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Physics Abstracts

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Paramagnetic spectral response of the Kondo lattice tetragonal compound CePt_2Si_2

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Résumé. — Des expériences de diffusion inélastique de neutrons ont été effectuées sur le réseau Kondo tétragonal CePt_2Si_2 . L'analyse de la réponse spectrale paramagnétique révèle l'existence d'une large diffusion de type quasi élastique et de deux transitions inélastiques attribuées au champ cristallin. CePt_2Si_2 apparaît comme intermédiaire entre un composé à fluctuation de valence et un fermion lourd. Les effets de champ cristallin sur les trois composés isostructuraux CeM_2Si_2 ($M = \text{Pt}, \text{Cu}, \text{Ru}$) sont comparés.

Abstract. — Inelastic neutron scattering experiments have been performed on the Kondo-lattice tetragonal compound CePt_2Si_2 . The magnetic spectral response is analysed in terms of a broad quasielastic scattering plus two weak inelastic transitions attributed to crystal field effects. It appears that CePt_2Si_2 is intermediate between valence-fluctuation and heavy-fermion compounds. Crystal field effects in the three isostructural compounds CeM_2Si_2 ($M = \text{Pt}, \text{Cu}, \text{Ru}$) are compared.

1. Introduction.

The inelastic neutron scattering technique has been widely used in recent years for investigating the behaviour of anomalous cerium systems such as heavy fermions, Kondo lattice and intermediate valence compounds. In these latter compounds, a broad quasielastic magnetic spectral response is usually observed at high temperature, with a half-width at half-maximum $\Gamma/2$ of typically 25-30 meV [1], while at low temperature, an inelastic hump appears around 40-50 meV, as in the case of CeSn_3 [2] or CePd_3 [3]. In Kondo lattice or heavy fermions systems, crystal field excitations are often observed, although their magnitude is much weaker than in normal cerium compounds, and a relatively narrow residual quasielastic distribution ($\Gamma/2 \sim 0.5$ meV) subsists at low temperature. Moreover, in a wide temperature range, its width follows a $T^{1/2}$ dependence as found in CeAl_3 [4], CeCu_2Si_2 [5] or CeCu_6 [6] compounds.

Intermediate situations have been found in the pseudo-ternary system $\text{CeSn}_{3-x}\text{In}_x$ where a continuous evolution occurs from the intermediate valence state ($x = 0$) towards a Kondo lattice behaviour when x is increased, as shown by susceptibility and specific heat

measurements [7]. Recent inelastic neutron scattering experiments have confirmed this continuous evolution, showing a Fermi-liquid scaling of the paramagnetic spectral response of these compounds with $T_{\max}$, the temperature where a maximum of susceptibility occurs, with χ ($T \rightarrow 0$) and with $\gamma = C/T$ [8].

In this respect, the compound CePt_2Si_2 appears an interesting candidate for study by inelastic neutron scattering because of its intermediate situation between heavy fermions (Kondo lattice with low T_K) and intermediate valence (IV) systems (high T_K), as shown by specific heat, susceptibility and resistivity measurements [9]. After experimental considerations (Sect. 2), the results of the paramagnetic spectral response are analysed (Sect. 3). Section 4 is devoted to discussion.

2. Experimental.

CePt_2Si_2 crystallizes in the CaBe_2Ge_2 -type tetragonal structure with $a = 4.25 \text{ \AA}$ and $c = 9.79 \text{ \AA}$ [10]. A polycrystalline material has been obtained by melting a stoichiometric amount of pure constituents (99.99 % purity cerium) in a cold crucible using a high-frequency induction furnace. A sample of the isomorphous non-magnetic compound LaPt_2Si_2 was also prepared in order to correct the spectra for phonon scattering.

The neutron scattering measurements were performed on the IN4 time-of-flight spectrometer of the Institut Laue-Langevin in Grenoble, with neutrons of incident energy $E_0 = 50.4 \text{ meV}$, and at various temperatures ranging from 6 K to 200 K.

A preliminary analysis of these data led us to perform an additional experiment at $T = 6 \text{ K}$ with a higher incident energy for the neutrons, i.e. $E = 67 \text{ meV}$. Spectra were collected for scattering angles 2θ ranging between 4° and 100° , in order to follow the Q -dependence of the magnetic scattering as well as that of the phonon scattering.

All the measured spectra were first corrected for the background signal using a standard procedure described previously [2] which yields the total normalized response, with reference to a vanadium standard.

A second correction of the spectra was then performed namely subtracting the phonon contribution. This is illustrated in figure 1. Due to the magnetic form factor, the magnetic response is very weak at high scattering angles ($2\theta = 93^\circ$) where the momentum transfer $Q/4\pi$ ranges from $0.6 \text{ \AA}^{-1}$ to $0.5 \text{ \AA}^{-1}$ depending upon the energy transfer (0-60 meV); the corresponding spectra are then mainly characteristic of the phonon scattering. However, in addition to the fact that the scattering lengths of Ce and La are different, the phonon energies and their dispersion may not be exactly the same for CePt_2Si_2 and the isomorphous non-magnetic compound LaPt_2Si_2 , as can be seen in figures 1a and 1b, preventing direct subtraction of the corresponding spectra, for phonon correction.

Instead, as explained in reference [2], a scaling function was first obtained for the phonons in LaPt_2Si_2 between high and low scattering angles, and the same function was applied to the high-angle data of CePt_2Si_2 in order to obtain the extrapolated low-angle phonon contribution in CePt_2Si_2 (Fig. 1c). This latter was then subtracted from the total spectra, giving the pure magnetic response $S(Q, \omega)$ (Fig. 1d). Finally, the spectra were corrected for variation of intensity with energy transfer $\hbar\omega$ for a fixed scattering angle θ , using the magnetic form factor $f(Q)$, in order to obtain the magnetic response $S(Q, \omega)$ at constant Q [1]. This process of correction assumes no intersite magnetic correlations.

3. Results and analysis.

The magnetic spectral response $S(Q, \omega)$ of CePt_2Si_2 has been analysed in reference [11] for the incident energy $E_0 = 50.4 \text{ meV}$. No well-defined crystal-field transition was observed in

Fig. 1. — Phonon correction procedure in CePt_2Si_2 at 6 K, for different scattering angles 2θ (see text); (a) spectra for LaPt_2Si_2 ; (b) total spectral response of CePt_2Si_2 ; (c) phonon spectral response for CePt_2Si_2 , extrapolated from the $2\theta = 93^\circ$ spectrum of (b); (d) magnetic spectral response of CePt_2Si_2 obtained by subtracting (c) from (b).

the whole energy range 0-40 meV. Rather a very broad magnetic scattering was evidenced with non negligible amplitude at the highest available energy, i.e. 40 meV. These results were qualitatively similar to those obtained by others [12].

These data were not very selective to determine the exact shape of the magnetic scattering. In particular, a satisfactory fit was obtained at $T = 6$ K with either a quasi-elastic scattering (halfwidth $\Gamma_{\text{QE}}/2 \sim 21$ meV), or an inelastic response (position $\omega_{\text{EL}} = 8$ meV, halfwidth $\Gamma_{\text{EL}}/2 = 20$ meV), or within an alternative model, as proposed by Kuramoto and Müller Hartmann [13] ($\tilde{E}_{\text{F}} \sim 18$ meV, $\alpha \sim 1$). In this latter model, the magnetic response is determined by a characteristic energy $\tilde{E}_{\text{F}}$ closely related to T_{K} , and by a parameter $\alpha = \pi n_{\text{f}}/N$ where n_{f} is 4f electron number ($n_{\text{f}} = 1$) and N the degeneracy of the 4f ground state multiplet.

The experiment with the incident neutron energy $E_0 = 67$ meV presented in this paper allowed us to better evaluate the shape of the magnetic response by extending the investigation range up to ~ 60 meV. Our previous assumption [11] of a purely quasielastic scattering at low temperature seems questionable now, since it leads to a systematic overestimation of the magnetic response in the range 40-60 meV (see Fig. 2). On the other hand a pure inelastic (Lorentzian-shape) scattering leads to better agreement in the high energy range, except that its halfwidth of about 16 meV is much larger than its position (~ 8 meV). We find that the best fit is obtained using the Kuramoto expression [13], with

Fig. 2. — One-component (a), two-component (b) and three-component (c) fits of the magnetic spectral response of CePt_2Si_2 at 6 K for an incident neutron energy $E_0 = 67$ meV and a scattering angle $2\theta = 12^\circ$, after correction for the Ce^{3+} form factor; (a) quasielastic model, $\Gamma_{\text{QE}}/2 = 16.8$ meV (continuous line); inelastic model, $\Gamma_{\text{IN}}/2 = 15.9$ meV, $\omega_{\text{IN}} = 8.4$ meV (hatched line); Kuramoto model, $\tilde{E}_{\text{F}} = 16.8$ meV, $\alpha = 1.57$ (dotted line); (b) quasielastic + inelastic model, $\Gamma_{\text{QE}}/2 = 16$ meV, $\Gamma_{\text{IN}}/2 = 2.35$ meV, $\omega_{\text{IN}} = 36.3$ meV (continuous line); inelastic model, $\Gamma_1/2 = 13.9$ meV, $\omega_1 = 9.6$ meV, $\Gamma_2/2 = 4.2$ meV, $\omega_2 = 36.8$ meV (hatched line); (c) quasielastic plus inelastic model, $\Gamma_{\text{QE}}/2 = 12.5$ meV, $\Gamma_1/2 = \Gamma_2/2 = 8$ meV, $\omega_1 = 17.4$ meV, $\omega_2 = 36$ meV (continuous line).

$\tilde{E}_{\text{F}} \sim 17$ meV and $\alpha \sim 1.6$. This latter value leads to a degeneracy close to $N = 2$, quite consistent with a doublet crystal field ground state, as expected in the tetragonal symmetry.

The above results were obtained assuming a single component in the magnetic response. However a more detailed analysis of the experimental curve suggests the existence of a second small magnetic component around $\omega \sim 36$ meV. Although a phonon contribution is present in the range 30-50 meV, its extrapolated magnitude has been slightly overestimated (see Fig. 1c) because the experimental results have shown *a posteriori* that a small magnetic contribution is still present in the uncorrected data of CePt_2Si_2 for $2\theta = 93^\circ$ (Fig. 1b). The corresponding two-component fits are drawn in figure 2, assuming an additional inelastic-type contribution to the three fits described above. Since the Kuramoto model does not include explicitly the crystal field effects it cannot explain the existence of two structures in the magnetic response. Among the other two models, the one with two inelastic components gives excellent agreement with the experimental data and is quite consistent with crystal field effects in tetragonal symmetry, which lead to a raising of degeneracy of the $J = 5/2$ multiplet into 3 doublets. The two components therefore should correspond to transitions from the ground state toward the two excited doublets situated respectively at $\Delta_1 \approx 10$ meV and $\Delta_2 = 37$ meV above the ground state. However, there subsists a large disparity between the linewidth of the two transitions ($\Gamma_1/2 \sim 14$ meV, $\Gamma_2/2 \sim 4$ meV).

That leads us to consider now a three-component model for describing the magnetic response, namely two inelastic lines with *similar* linewidth plus one quasi-elastic magnetic contribution. Although the selectivity is not high, a reasonably good agreement is obtained with inelastic transitions at about $\Delta_1 \sim 17$ meV and $\Delta_2 \approx 36$ meV, the linewidth ranging from $\Gamma_1/2 = \Gamma_2/2 = 6$ meV, $\Gamma_{\text{QE}}/2 = 14$ meV to $\Gamma_1/2 = \Gamma_2/2 = 10$ meV, $\Gamma_{\text{QE}}/2 = 9$ meV (see Fig. 2). This halfwidth of the quasi-elastic magnetic scattering appears more consistent with the values at higher temperature [11]. In particular its temperature dependence is similar to that found in intermediate valence compounds, i.e. nearly constant. The anomalous variation reported in reference [11] is then an artefact due to an incorrect definition of the different component of the magnetic response. This emphasizes the fundamental importance to obtain

the high energy part of $S(Q, \omega)$ in order to properly account for its shape in this type of compound with a broad scattering. The presence of the two inelastic lines will be discussed below in relation with the crystal field effects.

4. Discussion.

From the previous analysis, it appears that the experimental results of paramagnetic spectral response of CePt_2Si_2 can be satisfactorily accounted for by the superimposition of a wide quasielastic contribution ($\Gamma_{\text{QE}}/2 \sim 12$ meV) and two inelastic crystalline electric field (CEF) transitions ($\Delta_1 \sim 17$ meV, $\Delta_2 \sim 36$ meV). The latter have also a rather large halfwidth ($\Gamma_{\text{IN}}/2 \sim 8$ meV), as expected in Kondo lattice compounds [14]. In addition, as the temperature is raised, the intensity of the inelastic transitions decreases relative to the quasielastic part [14] such that at high temperature, the magnetic spectral response can be well described by a single quasielastic behaviour with a halfwidth nearly constant ($\Gamma_{\text{QE}}/2 \sim 14$ meV, see Ref. [11]).

The present interpretation of the low temperature results appears more realistic than previously with regard to the temperature dependence of Γ_{QE} . The latter appears to be nearly constant in the whole temperature range, such a behaviour being similar to that observed in intermediate valence compounds such as CePd_3 or CeSn_3 [15]. A slight increase of Γ_{QE} between 6 and 40 K cannot be completely excluded since the superposition of the different magnetic components at 6 K prevents any certainty. The comparison with other compounds however shows that CePt_2Si_2 is in an intermediate situation between IV compounds (high T_K) and heavy fermions or Kondo lattice compounds (low T_K). In this respect, CePt_2Si_2 can be compared to the cubic $\text{CeSn}_x\text{In}_{3-x}$ system which evolves continuously from an IV regime ($x = 3$) towards a Kondo behaviour ($x = 2$) [8]. Among the different compounds of this series, $\text{CeSn}_{2.5}\text{In}_{0.5}$ behaves similarly to CePt_2Si_2 , with a paramagnetic spectral response $S(Q, \omega)$ exhibiting also a broad but somewhat better pronounced maximum around 20 meV at 5 K, and a similar bulk susceptibility behaviour. The value of T_K obtained from the present data ($T_K \sim \frac{1}{2} \Gamma_{\text{QE}}(T = 0) = 130$ K) seems much larger than from other determinations ($T_K \sim 60$ K, see Ref. [16]), but as mentioned above the uncertainty in determination of Γ_{QE} is fairly large.

From the present results, an estimation of the crystal field can be also undertaken. Indeed the two inelastic transitions evidenced at 6 K in the spectra correspond to transitions from the doublet ground state to the two excited CEF states, as expected for the tetragonal symmetry which splits the $J = 5/2$ multiplet into 3 CEF doublets. The anisotropy of the magnetic susceptibility [16], as well as the values of Δ_1 and Δ_2 allow us to determine the three CEF parameters [17], namely $B_2^0 = 7.4$ K, $B_4^0 = -1.2$ K, $B_4^4 = -1.8$ K. This leads to a $|\pm 1/2\rangle$ ground state Γ_6 , well isolated from the two excited doublet $\Gamma_7^{(1)}$ and $\Gamma_7^{(2)}$. These two latter levels are not pure $|\pm M_J\rangle$ states because the B_4^4 term slightly mixes the $|\pm 5/2\rangle$ and $|\mp 3/2\rangle$ components (see Tab. I), giving a weak anisotropy in the basal plane of the tetragonal structure, in favour of the [100] direction [16]. On the other hand, this plane is strongly favoured compared to the c-axis, mainly due to the B_2^0 term.

The CEF level scheme of CePt_2Si_2 is then noticeably different from that found in other tetragonal compounds like CeCu_2Si_2 or CeRu_2Si_2 . In CeCu_2Si_2 , the crystal field anisotropy is opposite to that of CePt_2Si_2 , and favours the c axis. The CEF parameters given in reference [5] are however not consistent with magnetic measurements performed later on single crystals [18, 19]. In particular they lead to an anisotropy of the reciprocal susceptibility about three times weaker than that found experimentally. We tried to interpret again the neutron scattering results [5] together with those of magnetic measurements, keeping the doublet

Table I. — *Proposed CEF level scheme in CeM_2Si_2 , where $M = \text{Pt, Cu, Ru}$ (see text) ; the associated CEF wavefunctions are $|\pm 1/2\rangle$ for Γ_6 , $a|\pm 5/2\rangle + b|\mp 3/2\rangle$ for $\Gamma_7^{(1)}$ and $b|\pm 5/2\rangle - a|\mp 3/2\rangle$ for $\Gamma_7^{(2)}$; the CEF energies are shown in brackets ; the CEF parameters (in K) are : $B_2^0 = 7.4$, $B_4^0 = -1.2$, $B_4^4 = -1.8$ for CePt_2Si_2 , $B_2^0 = -13.5$, $B_4^0 = 0.73$, $B_4^4 = -2.6$ for CeCu_2Si_2 and $B_2^0 = -39$, $B_4^0 = 1.2$, $B_4^4 = 2$ for CeRu_2Si_2 .*

CePt_2Si_2	CeCu_2Si_2	CeRu_2Si_2
		Γ_6
$\Gamma_7^{(2)}$ (415 K)		
	Γ_6 (364 K)	
		$\Gamma_7^{(2)}$ (220 K)
$\Gamma_7^{(1)}$ (194 K)	$\Gamma_7^{(2)}$ (140 K)	
Γ_6	$\Gamma_7^{(1)}$	$\Gamma_7^{(1)}$
$a = 0.97, b = 0.24$	$a = 0.67, b = 0.74$	$a = 0.97, b = -0.24$

Γ_6 as the first excited state at $\Delta_1 = 140$ K above the ground state, and the second excited doublet $\Gamma_7^{(2)}$ at $\Delta_2 = 364$ K.

Within this assumption, the calculated ratio I_2/I_1 of the two transitions seen with the neutrons is about three times larger than that found experimentally, and the shape of the temperature variation of the reciprocal susceptibility noticeably differs from the experimental one. Considering Γ_6 as the second excited level, we found a much better agreement for the intensity ratio and for the temperature variation of the magnetic susceptibility, at least above 50 K [20]. This solution (see Tab. I) needs to be carefully checked by other experiments in particular with respect to recent results on $\text{CeCu}_{2 \pm 0.2}\text{Si}_2$ [21]. In any case, it does not change the previous consideration of a CEF anisotropy opposite to that in CePt_2Si_2 .

The situation of CeRu_2Si_2 is also opposite to that of CePt_2Si_2 , with an even stronger anisotropy than in CeCu_2Si_2 favouring also the c axis [22]. From magnetic measurements, it has been shown that the CEF ground state is an almost pure $|\pm 5/2\rangle$ doublet [23]. Specific heat experiments on CeRu_2Si_2 [24] have been interpreted by considering an excited doublet situated at about $\Delta_1 = 220$ K above the ground state, while inelastic neutron scattering experiment indicates a weak transition around 270 K [12]. From these considerations, and taking into account the experimental anisotropy of the reciprocal susceptibility [22], we found that CEF parameters $B_2^0 \sim -39$ K, $B_4^0 \sim 1.2$ K and $B_4^4 \sim 2$ K provide a satisfactory agreement of the overall variation of $1/\chi$ along and perpendicular to the c axis. Note that the agreement is better than with parameters given in reference [25] or [26]. As a consequence, the second excited CEF level should lie at about 790 K above the ground state (see Tab. I).

Among the three compounds, CePt_2Si_2 is the only one exhibiting a $|\pm 1/2\rangle$ doublet ground state and consequently a CEF anisotropy favouring the basal plane. As for CeRu_2Si_2 , because of the tetragonal symmetry, the $|\pm 5/2\rangle$ and $|\mp 3/2\rangle$ states are mixed, although weakly compared with CeCu_2Si_2 . However the anisotropy is less strong in CePt_2Si_2 than in CeRu_2Si_2 because of the different ground states for the two compounds.

In summary, the present results confirm the intermediate situation of CePt_2Si_2 , between mixed valence and heavy fermion systems. We emphasize that in this compound with its very broad magnetic scattering as seen by the present inelastic neutron scattering experiments, it

was important to measure the high energy part in order to more reliably describe its energy variation. In particular, it has been shown that the high energy part of the response clearly indicates that it cannot be due to a single quasielastic contribution, as we previously assumed.

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