Universal linear-temperature dependence of static magnetic susceptibility in iron pnictides

G. M. Zhang\textsuperscript{1(a)}, Y. H. Su\textsuperscript{2}, Z. Y. Lu\textsuperscript{3}, Z. Y. Weng\textsuperscript{4}, D. H. Lee\textsuperscript{5} and T. Xiang\textsuperscript{6,7}

1 Department of Physics, Tsinghua University - Beijing 100084, China
2 Department of Physics, Yantai University - Yantai 264005, China
3 Department of Physics, Renmin University of China - Beijing 100872, China
4 Center for Advanced Study, Tsinghua University - Beijing 100084, China
5 Department of Physics, University of California at Berkeley - Berkeley, CA 94720, USA
6 Institute of Physics, Chinese Academy of Sciences - Beijing 100190, China
7 Institute of Theoretical Physics, Chinese Academy of Sciences - Beijing 100190, China

received 11 November 2008; accepted in final form 17 April 2009
published online 18 May 2009

PACS 74.25.Ha – Magnetic properties
PACS 71.27.+a – Strongly correlated electron systems; heavy fermions
PACS 75.30.Fv – Spin-density waves

Abstract – A universal linear-temperature dependence of the uniform magnetic susceptibility has been observed in the non-magnetic normal state of iron pnictides. This non-Pauli and non-Curie-Weiss-like paramagnetic behavior cannot be understood within a simple mean-field picture. We argue that it results from the existence of a wide antiferromagnetic fluctuation window in which the local spin-density-wave correlations exist but the global directional order has not been established yet.

Copyright © EPLA, 2009

The recent discovery \cite{1} of superconductivity in LaFe\textsubscript{As}O\textsubscript{1-x}F\textsubscript{x} has generated strong interest in the investigation of iron-based pnictide materials. There are mainly two types of materials synthesized: the rare-earth pnictide oxide layered systems, ReFeAsO, denoted as “1111” and the so-called “122” systems, MFe\textsubscript{2}As\textsubscript{2} with M = Ca, Ba, Sr, etc. Both the 1111 and 122 parent compounds are metals and have shown a spin-density-wave (SDW) ordering at $T \sim 130$ K, accompanying a tetragonal-orthorhombic structure phase transition \cite{2}. The fact that the parent compounds of the iron pnictides are antiferromagnetic (AF) has attracted a lot of attention, because of the close analogy with the cuprates. Indeed, in the most interesting scenario, this suggests that the AF correlation is intimately connected to the high $T_c$ in both materials. Therefore, a deeper understanding of the AF correlation in the iron pnictides is of particular importance. The purpose of this paper is to take a first step in this direction.

In order to establish a microscopic theory for these materials, two different scenarios, starting from either the weak or the strong coupling limit, have been proposed. The first scenario invokes an itinerant electron approach in which the commensurate SDW ordering as well as the structural transition is believed to be solely induced by the Fermi surface nesting \cite{3,4}. In contrast, the second scenario emphasizes an As-bridged superexchange antiferromagnetic interaction between the nearest- and next-nearest-neighbor local moments of iron, which serve as the basic driving force for both transitions \cite{5–7} without the critical involvement of the Fermi surface nesting. To distinguish the above two scenarios, understanding of the origin of the SDW ordering is the key.

Like any ordering phenomena, one can use an order parameter $\vec{n}$ to describe the SDW order of iron pnictides. In the simplest mean-field picture, $\vec{n}$ is independent of space and time. Above $T_{\text{SDW}}$, $|\vec{n}| = 0$ and there is no trace of magnetism whatsoever. At $T_{\text{SDW}}$ two processes occur simultaneously: a finite $|\vec{n}|$ develops and the directional long-range order establishes. In a more realistic picture $\vec{n}$ is space (and time) dependent. Above $T_{\text{SDW}}$ even though locally $|\vec{n}| > 0$, due to the lack of directional order, global antiferromagnetism is absent. In the latter picture the SDW transition is controlled by the onset of directional long-range order. In the following we shall refer to this as “SDW moment fluctuation scenario”.

When applying the mean-field picture to the iron pnictides, one expects normal metallic behavior with no
trace of antiferromagnetic correlation above $T_{\text{SDW}}$. As a result the uniform magnetic susceptibility, $\chi_u$, should be Pauli paramagnetic like. However, the experimental data of $\chi_u$ for both the 1111 and 122 compounds are shown in fig. 1 as a function of temperature [8–12]. Interestingly, they exhibit a universal linear-temperature dependence in both the undoped and the F-doped LaFeAsO$_{1-x}$F$_x$ [11] compounds. Clearly, this is inconsistent with the mean-field approach expectation.

In this paper, we will argue that this linear-$T$ uniform susceptibility is a strong evidence for the existence of a wide antiferromagnetic fluctuation window of local magnetic moments. It is important to emphasize, however, that the metallic behavior of these compounds makes the present local magnetic moments not quantized as those local atomic moments as in a Mott insulator. Interestingly, in undoped or highly underdoped cuprates La$_{2-x}$Sr$_x$CuO$_4$, $\chi_u$ increases linearly with temperature, just like that in iron pnictides, before reaching a broad peak at a temperature $T_{\text{max}}$ [13]. Moreover, the experimental curves can be scaled onto a universal curve independent of doping. This universal curve agrees with the theoretical result [14–16] obtained for the two-dimensional Heisenberg model with the nearest-neighbor AF coupling.

It is important to note that there are other metallic SDW systems which also show the linear-$T$ susceptibility above $T_{\text{SDW}}$. The best example is chromium and some of its alloys [17]. In the case of pure Cr, diffusive commensurate AF magnetic scattering peak had been observed up to temperatures $T > 2T_{\text{SDW}}$, from which a very small effective magnetic moment ($\mu = 0.16–0.28\mu_B$) can be exacted [18]. This suggests that the local AF SDW correlations extend to rather high temperatures. Another metallic AF system that shows the above linear-$T$ susceptibility above $T_N$ is Na$_{0.5}$CoO$_2$ which is a poor metal with a Néel transition at 86 K [19]. Thus the linear-$T$ susceptibility clearly cannot be used as an evidence for quantized atomic moments as in Mott insulators.

Put it simply, such a phenomenon just implies a non-mean-field transition into the SDW ordered state. The temperature range showing linear-$T$ susceptibility is the fluctuation window in the Ginzburg sense. A more appropriate way of thinking is through Ginzburg-Landau-Wilson theory which captures the fluctuation of the SDW order parameter. To mimic such a theory one can write down an effective lattice model of fixed magnitude spin moments and perform statistical mechanics on it. If one takes a classical antiferromagnetic Heisenberg model on a non-frustrated two-dimensional lattice, it can be shown that the above linear-$T$ susceptibility exists in the temperature range $0 < T < T_{\text{MF}}$, with $k_B T_{\text{MF}}$ of order the nearest-neighbor exchange constant [20].

In the present paper, we prefer to start from a quantum spin model and perform finite temperature statistical mechanics. For the iron pnictides, the following two-dimensional frustrated antiferromagnetic $J_1$-$J_2$ Heisenberg model is assumed

$$H = J_1 \sum_{\langle i,j \rangle} S_i \cdot S_j + J_2 \sum_{\langle\langle i,j \rangle\rangle} S_i \cdot S_j,$$

(1)

where $\langle i, j \rangle$ and $\langle\langle i, j \rangle\rangle$ denote the summations over the nearest and next-nearest neighbors, respectively. With $J_2 > J_1/2$, this model captures the $\langle \pi, 0 \rangle$ and $\langle 0, \pi \rangle$ collinear ordering tendencies of the iron pnictides. Here we assume that at $T = 0$ K the spins are ferromagnetic ordering along the $x$-direction and antiferromagnetic ordering along the $y$-direction. So the lattice is bipartite and divided into $A$ and $B$ sublattices. On the $A$ ($B$) sublattice, the vacuum state is the $S^z = S (-S)$ state. There are two spins in each unit cell.

We then use the antiferromagnetic Dyson-Maleev transformation to represent the spin operators:

$$S_i^- = a_i^\dagger, \quad S_i^z = (2S - a_i^\dagger a_i) a_i,$$

$$S_i^\dagger = S - a_i^\dagger a_i, \quad l \in A,$$

$$S_m^z = b_m, \quad S_m^+ = b_m^\dagger (2S - b_m^\dagger b_m),$$

$$S_m^- = -S + b_m^\dagger b_m, \quad m \in B,$$

(2)
where $a_k^\dagger$, $a_k$, $b_m^\dagger$, and $b_m$ are bosonic operators. Different from the variational approach used by Takahashi [21], we approximate the model Hamiltonian by keeping the quadratic interactions of the boson operators only. Then the model Hamiltonian is hermitian, and it can be expressed after Fourier transform as [22]

$$H \approx \sum_k \left[ \eta_k \left( a_k^\dagger a_k + b_k^\dagger b_k \right) + \Lambda_k \left( a_k b_{-k} + a_{-k}^\dagger b_{-k}^\dagger \right) \right] - 2N S (J_2 S + \lambda),$$

where $\eta_k = 2 J_1 S \cos k_x + 2 J_2 S + \lambda$, $\Lambda_k = 2 J_1 S \cos k_x^2 + 2 J_2 S \gamma_k$, $\gamma_k = \cos k_x \cos k_y$, and a chemical potential term $\lambda$ has been introduced to make the local magnetization vanish at finite temperatures. By using the Bogoliubov transformation, we can diagonalize the Hamiltonian as

$$H = \sum_k \epsilon_k \left( a_k^\dagger a_k + b_k^\dagger b_k \right) + \epsilon_g N,$$

where $\epsilon_k = \sqrt{\eta_k^2 - \Lambda_k^2}$ and $\epsilon_g$ is the ground state energy per site. From the free energy, the chemical potential $\lambda$ is determined by the following equation:

$$\frac{1}{N} \sum_k \frac{\eta_k}{\epsilon_k} \coth \left( \frac{\epsilon_k}{2 k_B T} \right) = 2S + 1.$$  (5)

Moreover, when a magnetic field is applied, the above treatment with a Zeeman term can still be carried out, and the static uniform magnetic susceptibility is derived as

$$\chi_u = \frac{(g u B)^2}{4k_B T N} \sum_k \frac{1}{\sinh^2 \left( \frac{\epsilon_k}{2 k_B T} \right)}.$$  (6)

However, compared to the static uniform susceptibility expression obtained from the spin correlation function, a factor of $\frac{1}{2}$ has to be multiplied [22].

Numerical calculations for the static uniform magnetic susceptibility $\chi_u$ can be performed at finite temperatures. Surprisingly, it has been found that $\chi_u$ behaves as linearly temperature dependent before it reaches a broad peak, then it changes to the Curie-Weiss behavior [22]. The broad peak just corresponds to the mean-field crossover connecting two different behaviors of $\chi_u$. Moreover, as the coupling ratio of $J_2/J_1$ is increased, the window of the linear magnetic susceptibility becomes wider and wider. In other words, the broad peak position is shifted as increasing the coupling ratio of $J_2/J_1$. Of course, such a treatment is just a qualitative description of the non-magnetic state of this frustrated Heisenberg model. In fig. 2, we present the numerical results of $\chi_u$ at finite temperatures for $S = 1$ and $J_2/J_1 = 1.0$, 1.5. For the case of $J_2/J_1 = 1.0$, the uniform susceptibility in the temperature range between 0 and 0.9$J_1$, $\chi_u$ can be fit as

$$\chi_u = \chi_0 \left[ 1 + c \left( \frac{T}{J_1} \right) \right], \quad c > 0.$$  (7)

Quantitatively by taking $J_1 \sim J_2 = 55$ meV estimated by the local density approximation (LDA) calculation [5], we find $\chi_0 \sim 3 \times 10^{-4}$ emu/mol, which is very close to the experimental values extrapolated from the linear-$T$ regime in fig. 1.

It is noted that a wide range of temperatures showing linear-$T$ susceptibility can be attributed to the Mermin-Wagner theorem, which says that a two-dimensional Heisenberg system cannot order at non-zero temperatures. As a result, all temperatures below the mean-field crossover are in the fluctuation regime. The Curie-Weiss-like behavior will eventually recover at higher temperatures beyond $T_{\text{max}} \sim J_2/k_B$ roughly, where the correlation length is less than a lattice constant and the moments become effectively free, similar to the cuprates [13]. Of course the true iron pnictide systems show a finite temperature SDW ordering transition. This is due to the inter-layer coupling $J_z$. In this case, we expect the fluctuation window to lie between the mean-field crossover and the temperature $T_{\text{SDW}}$. In addition, due to the presence of competing interactions ($J_1$ and $J_2$), it is argued that above the SDW ordering transition, there should be an Ising-like transition where the symmetry between the $(\pi, 0)$ and $(0, \pi)$ SDW patterns is broken. Such a transition necessarily breaks the lattice rotation symmetry, and as a result can trigger the tetragonal-orthorhombic distortion [23,24]. The persistence of the linear-$T$ susceptibility into the doped regime implies that the SDW correlation is strong in the superconducting samples. This can be used as indirect evidence for the involvement of antiferromagnetic correlation in Cooper pairing.

We cannot overemphasize that the above quantum model is merely used to mimic the Ginzburg-Landau-Wilson description of the SDW moment fluctuation scenario. It should not be used to imply that we believe that the quantized $S = 1$ atomic moment exists in the system.

Apparently, there is a coupling term between the SDW moments and conduction electrons near the Fermi surface. For example, the SDW transition induces abrupt changes

37006-p3
of the Drude weight [25], magneto-resistance [26], and Hall coefficient [26]. In addition, angle-resolved photoemission experiment has shown a change of the electronic structure near the Fermi energy at $T_{\text{SDW}}$ [27]. These experiments suggest that as the SDW moment orders, magnetic scatterings further gap out parts of the Fermi surface and as a result some itinerant carriers are lost.

Let us now switch to the electron origin of the SDW order. A popular point on this issue says that the SDW moments form because of the Fermi surface nesting effect [3,4]. First of all, the Fermi surface nesting is an “instability” concept. To be precise, in the presence of Fermi surface nesting even infinitely weak SDW channel quasiparticle scattering can open the SDW gap. For a strong scattering, however, nesting is not required. According to the band calculations, the Fermi surface is not well nested by the magnetic ordering wave vectors $(\pi, 0)$ or $(0, \pi)$. In addition, the long-ranged ordering moment, which is a lower bound of the preformed SDW moment, is about $0.365\mu_B$ for LaFeAsO [2], and $0.873\mu_B$ for BaFe$_2$As$_2$ [28]. These moments are rather big since they are comparable with the $T = 0$ ordering moment $0.6\mu_B$ of the spin-$1/2$ Heisenberg model on the square lattice.

Such a large magnetization moment also rules out the SDW transition being the mean-field SDW moment formation temperature. If it were the case, one expects only the electronic states at the energy $k_BT_{\text{SDW}}$ away from the Fermi energy would be perturbed. Given $T_{\text{SDW}}$~130 K and the band structure results, we estimate an upper bound of the ordering moment to be $\sim 0.02\mu_B$, which is more than one order of magnitude less than the measured value.

In our opinion, the SDW fluctuation moment is more likely due to the strong short-range repulsion between the electrons. For example, refs. [5,6] emphasize an As-bridged antiferromagnetic interaction between the nearest- and next-nearest-neighbor iron electrons, which serves as the basic driving force for SDW moment formation without the critical involvement of the Fermi surface nesting. In addition, ref. [29] takes the un-nested LDA band structure adding moderate strong Hubbard-like and Hund-like interactions, and obtains a good fraction of $\mu_B$ for the SDW ordering moment in a mean-field theory [29]. Finally the LDA-based SDW mean-field calculations have yielded the ordering moment between 2.2 and 2.6$\mu_B$ [5]. However it is typical that all such mean-field calculations overestimate the ordering moment since it does not capture the long-wavelength directional fluctuations.

The experimental evidences as well as the theoretical considerations all lead us to conclude that the SDW moment formation temperature for the iron pnictide materials should occur at much higher temperature than $T_{\text{SDW}}$. Thus there should be a “pseudogap” temperature for iron pnictides as well. Below such a pseudogap temperature, it is appropriate to consider an effective lattice spin model with fixed moments such as the one given by eq. (1) to describe the magnetic properties of the system. By comparing the energy of a variety of magnetic structures, Ma et al. [5] have estimated $J_1 \sim J_2$ to be about 55 meV for LaFeAsO and 35 meV for BaFe$_2$As$_2$.

In conclusion, we have argued that the universal linear-temperature dependence of the susceptibility provides a strong evidence for the SDW fluctuation moments with strong antiferromagnetic interactions above the SDW transition temperature in iron pnictides. This linear static uniform susceptibility can be effectively described as the finite temperature behavior of a Heisenberg model with the nearest- and next-nearest-neighbor AF interactions. Further investigations are certainly needed to put our conclusion on a solid ground.

***

This work is partially supported by NSFC-China and the National Program for Basic Research of MOST, China.

REFERENCES

[1] Kamihara Y., Watanabe T., Hirano M. and Hosono H., J. Am. Chem. Soc., 130 (2008) 3296.
[2] de la Cruz C., Huang Q., Lynn J. W., Li J., Ratcliff II, Zarestky J. L., Mook H. A., Chen G. F., Luo J. L., Wang N. L. and Dai P., Nature (London), 453 (2008) 899.
[3] Dong J., Zhang H. J., Xu G., Li Z., Li G., Hu W. Z., Wu D., Chen G. F., Dai X., Luo J. L., Fang Z. and Wang N. L., EPL, 83 (2008) 27006.
[4] Mazin I. I., Singh D. J., Johannes M. D. and Du M. H., Phys. Rev. Lett., 101 (2008) 057003.
[5] Ma F., Lu Z. Y. and Xiang T., Phys. Rev. B, 78 (2008) 224517; arXiv:0806.3526.
[6] Yildirim T., Phys. Rev. Lett., 101 (2008) 057010.
[7] St Q. and Ahrahams E., Phys. Rev. Lett., 101 (2008) 076401.
[8] Wu G., Chen H., Wu T., Xie Y. L., Yan Y. J., Liu R. H., Wang X. F., Ying J. J. and Chen X. H., J. Phys.: Condens. Matter, 20 (2008) 422201.
[9] Wang X. F. et al., Phys. Rev. Lett., 102 (2009) 117005.
[10] Yan Y. Q., Kreyssig A., Nandi S., Ni N., Bud’ko S. L., Kracher A., McQueeney R. J., McCallum R. W., Lograsso T. A., Goldman A. I. and Canfield P. C., Phys. Rev. B, 78 (2008) 024516.
[11] Klingeler R., Lesp N., Hellmann I., Popa A., Hess C., Kondrat A., Hamann-Borrero J., Behr G., Kataev V. and Buechner B., arXiv:0808.0708.
[12] Chen G. F., Luo J. L. and Wang N. L., unpublished.
[13] Nakano T., Oda M., Manabe C., Momono N., Miura Y. and Ido M., Phys. Rev. B, 49 (1994) 16000.
[14] Chuubukov A. V. and Sachdev S., Phys. Rev. Lett., 11 (1993) 169.
[15] Makivic M. and Ding H.-Q., Phys. Rev. B, 43 (1991) 3562.
[16] Kim J.-K. and Troyer M., Phys. Rev. Lett., 80 (1998) 2705.
[17] Fawcett E., Rev. Mod. Phys., 60 (1988) 209; Fawcett E., Alberts H. L., Yu Galkin V., Noakes D. R. and Yakhmi J. V., Rev. Mod. Phys., 66 (1994) 25.
[18] Grier B. H., Shirane G. and Werner S. A., Phys. Rev. B, 31 (1985) 2892.
[19] Foo M. L., Wang Y., Watauchi S., Zandbergen H. W., He T., Cava R. J. and Ong N. P., Phys. Rev. Lett., 92 (2004) 247001.
[20] Hinzke D., Nowak U. and Garanin D. A., Eur. Phys. J. B, 16 (2000) 435.
[21] Takahashi M., Phys. Rev. B, 40 (1989) 2494.
[22] Su Y.-H. et al., in preparation.
[23] Fang C., Yao H., Tsai W. F., Hu J. P. and Kivelson S. A., Phys. Rev. B, 77 (2008) 224509.
[24] Xu C., Muller M. and Sachdev S., Phys. Rev. B, 78 (2008) 020501 (R).
[25] Hu W. Z., Dong J., Li G., Li Z., Zheng P., Chen G. F., Luo J. L. and Wang N. L., Phys. Rev. Lett., (101) 2008257005.
[26] Cheng P., Yang H., Jia Y., Fang L., Zhu X., Mu G. and Wen H.-H., Phys. Rev. B, 78 (2008) 134508.
[27] Yang L. X. et al., Phys. Rev. Lett., 102 (2009) 107002.
[28] Huang Q., Qiu Y., Bao W., Lynn J. W., Green M. A., Gasparovic Y. C., Wu T., Wu G. and Chen X. H., Phys. Rev. Lett., 101 (2008) 257003.
[29] Ran Y. et al., Phys. Rev. B, 79 (2009) 014505.