Towards a Unified Description of the Electronic Orders in Iron-Based Superconductors: Insights from FeSe

1) Nature Communications **7**, 12182 (2016), by Qisi Wang, Yao Shen, Bingying Pan, Xiaowen Zhang, K. Ikeuchi, K. Iida, A. D. Christianson, H. C. Walker, D. T. Adroja, M. Abdel-Hafiez, Xiaojia Chen, D. A. Chareev, A. N. Vasiliev, Jun Zhao

2) arXiv:1603.04589, by P. Wang, S. Sun, Y. Cui, W. Song, T. Li, Rong Yu, Hechang Lei, Weiqiang Yu

3) arXiv:1603.04135 by K. Kothapalli, A. E. Böhmer, W. T. Jayasekara, B. G. Ueland, P. Das, A. Sapkota, V. Taufour, Y. Xiao, E. E. Alp, S. L. Bud'ko, P. C. Canfield, A. Kreyssig, A. I. Goldman

Recommended and a Commentary by Qimiao Si, Rice University

The 8-year-old field of iron-based superconductors (FeSCs) is quite young in the overall context of superconductivity. It was recognized from the beginning that superconductivity in the FeSCs is unconventional, with superconducting pairing driven by electron-electron Coulomb interactions instead of electron-phonon coupling. A notion that has played a central role in the field is that superconductivity in these systems occurs at the border of electronic orders, which are induced by the same electron correlations.

FeSCs have an extensive materials basis. Early work focused on the iron pncitides, such as SmFeAsO and $BaFe_2As_2$ with various chemical substitutions. More recently, the iron chalcogenides have occupied the center stage. The structurally simplest of these is the bulk FeSe, whose variants have provided a new record of T_c (about 65 K, according to the onset of the Meissner effect [1]) and a renewed hope of reaching even higher T_c. Across these FeSCs, the crystal structure is very similar but their electronic structure has a large variability. Comparing their properties provides important clues to the microscopic physics of the FeSCs.

A case in point is the magnetic and "nematic" properties. Consider, for example, the iron pnictide $BaFe_2As_2$. Structurally, it contains layers of FeAs, each of which includes a square lattice of Fe ions. Lowering the temperature leads to a tetragonal-to-orthorhombic distortion at T_s, which is closely followed by a Néel transition at $T_N \leq T_s$. The structural transition at T_s is driven by the transition into an electronic nematic state [3]. Because the AF order, at the wavevector $(\pi, 0)$, breaks the C_4 symmetry in the same way, the associated magnetic fluctuations are a driving force for the nematic order.

Compared to the above, FeSe might appear to be rather unusual. Here, a similar C_4-breaking transition is not accompanied by any magnetic transition. While proposals have been put forward that the underlying physics is very different in FeSe from the iron pnictides (invoking, in particular, an orbital order [2]), several theoretical studies have sought for a route towards a unified description of FeSe and the iron pnictides through the magnetic fluctuations. A possible starting point is the effective Hamiltonian describing quasi-localized magnetic moments with frustrated magnetic interactions:

$$H = \sum_{i,j} \left[J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + K_{ij} \left(\mathbf{S}_i \cdot \mathbf{S}_j \right)^2 \right] . \tag{A}$$

Here, $\mathbf{S}_i$ is taken as a spin-1 operator at site i of the Fe square lattice, and the interactions include both nearest-neighbor and further-neighbor terms. (A small interlayer interaction term should be added to properly describe the orders at nonzero temperatures.)

The magnetic frustration in this Hamiltonian leads to a variety of ground states. The $(\pi, 0)$ AF order, as appropriate for the iron pnictides, is only one of the ground states in the phase diagram. Others include a $(\pi, 0)$ antiferroquadrupolar (AFQ) order accompanied by a nematic order [4], and even a nematic quantum paramagnet [5].

In the context of this quest for a unified understanding, several recent experiments in FeSe have made significant progresses. In article #1, Q. Wang and collaborators reported inelastic neutron scattering measurements of $\chi''(\mathbf{q}, E)$, the dynamical spin susceptibility. They mapped out the magnetic excitation spectrum over a large range of energy E and an extended range of the wave vectors $\mathbf{q}$. Several results are obtained:

- In spite of the absence of any long-range magnetic order, the low-energy branch of the magnetic excitations emanate from $(\pi, 0)$, much like the AF-ordered iron pnictides. The twist is that the spectral weight is approximately linear in energy (up to about 50 meV). The fact that it vanishes in the zero energy limit is in accordance with the absence of dipolar static magnetic order, and the linear energy dependence is expected for the AFQ order at $(\pi, 0)$.

- The higher energy spin excitations bear even closer analogy with those of the AF-ordered iron pnictides, covering all the way to the wavevector (π, π) and extending to an energy of about 200 meV. These features are expected for both the AFQ order and the nematic quantum paramagnet.

- The total spectral weight, $\int dE d\mathbf{q} \chi''(\mathbf{q}, E)$, integrated over the Brillouin zone and the above energy range is about 5 μ_B^2/Fe. This exceeds the already large value of the AF-ordered iron pnictides, which is about 3 μ_B^2/Fe. Therefore, the magnetic excitations in FeSe are expected to be as important in interactions with fermions as in the iron pnictides.

The other two highlighted articles addressed the phase diagram by probing FeSe under pressure. It has been known for some time that pressure leads to quantum phase transition(s) into magnetic order. What has been left open is the nature of the magnetism in the pressurized FeSe. The two new studies go a long way to clarifying this issue:

- In article #2, P. Wang and collaborators carried out NMR measurements at different magnetic-field orientations, and concluded that the magnetic order is AF with wavevector $(\pi, 0)$. One of the important consequences is that the pressured FeSe must break the rotational symmetry of the square lattice.
- Indeed, in article #3, K. Kothapalli and collaborators demonstrated the C_4-symmetry breaking in the pressurized FeSe, showing that it undergoes a tetragonal to orthorhombic structural transition at a temperature comparable to the magnetic transition temperature.

What emerges from these two studies is a temperature-pressure phase diagram with pressure inducing a quantum phase transition (or, even a sequence of transitions) from the unusual nematic state at ambient pressure to a more standard $(\pi, 0)$ AF order at pressures on the order of 2 GPa. The fact that the pressurized FeSe orders magnetically with the same wavevector as that of the AF iron pnictides highlights the intimate connection between the electronic orders of the two classes of FeSCs.

Collectively, the new developments raise a number of new issues worthy of further explorations:

a) The results point to the viability of a unified understanding of the electronic orders in the iron-based systems in terms of local-moment magnetism. [The effective Hamiltonian Eq. (A) also describes the ordered ground state in another prototypical iron chalcogenide, FeTe.] In the case of the iron pnictides, a consistency check has been provided by the observation of a linear-temperature dependence of the bulk magnetic susceptibility, $\chi(T)$, with a large slope [6]. The observation of the large fluctuating spin moment in FeSe encourages a similar analysis, and the available experiments suggest that $\chi(T)$ of FeSe indeed behaves similarly [7].

b) At the ambient pressure, the nematic order in FeSe distorts the Fermi surface [8]. Under pressure, the $(\pi, 0)$ AF order is expected to not only distort the Fermi surface, but also reconstruct it. We then expect to see a sharp change to the Fermi surface in FeSe under pressure. Preliminary evidence for such an effect has emerged from quantum oscillation experiments [9], but further studies of the Fermi surface as a function of pressure in FeSe will certainly be very welcoming.

c) What is the relationship between the bulk FeSe and the single-layer FeSe on substrate, where superconductivity with the highest T_c in FeSCs has been demonstrated [1]? I could envision several ways that progress can be made to elucidate this connection. It would be very instructive to clarify the electronic orders in the case of multiple-layers of FeSe on similar substrates, in light of the new understandings on the bulk FeSe. Moreover, with point a) above in mind, it would also be instructive to determine the magnetic susceptibility of the single-layer FeSe. Finally, the intercalated FeSe systems (A. Fujimori, Commentary in the February 2016 issue of JCCM) may well serve as an important stepping stone in linking the new insights on the bulk FeSe to the much desired further understandings on the single-layer FeSe.

To reiterate, the three highlighted studies represent important steps towards a unified understanding of the magnetic and nematic correlations across the FeSCs. Given the close proximity of the superconductivity to the electronic orders, these types of new insights will surely be important to understanding the mechanism of the iron-based high temperature superconductivity.

In completing this commentary, I benefited from a discuss session of the Summer Program on Superconductivity currently ongoing at the Aspen Center for Physics. Readers are referred to the highlighted articles for more comprehensive references on this rapidly developing topic.

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[1] Z. Zhang *et al.*, Science Bulletin **60**, 1301-1304 (2015).

[2] S.-H. Baek *et al.*, Nat. Mater. **14**, 210 (2015); A. E. Böhmer *et al.*, Phys. Rev. Lett. **114**, 027001 (2015).

[3] J. H. Chu, H.-H. Kuo, J. G. Analytis, and I. R. Fisher, Science **337**, 710-712 (2012).

[4] R. Yu and Q. Si, Phys. Rev. Lett. **115**, 116401 (2015).

[5] F. Wang, S. A. Kivelson and D.-H. Lee, Nature Phys. **11**, 959-963 (2015).

[6] G. M. Zhang *et al.*, EPL **86**, 37006 (2009).

[7] G. E. Grechnev *et al.*, J. Phys.: Condens. Matter **25**, 046004 (2013).

[8] M. D. Watson *et al.*, Phys. Rev. B**91**, 155106 (2015).

[9] T. Terashima *et al.*, Phys. Rev. B**93**, 094505 (2016).