

## Tracking quantum phase transitions with continuously variable temperature cryo-STEM

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Recent advances in cryogenic scanning transmission electron microscopy (cryo-STEM) have enabled low temperature quantum phases to be probed at the atomic scale providing key insights into their microscopic nature. For instance, at liquid nitrogen temperature periodic lattice distortions (PLDs) can be spatially mapped with pm-precision in charge-ordered manganites revealing defects and nanometer-scale variations in the charge-ordering [1,2]. In more complex low temperature structures where lattice distortions are not coherently ordered along the beam direction, imaging alone may not be sufficient to extract the local order. In such cases, coupling cryo-STEM imaging with density-functional theory calculations and multislice image simulations may help to reveal the low-temperature state as we have shown for monoclinic 1T'-TaTe<sub>2</sub> [3].

While cryo-STEM is now a practical real-space probe for mapping lattice order (at least at liquid nitrogen temperature), understanding the emergence or melting of electronic phases necessitates atomic-scale imaging at a broad range of temperatures and over multiple temperature cycles. In continuously variable temperature (CVT) cryo-STEM, we have addressed this challenge by coupling a side-entry, dual-tilt cryo-holder with fast, local heating via a 6-pin microelectromechanical device [4]. This combination enables intermediate temperature studies and the ability to rapidly change temperature within a single experiment while maintaining sample stability for atomic resolution imaging. Here, we demonstrate the impact of such experiments for quantum materials using two examples.

The first example focuses on magnetic transitions and their layer-dependence in 2D materials, currently a major topic of interest in the quantum community. Nb<sub>3</sub>Br<sub>8</sub> is a 2D material exhibiting a paramagnetic to non-magnetic transition upon cooling [5]. Loss of magnetism at low temperature has been coupled to a structural transition with re-stacking of the van der Waals layers from a 2-layer ( $\alpha$ -phase) to a 6-layer ( $\beta$ -phase) unit cell—the structures of which are easily distinguished in plan view. CVT cryo-STEM allows atomic resolution imaging of the structure of exfoliated Nb<sub>3</sub>Br<sub>8</sub> while cycling the temperature through the full structural transition ( $\sim 723$  K to  $\sim 100$  K to  $\sim 723$  K). Our cryo-STEM temperature cycle experiments provide several significant insights into the nature of this structural phase transition, including thickness-dependent hysteresis and the presence of intermediate stacking order.

In the second example, we return to the manganite Bi<sub>1.35</sub>Sr<sub>1.17</sub>Ca<sub>0.48</sub>MnO<sub>3</sub> (BSCMO), a model charge ordered manganite system with a  $T_c$  near room temperature. Our previous work was limited to characterizing the endpoints of the commensurate-incommensurate charge order phase transition. Using

CVT cryo-STEM, we track the material's phase transition directly by measuring the PLDs over the same region of interest at intermediate temperatures between liquid nitrogen and room temperature. These measurements reveal motion of topological defects in the PLD upon heating as well as local restructuring of the distortion around these defects.

Our work demonstrates that cryo-STEM enables quantification of lattice symmetries, inhomogeneities and defects of low temperature quantum states. Nevertheless, key challenges remain. Probing atomic-scale variations in electronic degrees of freedom at low temperature using spectroscopic techniques such as electron energy loss spectroscopy (EELS), for example, is more difficult. The total signal in these measurements is typically low and the dwell times required are long (ms) compared to what is typically used in imaging ( $\mu$ s). Advances in hardware are therefore required to overcome limitations especially at cryogenic temperatures where the sample stability is reduced at least in presently available cryogenic sample holders.

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## References

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