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Classification of Collective Modes in a Charge Density Wave by Momentum-Dependent Modulation of the Electronic Band Structure

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We present time- and angle-resolved photoemission spectroscopy measurements on the charge density wave system CeTe₃. Optical excitation transiently populates the unoccupied band structure and reveals a gap size of $2\Delta = 0.59$ eV. The occupied Te-5*p* band dispersion is coherently modified by three modes at $\Omega_1 = 2.2$ THz, $\Omega_2 = 2.7$ THz and $\Omega_3 = 3$ THz. All three modes lead to small rigid energy shifts whereas Δ is only affected by Ω_1 and Ω_2 . Their spatial polarization is analyzed by fits of a transient model dispersion and DFT frozen phonon calculations. We conclude that the modes Ω_1 and Ω_2 result from in-plane ionic lattice motions, which modulate the charge order, and that Ω_3 originates from a generic out-of-plane A_{1g} phonon. We thereby demonstrate how the rich information from trARPES allows identification of collective modes and their spatial polarization, which explains the mode-dependent coupling to charge order.

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Interaction of multiple degrees of freedom in quantum materials can result in ordering phenomena that give rise to complex phase diagrams with competing or intertwined orders¹. Such broken-symmetry ground states host characteristic low-energy excitations, which are intimately related to the interactions responsible for ordering. Ultrafast non-equilibrium methods are a new and active field for the investigation of broken-symmetry phases as they grant direct access to collective modes in the time-domain²⁻⁷ and can provide strong evidence for the ordering mechanism operating in a particular material^{8,9}.

Charge density wave (CDW) formation is one of the most basic and widely studied cases for long-range ordering when only electronic and lattice degrees of freedom are interacting¹⁰. Below a critical temperature T_{CDW} a spontaneous lattice distortion Δz appears, generally originating via the complex interplay of the electronic structure and lattice. This results in a spatial charge modulation at a wave vector \vec{q} (spatial period $2\pi/q$) and opening of a bandgap 2Δ in the band structure, which minimizes the system's total energy by reducing the kinetic energy of the electronic system. One of the characteristic collective excitations in such Peierls-like CDWs is a gapped amplitude mode (AM), which modulates the magnitude $\Delta(t)$ of the order parameter¹⁰.

Quasi-2D rare-earth tritellurides $R\text{Te}_3$ ($R=\text{Y, La, Sm, Gd-Tm}$) form an incommensurate CDW along the crystallographic *c*-axis and have been studied extensively¹¹⁻¹⁹. At low temperatures, the heaviest members of the $R\text{Te}_3$ series ($R=\text{Dy-Tm}$) additionally order with a second CDW perpendicular to the first CDW^{15,19,20}. Angle-resolved photoemission spectroscopy (ARPES) reveals the Fermi surface (FS) topology and the momentum-dependent gap structure²⁰⁻²⁴.

Collective modes in $R\text{Te}_3$ were studied with

Raman spectroscopy^{19,25,26}, time-resolved optical reflectivity^{7,27,28}, and time-resolved ARPES (trARPES)^{4,29,30}. Yet even for this thoroughly investigated CDW system unambiguous identification of an AM is non-trivial. Lowering the translational symmetry in the CDW phase leads to the renormalization of the normal state optical and acoustic phonon branches at \vec{q} ²⁵⁻²⁷. Ubiquitously observed generic phonon modes add to the difficulty of mode identification. Direct comparison of modes observed in the CDW phase to calculated phonon spectra is hampered by the incommensurate CDW in $R\text{Te}_3$. These challenges are present in CDW materials in general and have led to conflicting reports regarding the AM in $R\text{Te}_3$ in particular. In DyTe₃, for instance, a 1.75 THz mode is observed in both temperature-dependent transient reflectivity²⁷, Raman spectroscopy²⁶ and a trARPES experiment using a pump-pump-probe scheme³⁰. Optical measurements attribute this mode to a phonon in the lower symmetry CDW state that is coupled to the AM, whereas trARPES³⁰ assigns it to a second AM.

In this paper, we report a trARPES study of the collective response of CeTe₃ in the CDW phase. The unoccupied band structure is transiently populated by optical transitions, which reveals a gap size of $2\Delta = 0.59(2)$ eV. The occupied Te-5*p* band dispersion near the CDW gap is coherently modified by three modes and analyzed within a transient tight-binding (TB) model. Momentum dependent analysis combined with DFT frozen phonon calculations reveals different coupling to the band structure for in-plane AMs and out-of-plane optical phonons.

Small rigid shifts of the band dispersion E_0 are introduced by all three modes, whereas much larger periodic modulations of Δ are driven by lattice motions $\Omega_1 = 2.2$ THz and $\Omega_2 = 2.7$ THz. We explain this ob-

servation by concluding that both modes Ω_1 and Ω_2 result from in-plane ionic motions which are coupled to the CDW and hence drive oscillations of Δ as AMs. The $\Omega_3 = 3$ THz mode originates from an out-of-plane optical A_{1g} phonon that does not couple to the CDW. Our work demonstrates how careful evaluation of the rich information from trARPES allows unambiguous identification of AMs in charge-ordered materials.

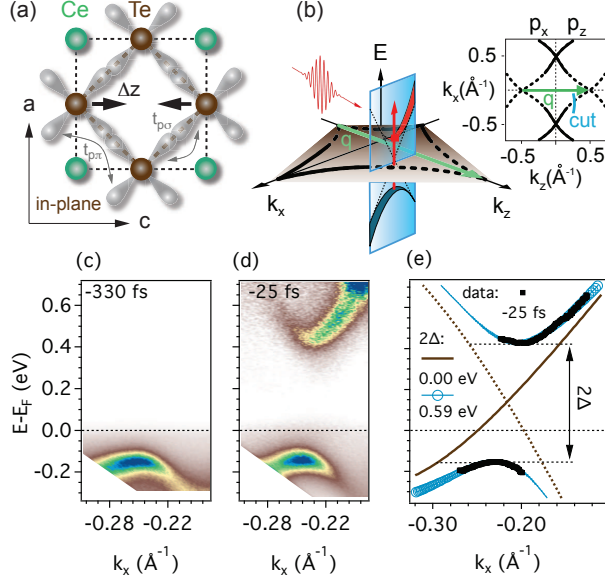


FIG. 1. (a) Structure of the Te-planes where the CDW forms along the c -axis with lattice displacement Δz indicated. The FS is described in a TB model, with the overlap integrals $t_{p\sigma}$ along and $t_{p\pi}$ perpendicular to Te- $5p$ orbitals. (b) Cartoon of the trARPES experiment on CeTe₃ in the CDW region of the FS (dotted lines). The blue plane illustrates an energy versus momentum cut with Te- $5p$ bands and a pump pulse exciting electrons into unoccupied states. The blue line indicates the momentum cut in the experiment across the gapped region (dotted lines) of the calculated 2D FS, green arrow denotes the CDW wave vector \vec{q} . trARPES spectra for selected pump-probe delays, 330 fs before the arrival of the pump pulse (c) and during the presence of the pump pulse (d). Intensities are rescaled exponentially as function of energy for enhanced visibility of the transiently occupied features above E_F . (e) TB fit (blue markers) according to Eqs. (1) and (2) to the extracted band dispersion (black markers) at -25 fs yields $2\Delta = 0.59(2)$ eV. Size of the blue markers is proportional to the calculated spectral weight of the TB bands²⁴. Brown lines indicate the calculated metallic bare band dispersion.

Our trARPES setup^{31,32} consists of an amplified laser system operating at 310 kHz repetition rate that drives an optical parametric amplifier to optically excite the sample with ~ 50 fs pump pulses at a photon energy of 1.03 eV. Electrons are photoemitted with ~ 150 fs probe pulses at 6 eV as function of pump-probe delay and collected with 50 fs delay steps in a hemispherical electron analyzer, yielding a total energy resolution of ~ 22 meV. CeTe₃ single crystals were grown by slow

cooling of a binary melt¹⁴. Measurements were performed on freshly cleaved CeTe₃ samples at $T = 100$ K in an ultrahigh vacuum chamber with a pressure of $< 1 \times 10^{-10}$ torr. Phonon eigenmodes of the normal state of RTe₃ were calculated from the dynamical matrix using the Quantum-ESPRESSO³³ package with ultrasoft pseudopotentials and the Perdew-Burke-Ernzerhof³⁴ exchange-correlation functional, a kinetic energy cutoff of 500 eV, and $13 \times 3 \times 13$ k -points. Incident pump fluences of $55 - 354 \mu\text{J}/\text{cm}^2$ correspond to absorbed energies of $16 - 103$ meV per unit cell at the surface³⁵. 2Δ decreases by no more than 15% and the system responds linearly³⁵ without melting of the charge order. This is in contrast to previous studies at higher excitation densities^{4,29,30}, where qualitative changes in the dispersion prevent decoupling the effects of coherent in-plane and out-of-plane motions.

Fig.1(b) illustrates the cut through the FS where the trARPES data, which are shown in Fig. 1(c) and (d) for selected pump-probe delays, have been taken. Due to an asymmetric band dispersion and a slightly curved cut in the $\vec{k} = \{k_x, k_z\}$ plane the Fermi momentum k_F does not exactly coincide with the maximum (minimum) position of the occupied (unoccupied) band. For simplicity, dispersions along \vec{k} are projected on k_x . At a pump-probe delay of -330 fs the Te- $5p$ band with the occupied part of the CDW gap is observed in equilibrium. At -25 fs, the leading edge of the pump pulse promotes electrons into unoccupied Te- $5p$ states without yet changing the band dispersion significantly, which occurs at later delays. The magnitude of the full CDW gap $2\Delta = 0.59(2)$ eV is directly given by the difference of the maximum of the lower band at $E - E_F = -0.16(1)$ eV and the minimum of the upper band at $E - E_F = 0.43(1)$ eV. This is in agreement with previous results from optical reflectivity^{13,16} while being smaller than extracted from ARPES²² and larger than estimated by tunneling experiments¹⁷.

To obtain a quantitative description of the electronic bands near E_F we consider a TB model of the in-plane Te- $5p_{x/z}$ orbitals^{19,24} as sketched in Fig. 1(a). Nesting of the metallic bare bands $\epsilon(\vec{k})$ via \vec{q} creates two branches $E_{1,2}(\vec{k})$, which are separated by 2Δ ¹⁰:

$$E_{1,2}(\vec{k}) = \frac{\epsilon_{\vec{k}} + \epsilon_{\vec{k}-\vec{q}}}{2} \pm \sqrt{\left(\frac{\epsilon_{\vec{k}} - \epsilon_{\vec{k}-\vec{q}}}{2}\right)^2 + \Delta^2}. \quad (1)$$

$\epsilon(\vec{k})$ is approximated by a non-interacting TB model^{19,24} of the in-plane Te- $5p$ orbitals, see Fig. 1(a).

$$\begin{aligned} \epsilon(\vec{k}) = & -2t_{p\sigma} \cos((k_x - k_z)a/2) \\ & - 2t_{p\pi} \cos((k_x + k_z)a/2) - E_F + E_0 \end{aligned} \quad (2)$$

The TB coupling parameters $t_{p\sigma} = -1.9$ eV along and $t_{p\pi} = 0.35$ eV perpendicular to the Te-chains (Fig. 1(a)) have been established by ARPES experiments^{20,22,24}. $E_F = -2t_{p\sigma} \sin(\pi/8)$ and $|\vec{q}| = 0.685(2\pi/a)$ are constant. The in-plane lattice spacing is given by $a =$

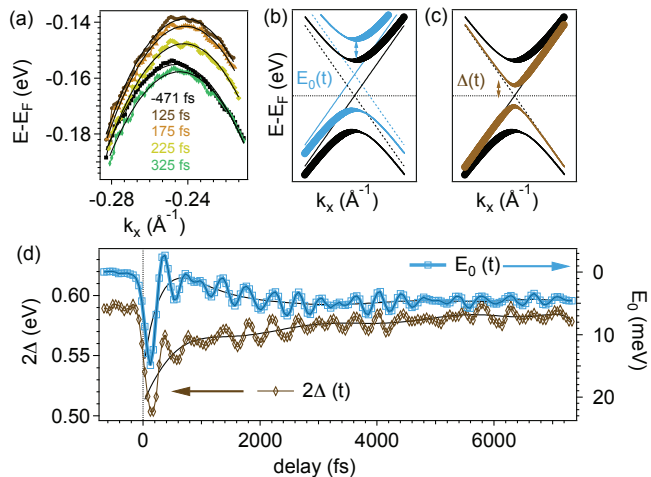


FIG. 2. (a) Measured dispersion of the occupied Te-5p band for selected delays (*markers*) and transient TB fits (*lines*). Sketched effect of a transient rigid energy shift $E_0(t)$ (b) and change of the CDW gap $\Delta(t)$ (c) on the band dispersion. (d) $2\Delta(t)$ and $E_0(t)$ with smooth backgrounds (*black lines*) of a 10^{th} order polynomial indicated.

4.34 \AA^{15} . The band dispersion is extracted by fitting a Gauss function to the spectra as function of \vec{k} , and yield the data points in Fig. 1(e). Eqs. (1) and (2) are combined and simultaneously fitted to the extracted dispersion above and below E_F at -25 fs as shown in Fig. 1(e). The rigid energy shift $E_0 = 0$ is kept fixed and the only free fit parameter is Δ . This yields $2\Delta = 0.59(1)$ eV, which is identical to the value derived from the difference of upper and lower band energies at -25 fs.

After having established that the TB model is well suited to describe the gapped dispersion near E_F , it is applied to the occupied band for all delays, as shown in Fig. 2(a), with $\Delta(t)$ and $E_0(t)$ now being free time-dependent fit parameters. The low excitation regime allows for holding the TB parameters $t_{p\sigma}$ and $t_{p\pi}$ constant. $\Delta(t)$ leads to momentum-dependent modulations in the spectra, and is naturally sensitive to the AM¹⁰ as illustrated in Fig. 2(c). In contrast, $E_0(t)$ is a momentum-independent energy shift of the whole band structure, as sketched in Fig. 2(b). As we will show, these parameters capture the two types of modes observed.

Results for $2\Delta(t)$ and $E_0(t)$ are shown in Fig. 2(d). The magnitude of $E_0(t)$ amounts to a few meV and is much smaller than the changes in $2\Delta(t)$, which are on the order of several 10 meV. Within the pump pulse duration, 2Δ drops by 15 % yet the charge order is only perturbed and not destroyed³⁵. After ~ 3 ps, the system has thermalized at reduced gap size, which agrees with the timescale of the suppression of the structural order parameter satellite observed in time-resolved electron diffraction^{36,37}.

The pronounced beating patterns of $2\Delta(t)$ and $E_0(t)$ exhibit an oscillatory response with similar yet distinct

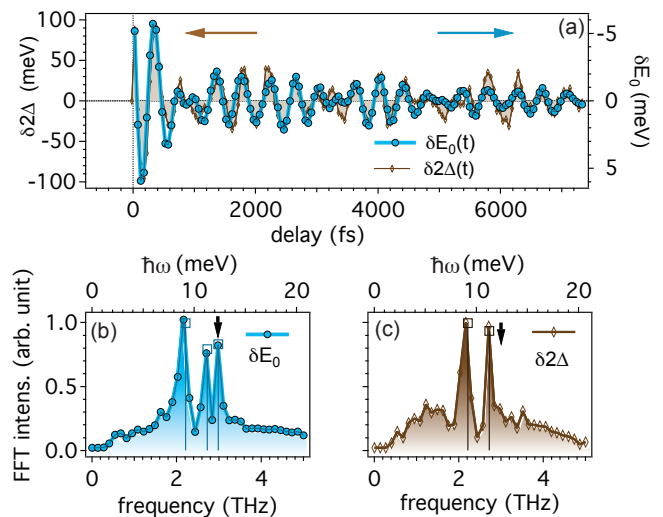


FIG. 3. (a) Residuals of the polynomial background fit shown in Fig. 2(d). (b) and (c) FT of (a). The *vertical lines* and *markers* at $\Omega_1 = 2.19(1)$ THz, $\Omega_2 = 2.69(1)$ THz and $\Omega_3 = 2.98(1)$ THz indicate mode frequencies and amplitudes independently obtained from fitting damped cosine functions³⁵.

frequencies. For a more detailed analysis smooth backgrounds (10^{th} order polynomials) are subtracted and the residuals $\delta 2\Delta(t)$ and $\delta E_0(t)$ are plotted in Fig. 3(a). Analysis via Fourier transformation (FT) of $\delta 2\Delta(t)$ reveals two dominant frequencies at $\Omega_1 = 2.2(1)$ THz and $\Omega_2 = 2.7(1)$ THz, as shown in Fig. 3(c). In contrast, the oscillatory response and particularly the beating pattern of $\delta E_0(t)$ differs from $\delta 2\Delta(t)$. In addition to Ω_1 and Ω_2 , the FT of $\delta E_0(t)$ exhibits a third pronounced peak at $\Omega_3 = 3.0(1)$ THz, marked by a vertical arrow in Fig. 3(b). These findings are robust regardless of the background subtraction³⁵. Fitting damped cosine functions to $\delta 2\Delta(t)$ and $\delta E_0(t)$ ³⁵ yields the same frequencies albeit with higher resolution as indicated in Fig. 3.

The mode at Ω_1 is identified as a previously observed AM of $R\text{Te}_3$. As sketched in Fig. 4(a), coherent in-plane atomic motions modulate the lattice distortion Δz and thus directly affect Δ . This assignment is supported by comparing frequency Ω_1 to the soft mode observed in temperature-dependent transient optical reflectivity^{7,27}, temperature-dependent Raman spectroscopy^{19,25,26}, and temperature- and fluence-dependent trARPES^{4,29,30}.

The mode at Ω_2 is missing at the zone center of the frozen phonon calculations, suggesting it originates either from a normal state optical or acoustic branch which is renormalized by coupling to the charge modulation at \vec{q} . A finite projection of the ionic motion on the lattice displacement Δz associated with the CDW then naturally explains its presence in $\Delta(t)$ and the temperature-dependent softening in^{26,27}. We thus assign Ω_2 as a 2nd AM.

The mode at Ω_3 is assigned to an out-of-plane A_{1g}

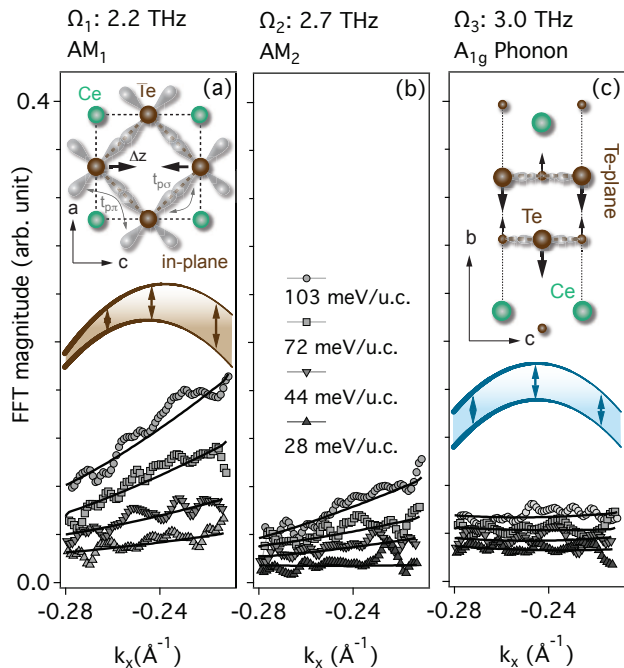


FIG. 4. Mode-dependent modulations of the Te-5p bands (*markers*) by the three dominant modes is derived from momentum-dependent FT amplitudes for deposited energies of 28 – 103 meV per unit cell. *Solid lines* are obtained from the corresponding TB fits of the transient Te-5p bands³⁵. (a) The AM at $\Omega_1 = 2.2$ THz modulates the in-plane lattice displacement Δz and leads to a strongly momentum- and fluence-dependent response. (b) The AM at $\Omega_2 = 2.7(1)$ THz exhibits a weaker momentum- and fluence-dependence, which indicates a weaker influence on the in-plane CDW. (c) The out-of-plane A_{1g} mode at $\Omega_3 = 3$ THz only shifts the band rigidly as it does not couple to the in-plane charge order.

phonon, which is not related to the CDW by comparison to frozen phonon calculations and transient reflectivity²⁷. The calculated displacement of the Te-atoms is sketched in Fig. 4(c). The response of the Te-5p band is remarkably mode-dependent; Ω_3 mainly affects E_0 , and has a much smaller influence on Δ even though Ω_3 involves the motion of the Te atoms, which form the CDW bands. However, the out-of-plane motion couples only weakly to the charge order, with correspondingly small influence on the order parameter Δ .

To further investigate this explanation for mode-dependent coupling we plot FT amplitudes for all three modes in Fig. 4 as function of fluence and momentum. These are derived from an FT analysis of the oscillatory

binding energies, which have been extracted from transient spectra as function of \vec{k} ³⁵. The influence on the dispersion is illustrated in Fig. 4(a) and (c). The AM at Ω_1 is strongly fluence- and momentum-dependent as expected for a reduction of Δ . The AM at Ω_2 exhibits a qualitatively similar behavior albeit of smaller magnitude. This is consistent with a smaller coupling between the phonon with the charge density or a smaller projection of the ionic motion on the lattice distortion Δz .

In contrast, the out-of-plane A_{1g} phonon at Ω_3 causes a strikingly k -independent shift, which is only weakly dependent on fluence. This suggests a small fluence dependent atomic displacement $\mu(t)$ and a constant deformation potential $D_{5p}(\vec{k})$ that explains the rigid energy shift via $E_0(t) \sim D_{5p}(\vec{k})\mu(t)$ ^{38,39}. We confirm the approximation of a k -independent deformation potential by DFT calculations for atomic displacements μ of up to 0.13 Å, shown in Ref.³⁵.

In summary, trARPES measurements on CeTe₃ reveal a CDW gap size of $2\Delta = 0.59$ eV. Coherent excitations of two AMs at $\Omega_1 = 2.2$ THz and at $\Omega_2 = 2.7$ THz and a generic A_{1g} phonon at $\Omega_3 = 3$ THz are identified by fits of a time-dependent model dispersion and comparison to normal state frozen phonon calculations. The out-of-plane A_{1g} phonon leads to small rigid shifts of the entire Te-5p band but does not affect Δ . In contrast, Ω_1 and Ω_2 result in pronounced changes of Δ . We point out that our k -dependent analysis combined with low excitation densities is key to identifying and assigning AMs. We have generalized our approach and introduced a model-free evaluation (Fig 4), which can be applied to materials where a detailed description of the band dispersion may not be available. More specifically, our approach can be transferred to charge-ordering in striped cuprates⁴⁰, which has identical symmetry to $R\text{Te}_3$ ⁴¹.

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