

Pressure Dependence of the Charge-Density-Wave Gap in Rare-Earth Tritellurides

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We investigate the pressure dependence of the optical properties of CeTe_3 , which exhibits an incommensurate charge-density-wave (CDW) state already at 300 K. Our data are collected in the midinfrared spectral range at room temperature and at pressures between 0 and 9 GPa. The energy for the single particle excitation across the CDW gap decreases upon increasing the applied pressure, similarly to the chemical pressure by rare-earth substitution. The broadening of the bands upon lattice compression removes the perfect nesting condition of the Fermi surface and therefore diminishes the impact of the CDW transition on the electronic properties of $R\text{Te}_3$.

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The physical properties of low-dimensional systems have fascinated researchers for a great part of the last century, and have recently become one of the primary centers of interest in condensed matter research. Low-dimensional systems not only experience strong quantum and thermal fluctuations, but also admit ordering tendencies which are difficult to realize in three-dimensional materials. Prominent examples are spin- and charge-density waves in quasi-one-dimensional compounds [1]. Moreover, the competition among several possible order parameters leads to rich phase diagrams, which can be tuned by external variables as temperature, magnetic field, and both chemical and applied pressure [1,2]. Tunable external parameters also affect the effective dimensionality of the interacting electron gas and thus the intrinsic electronic properties of the investigated systems.

The rare-earth tetratellurides $R\text{Te}_3$ ($R = \text{La-Tm}$, excepting Eu) are the latest paramount examples of low dimensional systems exhibiting the formation of an incommensurate charge-density-wave (CDW) state [3,4]. The average crystal structure is layered and weakly orthorhombic, consisting of double layers of nominally square-planar Te sheets, separated by corrugated $R\text{Te}$ slabs [5]. The lattice constant decreases on going from $R = \text{La}$ to $R = \text{Tm}$ [6], i.e., by chemically compressing the lattice, as consequence of the reduced ionic radius of the rare-earth atom [7]. The CDW state in $R\text{Te}_3$ can be then investigated as a function of the in-plane lattice constant a , which is directly related to the Te-Te distance in the Te layers.

Recently, we have reported on the first optical measurements of $R\text{Te}_3$ [8]. Our data, collected over an extremely broad spectral range, allowed 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 excitation across the CDW gap, respectively. We established a diminishing impact of the CDW condensate on the

electronic properties of $R\text{Te}_3$ with decreasing a across the rare-earth series [8]. On decreasing a , a reduction of the CDW gap together with an enhancement of the metallic (Drude) contribution were observed in the absorption spectrum. This is the consequence of a quenching of the nesting condition, driven by the modification of the Fermi surface (FS) because of the lattice compression [8].

We present in this Letter infrared optical investigations of the pressure dependence of the optical reflectivity on CeTe_3 at 300 K, i.e., below the CDW transition temperature [4,9]. The motivation of this work originates from the fact that $R\text{Te}_3$ generally provides an adequate playground to study the effect of chemical pressure and externally applied pressure in shaping the predisposition of these materials to undergo a CDW phase transition. Upon increasing pressure the excitation due to the CDW gap decreases in a quite equivalent manner when compressing the lattice by substituting large with small ionic radius rare-earth elements (i.e., by reducing a). These results demonstrate that chemical and applied pressure similarly affect the electronic properties and equivalently govern the onset of the CDW state in $R\text{Te}_3$.

Single crystals of CeTe_3 were grown by slow cooling a binary melt, as described elsewhere [9]. A small piece of CeTe_3 (i.e., $50 \times 50 \mu\text{m}^2$) was cut from the same sample previously used in Ref. [8] and was placed on the top surface of a KBr pellet presintered in the gasket hole of the pressure cell. The gasket was made of stainless steel, 50 μm thick and with a 200 μm diameter hole. A clamp-screw diamond anvil cell (DAC) equipped with high-quality type IIa diamonds (400 μm culet diameter) was employed for generating high pressure up to 9 GPa. Pressure was measured with the standard ruby-fluorescence technique [10]. Because of the metallic character of the sample, absorption measurements are not possible on this compound. Therefore, we carried out optical reflectivity

measurements exploiting the high brilliance of the infrared beam line at ELETTRA synchrotron in Trieste [11]. The incident and reflected light were focused and collected by a cassegrainian-based optical microscope equipped with a (HgCdTe)-detector and coupled to a Bruker Michelson interferometer, which allows to explore the 600–8000 cm^{-1} spectral range. At each pressure, we measured the light intensity reflected by the sample $I_S(\omega)$ and by the external face of the diamond window $I_D(\omega)$, thus obtaining the quantity $R_D^S(\omega) = I_S(\omega)/I_D(\omega)$. At the end of the pressure run, we also measured the light intensity reflected by a gold mirror [$I_{Au}(\omega)$] placed between the diamonds at zero pressure and again $I_D(\omega)$, acting as a reference. One achieves $R_D^{Au}(\omega) = I_{Au}(\omega)/I_D(\omega)$, which is assumed to be pressure independent. This procedure allows us to finally obtain the sample reflectivity $R(\omega) = R_D^S(\omega)/R_D^{Au}(\omega)$ at each pressure, which takes into account the variations in the light intensity due to the smooth depletion of the current in the storage ring. The strong diamond absorption at about 2000 cm^{-1} and the presence of diffraction effects (see below) prevent data reliability at low frequencies. Therefore, we display the data in the 2700–8000 cm^{-1} range.

$R(\omega)$ of CeTe_3 at 300 K and 0.3 GPa is shown in Fig. 1(a), together with the corresponding $R(\omega)$ at ambient pressure (i.e., outside the cell) [8]. Figure 1(a) also reproduces the expected $R(\omega)$ of CeTe_3 calculated from the complex refractive index at zero pressure [8] and assuming the sample inside the DAC [12–14]. We immediately observe that the expected $R(\omega)$ spectrum inside the DAC is lower than the one at 0 GPa in air but still considerably higher than the experimental finding. We ascribe this difference to diffraction effects induced by the nonperfectly flat shape of the sample. In order to take into account these diffraction effects, we define a smooth correction function [Fig. 1(a)] which is then applied to all spectra. We justify our choice for the correction function by pointing out that it is somehow more effective at high frequencies (as expected for diffraction effects) and that it shows a strong frequency dependence only below 2700 cm^{-1} . Furthermore, we checked that the final corrected spectra as well as the data analysis do not substantially change when correction procedures based on a simple scaling by a constant factor or on adding a constant background to the measured spectra are employed.

Figure 1(b) reproduces the corrected spectra of CeTe_3 at selected pressures. Although the light spot was precisely limited (by means of fissures) to the sample area, there is still some diffused light giving rise to the interference pattern (between the diamond windows) observed in the spectra. The striking feature is the filling in of the deep minimum in $R(\omega)$ at about 3500 cm^{-1} with increasing pressure, quite similar to the behavior of $R(\omega)$ across the rare-earth series (inset of Fig. 1 in Ref. [8]). The depletion at 3500 cm^{-1} was ascribed to the charge excitation across the CDW gap into a single particle (SP) state [8]. A more

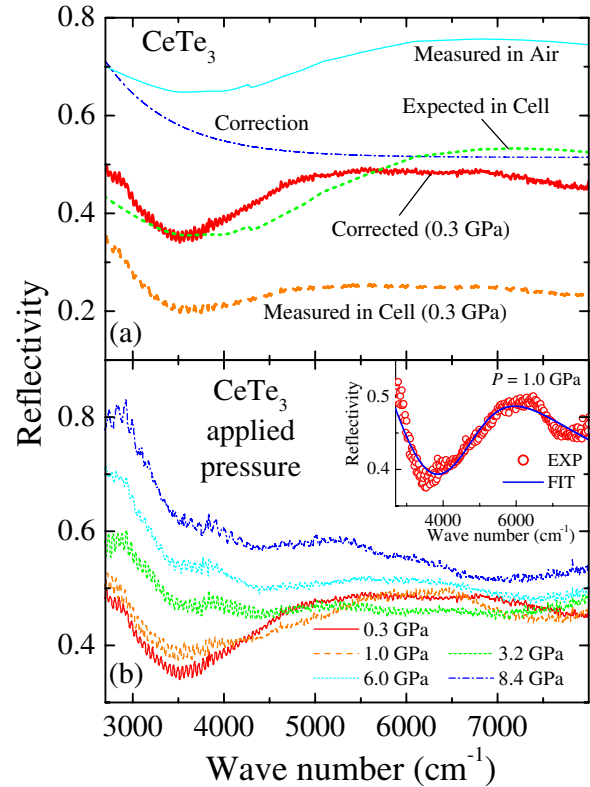


FIG. 1 (color online). (a) Raw $R(\omega)$ data of CeTe_3 at 300 K and 0.3 GPa compared with the measured spectrum in air [8] and its expectation inside the DAC. The correction function accounting for the diffraction effects is reproduced, as well as the corrected reflectivity of CeTe_3 at 0.3 GPa. (b) $R(\omega)$ of CeTe_3 at 300 K and selected applied pressures. The inset shows the very good reproducibility of the data at 1 GPa within the Drude-Lorentz model (see text).

compelling comparison is given in Fig. 2, displaying the ratio of the $R(\omega)$ spectra of CeTe_3 at selected pressures with respect to the spectrum at the highest measured pressure [Fig. 2(a)] and the ratio of the $R(\omega)$ spectra for selected rare-earth compounds with respect to $R(\omega)$ of DyTe_3 [Fig. 2(b)] [8]. The obvious similarity between the $R(\omega)$ ratios upon increasing pressure and when moving from the La to the Dy compound suggests the equivalence between chemical and applied pressure.

The optical findings on the rare-earth series were systematically reproduced within the Drude-Lorentz fit: the most relevant components were the Drude term ascribed to the effective metallic contribution and three Lorentz harmonic oscillators (HO) which are necessary for reproducing the broad SP absorption. Therefore, the SP peak is composed by the superposition of several excitations, which mimic the continuous distribution of gaps, as observed in angle resolved photoemission spectroscopy (ARPES) [8,15]. The same fit procedure [Fig. 3(a)] is applied here to the pressure dependent $R(\omega)$ spectra of CeTe_3 [14]. Only the parameters of the three Lorentz HO's, describing the SP excitation, were allowed to change as a function of pressure. All other components (Drude term

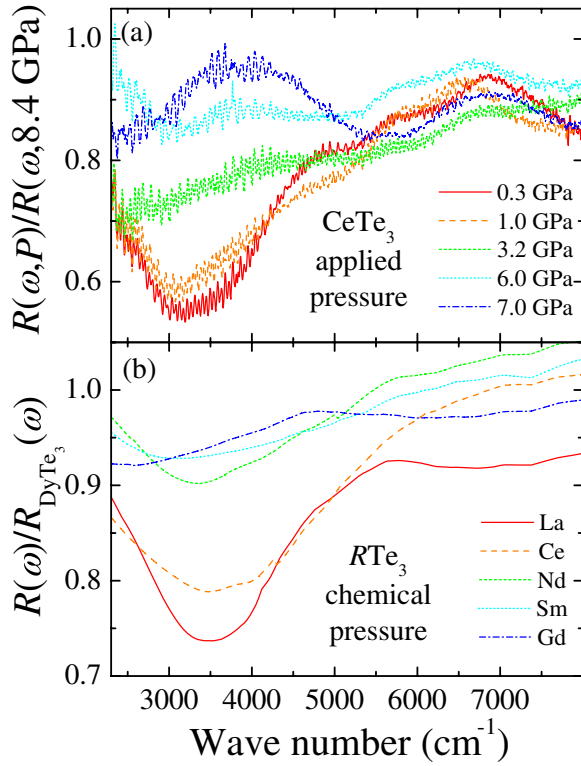


FIG. 2 (color online). (a) $R(\omega)$ of CeTe_3 at 300 K and selected applied pressures, normalized by the spectrum at 8.4 GPa. (b) $R(\omega)$ of RTe_3 at 300 K and ambient pressure, normalized by the spectrum of DyTe_3 [8].

and electronic interband transitions) were left fixed, by exploiting the best fit of CeTe_3 at ambient pressure outside the DAC [8]. We also tested slightly different fitting procedures obtaining similar results. By fitting $R(\omega)$ spectra in the energy interval displayed in Fig. 1(b), we can then reconstruct the real part $\sigma_1(\omega)$ of the optical conductivity of CeTe_3 at selected pressures. This is shown in Fig. 3(a), while the inset of Fig. 1(b) exemplifies the good fit quality of $R(\omega)$ at 1 GPa. There is an overall good correspondence with $\sigma_1(\omega)$ of the rare-earth series [Fig. 3(b)] [8].

In order to push further the comparison between chemical (i.e., rare-earth dependence) and applied pressure, we first need to establish the pressure dependence of the lattice constant $a(P)$. A direct experimental determination of $a(P)$ is still missing, but we can extract this latter quantity from the zero-pressure bulk modulus B_0 . First of all, from the β value [$\beta = (2\pi^2/5)k_B(k_B/\hbar v_s)^3$] of the phononic part of the specific heat in LaTe_3 [9,16] one achieves the sound velocity $v_s = 1923$ m/s [17]. Knowing that $B_0 = \rho v_s^2$, $\rho = 6837$ kg/m³ being the density, one gets $B_0 = 25$ GPa. We can then assume a linear pressure dependence of the bulk modulus $B(P) = B_0 + B'P$, where B' usually ranges between 4 and 8 [18]. This leads to the so-called Murnaghan equation for the pressure dependence of the volume [19]:

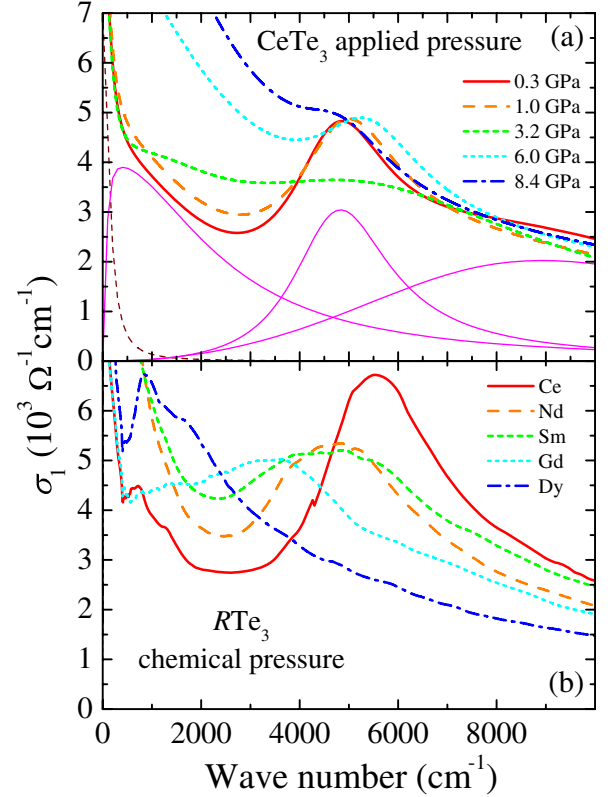


FIG. 3 (color online). (a) Real part $\sigma_1(\omega)$ of the optical conductivity of CeTe_3 at 300 K and selected pressures, calculated from the fit of $R(\omega)$ within the Drude-Lorentz model (see text). The Drude-Lorentz fit components at 0.3 GPa are also displayed. (b) $\sigma_1(\omega)$ at 300 K of RTe_3 ($R = \text{Ce}, \text{Nd}, \text{Sm}, \text{Gd}, \text{and Dy}$), obtained through Kramers-Kronig transformation of the measured $R(\omega)$ spectra [8].

$$V(P) = V(0) \left(1 + \frac{B'}{B_0} P \right)^{-1/B'} \quad (1)$$

from which we can immediately obtain $a(P) = a(0) \times [V(P)/V(0)]^{1/3}$. The inset of Fig. 4 shows the range within which the pressure dependence of a can evolve for the two limits of B' .

The three HO's for the SP excitation [Fig. 3(a)] allow us to define the single energy scale ω_{SP} at each pressure [20], describing the overall gap excitation. The main panel of Fig. 4 displays the resulting dependence of ω_{SP} on the lattice constant; $\omega_{SP}(a)$ for CeTe_3 at different pressures is determined for the average of $a(P)$ between the $B' = 4$ and 8 curves (inset of Fig. 4), while $\omega_{SP}(a)$ for the rare-earth series is reproduced from Fig. 3b of Ref. [8]. There is again a similar trend between the two sets of data, even though the two curves do not fully overlap. A perfect correspondence is anyhow not expected in view of the approximations, employed for the determination of $a(P)$. In particular, the observed discrepancy could be ascribed to an underestimate of B_0 , as well as to the assumption of isotropic compression [$a(P) \propto V^{1/3}(P)$]. It appears that ω_{SP} gets smaller upon decreasing a . Analogous to the rare-earth series [8], such a reduction of ω_{SP} on decreasing

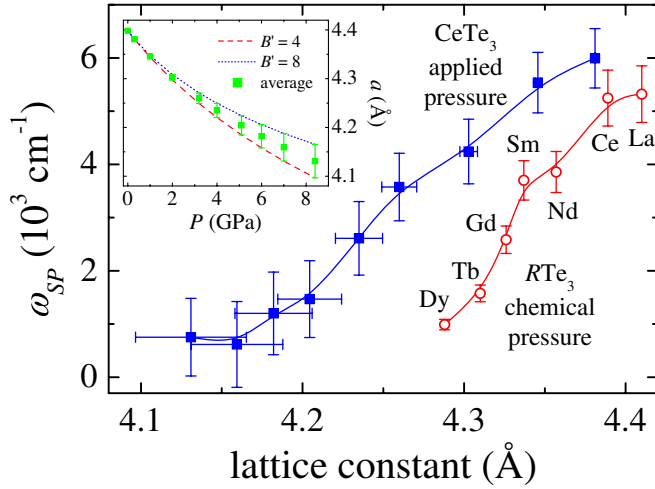


FIG. 4 (color online). Single particle excitation energy ω_{SP} as a function of the lattice constant a for CeTe_3 under applied pressures and for the $R\text{Te}_3$ series [8]. Solid lines are guides to the eye. Inset: calculated pressure dependence of a (see text).

a may be considered as an indication for the lesser impact of the CDW state upon increasing pressure. Pressure broadens the bands and changes the shape of FS in such a way to alter the favorable nesting conditions, which are the prerequisite for the formation of the CDW condensate [1]. The decrease of ω_{SP} upon compressing the lattice is then consistent with the reduction of the perfectly nested FS regions (with large CDW gap) in favor of the non-perfectly nested ones (with small CDW gap). This induces moreover an increase of spectral weight (Fig. 3) at low frequencies. Although our data in pressure do not extend to the far infrared it is safe to say that part of this spectral weight merges into the effective metallic component of $\sigma_1(\omega)$, as observed with chemical pressure [8]. There is an indirect support to these conclusions by a recent ARPES experiment [15], where the CDW gap decreases with a for several $R\text{Te}_3$ compounds.

In conclusion, we have reported the first optical investigation of the pressure dependence of the single particle excitation across the CDW gap in CeTe_3 . Pressure affects the gapping of FS so that the CDW gap is progressively suppressed on decreasing the lattice constant. Therefore, our findings confirm the equivalence between applied and chemical pressure in the rare-earth tritelluride series. The formation of the CDW state in $R\text{Te}_3$ was also considered as an indication for a hidden one-dimensional behavior in these quasi-two-dimensional compounds [8,21]. This work does not address to which extent the applied pressure might influence the effect of electron-electron interactions and umklapp processes, as suggested in Ref. [8], as well as the dimensionality crossover, in driving the CDW transition. This awaits for further experimental effort, allowing the extension of the measured spectral range under pressure up to higher as well as to lower energies than the energy window presented here. This could open new perspectives to a comprehensive study about the pressure

dependence of the characteristic power law behavior, seen in the absorption spectrum of the $R\text{Te}_3$ series [8], and more generally about the influence of pressure in the formation of the Luttinger liquid state in quasi-one-dimensional systems.

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