SPIN RELAXATION DYNAMICS IN HEAVY-FERMION SYSTEMS

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The magnetic relaxation rates Γ as determined via inelastic neutron scattering experiments are reported for the ternary compounds CeM_2X_2 (M = Ni, Cu, Ag, Au, Ru; X = Si, Ge). Strongly enhanced values of Γ and deviations from a Korringa-type of behaviour were found. A close correlation between the unit cell volume and the 4f-local spin / conduction electron hybridization is demonstrated.

Heavy-fermion systems (HFS) are stoichiometric metallic compounds containing Ce- or U-ions which exhibit very unusual low-temperature properties: a very large linear term in the specific heat and an almost temperature-independent Pauli-spin susceptibility correspond to high effective masses of the electronic quasiparticles. These systems are thought to be on the borderline between localized magnetic 4f- (5f-) systems and valence fluctuating compounds with extended electronic f-states.

We performed a systematic investigation of the magnetic relaxation rates in CeM_2X_2 (M = Cu, Ag, Au, Ru, Ni; X = Si, Ge) compounds. These Ce-based ternary compounds, which exhibit the $ThCr_2Si_2$ structure, include systems with very different ground states: $CeNi_2Si_2$ [1] is a mixed valent system, $CeCu_2Si_2$ [2] was the first HFS reported to exhibit superconductivity and $CeCu_2Ge_2$ [3, 4] is a heavy-fermion compound with a magnetic ground state. $CeRu_2Si_2$ [5] and $CeNi_2Ge_2$ [6] are systems characterized by high effective electron masses which undergo neither a magnetic nor a superconducting phase transition. Finally, there are a number of compounds with well-localized 4f-electrons [7, 8] exhibiting a pure magnetic ground state, where hybridization effects with the conduction electrons play a minor role only.

The width of the quasielastic line Γ (half width at half maximum) of the Lorentzian component in the magnetic neutron scattering spectrum measures the magnetic relaxation rate and determines the strength of the hybridization between the 4f- and the conduction electrons. In rare earth compounds with a magnetically stable 4f-configuration one expects a Korringa-type of behaviour for the quasielastic line width, namely $\Gamma = \alpha k_B T$, where α is typically 10^{-3} [9]. Cox et al. [10] calculated the magnetic relaxation rate for systems with non-stable f-electrons. They found that i) Γ is non-monotonic with a minimum near the Kondo temperature T_K and reaches a finite low-temperature value of $1.35T_K$, ii) the magnetic relaxation rate shows a non-linear high-temperature behaviour which is well fit by a square root law and iii) significant deviations from a Lorentzian line shape appear for $T \ll T_K$.

The measurements of the quasielastic line widths have been performed on the time-of-flight spectrometers IN4 (located on a thermal neutron source) and IN6 (located on a cold source) at the high flux reactor at the Institut Laue-Langevin in Grenoble, France. Incoming neutron energies of 12.5 meV (IN4) and 3.15 emV (IN6) have been chosen depending on the inverse lifetime of the ground state. Some of the data as obtained at IN6 in $CeNi_2Ge_2$ and $CeAg_2Si_2$ are shown in fig. 1. Well-defined quasielastic magnetic intensities were detected in both compounds. The magnetic relaxation rate Γ in $CeNi_2Ge_2$ is much larger than in $CeAg_2Si_2$, pointing towards a higher Kondo temperature and indicating a stronger

4f- / conduction electron hybridization in the former compound. The measured spectra were fitted with Lorentzian lines including a detailed balance factor. The results for $CeNi_2Ge_2$ are shown as solid lines in fig. 1. The temperature of the minimum value in $\Gamma(T)$ is a reliable estimate of the Kondo temperature $T_K \approx 30\,K$ [6, 10]. Even at the lowest temperatures ($T \ll T_K$), the scattering intensities are well described by Lorentzian line shapes, in contradiction with the theoretical predictions (this might be due to the fact, that according to the theory of ref. [10] the deviations from a Lorentzian quasielastic line shape become pronounced when the degeneracy of the ground state $N_f \geq 4$, while the systems under consideration are characterized by doublet ground state).

In $CeAg_2Si_2$ which orders magnetically at $T_M = 10\,K$ [7] significant deviations from the Lorentzian line shapes appeared for temperatures $T \leq 30\,K$. These additional contributions can be described by quasielastic Gaussian lines in the paramagnetic and with inelastic Gaussian lines in the magnetically ordered state. The results of these fits using Lorentzian (dashed lines) and Gaussian profiles (dashed-dotted lines) are indicated in fig. 1. Following the authors of ref. [11] we associate these Gaussian contributions in the paramagnetic state with critical spin fluctuations which occur as precursors of the magnetic order. In the magnetically ordered phase the inelastic scattering intensities describe magnons. The quasielastic Lorentzian intensities are a measure of the hybridization strength between the 4f- and the conduction electrons and prove the existence of heavy quasiparticles which exist even below T_M. They indicate the existence of a (medium) heavy-fermion state with the magnetic order and point towards an enhanced linear term in the specific heat [8]. Lorentzian quasielastic contributions and an enhanced linear term in the specific heat have been observed in the magnetically ordered state of the HFS $CeCu_2Ge_2$ [3] and $CeAg_2Ge_2$ [8].

In fig. 2 we show a summary of the magnetic relaxation rates Γ versus temperature for $T \geq T_M$ of all compounds investigated. The upper frame of fig. 2 shows $\Gamma(T)$ as obtained in $CeNi_2Ge_2$ and $CeRu_2Si_2$ (the present investigation of the RuSi compound is limited to temperatures $T \geq 10\,K$; a detailed study down to the lowest temperatures is presented in ref. [12]). For comparison we included the temperature dependence of the linewidth as measured in $CeCu_2Si_2$ [13]. The lower frame shows the results for

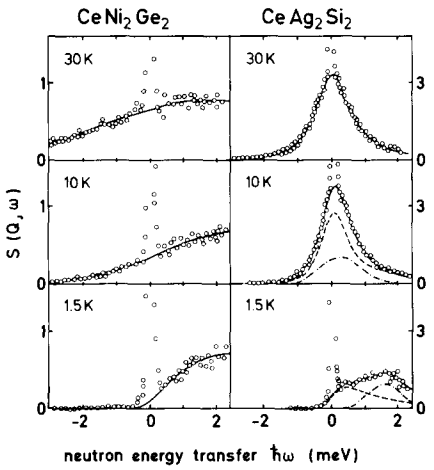

Fig. 1. Scattering function versus neutron energy transfer for an average scattering angle of $\Theta = 19$ deg, as obtained for $CeNi_2Ge_2$ and $CeAg_2Si_2$ at different temperatures. The solid lines are results of fits as described in the text.

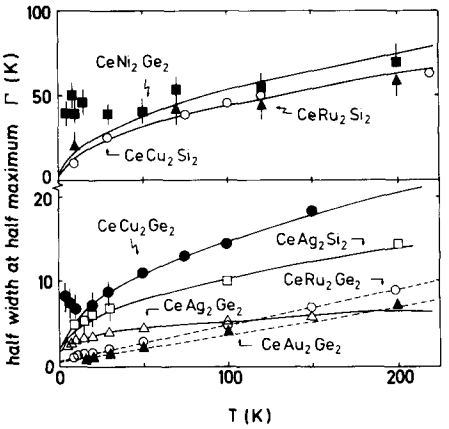

Fig. 2. Temperature dependence of the quasielastic Lorentzian line width in CeM_2X_2 compounds: upper frame: $CeNi_2Ge_2$ (■) and $CeRu_2Si_2$ (▲); for comparison: $CeCu_2Si_2$ (○) (ref. [13]); lower frame: $CeCu_2Ge_2$ (●); $CeAg_2Si_2$ (□); $CeAg_2Ce_2$ (△); $CeAu_2Ge_2$ (▲) and $CeRu_2Ge_2$ (○). The solid lines represent fits using a square root dependence of $\Gamma(T)$.

$CeCu_2Ge_2$, $CeAg_2Si_2$, $CeAg_2Ge_2$, $CeAu_2Ge_2$ and $CeRu_2Ge_2$ which exhibit considerably lower values of the magnetic relaxation rate. The most signigicant results of fig. 2 can be summarized as follows: i) $\Gamma(T)$ in the RuGe and AuGe compounds follow a linear T-dependence. All the other systems exhibit distinct deviations from a Korringa-type of behaviour of the spin relaxation rates; ii) at high temperatures $T > T_K$, $\Gamma(T)$ can be described roughly by a square-root dependence $\Gamma = A\sqrt{T}$ as indicated by the solid lines in fig. 2. The coefficient A approximately scales with the Kondo temperature.

Fig. 2 shows the existence of a close relationship between the solid state environment of the Ce-ions and the stability of the 4f-electrons. Obviously, when moving from Ni→Cu or from Cu→Ag→Au and 4f- / conduction electron hybridization becomes smaller. The same trends can be detected when Si is substituted by Ge. We believe that these trends reflect variations in the electronic properties [14], e.g. an increase of the band width of the conduction electrons when moving from the Si to the Ge series. In this paper we demonstrate that within the $ThCr_2Si_2$ compounds these trends in the electronic properties are correlated with the unit cell volume. In fig. 3 we plotted the magnetic ordering temperatures (T_M) and the estimated Kondo temperatures (T_K) versus the unit cell volume V. This results in a phase diagram analogous to that proposed by Doniach [15]. T_K was determined by the temperature $\Gamma_{min}(T)$ (NiGe, CuGe) or by

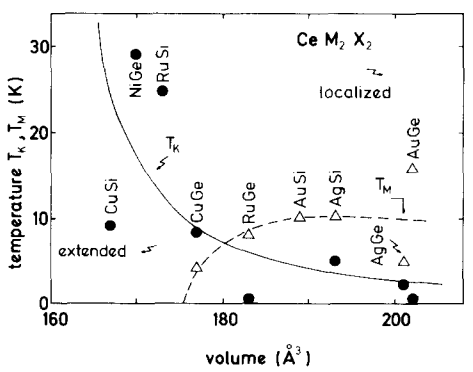

Fig. 3. Kondo temperatures T_K (●) and magnetic ordering temperatures T_M (△) [4, 7, 8] versus the unit cell volume V. The lines are guides to the eye.

values of $\Gamma(T)$ just at T_M (AgSi, AgGe). For the CuSi and RuSi compounds we took $T_K = \Gamma(10\,K)$. Fig. 3 demonstrates that for compounds with large volumes the RKKY interaction between well-localised f-electrons dominates. In this regime ordinary magnets are found, whereas the Kondo effect plays a minor role. For small volumes and low temperatures we find almost extended 4f-electrons. $CeNi_2Si_2$ ($V = 155.6$ Å^3) is a typical intermediate valent system. In a narrow range of unit cell volumes (165 Å$^3 \leq V \leq 185$ Å^3) heavy-fermion systems can be found characterized by a delicate balance between the binding energies of the RKKY antiferromagnetic state and of the Kondo singlet.

Acknowledgements

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