

# Spectroscopic signatures of phase transitions in a charge-density-wave system: 1T-TaS<sub>2</sub>

B. Dardel, M. Grioni, D. Malterre, P. Weibel, and Y. Baer  
*Institut de Physique, Université de Neuchâtel, CH-2000 Neuchâtel, Switzerland*

F. Lévy

*Laboratoire de Physique Appliquée, Ecole Polytechnique Fédérale, CH-1015 Lausanne, Switzerland*

(Received 7 April 1992)

Photoelectron spectroscopy with high-energy resolution has been utilized to investigate the charge-density wave (CDW) in 1T-TaS<sub>2</sub> between 20 and 360 K. Constant binding-energy curves reveal discontinuities in the temperature dependence of the photoelectron spectral function  $\rho(E)$ , and demonstrate that sudden modifications of the electronic structure, namely, in the vicinity of the Fermi level  $E_F$ , mark the first-order CDW transitions. In the commensurate phase, below 180 K,  $\rho(E_F)$  reflects the formation of a correlation pseudogap. On disordered surfaces, however, the long-range coherence typical of the commensurate phase is lost, and a normal metallic behavior is recovered.

## I. INTRODUCTION

1T-TaS<sub>2</sub> has attracted much interest because of peculiar physical properties, unique among the transition-metal dichalcogenides.<sup>1-3</sup> Its layered structure consists of strongly bound S-Ta-S planar building layers coupled by weaker forces. As a result, several macroscopic properties, like the electrical resistivity,<sup>4</sup> are anisotropic and 1T-TaS<sub>2</sub> is generally considered as a quasi-two-dimensional (2D) material. The marked 2D character is exemplified by the calculated electronic structure<sup>5-7</sup> and it has been confirmed by angle-resolved photoemission experiments.<sup>8-12</sup> Suitable nesting conditions of the quasi-2D Fermi surface favor the appearance (at  $\sim 550$  K) of a periodic lattice distortion with a complex charge-density wave (CDW).<sup>1,13-18</sup> The CDW is incommensurate (I) above  $\sim 350$  K, and commensurate (C) with periodicity  $\sqrt{13} \times \sqrt{13}$  below 180 K, while between 350 and 180 K (between  $\sim 230$  and 350 K upon heating), it is *on the average* incommensurate, or quasicommensurate (QC), with commensurate domains arranged in a hexagonal lattice and separated by discommensurations.

Unlike other layered materials, the properties of 1T-TaS<sub>2</sub> present an unusual and strong temperature dependence. The electrical resistivity, for instance, exhibits sharp steps at the I-QC and QC-C transitions, and an unbounded rise below  $\sim 60$  K,<sup>4,19</sup> suggesting large changes in the electronic density of states (DOS). This observation contrasts with the usual assumption that CDW-related effects should be small in a 2D system, since a Peierls gap can only appear over limited portions of the Fermi surface. Additional mechanisms, acting with or besides the CDW, have therefore been invoked. Low-temperature resistivity data, suggestive of variable range hopping conduction,<sup>20,21</sup> have prompted a model predicting the occurrence of electron (Anderson) localization in the random field created by impurities or defects. Fazekas and Tosatti,<sup>22</sup> on the other hand, have stressed the importance of the electronic correlations. Their model maintains that the QC-C transition is accompanied by a

Mott transition, which causes a sudden reduction of the electron density of states at the Fermi level and accounts for the observed resistivity jump.

Photoemission spectroscopy (PES) and inverse photoemission spectroscopy (IPES) measurements have shown that the whole valence band of 1T-TaS<sub>2</sub> is affected by the CDW and that clear differences in the electronic density of states characterize the various CDW phases.<sup>10-12,23,24</sup> These results generally support the model of Fazekas and Tosatti. However, surprisingly, estimates of the energy gap (150–200 meV) based on spectroscopic data are two orders of magnitudes larger than values deduced from transport measurements.<sup>19</sup> Recently, we have shown<sup>25</sup> that this inconsistency actually reflects the different sensitivity of the various techniques to the two distinct energy scales of this material. PES and IPES data, in fact, directly display the characteristic energy of the Mott transition, the Coulomb correlation energy  $U \sim 200$  meV, but a much smaller energy scale, of the order of a few meV controls the low-temperature properties of the material. This small energy scale, which conventional spectroscopic measurements cannot reveal, emerges from measurements performed with state-of-the-art energy resolution. In this paper we present the results of a photoemission investigation of 1T-TaS<sub>2</sub> combining high-energy resolution and an accurate temperature control over a wide range (20–370 K), and show that with these joint capabilities it is possible to identify clear fingerprints of the structural phase transitions.

## II. EXPERIMENT

Single crystalline samples of 1T-TaS<sub>2</sub> in the form of platelets of approximately  $5 \times 5 \times 0.5$  mm<sup>3</sup> have been prepared from the elements by reversible chemical reaction with iodine as a transport agent, as described elsewhere.<sup>25</sup> The electrical resistance, measured between RT and 20 K with a standard fourpoint technique, presented sharp steps at 180 K on cooling and at 230 K on warming.

The samples, mounted on a He closed-cycle refrigerator, could be indirectly heated, and their temperature, monitored by a Rh-Fe resistance, could be controlled between 20 and 370 K, with an accuracy of  $\pm 1^\circ$ . Clean mirrorlike surfaces were prepared *in situ* by cleaving the crystals with a sharp blade, at a pressure better than  $1 \times 10^{-10}$  Torr. Under these conditions the surface remained spectroscopically clean over a period of days. Only after prolonged exposure to the residual vacuum at the lowest temperature ( $\sim 20$  K) could spurious peaks from adsorbates be observed, but these features readily disappeared after flash heating the sample to 100 K. Photoelectron spectra were collected at photon energies of 21.2 and 1486 eV, with energy resolutions of, respectively, 15 and 300 meV. The estimated angular resolution in the valence-band spectra was  $\pm 3^\circ$ .

### III. RESULTS AND DISCUSSION

Near-normal emission photoelectron spectra of the top 1.5 eV of the valence band of 1T-TaS<sub>2</sub>, measured at increasing temperatures between 20 and 356 K, are shown in Fig. 1. The bottom spectrum (356 K) is representative of the incommensurate phase: It exhibits a featureless band cut by a metallic edge whose width is compatible with the width of the Fermi function at that temperature. New structures appear in the QC phase (230 K  $< T < 350$  K) initially at binding energies of 100, 300, and 800 meV. These structures sharpen and shift towards higher binding energy with decreasing temperature, and at 230 K the metallic Fermi edge is clearly separated from the first peak. Below 230 K, in the commensurate CDW phase, a substantial growth of the shallowest peak is accompanied

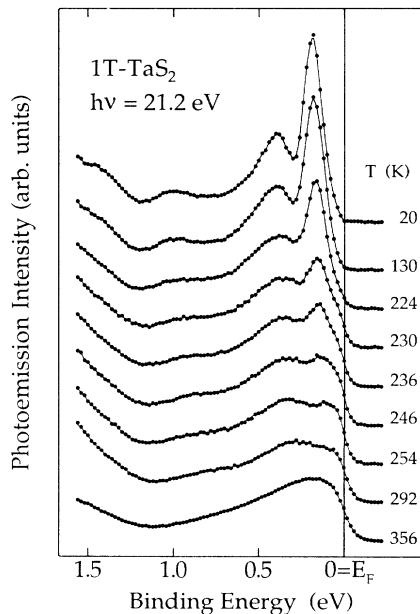


FIG. 1. Near-normal emission valence-band photoelectron (PES) spectra of 1T-TaS<sub>2</sub> collected at increasing temperatures between 20 (commensurate CDW) and 356 K (incommensurate CDW). The solid lines are guides to the eye.

by a rapid decrease of intensity at  $E_F$ . At 20 K the valence-band emission is dominated by a sharp peak at 180 meV, and the emission at the Fermi level is vanishingly small. The whole low-temperature spectrum exhibits a strong angular dependence (not shown), but the intensity at  $E_F$  remains very small at all angles. These results are consistent with published data, available for  $T \geq 120$  K.<sup>10–12,23</sup> The evolution of the spectral function reflects the splitting of the Ta  $d_{z^2}$  band into three manifolds (with occupancies of six, six, and one electron per  $\sqrt{13} \times \sqrt{13}$  unit cell), under the effect of the CDW,<sup>11,24</sup> and the occurrence of a Mott localization in the C phase. The prominent peak centered at 180 meV is interpreted as the lower Hubbard subband. No major qualitative changes are observed when the temperature is lowered from 120 to 20 K, but all spectral features become sharper. We shall discuss this point in a subsequent section.

#### A. The incommensurate-quasicommensurate transition

Although Fig. 1 may suggest a smooth temperature dependence of the PES spectral function, a more accurate analysis reveals measurable discontinuities at both the I-QC and the QC-C transitions. In Fig. 2, we compare valence-band spectra measured on both sides of the I-QC transition, at 356 and 347 K. A shift of  $\sim 100$  meV in the peak position and a 15% reduction of the intensity of  $E_F$  can be observed at the lower temperature. Given the small difference of less than 10 K, these differences cannot be attributed to normal temperature effects and must therefore be correlated with the CDW transition. We have noticed that this correlation can be established in a very direct way by recording, at a fixed binding energy,

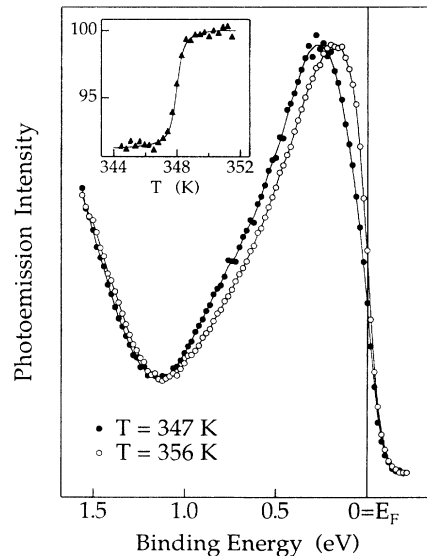


FIG. 2. PES spectra recorded above (open symbols) and below (solid symbols) the I-QC CDW transition temperature. The inset shows temperature dependence of the photoemission intensity (in arbitrary units) at a binding energy of 180 meV, corresponding to the peak in the 356-K spectrum.

the temperature dependence of the photoelectron signal. The inset of Fig. 2 reproduces such a temperature-dependent-constant-energy (TCE) curve collected, around the transition temperature, at the peak's binding energy (180 meV) of the 356-K spectrum. The clear step indicates that, in line with the first-order character of the I-QC transition, the photoelectron intensity presents a discontinuity at the critical temperature. A similar decrease of the photoemission intensity at  $E_F$  suggests a reduction of the electronic DOS which can be correlated with the stepwise increase of the electrical resistivity. From Fig. 2 we can then conclude that the I-QC CDW transition brings about a sudden rearrangement of the electronic states of 1T-TaS<sub>2</sub> over about 1 eV.

### B. The quasicommensurate-commensurate transition

The large anomaly observed in the electrical resistivity at the QC-C transition suggests important modifications of the electronic DOS, which should manifest themselves in the photoemission spectra. This is confirmed by the large changes we have observed in temperature-dependent scans. In Fig. 3(a) we present, in the temperature range 150–300 K, a TCE curve collected at a binding energy of 180 meV (corresponding to the main peak in the low-temperature spectra) during a complete cooling and warming cycle 300 → 20 → 300 K. We observe a 100% rise of intensity at 187 K upon cooling and a corresponding intensity drop at 230 K upon warming. The transition at 230 K is not as sharp as the one at 187 K, and the main step is followed by a second one around 245

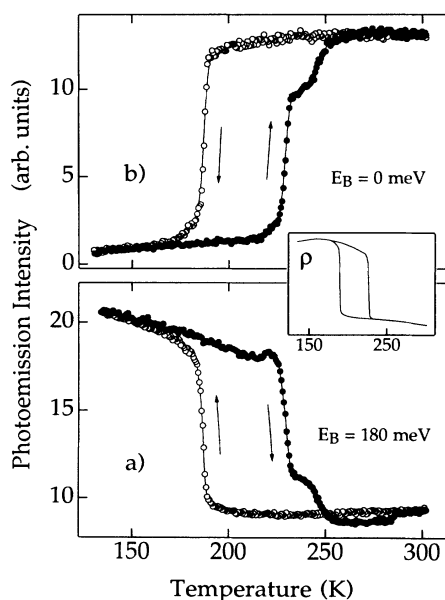


FIG. 3. Temperature-dependent constant-energy (TCE) photoemission intensity curves, collected during a complete cooling (open symbols) and warming (solid symbols) cycle, between room temperature and 20 K: (a) at a binding energy of 180 meV and (b) at the Fermi level. The inset shows the electrical resistivity of 1T-TaS<sub>2</sub> (arbitrary units) in the same temperature range.

K. Following this second step, the “warm-up” curve falls below the “cool-down” curve, until a new step at 285 K equals again the two curves.

The curve of Fig. 3(a) reproduces, with the exception of the double step in the “warm-up” curves, the characteristic temperature dependence of bulk parameters. Previous authors<sup>23</sup> had suggested, on the basis of spectroscopic results, that the backtransformation from the correlated low-temperature phase to the normal metallic state should be identified with a structural transition occurring at 285 K from the T (triclinic) phase<sup>26</sup> to the QC phase. This conjecture was rather disturbing because it implied that metallic conductivity was recovered  $\sim 50$  K below the Mott transition, when electrons are still localized. The results of Fig. 3 demonstrate unambiguously that the critical temperature coincides with that determined from the electric resistivity. The structural transition at 285 K, which mainly concerns the  $c$  axis (perpendicular to the Ta planes), is revealed in Fig. 3 by a small jump in the warm-up curve. The fact that the TCE bears a signature of this transition actually demonstrates that the sensitivity of PES is well adapted to study the close relationship between structural phase transitions and subtle changes in the electronic structure in 1T-TaS<sub>2</sub>.

On the other hand, the presence of two steps in the warm-up curves of Fig. 3, but not in the resistivity, calls for an explanation. The large, sample-dependent hysteresis, indicative of the influence of defects on the QC-C transition, and the high surface sensitivity (10–20 Å) of PES, suggest that our observations might reflect different pinning mechanisms acting in the bulk and at the surface. In order to verify whether the two steps could be assigned to distinct surface and bulk transitions, we have repeated the measurement with a different photon energy (40.8 eV) and therefore a slightly different probing depth, but the new curves exactly reproduced the results of Fig. 3. We must therefore conclude that both transitions occur at the surface. In the absence of a more systematic study, we can only speculate that cleavage defects could act as pinning centers for the CDW. A cleaved surface will inevitably present both regions with a low density of defects, insufficient to oppose the bulk-driven transition, and regions where the density of defects might be large enough to locally retard the transition to the C phase. The curves of Fig. 3 should therefore be regarded as accounting for a distribution of such surface domains, and the observation of two sharp steps, rather than a broad, continuous transition, would then suggest that these surface defects are only effective above a well-defined threshold density. A combined use of PES and of a surface-sensitive structural technique like scanning tunneling microscopy (STM) should be extremely valuable to elucidate this point.

The temperature dependence of the photoemission signal at the Fermi level, shown in Fig. 3(b), is very similar to that of the main peak [Fig. 3(a)], with sharp edges at 187 K on cooling and steps at 230 and at 245 K on warming. However, the relative intensity variations are about ten times as large, and of opposite sign; moreover, the transition at 285 K is not visible. Opposite temperature dependences of the spectral function at  $E_F$  and at 180

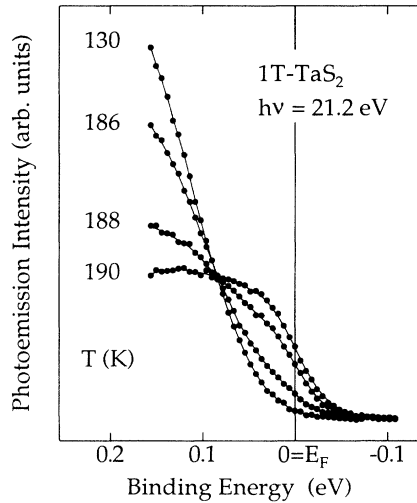


FIG. 4. PES spectra of a small energy region around the Fermi level at the QC-C CDW transition upon cooling. The transition is characterized by a sudden loss of intensity at  $E_F$  and by the growth of a peak at a binding energy of 180 meV.

meV were already suggested by the analysis of Fig. 1, which indicates a transfer of spectral weight from the Fermi level to the main peak. It is quite interesting to observe here that this transfer abruptly takes place at the QC-C transition temperature. The sharp 90% intensity drop at 187 K, and similar observations made at different emission angles, mark a collapse of the Fermi surface explaining the related tenfold increase in resistivity. These rapid modifications can be well appreciated from the raw spectra of Fig. 4. Again it must be stressed that since the instrumental broadening is smaller than the intrinsic thermal broadening, these spectra provide a faithful image of the (temperature-dependent) spectral function.

### C. The low-temperature correlated state

The sudden collapse of the Fermi surface implied by the TCE's of Fig. 3(b) is consistent with the prediction, by Fazekas and Tosatti,<sup>22</sup> of a Mott transition taking place at the QC-C critical temperature. This transition corresponds to the localization of electrons from a narrow band straddling the Fermi level into molecularlike orbitals of the star-shaped 13-atom clusters that constitute the fundamental units of the distorted structure.<sup>22</sup> The separation between the center of mass of the occupied lower and the unoccupied upper Hubbard subbands, which yields a good estimate of the strength of the on-site Coulomb correlation  $U$ , can be determined from PES (e.g., Fig. 1) and IPES (Ref. 24) spectra to be approximately 200 meV. This circumstance has led a number of spectroscopists to conclude that 1T-TaS<sub>2</sub>, in the commensurate phase, is a semiconductor with a gap of the order of 200 meV, in contradiction with the resistivity measurements. The results of Figs. 3 and 4, obtained with high-energy resolution, show, on the contrary, that the density of states at the Fermi level is small but finite well beyond the QC-C transition temperature. This point has

been discussed in a recent paper.<sup>25</sup> The photoemission intensity at  $E_F$ ,  $\rho(E_F)$ , decreases linearly with decreasing temperature, but a real gap never opens, even at temperatures much lower than the resistivity minimum ( $\sim 60$  K). This observation is crucial to understanding the physical properties of 1T-TaS<sub>2</sub>, because it indicates that the Fermi surface does not completely disappear at the transition temperature and that the QC-C transition must be regarded as a gapless Mott transition, a possibility already envisaged by Fazekas and Tosatti.<sup>22</sup> 1T-TaS<sub>2</sub> would then exhibit a weak metallic character even at low temperature if a disorder-driven localization, which is bound to occur in the deep pseudogap formed by the overlapping tails of the Hubbard subbands,<sup>27,28</sup> did not prevent metallic conduction. The low-temperature electrical conductivity is therefore controlled by the energy separation between the mobility edge and the Fermi level. A rough estimate, based on the temperature of the resistivity minimum, yields  $\sim 5$  meV for this characteristic energy.<sup>25</sup>

The modifications of the PES spectrum in the C phase are not limited to the crucial region around the Fermi level. Changes in the valence-band emission over more than 1.5 eV can be observed in Fig. 5, where we compare spectra taken at 165 and 20 K. All the spectral structures appear sharper and more intense in the 20-K spectrum (solid symbols), while the integrated intensity remains, within the experimental accuracy, constant. In particular, the progressive contraction of the main peak around its center accounts for the low-temperature behavior of the TCE's of Fig. 3. It is tempting to interpret the measured contraction as the spectroscopic consequence of an increasing CDW amplitude. The growth of the CDW below the QC-C transition, suggested by the temperature dependence of the Hall coefficient<sup>29</sup> and by x-ray-photoelectron-spectroscopy data on the Ta 4*f* core levels,<sup>30,31</sup> should in fact lead to a further localization of the conduction electron wave functions within the 13-

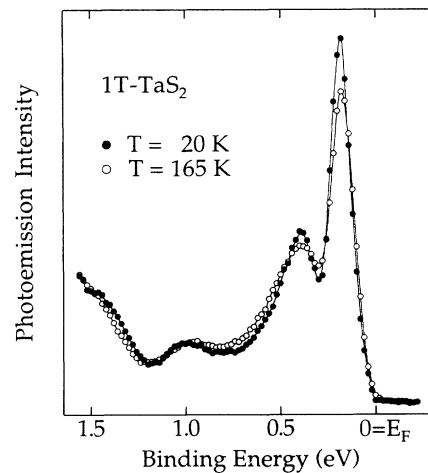


FIG. 5. Valence-band PES spectra of 1T-TaS<sub>2</sub> in the C CDW phase. The spectral features are sharper and more intense in the lower temperature (20-K) spectrum.

atom clusters. The progressive development of a molecular like situation would necessarily result in sharper spectral features. However, additional temperature-dependent sources of broadening that cannot be easily quantified, like the influence of electron-phonon scattering on the ( $k$ -dependent) spectral function,<sup>32,33</sup> and more general temperature effects in the electronic structure,<sup>34</sup> cannot be excluded.

#### D. The effect of surface disorder

As a last point, we consider the influence of surface disorder on the evolution of the charge-density wave. Disorder, as a driving force for localization, has been regarded in the past as the primary cause of the diverging low-temperature electrical resistivity in 1T-TaS<sub>2</sub>. Subsequent measurements on irradiated samples,<sup>35</sup> where the density of defects, and therefore the amount of disorder, could be accurately controlled, have, however, demonstrated that added disorder reduces the low-temperature resistivity by suppressing the transition to the commensurate phase. The suppression of the QC-C transition can be understood as the consequence of local pinning of the CDW phase by defects, which prevents the development of a coherent low-temperature ground state. From the previous discussion, we can expect the disorder-driven suppression of the commensurate phase, and the appearance of metallic conduction, to be accompanied by visible changes in the electronic DOS, and therefore in the PES spectral function.

In an attempt to test this hypothesis, we have examined surfaces prepared by cleavage in UHV immediately followed by scraping with a thin-wired tungsten brush. The amount and the nature of disorder thus induced in the sample is admittedly ill defined, but our procedure can find some justification in its simplicity and, *a posteriori*, in the striking results reported in Fig. 6. There we compare the PES spectrum of an as-cleaved surface with the spectrum of the same sample after scraping; similar curves have been obtained on all surfaces prepared with the same procedure. The disappearance of the main peak at 180 meV coincides with a redistribution of spectral weight over the whole band and with the recovery of a clear metallic edge. Structures at 400 and 900 meV are indicative of a persisting CDW distortion, and, overall, the spectrum of the “disordered” surface closely resembles spectra collected in the QC phase, although, because of the lower temperature (20 K), the Fermi edge is considerably sharper. Since the low-temperature spectra of cleaved surfaces exhibit a vanishingly small intensity at  $E_F$  at all emission angles, macroscopic effects (e.g., misoriented crystallites) can be ruled out, and we can conclude that the perturbation acts at a microscopic level in such a way that a metallic phase has been stabilized at low temperature.

The nature of this low-temperature phase is clarified by the analysis of the Ta  $4f$  core lines, whose shape is highly sensitive to the CDW. The inset of Fig. 6 shows the Ta  $4f_{7/2}$  line for the as-cleaved and the disordered surface. The splitting of the  $4f$  line into two well-resolved com-

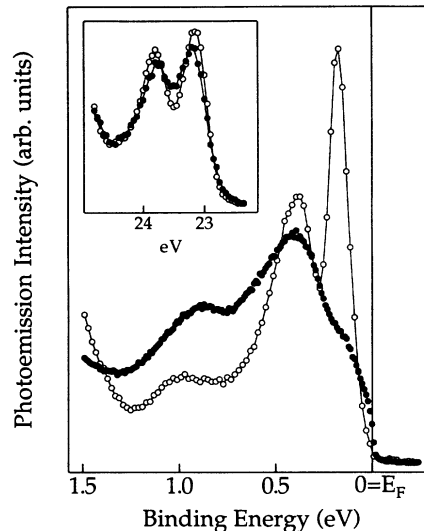


FIG. 6. Valence-band PES spectra of a cleaved 1T-TaS<sub>2</sub> surface at 20 K (open symbols) and of the same sample, mechanically disordered by brushing (solid symbols). The inset shows x-ray-photoelectron spectra of the Ta  $4f_{7/2}$  core level for the cleaved (open symbols) and the disordered (solid symbols) surface.

ponents in the spectrum of the as-cleaved sample reflects nonequivalent Ta sites, and it is consistent with previous measurements of the C phase. In the spectrum of the disordered surface, instead, the two components are broader, and their apparent separation is noticeably reduced. This is exactly the evolution expected for a transition from the long-range order of the C phase to the domain structure of the QC phase.<sup>30,31</sup> Our results therefore confirm that disorder inhibits the formation of the coherent state represented by the C phase, and yield a direct image of the disorder-stabilized metallic phase.

#### IV. CONCLUSIONS

The physical properties of 1T-TaS<sub>2</sub> are determined by the intrinsic instability of its quasi-2D Fermi surface. We have investigated the rearrangement of the electronic density of states that accompanies the transitions between different CDW phases, and we have identified precise spectroscopic signatures of each phase. We have shown that photoelectron spectroscopy, in the temperature-dependent constant-energy mode, reveals an impressive correspondence between structural and electronic properties of this material, which was not fully recognized in previous investigations. The high-energy resolution of our measurements allows us to demonstrate the formation of a correlation pseudogap in the commensurate CDW phase. This observation is not accessible to more conventional experiments where the energy resolution is typically larger than the intrinsic breadth of the metallic Fermi edge at the transition temperature. The stabiliza-

tion of a low-temperature metallic ground state by defects has also been briefly discussed. These results demonstrate that the bringing into play of high resolution and of an accurate temperature control establishes photoemission as a powerful and promising tool for the investigation of the subtle relationship between the low-energy properties of solids and the relevant electronic states.

#### ACKNOWLEDGMENTS

This work has been supported by the Fonds National Suisse de la Recherche Scientifique. We gratefully acknowledge discussions with Y. Pétroff. Many thanks are due to C. Paladión and G. Montenegro for a critical reading of the manuscript.

- <sup>1</sup>J. A. Wilson, F. J. Di Salvo, and S. Mahajan, *Adv. Phys.* **24**, 117 (1975).
- <sup>2</sup>P. M. Williams, in *Physics and Chemistry of Materials with Layered Structures*, edited by F. Lévy (Reidel, Dordrecht, 1976), p. 51.
- <sup>3</sup>R. L. Whithers and J. A. Wilson, *J. Phys. C* **19**, 4809 (1986).
- <sup>4</sup>P. D. Hambourger and F. J. Di Salvo, *Physica B* **99**, 173 (1980).
- <sup>5</sup>L. F. Mattheis, *Phys. Rev. B* **8**, 3719 (1973).
- <sup>6</sup>H. W. Myron and A. J. Freeman, *Phys. Rev. B* **11**, 2735 (1975).
- <sup>7</sup>A. M. Woolley and G. Wexler, *J. Phys. C* **10**, 2601 (1977).
- <sup>8</sup>N. V. Smith and M. M. Traum, *Phys. Rev. B* **11**, 2087 (1975).
- <sup>9</sup>R. Mamy, P. Thiry, G. Vachier, and A. Couget, *J. Phys. (Paris) Lett.* **42**, L79 (1981).
- <sup>10</sup>R. A. Pollak, D. E. Eastman, F. J. Himpsel, P. Heimann, and B. Reihl, *Phys. Rev. B* **24**, 7435 (1981).
- <sup>11</sup>N. V. Smith, S. D. Kevan, and F. J. Di Salvo, *J. Phys. C* **18**, 3175 (1985).
- <sup>12</sup>R. Manzke, O. Anderson, and M. Skibowski, *J. Phys. C* **21**, 2399 (1988).
- <sup>13</sup>C. B. Scruby, P. M. Williams, and G. S. Parry, *Philos. Mag.* **31**, 255 (1975).
- <sup>14</sup>R. Brouwer and F. Jellinek, *Physica B* **99**, 51 (1980).
- <sup>15</sup>G. Boato, P. Cantini, and R. Colella, *Phys. Rev. Lett.* **42**, 1635 (1979).
- <sup>16</sup>R. V. Coleman, B. Drake, P. K. Hansma, and G. Slough, *Phys. Rev. Lett.* **55**, 394 (1985).
- <sup>17</sup>X.-L. Wu, P. Zhou, and C. M. Lieber, *Phys. Rev. Lett.* **61**, 2604 (1988).
- <sup>18</sup>B. Burk, R. E. Thomson, A. Zettl, and J. Clarke, *Phys. Rev. Lett.* **66**, 3040 (1991).
- <sup>19</sup>A. H. Thompson, F. R. Gamble, and J. F. Revelli, *Solid State Commun.* **9**, 981 (1971).
- <sup>20</sup>F. J. Di Salvo and J. E. Graebner, *Solid State Commun.* **23**, 825 (1977).
- <sup>21</sup>N. Kobayashi and Y. Muto, *Solid State Commun.* **30**, 337 (1979).
- <sup>22</sup>P. Fazekas and E. Tosatti, *Philos. Mag. B* **39**, 229 (1979); *Physica B* **99**, 183 (1980).
- <sup>23</sup>R. Manzke, T. Buslaps, B. Pfalsgraf, M. Skibowski, and O. Anderson, *Europhys. Lett.* **8**, 195 (1989).
- <sup>24</sup>R. Claessen, B. Burandt, H. Carstensen, and M. Skibowsky, *Phys. Rev. B* **41**, 8270 (1990).
- <sup>25</sup>B. Dardel, M. Grioni, D. Malterre, P. Weibel, Y. Baer, and F. Lévy, *Phys. Rev. B* **45**, 1462 (1992).
- <sup>26</sup>S. C. Bayliss, A. M. Gorayeb, and D. R. P. Guy, *J. Phys. C* **17**, L533 (1984).
- <sup>27</sup>N. F. Mott, *Metal-Insulator Transitions* (Taylor & Francis, London, 1974), pp. 44ff.
- <sup>28</sup>D. J. Thouless, *J. Phys. (Paris) Colloq.* **37**, C4-349 (1976).
- <sup>29</sup>R. Inada, Y. Onuki, and S. Tanuma, *Phys. Lett.* **69A**, 453 (1979).
- <sup>30</sup>H. P. Hughes and R. A. Pollak, *Philos. Mag.* **34**, 1025 (1976).
- <sup>31</sup>G. K. Wertheim, F. J. Di Salvo, and S. Chiang, *Phys. Rev. B* **13**, 5476 (1976).
- <sup>32</sup>R. S. Williams, P. S. Wehner, J. Stöhr, and D. A. Shirley, *Phys. Rev. Lett.* **39**, 302 (1977).
- <sup>33</sup>R. C. White, C. S. Fadley, M. Sagurton, P. Roubin, D. Chandris, J. Lecante, C. Guillot, and Z. Hussain, *Phys. Rev. B* **35**, 1147 (1987).
- <sup>34</sup>P. B. Allen and V. Heine, *J. Phys. C* **9**, 2305 (1976); P. B. Allen and M. Cardona, *Phys. Rev. B* **27**, 4760 (1983).
- <sup>35</sup>H. Mutka, L. Zuppiroli, P. Molinié, and J. C. Bourgouin, *Phys. Rev. B* **23**, 5030 (1981).