetic metals. For a determination of the ground state and of the heavy-fermion behaviour, the excited states due to crystal-field or spin–orbit interactions can be neglected in many cases. Apparently, the situation will change when the hybridization strength is strong enough that the Kondo-lattice temperature becomes comparable to the CF splitting energies Δ. Then the CF state will collapse and the corresponding electronic state recovers its full degeneracy. (In the present case, the $^2F_{5/2}$ state of the Ce^{3+} ion would become sixfold degenerate.) Indeed, HFS with $T^* \geqslant 30$ K and

valence fluctuation compounds often show a structureless and broad quasielastic response rather than well defined CF excitations.

The surprising result of our present INS study is that the magnetic intensities of the CF transitions are strongly suppressed for $x = 0.5$ and $x = 0.65$, even though the Kondo-lattice temperatures are at least one order of magnitude below the typical CF energies Δ. Quasielastic neutron scattering revealed [3] that within the actual composition range $0 \leqslant x \leqslant 0.65$, the Kondo temperature varied almost linearly with concentration from $T^* = 8$ K ($CeCu_2Ge_2$, $x = 0$) to $T^* = 14.4$ K ($CeCu_{0.7}Ni_{1.3}Ge_2$, $x = 0.65$). The typical CF splitting energy Δ, as found for $x = 0$, 0.1 and 0.28, is $\Delta = 187 \pm 5$ K.

Table 3. The Kondo-lattice temperature T^*, overall CF splitting Δ_{CF} and unit-cell volume (at $T \leqslant 20$ K) of some CeM_2X_2 (M = transition metal, X = Si, Ge) heavy-fermion systems where the CF level scheme has been determined, in comparison with $Ce(Cu_{1-x}Ni_x)_2Ge_2$. The compounds are listed with decreasing unit-cell volume.

CeM_2X_2				
M (X)	T^* (K)	Δ_{CF} (meV)	V_{unit} (Å^3)	Reference
Ag (Ge)	3	11.0	200.11	[5]
Au (Ge)	$\leqslant$1	16.9	198.47	[5]
Ag (Si)	6	18.0	190.55	[15]
Au (Si)	1.7	20.5	188.80	[15]
Pd (Si)	10	20.9	176.58	[15]
Cu (Si)	15	31.4	167.34	[14]
$Ce(Cu_{1-x}Ni_x)_2Ge_2$				
x	T^* (K)	Δ_{CF} (meV)	V_{unit} (Å^3)	
0	8	16.5	176.70	
0.1	9	15.7	174.18	
0.28	10	16.1	172.56	
0.5	12.3	—	171.14	
0.65	14.4	—	170.60	

A comparison with other isostructural 1:2:2 compounds is very intriguing. In all CeM_2X_2 (M = transition metal, X = Si, Ge) HFS, where the CF level schemes have been determined [2, 14, 15, 5] the ratio of the different CF parameters is very similar. In particular, $CeCu_2Ge_2$ and $CeCu_2Si_2$ have an almost identical level scheme except for as regards the overall CF splitting [14] that differs roughly by a factor of 2. As pointed out by Goremychkin and Osborn [14], the small values of B_4^0 and the fact that the values of B_4^4 lie between 0.23 and 0.46 meV [15, 5] for the CeM_2X_2 compounds reveal that the X–ligand nearest-neighbour shell has the strongest influence on the CF potential along with an f–p hybridization. From this argument, the CF potential of $Ce(Cu_{1-x}Ni_x)_2Ge_2$ should mainly depend on the Ce–Ge distance along with the increase of the f–p hybridization strength, which in turn depends almost linearly on the Ni content. The differences of the CF potentials within the CeM_2X_2 series would be essentially the overall CF splitting energy Δ. The Kondo temperatures, the total CF splitting and the unit-cell volumes of CeM_2X_2 (M = Ag, Au, Pd, Cu and X = Ge, Si) are listed in table 3 and compared to the corresponding values for $Ce(Cu_{1-x}Ni_x)_2Ge_2$. As is evident, the total CF splitting increases with decreasing unit-cell volume and hence with increasing hybridization. A CF splitting energy of approximately 16 meV as determined in $Ce(Cu_{1-x}Ni_x)_2Ge_2$ is also found in $CeAg_2Ge_2$, which is characterized by a significantly larger unit-cell volume and a correspondingly small Kondo temperature.

On the basis of the value of the unit-cell volume one would expect a total CF splitting of at least 20 meV in $Ce(Cu_{1-x}Ni_x)_2Ge_2$. The possibility of a transfer of spectral weight beyond the 69 meV window is contrasted by predictions of macroscopic and in particular resistivity measurements [3]. Two clearly resolved peaks corresponding to the onset of coherent scattering at $T \leqslant T^*$ and CF excitations at around 100 K have been recorded. With increasing Ni content up to $x \leqslant 0.65$, this double-peak structure is essentially preserved, though the maximum due to CF excitations becomes slightly less pronounced. Thermopower and susceptibility data confirm the results of the resistivity. All of these macroscopic measurements suggest well defined CF states with an almost unaltered level scheme up to $x \approx 0.65$. The dominant role of the hybridization has been demonstrated recently by Jaccard *et al* [16]. By applying pressure on $CeCu_2Ge_2$, a superconducting state has been induced above 70 kbar, when the unit cell has been compressed to the volume of $CeCu_2Si_2$ at ambient pressure. In contrast, when compressing the unit cell of $CeCu_2Ge_2$ by alloying with Ni, the magnetic order is suppressed without being replaced by superconductivity. As discussed in detail in [3], an itinerant form of magnetism formed by the band states of the heavy quasiparticles has been found in $Ce(Cu_{1-x}Ni_x)_2Ge_2$ above the critical concentration of Ni. This new phase starts to develop at $x = 0.5$ and seems to be fully established at $x = 0.8$ [4]. The corresponding suppression of the CF states despite the much lower Kondo-lattice temperature might be a further signature of heavy-fermion band magnetism, especially since the hybridization strength seems to be more strongly influenced by the Ge than by the Ni ligands [14]. The situation is different in $CeIn_{3-x}Sn_x$, the only system in which a progressive broadening and a collapse of CF states upon increasing the hybridization strength has been demonstrated so far [7]. As described in detail in [7], in this system the width (i.e. the hybridization strength) of the CF level is comparable to the splitting energy Δ.

To conclude, we have performed INS experiments on $Ce(Cu_{1-x}Ni_x)_2Ge_2$ to study the evolution of the CF states upon increasing the hybridization. For $x = 0.1$ and 0.28, the data could be described in terms of CF wave functions that are very similar to those of pure $CeCu_2Ge_2$. For higher concentrations $x = 0.5$ and 0.65, no well defined CF transitions could be detected. This suppression of CF states in $Ce(Cu_{1-x}Ni_x)_2Ge_2$ is in contrast to the behaviour of other (isostructural) heavy-fermion compounds where the magnetic behaviour is really dominated by the interplay between the magnetic interactions and the hybridization alone, i.e. those which may be described within the framework of Doniach's phase diagram [17].

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