High energy spin excitations in BaFe$_2$As$_2$

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We report neutron scattering measurements of cooperative spin excitations in antiferromagnetically ordered BaFe$_2$As$_2$, the parent phase of an iron pnictide superconductor. The data extend up to $\sim 100$ meV and show that the spin excitation spectrum is sharp and highly dispersive. By fitting the spectrum to a linear spin-wave model we estimate the magnon bandwidth to be in the region of 0.17 eV. The large characteristic spin fluctuation energy suggests that magnetism could play a role in the formation of the superconducting state.

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One of the greatest challenges presented by the recently discovered iron pnictide superconductors ¹ is to identify the electron pairing interaction which permits the formation of a superconducting condensate. In conventional superconductors this interaction is provided by the exchange of a phonon. For the iron pnictides, however, theoretical calculations ² indicate that the electron–phonon coupling is too weak to account for the observed high critical temperatures. Attention has therefore turned to other types of bosonic excitations which could mediate the pairing interaction.

One such candidate is spin fluctuations.³ In common with the layered cuprates, superconductivity in the pnictides is found in close proximity to parent phases which exhibit long-range antiferromagnetic order.⁴ However, unlike the cuprates, whose magnetic properties are governed by strong superexchange interactions between localized spin–$\frac{3}{2}$ moments in a single Cu $3d_{x^2−y^2}$ orbital, magnetism in the pnictides is more itinerant in character and derives from multiple d orbitals. It may also involve a degree of frustration. In magnetically ordered materials the dominant magnetic excitations are coherent spin waves. Wavevector-resolved measurements of the spin-wave spectrum by inelastic neutron scattering provide information on the fundamental magnetic interactions and can also reveal effects due to itinerancy and frustration. Such studies on the magnetically ordered parent phases of unconventional superconductors like the cuprates and iron pnictides are important to establish the characteristic energy scales of the spin fluctuations and also to provide a reference against which changes associated with superconductivity can be identified.

Here we present neutron scattering data on the collective spin excitations in antiferromagnetic BaFe$_2$As$_2$. We find that the spin excitation spectrum has a very steep dispersion within the FeAs layers with a bandwidth in the region of 0.17 eV, not much less than that in the cuprates. Such a high characteristic energy suggests that spin fluctuations are a serious candidate to mediate high temperature superconductivity in the iron pnictides.

The parent phase BaFe$_2$As$_2$ becomes superconducting on doping with holes ² or on application of pressure ². At $T_s = 140$ K, BaFe$_2$As$_2$ undergoes a structural transition from tetragonal to orthorhombic and simultaneously develops three-dimensional long-range antiferromagnetic order.⁶,⁷ On cooling through $T_s$, the space group changes from $I4/mmm$ (lattice parameters $a = 3.96$ Å, $c = 13.0$ Å) to $Fmmm$ (lattice parameters $a = 5.61$ Å, $b = 5.57$ Å, $c = 12.9$ Å). The magnetic structure of BaFe$_2$As$_2$, shown in Fig. 1, is a collinear antiferromagnet with propagation vector $\mathbf{Q}_{\text{AF}} = (1, 0, 0)$. The ordered moments on the Fe atoms are of approximate magnitude 0.9 $\mu_B$ and point along the orthorhombic $a$ axis.

Polycrystalline BaFe$_2$As$_2$ was prepared by reacting stoichiometric amounts of the elements in a tantalum ampoule sealed under argon (800°C for 2 days, then 900°C for 2 days after regrounding). Phase purity was confirmed using X-ray powder diffraction. The neutron

![Image](326x387 to 553x519)

FIG. 1: (Color online) Crystal and magnetic structure of BaFe$_2$As$_2$. On the left is the crystal structure with the conventional unit cell for the low temperature orthorhombic $Fmmm$ structure. The three-dimensional antiferromagnetic (AFM) ordering of Fe spins is indicated. On the right is a single layer of Fe spins showing the in-plane AFM order and the nearest- and next-nearest-neighbour exchange interactions.
scattering experiments were performed on the MERLIN chopper spectrometer at the ISIS Facility. Approximately 8 g of the BaFe$_2$As$_2$ powder was sealed inside a cylindrical aluminium can mounted in a top-loading closed-cycle refrigerator. Spectra were recorded at a temperature of 7 K with four different neutron incident energies: $E_i = 25$ meV, 50 meV, 200 meV and 400 meV. The scattering from a standard vanadium sample was used to normalize the spectra and to place them on an absolute intensity scale with units mb sr$^{-1}$ meV$^{-1}$ f.u.$^{-1}$, where f.u. stands for ‘formula unit’ (of BaFe$_2$As$_2$). The spectra were azimuthally-averaged and transformed onto a $(Q, E)$ grid, where $Q = |Q|$ is the magnitude of the neutron scattering vector. The presented intensity is the partial differential cross-section $d^2\sigma/d\Omega dE$ multiplied by the factor $k_i/k_f$, where $k_i$ and $k_f$ are the initial and final neutron wavevectors and $E_f$ is the final neutron energy.

Figure 2(a) illustrates the general features of the data. At low energies there is strong diffuse scattering due to the elastic peak and scattering from phonons, the latter of which increases with $Q$. The phonon signal drops off sharply above 40 meV, which is the upper limit of the vibrational density of states. Two distinct features stand out from the phonon signal. One is a narrow pillar of scattering at $Q = 1.2\AA^{-1}$, and the second is a plume of intensity centred on $Q = 2.6\AA^{-1}$. The latter extends in energy to at least 90 meV where it disappears out of the accessible region of $(Q, E)$ space. The 1.2 A$^{-1}$ feature is followed to lower energies in Fig. 2(b), which was obtained with a higher resolution configuration.

The origin of the 1.2 A$^{-1}$ and 2.6 A$^{-1}$ features is the cooperative spin wave excitations (magnons) associated with the antiferromagnetic (AFM) zone centres $(1, 0, l)$ and $(1, 2, l)$. Since the structure is layered we expect only a weak variation in the inelastic scattering with $l$. The effect of powder-averaging and resolution-folding makes the 2D magnon scattering appear at slightly higher $Q$ than the 2D AFM wavevectors $Q_{(1,0)} = 1.1\AA^{-1}$, $Q_{(1,2)} = 2.5\AA^{-1}$, as observed in Fig. 2.

Figure 3 shows examples of a series of cuts taken through the data at different energies. At the higher energies the signal is seen to broaden (right panel). This is due to dispersion of the spin waves. Below ~15 meV the magnetic signal decreases in intensity. We fitted the cuts through $Q = 1.2\AA^{-1}$ with a Gaussian line shape on a quadratic background and plot the integrated intensities of the fitted peaks at each energy in the insert to Fig. 3. The data show that the magnetic excitations are gapped, with no detectable signal below 5 meV. However, the gap is not sharp since a sharp gap would produce the resolution-broadened step in energy shown in Fig. 3. The broadening of the step could be due to dispersion of the gap in the $c$ direction and/or the existence of two or more gaps in the 5–15 meV range. In orthorhombic symmetry two gaps are expected at $Q_{\text{AF}}$ as a result of magnetic anisotropy which splits the spin waves into two non-degenerate branches with predominantly in-plane and out-of-plane character, respectively.

We now compare our data with a linear spin wave model for BaFe$_2$As$_2$ based on an effective Heisenberg spin Hamiltonian. The suitability of such a model is perhaps questionable in view of the itinerant character of the magnetism, but at the very least it will provide an estimate of the scale of the magnetic interactions. We calculated the spectrum by the same method as Yao and Carlson but extended the Hamiltonian to include terms that represent the single-ion anisotropy:

$$H = \sum_{\langle jk \rangle} J_{jk} \mathbf{S}_j \cdot \mathbf{S}_k + \sum_j \{ K_c (S^2_z)_j + K_{ab} (S^2_y - S^2_z)_j \}. \quad (1)$$

The first summation is over nearest-neighbour and next-nearest-neighbour pairs with each pair counted only once. The $J_{jk}$ are exchange parameters as defined in Fig. 1 and $K_{ab}$ and $K_c$ are in-plane and out-of-plane anisotropy constants, respectively. Diagonalisation of equation (1)
leads to two non-degenerate branches with dispersion

$$\hbar \omega_{1,2}(Q) = \sqrt{A_Q^2 - (C \pm D_Q)^2},$$  

(2)

where

$$A_Q = 2S \{ J_{1b} \cos(\frac{Q \cdot a}{2}) - 1 \} + J_{1a} + 2J_2 + J_c$$

$$+ S(3K_{ab} + K_c)$$

$$C = S(K_{ab} - K_c)$$

$$D_Q = 2S \{ J_{1a} \cos(\frac{Q \cdot a}{2}) + 2J_2 \cos(\frac{Q \cdot a}{2}) \cos(\frac{Q \cdot b}{2})$$

$$+ J_c \cos(Q \cdot c) \}.$$  

(3)

The neutron scattering cross section may be written

$$\frac{d^2 \sigma}{d\Omega dE_i} = \frac{k_i}{k_f} \left( \frac{\gamma r_0}{2} \right)^2 g^2 f^2(Q) \exp(-2W)$$

$$\times \sum_{\alpha\beta} (\delta_{\alpha\beta} - \hat{Q}_\alpha \hat{Q}_\beta) S^{\alpha\beta}(Q, \omega),$$  

(4)

where $\gamma r_0/2 = 72.8$ mb, $g$ is the $g$-factor of iron, $f(Q)$ the form factor of iron, $\exp(-2W)$ is the Debye–Waller factor which is close to unity at low temperatures, $\hat{Q}_\alpha$ is the $\alpha$ component of a unit vector in the direction of $Q$, and $S^{\alpha\beta}(Q, \omega)$ is the response function describing $\alpha\beta$ spin correlations. Only the transverse correlations ($yy$ and $zz$ for BaFe$_2$As$_2$) contribute to the linear spin wave cross section and the response functions (per BaFe$_2$As$_2$ formula unit) for magnon wave are given by

$$S^{yy}(Q, \omega) = S_{eff} \frac{A_Q - C - D_Q}{\hbar \omega(Q)} \{ n(\omega) + 1 \} \delta[\omega - \omega_1(Q)]$$

$$S^{zz}(Q, \omega) = S_{eff} \frac{A_Q + C - D_Q}{\hbar \omega(Q)} \{ n(\omega) + 1 \} \delta[\omega - \omega_2(Q)]$$

(5)

where $S_{eff}$ is the effective spin and $n(\omega)$ is the boson occupation number. In linear spin wave theory $S_{eff} = S$, but we keep them distinct here because in the analysis they are obtained essentially independently (see below).

Because our data do not extend over the full spin wave dispersion we were unable to determine $J_{1a}$, $J_{1b}$ and $J_2$ independently. However, several authors$^{17,18,19}$ have made predictions of effective Heisenberg exchange parameters from first-principles electronic structure calculations. In some cases$^{17,18,19}$ $J_2$ is predicted to exceed $J_{1a}$ and $J_{1b}$ by about a factor of two, with $J_{1a}$ and $J_{1b}$ either both ferromagnetic or both AFM (providing $J_{1a}$ and $J_{1b}$ are the same sign the spectrum at low energies is not very sensitive to which sign it is$^{15}$). Alternatively,$^{20}$ $J_{1a} \gg J_{1b}$ and $J_{1a} \approx 2J_2$. Guided by these predictions we fixed the ratios of the exchange parameters to be either (i) $J_{1a} = J_{1b} = J_2/2$, or (ii) $J_{1a} = 2J_2 = -5J_{1b}$. Since we cannot resolve more than one gap in the data we set $K_{ab} = K_c$ so that the in-plane and out-of-plane gaps are the same, and we neglected the $c$-axis coupling, which is expected to be much smaller than the in-plane coupling. The $g$-factor was set to 2.

We computed the powder-averaged spin wave spectrum convoluted with the instrumental resolution and fitted it to the experimental data near $Q = 2.6$ Å$^{-1}$ allowing only $SJ_2$ and $S_{eff}$ to vary. $S_{eff}$ is essentially determined by the absolute intensity, and $SJ_2$ by the dispersion. Data near $Q = 1.2$ Å$^{-1}$ were excluded as the peak widths are dominated by instrument resolution and consequently are insensitive to $SJ_2$. We obtained equally good fits to the data with both sets of parameter ratios. The best-fit parameters are, for case (i) $SJ_2 = 35 \pm 3$ meV and $S_{eff} = 0.28 \pm 0.04$, and for case (ii) $SJ_2 = 18 \pm 1$ meV and $S_{eff} = 0.54 \pm 0.05$. The highest resolution data reveals a gap of $7.7 \pm 0.2$ meV — Fig. 3 (insert) — which yields $SK_c = SK_{ab} = 0.042 \pm 0.004$ meV [case (i)] and $0.053 \pm 0.005$ meV [case (ii)]. For case (i) we tried different values of $J_1/J_2$, but the best-fit value of $SJ_2$ was relatively insensitive, varying from 33 meV ($J_1/J_2 = 0.25$)

FIG. 3: (Color online) The left and right panels show a series of constant-energy cuts through the magnon signals at $Q = 1.2$ Å$^{-1}$ and $Q = 2.6$ Å$^{-1}$, respectively. The data were averaged over the energy ranges indicated. Successive cuts are displaced vertically for clarity. The symbols represent different neutron incident energies: 25 meV (red circles), 50 meV (blue squares), 200 meV (green triangles), 400 meV (grey inverted triangles). In the left panel the lines are fits to Gaussian peaks on a sloping background. In the right panel the lines are constant-energy cuts (averaged over the same energy ranges as the data) through the powder-averaged spin wave spectrum calculated with parameters $SJ_2 = 2SJ_{1a} = 2SJ_{1b} = 35$ meV, $J_c = 0$, $SK_{ab} = SK_c = 0.042$ meV and $S_{eff} = 0.28$ (see Fig. 1). The insert shows the integrated intensities of the magnon scattering measured at $Q = 1.2$ Å$^{-1}$ as a function of energy. The line indicates the expected step in intensity for a clean gap folded with the experimental resolution.
to 46 meV ($J_1/J_2 = 1$). Aside from the uncertainty in $J_1/J_2$, the major error in $S_{\text{eff}}$ is the statistical error on the fit. The error in $S_{\text{eff}}$ comes from estimates of the background and instrumental resolution.

To illustrate the level of agreement, Fig. 4 shows the powder-averaged spin wave spectrum of BaFe$_2$As$_2$. The simulation covers the same $Q$ and energy range as the data in Fig. 2(a). The parameters used for the simulation are given in the caption to Fig. 3.

Our results show that spin fluctuations in the parent phase of the pnictides exist over a wide energy range extending up to ~175 meV, not much less than that found in the cuprate high temperature superconductors. Recent neutron scattering studies on single crystals of SrFe$_2$As$_2$ and CaFe$_2$As$_2$, though restricted to energies below 25 meV, also find steep magnon dispersion relations. For high $T_c$, it is natural to look for a pairing boson with a large characteristic energy. The data here show that spin fluctuations satisfy this requirement. However, if magnetism is to play a role in the superconducting state then there must also be a coupling between spin fluctuations and the electrons involved in pairing.

One piece of evidence for this is the recent observation of a resonant spin excitation in the superconducting state of Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. More generally, the itinerant character of magnetism in the pnictides has been inferred from the nesting of electron and hole Fermi surface pockets with nesting vector $Q_{\text{AF}}\approx 0.21,0.25$ and by the observed effect of AFM order on the Fermi surface as revealed for example by optical spectroscopy, angle-resolved photoemission spectroscopy, and quantum oscillations.

On the other hand, the dynamic magnetic response measured here does not show any obvious fingerprints of itinerant magnetism, such as damping due to a Stoner continuum. It will be interesting to follow the spin excitation spectrum to still higher energies where itinerant effects generally have the largest influence.

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