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Nematic Fluctuations in the Non-Superconducting Iron Pnictide BaFe_{1.9-x}Ni_{0.1}Cr_xAs₂

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The main driven force of the electronic nematic phase in iron-based superconductors is still under debate. Here, we report a comprehensive study on the nematic fluctuations in a non-superconducting iron pnictide system BaFe_{1.9-x}Ni_{0.1}Cr_xAs₂ by electronic transport, angle-resolved photoemission spectroscopy (ARPES), and inelastic neutron scattering (INS) measurements. Previous neutron diffraction and transport measurements suggested that the collinear antiferromagnetism persists to x = 0.8, with similar Néel temperature T_N and structural transition temperature T_s around 32 K, but the charge carriers change from electron type to hole type around x = 0.5. In this study, we have found that the in-plane resistivity anisotropy also highly depends on the Cr dopings and the type of charge carriers. While ARPES measurements suggest possibly weak orbital anisotropy onset near T_s for both x = 0.05 and x = 0.5 compounds, INS experiments reveal clearly different onset temperatures of low-energy spin excitation anisotropy, which is likely related to the energy scale of spin nematicity. These results suggest that the interplay between the local spins on Fe atoms and the itinerant electrons on Fermi surfaces is crucial to the nematic fluctuations of iron pnictides, where the orbital degree of freedom may behave differently from the spin degree of freedom, and the transport properties are intimately related to the spin dynamics.

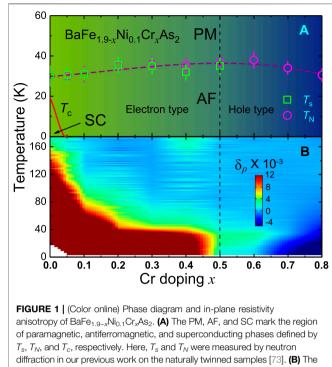
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1

1 INTRODUCTION

Electronic nematic phase breaks the rotational symmetry but preserves the translational symmetry of the underlying lattice in correlated materials [1-4]. In iron-based superconductors, the nematic order associated with a tetragonal-to-orthorhombic structural transition at temperature T_s acts as a precursor of the magnetic order below T_N and the superconducting state below T_c [5–10]. The nematic fluctuations can be described by the electronic nematic susceptibility, which is defined as the susceptibility of electronic anisotropy to the uniaxial in-plane strain [11]. Divergent nematic susceptibility upon approaching T_s from high temperature is revealed by the elastoresistance and elastic moduli measurements, suggesting nematic fluctuations well above T_s [12–16]. The nematic fluctuations commonly exist in iron-based superconductors and are even present in compounds with tetragonal crystal symmetry without any static nematic order [17]. Accumulating evidence suggests that the optimal superconductivity with maximum T_c usually occurs near a nematic quantum critical point where the nematic fluctuations are the strongest [18-29]. However, the charge, spin, and orbital degrees of freedom are always intertwined in the presence of nematic fluctuations [30-39], giving a twofold rotational (C_2) symmetry in many physical properties [5–11, 40-44] including anisotropic in-plane electronic resistivity and optical conductivity [45–51], lifting of degeneracy between d_{xz}/d_{yz} orbitals [52-58], anisotropic spin excitations at low energies [59-63], phonon-energy split in lattice dynamics [64, 65], and splitting of the Knight shift [66, 67]. In addition, it has been proposed that the local anisotropic impurity scattering of chemical dopants likely induces the twofold symmetry in the transport properties [68-70]. Such complex cases make it is difficult to clarify the main driven force of nematic phase by a single experimental probe.

Our previous works suggest that the Cr substitution is an effective way both to suppress the superconductivity and to tune the magnetism in iron-based superconductors [27, 71-73]. Specifically, in the BaFe_{1.9-x}Ni_{0.1}Cr_xAs₂ system, by continuously doping Cr to the optimally superconducting compound $BaFe_{1,9}Ni_{0,1}As_2$ with $T_c =$ 20 K, the superconductivity is quickly suppressed above x = 0.05, but the magnetic transition temperature T_N and the structural transition temperature T_s remain between 30 and 35 K as shown by neutron diffraction results on naturally twinned samples (Figure 1A). Moreover, the effective moment m is significantly enhanced first and then suppressed for dopings higher than x = 0.5, where the charge carriers change from electron type to hole type as shown by the sign of Hall and Seebeck coefficients [73]. These make $BaFe_{1,9-x}Ni_{0,1}Cr_xAs_2$ a rare example to separately tune the magnetically ordered temperature T_N by the local spin interactions and the magnetically ordered strength by the scattering of itinerant electrons on Fermi surfaces, respectively. The extra holes introduced by Cr substitutions compensate the electron doping thus may drive those nonsuperconducting compounds to a half-filled Mott insulator similar to the parent compounds of cuprate and nickelate superconductors [74-79]. It would be interesting to monitor



of paramagnetic, antiferromagnetic, and superconducting phases defined by T_s , T_N , and T_c , respectively. Here, T_s and T_N were measured by neutron diffraction in our previous work on the naturally twinned samples [73]. (B) The gradient color maps the in-plane resistivity anisotropy δ_ρ measured on detwinned samples. The vertical dashed line divides the regions for electron-type and hole-type charge carriers as determined by the sign of Hall and Seebeck coefficients [73].

the evolution of the nematic fluctuations starting from a metallic state toward to a localized insulating state [79–81], especially on the detwinned samples (**Figure 1B**).

In this paper, we further report a multi-probe study on the nematic fluctuations in the non-superconducting compounds $BaFe_{1.9-x}Ni_{0.1}Cr_xAs_2$ (x = 0.05 ~ 0.8) by electronic transport, angle-resolved photoemission spectroscopy (ARPES), and inelastic neutron scattering (INS) measurements. The in-plane resistivity anisotropy measured in the detwinned samples under uniaxial pressure shows a strong dependence on the Cr content with a clear sign change above x = 0.6. By focusing on two compounds with x = 0.05 and 0.5, ARPES measurements suggest possible band shifts induced by orbital anisotropy near T_s/T_N for both dopings, but INS experiments reveal clearly different behaviors on the spin nematicity. The onset temperature of low-energy spin excitation anisotropy between Q = (1, 0, 1)and Q = (0, 1, 1) for x = 0.05 is about 110 K, but for x = 0.5, it is much lower, only about 35 K near the magnetic transition. Such temperature dependence of spin nematicity is consistent with the results of in-plane resistivity anisotropy. At high energies, the spin nematicity for x = 0.05 extends to about 120 meV, much larger than the case for x = 0.5 (about 40 meV), suggesting a possible linear correlation between the highest energy scale and the onset temperature of spin nematicity. Therefore, the nematic behaviors in iron pnictides are highly related to the interplay between local moments and itinerant electrons. While the C2-type anisotropies in spin excitations and

in-plane resistivity are strongly correlated with each other [60], the orbital anisotropy induced band splitting may behave differently as affected by the complex Fermi surface topology [82–91].

2 EXPERIMENT DETAILS

High-quality single crystals of BaFe_{1.9-x}Ni_{0.1}Cr_xAs₂ were grown by the self-flux method [71-73, 92-95]; the characterization results of our sample can be found in previous reports [71, 73]. The crystalline directions of our sample were determined by an X-ray Laue camera (Photonic Sciences) in the backscattering mode with incident beam along the c – axis. After that, the crystals were cut into rectangle shapes (typical sizes: 1 mm × 2 mm) by a wire saw under the directions $[1, 0, 0] \times [0, 1, 0]$ in orthorhombic lattice notation (a = b = 5.6 Å). By applying a uniaxial pressure around 10 MPa, the crystal can be fully detwinned at low temperature, where the direction of pressure was defined as the *b* direction, and the pressure-free direction was defined as the *a* direction [60–63, 96–99]. The in-plane resistivity $(\rho_{a,b})$ was measured by the standard four-probe method with the Physical Property Measurement System (PPMS) from Quantum Design. To compare the temperature dependence of resistivity at different directions, we normalized the resistivity $\rho_{a,b}(T)$ data at 150 K for each sample. The in-plane resistivity anisotropy was defined by $\delta_{\rho} = (\rho_b - \rho_a)/(\rho_b + \rho_a)$ same as other literature [45–47].

ARPES experiments were performed at beamline 10.0.1 of the Advanced Light Source and beamline 5-4 of the Stanford Synchrotron Radiation Light source with R4000 electron analyzers. The angular resolution was 0.3°, and the total energy resolution was 15 meV. All samples were cleaved insitu at 10 K and measured in ultra-high vacuum with a base pressure lower than 4×10^{-11} Torr. We note that we used twinned samples without uniaxial pressure for the ARPES experiments. INS experiments were carried out at two thermal triple-axis spectrometers: PUMA at Heinz Maier-Leibnitz Zentrum (MLZ) [100], Germany, and TAIPAN at the Australian Centre for Neutron Scattering (ACNS) [101], ANSTO, Australia. The wave vector **Q** at (q_x, q_y, q_z) was defined as $(H, K, L) = (q_x a/2\pi, q_y)$ $q_v b/2\pi$, $q_z c/2\pi$) in reciprocal lattice units (r.l.u.) using the orthorhombic lattice parameters $a \approx b = 5.6$ Å and $c \approx 13$ Å. All measurements were done with fixed final energy $E_f = 14.8$ meV, and a double focusing monochromator and analyzer using pyrolytic graphite crystals. To gain a better signal-noise ratio, eight pieces of rectangularly cut crystals (typical sizes: $7 \text{ mm} \times$ 8 mm \times 0.5 mm) were assembled in a detwinned device made by aluminum and springy gaskets [60–63]. To reach both Q = (1, 0, 0)1) and Q = (0, 1, 1), the sample holder was designed to easily rotate by 90°, thus the scattering plane can switch from $[H, 0, 0] \times$ [0, 0, L] to $[0, K, 0] \times [0, 0, L]$. The total mass of the crystals used in INS experiments was about 2 g from each sample set of x = 0.05and x = 0.5. Time-of-flight neutron scattering experiments were carried out on the same sample sets at 4SEASONS spectrometer (BL-01) at J-PARC [102, 103], Tokai, Japan, with multiple incident energies $E_i = 250, 73, 34, 20 \text{ meV}, k_i$ parallel to the c axis, and chopper frequency f = 250 Hz. The data were only corrected by the efficiency of detectors from the incoherent scattering of vanadium with white beam. As we were comparing two samples with similar mass under the same measured conditions at the same spectrometer, it was not necessary to do the vanadium normalization with mono-beam. The data were analyzed by the Utsusemi and MSlice software packages [104, 105].

3 RESULTS AND DISCUSSIONS

We first present the resistivity results in Figures 1-3. Apparently, the in-plane resistivity anisotropy show a strong dependence on the Cr doping level. In the Cr free sample BaFe_{1.9}Ni_{0.1}As₂, the difference between ρ_a and ρ_b presents above the superconducting transition temperature $T_c = 20$ K, where ρ_a is metallic and ρ_b is semiconducting-like with an upturn at low temperature (namely, $\rho_a < \rho_b$) (Figure 2A). The superconductivity is completely suppressed at x = 0.05, and there is a dramatic difference between ρ_a and ρ_b with an anisotropy δ_o persisting to about T = 110 K (**Figure 2B**). By further increasing Cr doping, both ρ_a and ρ_b become semiconducting-like even insulating-like above x = 0.1, and the resistivity anisotropy gets weaker and weaker, until it nearly disappears at x = 0.5 and 0.6 compounds (Figure 2C-H). For those high doping compounds x = 0.7and 0.8, it seems that δ_{ρ} changes sign with $\rho_a > \rho_b$ at low temperatures (Figure 2I,J). To clearly compare the resistivity anisotropy upon Cr doping, we plot δ_{ρ} as gradient color mapping in Figure 1B and show its detailed temperature dependence in **Figure 3**. Interestingly, the sign of δ_{ρ} is also related to the type of charge carriers. δ_{ρ} keeps strong and positive in the electron-type compounds but changes to negative and weak (<1%) in the hole-type compounds (Figure 1B and Figure 3B). This is consistent with the results in the electron doped $BaFe_{2-r}(Ni)$, $Co)_x As_2$ and the hole doped $Ba_{1-x} K_x Fe_2 As_2$, $Ca_{1-x} Na_x Fe_2 As_2$, and BaFe_{2-x}Cr_xAs₂ systems [45-48, 106-109]. However, in those cases, the onset temperature of δ_{ρ} decreases with the structural transition temperature T_s when increasing the doping level from the non-superconducting parent compounds to optimally doped superconducting compounds. Here, in the BaFe_{1.9-x}Ni_{0.1}Cr_xAs₂ system, both T_N and T_s are actually within the range 32 ~ 35 K for all probed dopings [73], but the onset temperature of δ_{ρ} still extends to high temperatures, and it is then strongly suppressed by Cr doping (**Figure 3A**). In those hole-type compounds, δ_{ρ} shows a peak feature (for x = 0.5 and 0.6) or a kink (for x = 0.7 and 0.8) responding to the magnetic and structural transitions (Figure 3B). The non-monotonic behavior of δ_{ρ} may come from the competition between the scattering from hole bands and electron bands, and similar behaviors were observed in the nematic susceptiblity of the Cr doped $BaFe_2(As_{1-x}P_x)_2$ system [27].

Next, we focus on the electronic structure and the spin excitations in two typical dopings x = 0.05 with $T_N = 32$ K and x = 0.5 with $T_N = 35$ K. The Fermi surface topology and band structure measured by ARPES on naturally twinned samples are shown in **Figure 4**. From the Fermi surface mapping in **Figure 4A,B**, we can find typical hole pockets

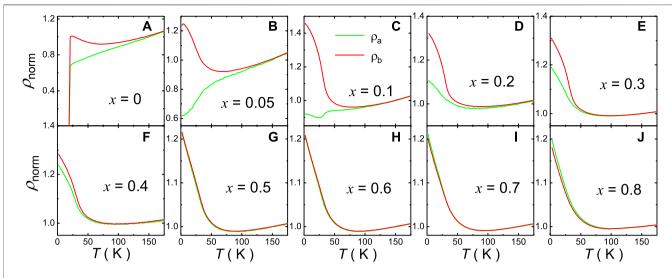
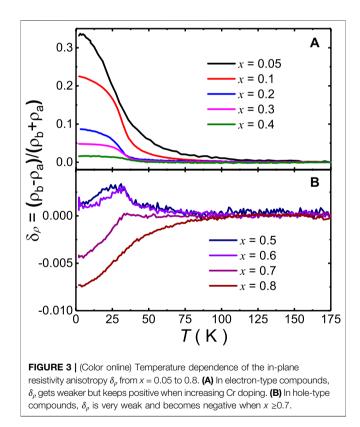


FIGURE 2 (Color online) In-plane resistivity anisotropy of BaFe_{1.9-x}Ni_{0.1}Cr_xAs₂ under uniaxial pressure. Here, ρ_b is the in-plane resistivity along the direction of uniaxial pressure, and ρ_a is the in-plane resistivity perpendicular to the direction of uniaxial pressure. For easy comparison, each curve is normalized by its resistivity at 150 K, and there is no resistivity anisotropy above this temperature.



around the zone center Γ point. Near the *X* point, an electron pocket is observed for x = 0.05. For x = 0.5, however, the Fermi surface resembles that of the hole-doped (Ba,K)Fe₂As₂ [53]. This is due to the hole doping introduced by the Cr substitution, which also introduces disorder directly in the Fe-planes, thus resulting in spectral features that appear broad [82]. **Figure 4C,D** show the

second energy derivatives of the spectral images along the high symmetry direction (Γ -X). Larger hole pockets can indeed be seen for x = 0.5 compared to x = 0.05. As has been demonstrated previously on BaFe₂As₂, NaFeAs, and FeSe, the onset of T_s is associated with the onset of an observed anisotropic shift of the d_{xz} and d_{vz} orbital-dominated bands where the d_{xz} band shifts down and the d_{vz} band shifts up [52-56]. This shift is most prominently observed near the X point of the Brillouin zone. Moreover, such band splitting as measured on uniaxially strained crystals can be observed above T_s in the presence of this symmetry-breaking field. On a structurally twinned crystal, the anisotropic band shifts would appear in the form of a band splitting due to domain mixing. While we do not observe clearly the band splitting as shown in Figure 4C,D, we can clearly observe the lower branch with dominant intensity that shifts with temperature. This can be understood as the lower d_{xz} band. We can fit the energy position of the band extracted from the X point and plot as a function of temperature. The temperature evolution clearly identifies a temperature scale associated with an onset of the band shift [53–55]. As shown in Figure 4E,F, the X band shifts at low temperature $T \approx 25$ K for x = 0.05 and $T \approx 45$ K for x = 0.5, respectively, closing to their structural or magnetic transition temperatures. We do note that while we cannot conclusively state that this represents the orbital anisotropy, the behavior we observe here on these twinned crystals is consistent with the expectation of the onset of orbital anisotropy [52, 57, 62]. We note here that the observed onset temperature of band splitting is close to the T_s (or T_N), in contrast to the much higher onset in the resistivity anisotropy shown in Figure 3 measured on a strained crystal.

We then turn to search the connection between the resistivity anisotropy and the spin excitation anisotropy. The first evidence of spin nematicity was observed in $BaFe_{2-x}Ni_xAs_2$ (x = 0, 0.065, 0.085, 0.10, 0.12) [60–63], where $BaFe_{1.9}Ni_{0.1}As_2$

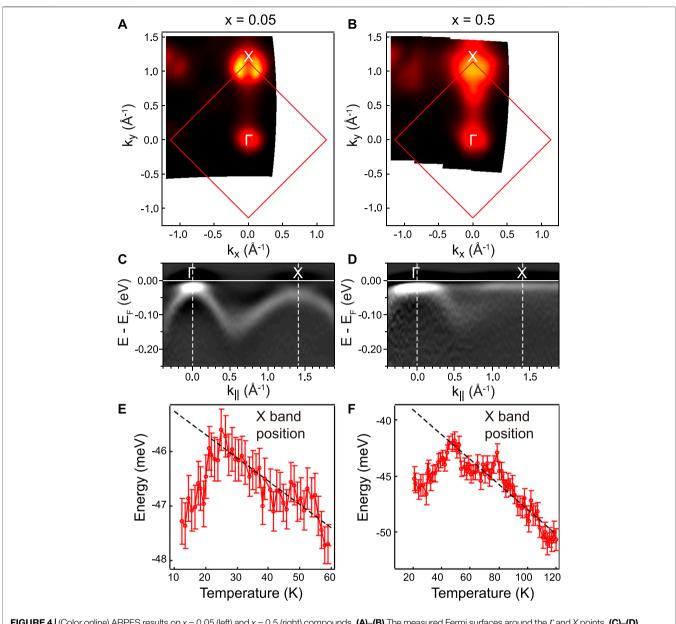


FIGURE 4 (Color online) ARPES results on x = 0.05 (left) and x = 0.5 (right) compounds. (A)–(B) The measured Fermi surfaces around the Γ and X points. (C)–(D) Band dispersions along the high symmetry direction Γ -X obtained from the second derivatives in the energy direction. (E)–(F) Temperature dependence of the fitted band position from the X point. All dashed lines are guides for eyes.

is the starting compound of this study. Low-energy spin excitations are measured on the detwinned $BaFe_{1.9-x}Ni_{0.1}Cr_xAs_2$ (x = 0.05 and 0.5) samples by INS experiments using two triple-axis spectrometers. The results of constant-energy scans at E = 3, 6, 9, and 12 meV are summarized in Figure 5. With convenient design of the detwinned device and sample holder, we can easily perform constant-energy scans (Q - scans) either along the [H, 0, 1] or [0, K, 1] direction after rotating the whole sample set by 90°. For the x = 0.5 sample, we instead do the S1 rocking scans at Q = (1, 1)0, 1) and (0, 1, 1). It should be noticed that the Néel temperature T_N is slightly enhanced by the applied uniaxial pressure in the x = 0.05 sample from 32 to 40 K (so does T_s) but does not change for the x = 0.5 sample ($T_N \approx T'_N = 35$ K) (**Figure 6A,B**). Such an effect has been detected in the BaFe_{2-x}(Ni, Co)_xAs₂ system [110]. The detwinned ratio can be estimated by comparing the integrated intensities of magnetic Bragg peak between Q = (1, 0, 1) and Q = (0, 1, 1) positions, which is about 10:1 for the x = 0.05 samples, and 4:1 for the x = 0.5 samples, respectively. Such a large ratio means successful detwin for both sample sets. At the first glance, it is very clear for the difference of the spin excitations between Q = (1, 0, 1) and Q = (0, 1, 1)especially at low temperatures, which could be attributed to the spin Ising-nematic correlations (so-called spin nematicity).

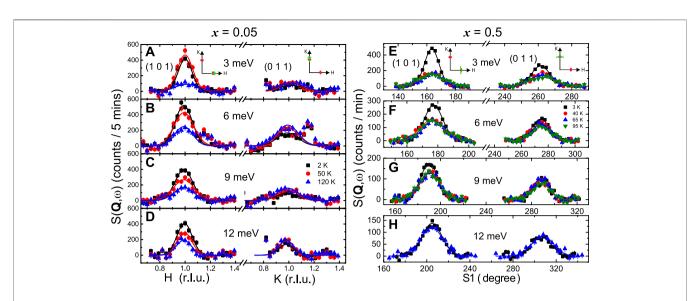


FIGURE 5 (Color online) Inelastic neutron scattering results on the spin excitations of uniaxially detwinned samples for x = 0.05 (left) and x = 0.5 (right) compounds measured by two triple-axis spectrometers TAIPAN and PUMA. We compared the constant-energy scans (Q –scans along [H,0,1] or [0, K,1], S1 rocking scans at Q = (1,0,1) or (0, 1, 1)) for E = 3, 6, 9, 12 meV, respectively. All data are corrected by a linearly Q –dependent background, and the solid lines are Gaussian fittings. The spurious signals in 6 meV data are ignored.

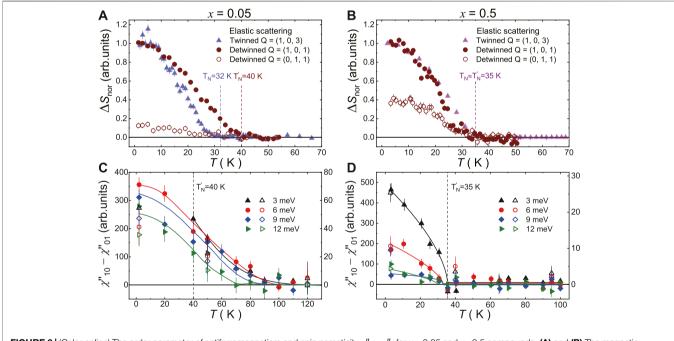


FIGURE 6 (Color online) The order parameter of antiferromagnetism and spin nematicity $\chi_{10}^{\prime\prime} - \chi_{01}^{\prime\prime}$ for x = 0.05 and x = 0.5 compounds. (A) and (B) The magnetic order parameters measured at Q = (1,0,3) on twinned samples, Q = (1,0,1) and Q = (0,1,1) on detwinned samples by elastic neutron scattering. All data are subtracted by the normal state background and normalized by the intensity at base temperature for Q = (1,0,3) or Q = (1,0,1). (C) and (D) Spin nematicity measured by inelastic neutron scattering. The solid symbols are the differences of local susceptibility χ'' between Q = (1,0,1) and Q = (0,1,1) (left *y*-axis), and the open symbols are similar but obtained by integrating the constant-energy scans in **Figure 5** corrected by the Bose population factor (right *y*-axis). The vertical dash lines mark the magnetic transition temperature T_N on twinned samples and T_N' on detwinned samples. All solid lines are guides to eyes.

After warming up to high temperatures, the spin excitations at Q = (1, 0, 1) decrease and become nearly identical to those at Q = (0, 1, 1). The nematic order parameter for the spin system

can be approximately represented by $\chi''_{nematic} = \chi''_{10} - \chi''_{01}$, in which χ''_{10} (or χ''_{01}) is the local spin susceptibility at Q = (1, 0, 1) (or Q = (0, 1, 1)). **Figure 6C,D** show the temperature

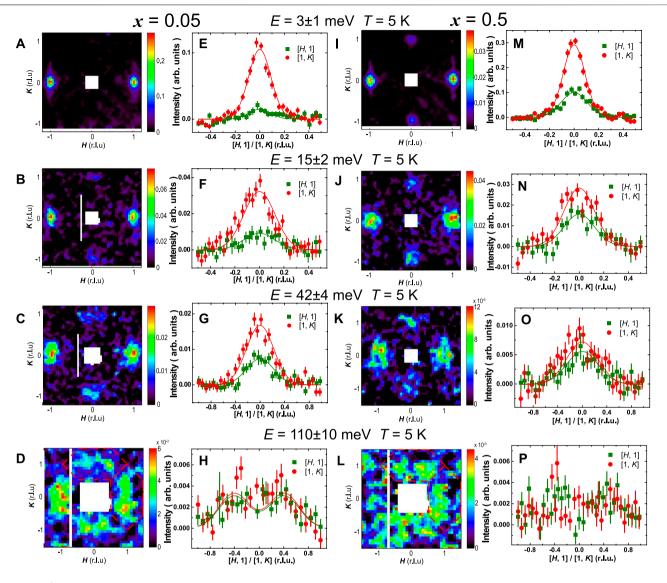
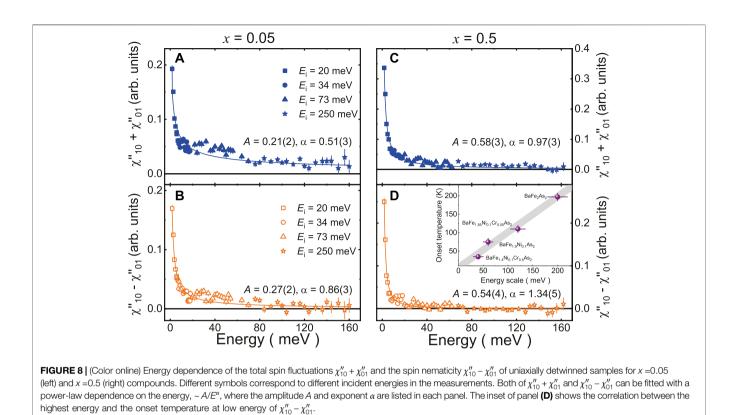


FIGURE 7 | (Color online) Inelastic neutron scattering results on the spin excitations of the uniaxially detwinned samples for x = 0.05 (left) and x = 0.5 (right) compounds measured at T = 5 K by time-of-flight spectrometer 4SEASONS. All data are presented in both 2D slices for the [*H*, *K*] plane and 1D cuts along [*H*,0] or [0, *K*] at typical energy windows $E = 3\pm1,15\pm2,42\pm4,110\pm10$ meV. The solid lines are Gaussian fittings guiding for eyes, which are not shown in panel (**P**) due to poor data quality. The dashed diamonds in panel (**D**) and (**L**) illustrate the integrated Brillouin zone for spin excitations around Q = (1,0) and Q = (0,1), respectively.

dependence of $\chi_{10}'' - \chi_{01}''$ for both compounds, where the Bose population factor is already corrected. We also plot the data (open symbols) obtained from the integrated intensity of those Q - scans in **Figure 5**. For the x = 0.05 compound, the spin nematicity decreases slightly upon increasing energy and terminates well above $T'_N = 40$ K (**Figure 6C**)⁷³. For the lowest energy we measured (3 meV), the onset temperature of spin nematicity is about 110 K, similar to the in-plane resistivity anisotropy in **Figure 3A**. The results for x = 0.5compound show markedly differences, where $\chi_{10}'' - \chi_{01}''$ quickly decreases with both energy and temperature, and the onset temperature is around $T'_N = 35$ K (**Figure 6D**). No spin anisotropy can be detected above 40 K for both Q - scans

and energy scans, and this is also consistent with the very weak in-plane resistivity anisotropy for x = 0.5 (**Figure 3B**). The spin nematic theory predicts that the nematic fluctuations enhance both the intensity and the correlation length of spin excitations at $(\pi, 0)$ but suppress those at $(0, \pi)$ even above T_s . This was firstly testified in the detwinned BaFe_{1.935}Ni_{0.065}As₂ and can also be seen here in **Figure 5**⁶¹. Although the peak intensities at Q = (1, 0, 1) seem stronger than those at Q = (0, 1, 1) in **Figure 5G,H**, the peak width is smaller, and the integrated intensity of the *Q*-scans are closed to each other. The above results of spin nematicity in BaFe_{1.9-x}Ni_{0.1}Cr_xAs₂ (x = 0.05 and 0.5) resemble to those in BaFe_{2-x}Ni_xAs₂, where spin excitations at low energies change from C_4 to C_2 symmetry in the tetragonal



phase at temperatures approximately corresponding to the onset of the in-plane resistivity anisotropy.

Moreover, INS experiments on detwinned BaFe2As2 and BaFe1.9Ni0.1As2 suggest that the spin anisotropy can persist to very high energy [62, 63], even in the later case the splitting of the d_{xz} and d_{yz} bands nearly vanishes [57]. To quantitatively determine the energy dependence of spin excitation anisotropy, we have performed time-offlight INS experiments on the uniaxially detwinned $BaFe_{1.9-x}Ni_{0.1}Cr_xAs_2$ (x = 0.05 and 0.5), and the results are shown in Figures 7, 8. It should be noted that for such experiments, the energy transfer is always coupled with L due to $k_i \parallel c$ [102, 103]. The twodimensional (2D) energy slices and one-dimensional (1D) cuts along [H, 0] and [0, K] at various energies are presented in Figure 7. Indeed, the spin excitations are twofold symmetric below 100 meV for both compounds. The spin excitations at E = 3 meV, $Q = (0, \pm 1)$ are very weak in the x = 0.05 compound, then continuously increase upon energy, and become nearly the same as $Q = (\pm 1, 0)$ around 110 meV. For the x = 0.5 compound, although the spin excitations at $Q = (0, \pm 1)$ can be initially observed at E = 3 meV, the spin anisotropy still exists at 15 meV and then disappears above 42 meV. To further compare the spin excitations in both compounds, we have calculated the total spin fluctuations $\chi_{10}'' + \chi_{01}''$ and the spin nematicity $\chi_{10}'' - \chi_{01}''$ from the integrated intensity marked by the dashed diamonds in Figure 7D,L. In principle, the local dynamic susceptibility χ'' can be estimated from the integration outcome of the spin excitations within one Brillouin zone, and here χ'' can be simply calculated through dividing the integration signal in the Q = (0, 0) (1, 1), (2, 0), (1, -1) boxes, giving the diamond shape integration zone [8]. The total spin susceptibility χ_{10}'' + χ_{01}'' in the x = 0.5 compound is stronger than that in x = 0.05 but

decays much quickly with energy (Figure 8A,C). The spin nematicity $\chi_{10}'' - \chi_{01}''$ apparently has different energy scales for two compounds, where it is about 120 meV for x = 0.05 but only 40 meV for x = 0.5, respectively. The energy scale of $\chi_{10}'' - \chi_{01}''$ in the superconducting compound BaFe1.9Ni0.1As2 is 60 meV [62], and for the parent compound BaFe2As2, it is about 200 meV up to the band top of the spin waves [63]. These facts lead to a possible linear correlation between the highest energy and the onset temperature of spin nematicity at low energy (inset of **Figure 8D**). Within the measured energy range, both $\chi_{10}'' + \chi_{01}''$ and $\chi_{10}'' - \chi_{01}''$ can be fit with a power-law dependence on the energy, ~ A/E^{α} , where the amplitude A and exponent α are listed in each panel of Figure 8. Indeed, the larger value of α for x = 0.5 in comparison to that for x =0.05 suggests faster decay with energy for both the spin fluctuations and the spin nematicity. Similar fitting on the results of BaFe_{1.9}Ni_{0.1}As₂ gives parameters in between them [62]. Although the low energy data below 10 meV may be affected by the L-modulation of spin excitations, and by the superconductivity in BaFe_{1.9}Ni_{0.1}As₂, the similar quantum critical behavior both for $\chi_{10}'' + \chi_{01}''$ and $\chi_{10}'' - \chi_{01}''$ in these three compounds is expected by the Ising-nematic scenario [60-63].

In our previous neutron diffraction results on the BaFe_{1.9-x}Ni_{0.1}Cr_xAs₂ system, the Cr dopings have limited effects on the magnetically ordered temperature T_N but significantly enhance the effective ordered moment *m* by reaching a maximum value at x = 0.5 [73]. The Néel temperature T_N is mostly determined by the local magnetic coupling related to the local FeAs₄ tetrahedron structure. The evolution of ordered moment probably induced by the changes of the density of states and the orbital angular momentum from itinerant electrons on the Fermi surfaces. The Cr doping

introduces both local distortion on the lattices and hole doping on the Fermi pockets, yielding a non-monotonic change of the conductivity of charge carriers. As shown in **Figure 2**, the low-temperature upturn of resistivity is enhanced by Cr doping first but then weakens in those hole-type compounds. Among these dopings, x = 0.5 has the most insulating-like behavior, and thus strongly localized charge carriers and maximum ordered moment, but its spin nematicity quickly drops down for both the temperature and energy dependence. In contrast to the magnetically ordered strength, both the structural transition temperature T_s and the lattice orthorhombicity $\delta = (a - b)/(a + b)$ are nearly Cr doping independent [73]. This means the static nematic order is also nearly Cr independent in this system, as opposed to the case for dynamic nematic fluctuations.

The nature of the iron-based superconductor can be theoretically described as a magnetic Hund's metal, in which the strong interplay between the local spins on Fe atoms and the itinerant electrons on Fermi surfaces gives correlated electronic states [80, 81]. Indeed, time-of-flight INS experiments on the detwinned BaFe2As2 suggest that the spin waves in the parent compound are preferably described by a multi-orbital Hubbard-Hund model based on the itinerant picture with moderate electronic correlation effects, instead of a Heisenberg model with effective exchange couplings from local spins. Upon warming up to high temperatures, the intensities of spin excitation anisotropy decrease gradually with increasing energy and finally cut off at an energy away from the band top of spin waves [63]. Therefore, the energy scale of spin nematicity sets an upper limit for the characteristic temperature for the nematic spin correlations, as well as the onset temperature of resistivity anisotropy. Here, by adding up the results on the in-plane anisotropies of resistivity, orbital energy, and spin excitations in BaFe_{1.9-x}Ni_{0.1}Cr_xAs₂, they clearly suggest that the electronic nematicity is intimately related to the spin dynamics, which seems consistent with Hund's metal Specifically, by doping Cr to suppress picture. the superconductivity in BaFe_{1.9}Ni_{0.1}As₂, it makes the charge carriers initially localized with enhanced electron correlations [73], which may enhance the electronic correlations by increasing the intra- and inter-orbital onsite repulsion U as well as Hund's coupling J_H [63], and thus gives rise to stronger spin excitations and larger spin anisotropy in the Cr doping x = 0.05 compound. Another effect is the lifting up of d_{yz} and d_{xy} along the Γ -X direction to the Fermi level, which primarily contributes to the effective moments [80]. The orbital-weight redistribution triggered by the spin order suggests that the orbital degree of freedom is coupled to the spin degree of freedom [111]. By further increasing Cr doping to x = 0.5, the localization effect is so strong that the electron system becomes insulating at low temperature. In this case, the itinerant picture based on Hund's metal may not be applicable anymore. The low density of itinerant electrons weakens the nematic fluctuations and probably limits them inside the magnetically ordered state. In either case for x =0.05 or x = 0.5, the band splitting does not directly correspond to the spin nematic correlations but only present below the nematic ordered temperature. This may attribute to the weak spin-orbit coupling in this system, as the spin anisotropy in spin space can only present at very low energies [59]. In addition, our results can rule out the picture of local impurity scattering driven nematicity since the impurity scattering from Cr substitutions is certainly stronger in the x = 0.5 compound, but it does not promote the nematic fluctuations.

4 CONCLUSION

In conclusion, we have extensively studied the in-plane resistivity anisotropy, orbital ordering, and spin nematicity in a nonsuperconducting $BaFe_{1.9-x}Ni_{0.1}Cr_xAs_2$ system. We have found that the Cr doping strongly affect the anisotropy of resistivity and spin excitations along with the itinerancy of charge carriers. While the onset temperatures of resistivity anisotropy and spin nematicity are similar and correlated with the energy scale of spin anisotropy, the orbital anisotropy shows an onset temperature irrelevant to them. These results suggest that the electronic correlations from the interplay between local moments and itinerant electrons are crucial to understand the nematic fluctuations, thus inspiring the quest for the driven force of the electronic nematic phase in iron-pnictide superconductors.

DATA AVAILABILITY STATEMENT

The datasets presented in this article are not readily available because the datasets are currently private. Requests to access the datasets should be directed to HL, hqluo@iphy.ac.cn.

AUTHOR CONTRIBUTIONS

HL and DG proposed and designed the research. DG, TX, WZ, and RZ contributed in sample growth and resistivity measurements. MY, MW, S-KM, MH, DL, and RB contributed to the ARPES measurements. DG and HL carried out the neutron scattering experiments with SD, GD, XL, JP, KI, and KK. DG, HL, SL, and PD analyzed the data. HL, DG, and MY wrote the paper. All authors participated in discussion and comment on the paper.

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are performed at thermal triple-axis spectrometer PUMA at Heinz Maier-Leibnitz Zentrum (MLZ), Germany, thermal triple-axis spectrometer TAIPAN at Australian Centre for Neutron Scattering (ACNS), Australian Nuclear Science and Technology Organisation (ANSTO), Australia (Proposal No. P4263), and time-of-flight Fermi-chopper spectrometer 4SEASONS (BL-01) at the Materials and Life Science Experimental Facility of J-PARC (Proposal Nos. 2015A0005, 2016A0169). ARPES measurements were performed at the Advanced Light Source and the Stanford Radiation Lightsource, which are both operated by the Office of Basic Energy Sciences, U.S. DOE.

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