Time-resolved Fermi surface mapping of the charge density wave material DyTe$_3$

L. Rettig[1,2], R. Cortés[1,3], J.-H. Chu[4,5], I. R. Fisher[4,5], F. Schmitt[4], P. S. Kirchmann[3,5], R. G. Moore[4], Z.-X. Shen[4,5], M. Wolf[3], and U. Bovensiepen[2,a]

[1] Fachbereich Physik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany
[2] Fakultät für Physik, Universität Duisburg-Essen, Lotharstraße 1, D-47048 Duisburg, Germany
[3] Abteilung Physikalische Chemie, Fritz-Haber-Institut der MPG, Faradayweg 4-6, D-14195 Berlin, Germany
[4] Department of Applied Physics, Via Pueblo Mall, Stanford, CA 94305, USA
[5] Stanford Institute for Materials and Energy Science, 476 Lomita Mall, Stanford, CA 94305, USA

Abstract. The femtosecond dynamics of the Fermi surface of DyTe$_3$ and its band structure are investigated by time- and angle-resolved photoemission spectroscopy. We directly monitor the ultrafast collapse of the charge density wave gap within 200 fs.

1 Introduction

The charge density wave (CDW) formation in the family of rare earth tritellurides RTe$_3$ presents an excellent model system to study the effects of charge and lattice order and broken symmetry in a low-dimensional material. Below the CDW transition temperature, large energy gaps open in the electronic band structure leading to the suppression of large areas of the Fermi surface (FS), as revealed by angle-resolved photoemission spectroscopy (ARPES) [1]. Time-resolved spectroscopies can provide complementary information on the dynamics of the electronic structure after photoexcitation and on the underlying processes like electron-electron or electron-phonon interaction [2]. Here, time-resolved ARPES (trARPES) is employed to provide insight into the ultrafast dynamics of the electronic structure linked to collective excitations of the charge ordered state, as already demonstrated in first experiments observing the ultrafast melting of the CDW gapped state and the excitation of the amplitude mode in TbTe$_3$ [3,4]. In addition, trARPES enables the direct investigation of the occupied and unoccupied band structure close to the Fermi level.

2 Experimental details

The experimental geometry is shown in Figure 1 (a). Samples of DyTe$_3$ mounted on slanted sample holders are cleaved in ultrahigh vacuum prior to the measurements. The electronic system is excited by femtosecond (fs) laser pulses ($h\nu_1 = 1.5$ eV, 55 fs) from a regenerative amplifier operating at 300 kHz. A subsequent time-correlated ultraviolet probe pulse ($h\nu_2 = 6.0$ eV, 80 fs) generates photoelectrons, which are detected by a novel position-sensitive Time-of-Flight spectrometer (pTOF) [5]. Its momentum resolution of < 0.01 Å^{-1} and 2D photoelectron detection scheme for simultaneous detection of both in-plane momentum components k_x and k_z allow to investigate the dynamics of both occupied and unoccupied electronic states of DyTe$_3$ over a contiguous area of the reciprocal space.

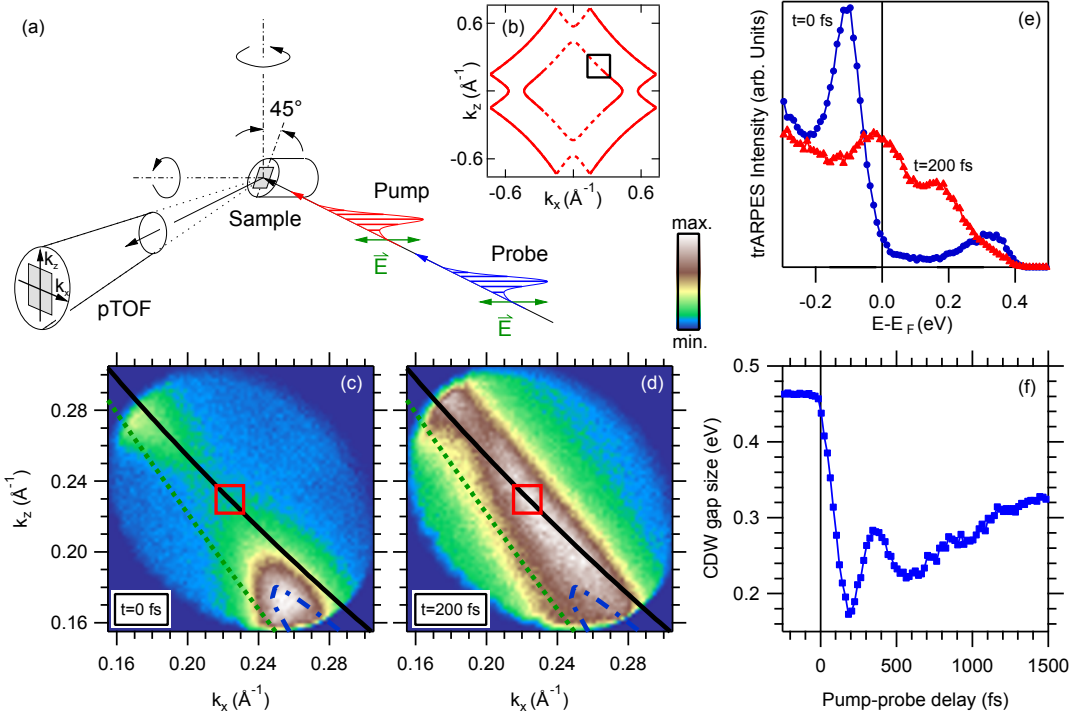[a] e-mail: uwe.bovensiepen@uni-due.de

Fig. 1. (a) Experimental setup using slanted sample holders and the pTOF spectrometer for simultaneous detection of both in-plane momentum components. (b) Schematic FS of $DyTe_3$. The CDW gap opens on the dashed regions. (c) trARPES FS map of $DyTe_3$ within the black box in (b) at $T = 30$ K in a false color representation, obtained by integrating the trARPES intensity at ± 25 meV about the Fermi level, and at zero pump-probe delay with an incident pumping fluence of $F = 1.1$ mJ/cm^2. Lines are bands of a TB model: main TB band (solid line), CDW shadow band (dashed line) and residual FS (dash-dotted line). The box marks the integration area for data shown in (e). (d) At $t = 200$ fs, the CDW gap is closed and a continuous FS is observed. (e) Transient EDCs integrated over the boxes in (c) and (d) at $t = 0$ fs (circles) and $t = 200$ fs (triangles). (f) Transient CDW gap size as function of pump-probe delay.

3 Results

The schematic FS of $DyTe_3$ is shown in Figure 1 (b), with the CDW gap indicated by dashed lines. Panel (c) and (d) show the experimental time-resolved FS within the black box in Figure 1 (b), obtained at $t = 0$ fs and $t = 200$ fs, respectively. The CDW gap closes within 200 fs resulting in an ungapped (metallic) FS, well described by a Tight-Binding (TB) model (lines). The determination of the energetic position of the CDW gap along the normal state FS by analyzing the lower and upper band edge shows a continuous shift of the gap center determined by the nesting condition and confirms the FS nesting driven CDW formation in RTe$_3$ [1,6].

To quantify the ultrafast closing of the CDW gap, we analyze transient energy distribution curves (EDCs) integrated around the center of the gap, as shown in panel (e) for selected pump-probe delays. The transient peak positions representing the occupied and unoccupied CDW bands are determined by Lorentzian line fits and allow to determine the transient CDW gap size, shown in panel (f). We find an ultrafast reduction of the gap size within 200 fs where the gap is reduced from $\Delta_{CDW} \sim 460$ meV at $t = 0$ fs to $\Delta_{CDW} \sim 170$ meV at $t = 200$ fs. For later times, Δ_{CDW} increases again and shows anharmonic, strongly damped oscillations. These oscillations are determined by the atomic rearrangements that govern the collapse of long-range CDW order. Thus, they are a direct fingerprint of the transient potential energy surface, which determines the transition to a transient metallic state of the highly excited system.

In addition, the pTOF spectrometer allows analyzing the momentum dependence of the CDW dynamics along the FS. Details of this analysis are beyond the scope of this short report and in the following we limit the discussion to a qualitative description of our findings. We observed an asymmetric closing of the CDW gap which manifests in a pronounced asymmetry of the relative shift of the upper and lower CDW band. This asymmetry is found to depend on the position on the FS and leads to a transient shift of the gap center along the FS. As the energetic position of the gap center strongly depends on the FS nesting condition [1], this shift might be related to a transient modification of the FS nesting, which could occur due to changes of the transient electronic band structure.

4 Conclusion

We have analyzed the ultrafast evolution of the electronic structure of $DyTe_3$ upon optical excitation by trARPES using a position-sensitive electron spectrometer. The unique capability of this technique allows to monitor both the occupied and unoccupied band structure in great detail and in particular the ultrafast closing of the CDW gap within 200 fs. The observed gap dynamics provides rich information about the transient loss of long-range CDW order in the system. In addition, the asymmetric closing of the CDW gap along the FS suggests a transient modification of the nesting condition.

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References

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