Quasiparticle interference in antiferromagnetic parent compounds of iron-based superconductors

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Recently reported quasiparticle interference imaging in underdoped $Ca(Fe_{1-x}Co_x)_2As_2$ shows pronounced C_2 asymmetry, which is interpreted as an indication of an electronic nematic phase with a unidirectional electron band, dispersive predominantly along the b axis of this orthorhombic material. On the other hand, even more recent transport measurements on untwinned samples show near isotropy of the resistivity in the ab plane, with slightly larger conductivity along a (and not b). We show that, in fact, both sets of data are consistent with the calculated *ab initio* Fermi surfaces, which have a decisively broken C_4 and yet similar Fermi velocity in both directions. This reconciles completely the apparent contradiction between the conclusions of the scanning tunneling microscopy and the transport experiments.

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The Fe-based superconductors present a new paradigm for high-T_C superconductivity, as Cooper pairs appear to emerge upon chemical doping from a metallic ground state rather than from a Mott insulator as found in the celebrated high-T_C cuprates.[1] Despite this difference of parent ground state between the Fe- and Cu-based superconductors, similarities lie in the fact that, in both cases, superconductivity emerges after the suppression of static ordered magnetism.[2] Although band theory has correctly predicted the unusual antiferromagnetic (AFM) order in the parent compounds of the Fe-based superconductors, it consistently overestimates the tendency to magnetism and underestimates the electronic mass. Therefore, there is no doubt that electronic interactions can not be ignored in quantitative descriptions, and that they play a different role compared to cuprates. The exact role of correlations, especially once the parent phase of the Fe superconductors is doped, has been the focus of much debate and controversy.

An almost universal feature of the Fe superconductors is that, in the parent phases, there is a tetragonal-to-orthorhombic structural phase transition that is closely associated with the onset of antiferromagnetic order.[3] Upon chemical doping x, the onset of the structural and magnetic transitions (T_S and T_N, respectively) decrease with x and superconductivity emerges. The physical nature of the crossover from antiferromagnetic order to superconductivity varies between specific materials. In some cases, both T_S and T_N coincide, while, in others, T_S is a few degrees higher than T_N.[3]

Band-structure calculations have indicated that the AFM ordering is accompanied by a strong restructuring of the Fermi surface, with the Fermi surface area being reduced by roughly an order of magnitude. This has been confirmed by optical and Hall measurements, which register a drastic reduction of the carrier concentration in the AFM state.[4–6] The calculated AFM Fermi surface consists of several small pockets, which are arranged in the Brillouin zone in a way that strongly breaks the tetragonal symmetry, but each of them is rather isotropic.[7] This led to a prediction of small transport anisotropy. An alternative point of view, which associates the orthorhombic transition with orbital (charge) degrees of freedom, suggests a double-exchange (metallic) ferromagnetic interaction along one crystallographic direction and a super exchange along the other direction. This picture is also consistent with the observed AFM order and naturally suggests a metallic conductivity along the ferromagnetic chains and a substantially reduced conductivity in the other direction.

Recent experiments on detwinned single crystals support the former point of view: they demonstrate a small anisotropy with the AFM direction being *more*, not *less*, metallic. However, transport measurements are integrated probes, and also involve possibly anisotropic scattering rate. Therefore, experiments that directly probe the topology of the Fermi surface in the AFM state are highly desirable.

One such experiment has been performed recently by Chuang *et al.*[8] They have reported quasiparticle interference (QPI) imaging of a lightly cobalt-doped sample of a $CaFe_2As_2$ compound. They interpreted their result in terms of a quasi-one-dimensional (unidirectional) electronic structure, metallic only along the FM, which is consistent with the above-mentioned orbital picture. On the the other hand, their argumentation was rather indirect, based largely on the fact that directly measured dispersion of the QPI maxima (which was indeed one dimensional) coincided with the angle-resolved photoemission spectroscopy (ARPES) measured band dispersion along the the same direction.

In this paper, we show that, in reality, the data of Ref. 8 are consistent with the calculated *ab initio* Fermi surfaces, and not with the one-dimensional bands implied in that work. This reconciles completely the apparent contradiction between the conclusions of Ref. 8 and the transport measurements on untwinned samples.

The reported scanning tunneling microscopy (STM) examination shows a QPI pattern in the momentum space that completely breaks the C_4 symmetry, with the main features being two bright spots along the y (crystallographic b) direction, with no counterparts along the x direction (note that y is the *ferromagnetic* direction, and x is the *antiferromagnetic* one). Reference 8 insists "that the scattering interference modulations are strongly unidirectional, which should occur if the k-space band supporting them is nematic." However, it should be kept in mind that this occurs in that part of the phase diagram where the long-range antiferromagnetic order is fully established, as reflected by the fact that the lattice symmetry is orthorhombic, and that the C_2 symmetry is already completely broken. Indeed, the size of the orthorhombic distortion is

not "minute," as Ref. 8 posits, with $b/a \sim 1\%$, and is instead comparable with distortions seen in various iron-oxides systems. For instance, in the Verwey transition, the Fe–O bond dilation is ~0.6% with Fe atoms in the same tetrahedral symmetry as in the ferropnictide superconductors,[9] and this is usually considered to be a strong distortion. Similarly, in the antiferromagnetic phase of FeO, where the cubic symmetry is completely broken, the structural effect is also of the same order.[10]

Since the sample under study is orthorhombic, it is misleading to call its electronic structure nematic, as the lattice orthorhombic distortion here is substantial. Nematic phases are frequently found in organic matter. The defining characteristic of these phases is orientational order in the absence of long-range positional order, resulting in distinctive uniaxial physical properties. It has also been proposed that nematic order exists in some electronic systems, and may even play a role in mediating high-temperature superconductivity.[11] Borzi *et al.*[12] demonstrated the presence of another interesting phase in $Sr_3Ru_2O_7$ at millikelvin temperatures and high magnetic fields, which has also been called nematic. In this case, the crystallographic planes were shown to remain strictly tetragonal (within 0.01%) with C_4 structural symmetry, while a pronounced C_2 asymmetry in electronic properties was measured. This breaking of the electronic symmetry compared to that of the underlying lattice is now conventionally referred to as electronic nematicity. (In fact, even in those cases, one has to be careful to distinguish between nematic physics and simply an unusually weak electron-lattice coupling, but this goes beyond the scope of this paper and, in any event, is not a concern for Fe pnictides where this coupling is strong.)

Since the tetragonal symmetry is decisively broken at the onset of the magnetic order in this ferropnictide, it is clear that the symmetry of the electronic structure defining the structural distortion is also completely broken. What is more important is that, while the observed QPI pattern does violate the C_4 symmetry, it is clearly not one dimensional, in the sense that it varies equally strongly along the k_x and k_y directions. Thus, interpretation of the data in terms of a unidirectional electron band does not appear to be possible. To understand this experiment, one needs to start with a realistic model for the electronic structure and actually calculate the QPI pattern.

Such a calculation has recently been presented by Knolle *et al.*[13] They used a weak-coupling theory, which interprets the antiferromagnetic state as resulting from a spin-Peierls transition, with a correspondingly small magnetic moment. Knolle *et al.* have been able to describe qualitatively the experimental data obtained by Chuang *et al.* in the sense that their calculated QPI pattern strongly breaks the C_2 symmetry, while the band dispersion, on average, remains fairly isotropic in plane. Note that one should not be looking for a *quantitative* interpretation, since the STM experiment in question did not detect any Ca atoms on the surface; so, the sample surface is likely charged with up to 0.5 hole per Fe, and thus any bulk calculation can only be applied to this experiment in a qualitative way. Besides, it was recently shown[14] that Fe pnictide systems feature surface states quite different from the bulk states, which should undoubtedly affect the STM spectra.

However, this result, as mentioned, has been obtained in a weak-coupling limit, corresponding to small magnetization, while in this system, the ordered magnetic moments are on the order of 1 μ_B, and local moments are even larger.[15–17] Not surprisingly, their Fermi surface is rather far from that measured recently on untwinned samples by Wang *et al.*[18] while the local density approximation (LDA) Fermi surface reproduces it quite well.[19] Indeed, this is a known problem in the weak-coupling approach: While being physically justified for the paramagnetic parts of the phase diagram, the Fe magnetism in the ordered phases is driven by the strong local Hund rule coupling, and not by the Fermi surface nesting, as assumed in the weak-coupling models.

Therefore, we have calculated the QPI images for antiferromagnetic $CaFe_2As_2$ entirely from first principles,[19] using the LDA magnetic moment (somewhat larger than the experimental moment at zero doping). We used the standard linear augmented plane-wave method as implemented in the WIEN2K code.[20] The corresponding Fermi surface is shown in Fig. 1. We see that the magnetism has a drastic effect on the fermiology, and the resulting Fermi surfaces are completely breaking the C_4 symmetry. Apart from small quasi-two-dimensional tubular pockets, originating from Dirac cones, there is one hole pocket around Z $(0,0,\pi/c$ or $2\pi/a,0,0)$ and two electron pockets between Z and $0,\pi/b,\pi/c$. It is immediately obvious that the QP scattering between these pockets must exhibit strong interference for scattering along b, but not along a.

Indeed, we have calculated the QPI function Z, using the known expression [Ref. 21, Eq. (S9)]

$$|Z(\mathbf{q},E')|^2 \propto \int \frac{dE'}{E-E'} \sum_{\mathbf{k}} \delta(E-E_{\mathbf{k}})\delta(E'-E_{\mathbf{k}+\mathbf{q}}), \quad (1)$$

where we assumed a constant impurity scattering rate and a constant tunneling matrix element. This approximation is sufficient for a qualitative or semi-quantitative comparison. As explained, given that the surface in the experiment in question was charged compared to the bulk, a quantitative comparison is meaningless.

A calculated pattern (there is some dependence on q_z and on E, but we are interested in the qualitative features only) are shown in Fig. 2. One can see immediately that, very similar to the patterns obtained in Ref. 8, two sharp maxima appear

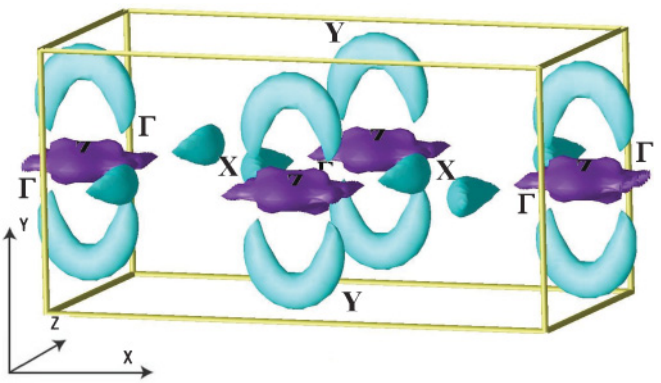

FIG. 1. (Color online) Calculated LDA Fermi surface for $CaFe_2As_2$ in the antiferromagnetic state.

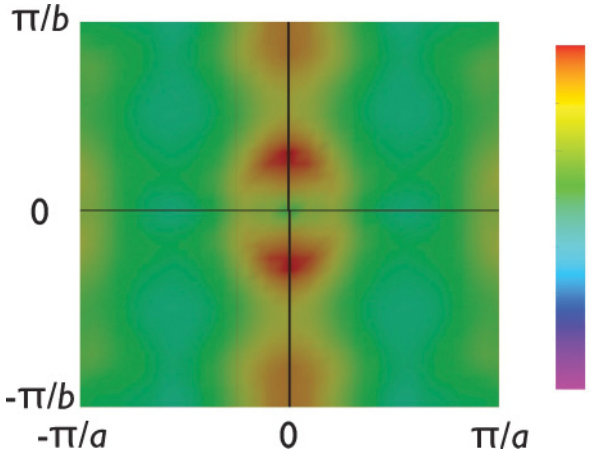

FIG. 2. (Color online) Quasiparticle interference pattern (in arbitrary units) for zero bias and $q_z \sim 0$, calculated using the same electronic structure as in Fig. 1 and Eq. (1). The color coding, shown on the right, goes linearly from zero to the maximum value.

at $\mathbf{q} = 0, \pm\xi, 0$, where $\xi \sim \pi/4b$. The origin of these QPI features is obvious from the Fermi surface (Fig. 1). Note that these LDA calculations have no adjustable parameters, and yet are in excellent qualitative agreement with the QPI images.

It is also worth noting that, while the calculated Fermi surfaces completely break the tetragonal symmetry, which is fully reflected in the QPI images, the individual pockets are very three dimensional, so that the calculated conductivity is comparable for all three directions.[7] While, experimentally, there is up to a 20% a/b charge transport anisotropy[7] close to tetragonal-to-orthorhombic phase boundary in $CaFe_2As_2$, it is much less than what would be predicted for a quasi-one-dimensional electronic band, and of the opposite sign.[22]

It may be worthwhile at this point to explain at some length why a quantitative comparison between a Fourier transform of a tunneling current map, and theoretical calculations (whether ours or any other) is impossible at this stage. Quasiparticle interference, as discussed in many papers, manifests itself in tunneling in a very indirect way. In a sense, it is a multistage process. First, a defect existing near the metal surface is screened by the conducting electrons. This creates Friedel oscillations in the real space. This oscillations are formed by all electrons (mostly those near the Fermi surface, but not only those). In a multiband system, it includes electrons originating from different atomic orbitals, such as xy, xz, yz, z^2, and $x^2 - y^2$. As is well known in the theory of tunneling, the rate at which electrons tunnel through a vacuum depends drastically on their orbital symmetry, especially on their parity (see, e.g., Ref. 23). Indeed, tunneling through a wide barrier mainly proceeds through electrons with zero-momentum projection onto the interface plane (such electrons have to travel the shortest lengths in the sub-barrier regime). If such electrons belong to an odd two-dimensional representation (for d electrons, all but z^2, if z is the normal direction), the tunneling rate is suppressed. This effect is well known in spintronics, where it can drastically change the current spin polarization. On the other hand, for a thin barrier, the tunneling conductance depends on the number of the conductivity channels, which is given by the density of states (DOS) times normal velocity. In both cases, it is not just the density of quasiparticles, as assumed in Eq. (1) (and in Ref. 13), but the DOS weighed by a strongly **k**-dependent, unknown function.

Nothing is known about the nature of the scattering centers, which produce the above-mentioned Friedel oscillations. In this particular experiment, they may be magnetic or non-magnetic defects, twin domain boundaries, antiphase domain boundaries, remaining surface Ca ions, and more. Some of these scatterers are strongly anisotropic by nature, while others are strongly dependent on the orbital character. We have dropped the scattering matrix elements completely form our consideration. Knolle *et al.*[13] instead have chosen a specific model for the scattering centers. We believe that, without any knowledge about the actual scattering centers in the system, any QPI using a particular model obscures the actual physics, compared to the simplest constant matrix elements approximation, rather than clarifies it.

Finally, there are several issues specific to this particular experiment, such as unknown, but strongly different from the bulk, charge state. As opposed to Ba122, and Sr122, where 1/2 of the alkaline earth atoms stay on the surface, providing charge neutrality, in Ca122, STM does not detect any Ca on the surface, suggesting a strongly charged surface. A corollary of that is the appearance of a surface reconstruction (as indeed observed), a surface relaxation, and, importantly (since tunneling proceeds largely through the surface states), surface bands (as demonstrated, for instance, in Ref. 24.

While these considerations preclude a quantitative comparison and an extraction of quantitative analysis of the experiment in question, we see, particularly when comparing our calculations with those of Knolle *et al.*,[13] that the C_2 QPI structure observed in Ref. 8 is a very universal consequence of the long-range stripe-type antiferromagnetic ordering. Indeed, the calculations by Knolle *et al.* were built upon a basically incorrect band structure and Fermi surfaces, and used a weak-coupling nesting scenario for the antiferromagnetism; in reality, the magnetism in pnictides is a strong coupling phenomenon; yet, their calculations produced a "unidirectional" QPI pattern just as well. Together with the strong-coupling LDA calculations, this spans a large range of possible models, indicating that the C_4 symmetry is strongly broken in QPI images simply by virtue of the long-range AFM order, regardless of the origin of this order.

Last, but not least, we can also predict from our calculations that this symmetry will be also broken, although the peaks are likely to be substantially broader, in the truly *nematic* phase (see Ref. 15 for a discussion), that is to say, the phase between the long-range magnetic transition and the structural orthorhombic transition.

[1] P. A. Lee, N. Nagaosa, and X.-G. Wen, Rev. Mod. Phys. **78**, 17 (2006).
[2] I. I. Mazin, Nature (London) **464**, 183 (2010).
[3] D. C. Johnston, Adv. in Phys. **59**, 803 (2010).
[4] W. Z. Hu, J. Dong, G. Li, Z. Li, P. Zheng, G. F. Chen, J. L. Luo, and N. L. Wang, Phys. Rev. Lett. **101**, 257005 (2008).

[5]L. Fang, H. Luo, P. Cheng, Z. Wang, Y. Jia, G. Mu, B. Shen, I. I. Mazin, L. Shan, C. Ren, and H.-H. Wen, Phys. Rev. B **80**, 140508(R) (2009).
[6]F. Rullier-Albenque, D. Colson, A. Forget, and H. Alloul, Phys. Rev. Lett. **103**, 057001 (2009).
[7]M. A. Tanatar *et al.*, Phys. Rev. B **81**, 184508 (2010).
[8]T.-M. Chuang, M. P. Allan, J. Lee, Y. Xie, N. Ni, S. L. Bud'ko, G. S. Boebinger, and P. C. Canfield, J. C. Davis, Science , **327**, 181 (2010).
[9]J. P. Wright, J. P. Attfield, and P. G. Radaelli, Phys. Rev. Lett. **87**, 266401 (2001).
[10]D. G. Isaak, R. E. Cohen, M. J. Mehl, and D. J. Singh, Phys. Rev. B **47**, 7720 (1993).
[11]S. A. Kivelson, E. Fradkin, and V. J. Emery, Nature (London) **393**, 550 (1998).
[12]R. A. Borzi, S. A. Grigera, J. Farrell, R. S. Perry, S. J. S. Lister, S. L. Lee, D. A. Tennant, Y. Maeno, and A. P. Mackenzie, Science **315**, 214 (2006).
[13]J. Knolle, I. Eremin, A. Akbari, and R. Moessner, Phys. Rev. Lett. **104**, 257001 (2010).
[14]E. van Heumen, J. Vuorinen, K. Koepernik, F. Massee, Y. Huang, M. Shi, J. Klei, J. Goedkoop, M. Lindroos, J. van den Brink, and M. S. Golden, e-print arXiv:1009.3493 (unpublished).
[15]I. I. Mazin and J. Schmalian, Phys. C (Amsterdam) **469**, 614 (2009).
[16]M. D. Johannes, I. I. Mazin, and D. S. Parker, Phys. Rev. B **82**, 024527 (2010).
[17]M. D. Johannes and I. I. Mazin, Phys. Rev. B **79**, 220510(R) (2009).
[18]Q. Wang *et al.*, e-print arXiv:1009.0271 (unpublished).
[19]Compared to Ref. 18, both $\beta 1$ and $\beta 2$ bands are present, their nontrivial crescent shape is reproduced, and their size and location (around the Z point) are consistent with the calculation. Note that these β pockets are mainly responsible for the QPI peak in our Fig. 2. The flattish γ pocket is also in excellent agreement with the calculation, although in the experiment it is split into $\gamma 3$ and $\gamma 4$ (probably an effect of the surface reconstruction). The claimed experimental bands ($\alpha 1, \alpha 2, \gamma 1$, and $\gamma 2$) along Z-X are quite messy. The calculations predict small pockets, located roughly where ARPES sees some bands. These are formed by the famous "Dirac cones." The only feature that does not find any correspondence in the calculation is the long segment "$\gamma 2$"stretched along k_y. This may be a surface state similar to those discovered in Ref. 14 (note that this band is drawn rather speculatively, since the corresponding signal is really weak). It is worth noting that the Fermi surface shown in Fig. 1 was first published in Ref. 7, well before any untwinned ARPES data became available.
[20]P. Blaha *et al.* computer code WIEN2K, Technische Universit at Wien, Austria, 2001.
[21]T. Hanaguri *et al.* Science **323**, 923 (2009).
[22]Measurement of charge transport in Co-doped $BaFe_2As_2$ reported by J.- H. Chu, J. G. Analytis, K. De Greve, P. L. McMahon, Z. Islam, Y. Yamamoto, and I. R. Fisher, Science **329**, 824 (2010), show a larger a/b anisotropy close to the orthorhombic transition, which we believe not to be representative of the $CaFe_2As_2$ upon which the STM measurements of Ref. 8 were taken. Regardless, the values of a factor of 2 anisotropy for doped samples in that study still remain too low to be associated with the large anisotropies expected from a unidirectional band structure suggested in Ref. 8, and are of the opposite sign.
[23]I. I. Mazin, Europhys. Lett., **55**, 404, (2001).
[24]E. van Heumen *et al.* (unpublished).