Neutron-Scattering Measurements of Spin Excitations in LaFeAsO and Ba(Fe0.953Co0.047)2As2: Evidence for a Sharp Enhancement of Spin Fluctuations by Nematic Order

Qiang Zhang
*Iowa State University*

Rafael M. Fernandes
*University of Minnesota*

Jagat Lamsal
*Iowa State University*

Jiaqiang Yan
*Oak Ridge National Laboratory*

Songxue Chi
*Oak Ridge National Laboratory*
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Qiang Zhang,1,2 Rafael M. Fernandes,3 Jagat Lamsal,1,2 Jiaqiang Yan,4 Songxue Chi,4 Gregory S. Tucker,1,2 Daniel K. Pratt,5 Jeffrey W. Lynn,5 R. W. McCallum,1,6 Paul C. Canfield,1,2 Thomas A. Lograsso,1,6 Alan I. Goldman,1,2 David Vaknin,1,2 and Robert J. McQueeney1,2,4,*

1Ames Laboratory, Ames, Iowa 50011, USA
2Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011, USA
3School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455, USA
4Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA
5NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899-6102, USA
6Department of Materials Sciences and Engineering, Iowa State University, Ames, Iowa 50011, USA

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Inelastic neutron scattering is employed to investigate the impact of electronic nematic order on the magnetic spectra of LaFeAsO and Ba(Fe0.953Co0.047)2As2. These materials are ideal to study the paramagnetic-nematic state, since the nematic order, signaled by the tetragonal-to-orthorhombic transition at \(T_S\), sets in well above the stripe antiferromagnetic ordering at \(T_N\). We find that the temperature-dependent dynamic susceptibility displays an anomaly at \(T_S\) followed by a sharp enhancement in the spin-spin correlation length, revealing a strong feedback effect of nematic order on the low-energy magnetic spectrum. Our findings can be consistently described by a model that attributes the structural or nematic transition to magnetic fluctuations, and unveils the key role played by nematic order in promoting the long-range stripe antiferromagnetic order in iron pnictides.

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One of the most interesting features of the “122” (e.g., BaFe2As2) and “1111” (e.g., LaFeAsO) families of iron-based superconductors is the intimate coupling between superconductivity (SC), stripe antiferromagnetic (AFM) order, and the tetragonal-to-orthorhombic structural transition \([1–5]\). For example, in both families, chemical substitutions on the transition metal site, such as Co and Ni, suppress the AFM ordering and the structural transition and, over a limited range of doping, promote SC \([4]\). For underdoped BaFe2As2, evidence of a direct competition between AFM and SC has been presented \([6–10]\) in addition to a suppression of the orthorhombic distortion below the superconducting transition temperature \(T_C\) \([11,12]\). Despite this competition between SC and long-range magnetic or orthorhombic order, SC generally arises when large AFM or structural fluctuations are present \([13]\), a feature that attests the intricate relationship between these intertwined phases \([14]\).

While these previous studies have focused on the impact of SC on the magnetic and orthorhombic phases, the interplay between these two ordered states has been a topic of intense debate \([15]\). For the parent compounds of the 122 family, the magnetic transition temperature \(T_N\) practically coincides with the structural distortion at \(T_S\) \([16–18]\), whereas in the Co-underdoped BaFe2As2 and in the parent compounds of the 1111 family, such as LaFeAsO, the orthorhombic distortion occurs well above \(T_N\) \([19,20]\). The structural transition has been proposed to be driven by electronic correlations \([21]\)—associated with either spin \([22–26]\) or charge or orbital degrees of freedom \([27–30]\)—giving rise to the so-called nematic phase in the temperature range between \(T_S\) and \(T_N\). This electronic nematic phase is characterized not only by a weak in-plane structural anisotropy manifested by distinct \(a\) and \(b\) lattice constants \([11]\), but also by large in-plane anisotropies in many electronic properties, such as resistivity \([21,31,32]\), optical conductivity \([33–35]\), thermopower \([36]\), uniform susceptibility \([37,38]\), and charge correlations \([19,39,40]\). Previous angle-resolved photoemission spectroscopy \([41–44]\), scanning tunneling microscopy \([45,46]\), and Raman \([47]\) studies focused on how nematic order affects the normal-state electronic spectrum and, in particular, the charge and orbital degrees of freedom. However, little is known about how nematic order affects the low-energy magnetic fluctuations \([48–52]\), which are particularly important for the formation of the SC state \([13]\).

Here we perform inelastic neutron scattering experiments to elucidate the evolution of the magnetic spectrum across the nematic transition in single crystals of LaFeAsO and Ba(Fe0.953Co0.047)2As2, focusing on the behaviors of the imaginary part of the dynamic magnetic susceptibility \(\chi'(Q, E)\) and of the spin-spin correlation length \(\xi\) as a function of temperature. These two systems exhibit an orthorhombic distortion whose onset is well separated from...
the stripe AFM ordering \[6,19,53,54\], enabling the survival of the nematic phase over a considerable temperature range. Our measurements in twinned samples find clear anomalies in the magnetic spectrum at \(T_S\). In particular, we find that not only is the overall low-energy magnetic intensity enhanced below \(T_S\), but also that the spin-spin correlation length undergoes a sharp increase at the nematic transition temperature, in contrast with what one expects from a typical AFM system. This effect reveals a cooperative relationship between nematicity and magnetism, in agreement with theoretical predictions from models that attribute the nematic transition to a spontaneous symmetry breaking driven by magnetic fluctuations \[22–26\].

The LaFeAsO and \(\text{Ba(Fe}_{0.953}\text{Co}_{0.047})_2\text{As}_2\) crystals were grown using a flux technique as previously described \[53,54\]. Dozens of small single crystals of LaFeAsO with a total mass of approximately 600 mg were co-aligned in the \((\text{HOL})\) plane within \(\sim 2^{\circ}\) mosaicity. Hereafter, unless otherwise noted with a subscript "T," we use orthorhombic notation. A large single crystal of \(\text{Ba(Fe}_{0.953}\text{Co}_{0.047})_2\text{As}_2\) with a mass of \(\approx 700\) mg was also aligned in the \((\text{HOL})\) plane for the investigation. The elastic and inelastic neutron measurements on LaFeAsO and \(\text{Ba(Fe}_{0.953}\text{Co}_{0.047})_2\text{As}_2\) were performed on the HB3 spectrometer (located at the High Flux Isotope Reactor at Oak Ridge National Laboratory) and the BT-7 triple-axis neutron spectrometer at the NIST Center for Neutron Research \[55\], respectively.

In LaFeAsO, neutron diffraction measurements of the \((1\ 0\ 3/2)\) magnetic Bragg reflection and the \((4\ 0\ 0)/(4\ 0\ 4)\) nuclear Bragg reflection as a function of temperature show a structural transition at \(T_S = 165\) K split from the magnetic transition at \(T_N = 145\) K, as illustrated in Fig. 1(a), and consistent with previous reports \[19,53,56,57\]. The \((4\ 0\ 0)/(4\ 0\ 4)\) reflection, which develops from the \((2\ 2\ 0)_{\text{R}}\) tetragonal Bragg reflection, was used to monitor the structural transition indirectly by virtue of secondary extinction changes resulting from the structural transition. Similarly, in \(\text{Ba(Fe}_{0.953}\text{Co}_{0.047})_2\text{As}_2\), the intensity of the \((2\ 0\ 2)/(0\ 2\ 2)\) nuclear Bragg reflection indicates that the structural transition occurs at \(T_S = 60\) K, which is split from the magnetic transition at \(T_N = 47\) K according to the \((1\ 0\ 1)\) magnetic Bragg reflection. The anomalous decrease of the intensity of the \((1\ 0\ 1)\) magnetic peak below \(T_C \approx 17\) K marks the reduction of the AFM order parameter due to competition with the SC state \[6\]. The locations of these three transitions in \(\text{Ba(Fe}_{0.953}\text{Co}_{0.047})_2\text{As}_2\) are also consistent with previous reports \[39,54\].

To determine the impact of nematic order on the magnetic spectrum, we explore the dependence of the imaginary part of the dynamic susceptibility \(\chi''(Q,E)\) on the energy \(E\), the momentum \(Q\), and the temperature \(T\). This quantity is extracted via the relationship

\[
S(Q,E) \propto f^2(Q)\chi''(Q,E)(1-e^{-E/k_B T})^{-1},
\]

where \(S(Q,E)\) is the measured background-subtracted intensity \(I(Q,E)-B(Q',E)\), \(f(Q)\) is the magnetic form factor of \(\text{Fe}^{2+}\), and \(k_B\) is the Boltzmann constant. Figure 2 shows \(\chi''(Q_{\text{AFM}},E)\) at the magnetic reflection \(Q_{\text{AFM}} = (101/2)\) in LaFeAsO and \(Q_{\text{AFM}} = (101)\) in \(\text{Ba(Fe}_{0.953}\text{Co}_{0.047})_2\text{As}_2\) at several temperatures. Below \(T_N\), the spectra in LaFeAsO exhibit the onset of an energy gap \(\sim 5\) meV, consistent with previous reports \[56\]. In \(\text{Ba(Fe}_{0.953}\text{Co}_{0.047})_2\text{As}_2\), a heavily overdamped energy gap \(\sim 10\) meV \[58\] is observed. It has been reported \[58\] that upon increase of Co substitution in \(\text{Ba(Fe}_{1-x}\text{Co}_x)_{2}\text{As}_2\), the spin gap appears to close gradually and is completely absent at \(x = 0.055\) due to the crossover from well-defined spin waves to overdamped spin excitations. The spin gaps in both systems vanish above \(T_N\) and the energy-dependent damping also increases above \(T_N\). These results guide us to measure \(\chi''(Q,E)\) at a fixed energy transfer of \(E = 5\) meV in LaFeAsO and \(E = 3\) meV in \(\text{Ba(Fe}_{0.953}\text{Co}_{0.047})_2\text{As}_2\) to obtain both the spin-spin correlation length and the magnetic intensity as a function of temperature, according to the model for spin fluctuations described in Ref. \[58\].

Representative longitudinal \(H\) scans through \(Q_{\text{AFM}} = (101/2)\) in LaFeAsO and \(Q_{\text{AFM}} = (101)\) in \(\text{Ba(Fe}_{0.953}\text{Co}_{0.047})_2\text{As}_2\) at low energy transfers are shown in Fig. 3. The solid lines represent Gaussian fits to the data,
FIG. 2 (color online). Low-energy spin excitation in LaFeAsO at (a) 170, (b) 147, and (c) 4 K and in Ba(Fe$_{0.953}$Co$_{0.047}$)$_2$As$_2$ at (d) 70, (e) 47, and (f) 2.5 K. The results are derived from the difference between a constant-$Q$ energy scan at $Q_{\text{AFM}}$ and a background scan at $Q = (0.701)$ for Ba(Fe$_{0.953}$Co$_{0.047}$)$_2$As$_2$ and at $Q = (0.83 \pm 0.098)$ after the crystal was rotated from nominal $Q_{\text{AFM}}$ by 20° for LaFeAsO. The intensities have been normalized to reflect a counting time of approximately 5 min. Error bars where indicated represent one standard deviation.

as justified in the Supplemental Material [59]. We note that upon decreasing the temperature below $T_N$, the line shape narrows and the peak amplitude increases. The dynamic susceptibility and linewidth (full width at half maximum) versus temperature are shown in Fig. 4. Note that the reasonable mosaicity within $\sim 2^\circ$ of the coaligned LaFeAsO samples does not appreciably affect the linewidth of longitudinal scans and thus the linewidth reflects the intrinsic behavior of the spin-spin correlation length similar to that of Ba(Fe$_{0.953}$Co$_{0.047}$)$_2$As$_2$. The dynamic susceptibility shows a discontinuous increase below $T_S$ (much stronger for LaFeAsO) and exhibits a maximum at the AFM ordering temperature $T_{N}$, followed by a gradual decrease below $T_N$ due to the opening of the spin gap. As shown in Figs. 4(b) and 4(d), the linewidth decreases as $T$ approaches $T_N$, which is expected for a classic second-order AFM phase transition. The striking result of this study is the observation of a sharp decrease in the linewidth below $T_S$ in both LaFeAsO and Ba(Fe$_{0.953}$Co$_{0.047}$)$_2$As$_2$ systems, which signifies a strong effect of nematic order on the approach to AFM order.

Above the magnetic transition temperature $T_N$, the linewidth of the constant-$Q$ scans is proportional to the inverse magnetic correlation length $\xi^{-1}$ associated with the paramagnetic fluctuations [58,64] (also see the Supplemental Material [59]). Therefore, the onset of long-range nematic order promotes a strong increase of this correlation length, enhancing the tendency of the system towards long-range magnetic order. Such a cooperative interplay between nematicity and magnetism can be understood qualitatively within models that attribute the tetragonal symmetry breaking to magnetic fluctuations emerging from either localized [22, 23] or itinerant spins [25]. To illustrate the corresponding microscopic mechanism, we show schematically in Fig. 5(a) the evolution of the magnetic fluctuations across $T_S$ and $T_N$ both in real space (upper panels) and in spin space (lower panels). The crucial point behind this mechanism is that the iron pnictides display two degenerate stripe AFM ground states, with ordering vectors $Q_1 = (10L)$ and $Q_2 = (01L)$. Thus, the magnetic ground state can be described in terms of two interpenetrating square sublattices—associated with the two distinct Fe atoms in the unit cell—that tend to order magnetically in Néel-like configurations [blue and red dashed lines in Fig. 5(a)].

Above $T_S$, where there is no long-range magnetic order, these two sublattices are essentially independent [as shown in the upper left panel of Fig. 5(a)], and their fluctuations are uncoupled (as shown in the lower left panel). As a result, the system has multiple possible ground states, a
feature commonly seen in frustrated spin systems with low magnetic transition temperatures. However, below \( T_S \) but above \( T_N \), nematic order emerges as a coupling between the two sublattices [upper middle panel in Fig. 5(a)], enforcing the two corresponding Néel order parameters to fluctuate coherently either antiparallel (as shown in the lower middle panel) or parallel to each other. There is still no long-range magnetic order, since the spins can point at any direction in spin space. However, the tetragonal symmetry of the system is broken, since nearest-neighbor spins are locked in a ferromagneticlike or an antiferromagneticlike configuration. Furthermore, by breaking the tetragonal symmetry, nematic order reduces the number of possible magnetic ground states to only one—either the \( \mathbf{Q}_1 = (1 \ 0 \ 1) \) stripe if the \( a \) direction is selected along the \( x \) axis, or the \( \mathbf{Q}_2 = (0 \ 1 \ 0) \) stripe if the \( a \) direction is selected along the \( y \) direction. Thus, the frustration, resulting from two degenerate magnetic stripe states present at higher temperatures, is lifted by nematic order, leading to an enhancement of the spin-spin correlation length \( \xi \), and therefore of \( T_N \), which sets in when \( \xi \) diverges (right panels). Note that this phenomenon can be observed even in twinned samples as the ones studied here, since magnetic fluctuations are enhanced regardless of the type of nematic domain selected.

To go beyond this qualitative analysis, we calculate \( \xi \) using a low-energy action for the magnetic degrees of freedom that accounts for the existence of two symmetry-related magnetic instabilities, giving rise to a preemptive nematic phase at \( T_S > T_N \) (see Ref. [25] for a microscopic derivation from an itinerant three-band model). The equations for \( \xi \) and the parameters used here are presented in the Supplemental Material [59]. To take into account the resolution limitations in the linewidth \( W \) imposed by the instrument and by the fact that the measurements are performed at nonzero energy, we shift \( \xi^{-1} \) by a temperature-independent term \( \delta_{\text{res}} > 0 \), \( W \propto \xi^{-1} + \delta_{\text{res}} \). The results are shown in Fig. 5(b). Because our model is based on an expansion near \( T_N \), it systematically underestimates the correlation length at higher temperatures. Yet, it captures the main qualitative feature observed experimentally, namely, the sharp enhancement of \( \xi \) below \( T_S \) due to the onset of long-range nematic order. This is shown explicitly in Fig. 5(b) by comparing the hypothetical behavior of \( \xi \) in the absence of nematic order (dashed lines) with the behavior in the presence of nematicity (solid lines). We emphasize that this theoretical calculation is
intended to highlight the strong feedback effect of nematic order on the magnetic fluctuations, and not to capture the full quantitative dependence of $\xi$ on temperature, which will be affected by other features such as domains, mosaicity, etc.

In summary, we have reported unambiguous evidence for the feedback effect of nematic order on the magnetic spectrum in both “1111” and underdoped “122” families of the iron pnictides with $T_S > T_N$, manifested by the sharp enhancement of the spin-spin correlation length below $T_S$, revealing a key impact of this elusive electronic order on the normal-state properties of the iron arsenides. Since magnetic fluctuations are believed to be important for the formation of the SC state [13], and our results provide evidence that nematic order enhances them, this suggests that nematicity may be more than another competing order, as previously reported [11,12], and may even help enhancing $T_C$ in some circumstances [65,66].

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*mcqueeneyrj@ornl.gov*


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