Electronic structure of metallic tetra-boride TmB₄: An LDA+DMFT study

Nandan Pahhira,¹,² Jyoti Krishna,³ S. Nandy,¹ T. Maitra,⁴ and A Taraphder¹,4,5

¹Department of Physics, Indian Institute of Technology, Kharagpur, West Bengal 721302, India
²Department of Physics, Kazi Nazrul University, Asansol, West Bengal 713340, India
³Department of Physics, Indian Institute of Technology, Roorkee, Uttarakhand 247667, India
⁴Center for theoretical studies, Indian Institute of Technology, Kharagpur, West Bengal 721302, India
⁵School of Basic Sciences, Indian Institute of Technology Mandi, HP 175001, India

Recent experimental observations of magnetization plateau in metallic tetraboride TmB₄ have created a lot of interest in these class of materials. Hysteretic longitudinal resistance and anomalous Hall Effect are other remarkable features in the rare-earth tetraborides which represent experimental realizations of Archimedean Shastry-Sutherland (SSL) lattice. Electronic band structures, calculated under GGA and GGA+SO approximations, show that TmB₄ is a narrow band system with considerable correlation in its f-level. Strong correlation effects in this system are studied under single-site dynamical mean field theory (DMFT) [LDA+DMFT scheme] using multi-orbital generalization of iterated perturbation theory (MO-IPT). Pseudo-gap behaviour in spectral function and non-Fermi liquid behaviour of self-energy shows non-trivial strong correlation effects present in this geometrically frustrated metallic magnets. We also consider the extent, heather-to-neglected, strong atomic spin-orbit coupling (SOC) effects. While there is a significant change in the topology of the Fermi surface in the presence of SOC, the non-Fermi liquid behavior survives. The system can be modelled by an effective two orbital spinless Falicov-Kimball model together with two free band like states.

I. INTRODUCTION

Interplay between geometric frustration and strong correlation effects can lead to exotic quantum phases like quantum spin liquid in frustrated magnets and superconductivity in organic materials. While the role of geometric frustration in insulating magnets is well studied, its role in metallic systems is not. Rare-earth tetraborides are one such class of materials with chemical formula RB₄ [R being rare-earth atom]. They are frustrated metallic magnets with long range ordered magnetic ground states. It is interesting to mention that the position of the rare-earth atoms on two dimensional sheets perpendicular to the tetragonal c-axis closely resembles the Archimedian SSL.

Recent experimental observation of fractional magnetization plateau at low temperature has drawn significant interest in the insulating materials of this series. The magnetization plateaus occurring at a fraction of saturation magnetization with stable plateau at 1/2 and fractional plateaus at 1/7, 1/8, · · · etc. closely mimic the plateaus observed in the Hall resistivity of a two dimensional degenerate electron gas. The observation of magnetization plateau in TmB₄ has been attributed to huge spin degeneracy present in nearest neighbour large spin model on an SSL. However, for a metallic system screening effects are significant and the localised spins interact via long range RKKY like interactions. Hence the mapping of interacting fermionic model onto an effective spin model on SSL with nearest neighbour interaction is highly non-trivial.

The rich phase diagram, hysteretic longitudinal magnetoresistance and anomalous Hall resistivity are other notable experimental observations in TmB₄. First-principles study of the electronic structure of rare-earth tetraborides are few in number. LDA+U calculations of a few of the tetraboride systems were performed by Yin et al. For TmB₄, a first-principles calculation showed its band characters recently.

Using first principles (LDA+U approximation) Shin et. al. argued that the electronic structure of TmB₄ admits of a local Kondo-Ising type model. It is important to mention that Tm is nominally in a trivalent state and cannot be treated within the LDA+U approximation where the only effect of U is to shift the local chemical potential. Also it is well known that interplay of strong correlation effects and Hund’s coupling in a multi-orbital system can lead to unusual effects, like orbital selective Mott transition (OSMT). In this paper, using dynamical mean field theory (DMFT), we study the strong correlation effects on metallic TmB₄, modelled by realistic bands obtained from density functional theory (DFT). This method is known as LDA+DMFT in the literature.

We consider a model Hamiltonian with four spin-degenerate orbitals, developed using density functional theory (DFT) under GGA, and we study strong electronic correlation effects in this narrow band system by applying DMFT with MO-IPT as the impurity solver. Spectral function obtained from self-consistent local Green’s function consists of a negative energy charge fluctuation peak and broad peaks around the Fermi energy. Spectral features also suggest that for low energy properties we can integrate out one of the orbitals whose integrated spectral weight is predominantly in the positive energy region. Angle resolved photo-emission spectra (ARPES) shows significant dispersion along various
high symmetry directions except along $X-M$ due to flat bands along this direction. The imaginary part of the self-energy shows asymmetric behaviour about the Fermi energy. Most importantly, the imaginary part of self-energy for one orbital shows non-Fermi liquid behaviour whose origin is possibly linked to pseudo-gap behaviour of corresponding spectral function.

In addition, due to large atomic mass of Tm, the atomic spin orbit coupling (SOC) effect is quite strong. To the best of our knowledge, the effect of strong SOC on the bands of TmB$_4$ has not been considered in any previous study. In this paper we include its effect on correlated bands. Due to strong SOC effect, the topology of the Fermi surface gets significantly modified. Interestingly even in the presence of SOC we get 3 two-fold degenerate bands. Even more interestingly, strong correlation effects and finite Hund’s coupling shows degeneracy lifting at low temperature for certain degenerate manifold. The orbitals in other manifold still remain degenerate. Finally, non-Fermi liquid characteristics of some bands remain unchanged.

The organization of the rest of the paper is as follows. In Sec. II we discuss crystal structure of the system. In Sec. III we explain computational details for band structure. Then we discuss electronic band structure in Sec. IV. In Sec. V we introduce the model involving strong correlation effects and discuss its solution under LDA+DMFT scheme. In the same section we describe results on the spectral function, self-energy, ARPES and constant energy surfaces. In Sec. VI we introduce the model involving strong correlation and strong spin-orbit coupling effects. We also discuss SOC effects on spectral properties of the model as well as its effect on the constant energy surfaces. In Sec.VII we introduce an effective low energy model for the system and briefly discuss its connection with TmB$_4$. Finally in Sec. VIII we summarize our results and conclude.

II. CRYSTAL STRUCTURE

TmB$_4$ crystallizes in the tetragonal symmetry with space group P4/mmb. It consists of alternate layers of Tm and B ions stacked along c-axis. Fig. 1 shows the crystal structure of TmB$_4$ from different perspectives. Fig.1(a) displays the full three dimensional tetragonal structure. Fig.1(b) shows only the Tm sublattice in the ab-plane which has the same topology as SSL. Fig.1(c) shows the top view of the crystal structure. There are two distinct types of B ions (1) planar and (2) octahedral present in the structure. Boron atoms form octahedra as well as 7-atom rings as shown in Fig.1(c). In Fig.1(d) one unit formed by four such B octahedra is shown. B atoms lying in the ab-plane also form dimers.

FIG. 1. (Color online) Tetragonal crystal structure of TmB$_4$. Fig.1(a) represents the full structure showing different layers of Tm (blue) and B (green) ions stacked along the c-axis. (b) The Tm ions in the ab plane can be mapped to the Sashtrry-Sutherland lattice. (c) The sub lattice of B as viewed along ab plane comprising of 7 atom ring and a square .(d) Showing two different types of B; one forming dimer(shown in red) and other forms a part in B octahedra (shown in green).
(shown in red), which are arranged in a regular pattern (see Fig. 1(c)).

III. METHODOLOGY

We performed first principles density functional theory calculations within GGA and GGA+SO approximations to study the electronic structure of TmB₄. These calculations were carried out using Perdew-Burke-Ernzerhof Generalized Gradient Approximation (PBE-GGA) exchange-correlation functional within the full potential linearized augmented plane wave (FP-LAPW) method as implemented in WIEN2k. Calculations were performed with 168 \( \mathbf{k} \) points in the irreducible wedge of the Brillouin zone (BZ). The muffin tin radii of Tm and B were taken to be 2.5 a.u. and 1.52 a.u. respectively and the plane wave expansion in spherical harmonics was considered up to angular momentum quantum number \( l = 10 \). Because of the presence of localized f-electrons, we have included the spin-orbit coupling (SOC) within the second variational method with scalar relativistic wave functions. We have performed the calculations for A-type antiferromagnetic (A-AFM) configuration. Wannier orbital fitting of the DFT bands (which are used as inputs in DMFT calculations) are obtained using the interface package WIEN2WANNIER along with the Wannier90 package.

IV. ELECTRONIC STRUCTURE

We discuss below the electronic structure calculated within GGA and GGA+SO approximation for A-type AFM configuration of TmB₄ where Tm moments are ferromagnetically aligned in the a-b plane and anti-ferromagnetically aligned c-direction. In Fig. 2, we present the spin-polarized partial density of states (DOS) of Thulium(Tm) f, d and Boron(B) p states. DOS is clearly metallic in nature with Tm f-states dominantly present at the Fermi level (FL). The itinerant carriers from B p and Tm d states also present at the FL and their overlap (hybridization) with Tm f-states make them slightly dispersive which is clearly seen in the band structure plots presented in Fig. 3. The band structure calculated within GGA and GGA+SO approximations are plotted along the high symmetry path \( \Gamma(0,0,0)-X(0.5,0,0)-M(0.5,-0.5,0)-Z(0,0,-0.5)-R(0.5,0,-0.5)-A(0.5,0.5,-0.5)-\Gamma(0,0,0) \). Within GGA (see Fig 3(a)) four bands cross the FL (shown in color) and are mostly of Tm f-character with very small hybridization with B p (mostly along \( \Gamma-X \) and \( M-Z-R \)) and Tm d states. At high symmetry points degeneracy is observed among some of these bands. In Fig. 4 we present the Fermi surfaces for each of these 4 bands.

With the inclusion of SO interaction, each of these bands crossing FL becomes doubly degenerate and form a set of 8 bands out of which 6 bands (3,4; 5,6; 7,8) now cross the FL and remaining two (1,2) are pushed below the FL. We note that at \( \Gamma \), along M-Z and at R, the previously seen degeneracy (within GGA) of band 1 and band 2 has now been lifted. New degeneracies have appeared though. The overall SO splitting has been found to be larger at \( \Gamma \), Z and R. The corresponding Fermi surfaces are shown in Fig. 5. The Fermi surfaces corresponding to degenerate bands (3,4) consists of disconnected hole pockets along \( Z-\Gamma-Z \) direction. The Fermi surface corresponding to degenerate bands (5,6) consists of hole pockets at \( R \) points and electron like pockets and Fermi surfaces in the interior region. The Fermi surface corresponding to degenerate bands (7,8) consists of hole pockets at \( R \) points. The volume of these hole pockets are much smaller than the volume enclosed by the Fermi surfaces corresponding to degenerate bands (3,4) and (5,6).

In all the previously reported electronic structure calculations, the Coulomb correlation was included within a DFT+U approach whereas here we look at the effect of correlation through DMFT. In a recently reported electronic structure calculation, the effects of Coulomb correlation and SO interaction were considered within GGA+U and GGA+U+SO approximations. Due to the application of large U, Tm f-states are pushed about 8-10 eV below Fermi level and only Tm d and B p states straddle the FL. The so called B1 and B3 Boron atoms are seen to have a clear bonding-antibonding splitting of a few eV. The bonding states are nearly filled and anti-bonding states empty. The metallic property is due to the fact that one of the three valence electrons resides in the conduction band. On the other hand Shin et al. from their GGA+U calculations find some Tm f-states (strongly hybridized with B p states) are present at the FL giving rise to the metallic property. They further observe...
FIG. 3. (Color online) The band structure for TmB$_4$ within GGA (left) and GGA+SO (right). The bands crossing the Fermi energy level ($E_F$) are shown in colors. Degeneracy at the $M$ point is maintained even in the presence of spin-orbit coupling effect. See text for more details.

FIG. 4. (Color online) Panel (a)-(d) : Fermi surface plots for bands crossing the Fermi level within GGA with band index 1,2,3 and 4 [as shown in Fig. 3(a)], respectively. The $\Gamma$ point is at the zone centre and various other symmetry points are identified in panel (a).

that 1.97 holes reside in the Tm f-states giving rise to large hole surfaces of Tm f character and smaller electron surfaces of B p character.

V. LDA+DMFT

The non-interacting Hamiltonian for TmB$_4$ can be expressed as

$$H_0 = \sum_{\mathbf{k}\alpha\sigma} (\epsilon_{\mathbf{k}\alpha\sigma} - \mu) c_{\mathbf{k}\alpha\sigma}^\dagger c_{\mathbf{k}\alpha\sigma} + \sum_{\mathbf{k}\alpha\neq\beta} V_{\alpha\beta} \left( \frac{1}{2} (c_{\mathbf{k}\alpha\sigma}^\dagger c_{\mathbf{k}\beta\sigma} + H.c.) \right) \tag{1}$$

where $\epsilon_{\mathbf{k}\alpha\sigma}$, $\alpha = 1, \cdots, 4$ are the 4 bands crossing the Fermi-energy and $\sigma = \uparrow, \downarrow$ are the spin indices of the electrons in each orbital, $\mu$ is the chemical potential of the system and $V_{\alpha\beta}$ is the residual inter-orbital hybridization which has not been included at the level of GGA. For simplicity we take $V_{\alpha\beta} = V$. It is important to mention that non-interacting dispersion $\epsilon_{\mathbf{k}\alpha\sigma}$ is obtained by diagonalizing band Hamiltonian, $H(\mathbf{k})$, at each $\mathbf{k}$-point. The band Hamiltonian $H(\mathbf{k})$ is obtained by fitting low energy bands around the Fermi-energy to 22 Wannier orbital basis. The non-interacting partial DOS for each of the 4 Fermi energy crossing bands are shown in Fig. 6. From Fig. 6 it is noticeable that total band width of each band is less than 0.2 eV which makes TmB$_4$ a narrow band system. It is well known that in narrow band systems electronic correlation effects are significantly large. We incorporate such strong correlation effects under single site dynamical mean field theory (DMFT) approximation. The Hamiltonian describing strong correlation
FIG. 5. (Color online) Panel (a) - (c) : Fermi surface plots for bands crossing the Fermi level within GGA+SO with band index 3, 5 and 7 [as shown in Fig. 3(b), respectively. The Fermi surfaces for bands with band index 4, 6, and 8 are degenerate with bands with band index 3, 5 and 7 respectively and are not shown explicitly.

We choose intra-orbital correlation strength $U = 0.1$ eV which will correspond to $U \sim W/2$, $W$ being the band width of the system. This choice of $U$ is appropriate for systems with strong correlation effects. We also choose inter-orbital correlation strength $U' = 0.05$ eV, inter-orbital hybridization strength $V = 0.02$ eV and the Hund’s coupling energy $J_H = 0.01$ eV. The chemical potential of the system $\mu$ is fixed from the total particle number $n_{tot} = \sum_{\alpha,\sigma} n_{\alpha\sigma} = n_{\text{DFT}}$. Where $n_{\text{DFT}}$ is the total particle number obtained from DFT calculation. For $\text{EuB}_5$ $n_{\text{DFT}} = 4.0$. We solve the single site impurity problem using multi-orbital generalization of iterated perturbation theory (MO-IPT) \cite{31}. MO-IPT has been bench marked against other impurity solver like continuous time quantum monte carlo (CT-QMC) and has been shown to reproduce photo-emission spectra in 3$d$ transition metal oxide compound SrVO$_3$ better than other impurity solvers \cite{31}.

FIG. 6. (Color online) Partial DOS of 4 Fermi energy crossing bands as shown in Fig. 3. The total band width of each band is less than 0.2 eV.

has been bench marked against other impurity solver like continuous time quantum monte carlo (CT-QMC) and has been shown to reproduce photo-emission spectra in 3$d$ transition metal oxide compound SrVO$_3$ better than other impurity solvers \cite{31}.

A. Spectral function

From DMFT self-consistency we obtain local Green’s function $G_{\mu\alpha}^\alpha(\omega)$ for each of the orbitals. In Fig. 7 we show the corresponding spectral function $A_{\mu\alpha}^\alpha(\omega) \equiv -\text{Im}[G_{\mu\alpha}^\alpha(\omega^*)]/\pi$ for each of the 4 degenerate orbitals. Spectral function of each of the orbitals consists of a low energy charge fluctuation peak and broad peak(s) near Fermi energy. Integrated spectral weight under low energy charge fluctuation peak is much smaller than that under broad peaks and strongly temperature dependent - at low temperature spectral weight gets transfered to broad peak(s) near Fermi energy. Total bandwidth for broad peak(s) near Fermi energy for each of the orbitals is about 0.15 eV. The spectral function for orbital 1 and 2 shows two peak structure broadened by correlation effect. Orbital 1 shows a peak at the Fermi energy while orbital 2 shows a dip at the Fermi level. With the reduction of temperature the spectral weight gets transfered to lower energy. As a result the peak (orb 1) becomes sharper while the dip (orb 2) becomes deeper. This kind of behaviour is expected in multi-band systems with strong correlation effects. The spectral function for orbital 3 and 4 shows a single peak structure with incoherent spectral features in positive energy. With the reduction of temperature spectral weight gets transfered to lower energy as has been observed for the other two orbitals.
interestingly, at the lowest temperature $T = 1$ meV an additional peak near Fermi energy appears for orbital 3 but there is a dip in the spectral function at $\omega = 0$ [See inset of Fig. 7(c)]. The pseudo-gap behaviour of spectral function for orbital 3 is possibly a precursor to orbital selective Mott transition (OSMT) state. The behaviour arises mainly due to strong correlation effect in narrow band system TmB$_4$. Also, finite $J_H$ in multi-orbital systems with different band widths can energetically favour OSMT state. For orbital 4 we see that most of the integrated spectral weight is in the region $\omega > 0$. As a result orbital 4 will not have any significant effect in low temperature dc transport and thermodynamic properties of TmB$_4$.

B. Self Energy

From the DMFT self consistency we also get self energy $\Sigma_\alpha(\omega)$ for each of the orbitals. In Fig. 8 we show the imaginary part of self consistent self energy, $\Sigma_2^\alpha(\omega) \equiv \text{Im}[\Sigma^\alpha(\omega)]$, for different orbitals labeled by orbital index $\alpha$ for various temperatures. $\Sigma_2^\alpha(\omega)$ for all the 4 orbitals shows peak in the negative energy region corresponding to the charge fluctuation peak observed in the local spectral function $A_\alpha^\text{loc}(\omega)$. Inset of each plot shows low frequency behaviour of $\Sigma_2^\alpha(\omega)$. $\Sigma_2^\alpha(\omega)$ for orbital 4 shows Fermi-liquid behaviour : $\Sigma_2^\alpha(\omega) \propto -|\omega|^2 + (\pi k_B T)^2$ at the lowest temperature $T = 1$ meV. For orbital 1 and 2 $\Sigma_2^\alpha(\omega)$ is asymmetric about $\omega = 0$ and there is possible finite intercept at the Fermi energy. Most importantly, the low energy behaviour of $\Sigma_2^\alpha(\omega)$ for orbital 3 shows behaviour contrary to what one would expect for a coherent Fermi liquid state. Inset of Fig. 8(c) clearly shows that $\Sigma_2^\alpha(\omega)$ for orbital 3 is finite at $\omega = 0$ instead of vanishing for a coherent Fermi liquid state. Also the slope of $\Sigma_2^\alpha(\omega)$ is opposite to what one would expect for a Fermi liquid state. This kind of behaviour is possibly linked to pseudo-gap behaviour of spectral function for orbital 3 as has been discussed in the previous section. The non-Fermi liquid behaviour of $\Sigma_2^\alpha(\omega)$ is possibly a precursor to OSMT in narrow band multi-orbital system such as TmB$_4$.

FIG. 7. (Color online) Local spectral function, $A_\alpha^\text{loc}(\omega)$, for spin degenerate orbital $\alpha$ for various temperatures. The parameters chosen are $U = 0.1$, $U' = 0.05$, $V = 0.02$, $J_H = 0.01$. All energies are measured in units of eV.

FIG. 8. (Color online) Imaginary part of impurity self-energy, $\Sigma_2^\alpha(\omega)$, for spin degenerate orbital $\alpha$ for various temperatures. Inset of each plot shows behaviour of $\Sigma_2^\alpha(\omega)$ in the low frequency region. The parameters chosen are same as in Fig. 7 and all the energies are measured in eV.

FIG. 9. (Color online) ARPES intensity [in arbitrary units] spectra along the high symmetry directions : (a) A - $\Gamma$ (b) $\Gamma$ - X (c) X - M (d) M - Z (e)Z - R (f) R - A at high temperature, $T = 0.05$ eV. The parameters chosen are same as in Fig. 7.
nature ARPES spectra is sharpening of spectral peaks at high temperatures. Also, as we compare with ARPES spectra in Fig. 9 disappearance of some of the spectral peaks are clearly visible. The origin of these spectral features at high temperatures are due to charge transfer between thermally excited states in different bands. The peak at $\omega \sim -0.1$ eV can be identified as the spectral peak at $\omega \sim 0.0$ shifted by the chemical potential, $\mu$. This peak is found to be less dispersive over the entire Brillouin zone except in the $\Gamma$-$X$ direction. This corresponds to spectral weight transfer from $\Gamma$ point to $X$-point. The two closely placed peaks at $\omega \sim -0.15$ eV and $\omega \sim -0.17$ eV are due to orbital 1 and orbital 2. The $\omega \sim 0.05$ eV peak due to orbital 4 gets suppressed by the Fermi factor at low temperature but appears as a $\omega \sim -0.05$ eV bump at high temperature $T = 0.05$ eV.

D. Constant energy surface

It is also important to understand the strong correlation effects on the energies of the many-body quantum states. It can be shown that under single site DMFT approximation the energies of the many-body states, $\varepsilon_{\alpha}$, in orbital $\alpha$ is related to the energies of the corresponding non-interacting states, $\varepsilon_{\alpha 0}$, through

$$\varepsilon_{\alpha} = \varepsilon_{\alpha 0} - \mu + \text{Re} \left[ \Sigma_{\alpha}(\varepsilon_{\alpha}) \right], \quad (4)$$

where $\Sigma_{\alpha}(\varepsilon_{\alpha})$ is the selfconsistent self-energy corresponding to orbital $\alpha$.

We know that in the Fermi liquid state the system can be characterized by weakly interacting quasi-particle states at the Fermi surface. In order to understand strong correlation effect on these quasi-particle states at the Fermi level we calculate correlated Fermi surface and compare against the noninteracting Fermi surface. In particular we can calculate interacting Fermi surface corresponding to orbital $\alpha$ from the solution of

$$\varepsilon_{\alpha} + \mu - \text{Re} \left[ \Sigma_{\alpha}(\varepsilon_{\alpha}) \right] = 0, \quad (5)$$

as the constant energy surface $E_{\varepsilon_{\alpha} 0} = \varepsilon_{\alpha}$. It is important to mention that in a non-Fermi liquid state we can still solve Eq. 4 and define a constant energy surface. We are interested in particular constant zero energy surface as the solution of Eq. 5. In the weakly correlated regime the zero energy surface evolves into interacting Fermi surface.

In Fig. 11 we show the constant zero energy surface for various orbitals (bands) in the first Brillouin zone. Appearance of hole pockets and shrinkage of Fermi surface volume are the most important noticeable features of the interacting Fermi surface for orbital 1 (band 1). The interacting Fermi surface for orbital 2 (band 2) gets distorted but its topology remains largely preserved. It is interesting to mention that strong correlation effects have negligible effect on orbital 4 (band 4). However due to strong correlations and inter orbital hybridization effects the topology of the zero energy surface for orbital 3 (band 3) gets significantly affected [see Fig. 11].
VI. SPIN ORBIT COUPLING EFFECT

Due to quite large atomic mass of Tm ions spin-orbit coupling (SOC) effect in 4f systems like TmB4 is quite important. It is important to mention that in previous studies of rare-earth tetra-borides SOC effects have been neglected altogether. SOC effect on GGA band structure is shown in the right panel of Fig.3. It is important to mention that in the presence of SOC effect spin and orbital degrees of freedoms gets coupled and hence electron spin is no longer a good quantum number. We now consider a 8 orbital non-interacting Hamiltonian

$$H_0^{SO} = \sum_{k\alpha} (\epsilon_{k\alpha}^{SO} - \mu^{SO}) d_{k\alpha}^\dagger d_{k\alpha} + \sum_{k,\alpha\neq\beta} V_{\alpha\beta}^{SO} \left(d_{k\alpha}^\dagger d_{k\beta} + H.c.\right)$$  \hspace{1cm} (6)

where \(d_{k\alpha}^\dagger\) creates an electron in orbital \(\alpha\) at \(k\) in the first Brillouin zone and \(\epsilon_{k\alpha}^{SO}\) is the non-interacting band dispersion in the presence of SOC. It is important to mention that \(\epsilon_{k\alpha}^{SO}\) for \(\alpha = 1, 2\) corresponds to two fold degenerate orbitals labeled (1,2) below the Fermi level as shown in Fig. 3. We consider these two bands because of their degeneracy with other bands at the \(M\) point. As a result any residual inter-orbital hybridization can cause spectral weight transfer to other orbitals. \(\epsilon_{k\alpha}^{SO}\) is obtained by diagonalizing a Hamiltonian \(H_k^{SO}\) at each \(k\)-point. \(H_k^{SO}\) is obtained by fitting low energy bands around Fermi level to 44 Wannier orbital basis. \(\mu^{SO}\) is the new chemical potential of the system in the presence of SOC. We choose \(V_{\alpha\beta}^{SO} = V_{\alpha\beta}^{SO}\) if \(\beta > \alpha + 1\) and \(V_{\alpha\beta}^{SO} = 0\) otherwise. This choice of \(V_{\alpha\beta}^{SO}\) ensures hybridization between widely separated orbitals only.

We include electron-electron correlation effect using a eight orbital Hamiltonian given by\(^{33}\)

$$H_U^{SO} = \sum_{\alpha,\alpha'} U_{\alpha\alpha'} n_{\alpha} n_{\alpha'} + \sum_{\alpha\beta} U_{\alpha\beta} n_{\alpha} n_{\beta}$$  \hspace{1cm} (7)

where \(U_{\alpha\alpha'}\) is the interaction energy between electrons in two degenerate orbitals with orbital indices \(\{\alpha = 1, 3, 5, 7; \alpha' = \alpha + 1\}\) while \(U_{\alpha\beta}\) is the interaction energy between electrons in widely separated orbitals. It is well known that interaction energy between electrons in degenerate orbitals is stronger than that between electrons in widely separated orbitals. For notational convenience we choose \(U_{\alpha\alpha'} = U\) for the rest of the paper. We choose \(U_{\alpha\beta}\) non-zero only if \(\beta > \alpha + 1\). Further \(U_{\alpha\beta} = U'\) if the pair of indices \(\alpha, \beta\) are different kind i.e. \(\alpha\) being odd and \(\beta\) even and vice versa and \(U_{\alpha\beta} = U' - J_H^{SO}\) if the pair of indices \(\alpha, \beta\) are of same kind i.e. both being odd or even. Inclusion of \(J_H^{SO}\) closely mimics the role of \(J_H\) between spin degenerate orbitals in atomic physics. Inclusion of \(J_H^{SO}\) reduces some of the correlation effects and hence promotes gain in the kinetic energy and metallicity. We study the effective Hamiltonian \(H_{eff}^{SO} = H_0^{SO} + H_U^{SO}\) under single site DMFT approximation and MO-IPT is the impurity solver. We choose \(U = 0.1\ \text{eV}, U' = 0.05\ \text{eV}, V^{SO} = 0.02\ \text{eV} and J_H^{SO} = 0.01\ \text{eV}.

A. Spectral function

FIG. 12. (Color online)Spin-orbit (SO) coupling effect on local spectral function, \(A_{loc}(\omega)\), at high temperature \(T = 0.05\). Degeneracy lifting effect is compensated by the thermal broadening effect. The parameters chosen are \(U = 0.1\ \text{eV}, U' = 0.05\ \text{eV}, V^{SO} = 0.02\ \text{eV} and J_H^{SO} = 0.01\ \text{eV}[\text{see text for details}] and all energies are measured in eV.

In Fig. 12 we show SOC effect on the spectral function at high temperature \(T = 0.05\ \text{eV}. In each panel
we show spectral function for degenerate orbital pairs. The most important feature of all the plots is that the degenerate orbital pairs remain degenerate. This is possibly due to thermal broadening effects compensating any possible degeneracy lifting effects. Spectral function for each of the degenerate pairs of orbitals consists of peaks around Fermi energy and a charge fluctuation peak around \( \omega \sim -0.2 \) eV. Orbital 1 and 2 are completely filled and will have no significant contribution to thermodynamic and dc transport properties. For orbital 3 and 4 we see two sharp peaks around Fermi energy and incoherent features extending up to \( \omega \sim -0.075 \) eV. The negative energy peak is quite sharp while the positive energy peak is less prominent and more of a sub peak also there is a dip in the density of states at the Fermi level. For orbital 5 and 6 there is a single peak around Fermi energy and the band width for this peak is smaller than all other orbitals. Spectral function for orbital 7 and 8 consists of a broad peak in the positive energy with a possible splitting due to SOC effect.

In Fig. 13 we show SOC effect on the spectral function at low temperature \( T = 0.001 \) eV. As in the case without SOC effect the negative energy charge fluctuation peak gets strongly suppressed and the spectral weight under this peak is transferred to the region around Fermi energy. For all other orbitals except orbital 5 and 6 there is no significant change in the spectral features except the peaks get sharper and there is appearance of additional kinks on the spectral function. The most significant effect of SOC effect is observed in spectral function for orbital 5 and 6. There is clear signature of degeneracy lifting between orbital 5 and 6. It is important to mention that interplay between SOC effect and Hund’s coupling leads to this degeneracy lifting effect. The degeneracy lifting between orbital 5 and 6 can give rise to magnetic instability at low temperature.

B. Selfenergy

In the last section we showed that at low temperature SOC effect can lift degeneracy between certain orbital pairs (orbital 5 and 6). Therefore it is important and interesting to understand SOC effect on the self energy of degenerate orbital pairs at low temperature. In Fig. 14 we show SOC effect on the imaginary part of self energy, \( \Sigma_2(\omega) \), of degenerate orbital pairs at \( T = 1 \) meV. Imaginary part of self energy for all degenerate orbital pair is dominated by a peak in the negative frequency region. This peak corresponds to charge fluctuation peak observed at around \( \omega \sim -0.2 \) eV. The inset of each panel shows detail behaviour of \( \Sigma_2(\omega) \) in the low frequency region. Except for orbital pair 5 and 6 no other degenerate orbital pairs show any significant degeneracy lifting effect due to SOC. \( \Sigma_2(\omega) \) for all orbital pairs are highly asymmetrical about \( \omega = 0 \) with possible finite intercept at the Fermi level. \( \Sigma_2(\omega) \) for orbital 5 and 6 shows degeneracy lifting effect as expected but the behaviour of \( \Sigma_2(\omega) \) at \( \omega = 0 \) is contrary to Fermi liquid characteristics - \( \Sigma_2(\omega) \) is finite at \( \omega = 0 \) instead of vanishing. Also the sign of the slope of \( \Sigma_2(\omega) \) is opposite to what one would expect in a conventional Fermi liquid state. More interestingly, corresponding spectral function for orbital 5 and 6 shows no pseudo-gap like behaviour as in the case without any SOC effect [see inset of Fig. 7 (c)]. These puzzling features points towards the existence of non-Fermi liquid band.
C. ARPES

FIG. 15. (Color online) Spin-orbit coupling effect on ARPES intensity [in arbitrary units] spectra at high temperature, \( T = 0.05 \) eV. The spectra is calculated along the high symmetry directions : (a) \( A - \Gamma \) (b) \( \Gamma - X \) (c) \( X - M \) (d) \( M - Z \) (e) \( Z - R \) (f) \( R - A \). The parameters chosen are same as in Fig. 12.

In Fig. 15 we show the effect of SOC on ARPES intensity plot at high temperature \( T = 0.05 \) eV. The first noticeable feature in Fig. 15 is that the spectral features in almost all the symmetry directions are highly dispersive as compared to ARPES intensity plots without SOC effect [see Fig. 9]. This is due to the fact that band dispersion becomes highly dispersive due to the presence of SOC. The spectral features arises mainly due to degenerate orbital manifolds (1,2) and (3,4). The contribution of orbital (7,8) is negligible due to Fermi function. Fermi function chop off effect is less severe for degenerate manifold (5,6) at high temperature \( T = 0.05 \) eV. The peak just above Fermi energy will appear as a peak around \( \omega \sim -0.1 \) eV. Another noticeable feature is that the convergence of three peak structure into a single peak at \( M \) point. This is due to the fact that between \( X \) and \( M \) 4 fold degenerate bands organize into two manifolds and most interestingly the gap between the two manifolds is minimum at \( M \)-point.

Finally in Fig. 16 we show the effect of SOC on ARPES at low temperature \( T = 1 \) meV. The spectral features becomes sharp due to reduced thermal broadening effects. Also some of the spectral peaks disappears. This is mainly due to Fermi function chopping effect at low temperature as well as shifting of self-consistent local chemical potential, \( \mu(T) \), to lower energy.

D. Constant energy surface

Finally, we consider strong correlation effects on constant energy surfaces of spin-orbit split bands at low temperature. In Fig. 17 we show interacting constant zero energy surface of various spin-orbit split bands at \( T = 1 \) meV. As in the case without SOC effect we first solve Eq. 5 at \( T = 1 \) meV and then find the constant energy surface \( E_{\text{SO}}(k) = \epsilon_{\text{SO}} \) in the first Brillouin zone. It is important to mention that in the Fig. 17 we do not show interacting zero energy surface for band 1 and 2 (orbital 1 and 2). As these orbitals are completely filled and does not cross the Fermi energy. The interacting zero energy surface for band 3 and 4 (orbital 3 and 4) shows dramatic change in comparison with non-interacting Fermi surfaces of the corresponding nearly degenerate bands (orbitals). The zero energy surfaces are extremely small and dispersion less which will correspond to nearly localized bands (orbitals). These strong correlation effects are also evident in the pseudo-gap like behaviour in the corresponding local spectral function as well. The topology of the interacting zero energy surfaces for band 5 and 6 (orbital 5 and 6) shows dramatic change in comparison with the corresponding non interacting Fermi surfaces. The zero energy surface for band 5 is multiply connected while the zero energy surface of band 6 consists of simply connected electron and hole pockets. The interacting Fermi surface of band 7 and 8 (orbital 7 and 8) shows no significant change in comparison with corresponding nearly degenerate non interacting Fermi surfaces as shown in Fig. 5.

VII. EFFECTIVE MODEL FOR TmB₄

In the preceding sections we have shown frequency and temperature dependence of spectral function and self-energy for TmB₄ both in the presence and absence of spin-orbit coupling. The imaginary part of self-energy
FIG. 17. (Color online) Effect of spin-orbit coupling on the interacting Fermi surfaces for various orbitals in the first Brillouin zone. The calculation is done at $T = 1$ meV and all other parameters are same as in Fig. 12 for certain orbitals clearly shows non-Fermi liquid (nFL) characteristics though the corresponding spectral functions are gapless at the Fermi energy. In Fig. 11 and Fig. 17 we show constant zero energy surface of various orbitals in the absence and presence of spin-orbit coupling. As we already discussed in the last section that the interacting zero energy surface for certain orbitals [see Fig. 17(a) and (b)] show dispersionless and nearly vanishing characteristics in the first Brillouin zone which indicates nearly localized nature of the orbitals. Also, we observed that at low temperature the two fold degeneracy between orbital pairs (5,6) is being lifted. The nFL characteristics of band 5 and 6 (orbital 5 and 6) is possibly due to unquenched scattering from the nearly localized two fold degenerate orbital pairs (3,4). Interestingly, nearly degenerate orbital pairs (7,8) do not show any significant correlation effects [see Fig. 17(e) and (f)] as evident from the nearly unchanged zero energy surface in comparison with the corresponding non interacting Fermi surfaces. These two orbitals also show Fermi liquid characteristics [see Fig. 14(e) and (f)] in their self-energies. The reduced correlation effect is possibly due to much smaller spectral weight of these bands at the Fermi level and hence much weaker scattering from the nearly localized bands (orbitals). Based on these observations, we propose an effective low energy Hamiltonian which will include doubly degenerate localized orbitals [corresponding to orbital pair (3,4)], doubly degenerate free band like states [corresponding to orbital pair (7,8)] and doubly degenerate orbitals [corresponding to orbital pairs (5,6)] which interacts with the localized orbitals. We propose an effective low energy Hamiltonian for $\text{TmB}_4$ as:

$$H_{\text{FKM}} = - \sum_{\langle ij \rangle, \alpha} \left( t_{\alpha}^d l_{i\alpha}^d l_{j\alpha} + H.c. \right) - \mu \sum_{i, \alpha} \hat{n}_{di\alpha} + \sum_{i, \alpha} \left( \epsilon_{\alpha} - \mu \right) \hat{n}_{li\alpha} + \sum_{i, \alpha} U \hat{n}_{li\alpha} \hat{n}_{di\alpha},$$

where $\alpha = 1, 2$ and $\hat{n}_{li\alpha} \equiv l_{i\alpha}^d l_{i\alpha}, \hat{n}_{di\alpha} \equiv d_{i\alpha}^d d_{i\alpha}$. The model consists of two different type of orbital states. At a given lattice site $i$, $l_{i\alpha}$ ($d_{i\alpha}$) creates (destroys) localized electrons in orbital $\alpha$ and onsite energy $\epsilon_{\alpha}$ where as $d_{i\alpha}$ ($d_{i\alpha\sigma}$) creates (destroys) extended states in orbital $\alpha$ with nearest neighbour hopping amplitude $t_{\alpha}^d$ and interacts with the localized states through onsite Coulomb correlation $U$. $\mu$ is the chemical potential of the system and $\hat{n}_{di\alpha}, \hat{n}_{li\alpha}$ are the occupation number operators for $d$ and $l$ states at a given site $i$, respectively. The model as described above is essentially a two orbital Falicov-Kimball model. It is important to mention that we do not include free band like states in our Hamiltonian as they are merely spectators and their only role is to conserve filling fraction. Also, we do not include spin in our model as in the presence of spin-orbit coupling spin is no longer a good quantum number hence the model described above is a spin less multi orbital Falicov-Kimball model. It is important to mention that a similar kind of multi-orbital Falicov-Kimball model was introduced by Taraphder et. al.\textsuperscript{34} as a possible low energy effective model for two dimensional colossal magneto-resistive system, $\text{GdI}_2$.

It is well known that the metallic state of Falicov-Kimball model is non-Fermi liquid like. The non-Fermi liquid state arises due to unquenched scattering from the
static species. Similarly, scattering of two band like states from two static species can give rise to two non-Fermi liquid bands. This feature is consistent with the spectral properties of TmB₄. However the Fermi surface for the localized species will completely disappear in contrast to observed small [and dispersionless] Fermi surface. This feature can easily be incorporated if we introduce a hopping term for the localized species with hopping amplitude $t_{\text{h}} \ll t_{\text{ed}}$. However inclusion of such a term will destroy the symmetry of the Falicov-Kimball model and can be neglected under lowest order approximation.

VIII. SUMMARY AND CONCLUSION

To summarize, we have calculated electronic structure of metallic tetra-boride system TmB₄ using LDA+DMFT scheme. We have studied strong correlation effect in this narrow band system by constructing a model Hamiltonian of 4 spin degenerated orbitals which were obtained from DFT calculation [GGA scheme]. From the DMFT self-consistency we have calculated spectral function and ARPES along various high symmetry directions. The spectral function consists of a negative energy charge transfer peak along with broad peaks around Fermi surface. The integrated spectral weight under one of the orbitals is predominantly above the Fermi energy. Hence we can neglect this orbital for thermodynamics as well as low temperature dc transport properties. The imaginary part of self-energy for one of the orbitals show strong non-Fermi liquid like characteristics which is possibly linked to pseudo-gap like behaviour of spectral function. It is interesting to mention that in multi-orbital systems interplay between strong correlation and Hund’s coupling can lead to orbital selective Mott transition(OSMT). The non-Fermi liquid like behaviour is possibly a precursor to OSMT. We have also included strong spin-orbit coupling effects present in this system. The inclusion of SOC dramatically changes the topological properties of Fermi surface of some of the orbitals. Interestingly degeneracy of orbitals are still preserved which is linked to the nature of the magnetic ground state. Inclusion of Hund’s coupling effect can lead to degeneracy lifting effects of some degenerate manifold while the other orbitals still remains degenerate. The non-Fermi liquid behaviour of certain orbitals are still preserved. Finally we have introduced two orbital Falicov-Kimball model with two free band like states as an effective low energy model for TmB₄. We have discussed in detail the connection of this model with the observed spectral properties of the more general 8 band model for TmB₄.

The interplay of strong spin orbit coupling effect and strong correlation effect in the narrow band system like TmB₄ will have experimentally observable consequences. It is quite natural to expect that in the presence of external magnetic field this system will show Lifshitz transitions. As a result magneto transport properties of this system like longitudinal magneto resistance and Hall effect can show non linear behaviour. The chemical potential tuned by the external magnetic filed can lead to partially filled electron like and hole like bands. As a result both semi classical electron like and hole like orbits can contribute towards magneto transport. A detailed study on magneto transport of this material is still under investigation.

ACKNOWLEDGEMENTS

AT acknowledges Pinaki Sengupta for introducing the material to the group. N.P. would like to acknowledge Sudipta Koley, N. S. Vidyadhira, N. Dasari Rao, Monodeep Chakraborty, Subhasee Pradhan and Swastika Chatterjee for many stimulating discussions. One of us N. P. would like to acknowledge financial and computational support from IIT, Kharagpur.

1. C. Lacroix, P. Mendels and F. Mila eds., Introduction to Frustrated Magnetism (Springer, 2011).
2. B. J. Powell and R. H. McKenzie, Rep. Prog. Phys. 74, 056501 (2011).
3. B. S. Shastry and B. Sutherland: Physica B 108 1069 (1981).
4. S. Yoshii, T. Yamamoto, M. Hagiwara, S. Michimura, A. Shigekawa, F. Iga, M. Sera, T. Takabatake, K. Ohoyama, and Y. Okabe, Physica B 396, 378 (2006).
5. S. Yoshii, T. Yamamoto, M. Hagiwara, S. Michimura, A. Shigekawa, F. Iga, T. Takabatake, and K. Kindo, Phys. Rev. Lett. 101, 087202 (2008).
6. K. Siemensmeyer, E. Wulf, H.-J. Mikeska, K. Flachbart, S. Gabani, S. Matías, P. Priputen, A. Efokimova, and N. Shitsevalova, Phys. Rev. Lett. 101, 177201 (2008).
8. S. Matías, K. Siemensmeyer, E. Wheeler, E. Wulf, 10 R. Beyer, T. Hermannsdörfer, O. Ignatchik, M. Uhlarz, K. Flachbart, S. Gabani, P. Priputen, A. Efokimova, and N. Shitsevalova, J. Phys.: Conf. Ser. 200, 032041 (2010).
9. M. Moliner, D. C. Cabra, A. Honecker, P. Pujol, and F. Stauffer, Phys. Rev. B 79, 144401 (2009).
10. M. H. Qin, G. Q. Zhang, K. F. Wang, X. S. Gao, and J.-M. Liu, J. Appl. Phys. 109, 07E103 (2011).
11. V. V. Slavin and A. A. Krivchikov, Fizika Nizkikh Temperatur 37, 1264 (2011). [Low. Temp. Phys. 37, 1006 (2011)]
12. Alexei Grechnev, Phys. Rev. B 87, 144419 (2013).
13. Z. P. Yin and W. E. Pickett, Phys. Rev. B 77, 035138 (2008).
14. S Pradhan, A Taraphder, Materials Today: Proceedings 4, Part E, 5532 (2017).
15. John Shin, Zack Schlesinger, and B. Sriram Shastry, Phys.
Rev. B 95, 205140 (2017).
16 V. Anisimov, I. Nekrasov, D. Kondakov, T. Rice, and M. Sigrist, Eur. Phys. J. B 25, 191 (2002).
17 L. d’É Medici, A. Georges, and S. Biermann, Phys. Rev. B 72, 205124 (2005).
18 P. Werner and A. J. Millis, Phys. Rev. Lett. 99, 126405 (2007).
19 A. Koga, N. Kawakami, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. 92, 216402 (2004).
20 M. Ferrero, F. Becca, M. Fabrizio, and M. Capone, Phys. Rev. B 72, 205126 (2005).
21 A. Liebsch, Phys. Rev. Lett. 95, 116402 (2005).
22 S. Biermann, L. d’É Medici, and A. Georges, Phys. Rev. Lett. 95, 206401 (2005).
23 L. d’É Medici, Phys. Rev. B 83, 205112 (2011).
24 A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Rev. Mod. Phys. 68, 13 (1996).
25 G. Kotliar, S.Y. Syrakov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. 78, 865 (2006).
26 J. P. Perdew, A. Ruzsinszky, G. I. Csonka, O. A. Vydrov, G. E. Scuseria, L. A. Constantin, X. Zhou, and K. Burke, Physical Review Letters 100, 136406 (2008).
27 P. Blaha, K. Schwarz, G. K. H. Madsen, D. Kvasnicka, and J. Luitz, WIEN2k An Augmented Plane Wave + Local Orbitals Program for Calculating Crystal Properties (Karlheinz Schwarz, Techn. Universität Wien, Austria (2001).
28 Z. Fisk, A. S. Cooper, P. H. Schmidt, and R. N. Castellano, Material Research Bulletin 7, 285 (1972).
29 A. A. Mostofi, J. R. Yates, Y.-S. Lee, I. Souza, D. Vanderbilt, and N. Marzari, Computer Physics Communications 178, 685 (2008).
30 J. Kunes, R. Arita, P. Wissgott, A. Toschi, K. Helda, Computer Physics Communications 181, 1888 (2010).
31 N. Dasari, Wasim Raja Mondal, N. S. Vidyadhiraja, P. Zhang, J. Moreno, and Mark Jarrell, Eur. Phys. J. B 89, 202 (2016).
32 Yu Fu, Zhong Huang, Xun Wang, and Ling Ye, J. Phys.: Condens. Matter 15, 1437 (2003).
33 S. Nandy, N. Mohanta, S. Acharya, and A. Taraphder, Phys. Rev. B 94, 155103 (2016).
34 A. Taraphder, M. S. Laad, L. Craco, and A. N. Yaresko, Phys. Rev. Lett. 101, 136410 (2008).