Block magnetic excitations in the orbitally-selective Mott insulator BaFe$_2$Se$_3$

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Broad-band inelastic neutron scattering on the two-leg ladder BaFe$_2$Se$_3$ provides a detailed understanding of magnetic excitations originating from the Fe$_4$ block ground-state. Consisting of a 45 meV wide band of quasi-1D acoustic spin-waves and three high-energy modes at 89 meV, 108 meV and 198 meV, the spin fluctuations and the static moment carry a total squared magnetic moment of 16(3) $\mu_B^2$ per Fe, indicative of orbital selectiveness for localized spins. An effective Heisenberg model accounts for the observed spectrum and provides a set of exchange interactions to understand how exotic magnetism stems from strong lattice, orbital and electronic correlations in iron chalcogenides.

Magnetism in iron-based superconductors is a complex many-body phenomenon and understanding it is now a central challenge in condensed-matter physics [1, 2]. The parent compounds of a large majority of iron-based superconductors are constructed from quasi two-dimensional (2D) FeAs or FeSe layers and host a range of metallic, semi-metallic, and insulating behaviors originating from the interplay between structural, orbital, magnetic and electronic degrees of freedom [3–6]. Their magnetic ground-states and excitations have been extensively studied by neutron scattering [7–12] but a unified theoretical description that accounts for the role of Coulomb repulsion and Hund’s coupling on electrons occupying multiple active 3d-orbitals remains a challenging task. To understand these unfamiliar Fe-based magnets it is necessary to probe electronic correlations at the atomic scale in chemically and structurally related materials.

Inspired by the successful description of magnetic, electronic and orbital phenomena in various insulating chain- and ladder-based cuprates [13–15], recent experimental work explored the properties of quasi one-dimensional (1D) Fe-based compounds such as KFe$_2$Se$_3$ [16], CsFe$_2$Se$_3$ [17], BaFe$_2$Se$_2$O [18], TaFe$_{1+y}$Te$_3$ [19] and the two-leg ladder BaFe$_2$Se$_3$ [20–25]. Unlike the former materials, BaFe$_2$Se$_3$ hosts an exotic form of magnetic order, the Fe$_4$ block state, that has also been observed in the $\sqrt{5} \times \sqrt{5}$ vacancy-ordered quasi-2D compound Rb$_{0.88}$Fe$_{1.58}$Se$_2$ [11, 26] and reproduced by first-principles electronic structure calculations [27]. Facilitated by low-dimensionality, exact diagonalization (ED) and density-matrix renormalization group (DMRG) analysis [28, 29] of multi-orbital Hubbard models relevant for BaFe$_2$Se$_3$ indicate the exotic block state is stabilized by sizable Hund’s coupling [28, 29]. It is proposed that BaFe$_2$Se$_3$ forms an orbital-selective Mott phase [29] where narrow-band localized electrons coexist with wide-band itinerant electrons originating from different 3d atomic orbitals [27, 30].

In this work, we determine the magnetic excitation spectrum of BaFe$_2$Se$_3$ through broad band inelastic neutron scattering from a powder sample. We provide direct spectroscopic evidence for the Fe$_4$ block state [Fig. 1] and develop an effective Heisenberg model that accounts for all observed acoustic and optical spin-wave modes. We also determine the effective moment in the energy range below 300 meV to be $\mu_{eff}^2 \approx 16 \mu_B^2$ per Fe, which is indicative of spin-orbital magnetism in BaFe$_2$Se$_3$.

The crystal structure of BaFe$_2$Se$_3$ [Fig. 1] comprises

![Crystal structure of BaFe$_2$Se$_3$](image)

FIG. 1. (Color online) (a) Crystal structure of BaFe$_2$Se$_3$ with Ba-atoms omitted for clarity. The Fe$_4$ block ground-state is represented with light (spin-down) and dark (spin-up) bold arrows. (b) Structure of an individual ladder in the $Pnma$ space-group determined from a pair-distribution-function analysis of powder neutron diffraction data. (c) Values of exchange interactions determined from our BaFe$_2$Se$_3$ data using an effective Heisenberg model.
edge-sharing FeSe$_4$ tetrahedra forming two-leg Fe-ladders along the $b$ axis of the orthorhombic space-group $Pnma$ determined by low-temperature neutron powder diffraction (NPD) [20, 21, 23, 31]. Individual ladders are separated by Ba atoms with a face-centered organization in the $ac$ plane. The environment of the Fe site is distorted with four distinct distances to coordinating Se atoms. BaFe$_2$Se$_3$ is a magnetic insulator with a resistivity activation-energy of $E_a \approx 0.13-0.18$ eV [21, 23]. Assuming a high-spin electronic configuration in the tetrahedral crystal field leads to $S = 2$ per Fe$^{2+}$ 3$d^6$ ions. Long-range magnetic order develops below $T_N \approx 255$ K with a large saturated moment of $2.8 \mu_B$ per Fe [20, 21] and a propagation-vector $\kappa = (1/2, 1/2, 1/2)$. The corresponding magnetic structure consists of Fe$_4$ blocks (plaquettes) in which four nearest-neighbor spins co-align parallel to the $a$ direction [20, 21]. In turn, the plaquettes arrange in a staggered fashion along the ladder and inter-ladder directions [Fig. 1(a)] with no net magnetization.

It is not easy to reconcile the Fe$_4$ block ground-state with the $Pnma$ lattice structure because the two inequivalent Fe–Fe distances are staggered between the two legs of the ladder. This hints at the possible role of exchange frustration, orbital ordering and/or spin-lattice coupling to stabilize the exotic magnetic state. Detailed neutron pair-distribution-function (NPDF) analysis [16, 20] reveals gradual Fe displacements upon cooling resulting in structural Fe$_4$ blocks [Fig. 1(b)]. The local displacements lead to two inequivalent Fe sites in the $Pnma_2_1$ space-group although a Rietveld refinement of the NPD data cannot discriminate between the two lattice structures. Due to the large magneto-elastic coupling, it was recently proposed that BaFe$_2$Se$_3$ may host a ferrielectric polarization driven by exchange striction [32].

To search for magnetic excitations associated with the Fe$_4$ block spin structure, our inelastic neutron scattering experiment was performed on the ARCS [33] time-of-flight spectrometer at the Spallation Neutron Source (SNS), Oak Ridge National Laboratory (ORNL). A $m \approx 9.9$ g power sample of BaFe$_2$Se$_3$, synthesized using the method of Ref. [20], was mounted in an aluminum can, sealed under 1 atm of $^3$He, and cooled to a base temperature of $T = 5$ K in a close-cycled cryostat. To reduce multiple scattering, the can contained horizontal sheets of neutron adsorbing Cd inserted every centimeter between layers of BaFe$_2$Se$_3$ powder. Data were acquired with the incident neutron energy set to $E_i = 20$ meV, 50 meV, 150 meV and 450 meV with full-width at half-maximum (FWHM) elastic energy-resolution of $0.8$ meV, $2.0$ meV, $5.8$ meV and $40$ meV, respectively. The intensity measured as a function of momentum $h\mathbf{Q} = h(Q)$ and energy-transfer $E = h\omega$, $I(Q, E) = k_i/k_f (d^2\sigma/dQdE)$, was normalized to absolute units (mbarns sr$^{-1}$ meV$^{-1}$ Fe$^{-1}$) using the intensity of the nuclear elastic scattering in the paramagnetic phase at $T = 300$ K.

In Fig. 2, we discuss the elastic scattering and spectrum of low-energy excitations measured in BaFe$_2$Se$_3$. At $T = 5$ K [Fig. 2(a)], we observe an intense ridge of inelastic signal which extends from $E \approx 4$ meV and is characterized by a sharp onset at $Q \approx 0.7$ Å$^{-1}$ and an asymmetric lineshape towards larger $Q$. A similar signal is observed for $Q \approx 1.8$ Å$^{-1}$ and 2.5 Å$^{-1}$. The corresponding $I(Q)$ obtained by $E$-integration over the range $4 \leq E \leq 12$ meV can be compared to the scaled elastic signal $I_0(Q)$ integrated over $E = \pm 2$ meV [Fig. 2(b)]. The coherent elastic signal can be reproduced without any free parameter using the known magnetic propagation vector $\kappa$ and Fe$_4$ block spin structure, and a static moment of $\langle m \rangle = 2.7(1) \mu_B$ per Fe. The latter value, obtained from an order-parameter fit to the temperature dependence of the magnetic intensity using $E_i = 20$ meV and $E = \pm 0.8$ meV [Fig. 2(c)], agrees very well with previous diffraction results [16, 20, 21]. As maxima in $I(Q)$ are observed around strong magnetic Bragg reflections of...
$\tilde{I}_0(Q)$, the former signal clearly originates from acoustic spin-waves and contains information particularly about inter-plaquette magnetic interactions in BaFe$_2$Se$_3$.

To determine the dimensionality of magnetism in BaFe$_2$Se$_3$, we use an approach previously employed to determine the spatial correlations in short-range ordered states [34] and compare the $E$-integrated inelastic signal to the powder-average form

$$I(Q) \propto \int \frac{d\Omega}{4\pi} |f(Q)|^2 \sum_{\tau_m} \frac{|F_\perp(\tau_m)|^2}{1 + \sum_{\alpha=1}^3 c_{\alpha}^2 |(Q - \tau_m) \cdot \hat{e}_{\alpha}|^2},$$

where $f(Q)$ is the magnetic form-factor of Fe$^{2+}$, $F_\perp(Q)$ is the magnetic scattering amplitude perpendicular to the momentum-transfer $Q$, and $\tau_m = \tau \pm \kappa$ with $\tau = ha^* + kb^* + ic^*$ a reciprocal-lattice vector. The parameters $\xi_\alpha$ with $\alpha = 1, 2, 3$ correspond to pseudo-correlation lengths in the crystallographic directions $a$, $b$ and $c$, respectively. After convolution with the $Q$-resolution of the instrument, the best fit to the inelastic data is obtained for $\xi_a = 3(1)$ Å, $\xi_b = 22(5)$ Å and $\xi_c = 13(3)$ Å [Fig. 2(b)]. Qualitatively, this reveals a quasi-1D character for magnetism in BaFe$_2$Se$_3$ with longest range correlations along $b$, the ladder direction, but also sizable correlations along $c$. Correlations along $a$ are on the inter-atomic length scale only.

Our higher-resolution $E_i = 20$ meV data reveal an apparent gap $\Delta \approx 5$ meV in the spectrum for $T = 5$ K [Fig. 2(d)]. This gap closes when warming to $T = 300$ K but the $\Delta$ energy scale remains apparent. The signal’s lineshape changes from a Gaussian peak centered at $Q \approx 0.72$ Å$^{-1}$ for $E < \Delta$ to an asymmetric peak-shape that onsets at $Q \approx 0.68$ Å$^{-1} = |(0, \frac{1}{2}, \frac{1}{2})|$ for $E > \Delta$, where it resembles the lineshape of the low temperature spectrum. This behavior can be qualitatively understood by the dimensionality of the inter-ladder interactions and a small single-ion or exchange anisotropy responsible for $\Delta$.

To determine the bandwidth of the acoustic spin-waves in BaFe$_2$Se$_3$, we turn to the $E$-dependence of the low-energy signal $\tilde{I}(E)$ integrated over $0.6 \leq E \leq 2.1$ Å with $E_i = 150$ meV [Fig. 2(f)]. The low-energy excitations extend continuously from $E \approx \Delta$ up to $E \approx 50$ meV, with a small peak at $E_1 = 46(1)$ meV indicating the top of the acoustic spin wave band [see also Fig. 3(c)]. As we shall see, this conventional 45 meV wide spectrum of acoustic spin waves belies the exotic Fe$_4$ block state.

It is the higher-energy excitations of BaFe$_2$Se$_3$ [Fig. 3] that offer salient signatures of Fe$_4$ block magnetic order. With $E_i = 450$ meV [Fig. 3(a)] and $E_i = 150$ meV [Fig. 3(b)], the experiment covers a large dynamical range and reveals three additional bands of excitations, labeled $n = 2, 3$, and $4$ in the following. Two of these are centered around $E \approx 100$ meV with $E_2 = 88.9(1)$ meV and $E_3 = 108.2(5)$ meV while the higher energy excitations is found at $E_4 = 198(1)$ meV [Fig. 3(c)]. Their corresponding widths (Lorentzian full-width at half-maximum), $\Gamma_n = 1.7(2)$ meV, $4(1)$ meV, and $15(3)$ meV, respectively, can be compared with predictions for the energy-resolution of the spectrometer at $E = E_n$, $\delta E_n = 2.2(4)$ meV, $1.8(4)$ meV and $18(4)$ meV, respectively. Although these calculations over-estimate the resolution width by 20%, they indicate that the $n = 2$ and $n = 4$ modes are close to be resolution-limited while the $n = 3$ excitation is intrinsically broad. The $Q$-dependence of all four bands of magnetic excitations [Fig. 3(d)] compare well with the Fe$^{2+}$ form-factor for $Q \geq 3\sim 4$ Å$^{-1}$, $\tilde{I}(Q) \propto |f(Q)|^2$ and contrasts with the approximatively $Q$-independent background. Given the form-factor and the fact that charge and intra-orbital $dd$-excitations (crystal-field excitations) have been observed by resonant inelastic X-Ray scattering (RIXS) [25] at higher-energies, $E \approx 0.35$ eV and $E \approx 0.65$ eV respectively, we infer the signal arises from

![FIG. 3.](image-url) (Color online) High energy magnetic spectrum of BaFe$_2$Se$_3$. (a-b) Intensity plot of $\tilde{I}(Q,E)$ at $T = 5$ K with (a) $E_i = 450$ meV and (b) $E_i = 150$ meV. (c) Momentum-integrated inelastic scattering $\tilde{I}(E)$ (open symbols) for various ranges of $Q$ and fits to Lorentzian lineshapes (blue solid lines). (d) Energy-integrated inelastic scattering $\tilde{I}(Q)$ for the $n = 1, 2, 3$ and 4 spin-wave modes (open symbols) compared to $B(Q)$, the momentum-dependence of the nearby background (full symbols), and to the Fe$^{2+}$ form-factor plus background $\tilde{I}(Q) = A|f(Q)|^2 + B(Q)$ (blue solid lines). (e-f) SWT prediction $\tilde{I}_{SWT}(Q,E)$ for (e) $E_i = 450$ meV and (f) $E_i = 150$ meV in arbitrary intensity units.
intra Fe₄ block excitations (optical spin waves).

We can then extract the (total) dynamical spin correlation function for each band of scattering, \( g^2 \tilde{S}_n(Q, E) = 2I_n(Q, E)/|r_0f(Q)|^2 \) with \( r_0 = 0.539 \times 10^{-12} \text{ cm} \), and subsequently obtain the inelastic spectral weight per Fe and per mode \( m_n^a = \mu_B^2 B \int Q^2 |g^2 \tilde{S}_n(Q, E)| \mathrm{d}Q \mathrm{d}E/\int Q^2 \mathrm{d}Q \). After background subtraction and adapting the integration range to the bandwidth of each mode we obtain Tab. I. The total integrated moment, \( m_n^{\text{tot}} = 16(3) \mu_B^2 \) per Fe is significantly smaller than \( g^2 S(S + 1) \mu_B^2 = 24 \mu_B^2 \) expected for \( g = 2 \) and \( S = 2 \). However, it agrees remarkably well with the prediction of \( 16 \mu_B^2 \) per Fe obtained for the Fe₄ block in Ref. [28] from a Hartree-fock treatment of a five-band Hubbard model.

For a more detailed understanding of the magnetic excitations, we develop an effective spin-S Heisenberg model for BaFe₂Se₃. We start from an isolated rectangular Fe₄ block with ferromagnetic \( J_R \) and \( J_L \) exchange interactions along its rungs and legs, respectively [Fig. 4(inset)]. An elementary diagonalization yields four localized excitations at energies \( \varepsilon_n = 0, 2SJ_L, 2SJ_R \) and \( 2S(J_R + J_L) \). These resemble our observations of high-energy optical spin-waves with the ferromagnetic exchange parameters \( SJ_L \approx -44 \text{ meV} \) and \( SJ_R \approx -54 \text{ meV} \) or their permutation. The long range ordered state, the wide acoustic band, and the broadened 108 meV mode imply intra-ladder and inter-ladder exchange interactions that we parametrize consistent with the \( Pnmn_1 \) structure and the effects of which we describe with linear spin-wave theory (SWT) [Fig. 1(b)]. These interactions can originate from Fe-Se-Fe and Fe-Se-Fe-Se-Fe super-exchange paths or from electronic ring-exchange terms that are indistinguishable from further-neighbor exchange at the level of linear SWT [35].

Considering an isolated single-ladder with Fe₄ block spin structure, linear SWT yields four spin-wave modes \( \tilde{\varepsilon}_n \) that directly stem from the above localized modes \( \tilde{\varepsilon}_n \). Their energies are \( \tilde{\varepsilon}_n(k) = S \sqrt{A_n^2 - B_n^2 \cos(4\pi k)} / \sqrt{2} \) where \( A_n \) determines the average energy of each mode and \( B_n = (\pm J_1 + J_2 \pm 2J_3) \) controls the bandwidth of their dispersion, with sign combinations \((+++)\), \((+-)\), \((+--)\) and \((-+++)\) for \( n = 1, 2, 3, \) and \( 4 \), respectively. The constant \( A_n \) depends on \( J_R, J_1, J_1^*, J_2, J_2^* \) and \( J_3 \). As for the isolated Fe₄ block, \( J_R \) and \( J_1 \) (\( \equiv J_L \)) control the overall energy scale and the splitting between \( \varepsilon_2 \) and \( \varepsilon_3 \). As the lowest energy mode acquires a bandwidth controlled by \( J_1^* + J_2^* + 2J_3 \), we expect reduced values for \( J_R \) and \( J_1 \) compared to an isolated plaquette. We also anticipate a sizable \( J_2 \) to tune the relative position of \( \varepsilon_2 \) and \( \varepsilon_3 \) with respect to \( \varepsilon_4 \). As \( J_1^* \) and \( J_2^* \) constrain the bandwidth of the high-energy modes, their values are important to allow a broad \( \varepsilon_2 \) while keeping the widths of \( \varepsilon_2 \) and \( \varepsilon_4 \) limited by the resolution of the instrument.

To obtain realistic values for these exchanges, we compared the experimental energies \( E_{n=1,2,3,4} \) and intrinsic widths \( \Gamma_{n=2,3,4} \) to predictions from the analytical SWT model including instrumental resolution. A least-squares fit to the above experimental constraints yields \( SJ_R = -43.2 \text{ meV}, SJ_1 = -33.2 \text{ meV}, SJ_1^* = 8.4 \text{ meV}, SJ_2 = 11.3 \text{ meV}, SJ_2^* = 6.4 \text{ meV} \) and \( SJ_3 = 15.4 \text{ meV} \), see also Fig. 1(b-c). Our model goes beyond a single-ladder and includes a small easy \( \alpha \)-axis anisotropy \( |SD_\alpha| = 0.08 \text{ meV} \) to account for the spin gap.

In addition, the steep spin-wave dispersion and the absence of enhanced density-of-states (typically associated with inter-chain coupling) below 20 \text{ meV} [Fig. 2(a)], indicates inter-ladder exchanges greater than 3 \text{ meV}. The latter is included in our analytical model with \( SJ_2 = 4(1) \text{ meV} \) while data do not provide evidence for other interactions such as \( J_4 \) and \( J_6 \) [Fig. 1(a)]. Using the numerical implementation of linear SWT [36, 37] in the SpinW program [36], the powder averaged scattering intensity \( I_{\text{SWT}}(Q, E) \) for the model and exchanges of Fig. 1(b-c) is shown in Fig. 2(g) and Fig. 3(e-f). The model accounts for all significant aspects of the data [Fig. 2(a) and Fig. 3(a-b)]. This allows us to present predictions for the dispersion and intensity of the spin-waves out of the Fe₄ block ground-state in a hypothetical single-crystal of BaFe₂Se₃ [Fig. 4].

We have shown that BaFe₂Se₃ is a nearly isotropic quasi-one-dimensional antiferromagnet with a sizable ratio between inter-ladder and intra-ladder interactions \( 4J_2/(J_1^* + J_2^* + 2J_3) \approx 0.45 \). We have determined a set of exchange interactions, compatible with the \( Pnmn_1 \) structure, that stabilizes the Fe₄ block ground-state and produces the peculiar excitation spectrum that we detected. Our experiment recovers a large fraction of, but not the entire, total spectral weight expected for localized \( S = 2 \) spins. The reduced effective moment is evidence for or-
bital selectiveness [29] by which only \( \approx 2/3 \) of the Fe \( 3d \) electrons participate in the formation of local moments. In this respect there are significant similarities to superconducting \( \text{Rb}_{0.89}\text{Fe}_{1.58}\text{Se}_2 \) [11]. The sign reversal between effective intra-block (\( J_1 \)) and inter-block (\( J_2 \)) exchanges interactions is clear evidence for the orbital degrees of freedom that underlie a wealth of exotic magnetic and electronic ground-states in this class of materials. The accurate spin Hamiltonian that we can report for quasi-one-dimensional \( \text{BaFe}_2\text{Se}_3 \), will advance a quantitative understanding of short range spin-orbital interactions in iron bearing square lattices and their potential role in promoting superconductivity.

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