Photoinduced ultrafast charge-order melting: charge-order inversion and non-thermal effects

Michel van Veenendaal*[1]

[1]United States

Using a pump-probe set-up, the melting of charge order has been observed in a wide variety of compounds.[1] Often, the X-ray intensity shows oscillations, which are taken as an indication of coherent behavior. Here, we demonstrate the melting of charge-order in a model system with Ising-like bond charges and classical oscillators representing the ligands. The number of sites is over 100,000 allowing the system to reach a thermodynamic equilibrium. The photoexcitation directly affects the electronic order, but not the local Jahn-Teller like distortions of the ligands. The charge-order melts on the order of a few hundred femtoseconds. The system is coupled to a bath which is kept at a fixed temperature. The recovery times depend strongly on the initial excitation and the presence of long-range restoring forces in the material. The possibility of an inversion of the charge order is demonstrated. Additionally, it is shown that there are substantial differences between photoexcitation and simple heating.

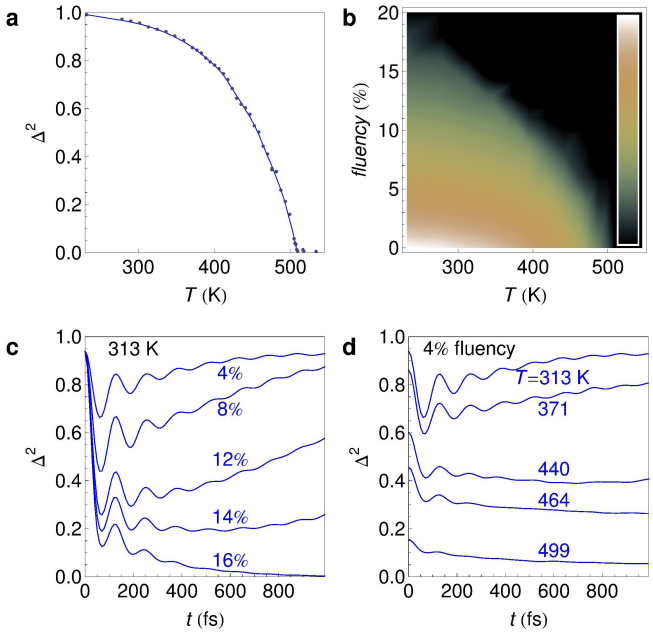

Figure 1: (a) The distortion order parameter Δ^2 as a function of temperature. (b) Density plot of the minimum value of Δ^2 as a function of initial temperature and fluence of the pump. The fluence is given as the percentage of flipped bond charges. The inset gives the change in color on a linear scale from 0 to 1. (c) Dependence of Δ^2 as a function of time Δ^2 after the initial pump. The different curves give the dependence on the fluence of the pump for the same initial state at 313 K. (d) Time dependence of Δ^2 for different initial temperatures, but at a fixed fluence.

References

[1] see for example, P. Beaud *et al.*, Nat. Mat. **13**, 923 (2014).

*Corresponding author: veenendaal@niu.edu