Magneto-elastically coupled structural, magnetic and superconducting order parameters in BaFe$_2$(As$_{1-x}$P$_x$)$_2$

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We measure the transport properties of mechanically strained single crystals of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ over a wide range of $x$. The Néel transition is extremely sensitive to stress and this sensitivity increases as optimal doping is approached, even though the transition itself is strongly suppressed. Furthermore, we observe significant changes in the superconducting transition temperature with applied strain, which mirror changes in the composition $x$. These experiments are a direct illustration of the intimate coupling between different degrees of freedom in iron-based superconductors, revealing the importance of magneto-elastic coupling to the magnetic and superconducting transition temperatures.

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I. INTRODUCTION

Materials that exhibit unconventional superconductivity are almost always in the proximity of alternative, often magnetic ground states. Each ground state is characterized by a broken symmetry and an associated order parameter which acquires a finite value at the critical temperature, indicating the transition has occurred. The iron based superconductors fall within a broad family of correlated electron materials which are related to anti-ferromagnetism, joining the cuprate, heavy fermion and layered organic superconductors. The relationship of the magnetic, structural and other (sometimes unknown) order parameters, and particularly how these conspire to give rise to high superconducting critical temperatures $T_c$, is one of the most important experimental challenges in understanding the mechanism behind high-temperature superconductivity.

In the present work, we reveal the intimate relationship between different broken symmetry ground states in the BaFe$_2$As$_2$ superconducting family of iron-pnictides. When left chemically unmoled, these materials are characterized by high temperature phase that is tetragonal (Tet) and paramagnetic, transitioning at $\sim 138K$ to an orthorhombic (Ort), collinear antiferromagnet (AFM). In this case, the structural transition breaks tetragonal symmetry ($C_4 \rightarrow C_2$), and the shear strain $u_{xy} \equiv \partial_y u_x + \partial_x u_y$ plays the role of the order parameter. The magnetic order breaks both spin-rotational and tetragonal symmetry, characterized by an order parameter corresponding to the staggered sublattice magnetization $M_i$ where $i = 1, 2$ refers to the magnetization of each sublattice. When BaFe$_2$As$_2$ is electron, hole or isovalently ‘doped’, these transitions are suppressed and superconductivity (SC) emerges, with optimal $T_c$ appearing when magnetism is completely absent, indicating that AFM and SC order parameters compete. While for electron doped materials the structural ($T_S$) and magnetic ($T_N$) transitions separate with $T_S > T_N$, in the present isovalently substituted BaFe$_2$(As$_{1-x}$P$_x$)$_2$ materials, no such splitting is observed at any composition.

Several theoretical descriptions have attempted to explain the coupling between the structural and magnetic transition, based on pure ferroelastic phenomenology or inclusion of a nematic order parameter. Despite the different approaches of each of the works, all of them highlight the importance of the magneto-elastic coupling, which can cause the two transitions to split or to occur simultaneously. Here, we investigate the role of the magneto-elastic coupling by studying the thermodynamic response of the material BaFe$_2$(As$_{1-x}$P$_x$)$_2$ to a small mechanical strain applied along the tetragonal [110] direction, or equivalently the orthorhombic $b$ axis when $T < T_N$. We find that a small shear stress $\sigma$ can significantly alter the Néel transition $T_N$ and superconducting $T_c$, in a manner akin to changing $x$. Surprisingly, even though the magnetism is almost completely suppressed at optimal doping, the effect on the magnetic transition grows, suggesting that magneto-elastic fluctuations substantially increase near optimal $T_c$.

II. EXPERIMENTAL METHODS

The growth of single crystals of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ is described elsewhere. It is worth noting however that the quality of the crystals can be improved by annealing within the growth for a week at 900°C. Samples were mechanically strained along either the [100]$_T$ or [110]$_T$ direction using a custom built mechanical device.
FIG. 1. Temperature dependence of the normalised resistivity, $\rho/\rho(300K)$, for unstressed (blue), and uni-axially stressed along [110]$_{\parallel}$ (red) BaFe$_2$(As$_{1-x}$P$_x$)$_2$ crystals with $x = 2.6\%$ to $x = 32\%$. $T_N$ determined from $d\rho/dT$ are labeled and denoted by vertical lines with corresponding colors (see Appendix A for details).

described in Ref. [2]. A cantilever was pressed against the sample by adjusting a turnable screw, applying < 10MPa. Each sample was cut to have similar dimensions (300×200×80µm$^3$ - the largest dimension of the batch with the smallest samples). To reduce the errors associated with differences in pressure, the experiments were repeated on 3 samples from each batch. This gives us confidence that we are able to apply a similar stress for all samples and hence that the changes we detect between samples from different batches (different P content $x$) are in fact systematic. This study is distinct from our earlier investigations of the transport anisotropy, which is a non-equilibrium property, whereas we presently focus to the effect of mechanical strain on the temperature of the phase transitions themselves.

III. RESULTS

Figure 1 (a)-(h) illustrates the main experimental data. In each panel we show the normalised resistivity vs temperature for an unstressed (blue) and stressed (red) crystal at a given doping, where the stress has been applied along the [110]$_{\parallel}$. The vertical lines denote the assigned Néel transitions for each curve which have been determined by the minimum in the resistivity derivative (Appendix A). Note that in contrast to electron doped materials where two anomalies are observed in $d\rho/dT$ in the unstrained samples, we only observe one in BaFe$_2$(As$_{1-x}$P$_x$)$_2$, indicating that $T_N \approx T_s$ for all $x$.

This may suggest that the magneto-elastic coupling in these materials is perhaps larger [10], though we point out that other studies have observed split transitions in these compounds [4]. In the presence of mechanical strain, the structural transition will naturally broaden to higher temperatures, but by continuity, the anomaly seen in the data of mechanically strained samples must be associated with the magnetic order.

In all the samples we studied, $\Delta T_N = T_N(\sigma) - T_N(\sigma = 0) > 0$, where $\sigma$ indicates the mechanical strain field (the stress). Intriguingly, for the unstressed optimally superconducting $x = 32\%$ sample (blue curve in Figure 1 (h)) there is no detectable magnetic transition, but after application of stress a distinct minimum arises at 45K, almost identical to the minimum seen in the unstrained samples at lower doping (consider blue curve in Figure 1 (g)). It appears that the magnetic transition has been summoned from beneath the superconducting dome by the application of mechanical strain. Even though the magnetic order itself vanishes, the strong magneto-elastic coupling as well as magnetic and elastic fluctuations remain.

In Figure 2 we show the same data presented in Figure 1 but focus around the superconducting critical temperature at each composition. In this case $T_c$ is defined as the midpoint in the superconducting transition. Even though the transition likely gets broader with the application of strain, the superconducting $T_c$ nevertheless can be seen to decrease as strain is applied until compositions beyond optimal $x$, where we observe $T_c$ to increase.
We plot the change in the Néel temperature $\Delta T_N$ as a function of doping for nominally the same strain at each doping. Surprisingly, even though the magnetic and the structural transitions are suppressed as a function of doping, the amount that $T_N$ can be changed by stress monotonically increases with P content, within our error bars. This is in contrast to the value of the resistivity anisotropy itself, which is highly non-monotonic with doping (Appendix C). If we also include data of $T_N(\sigma)$ at $x = 32\%$, $\Delta T_N$ appears to have the largest response at optimal doping, as shown in Figure 4(a). This large enhancement of $\Delta T_N$ implies an enhanced susceptibility of the AFM ground state to shear stress.

In Figure 4(b), we illustrate the equivalent plot of changes in the superconducting critical temperature $\Delta T_c$ as a function of doping for nominally the same strain. $\Delta T_c$, unlike $\Delta T_N$, is not monotonic with $x$. Comparing the dependence with the evolution of the unstressed superconducting transition with $x$, it appears that the magnitude of $\Delta T_c$ is largest where the dome is steepest and small otherwise; mathematically $\Delta T_c \propto -dT_c/dx$. Indeed, considering Figure 4(a) a very similar equation likely applies to the magnetic transition, so that $\Delta T_N \propto -dT_N/dx$. As $T_N$ is increased, there are likely fewer electrons available to participate in superconductivity, and so the fact that $T_c$ decreases with applied stress in the underdoped region is not surprising, since $T_N$ increases. However, we note that an unstrained sample with a given $T_N$ has always a $T_c$ that is lower than a strained sample with the same $T_N$, i.e. $T_c(T_N, 0) < T_c(T_N, \sigma)$. Therefore, there is an intrinsic effect of stress on $T_c$, beyond the indirect effect due to the competition between AFM and SC.

We employ a Ginzburg-Landau (GL) approach to obtain further insight into our observations. The GL model has been applied to the ferro-pnictides by several authors already to describe the coupling between structure and magnetism[2,14,18]. From symmetry considerations, the coupling between magnetic and elastic degrees of freedom enters the free energy via:

$$F_{ME} = g (M_1 \cdot M_2) u_{xy},$$

(1)

where $g$ is the magneto-elastic coupling. In order to describe the present experiment, we need to add also the term $-\sigma u_{xy}$, where $\sigma$ is the applied mechanical stress. If one assumes that the structural and magnetic transitions occur independently, the coupling effectively ties them together. Alternatively, it has been proposed that

\[ \Delta T_N = T_N(\sigma) - T_N(\sigma = 0) \]

switches sign beyond optimal $x$. Finally, the effect on $T_N$ and $T_c$ is proportional to the amount of applied strain, and this can be demonstrated by applying a systematically increasing amount (Appendix B). These data are suggestive that the application of shear stress has a similar affect to decreasing $x$ across the phase diagram.

Figure 2(c) illustrates that $\Delta T_c = T_c(\sigma) - T_c(\sigma = 0)$ switches sign beyond optimal $x$. Finally, the effect on $T_N$ and $T_c$ is proportional to the amount of applied strain, and this can be demonstrated by applying a systematically increasing amount (Appendix B). These data are suggestive that the application of shear stress has a similar affect to decreasing $x$ across the phase diagram.

To ensure that this effect is indeed intrinsic to pressure along one of the orthorhombic axes, we also apply pressure along [100]$_T$, shown for comparison on two samples from the same batch in Figure 2(a) and (b). Mechanical strain in this direction results in small changes in both magnetic and superconducting transitions. We furthermore apply pressure in the inter-layer $c$ direction (Figure 2(c)) and found the opposite behavior whereby $\Delta T_N < 0$ and $\Delta T_c > 0$. This suggests that the ratio $c/b$ whether directly or indirectly, likely plays a role in the superconducting mechanism.

\[ \frac{\partial \rho}{\partial T} \propto -c \]

FIG. 2. Expanded view of the data shown in Figure 1 around to $T_c$ for $x = 21.3\%$ to $x = 32\%$ ((a) to (d), underdoped) and $x = 21.3\%$ ((e), overdoped). Red and blue curves correspond to the normalized resistivity of unstressed, and uni-axially stressed (along [110]$_T$) respectively. For underdoped samples, $\Delta T_c > 0$; for overdoped samples, $\Delta T_c < 0$.

\[ \frac{\partial \rho}{\partial T} \propto -c \]

FIG. 3. Temperature dependence of normalized in-plane resistivity $\rho/\rho(300K)$, for unstressed crystals and (a) uni-axially stressed along [110]$_T$ (red), (b) uni-axially stressed along [100]$_T$ (green), and (c) uni-axially stressed along the crystal c axis (yellow) BaFe$_2$(As$_{1-x}$P$_x$)$_2$ crystals with $x=28.7\%$. The upper inserts show $d\rho/dT$ vs $T$ of each crystal around $T_N$ with corresponding colors. For (a), $\Delta T_N \sim 27K$, (b), $\Delta T_N \sim 5K$, and (c), $\Delta T_N \sim -5K$. The lower inserts are magnified plots of $\rho$ vs $T$ around $T_c$. For (a), $\Delta T_c \sim -2.5K$, (b), $\Delta T_c \sim -0.3K$, and (c), $\Delta T_c \sim 4K$.

\[ \frac{\partial \rho}{\partial T} \propto -c \]
the structural transition is a secondary consequence of an underlying electronic order dubbed nematic. In this case, an independent order parameter \( \eta \propto (M_1 \cdot M_2) \) condenses and triggers the structural transition via the coupling. Within this approach, the elastic degrees of freedom are not intrinsically soft and can be integrated out from the partition function (see Supplemental Material for more details), yielding the contribution to the free energy \( \propto \frac{\sigma^2}{\zeta} (M_1 \cdot M_2) \), where \( \zeta \) is the bare elastic shear constant (see also ). This term shows that the mechanical stress is converted into a conjugate field \( \sigma g / \zeta \) to the electronic order parameter \( \eta \). Furthermore, it also changes the magnetic part of the free energy, resulting in an increase of the magnetic transition temperature \( T_N(\sigma \neq 0) > T_N(\sigma = 0) \).

As \( x \) approaches optimal compositions, we observe that \( g / \zeta \) increases substantially and is strongest at optimal doping. In contrast, our previous mechanical strain studies on Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)AS\(_2\) did not show significant changes in \( T_N \), and changes have only been observed for pressures \( \sim 5 \times \) those used here, though a recent neutron study in the parent compound could detect changes in \( T_N \) at much smaller pressures. Nevertheless, as a function of Co doping the difference between \( T_N \) and \( T_S \) grows, which could be interpreted in terms of a decreasing \( \zeta \). Furthermore, even though the lattice softens as a function of temperature, the average value of \( C_0^0 \) in fact increases as a function of Co doping. Here, in contrast, the effect of stress on \( T_N \) is strongly enhanced as \( x \) increases in BaFe\(_2\)(As\(_{1-x}\)P\(_x\))\(_2\), suggesting that either \( g \) becomes larger or \( C_0^0 \) smaller, or both. Another possibility is that, near optimal doping, where there is no structural or magnetic transitions, the nematic susceptibility \( \chi_{\text{nem}} \) is enhanced, providing an additional contribution that enhances the “conjugate field” \( \frac{\sigma g}{\zeta} \chi_{\text{nem}}(M_1 \cdot M_2) \) and, consequently, \( \Delta T_N \) (see Appendix [10]). Interestingly, experiments have indicated that magnetic fluctuations are critical at optimally-doped BaFe\(_2\)(As\(_{1-x}\)P\(_x\))\(_2\), which could suggest a close connection between nematic and magnetic fluctuations in these compounds.

Furthermore, magnetic fluctuations enhance the repulsive inter-band pairing interaction that can lead to an unconventional superconducting state. Nematic fluctuations, on the other hand, give rise to an attractive intra-band pairing interaction, which can potentially enhance the transition temperature of the unconventional SC phase.

Previous x-ray studies on Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)AS\(_2\) showed that \( u_{xy} \) is strongly suppressed below \( T_N \), indicating that the SC and orthorhombic phases compete. One would then expect that by applying stress and inducing a non-zero \( u_{xy} \), \( T_c \) would decrease. However, our observations that \( T_c(\sigma) > T_c(0) \) in the overdoped region and \( T_c(T_N, \sigma) > T_c(T_N, 0) \) in the underdoped region suggest that the applied stress may actually lead to an intrinsic increase of \( T_c \). To understand the effect of mechanical stress on superconductivity, we can compare to the case of high-\( T_c \) copper-oxide based materials. Though the effects vary between different compounds, Hardy et al. proposed a unified picture of the influence of uniaxial strain (excluding YBCO), whereby changes in \( T_c \) could be accounted for by changes in the ratio \( c/a \). In the present study, \( c/b \) will always increase when stress is applied along [110], and by a smaller amount when applied along [100], but will decrease when applied along \( c \) (see Figure [4]). The changes we are able to invoke on the under-doped samples follow this trend, so that qualitatively \( \Delta T_c \propto -\Delta(c/b) \). However, one cannot say whether it is the lattice parameters alone, their ratio or some other systematically adjusted internal parameter which is most important (such as the As-Fe-As bond angle). Direct structural measurements as a function of mechanical strain are required to answer this question.

V. CONCLUSIONS

In conclusion, we have found a strongly enhanced magneto-elastic response in BaFe\(_2\)(As\(_{1-x}\)P\(_x\))\(_2\) as \( x \) approaches optimal doping, which may be related to the superconducting mechanism itself. We also find that mechanical strain can directly couple to the superconducting order parameter in a manner that is similar to decreasing the P concentration \( x \). These experiments are therefore a direct illustration of the subtle coupling between different degrees of freedom in BaFe\(_2\)(As\(_{1-x}\)P\(_x\))\(_2\).

VI. ACKNOWLEDGMENTS

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Appendix A: Determination of the Néel temperature from the resistivity derivative

The Fermi surface reconstruction associated with the Néel order at \( T_N \) appears as a pronounced minimum in the derivative with respect to temperature. After applying strain, we observe an increase in \( T_N \), as shown in Fig. \( \Box \). Blue and red represent unstressed and uni-axially stressed (along [110]) \( \text{BaFe}_2(\text{As}_1-x\text{P}_x)_2 \) single crystals respectively. Even though the stress broadens the transition, the increase in \( T_N \) can be easily resolved.

Appendix B: Systematic response of \( T_N \) to pressure

Although we cannot determine the absolute value of stress applied, we can nevertheless tune the amount of pressure applied by gradually tightening the screw on the device. As a typical example, two samples of \( \text{BaFe}_2(\text{As}_1-x\text{P}_x)_2 \) were measured with a different amount of stress applied along [110]. |\( \Delta T_N \)| and |\( \Delta T_c \)| increase with increasing stress.

Appendix C: Resistivity Anisotropy

The in-plane resistivity anisotropy of \( \text{BaFe}_2(\text{As}_1-x\text{P}_x)_2 \) was obtained by measuring the resistivity of mechanically detwined crystals as described elsewhere. Clearly, the resistivity anisotropy has a highly non-monotonic dependence on doping. This is in contrast to the trend of the response of \( T_N \) at constant stress as a function of doping, which is a monotonic increase with the concentration \( x \), as shown in Figure \( \Box \).

Appendix D: Ginzburg-Landau analysis

To understand how the different degrees of freedom affect \( T_N \) in mechanically stressed samples, we use a phenomenological Ginzburg-Landau model for the magnetic, nematic, and elastic degrees of freedom. For the magnetic part, we have:

\[
F_{\text{mag}} = \frac{r_0}{2} (M_1^2 + M_2^2) + \frac{u}{8} (M_1^2 + M_2^2)^2 - \frac{\lambda}{2} (M_1 \cdot M_2)^2
\]

where \( M_1 \) and \( M_2 \) are the staggered magnetization of the two interpenetrating Néel sublattices. Here, we defined \( r_0 = a (T - T_{N,0}) \), with \( T_{N,0} \) denoting the mean-field magnetic transition temperature. The coupling constants satisfy \( u > 0 \) and \( \lambda > 0 \), such that in the free-energy minimum \( M_1 \) and \( M_2 \) are either parallel or anti-parallel, corresponding to the two possible magnetic stripe configurations with ordering vectors \((\pi, 0)\) and \((0, \pi)\) in the 1-Fe unit cell Brillouin zone. It is convenient to introduce the order parameters of these two magnetic stripe states \( \Delta_1 \) and \( \Delta_2 \), such that \( \Delta_1 = (M_1 + M_2)/2 \) and \( \Delta_2 = (M_1 - M_2)/2 \). Notice that \( M_1^2 + M_2^2 = 2 (\Delta_1^2 + \Delta_2^2) \) and \( M_1 - M_2 = \Delta_1 - \Delta_2 \).

We now consider the nematic part in a phenomenological way. The nematic order parameter \( \varphi \) breaks the tetragonal symmetry of the lattice. At high enough temperatures (compared to the structural transition temperature), such as those for the optimally-doped compounds, we consider the free energy expansion only up to second-order in the nematic order parameter:

\[
F_{\text{nem}} = \frac{1}{2} \left( \chi_{\text{nem}}^{(0)} \right)^{-1} \varphi^2 - \kappa \varphi \left( \Delta_1^2 - \Delta_2^2 \right)
\]

where \( \kappa \) is the coupling between nematic and magnetic degrees of freedom, and \( \chi_{\text{nem}}^{(0)} \) is the static nematic susceptibility. For the elastic part, we consider a harmonic lattice

\[
F_{\text{el}} = \frac{C_s}{2} u_{xy}^2 - g u_{xy} \varphi - \sigma u_{xy}
\]

where \( g \) is the magneto-elastic coupling, \( C_s \) is the shear modulus, and \( \sigma \) is the applied stress.

To study how \( T_{N,0} \) changes as function of \( \sigma \), we first integrate out the elastic degrees of freedom from the partition function

\[
\int du_{xy} e^{-\frac{C_s}{2} u_{xy}^2 + \varphi \left( g u_{xy} \varphi - \sigma u_{xy} \right) \propto }
\]

\[
\exp \left[ \frac{(g \varphi + \sigma)^2}{2C_s^2} \right]
\]

Substituting in Eq. (D2), the nematic free energy becomes

\[
F_{\text{nem}} = \chi_{\text{nem}}^{-1} \varphi^2 - \varphi \left[ \kappa \left( \Delta_1^2 - \Delta_2^2 \right) + \frac{g \sigma}{C_s^2} \right]
\]

where we defined the renormalized nematic susceptibility \( \chi_{\text{nem}}^{-1} = \left( \chi_{\text{nem}}^{(0)} \right)^{-1} - \frac{g^2}{2C_s^2} \). If we consider the regime where the nematic free energy can be approximated by the quadratic expansion (D2), we can also integrate out the nematic degrees of freedom, obtaining

\[
\int d\varphi e^{-\frac{\varphi^2}{2\chi_{\text{nem}}^{-1}}} \varphi^2 \left[ \kappa \left( \Delta_1^2 - \Delta_2^2 \right) + \frac{g \sigma}{C_s^2} \right] \propto
\]

\[
\exp \left[ \frac{\left( \kappa \left( \Delta_1^2 - \Delta_2^2 \right) + \frac{g \sigma}{C_s^2} \right)^2}{2\chi_{\text{nem}}^{-1}} \right]
\]
FIG. 6. Temperature dependence of normalized resistivity of BaFe$_2$(As$_{1-x}$P$_x$)$_2$ crystals with $x = 2.6\%$ to $x = 32\%$. The normalized resistivity $\rho$ becomes systematically increased amounts of stress. The normalized resistivity $\rho_i$ with $i = \{0, \sigma 1, \sigma 2\}$ stand for unstressed (blue), and uni-axially stressed along [110] respectively. The upper inserts show $d\rho_i/dT$ vs $T$ near $T_N$, and the lower inserts are expanded plots of $\rho_i$ near $T_c$.

FIG. 5. Derivative of normalized resistivity with respect to temperature, $d\rho/(\rho(300K))$, for unstressed (blue), and uni-axially stressed along [110] (red) BaFe$_2$(As$_{1-x}$P$_x$)$_2$ crystals with $x = 2.6\%$ to $x = 32\%$. The minimum in $d\rho/dT$ is shown in corresponding color, and is interpreted as the Néel temperature $T_N$.

Substituting in Eq. (D1), the magnetic free energy becomes

\[
F_{mag} = r_0 \left( \Delta_1^2 + \Delta_2^2 \right) + \frac{u}{2} \left( \Delta_1^2 + \Delta_2^2 \right)^2 - \frac{1}{2} \left( \lambda + \frac{\kappa^2}{\lambda_{nem}} \right) (\Delta_2^2 - \Delta_1^2)^2 - \frac{g\kappa\sigma}{\lambda_{nem} C_0} (\Delta_1^2 - \Delta_2^2)
\]  

(D7)
The last term acts as a conjugate field and breaks the tetragonal symmetry, selecting the magnetic stripe configuration corresponding to the \( \Delta_1 \) order parameter (ordering vector \( (\pi, 0) \)). Since \( r_0 = a (T - T_{N,0}) \), the magnetic transition temperature is given by

\[
T_N = T_{N,0} + \left( \frac{\partial \chi_{nem}}{\partial C_0^s} \right) \sigma \quad (D8)
\]

Hence, the increase in \( T_N \) is proportional to the applied strain \( \sigma \). The enhanced response at optimal doping can be due to one (or a combination) of the following features: an intrinsic softening of the lattice (i.e. decrease \( C_0^s \)), an enhancement to the magneto-elastic coupling (i.e. increase of \( g \) and/or \( \kappa \)), and an enhancement of nematic fluctuations (i.e. increase of \( \chi_{nem} \)). A similar enhancement in \( T_N \) is also expected even in the absence of a nematic order parameter, as pointed out recently by Cano and Paul.\(^{19}\)

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