Strong electronic correlations and Fermi surface reconstruction in the quasi-one dimensional iron superconductor \( \text{BaFe}_2\text{S}_3 \)

J.M. Pizarro\(^1\) and E. Bascones\(^1,\,*\)

\(^1\)Materials Science Factory. Instituto de Ciencia de Materiales de Madrid, ICMM-CSIC, Cantoblanco, E-28049 Madrid (Spain).

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\( \text{BaFe}_2\text{S}_3 \) is a special iron superconductor with two-leg ladder structure which can help to unravel the role played by the electronic correlations in high-T\(_c\) superconductivity. At zero pressure it is insulating with stripe antiferromagnetic (AF) order and superconductivity emerges under pressure. We use a slave-spin technique to analyze the strength of the local correlations in \( \text{BaFe}_2\text{S}_3 \). We find that at the pressure at which the superconductivity appears the electronic correlations in \( \text{BaFe}_2\text{S}_3 \) are similar to the ones measured in other iron superconductors. At zero pressure the strength of the correlations is strongly enhanced being particularly severe for the two orbitals with the largest weight at the Fermi level what invalidates nesting as the mechanism for AF. At zero temperature the system is not a Mott insulator, but these two orbitals with mass enhancements \( m^* \sim 12 - 15 \) will become incoherent at higher temperatures. Different from what happens in other iron superconductors, at both pressures, the Fermi surface is reconstructed by the electronic correlations.

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Ten years after the discovery of high-T\(_c\) superconductivity in iron materials its origin is not understood\(^1\). AF interactions are believed to play a key role in the emergence of superconductivity. Whether the magnetism is due to a Fermi surface instability\(^2\) of itinerant carriers, to exchange interactions between localized electrons \(^3\) or to double exchange physics\(^4\) is controversial\(^5\).

Well studied iron superconductors have a planar square lattice. Recently superconductivity has been found in the two-leg ladder 123-compounds \( \text{BaFe}_2\text{S}_3 \) and \( \text{BaFe}_2\text{Se}_3 \) up to 24 K and 11 K respectively\(^6–8\). As in planar iron superconductors, in these compounds Fe is tetrahedrally coordinated, but every third Fe row in the layer is missing, see Fig.1. The nominal occupation of Fe, 6 electrons in 5 orbitals, equals the one in undoped iron superconductors. However these ladder materials are AF insulators at zero pressure and superconductivity emerges under pressure. In \( \text{BaFe}_2\text{Se}_3 \) the ladders are tilted and the iron atomic distances distort as a plaquette AF ordering with \( \mu = 2.8\mu_B \) emerges\(^9–11\). Pressure suppresses the tilting of the ladders, induces a metal-insulator transition and probably a change to AF stripe order before superconductivity appears\(^8, 12, 13\).

\( \text{BaFe}_2\text{S}_3 \) shows stripe order, AF along the ladder and ferromagnetic along the rung, with Neel temperature and magnetic moment \( T_N \sim 120 \text{ K} \) and \( \mu \sim 1 - 1.2\mu_B \), close to the ones of other iron superconductors\(^3, 7, 14, 15\). Insulating character is observed above and below \( T_N \). A resistivity anomaly around 180 K could indicate orbital order\(^7, 15\). Metallicity and superconductivity appear at \( P \sim 11 \text{ GPa} \). Ab-initio calculations reproduce the AF order but with larger moment \( \mu \sim 2.1\mu_B \) and nesting arguments have been used to explain intraladder AF\(^18\). Other authors have suggested that \( \text{BaFe}_2\text{S}_3 \) is a Mott insulator in which pressure suppress the Mott gap and induce superconductivity\(^6, 7\). However, the activation gap is only 70 meV and the magnetic moment is much smaller than expected from saturated spins \( \mu = 4.0\mu_B \)\(^14\). Photoemission and X-ray experiments suggest the presence of local and itinerant electrons\(^19, 20\).

In this paper we use a slave spin technique to study the strength of electronic correlations in \( \text{BaFe}_2\text{S}_3 \) and shed light on the nature of the superconducting and AF instabilities. We find that at pressures for which superconductivity appears the electronic correlations are similar to those found in other iron superconductors. However
at zero pressure the correlations are much stronger, especially for the orbitals with the largest weight at the Fermi level. Such strong correlations invalidate nesting as the origin of AF in this material with local and itinerant electrons. We also show that electronic correlations reconstruct the Fermi surface at both pressures.

Model and methods We consider a multi-orbital model for Fe atoms. Interactions, restricted to orbitals in the same atom, include intraorbital $U$, interorbital $U'$, Hund’s coupling $J_H$, and pair hopping $J'$ terms, see the supplementary information (SI) and [5]. We start from the tight-binding models for BaFe$_2$S$_3$ derived from ab-initio calculations at pressures $P = 0$ and $P = 12.4$ GPa[13]. They include 20 orbitals, 5 per each of the four iron atoms in the unit cell. Orbital and unit cell axis differ among them and from the ones commonly used in iron superconductors. X and Y axis connect ladders in adjacent planes and Z runs along the ladders, see Fig.1. The orbitals $xz$, $yz$, $xy$ and $3z^2−r^2$, $x^2−y^2$ are defined with $z$ along the ladders, $x$ connecting ladders in the same Fe plane and $y$ axis perpendicular to the Fe-ladders plane. In this basis there are finite onsite non-diagonal terms among these orbitals. We change to a basis with on-site diagonal terms only $w_{\alpha}=w_{xz}$, $w_{yz}$, $w_{xy}$, $w_{3z^2−r^2}$, $w_{x^2−y^2}$. Here the subscript $\alpha$ indicates which orbital in the original basis has the largest weight, see SI.

The interaction terms are defined in the $w_{\alpha}$ basis. We take atomic filling $n=6$, $U' = U − 2J_H$, $J' = J_H$, assume $J_H = 0.25U$ and study the electronic correlations as a function of $U$[21]. We quantify the strength of the local correlations by the orbital dependent quasiparticle weight $Z_{\gamma}$ calculated with the U(1) slave-spin technique[22]. $Z_{\gamma} = 1$ in non-correlated materials and decreases as the electronic correlations increase. It has the same value in the four Fe atoms of the unit cell. In the approximation used, $Z_\gamma$ equals the inverse of the mass enhancement factor of each orbital $m_\gamma^*$. Using constraint RPA the strength of the interactions in BaFe$_2$S$_3$ was estimated to be similar to that in LiFeAs, i.e. larger than the interaction in BaFe$_2$As$_2$ and smaller than in FeSe, and to be reduced a 6% under a pressure of 12.4 GPa[18,23]. Slave-spin calculations for BaFe$_2$As$_2$ and FeSe using $J_H = 0.25U$ compare well with experiment if the interactions $U_{BaFe_2As_2} = 2.7$ eV and $U_{FeSe} = 3.0$ eV are used[24,25]. Therefore to study the electronic correlations in BaFe$_2$S$_3$ we take $U_{P=0} = 2.90$ eV for $P = 0$ and $U_{P=12.4} = 2.75$ eV for $P = 12.4$ GPa.

Electronic correlations Fig.2 shows the quasiparticle weight $Z_{\gamma}$ as a function of $U$. At $P = 0$, $Z_{\gamma}$ is strongly suppressed at $U^* \sim 2.1$ eV. Beyond this crossover the system enters into a correlated state, the Hund’s metal, with well formed local spins satisfying Hund’s rule[3,22,26-30]. Two of the orbitals, $w_{xz}$ and $w_{yz}$, become very strongly correlated, $Z_{\gamma} < 0.1$, while the other three orbitals $w_{xy}$, $w_{3z^2−r^2}$ and $w_{x^2−y^2}$ show intermediate correlations $Z_{\gamma} \sim 0.3–0.6$. The suppression of the quasiparticle weight $Z_{\gamma}$ is concomitant with a reorganization of the orbital filling $n_{\gamma}$, see inset in Fig.2(a). The strongly correlated orbitals $w_{xz}$ and $w_{yz}$ approach half-filling in the Hund’s metal state. The interaction at which the Hund’s metal crossover appears in Fig.2(a) is reduced with respect to the one found in other iron superconductors $U^* \sim 2.6–2.7$ eV [22,24,25]. Behind this reduction it is the smaller value of the bandwidth: 4 eV in BaFe$_2$S$_3$, see[18] and SI, and close to 5 eV in other compounds.

BaFe$_2$S$_3$ with $U_{P=0} = 2.90$ eV is a strongly correlated Hund metal. At this interaction, the electronic correlations reduce the bandwidth to less than 2 eV, see SI. The narrowing of the bands is especially prominent close to the Fermi level. This happens because the bands close to
the Fermi level are dominated by $w_{yz}$ and $w_{zx}$ orbitals with a very small $Z_w \sim 0.06$, i.e. large mass enhancements $m^*_w \sim 12 - 15$ are expected. In the other three orbitals the strength of correlations and the mass enhancements are intermediate $Z_m \sim 0.3 - 0.5$ and $m^*_m \sim 2 - 3.5$.

At zero pressure BaFe$_2$S$_3$ is not a Mott insulator. Several bands cross the Fermi level and reconstruct the Fermi surface. However, due to the small quasiparticle weight, if the temperature is not very low the electrons

**Summary and discussion** We find that at zero pressure BaFe$_2$S$_3$ is a strongly correlated Hund metal with orbital selective correlations. The two orbitals with the largest weight at the Fermi level have very small quasiparticle weight $Z_{w_{zx},w_{yz}} \sim 0.06$, i.e. large mass enhancements $m^*_{w_{zx},w_{yz}} \sim 12 - 15$ are expected. In the other three orbitals the strength of correlations and the mass enhancements are intermediate $Z_m \sim 0.3 - 0.5$ and $m^*_m \sim 2 - 3.5$.
FIG. 4: (Color online) Reconstruction of the Fermi surface due to the electronic correlations at $P = 0$ GPa and $P = 12.4$ GPa. On spite of the quasi-one dimensional lattice the Fermi surface has three dimensional character. When correlations are included enhanced scattering at $Q \sim (0,0,2\pi)$ ($Q^* \sim (0,0,\pi)$ if the Brillouin zone is unfolded along $k_z$) between hole pockets along $(k_x,-k_x,\pm\pi)$ is expected.

close the Fermi level will behave as incoherent particles: the quasiparticle peak is expected to be absent in photoemission experiments and the resistivity could show insulating behavior as observed experimentally. Short range AF correlations above the Neel temperature will enhance the insulating tendencies localizing these very weakly coherent electrons. In agreement with photoemission and X-ray experiments the system can be described in terms of itinerant and localized electrons [19, 20].

The reconstructed Fermi surface shows enhanced scattering at $Q \sim (0,0,2\pi)$ between the hole pockets. In a Brillouin zone unfolded along $k_z$, $Q$ would become $Q^* \sim (0,0,\pi)$ coincident with the intra-ladder AF moment. These pockets are very shallow and therefore very sensitive to other effects not included in this work. But overall the small quasiparticle weight prevents any explanation of the antiferromagnetic state in terms of nesting. This can have important consequences for the understanding of the superconducting phase which it is believed to be mediated by the AF correlations. Whether the AF order in this material is better explained by a double exchange mechanism or with local moments is an interesting issue beyond the scope of this work.

Interestingly, at $P = 12.4$ GPa, at which the superconductivity is found, the strength of the correlations is reduced to values similar to the ones measured in other iron superconductors. Intermediate correlations could be beneficial for high-Tc temperature superconductivity in iron superconductors [32].

As in the $P = 0$ case, at $P = 12.4$ GPa the reconstructed Fermi surface shows enhanced scattering at $Q \sim (0,0,2\pi)$, $Q^* \sim (0,0,\pi)$ in the unfolded Brillouin zone. At this pressures the Fermi pockets are not so shallow and the larger quasiparticle weight could, in principle, justify an explanation of the superconductivity in terms of the processes at the Fermi surface, or at least some contribution to it. Such mechanism would predict an order parameter with sign change between the two hole-pockets. An interesting issue which requires further work is whether this weak coupling mechanism for superconductivity is invalidated by the non-nesting character of the AF phase suppressed by pressure or not.

In conclusion our work shows that the quasi-one dimensional 123 family can be very useful to decipher the nature of high-Tc superconductivity in iron based materials. Further experimental and theoretical work is desired.
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* Electronic address: leni.bascones@icmm.csic.es

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SUPPLEMENTARY INFORMATION

Methods

We start from a multi-orbital model with local interactions including: intraorbital $U$, interorbital $U'$, Hund’s coupling $J_H$, and pair hopping $J'$ terms,

$$H = \sum_{k,\gamma,\beta,\sigma} \epsilon_{k,\gamma,\beta} c_{k,\gamma,\sigma}^\dagger c_{k,\gamma,\sigma} + \text{h.c.} + \sum_{j,\gamma,\sigma} \epsilon_{\gamma} n_{j,\gamma,\sigma}$$

$$+ U \sum_{j,\gamma} n_{j,\gamma,\uparrow} n_{j,\gamma,\downarrow} + \left( U' - \frac{J_H}{2} \right) \sum_{j,\{\gamma>\beta\}} \sum_{\sigma,\tilde{\sigma}} \epsilon_{\gamma} n_{j,\gamma,\sigma} n_{j,\gamma,\tilde{\sigma}}$$

$$-2J_H \sum_{j,\{\gamma>\beta\}} \tilde{S}_{j,\gamma} \tilde{S}_{j,\beta} + J' \sum_{j,\{\gamma\neq\beta\}} \sum_{\sigma,\tilde{\sigma}} \epsilon_{j,\gamma,\sigma}^4 c_{j,\gamma,\sigma}^\dagger c_{j,\beta,\tilde{\sigma}} c_{j,\beta,\tilde{\sigma}} c_{j,\gamma,\sigma}$$

where $j$ label the Fe atoms. Each unit cell contains 4 atoms. X and Y axis connect ladders in adjacent planes and Z runs along the ladders. $k$ is the momentum in the 4 Fe Brillouin zone, $\sigma$ the spin and $\gamma$, and $\beta$ the orbitals. We include five orbitals per Fe atom. There are 20 orbitals in the unit-cell. Curly brackets $\{\}$ in the sum subscript indicate that the sum is restricted to orbitals in the same atom, i.e., both orbitals are between 1 and 5 or between 6 and 10, and so on. The model is defined in $w_\alpha$ orbital basis. In this basis there are no hybridization terms between different orbitals within the same atom, only a diagonal on-site contribution $\epsilon_{\gamma}$. We assume $U' = U - 2J_H$ \[21\] and $J' = J_H$, as in rotationally invariant systems, leaving only two independent interaction parameters, $U$ and $J_H$. We take $J_H = 0.25U$.

To obtain $\epsilon_{k,\gamma,\beta}$ and $\epsilon_{\gamma}$ we start from the 20-orbital tight-binding models calculated in \[18\] from a Wannier projection of ab-initio results for BaFe$_2$Sc$_3$ at $P = 0$ and $P = 12.4$ GPA. These models are obtained using as orbital basis: $zx, yz, x^2 - y^2, 3z^2 - r^2$ and $xy$ defined with $z$ along the ladders, $x$ connecting ladders in the same Fe plane and $y$ axis perpendicular to Fe-ladders plane. $zx$ and $xy$ along these axis are respectively equivalent to $xy$ and $zz$ in the basis frequently used in tight-binding models for iron superconductors (with $x$ and $y$ along Fe bonds and $z$ perpendicular to the Fe plane), i.e. the two orbitals are exchanged; $yz$ is equivalent in both basis and $x^2 - y^2$ and $3z^2 - r^2$ do not have direct analogues. They are linear combinations of the orbitals defined with the axis exchanged.

In this basis there are inter-orbital on-site terms. We perform a change to a basis $w_\alpha$ in which the on-site terms of these tight-binding models are diagonal. The subscript $\alpha$ corresponds to the orbital of the original basis which gives a larger contribution to $w_\alpha$. The matrix elements of the change of basis have the same absolute value in the four atoms, but their sign can differ. Their value depend on pressure. An example is given below. For $P = 0$:

$$w_1^{12z-r^2} = 0.95|3z^2 - r^2 > + 0.16|yz > - 0.25|x^2 - y^2 >$$

$$w_3^{3z-r^2} = 0.95|3z^2 - r^2 > - 0.16|yz > - 0.25|x^2 - y^2 >$$

where the superscript label the atom in the unit cell. For $P = 12.4$ GPA

$$w_1^{12z-r^2} = 0.92|3z^2 - r^2 > + 0.22|yz > - 0.33|x^2 - y^2 >$$

$$w_3^{3z-r^2} = 0.92|3z^2 - r^2 > - 0.22|yz > - 0.33|x^2 - y^2 >$$

We then write the onsite and hopping terms of the tight-binding models are the basis $w_\alpha$. To address the role of the electronic correlations we use $U(1)$ slave spin theory \[22\] and keep only density-density terms. That is, pair hopping and spin-flip terms do not enter into the calculation. With this technique we calculate the quasiparticle weight and the onsite energy shifts which are generated by the electronic correlations.

Bands and Fermi surface reconstruction

Below we provide some figures, complementary to Fig[3] and Fig[4] which show the effect of the electronic correlations in the band structure and in the Fermi surface at $P = 0$ and $P = 12.4$ GPA.
FIG. S1: (Color online) Band structure for $P = 0$ and $P = 12.4$ GPa with and without including the electronic correlations. The bandstructure in (a) and (c) for $P = 0$ and $P = 12.4$ GPa is calculated using the tight-binding model from [18] and mimics the one obtained in ab-initio calculations, see text. The bandwidth is approximately 4 eV and 5 eV respectively for $P = 0$ and $P = 12.4$ GPa. As shown in (c) and (d) the electronic correlations reduce these bandwidth below 2 eV and 3.5 eV, respectively. Band narrowing is more evident close to the Fermi level.
FIG. S2: (Color online) Fermi surface cuts for $P = 0$ and $P = 12.4$ GPa with and without including the electronic correlations plane, along $k_z - k_x = k_y$ reciprocal to the Fe-ladder plane. At both pressures the Fermi surface is reconstructed due to electronic correlations.

FIG. S3: (Color online) Fermi surface cuts for $P = 0$ and $P = 12.4$ GPa with and without including the electronic correlations plane, along $k_z - k_x = k_y$ plane, reciprocal to the plane perpendicular to the Fe-ladder plane.