Experimental report

16/02/2020

Proposal: **4-04-482** **Council:** 4/2016

Title: Hyperfine field study on Sr2-xLaxFeCoO6 (x = 0.0, 1.0, 2.0)

Research area: Physics

This proposal is a new proposal

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Samples: Sr2FeCoO6
La2FeCoO6
LaSrFeCoO6

Instrument	Requested days	Allocated days	From	To
IN16B	4	4	28/05/2016	30/05/2016
			27/08/2016	29/08/2016

Abstract:

With the advent of high-resolution back-scattering neutron spectrometers, Heidemann and co-workers investigated the hyperfine fields in Co and V based compound. So far hyperfine interaction study by high-resolution inelastic neutron scattering has been undertaken only on long-range ordered magnetic systems. We now wish to extend this study to short-range correlated magnetic systems and also to the spin-glass and spin-ice systems. We propose to investigate hyperfine interaction on double-perovskite compounds Sr2FeCoO6 SrLaFeCoO6 and La2FeCoO6 systematically. We know beyond doubt that Sr2FeCoO6 is a spin glass with a spin freezing temperature of Tsf = 100 K. We also know that SrLaFeCoO6 although does not show long-range magnetic order, has strong ferromagnetic correlations below Tc = 75 K. We know from neutron powder diffraction experiments that La2FeCoO6 undergoes a structural and magnetic phase transition at about T = 225 K. In order to study magnetic properties through hyperfine interaction we propose to measure hyperfine interaction on double-perovskite compounds Sr2FeCoO6 SrLaFeCoO6 and La2FeCoO6.

Data from proposal 4-04-482 are published in PRB [1][2] and where classified as Editor's suggestion. Here we focus on some of the information which was contributed by our IN16B experiments.

ABSTRACT *(of the above mentioned PRB article)*

The study of hyperfine interaction by high-resolution inelastic neutron scattering is not very well known compared to the other competing techniques viz. nuclear magnetic resonance, Mössbauer, perturbed angular correlation spectroscopy, etc. Also, studies have been limited mostly to magnetically ordered systems. Here, we report such a study on Sr_{2-x} La_x $FeCoO_6$ (x = 0, 1, 2) of which the first (Sr_2 $FeCoO_6$ with x = 0) has a canonical spin-glass state, the second ($SrLaFeCoO_6$ with x = 1) has a so-called magnetic glass state, and the third (La_2FeCoO_6 with x = 2) has a magnetically ordered ground state. Our present study revealed a clear inelastic signal for $SrLaFeCoO_6$, a possible inelastic signal for Sr_2FeCoO_6 below the spin freezing temperatures T_{sf}, but no inelastic signal at all for the magnetically ordered La_2FeCoO_6 in the neutron-scattering spectra. The broadened inelastic signals observed suggest hyperfine field distributions in the two disordered magnetic glassy systems, whereas the absent inelastic signal for the third compound suggests no, or a very small, hyperfine field at the Co nucleus due to Co electronic moment. The hyperfine splitting on the Co nucleus is induced by the electronic spin state of the magnetic sample atom, and our experiments add information concerning the timescale of electronic spin fluctuations by the appearance of quasielastic broadening in the μeV range at low Q and spin freezing on the nanosecond timescale below T_{sf}. Whereas these features are observed at low Q for x = 0 and 1, they are absent for La_2FeCoO_6, which evidences a gradual increase of the elastic intensity only at large Q near an emerging Bragg peak. Thus both electronic magnetic spin freezing and inelastic excitations arising from nuclear hyperfine splitting at the Co site consistently indicate a different behavior for x = 2.

EXPERIMENTAL

Powder samples of Sr2FeCoO6, SrLaFeCoO6, and La2FeCoO6 were prepared by a sol-gel method [3] and about 3 g of material were placed in Al sample holders, which were placed in a cryofurnace.
High-resolution inelastic-neutron-scattering experiments were carried out on SPHERES, operated by Jülich Center for Neutron Science at the MLZ in Garching, Germany, and IN16B at the Institut Laue-Langevin, Grenoble in their standard configurations with Si 111 backscattering crystals, a momentum transfer (Q) range between $0.2 < Q < 1.9$ $Å^{-1}$ and the maximum energy transfer near 30 μeV. For most measurements, the energy range was deliberately restricted to the range where the hfs is expected which optimized the count rate.

On IN16B we employed different instrument configurations, i) the standard configuration ('deformed' Si 111) which gives the highest flux, ii) the 'high signal-to-noise ratio' mode (HSNR) which uses the same crystals but only every second neutron pulse10 and iii) the higher energy resolution configuration. The SrLaFeCoO6 sample was measured in both the standard high flux configuration (FWHM ≈ 0.75 μeV) and in the HSNR mode (same resolution, lower flux and nearly a factor 10 better signal-to-noise ratio) at a few temperatures. Sr2FeCoO6 and at two temperatures also La2FeCoO6 and SrLaFeCoO6 were measured with high resolution 'polished Si 111 setup. For the latter a 'polished'

monochromator and 'polished' analysers were used in the low Q-range below 1.06 Å^{-1} which results in an energy resolution of FWHM ≈ 0.31 μeV . In this experimental setup we have completed the analysers at large scattering angles with strained Si 111 crystal analysers (Q >~ 1.06 Å^{-1}) for which a resolution of FWHM ≈ 0.6 μeV is obtained due to unmatched monochromators and analysers. On IN16B both full spectra and 'elastic fixed window scans' (efws) were carried out. In efws only neutrons scattered without energy change are counted as function of temperature.

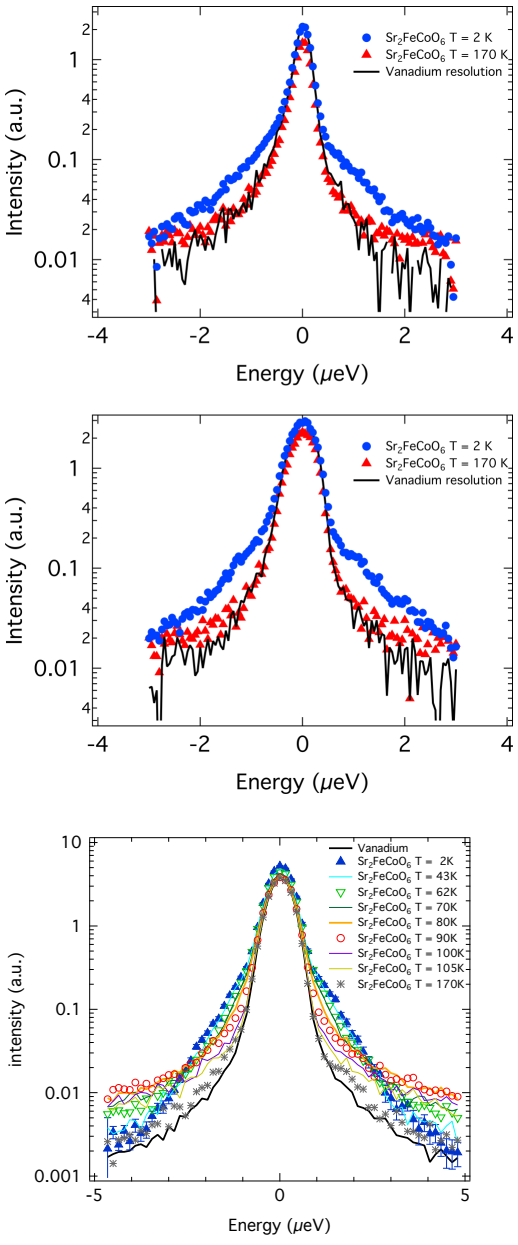

Fig1.: Spectra of Sr2FeCoO6 measured on IN16B with higher energy resolution: (a) top spectra summed in the lower Q range and resolution of FWHM ≈ 0.31 μeV and (b) bottom: at higher Q values with the "unpolished" analyzers (see text) and a resolution of FWHM ≈ 0.6 μeV. At T = 2 K one sees clearly the signal from hyperfine splitting, whereas at T = 170 K the linewidth is identical with the measured Vanadium resolution, which is scaled to the elastic peak intensity of the sample at 2 K

A few selected examples are shown in Fig.1 to Fig. 3. Fig1 illustrates the mixed analyser setup with 'polished' and 'unpolished' analysers and a 'polished' monochromator. Clearly inelastic excitations can be seen which were not resolved with the lower resolution of SPHERES.

Fig.2 show how the enhanced signal-to-noise ration of IN16B (HSNR mode) [4] allowed to evidence the presence of hyperfine splitting. Finally Fig.3a-c show that a careful investigation of the elastic scans as a function of temperature and Q can provide additional information concerning the electronic magnetism in these systems.

More detailed information can be found in [1] and [2].

Fig2.: Temperature dependence of the Sr2FeCoO6 spectra between 2 K and 170 K compared to the Vanadium resolution (summed from Q = 0.65 Å–1 to 1.9 Å–1). Due to a factor 10 improved signal-to-noise ratio of the IN16B HSNR mode and the energy range extending to 5 μeV the presence of additional quasielastic scattering between T = 62 K and 170 K becomes clear.

[1] T. Chatterji, B. Frick, M. Zamponi, M. Appel, H.S. Nair, R. Pradheesh, G.R. Hariprya, V. Sankaranarayanan, and K. Sethupathi, Phys Rev B 98, 094429 (2018). [2] Supplemental Material at http://link.aps.org/supplemental/ 10.1103/PhysRevB.98.094429.

[3] R. Pradheesh, H. S. Nair, C. M. N. Kumar, J. Lamsal, R. Nirmala, P. N. Santhosh, W. B. Yelon, S. K. Malik, V. Sankaranarayanan, and K. Sethupathi, J. Appl. Phys. 111, 053905 (2012).

[4] M. Appel and B. Frick, Rev. Sci. Instrum. 88, 036105 (2017).

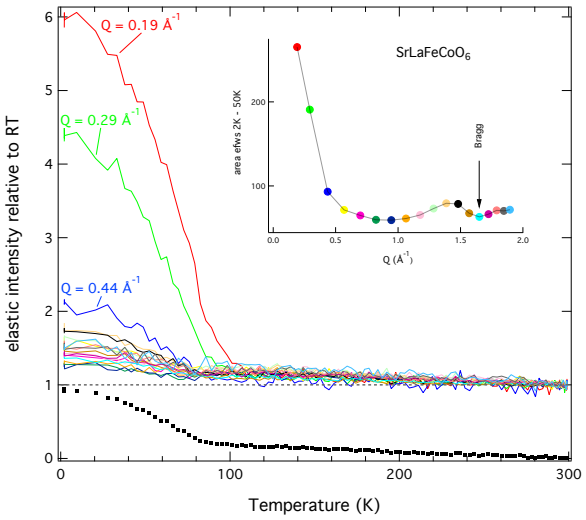

Fig.3a.: Temperature-dependence of the elastic intensity of SrLaFeCoO6 normalized to its high-temperature average value, measured on the backscattering spectrometer IN16B. Main panel, bottom (black squares): sum over all Q-values; for clarity offset by –1. Lines in the main panel: elastic intensity for different Q values, with the three lowest Q-values labeled. A strong increase of the elastic intensity is observed at low Q for temperatures below T = 80 K. The inset displays the Q dependence of the integrated intensity ("area") between T = 2 and 50 K for these curves, where the colors of the symbols in the inset correspond to the line colors of the curves in the main panel. Besides the low temperature intensity increase at low Q, another small relative intensity increase of an elastic diffuse contribution can be observed below the high-temperature Bragg peak position (indicated by an arrow) at Q = 1.57 Å–1. Note that the intensities are scaled to T = 300 K.

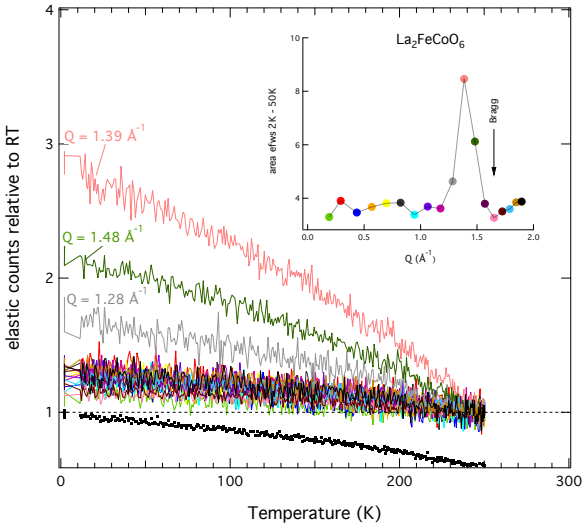

Fig.3b: Temperature-dependence of the elastic intensity of Sr2FeCoO6 normalized to its high temperature average value for all Q values measured. Main panel, bottom (black squares): sum over all Q values; for clarity offset by –0.57. Lines in main panel: elastic intensity for different Q values. A strong increase of the elastic intensity is observed below T = 80 K. Inset: The Q dependence of the area under the efws curves between T = 2 and 50 K, where the colors of the symbols in the inset correspond to the line colors of the curves in the main panel. Besides the low temperature intensity increase at low Q, another small relative intensity increase of an elas- tic diffuse contribution can be observed below the high temperature Bragg peak position (indicated by an arrow) at Q = 1.65 Å–1. Note that the intensities are scaled to T = 300 K.

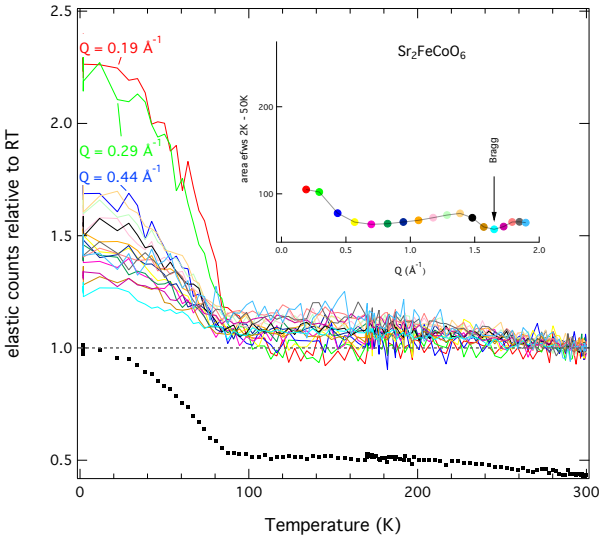

Fig.3c: Temperature-dependence of the elastic intensity of La2FeCoO6 normalized to its high temperature average value. Main panel, bottom (black squares): sum over all Q values; for clarity offset by –0.6. Lines in the main panel: elastic intensity for different Q values, with the three Q values of the low-temperature Bragg peak labeled. Besides, at the additional Bragg peak the elastic intensity increases only weakly with decreasing temperature. Inset: The Q dependence of the area under the efws curves between T = 2 and 50 K, where the colors of the symbols in the inset correspond to the line colors of the curves in the main panel. No additional increase at low Q is observed, only the appearance of an additional Bragg peak (011) at Q ≈ 1.4 Å–1 below the high-temperature Bragg peak position marked with an arrow at Q = 1.61 Å–1. Note that the intensities are scaled to T = 250 K.