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μSR magnetic studies of $CeNi_{1-x}Cu_x$ ☆

G.M. Kalvius[a,*], E. Schreier[a], A. Kratzer[a], D.R. Noakes[b], R. Wäppling[c], J.I. Espeso[d], J.C Gómez Sal[d], A. Amato[e], Ch. Baines[e]

[a] *Physik-Department, Technical University Munich, James-Franck-Strasse, 85747 Garching, Germany*
[b] *Physik-Department, Viginia State University, Petersburg, VA 23806, USA*
[c] *Physik-Department, Uppsala University, 75121 Uppsala, Sweden*
[d] *Faculty of Science, University of Cantabria, 39005 Santander, Spain*
[e] *Paul Scherrer Institute, 5232 Villigen-PSI, Switzerland*

Abstract

$CeNi_{0.8}Cu_{0.2}$ and $CeNi_{0.4}Cu_{0.6}$ were studied by μSR spectroscopy. They showed two magnetic transitions at low temperatures. The upper one leads to a spin-glass-like state which is not a conventional spin frozen state but a magnetically inhomogenous dynamic spin cluster system with the magnetic inhomogeneities being on a scale of a few lattice constants. The lower transition leads into a long-range ordered state with a high degree of local spin disorder. The local field in $CeNi_{0.4}Cu_{0.6}$ is ~ 500 G, in agreement with dipolar field sums for the most likely muon stopping site derived from crystal potential calculations. The ordered moment in $CeNi_{0.8}Cu_{0.2}$ was estimated to be on the order of $0.1\mu_B$. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Kondo metal; Spin glass; μSR

Strongly correlated electron systems are often characterized by a competition between the on-site Kondo interaction favoring a local nonmagnet state and the indirect RKKY interaction trying to create long-range magnetic order (LRO). Short-range spin correlations (SRC) may also come into play. The $CeNi_{1-x}Cu_x$ system is a good example for this situation. CeNi is nonmagnetic and crystallizes in the CrB structure. In contrast, CeCu is an antiferromagnet ($T_N \approx 4$ K) and possesses the FeB structure (Pnma) which is maintained in $CeNi_{1-x}Cu_x$ for $x > 0.15$. LRO was established for $x \geqslant 0.4$, but to reach this state the compounds first pass from the paramagnetic (PM) state through a spin-glass-like (SGL) regime [1]. The main goal of the μSR study was to gain more information on the magnetic states of these compounds down to very low temperatures (0.1 K), this information being supplemented by further macroscopic measurements [2]. The μSR measurements were carried out at the Paul Scherrer Institute (Switzerland). The samples were polycrystalline ingots melted in an arc furnace under a protective argon atmosphere.

Fig. 1 shows the temperature dependence of the ZF muon spin relaxation rate in $CeNi_{0.8}Cu_{0.2}$. Three regions can be distinguished. For $T \geqslant 20$ K relaxation is very weak reflecting the rapid spin fluctuations in a typical PM regime. Between 10 and 4 K the relaxation rate rises, indicating the formation of spin correlations leading to the SGL state. Below 4 K down to 1 K this state is completely formed and the rate remains constant but is now dependent on field cooled (FC) or zero field cooled (ZFG) measurement conditions. The spectral shape is an exponential decay of muon spin polarization which implies that the SGL regime is not a spin-frozen state but rather a SRC dynamic spin system. LF spectra do not show static 'decoupling' behavior even in fields as large as 4 kG which is characteristic for magnetically inhomogenous states such as a spin cluster system. Before spin freezing can take place the RKKY interaction wins and a transition into a LRO state occurs at

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*Corresponding author. Tel.: +49-89-289-12501; fax: +49-89-320-6780.

E-mail address: kalvius@ph.tum.de (G.M. Kalvius).

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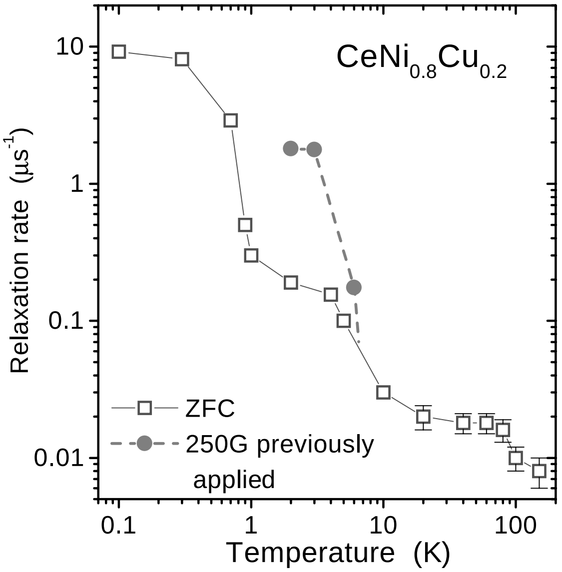

Fig. 1. Temperature dependence of the ZF-μSR relaxation rate in $CeNi_{0.8}Cu_{0.2}$. The lines are guides to the eye. Below 1 K, Λ_t (see Eq. (1)) is plotted.

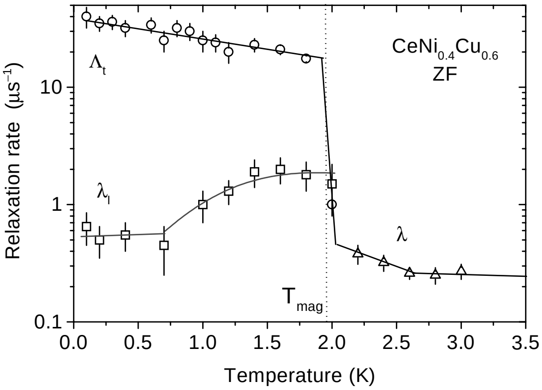

Fig. 2. Temperature dependence of the ZF-μSR relaxation rate in $CeNi_{0.4}Cu_{0.6}$. See text for details.

1 K. For LRO magnetism the μSR response function in case of a (texture free) powder sample is:

$$A(t) = (2a_0/3)\exp[-\Lambda_t t]\cos(\gamma_\mu B_\mu t) + (a_0/3)\exp[-\lambda_l t], \quad (1)$$

with $\gamma_\mu/2\pi = 13.5$ MHz/kG. The transverse relaxation rate Λ_t reflects the static distribution of the field B_μ at the muon site while the longitudinal rate λ_l is proportional to the spin fluctuation rate. In $CeNi_{0.8}Cu_{0.2}$ we find $\Lambda_t \gg \gamma_\mu B_\mu$ and hence an oscillatory pattern is not seen, revealing that despite LRO, considerable *local* spin disorder is present. The Brillouin-like temperature dependence of Λ_t reflects the rise of effective ordered moment on Ce for $T \to 0$. The large value of Λ_t makes a precise determination of B_μ difficult. The least-squares fit gives $B_\mu \sim 100$ G. The longitudinal rate λ_l is always rather low ($\leqslant 0.05\ \mu s^{-1}$) but never truly zero. Such persistent slow spin fluctuations are characteristic for magnetically frustrated systems [3]. TF data established that all muons implanted in the sample contribute to a single spectral pattern for all compounds over the whole temperature ranges scanned. It means that all muons see a similar magnetic surrounding, albeit with widely varying local field magnitude.

For the $CeNi_{0.4}Cu_{0.6}$ compound, the spectrum at 2.6 K was fitted with a 'power-exponential' relaxation ($\exp[-(\lambda t)^p]$), but the spectrum at 0.2 K needed the function given in Eq. (1). The power p was found around 0.5, a value typical for a dynamic random spin system. At 0.2 K a weak indication of an oscillatory signal is discernable. The spin precession frequency corresponds to $B_\mu \approx 500$ G. Also clearly visible is the presence of longitudinal relaxation, stressing once more that slow spin fluctuations persist down to base temperature. The temperature dependence of the relaxation rates λ (for $T > 1.8$ K) and Λ_t (for $T \leqslant 1.8$ K) are plotted in Fig. 2. Overall the μSR results for $CeNi_{0.4}Cu_{0.6}$ are quite similar to those described for $CeNi_{0.8}Cu_{0.2}$. The transition into the SGL state is less apparent (probably around 2.5 K), mainly because of the lack of high temperature data. The transition to the ordered state occurs near 1.6 K. Below this transition the main difference to $CeNi_{0.8}Cu_{0.2}$ is a roughly five times larger transverse relaxation rate due to the increase of the ordered moment with Cu concentration [4]. Neutron diffraction established a simple ferromagnetic spin structure in $CeNi_{0.4}Cu_{0.6}$. μSR data in an external field were compatible with a ferromagnetic component of the spin structure but required a low saturation field (on the order of 500 G) and a weak saturation magnetization. Significantly, μSR finds that despite LRO, the spin system shows considerable disorder on a short-range scale. These two features must be combined in the final evaluation of the proper spin structure.

Quantitative analysis of μSR data requires the knowledge of the muon stopping site. Its experimental determination requires a single crystal specimen which was not available. In lieu, we performed crystal field potential calculations assuming that the muon selects the largest and 'deepest' interstitial hole. The most likely stopping site has the unit cell coordinates $(0.346, 1/4, 0.538)$. It has two nearest Ce neighbors in the $y = \frac{1}{4}$ plane 2.64 Å distant and one nearest neighbor each in the plane above ($y = \frac{3}{4}$) and below ($y = -\frac{1}{4}$) 2.34 Å away, thus corresponding to an almost tetrahedral coordination. Dipole field calculations based on the ferromagnetic spin structure proposed by Espeso et al. [4] gave $B_\mu \sim 940$ G per μ_B. Using $\mu_{ord} = 0.6\mu_B$

from neutron data we obtain $B_\mu \sim 560$ G in acceptable agreement with the μSR results. Taking $B_\mu \sim 100$ G for $\mathrm{CeNi_{0.8}Cu_{0.2}}$ then gives $\mu_{\mathrm{ord}} \sim 0.1\mu_{\mathrm{B}}$ which fits well into the general trend of μ_{ord} vs. Cu concentration and extends this systematics to lower x values. The lower transition temperature in $\mathrm{CeNi_{0.8}Cu_{0.2}}$ is ~ 1 K and differs little from the values found up to $x = 0.6$. As stated, the SGL state must be a magnetically inhomogenous random spin system, most probably a disordered spin cluster state. A system of magnetic clusters embedded in a nonmagnetic matrix for richer Ni compounds had been previously proposed in Ref. [5]. The single response μSR pattern restrict the inhomogeneities to a range of about three lattice constants. Hence the clusters must nearly touch each other and the nonmagnetic matrix, if it exists at all, must be very thin.

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