Experimental report

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Proposal: **4-02-576** **Council:** 10/2019

Title: Superconductivity induced gain of exchange energy in optimum Co-dopedBaFe2As2

Research area: Physics

This proposal is a new proposal

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Samples: $Ba(Fe_{0.94}Co_{0.06})_2As_2$

Instrument	Requested days	Allocated days	From	To
IN20	7	7	12/08/2020	24/08/2020

Abstract:

We propose to study the change in exchange energy – or equivalently the change in fluctuating momen – for an optimum electron doped Fe-based superconductor, $Ba(Fe_{0.94}Co_{0.06})_2As_2$. So far rough estimates were obtained for hole doped Fe-based superconductors, but a precise determination should take the anisotropies, the L-dispersion and the change in Q widths into account. $Ba(Fe_{0.94}Co_{0.06})_2As_2$ is best studied due its high Tc, good knowledge from previous experiments and due to the availability of a large crystal.

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Superconductivity induced gain of exchange energy in optimum Co-doped$BaFe_2As_2$

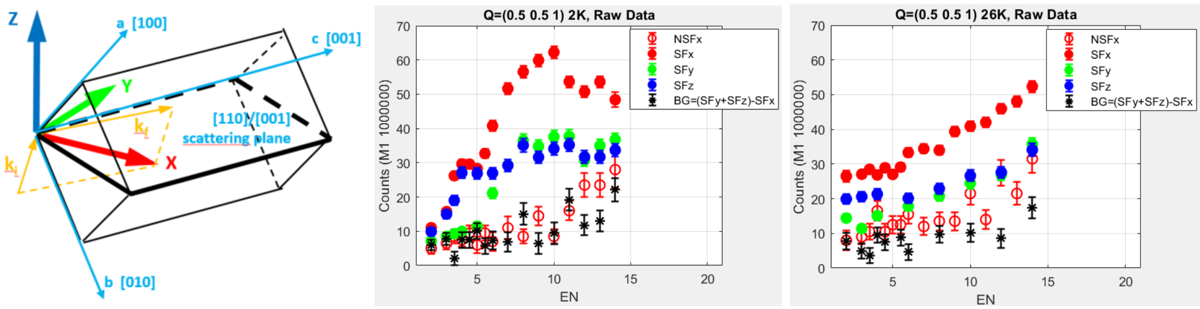
The emergence of the spin-resonance modes (SRM) in the superconducting (SC) states of unconventional superconductors is taken as one of the main arguments in favor of magnetically mediated pairing [1]. In Fe-based SCs inelastic neutron scattering (INS) experiments reveal a complex character of the SRMs in almost every studied compound; for a recent summary see [2,4]. For optimum 6% Co-doped $BaFe_2As_2$ there is a splitting in an anisotropic lower-energy SRM with some three-dimensional character (as its amplitude sharply depends on the l-component of the scattering vector) and a broader SRM at higher energies, E, that is isotropic [2,3]. Similar observations were reported for Co-doped NaFeAs, for K-doped Ba Fe_2As_2 as well as for Ni- and P-doped Ba Fe_2As_2, see [2-5] and references, so that split and anisotropic SRMs must be considered as the ubiquitous response. The anisotropy of SRMs must be attributed to spin-orbit coupling, which also manifests itself in the reorientation of magnetic structure in hole underdoped Ba Fe_2As_2 [6].

Some understanding of the anisotropic and split SRMs in doped $BaFe_2As_2$ has been obtained by full treatment of spin-orbit coupling in the electronic structure [7]. Scherer et al. qualitatively reproduce the main findings of the anisotropic SRMs, such as the primarily c-orientation, splitting of the modes and enhancement of anisotropy in the SC state [7], but the agreement is not quantitative at all. For example, they also find strong anisotropy for the high-energy SRMs, which are nearly isotropic in all systems studied. Furthermore, the relation between the anisotropic SRMs and the coexisting antiferromagnetic ordering has not been addressed, but our most recent experiments on Na-doped Ba Fe_2As_2 demonstrate a very close relation [4].

The SRMs represent a clear enhancement of spectral weight in the SC state and thus a gain of magnetic exchange energy, Eexch [8]. In the Heisenberg model E_{exch} corresponds to $Eexch=\Sigma J_{ij}<S_iS_j>$ summing over pairs of spins interacting through J_{ij}; and the mean value $<S_iS_j>$ can be obtained from the integration of the generalized dynamic susceptibility χ''(q,E) over the Brillouin zone and energy. The gain in E_{exch}, ΔE_{exch}, has been determined for cuprates and $CeCu_2Si_2$ and it is extensively discussed in the review article by Scalapino [8]. In both cases ΔE_{exch} is about one order of magnitude larger compared to the SC condensation energy (determined by specific heat) underlining the large pairing potential of magnetic fluctuations.

For Fe-SCs the determination of ΔE_{exch} is difficult, because fluctuations are anisotropic and L-dispersive, and because they sharpen across Tc. Neglecting anisotropies and sharpening Wang et al. report ΔE_{exch} =0.7meV for 33% hole K-doped Ba Fe_2As_2 using TOF data; again ΔE_{exch} is almost an order of magnitude higher than the condensation energy. For 39% Na hole-doped $BaFe_2As_2$ we made a rough estimate with unpolarized INS data again resulting in an order of magnitude compared to the condensation energy [4]. A more accurate determination taking into account the anisotropies, the finite L-dispersion and the q-sharpening of χ''(q,E) in the SC phase seems highly desirable in particular for an electron doped material not analyzed at all so far. The theoretical understanding of SRMs in Fe-based SCs needs to be improved, see above, and the changes in fluctuating moment (in absolute units) and in E_{exch} are well-defined entities to verify any theories. The aim of the new experiment was to obtain a quantitative estimation of the ΔE_{exch} by using neutron polarization analysis. Only with the polarizations analysis it seems possible to determine the background in sufficient precision. In order to obtain a quantitative result it is necessary to study the E dependence of the magnetic correlations not only at the AFM zone centers but also at different q_L values of the scattering vector. In addition one needs information about the width of the correlations parallel to the planes.

For the determination of the gain of antiferromagnetic exchange energy ΔE_{exch} as an essential part of the superconducting condensation energy, we performed an INS experiment on the optimum electron Co-doped $Ba(Fe_{1-x}Co_x)_2As_2$ sample with x=0.06. The large sample consists of three coaligned crystals with a total mass of ~2.85g and was already used in previous INS experiments; it was mounted in the [110]/[001] scattering plane. The longitudinal polarization analysis using the CryoPAD setup was used for separating the nuclear cross-section and individual components of the magnetic cross-section. The frame of the experiment seen in Fig.1(a) consists of X-axis directed parallel to the scattering vector **Q**, Z-axis perpendicular to the scattering plane and Y-axis ranged within the scattering plane and perpendicularly to both X and Z axes. One non-spin-flip (NSFx) and three spin-flip (SFx, SFy and SFz) cross-sections were collected. As the superconducting transition temperature for this sample was determined at T_c= 24K [3], the two sets of E- and Q-scans were collected at temperatures of 2 and 26K, above and below T_c respectively. The experiment ran in remote-mode due to Corona-Virus restrictions at the ILL.



(a) (b) (c)

Figure 1. (a) Scattering geometry with reference frame for longitudinal polarization analysis. (b) and (c) Examples of polarized raw-data E-scans collected at Q=(0.5,0.5,1) at 2 and 26K, respectively.

A typical set of four cross-sections (NSFx, SFx, Sfy and SFz) collected at Q=(0.5,0.5,1) at temperature of 2K is shown in Fig.1(b). The NSFx channel measured for polarization parallel to X-axis was used for check of eventual nuclear contamination, as the nuclear scattering is always a NSF process. For all collected sets of data the NSFx intensities revel only a smooth variation without pronounced features indicating the absence of nuclear contributions in the studied (**Q**, ω) regions. Black stars in Figs.1(b,c) represent background in the SF channels, which was calculated by linear combination of the SF cross-sections. Note, that an excellent flipping ratio (FR) of 29, corresponding to high beam polarisation of 0.93, was measured at (004) Bragg peak. As a result, the FR corrections in calculations of background and magnetic susceptibilities can be omitted. The SF background resembles the signal in NSFx channel, smoothly varies with the energy transfer and is well described by a polynomial function of the 4th order. Similar data were taken at (0.5,0.5,3), (0.5,0.5,5), (0.5,0.5,1.5) and (0.5,0.5,2). We always record the SFx and NSFx data but had to restrict the other SF channels to the most important positions.

Brief inspection of the spectra shown in Fig.1(b) confirms the two-component structure of the magnetic excitations appearing in the superconducting state [2,3]. One can distinguish the anisotropic low-E spin-resonance mode (SRM) clearly seen at about 4meV and the broad isotropic high-E SRM appearing at about 9 meV.

Fig. 2 shows Q scans in in-plane direction across the maxima of magnetic fluctuations at various energies and Q_L components recorded at 2K. The same scans were also recorded at 26K so that the sharpening of magnetic excitations can be taken into account for the determination of the total change of spectral weight in the magnetic excitations. The main information of our experiment is shown in Fig. 3 comparing the data below and above the SC transition. Our measurements extend over the range where temperature changes occur and yield the difference in good statistics.

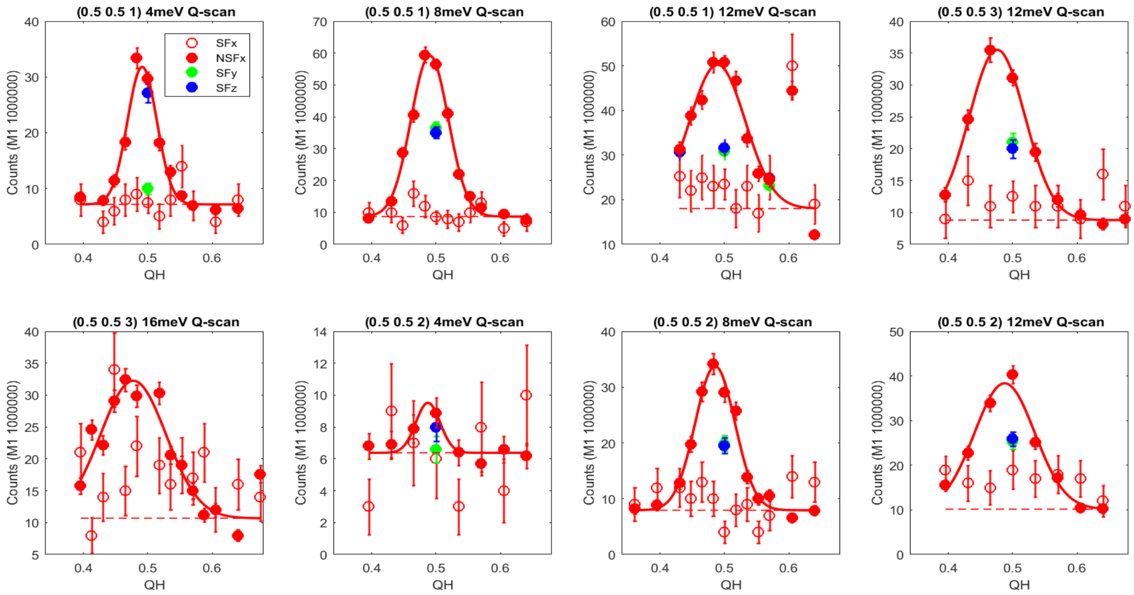


Figure 2. Q scans in in-plane direction across the maxima of magnetic fluctuations at various energies and Q_L components performed at T=2K. SFx and NSFx were always recorded by the SFy and SFz measurements were restricted to the zone centers. The same scans were also recorded at 26K.

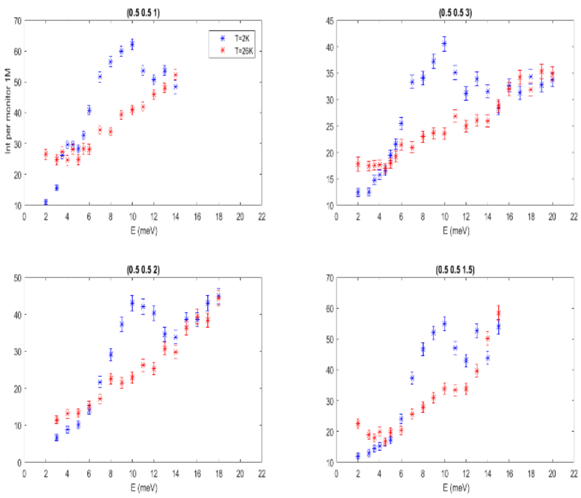


Figure 3. Comparison of the energy scans (SFx data) taken at 2 and 26K. The range of the measurements covers the energies where SC induced changes occur. And in spite of applying polarization analysis high statistics could be achieved.

In conclusion the analysis of magnetic excitations in $Ba(Fe_{0.94}Co_{0.06})_2As_2$ by polarized INS results in a fully characterization of the spectral weight as function of energy, width and q_L dependence for both temperatures. By measuring SFy and SFz at characteristic points we can model the anisotropic components to reliably determine the change in exchange energy.

[1] Hirshfeld et al. Rep. Prog. Phys. **74**, 124508 (2011). [2] F. Wasser et al., Sci. Rep. **7**, 10307 (2017) [3] P. Steffens et al., PRL **110**, 137001 (2013) [4] F. Waßer et al., PhD dissertation Cologne Univ. (2018); and npj Quantum Materials **4**, 59 (2019). [5] J. Guo et al., PRL **122**, 017001 (2019). [6] F. Waßer et al., PRB B **91**, 060505 (2015). ; ibid. physica status solidi (b) 254, 1600181 (2017); Avci et al. nature comm. **5**, 3845 (2014). [7] D. D. Scherer and B. M. Andersen arXiv1906.08566. [8] D. J. Scalapino, Rev. Mod. Phys. **84**, 1383 (2012). [9] M. Wang et al., Nat Commun. **4**, 2874 (2013).