ICFO



PhD THESIS DEFENSE: Inhomogeneity and Disorder in Ultrafast Phase Transitions

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10:00 to 13:00
Online (Teams) and ICFO Auditorium

In the recent decades, there has been a surge of interest in the wide array of emergent phenomena found in strongly correlated materials. Understanding the inner workings of this type of systems is a major challenge due to the complex way in which multiple degrees of freedom, such as electronic, structural and spin, interact with each other and themselves in non-trivial fashion. One of the most striking outcomes of these subtle interactions in correlated materials is the richness of their phase diagrams, which include exotic states that still elude a complete physical description. It is in the transition from one phase to another where the insights into the complex microscopic mechanisms may be most readily found, and so the study of phase transitions has become a staple in correlated materials science.

The experimental techniques used to track phase transitions are steadily becoming more precise and, with the improvements, previously overlooked aspects of a phase transition become more apparent, such as inhomogeneity or disorder, which add another layer of complexity that may clash with our current understanding. This is particularly important in the relatively young field of ultrafast studies of correlated materials, which tackles these systems in a largely uncharted territory: non-equilibrium situations. In this thesis, we develop novel experimental techniques which push towards an assessment of disorder and/or inhomogeneity in non-equilibrium phase transitions, while still being able to accurately track the dynamics of the degrees of freedom involved. We then apply these techniques in two systems of current interest: La0.5Sr1.5MnO4, a prototypical layered manganite, and VO2, one of the most emblematic correlated materials.

For La0.5Sr1.5MnO4, we introduce an all-optical tabletop pump-probe setup that is able to track the ultrafast melting of charge- and orbital-order parameter with high accuracy. We show how, in contrast with previous descriptions, the transition is incoherent and fits with the paradigm of an order-disorder process. A key factor in these dynamics, which is sometimes overlooked, is spatial phase separation into the depth of the material. With our setup, the role of initial inhomogeneity and its evolution can be readily tested.

For VO2, we employ facility-scale X-ray sources to directly image phase inhomogeneity in the metal-to-insulator transition with coherent X-ray diffraction techniques. We show quantitative imaging of phase separation and domain growth statically, which in our experimental setups should be able to distinguish intermediate phases appart from the usual monoclinic insulator and rutile metal. We find no evidence of previously claimed intermediate phases such as monoclinic metal VO2. Finally, we show the first non-scanning spatially-resolved observation of the ultrafast phase transition in VO2 with nanometer resolution, where we identify a global, prompt change in the domain pattern in the femtosecond scale.

Hosted by: Simon Wall