Supplementary information for: Ultrafast Optical Control of the Electronic Properties of $ZrTe_5$

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I. MULTIPLE DOMAINS IN THE SURFACE TERMINATION

As a consequence of the low dimensional nature of the prismatic chains and the weak interlayer binding energy, the cleaved surface of $ZrTe_5$ can result in a multiple domains structure, characterized by slightly different out-of-plane chain orientations. The existence of different domains results in the appereance of band replica in the ARPES images.
A proper attention has been paid for the ARPES measurements in order to avoid such spurious signal. Figure S1 shows a comparison between two ARPES images collected under the same experimental conditions but for two different sample regions. Panel (a) shows a pair of bands, disperding simmetrically with respect the high simmetry Γ point and resulting from a single domain surface termination. Conversely, panel (b) shows a weak clear replica of the bands, which is ascribed to the presence of a second domain with $\sim 4.3°$ misalignment. The angle corresponds to an out-of-plane rotation, along the chain axis. We found the ARPES signal from a single domain termination more probable than the multiple domains signal. This suggests an average domain dimension larger than beam spot size ($\sim\ 150\mu m\ \times\ 150\mu m$) used in the experiment.

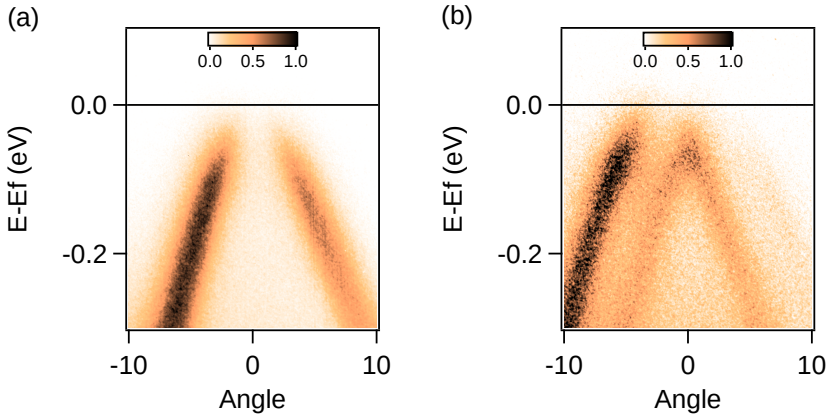

FIG. 1: (Color online) Electronic band structure of $ZrTe_5$ measured with s-polarized light (a) on a single-domain region of the sample along the chain direction ΓX at 300 K, (b) on a multi-domain region of the sample along the chain direction ΓX at 300 K. A replica of the bare band dispersion is clearly evident with an angular displacement equal to $\sim 4.3°$.

II. ANALYSIS OF THE TEMPERATURE SCAN

Here, we detail the procedure used for obtaining the energy shift of the band structure from the temperature scan in the300 K - 125 K range. In order to track the evolution of the band structure, we cut a momentum distribution curve (MDC) at -0.115 eV, integrated over 20 meV, from the ARPES image taken at 300 K as shown by the red line in Figure S2 (a). The resulting spectrum is fitted with a function which is the sum of two Lorentzians and a second order polynomial background. Then the MDCs are extracted for all the other temperatures at increasing binding energies keeping the k position of the peaks fixed, with an energy shift at 125 K equal to $\sim 60meV \pm 10meV$, as shown in Fig. S2 (b).
Figure S2(d) shows the resulting stack of MDCs as a function of the temperature along with the best fit, vertically shifted for

ease the reading. We repeat the same procedure also starting from other energies (-0.07 eV and -0.2 eV at 300 K), thus tracking the peak position at different fixed k value.The results are shown in Fig. S2 (c), with colored circle markers for -0.115 eV, and purple triangle and green square markers for -0.07 and -0.2 eV, respectively. The temperature evolution of the binding energies is the same within the experimental energy resolution.
Besides the precise peak position, the MDCs fitting provides also information about the evolution of the peak width. The full widths at half maximum (FWHM) has been used to evaluated the temperature evolution of the quasiparticle lifetime shown in Fig. 1(f) of the Letter.

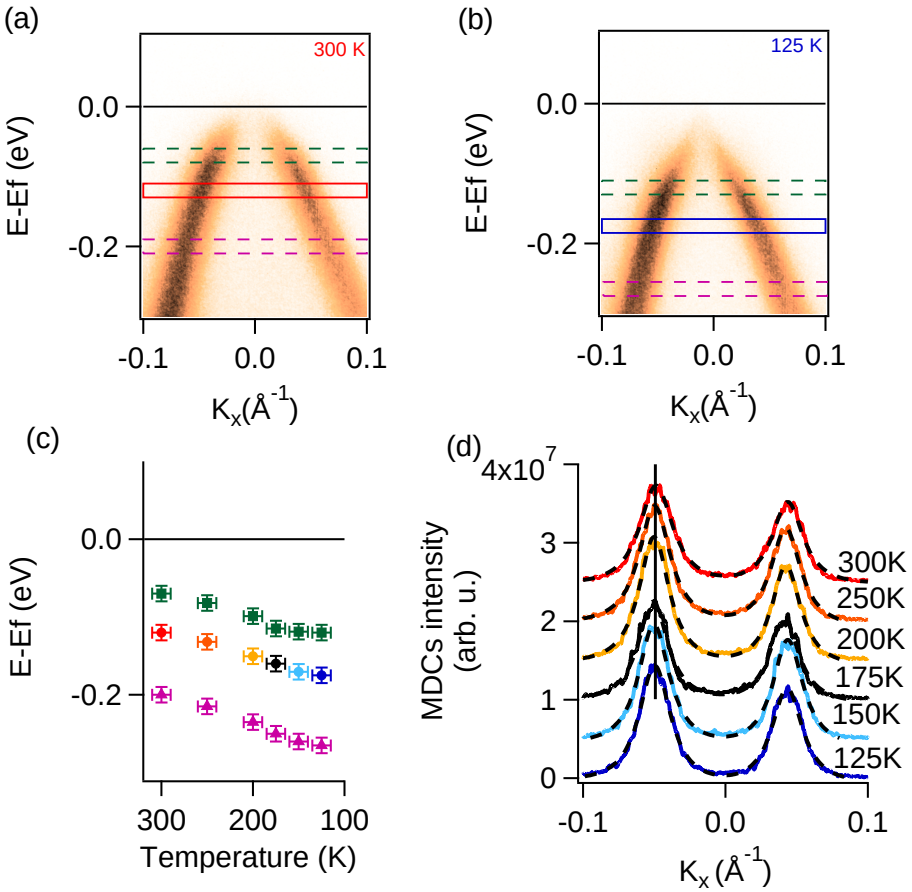

FIG. 2: (Color online) (a) - (b) Electronic band structure of $ZrTe_5$ measured with s-polarized light along the chain direction ΓX at 300 K and 100 K, respectively. (c) Energy of the MDCs cut versus sample temperature (colored), starting energy at 300 K: -0.115 eV; energy of MDCs cut done at different starting energies (-0.07eV, green, and -0.2 eV, purple). (d) MDCs cuts integrated over 20 meV, colored line in (a) and (b), and positively shifted along the vertical axis for the different sample temperatures. The traces are fitted (black dashed lines) with two lorentzian plus a polynomial background.

III. ANALYSIS OF THE TIME RESOLVED SIGNAL

We have analyzed the temporal evolution of the electron population for both valence and conduction bands, below and above the Fermi energy. As shown in Figure 2 (d) of the Letter, each population dynamic is fitted with a single exponential decay $Ae^{-(t-t_0)/\tau}$, where A is the intensity, τ the characteristic decay time and t_0 indicates the time zero, *i.e.* the temporal overlap between pump and probe. The function is convoluted with a Gaussian accounting for the finite experimental temporal resolution, equal to $\sim 250 fs$.

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