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Atomic Resolution CryoSTEM Across Continuously Variable Temperatures

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Cryogenic STEM has developed in recent years as a powerful tool to explore novel quantum materials phenomena, including low temperature spin states [1], emergent charge density phases [2], and interface charge transfer [3]. While these advances have begun to probe low-temperature materials physics, experiments reported thus far have been limited to single temperatures set by the choice of cryogen, i.e., liquid nitrogen or helium. However, exploration of many novel quantum materials will require experiments across continuously variable temperatures in order to track phase transitions and to tune into phases with narrow temperature windows of stability.

Zandbergen, et al. have developed a novel side-entry variable-temperature liquid nitrogen cryo-holder designed specifically to address this opportunity. Previous measurements with a single-tilt version showed single atom imaging at liquid nitrogen temperature [4], however, dual-tilt capabilities are critical for crystalline quantum materials. Here, we assess the performance of the dual-tilt variable-temperature cryo-holder in a Titan Themis 300. Compared to existing cryo-holders which allow for coarse temperature control via resistive heating of the holder tip, in the HennyZ holder the heating is accomplished locally at the sample using a MEMS chip. This permits continuously variable temperature control between ~100-800 K. More importantly, because the temperature varies only at the sample rather than throughout the rod, rapid temperature changes of tens of Kelvin can be implemented without substantial stability degradation and requiring only a few minutes of settle time after each temperature change. Drift rates remain consistently near ~1 Å/sec independent of samples temperature, as shown in Figure 1. By maintaining a constant liquid nitrogen level throughout the experiment consistent drift rates of <1 nm/min can be obtained, as demonstrated by extensive tests performed on a Philips CM200. This advance in temperature control promises new opportunities for high resolution *in situ* STEM, allowing dynamic processes to be tracked truly through phase transitions rather than simply on either side.

In order to track such processes at the atomic scale, however, there can be no concession to imaging resolution at intermediate temperatures. Figure 2 shows single HAADF STEM scans, recorded at 2 μs/px and 2048x2048 px^2 (~8 sec/frame), demonstrating consistent resolution <1 Å at previously inaccessible intermediate temperatures of ~123, 173, and 223 K. Note that the calibration for accurate temperature reading on the MEMS chip has not yet been fully optimized. Thus, while the trends shown here are correct the absolute temperature values given may be slightly low. Ongoing efforts to further the capabilities of high resolution cryoSTEM will expand to regions of quantum materials' phase diagrams not only across variable temperatures but also to couple cryogenic temperatures with external stimuli, e.g., light [5] or external fields. The MEMS contacts in the HennyZ holder will allow for cryogenic electrical biasing experiments, an exciting prospect for many multiferroic and correlated electron systems [6].

[1] Klie, et al. PRL **99** (2007), p. 702-704.
[2] El Baggari, et al. PNAS **115** (2018), p. 1445-1450.

[3] Zhao, et al. Sci. Adv. **4** (2018), eaao2682.
[4] Hotz, et al. Microsc. Microanal. **24** (2018), p. 1132-1133.
[5] Kociak, Gloter, Stéphan. Ultramicroscopy **180** (2017), p. 81-92.
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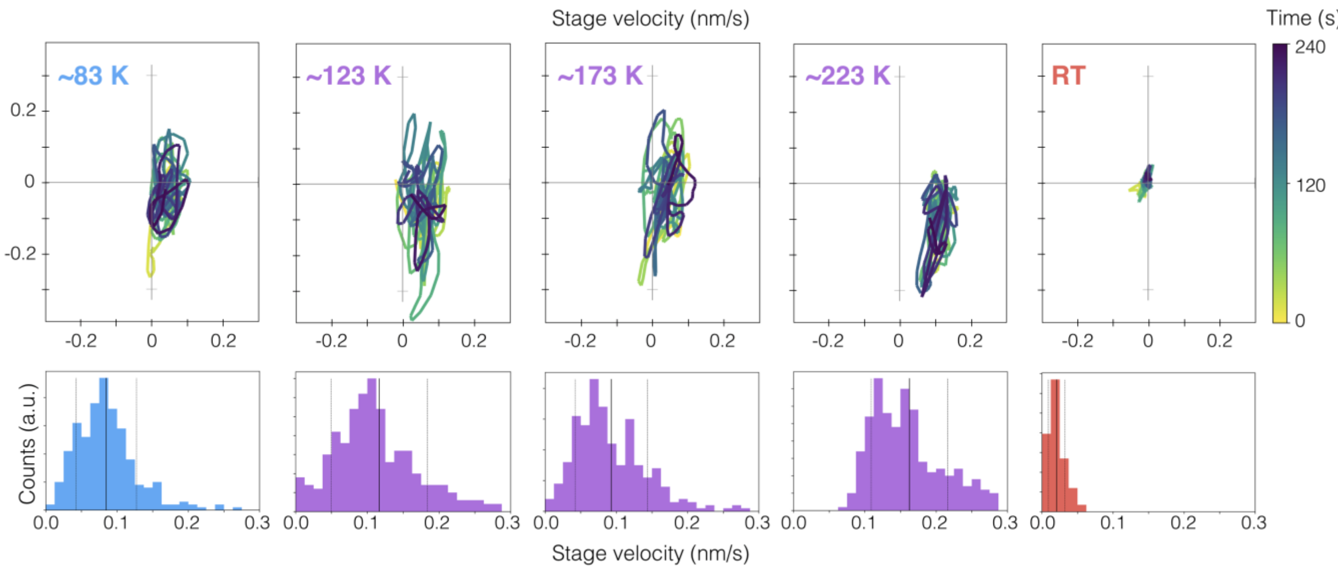

Figure 1. Stable imaging across a broad temperature range is achieved using the dual-tilt MEMS-heated variable temperature cryo-holder. Instantaneous stage drift velocities at ~ 83 K, 123 K, 173 K, 223 K, and room temperature tracked over 4 min from registered image stacks. Note that the actual temperatures at the sample may be slightly higher than the values quoted here due to inaccurate calibration prior to the experiment. Corresponding histograms show instantaneous velocity values with the mean (solid line) and standard deviations (dashed line) marked. Holder drift velocities can be further reduced by extending the setline time and/or by controlling the nitrogen level.

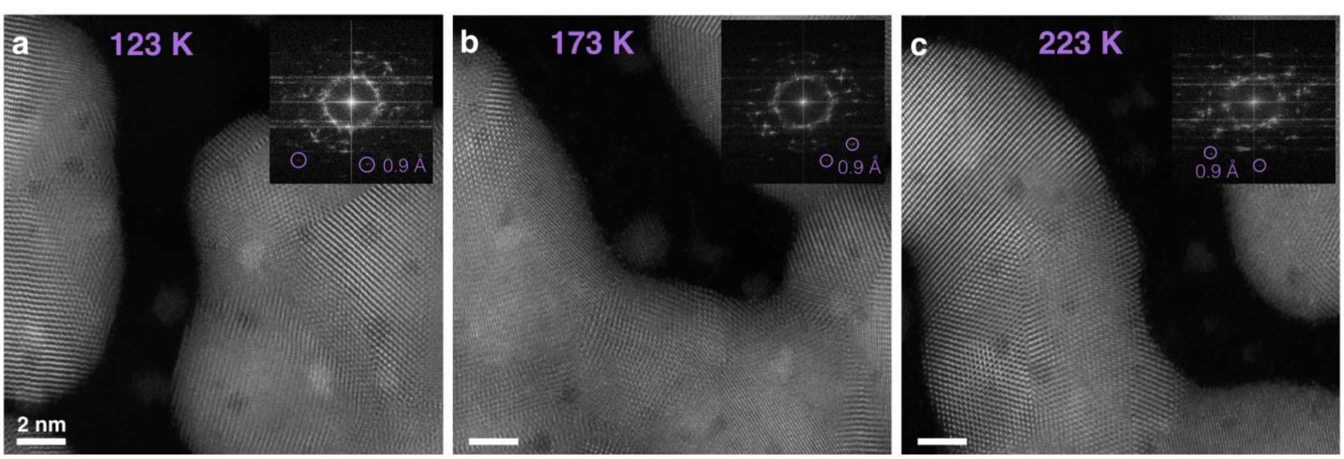

Figure 2. Atomic resolution single scan HAADF STEM images of gold particles on carbon support imaged at intermediate sample temperatures ~ (a) 123 K, (b) 173 K, (c) 223 K. Resolution is estimated from the respective FFT and listed on each; scale bars 2 nm. Further resolution improvement can be achieved with rigid registration of several rapid scan frames.

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