Experimental report

11/03/2019

Proposal: **4-04-495** **Council:** 4/2017

Title: CeCo1-xFexGe3 - inelastic excitations for x = 1 and near the Quantum Critical Point (x~0.6)

Research area: Physics

This proposal is a new proposal

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Samples: CeCoGe3
CeFeGe3
LaCoGe3, LaFeGe3, LaCo0.4Fe0.6Ge3
CeCo0.4Fe0.6Ge3
CeCu4Mn, LaCu4Mn

Instrument	Requested days	Allocated days	From	To
IN4	8	4	13/04/2018	17/04/2018

Abstract:

We have recently studied a transition between the CeCoGe3 antiferromagnetic compound (TN1 = 21 K, TN2 = 12 K, and TN3 = 8 K) and the nonmagnetic heavy fermion CeFeGe3. This series of compounds CeCo1-xFexGe3 is in a focus of our interest as for x~0.6 it exhibits a Quantum Critical Point. We have found that the crystal electric field (CEF) plays a crucial role in the properties of these alloys. The objective of the planned inelastic neutron scattering in combination with other methods (specific heat, magnetic susceptibility, thermopower) is to determine unambiguously the scheme of the CEF levels.

Experimental Report

Motivation:

Our recent results for the series of compounds $CeCo_{1-x}Fe_xGe_3$ show that it has complicated magnetic phase diagram [1]. It occurs because the transformation between the antiferromagnetic $CeCoGe_3$ and heavy fermion paramagnetic $CeFeGe_3$ is isostructural but not isoelectronic [2]. Moreover, a thorough interpretation of the magnetic, transport and thermal (heat capacity) properties requires a knowledge of the scheme of the crystal electric field (CEF) levels. Hence, the main aim of the carried out experiment was the verification of the CEF levels scheme by inelastic neutron scattering (INS) and comparison with the results of our specific heat and thermoelectric power measurements.

Experimental details:

The samples were prepared by induction melting of the stoichiometric amounts of the constituent elements under argon atmosphere and remelted several times to get better homogeneity. For the INS experiment the samples of the mass of about 6g were wrapped in aluminum foil. Additionally, Vanadium has been measured to enable the correction for background.

The inelastic neutron scattering experiments were performed on the IN4 time-of-flight instrument at the Institut Laue Langevin (ILL) in Grenoble using the incident neutrons wavelength of 1.5 Å and 3.0 Å. To extract the magnetic contribution for Ce-based compounds we have measured the La-based nonmagnetic reference compounds.

Preliminary results:

Both $CeCoGe_3$ and $CeFeGe_3$ compounds have the same non-centrosymmetric $BaNiSn_3$–type crystal structure (*I4mm* space group). $CeCoGe_3$ has three antiferromagnetic phase transitions at T_{N1} = 21 K, T_{N2} = 12 K, and T_{N3} = 8 K [3] with superconductivity found under hydrostatic pressure. $CeFeGe_3$ is a paramagnet with a high Kondo temperature (over 100 K). For $CeCo_{1-x}Fe_xGe_3$ a possibility of Quantum Critical Point (QCP) has been suggested [1,4] at x close to 0.6. To analyze the results of the magnetic measurements, specific heat, electrical resistivity and thermoelectric power one has to include the CEF effects in respective models.

Fig. 1 shows magnetic contribution to the specific heat for x=0.6 and analyzed with the Schottky contribution to the specific heat:

$$\frac{C_{\mathrm{CEF}}}{T} = \frac{R}{T^3}\left[\frac{\sum_{\mathrm{i=0}}^{\mathrm{n-1}} \Delta_{\mathrm{i}}{}^2\, \mathrm{e}^{-\frac{\Delta_{\mathrm{i}}}{\mathrm{T}}}}{\sum_{\mathrm{i=0}}^{\mathrm{n-1}} \mathrm{e}^{-\frac{\Delta_{\mathrm{i}}}{\mathrm{T}}}} - \left(\frac{\sum_{\mathrm{i=0}}^{\mathrm{n-1}} \Delta_{\mathrm{i}}\ \ \mathrm{e}^{-\frac{\Delta_{\mathrm{i}}}{\mathrm{T}}}}{\sum_{\mathrm{i=0}}^{\mathrm{n-1}} \mathrm{e}^{-\frac{\Delta_{\mathrm{i}}}{\mathrm{T}}}}\right)^2\right] \tag{1}$$

where Δ_{i} is energy distance in respect to the ground state, and with the Schotte-Schotte formula:

$$\frac{C_{\mathrm{Kondo}}}{T} = \frac{k_{\mathrm{B}} N_{\mathrm{A}} T_0}{\pi T^2}\left(1 - \frac{T_0}{2\pi T}\psi'\left(\frac{1}{2} + \frac{T_0}{2\pi T}\right)\right), \tag{2}$$

where ψ' is the derivative of the Digamma function. According to the expected split into three doublets, the excitations Δ_1 and Δ_2 were determined as equal to 81 K and 225 K, respectively. However, the fit in Fig. 1 is evidently not satisfactory. Instead of single anomaly due to the Shottky contribution an additional hump is visible.

Fig. 2 presents the INS spectrum at 50 K for the same composition, i.e. $CeCo_{0.4}Fe_{0.6}Ge_3$, where the magnetic ordering temperature is already dumped down to T = 0 K. Evidently more than the two expected CEF excitations are visible in the range 105 K-222 K, which implies that probably there are two different types of the Ce neighborhood and/or different local symmetries in some Ce positions. This type of disorder may be a source of the additional excitations and has to be included in our further analysis of the CEF contribution to the magnetic specific heat and other measurements. For the parent $CeCoGe_3$ compound our results are in good agreement with the previously published [5].

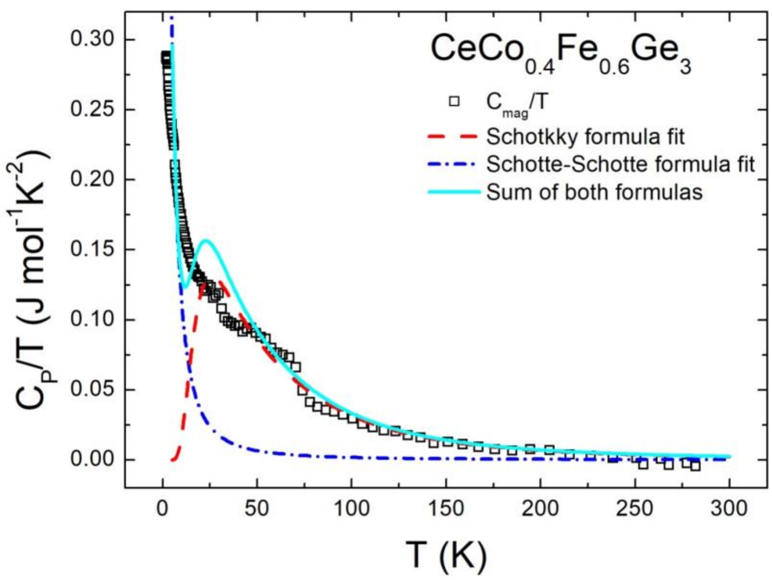

Fig. 1. Magnetic contribution to the specific heat fitted with a sum of Schottky and Schotte-Schotte formulas for sample x = 0.6.

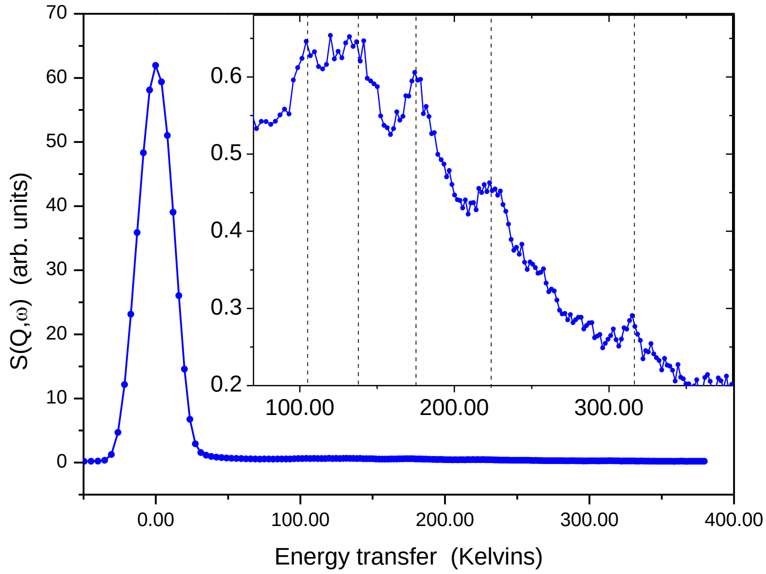

Fig. 2. Neutron time-of-flight spectrum for $CeCo_{0.4}Fe_{0.6}Ge_3$ at 50 K for the wave-length of 1.5 Å. Energy scale is recalculated from meV to Kelvins to facilitate comparison with the specific heat results for CEF.

References

[1] P. Skokowski, K. Synoradzki, T. Toliński, Complex magnetism and non-Fermi liquid state in the vicinity of the quantum critical point in the $CeCo_{\{1-x\}}Fe_xGe_3$ series, ArXiv180604656 Cond-Mat. (2018). http://arxiv.org/abs/1806.04656 (accessed March 10, 2019).

[2] P. Skokowski, K. Synoradzki, M. Werwiński, A. Bajorek, G. Chełkowska, T. Toliński, Electronic structure of $CeCo_{1-x}Fe_xGe_3$ studied by X-ray photoelectron spectroscopy and first-principles calculations, J. Alloys Compd. (2019). doi:10.1016/j.jallcom.2019.02.056.

[3] A. Thamizhavel, T. Takeuchi, T. D Matsuda, Y. Haga, K. Sugiyama, R. Settai, Y. Ōnuki, Unique Magnetic Phases in an Antiferromagnet $CeCoGe_3$, J. Phys. Soc. Jpn. 74 (2005) 1858–1864. doi:10.1143/JPSJ.74.1858.

[4] S.N. de Medeiros, S.L. Bud'ko, M.B. Fontes, M.A. Continentino, E.M. Baggio-Saitovitch, Phase diagram of $Ce(Co_{1-x}Fe_x)Ge_3$: from complex magnetic ordering to a non-magnetic Fermi liquid, J. Magn. Magn. Mater. 226–230 (2001) 152–154. doi:10.1016/S0304-8853(00)00653-3.

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