Neutron scattering study of the heavy-fermion compound $Ceku_2Si_2$

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> We have performed inelastic-neutron-scattering experiments on single crystals of the heavyfermion compound CeRu₂Si₂. At intermediate temperatures (25 K < T < 90 K) the magnetic response, Im (χ/ω) , is well described by a quasielastic Lorentzian as a function of energy. The halfwidth Γ of the Lorentzian follows a \sqrt{T} law. At low temperatures (T < 20 K), the magnetic response, Im (χ/ω) , seems to be better described by an inelastic peak. The half-width Γ remains temperature independent down to the lowest temperature ($T \approx 1.7$ K) and amounts about 1.2 meV yielding a Kondo temperature $T_K = 14$ K. The other important result we are presenting is the existence of magnetic correlations with a modulation characterized by two incommensurate wave vectors $\mathbf{k}_1 = (0.3,0,0)$ and $\mathbf{k}_2 = (0.3,0.3,0)$. These magnetic correlations start to develop already at rather high temperatures (\sim 60 K) but they saturate at $T \approx 15$ K when the magnetic response becomes inelastic. So the hybridation of $4f$ electrons with conduction electrons seems to prevent the divergence of the magnetic correlations which otherwise would give rise to a long-range magnetic ordering.

INTRODUCTION

In cerium and actinide compounds, the hybridization of the f electrons with electrons in the conduction band gives rise to various kinds of interesting phenomena. A magnetic or nonmagnetic ground state can be realized at low temperatures, depending on the strength of the hybridization, characterized by the so-called Kondo temperature T_K . With a small hybridization the system orders magnetically and often develops a complex magnetic ordering as in CeB_6 (Refs. 1 and 2) or anisotropic exchange interactions as in, e.g., $CeSb$.³ By increasing the hybridization, the ordered magnetic moment, decreases gradually, and a nonordered magnetic ground state is finally reached.

Usually cerium or actinide compounds, located at the borderline of the magnetic-nonmagnetic transition, exhibit the so-called heavy-fermion (HF) behavior. The best examples are, e.g., $CeCu₆$ and $UBe₁₃$. The HF behavior is characterized by a very large value of the linear term γ in the specific heat, which is attributed to the formation of quasiparticles, having a very large mass compared with the bare electron mass. Actually, these quasiparticles involve both f and conduction electrons. It is usual to define a second energy scale, called the coherence temperature T^* below which these quasiparticles would have a Fermi-liquid-like behavior.

A large amount of experimental work has been performed on many HF compounds. In particular a lot of neutron-scattering studies have been reported on polycrystalline samples, but only a few extensive studies have been reported on single crystals. The main reason is the difficulty in growing large single crystals. The most interesting results have been obtained on $CeCu₆$ (Refs. 4 and 5) and UPt_3 ⁶ CeCu₆ has a Kondo temperature $T_K \approx 3$ K, and the HF behavior is well established only below $T^* \approx 0.2$ K.⁷ Recent inelastic-neutron-scattering (INS) studies down to $T \approx 0.025$ K have given evidence for antiferromagnetic correlations, and the energy spectra cannot be described by a single quasielastic Lorentzian peak, but can only be accounted for by a broad inelastic peak. $5,8$

In view of this result, it seems quite interesting to investigate more carefully systems with a higher T_K , in order to check whether the behavior found in $CeCu₆$ still persists with a larger hybridization. One of the most suitable systems is $CerRu_2Si_2$, for which large single crystals are available. $Ceku_2Si_2$ is still a moderate HF system $[\gamma=385 \text{ mJ/K}^2 \text{mol})$ (Refs. 9 and 10)] with $T_K \approx 10-25$ \mathbf{K} .^{10,11}

EXPERIMENT

The ternary compound $Ceku_2Si_2$ crystallizes in the well-known tetragonal structure with space group I4/mmm and lattice parameters $a = b = 4.197$ Å and $c = 9.797$ Å. Large single crystals of about 0.4 cm³ have been grown in a three-arc furnace using the Czochralski method. In order to get a larger sample volume, an assembly of two of such crystals has been realized and oriented with the c axis vertical, in order to investigate the magnetic scattering within the (a^*, b^*) plane. The assembly was mounted in a standard cryostat, allowing temperature to be varied from 1.5 up to 300 K. The INS experiments were carried out on the triple-axis spectrometer DN1 installed in the reactor Siloe at CEN-Grenoble.

The experiments were performed in the constant- k_i mode with an incident energy $E_i = 14.3$ meV. The neutron flux was improved by means of vertically focusing pyrolitic graphite monochromator and analyzer. Higher-order contaminations were removed by a graphite filter. Collimations 40'-60'-60'-30' were used yielding a typical energy resolution (FWHM) of the incoherent elastic peak of about 0.7 meV.

RESULTS

Based on our experience with the INS experiment on $CeCu₆$, the most important points to be investigated are the energy width of the magnetic scattering and the q dependence of the magnetic intensity, in order to give evidence for magnetic correlations at low temperatures.

Several energy scans have been performed at $T = 1.7$ and 4.2 K. The most typical ones corresponding to the scattering vectors $Q = (0.7, 0.7, 0)$ and $(0.7, 1, 0)$ are reported in Fig. 1. On these scans we can identify qualitatively two contributions: a large incoherent, nonmagnetic scattering, centered around zero energy, with an energy width corresponding to the instrumental resolution and a magnetic scattering which is maximum for an energy transfer of about 1.4 meV and extends up to about 6 meV. A quantitative analysis of the experimental data will be given below. In this paper we have concentrated our attention mainly on the temperature and q dependence of the magnetic intensity. To do that we have followed the variations of the intensity at the maximum in energy, which was found to be weakly q dependent. For an energy transfer $\hbar \omega \approx 1.6$ meV several q scans have been performed through a few Brillouin zones indicated by thick lines in Fig. 2 representing the reciprocal-lattice space in the basal plane. These q scans give evidence for rather well-defined magnetic peaks in different Brillouin zones. Typical q scans reported in Fig. 3, performed along the direction [110] and [010] of the reciprocal space around the zone center $(1,1,0)$, show maxima of intensity for scattering vectors $Q = (0.7, 0.7, 0), Q = (1.3, 1.3, 0)$ and $Q = (0.7, 1, 0), Q = (1.3, 1, 0),$ respectively. This implies the existence of magnetic correlations characterized by two incommensurate wave vectors $\mathbf{k}_1 = (0.3, 0, 0)$ and $k_2 = (0.3, 0.3, 0)$. Such peaks are observed in each Brillouin zone and are indicated in Fig. 2 by dashed circles, the diameter of which corresponds to the FWHM. From the widths of these magnetic peaks, the correlation length can be determined using the relation $\xi = 2/\Delta q$. Thus, we get correlation lengths of about 4 and 5 A along [100] and [110], respectively, which indeed are relatively small values. However, the sign of the interactions cannot be determined unequivocally, but more likely both ferromagnetic and antiferromagnetic interactions coexist up to second nearest neighbors in the basal planes. Further information is obtained from scans along the [010] direction around the scattering vector $Q=(0, 1, 0)$, as reported in Fig. 4. In particular, around the reciprocal-lattice point $(0,1,0)$, the magnetic scattering is quite different from that observed around the reciprocal-lattice point $(0,2,0)$, which unambiguously demonstrates the 3d character of the correlations. Moreover if we remind that the scattering vector $Q=(0,1,0)$, associated with the

FIG. 1. Energy scans at low temperatures for scattering vectors $Q = (0.7, 1.0, 0)$ and $Q = (0.7, 0.7, 0)$. The open circles are corrected from the elastic incoherent scattering as described in the text. The dot-dashed line represents the elastic incoherent scattering. The dashed and solid lines are fits using Eqs. (1) and (2), respectively.

FIG. 2. Reciprocal lattice of CeRu₂Si₂ in the (a^*,b^*) plane. Dashed circles give the position where the magnetic scattering is peaked; the diameter represents the half-width at half maximum of the peak. The solid lines indicate the regions investigated in q scans.

FIG. 3. Typical q scans performed for an energy transfer $\hbar \omega = 1.6$ meV around the scattering vectors $Q = (1, 1, 0)$ along the [110] and [010] directions. Two incommensurate wave vectors k_1 and k_2 resulting from the modulation of the magnetic scattering can be identified. Solid and dashed lines are guides for the eye.

FIG. 4. q scan performed along the [010] direction for an energy transfer $\hbar \omega = 1.6$ meV around the scattering vector $Q = (0, 1, 0)$, showing the three-dimensional character of the magnetic correlations as explained in the text. The solid line is a guide for the eye.

Brillouin-zone center $(0,1,1)$, corresponds to the wave vector $q=(0, 0, 1)$, the maximum of intensity observed around (0,1,0) indicates an antiferromagnetic (AF) coupling between cerium moments in adjacent (001) planes. Such an AF interplanar coupling has also been observed in the isomorphous compound URu_2Si_2 , ¹² but in this case the in-plane ordering is mostly ferromagnetic. However, no 3d magnetic ordering has been detected in $CeRu₂Si₂$ down to 1.7 K.

The temperature dependence of the magnetic scattering has been investigated by performing both energy scans at $Q=(0.7,0.7,0)$ and q scans for an energy transfer of 1.6 meV, corresponding roughly to the peak maximum at low temperatures. For temperatures up to 90 K q scans along the [110] direction are reported in Fig. 5. The modulation of the magnetic scattering remains incommensurate $[k_2=(0.3,0.3,0)]$ and keeps the same intensity at least up to 10 K. However, above 20 K the q dependence of the magnetic scattering becomes less pronounced, but remains observable at least at temperatures as high as 60 K. These magnetic correlations are

FIG. 5. q scans performed along the [110] direction for an energy transfer $\hbar \omega = 1.6$ meV as a function of q in reduced lattice unit (r.l.u.) at various temperatures. The weak peak observed at $T = 90$ K is due to a phonon contribution. Solid and dashed lines are guides for the eye.

no more distinguished on the scan performed at $T = 90$ K. From this result we can conclude that magnetic correlations start to develop below about $T = 70$ K and increase down to $T = 15$ K, a temperature below which they become saturated.

We come back now to the analysis of the energy scans. As mentioned before, we have to correct the experimental data from a relatively high incoherent contribution which masks more or less the underlying magnetic contribution at low energy. This incoherent contribution has been estimated by fitting carefully the low-energy spectrum at $h\omega \lesssim 0.5$ meV to a resolution-limited Gaussian function. After correction of this contribution a good accuracy is obtained for data with energy transfers $\hbar \omega \gtrsim 0.7$ meV.

For $Q = (0.7, 0.7, 0)$, corresponding to the maximum in the magnetic scattering, several energy scans have been performed from 90 K down to the lowest temperature. A typical scan, performed at $T = 62.5$ K is given in Fig. 6, in which we can see that the magnetic scattering can be well accounted for by a single quasielastic Lorentzian peak (solid line in Fig. 6). This behavior persists down to $T\approx$ 20 K. The temperature dependence of the obtained width Γ of the Lorentzian is reported in Fig. 7. It is well described by a square-root behavior $\Gamma(T) \propto \sqrt{T}$, in agreement with the prediction of the single-impurity Kondo theory.¹³ However, below $T \approx 20$ K, the magnetic scattering can no longer be described by a single quasielastic Lorentzian, as shown by the dotted lines in Fig. ¹ for energy scans performed at $T = 1.7$ and 4.2 K.

FIG. 6. Energy scan at $Q = (0.7, 0.7, 0)$ for $T = 62.5$ K. The solid line corresponds to a fit using Eq. (1).

FIG. 7. Temperature dependence of the half-width Γ of the Lorentzian. The solid line corresponds to a \sqrt{T} law. Open circles are T_1 T values in arbitrary units as measured by NMR experiments (Ref. 11). T_1T , proportional to Γ , exhibits a quite similar temperature dependence.

The analysis with a single quasielastic Lorentzian peak for the dynamical susceptibility has been done with the following expression for scans performed in the constant k_i mode:

$$
I(\mathbf{Q}, \omega) = Ck_f^3(\cot \theta_a) f^2(\mathbf{Q}) \chi^{zz}(\mathbf{q})
$$

$$
\times \frac{1}{1 - \exp(-\beta \hbar \omega)} \left[\frac{\omega}{\Gamma} \right] \frac{1}{1 + (\omega/\Gamma)^2}
$$
 (1)

in which θ_a is the Bragg angle of the analyzer, C a scaling constant, and the other symbols have their usual meaning. The geometrical term $k_f^3 \cot \theta_a$, which arises from considerations on the scattered intensity measured on a three-axis spectrometer in a constant k_i ; mode, has been discussed by Dorner.¹⁴ By using Eq. (1) , we have implicitly assumed that corrections due to the instrumental resolution were negligible.

For temperatures much smaller than the excitation energy (here typically 15 K) the balance factor plays an important role, by depressing the scattering on the energygain side of the neutron, which even disappears completely below $T \approx 5$ K.

The purely magnetic contribution obtained after correction of the incoherent elastic scattering is given in Fig. ¹ by the open circles. Then, at low temperatures the magnetic scattering is strongly peaked at a finite energy \approx 1.4 meV. Within the framework of a single quasielastic Lorentzian peak, i.e., with only one single parameter Γ , it is not possible to account for both the energy width $\Delta\omega$ and the peak position ω_{max} which actually are related to Γ by the relations $\Delta \omega \simeq \sqrt{12}\Gamma$ and $\omega_{\text{max}} \simeq \Gamma$. Moreover the low-energy data clearly show the inadequacy of this model (see Fig. 1). Therefore, we propose to use a damped inelastic-scattering function, centered at a finite energy $\pm \hbar \omega_0$ with a Lorentzian shape as given by the expression:

$$
I(\mathbf{Q}, \omega) = Ck_f^3(\cot \theta_a) f^2(\mathbf{Q}) \chi^{zz}(\mathbf{q})
$$

$$
\times \frac{1}{1 - \exp(-\beta \hbar \omega)} (\omega/\Gamma)
$$

$$
\times \left[\frac{1}{1 + \left(\frac{\omega - \omega_0}{\Gamma} \right)^2} + \frac{1}{1 + \left(\frac{\omega + \omega_0}{\Gamma} \right)^2} \right].
$$
 (2)

The best fits with such an expression, represented by solid lines in Fig. 1, are, in general, in a better agreement with the experimental data. From $T \approx 20$ K down to $T = 1.7$ K the obtained parameters ($\hbar \omega_0$ and Γ) exhibit no temperature dependence (see Fig. 7) and take the typical values $\hbar \omega_0 = 1.2 \pm 0.2$ meV and $\Gamma = 1.2 \pm 0.2$ meV demonstrating both a tendency to some inelastic character of the scattering and a strong damping since $\Gamma \sim \hbar \omega_0$. So at low temperatures the analysis of the data indicates that the scattering is certainly not purely quasielastic. However, we cannot rule out the existence of both a broad quasielastic and inelastic contributions of comparable magnitude. Further experiments are in progress to clarify this point. A better description of the experimental results could be obtained by using a scattering function decreasing more rapidly on the lower-energy side, but necessarily with a finite value at zero energy to account necessarily with a finite value at zero energy to account
for the NMR results.¹¹ Above 20 K the inelasticity is progressively lost by increasing temperature, and the magnetic scattering becomes quasielastic with a Lorentzian shape as the magnetic correlations weaken.

DISCUSSION

One of the main results, deduced from these experiments, is the occurrence at low temperatures of magnetic correlations peaked at incommensurate wave vectors indicating competing couplings between first and second nearest neighbors. A typical feature of $Cer Ru₂Si₂$, which seems to be a general behavior of HF compounds, as, e.g., $CeCu₆,^{4,5}$ or $UPt₃,⁶$ is the development of magnetic correlations when the temperature is decreased. However, they do remain of short-range character even down to the lowest temperatures, in contrast with what is observed in URu_2Si_2 .¹² Actually the temperature depen dence of the magnetic intensity allows us to define three temperature regimes: (i) $T \gtrsim 70$ K, where magnetic corrections have collapsed; (ii) 20 K $\leq T \leq 70$ K, where magnetic correlations increase upon cooling; (iii) $T \lesssim 20$ K, where the magnetic scattering becomes temperature independent indicating that correlations are fully established but remain of short-range character.

These temperature regimes have been more or less observed in various magnetic and transport measure-These temperature regimes have been more or less observed in various magnetic and transport measurements.^{9–11,15} In particular, transport measurements give evidence for a characteristic temperature of about 20 K, at which the low-field Hall constant and the thermoelectric power reach a maximum, whereas the specific heat exhibits a maximum at 12 K which was first explained by single-site effects.¹⁰ In the light of our present results, we

think that these behaviors must be attributed to the building up of magnetic correlations and their saturation below 15—20 K. Indirect evidences for HF correlations are obtained from the large deviation to a Curie-Weiss law below 50 K and from the observation of a metamagneticlike transition in the magnetization curves at low
temperatures.¹⁵ The critical field value $H_{\sim} \approx 80$ The critical field value $H_c \approx 80$ kOe $\approx \hbar \omega_0/2\mu_B$ correlates well with the maximum energy of the peak observed in the INS experiments. This is also a confirmation for the existence of an energy gap in the magnetic response of $CeRu₂Si₂$.

The other important result which emerges from this study is the observation of a cross over in the magnetic response from a quasielastic behavior at high temperatures, to an inelastic one below about 20 K, i.e., around the temperature at which the correlations saturate. Moreover, a typical feature is that the peak position $\langle \hbar \omega_0 \rangle$ and the energy width (Γ) are almost constant from 15—20 K down to 1.7 K. A similar behavior has also been observed recently in the single crystal INS study on the HF compound $CeCu₆$.^{5,8} As mentioned above, the balance factor modifies strongly the scattering function when $\hbar \omega / kT \ll 1$, in such a manner that intensities at $\hbar\omega \simeq 0$ cannot be accurately determined by INS experiments. Fortunately this energy range can be investigated by NMR experiments which probe magnetic fluctuations at the Larmor frequency. In particular information on the Γ value can be extracted by measuring the nuclear relaxation time T_1 , which is related to Γ by the relation:

$$
\Gamma(T) \sim \sum_{\mathbf{q}} A(\mathbf{q}) \chi(\mathbf{q}) T_1 T \tag{3}
$$

in which $A(q)$ is the q-dependent hyperfine coupling constant.

The temperature dependence of T_1T in CeRu₂Si₂ has
en reported by Kitaoka *et al.*¹¹ Since $\chi(\mathbf{q})$ is found to been reported by Kitaoka et al.¹¹ Since $\chi(q)$ is found to be T independent up to about 20 K from neutron data, the scaling constant in Eq. (3} can be determined by adjusting $T_1 T$ with Γ as determined by neutron scattering at $T \approx 20$ K. A comparison between the Γ values determined from the INS and the rescaled NMR results is given in Fig. 7. A systematic deviation by about a factor 0.7 is observed at low temperatures (as $T\rightarrow 0$) which could indicate a slightly stronger decrease of $Im\chi(q, \hbar\omega)$ when $\hbar\omega \rightarrow 0$ than that predicted by Eq. (2). Nevertheless, the obtained orders of magnitude are consistent in both experiments, which make us confident in the analysis of the INS data. Thus, from these experimental results, the magnetic response below about 20 K is well described by an inelastic function, in particular no additional peak is developing at very low energy in the heavy-fermion regime. The inelastic character is, however, less pronounced in CeRu₂Si₂ ($\gamma \approx 380$ mJ/K² mol) than in URu₂Si₂ ($\gamma \approx 180 \text{ mJ/K}^2$ mol) for which welldefined inelastic peaks have been observed at low temperatures.¹² Moreover, the temperature dependence of the damping parameter Γ of the magnetic excitations does not show in URu_2Si_2 (Ref. 12) the saturation observed in $CeRu₂Si₂$, due essentially to the proximity of a transition to long-range order below $T_N \approx 17.5$ K. From the theoretical point of view the change from a quasielastic to an inelastic magnetic response upon cooling has been predicted by single-impurity models both for the intermediate valence case and the Kondo limit using the $1/N_f$ expansion (N_f being the Ce³⁺ ground-state degenera cv).¹⁶⁻¹⁸ Unfortunately, none of these calculations explain quantitatively the experimental data, at least without the introduction of unrealistic parameters. The main reason for this discrepancy is that a reasonable shape of the magnetic response function can only be obtained by taking a large degeneracy for the $4f$ groundstate level $(N_f \sim 6)$. In the case of a well-isolated ground-state doublet, as for $CeRu₂Si₂$, the response function is very close to a quasielastic Lorentzian whatever the value of T_K as was already underlined in the INS study of $CeCu₆$.⁵ Therefore we conclude that singleimpurity models cannot account for the inelastic peak and its temperature dependence at low temperatures, a result not so surprising in fact.

Many calculations have been reported concerning the Anderson lattice using various approaches.¹⁹⁻²² These calculations do not seem to explain the experimental results because they predict both a quasielastic and an inelastic peak corresponding to intra- and interband excitations of quasiparticles, respectively. If the inelastic peak at $\hbar\omega_0 \sim T_K$ may correspond to the observed peak, it is rather difficult to extract, at lower energy, an additional quasielastic contribution from our data. Besides such a peak should be strongly temperature dependent²³ below T_K , a behavior not observed at all in NMR experi- T_K , a behavior not observed at all in NMR experiments.¹¹ However, nothing is known on the damping of intraband quasiparticle excitations. Recently, Brandow reported a strong q dependence of both the quasielastic and the inelastic peaks for a square lattice which is actually the case of $CeRu₂Si₂$. This theoretical prediction is also not verified in the present INS experiments. Therefore, we conclude that the description of the Fermi liquid state at low temperatures must absolutely incorporate intersite magnetic correlations. Such a theory is not yet completely available. Attempts to take into account magnetic interactions have recently been done. In particular Shiba, 23 by means of a variational Monte Carlo method, has shown that the nature of magnetic correlations depends on band filling and can be peaked at incommensurate wave vectors. When intersite couplings are of the order of T_K , Fye et al.²⁴ have found that magnetic correlations build up when the temperature is lowered down to about T_K and then remain constant with moments retaining residual correlations at $T=0$. None of these calculations give a dependence in energy.

In conclusion the picture which emerges from these INS experiments is that $Cer(u_2Si_2$ would like to develop at low temperatures a long-range incommensurate magnetic ordering, but the hybridization ($-T_K$) is too large and prevents the divergence of the correlation lengths to occur. This behavior could be a general feature of heavy-fermion compounds which are actually located close to the magnetic-nonmagnetic transition. While $CeRu₂Si₂$ is not a very heavy HF compound, it is actually located very close to this transition because a dilution of Ce by only 13% La already induces a long-range order.²⁵ Preliminary elastic-neutron-scattering experiments have shown that the magnetic order is incommensurate²⁶ with a wave vector $k=(0.306,0,0)$ similar to that where magnetic fluctuations are peaked in pure $CeRu₂Si₂$. However, when Ru is replaced by Rh an antiferromagnetic ordering was found to develop. 27

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