



Relationship Between Nematicity, Antiferromagnetic Fluctuations, and Superconductivity in $\text{FeSe}_{1-x}\text{S}_x$ Revealed by NMR

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The S-substituted FeSe, $\text{FeSe}_{1-x}\text{S}_x$, under pressure (p), provides a versatile platform for studying the relationship among nematicity, antiferromagnetism, and superconductivity. Here we present a short review of the recent experimental evidence showing that nematicity has a remarkable impact on the relationship between antiferromagnetic fluctuations and superconductivity. This has been revealed by several ^{77}Se nuclear magnetic resonance studies that have tracked the variability of antiferromagnetic fluctuations and superconducting transition temperature (T_c) as a function of x and p . T_c is roughly proportional to antiferromagnetic fluctuations in the presence or absence of nematic order suggesting the importance of antiferromagnetic fluctuations in the Cooper pairing mechanism in $\text{FeSe}_{1-x}\text{S}_x$. However, the antiferromagnetic fluctuations are more effective in enhancing superconductivity in the absence of nematicity as compared to when it is present. These experimental observations give renewed insights into the interrelationships between nematicity, magnetism, and superconductivity in Fe-based superconductors.

Keywords: nematicity, unconventional superconductivity, NMR, magnetic correlations, quantum materials

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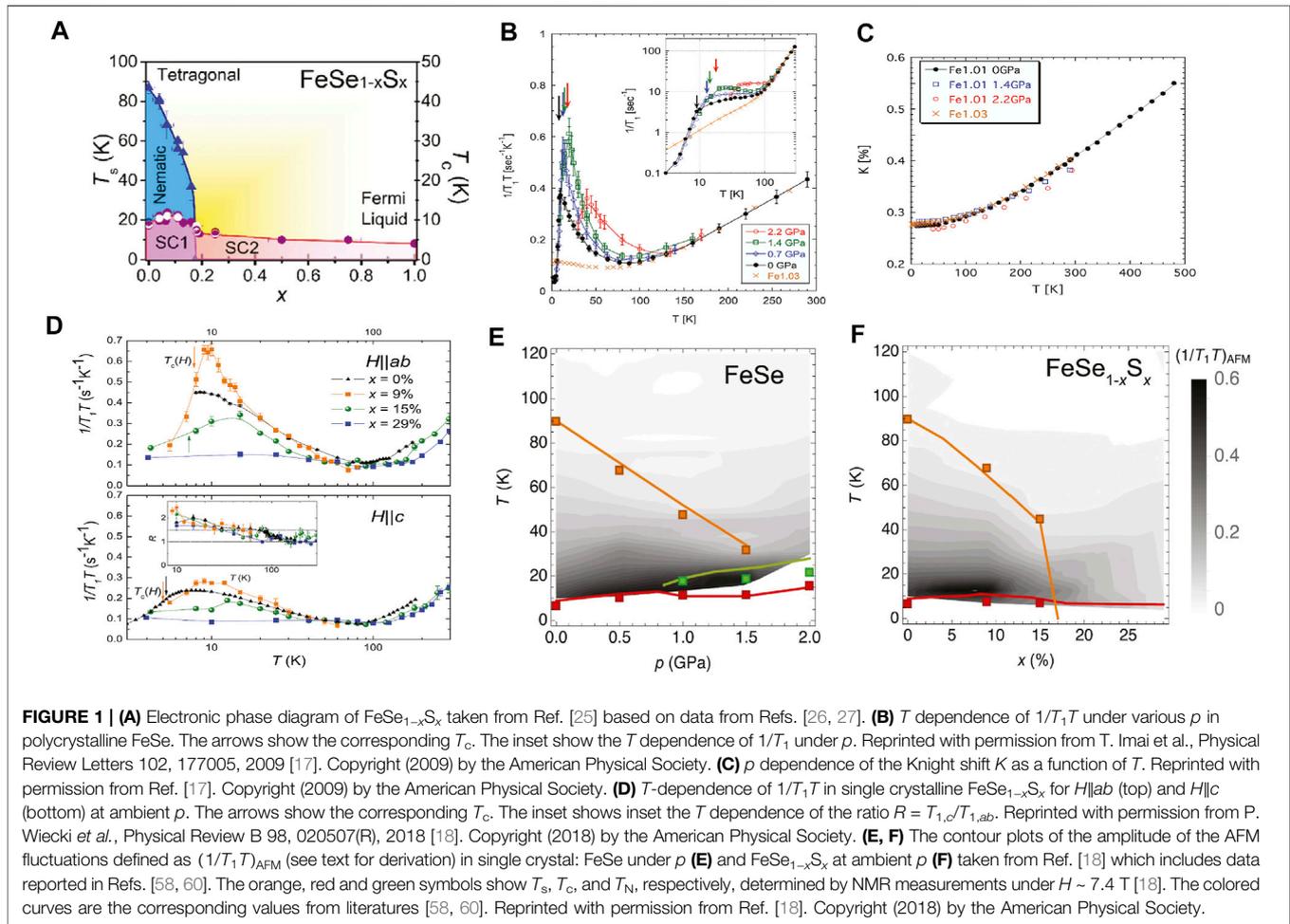
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1 INTRODUCTION

Suppressing the transition temperatures of long-range orders with a tuning parameter has led to the discovery of superconductivity (SC) in the associated quantum phase transition (QPT) regions of several classes of materials such as heavy-Fermion systems [1–3], itinerant ferromagnets [4, 5], high T_c cuprates and Fe-based superconductors [3, 6]. The quantum critical fluctuations of the suppressed long-range order parameter(s) could thus be responsible for the elusive Cooper pairing mechanism in those unconventional superconductors.

In most Fe-based superconductors, SC appears close to the quantum phase transitions of two long-range orders: the nematic order, which is an electronically driven structural transition from high-temperature tetragonal (C4 symmetry) to low-temperature orthorhombic (C2 symmetry), and the antiferromagnetic (AFM) order with spontaneously oriented electronic spins characterized by a wave vector [$\mathbf{q} = (\pi, 0)$ or $(0, \pi)$] [3, 7–9]. In those systems, the nematic transition temperature (T_n) is at or just above the Néel temperature (T_N), and both phases are simultaneously suppressed with carrier doping and/or the application of pressure (p), leading to two QPTs originating from the nematic and the AFM states. As SC in these compounds emerges around the two QPTs, AFM and nematic phases are believed to play important roles for the appearance of SC. However, the



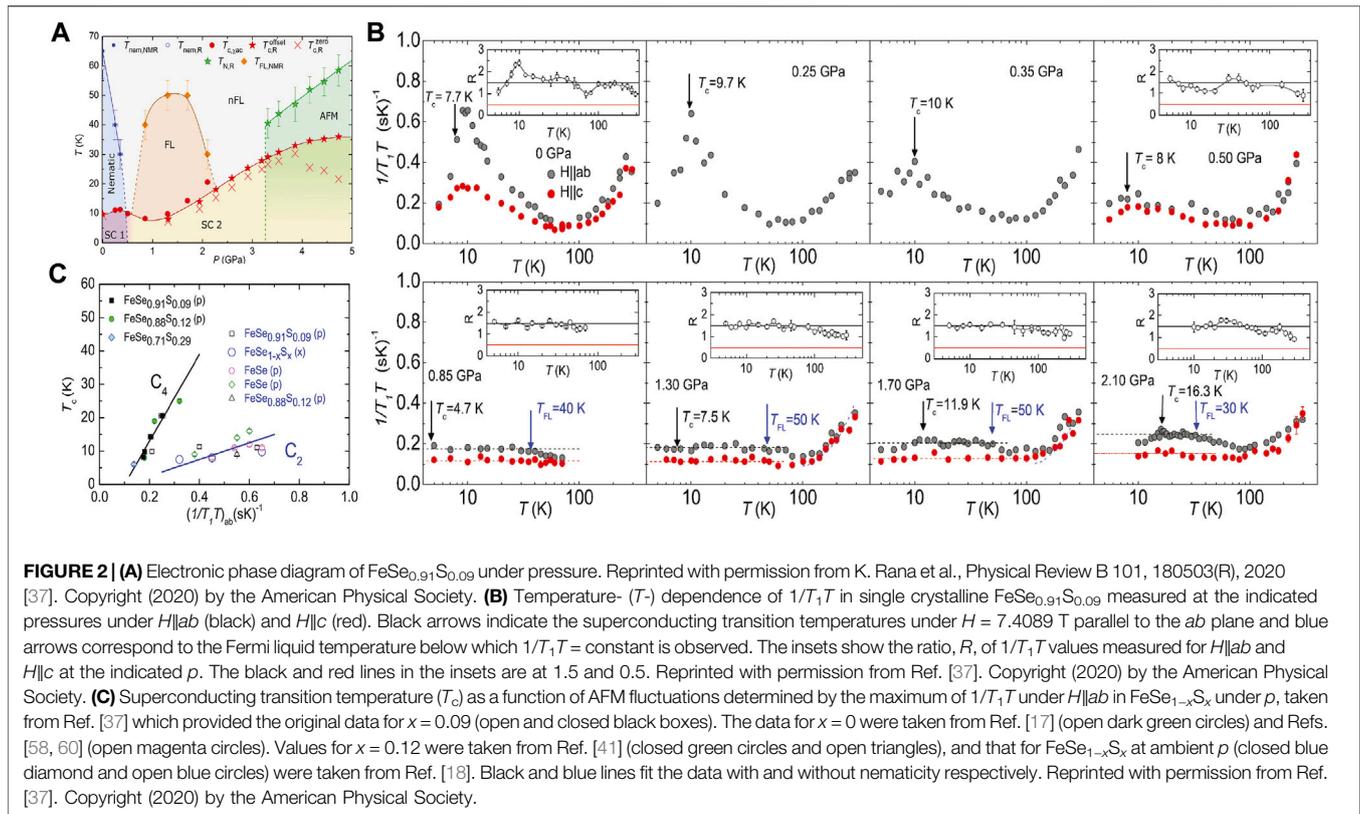
individual contribution to SC from these two phases becomes difficult to separate due to the close proximity of the two orders [10–12].

In this sense, the sulfur-substituted FeSe system, $\text{FeSe}_{1-x}\text{S}_x$, provides a favorable platform for the study of the role of nematicity or antiferromagnetism on SC independently [13]. $\text{FeSe}_{1-x}\text{S}_x$ has the simplest of crystal structures among the Fe-based superconductors, with a quasi-two dimensional $\text{FeSe}(\text{S})$ layer in the ab plane, stacked along the c axis. At $x = 0$, FeSe undergoes a nematic transition at $T_s \sim 90$ K followed by a superconducting transition at $T_c \sim 8.5$ K, but it does not show a long range AFM order at ambient p [13–16]. This allows the study of AFM fluctuations inside the nematic order and its relationship with SC [17]. The nematic phase in FeSe can be suppressed by pressure application, with T_s decreased down to 32 K at $p = 1.5$ GPa [18]. T_c shows a complex multi-domed structure with p , reaching a maximum $T_c \sim 37$ K at $p \sim 6$ GPa [19–21]. At the same time, an AFM ordered state appears above $p = 0.8$ GPa [22, 23], and T_s merges with T_N above $p = 1.7$ GPa [24], limiting the range for studying the effects of nematicity on SC without AFM state.

The nematic phase in FeSe can also be suppressed with the isoivalent S substitution for Se in $\text{FeSe}_{1-x}\text{S}_x$ as shown in **Figure 1A**

taken from Ref. [25] based on data from Refs. [26, 27], where T_s decreases to zero at the critical x value, $x_c \sim 0.17$. As no long-range AFM order appears in $\text{FeSe}_{1-x}\text{S}_x$ at ambient p , one can study the variability of T_c including near a nematic QPT without an AFM order. At x_c , diverging nematic fluctuations were reported from elasto-resistivity measurements [28], and a temperature- (T -) linear behavior of the resistivity was seen under high magnetic fields (H) [29]. As shown in **Figure 1A**, T_c first increases up to 10 K around $x = 0.09$ making a maximum and then decreases gradually at higher x without showing any clear change in T_c around x_c [18, 26, 30]. Nevertheless, the considerable change in the size and anisotropy of the SC gap is observed at the nematic QPT in spectroscopic-imaging scanning tunneling microscopy [26, 30, 31], thermal conductivity [32], and specific heat [33] measurements, implying different SC states inside (SC1) and outside (SC2) nematic states [25]. In addition, signatures of the crossover between Bardeen-Cooper-Schrieffer and Bose-Einstein-Condensate superconductivities at the nematic QPT were recently reported by laser-excited angle-resolved photoemission spectroscopy (ARPES) measurements [34].

The nematic phase in the S-substituted FeSe system is also controlled by pressure application and an AFM state appears at higher p [35–38]. The three-dimensional T - p - x phase diagram of



$\text{FeSe}_{1-x}\text{S}_x$ up to $p = 8$ GPa has been reported by Matsuura et al. [35] in which the AFM ordered phase shifts to higher p with increasing x . A typical p - T phase is shown in **Figure 2A** for the case of $x = 0.09$ [37]. In this case, with increasing p , the nematic phase disappears around $p \sim 0.5$ GPa corresponding to a putative nematic QPT, and the AFM state appears above $p \sim 3.5$ GPa. In addition to the nematic, AFM, and SC states, Fermi liquid behaviors were reported at low temperatures in $x = 0.09$ (see **Figure 2A**) [37] and 0.11 [39] after the suppression of the nematic order by applying p . The Fermi liquid phase was recently attributed to the presence of a quantum Griffiths phase close to the nematic QPT [40]. Similar to the T - x phase diagram of $\text{FeSe}_{1-x}\text{S}_x$, SC phase was shown to have two different states (SC1 and SC2) separated by the nematic QPT as shown in **Figure 2A**. Such two different SC states under p were also reported in $x = 0.11$ [39] and 0.12 [41], which is more apparent under H [41]. The presence of a series of nematic quantum phase transitions in the x - p phase diagram [35] allows the study of the correlation between T_c and AFM fluctuations in the presence and absence of the nematic order [37].

In this mini review, we show the positive correlation between AFM fluctuations and SC and the impact of nematicity on the relationship based on the nuclear magnetic resonance (NMR) studies of the $\text{FeSe}_{1-x}\text{S}_x$ system under p . After briefly introducing some basics of NMR which are used in ^{77}Se NMR studies to characterize the AFM fluctuations, we review the relationship between AFM fluctuations and SC in the presence of nematic order in FeSe under p and in $\text{FeSe}_{1-x}\text{S}_x$ at ambient p . Then, we

show the studies of $\text{FeSe}_{1-x}\text{S}_x$ system under p , where we review the relationship between AFM fluctuations and SC in the absence of nematic order. Finally, we end with a summary including the current research gaps and potential future developments in the field.

2 NUCLEAR MAGNETIC RESONANCE AND ANTIFERROMAGNETIC FLUCTUATIONS

NMR is one of the powerful techniques to study the magnetic and electronic properties of materials from a microscopic point of view and has been utilized to investigate the physical properties of Fe-based superconductors. Nuclei with finite angular momentum undergo Zeeman splitting in the presence of a magnetic field at the nuclear site (H_{nuc}). The energy difference between the nearest nuclear spin levels is given as $\Delta E = \gamma_N \hbar H_{\text{nuc}}$ where γ_N is the nuclear gyromagnetic ratio. In the NMR technique, nuclei are excited from lower energy states to higher ones by applying electromagnetic wave whose energy is equal to ΔE .

The resonance frequency is determined by H_{nuc} which is a sum of the external magnetic field (H) and the hyperfine field (H_{hf}) due to the interaction between nuclei and electrons. The shift of the resonance line due to the hyperfine interaction is defined by $K = H_{\text{hf}}/H$ which is the so-called Knight shift in metals. In general, the shift K has the T -independent orbital component, K_{orb} , and T -dependent spin component, K_s , which can be expressed as $K = K_{\text{orb}} + K_s$. K_s is proportional to the static and uniform magnetic

susceptibility (χ_s) with the wave vector $\mathbf{q} = 0$ and the frequency $\omega = 0$:

$$K_s \sim A_{hf} \chi_s(\mathbf{q} = 0, \omega = 0). \quad (1)$$

The T dependence of K_s gives us information of the magnetic properties of compounds at $\mathbf{q} = 0$. On the other hand, the nuclear relaxation rate ($1/T_1$) divided by T , $1/T_1T$, is sensitive to the \mathbf{q} -sum of the imaginary part of susceptibility ($\chi''(\mathbf{q}, \omega_N)$) at the NMR frequency (ω_N) [42] and is given as

$$1/T_1T \sim \gamma_N^2 k_B \sum_{\mathbf{q}} |A(\mathbf{q})|^2 \chi''(\mathbf{q}, \omega_N) / \omega_N \quad (2)$$

where $A(\mathbf{q})$ is the \mathbf{q} -dependent hyperfine form factor. $1/T_1T$ gives us information about the total magnetic correlations at all \mathbf{q} values. Therefore, one can obtain important insights about \mathbf{q} dependent magnetic correlations by comparing K_s and $1/T_1T$ data.

In simple metals, K_s is related to the density of states at the Fermi energy [$\mathcal{N}(E_F)$] where $K_s = A_{hf} (g\mu_B)^2 \mathcal{N}(E_F)/2$, and $1/T_1T$ is proportional to the square of $\mathcal{N}(E_F)$ as $1/T_1T = \pi\hbar A_{hf}^2 (g\mu_B)^2 \gamma_N^2 \mathcal{N}^2(E_F) k_B$ [43]. In a Fermi liquid picture, the ratio $S \equiv T_1TK_s^2$ becomes a constant [42, 44] which is called the Korringa relation. In real materials, an experimentally determined value of $T_1TK_s^2$ may deviate from S due to electron correlations. Thus, the deviation parameter defined as $\alpha = S/(T_1TK_s^2)$ provides information about electron correlations in materials. When AFM fluctuations are present, $\chi''(\mathbf{q}, \omega_N)$ with $\mathbf{q} \neq 0$ is enhanced with little or no effect on K_s which probes only the $\mathbf{q} = 0$ component of χ_s . Therefore, $1/T_1T$ is enhanced much higher than K_s and α becomes greater than unity. On the other hand, $\alpha < \text{unity}$ is expected for ferromagnetic correlations.

When the Korringa relation does not hold due to strong magnetic fluctuations (non-Fermi liquid picture), the T dependence of $1/T_1T$ could be different from that of K . When strong AFM fluctuations exist in systems, the contribution to $1/T_1T$ from AFM fluctuations will be the source of the different T dependence, and the experimentally observed $1/T_1T$ is sometimes decomposed as $1/T_1T = (1/T_1T)_{AFM} + (1/T_1T)_{\mathbf{q}=0}$ [18, 45, 46]. Here $(1/T_1T)_{AFM}$ denotes the AFM contributions from $\chi(\mathbf{q} \neq 0, \omega_N)$ and $(1/T_1T)_{\mathbf{q}=0}$ represents the contributions from $\mathbf{q} = 0$ components. By assuming $(1/T_1T)_{\mathbf{q}=0} = CK_s^2$, where C is the empirically determined proportionality constant, one can extract the AFM contribution to $1/T_1T$ by subtracting $(1/T_1T)_{\mathbf{q}=0}$ from the observed $1/T_1T$, providing insights into the magnetic fluctuations.

In the case of Fe-based superconductors, Kitagawa et al. proposed that anisotropy in $1/T_1$ at the chalcogen or pnictogen sites provides more detailed information about AFM fluctuations [47]. According to them, the ratio of $1/T_1$ values measured under H parallel to c axis ($1/T_{1,c}$) and parallel to ab plane ($1/T_{1,ab}$) [$R \equiv T_{1,c}/T_{1,ab}$] can determine the dominant \mathbf{q} for AFM fluctuations. In the case of isotropic AFM fluctuations, $R = 1.5$ is expected for stripe-type AFM fluctuations with $\mathbf{q} = (\pi, 0)$ or $(0, \pi)$, whereas when Néel type AFM fluctuations with $\mathbf{q} = (\pi, \pi)$ are present, $R = 0.5$. Such

analysis has been extensively used in Fe-based superconductors [18, 37, 47–51] and related materials [52, 53] to characterize the AFM fluctuations in those systems.

3 ANTIFERROMAGNETIC FLUCTUATIONS AND SUPERCONDUCTIVITY WITH NEMATICITY

Soon after the discovery of the Fe-based superconductors [54, 55], ⁷⁷Se ($I = 1/2$, $\gamma_N/2\pi = 8.1432$ MHz) NMR studies on polycrystalline FeSe were carried out [17, 56] and the importance of AFM fluctuations for superconductivity has been pointed out. **Figure 1B** shows the T dependence of $1/T_1T$ values in FeSe under various pressures reported by Imai et al. [17]. At higher temperatures above $T \sim 100$ K, $1/T_1T$ at all pressures decreases with decreasing T . This behavior is similar to the T -dependence of K shown in **Figure 1C** where K shows a monotonic decrease when cooling from 480 to ~ 100 K. The variations in both $1/T_1T$ and K above ~ 100 K were explained in terms of spin gap formation or a peculiar band structure near the Fermi level [57]. However, upon cooling below $T \sim 100$ K, the T dependences of $1/T_1T$ and K show quite different behaviors. Although K is nearly independent of both T and p below 50 K, $1/T_1T$ shows strong enhancements at all measured pressures at low temperatures where peaks are observed at the p -dependent T_c or T_N . As described above, K is proportional to $\chi(0, 0)$ and $1/T_1T$ reflects the T dependence of \mathbf{q} -summed $\chi''(\mathbf{q}, \omega_N)$. Therefore, the enhancements of $1/T_1T$ at low temperatures unequivocally establish the presence of AFM fluctuations at the T region, suggesting that the AFM fluctuations are relevant to the SC in FeSe. In fact, a close relationship between the AFM fluctuations and SC has been pointed out from the p dependences of T_c and $1/T_1T$ data: the maximum of $1/T_1T$ increases along with T_c as shown in **Figure 1B** where T_c s at different p are marked by downward arrows [17]. Broad humps in $1/T_1T$ observed at temperatures much higher than their respective T_c values at $p = 1.4$ and 2.2 GPa are due to magnetic orderings. It should be noted that, due to the occurrence of the AFM order under high pressures in FeSe, the relationship between T_c and the maximum of $1/T_1T$ can only be compared at low pressures in this system. A later single crystalline ⁷⁷Se NMR studies under $H\parallel ab$ and $H\parallel c$ characterized the AFM order and the AFM fluctuations at higher pressures to be of stripe type [51, 58].

A ⁷⁷Se NMR study of single crystalline FeSe_{1-x}S_x by Wiecki et al. [18] at ambient p also provided clear experimental evidence of the close relationship between the AFM fluctuations and SC in this system. **Figure 1D** show the T dependence of $1/T_1T$ in FeSe_{1-x}S_x for $H\parallel ab$ (upper) and $H\parallel c$ (lower), respectively, at ambient p [18], which includes the data from Ref. [58]. As in FeSe, K for all x shows monotonic decreases when lowering T from room T down to ~ 100 K, before leveling off at constant values [18] for both $H\parallel ab$ and $H\parallel c$. Although $1/T_1T$ showed a similar T dependence as K in all cases above 100 K, $1/T_1T$ shows a strong upturn below $T \sim 100$ K due to the growth of AFM fluctuations. The AFM fluctuations appear below 100 K for all samples of $x = 0, 0.09, 0.15$, and 0.29, however, the enhancement of the AFM fluctuations shows a strong x dependence. For x less

than $x_c \sim 0.17$, $1/T_1T$ increases with decreasing T showing a Curie-Weiss-like behavior expected for two-dimensional AFM fluctuations from the self-consistent renormalization theory [42]. On the other hand, for $x = 0.29$ greater than x_c , a subtle upturn on cooling below $T \sim 100$ K is observed, suggesting the tiny growth of the AFM fluctuations, followed by a nearly T independent behavior below $T \sim 25$ K without showing clear Curie-Weiss-like behaviors. At all measured x values, the ratios $R \equiv T_{1,c}/T_{1,ab}$ are found to be ~ 1.5 below $T \sim 100$ K shown in the inset of the lower panel of **Figure 1D**, indicating that the AFM fluctuations are characterized to be stripe type and do not change with x .

The x - T phase diagram (**Figure 1A**) of FeSe_{1-x}S_x at ambient p allowed Wiecki et al. to examine the correlation between AFM fluctuations and T_c , and it was shown to persist, despite the presence of a nematic QPT isolated from an AFM order. The maximum values of $1/T_1T$ first increased when x was changed from 0 to 0.09, then decreased for $x = 0.15$ and higher, similar to the x dependence of T_c shown in **Figure 1A**. **Figures 1E,F** taken from Ref. [18] are contour plots of the magnitude of AFM fluctuations determined by $1/T_1T$ data in FeSe under p (E) and in FeSe_{1-x}S_x at ambient p (F), respectively, along with their respective phase diagrams. It can be seen that T_c is enhanced at the p or x values where AFM fluctuations are stronger. This indicates the correlation between T_c and AFM fluctuations in both cases and also demonstrates the primary importance of AFM fluctuations to SC in FeSe_{1-x}S_x. It was also pointed out that, although nematic fluctuations are most strongly enhanced near the nematic QCP at $x \sim 0.17$ in the case of FeSe_{1-x}S_x, no clear correlation with T_c was observed [18].

4 ANTIFERROMAGNETIC FLUCTUATIONS AND SUPERCONDUCTIVITY WITHOUT NEMATICITY

With the firm establishment of the correlation between AFM fluctuations and SC in FeSe_{1-x}S_x, the question then arose about the role of nematicity on the relationship. As described above, FeSe_{1-x}S_x provides a suitable platform for the study of the role of nematicity on the relationship by changing samples as reported by Wiecki et al. [18]. The application of pressure on FeSe_{1-x}S_x also provides a versatile opportunity to study the effect of nematicity on the relationship. This has an advantage because p is known as one of the clean tuning parameters which control the ground state without changing the composition avoiding any additional effects of S substitutions such as homogeneity by changing x . Several ⁷⁷Se NMR studies on single crystalline FeSe_{1-x}S_x under pressure have been carried out [37, 38, 41, 59]. Here we show the results of NMR measurements under pressure up to 2.1 GPa on $x = 0.09$ whose p - T phase diagram is shown in **Figure 2A** reported in Ref. [37]. With p , the nematic phase is suppressed and disappears around the critical pressure $p_c \sim 0.5$ GPa, and an AFM state appears above 3 GPa with a dome-shaped Fermi-liquid phase between nematic and AFM phases. T_c shows a clear p dependence with a double dome structure with and without long-range nematicity, making the system suitable in investigating the role of nematicity on the relationship.

Figure 2B shows the T dependence of $1/T_1T$ for $x = 0.09$ under $H\parallel ab$ (black) and $H\parallel c$ (red) at several pressures, taken from the study by Rana et al [37]. Below $p_c = 0.5$ GPa, with decreasing T , $1/T_1T$ increases below ~ 70 K showing Curie-Weiss like behavior originating from two dimensional AFM fluctuations and starts to decrease around T_c (T_c for $H\parallel ab$ are shown by black arrows in the figures). On the other hand, above 0.5 GPa, $1/T_1T$ exhibits quite different temperature dependences in comparison with those observed at low pressures. Although $1/T_1T$ is slightly enhanced below ~ 70 K, indicating the existence of the AFM spin fluctuations, $1/T_1T$ s are nearly constant exhibiting the so-called Korringa behavior, expected for Fermi-liquid state below the temperature (defined as T_{FL}) marked by blue arrows. Thus the results indicate that the nature of AFM fluctuations changes below and above $p_c = 0.5$ GPa in FeSe_{0.91}S_{0.09}.

Similar T dependences of $1/T_1T$ have also been reported in ⁷⁷Se NMR studies of FeSe_{1-x}S_x by Kuwayama et al. [38, 41] under p up to 3.9 GPa. The authors pointed out that AFM fluctuations with different \mathbf{q} vectors may be responsible for the two distinct SC domes [41]. However, Rana et al found that the AFM fluctuations are characterized to be stripe type and p independent by showing the fact that the ratios R are close to ~ 1.5 at low temperatures for all measured p shown in the insets of **Figure 2B**.

Then what is the difference in the nature of AFM fluctuations in the presence and absence of nematic order? The idea that nematicity changes the relationship between T_c and AFM fluctuations was proposed by Rana et al [37] and can be clearly seen in **Figure 2C** taken from that study. Here, in the x axis, the maximum values of $1/T_1T$ with $H\parallel ab$ were taken as a representative of the magnitude of AFM fluctuations for different values of x and p in FeSe_{1-x}S_x. The corresponding x and p dependent T_c values were plotted in the y axis. The data included for $x = 0, 0.12$ and 0.29 were taken from Refs. [17, 58, 60], Ref. [41] and Ref. [18], respectively, while those for $x = 0.09$ were reported by Ref. [37]. These experimental data were classified into two groups: one that includes the data points where T_c and AFM fluctuations are in the nematic order, and another that includes those measured in the absence of nematic order. The slope for the linear fitting of the data points in the absence of nematicity was higher by a factor of ~ 5 compared to the slope for the linear fitting of those in the presence of nematicity. The results indicate that, for example, T_c is less sensitive to the strength of spin fluctuations in the tetragonal phase of FeSe_{1-x}S_x at ambient pressure for $x < 0.17$ while it is largely enhanced in the orthorhombic phase of FeSe_{0.91}S_{0.09} above 0.5 GPa even with a small increase in AFM fluctuations. When nematicity is absent, the AFM fluctuations in this system are present at both the wave vectors $\mathbf{q} = (\pi, 0)$ and $\mathbf{q} = (0, \pi)$ due to the four-fold rotational symmetry (C4) of the tetragonal state. However, in the presence of nematicity, the rotational symmetry is reduced to two-fold rotational symmetry (C2) and the AFM fluctuations are present at only one of the wave vectors, either vector $\mathbf{q} = (\pi, 0)$ or $\mathbf{q} = (0, \pi)$ [61, 62]. Based on those results, Rana et al. pointed out that the AFM fluctuations with C4 symmetry are more effective in enhancing T_c for the FeSe_{1-x}S_x system.

5 SUMMARY

We presented a brief overview of ⁷⁷Se NMR studies in FeSe_{1-x}S_x at ambient pressure and under pressure, especially focusing on the role of nematicity on the relationship between superconducting transition temperature T_c and antiferromagnetic (AFM) fluctuations. It was shown that T_c has a positive relationship with AFM fluctuations, suggesting the importance of AFM fluctuations in the pairing mechanism of superconducting electrons in FeSe_{1-x}S_x. Furthermore, nematicity is found to play a central role on the positive relationship. In the absence of nematic order, T_c can be greatly enhanced by AFM fluctuations. When the nematic order is present, this enhancement decreases by a factor of ~ 5 . The evidence of the impact of nematicity on the relationship between superconductivity and AFM fluctuations has emerged from various ⁷⁷Se NMR studies in the FeSe_{1-x}S_x system under pressure.

Although the findings provide a renewed insight on the relationships between nematicity, magnetism, and unconventional superconductivity in Fe-based superconductors, the origin for the strong impact of nematicity on the relationship between T_c and AFM fluctuations is still an open question. Further detailed experimental as well as theoretical investigations of the underlying reason behind the impact of nematicity on the relationships between superconductivity and AFM fluctuations

would bring us a step towards understanding the physical mechanism behind unconventional superconductivity.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct, and intellectual contribution to the work and approved it for publication.

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