Final Report: *Competing Orders in Correlated Materials: Impact of Disorder and Non-Equilibrium Perturbations*

PI: Dr. Rafael M. Fernandes (University of Minnesota)

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The goal of this project was to explore, understand, and ultimately control the competing electronic ordered states ubiquitously present in correlated materials, with particular emphasis in unconventional superconductors, such as iron-based and copper-based materials. While on the one hand the competition with different types of magnetic, orbital, and charge order limits the transition temperatures of these unconventional superconductors, on the other hand the enlarged ground-state degeneracy associated with these multiple many-body instabilities can give rise to unusual inhomogeneous correlated normal states, such as electronic smectic and nematic phases. To achieve the aforementioned goals, we employed a multi-faceted theoretical approach consisting of: (i) The investigation of relatively unexplored regimes with the potential to unveil novel behaviors – in particular, the study of competing phases taken out of equilibrium to explore the potential to enhance the transition temperatures of unconventional superconductors by optical pulses. (ii) The embracement of realistic features of correlated materials in their microscopic descriptions – in particular, the investigation of the impact that disorder, in its various forms, has on competing and emergent inhomogeneous states present in the phase diagrams of unconventional superconductors. (iii) The promotion of synergy with established and novel experimental probes (with particular emphasis on scanning tunneling microscopy and ultrafast spectroscopy) not only by using data as input of theoretical models, but also by providing concrete guidance for experiments.

A total of 68 manuscripts acknowledging this award have been published or submitted for publication [1–68]. The full list can be found in the end of this report, and includes papers published in *Nature Physics*, *Nature Communications, Physical Review Letters*, *PNAS*, *Physical Review X* and *Physical Review B,* among other journals. This three-page report provides an overview of the main accomplishments reported in these manuscripts, highlighting their impact on the elucidation of correlated materials. Overall, our studies shed new light on the intricate connection between the competing phases present in the phase diagrams of several correlated materials, unveiling novel routes to control and exploit them.

One of the highlights was the development of a unified theoretical framework to elucidate the several different ordered states that emerge in the underdoped phase diagram of the iron-based superconductors. In this framework, the rich landscape of ordered phases observed experimentally can be attributed to the same microscopic electronic interactions, which promote different types of magnetically ordered states as well as partially-melted magnetic phases with non-trivial orbital order. In particular, for a long time, it was believed that the magnetic ground state of the iron pnictides was the orthorhombic stripe-type magnetic state. Theoretical studies, including our own [2, 5], showed that the same low-energy electronic model also favors double-**Q** magnetic phases that preserve tetragonal symmetry. In [2], it was shown that these so-called C_4 magnetic phases can be stabilized by doping. The two types of C_4 double-**Q** phases were dubbed, in Ref. [13], spin-vortex crystal (a double-**Q** non-collinear phase) and charge-spin density-wave (a double-**Q** collinear phase). Both types of C_4 order were directly observed experimentally in Co-doped $CaKFe_4As_4$ and in Na-doped $SrFe_2As_2$, respectively, via joint experimental-theoretical collaborations [14, 38]. Interestingly, in Ref. [5] we found that the C_4 phases compete much more strongly with superconductivity than the stripe

phase, in agreement with experimental observations.

An important question motivated by experiments was why nearly-degenerate C_4 and stripe magnetic phases proliferate near the putative magnetic quantum critical point. In Refs. [42, 43], we attributed this behavior to the interplay between spin-orbit coupling and quantum fluctuations. In particular, spin-orbit coupling promotes spin anisotropy, which in our renormalization-group calculations was found to promote a stable Gaussian fixed point with essentially degenerate C_4 and stripe magnetic phases. The impact of spin-orbit coupling on the magnetic properties of the iron-pnictides was invoked by us to explain the spin-reorientation seen in some hole-doped pnictides [12], to address the experimentally-observed anisotropy of the so-called resonance mode inside the superconducting state [28], and to predict an unusual type of loop-current order that accompanies stripe magnetism [41].

An important property of the stripe magnetic phase is that it can melt in two stages, giving rise to an intermediate nematic state in which spin-rotational symmetry is restored, but tetragonal symmetry remains broken. In this framework, the nematic phase can be thus understood as a vestige of the stripe magnetic order, which naturally explains why the nematic and magnetic transition temperature lines follow each other closely in the phase diagrams of most iron-based superconductors. In Ref. [13], we showed that the C_4 magnetic states also support vestigial phases, characterized by emergent charge order (in the case of the charge-spin density-wave) or emergent spin-current order (in the case of the spin-vortex crystal). In the same way that nematic order triggers ferro-orbital order, we found that these vestigial phases are characterized by unusual types of orbital order, manifested for instance as Rashba/Dresselhaus spin-orbit interactions and staggered atomic spin-orbital coupling [60]. These investigations not only provided a simple and appealing unifying framework to explain the complexity of the normal-state instabilities of the iron-based superconductors, but also led to a much broader understanding of the concept of vestigial orders beyond nematicity or iron-pnictide compounds. As a result, the concept of vestigial phases was applied to explain a puzzling phase transition seen in titanium oxypnictides [31] and the unusual response to strain of certain triangular-lattice antiferromagnets [65]. The promising idea of vestigial order as a route to explain the intertwinement between different ordered states in correlated systems was reviewed by us in Ref. [57].

While this model of nematic order as a vestige of a stripe magnetic phase was employed to successfully explain a variety of experimental observations on several iron-based superconductors – including those arising from NMR, neutron scattering, and scanning tunneling microscopy measurements [4, 23, 39, 48, 55] – one challenge is the case of FeSe, where nematic order appears in the absence of long-range magnetic order. Importantly, FeSe in monolayer form displays the highest T_c of any iron superconductor. Our works in Refs. [7, 10, 27] proposed appealing theoretical scenarios in which magnetic fluctuations promote nematic order even when stripe magnetic order is completely suppressed. Different aspects of the interplay between nematicity and superconductivity in FeSe featured in a series of publications. In Ref. [24], we showed that pairing mediated by nematic fluctuations in thin-film FeSe leads to degenerate s-wave and d-wave gap states, with the degeneracy being lifted by spin-orbit coupling. Refs. [46, 50] focused on the situation where superconductivity emerges in a state with pre-existing long-range nematic order, such as the case of bulk FeSe. The impact of nematicity on superconductivity was invoked to explain the unusual anisotropy of the gap function observed experimentally, and to predict the possible formation, at low enough temperatures, of an exotic superconducting state that breaks time-reversal symmetry. Successful models were also developed to explain the origin of the resistivity anisotropy observed experimentally in the nematic phase of FeSe [21, 22], as well as the puzzling ARPES and scanning tunneling microscopy data on FeSe [66].

Beyond nematicity in FeSe, we also addressed how a novel B_{1g} nematic phase emerges in heavily hole-doped $BaFe_2As_2$ due to a change in the dominant magnetic interactions [62]. In this B_{1g} nematic phase, the Fe-As-Fe bonds become inequivalent, in contrast to the more standard B_{2g} nematic phase observed in other iron pnictides, where the Fe-Fe bonds become unequal. The transition from B_{2g} to B_{1g} nematics that is expected to take place upon doping led to our joint theoretical-experimental proposal of a nearly-XY nematic phase in hole-doped pnictides [54]. Besides iron-based superconductors, we also explored manifestations

of nematic order in nickel-arsenides [64], in f-electron systems [9, 35, 56, 68], and in cuprates [8].

The competition between unconventional superconductivity and antiferromagnetism was another topic intensively studied in this award, via phenomenological approaches [34], microscopic calculations [53], and joint theoretical-experimental investigations [18, 32, 45]. Interesting results came from extensive Quantum Monte Carlo (QMC) simulations of the spin-fermion model [33, 44], where electrons interact with each other only via the exchange of antiferromagnetic fluctuations, which can be tuned to become quantum critical. The two-band version of the spin-fermion model can be simulated with QMC without the infamous fermionic sign-problem. As a result, one can probe much lower temperatures than those accessible via QMC simulations that suffer from the sign problem, providing important benchmarks to compare with analytical field-theoretical calculations. In Ref. [33], we demonstrated that the superconducting instability of the spin-fermion model is governed by special states of the Fermi surface called hot spots, validating an analytical hot-spots Eliashberg approximate solution to the spin-fermion model. In Ref. [44], we established that the same antiferromagnetic fluctuations that favor superconductivity also promote an unusual d-wave charge order, which however is fragile against the emergence of the former, as it is strongly affected by states other than those near the hot spots.

Another set of studies shed new light on the impact of disorder on competing and vestigial orders. Ref. [47] showed that rare-region effects, typically found in inhomogeneous systems, have a very strong impact on the coupled vestigial nematic and stripe magnetic phase transitions at the putative quantum critical point. Specifically, while the two zero-temperature transitions are predicted to be simultaneous and first-order in a completely clean system, in disordered systems the effects of rare regions devoid of impurities split the two transitions, giving rise to a smeared nematic quantum critical point and to an inhomogeneous nematic state. The latter may offer an interesting avenue to understand the puzzling experimental observation of inhomogeneous nematic fluctuations in a doped iron pnictide superconductor [16]. The dramatic impact of disorder on quantum critical points was also explored in Ref. [19], where it was shown that pairing promoted by the exchange of quantum critical magnetic fluctuations is much more robust against pair-breaking impurity scattering than the case of classical fluctuations. Disorder was demonstrated to be a viable tuning knob to tilt the balance between competing orders, be it superconductivity and magnetism [58] or C_4 versus stripe magnetic phases [17].

Non-equilibrium perturbations were shown to be another promising way to manipulate phases that compete with superconductivity. In Ref. [37], we proposed to excite non-equilibrium acoustic phonons (via e.g. shock waves) to selectively melt density-wave orders that compete with superconductivity. The basis of this idea is that driven acoustic phonons generate a momentum-dependent effective temperature, making different regions of the Fermi surface subject to distinct effective temperatures. To control competing nematic orders, we proposed the excitation of an optical E_u phonon mode generically present in tetragonal systems [52]. Both proposals avoid excessive heating, which is very detrimental to superconductivity. Besides the possibility to control competing orders, non-equilibrium excitations can also be used to probe the structure of the superconducting condensate by providing access to the coherent dynamics of the Cooper pairs. In the joint pump-probe experimental-theoretical investigation reported in Ref. [59], we put forward a formalism to explain the puzzling observation of damping of the superconducting order parameter in the picosecond time scale, before lattice relaxation sets in. In Ref. [67], we developed a powerful self-consistent perturbative method that predicts unique signatures of multi-band superconductivity in the gap dynamics.

A final highlight is our recent strong-coupling theoretical model developed to describe the correlated phases of twisted bilayer graphene [51]. For twist angles near the so-called magic angle, the gate-voltage temperature phase diagram of this system was recently shown to display superconducting domes in the vicinities of correlated insulating phases. Our work derives a strong-coupling expansion of a low-energy model used to describe the nearly-flat bands that appear near the magic angle, resulting in an effective anisotropic SU(4) model.

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