Evolution of the nematic susceptibility in LaFe$_{1-x}$Co$_x$AsO

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The identification of electronic nematicity across series of iron-based superconductors raises the question of its relationship with superconductivity and other ordered states. Here, we report a systematic elastoresistivity study on LaFe$_{1-x}$Co$_x$AsO single crystals, which have well separated structural and magnetic transition lines. All crystals show Curie-Weiss-like nematic susceptibility in the tetragonal phase. The extracted nematic temperature is monotonically suppressed upon cobalt doping, and changes sign around the optimal doping level, indicating a possible nematic quantum critical point beneath the superconducting dome. The amplitude of nematic susceptibility shows a peculiar double-peak feature. This could be explained by a combined effect of different contributions to the nematic susceptibility, which are amplified at separated doping levels of LaFe$_{1-x}$Co$_x$AsO.

Unconventional superconductivity is intimately related to other electronic symmetry breaking states. A common feature among unconventional superconductors is that by varying tuning parameter $x$, such as pressure or chemical substitution, the superconductivity exists under a dome-like region in the $T - x$ phase diagram, and the extrapolation of some transition line hits zero temperature inside or close to that dome [1–3]. Formation of a spin density wave is the most recognized order in the normal state of unconventional superconductors, and its ubiquitousness stimulated theories to explain the origin of unconventional superconductivity mediated by fluctuations of the magnetic order [4–9]. Recently, the report of newly recognized electronic orders beside magnetism in unconventional superconductors is infectious [10]. Along with others, the electronic nematic order is currently under the spot light.

Strictly speaking, the term electronic nematicity denotes an electronic state that spontaneously breaks the rotational symmetry of its host crystal [10]. The existence of such a counterintuitive electronic state was first experimentally demonstrated in the two-dimensional electronic gas system [12]. In the case of iron-based superconductors, the $C_2$ order of electrons only exists when the crystal structure is in the orthorhombic ($C_2$) phase. However, because the electronic properties acquire very large anisotropy above the antiferromagnetically ordered state regardless of the small lattice anisotropy, the paramagnetic orthorhombic region in the phase diagram is still regarded as an electronic nematic state [13].

All ordered states of iron-based compounds are highly intertwined, making it hard to find the leading instability among them. In the tetragonal phase, these orders melt into pertinent fluctuations. Due to the existence of finite electronic-lattice coupling, the electronic nematic fluctuation can be probed by various approaches, such as lattice softening and elastoresistivity measurements [14–16]. Chu et al. introduced a method to probe the elastoresistivity in the small strain limit [17–18]. By adopting this technique, the divergent nematic susceptibility has been found in different families of iron-based compounds [19–21]. Remarkably, by driving the system to optimal doping level, the amplitude of nematic susceptibility becomes enhanced [19–20]. The divergence of such fluctuation clearly rules out a structural instability as the driving force for other transitions [16–17].

One prevailing understanding of the nematicity is to treat it as the result of fluctuating anti-ferromagnetic (AFM) order [22–25]. This scenario received strong support from the discovery of good scaling between lattice softening and magnetic fluctuating amplitude in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ over a wide doping range [26]. However, nematicity can also be the resultant of different orbital occupancy [27–31]. The orbital scenario gained its experimental favoritism mainly from experimental studies of FeSe [32–33]. As illustrated in Fig. 1, the phase diagram of iron-based superconductors is diverse. The structural and magnetic transition lines are very close to each other in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$. The extension of these lines penetrate into the superconducting dome. In stark contrast, there is no static magnetic order over the whole phase diagram of FeSe$_{1−x}$S$_x$, while the structural transition still exists and can be suppressed with doping. Recent studies on the newly synthesized LaFe$_{1-x}$Co$_x$AsO single crystals revealed the material’s structural and magnetic transitions being well separated [33–36]. Superconductivity in LaFe$_{1-x}$Co$_x$AsO emerges after the total suppression of the AFM order, making the phase diagram distinct from the aforementioned cases [35–37].

In this letter, by utilizing elastoresistivity measurements, we report the persistence of a Curie-Weiss-like nematic susceptibility deep in the tetragonal phase of LaFe$_{1-x}$Co$_x$AsO. The Curie-Weiss temperature changes sign around the optimal doping level $x = 0.06$, with an enhanced nematic susceptibility, similar to other iron-
Temperature

(a)

(b)

(c)

Dopant concentration

-2
-1
0
1
2
0
8
20
60
200
300

T = 150 K

V

P ZT

x = ΔLx / Lx (x 104)

ε

η = ΔP / L

0
1
2
3
4
5
6
7
8
9
10

T (K)

χnem

/ (χnem − χ0)

/ 1000

(χnem − χ0) (T − T nem) / 1000

LaFeAsO

Parent compound

0
1
2
0
8
20
60
200
300

T = 150 K

V

0
1
2
3
4
5
6
7
8
9
10

ε (x 100)

η (x 100)

T (K)

FIG. 1: (Color online). Schematic T – x phase diagrams of three representative iron based superconductors [20] [35] [37]: (a) Ba(Fe1-xCox)2As2, (b) FeSe1-xSx, (c) LaFe1-xCoxAsO. The orange area is the AFM ordered state, the green dome is the superconducting state, and the solid blue line shows the structural transition (with dashed line as a guide to the eye). The nematic order exists below the structural transition line and above the AFM region.

FIG. 2: (Color online). Elastoresistivity measurements of the parent compound LaFeAsO. (a) The relative change of strain (εx = Δlx / lx) and sample resistivity (η = Δρ / ρ) according to the voltage applied to the piezo actuator at a fixed temperature T = 150 K. The strain was measured along the current direction, which was aligned against the [110] axis of the crystal. (b) The relationship between εx and η at some representative temperatures. The nematic susceptibility was obtained from χnem = (−δη / δεx) at the εx = 0 limit. (c) The temperature dependence of χnem is shown as blue open squares in the upper panel. The structural transition temperature determined in Ref. [31] is indicated by the green arrow. The red curve represents a Curie-Weiss fitting to data in the temperature range between the vertical dotted lines. The inverse nematic susceptibility (x n em − x 0)−1 and the Curie constant C = (x n em − x 0)(T − T n em) are shown in the lower panel, indicating the fitting quality.

based superconductors [17] [19] [20]. Besides, another enhancement of the nematic susceptibility is resolved around x = 0.04, close to the end point of the AFM order.

Elastoresistivity measured along the [110] direction of LaFeAsO single crystal is shown in Fig. 2. By changing the voltage across a piezo actuator, one can precisely tune the uniaxial strain applied to the sample glued on the actuator’s edge. In the nematic ordered state, the resistivity anisotropy of the in-plane axes is very large [38, 39]. This anisotropy represents the electronic nematicity. Due to the electron-lattice coupling, strain can induce resistivity anisotropy above the nematic ordered state if nematic fluctuation exists [17]. As illustrated in Fig. 2 (b), the fractional change of sample resistivity (η = Δρ / ρ) shows perfect linear relationship to the strain (εx = Δlx / lx) applied along the current direction. As demonstrated in the references [17] [20], the slope of η(εx) curve probes the nematic susceptibility of the sample. The negative slope means the in-plane resistivity of LaFeAsO is higher along the shorter orthorhombic direction. For simplicity, we use (−δη / δεx) in the small strain limit as the definition of the measured nematic susceptibility χnem. The temperature dependence of χnem in LaFeAsO is shown in Fig. 2(c). A clear kink is resolved at the structural transition TS, above which the χnem(T) curve can be well fitted to the Curie-Weiss type temperature dependence

χnem = χ0 + C (T − T nem),

in which χ0 is the intrinsic piezoresistivity effect unrelated to electronic nematicity, T n em is the nematic transition temperature in the mean field theory, and C is the Curie constant which indicates the magnitude of nematic susceptibility.

Similar measurements and analyses were conducted for LaFe1-xCo2AsO single crystals across the substitution series. As shown in Fig. 3, for all the samples we have studied, their χnem(T) curves diverge in a clear Curie-Weiss-like form. The kink at TS becomes less pronounced with doping and is unresolvable in our data set for x > 0.03. One may attribute the smearing of the TS feature to the increasing impurity scattering effect. In such a case, according to Ref. [11], χnem should remain practically unaffected by the defect and impurity concentrations in the tetragonal phase. However, the blurred feature of TS could also be an intrinsic property of LaFe1-xCo2AsO, since the structural transition anomaly also gets broader with increasing x in a thermal expansion measurement [37]. Note that a spin-lattice relaxation study on the sister compound LaFeAsO1-xFx showed that competing local charge environments exist at the nanoscale in the under doped region [35] [30]. If the same mechanism also works in LaFe1-xCo2AsO, it is natural that the features of TS from averaged properties are muddled by doping. In order to choose the proper fitting range of the χnem(T) curves, especially for those without (a clear) TS, we followed the procedure described...
in Ref. [19]. Additionally, the fitting range is allowed to alter by 20 K for each sample in order to estimate the uncertainty caused by improper fitting ranges, set as error bars of the fitted parameters $T_{nem}$ and $C$ in the following discussion.

The information extracted from Fig. 3 is summarized in Fig. 4, plotted together with the phase diagram of LaFe$_{1-x}$Co$_x$AsO. First of all, one can see that the nematic transition temperature $T_{nem}$ is linearly suppressed by doping, and finally changes its sign around the optimal doping level $x \approx 0.06$. It is known that superconductivity could get boosted via fluctuations related to quantum critical points. The most studied case is the AFM fluctuations with certain wave vectors around a magnetic quantum critical point (mQCP). As shown in Fig. 1(c), the magnetic transition is totally suppressed before the superconductivity emerges in LaFe$_{1-x}$Co$_x$AsO [35, 37]. That means the presumptive mQCP does not contribute to the superconducting pairing directly. Theoretically, the nematic quantum critical point (nQCP) and its pertinent fluctuation can also enhance the superconductivity by promoting the attractive forces in all pairing channels [12, 45], or by reducing the intra-pocket repulsion [15, 46].

Our observation of the sign change of $T_{nem}$ around the optimal doping level strongly corroborates those theoretical expectations. As a remarkable system with convincing evidences for the existence of a nQCP under its superconducting region [20, 47], FeSe$_{1-x}$S$_x$ exhibits a relatively flat $T_c(x)$ curve. This has been explained by magnetic fluctuations, instead of nematic fluctuations, still dominating the $T_c$ value in FeSe$_{1-x}$S$_x$, although the magnetic order does not exist without pressure [48]. Since the magnetic fluctuation surrounds the presumptive mQCP does not cause superconductivity in LaFe$_{1-x}$Co$_x$AsO, it could be an alternative choice to search for the relationship between superconductivity and nematicity.

Interestingly, the nematic fluctuation $\chi_{nem}$ of LaFe$_{1-x}$Co$_x$AsO seems to exhibit a double-peak feature. It can be seen in the upper panels of Fig. 3(a-k) and also in the contour plot presented in Fig. 4. This trend is more clearly shown by the evolution of the Curie constant displayed in Fig. 4, which represents the divergency coefficient of the $\chi_{nem}(T)$ curve. On the one hand, the enhanced $\chi_{nem}$ on top of the critical point around $x = 0.06$ is a further supportive characteristic of a presumptive nQCP. On the other hand, the second peak in the underdoped region at $x \approx 0.04$ is close to the presumptive mQCP. An alternative nematic origin for this enhancement of $C$ can be ruled out because at that particular doping level, the signal of structural transition at around 60 K from other measurement techniques is still unambiguous [35, 37]. According to our fitting, the
The authors of that paper explained their results as separated QCPs masked below the widespread superconducting dome. We therefore suspect a similar double-peak $\chi^{\text{nem}}$ profile would also be detected in NaFe$_{1-x}$Co$_2$As.

To summarize, we found that the Curie-Weiss-like $\chi^{\text{nem}}$ also exists in LaFe$_{1-x}$Co$_2$AsO, in line with the other iron-based superconductors. The sign-change of $T^{\text{nem}}$ around $x = 0.06$ and the divergent amplitude of $\chi^{\text{nem}}$ by approaching this doping level fulfill the requirements of a possible nQCP at the optimal doping level. An additional peak of $\chi^{\text{nem}}$ in the under-doped region around $x = 0.04$ is present close to the end point of the AFM transition line in LaFe$_{1-x}$Co$_2$AsO. We interpret this as an evidence for a presumptive mQCP in the system, which influences the $\chi^{\text{nem}}$. However, the double-peak feature of $\chi^{\text{nem}}$ defy the understanding of nematicity merely as an induced effect of the vestigial magnetism.

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