Experimental report

03/03/2016

Proposal: **4-03-1707** **Council:** 10/2014

Title: Probing the crystal-field potential in CeFe2Al10 using substituted Ndand Pr ions

Research area: Physics

This proposal is a new proposal

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Samples: Ce0.85Nd0.15Fe2Al10
Ce0.95Nd0.05Fe2Al10
LaFe2Al10
Ce0.85Pr0.15Fe2Al10
Ce0.85Pr0.05Fe2Al10
La0.85Nd0.15Fe2Al10

Instrument	Requested days	Allocated days	From	To
IN4	7	6	26/06/2015	02/07/2015

Abstract:

The compounds from the CeT2Al10 family (T: Ru, Os, Fe) exhibit Kondo-insulator properties, with an unusual competition of strongly anisotropic crystal-field and exchange interactions, as well as c-f hybridization. We propose to probe the crystal-field potential in CeFe2Al10 by using Nd and Pr impurities, which possess a stable 4f shell, as a local sensor at the rare-earth site in inelastic neutron scattering experiments.

4-03-1707 - Probing the crystal-field potential in $CeFe_2Al_{10}$ using substituted Nd and Pr ions

The CeT_2Al_{10} family (T: Ru, Os, Fe) of orthorhombic (*Cmcm*, no 63) compounds exhibits a very unusual combination of transport, magnetic, and spectroscopic properties [1-4], which challenges our present understanding of Kondo insulators. In particular, for T = Ru and Os, long-range antiferromagnetic (AF) order, with strongly reduced Ce magnetic moments ($m_{AF} = 0.34\ \mu_B$), was found to set in at a surprisingly high Néel temperature $T_N \sim 30$ K, almost twice that of the isostructural compound $GdRu_2Al_{10}$, which has a much (more than 20 times) larger magnetic moment. In $CeFe_2Al_{10}$, on the other hand, no magnetic order was observed, but a pronounced Kondo-insulator character exists. In all three compounds, the formation of a spin gap [2, 3] at low temperatures has been reported from inelastic neutron scattering (INS) measurements, but its origin (anisotropy gap of magnonlike excitations, hybridization gap, or some more complex mechanism involving anisotropic hybridization) remains unknown. The interaction of the Ce *f*-electrons with the local crystal field (CF) potential is one of the key physical mechanisms for describing the ground state in CeT_2Al_{10} compounds. It is thought [1,4,5] to produce a strong single-ion magnetic anisotropy, which is dominant at high temperature, but competes with other sources of anisotropy (exchange, hybridization) at low temperature, especially in the AF ordered state of $CeRu_2Al_{10}$ and $CeOs_2Al_{10}$, where the orientation of the Ce magnetic moments (//*c*) does not correspond to the easy axis (//*a*) defined by the CF.

The idea of the present experiment was to use a "normal" magnetic rare earth ion with a stable 4*f* shell, such as Nd^{3+}, as a sensor of the CF potential in the $CeFe_2Al_{10}$ system, without the extra complexity caused, in the case of Ce, by the hybridization of *f*-electron states with the conduction band. This method was successively tested in previous studies of the CF potential in the mixed-valence compound $CeNi_5$ [7], the heavy-fermion compound $CeAl_3$ [6] and, more recently, the Kondo insulator YbB_{12} [8]. Here the use of a Nd impurity was chosen because of its rather large magnetic moment, and the reasonable number of CF-split levels (5 doublets connected by magnetic dipole transitions) in orthorhombic or lower local symmetry. This provides 8 independent pieces of experimental information whence to derive the coefficients of the CF Hamiltonian. Measurements at different temperatures will provide still additional information. Impurity-impurity interactions can be minimized by using low Nd concentrations. The order of magnitude of the overall CF splitting in $NdFe_2Al_{10}$, estimated from magnetization data to be about 24 meV [9], is well suited to thermal neutron spectroscopy. In order to ensure optimal conditions regarding statistical accuracy and counting time, a Nd concentration of 15% was selected for the experiment.

Measurements were performed on IN4C at two incoming energies E_0 of 16 and 38 meV, with energy resolutions ΔE_0 of 1.0 and 2.5 meV, respectively. This allowed us to measure the magnetic signal in a wide enough energy range with respect to the expected CF splitting of Nd^{3+}, and also to study the region of the spin gap in pure $CeFe_2Al_{10}$ with a better resolution. The higher incoming energy also provides better conditions for determining the phonon contribution because of the larger difference in momentum transfer between low and high scattering angles The compositions of the samples used in the present experiment were:

$CeFe_2Al_{10}$, $NdFe_2Al_{10}$, $Ce_{0.85}Nd_{0.15}Fe_2Al_{10}$, $La_{0.85}Nd_{0.15}Fe_2Al_{10}$, and $LaFe_2Al_{10}$. Sample containing Pr could not be measured due to the lack of experimental time. The temperatures of the samples were comprised between 2 and 80 K. The transmission coefficient depended on the thickness of the powder samples (ranging from 3 to 7 mm) but were always better than 85% at E_0 = 38 meV. The experimental spectra for E_0 = 38 meV, corrected for absorption, are presented hereafter in the form of the scattering function $S(Q, E)$, normalized to the monitor count and to the solid angle. Each data set was divided into 4 groups of scattering angles. The largest angles ($\langle\Theta\rangle = 112^{\circ}$) correspond to momentum transfers from 7 to 9 $Å^{-1}$, the lowest ones ($\langle\Theta\rangle = 18^{\circ}$) to momentum transfers from 1.4 to 2.5 $Å^{-1}$. Knowing the angle dependence is essential to properly subtract out the phonon contribution from low-angle data.

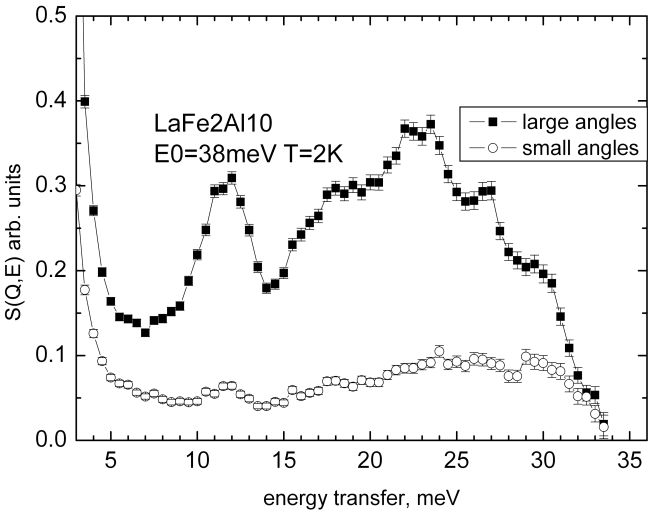

Fig. 1. *Spectra of $LaFe_2Al_{10}$ at T = 2 K measured at incident neutron energy E_0 = 38 meV, for the high-angle (closed circles: $\langle\Theta\rangle$ = 112°) and low-angle (open circles: $\langle\Theta\rangle$ = 18°) detector groups. The curves represent the energy dependence of the phonon density of states.*

In **Fig. 1**, we present the high-angle spectrum $S(Q, E)$ of $LaFe_2Al_{10}$ for E_0 = 38 meV, which provides the neutron weighed phonon density of state for these systems. This contribution was scaled to yield an estimate of the phonon background in the Ce, Nd samples measured at low scattering angles, and subtracted out. The remaining intensity is considered to result mainly from magnetic scattering, and the corresponding $S_m(Q, E)$, measured at T = 2 K and normalized per formula unit, are shown in the **Fig. 2** for the different compositions. It is seen that the magnetic intensity for the pure Ce sample is not strong and is distributed over a broad energy range up to approximately 20 meV. In the data obtained with E_0 = 16.6 meV, the phonon contribution in the accessible energy window is limited to some intensity around 12 meV. It is much weaker, for the lower-angle group of detectors ($0.9 \le Q \le 1.8$ Å^{-1}), than in the spectrum measured with E_0 = 38 meV, as expected from the much lower momentum transfer. Experimental spectra of $CeFe_2Al_{10}$ and Ce(Nd)Fe_2Al_{10} for E_0 = 16.6 meV are displayed in **Fig. 3**.

From the above, the following qualitative conclusions can be drawn. Firstly, there is clear evidence from **Fig. 2** that, for all compositions, the total CF splitting is of the order of 17 meV, which is less by almost a factor of 1.5 than expected from the susceptibility data [9]. A similar discrepancy is observed in the energies of the lower two excited states. Regarding a possible difference between the $CeFe_2Al_{10}$ (mixed-valence) and $NdFe_2Al_{10}$ (integral valence) “matrices”, the overall change in the CF potential seems to be moderate, as can be seen by comparing the spectrum for 15% of Nd^{3+} in $CeFe_2Al_{10}$ to that of pure $NdFe_2Al_{10}$. This could be expected in principle, since the lattice constants of all these compounds are very close to each other. However, a clear difference exists, in the energy and intensity of the upper CF peak, between the spectrum of $Ce_{0.85}Nd_{0.15}Fe_2Al_{10}$, on the one hand, and those of $NdFe_2Al_{10}$ and $La_{0.85}Nd_{0.15}Fe_2Al_{10}$, on the other hand (the peak positions for the latter two compounds practically coincide). This nontrivial deviation of the CF splitting of Nd^{3+} in Ce(Nd)Fe_2Al_{10} from that expected from a direct scaling suggests that the *f-sd* hybridization in Ce may significantly affect the CF potential probed by the Nd impurities.

The second point of interest is the effect on the Ce spectral response produced by the presence of Nd impurities. This effect is not obvious but could consist in the appearance of some extra intensity in the spin gap region between 2 and 10 meV (see **Fig. 3)**. A more complete analysis is in progress to trace this effect as a function of temperature up to 80 K. Let us just mention here a possible analogy with recent results on the influence of magnetic Tm impurities on the excitation spectrum of the Yb(Tm)B_{12} Kondo-insulator [8], which was previously ascribed to a "Kondo under-compensation" phenomenon.

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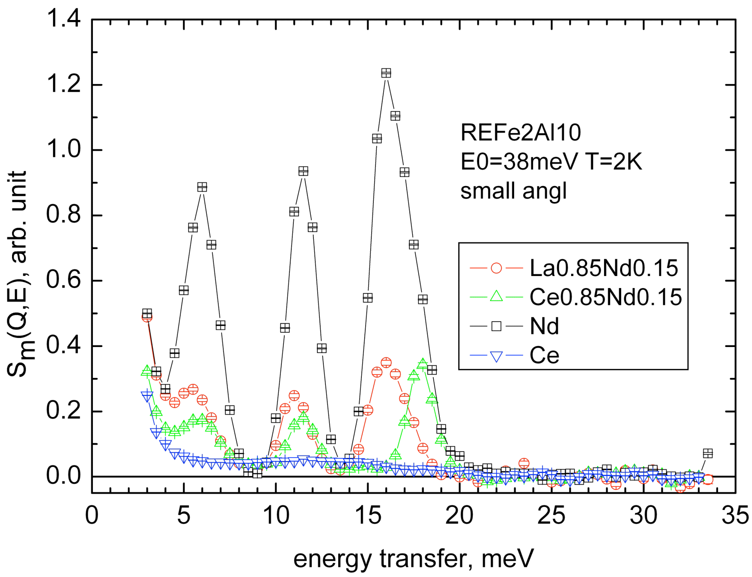

***Fig. 2**. Magnetic neutron scattering spectra (phonon subtracted, normalized to the number of RE ions) of $CeFe_2Al_{10}$, $NdFe_2Al_{10}$, $La_{0.85}Nd_{0.15}Fe_2Al_{10}$, and $Ce_{0.85}Nd_{0.15}Fe_2Al_{10}$ at T = 2 K, measured at an incoming energy E_0 = 38 meV using detectors at an average angle $\langle\Theta\rangle = 18^{\circ}$.*

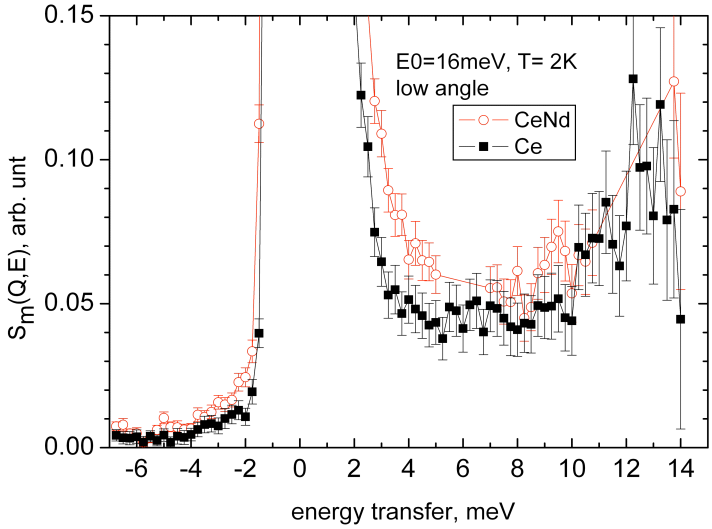

***Fig. 3.** Magnetic neutron scattering spectra of $CeFe_2Al_{10}$, and $Ce_{0.85}Nd_{0.15}Fe_2Al_{10}$ at T = 2 K, measured with a higher resolution at incident energy E_0 = 16.6 meV. CF excitation peaks from Nd have been removed.*