

Fermi-Liquid Line Shapes Measured by Angle-Resolved Photoemission Spectroscopy on 1-*T*-TiTe₂

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We have performed high-resolution angle-resolved photoemission spectroscopy (ARPES) on the layered compound 1-*T*-TiTe₂, whose low-energy properties are those of a normal metal, and analyzed the experimental line shapes in terms of the Fermi-liquid self-energy. We find excellent agreement between the measured and theoretical spectral weight distribution, while line profiles expected for other theoretical models such as the marginal Fermi liquid clearly fail to reproduce the experimental spectra. This demonstrates that ARPES line shapes are able to reflect the nature of an interacting electron system.

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Angle-resolved photoemission spectroscopy (ARPES) has provided deep insight into the electronic structure of high-temperature superconductors (HTS) by providing the first evidence of dispersing single-particle excitations and a Fermi surface in accordance with the Luttinger counting theorem. Furthermore, the unusual behavior of the ARPES linewidths and line shapes has been interpreted as an indication for a non-Fermi-liquid-like ground state [1] such as the marginal Fermi liquid or the Luttinger liquid. However, a detailed analysis of HTS line shapes [2] showed that conclusions of non-Fermi-liquid behavior hinge on arguments for a negligible inelastic background, and pointed out that the spectra have an unexplained steplike emission at the Fermi energy E_F . Also, *a priori* it is not at all clear to what extent ARPES line shapes are really dominated by the single-particle excitation spectrum and how much they may be affected by transition matrix elements or extrinsic effects like photoelectron scattering or diffraction. To date, the work by Kevan [3] on the anomalous broadening of a surface state in copper is the only systematic study on ARPES line shapes in the literature. Thus there is a clear need to calibrate angle-resolved photoemission spectroscopy on a system with a known many-body ground state. In this Letter we present high-resolution ARPES data on the layered compound 1-*T*-TiTe₂ and an analysis of the spectra in terms of Fermi-liquid line profiles, thereby demonstrating for the first time that ARPES line shapes can indeed reflect the character of an interacting many-body system.

It is important to assess the relation between the ARPES spectrum and the quantity of interest, the single-particle excitation spectrum $A(\mathbf{k}, \omega)$. Within the usual three-step model of photoemission and using the sudden approximation, the ARPES signal is proportional to

$|M_{if}|^2 A(\mathbf{k}, \omega)$, where M_{if} is the transition matrix element. Thus, apart from the modulations by the matrix elements, ARPES measures essentially the spectral function. However, when going beyond this simple photoemission theory there are additional complications. For example, in the more comprehensive one-step theory it is shown that the lifetime of the photoelectron adds to the total ARPES linewidth. Since the final-state energy width is mixed in with a weight factor of $v_{h\perp}/v_{e\perp}$, where $v_{h\perp}$ and $v_{e\perp}$ are the band velocities, perpendicular to the surface, of the photohole and the photoelectron, respectively, the effect of the final electron state broadening can be suppressed if $v_{h\perp} \ll v_{e\perp}$ [4]. This is why detailed photohole line-shape studies can only be done on surface states or layered systems like the HTS. Other mechanisms that may be capable of distorting the spectral weight information include scattering and diffraction of the outgoing photoelectron. Only if all these effects are negligible, or if their energy dependence is small on the scale of the intrinsic linewidths, can the ARPES signal be taken to be representative of the electron removal spectrum.

The spectral function $A(\mathbf{k}, \omega)$ itself is simply proportional to the imaginary part of the Green's function $G(\mathbf{k}, \omega) = [\omega - \epsilon_{\mathbf{k}} - \Sigma(\mathbf{k}, \omega)]^{-1}$ of the interacting electron system, where $\epsilon_{\mathbf{k}}$ is the free-particle energy and Σ the self-energy containing all many-body aspects of the system. In the case where the low-lying excitations can be well described by quasiparticles, i.e., in a Fermi liquid (FL), the self-energy near the Fermi energy is well known [5]:

$$\Sigma(\mathbf{k}, \omega) = \alpha\omega + i\beta\omega^2, \quad (1)$$

where we have assumed a three-dimensional isotropic electron system. Note that this is an approximation that

holds only for energies small compared to the occupied bandwidth ϵ_F . Using this self-energy the FL spectral function is given by

$$A(\mathbf{k}, \omega) \propto \beta' \omega^2 / [(\omega - \epsilon'_k)^2 + \beta'^2 \omega^4], \quad (2)$$

where $\epsilon'_k = Z_F \epsilon_k$, $\beta' = Z_F \beta$, and Z_F is the quasiparticle weight factor or wave-function renormalization constant on the Fermi surface, given by $(1 - \alpha)^{-1}$. In this approximation, the spectral function depends on momentum \mathbf{k} only through ϵ'_k . The effect of the FL self-energy (1), i.e., of the interaction in the system on the spectral function is threefold: (i) a renormalization of the quasiparticle energies, (ii) a finite lifetime width and an asymmetric tail on the high-binding-energy side in the line shapes (Breit-Wigner profile), and (iii) a transfer of spectral weight across the Fermi energy. While (i) is not directly accessible by measurements, because the noninteracting system cannot be realized, we will present evidence for the latter two effects from our ARPES data.

1-*T*-TiTe₂ is a layered material that, according to band-structure calculations [6,7], is a semimetal with overlapping bands of Te 5*p* and Ti 3*d* parentage. This picture has been confirmed by ARPES and inverse photoemission [7-9]. Here we will concentrate on the Ti 3*d*-derived band, which constitutes electron pockets around the *L* points of the Brillouin zone. It has a small but finite dispersion perpendicular to the layers, with a Fermi-level crossing also along the *z* direction of \mathbf{k} space. This allows us to use the self-energy (1) of the *three-dimensional* FL in the analysis of the line shapes. On the other hand, the dispersion is small enough for the condition $v_{h\perp} \ll v_{e\perp}$ to hold, enabling us to measure the intrinsic photohole line profiles. The FL character of 1-*T*-TiTe₂ can be seen from the temperature dependence of the resistivity, which is linear for $T > 60$ K and behaves as T^5 for smaller temperatures [10], as expected for a FL interacting with a phonon gas. The low-temperature T^2 behavior characteristic of the FL is obscured by a logarithmic dependence below 5 K, which has been attributed to localization [10].

The experiments were carried out at the University of Wisconsin Synchrotron Radiation Center using the high-resolution photoemission spectrometer on the Ames-Montana State beam line. The full acceptance angle of the analyzer was 2°, which at the photon energy of 21.2 eV used here translates into a \mathbf{k} resolution of $\Delta k = 0.07 \text{ \AA}^{-1}$. The overall energy resolution was determined to be 35 meV from the Fermi edge of a platinum foil measured at 20 K. The TiTe₂ single crystals were grown by the usual iodine vapor transport method and were characterized and oriented by Laue diffraction. Final orientation of the crystals was achieved by using the symmetry of the ARPES spectra about a high-symmetry line, which, due to the high resolution of the experiment and the good quality of the samples, was possible to within $\pm \frac{1}{4}^\circ$. The crystals were cleaved *in situ* and measured at a base pres-

sure of 3×10^{-11} torr. All measurements were taken at low temperatures (~ 20 K) in order to suppress thermal broadening of the spectra.

Figure 1 shows the raw ARPES spectra measured along the Γ -*M* line of the Brillouin zone normalized to the incoming photon flux. For small emission angles one can observe a Te 5*p*-derived band that disperses upwards towards the zone center and eventually crosses the Fermi level near $\vartheta = 0^\circ$. In the vicinity of $\vartheta = 14.75^\circ$ the Ti 3*d* band appears to cross E_F , moving to higher binding energy and becoming broader as the angle is increased. For $\vartheta \geq 19^\circ$ the maximum of the ARPES peak does not disperse much more but becomes rather broad and strongly asymmetric. Note that the inelastic background in the spectra is almost negligible. We also observe that, although the main weight of the 3*d* band has moved to above the Fermi level for $\vartheta \leq 14.5^\circ$, there still remains a small bump near E_F .

We have modeled the ARPES line shapes by using expression (2) for the FL self-energy, and taking into account the finite momentum resolution by integrating $A(\mathbf{k}, \omega)$ over the volume of \mathbf{k} space corresponding to the

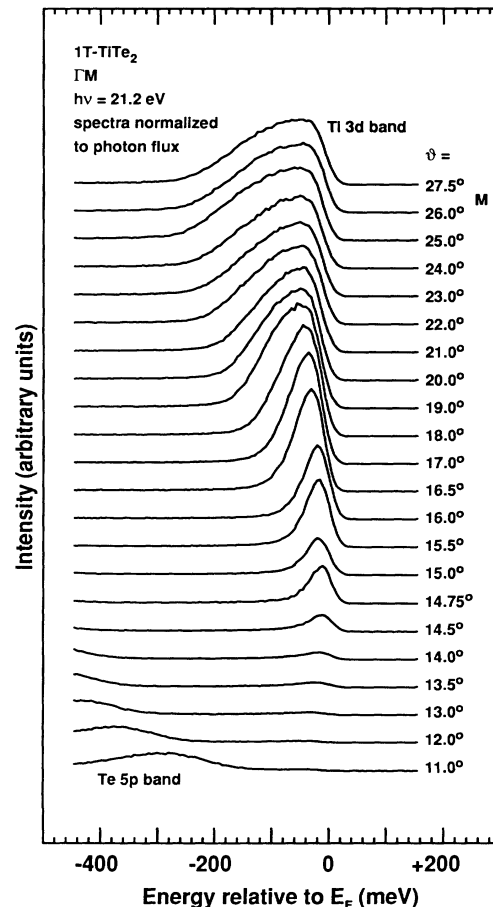


FIG. 1. Angle-resolved photoemission spectra of 1-*T*-TiTe₂ taken along the Γ -*M* direction.

analyzer acceptance cone. The energy resolution is taken into consideration by convolution with a Gaussian of 35 meV width. To fit the theoretical line profiles to the data we used the parameters β' and ϵ'_k , where β' is kept fixed for all spectra and ϵ'_k is varied for each spectrum to obtain the correct peak position. Finally, the FL line shape for each \mathbf{k} was scaled in intensity to the ARPES data by a factor which due to matrix element effects varies smoothly with \mathbf{k} . The results of these fits are shown in Fig. 2. All FL line shapes were calculated with a quasiparticle decay constant $\beta' = 40 \text{ eV}^{-1}$. Because (1) is an approximation for $\epsilon'_k \ll \epsilon_F$ (i.e., binding energy small as compared to the occupied bandwidth) the best agreement is to be expected for the spectrum at $\vartheta = 14.75^\circ$ corresponding to the Fermi-level crossing, and indeed we find an excellent correspondence. As ϑ is increased, i.e., as the $3d$ band is followed to higher binding energies, we obtain very good fits up to 16° . Note again that we did not have to include any inelastic background to obtain this agreement. For higher angles ($\vartheta \geq 16.5^\circ$) the high-

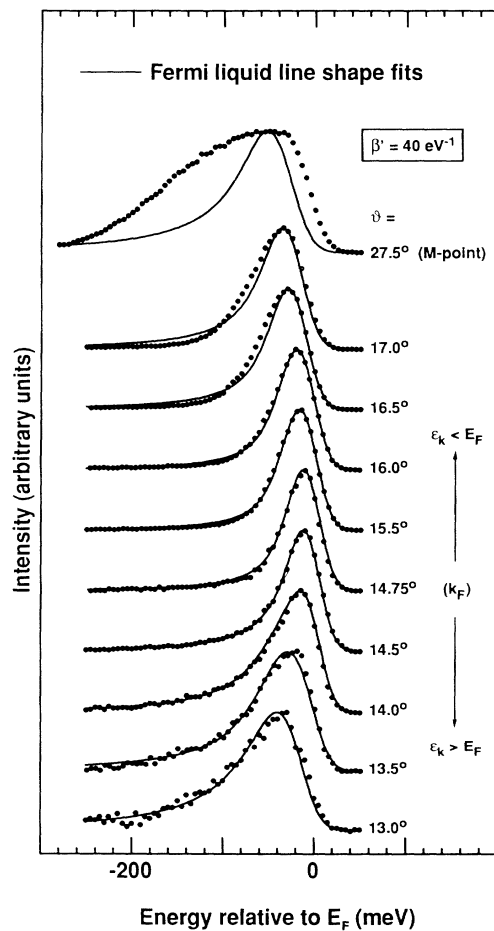


FIG. 2. ARPES spectra of Fig. 1 (dots) and Fermi-liquid line-shape fits (solid lines) for $\beta' = 40 \text{ eV}^{-1}$. Note that all spectra are normalized to the same size. Except for the lower four spectra there is no inelastic background subtracted.

binding-energy tail of the model clearly starts to deviate from the data, until at the zone boundary (27.5°) the spectral function (2) completely fails to describe the data correctly. This is due to the eventual breakdown of the self-energy approximation (1), as the binding energy ϵ_k cannot be considered small anymore in comparison to ϵ_F , which from band-structure calculations [6,7] is estimated to be $\sim 0.3 \text{ eV}$.

The lower four spectra ($\vartheta \leq 14.75^\circ$) in Fig. 2 show the small spectral weight remaining below E_F for angles where the band has already dispersed above the Fermi level. Note that in the figure these spectra are scaled up considerably with respect to the spectra with ϵ_k below E_F (e.g., for $\vartheta = 13^\circ$ by a factor of 16) and that for these very small intensities we had to account for the weak inelastic background. We find that the small structure near E_F and especially its apparent reversed dispersion can indeed be modeled quite accurately by a FL line shape with the quasiparticle energy lying *above* the Fermi energy. Thus, this feature can be identified as the part of the FL spectral weight that is transferred across the Fermi level.

In Fig. 3 we show the spectrum at the Fermi-level crossing on an enlarged scale in order to demonstrate the

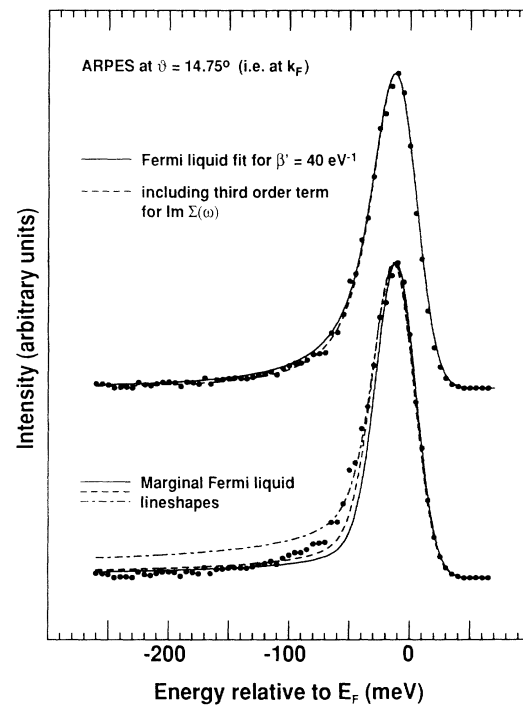


FIG. 3. Upper part: ARPES spectra at the Fermi-level crossing (dots) and FL line profiles calculated using approximation (1) (solid line) and the higher-order approximation of Ref. [11] (dashed line) for the self-energy. Lower part: Same experimental spectra, but fitted with line shapes of the marginal Fermi liquid for various model parameters (for details see Refs. [2,12]).

very high quality of the FL fit. Also shown is a theoretical line shape using a higher-order approximation for the FL self-energy in a Coulomb gas [$\text{Im}\Sigma(\mathbf{k},\omega) = \beta\omega^2(1 - \omega/\epsilon_F)$] [11], which has the advantage that it does not require a further fit parameter, but only the occupied bandwidth ϵ_F , which can be taken from band theory. For the spectrum in Fig. 3 this approximation leads to an excellent agreement of the theoretical and measured line shape. Also we found that it clearly diminishes the deviation in the high binding-energy tail observed at higher emission angles.

We should emphasize here that the excellent quality of the fits is really unique to the use of FL line shapes. Attempts to fit with other line profiles, e.g., that of the marginal Fermi liquid [12], failed to reproduce the experimental spectra, as is also shown in Fig. 3. In particular, we find distinct disagreement on the high-binding-energy side of the ARPES peak. This will be discussed in more detail in a forthcoming paper which will contain a comprehensive description of this line-shape analysis.

The FL fits yield for the renormalized quasiparticle decay constant β' a value of $40 \pm 5 \text{ eV}^{-1}$. For the Coulomb gas the bare β can be expressed in terms of the occupied bandwidth ϵ_F and the plasmon energy ω_p as $\beta = 0.067 \times \omega_p/\epsilon_F^2$ [13]. Using ϵ_F from above and the experimental value [14] $\omega_p = 18.2 \text{ eV}$, and extrapolating existing calculations [5] of the wave-function renormalization constant Z_F as a function of the electron gas parameter r_s [15] to $r_s \sim 10$, appropriate to the low $3d$ -electron density in TiTe_2 , we obtain a rough estimate $\beta' \leq 5 \text{ eV}^{-1}$. This theoretical estimate is based only on the $3d$ electrons, and since the existence of the Te $5p$ -derived sheets of the Fermi surface provides additional phase space for the decay of the quasiparticles, we expect a larger theoretical value for β' . Thus the experimentally determined β' does not seem to be unreasonable. For comparison, a similar estimate for the theoretical β' of the high-temperature superconductor $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ is smaller by 2 orders of magnitude than the value obtained by FL line-shape fits [2].

In conclusion, we have analyzed angle-resolved photoemission spectra of $1-T\text{-TiTe}_2$ in terms of Fermi-liquid line shapes and found excellent agreement between the

theoretical and experimental spectral weight distributions. This demonstrates for the first time that ARPES line profiles can reflect the nature of a many-body ground state.

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