PHYSICAL REVIEW B VOLUME 23, NUMBER 7 1 APRIL 1981

Magnetic neutron scattering and crystal-field states in $CeCu_2Si_2$

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(Received 28 July 1980)

The energy-loss spectra of $CeCu_2Si_2$ and $LaCu_2Si_2$ have been determined using neutrons of high incident energy. The magnetic part of the low-temperature spectra of $CeCu_2Si_2$ consists of a quasielastic line and two inelastic lines at 12.5 and 31 meV resulting from transitions between the Kramers ground state and two excited crystal-field doublets of Ce^{3+} in tetragonal symmetry. When positions as well as intensities of the inelastic lines are used, the crystal-field level scheme can be derived. The temperature dependence and magnitude of the quasielastic linewidth suggest that the contribution of Kondo-type spin fluctuations to the spin dynamics of $CeCu_2Si_2$ predominates over that of valence fluctuations. No indication of any magnetic ordering in $CeCu_2Si_2$ was found by neutron diffraction down to the mK range.

I. INTRODUCTION

The low-temperature behavior of the tetragonal compound $CeCu_2Si_2$ poses some challenging problems to experimentalists as well as theorists: according to its bulk properties, $CeCu_2Si_2$ may be classified as a nearly magnetic, strongly interacting Fermi system, which has been found to undergo a second-order phase transition around 0.5 K.[1] Although some of the phenomenologically related Ce intermetallics like $CeAl_2$ (Ref. 2) are known to order antiferromagnetically, it has been concluded from (i) the vanishing of the electrical resistivity, (ii) a large, static Meissner effect, and (iii) the temperature dependence of the critical magnetic field, that $CeCu_2Si_2$ assumes a superconducting state below $T_c \simeq 0.5$ K.[1]

There are two possible mechanisms which could explain the low-temperature formation of "Fermi-liquid effects" in such a Ce intermetallic, i.e., valence fluctuations[3] and the Kondo effect.[4] $CePd_3$ (Ref. 5) and $CeSn_3$ (Ref. 6) may serve as typical examples of homogeneous *intermediate valence* (IV) compounds while $CeAl_3$ (Ref. 7) and $CeAl_2$ (Ref. 2), for which no valence fluctuations could be found, may be considered as examples of "Kondo-lattice" systems.[8] As has been stated earlier, $CeCu_2Si_2$ seems to be close to the borderline between IV compounds and Kondo lattices.[9] This point of view has been supported by recent experiments: while room-temperature measurements of both the $3d$ and $4d$ x-ray photoelectron spectra[10] and the x-ray absorption edge[11] of the Ce ions show (within an experimental resolution of $\simeq 10\%$) that only the Ce^{3+} configuration and no Ce^{4+} configuration is present in $CeCu_2Si_2$; from lattice-parameter measurements, an increase of the average Ce valence by a few percent is inferred upon cooling the system down to helium temperature.[12] This latter observation supports results of the nuclear quadrupole resonance[13] which reveal an "intermediate" electric-field gradient at the Cu sites in $CeCu_2Si_2$.

Measurement of the magnetic neutron scattering is an appropriate tool to further characterize the status of $CeCu_2Si_2$. It was found that IV compounds like $CePd_3$ and $CeSn_3$ show an almost temperature-independent half-width $\Gamma/2$ of the central ("quasielastic") line in the neutron scattering spectra, while, for instance, $CeAl_3$ (Ref. 14) exhibits a temperature dependence of $\Gamma/2(T)$ similar to that found earlier for the dilute Kondo alloy *Cu*Fe.[15] We note that this difference in phenomenological behavior of IV and Kondo systems is — at least — qualitatively traced by corresponding theoretical results.[16] There exist further important differences between the two types of materials: (i) the residual ($T \rightarrow 0$) value of $\Gamma/2$ is 20–25 meV for $CePd_3$ and $CeSn_3$ and $\lesssim 0.5$ meV for $CeAl_3$,[14] pointing to a characteristic valence-fluctuation temperature of the order 200 K for the former compounds and to a characteristic spin-fluctuation ("Kondo") temperature of only a few K for $CeAl_3$; (ii) while no inelastic magnetic lines due to crystal-field (CF) transitions could be resolved for both $CePd_3$ and $CeSn_3$,[17] such CF contributions were seen in the neutron scattering spectra of $CeAl_3$.[18]

Preliminary magnetic neutron scattering of $CeCu_2Si_2$ has been measured by one of the authors.[6] These experiments were initiated to determine the temperature dependence of the quasielastic linewidth $\Gamma/2(T)$ and were therefore performed with neutrons of low incident energy, i.e., 3.53 and 12.6 meV. Although no CF-transitions could be detected in the low-temperature energy-loss spectra up to an energy transfer of 8 meV, an inelastic magnetic contribution (Lorentzian at 26 meV with half-width 12 meV) had to be assumed in order to obtain a reasonable fit of the high-temperature (220–300 K) energy-gain spectra. The existence of CF-transitions in $CeCu_2Si_2$ has also been inferred from measurements of the electronic transport properties.[9]

In order to directly observe such CF-transitions and thus to obtain unambiguously the temperature dependence of the quasielastic linewidth we have performed new neutron scattering experiments on $CeCu_2Si_2$ and $LaCu_2Si_2$ employing sufficiently high incident neutron energies. In addition, we have measured the neutron diffraction on $CeCu_2Si_2$ well above and below the transition temperature $T_c \simeq 0.5$ K in order to search for a possible magnetic structure in the low-temperature phase of this compound.

II. EXPERIMENT

$LaCu_2Si_2$ and $CeCu_2Si_2$ samples were prepared by induction melting and subsequent pulverization under argon atmosphere. The $CeCu_2Si_2$ sample was identical to No. 4 used for previous bulk measurements.[1,19]

Inelastic neutron scattering experiments were made using the IN4 time-of-flight instrument at the high-flux reactor of the Institut Laue-Langevin (ILL), Grenoble, providing an incident neutron energy of 51 meV.

Measurements of the neutron diffraction were performed with aid of the multidetector instrument D1b at ILL in the same way as in previous work on $CeAl_2$ (Ref. 20) as well as $CeAl_3$ and Ce_3Al_{11}.[21] The sample was filled in a cylindrical Cu cell with inner (outer) diameter of 8(9) mm and 60 mm length. This cell was attached to the mixing chamber of a ^3He-^4He dilution refrigerator. The lowest temperature accessible was 20 mK, as measured by two carbon resistors, located on different sides of the Cu cylinder and precalibrated by means of a ^{60}*Co*Co nuclear orientation thermometer.

III. RESULTS AND DISCUSSION

Figure 1 shows the energy-loss spectra of $CeCu_2Si_2$ taken at 10 and 100 K and $LaCu_2Si_2$ at 100 K. These spectra represent an average of 13 counters, corresponding to an average scattering angle $\theta = 14°$. They were obtained from the measured time-of-flight spectra after both correction for background and absorption and transformation to an energy-transfer ($\hbar\omega$) scale. The scattering law $\bar{S}(\hbar\omega, \theta)$ is given in absolute intensities as obtained by calibration with a vanadium standard. The spectrum of $LaCu_2Si_2$ indicates nuclear-incoherent, elastic scattering and phonon-inelastic scattering centered around $\hbar\omega = 15$ meV ($\theta = 14°$). Using the coherent scattering lengths[22] and the masses of Ce and La, the phonon contribution to the spectrum at 100 K is estimated to

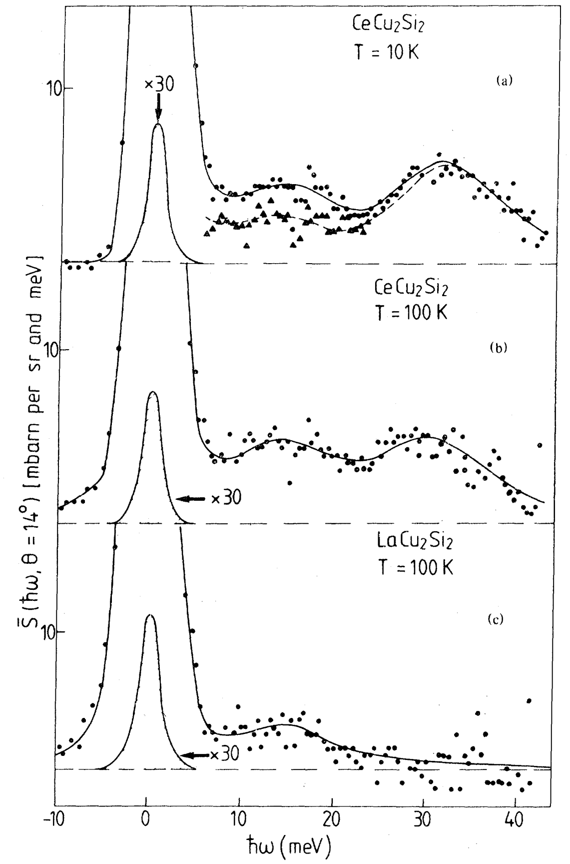

FIG. 1. Scattering law for average scattering angle $\theta = 14°$ as a function of energy transfer $\hbar\omega$ as obtained for $CeCu_2Si_2$ (a) and (b) and $LaCu_2Si_2$ (c) by IN4 instrument (full circles). Solid lines represent a fit to the data points as described in the text. The difference spectrum (triangles) $CeCu_2Si_2$ ($T = 10$ K) minus $(0.72)LaCu_2Si_2$ ($T = 100$ K) shows the existence of the inelastic magnetic line around $\hbar\omega = 12.5$ meV, which is masked by phonon scattering in the original spectrum. The dashed line represents the difference between the fit spectrum of $CeCu_2Si_2$ ($T = 10$ K) and 0.72 times the fit spectrum of $LaCu_2Si_2$ ($T = 100$ K). The factor 0.72 is estimated from the different nuclear coherent scattering lengths of $CeCu_2Si_2$ and $LaCu_2Si_2$ and the different phonon population at 10 and 100 K, see text.

be smaller by $\simeq 20\%$ for $CeCu_2Si_2$ than for $LaCu_2Si_2$. A further 10% reduction at 10 K is caused by the usual phonon-population factor.

After subtraction of this phonon part from the spectrum of $CeCu_2Si_2$ as measured at 10 K, we are left with considerable intensity in the energy range of 5 to 40 meV [see dashed line in Fig. 1(a)]. This intensity must be due to *magnetic* scattering. We have refrained from extracting the magnetic scattering intensity from the low-energy (< 5 meV) spectrum, which is dominated by the nuclear-elastic, incoherent contributions when using the IN4 instrument. However, from previous work at the D7 instrument using neutrons of low incident energy E_0 (3.53 meV) and rather high-energy resolution $\Delta E/E_0$ (0.3 meV), we know the half-width of the quasielastic magnetic line, i.e., $\Gamma/2 \simeq 1$ meV at $T = 10$ K.[6] We, therefore, conclude the magnetic intensity for energies > 5 meV to be *inelastic* in origin.

Evidently, this inelastic magnetic part of the spectrum consists of a well pronounced peak at $\hbar\omega = 31.5$ meV and, in addition, a less pronounced, albeit clearly visible, peak at $\hbar\omega = 12$ meV. These two contributions can be reasonably well fitted by two Lorentzians centered at $\hbar\omega = k_B(135 \pm 15)$ and $k_B(360 \pm 20)$ K. We attribute them to the two CF transitions from the ground-state doublet, $|0\rangle$, to the excited doublets, $|1\rangle$ and $|2\rangle$, as expected for Ce^{3+} ions on tetragonal sites. The ratio of the intensities for the two transitions is found from this fit to be 1:3.4. We note that the energy of the $|0\rangle \rightarrow |1\rangle$ transitions is in satisfactory agreement with the temperature (100 K) where the resistivity has a peak when plotted versus temperature.[9] No structure in the $\rho(T)$ curves of $CeCu_2Si_2$ is known so far to correspond to the $|0\rangle \rightarrow |2\rangle$ transition, simply because this is to expected to happen near 400 K, i.e., outside the temperature range of the transport experiments conducted hitherto.

At 100 K, the position of the low-energy inelastic line, which is obtained after subtraction of the phonon line derived, as explained above, from the $LaCu_2Si_2$ spectrum [Fig. 1(c)] remains unchanged, while the high-energy inelastic line appears shifted somewhat toward lower energies [Fig. 1(b)]. This reflects the fact that a finite part of the Ce ions are in the first excited CF state, so that transitions $|1\rangle \rightarrow |2\rangle$ with $E_2 - E_1 = k_B(220$ K) can contribute to the energy-loss spectrum.

In the following, we want to discuss quantitatively the measured spectra of $CeCu_2Si_2$ in terms of CF theory. The CF Hamiltonian for a rare-earth ion with tetragonal site symmetry is

$$H_{\mathrm{CF}} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^4 O_4^4 + B_6^0 O_6^0 + B_6^4 O_6^4 \quad , \tag{1}$$

where the c axis of the tetragonal cell has been chosen as quantization axis. For Ce^{3+} ($J = \frac{5}{2}$) the last two terms can be omitted.[23] The operators O_n^m and the coefficients B_n^m are as defined by Hutchings.[23] By diagonalization of Eq. (1), one can express the matrix elements $|\langle n|J_\perp|m\rangle|^2$ of the component of the total angular momentum operator J perpendicular to the scattering vector $\vec{Q}$ as well as the CF energies E_n as functions of the CF parameters B_2^0, B_4^0, and B_4^4. By simultaneously fitting the ratio of the intensities of the two inelastic lines, $|\langle 0|J_\perp|1\rangle|^2 / |\langle 0|J_\perp|2\rangle|^2$, and the splitting energies, E_1 and E_2 ($E_0 = 0$) to the measured values, we find the CF level scheme shown in Fig. 2 (with the CF parameters as noted in the caption). The matrix elements and CF energies as calculated with aid of the wave functions given in Fig. 2 agree well with the corresponding values as obtained from the measured spectra (cf. Table I). On the basis of this level scheme, the magnetic part of the neutron scattering spectra can be calculated and compared to the measured spectra of Fig. 1. As discussed in Ref. 5, the double differential magnetic cross section is given by (Ω: solid angle)

$$\frac{d^2\sigma}{d\Omega\, d(\hbar\omega)} = \frac{1}{2}\left(\frac{1.91 r_e}{\mu_B}\right)^2 \frac{k_f}{k_i} F_l^2(Q)\, \chi_l(T)\, P(\hbar\omega, T) \times \frac{\hbar\omega}{1-\exp(-\hbar\omega/k_B T)} \quad , \tag{2}$$

if one can assume that (i) the magnetic moments of the ions show relaxation behavior and (ii) no spatial correlations between them exist. In case of a CF-split ground state of Ce^{3+}, we may substitute in Eq. (2) (Ref. 24)

$$\frac{2}{\mu_B^2} \chi_l(T) P(\hbar\omega, T) = \sum_{n,m} \rho_n |\langle n|J_\perp|m\rangle|^2\, P^{nm}(\hbar\omega, T) \quad , \tag{2a}$$

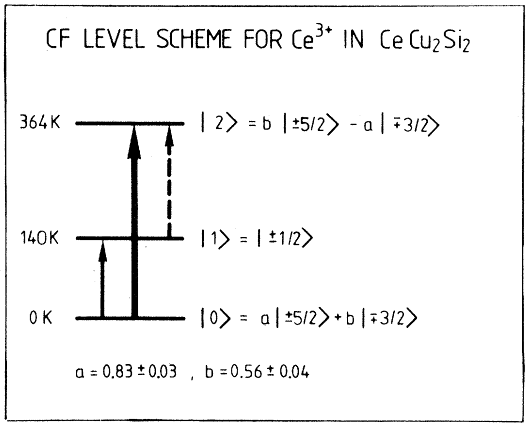

FIG. 2. Crystal-field level scheme of Ce^{3+} in the tetragonal compound $CeCu_2Si_2$ as obtained from the measured spectra of Fig. 1 and by diagonalization of Eq. (1), yielding $B_2^0 = -3.0 \pm 1.0$, $B_4^0 = -0.4 \pm 0.1$, $B_4^4 = -0.25 \pm 0.05$, and the transition energies and matrix elements as collected in Table I (for details see text).

TABLE I. Matrix elements $M_{nm} = |\langle n|J_{\perp}|m\rangle|^2$ and transition energies $\hbar\omega_{nm}$ as calculated and as measured, respectively (for details see text).

Transition	M_{nm}^{calc}	M_{nm}^{meas}	$\frac{\hbar\omega_{nm}^{\text{calc}}}{k_B}$ (K)	$\frac{\hbar\omega_{nm}^{\text{meas}}}{k_B}$ (K)
$n=0 \rightarrow m=1$	1.61	1.65	139	135 ± 15
$n=0 \rightarrow m=2$	5.1	5.6	364	360 ± 20
$n=1 \rightarrow m=2$	3.7	$\cdots$	225	$\cdots$

with

$$P^{nm}(\hbar\omega,T)=\frac{1-\exp(-\hbar\omega_{nm}/k_BT)}{\hbar\omega_{nm}} \times L(\Gamma_{nm},\hbar\omega-\hbar\omega_{nm}) \ . \tag{2b}$$

Here $L(\Gamma_{nm},\hbar\omega-\hbar\omega_{nm})$ are normalized Lorentzians, with linewidths [full width at half maximum (FWHM)] Γ_{nm}, $\hbar\omega_{nm}=E_n-E_m$ the CF-transition energies, $\rho_n=\exp(-E_n/k_BT)/Z$, where Z is the partition function, $E_l(Q)$ the local magnetic form factor, k_i,k_f the wave numbers of the initial and final states of the neutron, r_e the classical electron radius, and μ_B the Bohr magneton. For $\Gamma_{nm}\rightarrow 0$, the double differential cross section reduces to that given by Birgeneau[25] for sharp, i.e., infinitely lived, CF levels. If one folds $(k_i/k_f)[d^2\sigma/d\Omega\, d(\hbar\omega)]$ with the instrumental resolution function, one can calculate the scattering law $\bar{S}(\hbar\omega,\theta)$ (see Fig. 1). Using the widths Γ_{nm} as adjustable parameters, and employing the nuclear-elastic and phonon-inelastic contributions as discussed above, we obtain a very good fit of the $CeCu_2Si_2$ spectra as shown by the solid curves in Fig. 1.

In the absence of spatial correlations between magnetic moments, the local susceptibility $\chi_l(T)$ should coincide with the static bulk susceptibility. This has, in fact, been observed for the IV compounds $CePd_3$ (Ref. 5) and $CeSn_3$ (Ref. 6). However, in the case of $CeCu_2Si_2$, $\chi_l(T)$ as determined from the CF level scheme and employed in the above fit procedure exceeds considerably the static bulk susceptibility $\chi_m(T)$ as measured.[26] While at helium temperatures $\chi_l(T)$ is dominated by the Curie part of the CF ground state, $|0\rangle$, $\chi_m(T)$ may be approximated by a Curie-Weiss law.[26] If we introduce a "temperature-dependent Curie-Weiss temperature," $\theta(T)$, we can write in the whole temperature range up to 300 K

$$\chi_m(T)=\chi_l(T)\frac{T}{T+\theta(T)} \ . \tag{3}$$

As shown in Fig. 3, we find $\theta_0=\theta(T\rightarrow 0)\simeq 40$ K, a rather strong increase of $\theta(T)$ up to 100 K and some flattening off at higher temperature.

Since the local susceptibility is found to be larger than the bulk susceptibility, one could be tempted to infer antiferromagnetic correlations between the Ce^{3+} magnetic moments. From the value $\theta_0\simeq 40$ K one would then expect an antiferromagnetic phase transition at or below this temperature. As mentioned in the Introduction, a phase transition takes place in $CeCu_2Si_2$ at $T_c\simeq 0.5$ K, but has been attributed according to bulk measurements to the onset of superconductivity rather than antiferromagnetism.[1] This conclusion is strongly supported by the results of our neutron-diffraction experiments. Figure 4 shows the diffraction pattern of $CeCu_2Si_2$ at $T=1.5$ K, and the difference between the patterns, which were obtained after counting for 12 h in each case at $T=0.02$ and 1.5 K, i.e., well below and above T_c, respectively. At low temperature, there is no indication of either an increase in intensity at the nuclear Bragg reflections (peaks in the 1.5-K pattern) or additional magnetic Bragg reflections. This experiment confines the aver-

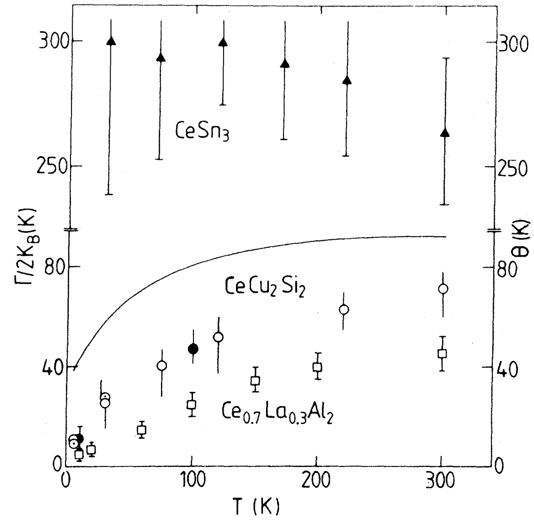

FIG. 3. Temperature dependence of phenomenological Curie-Weiss temperature θ as defined by Eq. (3) (solid line) and of half-width of quasielastic line divided by k_B for $CeCu_2Si_2$. Energies of incident neutrons used: $E_0=3.5$ meV (○) and 12.4 meV (⊙) (Ref. 6), $E_0=51$ meV (●) (this work). Also shown is $\Gamma/2k_B(T)$ for $CeSn_3$ ($E_0=3.5$ meV, Ref. 6) and $Ce_{0.7}La_{0.3}Al_2$ ($E_0=3.5$ meV, Ref. 28).

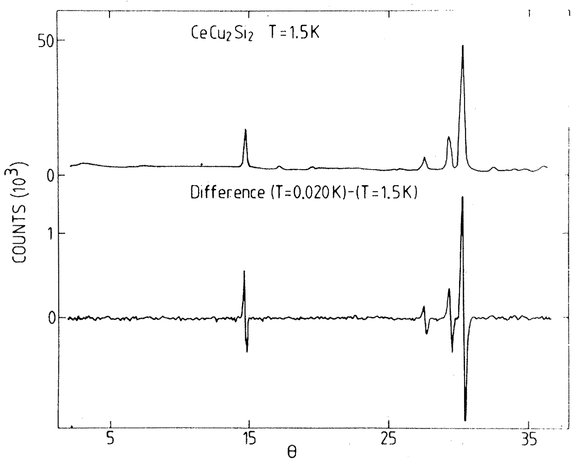

FIG. 4. Neutron diffraction results of $CeCu_2Si_2$ as obtained by multidetector instrument D1b. Top: diffraction pattern at $T = 1.5$ K; bottom: difference of diffraction patterns at $T = 0.02$ and 1.5 K (note change in vertical scale).

age Ce moment in any ferromagnetically or antiferromagnetically ordered phase of $CeCu_2Si_2$ at very low temperatures to be $\bar{\mu} < 0.1\mu_B$.

The neutron-diffraction results of Fig. 4 also suggest that the discrepancy between the local and bulk susceptibilities as found above helium temperature cannot be caused by antiferromagnetic correlations between the Ce ions, but indicates correlations between the $4f$ electron of Ce^{3+} and the conduction electrons mediated by "Kondo-type" spin fluctuations. For dilute Kondo alloys, the Curie-Weiss temperature may be correlated to the characteristic spin-fluctuation temperature, T_K, i.e., $\Theta_0 \simeq 4.5T_K$.[27] On the other hand, the spin-fluctuation temperature of $CeCu_2Si_2$ may be read off the residual half-width of the quasielastic neutron line, $\Gamma/2(T \rightarrow 0)$. In Fig. 3 is plotted the temperature variation $\Gamma/2k_B(T)$ obtained from the present neutron scattering experiments as well as from a reanalysis of the previous data,[6] which was done employing both positions and intensities of the CF transitions as given above. In fact, we find $\Gamma/2k_B(T \rightarrow 0) \simeq 10$ K, i.e., a spin-fluctuation temperature about four times smaller than the Curie-Weiss temperature Θ_0. In addition, we find the temperature dependencies of $\Gamma/2k_B$ and Θ to be quite similar.

For comparison we also show in Fig. 3 $\Gamma/2k_B(T)$ for the IV compound $CeSn_3$ (Ref. 6) lacking, within experimental error, significant temperature variation as well as for $La_{0.3}Ce_{0.7}Al_2$,[28] a well-defined Kondo system.[29] In the latter, the Ce^{3+} ions experience—in time average—a cubic site symmetry by which the $J = \frac{5}{2}$ state is CF split into a Γ_7 ground-state doublet and an excited Γ_8 quartet[30] separated from Γ_7 by $k_B \cdot 100$ K. The structure in the $\Gamma/2k_B(T)$ curve of this system around 70 K has been attributed to an increase of the relaxation rate of the Ce^{3+} magnetic moments when excited CF levels become populated.[28] A similar, albeit structureless, temperature dependence of the relaxation rate is found for $CeCu_2Si_2$ and, as we have mentioned before, is traced by the thermal variation of the phenomenological Curie-Weiss temperature $\theta(T)$.

IV. CONCLUSION

We have observed by means of inelastic neutron scattering experiments with high-energy neutrons transitions between the crystal-field sublevels of the $J = \frac{5}{2}$ configuration of Ce^{3+} in $CeCu_2Si_2$. The wave functions and energies of the CF levels could be derived from our experiments. Knowledge of the inelastic contributions allowed us to obtain accurately the quasielastic part of the magnetic neutron scattering spectra. The size of the half-width $\Gamma/2$ of the quasielastic line of $CeCu_2Si_2$ is close to that of typical Kondo systems on the one hand, but considerably smaller than that of typical IV systems on the other. Also, instead of being almost independent of temperature as in IV systems, $\Gamma/2$ depends on temperature as in Kondo systems. Our results suggest that, while valence fluctuations evidenced by other experiments[12,13] apparently contribute only a small part to the quasielastic linewidth of $CeCu_2Si_2$, if at all, the relaxation behavior of Ce is predominated in this compound by Kondo-type spin fluctuations. Our experiments prove that a peak in the temperature dependence of the resistivity as previously observed for $CeCu_2Si_2$ at 100 K (Ref. 9) is caused by CF effects. We expect to find additional structure due to the highest CF doublet in the $\rho(T)$ curve by extending the temperature range of the resistivity measurement to $T > 300$ K. This experiment is in preparation. On the other hand, the present work disproves our previous explanation of a pronounced $\rho(T)$ peak at 20 K as being due to CF splitting, too.[9] Rather, we now believe that this peak indicates the formation of a low-temperature, coherent state of Ce scattering centers, which is to be expected in the translationally invariant lattice of an intermetallic compound.

No magnetic structure could be detected in the neutron-diffraction pattern of $CeCu_2Si_2$ well below the transition temperature $T_c \simeq 0.5$ K. This lends additional support to the existence of a superconducting low-temperature phase in $CeCu_2Si_2$ as concluded from previous macroscopic experiments.[1]

ACKNOWLEDGMENT

Part of this work was supported by Sonderforschungsbereiche 65 and 125.

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