

INTERPLAY BETWEEN SUPERCONDUCTIVITY AND MAGNETISM IN IRON-BASED SUPERCONDUCTORS

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PROJECT SCOPE

The goal of the project was to understand theoretically the interplay between superconductivity, magnetism, and nematic order in Iron-based superconductors (FeSCs) which currently attract high attention in the physics community. The key goal was to understand the mechanism of superconductivity, the origin of magnetism and of the nematic order which accompanies structural distortion, the co-existence regimes of different orders, and the observed pseudogap-like behavior in the normal state. My research explores the idea that superconductivity in FeSCs is of electronic origin and is caused by the exchange of spin-fluctuations, enhanced due to close proximity to antiferromagnetism. Spin-fluctuation approach has been discussed over a number of years for the cuprates where it gives rise to a d-wave superconductivity. For the pnictides, which are multi-band materials, spin-fluctuation approach leads to an attraction in both d-wave and extended s-wave ($s+-$) channels, and it becomes an issue what type of order the system chooses at a given doping. Besides, due to the presence of multiple Fermi surface sheets, both s-wave and d-wave gaps can have quite non-trivial forms, e.g., an $s+-$ gap may have nodes. The understanding of magnetically-mediated superconductivity in FeSCs is quite important as it will contribute to a generic understanding of the pairing of fermions near quantum-critical points – the problems ranging from s-wave pairing by soft optical phonons to color superconductivity of quarks mediated by a gluon exchange. Antiferromagnetism and superconductivity are neighbors in many systems: cuprates, heavy-fermion materials, 5d- oxides, cobaltates, and my proposed study of the interplay between magnetic and superconductivity should have impact on these systems as well.

My specific research on Fe-pnictides was focused on several issues issues: a search for non-trivial superconductivity, chiefly the one which breaks time-reversal symmetry, on the understanding of superconductivity and magnetic properties in systems with only hole or only electron pockets, on the evolution of the superconducting gap structure with doping, and on possible novel s-wave superconductivity in LaFeAs . The key goal of these studies was to understand whether multi-orbital/multi-band structure of Fe-pnictides leads to qualitatively new physics, which is not present in other unconventional superconductors, like cuprates and heavy-fermion materials.

The project started on September 1, 2012 and ran for two years with no-cost extension for the third year. Approved budget – \$300,000 for two years.

HIGHLIGHTS

My main accomplishments during the grant period are

- With my student S. Maiti, we analyzed the evolution of the superconducting gap structure in strongly hole doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ between $x = 1$ and $x \sim 0.4$ (optimal doping). In the latter case, the pairing state is most likely $s\pm$, with different gap signs on hole and electron pockets, but with the same signs of the gap on the two Γ -centered hole pockets (a $++$ state on hole pockets). In a pure KFe_2As_2 ($x = 1$), which has only hole pockets, laser ARPES data suggested another $s\pm$ state, in which the gap changes sign between hole pockets (a $+-$ state). We analyze how $++$ gap transforms into a $+-$ gap as $x \rightarrow 1$. We found that this transformation occurs via an intermediate $s+is$, state in which the gaps on the two hole pockets differ in phase by ϕ , which gradually involves from $\phi = \pi$ (the $+-$ state) to $\phi = 0$ (the $++$ state). This state breaks time-reversal symmetry and has huge potential for applications. We computed the dispersion of collective excitations and showed that two different Leggett-type phase modes soften at the two end points of TRSB state.
- With my other student A. Hinijosa, we analyzed superconductivity in the coexistence region with spin-density-wave (SDW) order in weakly doped Fe-pnictides and argued that it differs qualitatively from the ordinary s^{+-} state outside the coexistence region, as it develops an additional gap component which is a mixture of intra-pocket singlet (s^{++}) and inter-pocket spin-triplet pairings (the t -state). The coupling constant for the t -channel is proportional to the SDW order and involves interactions that do not contribute to superconductivity outside of the SDW region.
- With M. Khodas from U. of Iowa we analyzed the pairing symmetry in Fe-based superconductors AFe_2Se_2 ($\text{A} = \text{K}, \text{Rb}, \text{Cs}$) which contain only electron pockets. We argued that the pairing condensate in such systems contains not only intra-pocket component, but also inter-pocket component, made of fermions belonging to different electron pockets. We analyzed the interplay between intra-pocket and inter-pocket pairing depending on the ellipticity of electron pockets and the strength of their hybridization. We showed that with increasing hybridization the system undergoes a transition from a d -wave state to an s^{+-} state, in which the gap changes sign between hybridized pockets. This s^{+-} state has the full gap and at the same time supports spin resonance, in agreement with the data. Near the boundary between d and s^{+-} states we found a long-sought $s + id$ state which breaks time-reversal symmetry. In subsequent papers we analyzed the excitation spectrum and neutron and Raman intensities in such novel s -wave state.
- With R. Fernandes and J. Schmalian, we analyzed in detail the origin of nematic order in Fe-pnictides. The existence of nematic order in iron-based superconductors is now a well-established experimental fact, yet its origin remains controversial. Nematic order breaks the discrete lattice rotational symmetry by making the x - and y -directions in the iron plane non-equivalent. This can happen because of a regular structural transition or due to a electronically-driven instability – in particular, orbital order and spin-driven Ising-nematic order. The latter is a magnetic state that breaks rotational

symmetry but preserves time-reversal symmetry. Symmetry dictates that the development of one of these orders immediately induces the other two, making the origin of nematicity a physics realization of the chicken and egg problem. We reviewed different scenarios and argued that the evidence strongly points to an electronic mechanism of nematicity, placing nematic order in the class of correlation-driven electronic instabilities, like superconductivity and density-wave transitions. We discussed different microscopic models for nematicity and linked them to the properties of the magnetic and superconducting states, what allowed us to provide a unified perspective on the phase diagram of the iron pnictides.

- With F. Ahn and others we analyzed the structure of the pairing interaction and superconducting gap in LiFeAs by decomposing the pairing interaction for various k_z cuts into s - and d -wave components and by studying the leading superconducting instabilities. We used the ten orbital tight-binding model, derived from *ab-initio* LDA calculations with hopping parameters extracted from the fit to ARPES experiments. We found that the pairing interaction almost decouples between two subsets, one consists of the outer hole pocket and two electron pockets, which are quasi-2D and are made largely out of d_{xy} orbital, and the other consists of the two inner hole pockets, which are quasi-3D and are made mostly out of d_{xz} and d_{yz} orbitals. Furthermore, the bare inter-pocket and intra-pocket interactions within each subset are nearly equal. In this situation, small changes in the intra-pocket and inter-pocket interactions due to renormalizations by high-energy fermions give rise to a variety of different gap structures. We focus on s -wave pairing which, as experiments show, is the most likely pairing symmetry in LiFeAs. We found four different configurations of the s -wave gap immediately below T_c : the one in which superconducting gap changes sign between two inner hole pockets and between the outer hole pocket and two electron pockets, the one in which the gap changes sign between two electron pockets and three hole pockets, the one in which the gap on the outer hole pocket differs in sign from the gaps on the other four pockets, and the one in which the gaps on two inner hole pockets have one sign, and the gaps on the outer hole pockets and on electron pockets have different sign. Different s -wave gap configurations emerge depending on whether the renormalized interactions increase attraction within each subset or increase the coupling between particular components of the two subsets. We discuss the phase diagram and experimental probes to determine the structure of the superconducting gap in LiFeAs. We argued that the state with opposite sign of the gaps on the two inner hole pockets has the best overlap with ARPES data. We also argued that at low T , the system may enter into a "mixed" $s + is$ state, in which the phases of the gaps on different pockets differ by less than π and time-reversal symmetry is spontaneously broken.
- With R. Fernandes and others I analyzed the type of spin-density wave (SDW) order in doped iron-pnictides and the discontinuities of the superconducting transition temperature T_c in the coexistence phase with SDW magnetism. By tracking the magnetic

transition line $T_N(x)$ towards optimal doping within an itinerant fermionic model, we found a sequence of transitions from the stripe-orthorhombic (C_2) SDW order to the tetragonal (C_4) order and then back to the C_2 order. We argued that the superconducting T_c has two discontinuities – it jumps to a smaller value upon entering the coexistence region with the C_4 magnetic phase, and then jumps to a larger value inside the SDW state when it crosses the boundary between the C_4 and C_2 SDW orders. We found the full agreement with the experimental phase diagram and argued that this provides a strong indication that the itinerant approach is adequate to describe the physics of weakly/moderately doped iron-pnictides.

- With S. Maiti and M. Sigrist we analyzed $s+is$ state proposed as a candidate superconducting state for strongly hole-doped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$. Such a state breaks time-reversal symmetry (TRS) but does not break any other discrete symmetry. We addressed the issue whether TRS breaking alone can generate spontaneous currents near impurity sites, which could be detected in, e.g., μSR experiments. We argued that there are no spontaneous currents if only TRS is broken. However, supercurrents do emerge if the system is put under external strain and C_4 lattice rotation symmetry is externally broken.
- With R. Fernandes and J. Schmalian we studied the origin of the 90 K nematic transition in the chalcogenide FeSe, which displays no magnetic order down to $T = 0$. We analyzed this problem in light of recent experimental data which reveal very small Fermi pockets in this material. We showed that the smallness of the Fermi energy leads to a near-degeneracy between magnetic fluctuations and fluctuations in the charge-current density-wave channel. While the two fluctuation modes cooperate to promote the same preemptive Ising-nematic order, they compete for primary order. We argued that this explains why in FeSe the nematic order emerges when the magnetic correlation length is smaller than in other Fe-based materials, and why no magnetism is observed. We discussed how pressure lifts this near-degeneracy, resulting in a non-monotonic dependence of the nematic transition with pressure, in agreement with experiments.
- With my student A. Hinojosa we studied the effects of hybridization between the two electron pockets in Fe-based superconductors with s -wave gap with accidental nodes. We argued that hybridization reconstructs the Fermi surfaces and also induces an additional inter-pocket pairing component. We analyzed how these two effects modify the gap structure by tracing the position of the nodal points of the energy dispersions in the superconducting state. We found three possible outcomes. In the first, the nodes simply shift their positions in the Brillouin zone; in the second, the nodes merge and disappear, in which case the gap function has either equal or opposite signs on the electron pockets; in the third, a new set of nodal points emerges, doubling the original number of nodes.
- I wrote three review articles on Fe-pnictides and related materials. One is on electronic itinerant mechanism of superconductivity and magnetism in Fe-pnictides. This is the

summary of my research on pnictides over the two years of DOE grant period. The two other reviews were written with my student S. Maiti, and are on general theoretical consideration of superconductivity in itinerant systems with repulsive interaction.

- With P. Hirschfeld I wrote an article in Physics Today about the present status of the research on iron-based superconductors. We summarized what we learned about these materials over the last seven years, what are open questions, and what new physics we expect to extract from studies of this new class of high-temperature superconductors.
- I also studied the behavior of the London penetration depth near a magnetic quantum-critical point, the effect of fluctuations in the Cooper channel on the spin resonance in s^{+-} superconductors, the interplay between nematicity and superconductivity, and on the development of a novel magnetic order near a critical doping where magnetism ends (the work done in collaboration with experimentalists from Argonne Natl. Lab)

I also did work on superconductivity and charge order in hole and electron-doped cuprates.

- With my student Y. Wang we analyzed charge order in hole-doped cuprates within the spin-fermion model. We show that magnetically-mediated interaction, which is known to give rise to non-Fermi liquid behavior in the normal state, d -wave superconductivity, and charge order with momentum along zone diagonal, also gives rise to charge order with momenta $Q_x = (2Q, 0)$ and $Q_y = (0, 2Q)$ consistent with the experiments. We showed that an instability towards $\Delta_k^Q = \langle c_{\mathbf{k}+\mathbf{Q}}^\dagger c_{\mathbf{k}-\mathbf{Q}} \rangle$ with $\mathbf{Q} = Q_x$ or Q_y is a threshold phenomenon, but the dimensionless spin-fermion coupling is above the threshold if the magnetic correlation length ξ exceeds a certain critical value. At a critical ξ , the onset temperature for the charge order terminates at a quantum-critical point distant from the magnetic one. We find that the charge order with Q_x or Q_y changes sign under $\mathbf{k} \rightarrow \mathbf{k} + (\pi, \pi)$, but $|\Delta_k^Q| \neq |\Delta_{k+(\pi,\pi)}^Q|$. In real space, such an order has both bond and site components, the bond one is larger. We further argued that Δ_k^Q and Δ_{-k}^Q are not equivalent, and their symmetric and antisymmetric combinations describe, in real space, incommensurate density modulations and incommensurate bond current, respectively. We derived Ginzburg-Landau functional for four-component $U(1)$ order parameters $\Delta_{\pm k}^Q$ with $\mathbf{Q} = Q_x$ or Q_y and analyzed it first in mean-field theory and then beyond mean-field. Within mean-field, we found two possible ordered states, depending on the interplay between system parameters. Both states spontaneously break C_4 lattice rotational symmetry down to C_2 . In the first state incommensurate density and current modulations emerge with different \mathbf{Q} (e.g., one with Q_x , another with Q_y) and time-reversal symmetry is unbroken. In the other state, density and current modulations emerge with the same \mathbf{Q} and, we argued, differ in phase by $\pm\pi/2$. The selection of $\pi/2$ or $-\pi/2$ breaks time-reversal symmetry. We extended the analysis beyond mean-field and argue that discrete symmetries get broken before long-range charge order sets in. For the second state, which, we argued, is most closely related to hole-doped cuprates, we showed that, upon lowering the temperature, the system first breaks Z_2 lattice rotational symmetry ($C_4 \rightarrow C_2$) at $T = T^*$ and develops a nematic

order, then breaks Z_2 time-reversal symmetry at $T_t < T^*$ and locks the relative phase between density and current fluctuations, and finally breaks $O(2)$ symmetry of a common phase of even and odd components of Δ_k^Q at $T = T_{cdw} < T_t < T^*$ and develops a true charge order. We argued that at a mean-field level T_{cdw} is smaller than superconducting T_c , but pre-emptive composite order lifts T_{cdw} and reduces T_c such that at large ξ charge order develops prior to superconductivity. We obtained the full phase diagram and presented quantitative comparison of our results with ARPES data.

- We extended this analysis in the work with A. Tsvelik, where we used other computational scheme to demonstrate that pre-emptive composite order, which breaks time-reversal symmetry, develops at temperatures higher than the ones where a true charge order develops.
- In a long PRB paper with Y. Wang and D. Agtenerg, we analyzed the interplay between charge-density-wave (CDW) and pair-density-wave (PDW) orders within the spin-fermion model for the cuprates. We specifically considered CDW order with transferred momenta $(\pm Q, 0)/(0, \pm Q)$, as seen in experiments on the cuprates, and PDW order with total momenta $(0, \pm Q)/(\pm Q, 0)$. Both orders have been proposed to explain the pseudogap phase in the cuprates. We showed that both emerge in the spin-fermion model near the onset of antiferromagnetism. Each order parameter is constructed out of pairs of fermions in “hot” regions on the Fermi surface, breaks $U(1)$ translational symmetry, and changes sign when the momenta of the fermions change by (π, π) . We further argue that the two orders are nearly degenerate due to an approximate $SU(2)$ particle-hole symmetry of the model. This near degeneracy is similar in origin to that relating conventional d -wave superconducting order and bond charge order with momentum $(Q, \pm Q)$. The $SU(2)$ symmetry becomes exact if one neglects the curvature of the Fermi surface in hot regions, in which case $U(1)$ CDW and PDW order parameters become components of an $SO(4)$ -symmetric PDW/CDW “super-vector”. We developed a Ginzburg-Landau theory for four PDW/CDW order parameters and find two possible ground states: a “stripe” state in which both CDW and PDW orders develop with either $(\pm Q, 0)$ or $(0, \pm Q)$, and a “checkerboard” state, where each order can develop with $(\pm Q, 0)$ and $(0, \pm Q)$. We showed that the $SO(4)$ symmetry between CDW and PDW can be broken by two separate effects. One is the inclusion of Fermi surface curvature, which selects a PDW order immediately below the instability temperature. Another is the overlap between different hot regions, which favors CDW order at low temperatures. For the stripe state, we showed that the competition between the two effects gives rise to a first-order transition from PDW to CDW inside the ordered state. We also argued that beyond mean-field theory, the onset temperature for CDW order is additionally enhanced due to feedback from a preemptive breaking of Z_2 time-reversal symmetry. We discussed the ground state properties of a pure PDW state and a pure CDW state, and showed in particular that the PDW checkerboard state yields a vortex-anti-vortex lattice. For the checkerboard state, we considered a situation when both CDW and PDW orders are present at low

T and showed that at small but finite Fermi surface curvature the presence of both condensates induces a long sought chiral $s + id_{xy}$ superconductivity.

- With Y. Wang and D. Agterberg we analyzed incommensurate charge-density-wave (CDW) and pair-density-wave (PDW) orders with transferred momenta $(\pm Q, 0)/(0, \pm Q)$ in underdoped cuprates within the spin-fermion model. Both orders appear due to exchange of spin fluctuations before magnetic order develops. We argued that the ordered state with the lowest energy has non-zero CDW and PDW components with the same momentum. Such a state breaks C_4 lattice rotational symmetry, time-reversal symmetry, and mirror symmetries. We argued that the feedback from CDW/PDW order on fermionic dispersion is consistent with ARPES data. We discussed the interplay between the CDW/PDW order and $d_{x^2-y^2}$ superconductivity and make specific predictions for experiments.
- With Y. Wang we presented a comparative analysis of superconducting and charge-density-wave orders in the spin-fluctuation scenario for the cuprates. That spin-fluctuation exchange gives rise to d-wave superconductivity is well known. Several groups recently argued that the same spin-mediated interaction may also account for charge-density-wave order with momenta $(Q, 0)$ or $(0, Q)$, detected in underdoped cuprates. This has been questioned on the basis that charge-density-wave channel mixes fermions from both nested and anti-nested regions on the Fermi surface, and fermions in the anti-nested region do not have a natural tendency to form a bound state, even if the interaction is attractive. We showed that anti-nesting is not an obstacle for charge order, but to see this one needs to go beyond the conventional Eliashberg approximation. We showed that in the perfect nesting/antinesting case, when the velocities of hot fermions are either parallel or antiparallel, the onset temperatures in superconducting and charge-density-wave channels are of comparable strength for any magnetic correlation length ξ . The superconducting T_{sc} is larger than T_{cdw} , but only numerically. When the velocities of hot fermions are not strictly parallel/antiparallel, T_{cdw} progressively decreases as ξ decreases and vanishes at some critical ξ .
- Other accomplishments on the cuprates include the calculation of superconducting T_c in electron-doped cuprates (with Y. Wang), the detailed analysis of optical conductivity and the derivation of a critical Fermi liquid theory near a magnetic quantum-critical point, and the analysis of superconductivity in fermionic systems on hexagonal lattice.

I also did work on quantum magnetism in anisotropic triangular antiferromagnets (with O. Starykh). In particular, we analyzed instabilities of the collinear up-up-down state of a two-dimensional quantum spin- S spatially anisotropic triangular lattice antiferromagnet in a magnetic field. We found within large- S approximation, that near the end point of the plateau, the collinear state becomes unstable due to condensation of two-magnon bound pairs rather than single magnons. The two-magnon instability leads to a novel 2D vector chiral phase with alternating spin currents but no magnetic order in the direction transverse to the field. This phase breaks a discrete Z_2 symmetry but preserves a continuous $U(1)$

one of rotations about the field axis. It possesses orbital antiferromagnetism and displays a magnetoelectric effect.

A list of papers in which DOE support is acknowledged.

- **Vertical loop nodes in iron-based superconductors** (with M. Khodas), Phys. Rev. B 86, 144519 (2012).
- **Superconductivity at the onset of spin-density-wave order in a metal** (with Yuxuan Wang) Phys. Rev. Lett. 110, 127001 (2013).
- **Enhancement of the London penetration depth in pnictides at the onset of SDW order under superconducting dome** (with A. Levchenko, M.G. Vavilov, and M. Khodas) Phys. Rev. Lett. 110, 177003 (2013).
- **s+is State with Broken Time Reversal Symmetry in Fe-Based Superconductors** (with Saurabh Maiti) Phys. Rev. B 87, 144511 (2013).
- **Spin-current order in anisotropic triangular antiferromagnets** (with Oleg A. Starykh) Phys. Rev. Lett. 110, 217210 (2013).
- **Superconductivity from repulsive interaction** (with Saurabh Maiti), 71 pages, Review article in "Proceedings of the XVII Training Course in the physics of Strongly Correlated Systems", Vietri sul Mare (Salerno), Italy, 2013. **Superconductivity from repulsion** (with Saurabh Maiti), Expanded version, 110 pages, Review article in "Superfluidity", vol. 2 (Bennemann and Ketterson eds) Oxford Univ. Press, 2014
- **How many quantum phase transitions exist inside the superconducting dome of the iron pnictides?** (with Rafael M. Fernandes, Saurabh Maiti, and Peter Wlfle), Phys. Rev. Lett. 111, 057001 (2013).
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- **Interplay between pair- and charge-density-wave orders in underdoped cuprates** (with Yuxuan Wang and Daniel F. Agterberg) Phys. Rev. B 91, 115103 (2015).
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- **Polar Kerr effect from chiral-nematic charge order** (with Yuxuan Wang and Rahul Nandkishore) Phys. Rev. B 90, 205130 (2014).
- **Time-Reversal Symmetry Breaking Superconductivity in the Coexistence Phase with Magnetism in Fe Pnictides** (with Alberto Hinojosa and Rafael M. Fernandes) Phys. Rev. Lett. 113, 167001 (2014).
- **Iron-based superconductors, seven years later** (with P. Hirschfeld) Phys. Today, 68, number 6, p. 46 (2015).
- **Fermi liquid on its way to a critical point – the role of planar diagrams** (with Sung-Sik Lee and Yong Baek Kim) - in preparation
- **Renormalization group analysis of Fe-pnictides – the role of Coulomb interaction** (with I. Aleiner and B. Altshuler), in preparation.

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