Local magnetic anisotropy in BaFe$_2$As$_2$: a polarized inelastic neutron scattering study

N. Qureshi,$^{1,2}$ P. Steffens,$^2$ S. Wurmehl,$^3$ S. Aswartham,$^3$ B. Büchner,$^3$ and M. Braden$^{1,*}$

$^1$II. Physikalisches Institut, Universität zu Köln, Zülpicher Strasse 77, D-50937 Köln, Germany
$^2$Institut Max von Laue-Paul Langevin, 6 rue Jules Horowitz, BP 156, 38042 Grenoble Cedex 9, France
$^3$Leibniz-Institut für Festkörper- und Werkstoffforschung (IFW) Dresden, D-01171 Dresden, Germany

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The anisotropy of the magnetic excitations in BaFe$_2$As$_2$ was studied by polarized inelastic neutron scattering which allows one to separate the components of the magnetic response. Despite the in-plane orientation of the static ordered moment we find the in-plane polarized magnons to exhibit a larger gap than the out-of-plane polarized ones indicating very strong single-ion anisotropy within the layers. It costs more energy to rotate a spin within the orthorhombic $a$-$b$ plane than rotating it perpendicular to the FeAs layers.

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The recent discovery of the iron-based oxypnictide superconductors has triggered enormous activity exploring the superconducting pairing mechanism in these materials. Density functional theory (DFT) calculations indicate that the conventional electron-phonon coupling is by far too weak in the iron pnictides to account for the observed high critical temperatures of up to $T_c = 55\,$K (Ref. 4). Instead, the phase diagrams of many FeAs-based families suggest a magnetic pairing mechanism, as superconductivity appears close to an antiferromagnetic spin-density wave (SDW) phase. Superconductivity can even coexist with the SDW order parameter which then slightly decreases upon entering the superconducting state. Furthermore, the onset of superconductivity in doped BaFe$_2$As$_2$ compounds is accompanied by the appearance of additional magnetic scattering at the SDW wave vector.

BaFe$_2$As$_2$ becomes superconducting on hole-doping on electron-doping or by the application of pressure. Pure BaFe$_2$As$_2$ exhibits a structural phase transition from the high-temperature tetragonal (14/mmm) to the low-temperature orthorhombic phase (Fmmm), see Fig. 1(a), at $T_S \sim 140\,$K (Ref. 17) accompanied by a magnetic phase transition into a collinear antiferromagnetic structure with an ordered Fe moment of $0.87\,\mu_B$ (Ref. 17). This orthorhombic antiferromagnetic phase exhibits remarkable properties. The magnetic structure shown in Fig. 1(b) breaks tetragonal symmetry, one may therefore expect some orthorhombic splitting in the crystal structure as it indeed occurs, but this splitting remains rather small. However, the signature of the orthorhombic structure in the electronic properties is much stronger than the small orthorhombic splitting in the lattice constants would suggest. Resistivity measurements on detwinned single crystals reveal pronounced anisotropy associated already with the precursors of the orthorhombic phase. The observation of a larger resistivity along the shorter axis with ferromagnetic spin coupling is further surprising. Also scanning tunnelling microscopy and optical conductivity analyses report essential in-plane anisotropy. Magnon excitations in the SDW phase have been studied by several inelastic neutron scattering experiments (unpolarized) and, again, the slope of the low energy dispersion is anisotropic. The nearest neighbor interaction along the $a$ and $b$ directions needed to model the magnon dispersion even show opposite signs. This remarkable anisotropy in magnetic exchange, however, can qualitatively be explained by DFT. The pronounced anisotropies in the electronic properties inspired a model of orbital polarization which gets support from recent ARPES experiments as well as speculations about the role of nematic properties. In this work we analyze the magnetic single-ion anisotropy which can be deduced from the gap in the magnetic excitations finding again remarkably strong in-plane anisotropy. It costs more energy to rotate the spin within the FeAs layers than aligning it perpendicular to them, in contrast to the simple expectation of an easy-plane system.

Large BaFe$_2$As$_2$ single crystals were obtained by a self-flux technique. The pre-reacted precursor materi-
als FeAs, Fe$_2$As, Co$_2$As and BaAs were mixed leading
to a Ba(Fe, Co)$_3$As$_3$ composition. This composition
was used to achieve a homogeneous melt at $T = 1463$ K.
The melt was cooled slowly under a temperature gradi-
ent in a double-wall crucible assembly to obtain large
and flux-free single crystals of BaFe$_2$As$_2$. Using this
technique, single crystals with lateral dimensions up to
$25 \times 10 \text{ mm}^2$ and thickness up to 1 mm were obtained.
Two plate-like single crystals with masses of 804 mg and
200 mg have been coaligned in order to decrease count-
ing time. The experiment has been carried out at the
thermal-beam three-axis spectrometer IN20 (ILL) using
polarizing Heusler (111) crystals as monochromator and
and flux-free single crystals of BaFe$_2$As$_2$. Using this
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thermal-beam three-axis spectrometer IN20 (ILL) using
polarizing Heusler (111) crystals as monochromator and
as analyzer. Longitudinal polarization analysis was per-
formed with a set of Helmholzt coils to guide and orient
the neutron polarization. The longitudinal polarization
analysis permits separating the nuclear cross-section
and the individual components of the magnetic cross-section
into the spin-flip (SF) and non-spin-flip (NSF) channels
by selecting the initial and the final polarization direction
of the neutrons. We use the conventional reference frame
for polarization analysis see Fig. 1(b), with the $x$ axis
parallel to the scattering vector $Q$, $y$ perpendicular to
$x$ within the scattering plane, and $z$ perpendicular to $x$
and $y$ (perpendicular to the scattering plane). The sam-
ple has been mounted in the $[100]/[001]$ scattering geometry
in orthorhombic notation, but note that the crystal is twinned
so that part of the crystal is in $[010]/[001]$ geometry and signals from both parts superpose.
Longitudinal polarization analysis adds additional se-
lection rules to the general law in neutron scattering that
only magnetic components perpendicular to the scattering
vector contribute. Only the magnetic components
parallel to the axis of polarization analysis contribute
to the SF scattering whereas the parallel components
contribute to the NSF scattering. Nuclear scattering is always a NSF process. We measured the NSF
cross-section for polarization parallel to $x$, serving as a reference for eventual contaminations, and all three SF
cross-sections keeping $k_f$ constant. Applying a correction
for the finite flipping ratio of $R = 7.4$ the magnetic cross-sections $\sigma_y$ and $\sigma_z$ (Eqs. 1 and 2) as well as the
background in the SF channel (Eq. 3) can be calculated
according to:

\begin{align}
\sigma_y &= \frac{R+1}{R-1}(I(SF_x) - I(SF_y)), \\
\sigma_z &= \frac{R+1}{R-1}(I(SF_z) - I(SF_x)), \\
BGR_{SF}(R) &= \frac{R}{R-1}[I(SF_y) + I(SF_z)] - \frac{R+1}{R-1}I(SF_x).
\end{align}

In Fig. 2 the maximum intensity of the magnetic
(1 0 1) reflection is shown as a function of temperature
for the two channels. A power law has been fitted to the
$\sigma_y$ data revealing a $T_N$ of approximately 137 K. The en-
tire elastic signal is found in $\sigma_y$ ($\sigma_z$ is essentially zero) in

FIG. 2: (Color online) Temperature dependence of the elastic magnetic scattering cross section at $Q=(1 0 1)$ revealing that
almost the entire magnetic signal stems from $\sigma_y$. The values
have been obtained by Eqs. 1 and 2. The fitting of a power
law to the $\sigma_y$ data yields a Neel temperature of 137 K.

FIG. 3: (Color online) Inelastic constant-Q scans at the mag-
netic Brillouin-zone centers $(1 0 l)$ for different $l$ values and at
$(1 0 0)$. (Blue) dots represent the magnetic scattering cross
section $\sigma_y$, while (red) squares depict $\sigma_z$. The cross sections
have been obtained by measuring the SF$_x$ and SF$_y$ channels,
respectively, and applying Eqs. 1 and 2. The solid lines are
spline-interpolated spin-wave theory calculations folded with
the experimental resolution.

perfect agreement with the well-accepted magnetic struc-
ture shown in Fig. 1 with moments parallel to the $a$ axis.

Inelastic constant-Q-scans were performed at the mag-
netic zone centers $Q=(1 0 l)$ with $l=1,3$ and 5, and
at $Q=(1 0 0)$ which is a magnetic zone boundary due
to the finite interlayer coupling (Fig. 3). The scan at
$Q=(1 0 0)$ does not indicate any magnetic scattering
in agreement with the published magnon dispersion for

\begin{align}
\sigma_y &= \frac{R+1}{R-1}(I(SF_x) - I(SF_y)), \\
\sigma_z &= \frac{R+1}{R-1}(I(SF_z) - I(SF_x)), \\
BGR_{SF}(R) &= \frac{R}{R-1}[I(SF_y) + I(SF_z)] - \frac{R+1}{R-1}I(SF_x).
\end{align}
indicating energies of the order of 100 meV for this \( q \) value. Due to the high zone-boundary energies the magnetic signals of the differently oriented twin domains do not superimpose in the scans at \( Q=(1\ 0\ l) \) with \( l=1,3 \) and 5, which are zone boundaries for the second domain orientation. The magnetic-zone-center scans sense the gap of the magnon dispersion arising from anisotropy. For a system with uniaxial anisotropy one expects a single gap for the two still degenerate transversally polarized magnons. This degeneracy, however, lifts for orthorhombic local symmetry causing an easy, a medium and a hard axis. We will see that such orthorhombic single-ion anisotropy is needed to describe magnetic excitations in BaFe\(_2\)As\(_2\). The cross sections \( \sigma_y \) and \( \sigma_z \) unambiguously show the splitting of the zone-center magnons in BaFe\(_2\)As\(_2\). The \( z \) direction of our reference system is always parallel to the \( b \) direction and therefore \( \sigma_z \) measures the transverse magnon polarized along the \( b \) direction. The \( y \) direction lies always in the \( a^*c^* \) plane but rotates with varying \( Q_l \). Since the ordered moment is parallel to \( a \), \( \sigma_y \) contains only the transverse magnon polarized along the \( c \) direction (see below for discussion of the contribution of the longitudinal magnon). Due to the geometry this signal is reduced by a factor 0.92, 0.61, and 0.42 for \( Q_l=1,3 \), and 5, respectively. The qualitative analysis of the scans at the magnon zone centers immediately shows that the in-plane magnon gap is larger than the out-of-plane one. This is remarkable in view of the Q-dependence of the magnetic scattering due to the form factor and the geometrical factor given above. The calculated values have been interpolated with a B-spline in order to obtain continuous curves which are depicted as solid lines in Fig. 3. The perfect description of the experimental data validates the assumption that the total magnetic scattering arises from the transversally polarized magnons. Using the obtained anisotropy values we determine the in-plane and the out-of-plane gap to amount to 16.4 meV and 10.1 meV, respectively.

The single-ion anisotropy parameters obtained in our experiment can be compared to the result of relativistic DFT calculations including spin-orbit coupling. The authors find that only 0.16 meV/Fe are necessary to turn the magnetic moment from the easy axis to the \( b \) direction, but that 0.20 meV/Fe are required to turn the moment perpendicular to the plane. These calculations cannot properly describe the local single-ion anisotropy in BaFe\(_2\)As\(_2\), as the sign of the anisotropy does not agree with the experimental values. DFT thus misses something in the description of the local symmetry of the SDW phase in BaFe\(_2\)As\(_2\).

In an itinerant system with non-saturated ordered moments, one may expect a longitudinal magnetic excitation, i.e. a magnon polarized along the ordered moment. This mode contributes to the \( \sigma_y \) channel, but by comparing the different \( Q \)-values studied we can fully rule out such an interpretation for the studied energy ranges. The longitudinal contribution should increase with increasing \( l \) component, but the experimental data show the opposite and are well described by the geometry factors for the transverse mode. We can thus exclude a longitudinal excitation below \( \sim 35 \) meV in agreement with DFT studies predicting them at significantly higher energies.

The temperature dependent measurement of the two magnetic cross sections at an energy transfer of 10 meV...
illustrates how the isotropic magnetic fluctuations in the paramagnetic phase transform into the anisotropic magnons below the Néel temperature (Fig. 4). There is no difference visible between the two channels at high temperature. Upon approaching the magnetic transition the intensity increases due to the softening of the magnetic response. Below the transition, the magnetic signal is almost completely suppressed in the $\sigma_z$ channel where the larger gap develops, but there is only moderate reduction in the $\sigma_y$ channel, where the energy transfer of 10 meV is just slightly below the low-temperature maximum.

In conclusion the polarized inelastic neutron scattering studies on the magnetic excitations in BaFe$_2$As$_2$ reveal a strong in-plane single-ion anisotropy which contrasts with the small structural orthorhombic distortion in this material. This pronounced local anisotropy corroborates the strong electronic signatures of the orthorhombic phase and gives further support for an important role of orbital degrees of freedom in the iron pnictides. The local anisotropy of Fe in the SDW phase apparently is insufficiently described in DFT theory even when including spin-orbit coupling.

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