Experimental Report

13/06/2014

Proposal: **4-01-1316** **Council:** 10/2012

Title: Study of Crystal-Electric Field (CEF) effects on Magnetism of Ce2NiGe3 using Inelastic Neutron Scattering

This proposal is a new proposal

Researh Area: Physics

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Local Contact: ROLS Stephane

Samples: Ce2NiGe3
La2NiGe3

Instrument	Req. Days	All. Days	From	To
IN4	4	4	08/07/2013	12/07/2013

Abstract:

The Kondo lattice compound, Ce2NiGe3 exhibits spin-glass like behavior at low temperatures due to magnetic frustration and disorder arising due to random intermixing at Ni and Ge sites. The specific heat studies show CEF excitation energies of Δ1 = 135 K and Δ2 =498 K from ground state. Keeping in mind the importance of CEF levels in understanding the physical properties of cerium intermetallics exhibiting Kondo behavior, we propose to carry out inelastic neutron scattering (INS) experiments to obtain energy spectra on IN4 at three different temperatures 2, 10 and 50 K (spanning the region of interest, as observed from magnetic susceptibility data) on Ce2NiGe3 as well as its non-magnetic counterpart, La2NiGe3. The study of both magnetic (Ce2NiGe3) and non-magnetic counterparts (La2NiGe3) will help in determining the variation of the quasi-elastic spectrum, magnetic excitation spectrum (CEF energies) as well as the phonon density of states as a function of temperature.

Study of Crystal Field Effect (CEF) effects on Magnetism of Ce_2NiGe_3 using Inelastic Neutron Scattering

Objective of the experiment:

The cerium intermetallic compounds are known to exhibit anomalous properties most likely due to the larger extent of 4*f* shells in cerium and a strong spin-orbit coupling [1]. In the ground state of Ce^{3+} ion, the orbital angular moment (L = 3) is antiparallel to the spin (S = ½), resulting in the total angular momentum of J = 5/2. Crystal-electric field (CEF) acting on J, is comparable or usually larger than the Kondo singlet energy (T_k), therefore, for systems exhibiting Kondo effect, crystal field effects should also be considered [2, 3] in interpreting the magnetic properties, as CEF determines the degeneracy of the f-level [4]. The Kondo lattice compound, Ce_2NiGe_3 exhibits spin-glass like behavior at low temperatures due to magnetic frustration and disorder arising due to random intermixing at Ni and Ge sites [5]. The specific heat studies show CEF excitation energies of Δ_1 = 135 K and Δ_2 =498 K from the ground state. Keeping in mind the importance of CEF levels in understanding the physical properties of cerium intermetallics exhibiting Kondo behavior, the aim was to carry out inelastic neutron scattering (INS) experiments to obtain energy spectra on IN4 at different temperatures between 2 and 150 K (spanning the region of interest, as observed from magnetic susceptibility data) on Ce_2NiGe_3 as well as its non-magnetic counterpart, La_2NiGe_3. The study of both magnetic (Ce_2NiGe_3) and non-magnetic counterparts (La_2NiGe_3) was expected to help in determining the variation of the quasi-elastic spectrum, magnetic excitation spectrum (CEF energies) as well as the phonon density of states as a function of temperature.

Details of the experiment carried out:

Temperature dependent inelastic neutron scattering measurements were carried out on the thermal time-of-flight (TOF) neutron spectrometer, IN4, at Institut Laue Langevin (ILL) to measure the crystalline electric field (CEF) splitting scheme, so that the magnetic properties of Ce_2NiGe_3 can be properly interpreted. The following experimental conditions were used:

Incident energy choosen	E_i = 67 meV (λ_i = 1.1Å), and
	E_i = 39 meV (λ_i = 1.45Å)
	Ei = 9 meV (λ_i = 3 Å)
Q-range covered,	1.5 – 9 $Å^{-1}$
Powder samples by weight	3.5 grams each
Temperature range:	1.8 K - 150 K

Standard corrections to the data were carried out and the phonon contribution was subtracted using the non-magnetic counterpart, La_2NiGe_3.

Summary of the results obtained:

Neutron diffraction measurements carried out on polycrystalline samples of Ce_2NiGe_3 and La_2NiGe_3 establishes the crystal structure as Er_2RhSi_3 type structure having hexagonal space group $P\text{-}6_2c$ [6]. The structure can be described as two dimensional network of Ni/Ge atoms leading to the formation of hexagonal layers with rare earth atoms (*RE*1 and *RE*2) sandwiched between them (see Fig. 1).

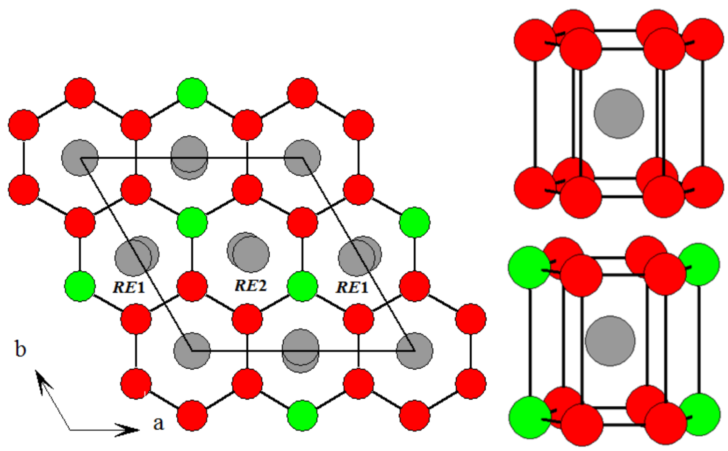

Fig.1. Crystal structure of RE_2NiGe_3 (*RE* = La, Ce) along the [001] direction and the coordination sphere for RE_1 and RE_2 are shown here (Ge is represented as solid red circles and Ni as bright green circles).

In Fig. 2 the magnetic scattering at E_i = 39 meV at 1.8 K is shown. The S(E) as a function of energy is shown in Fig. 3. The combination of Figs. 2 and 3 clearly shows the presence of CEF in the cerium compound.

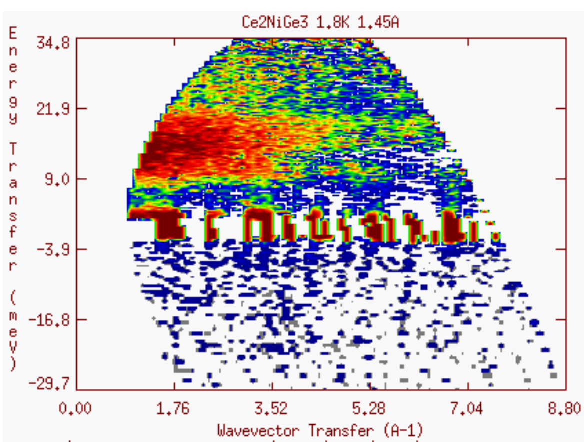

Fig. 2 Magnetic Scattering of Ce_2NiGe_3 at 1.8 K and 1.45 Å.

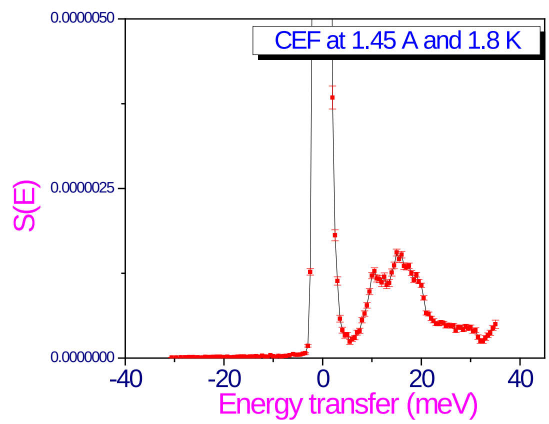

Fig. 3 CEF at 1.45 Å at 1.8 K (S(E) vs. Energy)

In Fig. 4, the plot of S(E) with energy is fitted with three Gaussians, which gives an indication of the splitting of the energy levels due to CEF. The energies at which the excitations (splittings) are seen are 10.6 meV, 15.5 meV, 19.3 meV.

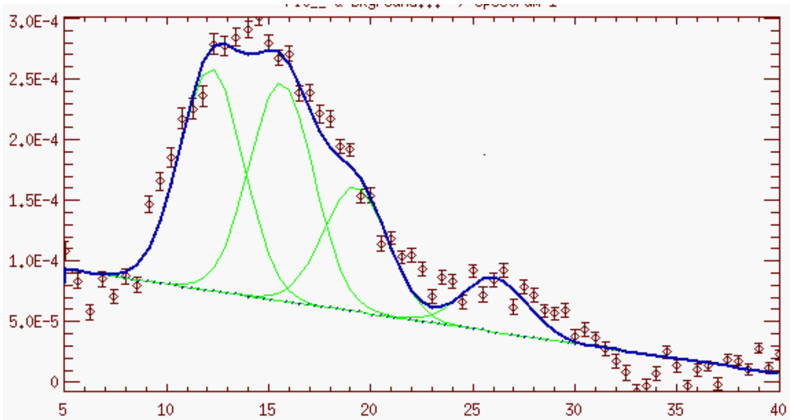

Fig.4. S(E) for Ce_2NiGe_3 after subtracting the phonon part.

The temperature dependence of the magnetic peak is plotted as a function of the energy transfer in Fig. 5. No shift was observed in the position of the magnetic peak with increasing temperature, thus giving convincing evidence for this crystalline electric field transition in Ce_2NiGe_3 to be from the ground state..

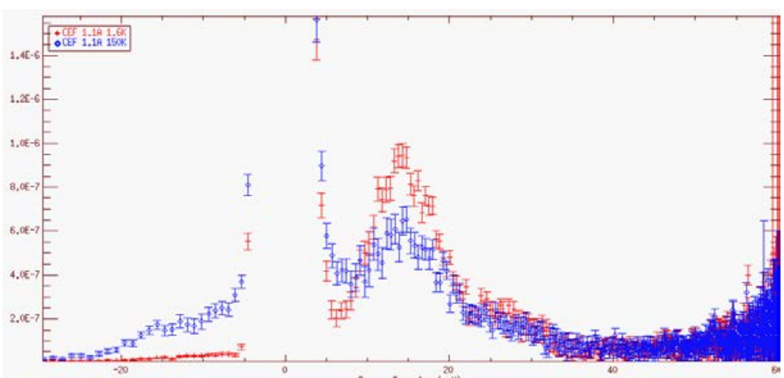

Fig.5. Temperature development of the CEF feature.

To conclude, we measured excitations of the cerium ion in Ce_2NiGe_3. Three transitions could be observed at 10.6 meV, 15.5 meV and 19.3 meV. High energy transition at 45 meV could not be observed. The initial results obtained from the INS data are quite encouraging and the detailed analysis of all the data is currently under progress which will be communicated shortly for publication.

References:

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