Edwards-Anderson parameter and local Ising-nematicity in FeSe revealed via NMR spectral broadening

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(Dated: July 21, 2021)

The NMR spectrum of FeSe shows a dramatic broadening on cooling towards the bulk nematic phase at \( T_s = 90 \) K, due to the formation of a quasi-static, short-range-ordered nematic domain structure. However, a quantitative understanding of the NMR broadening and its relationship to the nematic susceptibility is still lacking. Here, we show that the temperature and pressure dependence of the broadening is in quantitative agreement with the mean-field Edwards-Anderson parameter of an Ising-nematic model in the presence of random-field disorder introduced by non-magnetic impurities. Furthermore, these results reconcile the interpretation of NMR and Raman spectroscopy data in FeSe under pressure.

INTRODUCTION

The nucleation of local-symmetry-breaking order in nominally symmetry-preserving phases is an increasingly recognized phenomenon in correlated electron systems. Important examples include the formation of antiferromagnetic droplets in CeCoIn₅, high-\( T_c \) cuprates and low dimensional quantum magnets [1–4] as well as the nucleation of charge density wave order above the phase transition, in NbSe₂ [5–7], ZrTe₃ [8, 9], cuprates [10] and Sn/Ge(111)-α surfaces [11].

In the iron-based superconductors, signatures of \( C_4 \) symmetry breaking and short-range nematic order have often been found well above the bulk nematic phase transition temperature [12–15], especially from local probe measurements using scanning tunneling microscopy (STM) [10] and nuclear magnetic resonance (NMR) [17–24]. More recently, x-ray and neutron pair distribution function (PDF) studies have revealed the locally orthorhombic nature of the tetragonal paramagnetic phase in the (Sr,Na)Fe₂As₂ system [25]. Similarly, the re-entrant tetragonal antiferromagnetic state in this hole-doped system was also revealed to have short-range orthorhombic correlations [26]. Furthermore, inelastic x-ray scattering has recently produced insights into the spatial correlation length of nematic fluctuations in iron-based superconductors from the wavevector-dependent softening of acoustic phonons [27–30].

While the existence of local nematicity in the tetragonal phase has been rationalized in terms of residual strains in the crystal [31], it is most naturally explained as a consequence of impurities and vacancies that locally break the fourfold rotation symmetry and thus act as random-field impurities for nematicity. Recent theoretical work has addressed the impact of disorder on the emergence of nematicity in iron-based superconductors [32–33].

FeSe is a unique iron-based superconductor (\( T_c = 8.5 \) K) which has a bulk nematic phase below \( T_s = 90 \) K, but no corresponding magnetic phase. Several experiments have revealed a large nematic susceptibility in the high-temperature phase [34–40]. Recent NMR measurements [41–43] revealed a prominent broadening of the NMR spectrum on approaching \( T_s \) from above, which is attributed to the formation of locally-nucleated, short-range-ordered nematic domains. The local orthorhombicity of unstrained FeSe above \( T_s \) was later confirmed by x-ray and neutron PDF studies [44, 45] and has been used to rationalize properties of the tetragonal phase [46].

Under applied hydrostatic pressure \( p \), the temperature-dependent full-width-at-half-maximum (FWHM) of the NMR spectrum follows a universal curve, interrupted only at the nematic transition temperature \( T_s(p) \). This pressure-independent behavior of the NMR FWHM has been interpreted as evidence that the nematic fluctuations are robust against pressure application, despite the decrease of \( T_s(p) \) [41, 42]. However, this conclusion is in conflict with Raman spectroscopy measurements [17] which show a rapid suppression of nematic fluctuations with increasing pressure.

In order to resolve this discrepancy it is important to establish a quantitative understanding of the nematic broadening of the NMR spectrum and, in particular, its relation to the nematic susceptibility. Here, we find that the broadening of the NMR spectrum due to locally-nucleated nematic order is proportional to the Edwards-Anderson parameter of a random-field Ising model at the mean-field level. Within this picture, the pressure-independence of the NMR FWHM is seen to be a consequence of the pressure-independence of random-field de-
fects. We conclude that the NMR data are consistent with a suppression of nematic coupling with increasing pressure and are not in conflict with the Raman data.

**THEORY**

We consider an Ising nematic system characterized by the pseudo-spin variable $\tau_i^z$ on a lattice with site $i$. Our analysis is independent of the microscopic origin of nematicity and equally applies to systems with spin-induced nematicity \[58\]–\[61\], nematicity due to orbital ordering \[62\]–\[65\], or systems with a Pomeranchuk instability in the $\ell = 2$ angular momentum channel \[56\]–\[58\]. In a clean system a finite expectation value $\phi \equiv \langle \tau^z \rangle$ corresponds to nematic order.

We allow for random strain fields $\sim h_i$ that locally break the four-fold symmetry. To be specific we consider random strain characterized by the distribution function

$$p_\sigma(h_i) = \frac{1}{\sqrt{2\pi\sigma}} e^{-\frac{h_i^2}{2\sigma^2}}. \quad (1)$$

The width $\sigma$ is an energy scale that parameterizes the disorder strength. The order parameter is now $\bar{\phi} = \langle \tau^z \rangle$, where $\langle \rangle$ is a thermal average and the overbar is an average over disorder configurations. Despite the random strain one still expects a sharp nematic transition above which $\bar{\phi} = 0$, at least for three-dimensional systems and not too strong disorder \[59\]–\[61\]. However, for the problem at hand one expects at any finite randomness a finite Edwards-Anderson parameter

$$q_{EA} = \bar{\phi}^2 - \langle \phi \rangle^2, \quad (2)$$

that characterizes the strength of local disorder variations, even if $\phi$ vanishes on the average. Notice that this is different from the behavior in spin glasses where $q_{EA}$ serves, at least within mean field theory, as “order” parameter of the spin glass state \[61\]. Nevertheless, there are interesting analogies between our approach and the theory of NMR in spin glasses or relaxor ferroelectrics, where the determination of the Edwards-Anderson parameter has played an important role \[62\]–\[66\]. In what follows we will first discuss that $q_{EA}$ is directly related to the width of the NMR spectrum. In a second step we present and solve a simple mean-field model that allows for remarkable agreement between theory and experiment.

**Connection to NMR spectrum broadening**

We start our analysis with a brief discussion of the effect of random strain on the NMR spectrum. In a homogeneous case, the NMR spectrum of a system where the nucleus under consideration has one unique position in the lattice and occurs only in one isotope configuration can be expressed as

$$f(\omega) = \frac{1}{N} \sum_{i=1}^{N} \delta(\omega - \omega_0), \quad (3)$$

where $\omega_0$ is the Larmor frequency and $N$ is the number of nuclei in the sample. We assume that a defect-nucleated local order parameter $\phi_i(h_i)$ will give rise to a shift of the local NMR resonance frequency according to $\omega_0 \rightarrow \omega_0 + \alpha \phi_i(h_i)$. By averaging each site over the disorder distribution $p_\sigma(h_i)$ one obtains the NMR spectrum as

$$\overline{f(\omega)} = \frac{1}{N} \int \prod_{j=1}^{N} dh_j p_\sigma(h_j) \sum_{i=1}^{N} \delta(\omega - \omega_0 - \alpha \phi_i(h_i)). \quad (4)$$

The broadening $\nu$ of the NMR spectrum is given by the second moment of the distribution $\nu^2 = \int d\omega f(\omega)(\omega - \omega_0)^2$. Carrying through the integrations yields

$$\nu^2 = \frac{1}{N} \int \prod_{j=1}^{N} dh_j p_\sigma(h_j) \sum_{i=1}^{N} \alpha^2 \phi_i^2(h_i) \quad (5)$$

$$= \alpha^2 \overline{\phi^2} = \alpha^2 q_{EA}. \quad (6)$$

Therefore, the FWHM of the NMR spectrum is expected to be proportional to the square root of the Edwards-Anderson parameter:

$$\nu = \alpha q_{EA}^{1/2}. \quad (7)$$

This analysis was performed under the assumption of vanishing averaged order parameter $\bar{\phi}$. At finite $\bar{\phi}$ one easily finds that $\nu^2$ is proportional to the expression for $q_{EA}$ in Eq. (2).

**Random-Field Ising-nematic model**

In order to get a quantitative understanding of the temperature and strain dependence of the Edwards-Anderson parameter $q_{EA}$ of Eq. (2), we now perform a simple mean-field analysis of the corresponding random field Ising model. While the random-field Ising model is a theoretical problem of formidable complexity, we will confine ourselves to a mean-field analysis. As we will see, this already allows for a rather detailed understanding of the temperature and pressure dependence of the NMR broadening for FeSe. Mean field behavior of the nematic degrees of freedom is expected for clean systems. As was discussed in Ref. \[66\], long range strain forces drive the statistical mechanics of the system into a mean field regime in the entire temperature window where an appreciable softening of the shear modulus is observed. In systems with random strain, the situation is more subtle \[59\]–\[61\] and one expects disorder fluctuations...
at long distances and time scales. However on the local
length scale of the measurement of the NMR line-width,
a mean field analysis is a reasonable starting point. The
description of dynamical phenomena, as determined by
the NMR relaxation rate, is likely more subtle and may
requiring to go beyond the mean field theory.

The Hamiltonian is expressed as
\[ H = - \sum_{i,j} J_{i,j} \phi_i^* \phi_j - \sum_i (h_0 + h_i) \phi_i^* , \tag{8} \]
where \( h_0 \) is an external strain that breaks \( C_4 \) symme-
try globally, while \( h_i \) is a random field at each site. In
the spirit of the mean field analysis we approximate this
model by the infinite range interaction, where \( J_{i,j} \rightarrow J/N \). Then mean field theory becomes exact \[\text{[67]}\]. When
\( h_i = 0 \), we obtain the familiar equation of state for the
order parameter \( \phi = \langle \tau^z \rangle \) is \( \beta \equiv 1/k_B T \)
\[ \phi = \tanh(\beta(2J\phi + h_0)]. \tag{9} \]
The nematic transition temperature of the system with-
out disorder follows as \( T^{(0)}_s = 2J \).

If we now allow for random stress, the corresponding
mean-field equation of state for the order parameter be-
comes
\[ \overline{\phi} = \int dh\phi \tanh \left( \frac{2J\phi + h_0 + h}{k_BT} \right) , \tag{10} \]
which is solved self-consistently for \( \overline{\phi} \). To find the
nematic phase transition temperature \( T_s/J \) as a function
of disorder \( \sigma/J \), we take \( h_0 = 0 \) and expand the
hyperbolic tangent while keeping only terms linear in \( \overline{\phi} \) to
obtain
\[ \overline{\phi} = \frac{2J\overline{\phi}}{k_BT_s} C \left( \frac{\sigma}{k_BT_s} \right) , \tag{11} \]
where the function \( C \) is given by
\[ C(t) = \int dx \frac{e^{-x^2}}{\sqrt{2\pi}} \tanh^2 (xt) . \tag{12} \]
The transition temperature \( T_s(\sigma) \) of the system with
disorder is determined by the condition
\[ T_s = T^{(0)}_s C \left( \frac{\sigma}{k_BT_s} \right) . \tag{13} \]
Random strain reduces the transition temperature, but
the transition itself remains sharp. In distinction, global
external strain \( h_0 \) would smear the transition. The
nematic transition reaches \( T_s = 0 \) when \( \sigma/J = \pi/2 \). As
we will see, NMR experiments for FeSe suggest that \( \sigma \) is
significantly smaller than \( \pi J/2 \) where disorder destroys
nematic order completely.

The nematic susceptibility above \( T_s \) is given by
\[ \chi_{nem}(T) = \frac{\partial \overline{\phi}}{\partial \sigma} \bigg|_{h_0=0} = \frac{C \left( \frac{\pi T}{\pi T^*} \right)}{T - T^{(0)}_s C \left( \frac{\pi T}{\pi T^*} \right)} , \tag{14} \]
As mentioned above, we have to interpret the effective
Ising model as the one that includes all allowed couplings,
including the ones that are mediated by the lattice. In
fact the mean-field treatment of the clean system is pos-
sible because we have included this lattice coupling which
leads to an effective long-range interaction \[\text{[66]}\]. Hence,
the nematic susceptibility \( \chi_{nem} \) is proportional to the in-
verse elastic constant \( C_{66} \) \[\text{[50]}\]. It diverges at the ac-
tual thermodynamic phase transition, not at the lower
Curie-Weiss temperature that one deduces from Raman
or elasto-resistivity measurements \[\text{[68]}\].

Finally, we determine the Edwards-Anderson order pa-
rameter. Taking \( h_0 = 0 \) above \( T_s \) where \( \phi = 0 \), we have
\[ q_{EA}(T) = \int dh\phi \tanh \left( \frac{h}{k_BT} \right) \]
\[ = 1 - C \left( \frac{\sigma}{k_BT} \right) . \tag{15} \]
Obviously, \( q_{EA} \) above \( T_s \) only depends on temperature
and the strength of random strain \( \sigma \). As we will see be-
low, this explains the “scaling” of the NMR line width
as function of pressure. Changing the nematic coupling
constant will then not change the \( T \) dependence of \( q_{EA} \),
but merely set the temperature \( T_s = T^{(0)}_s (1 - q_{EA}(T_s)) \)
where it deviates from its high-temperature behavior.
Clearly, the finding that \( q_{EA}(T > T_s) \) is independent of
\( J \) is only valid within the mean-field approximation. We
can turn this reasoning around and conclude that a tem-
perature dependent NMR line width that is independent
of the value of \( T_s \) strongly supports that the inhomoge-
neous nematic state above \( T_s \) is well captured within a
mean field treatment.

We also note that the Edwards-Anderson parameter is
not proportional to the nematic susceptibility. However,
both are related via
\[ \chi_{nem}(T) = \frac{1 - q_{EA}(T)}{T - T^{(0)}_s (1 - q_{EA}(T))} . \tag{16} \]
Thus, in principle it is possible to determine the
Edwards-Anderson parameter from the nematic suscep-
tibility. In practice, the available accuracy of data for
\( \chi_{nem}(T) \) turns out not to be sufficient to determine
\( q_{EA}(T) \). The reason is that \( q_{EA}(T) \) for FeSe turns out
to be significantly smaller that unity. This also ex-
plains that the \( T \)-dependence of \( q_{EA} \) does not signifi-
cantly change the Curie-Weiss behavior of the nematic
susceptibility. Thus \( \chi_{nem} \) is not a sensitive indicator
of inhomogeneous nematicity while local probes, such as
NMR line-width measurements, are able to reveal the
existence and temperature dependence of inhomogeneous
nematic regions.
underdoped Ba
distortion corrected for the effect of the applied force,
result implies that the strong specific-heat peak in zero-field at
superconducting transition. Note the larger specific-heat
for a sample with 30% K content, which undergoes only a
phase occurs at 
(ref. 23), is a sign of excited quasiparticles far below
mains when local nematic order is present. In (a), a magnetic
field \( H \) is applied such that the nuclei in the two domains
experience symmetry-inequivalent field directions. Due to the
anisotropy of magnetic susceptibility in the nematic state,
the nuclei in the two domains see different local hyperfine
fields, resulting in a two-peak NMR spectrum, as illustrated
schematically. In contrast, when the field is rotated 45° as
in (b), the nuclei experience symmetry equivalent hyperfine
fields, and the NMR spectrum appears as a single peak.

![Diagram](image)

**SUMMARY OF NMR RESULTS**

Since \( ^{77}\text{Se} \) is a \( I = 1/2 \) nucleus, there are no
quadrupole satellite lines or quadrupole shifts of the spectrum.
Therefore, in the high-temperature tetragonal state, the NMR spectrum is a single peak. In the nematic state below \( T_c \), there are nematic-twin domains. As illustrated in Fig. 1(a), half of the domains experience \( H || a_O \) and the other half experience \( H || b_O \), where \( a_O > b_O \). Due to the anisotropic magnetic susceptibility of the nematic phase \( \chi\text{rem} \), the hyperfine field at the nucleus will be different in the two domains, causing the NMR spectrum to split into two peaks in the nematic ordered state. If the field is instead applied along \( H || 110 \) (Fig. 1(b)), then both types of domain see a symmetry-equivalent magnetic field, and there is only a single NMR peak in the nematic state, even though there are still domains. Recent NMR measurements under mechanical strain have revealed that the higher frequency NMR peak comes from the domains which experience \( H || a_O \), while the lower frequency peak comes from the \( H || b_O \) domains \( \chi\text{rem} \).

In the tetragonal phase, the FWHM of the single NMR peak increases on cooling towards the nematic state. However, this broadening is observed only when \( H || 100 \) and not when \( H || 110 \). This observation provides clear evidence that the broadening is related to nematicity and implies the existence of a short-range nematic domain structure in the tetragonal state of FeSe. Since this effect is observed in the NMR spectrum, the fluctuating nematic domain structure is static on the time scale set by the inverse NMR linewidth \( \sim 1/(1\text{kHz}) \).

**COMPARISON OF THEORY AND EXPERIMENT**

For a direct comparison between theory and experiment FeSe offers several clear advantages over other nematic iron-based superconductors. First, FeSe has no bulk magnetic phase at ambient pressure, and thus the pure Ising-nematic model is expected to be relevant in this system. Secondly, the \( ^{77}\text{Se} \) nucleus has \( I = 1/2 \) so that no complications arise from nuclear quadrupole couplings. Finally, no dopants are present, which can introduce additional lines into the NMR spectrum [17 22].

Fig. 2(a) shows the NMR FWHM data for FeSe under pressure. The FWHM increases on cooling towards \( T_c \). The data at different pressures follow a universal curve, simply interrupted at the appropriate nematic transition temperatures \( T(\rho) \). Below \( T_0(\rho) \) in the nematic state, the FWHM of each of the two NMR peaks is shown separately. The FWHM of the low-frequency NMR peak (open symbols) is greater than that of the high-frequency peak (hatched symbols). The different FWHM of the two NMR peaks in the nematic state of FeSe has been noted in independent measurements and is currently not understood [17].

Within the Ising-nematic model, the experimental data under pressure is expected to correspond to a constant disorder strength \( \sigma \) (determined only by the disorder of the particular crystal), but decreasing nematic coupling \( J_0 \), starting from a value \( J_0 \) that corresponds to ambient pressure. Figs. 2(b),2(c) show the mean-field behavior of the Edwards-Anderson parameter \( q_{EA}^{1/2} \) and the nematic susceptibility \( \chi_{\text{rem}} \) under these assumptions. As expected, the NMR data are well described by \( q_{EA}^{1/2} \) and not \( \chi_{\text{rem}} \) (Eq. 7). The universal FWHM curve followed at all pressures is seen to be the result of the constant \( \sigma \). The existence of a universal curve does not mean that the nematic tendency is somehow independent of pressure, as proposed in both NMR papers [11 32]. Rather this reflects the fact that \( q_{EA}(T > T_c) \) is independent of \( J \) within the mean-field approximation, as discussed above. Therefore, the NMR data are consistent with the Raman data, showing a suppression of nematic fluctuations (Fig. 2(c)) under pressure [47].

The FWHM data in Fig. 2(a) show a deviation from \( q_{EA}^{1/2} \) at high temperature. This deviation is easily understood. In a usual PM state, the broadening reflects the spatial distribution of \( K \) values due to sample inhomogeneity, and the temperature dependence of the broadening typically follows the temperature dependence of the
CONCLUSIONS

FeSe features a local, orthorhombic nematic order in its high-temperature, nominally tetragonal phase. The observed agreement between the NMR line broadening and the Edwards-Anderson parameter of the disordered, random-field Ising-nematic model implies that this local nematic order is primarily nucleated by crystal defects. The origin of the local orthorhombicity was not explic-
ity considered in the PDF studies of FeSe [44, 45]. We note that this short-range ordered nematicity in the high-temperature tetragonal state is distinct from the quantum Griffiths phase recently proposed in Fe(Se,S) under pressure, where rare but large regions of local nematic droplets undergo quantum fluctuations [74]. In this context it is important to keep in mind that in metallic systems such quantum fluctuations are known to be strongly suppressed due to the coupling to the particle-hole continuum [75-77], a behavior specific to Ising degrees of freedoms [78]. In fact it is this suppression of quantum fluctuations that is consistent with the effectively classical description of local nematic order of our approach. Finally, our results demonstrate that the nematic fluctuations in FeSe are suppressed by hydrostatic pressure, consistent with Raman studies [47], despite the pressure-suppressions in FeSe are suppressed by hydrostatic pressure, [79].

We thank B. M. Andersen, S. A. Kivelson, R. M. Fernandes and Y. Furukawa for valuable discussions. We acknowledge support by the Helmholtz Association and B. M. Andersen, S. A. Kivelson, R. M. Fernandes, L. Y. Xing, X. C. Wang, C. Q. Jin, A. J. Millis, and A. N. Pasupathy, “Visualization of electron nematicity and unidirectional antiferroic fluctuations at high temperatures in NaFeAs,” [Nature Physics 10, 225–230 (2014)].

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