Abstract submitted to the
Conference on Concepts in Electron Correlation
September 24 - 30, 2008 Hvar, Croatia

Infrared and Raman study of the charge-density-wave ground state

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Submitted : 25-03-2008
Keywords : charge-density-wave, optical properties, pressure dependence

The question of the relationship between the effective dimensionality of a physical system and the symmetry of its ground state is an important issue in order to figure out the relevant mechanism driving materials into peculiar charge ordering. In this respect, the rare-earth polychalcogenides $R\mathrm{Te}_n$ (where R is the rare earth element and n=2, 2.5, 3) have recently attracted great interest because of their low dimensionality. Among the $R\mathrm{Te}_n$ families are members that variously host large commensurate distortions, ordered and disordered vacancy structures, and Fermi surface driven charge-density-wave (CDW). They supply furthermore a playground to study the interplay between CDW state, peculiar magnetic order and eventually pressure induced superconductivity. Optical spectroscopic methods (infrared reflectivity and Raman scattering) were applied as a function of both temperature and external pressure, in order to address the complete excitation spectrum in these CDW materials. We establish the energy scale of the single particle excitation across the CDW gap and find that the CDW collective state gaps a large portion of the Fermi surface. The CDW gap decreases upon compressing the lattice (both with chemical and applied pressure, Fig. 1). The suppression of the CDW gap leads to a release of additional charge carriers, manifested by the shift of weight from the gap feature into the metallic component of the optical response (inset Fig. 1). Furthermore based on the observation of a power law behavior in the optical conductivity, we suggest that interactions and Umklapp processes may play a role in the onset of the CDW broken symmetry ground state. We discuss our optical conductivity at high frequencies with respect to predictions based on the Tomonaga-Luttinger liquid scenario. We will moreover present our Raman scattering experiments as a function of chemical and applied pressure. The observed spectra display five peaks (Fig. 2), four of which we can assign to A_{1g} Raman active phonon modes by comparing their frequencies to those obtained from a first principles calculation. The latter also produces the Kohn anomaly in the phonon dispersion, eventually responsible for the formation of the CDW condensate. Furthermore, distorting the lattice along the displacements of the soft phonon, modes of B_{1g} symmetry become Raman active in our experimental geometry. Our Raman scattering results give clear cut evidence for a coupling between the CDW condensate and the lattice vibrational modes.

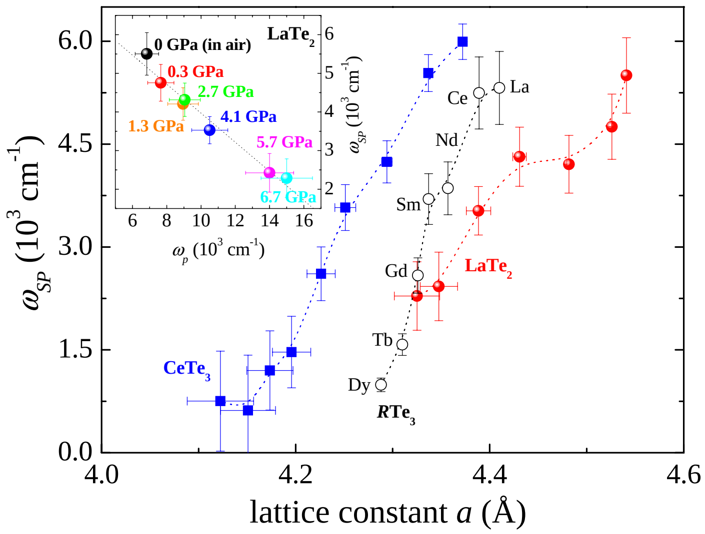

Figure 1: Single particle peak energy ω_{SP} (i.e., the CDW gap) as a function of the lattice constant a for $LaTe_2$, $CeTe_3$ and the RTe_3 series. Inset: single particle peak energy ω_{SP} versus plasma frequency ω_p for $LaTe_2$, as a function of pressure. Pressure is here an implicit variable.

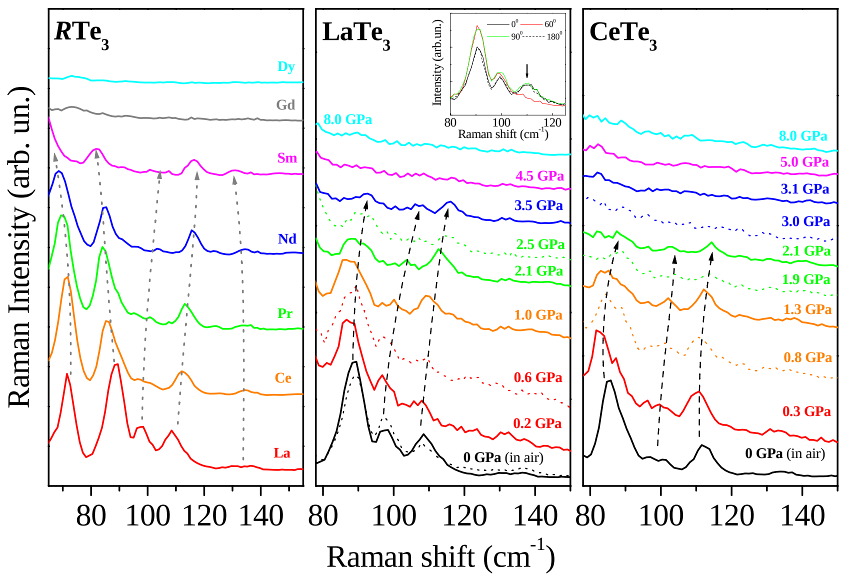

Figure 2: Raman scattering spectra for the rare-earth tri-tellurides series RTe_3 (R=La, Ce, Pr, Nd, Sm, Gd and Dy) at ambient pressure (a), and for $LaTe_3$ (b) and $CeTe_3$ (c) both for increasing (continuous line) and decreasing (dashed line) pressure. All spectra have been shifted for clarity. The pressure dependence of all modes is fully reversible. All modes slightly disperse (thin dotted lines as guides to the eye) and disappear upon compressing the lattice. The inset in Fig. 2b shows the polarization dependence of the Raman spectra of $LaTe_3$ for parallel incident and scattered light polarizations and at different orientations of the incident polarization. The origin of the angle scale is arbitrary.