Reduced Dimensionality and Magnetic Frustration in KCr$_3$As$_3$

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Abstract

We study the electronic and magnetic structures of the newly-discovered compound KCr$_3$As$_3$. The non-magnetic state has five Fermi surface sheets involving respectively three quasi-one-dimensional and two three-dimensional energy bands. However, the ground state is magnetic, exhibiting a novel interlayer antiferromagnetic order where the basic block-spin state of a unit Cr triangle retains a high spin magnitude. Moreover, its Fermi surface involves three one-dimensional sheets only, providing evidence for local moments in this compound due to the reduced dimensionality. By fitting a twisted spin tube model the magnetic frustrations caused by local moments are found to be relaxed, leading to gapless spin excitations. A frustration-induced transition to the disordered low block-spin state is expected upon increasing the intralayer exchange interaction.

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Introduction. The interplay between superconductivity and magnetism or other competing density-waves has been a major theme in modern condensed matter physics, particularly in transition metal compounds with both strong electron correlations and reduced dimensionality. Although the development of long-range orders at finite temperature is forbidden in strictly one-dimensional system with short-range interactions, such restriction can be lifted for quasi-one-dimensional (Q1D) systems. Typical examples include the Q1D superconductors (TMTSF)$_2$X$_3$, $M_2$Mo$_6$Sb$_6$ ($M$=Tl,In)$^4$, Li$_{0.9}$Mo$_6$O$_{17}$, etc. A "universal" phase diagram for the Q1D superconductors suggests that the superconductivity is in close proximity to the density-waves$^5$.\footnote{9.11}

The recently discovered Q1D superconductors A$_2$Cr$_3$As$_3$ (A=K, Rb, Cs) provide a new setting for understanding the interplay of unconventional superconductivity and density wave instabilities in reduced dimensions. The building blocks of these compounds consist of Q1D [CrAs]$_\infty$ double-wall nano-tubes intercalated by A$^+$ cations. The first principle calculations using the density functional theory (DFT)$^{12,13}$ predicted a three-dimensional (3D) Fermi surface (FS) sheet and two Q1D FS sheets, all contributed mainly by the Cr-3d electrons. The penetration depth measurement has evidenced a nodal line in the pairing state below $T_c$. Meanwhile, the NMR experiment$^{15}$ has revealed a power law behavior of the spin-lattice relaxation rate, manifesting the 1D Luttinger liquid feature at the normal state. A couple of theoretical proposals have been devoted to the possible pairing symmetries of the superconducting state, concerning the respective relevance of the Q1D and 3D bands$^{16,17}$, while the formation of molecular orbital bands has been detailed in a twisted Hubbard tube.$^{18}$ These studies suggest possible competing superconducting and density wave ordering tendencies in K$_2$Cr$_3$As$_3$. In fact, superconductivity associated with a magnetic quantum critical point has been revealed in the bulk materials of chromium arsenides$^{19,20}$. The material realization of density-wave instabilities competing with superconductivity in the Q1D chromium arsenide compounds should be very interesting and require more surveys.

Motivated by this issue, we will study the electronic and magnetic structures of a related compound, KCr$_3$As$_3$. This 133-type of chromium arsenide has been recently synthesized experimentally$^{21}$. It consists of the Q1D [CrAs]$_\infty$ structure similar to K$_2$Cr$_3$As$_3$. The compound is metallic down to the lowest measured temperature, but no superconductivity has been observed. Instead, the polycrystal sample displays a spin glassy feature at low temperatures$^{21}$. In this paper, we find based on DFT calculations that the electronic and magnetic structures of KCr$_3$As$_3$ exhibit new properties in contrast to the superconducting 233-type compound. The most prominent features are the lack of three-dimensional Fermi surface sheets and emergence of the interlayer antiferromagnetic order. These numerical results indicate coexistence of local moments and itinerant electrons in the KCr$_3$As$_3$ compound due to the reduced dimensionality. Such exotic metallic magnetic phase is understood as due to the formation of high spin states in each Cr triangles via the orbital selective Mott transition. A frustration-induced quantum phase transition to the disordered low block-spin state with a spin gap is expected by tuning the intralayer exchange to the antiferromagnetic side. Our study thus reveals a possible interplay between unconventional superconductivity and magnetic order in this class of materials.

Calculation Method and Details. All reported results are obtained using the DFT as implemented in the Vienna Abinit Simulation Package (VASP)$^{22,23}$, where the valence electron-ion interactions are modeled using projected augmented wave (PAW) method$^{23,24}$ and the exchange-correlation effects are approximated with Perdew-Burke-Ernzerhoff flavor of general gradient approximation (PBE). To ensure the convergence of the calculations, a 540 eV energy cutoff to the plane wave
basis and a $6 \times 6 \times 12$ $\Gamma$-centered $K$-mesh is employed; whereas a $12 \times 12 \times 24$ $\Gamma$-centered $K$-mesh and tetrahedron method are employed to perform the DOS calculations.

FIG. 1: Crystal structure of KCr$_3$As$_3$. (a) Experimentally determined geometry with space group P6$_3$/m (No. 176); (b) Hypothetic geometry closer to K$_2$Cr$_3$As$_3$ with space group P6/m2 (No. 187). In both panels, the red, blue and green balls represents As, Cr and K, respectively; larger and smaller balls indicate atoms at $z=0.75$ ($X^I$) and $z=0.25$ ($X^{II}$), respectively ($X=K$, Cr, and As).

Experimentally, the crystal structure of KCr$_3$As$_3$ shown in FIG. 1(a), was determined to be centrosymmetric with space group P6$_3$/m (No. 176). Unlike the K$_2$Cr$_3$As$_3$ structure, the Cr$^4$-Cr$^4$ bond lengths are identical to the Cr$^{II}$-Cr$^{II}$ bond lengths in KCr$_3$As$_3$. Apart from this apparent difference, the [CrAs]$_6$ sub-nanotubes in KCr$_3$As$_3$ are rotated $\sim 10^\circ$ with respect to their central axis compared to the K$_2$Cr$_3$As$_3$ structure. In order to make connections with the 233-compound, we have also performed calculations on a hypothetical crystal structure of KCr$_3$As$_3$ which is initially with space group P6$_3$/m (No. 176) but without the rotation of the [CrAs]$_6$ sub-nanotubes (FIG. 1(b)). Interestingly, this structure spontaneously turns into P6/m2 (No. 187) after optimization. So the loss of symmetric center is not due to the distribution of K atoms between $z=0.25$ and $z=0.75$ planes, but rather relates to the rotation of the [CrAs]$_6$ sub-nanotubes.

Table I lists the total energies of KCr$_3$As$_3$ with different magnetic configurations. Unlike the K$_2$Cr$_3$As$_3$ compound, where multiple magnetic states are energetically almost degenerate in DFT calculations, the inter-layer antiferromagnetic (IAF) configuration, a kind of block spin magnetic order, is apparently energetically much lower and is stable against structural optimization. Also, it is worthy noting that both the non-collinear phases (IOP and NCL) are energetically much higher than the collinear phases (FM and IAF). Upon optimization, the collinear phases are energetically stable while the non-collinear ones are not. For the lowest energy state IAF, the magnetic moment on each Cr is calculated to be $\sim 0.77 \mu_B$, in good agreement with the experimental observation of 0.68 $\mu_B$.

In order to understand the formation of magnetic moments, we first examine the electronic structure of KCr$_3$As$_3$ in the non-magnetic phase. As shown in FIG. 2 (a), the band structure of KCr$_3$As$_3$ near the Fermi level is quite similar to that of K$_2$Cr$_3$As$_3$, including the orbital characters, but the degeneracies at $\Gamma$ and $K$ are completely different. Indeed, the degeneracies at $K$ of KCr$_3$As$_3$ and the double degeneracy of the bands around $E_F$-0.5 eV at $k_z=0.5$ (A-H-L-A) are direct consequences of the P6$_3$/m symmetry which is broken in K$_2$Cr$_3$As$_3$. Furthermore, the reduction of 50% potassium atoms causes hole doping to the system, resulting in a shift of the Fermi level. As a result, the number of bands cross the Fermi level becomes five, among which three are quasi-1D and two are 3D. The DOS and the orbital contributions are shown in FIG. 2 (b). The quasi-1D signature van Hove singularities are apparent around -1.6 eV, -0.5 eV, -0.4 eV, 0.5 eV relative to $E_F$. The DOS around $E_F$ is dominated by the Cr-3d orbitals, in particular, Cr-3d$_{xy}$ and 3d$_{x^2-y^2}$ orbitals. The contribution of Cr-3d$_{xy}$ orbital is small, but non-negligible since the $\alpha$-band is almost exclusively contributed by Cr-3d$_{x^2}$ (FIG. 2(a)). The Fermi surface (FS) of KCr$_3$As$_3$ constitutes
TABLE I: Total energies of KCr$_3$As$_3$ with different magnetic configurations. Column Exp, Opt and P6m2 list the results with experimental lattice constants and coordinates, the optimized lattice constants and coordinates, and the hypothetical structure after optimization, respectively. Please be noted that the hypothetical structure was initially centrosymmetric P6$_3$/m (No. 176), but spontaneously turned into P6m2 (No. 187) after optimization.

|     | FM | IAF | TOP | NCL | NM | BIAF | UUD | FM | IAF | NM |
|-----|----|-----|-----|-----|-----|------|-----|-----|-----|-----|
| Exp |    |     |     |     |     |      |     |     |     |     |
| $\Delta E$ (meV) | $m_C$ (µB) |  |     |     |     |      |     |     |     |     |
| $a$ (c) | -41.3 (-43.9) | 0.36 | 9.0886 (4.1801) |     |     | -20.7 | -23.4 | -50.7 | -129.7 | 0.0 |
| Opt | 9.3442 (4.0862) | 0.14 | 9.7109 (4.0826) |     |     | -7.0 | 9.7067 (4.0901) |     |     | 0.0 |
| $a$ (c) | -0.8 (-6.0) | 0.29 | 9.3447 (4.0826) |     |     | 0.0 | 9.3442 (4.0862) |     |     | 0.0 |
| P6m2 | 9.7091 (4.0845) | 0.1 | 0.24(-0.29) |     |     | 0.0 | 9.7109 (4.0826) |     |     | 0.0 |

of three quasi-1D sheets and two 3D sheets as shown in the Supplemental Material (SM). If the system is doped with one electron per formula (i.e. K$_2$Cr$_3$As$_3$), the FS sheets will be reduced to two quasi-1D and one 3D, as in the original K$_2$Cr$_3$As$_3$ compound. Of course, the shape of the 3D FS sheet will be different from the one in the original K$_2$Cr$_3$As$_3$ due to the local structure difference.

We now show the electronic structure of the lowest interlayer antiferromagnetic phase (IAF) in Figure 3. The orbital characteristics of bands near $E_F$ remains the same as those in the NM state. The four Cr-3d$_{xy}$/Cr-3d$_{yz}$ bands near $E_F$ are separated by ~0.4 eV in the IAF phase, presumably due to formation of local moment on Cr; whereas these bands are very close to each other in the NM state. Only spin up channel is shown for clarity purposes. Please be noted that the hypothetical structure was initially centrosymmetric P6$_3$/m (No. 176), but spontaneously turned into P6m2 (No. 187) after optimization.

Local Moments and Magnetic Frustration. The above DFT calculations suggest the existence of local moments in Cr$_3$As$_3$, given the facts that the calculated magnetic moment $\sim 0.77\mu_B$ is close to the saturated value of $S = 1/2$ isolated local moment and quantum frustrations are not considered so far. The local moment phase is also hinted in the non-magnetic band structure FIG. 2 (a), where two upper degenerate bands along the $\Gamma \rightarrow A$ direction intersect the Fermi energy at the Fermi point $k_F$, $\sim 0.56\pi$. This value is very close to $\pi/2$, indicating the nearly half-filling case of the corresponding molecular orbital band. In 1D, any repulsive Coulomb interaction will induce the 4$k_F$ Umklapp scattering at half-filling, resulting in a finite charge gap. Therefore, a Mott transition in this band is naturally expected in the corresponding molecular orbital band. In fact, compared with the band structure of K$_2$Cr$_3$As$_3$, both the band narrowing effect and the electron correlations are enhanced due to the reduced dimensionality in KCr$_3$As$_3$, providing more evidence for a Q1D Mott phase via orbital selective Mott transitions as in iron pnictides.

FIG. 3: (a) Band structure and (b) DOS of KCr$_3$As$_3$ at the IAF state. Only spin up channel is shown for clarity purposes. In panel (a), the size of the red/blue/green circles are proportional to the orbital weight of the Cr-3d$_{xy}$/Cr-3d$_{yz}$/3d$_{z^2}$, respectively. The dashed horizontal line indicates the Fermi level corresponding to doping the system with 1 electron/formula, i.e. K$_2$Cr$_3$As$_3$. 




We are then led to consider a local moment at each Cr site described by a spin operator, $S_z$, with the magnitude $S = 1/2$. Assuming that the coupling between itinerant electrons and local moments is relatively weak, we consider the Hamiltonian of these localized spins as

$$H_{\text{spin}} = J_1 \sum_{n,I,I'} S_{n,I} \cdot S_{n,I'} + J_2 \sum_{n,I,I'} S_{n,I} \cdot S_{n+1,I'}$$

Where, the first two terms indicate the intra-layer nearest-neighbor couplings in the two different triangles, while the last two terms the inter-layer nearest-neighbor couplings between the two triangles within one unit cell and across two unit cells, respectively.

The spin model is an extended variant of twisted spin tubes previously proposed for the three leg systems [(CuCl$_2$taclH)$_2$]Cl$_2$ and CsCrF$_4$-33-34. The magnetic frustrations will arise not only due to the intralayer spin exchanges but also the interlayer or inter-unit-cell spin exchanges if they are all antiferromagnetic. Concerning the present KCr$_3$As$_3$ compound, we can assume $J_1 = J_1$, $J_2 = J_2$. Their values are estimated by fitting the corresponding energies in different magnetic structures with the moment per Cr atom being fixed at 0.77 $\mu_B$. The best fitting suggests $J_1 = -9.9$ meV and $J_2 = 18.5$ meV. It is worthy noting here that we have also determined the strength of the inter-tube interactions $J_t$ for the IAF state to be $< 0.1$ meV by comparing the DFT total energies of inter-tube-FM and inter-tube-AFM states. Thus the KCr$_3$As$_3$ system is a truly remarkable example of nearly one-dimensional spin chain, explaining why the IAF long range order was not observed in experiments. The ferromagnetic $J_1$ implies the suppression of the magnetic frustration in this 133-compound. In this case, the twisted tube is basically a rhombus lattice without frustration due to the bipartite nature of the lattice. A large block-spin $S_\Delta = 3/2$ state is then expected for each Cr triangle, involving the four polarization components: $|1/2\rangle = |\uparrow\uparrow\uparrow\rangle$, $| -1/2\rangle = |\downarrow\downarrow\downarrow\rangle$, $|1/2\rangle = \sqrt{2}/3(|\uparrow\downarrow\uparrow\rangle + |\uparrow\uparrow\downarrow\rangle + |\downarrow\uparrow\uparrow\rangle)$. By introducing a spin-3/2 operator $T$ acting on these $S_\Delta = 3/2$ states, we obtain the effective Hamiltonian

$$H_{S_\Delta=3/2} = \sum_n [K_1 T_n \cdot T_{n+1} + K_2 (T_n \cdot T_{n+1})^2]$$

with $K_1 = 2J_2/3 + J_2/(18|J_1|)$ and $K_2 = -J_2^2/(54|J_1|)$. The low temperature property is dominated by the $K_1$ term. With quantum fluctuations the interlayer antiferromagnetic order is only quasi-long-ranged, in the sense that the spin-spin correlation function exhibits a power-law behavior and the spin excitation spectrum is gapless.

This magnetic ground state remains stable in a wider parameter regime when $J_1$ moves toward the antiferromagnetic side. However, when $J_1$ is much larger than $J_2$, the ground state will be changed due to strong frustrations. In the limiting case, each triangle can be solved independently, giving rise to the doubly degenerated ground states involving $|\uparrow\uparrow\rangle = 1/\sqrt{3}(|\uparrow\uparrow\uparrow\rangle + \omega|\uparrow\downarrow\downarrow\rangle + \omega^{-1}|\downarrow\uparrow\uparrow\rangle)$, $|\uparrow\downarrow\rangle = 1/\sqrt{3}(|\uparrow\uparrow\downarrow\rangle + \omega|\uparrow\downarrow\uparrow\rangle + \omega^{-1}|\downarrow\uparrow\downarrow\rangle)$ (and the states with $|\uparrow\rangle \leftrightarrow |\downarrow\rangle$). Noted that these lower block-spin $S_\Delta = 1/2$ states are chiral in nature, associated with the $C_3$ rotation $\omega = e^{2\pi i/3}$. To the lowest order in $J_2$, the effective spin model describing these $S_\Delta = 1/2$ spin excitations is given by

$$H_{S_\Delta=1/2} = \frac{2J_2}{3} \sum_n T_n \cdot T_{n+1} [1 + 2(\omega_2^\sigma \tau^+ \tau^{-} + \text{h.c.})],$$

with $T$ being a spin-1/2 operator acting on the two different spin polarization components $\sigma = \uparrow, \downarrow$, while $\tau^\pm$ on the chirality degrees of freedom, namely, $\tau^\pm |\sigma L\rangle = 0$, $\tau^\pm |\sigma R\rangle = |\sigma L\rangle$, $\tau^- |\sigma R\rangle = 0$. According to the early numerical studies, this model has a disordered ground state, i.e., the dimerized ground state exhibiting gaps in both spin and chirality excitations. When $J_1$ moves toward the antiferromagnetic order, the IAF state is gapless, the lower spin excitations are therefore gapful. Noted that these lower block-spin $S_\Delta = 3/2$ defined in Eq.(2). The absence of superconductivity is likely due to the strong competition tendency of the quasi antiferromagnetic order in this 133-compound.

We should remark that due to the reduced dimensionality, a static long-range magnetic order is unstable against to enhanced quantum or thermal fluctuations, and strongly influenced by the disorder effect. So it is not very strange that the spin glass behavior, instead of the static magnetic order, is observed in the polycrystalline samples. We also remark here that although the ground state itself is gapless, the lower spin $S_\Delta = 1/2$ sector may still play some role at finite temperatures, exhibiting a crossover-to-spin-gap behavior as in the spin tube material [(CuCl$_2$taclH)$_2$]Cl$_2$. An intriguing situation with a large intralayer antiferromagnetic coupling $J_1$ is expected, hopefully by applying the planar pressure while keeping the symmetry of charge distributions among the two conjugated triangles. Such situation should be contrasted to the 233-compound, where the more $K$ concentration induces the non-centrosymmetric structural distortion and asymmetric charge distributions in the two conjugated triangles.
Because superconductivity may be also induced from the spin-gaped phase upon doping\textsuperscript{41} and given the recent pressure experiments on the K$_3$Cr$_2$As$_3$\textsuperscript{12,43}, we expect that the disordered phase with both spin and chirality gaps can be realized in further coming pressure experiments on KCr$_3$As$_3$ and the related compounds.

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SUPPLEMENTARY MATERIAL

Reduced Dimensionality and Magnetic Frustration in KCr$_3$As$_3$

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The Fermi surfaces are reconstructed by fitting the first-principles band structure to a 10-orbital tight-binding Hamiltonian using the maximally localized Wannier functions (MLWFs). Figure S1(a) shows the Fermi surface sheets of KCr$_3$As$_3$. Three quasi-1D sheets and two 3D sheets constitute the Fermi surface of pristine KCr$_3$As$_3$ as expected. If the system is doped with 1e per formula (i.e. K$_2$Cr$_3$As$_3$), the FS sheets will be reduced to two quasi-1D and one 3D, as in the original K$_2$Cr$_3$As$_3$ compound$^{12}$. However, the shape of the 3D FS sheet will be completely different from the one in the original K$_2$Cr$_3$As$_3$ due to the local structure differences.