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Communication

Pressure driven phase transition in 1T-$TiSe_2$, a MOIPT+DMFT study

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A B S T R A C T

The nature of unconventional superconductivity associated with charge density wave order in transition metal dichalcogenides is currently a debated issue. Starting from a normal state electronic structure followed by a charge ordered state how superconductivity in 1T-$TiSe_2$ arises with applied pressure is still under research. A preformed excitonic liquid driven ordered state mediated superconductivity is found in broad class of TMD on the border of CDW. Using dynamical mean field theory with input from noninteracting band structure calculation, I show a phase transition appearing near about 2 GPa pressure at a temperature of 2 K as a result of exciton-phonon coupling.

1. Introduction

Despite its advanced age, superconductivity is still one of the hottest topic in the strongly correlated materials [1–4]. It has been considered the most extraordinary and mysterious property of materials for a long time. High-T_c superconductors might look like an evergreen research theme [5]. The main reason behind this is the number of opened questions concerning the pairing mechanism which is strongly related to the materials' electronic structure [5]. The basic and till date most comprehensive theory of superconductivity was introduced by Bardeen, Cooper and Schrieffer [6]. Bardeen et al., discovered that an existing attractive force between two electrons makes energetically favorable bound two electron state and they can act as boson to condensate without violation of Pauli exclusion principle.

The low dimension and other structural symmetry make the transition metal dichalcogenide (TMD) system likely to be charge density wave (CDW) ordered. Energy minimization of these type of electronic system leads to the CDW transition. In the CDW ordered phase the structural periodicity of the system is reorganized to achieve stable state, which affects conductivity of the material [7–9]. Ordering in a CDW state and superconducting state are structurally two different phenomena but their coexistence and competitive nature is found in many systems [10,11]. Condensed matter physics has a lots of findings both theoretically and experimentally on these two competing ground states [12,13]. The findings like Fermi surface instabilities, electron-phonon coupling, orbital selectivity and antiferromagnetic ground state lead to both CDW and superconducting phase [14,15,8,12]. $TiSe_2$ was one of the first known compounds with CDW ground state, and also most studied material till now due to the puzzling nature of its CDW transition. Here the CDW transition at a temperature of 200 K is to a commensurate state with a 2×2×2 wavevector [16,17]. Most of the findings in $TiSe_2$ points that the idea of Fermi surface nesting as a reason for CDW transition is not applicable here. The normal state of this material is explained as a small indirect band gap semimetal [16,18–20]. Latest experiments on $TiSe_2$ like angle resolved photoemission spectra [21] explained that the CDW phase in $TiSe_2$ consists of larger indirect band gap at different momentum direction of brillouin zone. Theoretically CDW here is predicted by electron-phonon coupling and exciton induced orbital selectivity [8], though the CDW ground state and low temperature superconductivity still remains controversial.

Recent finding of superconductivity (SC) in doped 1T-$TiSe_2$ prompted large number of intense research activities [22–26] due to the questionable SC transition following charge density wave (CDW) [27,28]. The similarity in the pressure temperature phase diagram of 1T-$TiSe_2$ with other strongly correlated materials and the semi-metallic behavior of the compound in normal and density wave led to the idea of Overhauser type CDW transition [29], i.e. Bose-Einstein condensation of correlated excitons. The superconductivity in Cu_xTiSe_2 was found after the CDW phase due to doping of Cu confined in a region around critical doping [22]. Corresponding transition temperatures form a dome-like structure with copper doping. Moreover dome formation is mostly found as the important charaterstics of phase diagrams found in heavy fermion compounds, cuprate superconductors and layered materials [30,31]. However superconductivity in these high temperature superconductors is explained as a competing order with antiferromagnetic ground state [31,32] whereas in 1T-$TiSe_2$ there is a novel type of superconductivity, where emergence of SC ordering is not related to any magnetic degrees of freedom [24,33]. Alternatively the

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SC state close to the CDW state of the parent compound has no effect on each other and the superconductivity here is predicted as a conventional SC phase driven by phonon [23]. Around the transition temperature the dome formation with doping is then explained as a consequence of the shifting of chemical potential above Fermi level caused by the doping induced electrons [23]. Though these issues have been explained previously with different theoretical views but there is lack of any consistent picture how CDW state evolve into superconducting state with pressure. Main goal of this paper is to find out a consistent theory describing CDW state to superconducting state transition. What is behind the normal to density wave ordering in 1T-$TiSe_2$ has been already explained [8] from a preformed excitonic liquid (PEL) view which causes orbital selectivity and CDW phase. This PEL view [9] is now proved to be a novel alternative to conventional theories for TMD. I will start from this PEL view to find out high pressure phase at high temperature and then I have added a symmetry breaking term to the Hamiltonian to get low temperature ordered states.

2. Method

For 1T-$TiSe_2$, normal state followed by incoherent CDW state with two particle instability is explained from LCAO + dynamical-mean field theory (DMFT) calculation of a two band extended Hubbard model [8]. The two band model I used to find normal state is

$$H = \sum_{\mathbf{k},l,m,\sigma} (t_{\mathbf{k}}^{lm} + \mu_l \delta_{lm}) c_{\mathbf{k}l\sigma}^{\dagger} c_{\mathbf{k}m\sigma} + U \sum_{i,l=a,b} n_{il\uparrow} n_{il\downarrow} + U_{ab} \sum_i n_{ia} n_{ib} + g \sum_i (A_i + A_i^{\dagger})(c_{ia}^{\dagger} c_{ib} + h.\,c) + \omega_0 \sum_i A_i^{\dagger} A_i$$

It has been observed that in the normal state of 1T-$TiSe_2$ one-particle inter-band hopping is ineffective so the ordered states must now arise directly as two-particle instabilities. The two particle interaction, obtained to second order is proportional to t_{ab}^2, which is more relevant in ordered low T region. The interaction is $H_{res} \simeq -t_{ab}^2 \chi_{ab}(0,0) \sum_{\langle i,j\rangle,\sigma\sigma'} c_{ia\sigma}^{\dagger} c_{jb\sigma}^{\dagger} c_{jb\sigma'} c_{ia\sigma'}$, with $\chi_{ab}(0,0)$ the inter-orbital susceptibility calculated from the normal state DMFT results. Now the new effective Hamiltonian is $H = H_n + H_{res}^{HF}$, where $H_n = \sum_{k,\nu} (\epsilon_{k,\nu} + \Sigma_\nu(\omega) - E_\nu) c_{k,\nu}^{\dagger} c_{k,\nu} + \sum_{a\neq b,(k)} t_{ab} (c_{k,a}^{\dagger} c_{k,b} + h.\,c.)$, with $\nu = a, b$. The residual Hamiltonian H_{res}^{HF} is found by decoupling the intersite interaction in a generalised HF sense. Now this will produce two competing instabilities, one in particle-hole channel representing a CDW and another in particle-particle (SC) channel. The HF Hamiltonian is $H_{res}^{HF} = -p \sum_{\langle i,j\rangle,a,b,\sigma,\sigma'} (\langle n_{i,a}\rangle n_{j,b} + \langle n_{j,b}\rangle n_{i,a} - \langle c_{i,a,\sigma}^{\dagger} c_{j,b,\sigma'}^{\dagger}\rangle c_{j,b,\sigma'} c_{i,a,\sigma} + h.\,c.)$ (where p is proportionality constant). The CDW phase can be found directly from the particle hole instability with the order parameter $\Delta_{CDW} \propto \langle n_a - n_b\rangle$. I studied here the superconducting phase with the two particle instability in particle-particle channel (with parametrized p=0.1) and the superconducting order parameter can be calculated from $\Delta_{sc} \propto \langle c_{i,a,\sigma}^{\dagger} c_{j,b,\sigma'}^{\dagger}\rangle$ which yields multiband spin-singlet SC.

The input for DMFT is taken from the pressure dependent LDA DOS (the non-interacting DOS at three different pressure is shown in Fig. 1.) which can be calculated from bandwidth pressure relation $D = D_0 \exp(\Delta p/K)$ (where D is bandwidth, D_0 is set 1 for reference value, K is bulk modulus and Δp is change in pressure) [40,41]. The superconducting order parameter Δ_{sc} is self-consistently computed inside DMFT from the off-diagonal element of the matrix Green's function and it is found that for superconductivity the inter-orbital pairing amplitude is negligible due to the difference in their band energies, so I will be considering only the intra-orbital pairing amplitude. The total Hamiltonian now ($H = H_n + H_{res}^{(HF)}$) will be solved within multi orbital iterated perturbation theory (MOIPT)+DMFT following earlier approaches [34,35,8,7,36].

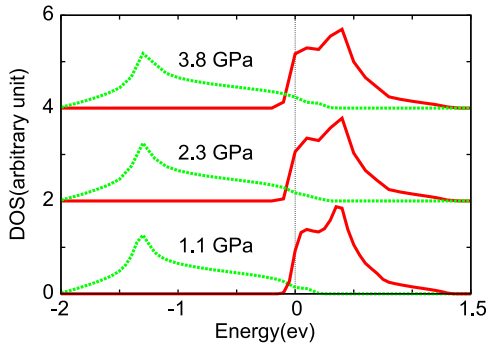

Fig. 1. (Color Online) Non-interacting Density of states at three different pressure. Red color denotes DOS of 'a' band while green color stands for 'b' band.

3. Dynamical mean field theory

To explore superconducting regime of 1T-$TiSe_2$, I used the MOIPT +DMFT with pressure dependent DOS. The anomalous Green's function: $F(k,\tau) \equiv -\langle T_\tau c_{k\uparrow}(\tau) c_{-k\downarrow}(0)\rangle$ satisfying $F(-k,-\tau) = F(k,\tau)$ for s wave pairing is introduced in the low temperature and high pressure phase of 1T-$TiSe_2$. Now in the presence of superconducting pairing the one-particle Green's function is

$$\widehat{G}(k,\tau) \equiv -\langle T_\tau \Psi(k,\tau)\Psi^{\dagger}(k,0)\rangle = \begin{pmatrix} G(k,\tau) & F(k,\tau) \\ F^{\dagger}(k,\tau) & -G(-k,-\tau) \end{pmatrix} \quad (1)$$

and all the interactions are described through self energy matrix,

$$\widehat{\Sigma}(k,i\omega_n) = \begin{pmatrix} \Sigma(k,i\omega_n) & S(k,i\omega_n) \\ S^{\star}(k,-i\omega_n) & -\Sigma^{\star}(k,i\omega_n) \end{pmatrix} \quad (2)$$

where $\omega_n = (2n+1)\pi/\beta$ are fermionic Matsubara frequencies and $S(i\omega_n)$ contains pairing information.

Within the infinite dimensional symmetry formalism the k-dependence of anomalous Green's function will be through ϵ_k and the self energy is purely k independent [37], so $\Sigma = \Sigma(i\omega_n)$. Furthermore, since the SC order parameter is considered real in this system, then $S(i\omega_n) = S^{\star}(-i\omega_n)$. Now the full lattice Green's function can be calculated using Dyson's equation as

$$\widehat{G}^{-1}(k,i\omega_n) = \begin{pmatrix} i\omega_n + \mu - \epsilon_k & 0 \\ 0 & i\omega_n - \mu + \epsilon_k \end{pmatrix} - \widehat{\Sigma}(i\omega_n) = \begin{pmatrix} i\omega_n + \mu - \epsilon_k - \Sigma(i\omega_n) & -S(i\omega_n) \\ -S(i\omega_n) & i\omega_n - \mu + \epsilon_k + \Sigma(-i\omega_n) \end{pmatrix} \quad (3)$$

The self-consistency is obtained by putting the condition that the impurity Green's function coincides with the onsite Green's function of the lattice.

Since in $TiSe_2$ the normal state and the CDW ordered state is having major dependence on Ti-d and Se-p state and preformed excitons drive the incoherent normal state to coherent CDW state I will now extend the Green's function here to take the inter-orbital hopping and Coulomb interaction into account and will put orbital indices on the Green's function and self energy.

Following the earlier formalism [34,37] to take inter-orbital hopping in Green's function I will now rewrite the full orbital dependent Green's function as

$$\widehat{G_\nu}^{-1}(k,i\omega_n) = \begin{pmatrix} G_{\nu\nu}^{-1} & -S(i\omega_n) \\ -S(i\omega_n) & -G_{\nu\nu}^{-1\star} \end{pmatrix}$$

where

$$G_{\nu\nu}^{-1} = i\omega_n + \mu - \epsilon_{k\nu} - \Sigma_\nu(i\omega_n) - \frac{(t_{\nu\nu'} - \Sigma_{\nu\nu'})^2}{(i\omega_n + \mu - \epsilon_{k\nu'} - \Sigma_{\nu'}(i\omega_n))}$$

Where $\nu\nu'$ stands for a(b),b(a) bands. In the above equation Σ_ν, $\Sigma_{\nu'}$, and $\Sigma_{\nu\nu'}$ are calculated following earlier procedure [8]. The electron

lattice interaction and inter-orbital coulomb interaction contributes in orbital dependent self energy. Now to solve the above mentioned model within DMFT I have redesigned the IPT which is already an established technique for the repulsive Hubbard model in paramagnetic phase [37,38]. Though it is an approximate method it gives good qualitative argument with the more exact methods to solve repulsive Hubbard model [39,8,34]. Also IPT has been successfully applied [36] in single band attractive Hubbard model. Reasonably this can be used in present context also. The IPT technique for superconductivity is constructed in a way that it should successfully reproduce the self energy leading order terms in both the weak coupling limit and large frequency limit and it must be exact in the atomic limit. Considering all this the IPT self energy is computed for superconductivity.

In the atomic limit, the second order self energy vanishes as discussed earlier [36]. Within this modified IPT approximation I have solved DMFT equation taking $U_{aa} = U_{bb} = U = 0.5$ eV, U_{ab}=0.1 and g=0.05 (This values were calculated after checking all possible realistic values and reproducing the normal state, CDW state and experimental results) and the order parameter is computed from the off diagonal element of the matrix Green's function in presence of an infinitesimal symmetry breaking field.

4. Result and discussion

I will show how the new DMFT approach described above explains a range of experimental data. DMFT renormalizes the LDA band position depending on its occupation by the intra and inter-orbital Hartree terms in self energy and dynamical correlations generically transfer spectral weight in larger energy scale. DMFT many body density of states (DOS) for Ti-d and Se-p band is shown at two different pressure in Fig. 2 at three different temperatures. These three temperatures correspond to predicted normal state (250 K), CDW state (100 K), [8] and superconducting state (2 K). A clear 'orbital selective' gap is found in the CDW state spectral function at both 1.1 GPa and 2.3 GPa but in the superconducting state the number density in the

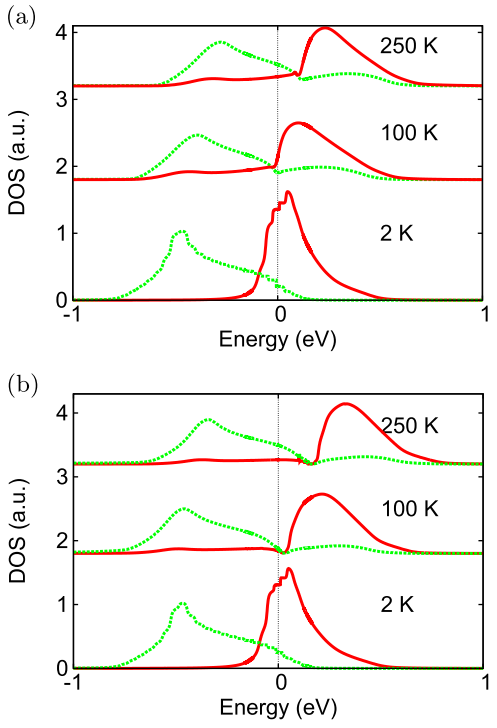

Fig. 2. (Color Online) Density of states at three different temperature and two pressure, (a) 1.1 GPa and (b)2.3 GPa. This two different pressure at lower temperature represents CDW state and superconducting state. Red color stands for DOS of 'a' band and green color stands for 'b' band. DOS for different temperatures are shifted in the y-axis.

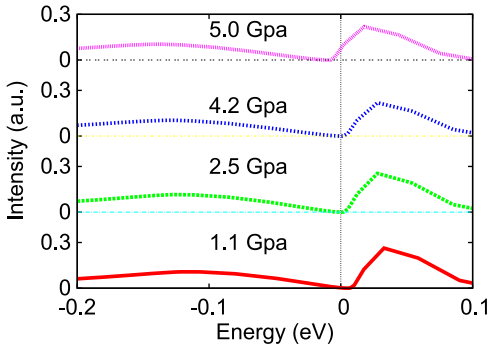

Fig. 3. (Color Online) Theoretical ARPES spectra at 'M' point in different pressure at 2 K temperature. As pressure increases from CDW state to normal state crossing a superconducting region the spectral density at Fermi level changes. A gap can be detected at the Fermi level at 2.5 and 4.2 GPa i.e. in the superconducting state.

Fermi level increases significantly. Also at 250 K temperature the orbital selectivity vanishes. Interestingly with increasing pressure orbital selective pseudogap of the CDW phase increases as well as in the SC phase the number density at the Fermi level increases. The increasing number density at the Fermi level also supports that not only increased pressure but also doping the system will help in superconductivity. Surprisingly the spectral functions are not showing any conventional energy gap in the Fermi level. In 1T-$TiSe_2$ SC phase is following an unconventional CDW state and an excitonic normal state. So superconductivity here can also be associated with unusual gap. In this scenario momentum-resolved one particle spectral function can prove if there is presence of any energy gap in the Fermi surface.

In Fig. 3 I show DMFT one-particle spectral functions, $A_{a,b}(\mathbf{k}, \omega) = -\mathrm{Im}\, G_{a,b}(\mathbf{k}, \omega)/\pi$ at M point at different pressure. Absence of infrared Landau Fermi liquid quasiparticle poles indicates existence of a incoherent excitonic features in $E_{k,a,b}$ which is associated with a pseudogap in angle resolved photoemission spectra (ARPES) lineshapes in CDW state. Here I have chosen 'M' point only because in the CDW state a 'quasi-particle peak' (originated due to electron phonon coupling) is found experimentally and theoretically [8,42] at this point. Below the superconducting transition temperature a clear 'gap' can be discernible in ARPES lineshapes at M point at both 2.3 GPa and 4.2 GPa pressure. This superconducting 'gap' closes with increasing pressure as 1T-$TiSe_2$ goes through a transition into a metallic state. Existence of such type of superconducting gap at M point was also found earlier in doped 1T-$Tise_2$ [26].

Next I have computed transport properties of the dichalcogenide in high pressure. DMFT resistivity here is calculated from dc conductivity. Remarkably the DMFT resistivity (Fig. 4) at different pressure shows a very good accord with the earlier experimental resistivity data [44]. At

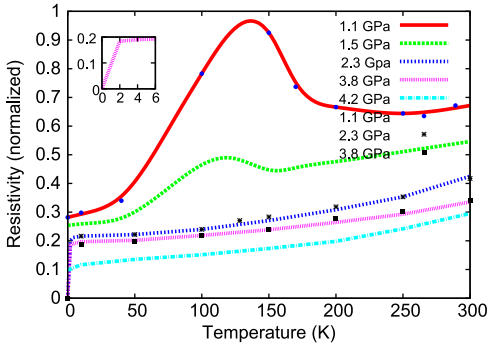

Fig. 4. (Color Online) Temperature dependent DMFT resistivity at different pressure. DMFT results agree well with previous experimental results (represented by three types of colored points for three different pressure after Kusmartseva et al. [44]). The inset shows the resistivity at low temperature at 3.8 GPa pressure in the superconducting phase. Resistivity numerical data is normalized with respect to maximum value.

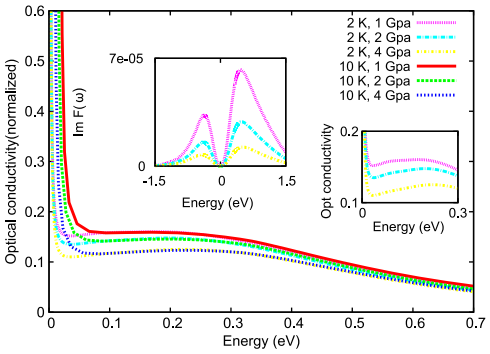

Fig. 5. (Color Online) Optical conductivity (normalized with respect to the highest value) at three different pressure and at two temperature for 1T-$TiSe_2$. The same (lower inset) at low energy presenting extended shoulder like feature appearing due to superconducting gap. Upper inset shows corresponding off diagonal Green's function at 2 K temperature.

low pressure region upto 1.1 GPa the resistivity curve resembles with the curve of ambient pressure. The CDW peak is prominent and resistivity increases from normal state to CDW state and decreases afterwards. The strong peak in the resistivity curve signaling the CDW transition becomes gradually weak with the increase in pressure, and the maxima in $-d\rho(T)/dT$ also moves to lower temperature. In the low pressure phase resistivity above transition temperature shows a semimetallic behavior and the same is suspected well below the transition temperature [43]. Further with increase in pressure in the high temperature region 1T-$TiSe_2$ undergoes metallic behavior where resistivity manifests almost linear dependence with temperature in full qualitative accord with experiment [44]. Also with increasing pressure the CDW peak in resistivity decreases and is destroyed completely above 2 GPa pressure and superconductivity is observed at about 2 K temperature. This excellently describes experimental resistivity data [44] at different pressure (Fig. 4). In the inset of (Fig. 4) the low temperature part of resistivity at 3.8 Gpa pressure is shown in an extended frame.

In Fig. 5 I show the normalized optical conductivity as a function of temperature and pressure. An extended shoulder like (right inset of Fig. 5) feature is observed at low temperature in the optical conductivity which reduces with increasing temperature to 6 K in the CDW state. This shoulder like feature is due to the superconducting gap found in DMFT ARPES spectra. Though there is still some sign of gap in optical conductivity at 1 GPa pressure but the off diagonal Green's functions shown in the inset display a comparatively larger integral than the superconducting ones which concludes that the gap is due to the CDW fluctuations. Optical conductivity is shown here till 0.7 eV without losing any important feature because I have considered only two band closer to the FL. Presence of superconducting gap in the spectral function and observation of sharpness of the low energy optical conductivity is highly contrary to each other: it is not a Drude peak because at lowest temperature also the compound shows orbital selective non Fermi liquid behavior in DMFT. Moreover it is due to the coherence which sets in due to the superconducting order. If I check the off-diagonal Green's function at three different pressure at 2 K which also gives superconducting order parameter, the decrease in both the peaks is perceptible with increasing pressure. This detectable change in order parameter confirms the presence of phase transition at particular pressure and temperature.

DMFT Fermi surface (FS) map at different temperature is given in Fig. 6. FS in 1T-$TiSe_2$ does not rebuild across transition from normal state to CDW state in 1T-$TiSe_2$. This was one of the pillar to suggest existence of a dynamically fluctuating excitonic liquid at high temperature which is being coherent at low temperature i.e. the system undergoes transition into CDW state. But still enough information can be collected from FS in transition from CDW to superconducting phase. The band pockets spread out at normal state and, more

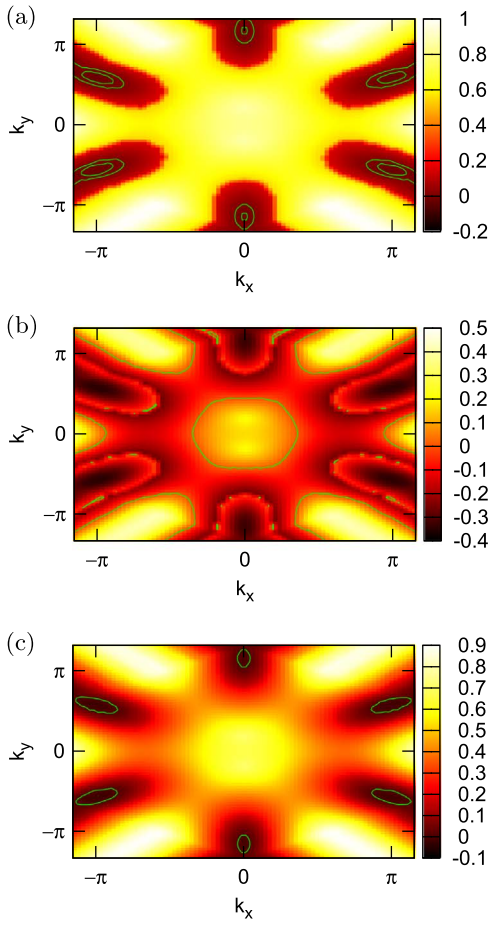

Fig. 6. (Color Online) DMFT Fermi surface map in the (a)superconducting, (b)CDW and (c)normal state.

importantly, a brighter ring structure appears at the M point in the CDW state. Now below superconducting transition temperature a much brighter elliptic structure in the middle of the hexagonal arms can be observed. Since these are found in the superconducting state so it may be a direct consequence of phase transition. Further the FS also shows isotropic s-wave superconducting gap. Moreover in the superconducting state there are electronlike pockets which develop from the sinking of band below FL. In case of doped samples of the same similar structure has been found in FS [26]. Now if this theory of superconductivity in this TMD is to be right then the superconducting order parameter which is determined self consistently within DMFT has to follow its nature of $(1 - T/T_c)^{1/2}$. In Fig. 7 main panel I have shown temperature dependent superconducting order parameter which is calculated self consistently from DMFT: $\Delta_{sc} \propto \langle c_{a\downarrow} c_{a\uparrow} \rangle = -\frac{|p|}{\pi} \int_{-\infty}^{0} \mathrm{Im}\, F(\omega) d\omega$. The order parameter is following its nature and becomes almost zero near 2 K, i.e at the transition temperature. Thus such quantitative argument built upon a novel PEL view between DMFT results and previous theoretical and experimental results support strongly the idea of a dynamically fluctuating excitonic liquid at high T giving way to a low-T superconducting ordered state.

However, to be a dependable theory the *same* formulation must also comprehensively describe transport as well. In DMFT, this task is simplified: it is an excellent approximation to compute transport coefficients directly from the DMFT propagators $G_{a,b}(k, \omega)$ [45], since (irreducible) vertex corrections rigorously vanish for one-band models, and turn out to be surprisingly small even for the multi-band case. In Fig. 4 resistivity curve at different pressure is shown which matches well with previous experiment both qualitatively and quantitatively.

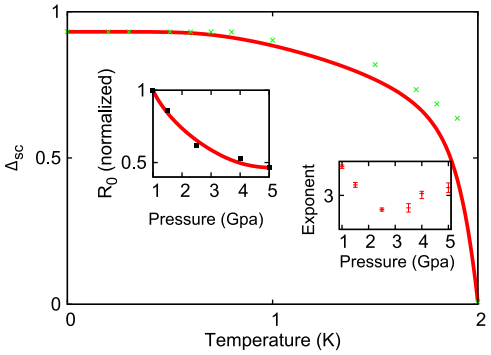

Fig. 7. (Color Online) Temperature dependency of superconducting order parameter in the main panel. The left inset shows the residual resistivity (normalized with respect to the highest value) at different pressure. The residual resistivity shows good agreement with the previous experimental result (represented by black points after Kusmartseva, et al. [44]). The right inset shows the resistivity exponent at different pressure.

Furthermore I have also fitted the resistivity curves from 3 K, above superconducting transition, to 30 K well below T_{CDW} to find out the actual procedure from CDW to superconducting transition. Until 4 GPa pressure the residual resistivity (see inset of Fig. 7) manifests a large change. Whereas only some marginal change in residual resistivity is found after superconducting phase. Detail study of the resistivity temperature exponent n, calculated from fitting the resistivity with $R(T) = R_0 + AT^n$, shows also huge change across the pressure range where superconductivity appears and at the pressure where transition temperature is maximum in the *dome* this exponent 'n' shows local minimum. Outside the superconducting dome the value of 'n' remains about 3.0, which is in stark conflict with the idea of electron-electron or electron-phonon scattering which gives value of n as 2 or 5 respectively. This fact is rather unusual but the same is found in experiment also [44]. Wilson [46] proposed this higher power law temperature dependence of the resistivity as scattering from low density band to a higher one. Though at the superconducting pressure region this exponent dovers around a value of 2.6 which represents a quantum critical scenario and quantum fluctuation around that region.

In the LDA bands of $TiSe_2$ there is negative indirect band gap. Increased pressure or doping closes the gap and some of the low energy levels cross the Fermi energy. Thus with increasing pressure CDW order is also destroyed. Increasing the number density at the FL enhances superconducting pairing correlations in a system. It has been observed that with increasing pressure the number density at the FL ($D(E_f)$) increases. Such an increase in $D(E_f)$ in turn increases superconducting transition temperature T_c in many systems as expected also from the formula $K_B T_c = \hbar\omega \exp -1/[D(E_f)V]$. Whereas increase in pressure beyond 4 GPa closes the superconducting gap found at M point and the dichalcogenide behaves like a metal.

5. Conclusion

In the light of these DMFT results, it can be concluded that with pressure the preformed excitons in the normal state drives the compound to undergo a CDW superconducting phase transition. Though conventional s-wave gap is absent in the density of states but ARPES spectra shows the presence of superconducting gap at *M* point, which is confirmed in FS plots also. An electronlike pocket is found to grow with applied pressure in the superconducting phase of this dichalcogenide. My approach highlights the role of pressure associated with excitonic correlation in small carrier density systems. Based upon a combination of high pressure LDA bandstructure and a DMFT treatment of dynamical interband excitons coupled to phonons, I have shown that this calulation describes a wide variety of features in this system. I have presented both qualitative and quantitative scenario for the emergence of unconventional superconductivity as a direct outcome of exciton phonon coupling at different pressure. Also in other parent or impurity doped TMDs superconductivity arises from normal states following a CDW order the present scenario should be relevant to all these cases.

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