Remnant Fermi surface in the presence of an underlying instability in layered 1T-TaS$_2$

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We report high resolution angle-scanned photoemission and Fermi surface (FS) mapping experiments on the layered transition-metal dichalcogenide 1T-TaS$_2$ in the quasi commensurate (QC) and the commensurate (C) charge-density-wave (CDW) phase. Instead of a nesting induced partially removed FS in the CDW phase we find a pseudogap over large portions of the FS. This remnant FS exhibits the symmetry of the one-particle normal state FS even when passing from the QC-phase to the C-phase. Possibly, this Mott localization induced transition represents the underlying instability responsible for the pseudogapped FS.

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The layered transition metal dichalcogenide 1T-TaS$_2$ is a model system being the first material where charge density waves (CDW) have been experimentally discovered by means of superlattice spots in X-ray diffraction (XRD) experiments [1]. It provides, however, one crucial difference to isostructural $\sqrt{3}$-type materials, because it shows, besides the Peierls transition at approximately 550 K, a Mott localization induced transition at 180 K, where electrons stemming from the Ta 5$d$-band manifold become more and more localized with decreasing temperature and, suddenly, yield a commensurate locked-in CDW [2]. As a consequence, 1T-TaS$_2$ exhibits a rich phase diagram, where several phases exist as a function of temperature [3]. The relevant phases here are, first, the quasi-commensurate (QC) phase, stable at room temperature (RT) and known to exhibit hexagonal arrays of commensurate domains with $(\sqrt{13} \times \sqrt{13})$ symmetry [4]. Second, the commensurate (C) phase below 180 K where the CDW is completely locked in. The electronic structure of 1T-TaS$_2$ is considerably influenced by the CDW expressed by the decay of the one-particle Ta 5$d$-band, which is split off into three dispersionless submanifolds already in the QC-phase [3,4]. Angle-resolved photoelectron spectroscopy (ARPES) investigations have given experimental evidence [4,5] for a so-called 'star-of-David' model of Fazekas and Tosatti (FT) [6]. Thirteen Ta atoms form two outerlying bonding shells with six Ta atoms each, displayed in ARPES spectra as the two low energetic manifolds. The shallow band containing the remaining thirteenth electron is susceptible to Mott localization and splits into a lower occupied (LHB) and upper unoccupied Hubbard subband. The LHB is manifest as a sharp, dispersionless peak near the Fermi level $E_F$ in near-normal emission ARPES spectra of the C phase [4,6]. Complementary tunneling spectroscopy data [10] indicates a symmetric splitting of the LHB and the unoccupied upper band with respect to $E_F$. Temperature dependent near normal emission measurements showed, that the C-phase reveals a pseudogap with residual spectral weight at $E_F$ down to very low temperatures, explaining low temperature resistivity data in terms of a variable range hopping mechanism [6]. Moreover, the local Coulomb correlation energy, the Hubbard $U_{dd}$, depends on random disorder [11].

Very recently Fermi surface (FS) measurements using ARPES have gained particular interest with respect to the mechanism behind high-temperature superconductivity [2,3,12-14]. In underdoped cuprates a remnant FS has been detected at temperatures around the transition temperature $T_c$ [13]. The important point seems to be that there is an underlying electronic instability which drives the pseudogap and the remnant FS behaviour [4,5]. One might ask whether to expect a comparable behaviour of the FS for other materials with underlying electronic phase transitions. As a possible candidate appears 1T-TaS$_2$, which has as an underlying instability the Mott localization derived transition at 180 K, where no new symmetry is dictated [6]. Above the transition temperature, in the QC phase an intact FS should be present except for new zone boundaries where a (Peierls) gap is opened [6].

In the present Letter we show, using temperature dependent scanned ARPES and FS mapping (FSM) measurements on 1T-TaS$_2$ that the FS is (pseudo)gapped although only a partial removal of the FS due to nesting is expected. Actually, it is a remnant FS yielding the symmetry of the normal (metallic) state (NS) even below 180 K with residual spectral weight at $E_F$.

Experiments have been performed in a modified VG ESCALAB Mk II spectrometer using monochromatized He-Io (21.2 eV) photons [17]. The FSM measuring mode including sequential motorized sample rotation has been outlined previously [18]. Note that this type of scanned ARPES measurement yields direct information about relevant points in k-space, in contrast to traditional ARPES work, where the datasets are intrinsically smaller and...
possibly omit information. ARPES and FSM experiments were performed with energy and angular resolution of 30 meV and ±0.5°, respectively. Pure 1T-TaS$_2$ samples were prepared by standard flux growing techniques [20] and cleaved in situ at pressures in the upper 10$^{-11}$ mbar region. Surface quality and cleanness have been checked by low energy electron diffraction (LEED) and X-ray photoelectron spectroscopy, respectively. X-ray photoelectron diffraction [21] allows us to determine the sample orientation in situ with an accuracy of better than 0.5°.

Figure 1 shows FSM data of 1T-TaS$_2$ taken with monochromatized He-Iα radiation (21.2 eV) in the QC-phase (a) and in the C-phase (b). The (1x1), i.e., NS-surfac Brillouin zones (SBZ) are given by the hexagons with the corresponding high symmetry points indicated. Additionally, in panel (b) the (expected) new SBZ due to the commensurate CDW is illustrated by the small hexagons. The raw data has been normalized [20] such that the overall polar intensity variation has been removed and symmetrized according to the space group D$_{3d}^1$ [22]. In all panels the measurements are given in a parallel-projected linear gray scale representation with maximum intensity corresponding to white. The center of the plots denotes normal emission (Γ, polar emission angle θ = 0°) and the outer circle represents grazing emission (θ = 90°). Elliptic branches, centered around M, as expected from band-structure calculations [21] are seen. A comparable shape has been found in a dichroism study of the valence band of 1T-TaS$_2$ [22]. When comparing the data at 295 K and 140 K, two features are surprising. First, at 295 K as well as at 140 K, the symmetry is threefold and not broken according to the (√13 × √13) reconstruction as indicated by the small hexagons in panel (b). This might be explained by a small Fourier component of the CDW potential [23], at least for 295 K. Nevertheless, this is surprising since 1T-TaS$_2$ is reported to have a very strong CDW amplitude [24]. Second, for 140 K, one would not expect to see any FS at all due to the reported rigid quasiparticle (QP) band shift of 180 meV in the C-phase [24]. Instead we observe small but finite spectral weight at E$_F$ all over the "FS".

In order to get more detailed information about the actual behaviour of the FS, ARPES spectra were measured along the FS contour. Spectra are given in Fig. 2 for 295 K and 140 K, respectively. The arrow on the right side indicates the k-space location of the spectra 1–41 as displayed in Fig. 3 (bottom) where the NS-FS is mimicked by ellipses. To obtain the exact FS contour locations, we measured the spectral weight at E$_F$ within the BZ wedge ΓMKΓ (see Fig. 3) with a very high point density including approximately 5400 angular positions. For 295 K one observes two broad QP peaks (Fig. 2), denoted A and B in the spectra at energy positions of about 220 meV and 700 meV, respectively. Both peaks show significant modulation along the FS contour but practically no dispersion. Most importantly, there is no clear Fermi edge visible. Instead we observe a leading edge shift of at least 30 meV as compared ot the Fermi edge of the polycrystalline Cu sampleholder. In the low temperature C-phase spectra (right panel) along the FS contour the situation is very similar expect that both peaks yield a rigid shift of about 120 meV and become narrower. Despite the energy shift the gap does not open completely and the ARPES spectra retain small but finite spectral weight at E$_F$. Note that the same k-space dispersion is observed as in (a), however with the maximal intensity reduced by approximately an order of magnitude. This means that the QC-C transition does not change the symmetry of the "FS" anymore. We interpret this in terms of a pseudogap due to finite hybridization of the overlapping tails of the two Hubbard subbands. The pseudogap remains open all over the "FS", indicating that there is no clear Fermi level crossing of a one-particle peak, even along the [ΓM] close to Γ, where the intensity is enhanced [24]. Instead one has a remnant Fermi surface (RFS) already at room temperature. Over all, this RFS has, as seen from Fig. 1(a) and (b) the symmetry retained from the elliptic one-particle NS-FS. For convenience, Fig. 3 shows a sketch of the situation in 1T-TaS$_2$. The upper part shows the E(k) of the one-particle NS Ta 5$d$ band with the Fermi level crossing in the normal phase at about 1/3 of the FM distance [21,22]. The "shady" areas indicate the practically dispersionless subbands due to the CDW potential splitting the Ta 5$d$ band [21]. The analogous E$_F$ (E$_F$) is plotted in the lower part as the NS-FS. In addition we superimposed on the NS-FS the Peierls gap region as a gray part on the ellipse. In both panels a single dispersing band can be seen sitting on an incoherent background which is built from non-dispersing QP peaks, corresponding to the decay of the NS band into three submanifolds [2]. At θ = 32° polar angle, the dispersing band reaches down to 0.95 eV bind-
ing energy in the $[\Gamma M]$ azimuth, and for $\theta = 50^\circ$ we have 0.5 eV as the maximum binding energy. The maximum binding energies fit well with the fact that the $a \rightarrow b$ scan crosses at the full depth of the band near M (see Fig. 3) whereas the $c \rightarrow d$ scan probes a more shallow part near the end of the ellipse. As a guide to the eye, the peak positions are shown, respectively, as white circles overplotted on the gray scale map and as small ticks in the spectra. Strikingly, one can see the backdispersing of the band when it reaches the region indicated by the arrows. However, there is no QP band crossing the Fermi level anywhere in the irreducible BZ wedge confirming that the original FS is completely pseudogapped and remains a RFS. These findings are remarkable insofar as one would expect a simple removal of FS portions due to nesting, either via large parallel areas (i.e., the gray shaded portion of the ellipse; bottom of Fig. 3) of the NS-FS or via a very strong electron-phonon coupling constant. Other nested FS regions than those indicated by the gray zones in Fig. 3 are not expected because XRD and LEED data clearly show the existence of a nesting vector corresponding to the superimposed ($\sqrt{3}x/\sqrt{3}$) symmetry (indicated by the small hexagons in Fig. 1(b)). Originally the lattice distortion may be driven by nesting, but the electronic structure is more and more influenced as the localization of star-centered Ta electrons sets in. This remaining thirteenth electron in the star, the LHB, becomes more and more localized and gives rise to the one order of magnitude increase of the in-plane resistivity at the QC-C transition. The QC-phase exhibits commensurate domains with an incoherent superposition of domains. Hence, the overlap of the QP peaks is considerable but decreases with temperature. At the QC-C transition there is probably a sudden increase of $U_{dd}$ which pops the two Hubbard subbands apart. Interestingly, the remaining spectral weight distribution keeps the same symmetry. From Fig. 4 we note that at RT we are left with one dispersing QP band whereas the other two bands are visible as shoulders in the QP peak, and already exhibit a dispersionless behaviour. Nonetheless we do not have a Fermi edge crossing. We may explain this when we consider the Mott transition at 180 K as an underlying transition. At RT the pseudogap might be interpreted as a kind of precursor. There is a striking similarity with underdoped cuprates where a crossover temperature scale $T^* > T_c$ has been introduced. Between $T^*$ and $T_c$ a pseudogap opens leading to the complete opening of the superconducting gap at $T_c$. One is tempted to introduce such a $T^*$ as well for 17-TaS$_2$, i.e. at the onset of the localization where the CDW becomes quasicommensurate. These findings should also motivate a reconsideration of isoelectronic systems, where electron-phonon and electron-electron interactions do interfere.

In summary, we have shown by means of temperature dependent scanned ARPES and FSM measurements that

at room temperature 17-TaS$_2$ yields a remnant Fermi surface which is not only affected by a comparatively small influence of the CDW formation, i.e., nesting, but rather it is the underlying Mott localization induced transition which seems to tune FS properties. This remnant FS is retained even below the QC-C transition with the same symmetry than in the metallic normal state phase. As in underdoped cuprates, the introduction of a crossover temperature has been suggested.

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**FIG. 1.** Fermi surface mapping results of 1T-TaS$_2$, taken with monochromatized He-I$_\alpha$ radiation (21.2 eV). Map (a) has been obtained at room temperature, i.e., in the quasi-commensurate CDW phase. Map (b) was measured at 140 K, in the commensurate CDW phase. The maps have been obtained by a simple renomalization process taking into account the space group symmetry to enhance outerlying weak features (see text).

**FIG. 2.** ARPES spectra along the Fermi surface contour, performed at room temperature (left side) and at 140K (right side). Enumeration of spectra is shown at the right by the arrow, corresponding to the drawing in Fig. 3. Raw data is shown without any normalization. The two quasiparticle peaks A and B are explained in the text.

**FIG. 3.** Simplifying sketch of the band dispersion and the Fermi surface in 1T-TaS$_2$. The upper panel mimicks the E(k) dispersion of the unperturbed Ta 5d band in the ΓM azimuthal direction. The CDW induced subbands are indicated by the shady areas. The lower panel sketches the inverse situation, the k$_\parallel$(E=E$_F$) dispersion, i.e., the Fermi surface, built up by the elliptic shape of the Ta 5d band. The arc along the FS ellipse denotes the location in k-space where the ARPES spectra of Fig. 2 have been measured. The two circular arcs show where the ARPES spectra of Fig. 4 have been measured.

**FIG. 4.** (a) Room temperature ARPES spectra measured along the circular arcs of Fig. 3, i.e., with two Fermi surface crossings, for a polar angle of $\theta = 32^\circ$. Location in k-space is indicated at the bottom, meaning spectra are from [ΓK] over [ΓM] to [ΓK]. The arrows indicate the Fermi surface crossings from the n(k) analysis (see text). In the spectra a→b on the right the dispersing quasiparticle peak is indicated by small ticks. (b) ARPES spectra as in (a), but at $\theta = 50^\circ$ polar angle, i.e., outside the Peierls gap region (gray region on the ellipse). As in (a), spectra are shown in the right panel from c to d, as indicated in the gray scale plot and in Fig.3.