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AN INELASTIC NEUTRON SCATTERING STUDY ON THE HEAVY FERMION SYSTEMS CeCu_2Ge_2 AND CeAg_2Ge_2

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An inelastic neutron scattering study is performed on the new magnetically ordered heavy-fermion systems CeCu_2Ge_2 and CeAg_2Ge_2 . The temperature dependence of the quasielastic line can be interpreted in terms of a Kondo type of behavior. In addition crystalline electric field transition energies are reported.

CeCu_2Ge_2 and CeAg_2Ge_2 are isostructural to the prototype heavy fermion system CeCu_2Si_2 . These ternary compounds crystallize in the tetragonal ThCr_2Si_2 structure [1]. The metallic linear T term in the specific heat $C = \gamma T$ is large pointing towards high effective masses m^* . However, these systems undergo a transition into a magnetically ordered ground state before a heavy Fermi liquid phase is fully built up. The results from specific heat [2], resistivity [1] and dc susceptibility [3] measurements indicate the onset of magnetic order in both systems with transition temperatures $T_N = (4.15 \pm 0.05)$ K and 7 K for CeCu_2Ge_2 and CeAg_2Ge_2 , respectively.

We performed inelastic neutron scattering experiments and investigated in detail

- (i) the splitting of the $4f^1$ ground state J -multiplet in the presence of the crystalline electric field (CEF) and
- (ii) the temperature dependence of the magnetic relaxation rate, as determined by the width of the quasielastic line in the magnetic neutron scattering spectra

(i) Crystal electric field splitting

The splitting of the ground state multiplet due to crystalline electric field effects was studied using the time-of-flight spectrometer IN4 of the Institute Laue-Langevin (ILL) in Grenoble utilizing incident neutron energies of $E_0 = 12.5, 30$ and 50 meV. Fig. 1 shows the neutron energy-loss spectra of CeCu_2Ge_2 and CeAg_2Ge_2 taken at an average scattering angle of 11.3° at low

temperatures ($T \geq T_N$). In each compound one CEF-transition can be clearly identified (CeCu_2Ge_2 : $\hbar\Delta = 16.5$ meV, CeAg_2Ge_2 : $\hbar\Delta = 11$ meV). An identification of the magnetic scattering is possible by comparing spectra at different temperatures and at low and high scattering angles. In addition, the nonmagnetic iso-

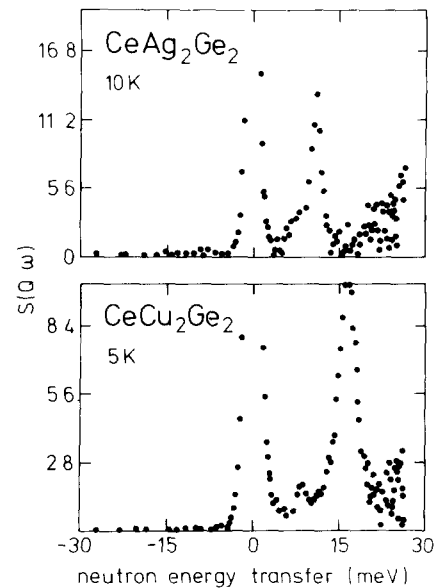


Fig. 1 Scattering law for an average scattering angle $\theta = 11.3^\circ$ as a function of neutron energy transfer $\hbar\omega$ as obtained for CeAg_2Ge_2 at 10 K and for CeCu_2Ge_2 at 5 K. The incident neutron energy for these measurements was 30 meV. The small peak at 10 meV is due to phonon scattering in both compounds.

structural compounds LaCu_2Ge_2 and LaAg_2Ge_2 have been measured. From the symmetry of the Ce-site one expects that the $J = 5/2$ multiplet is split into three doublets interconnected by at least two magnetic dipole transitions with non-zero matrix elements. After preliminary data reduction there is no evidence for a second or even a third crystal field transition in either compound within the range of energy transfers $0.5 \text{ meV} \leq \hbar\omega \leq 50 \text{ meV}$ accessible in the present experiment (a thorough analysis in terms of weighted differences between the spectra of the La- and the Ce-compounds as a function of temperature and scattering angle is in progress). Support for the view that CEF splittings of the present series of isomorphous Ce-compounds can be regarded as an effective two level system comes from specific heat measurements on CeCu_2Ge_2 [2] and on CeCu_2Si_2 [4] which show that the multiplicity of the excited CEF states is four, grouped either in one quartet or two closely spaced doublets. The interpretation of previous neutron measurements on CeCu_2Si_2 [5] is of course at variance with this view. There, two CEF-transitions have been identified, a strong one at 33 meV and a second weaker one at 15 meV. As this weaker transition falls into a range of high phonon density this identification could be somewhat questionable as it might depend on the subtleties of the data reduction, see also ref [6].

It is worthwhile to mention that the line width of the CEF transition in these compounds is very different. The relative full width at half maximum $\Delta E/E \approx 10\%$ (CeAg_2Ge_2), $\approx 25\%$ (CeCu_2Ge_2) and $\approx 35\%$ (CeCu_2Si_2) [5]. At present it is unclear whether this line width effect is due to two closely spaced doublets, due to structural properties, due to magnetoelastic coupling or due to correlation effects of the 4f electrons.

(ii) *Temperature dependence of the line width of the quasielastic line*

In rare-earth compounds with a stable 4f ground-state multiplet one expects a Korringa type of behaviour for the quasielastic line width, namely $\Gamma = \alpha k_B T$. For rare-earth ions with a stable moment α is typically 10^{-3} [7]. Due to a

strong coupling between conduction electrons and the localized 4f electrons some Ce and Yb compounds behave completely differently [7]. In valence fluctuation systems Γ is large and temperature independent. There, the width of the quasielastic line is a measure of the characteristic temperature. In dilute and dense Kondo systems one expects the spin relaxation rate of a Fermi liquid with $\Gamma(T) = \Gamma_0 + \gamma\sqrt{T}$ [7]. The residual line width Γ_0 is a measure of the characteristic Kondo temperature T_K . This behavior has indeed been observed for several Ce-based Kondo systems [8].

The measurements of the quasielastic line

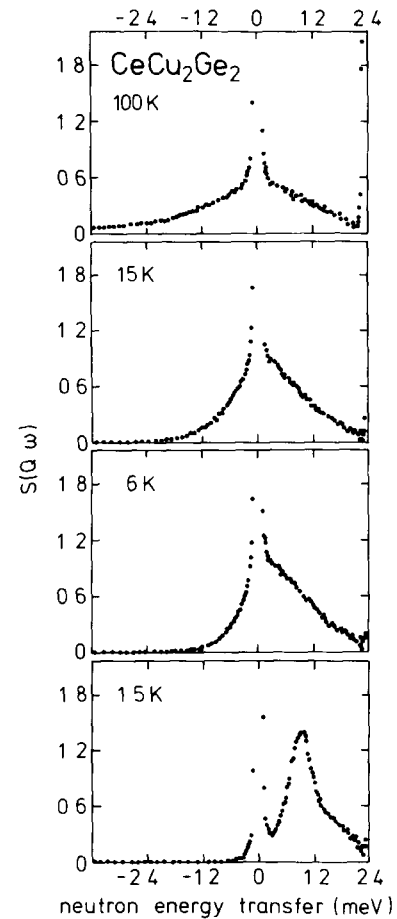


Fig. 2 Scattering law versus $\hbar\omega$ for an average scattering angle of $\theta = 19^\circ$ as obtained for CeCu_2Ge_2 at a series of temperatures ($T_N = (4.15 \pm 0.05) \text{ K}$)

width have been performed at the time-of-flight spectrometer IN6 located at the cold source of the HFR at the ILL. Incident neutron energies of 2.5 meV have been used. Some of the data as obtained in CeCu_2Ge_2 are shown in fig. 2.

At 1.5 K, which is well below T_N , a well defined excitation at 1 meV is clearly observable, which we interpret as a magnon-like excitation in the magnetically ordered phase. With increasing temperatures these inelastic scattered intensities merge into a quasielastic line. In the paramagnetic state ($T > 4.2$ K) this quasielastic line can – in a first approximation – be described by a Lorentzian line shape weighted with a detailed balance factor. For $T \leq 25$ K there appear significant deviations from a single Lorentzian line shape indicating the importance of short-range order correlations.

No well defined magnon lines were detectable in CeAg_2Ge_2 , even at 1.5 K. However, below 7 K Gaussian contributions in addition to the Lorentzian line shapes are important and indicate at least short-range order effects. For $T > 7$ K a single Lorentzian gives a reasonably

well description of the experimentally observed line shapes. Fig. 3 shows the temperature dependence of the half width at half maximum $\Gamma(T)$ of the Lorentzian line for both compounds investigated. The experimental data can well be described using a Kondo type of behaviour with residual line widths Γ_0 of 0.35 meV (CeCu_2Ge_2) and 0.15 meV (CeAg_2Ge_2) yielding characteristic temperatures $T_K = 4$ and 2 K, respectively. The Kondo temperatures in these systems are, therefore, smaller than the magnetic ordering temperatures so that a heavy Fermi-liquid phase cannot be fully developed. Yet, medium-heavy fermions have recently found to exist in the antiferromagnetically ordered phase of CeCu_2Ge_2 [3].

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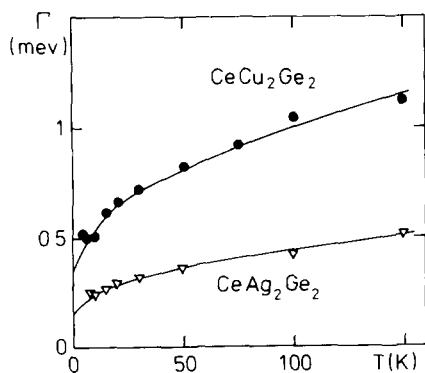


Fig. 3 Temperature dependence of the quasielastic line width Γ (hwhm) for CeCu_2Ge_2 and CeAg_2Ge_2 . The lines are calculated using a Kondo type of behaviour $\Gamma(T) = \Gamma_0 + \gamma\sqrt{T}$.