






Impact of nematicity on the relationship between antiferromagnetic fluctuations and superconductivity in $\text{FeSe}_{0.91}\text{S}_{0.09}$ under pressure

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The sulfur-substituted FeSe system, $\text{FeSe}_{1-x}\text{S}_x$, provides a versatile platform for studying the relationship among nematicity, antiferromagnetism, and superconductivity. Here, by nuclear magnetic resonance (NMR) and resistivity measurements up to 4.73 GPa on $\text{FeSe}_{0.91}\text{S}_{0.09}$, we established the pressure- (p -) temperature (T) phase diagram in which the nematic state is suppressed with pressure showing a nematic quantum phase transition (QPT) around $p = 0.5$ GPa, two superconductivity (SC) regions separated by the QPT appear, and antiferromagnetic (AFM) phase emerges above ~ 3.3 GPa. From the NMR results up to 2.1 GPa, AFM fluctuations are revealed to be characterized by the stripe-type wave vector which remains the same for the two SC regions. Furthermore, the electronic state is found to change in character from non-Fermi liquid to Fermi liquid around the nematic QPT and persists up to ~ 2.1 GPa. In addition, although the AFM fluctuations correlate with T_c in both SC states, demonstrating the importance of the AFM fluctuations for the appearance of SC in the system, we found that, when nematic order is absent, T_c is strongly correlated with the AFM fluctuations whereas T_c weakly depends on the AFM fluctuations when nematic order is present. Our findings on $\text{FeSe}_{0.91}\text{S}_{0.09}$ were shown to be applied to the whole $\text{FeSe}_{1-x}\text{S}_x$ system and provide an insight into the relationship between AFM fluctuations and SC in Fe-based superconductors.

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The interplay among magnetic fluctuations, electronic nematicity, and the unconventional nature of superconductivity (SC) has received wide interest after the discovery of high- T_c SC in iron pnictides [1]. In most of the iron pnictide superconductors, by lowering the temperature, the crystal structure changes from high-temperature tetragonal (HTT), C_4 symmetry, to low-temperature orthorhombic (LTO), C_2 symmetry, at, or just above, a system-dependent Néel temperature T_N , below which long-range stripe-type antiferromagnetic (AFM) order emerges [2–5]. SC in these compounds emerges upon suppression of both the structural (or nematic) and the magnetic transitions by carrier doping and/or the application of pressure (p). Although this clearly suggests a close relationship between AFM and nematic phases, the individual contribution to SC from these two phases becomes difficult to separate.

In this context, the sulfur-substituted FeSe system, $\text{FeSe}_{1-x}\text{S}_x$, provides a favorable platform for the study of the impact of nematicity or antiferromagnetism on SC independently [6]. The superconductor FeSe ($x = 0$) with a critical temperature of $T_c = 8.5$ K exhibits only a HTT-LTO structural phase transition, corresponding to a nematic phase transition, at $T_{\text{nem}} = 90$ K without AFM ordering under ambient pressure [6–8]. With increasing x , the nematic phase is suppressed, and a nematic quantum phase transition (QPT) was reported to be around $x = 0.17$ [9]. In contrast, T_c first increases from $T_c = 8.5$ K up to 10 K around $x = 0.09$ [10–12] then is suppressed at higher x , whereas the fully replaced FeS is still

a superconductor with $T_c = 5$ K [13]. As in the case of FeSe, no AFM state has been observed in $\text{FeSe}_{1-x}\text{S}_x$ at ambient pressure, making this a suitable system to study the effects of nematicity on SC [10–12]. Spectroscopic-imaging scanning tunneling microscopy [14], thermal conductivity, and specific heat [15] showed that the gap anisotropy and its size change drastically at the nematic QPT. Shubnikov–de Haas oscillation measurements indicate a change in both the topology of the Fermi surface and the degree of electronic correlations across the nematic QPT [16]. These results suggest that the presence or absence of nematicity results in two distinct superconducting states. Although no AFM state is observed in $\text{FeSe}_{1-x}\text{S}_x$ under ambient pressure, the correlations between T_c and AFM fluctuations have been pointed out from nuclear magnetic resonance (NMR) measurements [17,18].

With the application of pressure on $\text{FeSe}_{1-x}\text{S}_x$, the nematic state can also be suppressed, and an AFM state is induced [19,20]. The three-dimensional T - p - x phase diagram up to $p = 8$ GPa has been reported by Matsuura *et al.* [19] in which the AFM ordered phase shifts to higher p with increasing x , although a different phase diagram of $\text{FeSe}_{0.89}\text{S}_{0.11}$ having a wide AFM region was recently reported [21]. Recent resistivity measurements under high magnetic fields on $\text{FeSe}_{0.89}\text{S}_{0.11}$ under pressure reported a lack of nematic quantum criticality and the presence of Fermi-liquid behavior [22]. In addition, two SC domes separated by the nematic QPT under magnetic field have been reported in $\text{FeSe}_{1-x}\text{S}_x$ with $x = 0.12$ [23] and 0.11 [22] under pressure, which was not reported in the first

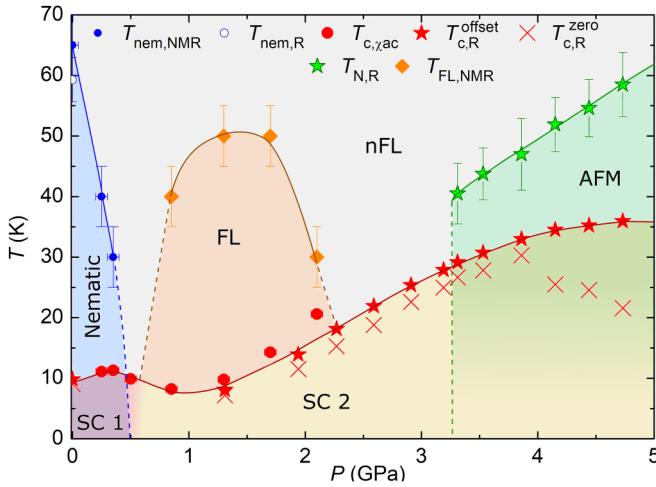


FIG. 1. p - T phase diagram of $\text{FeSe}_{0.91}\text{S}_{0.09}$. The nematic transition temperatures $T_{\text{nem,NMR}}$ and $T_{\text{nem,R}}$ are determined by the splitting of the NMR spectrum under $H \parallel ab$ and resistivity measurements at $H = 0$, respectively. $T_{c,\chi_{ac}}$ (red solid circles) denotes T_c under zero magnetic field, determined by *in situ* ac susceptibility measurements using the NMR coil. $T_{c,R}^{\text{zero}}$ (red stars) and $T_{c,R}^{\text{offset}}$ (crosses) denote T_c at $H = 0$ determined by zero resistivity and the offset point, respectively, in resistivity measurements (see the Supplemental Material [24]). The AFM transition temperature (T_N) was determined by resistivity measurements (see the Supplemental Material [24]). $T_{\text{FL,NMR}}$ represents a crossover temperature between non-Fermi-liquid (nFL) and Fermi-liquid (FL) states determined by $1/T_1$ measurements: Curie-Weiss-like behavior of $1/T_1 T$ for nFL and $1/T_1 T = \text{constant}$ Korringa behavior for FL. The solid and dotted lines are guides for the eyes.

phase diagram [19]. To clarify this, it is crucial to establish the p - T phase diagram and to investigate the change in the character of AFM fluctuations and its relationship with SC across a nematic QPT in $\text{FeSe}_{1-x}\text{S}_x$ under pressure.

In this Rapid Communication, we have carried out NMR and resistivity measurements on $\text{FeSe}_{0.91}\text{S}_{0.09}$ under pressure to investigate its physical properties from a microscopic point of view, especially focusing on the differences in the AFM fluctuations between the two different SC domes and their relationship with T_c . Based on the present NMR and resistivity data (see the Supplemental Material [24]), we established the phase diagram as a function of p shown in Fig. 1. Similar to the cases of $x = 0.11$ and 0.12 , a double SC dome structure is observed. From the temperature dependence of nuclear spin-lattice relaxation rate ($1/T_1$), we found a crossover from nFL to FL states with pressure and a dome-shaped FL phase between nematic and AFM phases. In addition, although we inferred that the wave vector of AFM fluctuations is a stripe type for both superconducting domes and does not change with pressure, the symmetry (C_4 or C_2) of the AFM fluctuations has been revealed to play an important role for superconducting transition temperature.

Single crystals of $\text{FeSe}_{0.91}\text{S}_{0.09}$ were prepared using the vapor transport method as outlined in Ref. [32]. The details of the single crystals used for NMR measurements were described in Ref. [17]. NMR measurements of ^{77}Se nuclei ($I = 1/2$, $\gamma_N/2\pi = 8.1432$ MHz) under a fixed magnetic

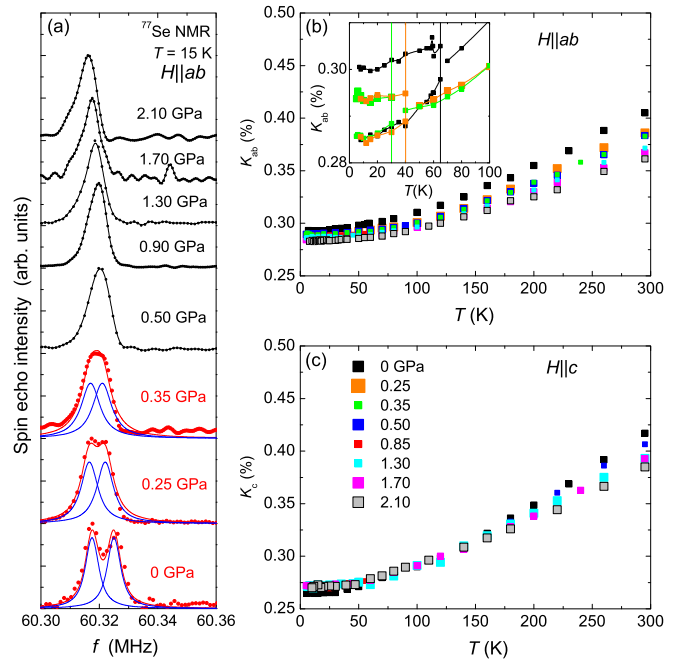


FIG. 2. (a) Pressure dependence of ^{77}Se NMR spectra of $\text{FeSe}_{0.91}\text{S}_{0.09}$ at 15 K for $H \parallel ab$. Below 0.50 GPa, the clear double peak structures (shown in red) are observed due to nematic phase transition, which can be well reproduced by the two Lorentzian curves shown in blue. (b) Temperature dependence of ^{77}Se NMR Knight-shift (K) at various pressures with $H \parallel ab$. When splitting of the line was present in the nematic state, the average values of K were plotted. The inset shows K values for the split lines below the nematic temperatures. (c) K for all measured pressures with $H \parallel c$. For this H direction, no splitting of spectra was observed.

field of $H = 7.4089$ T [33] have been carried out by using a laboratory-built spin-echo spectrometer up to a pressure of 2.10 GPa with a NiCrAl/CuBe piston-cylinder cell using Daphne 7373 as the pressure transmitting medium. Pressure calibration was accomplished by ^{63}Cu nuclear quadrupole resonance in Cu_2O [34,35] at 77 K. Resistivity measurements under higher pressures up to 4.73 GPa were carried out in a modified Bridgeman-anvil-type cell [25] using a 1:1 mixture of isopentane:*n*-pentane as the pressure medium.

Figure 2(a) shows the ^{77}Se NMR spectra of $\text{FeSe}_{0.91}\text{S}_{0.09}$ measured at 15 K under various pressures ($p = 0$ –2.10 GPa) with H parallel to the ab plane ($H \parallel ab$). Here, we applied a magnetic field along the [110] direction in the HTT phase. As reported in Ref. [17], a clear splitting of the line due to nematic order is observed at ambient pressure below $T_{\text{nem}} \sim 60$ K (see the Supplemental Material [24]).

Although the splitting becomes small with increasing p , the two-peak structure can be observed up to 0.35 GPa as shown in red in Fig. 2(a) where the spectra are well reproduced by the sum of two peaks shown in blue, evidencing the nematic order up to 0.35 GPa. On the other hand, no clear splitting of the line can be observed above 0.5 GPa. Even at $T = 4$ K, we do not observe the splitting, indicating no nematic order above 0.5 GPa. From the smooth extrapolation of the p dependence of T_{nem} described below [also, see Fig. 1], we found a nematic QPT around 0.5 GPa in $\text{FeSe}_{0.91}\text{S}_{0.09}$.

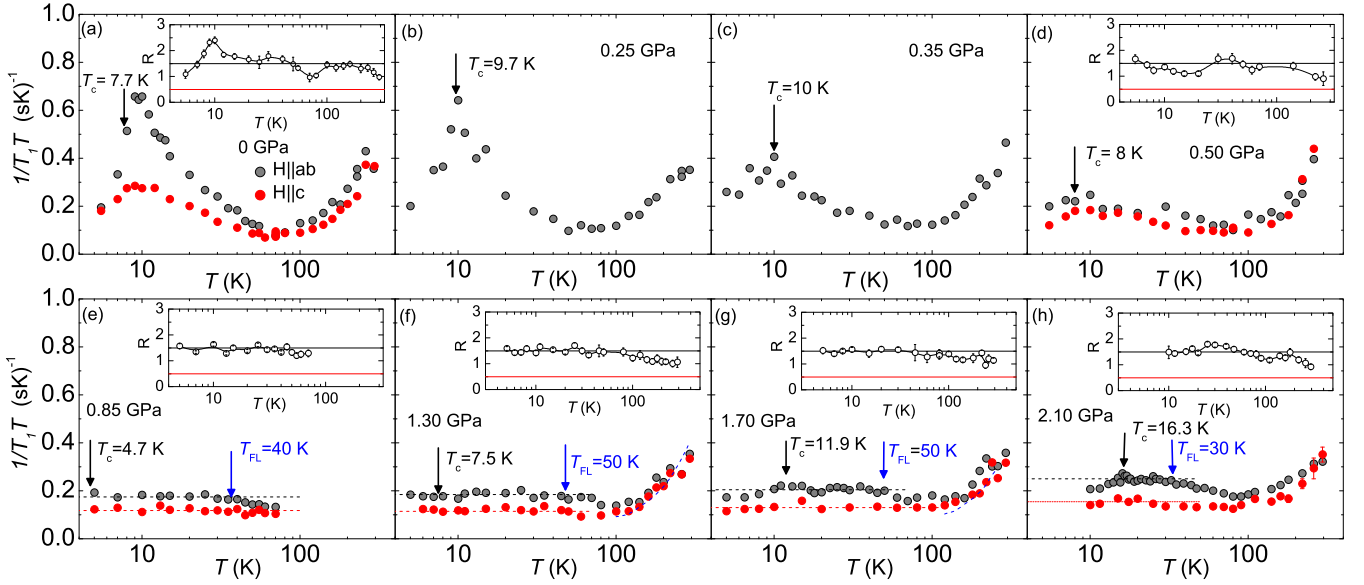


FIG. 3. Temperature dependence of ^{77}Se NMR $1/T_1T$ at various pressures with $H \parallel ab$ (gray circles) and $H \parallel c$ (red circles). Black arrows show T_c under $H \parallel ab = 7.4089$ T determined by the *in situ* ac susceptibility measurements. Blue arrows show the temperature below which $1/T_1T = \text{constant}$ behavior is observed, defined as T_{FL} . The inset of each panel shows the temperature dependence of the ratio $R \equiv (1/T_1T)_{ab}/(1/T_1T)_c$. The two horizontal lines represent the expected values for stripe-type ($R = 1.5$) and Néel-type ($R = 0.5$) AFM fluctuations, respectively.

Figures 2(b) and 2(c) show the temperature dependence of the Knight-shift (K) for $H \parallel ab$ and H parallel to the c axis ($H \parallel c$), respectively. The inset in Fig. 2(b) shows two values of K for the two peaks observed in the nematic state from which T_{nem} is determined to be ~ 65 , ~ 40 , and ~ 30 K for ambient, 0.25, and 0.35 GPa, respectively. The estimated values of T_{nem} are consistent with the previous report [20]. In the main panel of Fig. 2(b), the average values of K for the two peaks were plotted. When $H \parallel c$, no splitting of the line was observed. Throughout all pressures and both H directions, the values of K are nearly independent of p , although K seems to be suppressed very slightly with p (see the Supplemental Material [36]). As shown, K values are nearly constant below ~ 50 K and then increase with temperature above 100 K.

The nearly p -independent behavior of K indicates that static uniform magnetic susceptibility is nearly independent of p , especially at low temperatures. This also suggests that the application of pressure up to 2.10 GPa does not produce significant change in the density of states at the Fermi energy $N(E_F)$ [37], even though T_c varies significantly. This is in contrast to conventional BCS superconductors in which $N(E_F)$ generally correlates with T_c . These results strongly indicate that AFM fluctuations play an important role in the appearance of SC in $\text{FeSe}_{1-x}\text{S}_x$ as will be discussed below.

Figures 3(a)–3(h) show the temperature dependence of $1/T_1T$ at various pressures for $H \parallel ab$ (gray circles) and $H \parallel c$ (red circles). First, let us discuss the temperature dependence of $1/T_1T$ measured for $H \parallel ab$, $(1/T_1T)_{ab}$. In general, $1/T_1T$ is related to the dynamical magnetic susceptibility as $1/T_1T \sim \gamma_N^2 k_B \sum_{\mathbf{q}} |A(\mathbf{q})|^2 \chi''(\mathbf{q}, \omega_N)/\omega_N$, where $A(\mathbf{q})$ is the wave-vector \mathbf{q} -dependent form factor and $\chi''(\mathbf{q}, \omega_N)$ is the imaginary part of $\chi(\mathbf{q}, \omega_N)$ at the Larmor frequency ω_N [38]. Therefore, by comparing the temperature dependences

between $1/T_1T$ and K which measures the $\mathbf{q} = 0$ uniform magnetic susceptibility, one can obtain information on the temperature evolution of $\sum_{\mathbf{q}} \chi''(\mathbf{q}, \omega_N)$ with respect to that of $\chi'(0, 0)$. Above ~ 100 K, $(1/T_1T)_{ab}$ shows a similar T dependence as K for all measured pressures. On the other hand, below ~ 70 K, the temperature dependence of $(1/T_1T)_{ab}$ clearly deviates from that of K , although the enhancement of $(1/T_1T)_{ab}$ becomes less pronounced at higher pressures. This deviation of $1/T_1T$ at low T , therefore, evidences the existence of AFM fluctuations with $\mathbf{q} \neq 0$.

Below 0.5 GPa, with decreasing T , $(1/T_1T)_{ab}$ increases below ~ 70 K and starts to decrease around T_c , making a broad maximum. T_c 's for $H \parallel ab$ are shown by black arrows. The Curie-Weiss-like behavior of $(1/T_1T)_{ab}$ above the maxima can be associated with two-dimensional AFM fluctuations [17,23].

On the other hand, above 0.5 GPa, $(1/T_1T)_{ab}$ exhibits quite different temperature dependence in comparison with those observed at low pressures. Although $(1/T_1T)_{ab}$ is slightly enhanced below ~ 70 K, indicating the existence of the AFM spin fluctuations, we observe $1/T_1T = \text{constant}$, so-called Korringa behavior, expected for the Fermi-liquid state such as exchange enhanced metals [38,39] below the temperature (defined as T_{FL}) marked by blue arrows. T_{FL} seems to increase from 40 K at $p = 0.9$ GPa to 50 K at 1.70 GPa and then decreases to 30 K at 2.10 GPa. The suppression of T_{FL} at higher pressure may be due to the appearance of the AFM state under high pressures. It is important to point out that our NMR data do not indicate any quantum critical behavior due to nematicity around 0.5 GPa. These results seem to be consistent with the recent resistivity studies under high magnetic fields [22] which reported a lack of nematic quantum criticality and the presence of FL behavior in $\text{FeSe}_{0.89}\text{S}_{0.11}$

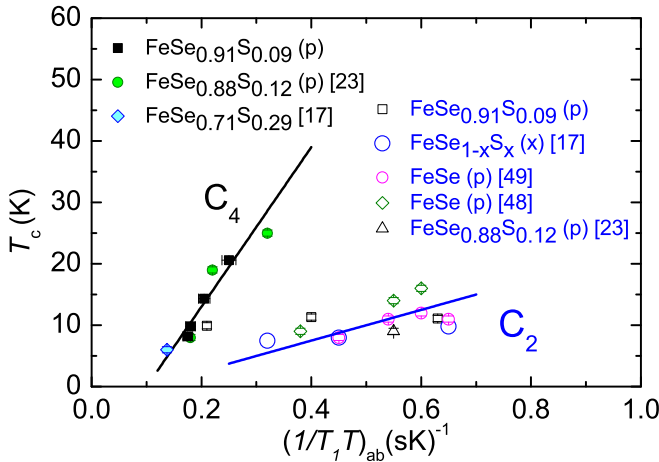


FIG. 4. Plot of T_c at zero field versus maximum values of $(1/T_1T)_{ab}$. For $p \leq 0.5$ GPa, the values of $(1/T_1T)_{ab}$ are taken at the peak positions nearly just above T_c . Above $p = 0.5$ GPa, the constant values of $(1/T_1T)_{ab}$ below T_{FL} were used. The solid and open squares are data from the present Rapid Communication. The values for FeSe under p were taken from Imai *et al.* [48] and Wiecki *et al.* [49]; for $\text{FeSe}_{1-x}\text{S}_x$ under ambient p from Wiecki *et al.* [17]; for $\text{FeSe}_{0.88}\text{S}_{0.12}$ under p from Kuwayama *et al.* [23]. The black and blue lines show linear relations for AFM fluctuations with C_4 and C_2 symmetries, respectively.

under pressure. It is also worth mentioning that no signatures of an AFM order were observed in $1/T_1T$ as well as the NMR spectra, in contrast to the recent muon spin rotation (μSR) report on $\text{FeSe}_{0.89}\text{S}_{0.11}$ under pressure [21]. It is not clear, at present, the reason why the AFM state reported by the μSR measurements is not detected by our NMR and resistivity measurements. Other experiments, such as neutron-diffraction measurements are highly required to elucidate the issue.

Our results indicate that the nature of AFM fluctuations changes below and above 0.5 GPa in $\text{FeSe}_{0.91}\text{S}_{0.09}$. According to Kuwayama *et al.* [23], AFM fluctuations with different \mathbf{q} vectors may be responsible for the two distinct SC domes. Therefore, it is important to reveal the nature of the AFM fluctuations in the different pressure ranges.

Based on previous NMR studies on Fe pnictides [40–42] and related materials [43–45], the ratio $R \equiv (1/T_1T)_{ab}/(1/T_1T)_c$ provides valuable information on \mathbf{q} of the spin fluctuations. In the case of isotropic spin fluctuations, $R = 1.5$ is expected for stripe-type $[\mathbf{q} = (\pi, 0)$ or $(0, \pi)]$ fluctuations whereas $R = 0.5$ for Néel-type $[\mathbf{q} = (\pi, \pi)]$ fluctuations [41]. Therefore, to determine the p and T dependences of R , we have measured $1/T_1T$ at several pressures for $H \parallel c$ (shown by red circles in Fig. 3). As plotted in the inset of each panel of Fig. 3, R is ~ 1 at temperatures above 200 K and increases to $R \sim 1.5$ at low temperatures below 100 K throughout all measured pressures, although the data are slightly scattered, especially for 0.5 GPa. It is important to note that R never decreases down to 0.5 at any pressures. Thus, one can conclude that the AFM fluctuations are characterized

to be stripe type and do not change in the lower and higher SC domes.

What then is the difference in AFM fluctuations between the SC1 and the SC2 domes? One of the important changes in the character of AFM fluctuations is the presence or absence of nematic order as has been discussed previously [46,47]. Below 0.5 GPa, the SC state arises from the nematic phase with C_2 symmetry. In this case, the amplitude of AFM fluctuations with $\mathbf{q}_x = (\pi, 0)$ and $\mathbf{q}_y = (0, \pi)$ must be inequivalent. On the other hand, since SC appears from the tetragonal phase above 0.5 GPa, the magnetic fluctuations with \mathbf{q}_x and \mathbf{q}_y are degenerate due to the C_4 symmetry.

In order to see how the relationship between SC and stripe-type AFM fluctuations changes with the symmetry, we plotted the T_c at zero field versus the maximum value of $(1/T_1T)_{ab}$ below 100 K in Fig. 4, together with data available from the literature. When SC emerged from the nematic state with decreasing temperature as in the case of FeSe for $p < 1.5$ GPa [48,49], $\text{FeSe}_{0.88}\text{S}_{0.12}$ at ambient p [23] and $\text{FeSe}_{1-x}\text{S}_x$ for $x < 0.17$ [17] at ambient p , the AFM fluctuations are labeled as C_2 . When SC emerged in the tetragonal phase for $\text{FeSe}_{0.71}\text{S}_{0.29}$ [17] at ambient p and $\text{FeSe}_{0.88}\text{S}_{0.12}$ for $p > 0.5$ GPa [23], the AFM fluctuations are labeled as C_4 . This plot shows two different correlations between T_c and stripe-type AFM fluctuations with and without nematic order, indicating that the correlations hold for the whole $\text{FeSe}_{1-x}\text{S}_x$ system. When nematic order is absent, a clear and strong correlation between T_c and the stripe-type AFM fluctuations with C_4 symmetry exists as represented by the straight black line. In contrast, when nematic order is present, T_c weakly depends on the stripe-type AFM fluctuations with C_2 symmetry as represented by the blue line with a slope about five times smaller than that of the black line. These results indicate that the AFM fluctuations with C_4 symmetry are more effective in enhancing the superconducting transition in the $\text{FeSe}_{1-x}\text{S}_x$ system.

In conclusion, by NMR and resistivity measurements under pressure, we have established the p - T phase diagram of $\text{FeSe}_{0.91}\text{S}_{0.09}$ exhibiting a nematic quantum phase transition around 0.5 GPa, two SC domes, and an AFM phase above ~ 3.3 GPa. The AFM fluctuations evolve from non-Fermi liquid (Curie-Weiss-like behavior of $1/T_1T$) to a Fermi-liquid behavior ($1/T_1T = \text{constant}$ behavior) across the nematic QPT. The stripe-type wave vector for the AFM fluctuations is revealed to be unchanged in the two SC domes, but the symmetry in the fluctuations is raised from C_2 to C_4 across the nematic QPT. Although both AFM fluctuations are found to be correlated with T_c in $\text{FeSe}_{1-x}\text{S}_x$ under pressure, our results clearly show that T_c is more sensitive to AFM fluctuations with C_4 symmetry than those with C_2 symmetry.

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