

Electronic instabilities in iron-based superconductors: A variational Monte Carlo study

Fan Yang,^{1,2} Hui Zhai,³ Fa Wang,⁴ and Dung-Hai Lee^{1,5}

¹*Department of Physics, University of California at Berkeley, Berkeley, California 94720, USA*

²*Department of Physics, Beijing Institute of Technology, Beijing 100081, People's Republic of China*

³*Institute for Advanced Study, Tsinghua University, Beijing 100084, China*

⁴*Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA*

⁵*Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA*

(Received 16 February 2011; published 5 April 2011)

We report a variational Monte Carlo (VMC) study of the iron-based superconductors. We use realistic band structures, and the ordering instabilities/variational ansatz are suggested by previous functional renormalization group calculations. We examine antiferromagnetism, superconducting pairing, normal state Fermi surface distortion, and orbital order in the antiferromagnetic state.

DOI: [10.1103/PhysRevB.83.134502](https://doi.org/10.1103/PhysRevB.83.134502)

PACS number(s): 74.20.Mn, 74.25.Gz, 74.72.-h

The variational approach has a glorious history in condensed matter physics. Examples of successful wave functions include the BCS wave function, Laughlin wave function, Jastrow wave function, etc. In the study of the cuprate superconductivity, Anderson's projected BCS wave function has been shown to capture many important aspects of the cuprates.¹ Despite the successes, all variational approaches have the drawback of being biased; i.e., they are based on the assumption that the few variational parameters built into the variational ansatz enable it to capture the essence of the true ground state wave function.

Understanding the pairing mechanism and possible electronic instabilities in iron-based superconductors has been a focus of interests in the past few years. This is not only for their high T_c , but also for many similarities they share with the cuprates. Presently most of the works attribute the electron pairing to the antiferromagnetic fluctuations, and it is widely believed that the superconducting (SC) gap function takes opposite signs on the electron and hole Fermi surfaces. This is partly because important consequences of the opposite pairing sign predicted for the neutron^{2,3} and STM experiments^{4,5} have now received experimental support.^{6,7}

On the theory side, except for Ref. 8 which assumes the iron-based compounds are doped Mott insulator, most of the studies are based on weak coupling approximations.^{9–20} Experimentally there are reports favoring weak²¹ and intermediate²² electronic correlation in this class of compounds. Among Refs. 9–20 the functional renormalization group (FRG)¹¹ approach has the virtue of being unbiased. It sums all one-loop particle-particle, particle-hole diagrams as well as the vertex corrections. The results predict that in addition to the AFM and SC instabilities, the iron-based compounds have a propensity toward Fermi surface distortion, magnetically coupled orbital order, and charge density wave orderings.¹⁸ However given the fact that there is an evidence that the pnictides are intermediate coupling materials,²² we must ask which, if any, of the weak coupling results in Refs. 9–20 are valid.

One of the main purpose of this paper is to address the above question by performing a variational Monte Carlo (VMC) calculation. In addition, while FRG only allows us to access the ordering tendencies, VMC allow us to quantitatively study the ordered state. We use the realistic band structures, and the

variational ansatz are guided by the predictions of Ref. 18. We use partially projected wave functions where both the degree of projection and the order parameters are variational degrees of freedom. We shall focus on normal state Fermi surface distortion, AFM coupled orbital order, and superconducting pairing. (In FRG the tendency toward charge density wave order is the weakest.)

The band structure we use is that of Kuroki *et al.*¹⁵ We model the electronic correlations by the Hubbard and Hunds types of local interactions. The Hamiltonian is given as

$$H = H_{\text{band}} + U_1 \sum_{i\mu} n_{i\mu\uparrow} n_{i\mu\downarrow} + U_2 \sum_{i,\mu<\nu} n_{i\mu} n_{i\nu} + J_H \times \left[\sum_{\mu<\nu} \sum_{\sigma\sigma'} c_{i\mu\sigma}^+ c_{i\nu\sigma'}^+ c_{i\mu\sigma'} c_{i\nu\sigma} + c_{i\mu\uparrow}^+ c_{i\mu\downarrow}^+ c_{i\mu\downarrow} c_{i\mu\uparrow} + \text{H.c.} \right], \quad (1)$$

where H_{band} and its parameters can be found in Ref. 15 and its Table I. In most of the paper we shall use $(U_1, U_2, J_H) = (4, 2, 0.7)$ eV. While these parameters are compatible with Ref. 22 it is larger than what is reported in Ref. 21. The additional reasons for doing so are (1) similar large correlation parameters are concluded from LDA + DMFT calculations²³ although with controversy,²⁴ (2) smaller interaction parameters will make the energy gain associated with the superconducting and orbital order too small for our VMC accuracy. As the energy reduction due to normal state FS distortion is relatively larger, smaller interaction parameters, e.g., $(U_1, U_2, J_H) \sim (2, 1, 0.35)$ eV, were also used.

We use the following partially projected variational wave functions:

$$|\psi\rangle = g_1^{\hat{N}_1} g_2^{\hat{N}_2} \hat{P}_{N_e} |\psi_{MF}\rangle, \quad (2)$$

where

$$\hat{N}_1 = \sum_{i,\mu} \hat{n}_{i\mu\uparrow} \hat{n}_{i\mu\downarrow}, \quad \hat{N}_2 = \sum_{i,\mu<\nu} \hat{n}_{i\mu} \hat{n}_{i\nu}. \quad (3)$$

\hat{P}_{N_e} fixes the total number of electron to N_e , and $0 \leq g_1, g_2 \leq 1$ suppress configurations according to the number of electrons residing in the same and different on-site orbitals. The mean-field wave function $|\psi_{MF}\rangle$ depends on the type of electronic order we are studying, and it will be discussed in detail later.

The VMC was carried out on $L^2 = 10 \times 10$ and 18×18 lattices with periodic boundary condition. In choosing the doping level, we were careful to avoid degeneracy in the band energy between different Slater determinants. A standard Markovian chain Monte Carlo approach was used with METROPOLIS update algorithm. After an initial “equilibration” using 10^5 Monte Carlo steps, about 10^5 , 10^6 , and 10^7 samples were used in the study of Fermi surface distortion, orbital order and SC pairing, respectively. The adjacent samples are separated by $5L^2$ Monte Carlo steps with each showing an acceptance ratio of about 0.23, which is enough to eliminate autocorrelation and thus guarantee an efficient sampling. Due to the higher demand of accuracy when studying SC pairing, we have applied the “reweighting” scheme.²⁵ The error bars are estimated by calculating the variance of the energy expectation.

Normal state FS distortion. FRG predicts two leading FS distortion: one preserves the 90° rotation symmetry and the other breaks it.¹⁸ The former shrinks both the electron and hole pockets and produces a relative energy shift of the electron and the hole bands.²⁶ The latter is the band version of orbital ordering, and is suggested to be stabilized by the AFM.¹⁸ The mean-field state $|\psi_{MF}\rangle$ for the two types of FS distortion are the ground state of the following quadratic Hamiltonian:

$$H_{MF} = \sum_{\mathbf{k}\alpha\sigma} [\varepsilon_{\mathbf{k}}^\alpha + \chi(\mathbf{k})] n_{\mathbf{k}\alpha\sigma}. \quad (4)$$

where $\varepsilon_{\mathbf{k}}^\alpha$ is the bare band dispersion, $n_{\mathbf{k}\alpha\sigma} = c_{\mathbf{k}\alpha\sigma}^\dagger c_{\mathbf{k}\alpha\sigma}$ and $\chi(\mathbf{k})$ can be $\chi_0 \cos k_x \cos k_y$ (90° preserving) or $\chi_0(\cos k_x - \cos k_y)$ (90° breaking).

In the absence of AFM order, the results for 0.6% hole-doping on a 18×18 lattice using $(U_1, U_2, J_H) = (4, 2, 0.7)$ eV are shown in the main panel of Fig. 1. The black and red symbols represent $\bar{E}(\chi_0) - \bar{E}(0)$ as a function of χ_0 with $g_{1,2}$ fixed at their optimized values. (The optimized $(g_1, g_2) \approx [0.31(4), 0.76(4)]$ are nearly independent of χ , as the energy gain associated with their optimization is much larger than that associated with optimization of χ .) From Fig. 1, it is clear that while both $\cos k_x \cos k_y$ and $\cos k_x - \cos k_y$ FS distortions gain energy, the former is the most energetically favorable one. This is consistent with the FRG prediction. The green symbols in Fig. 1 represent the energy reduction due to a real space version of orbital order (see later). We note that it is not energetically favorable in the nonmagnetic state. At the optimal χ_0 the total energy gain is about 27 meV per site. The negative χ_0 moves the bands near Γ downward and the bands near M upward. As a result it shrinks both the electron and hole pockets (it actually splits each electron pocket into two smaller ones). The distorted Fermi surface is compared with the undistorted one in Fig. 2(a)–2(b). The above results are completely consistent with the FRG prediction.¹⁸ In the inset of Fig. 1, we show the $\bar{E}(\chi_0) - \bar{E}(0)$ versus χ_0 plot for $U_1 = 2, U_2 = 1, J_H = 0.35$ eV with $\cos k_x \cdot \cos k_y$ form factor at 13% hole doping. The resulting relative energy shift of the bands near Γ and M is $90 - 126$ meV which is in good quantitative agreement with the experimentally observed ~ 100 meV relative shift.^{26,29}

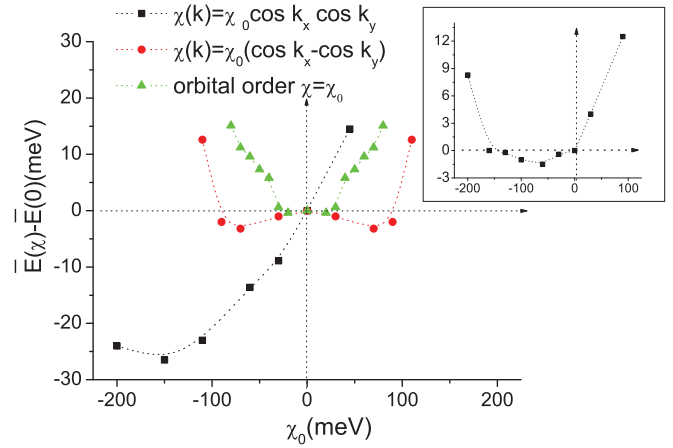


FIG. 1. (Color online) Energy gain per site as functions of χ_0 for the normal state FS distortion, and the band and real space version of orbital ordering. The results were obtained on a 18×18 lattice under periodic boundary condition. The parameters g_1, g_2 are fixed at their optimal values. The size of points represents the error bar and the dotted lines are a guide to the eyes. The main panel is for $U_1 = 4, U_2 = 2, J_H = 0.7$ eV at 0.6% hole doping. In the inset the result for at 13% hole doping, with $U_1 = 2, U_2 = 1, J_H = 0.35$ eV and $\cos k_x \cdot \cos k_y$ form factor is shown.

Orbital order in the AFM state. The $|\psi_{MF}\rangle$ for the AFM ordered state is the ground state of the following mean-field Hamiltonian:

$$H_{MF} = H_0 + \sum_{i\mu\nu\sigma} (c_{i\mu\sigma}^\dagger c_{i\nu\sigma}) M_{\mu\nu} e^{i\mathbf{Q} \cdot \mathbf{R}_i}, \quad (5)$$

where H_0 is given by either Eq. (4) or Eq. (6). Here $\mathbf{Q} = (\pi, 0)$ is the AFM ordering wave vector. As for $M_{\mu\nu}$ we used the mean-field result of Ref. 30 where the nonzero $M_{\mu\nu}$ are $M_{\mu\mu}, \mu = 1, \dots, 5$, and $M_{15}, M_{51} (= M_{15})$. The variational study is performed by keeping the ratio between the optimal mean-field parameters, $(M_{22}, M_{33}, M_{44}, M_{55}, M_{15})/M_{11} = (0.95, 1.03, 1.1, 0.76, -0.096)$, while allowing M_{11} to vary. For $(U_1, U_2, J_H) = (4, 2, 0.7)$ eV at 0.6% electron-doping (on 18×18 lattice) we found $(g_1, g_2) = [0.49(0), 0.76(6)]$, $M_{11} = 1.4$ eV, with a total energy reduction of about 3.0 eV per site, with nearly $2\mu_B$ ordering moment. This value is significantly larger than the measured ones for the stoichiometric compounds. The discrepancy can be due to the omission of the fluctuations in the orientation of magnetic moments and the

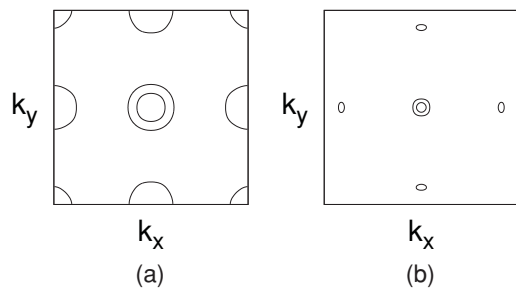


FIG. 2. The Fermi surfaces at 0.6% hole doping. (a) In the absence of FS distortion. (b) The distorted Fermi surfaces associated with $\chi(\mathbf{k}) = \chi_0 \cos k_x \cos k_y$.

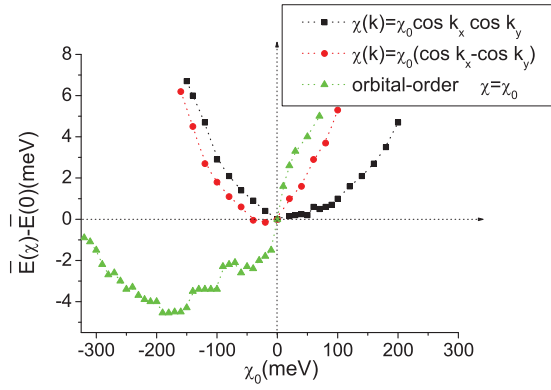


FIG. 3. (Color online) The energy gain per site in the AFM state as function of χ_0 for the three types of electronic orders studied in Fig. 1(a). The same lattice size and interaction parameters were used. The doping is 0.6%-electron doping. The variational parameters $g_1, g_2, M_{\mu\nu}$ are fixed at their optimized values. The size of points represents the error bar. The dotted lines are just guides to the eyes.

ordering wave vectors, and/or the large values of the interaction parameters.

In view of the strong atomic-like ordering moments in the AFM state obtained above, when studying the orbital ordering in the magnetic state we also adopt a real space version of orbital ordering,²⁸ where $|\psi_{MF}\rangle$ is the ground state of

$$H_{MF} = \sum_{\mathbf{k}\alpha\sigma} \varepsilon_{\mathbf{k}}^{\alpha} n_{\mathbf{k}\alpha\sigma} - \chi_0 \sum_{i,\sigma} (n_{i,yz\sigma} - n_{i,xz\sigma}). \quad (6)$$

Here $n_{i\alpha\sigma} = c_{i\alpha\sigma}^{\dagger} c_{i\alpha\sigma}$. We note that the orbital d_{xz}, d_{yz} we use are 45° rotated from the d_{xz}, d_{yz} orbitals in Ref. 15. From Fig. 3, we conclude that in the presence of AFM, the real-space d_{xz}/d_{yz} orbital order is the most energetically favorable (we suspect this is due to the fact that the large, localized, ordering moment in the AFM state). It produces a total energy gain of about 4.6 meV per site. At the optimal χ_0 the occupation-number difference between the xz and yz orbitals is $\bar{n}_{d_{xz}} - \bar{n}_{d_{yz}} = 0.20(1)$. This value is enhanced above the ~ 0.15 occupation difference already present in the pure AFM state. Our result agrees qualitatively with a recent photoemission result²⁷ and a first principle Wannier function calculation.²⁸ Comparing this result with the green symbols of Fig. 1(a), we conclude that this orbital order is stabilized by the AFM.

Superconducting pairing. The $|\psi_{MF}\rangle$ we use for studying pairing is the ground state of the following mean-field Hamiltonian:

$$H_{MF} = \sum_{\mathbf{k}\alpha\sigma} (\varepsilon_{\mathbf{k}}^{\alpha} - \mu_c) n_{\mathbf{k}\alpha\sigma} + \sum_{\mathbf{k}\alpha} (\Delta_{\mathbf{k}}^{\alpha} c_{\mathbf{k}\alpha\uparrow}^{\dagger} c_{-\mathbf{k}\alpha\downarrow}^{\dagger} + \text{H.c.}). \quad (7)$$

We have studied four different types of gap functions:

$$\begin{aligned} \Delta_{\mathbf{k}}^{\alpha} &= \Delta_0 (\cos k_x + \cos k_y), & \Delta_{\mathbf{k}}^{\alpha} &= \Delta_0 \cos k_x \cdot \cos k_y, \\ \Delta_{\mathbf{k}}^{\alpha} &= \Delta_0, & \Delta_{\mathbf{k}}^{\alpha} &= \Delta_0 (\cos k_x - \cos k_y). \end{aligned} \quad (8)$$

In addition we only allow pairing in bands that cross the Fermi energy. This is an excellent approximation because, as shown below, the gaps are quite small.

Our calculations for the SC state were carried out on a 10×10 lattice at 10% hole doping. The optimized $(g_1, g_2) \approx$

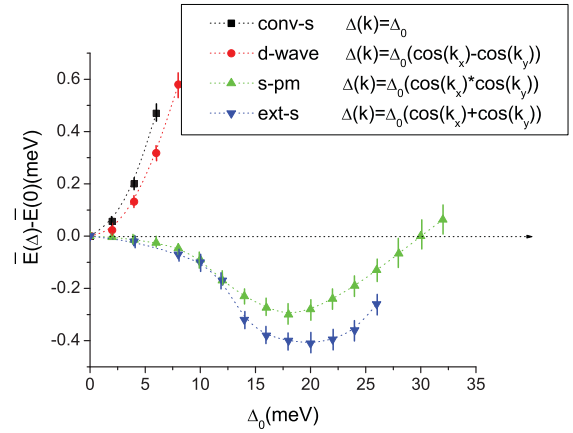


FIG. 4. (Color online) Energy gain per site as function of Δ_0 for four different pairing form factors. The lattice size is 10×10 , and 10% hole doping is studied. The other variational parameters g_1, g_2, μ_c are fixed at their optimized values. Error bars are shown. The dotted lines are to guide the eyes.

$[0.31(1), 0.76(2)]$. Again, to an excellent approximation, the optimal values of $g_{1,2}$ and μ_c do not depend on Δ_0 . The results for $\bar{E}(\Delta_0) - \bar{E}(0)$ with (g_1, g_2, μ_c) fixed at their optimal values are shown in Fig. 4. The results suggest that while both conventional- s and $d_{x^2-y^2}$ pairing raise the energy, s_{\pm} and extended s lower it.

Interestingly, our result suggests the extended s $[\Delta_0 (\cos k_x + \cos k_y)]$ form factor is slightly favored (by 0.1 ± 0.08 meV) over the s_{\pm} ($\Delta_0 \cos k_x \cdot \cos k_y$) one. On the surface this contradicts the predictions of s_{\pm} pairing form factor. However it is important to realize that the s_{\pm} gap function is not synonymous to $\cos k_x \cos k_y$. Indeed, the s_{\pm} form factor obtained from several weak coupling approaches^{11,14,15} has a strong variation around the electron Fermi surfaces. We believe the near degeneracy of the extended- s and the $\cos k_x \cos k_y$ form factors suggests the optimal pairing form factor is a linear combination of the two (hence is anisotropic on the electron pockets). However it is extremely computationally time consuming to verify this, and we have not been able to do it. In addition, we caution that the degree of the gap function variation on the electron pockets will depend on the values of the parameters. We cannot rule out that for other parameter sets the $\cos k_x \cos k_y$ can be the leading pairing form factor. Finally, the optimal gap amplitudes for the two symmetries are about 18 meV and 20 – 22 meV, respectively. Order-of-magnitude-wise these values are not far from the experimentally measured gap scales.

In conclusion, we have performed a variational Monte Carlo calculation to check the validity of our previous results based on a weak-coupling approximation. The results are qualitatively consistent. We caution that because of the time-consuming nature of the calculation we are only able to study two sets of interaction parameters. Clearly we cannot rule out the possibility that quantitative aspects of the above results will be sensitive to the precise values of the interaction parameters.

ACKNOWLEDGMENTS

We thank Zheng-Yu Weng for sharing computer resources and Hong Yao, Ying Ran, and Tao Li for helpful discussions.

We acknowledge the support by NSFC Grant No. 10704008 (FY); BRYs Program of Tsinghua University and NSFC Grant No. 10944002 (HZ); and DOE grant no. DE-AC02-

05CH11231 (DHL). This research also used the resources of NERSC supported by the Office of Science of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.

-
- ¹For a review and further references see P. W. Anderson, P. A. Lee, M. Randeria, T. M. Rice, N. Trivedi, and F. C. Zhang, *J. Phys. Condens. Matter* **16**, R755 (2004).
- ²T. A. Maier and D. J. Scalapino, *Phys. Rev. B* **78**, 020514(R) (2008).
- ³M. M. Korshunov and I. Eremin, *Phys. Rev. B* **78**, 140509(R) (2008).
- ⁴F. Wang, H. Zhai, and D.-H. Lee, *Europhys. Lett.* **85**, 37005 (2009).
- ⁵Y. Y. Zhang, C. Fang, X. Zhou, K. Seo, W. F. Tsai, B. A. Bernevig, and J. Hu, *Phys. Rev. B* **80**, 094528 (2009).
- ⁶M. D. Lumsden *et al.*, *Phys. Rev. Lett.* **102**, 107005 (2009); S. Chi *et al.*, *ibid.* **102**, 107006 (2009); S. Li, Y. Chen, S. Chang, J. W. Lynn, L. Li, Y. Luo, G. Cao, Z. Xu, and P. Dai, *Phys. Rev. B* **79**, 174527 (2009); D. S. Inosov *et al.*, *Nature Physics* **6**, 178 (2010); J. Zhao *et al.*, *Phys. Rev. B* **81**, 180505(R) (2010).
- ⁷T. Hanaguri, S. Niitaka, K. Kuroki, and H. Takagi, *Science* **328**, 474 (2010).
- ⁸K. Seo, B. A. Bernevig, and Jiangping Hu, *Phys. Rev. Lett.* **101**, 206404 (2008).
- ⁹I. I. Mazin, D. J. Singh, M. D. Johannes, and M. H. Du, *Phys. Rev. Lett.* **101**, 057003 (2008).
- ¹⁰Z. J. Yao, J. X. Li, and Z. D. Wang, *New J. Phys.* **11**, 025009 (2009).
- ¹¹F. Wang, H. Zhai, Y. Ran, A. Vishwanath, and D.-H. Lee, *arXiv:0805.3343*; *Phys. Rev. Lett.* **102**, 047005 (2009).
- ¹²A. V. Chubukov, D. V. Efremov, and I. Eremin, *Phys. Rev. B* **78**, 134512 (2008).
- ¹³H. Ikeda, *J. Phys. Soc. Jpn.* **77**, 123707 (2008).
- ¹⁴S. Graser, T. A. Maier, P. J. Hirschfeld, and D. J. Scalapino, *New J. Phys.* **11**, 025016 (2009).
- ¹⁵K. Kuroki, S. Onari, R. Arita, H. Usui, Y. Tanaka, H. Kontani, and H. Aoki, *Phys. Rev. Lett.* **101**, 087004 (2008).
- ¹⁶K. Kuroki, H. Usui, S. Onari, R. Arita, and H. Aoki, *Phys. Rev. B* **79**, 224511 (2009).
- ¹⁷T. A. Maier, S. Graser, D. J. Scalapino, and P. J. Hirschfeld, *Phys. Rev. B* **79**, 224510 (2009).
- ¹⁸H. Zhai, F. Wang, and D.-H. Lee, *Phys. Rev. B* **80**, 064517 (2009).
- ¹⁹F. Wang, H. Zhai, and D.-H. Lee, *Phys. Rev. B* **81**, 184512 (2010).
- ²⁰A. V. Chubukov, M. G. Vavilov, and A. B. Vorontsov, *Phys. Rev. B* **80**, 140515(R) (2009); R. Thomale, C. Platt, J. Hu, C. Honerkamp, and B. A. Bernevig, *ibid.* **80**, 180505(R) (2009).
- ²¹W. L. Yang *et al.*, *Phys. Rev. B* **80**, 014508 (2009).
- ²²See, e.g., M. M. Qazilbash, J. J. Hamlin, R. E. Baumbach, Lijun Zhang, D. J. Singh, M. B. Maple, and D. N. Basov, *Nature Physics* **5**, 647 (2009).
- ²³L. Craco, M. S. Laad, S. Leoni, and H. Rosner, *Phys. Rev. B* **78**, 134511 (2008); K. Haule, J. H. Shim, and G. Kotliar, *Phys. Rev. Lett.* **100**, 226402 (2008).
- ²⁴Some other LDA + DMFT studies listed below provide a different picture for the model parameter. Namely, that U 4 is relevant for a model including the As *p* orbitals, but that a reduced U should be used in the five-band model. V. I. Anisimov, Dm. M. Korotin, M. A. Korotin, A. V. Kozhevnikov, J. Kunes, A. O. Shorikov, S. L. Skornyakov, and S. V. Streltsov, *J. Phys. Condens. Matter* **7**, 075602 (2009); M. Aichhorn, L. Pourovskii, V. Vildosola, M. Ferrero, O. Parcollet, T. Miyake, A. Georges, and S. Biermann, *Phys. Rev. B* **80**, 085101 (2009).
- ²⁵D. Ceperley, G. V. Chester, and K. H. Kalos, *Phys. Rev. B* **16**, 3081 (1977); C. J. Umrigar, K. G. Wilson, and J. W. Wilkins, *Phys. Rev. Lett.* **60**, 1719 (1988).
- ²⁶Donghui Lu, private communication.
- ²⁷M. Yi *et al.*, e-print *arXiv:1011.0050*.
- ²⁸Chi-Cheng Lee, Wei-Guo Yin, and Wei Ku, *Phys. Rev. Lett.* **103**, 267001 (2009).
- ²⁹A. I. Coldea, J. D. Fletcher, A. Carrington, J. G. Analytis, A. F. Bangura, J.-H. Chu, A. S. Erickson, I. R. Fisher, N. E. Hussey, and R. D. McDonald, *Phys. Rev. Lett.* **101**, 216402 (2008).
- ³⁰Ying Ran, Fa Wang, Hui Zhai, Ashvin Vishwanath, and Dung-Hai Lee, *Phys. Rev. B* **79**, 014505 (2009).
- ³¹J. D. Fletcher, A. Serafin, L. Malone, J. Analytis, J.-H. Chu, A. S. Erickson, I. R. Fisher, and A. Carrington, *Phys. Rev. Lett.* **102**, 147001 (2009).
- ³²C. W. Hicks, T. M. Lippman, M. E. Huber, J. G. Analytis, J. H. Chu, A. S. Erickson, I. R. Fisher, and K. A. Moler, *Phys. Rev. Lett.* **103**, 127003 (2009).