Strong Coupling of Superconductivity and Quantum Nematic Fluctuations in BaFe$_{2-x}$Ni$_x$As$_2$

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There are increasing experimental evidences for the presence of nematic correlations in iron pnictide superconductors, where the in-plane electronic properties break the rotational symmetry of the underlying lattice. However, the role of nematic fluctuations in the formation of superconductivity is still unclear. Using a device that can continuously apply a uniaxial pressure on a thin sample, we are able to identify the nematic correlations throughout the superconducting dome by measuring the resistivity anisotropy in BaFe$_{2-x}$Ni$_x$As$_2$. The strong coupling between the superconductivity and nematic fluctuations is demonstrated by the nematic-like behavior of superconductivity under weak uniaxial pressure and the coincidence of the maximum $T_c$ and the nematic quantum critical point. Our results suggest that understanding the nematic fluctuations is essential to obtain the superconductivity in the iron-based superconductors.
In many cuprates and iron-based superconductors, it has been found that the rotational symmetry of the corresponding in-plane properties is broken due to the presence of the nematic order $^{1,2,3,4}$. The nematic correlations may intertwine with other orders such as superconductivity, antiferromagnetism (AF) and charge-density wave (CDW) that lead to many exotic properties in these materials$^{5,6}$. While the origin of the nematic correlations may be material dependent, it is suggested that quantum nematic fluctuations may induce attractive pairing interaction and thus enhance or even lead to superconductivity $^{7,8,9,10,11}$. As a result, superconductivity may exhibit strong nematic signatures in response to the uniaxial pressure. It is therefore essential to reveal such responses in order to establish the nature of superconductivity related to the quantum nematic fluctuations. In the absence of pseudogap and CDW, the iron-based superconductors provide an arguably good opportunity for studying the interplay between superconductivity and nematicity.

Most of the studies on the nematic order in the iron-based superconductors have focused on the BaFe$_2$As$_2$ and its doped compounds$^{12,13,14,15,16,17,18}$, where the establishment of the nematic phase is below a tetragonal-to-orthorhombic structural transition at $T_s$ and above an AF transition at $T_N < T_s$ $^3$. The presence of twinning domains in the orthorhombic phase requires a uniaxial pressure to detwin the sample to reveal the in-plane anisotropic properties in the nematic phase. The uniaxial pressure is also essential in detecting the nematic fluctuations above $T_s$, which persist above the optimal doping level, suggesting the presence of a nematic quantum critical point (QCP) $^{13}$. However, due to the quick decease of nematic signal with increasing doping, there is currently no study of nematic fluctuations in the heavily overdoped samples, which is
indispensable to understand the role of nematic fluctuations in the establishment of superconductivity.

In this paper, we use a new piezoelectric device to study the coupling between the superconductivity and nematic fluctuations in the BaFe$_{2-x}$Ni$_x$As$_2$ system by measuring the resistivity of the samples. In the overdoped regime, the unprecedented high resolution of our device allows us to observe nematic fluctuations up to very high doping levels that overshadow the whole superconducting dome, as shown in Fig. 1. Detailed analysis of the nematic susceptibility reveals a typical funnel shape for the nematic QCP that coincides with the optimal doping of maximum $T_c$. Most importantly, we find strong nematic response of the superconductivity to weak external uniaxial pressure even in the overdoped regime where no nematic order presents, demonstrating an intimate connection between the superconductivity and nematic quantum fluctuations in iron pnictides.

Starting from the long-range antiferromagnetism in undoped BaFe$_2$As$_2$, the Ni-doped sample shows superconductivity above $x \sim 0.04$ while both the antiferromagnetic and tetragonal-to-orthorhombic structural transitions vanish above $x \sim 0.08$\textsuperscript{19,20,21}. A simple phase diagram with doping is shown in Fig. 1. The resistivity anisotropy under constant pressure has been shown to be similar to that in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$\textsuperscript{14}.

The piezoelectric device to measure the resistivity under the uniaxial pressure is shown in Fig. 2a. Due to the large magnitude of the elastoresistivity coefficients in the iron-based superconductors\textsuperscript{22}, exceptional attention should be paid to avoid the residual strain caused by the glue \textit{etc.}, which may cause artificial effects and significantly reduce the measurement accuracy. There are two advantages of our setup: first, the part of the
sample within the electrical contacts is free of glue; second, a “zero” pressure should be achieved at some point with the movement of the piezoelectric bender since it can continuously change from pressing to stretching the sample (Fig. 2b). These two advantages make it possible to obtain very high resolution to measure the resistivity change under uniaxial pressure without suffering the influence of the built-in strain introduced by glue or thermal expansion of the frames etc. Moreover, it means that the results shown in this paper are intrinsic in the limit of zero pressure, which avoids any possible artificial effect from large pressure that may drive the system deep away from the equilibrium state.

Fig. 2c gives the results for the parent compound BaFe$_2$As$_2$ at different temperatures. Along with the change of pressure by tuning the voltage applied on the piezoelectric bender, the resistivity along the tetragonal (110) direction shows a positive pressure dependence. At high temperature, a fusiform hysteresis behavior is found due to the ferroelastic properties of the piezoelectric ceramic (supplementary materials). The hysteresis loop becomes ferromagnetism-like below $T_s$, which has also been observed in other lightly underdoped samples. Fig. 2d plots the temperature dependence of the normalized area $S_N$, defined as the area of the hysteresis loop divided by the minimum rectangular area that contains it. The value of $S_N$ decreases abruptly at $T_s$, suggesting that it is directly related to the order parameter of the nematic phase. In a ferromagnetic material, the hysteresis loop of the magnetization versus the magnetic field comes from the change of the magnetic domains. Similarly, the hysteresis loop observed here should come from the nematic domains, which clearly demonstrates the role of uniaxial pressure as an external field to the nematic order.
The response of the resistivity under the uniaxial pressure in the absence of the nematic order gives a measure of the nematic fluctuations. Similar to Ref. (13), one may use the slope of the resistivity change above $T_s$ to study the temperature dependence of the nematic fluctuations, i.e. $d(\Delta R/R_0)/dV \propto d\psi/dp$ along the (110) direction where $\psi$ and $p$ are the resistivity anisotropy and pressure, respectively. While the fusiform hysteresis makes it hard to obtain the precise pressure dependence of the resistivity, its average slope is still a good approximation to $d\psi/dp$. Here, we define $\zeta$ as $d(\Delta R/R_0)/dp$ to represent the nematic susceptibility. Fig. 2e and 2f give the temperature dependence of $\zeta_{(110)}$ at different doping levels. One can immediately see that the behaviors in the underdoped and overdoped regimes are distinctly different. For the samples with $x \leq 0.096$, we can fit the high-temperature data with a simple Curie-Weiss-like function $\zeta_{(110)} = A/(T-T') + y_0$, where $A$ and $y_0$ are temperature-independent constants (supplementary materials). $T'$ corresponds to the mean-field nematic transition temperature.

On the other hand, in the over-doped samples, we see for the first time the presence of nematic fluctuations in the highest doping level measured ($x = 0.24$) where superconductivity has already disappeared. The temperature dependence of $\zeta_{(110)}$ in these samples shows a broad hump peaked at $T_h$. The doping dependence of $T'$ and $T_h$ is shown in Fig. 1. The zero value of $T'$ indicates the disappearance of the nematic order, suggesting the presence of a nematic QCP at the optimal doping level. The zero value of $T_h$ extrapolated linearly from higher doping also happens at the QCP, indicating that it is the crossover temperature that is associated with the nematic disordered phase. We note that $T_h$ may be related to the $T*$ obtained in the thermal expansion measurement$^{23}$, suggesting that the strong coupling between the lattice and electronic system may persist.
for the whole doping range shown here. The doping dependence of the $T'$ and $T_h$ thus constitutes the funnel feature commonly found in a QCP system.

The above results suggest an intimate connection between the superconductivity and the nematic QCP. If nematic fluctuations indeed play a key role in the formation of the superconductivity, one may expect additional nematic responses from the superconductivity itself under the uniaxial pressure. Fig. 3a and 3c give the voltage dependence of the resistivity change along the (110) direction around $T_c$ for the underdoped $x = 0.068$ and overdoped $0.104$ samples, respectively. The change of the resistivity in the middle of the transition ($T = 16.9$ K) for the $x = 0.068$ sample is much larger than that above $T_c$ ($T = 18.9$ K), suggesting that the former is not due to the residual nematic signal from the part of the sample that is not superconducting yet. Moreover, the sign of the slope changes below $T_c$ ($T = 18.5$ K) for the $x = 0.104$ sample, clearly demonstrating it is directly related to the superconducting transition. Taking the values of the resistivity at +40 V and -40 V, we plot the temperature dependence of the resistivity for these two samples in Fig. 3b and 3d. Clearly, the slope of the voltage dependence of the resistivity corresponds largely to the change of $T_c$ under uniaxial pressure, with $dT_c/dp \propto (dR/dp)/(dR/dT)$, where $dR/dT$ is almost the same for different pressures.

The nematic behavior of the superconductivity is best seen by comparing Fig. 3c and Fig. 3e where the same measurement on the $x = 0.104$ sample is shown but along the (100) direction. A striking difference is that the pressure dependence of the resistivity along the (100) direction at the middle of the superconducting transition is almost linear while that along the (110) direction shows significant different slopes at the high positive
and negative voltages. Since there is only very weak resistivity anisotropy along the (100) direction above $T_c$, Fig. 3e represents an isotropic effect on the superconductivity that is the same for both negative and positive pressures. On the other hand, the variation of the slopes with pressure along the (110) direction should be attributed to a nematic effect on the superconductivity under the uniaxial pressure. As discussed above, the different pressure dependence of $dR/dV$ along the (100) and (110) directions suggests different pressure dependence of $dT_c/dp$, as sketched in the inset of Fig. 1. This may be better understood in comparison with the underdoped $x = 0.068$ sample (Fig. 3a) which resides underneath the nematic order. In the presence of nematic domains, the resistivity at the positive and negative high voltages (i.e., the positive and negative pressures) is thus measured along the $b_O$-dominant and $a_O$-dominant directions, respectively, where $b_O$ and $a_O$ represents the orthorhombic domains along the orthorhombic $b$ and $a$ axes, respectively. Since the slopes at the two sides are different, the values of $dT_c/dp$ are also different along the orthorhombic $a$ and $b$ axes. This may not be surprising since the underlying lattice also breaks the four-fold symmetry. However, the fact that this asymmetry of $dT_c/dp$ persists in the heavily overdoped samples (Fig. 3c) where neither the nematic phase nor orthorhombic structure presents is truly unexpected. In particular, the difference of $dT_c/dp$ along the (110) direction at the high positive and negative voltages for the $x = 0.104$ sample is about 1.11 mK/MPa, which is even larger than that along the (100) direction, 1.08 mK/MPa, suggesting that the superconductivity has exhibited strong nematic responses under weak uniaxial pressure. This can only be understood if there exists strong coupling between the superconductivity and nematic fluctuations. In cuprates, it has also been shown that the effects of pressure on $T_c$ along
the orthorhombic a and b axes are different\textsuperscript{24,25} while either stripes or nematic phase has been found\textsuperscript{26,27}, although it is not clear yet whether the latter plays an important role in the former or not.

The average value of $\frac{dT_c}{dp}$ is studied by simply comparing the temperature dependence of the resistivity at +40V and -40V as shown in Fig. 3b and 3d. Fig. 3f gives the $T'$ dependence of the average $\frac{dT_c}{dp}$ along the (110) direction around the optimal doping level. The sign change of $\frac{dT_c}{dp}$ in the underdoped ($T'>0$) and overdoped ($T'<0$) samples is consistent with the previous reports indirectly obtained by the thermal expansion and specific heat measurements\textsuperscript{28,29}. Therefore, we conclude that $\frac{dT_c}{dp}_{(110)}$ becomes zero near the nematic QCP despite the errors in our measurements and the mean-field nature of $T'$. The sign change of $\frac{dT_c}{dp}$ around the optimal doping suggests that the superconducting dome is pushed to higher doping under uniaxial pressure while $T_c$ at the optimal doping remains unchanged for sufficiently small pressure. This further demonstrates that the nematic QCP coincides with the maximum $T_c$ and the two are closely related. On the other hand, the nematic QCP in cuprates seems to be located at the slightly overdoped doping level\textsuperscript{27,30,31}, probably due to the presence of the pseudogap and other orders.

References


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Author contributions

Z. L. made the device and performed the measurements. S. L. conceived the idea and designed the experiments. Y. G., W. Z., X. M., X. Z., J. Z., C. R. L. S. X. Q. helped the measurements. D. G., W. Z., T. X., R. Z., P. D., H. L. grew the crystals. Y. Y. gave theoretical suggestions. S. L. wrote the manuscript.

Fig. 1. Phase diagram of BaFe$_{2-x}$Ni$_x$As$_2$. The superconductivity (SC) happens below the solid orange line. The color map shows the doping and temperature dependence of the nematic fluctuations $\zeta_{(110)}$ in the logarithmic scale, which persist well above the boundary of superconductivity at the overdoped side. The green circles, blue diamonds and red hexagons represent the actual structural transition temperature $T_s$, the fitted nematic transition temperature $T'$ and the hump temperature $T_h$, respectively. The error bars of the $T'$ and $T_h$ are determined from the fitting error and artificially set to 20 K, respectively. The
nematic QCP is located at 0.088 where several values of T’ are obtained from the samples coming from the same batch, suggesting the nematic QCP is very sensitive to the actual doping level. In the presence of uniaxial pressure along the (110) direction, the resistivity shows ferromagnetic-like hysteresis loop in the nematic ordered phase, as shown in the inset. The response of superconductivity is seen from the pressure dependence of superconducting transition temperature $T_c$, as sketched in the right lower inset. While $dT_c/dp$ changes little with pressure along the (100) direction (solid green line), it is different at the negative and positive pressures along the (110) direction (red dashed line), suggesting an asymmetric response of superconductivity to the uniaxial pressure that is intimately related to nematic fluctuations.

**Fig. 2. Measurement of nematic fluctuations.** a, The uniaxial pressure device, which is composed of a BeCu frame and a piezoelectric bender. The movement of the bender’s top (blue arrow) towards and away from the left frame corresponds to the pressing and stretching the sample, respectively. b, Top view of the Fe-As block and sketch of the measurement. The tetragonal axes $a_T$ and $b_T$ are along the next-nearest Fe-Fe direction. Below the structural transition temperature $T_s$, stretching and pressing the sample along the tetragonal (110) direction above the saturation pressure will measure the resistivity along the orthorhombic $a_O$ and $b_O$ axes, respectively. c, The voltage dependence of $\Delta R/R_0$ of BaFe$_2$As$_2$ along the (110) direction at several temperatures. Increasing and decreasing the voltage correspond to increasing and decreasing the pressure,
respectively. Here, \( R_0 \) is the average resistivity at zero voltage during increasing and decreasing the voltage process and we have \( \Delta R = R(V) - R_0 \). The asymmetric shape at low temperature could come from either the different pressure dependence of the resistivity along different orthorhombic axes or the residual strain that causes the zero pressure apart from zero voltage. d, The temperature dependence of the normalized hysteresis area \( S_N \) in the parent compound, which drops dramatically at \( T_s \). e & f, The temperature dependence of nematic susceptibility \( \zeta_{(110)} \) at different doping levels. The solid lines in E are fitted by the Curie-Weiss-like function as described in the main text. The arrows in F indicate the positions of the humps.

Fig. 3. Superconductivity response to the uniaxial pressure. a & c, The change of the resistivity with voltage along the (110) direction around the superconducting transition for the underdoped \( x = 0.068 \) and overdoped \( x = 0.104 \) samples, respectively. Three temperatures represent the data just above \( T_c \), in the middle of the superconducting transition and close to the zero-resistance temperature, respectively. b & d, Temperature dependence of the resistivity at +40 V and -40 V along the (110) direction transferred from the voltage scans. The vertical short bars indicate the temperatures of the data in a and c. e, The change of resistivity with voltage along the (100) direction around the superconducting transition for the overdoped \( x = 0.104 \) sample. The inset shows the transferred temperature dependence of resistivity at +40 V and -40 V.
f, $T'$ dependence of average $dT_c/dp$. The zero value of $T'$ and $dT_c/dp$ correspond to the nematic QCP and maximum $T_c$, respectively.
Fig. 2

a) Sample placement on a Piezo-Bender BeCu Frame.
b) Crystal structure of Fe and As atoms with specific axes and planes.

(c) Graph showing the change in resistance ($\Delta R/R_0$) with voltage for different temperatures (130 K to 295 K) for BaFe$_2$As$_2$.

(d) Graph showing the evolution of $T_s$ with temperature for BaFe$_2$As$_2$.

(e) Graph showing the evolution of $\zeta_{\tau(110)}$ with temperature for BaFe$_2$As$_2$.

(f) Graph showing the evolution of $\zeta_{\tau(110)}$ with temperature for BaFe$_2$As$_2$. 

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METHODS

Sample preparation

The BaFe$_{2-x}$Ni$_x$As$_2$ samples were grown by the self-flux method that has been reported elsewhere$^1$. The actual doping $x$ and the nominal doping $x'$ has a relationship of $x \approx 0.8 x'$. While we have used the actual doping in this paper, some of the neutron scattering works from our group have used the nominal doping. The crystal orientation is determined by x-ray Laue method and cut into thin plate along the desired direction by the diamond wire saw.

Uniaxial pressure device

The photo of the uniaxial pressure device is shown in Fig. 1a. Here we give some further information about the device and the resistivity measurement.

The displacement of the piezo-bender without any load is 0.6$\mu$m/V. Since the (100) direction has no nematic signal, any negative slope of $\Delta R/R_0$ should come from the change of the sample length due to the pressure. Since no negative slope has been observed along the (100) direction as shown later in Fig. S4, the relative movement of the length should be smaller than 1$\times$10$^{-4}$. We note that this value also set an upper limit of the strain induced in our measurement. Considering the length of the sample between the two voltage contacts is just several millimeters, the actual movement of the piezo-bender from the +40 V to -40 V should be smaller than 0.1 $\mu$m while it would be about 50 $\mu$m if the piezo-bender were free. Therefore, we conclude that the movement of the piezo-bender in our measurement is negligible so that the voltage applied to the piezo-bender has almost completely transferred to the external force. However, the force is transferred through glue, which makes the actual force applied on the sample smaller than the ideal blocking force. We have used another uniaxial pressure device with spring to calibrate our device. The pressure $p$ is thus calculated by $p = F/S$, where $F = 0.01383$ (N/V) $\times V$ and $S$ are the force and sectional area of the sample, respectively.

The resistivity has been measured by the four-point probe method with either a combination of a DC current source and a nanovoltmeter or a lock-in amplifier. In the case of DC measurement, the current is not reversed during the measurement to obtain high-quality data. Since the thermoelectric potential is at least 3 orders lower than the signal and does not change at a fixed temperature, it has negligible effect on the relative change of the resistivity with the change of voltage.

Due to the ferroelastic nature of the piezoelectric ceramic, the resistivity shows a fusiform hysteresis loop with the change of voltage. Fig. S1 gives the temperature dependence of the normalized area $S_N$ of such loop in several samples. Here $S_N$ is defined as the area of the hysteresis loop divided by the minimum rectangular area that contains it. While little temperature dependence is found at high temperature, its value decreases with decreasing temperature below 100 K.

The ferroelasticity of the piezo-bender also affects the determination of the slope of the pressure dependence of the resistivity. Fig. S2a shows that the value of the slope becomes smaller for smaller voltage range. Using the slope of the whole data to define $\Delta R/R_0$, which effectively equals to the slope of the line connecting the maximum and minimum points, we can plot the temperature dependence of $d(\Delta R/R_0)/dp$ as shown in Fig. S2b. While the values of $A$ and $y_0$ fitted by Eq. (1) may change due to the different way of defining $d(\Delta R/R_0)/dp$, the effects on the fitted temperature $T''$ and the ratio of –
$y_0/A$ are within the error bars. For example, for the particular data shown in Fig. S2b, the difference of $-y_0/A$ is less than 7%, while that of the fitted $T'$ is less than 6 K. More importantly, such difference only depends on the piezo-bender and is similar to all the samples, which will not alter our main results. In the main text, we have used the data with $\pm40$V range to obtain higher resolution.

The reliability of our device is further illustrated in Fig. S3, where the doping dependence of $-y_0/A$ is given (see main text). While the individual values of $A$ and $y_0$ may be affected by the strength of the glue, the way obtaining the slope and the cross-section area measured etc., the value of $-y_0/A$ decreases linearly with increasing doping for $x \leq 0.096$ except for the $x = 0.088$ samples, which may be influenced by the nematic QCP. It is beyond the scope of this paper to discuss the origin of $y_0$, but the linear doping dependence of $-y_0/A$ clearly demonstrates the reliability and advantages of our device.

**Results along the (100) direction**

We have measured the pressure dependence of the resistivity along the (100) direction. In the parent compound and heavily overdoped samples, no pressure dependence has been found as shown in Fig. S4a and S4b, respectively, i.e., $\zeta_{(100)} = 0$. However, it becomes non-zero around the optimal doping level. Fig. S4c gives the temperature dependence of $\zeta_{(100)}$ at different doping levels. Since there is no proper way to compare the data between the (110) and (100) directions, we simply give the ratio of $\zeta_{(100)}/\zeta_{(110)}$ at 30 K in Fig. S4d, which roughly suggests that anisotropy between these two directions is minimum at $x = 0.088$. It seems that the nematic QCP may be the key to understand the non-zero value of $\zeta_{(100)}$.

**Fig. S1** The effect of the intrinsic hysteresis of the piezo-bender. The normalized area $S_N$ shows little temperature dependence above 100 K but quickly decreases with decreasing temperature below 100 K.

**Fig. S2** The influence of the voltage range on the determination of the slope of $\Delta R/R_0$. **a,** The voltage dependence of $\Delta R/R_0$ for the $x = 0.068$ sample at 55 K. The voltage range is from $\pm 10V$ to $\pm 40V$. **b,** The temperature dependence of $d(\Delta R/R_0)/dp$ for the $x = 0.068$ sample with the voltage range of $\pm 40V$ (black square) and $\pm 10V$ (red circle).

**Fig. S3** Doping dependence of $-y_0/A$.

**Fig. S4 a & b,** The voltage dependence of $\Delta R/R_0$ along the (100) direction for the $x = 0$ and $x = 0.24$ samples, respectively. **c,** The temperature dependence of $\zeta_{(100)}$ at different dopings. **d,** Doping dependent of the ratio of $\zeta_{(100)}/\zeta_{(110)}$ at 30 K, which shows a peak around $x = 0.088$. The red line is guided to the eye.
Fig. S2

(a) $\Delta R/R_0 \times 10^{-2}$ vs Voltage (V) with $x=0.068$ and $T=55$ K. Lines represent different voltage values: ±40 V (solid), ±30 V (green), ±20 V (blue), ±10 V (red).

(b) $\zeta_{(110)} \times 10^{-8}$ MPa$^{-1}$ vs $T$ (K) with $x=0.068$. Symbols represent ±40 V (black) and ±10 V (red).
Fig. S3

![Graph showing a linear relationship between $-Y_0/A$ and Doping x]
Fig. S4