Splitting of resonance excitations in nearly optimally doped Ba(Fe_{0.94}Co_{0.06})_2As_2: an inelastic neutron scattering study with polarization analysis

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Magnetic excitations in Ba(Fe_{0.94}Co_{0.06})_2As_2 are studied by polarized inelastic neutron scattering (INS) above and below the superconducting transition. In the superconducting state we find clear evidence for two resonance-like excitations, at a higher energy of about 8 meV there is an isotropic resonance mode with weak dispersion along the c-direction. In addition we find a lower excitation at 4 meV that appears only in the c-polarized channel and whose intensity strongly varies with the l component of the scattering vector. These resonance excitations behave remarkably similar to the gap modes in the antiferromagnetic phase of the parent compound BaFe_2As_2.

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In various FeAs-based superconductors, INS studies have shown the existence of nearly antiferromagnetic excitations, and in particular the resonance-like intensity enhancement upon entering the superconducting phase suggests a close connection between superconductivity and magnetism [1]. Since the first observations of this resonant feature in nearly optimally-doped Ba_{0.6}K_{0.4}Fe_2As_2 [2] and Ba(Fe_{1-x}Co_x)_2As_2 (x=8%) [3], this signal has been observed at different doping levels in the underdoped, optimally doped, and overdoped region [2–16]. Near optimum doping, in the paramagnetic phase and above Tc the magnetic excitations are nearly antiferromagnetic spin fluctuations with anisotropic extension in longitudinal and transverse direction. These excitations can be followed up to high energy [8, 13, 16] where they strongly resemble those in the antiferromagnetic parent compound [8, 11, 14]. In the superconducting phase, spectral weight is suppressed at low energy, and a resonant enhancement of the intensity is seen at an energy depending on the doping level (about 9 meV near optimum doping [3, 7]). Comparing different Fe-based systems the resonance energy seems to follow a linear relation with Tc [12]. In doped BaFe_2As_2 the resonance mode appears to be quite broad and there is sizeable dispersion along (0.5,0.5,L) [3, 10] with the energy minima appearing at odd L.

In 5% Ni-doped BaFe_2As_2, signs of significant magnetic anisotropy have been observed in polarized INS experiments [17]. It was concluded that the resonance mode exhibits a pure in-plane polarization at this doping level while the out-of-plane excitations would exhibit a lower energy scale but no resonance mode. In contrast, at a Ni-doping of 7.5%, the magnetic excitations appear perfectly isotropic [18]. A polarized neutron study of FeTe_{0.5}Se_{0.5} also finds more isotropic behavior with the resonance mode appearing in both channels at the same energy, just the strength of the in-plane component is slightly larger [19]. Polarized INS has also been used to study the gap of magnetic excitations in pure BaFe_2As_2 that usually is attributed to single-ion anisotropy, but in contrast to a simple easy-plane system it costs more energy to rotate the spin within the layer than rotating it perpendicular to the layer [20].

Here, we present first results of polarized INS experiments on a 6% Co-doped BaFe_2As_2 crystal. In the superconducting phase we find sizeable anisotropy in the magnetic response and demonstrate the existence of an additional out-of-plane polarized resonance excitation appearing at lower energy. The spectra in the supercon-
We have measured the magnetic excitations by polarized neutron scattering on the IN20 and IN14 thermal and cold triple axis spectrometers at the Institut Laue-Langevin in Grenoble. The IN20 spectrometer was equipped with a Heusler monochromator and analyzer and a graphite filter, with the final wave vector of the neutron fixed to 2.662 Å\(^{-1}\), while on IN14 we have used 1.55 Å\(^{-1}\) with a cooled Be-Filter. In both cases the sample was mounted in the Cryopad, which insures a zero magnetic field environment of the sample. The flipping ratio, determined on a nuclear Bragg reflection both in the normal and in the superconducting phase amounts to R=12, corresponding to a beam polarization of 85% (and R~25 on IN14).

We have used a standard procedure to extract the magnetic signal from the spin-flip (SF) and non-spin-flip (NSF) cross sections, with the neutron spin along the different directions parallel and perpendicular to the scattering vector \(\vec{Q}\), as described for instance reference \[21\]. At all points, we have measured the three cross sections \(\sigma_{NSF}^{x,y,z}\), as well as \(\sigma_{SF}^x\). At selected positions we have also measured \(\sigma_{NSF}^{x,y,z}\) to check for consistency, which yielded good agreement. Except a smooth decrease with scattering angle, the scattering in the \(\sigma_{NSF}^x\) channel is featureless, indicating the absence of sizeable nuclear contributions in the studied regions of \(Q,\omega\) space. The background in the SF channels can be estimated by linear combination of the SF cross sections and showed no significant features neither. It turns out that in all data sets the background can be well described by a single smooth weakly scattering-angle dependent function, so we consider the extracted magnetic signal clean and free from contaminations. In order to obtain the imaginary part of the magnetic susceptibility we have divided the scattering intensity by the thermal population factor (Bose factor enhanced by one) and corrected for higher-order contributions in the incident beam monitor.

In Figure 1 we show the results of constant-energy scans across \(Q=(0.5,0.5,0)\) in transverse direction, where we superpose the signal stemming from magnetic excitations polarized perpendicular to the ab-plane and parallel to it. The width of the signal is larger than the instrumental resolution. At an energy transfer of about 9 meV, we observe the enhancement of intensity by the resonance in the superconducting state (\(I_{2K}\approx 2.3I_{30K}\)), consistent with previous studies\[3, 13\]. Our polarized data show that the magnetic excitations at this resonance energy are fully isotropic in spin space. At lower energy transfer, however, the component along the c-axis is significantly stronger. At 6 meV, the anisotropy factor is about 2.
FIG. 3: (Color online) Energy dependence of the two components of magnetic susceptibility at $Q=(0.5,0.5,1)$ at T=2 K (a) and T=30 K (c), and their difference at 2 K (b). The c-polarized component of the susceptibility exhibits a sharp resonant mode in the superconducting phase. Panel (d) and (e) show low-energy raw data (not corrected for thermal factor) of the Y- and Z-spin-flip channel collected in a separate measurement (IN14). The Z-sf channel contains mostly magnetic excitations polarized along 001, and Y-sf along 110. Lines are guides to the eye, and the dashed line the approximated background.

and at 3 meV, where most intensity is suppressed in the in-plane channel, still a very small component along c remains.

The energy scans shown in Fig. 2 confirm this behavior. At 2 K, the magnetic response consists of an isotropic component, giving rise to the resonance peak, and of an additional feature polarized along c, which appears at half the energy of the resonance. The difference of the two components shown in Fig. 2(b) clearly illustrates the isotropic character of the high-energy mode and the c-polarized nature of the lower signal. In the normal state (T=30K), the magnetic response is rather smooth and can be well described by a single relaxor formula, $\chi''(\omega) = \frac{\chi''_{110}}{\omega^2+\Gamma^2}$, with $\Gamma=12\pm2$ meV. Above $T_c$, we find no significant magnetic anisotropy at $Q=(0.5,0.5,0)$.

We have also performed measurements at $L=1$, see Fig. 3, using the [110]/[001] scattering geometry. The intensities cannot directly be compared with those in the other geometry, as the experimental resolution changes the integration of the magnetic signal. Due to the large width of the signal in $Q$-space, though, the effect on the intensity is not very large, and checking identical $Q$-positions shows that the count rate in the second orientation (110-001) is only slightly reduced. At the energy of the resonance signal at 8 meV, the count rate is nevertheless found to be $\sim 30\%$ higher at $L=1$ indicating sizeable dependence of the resonance intensity on $L$. Previous studies have found a stronger dependence for 4% Co-doping, and almost none (besides the magnetic form factor) for 8% Co. Most remarkably, the low-energy resonance feature is again observed in the c component of the susceptibility. For $L=1$, this low energy mode exhibits an even stronger signal than the c-axis polarized component of the high-energy resonance mode. The additional sharp peak in the $\chi''$ component parallel c possesses thus a pronounced dependence on $L$. At $L=1$, there is a small anisotropy also above $T_c$ (Fig. 3c,e). A focus on the low energy part of the spectrum using the IN14 spectrometer (Fig. 3c,e) proves, however, that the normal state response is smooth, and that the low-energy signal is intrinsically linked to the superconducting state.

The polarized INS experiment shows that magnetic excitations appearing in the superconducting state of Ba(Fe$_{0.94}$Co$_{0.06}$)$_2$As$_2$ possess an intrinsic structure and consist of two distinct components: the high-energy resonance which is magnetically isotropic and an additional low-energy component that is entirely polarized in the $c$ direction. This low-energy uniaxial mode has three-dimensional character, as shown by its strong $L$ dependence, while the $L$ dependence of the resonance is weaker, underlining a more two-dimensional character. The c-polarized excitation at low energy points to a fundamental property of the superconducting state near optimum doping.

By summing up the magnetic contributions, we obtain the curves shown in Fig. 4 which correspond to the signal expected in an unpolarized INS experiment. Indeed they closely resemble the published unpolarized data near
optimum doping \[ \text{[8, 10]} \]. Because the low-energy component contributes only to one channel, it has a relatively small weight in the summed response, rendering it hard to detect without polarization analysis. However, close inspection of published unpolarized data on Co and Ni doped BaFe\textsubscript{2}As\textsubscript{2} strongly suggests that this signal is present there as well but has been overlooked due to the superposition with the isotropic response \[ \text{[7, 11, 12]} \]. The dispersion of the resonance excitation has been so far determined by fitting the total magnetic response in the superconducting phase (or the difference between superconducting and normal-conducting phases) by a single peak \[ \text{[3, 10]} \]. Due to the strong variation of the low-energy signal with \( L \) this procedure partially hides the overlooked additional intensity in a more pronounced dispersion of the resonance energy. The analysis of our polarized data for the high-energy resonance yields a weak dispersion (8.4meV at \( L=0 \) and 7.8meV at \( L=1 \)) and there is no significant dispersion of the low-energy signal.

We also may compare the polarized INS result to the Ni-doped BaFe\textsubscript{2}As\textsubscript{2} superconductors, for which polarization analysis experiments have recently been performed. Near optimum Ni-doping \[ \text{[12]} \], a pronounced anisotropy has been found, but the interpretation disagrees with our analysis. Lipscombe et al. conclude that the high-energy resonance mode appears only in the in-plane channel while the out-of-plane susceptibility would be feature-less in this energy range and an exhibit a lower energy scale \[ \text{[17]} \]. The much better statistics of our data for Ba\((\text{Fe}_{0.94}\text{Co}_{0.06})\text{As}_{2}\) clearly disproves such an interpretation in our case. We show that the high-energy resonance mode exists in both channels and that the additional low-energy resonance mode appears in the out-of-plane channel. We think this interpretation is also consistent with the statistics of the experiment on BaFe\textsubscript{1.6}Ni\textsubscript{0.4}As\textsubscript{2} \[ \text{[17]} \], but an experiment with better statistics is highly desirable. In the Ni overdoped compound the magnetic response has been found to be completely isotropic \[ \text{[18]} \] excluding the possibility of a similar low-energy mode polarized along \( c \). The fact that the anisotropy in the magnetic excitations occurs most strongly around optimal doping may suggest that it stabilizes the superconducting state.

A paramagnetic itinerant system may exhibit a strongly anisotropic susceptibility when approaching a magnetic transition, as it has been observed in pure Sr\textsubscript{2}RuO\textsubscript{4} \[ \text{[23]} \] which is close to a spin-density wave (SDW) ordering driven by Fermi surface nesting \[ \text{[24]} \]. However, in the ruthenate, the channel which condenses into the SDW state exhibits the larger amplitude and the lower characteristic energies, while here we find the opposite. In Ba\((\text{Fe}_{0.94}\text{Co}_{0.06})\text{As}_{2}\), the out-of-plane polarization, which does not correspond to the order in the parent compound \[ \text{[27]} \], exhibits the lower resonance energy. Furthermore, Sr\textsubscript{2}RuO\textsubscript{4} exhibits the anisotropy in its normal state, whereas the anisotropy in Ba\((\text{Fe}_{0.94}\text{Co}_{0.06})\text{As}_{2}\) emerges with the opening of the superconducting gap. However, the magnetic response of superconducting Ba\((\text{Fe}_{0.94}\text{Co}_{0.06})\text{As}_{2}\) is remarkably similar to that of undoped antiferromagnetic BaFe\textsubscript{2}As\textsubscript{2}.

It has been outlined by several groups that the high-energy magnetic response in the superconducting phase in the energy range of 20 to 200 meV is very similar to that in the parent compounds in or above its Néel state \[ \text{[8, 11, 14, 28, 29]} \]. It was argued that the main difference consists in the fact that modes depart from the resonance excitation in the superconductor while they start at the gap modes in the antiferromagnetic phase in BaFe\textsubscript{2}As\textsubscript{2}. Comparing the polarized study of the gap modes in BaFe\textsubscript{2}As\textsubscript{2} \[ \text{[20]} \], with the results presented here reveals even more similarity. In both cases one finds a clear splitting of in-plane and out-of-plane spin gaps with the c-polarized response appearing at lower energy. (Since magnetic moments are aligned in the plane in BaFe\textsubscript{2}As\textsubscript{2}, the observed anisotropy is opposite to the expectation of a simple local-moment picture for the pure compound.)

We state that the opening of either a superconducting or of a SDW gap results in quite similar magnetic spectra in spite of the different nature of these phases. The energy scale of the spin gaps induced is however much smaller in the case of the superconducting compound.

The anisotropy in magnetic response indicates the presence of significant spin-orbit coupling. Orbital effects and the possible existence of nematic order have been proposed to explain the in-plane anisotropy of the physical properties in particular of the pure materials \[ \text{[30–32]} \]. The difference between in-plane and out-of-plane magnetic excitations in Ba\((\text{Fe}_{0.94}\text{Co}_{0.06})\text{As}_{2}\) also results from orbital degrees of freedom, and the smaller but qualitatively similar effect compared to that in pure BaFe\textsubscript{2}As\textsubscript{2} suggests similar orbital features to be involved. The isotropic response in Ni overdoped samples indicates that such orbital effects are strongest in the vicinity of the orthorhombic antiferromagnetic phase where the highest superconducting \( T_c \)'s are observed.

In conclusion, the polarization analysis of magnetic excitations in Ba\((\text{Fe}_{0.94}\text{Co}_{0.06})\text{As}_{2}\) has shown that the resonance excitations in the superconducting phase are more complex than previously assumed. A broad signal at higher energy exhibits no sizeable magnetic anisotropy in spin space, neither at \( L=0 \) nor at \( L=1 \). However, there is an additional low-energy component which is polarized along the c-direction of the lattice, and whose scattering strength depends on the \( L \) value: it is weak for \( L=0 \) and very strong for \( L=1 \). Besides a lower energy-scale, the two resonance excitations strongly resemble the low-energy response in pure antiferromagnetic BaFe\textsubscript{2}As\textsubscript{2} which is governed by two different spin gaps for the two transverse magnetic polarizations. The magnetic excitations in undoped and in optimally doped BaFe\textsubscript{2}As\textsubscript{2} appear thus much more similar than what one might expect for superconducting and antiferromagnetic phases.
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