SPIN RELAXATION DYNAMICS AND MAGNETIC FLUCTUATIONS IN KONDO LATTICES

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The spin relaxation rates Γ in magnetically ordered Kondo lattices have been determined using neutron scattering techniques. Enhanced values of Γ and distinct deviations from a Korringa-type of relaxation behaviour were found. Gaussian contributions to the quasielastic scattered intensities were interpreted as being due to magnetic fluctuations.

CeM_2X_2 compounds are Kondo lattices which exhibit very different ground states: $CeCu_2Si_2$ [1] was the first heavy-fermion system (HFS) reported, which undergoes a superconducting phase transition. $CeCu_2Ge_2$ is a HFS with a magnetic groundstate [2] and $CeNi_2Ge_2$ is characterized by a relatively high Kondo temperature ($T_K \approx 30$ K) and neither shows a magnetic nor a superconducting phase transition [3]. Here we present a systematic investigation of the magnetic relaxation rates Γ in isostructural compounds which undergo magnetic phase transitions: $CeCu_2Ge_2$ ($T_K \approx 8$ K; $T_N = 4.1$ K) [2,4], $CeAg_2Ge_2$ ($T_K \approx 3$ K; $T_N = 7$ K) [4] and $CeAg_2Si_2$($T_K \approx 5$ K; $T_N = 10$ K) [4,5] exhibit modulated magnetic structures below the Néel temperature T_N [5,6]. In these compounds the magnetic phase transition temperatures are close to the Kondo temperatures and thus a delicate balance between the Kondo effect and the RKKY interaction is expected. The width Γ (half width at half maximum: HWHM) of the quasielastic component of the magnetic neutron scattering spectrum determines the magnetic relaxation rate and is due to interactions of the 4f-moment with the conduction electrons. Γ is a direct measure of the stability of the 4f-electrons. In rare earth compounds with a stable 4f-configuration one expects a Korringa-type of behaviour for the temperature dependence of the relaxation rate, namely $\Gamma = \alpha k_B T$, where α is typically 10^{-3} [7]. In systems with non-stable 4f-electrons, Γ is nonmonotonic with a minimum near the Kondo temperature and a square root high temperature behaviour [8].

The measurements of the quasielastic linewidths have been performed on the time-of-flight spectrometer IN6 located on a cold source of the high flux reactor at the Institut Laue Langevin in Grenoble (F). Incoming neutron energies of 3.15 meV were utilized. The energy resolution of the experimental setup was 0.035 meV. Representative results as obtained in $CeAg_2Ge_2$ are shown in fig. 1. The magnetic quasielastic contribution can easily be separated from the narrow central line

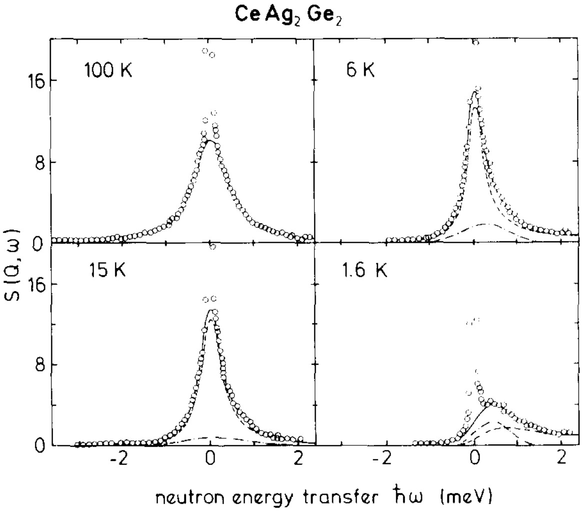

Fig. 1. Scattering law $S(Q, \omega)$ versus neutron energy transfer for an average scattering angle of $\theta = 19$ deg, as obtained for $CeAg_2Ge_2$ at various temperatures above and below the Néel temperature $T_N = 7$ K. The lines are results of fits as described in the text.

which is due elastic incoherent scattering processes. An identification of the magnetic scattering is possible by comparing spectra at different temperatures and different scattering angles. In addition, the neutron spectra of the nonmagnetic isostructural La-compounds allow a direct identification of phonon scattering contributions. At temperatures $T > 30$ K the total quasielastic scattered intensities (solid lines in fig. 1) can well be fitted by a single Lorentzian line (dashed lines). At lower temperatures significant deviations from the Lorentzian line shapes become apparent. These extra intensities can be described using Gaussian line shapes [9] (dashed-dotted lines in fig. 1). At 7 K $CeAg_2Ge_2$ undergoes a magnetic phase transition into a modulated magnetic structure [6]. Just below the phase transition the total magnetic scattering can still be described using two superimposed quasielastic lines (spectrum at 6 K in fig. 1). However, at the lowest temperatures the spectrum is characterized by an inelastic Gaussian contribution which describes magnetic excitations in the ordered state and in addition in quasielastic Lorentzian line which is a measure of the 4f-/conduction electron hybridization. This result demonstrates that magnetic relaxation still exists in the magnetically ordered state, well below the ordering temperature. The quasielastic component would be absent in a normal magnetic system in the ordered phase. Hence we suggest that the persistance of the quasielastic part is characteristic for the HFS.

In fig. 2 we show a summary of the results in the three compounds investigated. In the upper frame we show the quasielastic contributions in the paramagnetic state which can be fitted by a single Lorentzian component. The common signature is a square root behaviour of the magnetic relaxation rates at high and strongly enhanced values at low temperatures. $\Gamma(T)$ in $CeCu_2Ge_2$ exhibits a minimum which determines the Kondo temperature [8]. In the other two compounds, with $T_N > T_K$, the residual line width for $T \rightarrow 0$ K is screened by the magnetic ordering transitions. The overall behaviour of the magnetic relaxation rates, which are a measure of the strength of the 4f-/conduction-electron hybridization correlates nicely with the magnetic properties. These three compounds are characterized by a doublet ground state which is predominantly $|5/2\rangle$ and well separated from the higher CEF-levels [6]. The ordered

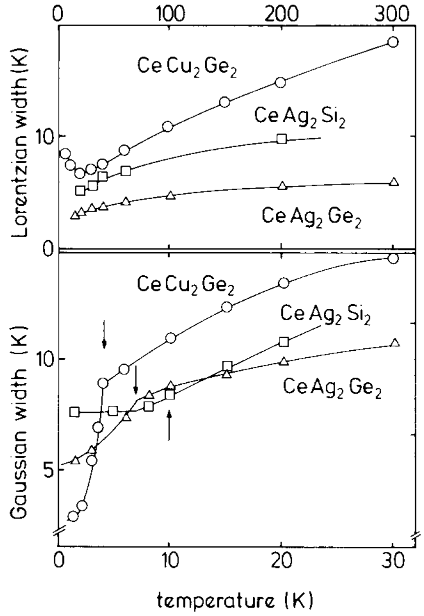

Fig. 2. Temperature dependence of the quasielastic and inelastic line width for $CeCu_2Ge_2$ (○), $CeAG_2Si_2$ (□) and $CeAg_2Ge_2$ (△). The lines are drawn to guide the eye. Upper frame: temperature dependence of the Lorentzian line width in the paramagnetic phase; lower frame: temperature dependence of the Gaussian line width in the paramagnetic and in the magnetically ordered state. The arrows indicate the magnetic phase transition temperatures.

magnetic moment is $0.74\mu_B$ in $CeCu_2Ge_2$ [2], 0.73–$0.93\mu_B$ in $CeAg_2Si_2$ [5] and $1.65\mu_B$ in $CeAg_2Ge_2$ [6]. Obviously, the RKKY interactions are still strong enough to yield magnetically ordered ground states, but the formation of Kondo singlet states, accompanied by the cancellation of the Ce-spins has started. The largest effect appears in $CeCu_2Ge_2$ where the reduction of the magnet moment of the CEF ground state is strongest [2,6]. The antiferromagnetic intersite correlations are demonstrated by the Gaussian contributions to the quasielastic intensities ($T > T_N$) [9], which can be detected already far above the ordering temperatures (lower frame in fig. 2). At the transition temperatures these Gaussian contributions are shifted towards finite frequencies and narrow in energy. For $T < T_N$ they characterize the magnetic excitations in the ordered state.

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