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Sr₂VO₃FeAs: A nanolayered bimetallic iron pnictide superconductor

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Abstract – One of the unifying concepts in the iron-pnictide superconductors, both for the mechanism of magnetic ordering and of unconventional order parameter character, has been the electron and hole Fermi surfaces that are approximately nested. Using the density functional methods that have predicted Fermi surfaces correctly in SrFe₂P₂, we find that the recently reported superconducting Sr₂VO₃FeAs, with $T_c = 37$ K and no apparent competition between magnetism and superconductivity, possesses different Fermi surface geometry and character than previous classes of iron pnictides. The intervening layer (a V bilayer) gives rise to bands that cross the Fermi level. Coupling to the FeAs layer is small except for interaction along the zone boundary, however that coupling degrades the Fermi surface nesting. Sr₂VO₃FeAs, with its alternating layers of open-shell atoms, deserves further close study that should help to understand the origin of the properties of iron pnictide compounds.

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Introduction. – Since Hosono and coworkers found superconductivity with $T_c = 27$ K in iron pnictides [1], the superconducting critical temperature T_c has been rapidly increased to 56 K, causing excitement in the community second only to the period following the discovery of high- T_c cuprates. So far, five distinct crystal systems in this class have been uncovered: $\mathcal{R}\text{OFeAs}$ (called “1111”) [2,3], $\mathcal{M}\text{Fe}_2\text{As}_2$ (denoted “122”, $\mathcal{M} = \text{Ca}, \text{Sr}, \text{Ba}$) [4–9], $\mathcal{A}\text{FeAs}$ (“111”, $\mathcal{A} = \text{Li}, \text{Na}$) [10–12], FeSe_{1– x} [13–15], and “21311” Sr₂ $\mathcal{M}'\text{O}_3\text{FeAs}$ (also called “42622” since the unit cell contains two formula unit, $\mathcal{M}' = \text{Sc}, \text{V}$) [16–18]. The superconductivity is expected to be based, broadly speaking, on the iron coordination and bonding, the band filling, and some details that are not yet clear. Some known results are: 1) although there seems to be nothing special about the band filling in stoichiometric $\mathcal{R}\text{OFeAs}$, it requires a small amount of doping (either electron or hole) to suppress magnetism and allow superconductivity; 2) in the 122 class, pressure alone, without doping, drives the magnetic-to-superconducting change, and in addition the required pressure is modest (~ 5 GPa) by modern standards; 3) in the $\mathcal{A}\text{FeAs}$, small

\mathcal{A} deficiency results in superconductivity; 4) in FeSe_{1– x} either a small amount of Se vacancies or modest pressure without doping leads to a high- T_c superconductor.

In the 21311 system, Ogino *et al.* synthesized superconducting Sr₂ScO₃FeP with $T_c = 17$ K [16]. Xie *et al.* prepared isovalent and isostructural Sr₂ScO₃FeAs [17]. In contrast to the 1111 system, in which substitution of P by As leads to enhancing T_c by ~ 20 K, this compound is a normal metal, showing Curie-Weiss behavior. Very recently, Zhu *et al.*, who synthesized superconducting Sr₂VO₃FeAs with $T_c = 37$ K, inferred primarily electron-like charge carriers through the Hall coefficient measurements and suggested that the strong temperature dependence of this coefficient may arise from multiband character [18].

Most of these Fe pnictides show a clear competition between superconductivity and magnetism, and the assortment of observed phenomena is leading to a profusion of ideas about what comprises the essential characteristics of these superconductors [19–24]. One of the most widely discussed features is the nesting of two cylinder Fermi surfaces (FSs) separated by large wave vector Q , being relevant both to magnetic ordering and to superconducting order parameter symmetry. In Sr₂VO₃FeAs,

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no indication of magnetic order nor even strong temperature dependence of the susceptibility has been observed, setting it apart from most other Fe pnictide superconductors. The saturation of the resistivity in high T regime [18] may represent “bad metal” behavior but the large residual resistivity suggests sample imperfections should be kept in mind.

This $\text{Sr}_2\text{VO}_3\text{FeAs}$ member brings new, distinctive features into consideration in Fe-based superconductors. First, a bilayer of open-shell V ions (presumably close to d^2 formal configuration that would leave the FeAs layer with a formal -1 charge as in other classes) lies between the FeAs layers, with centers separated by lattice constant $c = 15.7 \text{ \AA}$, whereas other members have only closed-shell ions in this layer. Secondly, $\text{Sr}_2\text{VO}_3\text{FeAs}$ superconducts at high temperature (37 K) without requiring *either* explicit doping or pressure. In the various classes of Fe-based superconductors, the degree of two-dimensionality and its importance has been much discussed. In $\text{Sr}_2\text{VO}_3\text{FeAs}$, the FeAs layers are very weakly coupled through conducting V bilayers, although (we will show) the one identifiable coupling is an important one.

Below we present the electronic structure and FSs, which show strong differences compared to other Fe pnictides, and discuss implications for the $\text{Sr}_2\text{VO}_3\text{FeAs}$ system and its superconductivity, using first-principles density functional theory. The FSs from such calculations have recently been verified by de Haas-van Alphen measurements in SrFe_2P_2 [25]. A study of the oxygen concentration dependence of the properties of $\text{Sr}_2\text{VO}_3\text{FeAs}$ indicate T_c is maximum (near 40 K) for the stoichiometric composition [26], so study of the stoichiometric compound is most relevant. The effects of the open-shell V bilayer on the properties must be addressed in tandem with the metallic Fe layer. The lack of any reported magnetic behavior [18,26] is an important clue. As in other Fe-pnictides, the tendency toward magnetic behavior is overemphasized with the local density approximation. The types of (overestimated or unphysical) magnetic solutions that can be obtained will be left to a separate report.

Structure and calculation. – In the tetragonal unit cell (space group: $P4/nmm$, No. 129) [18], displayed in fig. 1, Fe atoms lie at $2a$ sites $(\frac{1}{4}, \frac{3}{4}, 0)$, V, Sr, As, and O atoms lie at $2c$ sites $(\frac{1}{4}, \frac{1}{4}, z)$, and another O atom sits at the $4f$ sites $(\frac{1}{4}, \frac{3}{4}, z)$. In our calculations, the experimentally observed lattice parameters $a = 3.9296$ and $c = 15.6732 \text{ \AA}$ were used [18]. The experiment values of the internal parameters z also were used: 0.3081 for V, 0.8097 and 0.5855 for Sr sites, 0.0909 for As, 0.4318 for O at $2c$ sites, and 0.2922 for O at $4f$ sites. The Fe plane is sandwiched between two V planes (with As between) at a distance of 4.83 \AA , and the separation between V layers (in the c -direction) is 6.0 \AA . These large interplanar distances suggest very small dispersion perpendicular to the layers, as will be shown from the calculations.

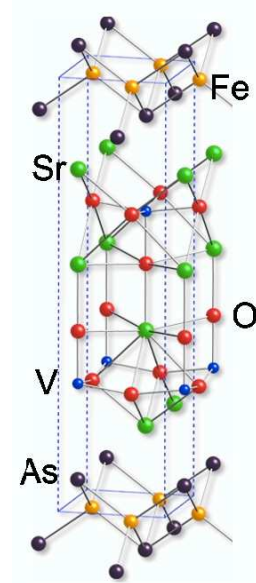


Fig. 1: (Color online) Crystal structure of $\text{Sr}_2\text{VO}_3\text{FeAs}$. The FeAs layers are separated by two perovskite SrVO_3 layers with incomplete O octahedra. The O ions are not coplanar with Sr or, respectively, V positions.

In this structure, the V ion is coordinated by five oxygens and one As ion. The bond lengths of V-O are 1.94 and 1.98 \AA in the a - b plane and c -direction, respectively, while the V-As separation is 3.40 \AA and breaks the local symmetry of the V t_{2g} orbitals. In the characterization scheme of Eschrig and Koepf [27], the Fe-Fe distance $d = 2.78 \text{ \AA}$ and the ratio r of the distance of the As layers from the Fe layer to d is $r = 0.511$. Both of these values lie nearly exactly between the small d , large r , values of FeSe and LiFeAs , and the high d , small r , values of LaOFeAs and BaFe_2As_2 . Noteworthy is that the r values lies closer to the ideal As tetrahedron value of $r = 0.50$ than in the other classes.

The accurate all-electron full-potential local orbital code, FPLO-7 [28], was used for all calculations. The Brillouin zone was sampled with a regular fine mesh up to 405 irreducible k points, necessary for sampling the FSs adequately.

Electronic structure and Fermi surface. – Shein and Ivanovskii have reported [29] a ferromagnetic electronic structure. We have obtained several magnetic solutions, differing by the relative orientation of Fe and V moments. However, since no sign of magnetism has been seen in the data, and LDA calculations also overestimate the tendency toward magnetism in several other Fe pnictide compounds, we focus here on the nonmagnetic solution. Even in the magnetic Fe pnictides, it is the electronic structure of the non-magnetic system that has received the most intense attention.

The band structure within 2 eV of the Fermi level (E_F), which is the regime of most of the Fe and V $3d$ character,

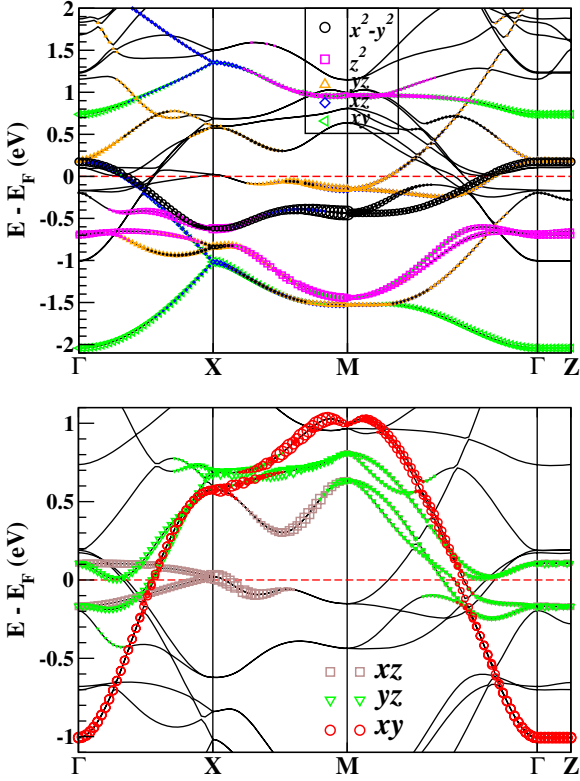


Fig. 2: (Color online) Band structure of nonmagnetic Sr₂VO₃FeAs. Top: Fe fatbands emphasis in the -2 to 2 eV range. Bottom: fatband representation of V t_{2g} states. The symbol size is proportional to fractional character of each orbital. The horizontal (red) dashed lines indicate the Fermi energy E_F , set to zero. Note that each band is either Fe or V, with the only substantial mixing occurring along the X-M line at E_F (see text).

is shown in fig. 2. The bands crossing E_F , and in fact throughout the d band complex, are easily identifiable as either V- or Fe-derived. With one exception (see below) there is little mixing of both characters into any single band, reflecting the weak electronic coupling between the V and Fe subsystems.

The most distinctive characteristic, compared with other iron pnictides [30,31], is the presence of *metallic* V t_{2g} bands in the “spacer layer” and these bands overlap the Fe $3d$ bands. The projected density of states in fig. 3 indicates that, while a large majority of the V $3d$ states are unfilled, there is a substantial contribution at the Fermi level and for several tenths of eV below. The V d_{xz} , d_{yz} bands are centered (fig. 2) near 0.3 eV with a bandwidth of 1 eV, while the d_{xy} band is centered at E_F (*i.e.* is half-filled) with twice larger bandwidth 2 eV. This d_{xy} band can be represented by an independent tight-binding band with near-neighbor hopping amplitude $t = 0.24$ eV and second neighbor $t' = 0.07$ eV. Without mixing to other bands, this d_{xy} band would give rise to two nearly circular and nearly degenerate Fermi surfaces. Note that the V d_{xy} band is doubly degenerate corresponding to the

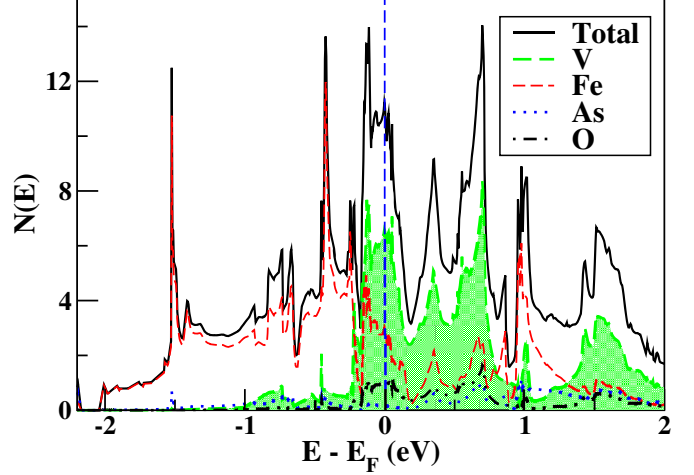


Fig. 3: (Color online) Total and atom-projected densities of states (DOSs) near E_F for nonmagnetic Sr₂VO₃FeAs. The t_{2g} - e_g crystal field splitting of V is about 1.3 eV. DOS at E_F $N(E_F) = 11.2$ states/eV per Fe, composed roughly of 60% V, 25% Fe, and 10% from O ions. The Fermi energy is the zero of energy.

two V layers, for which the d_{xy} orbitals are uncoupled to high precision (the splitting is not visible in the plot). The minor difference in band centers of the symmetry-split V t_{2g} bands reflects the non-zero but small V site tetragonal crystal field. There are also four d_{xz} , d_{yz} bands because of the two V ions; these are not degenerate but are split by up to 0.5 eV in various parts of the zone, indicating some coupling of these orbitals between the V layers or to the FeAs layers.

The half-filled d_{xy} band contains about one electron (probably less), while the two partially filled bands contribute roughly another electron, for each of the two V ions in the unit cell. However, since band characters are mixed, these are only rough estimates. The projected DOS of fig. 3 provide a better guide. The V t_{2g} DOS lies in the range -0.3 to 1 eV, and seems to be rather less than $1/3$ filled, which would indicate less than two $3d$ electrons per V ion. If this is indeed true (and it is not possible to divide the charge in an unambiguous way), then the FeAs layer would be electron-doped in stoichiometric Sr₂VO₃FeAs compared to other undoped FeAs compounds. Then no extrinsic doping would be required to give a high- T_c superconducting state, as seems to be the case experimentally.

The Fermi surfaces are presented in fig. 4. The fatbands representation of fig. 2 indicate a Fe $d_{x^2-y^2}$ band less than 1 eV wide straddling E_F and giving rise to one hole FS centered at Γ , while both d_{xy} and d_{z^2} bands have negligible character at or very near E_F . There is strong Fe d_{xz} , d_{yz} character at E_F , giving rise to the smallest (also hole) FS cylinder centered at the Γ -point.

The other FSs (hole bow-ties centered at X, a hole rounded-triangle FS lying along the (110) direction, and a

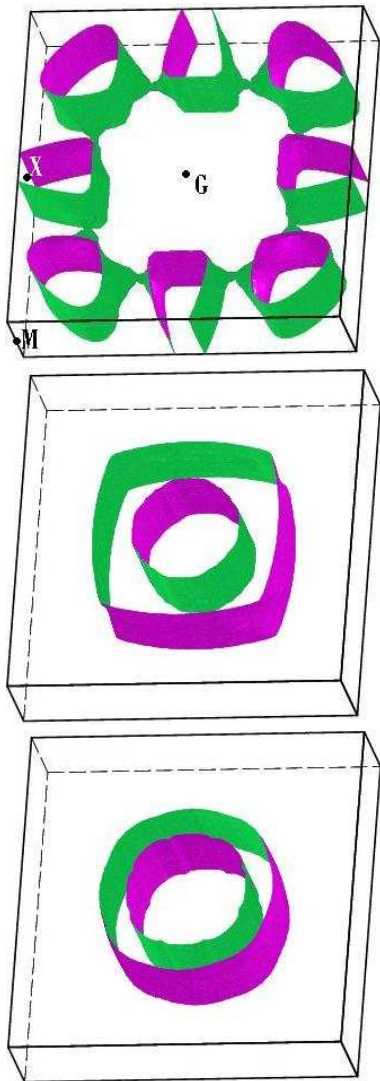


Fig. 4: (Color online) Fermi surfaces of $\text{Sr}_2\text{VO}_3\text{FeAs}$, showing the two-dimensionality. Top panel: X-centered V hole bowties and rounded triangles lying along the (1,1) directions. G denotes the Γ -point. Middle and bottom: larger surfaces in both panels are V character and contain electrons; smaller cylinders enclose Fe-derived holes. Some tiny surfaces are not shown.

squarish electron FS centered at Γ) provide Fermi surfaces not seen in previous Fe-pnictide classes. The triangle FSs contain Fe xy , yz character along the Γ -M line, and in other Fe pnictides this character dominates the electron cylinder at M. However, as a result of the only mixing of V and Fe bands that is visible along symmetry lines, a hybridization gap of 0.4 eV opens up along the X-M direction so no M-centered cylinders emerge and the approximate nesting observed in other classes is substantially degraded. LDA calculations reproduce the FSs reliably in the nonmagnetic phase, as shown recently for SrFe_2P_2 [25].

The difference in electronic structure compared to other FeAs classes has been addressed by Mazin [32],

who graphically showed the change in the M-centered, Fe-derived Fermi surfaces. He found not only degraded nesting but also a strong minimum in the (matrix-element-less) generalized susceptibility $\chi_o(q)$. However, if the processes contributing to $\chi_o(q)$ are confined to Fe (*i.e.* excluding V), then $\chi_o^{\text{Fe}}(q)$ has a broad maximum around the M-point similar to other FeAs compounds, even without Fermi surface nesting. The magnetic behavior within the Fe layer should be modeled more realistically by $\chi_o^{\text{Fe}}(q)$ than by $\chi_o(q)$, and can be expected to be much the same in $\text{Sr}_2\text{VO}_3\text{FeAs}$ as in other FeAs compounds. Full matrix elements should be included to understand the susceptibility more completely; when included in LaFeAsO , the peak around the M-point remains but is reduced in magnitude [33].

The contribution of Fe ions to the DOS at E_F $N(E_F)$ is about 50% larger than in other iron pnictides, due partially to the different band filling but probably also to the different angular structure of the FeAs_4 tetrahedron discussed in the introduction. Note also that 60% of the total $N(E_F)$ arises from the roughly one-third filled t_{2g} manifold of V bands.

The Fe FS cylinders consist of two cylinders with radii of $0.28\frac{\pi}{a}$ and $0.35\frac{\pi}{a}$, containing 0.50 and 0.76 holes, respectively. The two Fe-based cylinder FSs are in fact much like the important surfaces in MgB_2 : two similarly sized concentric cylinders at the zone center. The generalized susceptibility of such FSs is known [34]: there is structure at $2k_F$, and the “nesting function” has a weak divergence at $2k_F$. One notable feature of such FSs is that for boson exchange scattering through wave vector q , the strength is distributed over the entire region $|q| < 2k_F$, which here comprises more than a third of the area of the zone.

Discussion. – Now we collect our findings and discuss implications. The metallic V bilayer separating FeAs layers is a feature not present in other Fe pnictide classes. This primary distinction leads to several secondary differences. The coupling is weak except between V d_{xz} , d_{yz} to Fe d_{xz} , d_{yz} along the zone boundary X-M line, but this coupling seriously degrades the approximate nesting that is seen in other Fe pnictides. More study is needed to determine whether this change of nesting accounts for the lack of magnetic ordering [18]. Also important is that this coupling is sufficient to establish a common Fermi level, so that an integer number of electrons is not required in either subsystem, and the Fe band filling deviates somewhat from related materials. Superconductivity in the Fe layer will induce a non-zero order parameter in the V layers via the proximity effect, but a likely result is that the gaps in the two layers will be very different in size, leading to multi-gap (“multiband”) superconducting character. The strong differences between $\text{Sr}_2\text{VO}_3\text{FeAs}$ and other Fe pnictides should be of importance in identifying the pairing mechanism of these impressive high-temperature superconductors.

The broader consequences for Sr₂VO₃FeAs that can be expected due to the metallic V-O layer — a new conduction channel between the Fe layers, altered band filling and Fermi surface, etc.— deserve comment. Because Sr₂VO₃FeAs superconducts at 37 K, in the same temperature range as several other Fe pnictides, it seems highly likely that its superconducting mechanism is the same. The question of gap symmetry becomes more interesting as the Fermi surface develops more sheets and has strong variation of (Fe, V) character, and thereby of magnitude of gap. The gap could be extremely weak on the V sheets (although magnetic fluctuation may be strong in that layer as well), leading to an extreme two-gap scenario in addition to possible phase changes between different surfaces.

The occurrence of a periodic arrangement of strongly superconducting and weakly superconducting (proximity-induced) conducting layers within 5 Å of each other may be one of the more unusual aspects of Sr₂VO₃FeAs, altering other properties if not T_c . For example, a modest magnetic field might drive the V layer normal while the Fe layer remains strongly superconducting, leading to unexpected transport and spectroscopic behaviors. We expect Sr₂VO₃FeAs to display a number of properties, in the normal state as well as in the superconducting, that are different from the previous Fe pnictides, and which should help to clarify the microscopic mechanisms underlying the remarkable properties of the Fe pnictides.

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Additional remark: Since the submission of this paper, Wang *et al.* [35] have reported the electronic structure of Sr₂VO₃FeAs. That work uses theoretical atomic positions rather than the experimental ones, and differs in some respects from those presented here and by Mazin [32].

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