The double life of electrons in magnetic iron pnictides as revealed by NMR

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Abstract – We present a phenomenological, two-fluid approach to understanding the magnetic excitations in Fe pnictides, in which a paramagnetic fluid with gapless, incoherent particle-hole excitations coexists with an antiferromagnetic fluid with gapped, coherent spin wave excitations. We show that this two-fluid phenomenology provides an excellent quantitative description of NMR data for magnetic “122” pnictides, and argue that it finds a natural justification in LSDA and spin density wave calculations. We further use this phenomenology to estimate the maximum renormalisation of the ordered moment that can follow from low-energy spin fluctuations in Fe pnictides. We find that this is too small to account for the discrepancy between \textit{ab initio} calculations and neutron scattering measurements.

Introduction. – The discovery that, suitably doped, Fe pnictides can superconduct at temperatures greater than 50K \cite{1} has sparked a sudden rush of interest in these materials. As with the high-$T_c$ cuprates, the undoped parent compounds are magnetic. Neutron scattering and $\mu$SR experiments suggest a direct competition between the two states, with the magnetism winning at low doping and the superconductivity taking over as the doping is increased \cite{2–4}. Understanding the magnetic excitations in these materials is therefore widely believed to be an important step towards understanding their superconductivity, as well as an interesting problem in its own right.

To date, most theoretical approaches to this problem have stressed either the itinerant nature of electrons in Fe pnictides \cite{5–7}, or used strong electronic correlation to justify mapping them onto a frustrated local-moment model \cite{8–10}. In this paper we embrace the fact that Fe pnictides are both metals and magnets, proposing a simple, phenomenological, two-fluid description of their magnetic excitations. We argue that spin excitations at low energies and temperatures are dominated by the gapless, incoherent particle-hole excitations, characteristic of a metallic paramagnet, while for energies and temperatures comparable with a spin gap $\Delta_\sigma$, coherent, collective excitations of the magnetic order come into play. It follows naturally from experimental and theoretical determinations of the band structure that these two fluids are essentially independent.

While this two-fluid phenomenology is not tied to any particular microscopic model, it finds a natural justification in recent spin density wave (SDW) calculations \cite{11–17}, ARPES experiments \cite{7,18} and LSDA calculations \cite{7}. In this paper we further show that our phenomenology provides an excellent description of NMR experiments on Fe pnictides with 122 structure \cite{19,20}.

We go on to address a second major issue in the pnictide materials, namely the role of frustration. It has been suggested that the large discrepancy in the size of the ordered moment between \textit{ab initio} calculations and neutron scattering measurements can be understood by fine tuning a frustrated local-moment model \cite{8–10}. We critically re-examine such a model in terms of our two-fluid phenomenology and conclude that, while it does not rule out frustration \textit{per se}, quantum fluctuations cannot account for the observed reduction of the ordered moment relative to LDA calculations \cite{5–7}.

Both the magnetic and metallic properties of Fe pnictides originate in outer-shell Fe 3$d$-electrons. Band structure calculations \cite{5,6,21}, supported by photo-emission \cite{22,23} and quantum oscillation \cite{24,25} experiments, suggest that these hybridize with As 4$p$-orbitals.
to form a Fermi surface with two electron-like and three hole-like pockets, when viewed in a “natural” unfolded Brillouin zone based on Fe sites. SDW calculations [11–17], ARPES experiments [7,18] and LSDA calculations [7] find general agreement on a number of points. There are observed to be five bands crossing the Fermi surface in the paramagnetic state. These undergo a non-trivial reconstruction at the magnetic ordering transition, with some of the bands mixing to form a gapped SDW state and the rest remaining metallic. Furthermore, there is no pair of metallic bands in the ordered state that is nested with spanning vector (π, 0). For example, the calculations presented in [15] consider four of the five bands, two hole-like and two electron-like, and show that (π, 0) order arises most naturally when only one of the hole bands takes part in the Fermi surface mixing. Therefore the magnetically ordered state retains a hole-like sheet of Fermi surface, which will support metallic, particle-hole excitations. Similarly, by comparing ARPES experiments and LSDA calculations, ref. [7] shows that below Ts,DW the Fermi surface reconstructs to form (π, 0) SDW order, but ungapped, metallic Fermi surface pockets remain, centred on the Γ-point.

The fact that magnetic Fe pnictides are metals implies that some part of this complex Fermi surface remains gapless, and will support incoherent particle-hole excitations with vanishing energy. We treat this as the first of our fluids, characterised simply by an averaged density of states at the Fermi energy, n0, which can be estimated from heat capacity measurements.

Neutron scattering experiments [26–29], meanwhile, reveal a commensurate, collinear, antiferromagnetic (AF) ground state with ordering vector k* = (π, 0, π), and ordered Fe moment mS ≈ 1 μB, much smaller than predicted by ab initio calculations [5,6]. A single branch of spin wave excitations with dispersion,\[ \omega k' = \sqrt{\Delta^2 + v_{k''}^2 k_x'^{2} + v_{k''}^2 k_y'^{2} + v_{k''}^2 k_z'^{2}} \] (1)
is found above a gap Δσ ≈ 10 meV at the ordering vector k' = k − k* = (0, 0, 0). Spin wave velocities v = (vx, vy, vz) are anisotropic, with vx ≈ vy ≫ vz. The collective excitations of this magnetic order form our second fluid, and, following [30], we characterise them using a quantum nonlinear sigma model,

\[ S = \frac{1}{2abc} \int \text{d}x \text{d}t \left[ \hbar^2 \chi_\perp \partial_t n^2 - \rho_x \left( \partial_x n \right)^2 - \rho_y \left( \partial_y n \right)^2 - \rho_z \left( \partial_z n \right)^2 + \chi_\perp \Delta^2 n^2 \right], \] (2)

where \( \chi_\perp \) is the static perpendicular susceptibility, \( \rho_x, \rho_y, \rho_z \) are spin stiffnesses along the Fe-Fe crystal axes a, b, c, and \( \Delta^2 \) is an easy-axis anisotropy.

For \( \Delta \sigma \to 0 \), this action describes the long-wavelength Goldstone modes, which follow from the symmetry of the magnetic order. As such, it can be derived from any microscopic model that respects these symmetries, whether localised or itinerant. For finite anisotropy \( \Delta \sigma > 0 \), eq. (2) predicts a gapped, twofold degenerate cone of spin wave excitations with exactly the form of eq. (1), where \( v_0 = \sqrt{\rho_0 / \chi_\perp} \).

For collinear order it is natural to consider a Z2 symmetry [31,32] between (π, 0) and (0, π) states, which is not encoded in the non-linear sigma model. However, in the pnictides, this symmetry is broken by a tetragonal-to-orthorhombic phase transition that typically occurs at or very close to the magnetic ordering temperature [33]. Below this temperature the simpler non-linear sigma model description is sufficient, provided there are well-defined cones of spin wave excitations.

Within a spin density wave picture, eq. (2) should remain valid up to an energy scale of the spin density wave gap, estimated to be \( \Delta_{\text{SDW}} \approx 31 \text{ meV} \) for LaFeAsO [16]. For the specific case of CaFe2As2, it breaks down at energies of approximately 150 meV, where the spin wave branch is seen to enter a continuum of excitations [27].

Our final approximation is to ignore all coupling between these two fluids. This appears justified for two reasons. Firstly neutron scattering studies [27–29] observe sharp cones of low-energy magnetic excitations with no evidence of the damping that would be expected if the spin waves could scatter from the metallic fluid. Secondly LSDA calculations [7], photoemission studies [7] and SDW theory [11–17] show no evidence, in the magnetic ordered state, for a nested pair of Fermi surfaces with spanning vector (π, 0). Hence, within a band picture, there are no available particle-hole states close to the ordering vector for the spin waves to decay into.

We note that there is evidence [7,13] for a node in the SDW gap. However, this is not situated at the ordering vector, is not nested with any other Fermi surface with spanning vector (π, 0) and has vanishing density of states. Thus it does not appear relevant to our low-temperature model.

Sublattice magnetisation. - A simple test of our phenomenology is provided by the temperature dependence of the ordered moment \( \delta m_S(T) \). This renormalises the zero-temperature moment according to \( m_S = m_0 - \delta m_S - \delta \tilde{m}_S(T) \), where \( m_0 \) is the bare moment and \( \delta \tilde{m}_S \) describes the reduction due to quantum fluctuations. Within the two-fluid picture, \( \delta \tilde{m}_S(T) \) is controlled by the thermal excitation of spin waves, as described by eq. (2). For \( T < \Delta \sigma \) we find activated behaviour,

\[ \delta \tilde{m}_S(T) = \frac{m_0 (abc) \sqrt{\Delta \sigma}}{8 \chi_\perp v_s^2} \left( \frac{2 k_B T}{\pi} \right)^{1/2} e^{-\frac{\Delta \sigma}{k_B T}}, \] (3)

while for \( T \gg \Delta \sigma \) we find the power law behaviour,

\[ \delta \tilde{m}_S(T) = \frac{m_0 (abc)}{12 \chi_\perp v_s^2} (k_B T)^2 = M_{HT} T^2. \] (4)

The form of corrections depends only on the gap, \( \Delta \sigma \). The prefactor is determined by the geometric mean spin...
wave velocities, \( \bar{v}_s = v_x v_y v_z \) [(meV \AA\(^{-3}\)]

Fig. 1: (Colour on-line) Temperature dependence of the ordered moment \( \delta m_{S}(T) \) as determined by NMR measurements on BaFe\(_2\)As\(_2\) [19] (blue circles) and SrFe\(_2\)As\(_2\) [20] (black squares). Data is plotted as \( \ln [\delta m_{S}(T) / T^2] \) vs. \( \Delta_{c} / k_{B}T \), where the values of \( \Delta_{S} = 114 \) K and \( \Delta_{S} = 75 \) K are taken from inelastic neutron scattering experiments [26,29]. Straight lines show the expected form of corrections at low temperatures. The intercept gives the prefactors \( M_{LT}^{BA} \approx 1.6 \times 10^{-4} \mu_{B} K^{-\frac{3}{2}} \) and \( M_{LT}^{SR} \approx 5 \times 10^{-5} \mu_{B} K^{-\frac{3}{2}} \).

Spin-lattice relaxation rate. – NMR experiments also probe spin excitations through the nuclear spin-lattice relaxation rate, \( 1/T_{1} \). This has been measured for As nuclei in BaFe\(_2\)As\(_2\) [19] and SrFe\(_2\)As\(_2\) [20]. For hyperfine interactions, the relaxation rate is given by [36,37],

\[
\frac{1}{T_{1}} \approx \frac{\gamma_{N}}{2} k_{B} T \lim_{\omega_{0} \to 0} \frac{1}{N} \sum_{\mathbf{q}} |A_{\mathbf{q}}|^2 \frac{\chi''(\omega_{0}, \mathbf{q})}{\hbar \omega_{0}},
\]

where \( \gamma_{N} \) is the gyromagnetic ratio of the nucleus in question, \( \omega_{0} \) is the nuclear excitation energy, \( |A_{\mathbf{q}}|^2 \) is a form factor describing the coupling between the surrounding electrons and the nuclear spin and \( \chi''(\omega_{0}, \mathbf{q}) \) is the imaginary part of the longitudinal, dynamic susceptibility of the electron system. Both fluids contribute to \( 1/T_{1} \), but at low temperatures the leading contribution will come from gapless particle-hole pairs within the paramagnetic fluid. We assume a roughly constant contact interaction between the nucleus and the metallic electrons, \( |A_{\mathbf{q}}|^2 \approx |A_{0}|^2 [(T/\mu_{B})^2] \), over the relevant sheets of the Fermi surface. This leads to a contribution to \( 1/T_{1} \) which is linear in \( T \) [37],

\[
1/T_{1}^{inc} \approx \frac{1}{4} \pi \hbar \gamma_{N} |A_{0}|^2 g_0^2 \langle ab \rangle^2 n_0^2 k_{B} T = C_{inc} T,
\]

where \( g_0 \) is the Landé g-factor, \( n_0 \) is the density of states at the Fermi surface and we use units where \( \mu_{B} = 1 \).

At higher temperatures, the Raman scattering of thermally excited spin waves also plays a role in nuclear spin relaxation. This is dominant over single spin wave excitations since \( \Delta_{c} \gg \hbar \omega_{0} \). NMR probes the longitudinal susceptibility \( \chi''(\omega_{0}, \mathbf{q}) \), which can be calculated directly from eq. (2). We consider relaxation due to coupling of the As nucleus to a four-site plaquette of nearest-neighbour Fe sites, shown in fig. 2, in the same spirit as for the Y nucleus in YBa\(_2\)Cu\(_3\)O\(_{6+x}\) in [38].

In both cases, the antiferromagnetically ordered electron moments create an internal field at the nuclear site, and this is that dictates the behaviour of the form factor, \( |A_{\mathbf{q}}|^2 \). In the case of YBa\(_2\)Cu\(_3\)O\(_{6+x}\), the combined symmetry of the crystal structure and magnetic order causes the internal field at the Y site to disappear, and spin fluctuations are filtered by a form factor which vanishes at the magnetic ordering vector [38]. In contrast, the As site in a pnictide such as BaAs\(_2\)Fe\(_2\) experiences a finite internal field directed along the c-axis [19], and for NMR fields applied in the ab-plane, longitudinal fluctuations of the ordered Fe moment couple efficiently to the nuclear spin.

In this case, the appropriate form factor is

\[
|A_{\mathbf{q}}|^2 = 4B_{ac}^2 (1 - \cos q_x + \cos q_y - \cos q_x \cos q_y),
\]

where \( B_{ac} \) is the matrix element relevant for Raman relaxation processes. The form factor and the imaginary part of the longitudinal susceptibility are simultaneously peaked at the ordering vector, \( \mathbf{q} = (\pi, 0, \pi) \). The form factor is very slowly varying in comparison with the susceptibility, and hence we approximate it with the constant \( |A_{\mathbf{q}}|^2 \approx 16B_{ac}^2 \).

The more complex case of external magnetic field parallel to the c-axis will be discussed elsewhere [39].
Making these approximations, we find,

\[
\frac{1}{T_{\text{coh}}} \approx \frac{2B^2m_0^2\hbar(abc)^2}{\pi^3\chi_\perp^2 \xi^6} \Delta_S^2 \Phi \left( \frac{k_BT}{\Delta_\sigma} \right) 
\]

\[
\approx C_{\text{coh}} \Phi \left( \frac{k_BT}{\Delta_\sigma} \right),
\]

where,

\[
\Phi(x) = x^2 \text{Li}_1(e^{-1/x}) + x^3 \text{Li}_2(e^{-1/x}),
\]

and \(\text{Li}_n(z) = \sum_{k=0}^\infty \frac{z^k}{k^n}\) is the \(n\)-th polylogarithm of \(z\).

**Comparison to experiment.** – We are now in a position to compare directly with experiment, and, in fig. 3, we show the results of simultaneous fits to NMR data for \(\delta m_S(T)\) and \(1/T_1\) with field in the \((1,1,0)\) direction in BaFe\(_2\)As\(_2\) \cite{29} and SrFe\(_2\)As\(_2\) \cite{20}. We treat the total relaxation rate as the sum of the contributions of the two fluids, eq. (6) and eq. (8) and fit the prefactors \(M_{\text{LT}}\) (eq. (3)), \(C_{\text{inc}}\) (eq. (6)), and \(C_{\text{coh}}\) (eq. (8)), using the experimental value \(\Delta_{\text{Ba}} = 114 K\) \cite{29} for the gap. The agreement for these two parameter fits is excellent.

It is possible to make independent, quantitative estimates of these fit parameters by substituting known values of the spin wave velocities, hyperfine interactions, density of states at the Fermi surface, Fe-Fe lattice parameters, perpendicular susceptibility and zero-temperature sublattice magnetisation directly into eq. (6), eq. (8) and eq. (3). In table 1 we show that, within experimental error, this quantitative approach to the prefactors is consistent with the fits to NMR data for BaFe\(_2\)As\(_2\). Uncertainty in the model parameters for SrFe\(_2\)As\(_2\) are currently too great for a quantitative comparison.

**The ordered moment at zero temperature.** – One of the important issues in Fe pnictide magnetism has been the size of the ordered moment \(m_S\). Fe and its oxides typically show a large ordered moment at low temperatures. First-principles calculations for magnetic Fe pnictides suggest that \(m_S \approx 1.5–1.7 \mu_B\) \cite{5,6}. The moment measured by neutron scattering, in contrast, ranges from 0.25\(\mu_B\) (NdFeAsO) \cite{43} to 1\(\mu_B\) (SrFe\(_2\)As\(_2\)) \cite{44}. The AF “stripe”

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**Table 1**: Quantitative analysis of \(\delta m_S(T)\) and \(1/T_1\) in BaFe\(_2\)As\(_2\). We determine the prefactors \(M_{\text{LT}}\) (eq. (3)), \(C_{\text{inc}}\) (eq. (6)), and \(C_{\text{coh}}\) (eq. (8)) by fitting NMR experiments (cf. figs. 1, 3) and compare these with the quantitative estimates which follow from known values of the hyperfine interactions \(A_0 \approx 1.88 T/\mu_B\) and \(B_{\perp} = 0.43 T/\mu_B\) \cite{19}, spin wave velocities \(95 < \bar{v}_s < 228 \text{meV} \AA\) \cite{29}, spin gap \(\Delta_\sigma = 9.8(4) \text{meV}\) \cite{29}, ordered moment \(n_0 = 0.87 \mu_B\) \cite{40}, perpendicular susceptibility \(\chi_\perp = 10^{-4} \text{emn/mol}\) \cite{41}, lattice constants \([a,b,c] = [2.80, 2.79, 6.47] \AA\) \cite{40}, and density of states \(n_0 = 5.8 \times 10^{24} \text{J}^{-1} \text{m}^{-3}\) \cite{42}.

| Parameter          | Value           |
|--------------------|-----------------|
| \(M_{\text{LT}}\) | \(1.6 \times 10^{-4}\) |
| \(C_{\text{inc}}\) | 0.032           |
| \(C_{\text{coh}}\) | 7.5             |
| Estimate           | \((0.15–2.2) \times 10^{-4}\) | ~0.015 | 0.14–33 |

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Fig. 3: (Colour on-line) Simultaneous fits to nuclear relaxation rate \(T_1^{-1}\) and sublattice magnetisation \(m_S(T)\) for BaFe\(_2\)As\(_2\) \cite{19} (blue dots) and SrFe\(_2\)As\(_2\) \cite{20} (black squares). The external field is applied in the \((1,1,0)\) direction. The dashed lines show the contribution of incoherent particle-hole pairs of eq. (6); the full lines show the combined fit including the contribution of coherent, thermally activated spin waves of eq. (8). The gap values \(\Delta_{\text{Ba}} = 114 K\) and \(\Delta_{\sigma} = 75 K\) are taken from inelastic neutron scattering experiments \cite{26,29}. The inset shows simultaneous fits to the sublattice magnetisation, \(m_S\). The prefactors are determined to be, \(C_{\text{inc}}^{\text{Ba}} \approx 0.032 \text{s}^{-1}K^{-1}\), \(C_{\text{coh}}^{\text{Ba}} \approx 7.5 \text{s}^{-1}\), \(C_{\text{inc}}^{\text{Sr}} \approx 0.062 \text{s}^{-1}K^{-1}\) and \(C_{\text{coh}}^{\text{Sr}} \approx 2.28 \text{s}^{-1}\).
order found in Fe pnictides has also been observed in quasi–two-dimensional insulating oxides with frustrated exchange interactions, where the ordered moment is strongly renormalised by quantum fluctuations [45]. By analogy, it has been suggested that magnetic excitations in Fe pnictides can also be understood in terms of a frustrated local-moment model,

\[ \mathcal{H} = J_{1x} \sum_{\langle ij \rangle_{1x}} S_i \cdot S_j + J_{1y} \sum_{\langle ij \rangle_{1y}} S_i \cdot S_j + J_{1z} \sum_{\langle ij \rangle_{1z}} S_i \cdot S_j + J_{2} \sum_{\langle ij \rangle_{2}} S_i \cdot S_j - K_{xy} \sum_i (\langle S_i^x \rangle^2 - \langle S_i^y \rangle^2) + K_z \sum_i \langle S_i^z \rangle^2, \]

where \( \langle ij \rangle_{1a} \) counts first-neighbor bonds in the \( \alpha \)-direction, \( \langle ij \rangle_{2} \) second-neighbor bonds in the \( xy \)-plane, and \( K_{xy} \) is a single-ion anisotropy. It is interesting, therefore, to ask what constraints our two-liquid phenomenology places on this effective local-moment picture?

A telling, and direct, comparison can be made in the context of the ordered moment. At a mean field level, the collinear “stripe” phase of the square-lattice \( J_1-J_2 \) Heisenberg model,

\[ \mathcal{H} = J_1 \sum_{\langle ij \rangle_{1}} S_i \cdot S_j + J_2 \sum_{\langle ij \rangle_{2}} S_i \cdot S_j, \]

becomes unstable for \( J_2 < |J_1|/2 \) [46], or equivalently \( v_y < 0 \). Approaching this transition, quantum corrections to the ordered moment diverge, as illustrated in fig. 4, and the sublattice magnetisation becomes zero before reaching the classical transition point. For AF \( J_1 \), the dominant correction to \( m_S \) comes from spin waves near the ordering vector. These are described by eq. (2) with \( v_z = \Delta_\sigma = 0 \), and we find,

\[ \delta m_S = \frac{m_0}{2\chi_\perp} \frac{a^2}{(2\pi)^2} \int_{|k|<\Lambda} \frac{dk}{\omega_k} = \frac{m_0 a^2 \Lambda}{4\pi^2 \chi_\perp} K_1(\kappa), \]

where \( \Lambda \) is a momentum cut-off reflecting the size of the spin wave “cone”, \( K_1 \) is a complete elliptic integral of the first kind, and \( \kappa = \sqrt{1 - (v_x/v_y)^2} \). At the limit of the \( (\pi,0) \) AF phase, \( v_y \to 0 \), and \( \delta m_S \) diverges logarithmically [47].

The contribution of spin waves at higher energies must be determined separately, but for present purposes can be approximated by a constant offset \( \pm 0.1 \mu_B \).

At first sight, fine tuning a \( J_1-J_2 \) model into a region with \( v_y < v_x \) offers the possibility of achieving any desired renormalisation of the ordered moment, \( m_S \), cf. [8–10]. The same would hold in any itinerant-electron model which could be mapped onto eq. (2). However, neutron scattering results for Fe pnictides suggest that \( v_y \approx v_x \) [28]. Moreover, they clearly show a spin gap \( \Delta_\sigma \), and out-of-plane dispersion \( v_z \), both of which act to cut-off the divergence in \( \delta m_S \).

For a gapped, three-dimensional dispersion with \( v_y > \sqrt{v_x v_z (\Lambda/\pi)^2} \), eq. (2) predicts

\[ \delta m_S \approx \frac{m_0 a^2 \Delta_\sigma}{8\pi^2 \chi_\perp v_x^3} \left( \frac{\Lambda v_x}{\Delta_\sigma} \right)^2 \left( 1 + \frac{\Lambda^2 v_x^2}{\Delta_\sigma^2} \right) - \Delta_\sigma \arcsinh \left( \frac{\Lambda v_x}{\Delta_\sigma} \right), \]

where an energy cut-off \( \epsilon = \Lambda v_x \) has been imposed. For the purpose of comparison with experiment, the cut-off \( \Lambda \) can be determined by the extent of the cone of linearly dispersing spin wave excitations seen in neutron scattering experiments. For parameters relevant to BaFe\(_2\)As\(_2\), where \( \Lambda \approx 0.2\pi/\alpha \) [29], eq. (13) implies \( \delta m_S \approx 0.1 \mu_B \), a value too small to explain the gulf between \textit{ab initio} calculations and experiment, although comparable with the smaller discrepancy with model-based SDW theory [12].

In the highly frustrated region \( v_y \to 0 \), the approximation made in eq. (13) begins to break down, since the ellipsoidal integration region becomes longer and thinner, eventually escaping from the Brillouin zone. Nevertheless eq. (13) does provide a finite bound,

\[ \delta m_S < m_0 \Lambda^3 abc/(16\pi^2 \chi_\perp \Delta_\sigma), \]

on the maximum correction to the ordered moment from low-energy spin waves in a gapped, three-dimensional model\(^{1}\). To illustrate how this works, in fig. 4 we compare the predictions of two non-linear sigma model, eq. (2),

\(^{1}\)Cylindrical choices of integration region provide better approximations for highly frustrated parameters.
and the Heisenberg model, eq. (10), for the sublattice magnetisation, $m_S$, as a function of $J_2$ —and thereby $v_y$. Remaining parameters for eq. (10) are taken from experiments on CaFe$_2$As$_2$ [28]. Following LDA calculation [5], we set the bare moment $m_0 = 1.51\mu_B$. A constant offset $\delta m_S = -0.3\mu_B$ is added to eq. (13) to correct for high-energy spin waves, and the value of $\Lambda$ is chosen so that the non-linear sigma model predictions agree with the predictions of the Heisenberg model at large $J_2/J_1$ (equivalently, large $v_y$). The agreement between these two approaches is excellent for a wide range of $J_2$. Even at the maximally frustrated point, the correction $\delta m_S \approx 0.5\mu_B$ is smaller than the $\delta m_S \approx 0.7\mu_B$ needed to explain the discrepancy with experiment. We anticipate that this conclusion will hold for any spin model with realistic parameters [48], and conclude that the failure of LDA to accurately describe the size of the ordered moment lies in high-energy electronic correlation effects, not the zero point motion of low-energy spin waves.

**Conclusion.** — In conclusion, magnetic excitations in Fe pnictides are well described by a simple two-fluid phenomenology in which gapped, three-dimensional spin waves co-exist with gapless, but incoherent particle-hole pairs. These two fluids can be treated as independent. This is evidenced by quantitative fits to NMR. Our phenomenology is blind as to microscopic details of the real materials, but seems to fit naturally with spin density wave calculations which assign magnetism and metallicity to different, weakly coupled, portions of the Fermi surface.

Furthermore it follows from explicit calculation that collective low-energy spin fluctuations, of the type found in highly frustrated two-dimensional quantum magnets, cannot be invoked to explain the discrepancy in the LSDA and neutron scattering values for the sublattice magnetisation.

**REFERENCES**

[1] Kamihara Y. et al., J. Phys. Soc. Jpn., 130 (2008) 3296.
[2] Lester C. et al., Phys. Rev. B, 79 (2009) 144523.
[3] Uemura Y., Nat. Mater., 8 (2010) 253.
[4] For an alternative view, see Wilson J., J. Phys.: Condens. Matter, 22 (2010) 203201.
[5] Han M. J. et al., Phys. Rev. Lett., 102 (2009) 107003.
[6] Yaresko A. N. et al., Phys. Rev. B, 79 (2009) 144421.
[7] Yi M. et al., Phys. Rev. B, 80 (2009) 174510.
[8] St Q., et al., Phys. Rev. Lett., 101 (2008) 076401.
[9] Yao D.-X. et al., Phys. Rev. B, 78 (2008) 052507.
[10] Uhrig G. S. et al., Phys. Rev. B, 79 (2009) 092416.
[11] Korshunov M. M. and Eremin I., Phys. Rev. B, 78 (2008) 140509(R).
[12] Klauss H.-H. et al., Phys. Rev. Lett., 101 (2008) 077005.
[13] Ying Ran et al., Phys. Rev. B, 79 (2009) 014505.
[14] Kaneshita E. et al., Phys. Rev. Lett., 103 (2009) 247202.
[15] Eremin I. and Chubukov A. V., Phys. Rev. B, 81 (2010) 024511.
[16] Knolle J. et al., Phys. Rev. B, 81 (2010) 140506.
[17] Kaneshita E. et al., Phys. Rev. B, 82 (2010) 094441.
[18] Hsieh D. et al., arXiv:0812.229v1.
[19] Kitagawa K. et al., J. Phys. Soc. Jpn., 77 (2008) 114709.
[20] Kitagawa K. et al., J. Phys. Soc. Jpn., 78 (2009) 063706.
[21] Singh D. J. and Du M.-H., Phys. Rev. Lett., 100 (2008) 237003.
[22]Evtushinsky D. V. et al., Phys. Rev. B, 79 (2009) 054517.
[23] Liu C. et al., Phys. Rev. Lett., 101 (2008) 177005.
[24] Coldea A. I. et al., Phys. Rev. Lett., 101 (2008) 216402.
[25] Analytis J. et al., Phys. Rev. Lett., 103 (2009) 076401.
[26] Zhao J. et al., Phys. Rev. Lett., 101 (2008) 167203.
[27] Zhao J. et al., Nat. Phys., 5 (2009) 555.
[28] Diallo S. O. et al., Phys. Rev. Lett., 102 (2009) 187206.
[29] Matan K. et al., Phys. Rev. B, 79 (2009) 054526.
[30] Ong A. et al., Phys. Rev. B, 80 (2009) 014514.
[31] Chandra P. et al., Phys. Rev. Lett., 64 (1990) 88.
[32] Lante V. and Parola A., Phys. Rev. B, 73 (2006) 094427.
[33] Jesche A. et al., Phys. Rev. B, 78 (2008) 180504(R).
[34] Carlo J. P. et al., Phys. Rev. Lett., 102 (2009) 087001.
[35] Uemura Y. J., Physica B, 404 (2009) 3195.
[36] Moriya T., Prog. Theor. Phys., 16 (1956) 641.
[37] Moriya T., J. Phys. Soc. Jpn., 18 (1963) 4.
[38] Mila F. and Rice T. M., Phys. Rev. B, 40 (1989) 11382.
[39] Smerald A. and Shannon N., in preparation.
[40] Huang Q. et al., Phys. Rev. Lett., 101 (2008) 257003.
[41] Ning F. et al., J. Phys. Soc. Jpn., 78 (2009) 013711.
[42] Rotter M. et al., Phys. Rev. B, 78 (2008) 020503.
[43] Yaresko A. N. et al., Phys. Rev. B, 79 (2009) 212502.
[44] Chen Y. et al., Phys. Rev. B, 78 (2008) 064515.
[45] Skoulatos M. et al., EPL, 88 (2009) 57005.
[46] Shannon N. et al., Eur. Phys. J. B, 38 (2004) 599.
[47] Chandra P. et al., Phys. Rev. B, 38 (1988) 9335(R).
[48] Schmidt B. et al., Phys. Rev. B, 81 (2010) 165101.