Optical properties of the charge-density-wave rare-earth tri-telluride compounds: A view on PrTe$_3$


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We report on our recent optical measurements of the Pr tri-telluride charge-density-wave system. Our data, collected over an extremely broad spectral range, allow us to observe both the Drude component and the single-particle peak, ascribed to the contributions due to the free charge carriers and to the charge-density-wave gap excitation, respectively. Our findings perfectly fit within the scenario based on a diminishing impact of the charge-density-wave condensate on the electronic properties upon compressing the lattice, as evinced from our previous investigations on these systems both with chemical as well as with applied pressure.

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1. Introduction

The controlled design of novel materials requires a thorough understanding of the interplay between their electronic and lattice degrees of freedom. One class of materials in which such an interplay is particularly apparent are the charge-density-wave (CDW) systems, the existence of which was first predicted by Peierls [1]. Peierls argued indeed that one-dimensional (1D) metals are intrinsically unstable, and that a new broken-symmetry ground state results from the self-consistent rearrangement of the electronic charge density in response to the (static) modulation of the ionic positions [12]. The new lattice periodicity leads moreover to the opening of a gap at the Fermi level. The consequences of this intimate connection between electronic properties and lattice dynamics have been intensively investigated in a number of prototype quasi-1D materials [3].

CDWs have been observed in transition metal di- and trichalcogenides [4,5], in the ladder compounds Sr$_{14}$Ca$_4$Cu$_{24}$O$_{41}$ [6] and in some copper oxide high temperature superconductors [7] (where they are known as “stripes”) as well, suggesting that similar effects are to be expected also in layered quasi-2D systems. The rare-earth tri-tellurides RT$_3$ ($R$ = La–Tm, excepting Eu) are the latest paramount examples of 2D dimensional systems exhibiting the formation of an incommensurate CDW state [8,9]. The average crystal structure is layered and weakly orthorhombic, consisting of double layers of nominally square-planar Te sheets, separated by corrugated RT$_3$ slabs [10]. The lattice constant decreases on going from $R$ = La to $R$ = Tm [11], i.e. by chemically compressing the lattice, as a consequence of the reduced ionic radius of the rare-earth atom. The formation of the CDW condensate in RT$_3$ only partially gaps the Fermi surface [12] and therefore these materials remain metallic even well below the critical temperature $T_{CDW}$ at which the CDW appears [9].

Optical spectroscopy is in general an ideal tool to study CDW systems [3]. In our first optical investigations, we have established the excitation across the CDW gap and discovered that this gap is progressively reduced upon compressing the lattice either with chemical substitution (i.e., by changing $R$) or with externally applied pressure [13,14]. Moreover, because of the imperfect nesting, as typical for quasi-2D systems like RT$_3$, the optical data reveal the free-carriers contribution in terms of a Drude peak. The major goal of this short communication is to place our new data on PrTe$_3$ in the context of the results achieved on the chemical series of RT$_3$, thus generalizing our previous concepts.

2. Experiment and results

We report on optical reflectivity measurements carried out on PrTe$_3$, which is deep in the CDW ground state already at 300 K. Single crystals of the title compound were grown by slow cooling a binary melt, as described elsewhere [9]. Plate-like crystals up to several mm in diameter were removed from the melt by decanting in a centrifuge. The crystals could be readily cleaved between Te layers to reveal clean surfaces for the reflectivity measurements. Exploiting several spectrometers and interferometers, the optical reflectivity $R(\omega)$ was measured from the far-infrared (6 meV) up to the ultraviolet (6 eV) spectral range, with light polarized parallel...
to the Te-planes. Details pertaining to the experiments can be found elsewhere [15,16].

The inset of Fig. 1 shows the measured optical reflectivity of PrTe₃ over the whole spectral range, which is the prerequisite in order to perform reliable Kramers–Kronig (KK) transformations. To this end, $R(\omega)$ was extended towards zero frequency (i.e., $\omega \to 0$) with the Hagen–Rubens extrapolation ($R(\omega) = \frac{1}{\omega^2}$) and with standard power-law extrapolations at high frequencies [15,16]. The DC conductivity values employed in the Hagen–Rubens extension of $R(\omega)$ are consistent with transport measurements [9]. The KK transformations allow us to extract the real part $\sigma_1(\omega)$ of the optical conductivity, displayed in the main panel of Fig. 1.

No temperature dependence of the spectrum was observed between 2 and 300 K. At lower frequency a bump is apparent in $R(\omega)$, leading to a depletion around 4000 cm⁻¹, in agreement with previous data on the whole rare-earth series [13]. As expected from the presence of ungapped regions of the FS, all samples exhibit a metallic $R(\omega)$, tending to total reflection at zero frequency (i.e., $R(\omega) \to 1$, for $\omega \to 0$), and the appearance of a plasma edge around 10 000 cm⁻¹. Above the plasma edge, several spectral features are also observed.

3. Discussion

The $\sigma_1(\omega)$ spectrum in Fig. 1 is characterized by the presence of two main features, namely a zero-energy Drude peak, revealing metallic conduction due to the free charge carriers, and a mid-infrared peak centered at finite frequency (arrow in Fig. 1), corresponding to the bump observed in $R(\omega)$ (inset of Fig. 1). According to our previous findings [13], the depletion in the $\sigma_1(\omega)$ spectrum between these two features is identified with the CDW gap, namely to the charge excitation out of the CDW condensate into a single particle (SP) state. In the following we will refer to this peak as the SP peak.

A common procedure, in order to get more quantitative information, consists in exploiting the phenomenological Drude–Lorentz (LD) model (for details see Refs. [13,15,16]). The fit procedure was extended up to 50 000 cm⁻¹. Besides the Drude contribution, five Lorentz harmonic oscillators (h.o.) are required to fit the finite-frequency features. This is explicitly shown in Fig. 1 for PrTe₃, where the single fit components are displayed. The resulting fit on PrTe₃ is totally consistent with our previous analysis on the whole RE₃ series. The three low frequency oscillators allow to reproduce the rather broad absorption, ascribed to the SP peak. This choice is motivated by the fact that the SP peak cannot be fitted with a single Lorentz oscillator. Therefore, the SP peak in our compound can be thought as composed of the superposition of several excitations, which mimic the continuous distribution of gap values, as observed by ARPES [12]. The remaining two high frequency h.o.’s account for the optical (electronic interband) transitions.

There are several interesting parameters (Fig. 2) which can be extracted from such a phenomenological fit. First of all we note the plasma frequency $\omega_p$ (Fig. 2a), the square of which represents the total spectral weight of the Drude peak. The larger the $\omega_p$, the higher the metallic degree of the system. As to the SP peak, since it is composed of three Lorentz h.o.’s, we define the averaged quantity $\omega_{SP}$:

$$\omega_{SP} = \frac{\sum_{j=1}^{3} \omega_j^2 \sigma_j^2}{\sum_{j=1}^{3} \sigma_j^2},$$

Fig. 1. $\sigma_1(\omega)$ of PrTe₃ at 300 K. The arrow marks the position of the SP peak (see text). The Drude–Lorentz (LD) fit, showing the fitted curve, the Drude and the Lorentz components, is also displayed. Inset: $R(\omega)$ of PrTe₃ at room temperature.

Fig. 2. Plasma frequency ($\omega_p$) (a) and single-particle peak frequency ($\omega_{SP}$) (b) as a function of the in-plane lattice constant for the whole rare-earth series [11,13]. The rare-earth atom for each compound is shown in the plot. The solid lines are guides to eyes. The new data presented here on PrTe₃ are highlighted with a dedicated symbol.
which represents the center of mass of the SP excitation and thus provides an optical estimate for the CDW gap [13]. In Fig. 2b, \( \omega_{SP} \) is plotted as a function of the lattice constant \( a \) [11]. The decrease of \( \omega_{SP} \) (thus of the average CDW gap value) is well evident on going from LaTe\(_3\) to DyTe\(_3\), oppositely to the observed increase of \( \omega_p \) (thus of the Drude spectral weight, Fig. 2a). The reduction of \( \omega_{SP} \) on decreasing \( a \) (Fig. 2b) may be considered as an indication for the diminishing impact of the CDW state, when going from La to Dy. In recent ARPES experiments [12] a reduction of the CDW gap-value with decreasing \( a \) [13] of the perfectly nested regions (where the gap is close to zero). Within this scenario, additional charge carriers are then released in the normal metallic state leading to the increase in the ungapped portions of FS with decreasing \( a \) [13]. This induces an enhanced optical contribution due to the free charge carriers, therefore of \( \omega_{SP} \) (Fig. 2a). It is worth mentioning that the behavior of these optical parameters in general and specifically of \( \omega_{SP} \) is totally consistent with findings on related RTe\(_3\) (\( n = 2 \) and 2.5) compounds, as well [17,18].

Finally, we have also observed a power-law behavior \( (\sigma_1(\omega) - \sigma_0^n) \) in RTe\(_3\) for frequencies above \( \omega_{SP} \). This is shown in Fig. 3 for all investigated compounds, plotted on a bi-logarithmic scale. The power-law behavior extends over a rather limited energy interval, which at most is one order of magnitude wide for the Dy compound. The results on PrTe\(_3\) fit again perfectly within the trend given by the whole chemical series [13]. Power-law scaling is indicative of quasi-1D behavior [19,20]. Previously we argued that for RTe\(_3\) an exponent \( \eta \) of the order of \(-1\) is the consequence of direct interaction between electrons, as source forUmklapp scattering [13,21]. As a consequence of the direct electron–electron interaction there is a transfer of two particles from one sheet of the FS to the other, thus transferring \( 4q \) to the lattice [21]. Note that the advocated wave-vector [12] for the CDW modulation is indeed giving a value for \( 4q \) close to a reciprocal lattice vector, allowing such Umklapp processes to be effective. At least at high energy scales, there is then evidence of a typical Tomonaga–Luttinger liquid scenario, emphasizing a non-negligible contribution of 1D correlation effects in the physics of these 2D compounds [13,21]. In such a case, a standard electron–phonon mechanism may not be enough to fully account for the CDW phase transition, although further experiments will be necessary to firmly establish this hypothesis.

4. Conclusions

In summary, we reported on the complementary optical measurements of PrTe\(_3\). This latter experiment is totally consistent within the major findings collected on other members of the rare-earth tri-tellurides family. Our data allow for a detailed analysis of both the Drude contribution, ascribed to the free charge carriers resulting from the presence of ungapped regions of FS, and the SP peak, due to carriers’ excitation across the CDW gap. We also observe power-law behaviors in \( \sigma_1(\omega) \) which anticipates that interactions and Umklapp processes could play a significant role in the CDW formation in these compounds. Of course more studies both theoretically and experimentally would be useful to ascertain the respective roles of the interactions and of electron–phonon coupling with respect to the CDW state. Theoretically, a more careful treatment of the effects of the transverse warping of FS would be clearly needed, not only for the high energy behavior but even more for the low frequency part of the optical conductivity, much below the single particle peak. This is one of many challenging topics to be addressed for the time to come in relation to low dimensional interacting electron gas systems.

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