Photo-Induced Phase Transition between Charge Density Wave States in 1T-TaS_2

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1T-TaS_2 is a beautiful example of a 2D metal, which undergoes a series of electronically driven phase transitions. It is a lamellar compound formed by sheets of edge-linked TaS_6 octahedra. Within each sheet, the Ta-atoms form a hexagonal lattice which presents a series of structural modulations as the temperature decreases. The unmodulated structure observed at highest temperatures undergoes a first transition below 543 K with the formation of an incommensurate (I) Charge Density Wave (CDW), which further evolves into a nearly commensurate (NC) CDW below 353 K, and a commensurate (C) CDW below 183 K [1].

We will report on the photo-induced structural dynamics in the NC phase of 1T-TaS_2, on timescales ranging from fs to µs. The experimental technique used is pump-probe time-resolved diffraction, where the sample is excited by an infrared, 50 fs laser pulse and probed by 7 keV, 150 fs X-ray pulses delivered by the synchrotron slicing source of Swiss Light Source at Paul Scherrer Institute [2].

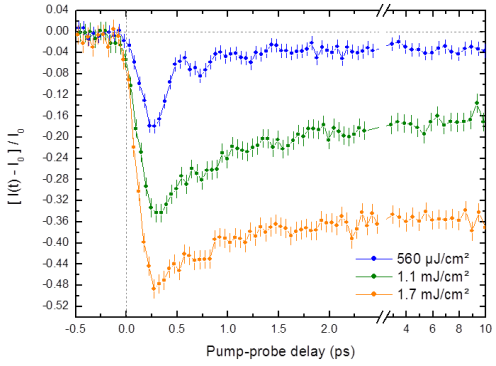

Figure 1

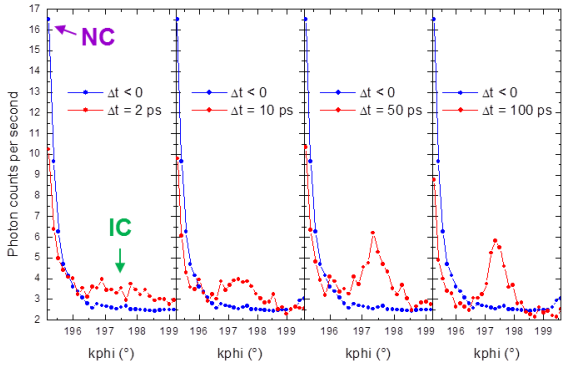

Figure 2

Figure 1 shows the time-evolution of the diffracted intensity at a NC satellite position of reciprocal space, for laser fluences ranging from 0.25 to 1.70 mJ/cm². The NC satellite peaks were chosen because their intensity is directly proportional to the square of the amplitude of the NC modulation. Coherent oscillations associated with the amplitude mode

of the NC-CDW are clearly visible at the lowest laser fluence only (560 µJ/cm²). At all fluences studied, a drop of diffracted intensity is observed, followed by a partial recovery within 3 ps. The higher the fluence, the less intensity recovers at the NC satellite peak position. The average NC modulation amplitude is thus reduced, for at least 250 ps after excitation (not shown).

Figure 2 shows diffracted intensity profiles on the line that joins the NC and I satellite peak positions, for various pump-probe delays in the [0 - 100 ps] range and at a fluence of 2.2 mJ/cm². The region shown is centered on the I satellite peak position, and only the base of the NC satellite peak is visible. One can observe that the reduction of intensity on the NC satellite peak is accompanied by an increase of diffuse scattering, which progressively condenses into a narrow satellite peak characteristic of a photo-induced I phase. Our measurements thus provide a unique view on the dynamics of a 1st order phase transition on its characteristic timescale. We will expose a tentative scenario of nucleation-growth processes for the presently studied NC → I photo-induced phase transition in 1T-TaS_2.

[1] T. Ishiguro and H.Sato, Phys. Rev. B **44** 2046 (1991)
[2] P. Beaud *et al.*, Phys. Rev. Lett. **99** 174801 (2007)