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

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The electronic instabilities of the Iron-based superconductors: a variational Monte-Carlo study

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We report the first variational Monte Carlo (VMC) study of the iron-based superconductors. We use realistic band structures, and the ordering instabilities/variational ansatzs 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.

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The variational approach has a glorious history in condensed matter physics. Examples of successful wavefunctions 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 variation approach have the draw back of being biased: i.e., it bases on the assumption that the few variational parameters built into the variational ansatz enable it to capture the essence of the true ground state wavefunction.

Understanding the pairing mechanism and possible electronic instabilities in iron-based superconductors have 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 exeriments[4, 5] have now received experimental supports[6, 7].

On the theory side, except 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 weak[21] and intermediate[22] electronic correlation in this class of compounds. Among Ref.[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 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 Ref.[9–20] are valid.

One of the main purpose of this letter 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 bandstructures, and the variational ansatzs 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 follows:

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

where $H_{\rm band}$ 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's reported in Ref.[21].The additional reasons for doing so are (1) similar large correlation parameters are concluded from LDA+DMFT calculations[23] although with controversery[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,

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

where

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

$\hat{P}_{N_e}$ 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 wavefunction $|\psi_{MF}\rangle$ depends on the type of electronic order we are studying, and it will be discussed in details 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. Standard Markovian chain Monte Carlo approach was used with Metropolis update algorithm. After an initial "equillibration" 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 guaranty an efficient sampling. Due to the higher demand of accuracy when studying SC pairing, we have applied the "re-weighting" 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^o 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 later 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

$$H_{MF} = \sum_{\mathbf{k}\alpha\sigma} (\varepsilon^{\alpha}_{\mathbf{k}} + \chi(\mathbf{k})) n_{\mathbf{k}\alpha\sigma}. \tag{4}$$

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

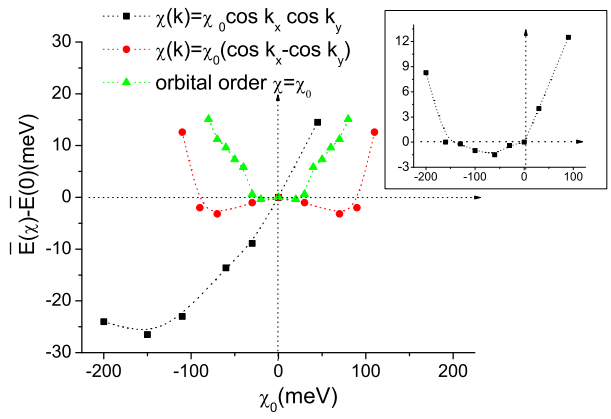

FIG. 1: (Color online) Energy gain per site as functions of χ_0 for the normal state FS distortion, and the band and real space version of orbital ordering. The results were obtained on a 18×18 lattice under periodic boundary condition. The parameters g_1, g_2 are fixed at their optimal values. The size of points represents the error-bar and the dotted lines are 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.

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 non-magnetic state. At the optimal χ_0 the total energy gain is about 27meV 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,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].

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,

$$H_{MF} = H_0 + \sum_{i\mu\nu\sigma} \left(\sigma c^{+}_{i\mu\sigma} c_{i\nu\sigma}\right) M_{\mu\nu} e^{i\mathbf{Q}\cdot\mathbf{R}_i}, \tag{5}$$

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 non-zero $M_{\mu\nu}$ are $M_{\mu\mu}, \mu = 1, .., 5$, and $M_{15}, M_{51} (= M_{15})$. The variational study is per-

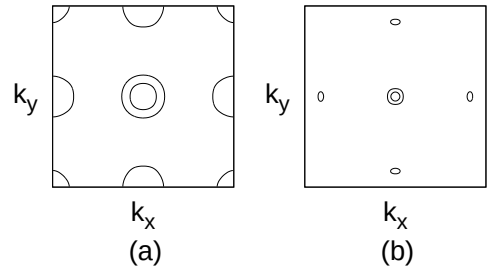

FIG. 2: The Fermi surfaces at 0.6% hole doping. (a) In the absence of FS distortion. (b) The distorted Fermi surfaces associated with $\chi(\mathbf{k}) = \chi_0 \cos k_x \cos k_y$.

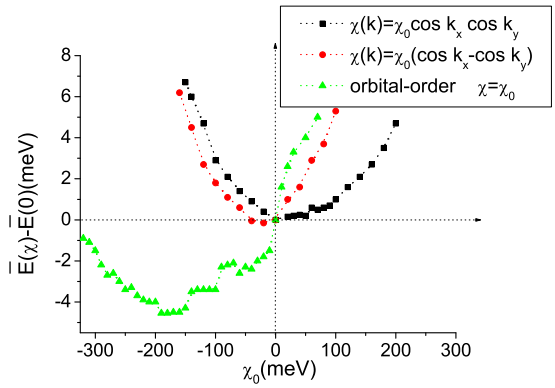

FIG. 3: (Color online) The energy gain per site in the AFM state as function of χ_0 for the three types of electronic orders studied in Fig.1(a). The same lattice size and interaction parameters were used. The doping is 0.6%-electron doping. The variational parameters $g_1, g_2, M_{\mu\nu}$ are fixed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

formed 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 $(g_1, g_2) = (0.49(0), 0.76(6))\,, M_{11} = 1.4eV$, with a total energy reduction of about 3.0eV 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 ordering wavevectors , 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

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

Here $n_{i\alpha\sigma} = c^{\dagger}_{i\alpha\sigma} c_{i\alpha\sigma}$. We note that the orbital d_{xz}, d_{yz} we use are 45^o 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 4.6meV 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 result[27] 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

$$H_{MF} = \sum_{\mathbf{k}\alpha\sigma} (\varepsilon^{\alpha}_{\mathbf{k}} - \mu_c) n_{\mathbf{k}\alpha\sigma} + \sum_{\mathbf{k}\alpha} \left(\Delta^{\alpha}_{\mathbf{k}} c^{+}_{\mathbf{k}\alpha\uparrow} c^{+}_{-\mathbf{k}\alpha\downarrow} + h.c. \right) . (7)$$

We have studied four different types of gap functions:

$$\Delta^{\alpha}_{\mathbf{k}} = \Delta_0 (\cos k_x + \cos k_y)\,, \ \Delta^{\alpha}_{\mathbf{k}} = \Delta_0 \cos k_x \cdot \cos k_y$$
$$\Delta^{\alpha}_{\mathbf{k}} = \Delta_0, \quad \Delta^{\alpha}_{\mathbf{k}} = \Delta_0 (\cos k_x - \cos k_y)\,. \qquad (8)$$

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 $(g_1, g_2) \approx (0.31(1), 0.76(2))$. Again, to an excellent approximation, the optimal value of $g_{1,2}$ and μ_c do not depends 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 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 computing 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 depends 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

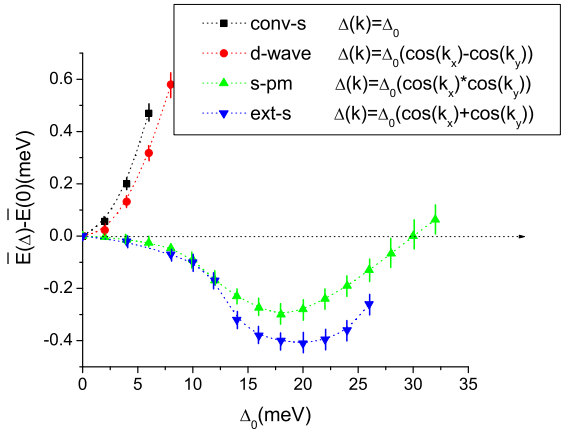

FIG. 4: (Color online) Energy gain per site as function of Δ_0 for four different pairing form factors. The lattice size is 10×10, and 10% hole doping is studied. The other variational parameters g_1, g_2, μ_c are fixed at their optimized values. Error-bars are shown. The dotted lines are guide to the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

leading pairing form factor. Finally, the optimal gap amplitudes for the two symmetries are about 18meV 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 variational Monte-Carlo calculation to check the validity of our previous results based on 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 can not rule of the possibility that quantitative aspects of the above results will be sensitive to the precise values of the interaction parameters.

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[1] For 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).
[2] T.A.Maier, and D.J.Scalapino, Phys. Rev. B **78**, 020514(R) (2008).
[3] M.M. Korshunov, and I. Eremin, Phys. Rev. B **78**, 140509(R) (2008).
[4] F. Wang, H. Zhai, and D.-H. Lee, Europhys. Lett. **85**, 37005 (2009).
[5] Y.Y.Zhang, C.Fang, X. Zhou, K. Seo, W.F.Tsai, B.A. Bernevig, and J. Hu, Phys. Rev. B **80**, 094528 (2009).
[6] M. D. Lumsden *et al*, Phys. Rev. Lett. **102**, 107005 (2009); S.Chi *et al*, Phys. Rev. Lett **102**, 107006 (2009); S. Li *et al*, Phys. Rev. B **79**, 174527 (2009); D. S. Inosov *et al*, Nature Physics 6, 178-181 (2010); J. Zhao *et al*, arXiv:0908.0954 (2009).
[7] T. Hanaguri, S. Niitaka, K. Kuroki, and H. Takagi, Science **328**, 474 (2010).
[8] K.Seo, B. A. Bernevig, Jiangping Hu, Phys. Rev. Lett. **101**, 206404 (2008).
[9] I.I. Mazin, D.J. Singh, M.D. Johannes, and M.H. Du, Phys. Rev. Lett. **101**, 057003 (2008).
[10] Z.J. Yao, J.X. Li, and Z. D. Wang, New J. Phys. **11**, 025009 (2009).
[11] F. Wang, H. Zhai, Y. Ran, A. Vishwanath, and D.-H. Lee, arXiv:0805.3343 (2008);Phys. Rev. Lett. **102**, 047005 (2009).
[12] A. V. Chubukov, D. V. Efremov, and I. Eremin, Phys. Rev. B **78**, 134512 (2008).
[13] H. Ikeda, J. Phys. Soc. Jpn. **77**, 123707 (2008).
[14] S. Graser, T. A. Maier, P. J. Hirschfeld, and D. J. Scalapino, New J. Phys. **11**, 025016 (2009).
[15] K. Kuroki, S. Onari, R. Arita, H. Usui, Y. Tanaka, H. Kontani, and H. Aoki, Phys. Rev. Lett. **101**, 087004 (2008).
[16] K. Kuroki, H. Usui, S. Onari, R. Arita, and H. Aoki, Phys. Rev. B **79**, 224511 (2009).
[17] T.A. Maier, S. Graser, D.J. Scalapino, and P.J. Hirschfeld, Phys. Rev. B **79**, 224510 (2009).
[18] H. Zhai, F. Wang, and D.-H. Lee, Physical Review B, **80**, 064517 (2009).
[19] F. Wang, H. Zhai, and D.-H. Lee, Phys. Rev. B **81**, 184512 (2010).
[20] A.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, Phys. Rev. B **80**, 180505(R) (2009).
[21] W.L. Yang *et al*, Phys. Rev. B **80**, 014508 (2009)
[22] See, e.g., M. M. Qazilbash, J. J. Hamlin, R. E. Baumbach, Lijun Zhang, D. J. Singh, M. B. Maple, D. N. Basov Nature Physics 5, 647 (2009).
[23] L. Craco *et al*, Phys. Rev. B **78**, 134511 (2008); K. Haule *et al*, Phys. Rev. Lett. **100**, 226402 (2008).
[24] Some 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 5-band model. V. I. Anisimov, Dm. M. Korotin, M. A. Korotin, A. V. Kozhevnikov, J. Kunes, A. O. Shorikov, S. L. Skornyakov, S. V. Streltsov, J. Phys.: Condens. Matter 21 No 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).
[25] D. 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).
[26] Donghui Lu, private communication.
[27] M. Yi, *et al*, arXiv 1011,0050.
[28] Chi-Cheng Lee, Wei-Guo Yin, and Wei Ku, Phys. Rev. Lett. 103, 267001 (2009)
[29] A.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, R.D. McDonald, Phys. Rev. Lett. 101, 216402

(2008).
[30] Ying Ran, Fa Wang, Hui Zhai, Ashvin Vishwanath, Dung-Hai Lee, Phys. Rev. B 79, 014505 (2009)
[31] J.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).
[32] C. 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).