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Diffraction-Ring Contraction as a Method of *In Situ* Thermometry in TEM

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In situ and *modus operando* transmission electron microscopy (TEM) enable visualization of atomic-scale phenomena occurring under conditions that mimic device operation and chemical reaction environments. These techniques have extended the capabilities of static TEM to new systems and dynamic processes previously experimentally inaccessible due to technical limitations. *In situ* heating experiments, made possible by specially-designed specimen holders, are being driven by interest in understanding heat transport, phase transitions, and thermal properties on the nanoscale [1-5]. For most heating holders, the specimen temperature is indirectly measured via incorporation of a thermocouple into the device. As such, the true steady-state temperature of the specimen in relation to the thermocouple or thermistor reading can be a potential source of error. Typically, *in situ* specimen thermometry methods involve calibrating the current applied to a resistively-heated holder to the melting point of a well-characterized material. As it is a binary measurement at the melting point, however, application over the entire temperature range accessible with the holder can be challenging. Thus, ideal methods for *in situ* lattice thermometry would enable continuous use over a wide temperature range and would directly reflect the true specimen temperature.

Here, we present a method for directly and continuously measuring specimen temperature in TEM via coefficient of thermal expansion (CTE) and sub-pixel analysis of parallel beam electron diffraction (PBED) patterns. To accurately measure lattice expansion, and thus temperature, from PBED patterns, one must determine the center position of the direct beam. This can be done using a circular Hough transform (CHT) allowing for sub-pixel determination of diffraction ring radii [6]. Here, we used polycrystalline aluminum films as a test of the method due to its large CTE and thus significant diffraction ring contraction. By comparing measured ring contraction at specific heating holder settings to that expected from the CTE of aluminum, an accurate calibration of the thermocouple reading with respect to lattice temperature from 20 to 340 °C was obtained. This indicates that, for the specific heating holder used, the thermocouple reading is an accurate indicator of true specimen temperature. In addition, the {200} and {111} planes follow the same trend with respect to the CTE, indicating the aluminum film is expanding isotropically over the temperature range measured.

The Gatan Model 652 Mark II Double Tilt Heating Holder used in the experiments described here has a resistively-heated crucible for changing specimen temperature. Due to the Debye-Waller effect, one would expect the intensity of Bragg spots or diffraction rings to decrease with increasing heating holder temperature. No such effect was observed over the experimental temperature range because measured intensity changes of the Bragg spots are a convolution of the Debye-Waller effect and a change in excitation error due to uncontrolled specimen holder tilting. Imprecision in the mechanical control of the β-tilt axis leads to uncontrolled tilting during crucible heating. By following the center-of-mass of a single-crystal silicon diffraction pattern over the course of heating, the real-space tilting of the specimen at each temperature can be estimated [7].

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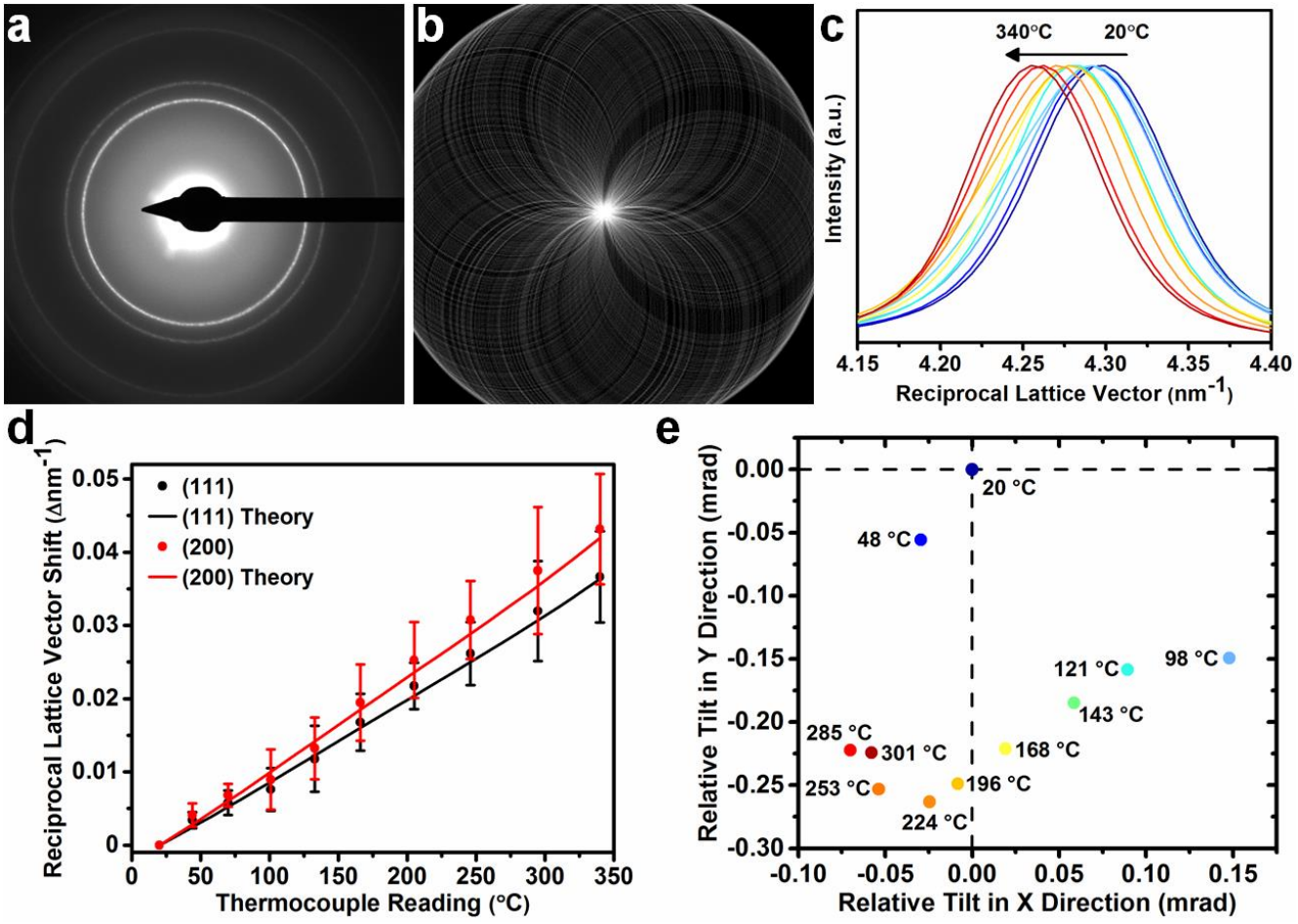

Figure 1. (a) Parallel-beam diffraction pattern of as-deposited aluminum film at 1.5 m camera length. (b) Hough transform of the (111) diffraction ring at a Hough radius, $r_{Hough} = r_{ring}$. (c) Full radial integration line scans of (111) peaks for ten temperature points of one trial showing the reciprocal lattice-vector contraction due to heating. (d) Comparison of fitted experimental diffraction ring shifts to theoretical shifts based on the CTE for aluminum. (e) Estimated uncontrolled tilting at each thermocouple reading for a single-crystal silicon specimen via center-of-mass determination.