



Electronic and Magnetic Anisotropies in FeSe Family of Iron-Based Superconductors

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Most parent compounds of iron-based superconductors (FeSCs) exhibit a tetragonal-to-orthorhombic lattice distortion below T_s associated with an electronic nematic phase that breaks the four-fold (C_4) rotational symmetry of the underlying lattice, and then forms collinear antiferromagnetic (AF) order below T_N ($T_N \leq T_s$). Optimal superconductivity emerges upon suppression of the nematic and AF phases. FeSe, which also exhibits a nematic phase transition below T_s but becomes superconducting in the nematic phase without AF order, provides a unique platform to study the interplay amongst the nematic phase and superconductivity. In this review, we focus on the experiments done on uniaxial pressure detwinned single crystals of FeSe compared to other FeSCs and highlight the importance of understanding the electronic and magnetic anisotropy in elucidating the nature of unconventional superconductivity.

Keywords: detwin, superconductivity, magnetism, nematicity, orbital selectivity

1. INTRODUCTION

In unconventional superconductors such as heavy fermions, copper- and iron-based materials, the observation that superconductivity often emerges from their antiferromagnetic (AF) ordered parent compounds suggests that magnetism plays an important role in the mechanism of high-transition temperature (high- T_c) superconductivity [1]. In addition to forming a collinear AF structure below T_N , most parent compounds of iron-based superconductors (FeSCs) exhibit a tetragonal-to-orthorhombic structural transition below T_s and form an electronic nematic phase that breaks the four-fold (C_4) rotational symmetry in the iron plane ($T_N \leq T_s$) [2, 3]. Since the tetragonal-to-orthorhombic structural transition for FeSCs occurs below room temperature ($T_s < 295$ K), the system forms 90° rotated twinned domains below T_s , making it impossible for a bulk probe to determine the intrinsic electronic and magnetic properties of the individual domains and the associated nematic phase. To alleviate this technical difficulty, mechanical detwin devices were developed first for the BaFe_2As_2 compounds [4, 5], and later adapted for other material families. These types of devices utilize a mechanical device to apply uniaxial pressure along one of the orthorhombic lattice directions, one can detwin single crystals of FeSCs and thus measure the intrinsic electronic and magnetic anisotropies present in the orthorhombic phase [4]. Therefore, uniaxial pressure detwinned FeSCs can provide a platform to study the interplay of the nematic phase, magnetic order, and superconductivity. Compared with other families of FeSCs, FeSe is highly unusual because FeSe exhibits an orthorhombic structural distortion at $T_s \approx 90$ K and superconductivity at $T_c \approx 9$ K [6] without magnetic order. As a consequence, one can directly probe the interplay between the nematic phase and superconductivity without the

complication of the static AF ordered phase. Moreover, unexpected phenomenon, say, extremely-high superconducting temperature in thin films of FeSe [7, 8], have been observed. And it is also proposed that exotic state like Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state is realized in this compound [9, 10].

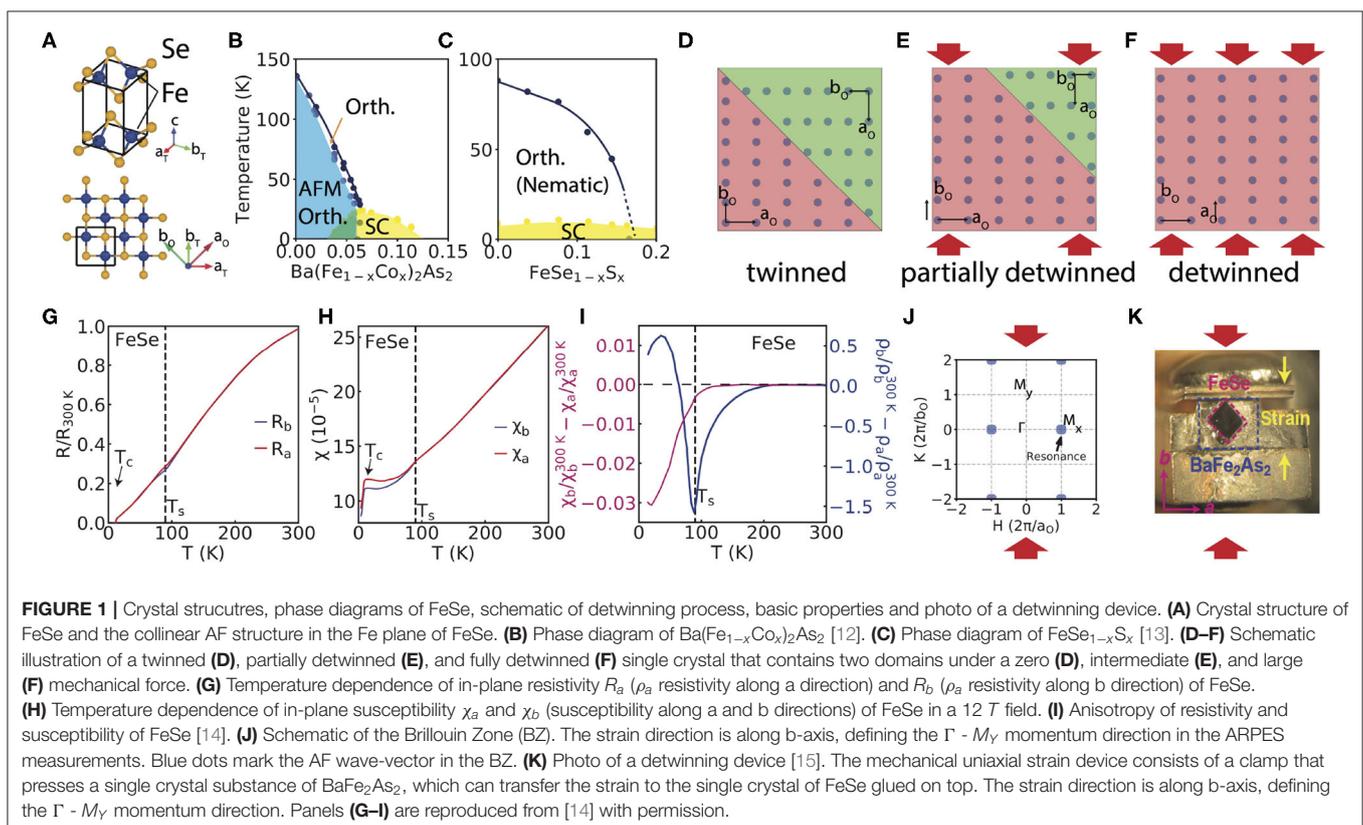
After the discovery of the unconventional superconductivity in F-doped LaFeAsO [11], many FeSCs were found and classified into RFeAsO (R = La, Ce, Pr, ..., the 1111 family), AFe₂As₂ (A = Ba, Sr, Ca, K, the 122 family), AFeAs (A = Li, Na, the 111 family, **Figure 1A**), Fe_{1+y}Te_{1-x}Se_x (the 11 family), and A_xFe_{2-y}As₂ (A = K, Rb, ..., the alkali iron selenide family) [16–18]. The 122 family, especially the electron- and hole-doped BaFe₂As₂, are the most intensively studied materials [19–22] because superconductivity can be induced by doping a large variety of chemicals including K/Na on Ba sites (hole-doping) [23, 24], Co/Ni on Fe sites (electron-doping) [25, 26] and P on As sites (isovalent doping) [27], and large-sized single crystals can be grown by self-flux methods in most cases [28]. In the phase diagram of Co-doped BaFe₂As₂, the static AF order is gradually suppressed and separated from the structural transition by Co-doping in the underdoped region [12, 29]. The optimal superconductivity appears when the nematic phase and AF order are suppressed, suggesting that the nematic and static AF orders are competing with superconductivity [30] (**Figure 1B**). However, FeSe does not follow this typical phase diagram (**Figure 1C**). Instead, in the S-doped FeSe, the structural (nematic) transition does not have an accompanying

magnetic transition [13, 31–33]. Moreover, superconductivity is not suppressed with increasing S-substitution, different from the superconductivity dome in the phase diagram of Ba(Fe_{1-x}Co_x)₂As₂. Given the contrasting behavior between Ba(Fe_{1-x}Co_x)₂As₂ and FeSe_{1-x}S_x, it will be interesting to study the relationship between nematic phase and superconductivity in these two families of materials.

2. NEMATICITY AND DETWINNING DEVICES

The electronic nematic phase refers to the in-plane rotational symmetry-breaking phase below the structural transition temperature T_s in FeSCs [3]. Transport measurements on uniaxial strain detwinned Ba(Fe_{1-x}Co_x)₂As₂ provided evidence of anisotropic transport properties that were attributed to the presence of a nematic phase because the small lattice distortion below T_s is not expected to induce such a large resistivity anisotropy [4]. Later, the electronic and magnetic anisotropies were observed by angle-resolved photoemission spectroscopy (ARPES) [34] and inelastic neutron scattering (INS) measurements on mechanically detwinned samples [35], highlighting nematicity as an essential degree of freedom that interplays with magnetism and superconductivity.

Since the iron-based materials naturally form two 90° rotated twin domains below the orthorhombic transition at T_s , experiments on twinned samples usually measure the average of anisotropic properties [36, 36]. Thus, to study the intrinsic



electronic and magnetic anisotropies, it is essential to detwin the sample (**Figures 1D–F,J,K**), which can be achieved by applying a uniaxial pressure through a mechanical clamp device that directly presses on the parallel edges of the crystal [4, 34, 37–42]. However, for materials that are structurally soft, such direct clamping is difficult to achieve. Instead, other types of detwinning strategy were developed that involve gluing the sample onto a substrate that can transfer a uniaxial strain to the sample. Piezoelectric materials [43, 44], glass-fiber-reinforced plastic [45], and even mechanical force detwinned BaFe_2As_2 (**Figure 1K**) [15, 46] are proved to be effective substrates to detwin the FeSCs. In this review, we discuss effective strain strategy on FeSe using BaFe_2As_2 as a substrate, which is clamped in a detwin device. Since BaFe_2As_2 itself undergoes tetragonal to orthorhombic transition at 138 K, the detwinned BaFe_2As_2 single crystal would provide a natural anisotropic strain on the FeSe glued on top.

3. ELECTRONIC AND MAGNETIC ANISOTROPIES

3.1. Resistivity and Susceptibility

Resistivity measurements on uniaxial strain detwinned $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ reveal clear evidence of in-plane anisotropy developing at a temperature above T_N and T_s [4, 43, 45]. In the undoped parent compound BaFe_2As_2 , the resistivity anisotropy increases when approaching $T_N = T_s \approx 138$ K from higher temperature, peaks at $T_N = T_s$, and then gradually decreases upon cooling. As a function of increasing electron (Co)-doping in $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$, the resistivity anisotropy increases and then vanishes near optimal doping, consistent with the spin nematic scenario in which the tetragonal-to-orthorhombic transition is driven by magnetic fluctuations at a temperature $T_s > T_N$. Since structural distortion in $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ is also associated with the lifting of degeneracy in the orbital degrees of freedom below T_s [34], the observed resistivity anisotropy could be a consequence of orbital order instead of magnetic fluctuations. In the magnetic susceptibility measurements on detwinned BaFe_2As_2 , χ_b becomes larger than χ_a below $T_N = T_s$, and the anisotropy monotonically increases upon cooling [45].

Since single crystals of FeSe are thin and fragile, more complicated detwinning strategies were developed to detwin FeSe, such as using a “horseshoe” device [47] or glue the single crystals on different substrates [14, 15, 46]. In resistivity and susceptibility measurements on detwinned FeSe, the behavior of the anisotropy is very similar to that of BaFe_2As_2 . The in-plane resistivity anisotropy in FeSe develops at a temperature above T_s , peaks at T_s , and then vanishes upon cooling (**Figures 1G,I**), while the absolute value of susceptibility anisotropy monotonically increases with decreasing temperature (**Figures 1H,I**) [14].

We note that the sign of $\rho_b - \rho_a$ and $\chi_b - \chi_a$, where $\rho_{a/b}$ and $\chi_{a/b}$ are resistivity and magnetic susceptibility along the lattice orthorhombic a/b directions, respectively, in detwinned FeSe is opposite to that of BaFe_2As_2 . The small magnitudes and the reversed sign of resistivity and susceptibility anisotropy in detwinned FeSe may be attributable to the small lattice orthorhombicity, which results in smaller orbital overlap along

the a -axis, while the static collinear AF order in the BaFe_2As_2 systems, coupled with related spin fluctuations, give rise to the overwhelming ρ_b and χ_b .

3.2. Angle-Resolved Photo-Emission Spectroscopy

The electronic structure of FeSe has been intensively studied by ARPES measurements [15, 48–60]. FeSe shares common features with other FeSCs, with three hole bands near the BZ center and two electron bands at the BZ corners. However, one distinction between FeSe and iron pnictides such as doped BaFe_2As_2 is that FeSe is the only compound that does not have the long range magnetic order that appears below the nematic ordering temperature to induce additional folding in the electronic structure and Fermi surfaces (**Figures 1B,C**). Hence FeSe provides an ideal system to study the effect of the nematic phase on the electronic structure.

Although FeSe has been studied by ARPES in great detail, the description of the electronic structure in the nematic state is still actively debated with important consequences regarding two central problems. First, while some reports conclude that the orbital anisotropy between d_{xz} and d_{yz} in FeSe has an energy scale comparable to those in the iron pnictides [15, 48–54, 60, 61], others report that it is much smaller in FeSe [55, 56]. The implication of the former is that the nematic order in FeSe is electronic in origin similar to the iron pnictides while for the latter, it is implied that the nematic order could be much more on par with the lattice distortion. The second debated question is the complete disappearance of an electron pocket in the nematic phase as observed by spectroscopy probes [56], which leads to strongly anisotropic superconducting gap [51, 53, 62]. Various scenarios have been proposed for the route in which the electron pocket disappears [62–65].

These issues are recently examined by studies on FeSe that are detwinned by gluing single crystals of FeSe on mechanically strained BaFe_2As_2 [15] (**Figures 2a–m**). The complete detwinning of FeSe via this method is demonstrated by comparing the signal from orthogonal directions of the crystal (**Figures 2a–e**). As seen at the same momentum point indicated by the arrows, the peaks from $\Gamma - M_X$ and $\Gamma - M_Y$ appear at distinct energies and the mixing of the two match that of the same measurement on a twinned crystal. Moreover, no portion of the signal from $\Gamma - M_X$ is leaked in the signal taken along $\Gamma - M_Y$. Hence the detwinning was complete. From photoemission matrix element effects [34], it was identified that the bands observed along $\Gamma - M_X$ and $\Gamma - M_Y$ originate from the d_{xz} and d_{yz} orbitals. The difference between the orbitals, as well as the anisotropy of d_{xy} orbital, cause the 50 meV energy difference at the BZ corners. [67] This result confirms that the nematic order in FeSe is electronic in origin and similar to that in the iron pnictides. Second, in addition to d_{xz} and d_{yz} , the d_{xy} orbital also shows a nematic band shift and participates in the nematic order, which could be explained by an anisotropic hopping term. Finally, the vanishing electron pocket that contains d_{xy} and d_{xz} characters in the tetragonal state disappears by shrinking in size due to a new hybridization between the d_{xy} and d_{xz} bands as they shift at the

onset of the nematic phase. Eventually deep in the nematic phase but above the superconducting phase, this electron pocket is lifted completely above the Fermi level, making way for a strongly anisotropic superconducting state.

In addition, in relation to the reversed anisotropy observed in susceptibility and resistivity mentioned above, it is interesting to note that FeSe has a much more prominent reversed orbital anisotropy at the Brillouin zone (BZ) center compared to BaFe_2As_2 , that is, the d_{xz} orbital is lifted up compared to d_{yz} , opposite to that of the large orbital anisotropy at the BZ corner [49, 68]. It is also interesting to note that a recent X-ray linear dichroism (XLD) measurement reveals a reversed orbital occupation imbalance between d_{xz} and d_{yz} for FeSe compared to BaFe_2As_2 [60].

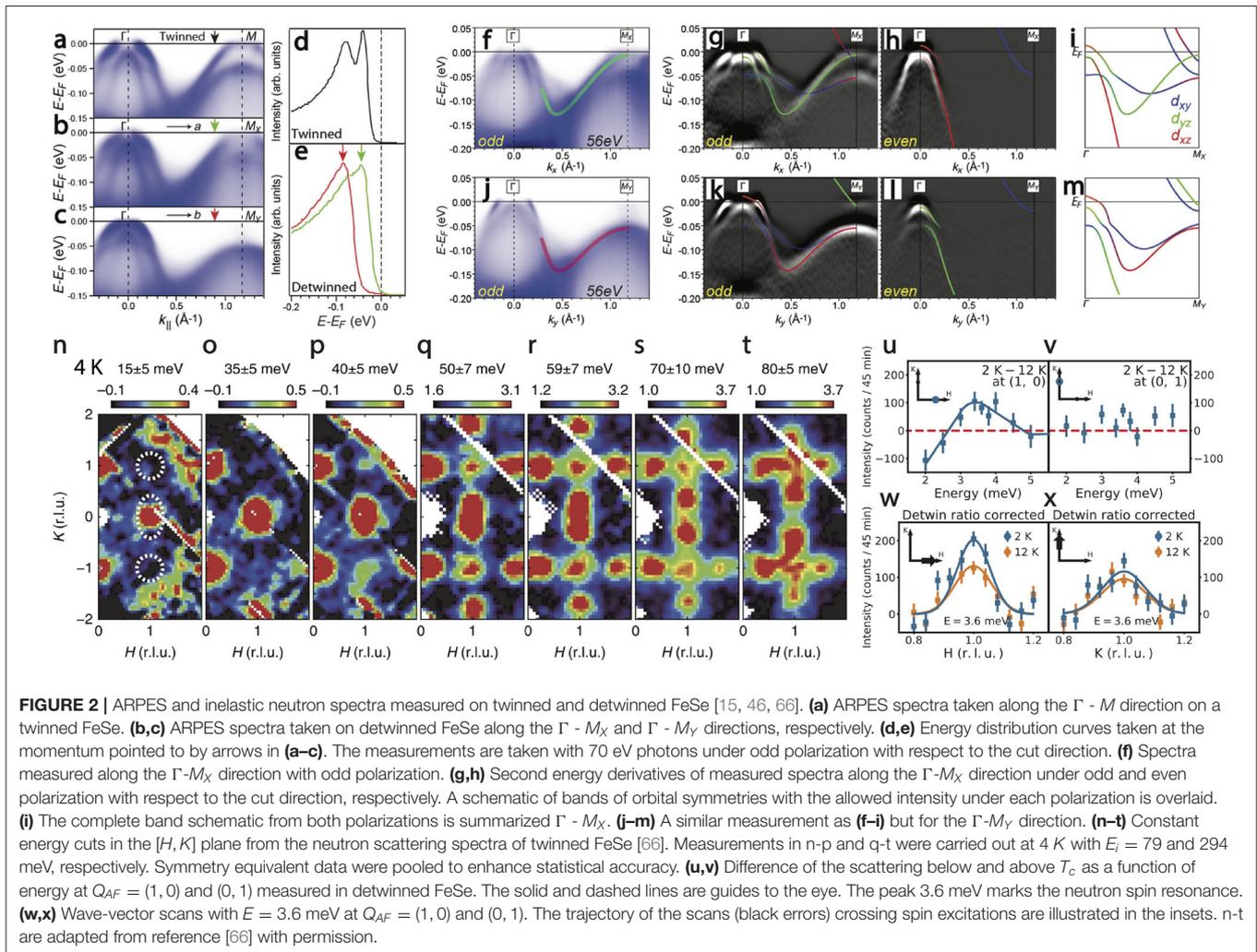
3.3. Neutron Scattering

Since superconductivity in unconventional superconductors usually emerges from AF ordered parent compounds, and a neutron spin resonance, a collective spin excitation with intensity tracking the superconducting order parameter, is widely observed by INS, magnetism is believed to be a common

thread to understand the microscopic origin of unconventional superconductivity [1, 2].

Spin excitations in parent compounds of FeSCs have been studied by neutron time-of-flight (TOF) chopper spectrometers soon after the availability of single-crystalline samples. Recently, the spin waves in fully detwinned BaFe_2As_2 are mapped out in the entire BZ using a TOF spectrometer [41]. It is shown that at low energies, spin waves are most intense at the AF wave-vectors $Q_{AF} = (\pm 1, 0)$. With increasing energy, spin waves become more transversely elongated and magnetic scattering starts to appear near $(0, \pm 1)$ to reduce the two-fold (C_2) anisotropy. Upon further increasing energy, the transverse excitations from $(\pm 1, 0)$ and $(0, \pm 1)$ merge together above 150 meV at $(\pm 1, \pm 1)$.

The neutron spin resonance, a signature of unconventional superconductivity, was studied in superconducting $\text{BaFe}_{1.915}\text{Ni}_{0.085}\text{As}_2$ [35]. INS experiments on twinned $\text{BaFe}_{1.915}\text{Ni}_{0.085}\text{As}_2$ have found order-parameter like spin resonance at wave vectors $Q_{AF} = (1, 0, 1)$ and $(0, 1, 1)$ with $E \approx 6$ meV below $T_c = 16.5$ K [69]. In the normal state in detwinned $\text{BaFe}_{1.915}\text{Ni}_{0.085}\text{As}_2$, well-defined peaks centered at both $(1, 0, 1)$ and $(0, 1, 1)$ are observed. On cooling below T_c ,



only the scattering at $(1, 0, 1)$ increases in intensity and forms a resonance, while it does not change across T_c at $(0, 1, 1)$, which shows that the spin resonance has the C_2 -symmetric, consistent with a highly anisotropic pairing state [35]. For a sample with slightly lower electron-doping, we find that spin excitations in the normal state are absent at $(0, 1, 1)$, indicating that resonance only appears at $Q_{AF} = (1, 0, 1)$ [70]. These results suggest that the superconductivity-induced resonance is orbital selective and arises from the electron-hole Fermi surface nesting of quasiparticles with the d_{yz} orbital characters [70].

Spin fluctuation spectra in twinned FeSe are similar to that of BaFe_2As_2 , as shown in **Figures 2n–t**, except that Néel spin fluctuations at $Q = (1, 1)$ coexist with the stripe spin fluctuations at $Q_{AF} = (1, 0)$ [66]. The crossover of their intensities at T_s indicates competition between each other, which is proposed to be the root cause for the absence of long-range magnetic order. In detwinned FeSe, spin excitations are most intense at $Q_{AF} = (1, 0)$ at low energies in the normal state, and the C_2 anisotropy is reduced at lower energies, 3 – 5 meV, indicating a gapped C_4 mode. At present, there are no measurement on detwinned FeSe for excitation energies above 20 meV, and it is therefore still an open question of how the anisotropy changes up to the band top in the system for a detwinned sample [46].

INS on twinned FeSe has found that superconductivity induces a spin resonance of $E = 3.6$ meV at $(1, 0)$ and $(0, 1)$ below T_c [66, 71, 72]. **Figures 2u,v** show temperature difference of spin excitations in detwinned FeSe below and above $T_c = 9$ K as a function of energy at $(1, 0)$ and $(0, 1)$, respectively [46]. While **Figure 2u** shows clear evidence of the resonance at 3.6 meV at $(1, 0)$ (negative intensities indicate the opening of a spin gap), the identical temperature difference plot (**Figure 2v**) yields no observable temperature dependence across T_c , indicating no spin resonance or spin gap at $(0, 1)$. **Figures 2w,x** show wave-vector scans after correcting for finite detwin ratio. As we can see, superconductivity induces a C_2 -

symmetric resonance on a background of C_4 -symmetric normal state scattering. These results are consistent with anisotropic superconducting gaps observed from STM measurements [62], and calculation based on anisotropic superconducting gaps from STM data can reproduce the observed anisotropic neutron spin resonance [46].

4. CONCLUSION

In this short review article, we focus on recent progress on detwinned FeSe, and compare it with detwinned electron-doped BaFe_2As_2 . Although optimal superconductivity appears at the expense of nematic and AF order in electron-doped BaFe_2As_2 and coexists with nematic order in FeSe, the basic behaviors of spin excitations in both classes of materials are similar. These results indicate that superconductivity in different families of FeSCs has the same microscopic origin, suggesting orbital selective superconductivity in the nematic region of the FeSCs.

AUTHOR CONTRIBUTIONS

The manuscript was written by TC, MY, and PD. All authors made the comments.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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