In-plane uniaxial pressure-induced out-of-plane antiferromagnetic moment and critical fluctuations in BaFe$_2$As$_2$

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A small in-plane external uniaxial pressure has been widely used as an effective method to acquire single domain iron pnictide BaFe$_2$As$_2$, which exhibits twin-domains without uniaxial strain below the tetragonal-to-orthorhombic structural (nematic) transition temperature $T_s$. Although it is generally assumed that such a pressure will not affect the intrinsic electronic/magnetic properties of the system, it is known to enhance the antiferromagnetic (AF) ordering temperature $T_N$ ($< T_s$) and create in-plane resistivity anisotropy above $T_s$. Here we use neutron polarization analysis to show that such a strain on BaFe$_2$As$_2$ also induces a static or quasi-static out-of-plane ($c$-axis) AF order and its associated critical spin fluctuations near $T_N/T_s$. Therefore, uniaxial pressure necessary to detwin single crystals of BaFe$_2$As$_2$ actually rotates the easy axis of the collinear AF order near $T_N/T_s$, and such effects due to spin-orbit coupling must be taken into account to unveil the intrinsic electronic/magnetic properties of the system.
Understanding the intrinsic electronic, magnetic, and nematic properties of iron pnictides such as BaFe$_2$As$_2$ form the basis to unveil the microscopic origin of high-temperature superconductivity because the system is a parent compound of iron-based superconductors. As a function of decreasing temperature, BaFe$_2$As$_2$ first exhibits a tetragonal-to-orthorhombic structural transition at $T_N$ and forms a nematic ordered phase, followed closely by a collinear antiferromagnetic (AF) order with moment along the $a$-axis of the orthorhombic lattice below the Néel temperature $T_N$ ($\leq T_c$) (Fig. 1a)\textsuperscript{6-9}. Since single crystals of BaFe$_2$As$_2$ form twin domains in the orthorhombic state below $T_N$ an external uniaxial pressure applied along one of the in-plane Fe-Fe bond direction has been widely used as an effective method to acquire single domains of iron pnictide crystals and determine their intrinsic transport\textsuperscript{10-15}, electronic\textsuperscript{16-18}, and magnetic\textsuperscript{19-21} properties (inset in Fig. 1b). Although uniaxial pressure necessary to detwin single crystals of BaFe$_2$As$_2$ is known to increase $T_N$ (Fig. 1b)\textsuperscript{22-25} and create in-plane resistivity anisotropy above $T_N$\textsuperscript{14}, it is generally assumed that it only induces a small strain on the sample and does not significantly modify the electronic and magnetic properties of the system\textsuperscript{10-19,21}. Recently, nuclear magnetic resonance (NMR) experiments have revealed that an in-plane uniaxial strain on BaFe$_2$As$_2$ induces an enhancement of the low-energy spin fluctuations along the $c$-axis in the paramagnetic state above $T_N$\textsuperscript{26}. However, it is unclear whether the applied uniaxial pressure can actually modify the collinear AF structure of the system (Fig. 1a)\textsuperscript{6,7}.

In this work, we use polarized neutron scattering and unpolarized neutron diffraction to demonstrate that an in-plane uniaxial pressure necessary to detwin BaFe$_2$As$_2$ also induces a $c$-axis ordered magnetic moment and changes the easy axis of the collinear AF structure around the magnetic/nematic critical scattering temperature regime where the applied pressure has a large impact on the crystal structure of the system (Fig. 1c-g)\textsuperscript{27}. In addition, we find that the applied pressure induces $c$-axis polarized critical spin fluctuations that diverge near $T_N/T_c$, confirming the results of NMR experiments\textsuperscript{26}. Therefore, uniaxial pressure on BaFe$_2$As$_2$ that breaks the tetragonal lattice symmetry also induces changes in the magnetic easy axis near the critical regime of the AF/nematic phase transitions, indicating that the intrinsic electronic and magnetic properties of the system near $T_N/T_c$ are much different from na"ive expectations.

**Results**

Collinear magnetic order in twinned BaFe$_2$As$_2$. Without external uniaxial pressure, BaFe$_2$As$_2$ exhibits separate weakly first-order magnetic and second-order structural phase transitions ($T_s > T_N$) by $\sim 0.75$ K\textsuperscript{7}. The spins within each FeAs layer are collinear and arranged antiferromagnetically along the $a$-axis and ferromagnetically along the $b$-axis of orthorhombic structure with lattice parameters of $a$ and $b$, respectively ($a > b$). Along the out-of-plane direction, spins are arranged antiferromagnetically within one chemical unit cell (lattice parameter $c$), but have no net magnetic moment along the $c$-axis (Fig. 1a)\textsuperscript{6,7}. For a collinear Ising antiferromagnet with second-order (or weakly first order) magnetic phase transition, magnetic critical scattering with moments polarized along the longitudinal (parallel to the ordered moment or $a$-axis) direction should diverge at $T_N$, while spin fluctuations with moments polarized transverse to the ordered moment ($b$- and $c$-axis) direction should not diverge\textsuperscript{26-32}. Unpolarized\textsuperscript{33} and polarized\textsuperscript{34} neutron scattering experiments on strain-free BaFe$_2$As$_2$ confirm this expectation. While the longitudinal component ($M_a$) of the magnetic critical scattering, defined as low-energy spin fluctuations polarized along the $a$-axis direction, diverges at $T_N$, the transverse components $M_b$ and $M_c$

along the $b$- and $c$-axis, respectively (Fig. 1c, d), do not diverge at $T_N$.

**Fig. 1** Summary of the effect of uniaxial pressure on crystalline lattice and magnetic structures of BaFe$_2$As$_2$. a Crystal and AF structure of BaFe$_2$As$_2$. The red arrows indicate the $a$-axis direction of magnetic moments. b Magnetic order parameters measured at $Q_f = (1, 0, 3)$ and $Q_y = (0, 1, 3)$ under uniaxial pressure, revealing $T_N = 143$ K. The red dashed curve denotes the magnetic order parameter measured on a strain-free sample with $T_N = 136$ K. c Scattering geometry of polarized neutron scattering experiment in the $[H, 0, L]$ plane. d The reciprocal space, where the fluctuating moments along the $a$, $b$, and $c$-axis directions are marked as $M_a$, $M_b$, and $M_c$, respectively. e Spin arrangements of BaFe$_2$As$_2$ in the paramagnetic (left), near $T_N$ (middle), and low-temperature AF state. f Temperature dependence of the static ordered magnetic moment $M_a$ and $M_c$ as determined from $\sigma_{2f}^a$ at $(1, 0, 1)$ and $(1, 0, 3)$. The vertical error bars are estimated errors from fits-to-order parameters. g Comparison of temperature dependence of the strain-induced lattice distortion from ref. 27 and our estimated $M_a$ at $-20$ (blue solid line) and $-45$ (pink solid line) MPa.

**Effect of uniaxial pressure on lattice parameters of BaFe$_2$As$_2$.** The in-plane uniaxial pressure-induced tetragonal symmetry-breaking lattice distortion $\delta(P \neq 0) - \delta(P = 0)$, where $\delta = (a - b)/(a + b)$] has a Curie–Weiss temperature dependence in the
paramagnetic state and peaks near $T_N/T_s$, but is greatly suppressed below $T_N/T_s$ when the intrinsic orthorhombic lattice distortion of BaFe$_2$As$_2$ sets in (Fig. 1g)\textsuperscript{27}. In the paramagnetic state, NMR experiments on BaFe$_2$As$_2$ suggest that an in-plane uniaxial strain can induce a diverging $c$-axis polarized spin susceptibility $\chi_c$, which equals to $M_s$ in the zero energy limit, on approaching $T_N/T_s$\textsuperscript{26}. Since $c$-axis polarized low-energy spin fluctuations do not diverge around $T_N/T_s$ in the strain-free BaFe$_2$As$_2$\textsuperscript{34}, it is important to confirm the NMR results and determine if the diverging $\chi_c$ is a precursor of a new magnetic order with a component along the $c$-axis (Fig. 1f)\textsuperscript{28}.

Neutron polarization analysis of spin excitations in detwinned BaFe$_2$As$_2$. Our polarized neutron scattering experiments were carried out on the CEA CRG-IN22 triple-axis spectrometer equipped with Cryopad capability at the Institut Laue-Langevin and the BT-7 triple-axis spectrometer at the NIST Center for Neutron Research. The experimental setup for IN22 has been described in detail before\textsuperscript{34-39}, while polarized neutrons were controlled and analyzed using a polarized $^3$He filter on BT-7\textsuperscript{40}. We have also carried out unpolarized neutron diffraction experiments on BT-7 using an in-situ uniaxial pressure device\textsuperscript{25}.

The wave vector transfer $Q$ in reciprocal space in $\text{Å}^{-1}$ is defined as $Q = H \mathbf{a} + K \mathbf{b} + L \mathbf{c}$, with $a' = (2\pi/a)\mathbf{a}$, $b' = (2\pi/b)\mathbf{b}$, and $c' = (2\pi/c)\mathbf{c}$, where $a = b = 5.6\text{Å}$, $c = 12.96\text{Å}$, and $H, K, L$ are Miller indices. In this notation, the collinear AF structure of BaFe$_2$As$_2$ in Fig. 1a gives magnetic Bragg peaks at $(H, K, L) = (1, 0, L)$ with $L = 1, 3, \ldots$. The magnetic responses of the system at a particular $Q$ along the orthorhombic lattice $a$, $b$, and $c$-axis directions are marked as $M_a$, $M_b$, and $M_c$, respectively (Fig. 1a–d). The scattering plane is $[H, 0, L]$. The incident neutrons are polarized along the $Q$ $(x)$, perpendicular to $Q$ but in the scattering plane $(y)$, and perpendicular to both $Q$ and the scattering plane $(z)$ (Fig. 1c).

In this geometry, the neutron spin-flip (SF) scattering cross sections $\sigma_{SF}$ and $\sigma_{y}$ and $\sigma_{z}$ are related to the components $M_a$, $M_b$, and $M_c$ via $\sigma_{SF} = \sigma_{y} G_M = \sigma_{z} G_M + G_{M_b}$, $\sigma_{SF} = \sigma_{y} G_M + G_{M_a}$, and $\sigma_{SF} = \sigma_{z} G_M + G_{M_c}$, where $R$ is the flipping ratio ($R = \sigma_{SF}^M / \sigma_{SF}^M \approx 13$), $B$ is the background scattering, $M_y = \sin^2 \alpha M_a + \cos^2 \alpha M_c$ with $\alpha$ being the angle between $[H, 0, 0]$ and $Q$, and $M_c = M_0$ (Fig. 1d)\textsuperscript{34-39}.

Figure 1b compares the temperature dependencies of the $(1, 0, 3)$ magnetic Bragg peak for strain-free and strained BaFe$_2$As$_2$. At zero external pressure ($P = 0$ and strain-free), the magnetic scattering shows an order parameter like increase below $T_N = 136\text{K}$\textsuperscript{34}. When an uniaxial pressure of $P = 20\text{MPa}$ is applied along the $b$-axis of BaFe$_2$As$_2$, the Néel temperature of the sample increases to $T_N = 143\text{K}$\textsuperscript{35}. The vanishingly small magnetic scattering intensity at $Q = (0, 1, 3)$ suggests that the sample is essentially $-100\%$ detwinned (Fig. 1b).

Figure 2a, b shows the energy dependence of $\sigma_{SF}^M$, $\sigma_{y}^M$, and $\sigma_{z}^M$ in the zero pressure paramagnetic state at $T = 0.105T_N = 138\text{K}$ for magnetic positions $(1, 0, 1)$ and $(1, 0, 3)$\textsuperscript{34}. Figure 2c, d shows identical scans as those of Fig. 2a, b, respectively, in the paramagnetic state at $T = 0.104T_N = 145\text{K}$ with uniaxial pressure of $P = 20\text{MPa}$. Comparison of the Fig. 2a, c reveals that $\sigma_{SF}^M$ is clearly larger than $\sigma_{SF}^M$ below $-5\text{meV}$ in the strained sample. Since the $(1, 0, 3)$ peak corresponds to $Q_x = 23.4^\circ$ giving $M_y = 0.16M_a + 0.84M_c$ (Fig. 1d)\textsuperscript{34}, the increased $\sigma_{SF}^M$ in strained BaFe$_2$As$_2$ is mostly due to the increased $M_c$. The $(1, 0, 3)$ peak, which corresponds to $Q_z = 52.4^\circ$, $M_y = 0.63M_a + 0.37M_c$, and the scattering is therefore much less sensitive to strain-induced changes in $M_c$. To conclusively determine the effect of uniaxial pressure on $M_s$, we consider $\sigma_{SF}^M - \sigma_{SF}^M \propto M_y - M_c$. Since $M_b$ (or $\sigma_{SF}^M$) does not diverge at $T_N$ or change as a function of uniaxial pressure as seen in NMR\textsuperscript{26} and neutron polarization analysis (Fig. 2a–d), the effect of uniaxial pressure can be seen directly from the energy dependence of $\sigma_{SF}^M - \sigma_{SF}^M$ at $(1, 0, 1)$ (Fig. 2e) and $(1, 0, 3)$ (Fig. 2f). Without uniaxial pressure, $\sigma_{SF}^M - \sigma_{SF}^M$ does not diverge at the $Q_1 = (1, 0, 1)$ position but diverges at $Q_2 = (1, 0, 3)$ at low energies consistent with the expectation that spin fluctuations at $(1, 0, 1)$ are mostly probing $M_c$. With uniaxial pressure, we see clear divergence of low-energy spin fluctuations at $(1, 0, 1)$ below $-5\text{meV}$, thus unambiguously confirming the uniaxial pressure-induced $M_c$ enhancement around $T_N$ observed in NMR experiments\textsuperscript{26}. To further clarify the energy dependence of $M_a$, $M_b$, and $M_c$, we estimate these components from measurements at the $(1, 0, 1)$ and $(1, 0, 3)$ positions as described in ref. 34. By comparing the energy dependence of $M_a$, $M_b$, and $M_c$ in strain-free (Fig. 2g) and strained (Fig. 2h) BaFe$_2$As$_2$, we again see that the effect of uniaxial strain is to enhance $M_c$ below about $4\text{meV}$, consistent with the NMR measurements which probe $M_b$ or $\chi_c$ in the zero energy limit\textsuperscript{26}.

To further demonstrate the effect of uniaxial strain on the magnetic critical scattering of BaFe$_2$As$_2$, we show in Fig. 3 the temperature dependence of $\sigma_{SF}^M$, $\sigma_{y}^M$, and $\sigma_{z}^M$ at $E = 2\text{meV}$ for the strain-free (Fig. 3a, b)\textsuperscript{34} and strained (Fig. 3c, d) samples. At...
Effect of uniaxial pressure on static AF order of BaFe$_2$As$_2$. In principle, a diverging dynamic spin susceptibility in the paramagnetic state of a system is an indication of the eventual magnetic order below $T_N$\cite{26,32}. For strain-free BaFe$_2$As$_2$, the magnetic ordered moment is along the $a$-axis with no net moment along the $b$-axis and $c$-axis directions\cite{26,32}. Therefore, only the $M_a$ component of the spin susceptibility diverges at $T_N$ (Fig. 3e-g)\cite{34}. The observation of a diverging $M_a$ in strained BaFe$_2$As$_2$, in addition to the usual diverging $M_a$ (Fig. 3e, f, h), suggests that the applied strain may induce static magnetic ordered moment along the $c$-axis. To test this hypothesis, we carried out polarized neutron diffraction measurements on BaFe$_2$As$_2$ as a function of uniaxial pressure, focusing on the temperature and neutron polarization dependence of the scattering at $\mathbf{Q} = (1, 0, 0)$ with $L = 0, 1, 2, 3$, and 5. At wave vectors $(1, 0, 0)$ and $(1, 0, 2)$, there is no evidence of magnetic scattering, consistent with uniaxial pressure-free BaFe$_2$As$_2$ (see Supplementary information for additional data and analysis).

Figure 4a,b shows 8/28 scans of $\sigma_{SF}$ around $\mathbf{Q}_1 = (1, 0, 1)$ and $\mathbf{Q}_2 = (1, 0, 3)$, respectively, at different temperatures. Since $\sigma_{SF}$ at these two wave vectors probes different combinations of $M_a$ and $M_c$, one can obtain magnitudes of the static ordered moments along the $a$-axis and $c$-axis directions at these temperatures. Figure 4c shows the temperature dependencies of the full-width-at-half-maximum (FWHM) of these peaks, indicating that the spin-spin correlation lengths are instrumental resolution limited and temperature independent. Figure 4d plots the magnetic scattering intensity ratio between $(1, 0, 1)$ ($I_{101}$) and $(1, 0, 3)$ ($I_{103}$), which measures the relative strength of $M_a$ and reveals a clear peak around $T_N/T_f$.

To further determine the effect of uniaxial pressure on $c$-axis ordered moment and its pressure dependence, we carried out unpolarized neutron diffraction measurements focusing on the magnetic scattering intensity ratio between $(1, 0, 1)$ ($I_{101}$) and $(1, 0, 3)$ ($I_{103}$) using an in-situ uniaxial pressure device. Since our polarized neutron diffraction measurements revealed no ordered moment $M_a$, we used unpolarized neutron diffraction on BT-7 to improve the statistics of the data across $T_N$. Figure 4e compares the measured $I_{101}/I_{103}$ from 130 to 150 K at $P = 0$ and 45 MPa uniaxial pressure. Consistent with earlier work\cite{67,25}, $I_{101}/I_{103}$ is approximately temperature-independent across $T_N$ at $P = 0$, thus indicating that the internal strain of the system does not induce a $c$-axis ordered moment. Upon applying an uniaxial pressure of $P \approx 45$ MPa, the identical measurement shows a dramatic peak at $T_N$, thus confirming the results of Fig. 4a-d. Figure 4f shows the uniaxial pressure dependence of the measured $M_a/M_c$ at $T_N$, suggesting that the ordered $c$-axis moment saturates with increasing pressure above 45 MPa.

Figure 4g shows the temperature dependencies of the magnetically ordered moments along the $a$-axis ($M_a$) and $c$-axis ($M_c$) directions obtained by comparing $\sigma_{SF}$ and $\sigma_{AF}$ at wave vectors $\mathbf{Q}_1 = (1, 0, 1)$ and $\mathbf{Q}_2 = (1, 0, 3)$ (see Supplementary information for additional data and analysis). In the low-temperature AF ordered state, the strain-free and strained BaFe$_2$As$_2$ have the standard collinear AF structure with no evidence of $M_c$ (right panel in Figs. 1c and 4e)\cite{67}. On warming to 143 K just below $T_{SP}$ the easy axis tilts from the $a$-axis toward the $c$-axis with an angle of $\approx 28^\circ$ (middle panel in Fig. 1e). Finally, on warming to temperatures well above $T_N$, there is no static AF order (left panel in Fig. 1e).

Figure 1g shows the temperature dependence of $M_a$ at $\approx 20$ (blue solid line) and $\approx 45$ (pink solid line) MPa uniaxial pressure, compared with the uniaxial strain-induced lattice distortion $\delta (P = 20$ MPa) $- \delta (P = 0)$ (green solid circles and lines) obtained from neutron Larmor diffraction experiments\cite{27}. The similarity of the data suggests that the $c$-axis aligned magnetic moment arises from the uniaxial pressure-induced lattice distortion.

**Fig. 3** Temperature dependence of the magnetic scattering across $T_N$ at $E = 2$ meV without and with uniaxial pressure. Temperature dependence of $\sigma_{SF}$, $\sigma_{AF}$, and $\sigma_{AF}^P$ at $E = 2$ meV of a, b, uniaxial pressure-free and c, d, pressurized $P = 20$ MPa) BaFe$_2$As$_2$ at a, c (1, 0, 1) and b, d (1, 0, 3). e, f Comparison of $P = 0$ and $P \approx 20$ MPa ($\sigma_{SF}^P - \sigma_{SF}^0$) at $Q = (1, 0, 1)$ and $Q_c = (1, 0, 3)$. Temperature dependence of $M_a$, $M_c$, and $M_{zC}$ at $E = 2$ meV for g, uniaxial pressure-free and h, pressure sample estimated from the data in (a-d). The dotted and solid lines guide to the eye and the error bars represent 1 standard deviation. The vertical dashed and solid lines mark $T_N/T_f$, at $P = 0$ and $P \approx 20$ MPa, respectively.
Discussion
Theoretically, the in-plane electronic anisotropy of the iron pnictides is expected to couple linearly to the lattice orthorhombicity by the Ginzburg-Landau free-energy formalism if one ignores the effect of spin-orbit coupling induced magnetic anisotropy. From this perspective, in-plane uniaxial strain should only induce in-plane electronic anisotropy. The discovery of a c-axis ordered magnetic moment coupled exclusively with uniaxial pressure-induced lattice distortion suggests that such an effect cannot be only associated with the lattice orthorhombicity of the system, as $M_z$ becomes vanishingly small in the low-temperature orthorhombic phase with large in-plane lattice distortion. This is also different from the c-axis moment AF structure in Ba$_4$K$_2$Fe$_2$As$_2$ in the sense that the c-axis order appears exclusively in the tetragonal phase, while the c-axis moment appears in BaFe$_2$As$_2$ only near the peak of the nematic susceptibility around $T_N/T_c$. Although there is currently no theoretical understanding of this observation, it must arise from spin-orbit coupling induced magnetic anisotropy. Our discovery opens a new avenue to control magnetic order in nematic materials using mechanical strain instead of magnetic fields. The strong coupling of the c-axis aligned magnetic order with an in-plane pressure-induced lattice distortion offers the potential for the next generation of mechanical-strain-controlled magnetic switches. One must consider the presence of the magnetically ordered moment along the c-axis in mechanically detwinned iron pnictides in order to understand their intrinsic electronic, magnetic, and nematic properties.

Alternatively, our observations are also consistent with strain inducing a proximate XY spin anisotropy near $T_N/T_c$. In this scenario, while a-axis is energetically favorable in terms of spin anisotropy, c-axis is very close in energy. This allows for a distribution of large (resolution-limited but not long-range ordered) and long-lived (quasi-static) collinear magnetic domains, with their collinear spin direction guided by the domain walls. $I_{01}/I_{103}$ (Fig. 4d, e) is then a measure of the distribution of domains, reflective of the difference in spin anisotropy energies along the a- and c-axis. Similar to when the easy axis tilts from a-axis toward c-axis under strain (Fig. 1e), the change to a proximate XY spin anisotropy under strain also indicates a large and highly unusual effect of strain on the spin anisotropy.

In conclusion, we have used polarized and unpolarized neutron scattering to study the magnetic structure and critical scattering in uniaxial strained BaFe$_2$As$_2$. We find that the uniaxial pressure necessary to make single domain samples of BaFe$_2$As$_2$ also induces c-axis polarized critical magnetic scattering and static magnetic order around $T_N/T_c$. The size of the c-axis ordered moment is consistent with the uniaxial pressure-induced lattice distortion, in contrast with the lattice orthorhombicity. These results indicate that in addition to detwinning BaFe$_2$As$_2$, uniaxial pressure applied on the sample actually modifies the magnetic structure of the system. Therefore, infrared angle resolved photoemission, and Raman spectroscopy experiments on mechanically detwinned BaFe$_2$As$_2$ near the magnetic and nematic phases should be reexamined to take into account the effect of strain-induced change to the spin anisotropy on the in-plane electronic and magnetic properties.

Methods
Sample preparation and experimental details. BaFe$_2$As$_2$ single crystals were grown by the self-flux method using the same growth procedure as described before. Our polarized inelastic neutron scattering experiments were carried out using the CEA CRG-IN22 triple-axis spectrometer at the Institut Laue-Langevin, Grenoble, France (All raw data from ILL will be published at https://doi.org/10.5291/ILL-DATA.4-02-531, and from NCNR will be provided upon request). Polarized neutrons were produced using a focusing Heusler monochromator and analyzer with pyrolytic graphite monochromator and analyzer with pyrolytic graphite filter in the beam. The experiments on twinned BaFe$_2$As$_2$ without external uniaxial pressure were performed on ~12-g aligned single crystals as described before. The polarized elastic neutron scattering experiments were carried out using the CEA CRG-IN22 triple-axis spectrometer at the Institut Laue-Langevin, Grenoble, France (All raw data from ILL will be published at https://doi.org/10.5291/ILL-DATA.4-02-531, and from NCNR will be provided upon request). Polarized neutrons were produced using a focusing Heusler monochromator and analyzer with a focusing Heusler analyzer with a final wave vector $\text{bQ} = 2.662$ $\text{Å}^{-1}$. The experimental setups for uniaxial pressured and pressure freed experiments are identical. However, it is difficult to directly compare the scattering intensity of these two experiments since the sample masses, their relative positions in the beam, and background scattering of these two experiments are different. Nevertheless, one can safely compare the relative intensity changes of these two experiments. The polarized elastic neutron scattering experiments were carried out on BT-7 utilizing $^3$He polarizers immediately before and after the sample at NIST center for neutron research, Gaithersburg, Maryland, USA. The unpolarized neutron diffraction experiments in Fig. 4e were carried out using a pyrolytic graphite monochromator and analyzer with pyrolytic graphite filter in the beam. The experiments on twinned BaFe$_2$As$_2$ without external uniaxial pressure were performed on ~12-g aligned single crystals as described before. The polarized inelastic neutron scattering experiments on uniaxial pressured detwinned BaFe$_2$As$_2$ were performed using 12 pieces cut single crystals (~3 g, Fig. S1). The BT-7 measurements were carried out on a single piece of BaFe$_2$As$_2$ mounted on a newly built in-situ uniaxial pressure device and the neutron wave vectors are set at $k_x = k_y = 2.662$ $\text{Å}^{-1}$. 

![Fig. 4 Uniaxial pressure dependence of the magnetic order and correlations](image-url)
Determination of $M_{\alpha}$, $M_{\beta}$, and $M_{\gamma}$. In our previous polarized neutron scattering studies of iron pnictides, we have established the method for determining the spin-fluctuation components $M_{\alpha}$ ($\alpha = a, b, c$) along the lattice axes via comparing the SF scattering $\sigma^2 (y = x, z)$ at two equivalent magnetic wave vectors (such as $Q_1 = (1, 0, 1)$ and $Q_2 = (1, 0, 3)$, as shown in Fig. 1 of the main text). The definition of the directions $x, y,$ and $z$ is described in Fig. 1. $\sigma^2 (y = x, z)$ is directly related to the spin-fluctuation components by:

$$\sigma^2 (y = x, z) = F^2(Q) |\sin \alpha Q | a_{\alpha} M_{\alpha} + F^2(Q) |\cos \alpha Q | a_{\beta} M_{\beta} + B(Q),$$
$$\sigma^2 (y = x, z) = F^2(Q) |\sin \alpha Q | a_{\alpha} M_{\alpha} + F^2(Q) |\cos \alpha Q | a_{\beta} M_{\beta} + B(Q),$$

where $a$ is the angle between $(1, 0, 0)$ and $Q$ (Fig. 1), $L(Q)$ is magnetic form factor of Fe$^{3+}$, $R$ is the flipping ratio ($R = d^2_{\text{앵고}}/d^2_{\text{앵고}} \approx 13$), and $B$ is the polarization-independent background scattering.

From Eq. (1), we can get four equations for our results on $Q_1$ and $Q_2$:

$$\sigma^2 (y = x, z) = \frac{1}{2} F^2(Q) |\sin \alpha Q | a_{\alpha} M_{\alpha} + \cos \alpha Q | a_{\beta} M_{\beta} + B(Q),$$
$$\sigma^2 (y = x, z) = \frac{1}{2} F^2(Q) |\sin \alpha Q | a_{\alpha} M_{\alpha} + \cos \alpha Q | a_{\beta} M_{\beta} + B(Q),$$

in which $r$ is the intensity ratio factor between $Q_1$ and $Q_2$ to account for the differences in sample illumination volume and the convolution with instrumental resolution. The third and fourth equations in Eq. (2) can be used to determine the ratio $r$ and $M_{\beta}$, and the first two equations for $M_{\alpha}$ and $M_{\gamma}$. More details concerning the determination of the spin-fluctuation components $M_{\alpha}$, $M_{\beta}$, and $M_{\gamma}$ can be found elsewhere. Although this method can determine the values of $M_{\alpha}$, $M_{\beta}$, and $M_{\gamma}$, it also results in large error bars of their values. To more accurately determine the effect of uniaxial pressure on $M_{\alpha}$ and $M_{\gamma}$, we consider the differences between $\sigma^2 (y = x, z)$ at $Q_1$ and $Q_2$:

$$\sigma^2 (y = x, z) = \frac{1}{2} F^2(Q) |\sin \alpha Q | a_{\alpha} M_{\alpha} + \cos \alpha Q | a_{\beta} M_{\beta} + B(Q),$$

in which $M_{\alpha}$ does not diverge in uniaxial pressure and pressure-free cases. A comparison of $\sigma^2 (y = x, z)$ at $Q_1$ and $Q_2$ raw data should be most sensitive to changes in $M_{\alpha}$, while $\sigma^2 (y = x, z)$ at $Q_1$ and $Q_2$ should be sensitive to changes in both $M_{\alpha}$ and $M_{\gamma}$. The outcome of this analysis is shown in Figs. 2e, f and 3e, f.

In our polarized neutron diffraction experiment at BT-7, we have only measured $\sigma^2 (y = x, z)$ and $\sigma^2 (y = x, z)$. In elastic channel, $M_{\beta}$ is proportional to the square of the ordered moment ($m_\beta$). The determination of $M_{\beta}$ follows the same method as described in Eqs. (1) and (2). But we need to apply the Lorentz factor ($L = \frac{m_\beta}{m_\alpha}$) as we use the integrated intensity of $\theta = 2\theta$ to perform a scan to calculate $m_\beta$ for a magnetic field of $40$ K (Fig. 3d-3f). Similar to the $\theta = 2\theta$ scans of $\sigma^2 (y = x, z)$ as described in Fig. 4 of the main text, the scans for $\sigma^2 (y = x, z)$ (Fig. S5a, b) exhibit temperature-independent FWHM from 40 to 143 K (Fig. S5c), indicating that the spin–spin correlation length is resolution limited even in the pressure range ranging from 0 to 1.3 kbar. Figure S5d plots the ratio between the scattering intensity at (1, 0, 1) and (0, 1, 0) at $Q_1 = (1, 0, 3)$ and $Q_2 = (0, 1, 3)$, which is greatly enhanced close to $T_N$. Since $\sigma^2 (Q_1) = 0.16 M_{\alpha} + 0.08 M_{\beta} + 0.37 M_{\gamma}$ at $Q_1 = (1, 0, 3)$, the enhancement of $I_{Q_1}/I_{Q_2}$ is consistent with the emergence of a $c$-axis magnetic moment induced by uniaxial strain. At temperatures below $M_\beta$, the intensity ratio $I_{Q_1}/I_{Q_2} = 0.16 M_{\alpha} + 0.08 M_{\beta} + 0.37 M_{\gamma} \approx 0.5$ (black dashed line in Fig. S5d), where $\frac{m_\beta}{m_\alpha}$ accounts for the Lorentz factor. The data points of $I_{Q_1}/I_{Q_2}$ in Fig. S5d show that $M_{\beta}$ is absent at 149 K and below 135 K but reaches a maximum at 143 K close to $T_N$. The unpolared data in Fig. 4f show similar behavior. In addition to the emergence of $M_{\gamma}$, it is also important to understand whether $M_{\alpha}$ forms a new periodicity along $c$-axis. The magnetic structure factor of the three-dimensional AF order of BaFe$_2$As$_2$ shows magnetic peaks at (1, 0, L) with $L = 1, 3, 5, \ldots$ and the absence of magnetic scattering at (0, 1, 0) with $L = 0, 2, 4, \ldots$. If the induced $M_{\alpha}$ forms a larger magnetic unit cell along $c$-axis that ensures the presence of $(1, 0, 1)$ and $(1, 0, 3)$, one can expect detectable magnetic scattering at $L = 0, 2$. However, the three-point $\theta = 2\theta$ across $(1, 0, 2)$ in Fig. 5S shows that the intensity for $(1, 0, 2)$ is smaller than 1/3000 of $(1, 0, 3)$, which rules out this possibility and further confirm our conclusion about the canting-moment picture as shown in Fig. 1 of the main text.

Data availability

The data that support the findings of this study are available from the corresponding authors on request, and all raw data from ILL will be published at https://doi.org/10.5291/IILL-DATA.4-02-531, and from NCNR will be provided upon request.

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Author contributions
P.D. and X.L. conceived the project. P.L., M.L.K., L.T., G.T. and X.L. prepared the samples. Polarized inelastic neutron scattering experiments at IN22 were carried out by P.L., L.T., X.L., K.S., I.T.P., Y.L., Y.X.S., and F.B. Neutron diffraction measurements at BT-7 were carried out by M.L.K., Y.S., D.W.T., J.W.L., Y.Z., and R.J.B. The entire project was supervised by P.D. The paper was written by P.D., P.L., X.L., and Y.S. All authors made comments.

Competing interests
The authors declare no competing interests.

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