

Triplet $p_z$-wave pairing in quasi one dimensional $\text{A}_2\text{Cr}_3\text{As}_3$ superconductors

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We construct minimum effective models to investigate the pairing symmetry in the newly discovered quasi-one-dimensional superconductor $\text{K}_2\text{Cr}_3\text{As}_3$. We show that a minimum three-band model based on the $d_{z^2}$, $d_{xy}$ and $d_{x^2−y^2}$ orbitals of one Cr sublattice can capture the band structures near Fermi surfaces. In both weak and strong coupling limits, the standard random phase approximation (RPA) and mean-field solutions consistently yield the triplet $p_z$-wave pairing as the leading pairing symmetry for physically realistic parameters. The triplet pairing is driven by the ferromagnetic fluctuations within the sublattice. The gap function of the pairing state possesses line gap nodes on the $k_x = 0$ plane on the Fermi surfaces.

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I. INTRODUCTION:

Searching for triplet superconductivity (SC) has been one of major research efforts recently partly due to its intrinsic connection to topologically related physics and quantum computations. Unconventional triplet SC is normally driven by electron-electron interactions. Until now, the candidates for triplet pairing superconductors are rather limited. For quasi-two dimensional electron systems, $\text{Sr}_2\text{RuO}_4$ is considered to be a possible good candidate. For quasi-one-dimensional (Q1D) systems, since the electron-electron correlation can be further enhanced, triplet pairing has also been proposed in previously discovered Q1D systems, such as Bechgaard salts $\text{K}_2\text{Cu}^{2+}\text{Be}^{16+}\text{C}_{28}^+$, $\text{Tl}_2\text{Mo}_6\text{Se}_9^-$ and $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$. However, in these Q1D materials, the correlation may be offset by the fact that their electronic physics is not attributed to the orbitals of chromium atoms.

The chromium-based superconductors have attracted great research interests recently. Their superconducting mechanism shown in many experimental results is very likely related to electron-electron interactions. Among the families of the chromium-based superconductors, the Q1D $\text{A}_2\text{Cr}_3\text{As}_3$ ($\text{A} = \text{K}, \text{Rb}, \text{Cs}$) superconductors family, which has $T_c$ up to 6.1 K at ambient pressure, is particularly intriguing. The crystal of this family consists of alkali metal ions separated by double-walled subnanotubes. The electronic structures calculated from Density Functional Theory (DFT) suggests that the $3d$ orbitals of Cr make major contribution near Fermi surfaces. The large electronic specific-heat coefficient, the non-fermi liquid behavior in the normal state and the abnormality in NMR relaxation-rate all suggest possible strong electron correlations in the family. The linearly temperature-dependent London penetration depth, the lack of the Herbal-Slichter peak in NMR relaxation-rate upon the superconducting transition, and the $\sqrt{H}$-dependence of $C_v$ in the mixed state suggest unconventional SC with line nodes in the system. More importantly, the upper critical field largely exceeds the Pauli limit, implying possible triplet pairings in the family.

The triplet pairings were also argued theoretically. The theoretical calculations predicted that the materials are very close to a new in-out coplanar (IOP) magnetic state. The magnetic state is caused by the strong antiferromagnetic couplings between the nearest neighbour (NN) Cr atoms and the weak ferromagnetic couplings between the next nearest neighbour (NNN) ones. Intriguingly, the magnetic fluctuations associated with such a magnetic state can favor triplet pairing. A unit cell of $\text{A}_2\text{Cr}_3\text{As}_3$ is composed of two types of inequivalent Cr atoms which will be referred as Cr1 and Cr2 atoms as shown in Fig. 1, which form two sublattices, A and B, respectively. It is shown that Q1D electronic structures are dominantly contributed by only the Cr2 sublattice. Within the Cr2 sublattice, the magnetic fluctuations are ferromagnetic (FM), which are caused by both the weak NNN ferromagnetic couplings and effective ferromagnetic fluctuations generated by the NN antiferromagnetism. Thus the triplet pairing is very possible in $\text{A}_2\text{Cr}_3\text{As}_3$. Furthermore, the materials do not have space inversion symmetry but it still has the mirror plane symmetry with respect to the $x−y$ plane. Therefore, along the Q1D chain direction, the triplet $p_z$-wave pairing, which is odd with respect to the mirror plane reflection, is a natural triplet candidate classified by the irreducible representations of the lattice symmetry.

In this paper, we construct low energy effective models to study the pairing symmetry of $\text{K}_2\text{Cr}_3\text{As}_3$. First, we construct the minimum tight binding model for the material. A six-band model are constructed with the $\text{Cr}-d_{z^2}$, $d_{xy}$ and $d_{x^2−y^2}$ orbitals of both Cr1 and Cr2 atoms. At low energy, this model can be further simplified to the $d$-orbital of the Cr2 atoms. Starting from the three-band tight-binding model, we investigate the pairing symmetries in two types of standard models, one with the standard electron-electron interaction terms including onsite Hubbard $U$ and Hund’s coupling $J_{H}$ and the other with the effective FM exchange couplings. We adopt combined random phase
approximation (RPA) and mean-field approaches for the weak and strong interaction limits respectively. Both approaches consistently yield the $p_z$-wave triplet pairing as the leading pairing symmetry for physically realistic parameters satisfying $\frac{J}{\mu^2} < r_c \approx \frac{1}{3}$ (for $\frac{J}{\mu^2} > r_c$, the RPA yields on-site interorbital $f_{9/2-3s2y}$-wave pairing, driven by the Hund’s rule coupling), driven by intra-sublattice FM spin fluctuations. The gap function of this pairing possesses line gap nodes on the $k_z = 0$ plane on the FS. This pairing is essentially Q1D and the pairing amplitudes on the Q1D $\beta$ band dominates those on the Q1D $\alpha$ band and the 3D $\gamma$-band. Finally, we shall address the experimental relevance of the $p_z$-wave triplet pairing obtained here.

II. MODEL AND APPROACH

A. The tight-binding Model

From the bandstructure of DFT calculations\textsuperscript{19,20}, it’s found that there are five bands near the FS and the orbital characters near the Fermi level are mainly of Cr-$d_{z^2}$ ($A_1'$ symmetry) and ($d_{xy}$, $d_{x^2-y^2}$) ($E'$ symmetry) components. To obtain an effective model, we consider a virtual atom at the center of each Cr triangle with three orbitals, which have the same symmetries as $A_1'$ and $E'$, as shown in Fig.1. Now the structure of K$_2$Cr$_3$As$_3$ is simplified as parallel placed Cr-chains, with each chain containing the A and B sublattices, which are inequivalent due to the asymmetry of K atoms and the absence of inversion symmetry. The point group of this new lattice model is the same with the original lattice and it is $D_{3h}$ not $D_{6h}$, where inversion and six-fold rotational operations are absent. The tight-binding Hamiltonian thus obtained in the momentum space can be written as,

$$H^{(0)}_{TB} = \sum_{k \mu \nu \sigma} h_{\mu \nu}^{m n}(k)c_{m \mu \sigma}^{\dagger}(k)c_{n \nu \sigma}(k), \quad (1)$$

where $m/n = A, B$ labels sublattice, $\mu/\nu = 1, \cdots, 3$ labels orbital and $\sigma = \uparrow, \downarrow$ labels spin. $c_{m \mu \sigma}^{\dagger}(k)$ creates a spin-$\sigma$ electron in the orbital $\mu$ in the $m$-th sublattice with momentum $k$. The elements of the matrix $h_{\mu \nu}^{m n}(k)$ and the resulting band structure of this model are given in Appendix.A. The band structure of this six-band tight-binding model well captures the main characters of that of DFT at low energy.

As the states near the Fermi level are dominantly contributed from the B sublattice, we can further simplify our model to a three-band one. Neglecting spin-orbit coupling, we obtain the following tight-binding Hamiltonian

$$H_{TB} = \sum_{k \mu \nu \sigma} h_{\mu \nu}(k)c_{\mu \sigma}^{\dagger}(k)c_{\nu \sigma}(k). \quad (2)$$

Here, the orbital index $\mu/\nu = 1, \cdots, 3$ represent the $d_{z^2}$ for 1, the $d_{xy}$ for 2, and the $d_{x^2-y^2}$ for 3, respectively. The matrix
TABLE I. The hopping parameters in the three-orbital tight-binding model. The onsite energies of $A_{1g}$ and $E_g$ orbitals are $\epsilon_1 = 2.0939$ and $\epsilon_2 = 2.0997$. They are given in unit of eV.

| $s^{\alpha\beta}_{i}$ | $i = x, 2$ | $i = z, 4$ | $i = z, 6$ | $i = y, 1$ | $i = 2y$ | $i = 11y$ | $i = 22y$ | $i = 12y$ | $i = y$ | $i = y^z$ |
|---------------------|----------|----------|----------|----------|--------|--------|--------|--------|--------|--------|
| $\alpha/\beta = 11$ | 0.2841   | -0.0505  | -0.0252  |          |        |        |        |        |        |        |
| $\alpha/\beta = 22$ | 0.1816   | -0.0469  | 0.0034   |          |        |        |        |        |        |        |
| $\alpha/\beta = 12$ |          |          |          | 0.0175   | 0.0095 |        |        |        |        |        |

FIG. 3. (color online). Density of states and susceptibility. (a). The DOS of the three-band tight-binding model. (b). Momentum-dependence of the eigen susceptibility $\chi(q)$ along the high-symmetry lines in the Brillouin-Zone.

elements $h_{\mu\nu}(k)$ are given by

$$h_{11} = \epsilon_1 + (s_{1y}^{11} + 2s_{1y}^{12}\cos 2z)(2\cos 2y + 4\cos x \cos y)$$

$$+ 2 \sum_{i=1}^{3} s_{2i,2i}^{12}\cos 2iz,$$

$$h_{12} = 2is_{1y}^{12}\sin 2y + 2is_{1y}^{12}\sin x \cos x + 2\sqrt{3}s_{2y}^{12}\sin x \sin y,$$

$$h_{21} = h_{12},$$

$$h_{13} = 2s_{2y}^{12}\cos 2y - 2i\sqrt{3}s_{1y}^{12}\cos x \sin x - 2s_{2y}^{12}\cos x \sin y,$$

$$h_{31} = h_{13}^*,$$

$$h_{22} = \epsilon_2 + 2s_{11y}^{22}\cos 2y + (s_{11y}^{22} + 3s_{12y}^{22})\cos x \cos y$$

$$+ 2 \sum_{i=1}^{2} s_{2i,2i}^{22}\cos 2iz,$$

$$h_{23} = \sqrt{3}(s_{11y}^{22} - s_{22y}^{22})\sin x \sin y + 2is_{12y}^{22}\sin 2y,$$

$$- 4is_{12y}^{22}\cos x \sin y,$$

$$h_{32} = h_{23}^*,$$

$$h_{33} = \epsilon_2 + 2s_{22y}^{22}\cos 2y + (3s_{11y}^{22} + s_{12y}^{22})\cos x \cos y,$$

$$+ 2 \sum_{i=1}^{2} s_{2i,2i}^{22}\cos 2iz. \quad (3)$$

Here $x = \sqrt{3}/2 k_x a_0$, $y = \frac{1}{2} k_y a_0$ and $z = \frac{1}{2} k_z c_0$, where $a_0$, $c_0$ are lattice constants within the $xy$-plane and along the $z$-axis respectively. The tight-binding parameters $\epsilon_{1,2}$ and $s_{\alpha\beta}^{\alpha\beta}$ are shown in Table I.

The band structure of the above introduced three-band tight-binding model is shown in Fig.2(a), in comparison with that of DFT. The chemical potential of our tight-binding model is $\mu_c = 2.3105 eV$, leading to a band filling of 4-electrons per unit cell. From the comparison, it’s clear that the three-band tight-binding band structure well captures the main characters of the DFT band structure at low energy. Two 1D bands (marked as $\alpha$ and $\beta$ in Fig.2 (a)) and one 3D band (marked as $\gamma$) are found near the FSs. The two 1D bands are degenerate along the $\Gamma - A$ line. The FSs of the tight-binding model are shown in Fig.2(b), where the FS sheets are marked corresponding to the three bands. The dominating orbital component on the $\alpha$- FS is $d_{z^2}$, while those on the $\beta$- and $\gamma$- FSs are $(d_{xy}, d_{z^2-y^2})$ (see Appendix A). Due to the flatness of the two 1D bands near the FS as shown in Fig.2(b), large density of states (DOS) is obtained at the Fermi level, as shown in Fig.3(a).

B. Interactions and Approaches

Since the interaction strength of the 3$d$-electrons of Cr might be moderate, we have investigated both the weak-coupling and strong-coupling limits of the electron interactions in the present work, with both limits giving consistent results. For the weak coupling limits, we adopted the Hubbard-Hund’s type of on-site interactions and performed the standard multi-orbital RPA calculations to study the pairing symmetry. For the strong coupling limit, we adopted the super-exchange type of interactions and performed a mean-field analysis on the $t - J$ type of model instead.

In the weak coupling limit, the following on-site Hubbard-Hund’s interactions are adopted

$$H_{\text{H-H}} = U \sum_{i,\mu} n_{i\mu}^\dagger n_{i\mu} + V \sum_{i,\mu<\nu} n_{i\mu} n_{i\nu} + J_H \sum_{i,\mu<\nu} \left[ \sum_{\sigma,\sigma'} c_{i\mu\sigma}^\dagger c_{i\nu\sigma'} c_{i\mu\sigma'} c_{i\nu\sigma} + (c_{i\mu\uparrow}^\dagger c_{i\downarrow}^\dagger c_{i\nu\downarrow} c_{i\nu\uparrow} + h.c.) \right]. \quad (4)$$

Here, the $U$-term, the $V$-term and the $J_H$-term denote the intra-orbital, inter-orbital Hubbard-repulsion and the Hund’s rule coupling as well as the pair hopping. For simplicity, we have assumed that the coupling constant $U$, $V$, $J_H$ doesn’t depend on the orbital indices in our effective model. General symmetry argument requires $U = V = 2J_H$. Note that physically the orbital bases $d_{z^2}, d_{xy}$ and $d_{z^2-y^2}$ building our effective model are not the maximally-localized Wannier wave functions around a real single Cr-atom, but the mixture of the three atomic wave functions around the virtual center. Therefore, the on-site interactions on these delocalized orbital bases are much weaker than that on a real single atom. In our study, we let $U$ and the ratio $r = J_H/U$ to be tuning parameters and...
study the parameter-dependence of the results.

To study the pairing symmetry of this Hubbard-Hund model, we adopt the standard multi-orbital RPA approach\textsuperscript{21-30}.

We first define and calculate the bare susceptibility tensor $\chi_{l_3,l_4}^{(0)}(q,\tau)$. After that, the renormalized spin(s) or charge(c) susceptibilities $\chi_{l_3,l_4}^{(s,c)}(q,\tau)$ are obtained in the RPA level. Then, through exchanging the spin or charge fluctuations, we obtain the effective pairing potential $V^{\alpha\beta}(k,q)$. Finally, solving the linearized gap equation for $V^{\alpha\beta}(k,q)$ as an eigenvalue problem, we obtain the leading gap pairing function as the eigenvector corresponding to the largest eigenvalue $\lambda$. Generally, the RPA only works well for weak interactions.

In the strong coupling limit, we consider super-exchange type of interactions. From DFT calculations on $K_2Cr_3As_3$\textsuperscript{28}, the NN and NNN exchange couplings along the Cr-chain are antiferromagnetic (AFM) and FM respectively. As only the Cr2 atoms are included in our three-band model, the super-exchange interactions between adjacent Cr2 along the chain direction (ie. the z-direction) are actually FM, which provides us the following exchange interaction term

$$H_J = -J_2 \sum_{\langle ij \rangle,\alpha} S_{\alpha i} \cdot S_{\alpha j},$$

with $J_2 > 0$. Here $\alpha \in \{d_{xz},d_{xy},d_{x^2-y^2}\}$, $\langle ij \rangle$ denotes NN bond along the $z$-direction in our model but NNN bond in real materials and $S_{\alpha i} = \frac{1}{2} \sum_{\sigma} c_{\alpha i \sigma}^\dagger c_{\sigma i \sigma}$ is the local spin operator. The inter-chain exchange coupling has been neglected here due to the Q1D character of the material. Note that only intra-orbital FM exchange coupling is considered because of the "IOP" spin configuration introduced in Ref.20.

Adding this interaction term into the tight-binding model, we obtain an effective $t-J$ model. Since the band filling here is far from making the system a Mott-insulator even in the large U limit, we shall omit the no-double-occupancy constraint and perform a mean-field analysis on the model.

### III. THE WEAK COUPLING LIMIT

For the weak-coupling limit, we adopt multi-orbital RPA approach\textsuperscript{21-30} to study the pairing symmetry of the Hubbard-Hund’s model of the system. The Hamiltonian of our model is,

$$H = H_{TH} + H_{H-H},$$

where the interaction term $H_{H-H}$ is give by Eq.(4).

#### A. Susceptibilities and magnetic phase

Let’s define the following bare susceptibility tensor,

$$\chi_{l_3,l_4}^{(0)}(q,\tau) = \frac{1}{N} \sum_{k_1,k_2} \langle T_\tau c_{l_3}^\dagger(k_1,\tau) c_{l_4}(k_1 + q,\tau) c_{l_3}^\dagger(k_2 + q,0) c_{l_4}(k_2,0) \rangle_0,$$

with $l_{i(i=1,\ldots,4)}$ denoting orbital indices. The explicit formulation of $\chi^{(0)}$ is given by

$$\chi_{l_3,l_4}^{(0)}(q,\omega_n) = \frac{1}{N} \sum_{k,\alpha,\beta} \xi_{l_3}^{\alpha}(k) \xi_{l_4}^{\alpha\dagger}(k) \xi_{l_3}^{\beta}(k + q) \xi_{l_4}^{\beta\dagger}(k + q)$$

$$\xi_{l_3}^{\alpha}(k+q) = n_F(\varepsilon_{k+q}) - n_F(\varepsilon_k^\alpha)$$

where $\alpha/\beta = 1,2,3$ are band indices, $\varepsilon_{k+q}^\alpha$ and $\varepsilon_k^\alpha$ are the $\alpha$-th eigenvalue and eigenvector of the $h(k)$ matrix respectively and $n_F$ is the Fermi-Dirac distribution function.

To detect the pattern of the dominating spin correlation in the system, we define the static susceptibility matrix $\chi_{l,l'}(m,m') = \chi_{l,l'}^{(0)}(q=0)$. The largest eigenvalue of this Hermitian matrix for each momentum $q$ is defined as $\chi(q)$, which represents the eigen susceptibility in the strongest channels. The distribution of $\chi(q)$ in the Brillouin-Zone reflects the pattern of spin correlations in the system. Fig.3(b) shows the momentum-dependence of $\chi(q)$ along the high-symmetry lines, where the peak at the $\Gamma$-point reveals strong FM spin correlations in the model. Note that in the building of our model, only one sublattice is taken from real material, which thus means that the spin correlation pattern found here is intra-sublattice FM.

When interactions turn on, the renormalized spin(s) or charge(c) susceptibilities are obtained in the RPA level as

$$\chi^{(s,c)}(q,\nu) = \left[I + \chi^{(0)}(q,\nu) U^{(s,c)}\right]^{-1} \chi^{(0)}(q,\nu)$$

Here $\chi^{(s,c)}(q,\nu_n)$, $\chi^{(0)}(q,\nu_n)$ and $U^{(s,c)}$ are operated as $9 \times 9$ matrices (the upper or lower two indices are viewed as one number) and the matrix $U^{(s,c)}$ are given in the Appendix.B Clearly, the repulsive Hubbard-interactions enhance(suppress) spin(charge) susceptibility. Note that the RPA approach only works for weak $U^{(s)}$ when all the eigenvalues of the denominator matrix $I - \chi^{(0)}(q,\nu) U^{(s)}$ are positive. At the critical interaction strength, the lowest eigenvalue of that matrix touches zero, which leads to divergence of the spin susceptibility and invalidates the RPA, suggesting the formation of magnetic order. The critical strength $U_c$, as function of $J_H/U$ is given in the phase-diagram in Fig.6 as the phase boundary separating the superconducting phase and the magnetic phase. For $U > U_c$, intra-sublattice FM phase emerges since the bare susceptibility shown in Fig.3(b) peaks at the $\Gamma$-point (the renormalized susceptibility also peaks there), consistent with the “IOP” phase obtained in Ref.20.

#### B. Pairing symmetry study

When the interaction strength $U < U_c$, there can be short-ranged spin or charge fluctuations in the system. Let’s consider a Cooper pair with momentum/orbital ($k'\tau',-k'\tau$), which could be scattered to ($k\tau,-k\tau$) via exchanging spin or charge fluctuations. This second-order perturbation process will contribute an effective interaction vertex $\Gamma^{\tau\tau'}_{\alpha\beta}(k,k')$. Considering only intra-band pairings, we project the effective
interaction $\Gamma_{st}^{\mu}(\mathbf{k}, \mathbf{k}')$ onto the FSs and obtain an effective pairing potential $V^{\alpha \beta}(\mathbf{k}, \mathbf{k}')$, where $\alpha/\beta = 1, \cdots, 3$ are band indices. The explicit formula of $V_{st}^{\mu}(\mathbf{k}, \mathbf{k}')$ and $V^{\alpha \beta}(\mathbf{k}, \mathbf{k}')$ are given in the Appendix.B.

Finally, one should solve the following linearized gap equation to find the pairing symmetry,

$$
- \frac{1}{(2\pi)^3} \sum_\beta \int_{FS} d^2k' \left\| \frac{V^{\alpha \beta}(\mathbf{k}, \mathbf{k}')}{\epsilon_F^{\mu}(\mathbf{k}')} \right\| \Delta_\beta(\mathbf{k}') = \lambda \Delta_\alpha(\mathbf{k}).
$$

Here, the integration and summation are along various FS patches. The $\epsilon_F^{\mu}(\mathbf{k})$ is Fermi velocity and $\epsilon_F^\mu$ represents the component within patch $\beta$. This equation can be looked upon as an eigenvalue problem, where the eigenvector $\Delta_\beta(\mathbf{k})$ represents the relative gap function near $T_c$, and the eigenvalue $\lambda$ determines $T_c$ via $T_c=\text{cut off energy } e^{-1/\lambda}$. The leading pairing symmetry is determined by the largest eigenvalue $\lambda$. Generally, the RPA study on pairing symmetry only works well for $U << U_c$, which will be our focus in the following.

The eigenvector($s$) $\Delta(\mathbf{k})$ for each eigenvalue $\lambda$ introduced above form an irreducible representation of the $D_{3h}$ point group of K$_2$Cr$_3$As$_3$. Altogether, there are 10 different pairing symmetries, including 5 singlet pairings and 5 triplet ones, which are listed in Table.II. Here, the $(d_{x^2-y^2}, d_{xy}), (d_{x^2-y^2}, d_{xy}) \cdot p_z, (p_x, p_y)$ and $(p_x, p_y) \cdot p_z$ symmetries each form a 2D representation of the point group with degenerate pairing eigenvalues. Generally, below $T_c$, the two degenerate eigenvectors of these representations would be mixed as $d_{x^2-y^2} \pm id_{xy}, (d_{x^2-y^2} \pm id_{xy}) \cdot p_z, p_z \pm ip_y$ and $(p_x \pm ip_y) \cdot p_z$. There are two different $f$-wave symmetries, i.e. $f_{y^3-3xy^2}$ and $f_{y^3-3xy^2}$. While the gap functions of both $f$-waves change sign with every 60° degree rotation, their nodal lines are different by 30° rotation. Due to the absence of spin orbital coupling (SOC) in our model, each of the five triplet pairings possesses three degenerate components, i.e. $\uparrow \uparrow, \downarrow \downarrow, (\uparrow \downarrow + \downarrow \uparrow)$, which we shall not distinguish in the following unless otherwise stated. Note that the concrete formulism of the gap function of each symmetry is not given a prior to the RPA calculation. Instead, they should be solved from the gap equation (10).

Our RPA calculations were performed on $18 \times 18 \times 200$ and $30 \times 30 \times 300$ lattices with periodic boundary conditions. The consistence between the two results indicates our RPA result converges in the thermodynamic limit. The $U$-dependence of the largest eigenvalue $\lambda$ for the five stronger pairing symmetries is shown in Fig.4 for (a). $J_H = 0.2U$ and (b), $J_H = 0.4U$. Clearly, all these eigenvalues increase promptly with the enhancement of $U$ and would diverge for $U \rightarrow U_c$. For $J_H = 0.2U$ shown in (a), the $p_z$-wave is the leading pairing symmetry and dominates other symmetries; for $J_H = 0.4U$ shown in (b), the $f_{y^3-3xy^2}$-wave becomes the leading one, with the $(p_z, p_y)$- and $p_z$- waves to be close candidates. Fig.5(a) shows the $\frac{1}{\lambda}$-dependence of these eigenvalues for fixed $U = 0.1eV$ for (a). RPA and (b). mean-field results.

| singlet | triplet |
|---------|---------|
| $s$     | $p_z$   |
| $(d_{x^2-y^2}, d_{xy})$ | $(d_{x^2-y^2}, d_{xy}) \cdot p_z$ |
| $(p_x, p_y)$ & $(p_x, p_y)$ |
| $\int_{p_x} f_{y^3-3xy^2} \cdot p_z$ | $\int_{p_x} f_{y^3-3xy^2} \cdot p_z$ |
| $\cdot p_z$ | $\cdot p_z$ |

TABLE II. The ten possible pairing symmetries for K$_2$Cr$_3$As$_3$, among which five are spin-singlet while the left are spin-triplet.

![FIG. 4.](image1.png) The $U$-dependence of the largest eigenvalues $\lambda$ for different pairing symmetries for (a), $J_H = 0.2U$ and (b), $J_H = 0.4U$. Only the five stronger symmetries are shown.

![FIG. 5.](image2.png) The $\frac{1}{\lambda}$-dependence of the largest eigenvalues $\lambda$ for the five stronger pairing symmetries for fixed $U = 0.1eV$ for (a). RPA and (b). mean-field results.

C. Triplet pairings

It’s interesting to find from the above shown RPA results that all the dominating pairing symmetries in different regimes, e.g. the $p_z$, $f_{y^3-3xy^2}$ and $(p_z, p_y)$ symmetries are triplet pairings. The ground state phase-diagram in the $U - \frac{J_H}{U}$ plane is shown in Fig.6, where three possible phases are present. For $U > U_c$ (which is around 0.16-0.24 eV and is $J_H/U$-dependent), the intra-sublattice FM SDW order emerges. For $U < U_c$, two triplet pairings, i.e. the $p_z$ and $f_{y^3-3xy^2}$-waves emerge, which are separated by the critical value $r_c$ (which is $U$-dependent) for $r = \frac{J_H}{U}$. For $r$ below or above $r_c$, the $p_z$- or $f_{y^3-3xy^2}$-wave pairing is the leading pairing symmetry respectively. In the limit of $U \rightarrow 0$, we have $r_c \rightarrow \frac{1}{2}$, with the enhancement of $U$, $r_c$ first decreases slightly and then drops near the SDW critical point. In our focused
regime of $U \ll U_c$ in RPA, we have $r_c \approx \frac{1}{3}$. While both gap functions have node lines, they are along $k_y = \pm \sqrt{3} k_x$, 0 directions for the $f_{y^3z^2-y^2}$-wave pairing and in the $k_z = 0$ plane for the $p_{z}$-wave pairing.

Note that the dominating part of the leading $f_{y^3z^2-y^2}$ and sub-leading $(p_x, p_y)$-wave pairings in the regime $r > r_c \approx \frac{1}{3}$ are on-site inter-orbital pairings driven by the Hund’s rule coupling. To clarify this point, let’s turn off the second-order perturbation process of RPA, and study the problem in the mean-field level. The Hubbard-Hund’s interaction (4) can be decoupled in the real space as the sum of an inter-orbital pairing part

$$H_{\text{int}} = (V + J_H) \sum_{i<\mu < \nu} \hat{\Delta}_{\mu \nu \sigma \sigma'} \langle i \mid \hat{\Delta}_{\mu \nu \sigma}^+ \rangle \langle \nu \mid \hat{\Delta}_{\mu \nu \sigma'}^+ \rangle$$

and an intraorbital pairing part

$$H_{\text{intra}} = U \sum_{i \mu < \nu} \hat{\Delta}_{\mu \mu \sigma \sigma}^\dagger \hat{\Delta}_{\mu \mu \sigma \sigma} \langle i \mid \hat{\Delta}_{\mu \mu \sigma}^+ \rangle \langle \mu \mid \hat{\Delta}_{\mu \mu \sigma}^+ \rangle$$

and its mean-field study below on the $t – J$ model of the system. Since the Hund’s rule coupling driven pairings don’t include the $p_{z}$-wave, as suggested by Fig.5(b), this pairing is purely driven by the intra-sublattice FM spin fluctuations reflected in Fig.3(b).

The distribution of the relative gap function of the leading $p_{z}$-wave pairing obtained by RPA for $U = 0.1 eV$, $J_H = 0.1 U$ is shown on the FSs in Fig.8(d), (e) and (f). This orbital-singlet spin-triplet pairing state is mirror-reflection even about the $z$-axis, causing line gap nodes in the direction of $k_y = 0, \pm \sqrt{3} k_x$. While the pairing amplitudes on the $\beta$- and $\gamma$-FSs are comparable, that on the $\alpha$-FS is much weaker as the dominating orbital component on that FS is $d_{z^2}$, which nearly doesn’t take part in the pairing, as suggested by the above mean-field analysis.

In real material, the situation $\frac{J_H}{U} > r_c \approx \frac{1}{3}$ can hardly be realized because the orbitals in our effective models are the molecule orbitals that are equally distributed on the three Cr$^2$ atoms and thus the effective value of $J_H$ should be small. Therefore, the realized pairing symmetry in K$_2$Cr$_3$As$_3$ might more probably be the $p_{z}$-wave pairing displayed in the regime $r < r_c$ in the phase-diagram Fig.6, which is consistent with the mean-field study below on the $t – J$ model of the system.

![FIG. 6. (color online). The ground state phase-diagram in the $U - \frac{J_H}{U}$ plane.](image)

for the strong-coupling limit, we consider the superexchange interaction Eq.(5). The Hamiltonian of our model is

$$H = H_{\text{TB}} + H_1,$$

where the interaction term $H_1$ is give by Eq.(5). Since this material is far from Mott-insulator, we neglect the no-double-
where $\Psi$ and $H$.

**FIG. 7.** (color online). (a) Dominating pairing pattern in the real space for the $f_{y^3-z^3}t_{2g}$-wave pairing shown in Fig.6. Note that only the $\uparrow\uparrow$ pairing component is shown. The $k_z$-dependence of the relative gap function of $p_z$-wave pairing averaged on the FSs for (b). RPA result for the Hubbard-Hund model with $J_{\mu}/U = 0.1$ and (c) Mean-field result for the $t-J$ model.

Occupance constraint on this $t-J$ model and perform a mean-field study on it.

In terms of real-space pairing operators, this interaction Hamiltonian can be written as,

$$
H_J = \frac{3}{4} J_2 \sum_{(ij),\mu} P^\dagger_{ij\mu} P_{ij\mu} - \frac{1}{4} J_2 \sum_{(ij),\mu\gamma} T^\dagger_{ij\mu,\gamma} T_{ij\mu,\gamma},
$$

$$
P^\dagger_{ij\mu} = \frac{1}{\sqrt{2}} (c^\dagger_{i\mu\uparrow} c^\dagger_{j\mu\downarrow} - c^\dagger_{i\mu\downarrow} c^\dagger_{j\mu\uparrow}),
$$

$$
T^\dagger_{ij\mu,\gamma} = \begin{cases} 
\frac{1}{\sqrt{2}} (c^\dagger_{i\mu\uparrow} c^\dagger_{j\mu\downarrow} + c^\dagger_{i\mu\downarrow} c^\dagger_{j\mu\uparrow}) & \gamma = 1, \\
\frac{1}{\sqrt{2}} c^\dagger_{i\mu\uparrow} c^\dagger_{j\mu\uparrow} & \gamma = 0, \\
\frac{1}{\sqrt{2}} c^\dagger_{i\mu\uparrow} c^\dagger_{j\mu\downarrow} & \gamma = -1.
\end{cases} 
$$

Here $P_{ij\mu}$ is the spin-singlet pairing operator and $T_{ij\mu,\gamma}$ is the spin-triplet one with $S_z = \gamma$. From Eq. (14), it’s clear that for $J_2 > 0$ ($J_2 < 0$), spin-triplet (singlet) pairing will be favorable. In $K_2Cr_3As_3$, FM intra-sublattice spin correlation leads to $J_2 > 0$, thus we only consider the triplet pairing terms. Due to the absence of SOC in our model, the pairing states in $S_z = -1, 0, 1$ channels are degenerate, which allows us to only consider the $S_z = 0$ channel in the following.

Defining $\Delta_{\mu} = -i J_2 (T_{ix+xz+0,\mu})/\sqrt{2}$ as the (real) pairing order parameters, the above $H_J$ term can be decoupled as

$$
H_J = \frac{\sqrt{2}}{2} \sum_{(ij),\mu} \left( -i \Delta_{\mu} T^\dagger_{ij\mu,0} + h.c. \right) + \frac{2N}{J_2} \sum_{\mu} |\Delta_{\mu}|^2.
$$

In combination with the tight-binding part, the total mean-field Hamiltonian in the momentum space is

$$
H_{mf} = \sum_k \Psi^\dagger(k) H(k) \Psi(k) + \frac{2N}{J_2} \sum_{\mu} |\Delta_{\mu}|^2,
$$

$$
H(k) = \begin{pmatrix} h(k) & \Delta_{\uparrow\uparrow}(k) \\ \Delta_{\uparrow\downarrow}(k) & -h^*(k) \end{pmatrix},
$$

$$
\Delta_{\uparrow\uparrow}(k) = \begin{pmatrix} \Delta_1(k) \\ \Delta_2(k) \\ \Delta_3(k) \end{pmatrix},
$$

where $\Psi^\dagger(k) = [c^\dagger_{1\uparrow}(k), c^\dagger_{2\uparrow}(k), c^\dagger_{3\uparrow}(k), c_{1\downarrow}(-k), c_{2\downarrow}(-k), c_{3\downarrow}(-k)]$ and $\Delta_{\uparrow\downarrow}(k) = \Delta_{\downarrow\uparrow}(k) = \Delta_{\uparrow\uparrow}(k)$. Note that a constant $\sum_k \{h_{\mu\mu}(k) - \mu_c\}$, which has no contribution to the dynamics, has been neglected on the above.

The above mean-field Hamiltonian can be solved via diagonalizing $H(k)$ by a unitary transformation: $U^\dagger(k) H(k) U(k) = \text{Diag}(E_i)$, with the eigenvalues $E_i = -E_{i+3} > 0 (i = 1, 2, 3)$. The self-consistent gap equations are,

$$
\Delta_{\mu} = -\frac{J_2}{2N} \sum_k \sum_{\alpha=1}^{6} U^\dagger_{\mu+3,\alpha}(k) U_{\mu,\alpha}(k) f[|E_\alpha(k)| \sin k_z]
$$

where $f(E)$ is the Fermi-Dirac distribution function and we only study the case of zero temperature. Solving these gap equations, we plot the ground state energy $E_G$ and the three gap amplitudes $\Delta_{\mu} (\mu = 1, \cdots, 3)$ as function of $J_2$ in Fig.9(a) and (b) respectively. Clearly, the gap amplitudes of $d_{x^2-y^2}$ and $d_{x\pm y}$ orbitals, belonging to the $E'$ irreducible representation, are equal and dominate that of $d_{z^2}$, which is consistent with the RPA result. Furthermore, there is no phase difference between the SC order parameters of the three orbitals.

Fig.10 shows the distribution of the pairing gap function $J_2 \langle c_{\mu\uparrow}(-k) c_{\alpha\uparrow}(k) \rangle = J_2 \sum_{\mu} \langle c_{\mu\uparrow}(-k) c_{\alpha\uparrow}(k) \rangle |\xi_{\mu\alpha}(k)|^2 = 4 \sum_{\mu} \Delta_{\mu} |\xi_{\mu\alpha}(k)|^2 \sin(k_z)$ over the FSs in the Brillouin-Zone. Here $\alpha$ is the band index and only intra-band pairing is shown. Clearly, due to the same sign among different $\Delta_{\mu}$, there is no sign change of the gap function all over the regime $k_z > 0$ and gap nodes only exist in the $k_z = 0$ plane in the 3D $\gamma$-band. Obviously, the pairing symmetry of this superconducting state is $p_z$ since its gap function is reflection odd respect to the mirror plane $k_z = 0$ and is rotation symmetric about the $z$-axis. What’s more, due to the dominance of $\Delta_{\alpha z}, \alpha$-band over $\Delta_1$, the pairing amplitude on the Q1D $\beta$-band is obviously stronger than that on the $\gamma$-band. Fig.11(a) shows the gap distribution over the FSs in the $k_{z} - k_{z'}$ plane with $k_{y} = 0$ and Fig.7(c) shows the averaged gap function $\langle \Delta_{\alpha z}(k) \rangle_{FS}$ on the FSs with fixed $k_z$ in comparison with the same quantity obtained from RPA in Fig.7(a). The obtained gap structure here is well consistent with the results of RPA introduced above. In Fig.11(b), we plot the DOS in the superconduct-
FIG. 8. (color online). The relative gap function distribution over the three FSs for the \( f_{y^-z^+} \) wave state (top panel) and the \( p_z \) wave state (bottom panel) in RPA calculations. Fig. (a) (d), (b) (e) and (c) (f) are for \( \alpha, \beta \) and \( \gamma \) FSs. With \( U = 0.1 \) eV, the adopted Hund couplings are \( J_H = 0.4U \) and \( J_H = 0.1U \) for \( f_{y^-z^+} \) and \( p_z \) wave states in the calculations.

FIG. 9. (color online). The ground state energy \( E_G \) (a) and gap amplitudes \( \Delta_\mu \) (b) for triplet pairing in \( S = 0 \) channel as a function of \( J_2 \).

The pairing state with \( \Delta_1 = 2.5\text{meV}, \Delta_2 = \Delta_3 = 10\text{meV} \) obtained for \( J_2 = 0.4\text{eV} \), scaled by a factor of 2 for clarity. The DOS displays a V-shaped structure, which can be detected by tunneling experiments.

Note that there can be another \( p_z \)-wave solution of the gap equation with the gap sign of \( A_1^\dagger - d_{z^2} \)-orbital different from that of \( E'(d_{xy}, d_{x^2-y^2}) \)-orbitals. Due to the phase difference and orbital characters, the gaps on the \( \alpha \)- and \( \gamma \)-FSs are strongly anisotropic and line nodes appear on the \( \alpha \)-FS; but the gap on the \( \beta \)-FS is quite uniform. Compared with the former case, there is an additional coherence peak in DOS. This state, however, is energetically unfavorable in our results, due to the extra gap nodes on the FS.\(^{32}\)

V. DISCUSSION AND CONCLUSION

In conclusion, we have performed combined weak-coupling RPA and strong coupling mean-field study on the pairing symmetry of \( \text{K}_2\text{Cr}_3\text{As}_3 \), starting from the three-band tight-binding model. The weak-coupling RPA study on the Hubbard-Hund model reveals two possible triplet pairings in different parameter regimes. For \( r \equiv \frac{J_H}{U} > r_c \approx \frac{1}{4} \), on-site inter-orbital pairing (between \( d_{xy} \) and \( d_{x^2-y^2} \)) triplet SC would be driven by the Hund’s rule coupling. This pairing has nodal lines in the direction of \( k_y = 0, \pm \sqrt{3k_z} \). However, as the orbitals in our effective models are the molecule orbitals that are equally distributed on the three Cr2 atoms, the effective value of \( J_H \) should be small. Therefore, \( r \leq r_c \) is a more realistic parameter. For \( r \leq r_c \), the Q1D \( p_z \)-wave pairing would be driven by the strong intra-sublattice FM spin fluctuations. The \( p_z \)-wave pairing state obtained by RPA for the Hubbard-Hund model is in well consistency with that obtained by mean-field study on the \( t-J \) model in the strong coupling limit. The gap function of this pairing state doesn’t change sign in the \( k_z > 0 \) (or \( k_z < 0 \)) regime on the FSs and the only line gap nodes lie on the \( k_z=0 \) plane on the 3D \( \gamma \)-FS. The pairing amplitude on the Q1D \( \beta \)-FS dominates those on the Q1D \( \alpha \)-FS and the 3D \( \gamma \)-FS, revealing the dominating part of this pairing to be the Q1D pairing within the \( E'(d_{xy}, d_{x^2-y^2}) \) orbitals along the \( z \)-axis.

In \( \text{K}_2\text{Cr}_3\text{As}_3 \), the weak SOC will lift up the degeneracy among the three components of the triplet pairings. The only symmetry allowed on-site SOC term added to our three-band tight-binding model takes the following formulism,

\[
H_{SOC} = i\lambda_{so} \sum_{k\sigma} \sum_{\kappa \sigma} \left[ c_{\kappa \sigma}(k) c_{\kappa \sigma}(k) - c_{\kappa \sigma}(k) c_{\kappa \sigma}(k) \right], \quad (18)
\]

where a tiny coupling constant \( \lambda = 2\text{meV} \) is adopted in our
Figure 10. (color online). The gap function distribution over the three FSs with $\Delta_{0}^{+1} = 0.0025$ eV, $\Delta_{x-y}^{\pm 1} = 0.01$ eV, obtained for $J_2 = 0.4$ eV, scaled by a factor of 2 for clarity. Fig. (a), (b) and (c) are for $\alpha$, $\beta$ and $\gamma$ FSs.

Figure 11. (color online). The SC gap function distribution over the FSs in the $k_y = 0$ plane (a) and density of states (b) with $\Delta_{0}^{+1} = 0.0025$ eV, $\Delta_{x-y}^{\pm 1} = 0.01$ eV, obtained for $J_2 = 0.4$ eV, scaled by a factor of 2 for clarity.

Figure 12. (color online). The $\frac{J_2}{U}$-dependence of the largest eigenvalues $\lambda$ for the $p_{y}$- and $f_{y}-y$ wave pairings for fixed $U = 0.1$ eV and $\Delta_{so} = 2$ meV for the $S_{z} = 0$ ($\uparrow\downarrow$) component and $S_{z} = \pm 1$ one.
below the critical one and the RPA is valid.
Appendix A: The tight-binding model

From the bandstructure of DFT, we find that there are five bands near the Fermi level and the orbital characters are mainly $d_{z^2}$, $d_{xy}$, and $d_{x^2−y^2}$. To obtain an effective model, we consider a virtual atom at the center of each Cr triangle with three orbitals, which have the same symmetry as $A'_1(d_{z^2})$, and $E'(d_{xy}, d_{x^2−y^2})$, as shown in Fig.1 in the main text. Now the structure of $K_2Cr_3As_3$ is simplified and every chain has two sublattices. Due to the asymmetry of K atoms and the absence of inversion symmetry, there are two inequivalent sublattices which are labeled as $A$ and $B$ respectively. The tight binding Hamiltonian is

$$h(k) = \begin{pmatrix}
    h_{11}^{AA} & h_{11}^{AB} & h_{11}^{AB} \\
    h_{11}^{BA} & h_{11}^{BB} & h_{11}^{BB} \\
    h_{11}^{AA} & h_{11}^{BB} & h_{11}^{AB}
\end{pmatrix},$$

(A1)

where the unshown matrix elements can be obtained from the shown ones by Hermicity of the $h(k)$ matrix. On the above, the orbital indices 1, 2 and 3 denote $d_{z^2}$, $d_{xy}$ and $d_{x^2−y^2}$ respectively.

Let $x = \frac{\sqrt{2}}{2}k_x a_0$, $y = \frac{1}{2}k_y a_0$ and $z = \frac{1}{2}k_z c_0$, the Hamiltonian matrix elements are as following.

(1) The hoppings between $A'_1(d_{z^2})$ orbital and $A'_1(d_{z^2})$ orbital lead to the following matrix elements of $h(k)$

$$h_{11}^{AA/BB} = \epsilon_{1/3} + 2 \sum_{i=1}^{3} s_{z_i}^{11} \cos 2iz + (s_y^{11} + 2s_{yz}^{11} \cos 2z)(2\cos 2y + 4\cos x\cos y)$$

$$h_{11}^{AB} = 2 \sum_{i=1}^{4} s_{z_i}^{11} \cos[(2i - 1)z] + 2s_{yz}^{11} \cos(2\cos 2y + 4\cos x\cos y)$$

$$h_{11}^{BA} = h_{11}^{AB\ast} = h_{11}^{AB}.$$  

(A2)

(2) The hoppings between $A'_1(d_{z^2})$ orbital and $E'(d_{xy}, d_{x^2−y^2})$ orbitals lead to

$$h_{12}^{AA/BB} = 2i s_{1y}^{12} \sin 2y + 2i s_{1y}^{12} \sin y \cos x + 2\sqrt{3} s_{2y}^{12} \sin y \sin x$$

$$h_{21}^{AA/BB} = h_{12}^{AB\ast}$$

$$h_{13}^{AA/BB} = 2s_{2y}^{12} \cos 2y - 2i\sqrt{3} s_{1y}^{12} \cos y \sin x - 2s_{2y}^{12} \cos y \cos x$$

$$h_{31}^{AA/BB} = h_{13}^{AB\ast}$$

$$h_{12}^{AB} = 2 \cos z(2i s_{1y}^{12} \sin 2y + 2i s_{1y}^{12} \sin y \cos x + 2\sqrt{3} s_{2y}^{12} \sin y \sin x)$$

$$h_{21}^{AB} = h_{12}^{AB\ast}, h_{12}^{BA} = h_{12}^{AB\ast}, h_{21}^{BA} = h_{12}^{AB\ast}$$

$$h_{13}^{AB} = 2 \cos z(2s_{2y}^{12} \cos 2y - 2\sqrt{3} s_{1y}^{12} \cos y \sin x - 2s_{2y}^{12} \cos y \cos x)$$

$$h_{31}^{AB} = h_{13}^{AB\ast}, h_{13}^{BA} = h_{13}^{AB\ast}, h_{31}^{BA} = h_{13}^{AB\ast}.$$  

(A3)
TABLE III. The hopping parameters (in unit of eV) along c axis to fit the DFT results in the six-orbital model. The onsite energies are $\epsilon_1 = 1.9080$eV, $\epsilon_2 = 2.0407$eV, $\epsilon_3 = 1.9722$eV and $\epsilon_4 = 1.9412$eV.

| $s^\alpha_1$ | $i = z, 1$ | $i = z, 2$ | $i = z, 3$ | $i = z, 4$ | $i = z, 5$ | $i = z, 6$ | $i = z, 7$ |
|-------------|------------|------------|------------|------------|------------|------------|------------|
| $\alpha = 11$ | 0.1749     | 0.1568     | 0.0301     | -0.0532    | -0.0097    | -0.0189    | 0          |
| $\alpha = 22$ | -0.0041    | 0.1734     | 0.0019     | -0.0452    | 0          | 0          | -0.004     |

TABLE IV. The in-plane hopping parameters (in unit of eV) to fit the DFT results in the six-orbital model.

| $s^\alpha_2$ | $i = 1y$ | $i = 2y$ | $i = 11y$ | $i = 22y$ | $i = 12y$ | $i = y$ | $i = yz$ |
|-------------|----------|----------|------------|-----------|-----------|--------|---------|
| $\alpha = 12$ | 0.0206   | 0        |   0       |   0       |   0       | 0      | 0       |
| $\alpha = 22$ | 0.0066   | 0.0133   | 0.0460    | -0.0191   | 0.0062    | 0.0159 | -0.0218 |
| $\alpha = 22$ | 0.0061   | -0.0109  | 0.0159    | -0.0218   | -0.0031   |        |          |

(3) The hoppings between $E'(d_{xy}, d_{x^2-y^2})$ orbitals and $E'(d_{xy}, d_{x^2-y^2})$ orbitals lead to

$$
h_{22}^{AA/BB} = \epsilon_2/4 + 2s_1^{22}\cos 2y + (s_1^{11} + 3s_2^{22})\cos x\cos y + 2\sum_{i=1}^{2} s_{2i,2i} \cos 2iz
$$

$$
h_{23}^{AA/BB} = \sqrt{3}(s_1^{11} - s_2^{22})\sin x\sin y + 2i s_{12y}^{22}\sin 2y - 4i s_{12y}^{22}\cos x\sin y
$$

$$
h_{32}^{AA/BB} = h_{23}^{AA/BB,*}
$$

$$
h_{33}^{AA/BB} = \epsilon_2/4 + 2s_2^{22}\cos 2y + (s_1^{11} + s_2^{22})\cos x\cos y + 2\sum_{i=1}^{2} s_{2i,2i} \cos 2iz
$$

$$
h_{22}^{AB} = 2\cos z \left[ 2s_1^{22}\cos 2y + (s_1^{11} + s_2^{22})\cos x\cos y \right] + 2\sum_{i=1}^{3} s_{i,2i-1}\cos (2i-1)z
$$

$$
h_{22}^{BA} = h_{22}^{AB,*} = h_{22}^{AB}
$$

$$
h_{23}^{AB} = 2\cos z \left[ \sqrt{3}(s_1^{11} - s_2^{22})\sin x\sin y + 2i s_{12y}^{22}\sin 2y - 4i s_{12y}^{22}\cos x\sin y \right]
$$

$$
h_{32}^{AB} = h_{23}^{AB,*}, h_{23}^{BA} = h_{23}^{AB,*}, h_{32}^{BA} = h_{23}^{AB,*}
$$

$$
h_{33}^{AB} = 2\cos z \left[ 2s_2^{22}\cos 2y + (s_1^{11} + s_2^{22})\cos x\cos y \right] + 2\sum_{i=1}^{3} s_{i,2i-1}\cos (2i-1)z
$$

$$
h_{33}^{BA} = h_{33}^{AB,*} = h_{33}^{AB}
$$

The hopping parameters are obtained by least-square-root fitting of the above tight binding model to the DFT band structure. These parameters are given in Table III and IV and the band structures of tight binding model and DFT are shown in Fig.13. Near the Fermi level, the tight binding band fits well with that of DFT.

As the states near the Fermi level are dominantly contributed by Cr2, we can drop Cr1 and further simplify our model to a three-band one. The Hamiltonian of the resulting tight-binding model is,

$$
h(\mathbf{k}) = \begin{pmatrix} h_{11} & h_{12} & h_{13} \\ h_{12} & h_{22} & h_{23} \\ h_{13} & h_{23} & h_{33} \end{pmatrix}
$$

(A5)

Again, the unshown matrix elements are obtained by Hermicity of the $h(\mathbf{k})$ matrix. The formula of the matrix elements of $h(\mathbf{k})$ are given in Eq.(3), with the hopping parameters in this model given in Table I in the main text. The band structure of this model is shown in Fig.2 in the main text, in comparison with that of DFT. This tight binding band also fits well with that of DFT near the Fermi level. Furthermore, the orbital characters, shown in Fig.14(a), are consistent with those of DFT\textsuperscript{20}. Fig.14 shows the weight distribution of $E'$ orbitals on Fermi surfaces in $k_y = 0$ plane.
FIG. 13. (color online). The band structures of DFT (gray lines) and tight binding (red lines).

FIG. 14. (color online). Orbital characters. (a) The orbital characters of the band structure of three-band tight binding model. (b) The weight distribution of $E'(d_{xy}, d_{z^2-y^2})$ orbitals on Fermi surfaces in $k_y = 0$ plane.

Appendix B: The multi-orbital RPA approach

The Hamiltonian adopted in our calculations is

$$H = H_{t-b} + H_{int}$$

$$H_{int} = U \sum_{i, \mu} n_{i \mu \uparrow} n_{i \mu \downarrow} + V \sum_{i \mu < \nu} n_{i \mu} n_{i \nu} + J_H \sum_{i, \mu < \nu} \left[ \sum_{\sigma} c_{i \mu \sigma}^+ c_{i \nu \sigma}^+ c_{i \nu \sigma} c_{i \mu \sigma} + (c_{i \mu \uparrow}^+ c_{i \mu \downarrow}^+ c_{i \nu \downarrow} c_{i \nu \uparrow} + h.c.) \right]$$  (B1)
Let's first study the case without SOC, then consider the inclusion of on-site SOC for $K_2Cr_3As_3$.

1. Without SOC

Let's define the following bare susceptibility for the non-interacting case ($U = V = J_H = 0$),

$$\chi^{(0)}_{l_1 l_2} (q, \tau) \equiv \frac{1}{N} \sum_{k_1, k_2} \left< T_{\tau} \xi_{c_{l_1}(k_1, \tau)} \xi_{c_{l_2}(k_1 + q, \tau)} c_{c_{l_1}(k_2 + q, 0)} c_{c_{l_4}(k_2, 0)} \right>_0,$$

where $l_i (i = 1, \cdots, 4)$ denote orbital indices. The explicit formulism of $\chi^{(0)}$ in the momentum-frequency space is,

$$\chi^{(0)}_{l_1 l_2} (q, i\omega_n) = \frac{1}{N} \sum_{k, \alpha, \beta} \xi_{l_1}^\alpha(k) \xi_{l_2}^\beta(k) \xi_{l_3}^\beta*(k + q) \xi_{l_4}^\alpha*(k + q) \frac{n_F(\varepsilon_k^\beta) - n_F(\varepsilon_{k+q}^\beta)}{i\omega_n + \varepsilon_k - \varepsilon_{k+q}},$$

where $\alpha/\beta = 1, \ldots, 3$ are band indices, $\varepsilon_k^\alpha$ and $\xi_{l_i}^\alpha (k)$ are the $\alpha-$th eigenvalue and eigenvector of the $h(k)$ matrix respectively and $n_F$ is the Fermi-Dirac distribution function.

FIG. 15. (color online). Feyman’s diagram for the renormalized susceptibilities in the RPA level.
When interactions turn on, we define the spin ($\chi^{(s)}$) and charge ($\chi^{(c)}$) susceptibility as follows,

$$\chi^{(c)}_{l_3,l_4}(q,\tau) = \frac{1}{2N} \sum_{k_1,k_2,\sigma_1,\sigma_2} \langle T_{c_{l_3}c_{l_4}}(k_1,\tau)c_{l_3\sigma_1}(k_1+q,\tau)c_{l_4\sigma_2}^+(k_2+q,0)c_{l_4\sigma_2}(k_2,0) \rangle,$$

$$\chi^{(s)}_{l_3,l_4}(q,\tau) = \frac{1}{2N} \sum_{k_1,k_2,\sigma_1,\sigma_2} \sigma_1\sigma_2 \langle T_{c_{l_3}c_{l_4}}(k_1,\tau)c_{l_3\sigma_1}(k_1+q,\tau)c_{l_4\sigma_2}^+(k_2+q,0)c_{l_4\sigma_2}(k_2,0) \rangle,$$

$$\chi^{(s^+)}_{l_3,l_4}(q,\tau) = \frac{1}{N} \sum_{k_1,k_2} \langle T_{c_{l_3}c_{l_4}^\dagger}(k_1,\tau)c_{l_3\sigma_1}(k_1+q,\tau)c_{l_4\sigma_2}^+(k_2+q,0)c_{l_4\sigma_2}(k_2,0) \rangle,$$

$$\chi^{(s^-)}_{l_3,l_4}(q,\tau) = \frac{1}{N} \sum_{k_1,k_2} \langle T_{c_{l_3}c_{l_4}^\dagger}(k_1,\tau)c_{l_3\sigma_1}(k_1+q,\tau)c_{l_4\sigma_2}^+(k_2+q,0)c_{l_4\sigma_2}(k_2,0) \rangle.$$  

(B4)

Note that in non-magnetic state we have $\chi^{(s)} = \chi^{(s^-)} = \chi^{(s^+)} \equiv \chi^{(s)}$, and when $U = V = J_H = 0$ we have $\chi^{(c)} = \chi^{(s)} = \chi^{(0)}$. In the RPA level, the renormalized spin/charge susceptibilities for the system are,

$$\chi^{(s)}(q,\nu) = \left[ I - \chi^{(0)}(q,\nu) U^{(s)} \right]^{-1} \chi^{(0)}(q,\nu),$$

$$\chi^{(c)}(q,\nu) = \left[ I + \chi^{(0)}(q,\nu) U^{(c)} \right]^{-1} \chi^{(0)}(q,\nu),$$

(B5)

where $\chi^{(s,c)}(q,\nu)$, $\chi^{(0)}(q,\nu)$ and $U^{(s,c)}$ are operated as $9 \times 9$ matrices (the upper or lower two indices are viewed as one number) with elements of the matrix $U^{(s,c)}$ to be

$$U^{(s)}_{l_3,l_4} = \begin{cases} U, l_1 = l_2 = l_3 = l_4 \\ J_H, l_1 = l_2 \neq l_3 = l_4 \\ V, l_1 = l_4 \neq l_3 = l_2 \end{cases}$$

(B6)

$$U^{(c)}_{l_3,l_4} = \begin{cases} U, l_1 = l_2 = l_3 = l_4 \\ 2V - J_H, l_1 = l_2 \neq l_3 = l_4 \\ 2J_H - V, l_1 = l_4 \neq l_3 = l_2 \end{cases}$$

(B7)

The Feynman’s diagram of RPA is shown in Fig. 15. For repulsive Hubbard-interactions, the spin susceptibility is enhanced and the charge susceptibility is suppressed. Note that there is a critical interaction strength $U_c$, which depends on the ratio $J_H/U$. When $U > U_c$, the denominator matrix $I - \chi^{(0)}(q,\nu) U^{(s)}$ in Eq.(B5) will have zero eigenvalues for some $q$ and the renormalized spin susceptibility diverges there, which invalidates the RPA treatment. This divergence of spin susceptibility for $U > U_c$ implies magnetic order. When $U < U_c$, the short-ranged spin or charge fluctuations would mediate Cooper pairing in the system.

Let’s consider a Cooper pair with momentum/orbital $(k't, -k's)$, which could be scattered to $(kp, -kq)$ by exchanging charge.
or spin fluctuations. In the RPA level, the effective interaction induced by this process is as follow,

\[
V_{\text{eff}}^{\text{RPA}} = \frac{1}{N} \sum_{pqst, kk'} \Gamma_{st}^{pq}(k, k') c_{p}^{\dagger}(k) c_{q}^{\dagger}(-k)c_{s}(-k')c_{t}(k'),
\]  

(B8)

We consider the three processes in Fig.16 which contribute to the effective vertex \(\Gamma_{st}^{pq}(k, k')\), where (a) represents the bare interaction vertex and (b) (c) represent the second order perturbation processes during which spin or charge fluctuations are exchanged between a cooper pair. In the singlet channel, the effective vertex \(\Gamma_{st}^{pq}(k, k')\) is given as follow,

\[
\Gamma_{st}^{pq(s)}(k, k') = \left(\frac{U^{(c)}}{4} + \frac{3U^{(s)}}{4}\right)_{qs}^{pt} + \frac{1}{4} \left[3U^{(s)}\chi^{(s)}(k - k') U^{(s)}(k - k') U^{(c)}(k')\right]_{qs}^{pt} + \frac{1}{4} \left[3U^{(s)}\chi^{(s)}(k + k') U^{(s)}(k + k') U^{(c)}(k')\right]_{qt}^{ps},
\]

(B9)

while in the triplet channel, it is

\[
\Gamma_{st}^{pq(t)}(k, k') = \left(\frac{U^{(c)} - U^{(s)}}{4}\right)_{qs}^{pt} - \frac{1}{4} \left[U^{(s)}\chi^{(s)}(k - k') U^{(s)}(k - k') U^{(c)}(k')\right]_{qs}^{pt} + \frac{1}{4} \left[U^{(s)}\chi^{(s)}(k + k') U^{(s)}(k + k') U^{(c)}(k')\right]_{qt}^{ps},
\]

(B10)

Notice that the vertex \(\Gamma_{st}^{pq}(k, k')\) has been symmetrized for the singlet case and anti-symmetrized for the triplet case. Generally, we neglect the frequency-dependence of \(\Gamma\) and replace it by \(\Gamma_{st}^{pq}(k, k') \approx \Gamma_{st}^{pq}(k, k', 0)\). Usually, only the real part of \(\Gamma\) is adopted\(^{25,26}\)

Considering only intra-band pairings, we obtain the following effective pairing interaction on the FSs,

\[
V_{\text{eff}} = \frac{1}{N} \sum_{\alpha\beta, kk'} \Gamma^{\alpha\beta}(k, k') C_{\alpha}\dagger(k) c_{\beta}(-k) c_{\beta}(-k') c_{\beta}(k'),
\]

(B11)

where \(\alpha / \beta = 1, \cdots, 3\) are band indices and \(\Gamma^{\alpha\beta}(k, k')\) is

\[
\Gamma^{\alpha\beta}(k, k') = \sum_{pqst, kk'} \Gamma_{st}^{pq}(k, k', 0) \xi_{\alpha}^{q}\dagger(-k) \xi_{\beta}^{p}(k') \xi_{\alpha}^{p}(k') \xi_{\beta}^{q}(k).
\]

(B12)

From the effective pairing interaction (B11), one can obtain the following linearized gap equation\(^{25,26}\) to determine the \(T_c\) and the leading pairing symmetry of the system,

\[
- \frac{1}{(2\pi)^3} \sum_{\beta} \int d^2 k' || V^{\alpha\beta}(k, k') \Delta_{\alpha}(k') = \lambda \Delta_{\alpha}(k).
\]

(B13)

This equation can be looked upon as an eigenvalue problem, where the normalized eigenvector \(\Delta_{\alpha}(k)\) represents the relative gap function on the \(\alpha\)-th FS patches near \(T_c\), and the eigenvalue \(\lambda\) determines \(T_c\) via \(T_c = \text{cut off energy} e^{-1/\lambda}\), where the cut off energy is of order band-width. The leading pairing symmetry is determined by the largest eigenvalue \(\lambda\) of Eq.(B13).

2. With on-site SOC

When the on-site SOC described by Eq.(18) turns on, the free electron part of our model becomes,

\[
H_{\text{band}} = H_{\text{TB}} + H_{\text{SOC}}.
\]

(B14)

Let’s rewrite the interaction part in the following formulism,

\[
H_{\text{int}} = \sum_{i \mu \nu \xi \sigma_1 \sigma_2} U_{i \xi \sigma_2}^{\mu \nu \sigma_1} c_{i \mu \sigma_1}^{\dagger} c_{i \nu \sigma_2}^{\dagger} c_{i \theta \sigma_2} c_{i \xi \sigma_2}.
\]

(B15)
Here $U_{\theta \xi \sigma}^{\mu \nu}$ can be defined to satisfy the following symmetry relation,

\[
U_{\theta \xi \sigma}^{\mu \nu} = U_{\theta \xi \sigma}^{\mu \nu} = U_{\theta \xi \sigma}^{\mu \nu} = U_{\theta \xi \sigma}^{\mu \nu}.
\]

(B16)

where the $\bar{U}, \bar{V}$ tensors are obtained as

\[
\bar{U}_{l_3 l_4}^{l_1 l_2} = \begin{cases} \frac{U_{\theta \xi \sigma}^{l_2-l_1}}{4}, & l_1 = l_2 \\ \frac{U_{\theta \xi \sigma}^{l_1-l_2}}{4}, & l_1 = l_3 = l_4 \end{cases}
\]

\[
\bar{V}_{l_3 l_4}^{l_1 l_2} = \begin{cases} \frac{U_{\theta \xi \sigma}^{l_1}}, & l_1 = l_2 = l_3 = l_4 \\ \frac{U_{\theta \xi \sigma}^{l_1}}, & l_1 = l_3 \\ \frac{U_{\theta \xi \sigma}^{l_1}}, & l_1 = l_4 \end{cases}
\]

(B17)

Due to the $S_z$-conservation of the on-site SOC, let’s define the following six susceptibilities,

\[
\chi_{l_1 l_2 l_3 l_4}(q, \tau) = \frac{1}{N} \sum_{k_1 k_2} \left< T c_{l_1 \uparrow}^\dagger (k_1, \tau) c_{l_2 \uparrow}^\dagger (k_1 + q, \tau) c_{l_3 \downarrow}^\dagger (k_2 + q, 0) c_{l_4 \uparrow}^\dagger (k_2, 0) \right>
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, \tau) = \frac{1}{N} \sum_{k_1 k_2} \left< T c_{l_1 \downarrow}^\dagger (k_1, \tau) c_{l_2 \uparrow}^\dagger (k_1 + q, \tau) c_{l_3 \downarrow}^\dagger (k_2 + q, 0) c_{l_4 \uparrow}^\dagger (k_2, 0) \right>
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, \tau) = \frac{1}{N} \sum_{k_1 k_2} \left< T c_{l_1 \uparrow}^\dagger (k_1, \tau) c_{l_2 \downarrow}^\dagger (k_1 + q, \tau) c_{l_3 \downarrow}^\dagger (k_2 + q, 0) c_{l_4 \uparrow}^\dagger (k_2, 0) \right>
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, \tau) = \frac{1}{N} \sum_{k_1 k_2} \left< T c_{l_1 \downarrow}^\dagger (k_1, \tau) c_{l_2 \uparrow}^\dagger (k_1 + q, \tau) c_{l_3 \downarrow}^\dagger (k_2 + q, 0) c_{l_4 \uparrow}^\dagger (k_2, 0) \right>
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, \tau) = \frac{1}{N} \sum_{k_1 k_2} \left< T c_{l_1 \uparrow}^\dagger (k_1, \tau) c_{l_2 \uparrow}^\dagger (k_1 + q, \tau) c_{l_3 \downarrow}^\dagger (k_2 + q, 0) c_{l_4 \downarrow}^\dagger (k_2, 0) \right>
\]

(B18)

For $(U, V, J_H) = (0, 0, 0)$, we find the following formula of the bare susceptibilities,

\[
\chi_{l_1 l_2 l_3 l_4}(q, i \omega_n) = \frac{1}{N} \sum_{k, i \omega_m} G_{l_4 l_3}^{(0)}(k, i \omega_m) G_{l_2 l_1}^{(0)}(k + q, i \omega_m + i \omega_n) + \frac{1}{i \omega_n} \sum_{k, \alpha, \beta} \xi_{l_4 \uparrow}^\alpha (k) \xi_{l_3 \downarrow}^{\alpha \dagger} (k + q) \xi_{l_2 \uparrow}^{\beta \dagger} (k + q + \omega_n) n_F\left(\frac{\epsilon_{k+q}}{i \omega_n} + \frac{\epsilon_k}{2} \right) - n_F\left(\frac{\epsilon_{k+q}}{i \omega_n} + \frac{\epsilon_k}{2} \right)
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, i \omega_n) = \frac{1}{N} \sum_{k, i \omega_m} G_{l_4 l_3}^{(0)}(k, i \omega_m) G_{l_2 l_1}^{(0)}(k + q, i \omega_m + i \omega_n)
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, i \omega_n) = \frac{1}{N} \sum_{k, i \omega_m} G_{l_4 l_3}^{(0)}(k, i \omega_m) G_{l_2 l_1}^{(0)}(k + q, i \omega_m + i \omega_n)
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, i \omega_n) = \frac{1}{N} \sum_{k, i \omega_m} G_{l_4 l_3}^{(0)}(k, i \omega_m) G_{l_2 l_1}^{(0)}(k + q, i \omega_m + i \omega_n)
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, i \omega_n) = \frac{1}{N} \sum_{k, i \omega_m} G_{l_4 l_3}^{(0)}(k, i \omega_m) G_{l_2 l_1}^{(0)}(k + q, i \omega_m + i \omega_n)
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, i \omega_n) = \frac{1}{N} \sum_{k, i \omega_m} G_{l_4 l_3}^{(0)}(k, i \omega_m) G_{l_2 l_1}^{(0)}(k + q, i \omega_m + i \omega_n)
\]

\[
\chi_{l_1 l_2 l_3 l_4}(q, i \omega_n) = \frac{1}{N} \sum_{k, i \omega_m} G_{l_4 l_3}^{(0)}(k, i \omega_m) G_{l_2 l_1}^{(0)}(k + q, i \omega_m + i \omega_n)
\]

(B19)

When interactions turn on, we obtain the following Dyson equations for these susceptibilities in the RPA level, represented
by the Feynman’s diagram shown in Fig. 15

\[
\begin{align*}
\chi^1 &= \chi^{1(0)} - \chi^{1(0)} \left(4\bar{U}\chi^1 + 2\bar{V}\chi^3\right) \\
\chi^2 &= -\chi^{1(0)} \left(4\bar{U}\chi^2 + 2\bar{V}\chi^4\right) \\
\chi^3 &= -\chi^{4(0)} \left(4\bar{U}\chi^3 + 2\bar{V}\chi^1\right) \\
\chi^4 &= \chi^{4(0)} - \chi^{4(0)} \left(4\bar{U}\chi^4 + 2\bar{V}\chi^2\right) \\
\chi^5 &= \chi^{5(0)} - \chi^{5(0)} \left(4\bar{U} - 2\bar{V}\right)\chi^5 \\
\chi^6 &= \chi^{6(0)} - \chi^{6(0)} \left(4\bar{U} - 2\bar{V}\right)\chi^6.
\end{align*}
\] (B20)

Here \(\bar{U}\) and \(\bar{V}\) are operated as matrices with the upper or lower two tensor indices taken as one number. These equations are easily solved.

The effective interaction vertex \(\Gamma_{\sigma_1\sigma_2}^{pq\sigma_1}(k, k')\) is obtained from the summation of the three processes listed in Fig. 16,

\[
\Gamma_{\sigma_1\sigma_2}^{pq\sigma_1}(k, k') = \Gamma_{\sigma_1\sigma_2}^{(a)pq\sigma_1}(k, k') + \Gamma_{\sigma_1\sigma_2}^{(b)pq\sigma_1}(k, k') + \Gamma_{\sigma_1\sigma_2}^{(c)pq\sigma_1}(k, k').
\] (B21)

Firstly, the contribution from (a) is,

\[
\Gamma_{\sigma_1\sigma_2}^{(a)pq\sigma_1}(k, k') = (U)_{\sigma_1\sigma_2}^{pq\sigma_1}.
\] (B22)

Then the contribution from (b) and (c) for \(\uparrow\uparrow\) and \(\uparrow\downarrow\) is,

\[
\begin{align*}
\Gamma_{\sigma_1\sigma_2}^{(b)pq\sigma_1}(k, k') &= -\left[16\bar{U}\chi^2(k - k') \bar{U} + 4\bar{V}\chi^3(k - k') \bar{V} + 8\bar{U}\chi^1(k - k') \bar{U} + 8\bar{V}\chi^4(k - k') \bar{U}\right]_{qs}^{\sigma_1} \\
\Gamma_{\sigma_1\sigma_2}^{(b)pq\sigma_1}(k, k') &= -\left[16\bar{U}\chi^1(k - k') \bar{U} + 8\bar{U}\chi^2(k - k') \bar{V} + 8\bar{V}\chi^3(k - k') \bar{U} + 4\bar{V}\chi^4(k - k') \bar{V}\right]_{qs}^{\sigma_1} \\
\Gamma_{\sigma_1\sigma_2}^{(c)pq\sigma_1}(k, k') &= 4 \left[(\bar{V} - 2\bar{U})\chi^6(k + k') (\bar{V} - 2\bar{U})\right]_{qs}^{\sigma_1} \\
\Gamma_{\sigma_1\sigma_2}^{(c)pq\sigma_1}(k, k') &= [16\bar{U}\chi^1(k + k') \bar{U} + 8\bar{U}\chi^2(k + k') \bar{V} + 8\bar{V}\chi^3(k + k') \bar{U} + 4\bar{V}\chi^4(k + k') \bar{V}]_{qs}^{\sigma_1}.
\end{align*}
\] (B23)

The \(\Gamma_{\sigma_1\sigma_2}^{pq\sigma_1}(k, k')\) for other spin configurations can be obtained by the relation \(\Gamma_{\sigma_1\sigma_2}^{pq\sigma_1}(k, k') = [\Gamma_{\sigma_1\sigma_2}^{pq\sigma_1}(-k, -k')]^*\) from time-reversal symmetry.

Finally, after the effective pairing potential \(V_{\sigma_1\sigma_2}^{\alpha\beta}(k, k')\) is obtained from \(\Gamma_{\sigma_1\sigma_2}^{pq\sigma_1}(k, k')\) via the relation (B12), we should solve the linearized gap equation (B13) for each spin configuration \(\sigma_1\sigma_2\).