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Magnetic form factor of the heavy fermion compounds $CeCu_2Ge_2$ and $CeCu_{1.9}Ni_{0.1}Ge_2$

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The heavy fermion compound $CeCu_2Ge_2$ orders magnetically in an incommensurate sinoidally modulated structure below $T_N = 4.15$ K. Doping with 5% Ni results in two superimposed incommensurate magnetic structures which are very similar to the structure of pure $CeCu_2Ge_2$ [1]. Due to the strongly localized character of 4f electrons in rare earths, the trivalent Ce ions are in its Hund's rule electronic configuration. The tetragonal crystalline environment in $CeCu_2Ge_2$ splits the sixfold degenerate $4f^1$, $J = \frac{5}{2}$ level of the Ce ions into three doublets. Inelastic neutron scattering on $CeCu_2Ge_2$ revealed a crystal field (CF) Γ^7-ground state separated from the (first) excited Γ^8state by 191 K [2]. The Γ^8-quartet has to be understood as two nearly degenerate doublets being closer in energy than the experimental resolution. Quasielastic neutron scattering experiments have been performed on $Ce(Cu_{1-x}Ni_x)_2Ge_2$ for $x = 0$, 0.1, 0.28, 0.5 and 0.65 [3]. In principle, the integrated magnetic intensity should be proportional to the neutron magnetic form factor. Due to the averaging over the Brillouin zone by measuring polycrystalline samples, as well as due to some possible variation of the scattering law, quasielastic neutron scattering experiments cannot be taken as a quantitative form factor measurements. Nevertheless, the Ni-doped compounds exhibited an unusual behaviour revealing a

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well-defined maximum of the magnetic intensity in dependence of momentum transfer Q around $1.3\,\text{Å}^{-1}$ [3].

To elucidate the unusual magnetic properties of these compounds, we have performed form factor measurements on single crystalline $CeCu_2Ge_2$ and $CeCu_{1.9}Ni_{0.1}Ge_2$. The neutron magnetic form factor is the structure factor of one single ion. The magnetic structure factors $F_M(\boldsymbol{Q})$, which are the Fourier components of the magnetization density are obtained most accurately by means of polarized neutron diffraction. The experiments have been performed on the diffractometer DN2 at the Siloë reactor of the CEN, Grenoble. The crystals with approximate dimensions of $7 \times 7 \times 0.5\,\text{mm}^3$ were mounted in a cryomagnet with a vertical field of 8 T and were oriented with the reciprocal lattice vectors (1 0 0) and (0 0 1) in the horizontal scattering plane. A Heusler monochromator provided a polarized neutron beam with $\lambda = 1.205\,\text{Å}$. First by measuring 54 ($CeCu_2Ge_2$) and 47 ($CeCu_{1.9}Ni_{0.1}Ge_2$) independent nuclear Bragg reflections, respectively, the lattice constants were refined for both compounds to $a = 4.190\,\text{Å}$ and $c = 10.000\,\text{Å}$. The width of the rocking curves of the nuclear Bragg peaks revealed extremely small mosaicities. Consequently, in such highly perfect crystals strong extinction effects have a major influence onto the measured intensities. The extinction g factors, as deduced from the integrated nuclear intensities have been refined as 2943 and 1268 for $CeCu_2Ge_2$ and $CeCu_{1.9}Ni_{0.1}Ge_2$, respectively.

The neutron magnetic form factor of $CeCu_2Ge_2$ has been established by recording the flipping ratios of 26 independent Bragg peaks at $T = 20\,\text{K}$ in the paramagnetic regime. To obtain the magnetic structure factors, the data have been corrected for secondary extinction, higher order contamination and incomplete polarization of the incoming beam. Only these reflections have been considered where the extinction was not too large, i.e. when a correction within the theory of an ideally imperfect crystal could be performed, leaving still 20 reflections for further analysis. The calculation of the magnetic form factor can be done by the tensor operator method [4]. The experimental results for $CeCu_2Ge_2$ and

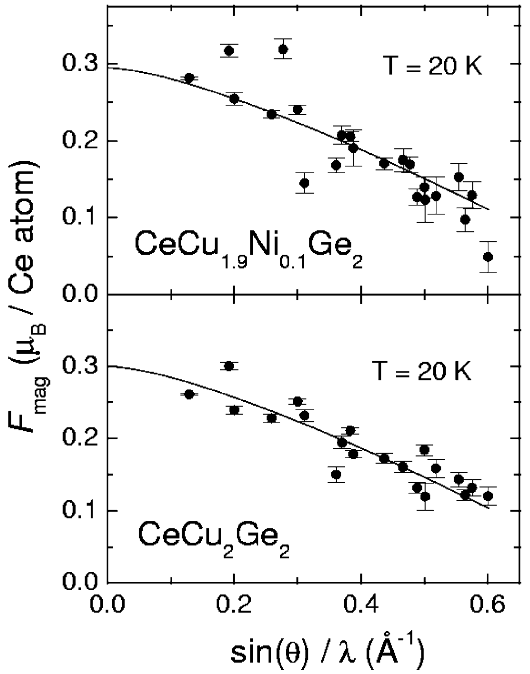

Fig. 1. Magnetic structure factors of $CeCu_2Ge_2$ (lower panel) and $CeCu_{1.9}Ni_{0.1}Ge_2$ (upper panel) at $T = 20\,\text{K}$ in the paramagnetic state in dependence of momentum transfer.

$CeCu_{1.9}Ni_{0.1}Ge_2$ are shown in Fig. 1. The magnetic structure factors of $CeCu_2Ge_2$ at $T = 20\,\text{K}$ in dependence of momentum transfer are compatible with both a Γ^7-ground state, as well as with the dipole approximation. The anisotropy due to CF effects is small. The extrapolation for $Q \rightarrow 0$ gives by definition the total magnetic moment at the Ce ion. The induced moment by the applied field of 8 T amounts to $0.29\mu_B$, which is consistent with high-field magnetization measurements on polycrystalline samples [3].

Almost the same results hold true for $CeCu_{1.9}Ni_{0.1}Ge_2$ (see Fig. 1). Despite a somewhat larger scattering of the data points, the dipole approximation again supplies a sufficiently appropriate description of the Q-dependence of the magnetic structure factors. Inelastic neutron scattering experiments revealed very similar CF level schemes and 4f-wave functions for pure and 10% doped $CeCu_2Ge_2$ [5]. The present measurements corroborate these results, showing that alloying $CeCu_2Ge_2$ with 5% Ni has virtually no effect on the shape of the 4f-wave function. Subsequently, the magnetic structure factors of $CeCu_2Ge_2$ have been measured at $T = 2\,K$ (the limited beam time did not allow to measure a sufficient number of structure factors for $CeCu_{1.9}Ni_{0.1}Ge_2$ at $T = 2\,K$ in order to establish its Q-dependence). Almost no difference could be observed as compared to the paramagnetic state. This shows that the internal magnetic field due to the onset of long-range magnetic order does not have a significant influence onto the 4f-electron wave function. The induced moment now reaches $0.4\mu_B$. Having measured the magnetic structure factors, the reconstruction of the magnetization density have been performed employing the maximum entropy method [6]. No magnetic contribution has been found at the transition metal site. Entering the magnetically ordered state of $CeCu_2Ge_2$ results in an increase of the magnetization density at the Ce ions, consistent with an increase of the induced magnetic moment from 0.29 to $0.4\mu_B$.

$CeCu_2Ge_2$ orders in an incommensurate modulated magnetic structure characterized by a propagation vector $\mathbf{k} = (0.284, 0.284, 0.534)$, as determined by powder and single crystal neutron diffraction [1,3]. For $CeCu_{1.9}Ni_{0.1}Ge_2$ two distinct magnetic phase transitions occur with, however, very similar magnetic structures like in pure $CeCu_2Ge_2$ differing only slightly in the z-component of the propagation vector [1]. The elevated counter technique of the DN2 diffractometer allowed us to monitor several magnetic Bragg reflections, thus investigating the influence of the vertically applied magnetic field of 8 T along the crystallographic b-axis onto the magnetic structure. Surprisingly, the external field did not seem to have any influence at all. The additional magnetic reflections all appeared at the same positions as expected from the well-known zero-field structure. The principal magnetic reflections $(0\,0\,0)^-$ of $CeCu_2Ge_2$ and $CeCu_{1.9}Ni_{0.1}Ge_2$ are shown in Fig. 2. For $CeCu_{1.9}Ni_{0.1}Ge_2$, the occurrence of two neighbouring satellites corresponds to the two different magnetic phases.

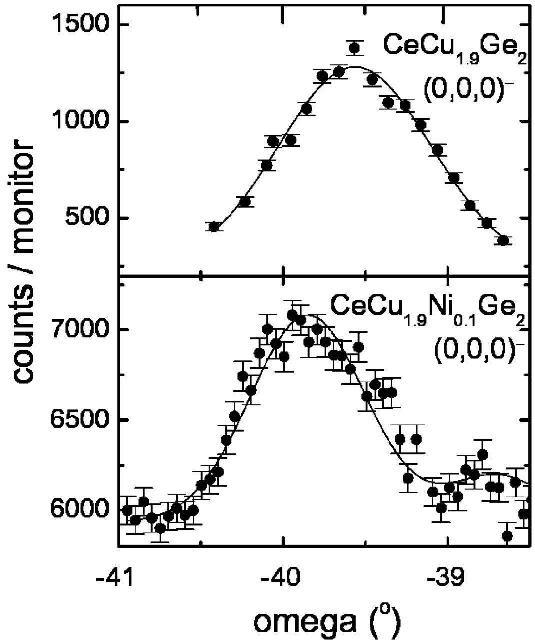

Fig. 2. Principal magnetic reflections $(0\,0\,0)^-$ of $CeCu_2Ge_2$ and $CeCu_{1.9}Ni_{0.1}Ge_2$ at $T = 2\,K$.

The present polarized neutron diffraction study therefore reveals an extraordinary stability of the magnetic moments of the heavy fermion compound $CeCu_2Ge_2$ against (i) a change of temperature from 2 to 20 K, thus passing through the magnetic ordering temperature, as well as through the Kondo lattice temperature $T^* = 8\,K$ [3] (ii) a change of an external magnetic field of 8 T, thus exceeding the magnetic correlation energy k_BT_N twice (iii) alloying with 5% Ni with a concomitant atomic disorder. This stability is not clear at present, in particular, since inelastic neutron scattering experiments demonstrated a strongly dynamic character of the magnetism of $CeCu_2Ge_2$ [3]. This is evidenced by the fact that the magnetic moment as deduced by integrating the quasielastic scattering intensities exceeds the value of the ordered magnetic moment for both, pure and Ni-doped $CeCu_2Ge_2$. Neutron diffraction studies on $CeCu_2Ge_2$ in high magnetic fields are necessary to establish its complete $B-T$ phase diagram as the next step in order to deeper understand its magnetic properties.

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