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Nematic spin correlations in the tetragonal state of uniaxial-strained $BaFe_{2-x}Ni_xAs_2$

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Understanding the microscopic origins of electronic phases in high-transition temperature (high-T_c) superconductors is important for elucidating the mechanism of superconductivity. In the paramagnetic tetragonal phase of $BaFe_{2-x}T_xAs_2$ (where T is Co or Ni) iron pnictides, an in-plane resistivity anisotropy has been observed. Here, we use inelastic neutron scattering to show that low-energy spin excitations in these materials change from four-fold symmetric to two-fold symmetric at temperatures corresponding to the onset of the in-plane resistivity anisotropy. Because resistivity and spin excitation anisotropies both vanish near optimal superconductivity, we conclude that they are likely intimately connected.

Superconductivity in iron pnictides can be induced by electron or hole-doping of their antiferromagnetic (AF) parent compounds (*1–6*). The parent compounds exhibit a tetragonal-to-orthorhombic structural phase transition at temperature T_s, followed by a paramagnetic to AF phase transition at T_N ($T_s \geq T_N$) (*4–6*). An in-plane resistivity anisotropy has been observed in uniaxially strained iron pnictides $BaFe_{2-x}T_xAs_2$ (where T is Co or Ni) above T_s (*7–9*). This anisotropy vanishes near optimal superconductivity and has been suggested as a signature of the spin nematic phase that breaks the in-plane four-fold rotational symmetry (C_4) of the underlying tetragonal lattice (*10–14*). However, such interpretation was put in doubt by recent scanning tunneling microscopy (*15*) and transport (*16*) measurements, which suggest that the resistivity anisotropy in Co-doped $BaFe_2As_2$ arises from Co-impurity scattering and is not an intrinsic property of these materials. On the other hand, angle-resolved photoemission spectroscopy (ARPES) measurements found that the onset of a splitting in energy between two orthogonal bands with dominant d_{xz} and d_{yz} character in the uniaxial-strain detwinned samples at a temperature above T_s (*17*, *18*), thereby suggesting the involvement of the orbital channel in the nematic phase (*19–22*). Here, we use inelastic neutron scattering (INS) to show that low-energy spin excitations in $BaFe_{2-x}Ni_xAs_2$ (x = 0, 0.085, and 0.12) (*23*, *24*) change from four-fold symmetric to two-fold symmetric in the uniaxial-strained tetragonal phase at temperatures corresponding to the onset of the in-plane resistivity anisotropy.

The magnetic order of the parent compounds of iron pnictide superconductors is collinear, with the ordered moment aligned antiferromagnetically along the a_o axis of the orthorhombic lattice (Fig. 1A), and occurs at a temperature just below $T_s \approx T_N \approx 138$ K for $BaFe_2As_2$ (*5*, *6*). Because of the twinning effect in the orthorhombic state, AF Bragg peaks from the twinned domains appear at the (±1, 0) and (0, ±1) in-plane positions in reciprocal space (Fig. 1B) (*3*). Therefore, one needs to prepare single domain samples by applying a uniaxial pressure (strain) along one axis of the orthorhombic lattice to probe the intrinsic electronic properties of the system (*7–9*). Indeed, transport measurements on uniaxial-strain detwinned samples of electron-underdoped $BaFe_{2-x}T_xAs_2$ (*7–10*) reveal clear in-plane resistivity anisotropy even above the zero pressure T_c, T_N, and T_s (Fig. 1C).

To search for a possible spin nematic phase (*12–14*), we carried out INS experiments in uniaxial-strain detwinned parent compound $BaFe_2As_2$ (T_N = 138 K), electron-underdoped superconducting $BaFe_{1.915}Ni_{0.085}As_2$ (T_c = 16.5 K, T_N = 44 K), and electron-overdoped superconducting $BaFe_{1.88}Ni_{0.12}As_2$ (T_c = 18.6 K, tetragonal structure with no static AF order) (Fig. 1C) (*23*, *24*) using a thermal triple-axis spectrometer. Horizontally and vertically curved pyrolytic graphite (PG) crystals were used as a monochromator and analyzer. To eliminate contamination from epithermal or higher-order neutrons, a sapphire filter was added before the monochromator, and two PG filters were installed before the analyzer. All measurements were done with a fixed final wave vector, k_f = 2.662 Å^{-1}. Our annealed square-shaped single crystals of $BaFe_2As_2$ (~120 mg), $BaFe_{1.915}Ni_{0.085}As_2$ (~220 mg), and $BaFe_{1.88}Ni_{0.12}As_2$ (~448 mg) were mounted inside aluminum-based sample holders with a uniaxial pressure of $P \approx 15$ MPa, ~7 MPa, and ~7 MPa, respectively, applied along the a_o/b_o axes direction (fig. S1A) (*25–27*). We define momentum transfer **Q** in three-dimensional reciprocal space in Å^{-1} as $\mathbf{Q} = H\boldsymbol{a}^* + K\boldsymbol{b}^* + L\boldsymbol{c}^*$, where H, K, and L are Miller indices and $\boldsymbol{a}^* = \hat{\mathbf{a}}_o 2\pi/a_o$, $\boldsymbol{b}^* = \hat{\mathbf{b}}_o 2\pi/b_o$, and $\boldsymbol{c}^* = \hat{\mathbf{c}}\, 2\pi/c_o$. In the AF ordered state of a 100% detwinned sample, the AF Bragg peaks should occur at (±1, 0, L) (L = 1, 3, 5, · · ·) positions in reciprocal space. In addition, the low-energy spin waves should only stem from the (±1, 0) positions with no signal at the (0, ±1) positions (*26*, *27*). By contrast, in the paramagnetic tetragonal phase ($T > T_s \geq T_N$) one would expect the spin excitations at the (±1, 0) and (0, ±1) positions to have equal intensities (*12*, *27*).

The results of our INS experiments on uniaxial-strain detwinned $BaFe_{2-x}Ni_xAs_2$ are summarized in Fig. 1C. The square red symbols indicate the temperature below which spin excitations at an energy transfer of E = 6 meV exhibit a difference in intensity between the (±1, 0) and (0, ±1) positions for undoped and electron underdoped $BaFe_{2-x}Ni_xAs_2$. For electron overdoped $BaFe_{1.88}Ni_{0.12}As_2$, the same uniaxial pressure has no effect on spin excitations at wave vectors (±1, 0) and (0, ±1) (*27*). A comparison to the transport measurements (*10*) in Fig. 1C indicates that the resistivity anisotropy occurs near the spin excitation anisotropy temperature T^* determined from INS.

Given that our experiments are performed in uniaxial-strain detwinned samples, it is important to establish how the structural and magnetic transition temperatures are affected by the applied pressure. Figure S2A compares the temperature dependence of the magnetic order parameters at (1, 0, 1)/(0, 1, 1) for $BaFe_2As_2$ in zero pressure (green symbols) and under uniaxial strain (red and blue symbols). We find that the $BaFe_2As_2$ sample is essentially 100% detwinned under the applied uniaxial strain without altering T_N (*27*). Similarly, the electron underdoped $BaFe_{1.915}Ni_{0.085}As_2$ is about 80% detwinned and has $T_N \approx 44$ K, unchanged from the zero-pressure case (fig. S2B) (*27*). To investigate whether the tetragonal-to-orthorhombic structural phase transition in $BaFe_{2-x}Ni_xAs_2$ is affected by uniaxial strain, we plot the temperature dependence of the (2, −2, 0) nuclear Bragg peak of $BaFe_2As_2$; both zero pressure (fig. S2C) and detwinned samples (fig. S2D) exhibit a steplike feature at $T_s \approx 138$ K resulting from the vanishing neutron extinction effect due to the tetragonal-to-orthorhombic structural transition (*28*, *29*).

In previous spin-wave measurements on twinned $BaFe_2As_2$, a spin gap of ~10 meV was found at the (1, 0, 1) and (0, 1, 1) positions (*30*). To probe spin excitations at the same wave vectors in the detwinned $BaFe_2As_2$, we aligned the sample in the [1, 0, 1] × [0, 1, 1] scattering plane (*27*). Figure 2, A, C, and E, shows constant-energy scans centered at (1, 0, 1) approximately along the [1, *K*, 1] direction. Whereas spin waves at (1, 0, 1) are clearly gapped at $E = 6$ meV in the AF ordered state ($T = 3$ K) in Fig. 2A, they are well-defined at $E = 15$ meV (Fig. 2C) and 19 meV (Fig. 2E), in line with the previous report (*31*). We find no evidence for spin waves at $E = 6$, 15, and 19 meV at (0, 1, 1) (Fig. 2, B, D, and F, respectively), which is consistent with a nearly 100% detwinned $BaFe_2As_2$. On warming the system to the paramagnetic tetragonal state at $T = 154$ K, the spin gap disappears and the $E = 6$ meV spin excitations at the AF wave vector (1, 0, 1) are clearly stronger than those at (0, 1, 1) (Fig. 2, A and B) (*27*).

To quantitatively study the energy dependence of the spin excitation anisotropy in $BaFe_2As_2$ at a temperature above T_s, we plot in Fig. 3A the energy scans at wave vectors (1, 0, 1) and (0, 1, 1) and their corresponding backgrounds at $T = 154$ K (*27*). The background-subtracted scattering at (1, 0, 1) is consistently higher than that at (0, 1, 1) (Fig. 3, C and E, left inset). When we warm up to $T = 189$ K, the corresponding energy scans (Fig. 3B) and the signals above background (Fig. 3D) reveal that the differences at these two wave vectors disappear (Fig. 3E, left inset). Figure 3E shows the temperature dependence of the spin excitations (signal above background scattering) across T_N and T_s. In the AF ordered state, we see only spin waves from the wave vector (1, 0, 1). On warming to the paramagnetic tetragonal state above T_N and T_s, we see clear differences between (1, 0, 1) and (0, 1, 1) that vanish above ~160 K, the same temperature below which anisotropy is observed in the in-plane resistivity (Fig. 3E, right inset) (*32*). We conclude that the four-fold to two-fold symmetry change in spin excitations in $BaFe_2As_2$ occurs alongside the resistivity anisotropy.

To see if spin excitations in superconducting $BaFe_{1.915}Ni_{0.085}As_2$ also exhibit the four-fold to two-fold symmetry transition, we study the temperature dependence of the $E = 6$ meV spin excitations at the (1, 0, 1) and (0, 1, 1) wave vectors. In previous INS experiments on twinned $BaFe_{1.92}Ni_{0.08}As_2$, a neutron spin resonance was found near $E \approx 6$ meV (*33*). Figure 4, A and C, shows approximate transverse and radial scans through (1, 0, 1) at various temperatures; one can clearly see the superconductivity-induced intensity enhancement from 48 K to 8 K. The corresponding scans through (0, 1, 1) (Fig. 4, B and D) have weaker intensity than those at (1, 0, 1). Figure 4E shows the temperature dependence of the magnetic scattering at (1, 0, 1) and (0, 1, 1). Consistent with constant-energy scans in Fig. 4, A to D, the scattering at (1, 0, 1) is considerably stronger than that at (0, 1, 1) above T_c. On warming through T_N and T_s (*24*), the spin excitation anisotropy between (1, 0, 1) and (0, 1, 1) becomes smaller, but reveals no dramatic change. The anisotropy disappears around $T^* \sim 80$ K, well above T_N and T_s (Fig. 4, E and F) but similar to the point of vanishing in-plane resistivity anisotropy (*10*). Finally, we find that uniaxial strain does not break the C_4 rotational symmetry of the spin excitations in electron-overdoped $BaFe_{1.88}Ni_{0.12}As_2$ (fig. S5) (*27*). In this compound, resistivity shows no a_o/b_o anisotropy (*10*).

Conceptually, once the C_4 symmetry of the electronic ground state is broken, the electronic anisotropy will couple linearly to the orthorhombic lattice distortion $\epsilon = a_0 - b_0$, so that the C_4 nematic transition should coincide with the tetragonal-to-orthorhombic transition at temperature T_s (*12–14*). How do we then understand the region $T_s < T < T^*$ in which the low-energy spin excitations develop an anisotropy? Theoretically, this is best understood in terms of the effective action for the electronic nematic order parameter Δ and magnetization $\mathbf{M}_{1/2}$ of the interpenetrating Néel sublattices (*14*, *34*, *35*):

$$S_{\mathrm{eff}}\left[\Delta,\mathbf{M}\right]=S_0\left[\mathbf{M}_1^2,\mathbf{M}_2^2\right] +\iint d\mathbf{q}d\omega\left[\alpha^{-1}\left(\mathbf{q},\omega\right)\left|\Delta\right|^2+v\left|\Delta\right|^4-g\left(\mathbf{M}_1\cdot\mathbf{M}_2\right)\Delta-\lambda\epsilon\Delta\right] +C_s\epsilon^2/2 \quad (1)$$

Here, S_0 is defined as part of the action that does not contain nematic correlations $\left(\mathbf{M}_1\cdot\mathbf{M}_2\right)$ (*36*), which have been decoupled in terms of the bosonic field $\Delta\left(\mathbf{q},\omega\right)$ characterized by the nematic susceptibility $\alpha\left(\mathbf{q},\omega\right)$, where energy $E=\hbar\omega$, g is a linear coupling between the Ising-spin variable and the bosonic field, v is the quartic coupling among the bosonic fields, λ is the linear coupling between the bosonic field and the orthorhombic lattice distortion ϵ, and $\mathbf{q}$ is the momentum transfer within one Brillouin zone. Minimizing the action with respect to ϵ, we arrive at $\epsilon=\lambda\left\langle\Delta\right\rangle/C_s$, where C_s is the shear modulus. In other words, the orthorhombic lattice distortion is proportional to the nematic order parameter $\left\langle\Delta\right\rangle$, and both are expected to develop nonzero expectation values below T_s (*12–14*). However, the nematic field Δ undergoes fluctuations in the tetragonal phase above T_s while lattice distortion ϵ remains zero. These fluctuations will be observable in dynamic quantities, such as the finite-energy spin fluctuations, and in transport measurements. We therefore conclude that the scale T^*, below which we observe anisotropy of low-energy spin fluctuations (Figs. 3E, 4E, and 4F) and where the resistivity anisotropy is observed (Fig. 3E, right inset), marks a typical range of the nematic fluctuations.

Several remarks are in order. First, the applied uniaxial pressure used to detwin the samples will induce a finite value of ϵ at any temperature, so that strictly speaking, the structural transition at T_s will be rendered a crossover. In practice, however, the applied pressure is too small to cause a perceptible lattice distortion, which is why the transition temperature T_s, as determined from the extinction effect of the nuclear (2, −2, 0) Bragg peak remains unchanged from the zero-pressure case [fig. S2, B and D] (*26*, *27*). On the other hand, the extent of nematic fluctuations may be sensitive to the shear strain, in agreement with the reported increase of T^* (as determined from resistivity anisotropy) with the uniaxial pressure (*37*). Second, in Eq. 1 the variable Δ could equally signify the orbital order $\Delta\propto\left(n_{xz}-n_{yz}\right)$ which lifts the degeneracy between the Fe d_{xz} and d_{yz} orbitals. In fact, the two order parameters will couple linearly to each other, $\left(\mathbf{M}_1\cdot\mathbf{M}_2\right)\propto\left(n_{xz}-n_{yz}\right)$, so that both will develop a nonzero value below T_s. In this respect, our findings are also consistent with the recent ARPES finding of an orbital ordering (*17*, *18*) in $BaFe_2As_2$. This underlines the complementarity of the spin-nematic and orbital descriptions of the C_4 symmetry breaking. Third, in the nearly optimally electron-doped superconductor, we observe anisotropy of the low-lying spin excitations in the tetragonal phase $T_s < T < T^*$, even though the orbital order is no longer detectable by ARPES (*17*, *18*). This is consistent with the absence of a static nematic order $\left\langle\Delta\right\rangle=0$ above T_s, whereas the observed spin anisotropy originates from Ising-nematic fluctuations. Because T_s is considerably suppressed for this doping, these fluctuations are quantum rather than thermal: They persist beyond the immediate vicinity of T_s, and the associated spin anisotropy should have sizable dependence on frequency that can be probed by future experiments. Fourth, when resistivity anisotropy under uniaxial strain disappears in the electron-overdoped sample (*10*), the uniaxial-strain–induced spin excitation anisotropy also vanishes (Fig. 1E and fig. S5), which suggests a direct connection between these two phenomena. Finally, our measurements in the spin channel do not necessarily signal a thermodynamic order at the temperature T^*. Rather, T^* likely signals a crossover, whereas the true nematic transition occurs at T_s (*9*). This implies that a static order above T_s inferred from recent measurements of magnetic torque anisotropy in the isovalent $BaFe_2As_{2-x}P_x$ (*38*) is most likely not in the spin channel accessible to the inelastic neutron scattering. A static order in other channels—such as, for instance, an octupolar order—would, however, not contradict our observations.

References and Notes

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$$S_0\left[\mathbf{M}_1^2,\mathbf{M}_2^2\right]=\iint d\mathbf{q}d\omega\left(r_0+\mathbf{q}^2+\gamma|\omega|\right)\left(\mathbf{M}_1^2+\mathbf{M}_2^2\right)+u\left(\mathbf{M}_1^2+\mathbf{M}_2^2\right)^2$$

where $r_0 \propto T-T_N$ describes distance from the Néel point.
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Supplementary Materials
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Materials and Methods
Figs. S1 to S5
References (*39*, *40*)

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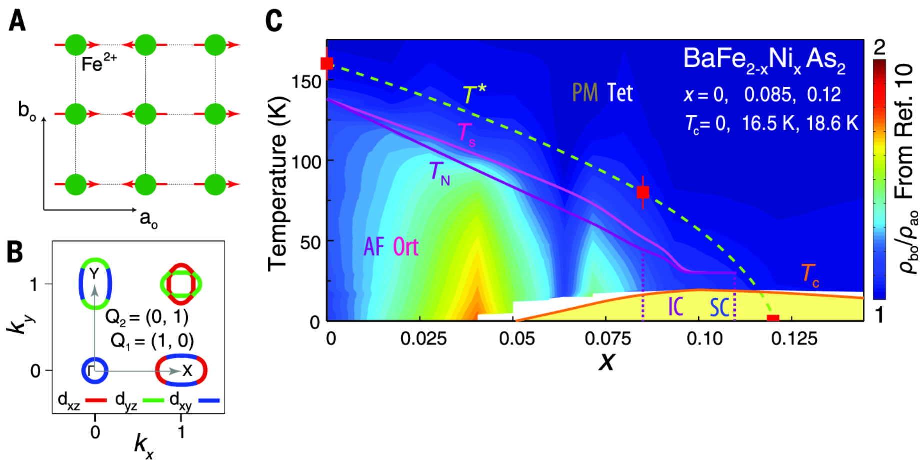

Fig. 1. Summary of the results. (**A**) The AF spin arrangement of iron in the FeAs layer of $BaFe_2As_2$. (**B**) The corresponding Fermi surfaces with one circular hole pocket around the zone center Γ point and two elliptical electron pockets at X and Y points (*3*). (**C**) The electronic phase diagram of $BaFe_{2-x}Ni_xAs_2$ from resistivity anisotropy ratio ρ_{bo}/ρ_{ao} obtained under uniaxial pressure (*10*). The spin excitation anisotropy temperatures are marked as T^*. The AF orthorhombic (Ort), incommensurate AF (IC) (*23*), paramagnetic tetragonal (PM Tet), and superconductivity (SC) phases are marked.

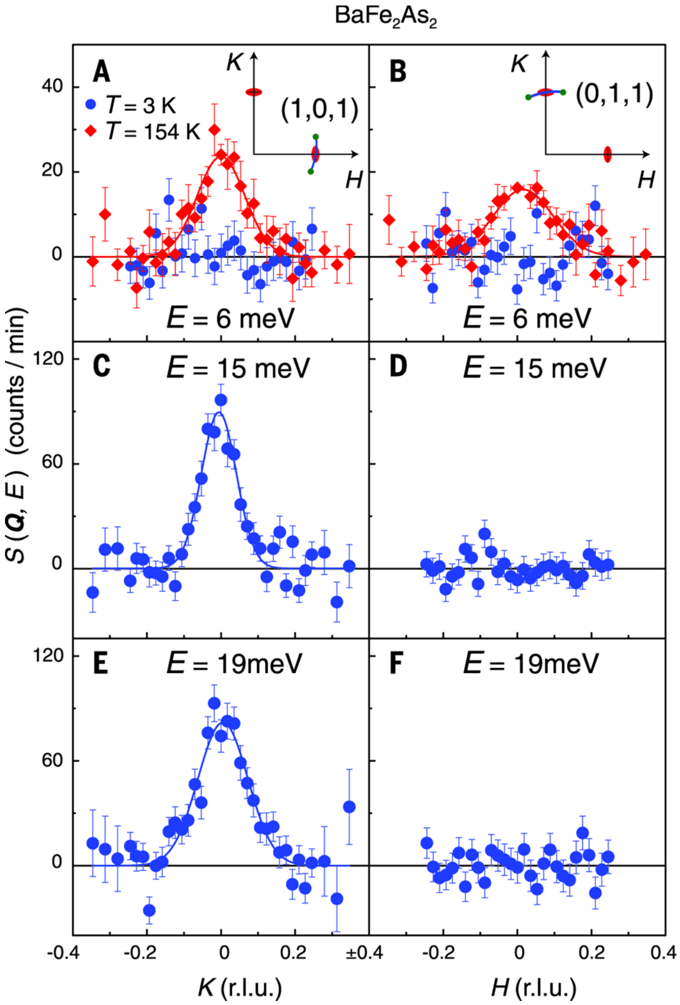

Fig. 2. Constant-energy scans for detwinned $BaFe_2As_2$. The E = 6 meV rocking scans measured at T = 3 K in the AF ordered state and T = 154 K in the paramagnetic tetragonal state centered at (**A**) (1, 0, 1) and (**B**) (0, 1, 1). T = 3 K scans at E = 15 meV for (**C**) (1, 0, 1) and (**D**) (0, 1, 1) and at E = 19 meV for (**E**) (1, 0, 1) and (**F**) (0, 1, 1). The in-plane projected trajectories of the rocking scans crossing (1, 0, 1) and (0, 1, 1) are illustrated by blue lines in the insets of (A) and (B), respectively. Solid lines are Gaussian fits.

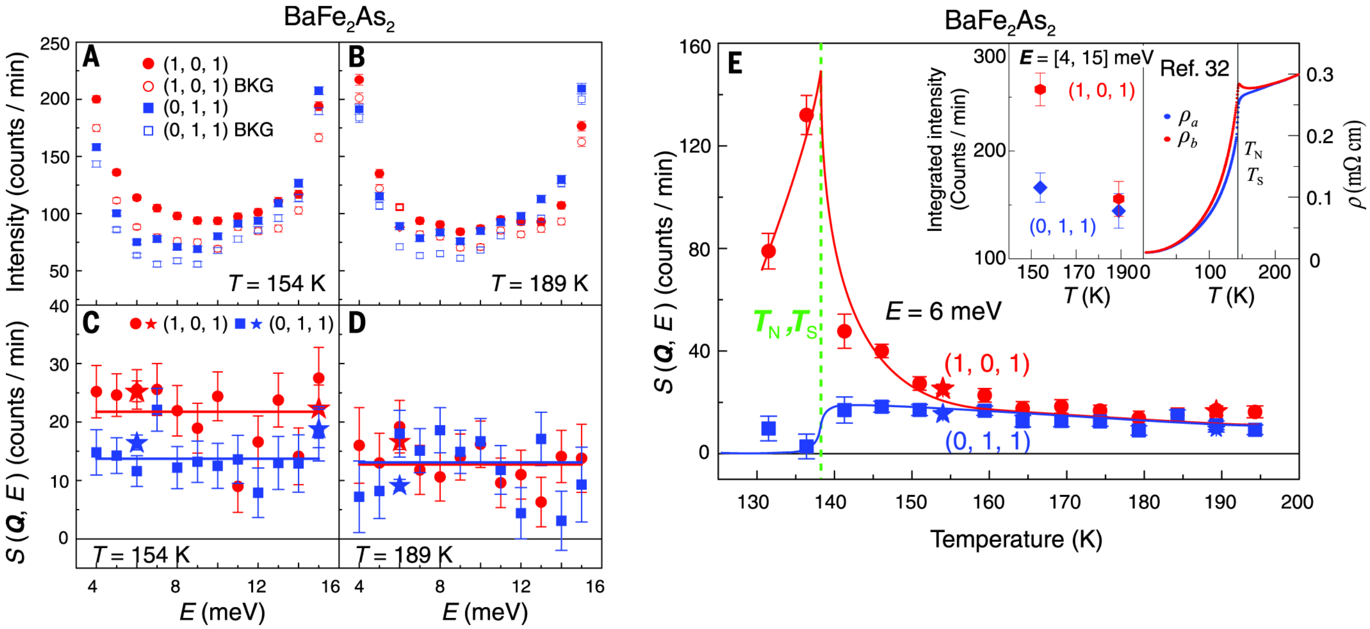

Fig. 3. Temperature dependence of spin excitations for $BaFe_2As_2$. Energy scans at wave vectors (1, 0, 1)/(0, 1, 1) and corresponding background positions at temperatures above T_s: (**A**) T = 154 K and (**B**) 189 K. (**C** and **D**) Magnetic scattering after subtracting the backgrounds. The solid lines are constant line fits. (**E**) Temperature dependence of the spin excitations at E = 6 meV for (1, 0, 1) and (0, 1, 1). The anisotropy in spin excitations vanishes around T = 160 ± 10 K. The data marked by filled squares and dots in (C) to (E) were obtained by subtracting the corresponding backgrounds. For each temperature, the background intensities at **Q** = (1, 0, 1) and (0, 1, 1) were obtained by averaging the data at the two wave vectors marked by green dots in the insets of Fig. 2, A and B, respectively. The data denoted by filled stars in (C) to (E) were obtained by fitting the rocking scans for E = 6 and 15 meV at different temperatures. The solid lines in (E) are guides to the eye. The left inset in (E) shows temperature dependence of the integrated intensity from 4 to 15 K; the right inset shows temperature dependence of the resistivity from (*32*).

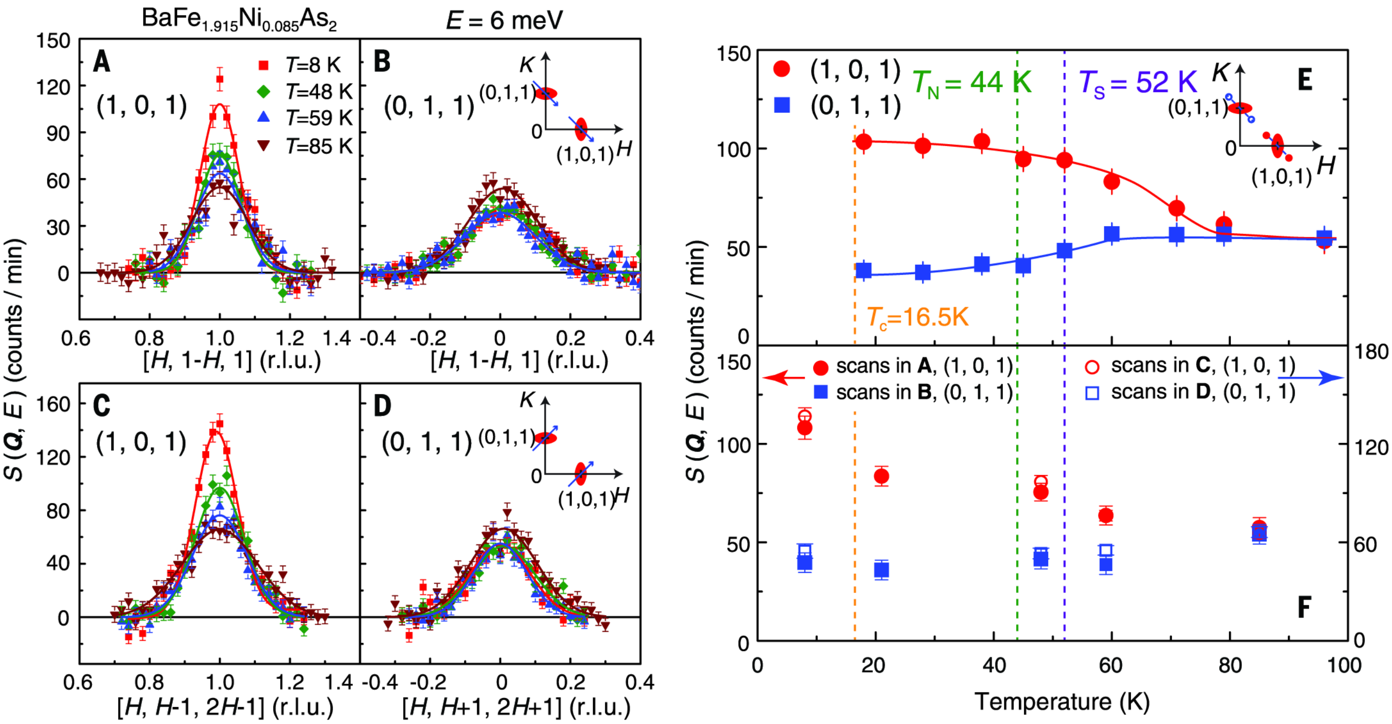

Fig. 4. Temperature dependence of spin excitations for $BaFe_{1.915}Ni_{0.085}As_2$. Background-subtracted **Q** scans at E = 6 meV around (**A** and **C**) (1, 0, 1) and (**B** and **D**) (0, 1, 1). The trajectory of the scans (blue line) crossing spin excitations (red ellipses) are illustrated in the insets of (B) and (D). (**E** and **F**) Temperature dependence of the spin excitations at (1, 0, 1) and (0, 1, 1) at E = 6 meV. The data in (E) were obtained by subtracting the background intensity from the peak intensity at every temperature; the data in (F) were obtained by fitting the **Q** scans. The wave vectors used for calculating the background (blue circles and red dots) are shown in the inset of (E). The solid curves in (A) to (D) are Gaussian fits and in (E) are guides to the eye. The temperatures for structural (purple), magnetic (blue), and SC (orange) transitions are marked by vertical dashed lines (*24*).