Magnetic Excitations in the High $T_c$ Iron Pnictides

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We calculate the expected finite frequency neutron scattering intensity based on the two-sublattice collinear antiferromagnet found by recent neutron scattering experiments as well as by theoretical analysis on the iron oxypnictide LaOFeAs. We consider two types of superexchange couplings between Fe atoms: nearest-neighbor coupling $J_1$ and next-nearest-neighbor coupling $J_2$. We show how to distinguish experimentally between ferromagnetic and antiferromagnetic $J_1$. Whereas magnetic excitations in the cuprates display a so-called resonance peak at $(\pi, \pi)$ (corresponding to a saddlepoint in the magnetic spectrum) which is at a wavevector that is at least close to nesting Fermi-surface-like structures, no such corresponding excitations exist in the iron pnictides. Rather, we find saddlepoints near $(\pi, \pi/2)$ and $(0, \pi/2)$ (and symmetry related points), which are not close to nesting the Fermi surfaces.

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The recent discovery of superconductivity exceeding 50K in a new class of materials holds tremendous potential for understanding the origin of high temperature superconductivity. Similar to the cuprate superconductors, the iron pnictides also have a layered structure, and display magnetism in the undoped parent compound. Both become superconducting upon doping. And like the cuprates, the transition metal layer is believed to play an important role in the superconducting pairing. On the other hand, the parent compound of LaOFeAs is a poor metal at room temperature, as opposed to a correlated insulator as in the cuprates.

Initially band structure calculations suggested the materials are nonmagnetic but close to a strong magnetic instability. However, subsequent calculations have shown that the antiferromagnetic state has lower energy than the nonmagnetic state because of Fermi surface nesting. In Ref. a stripe-like antiferromagnetic ground state was suggested based on strong nesting effects. Recent neutron scattering experiments have shown that the parent compound of LaOFeAs is a long-range ordered antiferromagnet with a type of spin stripe order (i.e. unidirectional spin density wave). However the magnetic moment was found to be $0.36(5)\mu_B$ per iron, which is much smaller than the calculated value of $\sim 2.3\mu_B$ per iron.

From an analysis of the superexchange interactions, Ref. suggested that the next-nearest-neighbor interaction $J_2$ is antiferromagnetic (AFM), while the nearest-neighbor interaction $J_1$ is ferromagnetic (FM). However a first-principles band structure calculation predicts that the nearest-neighbor interaction is also antiferromagnetic. They predict that $|J_2|$ is almost as twice large as $J_1$. In both cases, the competition between $J_1$ and $J_2$ leads to a type of stripe-ordered two-sublattice antiferromagnetic ground state when $|J_2/J_1|$ is larger than the critical value. While the interactions $J_1$ and $J_2$ can compete, the uniaxial SDW considered in Fig. is a classical ground state of the system, and it is thus not frustrated in the sense of having a macroscopic ground state degeneracy.

We use linearized spin wave theory to calculate the magnetic excitations and sublattice magnetization for the two-sublattice collinear antiferromagnet with nearest-neighbor superexchange coupling $J_1$ and antiferromagnetic next-nearest-neighbor superexchange coupling $J_2$. We present results for ferromagnetic coupling $J_1$ as well as for antiferromagnetic coupling $J_1$. (See Fig.) We find the results are quite different for the two cases, so that comparing our calculations with future neutron scattering results at finite frequency will be able to distinguish these two cases.

The model Hamiltonian is described by the Heisenberg spin model on the square lattice

$$H = J_1 \sum_{\langle ij \rangle_{nn}} S_i \cdot S_j + J_2 \sum_{\langle ij \rangle_{nnn}} S_i \cdot S_j$$

where $\langle ij \rangle_{nn}$ and $\langle ij \rangle_{nnn}$ mean the nearest-neighbor and next-nearest-neighbor spin pairs respec-
tively. There are two spins in each unit cell, as shown in Fig. 1. We study the elementary excitations of the classical ground state of this model by using the well-known Holstein-Primakoff boson method. The dispersion and intensities are calculated by quantizing the classical spin waves.

We use Holstein-Primakoff bosons to quantize about the collinear antiferromagnetic ground state found in recent neutron scattering. The associated spin wave velocities are

\[ v_x = 2S\sqrt{-J_1^2 + 4J_2^2}, \]

\[ v_y = 2S|J_1 + J_2|. \]

Notice that \( v_x \) becomes imaginary for \( |J_1| > 2|J_2| \), indicating a change in the classical ground state configuration.

Fig. 2 shows the spin wave band with the nearest neighbor coupling both antiferromagnetic (Fig. 2(a)) and ferromagnetic (Fig. 2(b)). The presence of saddlepoints can be seen, and we will return to this point later. In addition, because the \((\pi, \pi)\) point is a magnetic reciprocal lattice vector, the dispersion must have \( \omega \to 0 \) at this point, although as we will see there is no zero-frequency intensity associated with this part of the dispersion. This precludes finite frequency weight at the \((\pi, \pi)\) point from this band.

We calculate the zero-temperature dynamic structure factor using the same method, \[ S(k, \omega) = \sum_f \sum_{i=x,y,z} | < f | S^i(k) | 0 > |^2 \delta(\omega - \omega_f). \] Here \(| 0 >\) is the magnon vacuum state and \(| f >\) denotes the final state of the spin system with excitation energy \( \omega_f \). \( S^z \) does not change the number of magnons, contributing to the elastic part of the structure factor. \( S^z \) and \( S^y \) both contribute to the inelastic dynamic structure factor through single magnon excitations.

In Figs. 3 and 4, we show the expected neutron scattering intensity for constant energy cuts in \( k \)-space. We
We have integrated over an energy window of \( \pm 0.2 |J_1| S \). show our predictions from spin wave theory for both ferromagnetic and antiferromagnetic \( J_1 \). Fig. 3 shows the expected neutron scattering intensity from a single domain of the magnetic order (\textit{i.e.} for an untwinned case), and Fig. 4 shows the expected scattering intensity for the case where there is an equal contribution from domains with both orientations of the magnetic order (\textit{i.e.} for a twinned case).

For ferromagnetic \( J_1 \), at low frequency, the strongest diffraction peaks are located at \((0, \pi)\). (See Fig. 3.) However more intensity weight shifts to \((\pi, 0)\) when \( J_1 \) is antiferromagnetic. There is also a spin wave cone emerging from \((\pi, \pi)\), but the intensity is much weaker than the cones emanating from other magnetic reciprocal lattice vectors, since zero frequency weight is forbidden at \((\pi, \pi)\) for the magnetic order we consider. At high energy, the difference between ferromagnetic \( J_1 \) and antiferromagnetic \( J_1 \) becomes more apparent. For example, for FM \( J_1 \), there are two strong spots along the \((\pi, k_y)\) direction, whereas for AFM \( J_1 \), they are along the \((0, k_y)\) direction. In real materials, stripe order can be twinned due to, \textit{e.g.}, a finite correlation length, local disorder pinning, or crystal twinning. Therefore we show the twinned constant energy cut plots in Fig. 4 for both FM and AFM nearest neighbor coupling \( J_1 \).

As can be seen from the dispersion in Fig. 2 there are saddlepoints in the spin wave excitation spectrum at various points in \( k \)-space. For the case of both couplings antiferromagnetic, these occur at \((\pi/2, 0)\) and \((\pi, \pi/2)\) and symmetry related points. For ferromagnetic nearest neighbor coupling, saddlepoints can be seen at \((0, \pi/2)\) along with weak saddlepoints possible at \((\pi/2, 0)\) and \((\pi/2, \pi)\) and symmetry related points. The integrated intensity is generally large at such saddlepoints. In the cuprates, there is a saddlepoint in the magnetic excitations at \((\pi, \pi)\) which has been empirically connected to superconductivity, in that it increases in intensity at the onset of superconductivity, \textit{i.e.} the “resonance peak”. There has been much discussion concerning this scattering phenomenon in the cuprates, particularly because it is close to nesting vectors for the corresponding Fermi surface. However, in the case of the iron pnictides, the saddlepoints we find here are quite far from any nesting vectors.

Experimentally, the magnetic moment per iron was found to be 0.36(5)\( \mu_B \), which is much smaller than the expected value of \( \sim 2.3 \mu_B \) per iron site.\cite{10, 11, 12} The zero point energy of the spin waves reduces the sublattice magnetization. It was suggested in Ref. \cite{14} that the competition between \( J_1 \) and \( J_2 \) may be responsible for the small moment observed in experiment. The sublattice magnetization \( m \) is defined as

\[
m = < S_i^z > = S - \Delta m, \tag{13}
\]

where \( \Delta m \) is the deviation of sublattice magnetization from the saturation value,

\[
\Delta m = < a_i^+ a_i > = \sum_k < a_k^+ a_k > = \frac{1}{2V_k} \sum_k \frac{S A_k}{\omega(k)} - 1 + \frac{1}{V_k} \sum_k \frac{S A_k}{\omega(k)} e^{i/\omega(k)} - 1 = \Delta m^{quantum} + \Delta m^{thermal}. \tag{14}
\]

The first term \( \Delta m^{quantum} \) comes from quantum zero point fluctuations. The second term \( \Delta m^{thermal} \) comes from the classical thermal fluctuation, which is divergent.
at any finite temperature in agreement with the Mermin-Wagner theorem. (The very presence of the broken symmetry observed in experiment implies that there is some finite coupling between planes, however weak.)

Here we calculate $\Delta m_{\text{quantum}}$ by

$$
\Delta m_{\text{quantum}} = \frac{1}{2} \int_0^{2\pi} \int_0^{2\pi} \frac{dk_x}{2\pi} \frac{dk_y}{2\pi} \frac{SA_k}{\omega(k)} - \frac{1}{2}. \quad (15)
$$

It is difficult to get the analytical form of the integral. Thus we numerically calculate $\Delta m_{\text{quantum}}$. From the symmetry, the above integral does not change when $J_1$ changes sign. In Fig. 5, $\Delta m_{\text{quantum}}$ is plotted as a function of the superexchange coupling ratio $|J_2/J_1|$. It is $S$ -independent. If $S$ is in between 1 and $\sqrt{2}$, it will reduce the $m$ by 13% - 20%. $\Delta m_{\text{quantum}}$ decreases with increasing $J_2/J_1$ because stronger $J_2$ stabilizes the two-sublattice collinear antiferromagnet state. This deviation is not sufficient to explain the observed value of the sublattice magnetization.

In conclusion, we have used spin wave theory to calculate the magnetic excitations and sublattice magnetization for the two-sublattice collinear antiferromagnetic state of the new $La(O_1-xF_x)FeAs$ high-$T_c$ superconductors. We have studied both ferromagnetic and antiferromagnetic nearest-neighbor coupling $J_1$ with antiferromagnetic next-nearest-neighbor coupling $J_2$. We calculate the predicted inelastic neutron scattering pattern based on spin wave theory. Comparison with future inelastic neutron scattering studies can be used to distinguish the sign of $J_1$. We find that the sublattice magnetization can be reduced by the zero-point motion of spin waves, although not enough to account for the small moments observed in experiment. In addition, we identify several saddlepoints in the magnetic excitation spectrum. While magnetic excitations in these regions are expected to have extra intensity due to the saddlepoint structure, these corresponding wavevectors are not near nesting vectors of the Fermi surface.

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Note added: Some results from spin wave calculations have also been reported by Ref. [21].

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