Experimental report

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Proposal: 4-02-465 **Council:** 4/2015

Title: Spin resonance mode in optimally F-doped NdFeAsO

Research area: Physics

This proposal is a new proposal

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Samples: F-doped NdFeAsO

Instrument	Requested days	Allocated days	From	To
IN4	4	4	10/09/2015	14/09/2015

Abstract:

In iron pnictide superconductors there is well known empirical scaling law which relates the superconducting critical temperature with the spin resonance energy. Although the members of the 1111-family display higher Tcs than the other families the scaling law is predominantly deduced from those compounds. Here we propose to study by time-of-flight measurement the spin resonance energy of optimally F-doped NdFeAsO with a Tc = 49K.

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In iron-based superconductors (FeSC) the investigation of the spin resonance mode, which develops in the superconducting (SC) state, provided important information concerning the pairing mechanism in this class of materials. Furthermore, there is an empirical linear relation between the SC transition temperature T_C and the resonance energy $E_{res} \approx 4.6 k_B T_C$. So far the focus has been primary on the tertiary compounds, i.e. belonging to the "122" family, and binary compounds, i.e. belonging to the "11" family. For quaternary systems, i.e. the "1111" family, the investigation by neutron scattering is hampered, because large single crystals are not available and these compound contain a rare earth element, which in most cases possesses a huge neutron absorption cross section. However, the latter is not the case for Nd and there are several neutron scattering experiments reporting on the crystal electric field (CEF) excitations in $NdFeAsO_{1-x}F_x$ [1-3].

For this experiment we synthesised 10g of polycrystalline $NdFeAsO_{1-x}F_x$ ($x_{EDX} = 0.13$) with a TC of 49K, which would correspond, according to the scaling ratio, to a resonance energy at ≈20meV. To access this region in (Q,ω)-space we set the incident wavelength to 1.11Å ($E_i \approx$ 66meV) and measured the time-of-flight (TOF) spectra at 10K, 38K and 53K. All TOF-spectra were analysed with the ILL LAMP software, which includes a normalisation of each detector, subtraction of a linear background and correction for k_f/k_i. The Bose corrected spectra at 10K and 53K are displayed in figure 1 and show several CEF excitation lines. To analyse these spectra further we laid energy cuts at $Q=2.478Å^{-1}$ and fitted the CEF excitations basing on the reported CEF schemes in ref [2,3], c.f. figure 2 (a,b). Note that the three low energy CEF levels reported by Goremychkin *et al.* [2] cannot be resolved unambiguously within our resolution. Furthermore, the generalised phonon density of states (gdos) reported by Xiao et al., [1] is also plotted, figure 2 (a), for a qualitative comparison of CEF excitations and the phonon contribution. In particular the excitations at ~24meV and ~30meV seem to display a strong phononic contribution. However, the resonance mode, presumably located at 20meV, is superimposed by a CEF level, as reported from the undoped [1] and doped [2,3] compounds. In order to separate the spin resonance contribution to the signal we looked at the difference spectra of 10K and 53K, c.f. figure 1 (c). The remaining signal could indicate the gain of spectral weight as the resonance mode develops below T_C. On the other side the CEF excitations obey Boltzmann statistics and it is therefore expected that the intensity of some CEF signal decreases with increasing temperature, thus there would be signal in the difference spectra as well. In order to resolve this issue we analysed the Q-dependence of the difference spectra, c.f. figure 2 (d). The CEF excitations must follow the Nd^{3+} magnetic form factor dependence, while the spin resonance mode would follow the Fe^{2+} form factor. The data in figure 2 (d) can be described comparably well by the Nd^{3+} form factor, as well as, a weighted sum of the Nd^{3+} and Fe^{2+} form factors. However, the weight of the Fe^{2+} form factor at 20meV is the same as at other energies where the resonance mode is not expected, i.e. at 8meV, 14.6meV and 41meV. Note that the error bar of the weight is at least 50%. Since this does not allow us a conclusion about the resonance mode in more detail, the final step is to investigate the temperature dependence. For a CEF excitation the temperature dependence of the intensity is gradually, while for the resonance mode it should be an order parameter like behaviour. However, we could take spectra only at three different temperatures and thus cannot establish a proper temperature dependence, c.f. figure 2 (e). This would be a task for a future experiment.

Figure 2 (f) summarises the obtained CEF excitations which are in agreement with previous reports [2,3]. With increasing temperature an additional mode at ~11.9meV develops, c.f. figure 2 (b), (e). This mode can be identified as a transition between two excited CEF levels, as with increasing temperature more CEF levels become populated and thus transitions between them are more probable. The CEF level at ~36meV may originate from an unaffected electrostatic

crystal field by fluorine doping. As pointed out by Goremychkin et al. [2] 57% of the Nd CEFs are unaffected by 15% of F doping and therefore their CEF excitation scheme is unaltered. Finally, we would like to mention that the IN4 spectrometer performed well during the experiment.

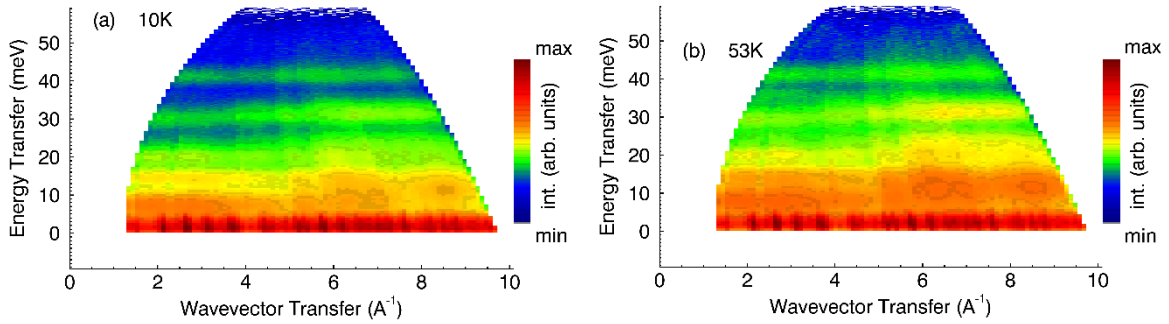

Figure 1 Bose-corrected TOF spectra at 10 K (a) and at 53 K (b), obtained by using an incident neutron wavelength of 1.11Å. For clarity a logarithmic intensity scale is applied.

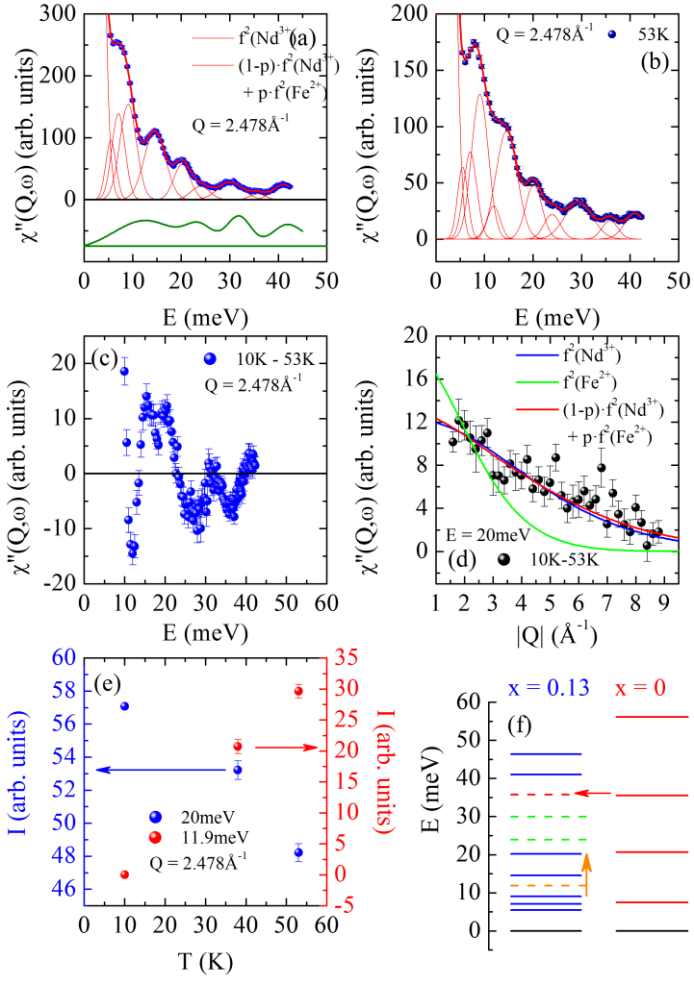

Figure 2 (a) Energy cut at Q=2.478Å^{-1} at 10K. The fit of the spectra is based on the reported CEF excitations in ref [2,3]. Note that the reported three low-energy excitations cannot be resolved within our resolution. Moreover, the general phonon density of states (gdos) reported by Xiao et al. [1] is inserted to qualitatively estimate the phononic contribution. Especially the excitations at ~24meV and at ~30meV seem to contain a strong phononic contribution. (b) The same as in (a) but at 53K. For the fit, the amplitudes were allowed to vary and an additional mode at 11.9meV is inserted. This additional mode could correspond to transition between two excited CEF level. (c) Difference of (a) and (b); below T_C there is an intensity gain around 20meV where the resonance mode would be expected. (d) Q cut at 20meV of the difference spectra at 10K and 53K. The solid lines are respectively the squared Nd^{3+} and Fe^{2+} magnetic form factor and a weighted sum of both form factors. While the Q-dependence of the CEF excitation should follow the Nd^{3+} form factor the spin resonance mode should follow the Fe^{2+} form factor. However, both fits describe the data equally well. (e) Temperature dependence of the 20meV and 11.9meV signal at Q=2.478Å^{-1}. The data corresponds to the fitted amplitude of the modes at 10K, 53K and 38K (not shown). (f) Scheme of the energy levels obtained for NdFeAsO$_{1-x}$F$_x$ with x=0.13. The CEF levels for x=0 are taken from ref. 1 and are shown for comparison. The dashed lines correspond either to a transition between two excited CEF levels (orange), have a strong phononic contribution (green), or to an unaffected CEF level, because 15% fluorine doping leaves 57% of the Nd CEF environment unaffected (red) [2].

[1] Y. Xiao et al., PRB **88**, 214419 (2013)
[2] E. A. Goremychkin et al., PRB 83, 212505 (2011)
[3] P. Cheng et al., Adv. Cond. Matter Phys. vol. 2015, Article ID 605160, 5 pages, 2015. doi:10.1155/2015/605160