Magnetism and exchange coupling in iron pnictides

J.J.Pulikkotil\textsuperscript{1}, L. Ke\textsuperscript{2}, M. van Schilfgaarde\textsuperscript{2}, T. Kotani\textsuperscript{2}, and V.P.Antropov\textsuperscript{1}

\textsuperscript{1} Condensed Matter Physics, Ames Laboratory, IA 50011 and
\textsuperscript{2} School of Materials, Arizona State University

(Dated: April 22, 2010)

Abstract

Using linear-response density-functional theory, we obtain the magnetic interactions in the several iron pnictides. The ground state has been found to be non-collinear in FeSe, with a large continuum of nearly degenerate states lying very close to the magnetic “striped” structure. The presence of non-collinearity also seems to be a generic feature of iron pnictides when the Fe moment is small. At small $R_{Fe-Se}$ the system is itinerant: strong frustration give rise to excess of spin entropy, long ranged interactions create incommensurate orderings and strong biquadratic (or ring) couplings violate the applicability of Heisenberg model. There is a smooth transition to more localized behavior as $R_{Fe-Se}$ increases: stable magnetic orbital order develops which favor long range AFM stripe ordering with strongly anisotropic in-plane exchange couplings. The stabilization of the stripe magnetic order is accompanied by the inversion of the exchange coupling.
Accumulated experimental studies indicate that the recently discovered iron oxyarsenide family (Fe-As-La-O) and iron chalcogenide (Fe-Se) superconductors\(^1\)\(^2\) similar itinerant antiferromagnetic metallic systems. This fact distinguishes these systems from traditional high-\(T_c\) superconductors where the local-moment picture and the importance of strongly correlated electrons is generally accepted. While the physics of these systems can be dramatically different they share an important common feature: antiferromagnetic (AFM) interactions on a (nearly) 2D lattice. One can hope that these materials have similar physics and analysis of new metallic systems will shed some light on a nature of magnetic fluctuations in both type of superconductors.

Description of magnetism in itinerant magnets is rather complicated, owing to the lack of a commonly accepted rigid spin picture and the disappearance of adiabaticity. The trivial use of localized models leads to oversimplification of the physics of itinerant magnets. As we show here, the degree of itineracy is critical to understand the ground states and magnetic excitations in iron pnictides. Owing to a strong competition between on-site Hund energy and Fe-As bonding in iron pnictides\(^3\)\(^4\), we adopt the local density approximation (LDA) as we believe it to be the most appropriate readily accessible method to describe these systems\(^6\).

We analyze the magnetic properties of iron pnictides, using a variety of implementations of the LSDA, including FLAPW, FPLMTO, LMTO-ASA. The latter includes calculations of non-collinear configurations following the original prescription\(^8\). Full-potential results are in excellent agreement with each other, and (where they can be compared) very similar to those obtained in Ref.\(^7\). The ASA results are also in reasonably good agreement with them. For the linear response calculations we adopted a Green’s function formalism within the ASA\(^9\)\(^10\). This technique has been extensively tested and applied to many different metallic and insulating systems\(^9\)\(^11\). A similar method was recently applied to LaFeAsO\(^12\). We have taken care to converge the calculations of parameters reported, e.g. using a 16\(\times\)16\(\times\)12 \(k\)-mesh and 24 energy points for the contour integration in the complex plane.

We begin our analysis with the low-temperature phase of the FeSe\(^2\). At low temperature FeSe assumes an orthorhombic structure, space group \(\text{Cm}m\text{ma}\), with distortions of FeSe slabs similar to those observed in FeAs layers in the iron oxyarsenide family. Experimental lattice parameters were employed with Fe at (4b) and As at (4g) Wyckoff positions. In both FeSe and LaFeAsO, there is an internal parameter \(z\) not fixed by symmetry, corresponding to the height of the chalcogen above the (possibly distorted) square of Fe atoms lying in the \(xy\) plane. \(z\) fixes the Fe-Se distance \(R_{\text{Fe-Se}}\), but we treat it here as a parameter because of the ambiguity in determining it even while the magnetic properties depend strongly on \(R_{\text{Fe-Se}}\). The similar dependencies of magnetic moment on volume have been observed in other iron based magnets (see Ref.\(^13\) for the review).

Our initial magnetic structure was obtained by relaxing a 32 Fe atom supercell (with \(z=0.25\)). Spins were initially put in random (noncollinear) orientations and their orientations relaxed along with the spin and charge densities, as described in Ref.\(^14\). In this way several magnetic structures have been identified (Fig.1). The self-consistent (minimum-energy) collinear solution was found to be an AFM configuration in the striped geometry, with a Fe moment \(M \approx 0.85\mu_B\). As Fig. 1 shows, \(M\) is very sensitive to \(R_{\text{Fe-Se}}\). The Néel state is the least stable among the structures indicated in Fig. 1: it is \(-i\cdot0\) mRy/Fe less stable than the striped structure (and nonmagnetic at the LSDA equilibrium geometry). All structures exhibit similar dependence on \(M(R_{\text{Fe-Se}})\). At very small moments \((M<0.2\mu_B)\) orderings \((b),(c),(d)\) in Fig. 1 have lower energy than the striped configuration \((a)\). For \(M>0.9\mu_B\) the dimer structure (Fig.1e) has lowest energy. Such sequence of ground states clearly indicate the significance of couplings beyond first two nearest neighbors.

The following picture emerges from studies of the pairwise exchange coupling parameters \(J_{ij}\) and non-magnetic susceptibility \(\chi_{ij}\) in different structures, as a function of \(R_{\text{Fe-Se}}\). We denote \(\chi_0\) as the on-site susceptibility and \(\chi_1\) and \(\chi_2\) that of the two nearest neighbors (NN). Much of the behavior we observe depends on these three quantities. Define \(\eta = \chi_2/\chi_0\) as the “magnetic frustration” parameter (see below). For small \(M\) (small \(R_{\text{Fe-Se}}\)), \(\chi_0\) is rather small \((-14\text{-}15\text{ Ry}^{-1})\). Noting that the Stoner parameter \(I\) is \(I \approx 0.068\) Ry for Fe, the local moment criterion \(I\chi_0 > 1\) is marginally, or not quite satisfied (see Ref.\(^10\)). Thus the system already at this stage can be classified as a marginally itinerant system: that is the magnetic moment depends on the environment for its stability as opposed to localized behavior, where the local moment is large and weakly dependent on environment. We find both \(\chi_1\) and \(\chi_2\) are large and negative. So, the criterion for AFM pair formation \(I(\chi_0 - \chi_1) > 1\) along \((1,0),(0,1)\) and \((1,1)\) is fulfilled while the criterion of FM pair formation \(I(\chi_0 + \chi_1) < 1\) is not. FM fluctuations are thus strongly suppressed while AFM interactions are frustrated, because both \(\chi_1 < 0\) and \(\chi_2 < 0\), with \(\eta \approx 0.4\). Increasing \(R_{\text{Fe-Se}}\) (or volume) from a small value, a magnetic moment appears as a consequence of the sum of exchange fields from all sites, supporting the itinerant nature of the ground state there. This is characterized by \(\alpha = |\chi(q=0)/\chi_0 - 1|\), which differentiates localized and itinerant systems\(^12\). For small \(M\), \(\alpha \approx 0.3\). As \(R_{\text{Fe-Se}}\) increases to the magnetic instability point \(I\chi = 1\), \(I\chi(q)\) approaches 1 in the small region of wave vectors corresponding to striped and dimer orderings. At low moments AFM states can be created and destroyed by arbitrarily weak interactions.

As \(R_{\text{Fe-Se}}\) increases further, \(M\) increases smoothly and \(\eta\) decreases, reaching \(\eta \approx 0.2\) for large \(M\). Thus the magnetic state evolves to a less itinerant and less frustrated condition and even FM fluctuations can exist. \(\alpha\) decreases smoothly with \(M\), but the magnetic behavior continues to be borderline between itinerant and localized for intermediate \(R_{\text{Fe-Se}}\),
and the frustrated nature of a weakly magnetic state is conserved for all reasonable distances $R_{Fe-Se}$. Only in the strongly magnetic state is the situation drastically changed.

In Table 1 we present both longitudinal and transverse components of the pairwise magnetic “exchange” parameters $J_{ij}$ for the striped structure with $M = 0.9 \mu_B$ (near the theoretically optimized LSDA value). Both components of the exchange matrix $J$ have similar amplitudes, indicating that both longitudinal and transverse spin fluctuations must be taken into account. The NN coupling between atoms with parallel spins (along $x$, $\tau = 1/2,0,0$) is negative (“frustrated”) while the exchange between pairs with antiparallel spins (along $y$, $\tau = (0,1,2,0)$) is positive (also for $\tau = (1/2,1/2,0)$). For more distant (1-5 to 1-9) in-plane neighbors all parameters have similar amplitudes with different signs. While two of the NN parameters indicate a stable pair configuration, Fig. 2 and the Table show the presence of rather unstable FM pair alignments in plane, and weak interaction along the $z$-axis (see $J_{001}$ in the Table 1), making this system very nearly a 2D AFM. However, even such weak interplanar coupling may lead to 3D long-range order and finite temperature transition. The insert of Fig. 3 shows how the Fe moment in the striped structure depends on anisotropy of the exchange coupling of NN atoms. The origin of this anisotropy is as follows. For the Néel structure both $J_{1x} = J_{100}$ and $J_{1y} = J_{010}$ are positive and equal (reflecting symmetry) while $J_2 = J_{110}$ is negative, reflecting instability of FM alignment along (1,1). For the striped structure at small $R_{Fe-Se}$ (“bad” nesting conditions) the system still reflects the frustrated $J_1$-$J_2$-$J_3$ model-like behavior and our $J_{1x}$ and $J_{1y}$ have opposite signs with similar amplitudes (Fig. 2). As $M$ increases $J_1$ becomes anisotropic, reflecting a broken symmetry induced by magnetic analog of Jahn-Teller effect\[\text{1}\]. For $M \sim 1.5 \mu_B$ the anisotropy is very large and soon ’frustrated’ exchange coupling along FM line disappears and the effective sign of this interaction becomes positive. Thus, large moment state does correspond to localized Heisenberg model and is absolutely stable with no sign of antiferromagnetic frustrations. At high temperature both $x$ and $y$ directions are equivalent, but as $T$ decreases the barrier between $x$ and $y$ orientations will prevent fluctuations between them and the spin symmetry-breaking phase— with attendant structural distortions—will be frozen in. Fig. (1c) depicts the ”rotator” structure — a state intermediate between $x$- and $y$- aligned striped phases. At $M \sim 1 \mu_B$ the LSDA energy of this state is 0.8 mRy/Fe relative to the striped state.

The spin wave velocities are not formed merely by NN interactions in such a system, and the contribution from distant Fe atoms (3-4 Å) with antiparallel spins is significant, as expected. We found significant in-plane and out-of-plane anisotropies of spin velocities. For instance $v_x/v_y = 1.6$ for $M = 1.07 \mu_B$. This anisotropy has not been determined experimentally. The calculated energy gap in the spin wave spectrum due to spin orbital coupling\[\text{1}\] is $\Delta = 0.15$ mRy. At larger $q$ we see deviations from the adiabatic magnon picture. However the adiabaticity parameter\[\text{18}\] for $M \sim 1 \mu_B$ is $\sim 0.2$; so we believe the system is already less itinerant and more adiabatic, therefore antiferromagnons can be defined not only at small $q$. However longitudinal fluctuations of similar strength are always present and should be seen in experiment. These features of spin excitation spectra will be reflected in the temperature dependence of the sublattice magnetization and will modify the enhancement of the linear specific heat. Also at those $R_{Fe-Se}$ where the spin fluctuations are very localized around a particular $Q$ one may expect weak temperature dependence of the susceptibility.

Because parameters $J_{ij}$ strongly depend on magnetic configuration (which may not be a ground state) and on symmetry-breaking, it is crucial to analyze the magnetic interactions for several magnetic orderings. The universal picture that emerges is that first and second NN interactions (both transverse and longitudinal) on the square Fe sublattice are AFM and very strong, with their absolute values and the ratio $\eta$ depending on $R_{Fe-Se}$. The role of longitudinal fluctuations is crucial at larger $M$. Even so the relatively small more distant neighbor couplings (and biquadratic terms) are not negligible and important for the appearance of non collinear fluctuations at small moments as we now describe.

To check the stability of our collinear ”ground” striped state and avoid uncertainties connected with the long-wave approximation\[\text{18}\], we performed spin dynamics simulations with small deviations between FM coupled atoms keeping first or second NN ordering antiferromagnetic. These excitations include $j_1$ and $j_2$ modes (Fig.1) and fictitious spin spiral (SS) orderings\[\text{8}\]. Our results demonstrate that for those $R_{Fe-Se}$ with $M < 1 \mu_B$ (Fig.3) spirals ($Q + (q,0,0)$) which rotate AFM planes against each other exist only in some limited spin phase space around $Q$ of the striped structure, clearly establishing the existence of the magnetic short range order (MSRO). This MSRO is significant and can exist far above the critical temperature. However, the phase volume of such long-wave excitations is rather small. As $R_{Fe-Se}$ increases the short ranged excitations with much larger volume of phase space become available and it leads to stabilization of the different magnetic ground state or changes the character of excitations.

The total energy of SS configurations which rotate FM spins along the stripes, i.e. ($Q + (q,0,0)$) appears very different. Fig.3 shows the energy is nearly independent of $q$ for a significant region of $q$. Such a flat surface in the itinerant AFM usually is a precursor of frustrated interactions. This is exactly the present case, as evident from the negative value of $J_{1x}$ (Fig. 2). Moreover SS calculations show that the collinear striped structure ($q = 0$), which has strong AFM coupling between stripes and no staggered magnetization at finite $q$, is not the minimum-energy state, although the stabilization of this particular SS is very small ($\sim 0.01$ mRy) with SS $q$ consistent with the amplitude of $J_3, J_4$ couplings. We also found that $j_1$ fluctuating mode (which has staggered magnetization) has very
low excitation energy, so states in this fluctuating direction are practically degenerate and only starting at 15-20 mRy do we see a small energy increase ~ 0.03 mRy. At larger angles the energy rapidly increases, so the “zig-zag” (45°) state has energy ~ 0.3 mRy/Fe. The nematic j2 mode has much higher energy at large moments (which quite natural for the biquadratic or ring type of exchange coupling), while at small moments it has lower energy than the j1 mode, supporting the increase of frustration parameter η. Both “zig-zag” and ”rotator” structures are indicators of the strength of biquadratic exchange (K) and its importance for the phase formation and spin wave spectrum gap. It is unusual to have biquadratic exchange as large as half of bilinear exchange parameter for the system with relatively small values of effective spin value (1/2 or 1). However, we obtained a similar behaviour in many other pnictides and believe that this is a generic feature of these systems. This fact, in turn, may create favorable conditions for the applicability of model, suggested in Ref.[15]. The biggest difference with that model consists in the origin of barrier energy. In our calculations this barrier exists already on adiabatic energy surface as a result of complicated spin-spin interactions in the ground state, providing even stronger support of conclusions of Ref.[15]. In addition, the low energy of j1 and corresponding ’nematic’ type of spin ordering (j2) can be responsible for the disappearance of LRO in plane due to developing at finite temperatures fluctuations between x and y stripes. This mechanism of magnetic transition can be considered as an alternative to usual formation of two-dimensional state which is widely used in high temperature superconductors study. Besides being important for the magnetic transition, these fluctuations must appear at the finite temperatures, may persist in the paramagnetic case and should be observable in the experiment. These type of excitations are very unusual: they do not exist at low temperatures in the ordered state, but will appear closer to the Neel temperature or above it.

Thus, our exchange coupling and SS calculations indicate the presence of two main competing mechanisms: one is an instability (driving force) towards formation of AFM order (“good” nesting conditions), and second frustration related to the strong negative couplings of first and second NN on a square lattice (with η ~ 0.2 – 0.4). While in the J1-J2 model[15] η is the only frustration parameter, our calculations reflect a more complex picture: the relative strength of all frustrations (as any interatomic coupling) is also determined by the “itineracy” characterized by α.

The latter mechanism gives rise to large spin fluctuations related to J3 or J4 pair interactions and is particularly important at small M (itinerant limit) with strong MSRO and limited available phase space with long-wavelength excitations (analogous non-collinearity we also found in CaFe2As2[10]). These fluctuations can even destroy the long range AFM order at lower RFexSe (or lower volume or c/a ratio). For larger RFeexSe improving nesting conditions and a naturally developing unstable longitudinal mode enhance the first effect: a stable, ordered AFM state forms with larger more localized M. The itineracy parameter α coincides with the “adiabaticity” parameter of Ref.[18] and indicates that strongly frustrated metallic systems simultaneously have strong non-adiabatic effects. Thus, our calculations shown that transverse frustrations are rapidly decreasing as the system becomes more localized.

To illustrate our result for FeSe we map the results obtained above onto Heisenberg J1-J2-J3 model[15]. Fig.4 clearly shows how relatively weak exchange coupling between third NN transforms the collinear ground state. Especially important are the third and forth NN coupling when J1=2J2, when the non-collinear state of different symmetry can be stabilized by a very small coupling beyond second NN. In strongly magnetic case (M=1μB) when NN coupling appear to be very anisotropic the ratio between main parameters of spin Hamiltonian are following (122 family): J1 ≈ 30 – 40 meV, J1y ≈ 0.2J1y, J2 ≈ 0.7J1y, J3 ≈ 0.15J1y, K1 ≈ 0.2 – 0.3 J1, and magnetic anisotropy ~0.1 meV.

Despite theory predicting very similar properties for many families of iron pnictides, experimental evidence for long range magnetic order in the normal state have not been found in all compounds. In particular, no long range order of any kind been found in LiFeAs or FeSe, nor have local moments been found so far. In Tab.2 we summarize the calculated exchange coupling obtained for the different iron pnictide systems, including some with no experimentally established LRO. All these systems clearly demonstrate a stable AFM stripe order at large moments. The exchange coupling shows rather itinerant behavior for smaller moments (large range of parameters), and more localized for the larger moment case. Due to large anisotropy between first NN exchange parameters the striped structure is very stable and the isotropic J1-J2 Heisenberg model is totally not applicable for iron pnictide systems at larger magnetic moments where this anisotropy is very large. However, this model might be relevant at smaller moments when J1 is close to J2. The similarity in the exchange coupling and the comparably stability of the stripe order clearly contradicts the available experimental data for these systems. Assuming that more experiments with better single crystals confirm that LRO in LiFeAs and FeSe is absent, the fact that theory contradicts these observations must be related to LSDA shortcomings. In particular, magnetic zero-point fluctuations (absent in LSDA) may well be strong enough to suppress the moment. Alternatively, there may be problems even in the LDA description of bonding in iron pnictides at the independent-particle level.

Finally, we calculate the imaginary part of the bare dynamical transverse spin susceptibility χ0+(q, ω) , which provides insight into Stoner excitations[19]. Calculations were performed for the Neé structure at z=0.25, which corresponds to a magnetic moment M=1.2μB. Along (110), χ0+(q, ω) is particularly interesting; see Fig.5a. Stoner excitations appear even for ω>0 for small q. As q increases they disappear completely; see q = (0.25, 0.25). Finally, they reappear at still higher q.
Most of these novel features can be explained in terms of the unique nature of the Fermi surface. The Fermi surfaces of FeSe (and the Fe pnictides) are approximately cylindrical, and are centered either at Γ or at the (π, π) point in the xy plane, as shown in Fig.3 of Ref.[7].

Cylinders at Γ are of hole character; those at the zone corner are of electron character. q is the connecting vector that links an electron at k + q to a hole at k. \( \chi^{0+-}(q, \omega) \) is obtained from a sum over all allowed k (i.e. any k for which an electron state at k + q and a hole state at k are separated by an energy difference \( \omega \)). We have identified the following facts, which can explain the behavior in Fig.5a.

(a) For \( q < (0.25, 0.25, 0) \), k points only couple to points k + q in the same pocket. 
(b) For \( q > (0.25, 0.25, 0) \), k points only couple to points k + q in other pocket. 
(c) For \( q = (0.25, 0.25, 0) \), no two k points can be connected. 
(d) For \( q > (0.375, 0.375, 0) \), there is a strong inter-pocket excitation.

Along (100) the story is different. Since the two pockets are separated by (1/2, 1/2, 0), they cannot communicate for \( q=(q00) \) (at least at low energy). So only intra-pocket excitations, Γ−Γ, M−M, are allowed. This picture is borne out in Im\( \chi^{0+-}(q, \omega) \), shown in Fig.5b. Im\( \chi^{0+-}(q, \omega) \) is already present for small \( \omega \), e.g. 50 meV, when q is small; but almost disappears when q reaches 0.25, and completely disappears for larger q.

While we have not yet attempted to calculate spin fluctuations, the fact that large Stoner excitations are present in a significant fraction of the Brillouin zone offer strong indications that low energy spin-flip excitations are important. They can strongly affect the single-particle description of the magnetism: in particular they reduce the magnetic moment M. These fluctuations may explain the discrepancy between observed magnetic order, and that predicted by the LDA in case of FeSe.

In conclusion, two competing mechanisms in the newly discovered superconductors have been identified: strong geometric frustrations dominant for small moments, which allow distant interactions lead to a symmetry breaking, forming a continuum of non-collinear states. As M increases, magnetic Jahn-Teller effect[10] creates better nesting conditions with stabilization of an AFM striped configuration. We have found third and fourth neighbor couplings \( J_3 \) and \( J_4 \) to be the driving force for non-collinear spin fluctuations. We believe that is the generic feature of iron pnictides when the Fe moment is small. The calculated exchange coupling and the corresponding spin wave excitations along z-direction are relatively small, can be easily destroyed by temperature and two-dimensional character of magnetism is likely to be realized at higher temperatures or paramagnetic state. We also find that strong frustration — a condition necessary for excess entropy in order to admit a superconducting state — are favored in the itinerant magnets when conditions for adiabaticity and long wavelength approximation are not well satisfied. Here longitudinal fluctuations make an additional contribution to the spin entropy. As moment increases, the in-plane exchange coupling along ‘ferromagnetic’ line changes its sign, so the stripe order becomes absolutely stable for such ‘no frustration’ localized magnetic moment case. We identified those magnetic excitations in pnictides that may exist below Neel temperature and contribute to observable properties. Together with traditional spin waves, in this system coexist Stoner type of excitations (electron-hole pairs) and longitudinal spin fluctuations, especially at small moment. The presence of low energy Stoner excitations ultimately describes this system as itinerant magnetic system. In addition to such traditional excitations, this system has very peculiar four fold symmetry planar spin fluctuations with the energy of 15 meV and below (modes (b)-(d) on Fig.1) with some of them being similar to ‘nematic’ spin order. Our results indicate that the localized spin description is somewhat academic for this system if magnetic moment does not exceed 1\( \mu_B \). Overall, the magnetic transformation in iron pnictides can be described as a transition from frustrated itinerant moment phase to symmetry broken orbitally ordered localized magnetic moment phase with “striped” order. The transition happened around M=1\( \mu_B \).

Since this paper has been submitted new neutron experimental data have been published for FeTe_{1-x}Se_{x}[20]. The authors observed the presence of two-dimensional incommensurate excitations which may reflect the non-collinear ordering identified above.

Work at the Ames Laboratory was supported by Department of Energy-Basic Energy Sciences, under Contract No. DE-AC02-07CH11358. ASU was supported by DOE contract DE-FG02-06ER46302 and ONR grant N00014-07-1-0479, and is grateful to the Fulton High-Performance Computing Center for use of their facilities.

[1] F.-C. Hsu, J.-Y. Luo, K.-W. Yeh, T.-K. Chen, T.-W. Huang, P. M. Wu, Y.-C. Lee, Y.-L. Huang, Y.-Y. Chu, D.-C. Yan and M.-K. Wu, Proc. Natl. Acad. Sci. U.S.A. 105, 14262 (2008), Y. Mizuguchi, F. Tomioka, S. Tsuda, T. Yamaguchi and Y. Takano, Appl. Phys. Lett. 93, 152505 (2008)
[2] S. Margadonna, Y. Takabayashi, M. T. McDonald, K. Kasperkiewicz, Y. Mizuguchi, Y. Takano, A. N. Fitch, E. Suard, and K. Prassides, Chem. Commun. (Cambridge) 5607 (2008); S. Margadonna, Y. Takabayashi, Y. Ohishi, Y. Mizuguchi, Y. Takano, T. Kugayama, T. Nakagawa, M. Takata, and K. Prassides, Phys. Rev. B 80, 064506 (2009)
[3] S. Mirbt, B. Sanyal, C. Isheden, and B. Johansson, Phys. Rev. B 67, 155421 (2003)
[4] J. Wu, P. Phillips and A. H. Castro Neto. Phys. Rev. Lett. 101, 126401 (2008)
\[ J_{100} \quad 2 \quad (\uparrow) \quad 0.497524 \quad 0.5, 0, 0 \quad -0.508 \quad -0.644 \\
J_{010} \quad 2 \quad (\downarrow) \quad 0.50 \quad 0, 0, 5, 0 \quad 0.773 \quad 0.828 \\
J_{110} \quad 4 \quad (\uparrow) \quad 0.70558 \quad 0.5, 0, 5, 0 \quad 0.145 \quad 0.299 \\
J_{000} \quad 2 \quad (\uparrow) \quad 0.995 \quad 1, 0, 0 \quad 0.045 \quad -0.045 \\
J_{020} \quad 2 \quad (\uparrow) \quad 1.0 \quad 0, 1, 0 \quad 0.046 \quad -0.043 \\
J_{001} \quad 2 \quad (\uparrow) \quad 1.028 \quad 0, 0, 1 \quad 0.001 \quad -0.009 \\
J_{120} \quad 4 \quad (\uparrow) \quad 1.1169 \quad 0.5, 1, 0 \quad 0.060 \quad -0.099 \\
J_{121} \quad 4 \quad (\uparrow) \quad 1.1136 \quad 1, 0, 5, 0 \quad -0.053 \quad -0.013 \\
\]

Table 1: The first eight Fe-Fe transverse \( J_T \) and longitudinal \( J_L \) exchange parameters in FeSe. Column \( n \) denotes the number of equivalent nearest neighbors, together with the spin orientation. \( R \) denotes the distance to the central atom and \( \overrightarrow{p} \) the connecting vector in units of the lattice parameter \( a \). \( J_T \) is related to the Heisenberg model \( J \) as \( J_S S_x = J_T \) and can be defined as a second derivative of the LSDA total energy with respect to moment variation.

\[ J_{100} \quad 1.61 \quad -1.5 \quad -16.1 \quad -12.1 \quad -2.0 \quad -5.0 \quad -3.3 \quad -0.3 \quad -0.8 \quad -0.2 \quad -0.4 \quad 91.5 \\
J_{010} \quad 1.78 \quad -8.9 \quad -41.8 \quad -7.8 \quad -0.5 \quad -0.5 \quad 3.7 \quad -0.2 \quad 0.0 \quad 0.0 \quad 102.6 \\
J_{110} \quad 1.28 \quad -11.2 \quad -35.5 \quad -7.6 \quad -3.2 \quad -3.1 \quad 3.6 \quad -0.5 \quad -0.8 \quad -0.3 \quad -0.5 \quad 91.2 \\
J_{000} \quad 1.07 \quad -45.3 \quad -70.9 \quad -13.5 \quad 0.8 \quad -0.1 \quad 4.5 \quad -2.2 \quad -0.3 \quad -0.2 \quad -0.1 \quad 35.9 \\
\]

Table 2: The Fe-Fe Heisenberg model parameters \( J \) in different families of iron pnictides (in meV). These parameters have been obtained from theoretically calculated \( J_T \) by normalizing it by scalar product \( S_x S_y \) where \( S_x = M_y/2 \).

Fig.1 The dependence of magnetic moment on Fe site as a function of Fe-Se distance (\( R_{Fe-Se} \)) for the stripped (lowest energy) and simple Néel (highest energy) magnetic structures. Also shown are schematics of the magnetic structures considered in the calculations: (a) striped structure (b) "zig-zag" structure. A transition from a striped to "zig-zag" structure (a 'nematic' type of state) leaves AFM ordering between first NN atoms unchanged (\( J_{11} \) mode). (c) "rotator" structure (or so called 2K structure). Here AFM ordering between second NN is unchanged (\( J_{22} \) mode) (d) mixed structure with AFM moments along diagonal and non magnetic NN atoms (e) dimer structure (f) simple Néel structure.

Fig.2 Magnetic interaction parameters in FeSe for the stripped structure, as a function of Fe magnetic moment \( M \) (controlled by varying \( R_{Fe-Se} \)). \( J_1 = J_{100} \) and \( J_2 = J_{010} \) are the NN interaction along \( x \) and \( y \) respectively; \( J_3 = J_{110} \) is the second-NN interaction in the plane (vectors are shown in the Table.) Interactions between planes are small (~6% of \( J_3 \)). Insert shows the anisotropy of the NN exchange coupling as a function of Fe moment. The inversion of the sign of the exchange coupling parameter \( J_{11} \) occurs at \( M \approx 1.88 \mu_B \).

Fig.3 The total energy of a spin spiral as a function of spiral wave vector \( q \) near the AFM striped structure. Top: SS for \( q = (0, q, 0)2\pi/a \). Near \( q = 0.25 \) (not shown), the moment disappears entirely. Bottom: SS along the stripe, for
\( \mathbf{q} = (q, 0) 2\pi / a \). The minimum energy occurs at \( q \approx 0.02 \), corresponding to a slow spiral along “frustrated” direction \( x \).

Fig. 4 The phase diagram of \( J_1-J_2-J_3 \) model and calculated non-magnetic susceptibilities ratio for FeSe (central red star). Other stars correspond to 2% and 4% of hole (right) and electron (left) doping of FeSe. In all cases the finite value of \( J_3 \) coupling leads to the stabilization of non-collinear order of different symmetry. The relative strength of \( J_3 \) is increased as moment decreases and the system becomes more itinerant.

Fig. 5 Bare transverse spin susceptibility \( \chi^{0+} (\mathbf{q}, \omega) \) as a function of \( \omega \) for different \( q \) points along (100) and (110).
The diagram shows the magnetic moment on Fe ($\mu_B$) as a function of $R_{Fe-Se}$ (Å). The data points are represented by black squares (stripes) and red circles (Neel). The magnetic moment increases as $R_{Fe-Se}$ increases. The inset illustrates different magnetic states:

- **a** and **d**: Upward and downward magnetic moments, respectively.
- **b** and **e**: Rightward and leftward magnetic moments, respectively.
- **c** and **f**: Circular magnetic moments, respectively.

The graph highlights the transition between the stripe state and the Neel state.
Exchange parameter (mRy)

Magnetic moment on Fe atom (µB)

|J²|/|J¹x| |J¹y|

M(µB) ∆E(mRy)

SS(x) SS(y)

|qₓ| |qᵧ|
\( \text{Im} \left\{ \chi_0^\pm(q, \omega) \right\} \) (1/Ryd)

(q,0,0) direction
$\text{Im} \left[ \chi_0^+(q, \omega) \right] (1/\text{Ryd})$

$(q, q, 0)$ direction