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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 ϕ , which undergoes a continuous transition towards nematic order with d -wave form-factor. We consider two cases: (a) when ϕ 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 $\text{FeSe}_{1-x}\text{S}_x$, $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ and BaFe_2As_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 $\Pi(\mathbf{q}, \Omega)$, which in turn is related to the bosonic susceptibility $\chi(\mathbf{q}, \Omega)$. Previous studies of $\chi(\mathbf{q}, \Omega)$ [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 $\chi(\mathbf{q}, \Omega)$ in the opposite limit of vanishing q and finite Ω [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 $\Pi(\mathbf{q}, \Omega)$ 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, ϕ , with a d -wave form-factor. If ϕ is a collective mode of fermions, the model describes an itinerant

fermionic system near a Pomeranchuk instability. If ϕ 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 $\text{FeSe}_{1-x}\text{S}_x$ [8, 9] and Fe-pnictides BaFe_2As_2 [10] and $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ [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 $\text{FeSe}_{1-x}\text{S}_x$, 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 $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ and BaFe_2As_2 we find a better agreement with the composite magnetic scenario of nematicity, consistent with the fact that nematic and magnetic ordering

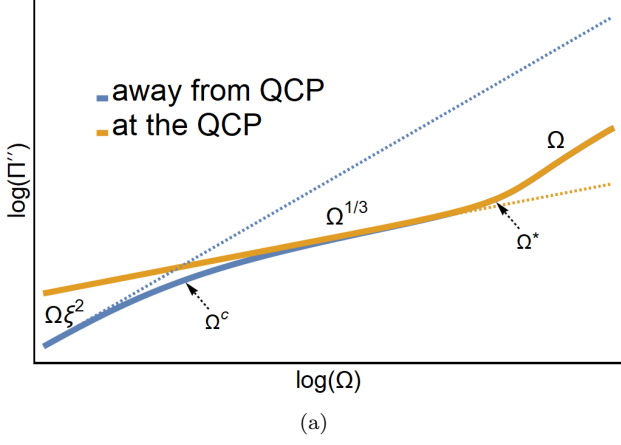


FIG. 1. The d-wave fermion polarization $\Pi''(q, \Omega)$ at $q = 0$ for a model of itinerant fermions near an Ising nematic quantum critical point. We use log-log scale to highlight different power-law regimes. At very low frequencies, $\Pi''(0, \Omega)$ scales as $\Omega\xi^2$. Above a crossover frequency $\Omega^c \propto \xi^{-3}$, $\Pi''(q, \Omega)$ becomes universal (independent of ξ) and has different forms in two regimes, depending on the ratio Ω/Ω^* , where $\Omega^* \ll \varepsilon_F$ is defined after Eq. (4). For $\Omega \ll \Omega^*$, $\Pi''(0, \Omega) \propto |\Omega|^{1/3}$. For $\Omega \gg \Omega^*$, $\Pi''(0, \Omega) \propto |\Omega|$. At even higher frequencies (not shown) $\Pi''(0, \Omega)$ passes through a maximum and decreases.

temperatures nearly coincide and in $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ 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 ϕ is coupled to the d -wave component of fermionic density as

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

where g is a coupling constant and $f(\mathbf{k})$ is a momentum dependent vertex with d -wave symmetry (e.g., $f(\mathbf{k}) = \cos k_x - \cos k_y$). Near the FS $f(\mathbf{k}) \approx f(\theta)$, where θ is an angle along the FS. This model has been discussed extensively in the regime where the characteristic bosonic frequency Ω 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

$$D(\mathbf{q}, \Omega) = \frac{\chi_0}{\xi_0^{-2} + \mathbf{q}^2 - \Omega^2/c^2 + \bar{g}\Pi(\mathbf{q}, \Omega)}. \quad (2)$$

Here $\bar{g} = g^2 \chi_0$ is the effective coupling constant (assumed much smaller than $\varepsilon_F \equiv v_F k_F/2$), ξ_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

$$\bar{g}\Pi(\mathbf{q}, \Omega) = -\frac{\bar{g}k_F^2}{4\pi\varepsilon_F} \left(\langle f^2 \rangle + i f^2(\hat{\mathbf{q}}') \frac{|\Omega|}{v_F |\mathbf{q}|} \right), \quad (3)$$

where $\varepsilon_F = v_F k_F/2$, $\langle f^2(\theta) \rangle = \int \frac{d\theta}{2\pi} f^2(\theta)$, and $\hat{\mathbf{q}}' = \hat{z} \times \hat{\mathbf{q}}$, \hat{z} is a unit vector in the direction perpendicular to the 2D plane [25, 26]. The constant term in $\bar{g}\Pi(\mathbf{q}, \Omega)$ accounts for the difference between ξ_0 and the actual nematic correlation length ξ : $\xi^{-2} = \xi_0^{-2} - \bar{g}k_F^2 \langle f^2 \rangle / (4\pi\varepsilon_F)$. The form of $D(\mathbf{q}, \Omega)$ at $\Omega \ll v_F |\mathbf{q}|$ determines the fermionic self-energy on the FS: at the QCP, $\Sigma(\Omega, \theta) \propto |f(\theta)|^{4/3} |\Omega|^{2/3} \omega_0^{1/3}$, where $\omega_0 \sim \bar{g}^2 / \varepsilon_F$.

Our goal is to obtain $\Pi(\mathbf{q}, \Omega)$ at $T = 0$ in the opposite limit of $\mathbf{q} = 0$ and finite Ω . We obtain $\Pi(0, \Omega)$ first along Matsubara axis $\Omega = i\Omega_m$, and then convert to real Ω . 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{g} , this is generally not the case. We evaluate $\Pi(0, \Omega_m)$ to leading order in \bar{g} 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

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

Here $\langle f_2 \rangle = \langle f^2 f'^2 + \frac{1}{2} f^3 f'' \rangle < 0$, and the functions A and C are

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

and

$$C(|\Omega_m|; k_F \xi) \sim \begin{cases} \frac{\bar{g} |\Omega_m|}{\varepsilon_F^2} (k_F \xi)^2 & |\Omega_m| \ll \Omega^c \\ \left(\frac{\bar{g} |\Omega_m|}{\varepsilon_F} \right)^{1/3} & \Omega^c \ll |\Omega_m| \ll \Omega^* \\ \frac{v_F}{c} \frac{|\Omega_m|}{\varepsilon_F} & \Omega^* \ll |\Omega_m| \ll \varepsilon_F \end{cases}. \quad (6)$$

The characteristic frequencies in (6) are $\Omega^c \sim \varepsilon_F^2 \bar{g}^{-1} (k_F \xi)^{-3}$ and $\Omega^* \sim \sqrt{\bar{g} \varepsilon_F (c/v_F)^3}$. The frequency Ω^c separates Fermi liquid and QC behavior, and Ω^* marks where the damping term starts to dominate over the bare Ω_m^2/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.

Nematic susceptibility When ϕ represents a collective mode of fermions near a d-wave Pomeranchuk instability, the nematic susceptibility is well approximated by $\chi_{Pom}(0, \Omega) = -\Pi(0, \Omega)/(1 + \bar{g}\xi_0^2 \Pi(0, \Omega))$ where $\bar{g}\xi_0^2$ is a four-fermion interaction in the d-wave channel. For $\Omega \ll v_F q$, one can verify that $\chi_{Pom}(\mathbf{q}, \Omega)$ has the same form as $D(\mathbf{q}, \Omega)$ in Eq. (2). For $\Omega \gg v_F q$, $\Pi(q \rightarrow 0, \Omega)$ is much smaller and $\chi_{Pom} \approx -\Pi(0, \Omega)$. When ϕ can

be considered an independent, near-critical bosonic field, the full bosonic susceptibility $\chi_{ind}(0, \Omega)$ predominantly comes from the ϕ field, i.e., $\chi_{ind}(0, \Omega) \approx D(0, \Omega)$, where D is given by Eq. (2). The behavior of $\chi_{ind}(0, \Omega)$ then depends on the scale ξ_0 as well as ξ . 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 behavior of $\delta\chi' = \chi'_{\xi=\infty}(0, \Omega \rightarrow 0) - \chi'_{\xi}(0, \Omega \rightarrow 0)$ and the slope of the imaginary part of the susceptibility $\Gamma = \chi''(0, \Omega)/\Omega|_{\Omega \rightarrow 0}$.

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

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

In the independent case, to leading order in \bar{g}/ε_F , $\chi'_{ind}(\Omega) = \chi_0 \xi_0^2$, and $\chi''_{ind}(\Omega) = -\chi_0 \bar{g} \xi_0^4 \Pi''(0, \Omega)$, where we recall that $\xi^{-2} = \xi_0^{-2} - \bar{g} k_F^2 (f^2)/(4\pi\varepsilon_F)$ [31] Therefore

$$\delta\chi'_{ind} \propto \xi^{-2} \xi_0^2, \quad \delta \left(\frac{1}{\chi'_{ind}} \right) \propto \xi^{-2}, \quad \Gamma_{ind} \propto \xi^2 \xi_0^4. \quad (8)$$

We emphasize that in both cases, $\chi'_{\xi=\infty}(\Omega \rightarrow 0)$ remains finite at the QCP because it differs from the thermodynamic nematic susceptibility $\chi'(\mathbf{q} \rightarrow 0, \Omega = 0)$, which scales as ξ^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 $\mathbf{q} \rightarrow 0$ 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 γ_{tr} . In this respect our $\chi'(0, \Omega \rightarrow 0)$ is actually the susceptibility for Ω much larger than γ_{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], Γ grows rapidly

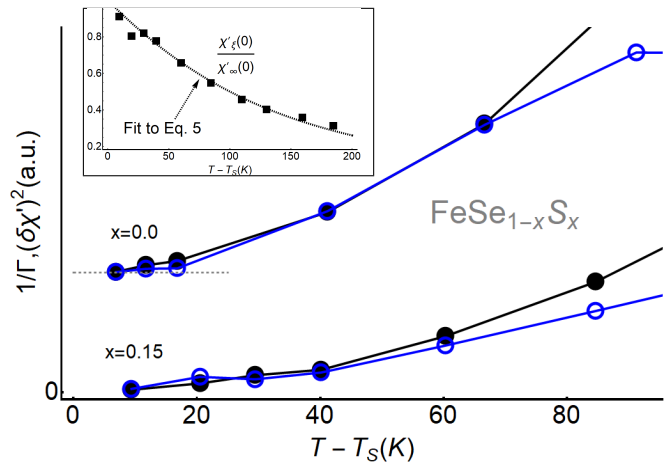


FIG. 2. B_{2g} Raman data on FeSe $_{1-x}$ S $_x$ for dopings $x = 0$ (shifted up for clarity) and $x = 0.15$, $(T - T_s)/T_s \lesssim 1$, taken from Ref. 9 [Similar data for $x = 0$ are in Ref. 8]. Filled circles - $1/\Gamma$, where $\Gamma = \chi''(0, \Omega)/\Omega$ is the slope of measured $\chi''(0, \Omega)$ at small frequencies. Open circles - $(\delta\chi')^2 = (\chi'_{\xi=\infty}(\Omega \rightarrow 0) - \chi'_{\xi}(\Omega \rightarrow 0))^2$, where $\chi'_{\xi}(\Omega \rightarrow 0)$ has been obtained from $\chi''(0, \Omega)$ by KK transform. The data for $(\delta\chi')^2$ have been rescaled by a constant factor [37]. The data show that $(\delta\chi')^2$ and $1/\Gamma$ scale together, i.e., their ratio is independent of ξ . Such behavior is consistent with the Pomeranchuk scenario described in the text. The inset shows the experimental $\chi'_{\xi}(\Omega \rightarrow 0)$ along with the fit to Eq. (5) using $\xi(T)$ extracted from the data for Γ [37].

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 χ'' are obtained at $\Omega \gg v_F q$, hence in $\chi'(0, \Omega \rightarrow 0)$ Ω/v_F 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 $\xi(x, T)$, 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 ξ^{-2} , Eq. (7). The data for FeSe $_{1-x}$ S $_x$ also show [9] that $\chi'_{KK}(0, 0)$ increases as the system approaches the nematic 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

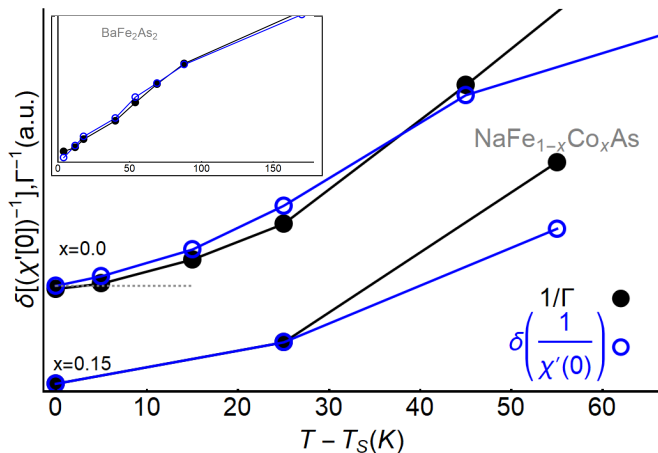


FIG. 3. B_{2g} Raman data for $\text{NaFe}_{1-x}\text{Co}_x\text{As}$ for dopings $x = 0$ (shifted up for clarity) and $x = 0.15$, $(T - T_s)/T_s \lesssim 1$ (taken from Ref. 11). Filled circles – $1/\Gamma$, open circles – $\delta(1/\chi')$. Inset – the same quantities for undoped BaFe_2As_2 (data from Ref. 10). The data show that $\delta(1/\chi')$ and $1/\Gamma$ scale together in both $\text{NaFe}_{1-x}\text{Co}_x$ and BaFe_2As_2 . Such behavior is consistent with the independent scenario, but the fact that the scaling holds in a wider range of T than expected requires further study.

k_F in this system is small for all pockets [19, 38, 39]), because in this case $\chi'(0, \Omega \rightarrow 0)$ increases as ξ^2 between ξ 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 Ω as $\Omega^{1/3}$, and therefore passes through a maximum at nonzero Ω . We consider the combination of these results as a strong indication that nematicity in $\text{FeSe}/\text{FeSe}_{1-x}\text{S}_x$ is caused by a d-wave Pomeranchuk instability.

For $\text{Ba}(\text{FeAs})_2$ and $\text{NaFe}_{1-x}\text{Co}_x\text{As}$, we found that the temperature dependence of $1/\Gamma$ 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 $1/\Gamma$ and $\delta(1/\chi')$ scale as ξ^{-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 $1/\Gamma \sim \delta(1/\chi')$ holds in our theory under the assumption that ξ_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 χ' varies significantly over the temperature ranges shown in Fig. 3 (Refs. [10, 11]).

Summary In this work we analyzed the dynamic response of a clean 2D system of itinerant fermions coupled to a scalar boson ϕ , 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 ϕ 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 $\text{FeSe}_{1-x}\text{S}_x$ and $\text{BaFe}_2\text{As}_2/\text{NaFe}_{1-x}\text{Co}_x\text{As}$. We argued that the data for $\text{FeSe}_{1-x}\text{S}_x$, which does not order magnetically down to $T = 0$, are well described by the d-wave charge Pomeranchuk scenario. The data for $\text{BaFe}_2\text{As}_2/\text{NaFe}_{1-x}\text{Co}_x\text{As}$ 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 $\chi(q \rightarrow 0, \Omega = 0)$ [16, 18, 34]) and therefore gives an additional contribution to the difference between ξ_0 and ξ . It also affects the momentum dependence of $D(q, \Omega)$ at $\Omega < v_F q$, 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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