Exchange Constants and Neutron Spectra of Iron Pnictide Materials

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We show that the neutron scattering spectra observed in CaFe$_2$As$_2$ by Zhao et al (arXiv:0903.2686v1) and the highly unusual spatially anisotropic exchange constants in the iron pnictides derived by Han et al (Phys. Rev. Lett. 102, 107003 (2009)), using electronic structure calculations, can be explained by assuming a role for orbital order in these materials. We write down a simple model Hamiltonian with tetragonal symmetry, whose spin-wave spectra describes the observed dispersion relations. We further argue that these materials have orbital selective Mott transition, which is driven by superexchange between neighboring iron atoms. We suggest that reduced spin and quasi-one dimensionality and not frustration are responsible for the reduced moments in these materials.

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The discovery of superconductivity at relatively high temperatures in the iron pnictide family of materials has invigorated condensed matter research. In order to develop a fundamental understanding of high temperature superconductivity, it is important to understand the relationship and differences between these pnictides and the even higher temperature superconductors, the cuprates. Both family of materials are layered quasi-two dimensional electronic systems, which at stochiometry develop antiferromagnetic order. Upon electronic doping of these layers, they become superconducting. One of the key differences between the two is that the cuprates, at stoichiometry, are in many respects classic Mott insulators. In local density approximation (LDA) based calculations they are found to be metallic and only strong correlation effects drive them to insulating and antiferromagnetic behavior. In contrast, the parent state of iron pnictide materials is a metal, whose conductivity increases upon the onset of antiferromagnetism. This metallic antiferromagnetic state is also reproduced in the LDA calculations.

Furthermore, the cuprate ions have a single d-hole, whereas the iron ions in the pnictides have multiple d electrons. Thus the former have spin-half, making up a system that has maximum quantum fluctuations and possibly exotic incipient quantum spin-liquid behavior whereas the local moment in the latter could be much larger and thus the magnetism more conventional. Any role of strong correlations in the pnictides has also been questioned, and this would have a strong bearing on any universal understanding of the mechanism of superconductivity in these family of materials.

We focus here on the parent insulating material. Magnetism in these materials has been highly debated. Some argue that these materials are spin-density-waves driven by nesting of fermi-surfaces and they should be viewed as weakly correlated itinerant magnets. Others have argued that despite the metallic behavior, strong correlations are present and the correct starting point for understanding the magnetism in these materials is a Heisenberg type model. Indeed, neutron scattering spectra can we be well understood by the latter approach.

The conventional strong correlation view has been that the magnetism is highly frustrated by having nearest neighbor antiferromagnetic exchange $J_1$ compete with second neighbor antiferromagnetic exchange $J_2$. It is well known that this leads to a $(\pi, 0)$ ordered state at large $J_2$ as observed in experiments. In this context, there is an additional Ising degree of freedom associated with the direction of antiferromagnetic and ferromagnetic neighbors, which leads to a finite temperature phase transition. At, or below this transition, small anisotropies and or weak three-dimensional coupling should lead to antiferromagnetic long-range order. The Ising transition should couple to lattice distortions and lead to a loss of tetragonal symmetry. Indeed, such structural and magnetic transitions are observed either at the same temperature, or with the structural transition being slightly higher in temperature, as expected from theory. The proximity to a quantum critical point near $J_2 = J_1/2$ can lead to a highly reduced moment which fluctuates with small changes in material parameters.

The key to distinguishing different scenarios is Neutron Scattering and the measurement of spin-wave dispersion in the full Brillouin zone. In this respect, the $J_1 - J_2$ model scenario is also of deep theoretical significance from the point of view of quantum magnetism, because the $(\pi, 0)$ order is selected by an order by disorder phenomena. The accidental degeneracy of the classical model leads to zero energy states near 4-points of the Brillouin zone $(0, 0)$, $(\pi, 0)$, $(0, \pi)$, $(\pi, \pi)$. Quantum fluctuations, which lift the accidental degeneracy, cause a gap at $(0, \pi)$ and $(\pi, \pi)$, leaving gapless excitations only at $(0, 0)$ and $(\pi, 0)$ as required by Goldstone’s theorem.

Recently Zhao et al have performed neutron scattering experiments on the material CaFe$_2$As$_2$. The experiments show sharply defined spin waves throughout the zone, not just at very low energies. The absence of a Stoner decay, leads them to strongly favor a local moment type picture for the system. However, a big surprise is the
finding that the spin-wave energy along the $K$ direction is a maximum at $(\pi, \pi)$. We will show below that this requires that the nearest neighbor exchange be strong and antiferromagnetic in one direction and weak and ferromagnetic in the other. Quantum fluctuations can lead to a gap at $(\pi, \pi)$ but they cannot turn it into a maximum. The materials clearly lack tetragonal symmetry and so such a result can be considered as resulting from lack of tetragonal symmetry. However, one needs to understand why a small distortion can lead to such a dramatic difference in exchange constants. Indeed, this is exactly what the electronic structure calculations of Han et al using local spin-density approximation (LSDA) find, within the tetragonal phase. These results have not yet found a simple explanation. The main purpose of this paper is to give a simple scenario by which this happens.

Rather than trying to write down a complete Hamiltonian for the low energy behavior of this system, we proceed in a step by step manner to uncover the physics behind the unusual neutron scattering spectra and the unusual LSDA results. We begin with a two orbital model (which we take as tetragonal) for the low energy behavior of this system. At the very least, this phenomenon of these materials.

Because this model has a local Ising like degree of freedom associated with orbital occupation, it should have a finite temperature phase transition. However, the energetics of the Ising order are mediated by ordering the spins. So, it is not clear that the universality class of the transition must necessarily be that of the 2D Ising model. There could be long-range effective interactions and that may lead to a first order phase transition. In future work, such details can be investigated numerically. Such an orbital order will clearly couple to lattice distortions and a lack of tetragonal symmetry. Note, that unlike the $J_1 - J_2$ model, this Ising parameter order is much less subtle here and the distortions should be more prominent. This is exactly what one finds in the phenomenology of these materials.

The model presented above gives a simple scenario for the observed magnetism and structural transition in the pnictide materials. However, it is certainly not a complete model for the system. At the very least, this phenomena must coexist with other $d$ electrons, which are itinerant and can contribute further to the magnetism. Electronic structure studies with local spin-density approximation (LSDA) provide further clues to the unusual physics. It is found that the local moment and magnetism is highly sensitive to the assumed spin configuration. Not all choice of LSDA patterns lead to well formed moments. The $(\pi, 0)$ pattern leads to particularly robust magnetism, whereas ferromagnetic ordering leads to almost no moment at all.

We can view such a calculation as providing further information on orbital occupations, when a spin-density is imposed on the system. Fixing the $(\pi, 0)$-state in LSDA

$$J_{1a} = J_1 - J_f$$

along $x$ and a ferromagnetic coupling $J_{1b} = - J_f$ along $y$. Thus the system will have Neel order. Linear spin-wave theory should be a good approximation for the spectra of this model. Using Holstein-Primakoff transformations, we obtain the spin-wave dispersion:

$$\omega_k = 4SJ_1 \sqrt{A_k^2 - B_k^2}$$

with

$$A_k = 1 + \alpha - \beta + \beta \cos k_y,$$

and,

$$B_k = \cos(k_x)(\cos(k_y) + \alpha).$$

Here, $\alpha = J_{1a}/(2J_2)$, and $\beta = J_{2a}/(2J_2)$. The condition for $(\pi, \pi)$ point to be a maximum along $k_y$ is that $\alpha > 1$ and $\beta$ be negative and small. A sketch of the resulting spectra is shown in Fig. 1. Parametrically this is the model obtained in the electronic structure calculations of Han et al. And, this is exactly the form of the model that gives an excellent fit to the neutron scattering spectra, where Zhao et al quote the values for CaFe$_2$As$_2$ $(SJ_{1a} = 49.9\pm9.9\text{meV}, SJ_{1b} = -5.7\pm4.5\text{meV}, S J_{2} = 18.9 \pm 3.4\text{meV})$.

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We can view such a calculation as providing further information on orbital occupations, when a spin-density is imposed on the system. Fixing the $(\pi, 0)$-state in LSDA
pattern drives the system to the orbital ordered magnetic state discussed above. In contrast, fixing a ferromagnetic spin configuration, not only need not cause orbital polarization, it fails to set the occupation of the $d_{xz}$ and $d_{yz}$ orbitals at a level needed for moment formation. This suggests that antiferromagnetic spin arrangement on neighboring sites, and the resulting gain in the exchange energy $J_1$ is necessary to stabilize the local moment. One way to view the situation is that electrons in other, perhaps weakly correlated, itinerant orbitals act as a reservoir for these strongly correlated orbitals. The gain in the superexchange energy, when neighboring spins are anti-parallel, alters their chemical potential in a way as to cause them to become Mott insulating, with occupancy close to unity and well formed moments. This is an example of an orbital selective Mott transition, which is driven by superexchange interaction between neighboring iron atoms. It would be interesting to further study this by Dynamical Mean Field Theory (DMFT) and its generalizations.\(^{26}\) The magnetic moment formed in these orbitals, via the local Hund’s coupling, can further polarize other d-electrons leading to an even enhanced moment as found in the electronic structure calculations.

An interesting aspect of the model is that while the system may be highly frustrated above the ordering temperature, the spins become unfrustrated below the ordering temperature. Yet, the ordered moment can be reduced significantly by quantum fluctuations because we are dealing effectively with a quasi-one dimensional spin-1/2 system may be highly frustrated above the ordering temperature, from usual local moment antiferromagnets. Rather than form moments around vacancies, one is more likely to consider a multi-band model with all the low energy orbitals at a level needed for moment formation. This suggests. The occupation of $d_{xz}$ orbitals may exceed the occupation of $d_{yz}$ orbitals by a small amount. Such a small symmetry breaking may be sufficient to cause the exchange interactions to be antiferromagnetic in one direction and ferromagnetic in the other. Nevertheless, our model serves as a good zeroth order starting point for understanding this behavior. It would be useful to look for such occupation numbers in more detail in the electronic structure calculations.

The orbital degrees of freedom should couple strongly to the lattice. Hence, they should have long time scales associated with them and may act like quenched impurities and give rise to glassy behavior. It would be interesting to look for such effects in experiments. Furthermore, the role of vacancies would be very different, in such a system, from usual local moment antiferromagnets. Rather than form moments around vacancies, one is more likely to destroy the Mott behavior altogether.\(^{14}\)

To get a complete picture for this material, one needs to consider a multi-band model with all the low energy electrons. However, this analysis shows that in some respects this system may be much closer to the cuprates than previously thought and hiding inside the multi-electron weakly correlated electronic system maybe a spin-half strongly correlated component which drives the magnetic phenomena. Whether this also plays a role in superconductivity remains to be seen.

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1. Y. Kamihara et al, J. Am. Chem. Soc. 130, 3296 (2008).
2. C. de la Cruz et al, Nature 453, 899 (2008); H. H. Klaus et al, PRL 101, 077005 (2008).
3. J. Dong et al PRL 83, 27006 (2008).
4. Z. P. Yin, et al, PRL 101, 047001 (2008).
5. C. Cao, P. J. Hirschfeld, and H. P. Cheng, PRB 77, 220506 (2008).
6. F. Ma and Z. Y. Lu, PRB 78, 033111 (2008).
7. T. Yildirim, PRL 101, 057010 (2008).
8. J. Wu, P. Phillips, A. H. C. Neto, PRL 101, 126401 (2008).
9. K. Haule, J. H. Sjim and G. Kotliar, PRL 100, 226402 (2008).
10. P. W. Anderson, Science 235, 1196 (1987).
11. Q. Si and E. Abrahams, Phys. Rev. Lett. 101, 076401 (2008).
12. C. Fang et al PRB 78, 052507 (2008).
13. C. Xu, M. Mueller and S. Sachdev, PRB 78, 020501 (2008).
14. I. I. Mazin and M. D. Johannes, Nat. Phys. 5, 141 (2009).
15. S. Raghu et al PRB 77, 220503 (2008).
16. Y. Ran et al, PRB 79, 014505 (2009).
17. P. Chandra, P. Coleman and A. I. Larkin, PRL 64, 88 (1990).
FIG. 1: Linear spin-wave spectra of the model through the Neel point ($\pi, \pi$). Case I refers to an antiferromagnetic coupling along x and a ferromagnetic coupling along y, whereas case II refers to equal antiferromagnetic coupling along both axes. In the latter case, order by disorder phenomena opens a gap at ($q_y = \pi$) but it leaves the point a minima.