Dynamical susceptibility of a near-critical nonconserved order parameter and quadrupole Raman response in Fe-based superconductors

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Dynamical susceptibility of a near-critical non-conserved order parameter and quadrupole Raman response in Fe-based superconductors

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We analyze the dynamical response of a two-dimensional system of itinerant fermions coupled to a scalar boson $\phi$, which undergoes a continuous transition towards nematic order with $d-$wave form-factor. We consider two cases: (a) when $\phi$ is a soft collective mode of fermions near a Pomeranchuk instability, and (b) when it is an independent critical degree of freedom, such as a composite spin order parameter. In both cases, the order-parameter is not a conserved quantity and the $d-$wave fermionic polarization $\Pi(q,\Omega)$ remains finite even at $q = 0$. The polarization $\Pi(0,\Omega)$ has similar behavior in the two cases, but the relations between $\Pi(0,\Omega)$ and the bosonic susceptibility $\chi(0,\Omega)$ are different, leading to different forms of $\chi''(0,\Omega)$, as measured by Raman scattering. We compare our results with polarization-resolved Raman data for the Fe-based superconductors FeSe$_{1-x}$S$_x$, NaFe$_{1-x}$Co$_2$As and BaFe$_2$As$_2$.

**Introduction.** - The behavior of strongly correlated fermions in the vicinity of a quantum critical point (QCP) is one of the most fascinating problems in many-body physics. A traditional way to treat the physics near a QCP is to study an effective low-energy model in which itinerant fermions are coupled to near-critical fluctuations of a bosonic order parameter [1]. The boson can be a collective mode of electrons, as in the case of a Pomeranchuk instability, or an independent degree of freedom (e.g., a phonon). In both cases, the boson-fermion coupling affects the bosonic dynamics. This effect is encoded in the fermionic polarization $\Pi(q,\Omega)$, which in turn is related to the bosonic susceptibility $\chi(q,\Omega)$. Previous studies of $\chi(q,\Omega)$ [1–5] focused primarily on the range $\Omega \ll v_F q$ ($v_F$ is the Fermi velocity), in which the scaling behavior holds in critical theories with a dynamical exponent $z > 1$. However, several experimental probes, most notably polarization-resolved Raman scattering, analyze $\chi(q,\Omega)$ in the opposite limit of vanishing $q$ and finite $\Omega$ [6]. The same regime has been probed in Quantum-Monte-Carlo studies [7]. If the order parameter is conjugate to a conserved quantity, e.g., the total fermion number-density or the total spin, the fermionic polarization $\Pi(q,\Omega)$ vanishes identically by the conservation law at $q = 0$ and, by continuity, is small at $\Omega \gg v_F q$. However, if the order parameter is conjugate to a quantity that is not constrained by conservation laws, $\Pi(0,\Omega)$ does not have to vanish and may give rise to a nontrivial frequency dependence of $\chi(0,\Omega)$.

In this letter we report the results of our study of $\Pi(0,\Omega)$ and $\chi(0,\Omega)$ for a clean system of 2D fermions coupled to fluctuations of a (charge) nematic order parameter, $\phi$, with a $d-$wave form-factor. If $\phi$ is a collective mode of fermions, the model describes an itinerant fermionic system near a Pomeranchuk instability. If $\phi$ is a separate degree of freedom, it softens on its own, but fermions still affect the critical behavior. In both cases, $\Pi(q,\Omega)$ is the same. However, in the Pomeranchuk case, the bosonic susceptibility represents the same collective excitations that determine the polarization and is proportional to $\Pi(q,\Omega)$ for all momenta and frequencies. We compare our results with the Raman data for Fe-chalcogenide FeSe$_{1-x}$S$_x$ [8, 9] and Fe-pnictides BaFe$_2$As$_2$ [10] and NaFe$_{1-x}$Co$_2$As [11], which all display nematic order in some range of temperature and doping. For these and related systems two different electronic scenarios for nematicity have recently been put forward [12]. One scenario is that nematicity is associated with a composite spin order [13, 14]. Another is that nematic order is a Pomeranchuk order in the charge channel with a $d-$wave form factor [15–18]. The two order parameters are linearly coupled, and if the susceptibility for one order parameter increases, it triggers the increase of the susceptibility for the other order parameter. Still, if the linear coupling is not strong, one can identify the primary mechanism, with the most strongly divergent correlation length. At the moment, there is no clear-cut experimental confirmation for either mechanism (for recent experimental reviews see [19, 20]).

For all systems, we find evidence of quantum critical behavior near the transition. For FeSe and FeSe$_{1-x}$S$_x$, we find good agreement with the Pomeranchuk scenario, consistent with the fact that magnetic order in these systems does not develop down to $T = 0$ (although some magnetic fluctuations have been detected [21–23]). For NaFe$_{1-x}$Co$_2$As and BaFe$_2$As$_2$ we find a better agreement with the composite magnetic scenario of nematicity, consistent with the fact that nematic and magnetic ordering
temperatures nearly coincide and in NaFe$_{1-x}$Co$_x$As follow each other as functions of doping.

The model.- We consider a clean two-dimensional system of itinerant fermions with a circular Fermi surface (FS) specified by Fermi momentum $k_F$ and Fermi velocity $v_F$, coupled to a scalar boson $\phi(q)$, which undergoes a continuous transition towards charge nematic order with $d$--wave form factor. The field $\phi$ is coupled to the $d$--wave component of fermionic density as

$$H_I = g \sum_{k,q} f(k) \phi(q) \psi^\dagger(k+q/2) \psi(k-q/2), \quad (1)$$

where $g$ is a coupling constant and $f(k)$ is a momentum dependent vertex with $d$--wave symmetry (e.g., $f(k) = \cos k_x - \cos k_y$). Near the FS $f(k) \approx f(\theta)$, where $\theta$ is an angle along the FS. This model has been discussed extensively in the regime where the characteristic bosonic frequency $\Omega$ is much smaller than $v_F q$, with $q$ the characteristic bosonic momentum [5, 23, 24]. In standard treatments of this regime, the boson propagator is

$$D(q, \Omega) = \frac{\chi_0}{\epsilon_0^2 + q^2 - \Omega^2/2 + i\epsilon_0} \pi \epsilon \bar{\Omega}(q, \Omega). \quad (2)$$

Here $\bar{\Omega} = q^2 \chi_0$ is the effective coupling constant (assumed much smaller than $\epsilon_F \equiv v_F k_F/2$), $\epsilon_0$ and $c$ are the bosonic correlation length and velocity at $g = 0$, i.e., in the absence of coupling to fermions, and $\Pi(q, \Omega)$ is the particle-hole polarization bubble, given by

$$\bar{\Pi}(q, \Omega) = -\frac{\bar{\Pi}(q, \Omega)}{4\pi\epsilon \epsilon_0} \left( f^2 + i f^2(q) \frac{\Omega}{v_F |q|} \right), \quad (3)$$

where $\epsilon_F = v_F k_F/2$, $(f^2(\theta)) = \int \frac{d\theta}{2\pi} f^2(\theta)$, and $q' = \hat{z} \times \hat{q}$, $\hat{z}$ is a unit vector in the direction perpendicular to the 2D plane [25, 26]. The constant term in $\bar{\Pi}(q, \Omega)$ accounts for the difference between $\Omega_0$ and the actual nematic correlation length $\xi^2 = \xi_0^2 - g k_F^2/(2\pi \epsilon_0)$. The form of $D(q, \Omega)$ at $\Omega \ll v_F |q|$ determines the fermionic self-energy on the FS: at the QCP, $\Sigma(\Omega, \theta) \sim |f(\theta)|^{4/3} \Omega^{2/3} \omega_0^{1/3}$, where $\omega_0 \approx g^2/\epsilon_0$.

Our goal is to obtain $\Pi(q, \Omega)$ at $T = 0$ in the opposite limit of $q = 0$ and finite $\Omega$. We obtain $\Pi(0, \Omega)$ first along Matsubara axis $\Omega = i\Omega_m$, and then convert to real $\Omega$. For free fermions, $\Pi(0, \Omega_m)$ vanishes for arbitrary $f(\theta)$ because the density of fermions at each momentum is separately conserved. At a finite $\bar{\Omega}$, this is generally not the case. We evaluate $\Pi(0, \Omega_m)$ to leading order in $\bar{\Omega}$ by computing the two-loop Maki-Thompson diagrams for the $d$-wave particle-hole bubble, along with the Aslamazov-Larkin diagrams, which contribute at the same order (see Ref. [27] for details). For a constant form-factor $f(\theta) = 1$, these diagrams cancel exactly, and the cancellation can be traced to the Ward identity for fermion number conservation [28]. For a non-conserved order parameter the diagrams do not cancel. Evaluating the diagrams, we obtain

$$\bar{\Pi}(q = 0, \Omega_m) = \left( \frac{\bar{\Pi}(0, \Omega)}{v_F} \right)^2 (f_2) \times \left[ A(k_F \xi) - C(m|\Omega_m|; k_F \xi) \right]. \quad (4)$$

Here $(f_2) = \langle f^2 f'^2 + \frac{1}{2} f'^2 (f'') \rangle < 0$, and the functions $A$ and $C$ are

$$A(k_F \xi) = 1 - (k_F \xi)^{-1} \tan^{-1}(k_F \xi) \quad (5)$$

and

$$C(m|\Omega_m|; k_F \xi) \sim \left\{ \begin{array}{ll} \frac{g^2 |\Omega_m|}{\epsilon_F} \xi_F^{-3} & |\Omega_m| \ll \xi_F^c \vspace{0.5em} \\
\left( \frac{g^2 |\Omega_m|}{\epsilon_F} \right)^{1/3} & |\Omega_m| \ll |\Omega|^* \vspace{0.5em} \\
\frac{g^2 |\Omega_m|}{\epsilon_F} & |\Omega_m| \ll |\Omega|^* \end{array} \right. \quad (6)$$

The characteristic frequencies in (6) are $\xi_F^c \sim \epsilon_F^c g^{-1}(k_F \xi)^{-3}$ and $\Omega^* \sim \sqrt{\bar{\Omega}F(c/v_F)}$. The frequency $\Omega^*$ separates Fermi liquid and QC behavior, and $\Omega^*$ marks where the damping term starts to dominate over the bare $\Omega^2 m/c^2$ in Eq. (2) (Refs. [29, 30]). The various dynamical regimes in real frequency, both at and near the QCP, are depicted qualitatively in Fig. 1.
be considered an independent, near-critical bosonic field, the full bosonic susceptibility $\chi_{\text{ind}}(0, \Omega)$ predominantly comes from the $\phi$ field, i.e., $\chi_{\text{ind}}(0, \Omega) \approx D(0, \Omega)$, where $D$ is given by Eq. (2). The behavior of $\chi_{\text{ind}}(0, \Omega)$ then depends on the scale $\xi_0$ as well as $\xi$. This opens an avenue to distinguish the “Pomeranchuk” and “independent” scenarios, by comparing low-energy properties of the real and imaginary parts of their corresponding susceptibilities. We concentrate on two such properties: the the behavior of $\delta \chi' = \chi'_f(0, \Omega \rightarrow 0) - \chi'_S(0, \Omega \rightarrow 0)$ and the slope of the imaginary part of the susceptibility $\Gamma = \chi''(0, \Omega)/\Omega|_{\Omega=0}$.

In the Pomeranchuk case, converting Eqs (5) and (6) to real frequencies we find

$$\delta \chi'_{\text{Pom}} \propto \xi^{-1}, \quad \Gamma_{\text{Pom}} \propto \xi^2.$$ \hspace{1cm} (7)

In the independent case, to leading order in $g/\varepsilon_F$, $\chi'_{\text{ind}}(0, \Omega) = \chi_0 \xi_0^2$, and $\chi''_{\text{ind}}(0, \Omega) = -\chi_0 g^2 \xi_0^2 \Omega^2/2(4\pi\varepsilon_F)$, where we recall that $\xi_0 = \sqrt{\varepsilon_F / g^2}$. Therefore

$$\delta \chi'_{\text{ind}} \propto \xi^{-2} \xi_0^2, \quad \delta \left( \frac{1}{\chi_{\text{ind}}} \right) \propto \xi^{-2}, \quad \Gamma_{\text{ind}} \propto \xi^2 \xi_0^2.$$ \hspace{1cm} (8)

We emphasize that in both cases, $\chi'_0(\Omega \rightarrow 0)$ remains finite at the QCP because it differs from the thermodynamic nematic susceptibility $\chi'(q \rightarrow 0, \Omega = 0)$, which scales as $\xi^2$ and diverges at the QCP. We recall in this regard that we consider a clean system. In the presence of weak disorder the limits $q \rightarrow 0$ and $\Omega \rightarrow 0$ indeed commute [16], but $\chi'(0, \Omega)$ nonetheless approaches its clean limit behavior for frequencies above an appropriate transport scattering rate $\gamma_{tr}$. In this respect our $\chi'(0, \Omega \rightarrow 0)$ is actually the susceptibility for $\Omega$ much larger than $\gamma_{tr}$, but well below any other energy scale.

**Comparison with experiments**

The d-wave bosonic susceptibility is directly measured in polarization-resolved Raman scattering experiments [6]. The momentum transfer in Raman experiments is very low, so that to high accuracy the susceptibility extracted from Raman measurements coincides with $\chi(q = 0, \Omega) = \chi(\Omega)$. Near the transition, the quadrupole response develops a broad peak. As $T$ approaches $T_s$, the peak position moves to a smaller frequency and the slope at $\Omega \rightarrow 0$ increases. Several explanations of the peak have been presented [16, 32–35]. Within our theory, this peak is a direct consequence of Eq. (6).

We compare our theoretical results for $\chi''(\Omega)$ with data for the Fe-chalcogenides FeSe/ FeSe$_{1-x}$S$_x$ (Refs. 8 and 9) and Fe-pnictides BaFe$_2$As$_2$ (Ref. 10) and NaFe$_{1-x}$Co$_x$As (Ref. 11). We assume that the relevant frequencies in Raman measurements are larger than the transport scattering rate, i.e., the data can be described within a clean limit, although disorder may be a source of systematic corrections[36]. From the data one can extract the slope $\Gamma = \lim_{\Omega \rightarrow 0} \chi''(\Omega)/\Omega$. Regardless of how the extrapolation to $\Omega \rightarrow 0$ is performed [37], $\Gamma$ grows rapidly in the vicinity of $T_S$. The real part of the susceptibility $\chi'(0, \Omega \rightarrow 0)$ was extracted [8, 9, 11] from the data for $\chi''(\Omega)$ via Kramers-Kronig: $\chi'(0, \Omega \rightarrow 0) = (2/\pi) \int_{W_0}^{\infty} d\Omega' \chi''(0, \Omega')/\Omega'$, where $W_0 \sim 3meV$ is the lower cutoff in the data. We emphasize that this is not a true static susceptibility, even if we set $W_0 = 0$, because the data for $\chi''$ are obtained at $\Omega > \pi/c$ and $\chi'(0, \Omega \rightarrow 0)$ is still much larger than momentum. The nematic transition temperature $T_S$ varies with $x$ in FeSe$_{1-x}$S$_x$ and NaFe$_{1-x}$Co$_x$As and vanishes at a particular S or Co doping. We assume that the $T$ dependence can be incorporated into $\xi_0(x, T)$ and $\xi(\xi, T)$, but do not otherwise incorporate finite temperature into our calculations. As such the results should be valid as long as typical $\Omega > T_S$, which is true for most of the relevant experimental frequency range.

For both sets of materials we examined possible scaling between $1/\Gamma$ and powers of $\delta \chi'$. For FeSe$_{1-x}$S$_x$ we found that $(\delta \chi')^2$ and $1/\Gamma$ scale together (Fig. 2). Such behavior is consistent with the Pomeranchuk scenario, as in this case both $(\delta \chi')^2$ and $1/\Gamma$ scale as $\xi^{-2}$, Eq. (7). The data for FeSe$_{1-x}$S$_x$ also show [9] that $\chi'_0(0,0)$ increases as the system approaches the nematic transition but deviates from Curie-Weiss behavior near the transition point. The deviation gets more pronounced with increasing $x$. Such behavior is also consistent with the Pomeranchuk scenario, Eq. (5), particularly given that
The scaling 1/χ′ is also a constant. This is the case near T_G (taken from Ref. 11). Filled circles – 1/χ′ (0, Ω = 0) (taken from Ref. 16, 18, 34) and therefore gives an additional contribution to the difference between ξ_0 and ξ. It also affects the momentum dependence of D(q, Ω) at Ω < v_F q, and eventually cuts off the critical behavior at T = T_s [32, 40]. Given that Γ and χ′ strongly increase as T approaches T_s, we conjecture that coupling to acoustic phonons affects the system’s dynamics only in a narrow range very near T_S, while our theory is applicable outside this range.

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Summary In this work we analyzed the dynamic response of a clean 2D system of itinerant fermions coupled to a scalar boson \( \phi \), which undergoes a continuous transition towards a d-wave charge nematic order. We obtained the form of \( \Pi(0, \Omega) \) both at and near the nematic transition and related it to the bosonic susceptibility \( \chi(0, \Omega) \) in the cases where \( \phi \) is a soft collective mode of fermions near a Pomeranchuk instability or an independent critical degree of freedom, such as a composite spin order parameter near an Ising-nematic transition. We compared our results with polarization-resolved Raman data for FeSe_{1−x}S_x and BaFe_2As_2/NaFe_1−xCo_xAs. We argued that the data for FeSe_{1−x}S_x, which does not order magnetically down to \( T = 0 \), are well described by the d-wave charge Pomeranchuk scenario. The data for BaFe_2As_2/NaFe_1−xCo_xAs at \( T \geq T_s \) are more consistent with the independent boson scenario (for which composite spin order is the primary candidate). For all compounds we found evidence for quantum critical behavior near \( T_S \).

Our analysis neglects the interaction with acoustic phonons. This interaction does not directly affect \( \chi(q = 0, \Omega) \), but it does contribute to \( \chi(q \to 0, \Omega = 0) \) [16, 18, 34] and therefore gives an additional contribution to the difference between \( \xi_0 \) and \( \xi \). The coupling to acoustic phonons affects the system’s dynamics only in a narrow range very near \( T_S \), while our theory is applicable outside this range.

\[ k_F \] in this system is small for all pockets [19, 38, 39]), because in this case \( \chi'(0, \Omega \to 0) \) increases as \( \xi^2 \) between \( \xi \) on order of the lattice constant and \( \xi \sim 1/k_F \). The data also show [8, 9] that the maximum in \( \chi''(0, \Omega) \) remains at a nonzero frequency at the nematic transition. This is consistent with the crossover to QC behavior because at \( \xi = \infty \), \( \chi''(0, \Omega) \) still increases at small \( \Omega \) as \( \Omega^{1/3} \), and therefore passes through a maximum at nonzero \( \Omega \). We consider the combination of these results as a strong indication that nematicity in FeSe/FeSe_{1−x}S_x is caused by a d-wave Pomeranchuk instability.

For Ba(FeAs)_2 and NaFe_{1−x}Co_xAs, we found that the temperature dependence of 1/Γ closely follows that of \( \delta(1/\chi') \) over several tens of kelvin near \( T_S \), as shown in Fig. 3. This observation is generally consistent with the “independent” scenario as there both 1/Γ and \( \delta(1/\chi') \) scale as \( \xi^{-2} \) (see Eq. (8)). A natural candidate for the independent order parameter is the composite Ising nematic operator derived from the magnetic order parameter [13], since in these materials the magnetic and nematic transitions are close to each other and show nearly identical doping dependence. We caution, however, that the scaling 1/Γ ~ \( \delta(1/\chi') \) holds in our theory under the assumption that \( \xi_0 \) is essentially a constant, and hence \( \chi' \propto \xi_0^2 \) is also a constant. This is the case near \( T_s \), but at higher \( T \), the measured \( \chi' \) varies significantly over the temperature ranges shown in Fig. 3 (Refs. [10, 11]).

Physics: Condensed Matter 24, 294205 (2012), and references therein; S.-S. Lee, ArXiv:1703.08172, and references within.


[28] We expect that Ω∗ is comparable to εF in systems where g ≲ E_F and c_v^σ. To estimate E_F we used the ARPES data for FeSe at k_F = 0 and k_z = π (Refs. [19, 38, 41–44] and averaged over k_z. This yields Ω∗ ≲ 20meV. For FeAs compounds we expect and somewhat higher Ω∗ because some E_F are larger [45–50].)

[29] The perturbative calculation leading to Eqs. (4)-(6) is only valid when |Σ(Ω_m)| ≪ |Ω_m|, which holds for |Ω_m| ≫ ω_0. At smaller Ω_m self-energy and vertex corrections cannot be neglected a priori. We computed gΠ(0,Ω_m) at Ω_m ≪ ω_0 by inserting a series of self-energy and vertex corrections and found that these higher order contributions cancel each other such that Eq. (4) remains unchanged (we checked cancellation to leading order in (g/εF)^4).

[30] We neglect the small correction to D(q = 0, 0) coming from the static part of Π(q = 0, 0). Although this term scales as (k_Fξ)^2 and asymptotically wins over the ξ^-2 term, it has an additional smallness in (g/εF)^4.


[39] The value γtr at T = 0 can be extracted from the resistivity data. For FeSe we used the data from Ref. [51] and obtained γtr ∼ 1meV. The data for Imχ(0, Ω) in Refs. 9 and 11 are for frequencies above roughly 3 meV, i.e. for all Ω the condition Ω > γtr is satisfied.)


