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Quantification of Thermal Interface Resistance Using Atomic Scale Debye-Waller Thermometry

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With the continuing miniaturization of electronic and functional devices, nanoscale temperature mapping capabilities are in demand for better thermal design and engineering of the devices. Thermal engineering must consider thermal interface resistance (TIR) since it can significantly influence the thermal conduction and dictate the performance and stability of devices. Quantification of TIR, however, requires very high spatial resolution and precision. Common thermometry techniques, such as Raman spectroscopy, time-domain thermo-reflectance and scanning thermal microscopy, are limited to ~100 nanometers in resolution. Non-contact temperature mapping has also been explored in (scanning) transmission electron microscopy [(S)TEM], including binary thermometry utilizing phase change, thermal strain mapping by electron energy loss spectroscopy and diffraction analysis achieving ~10 to 100 nanometer spatial resolution *e.g.* [1]. However, the spatial resolutions of these methods are still insufficient to probe TIR, as the temperature drop at the interface due to TIR is expected to be confined within the regions less than a few nanometer from the interface.

We present an atomic scale Debye-Waller thermometry based on quantitative analysis of STEM signals to directly detect and quantify TIR at oxide interfaces. STEM signals have been long known to be sensitive to the temperature of the materials being probed, which is commonly parametrized using Debye-Waller factor. With aberration correction, the electron probe can get to sub-angstrom level, allowing one to quantify the attenuation/enhancement of column intensity as a function of temperature with atomic resolution (Fig. 1a). By comparing with simulation, temperatures at each atomic column of a crystalline structure can be mapped. This approach should provide spatial resolution sufficiently high enough to resolve the temperature drop at the interface, which can then be used to calculate TIR.

Electrons scattered to all angles convey temperature information, but the sensitivity varies. Electron microscopy pixel array detector (EMPAD) [2] with high dynamic range (32 bit) and fast read-out speed enables us to plot the temperature sensitivity as a function of scattering angle, which help refine the simulation parameters and serves as a guideline for choosing collection angles for image formation. For example, the simulation shows that the angle range between 20 and 40 mrad should increase the scattering intensity from $SrTiO_3$ (Fig. 1b). This prediction is experimentally demonstrated in Fig. 1c and 1d. The images of two unit cells collected under different temperatures clearly shows the enhancement of column intensity at higher temperature with the collection angle range of 20 to 40 mrad. Meanwhile the collection angle range above ~ 70 mrad decreases the intensity, as shown in the simulation in Fig. 2a and the experimental data in Fig. 2b. We also note that the thermal signal can be affected by the amorphous layer on the surface, so we remove the amorphous layer using low energy ion mill (Fischione Nanomill) at 900 and 500 eV for at least 5 minutes each.

High voltage electron beam carries significant amount of energy that can be transferred to the material and induce extra phonon vibrations [3]. Such vibration will add background signals to temperature measurement and complicates the analysis. In order to minimize the extra vibrations, we apply low dose of electron beam for short dwell time albeit sacrificing the signal-to-noise ratio (SNR). The low SNR can

be compensated by application of non-rigid registration of fast-scanned images [4]. The method effectively increases the probe dwell time, which substantially improve SNR and the precision in scattering intensity. The uncertainty in the experimental data decreases to about 0.1% after registration using ~ 50 images (Fig. 2c). Based on our calculation, 0.1% uncertainty limits temperature resolution to ~ 2 K, which is sufficient for TIR measurement.

Quantification of TIR also requires the temperature gradient across the interface, which we realize using the in situ setup based on the DENS Wildfire heating holder (Fig. 2d). A separate calibration of the gradient was also performed using silver nanoparticles [5] (Fig. 2e), which confirms the gradient across the sample. This work is supported by NSF CAREER Program, DMR-1847964.

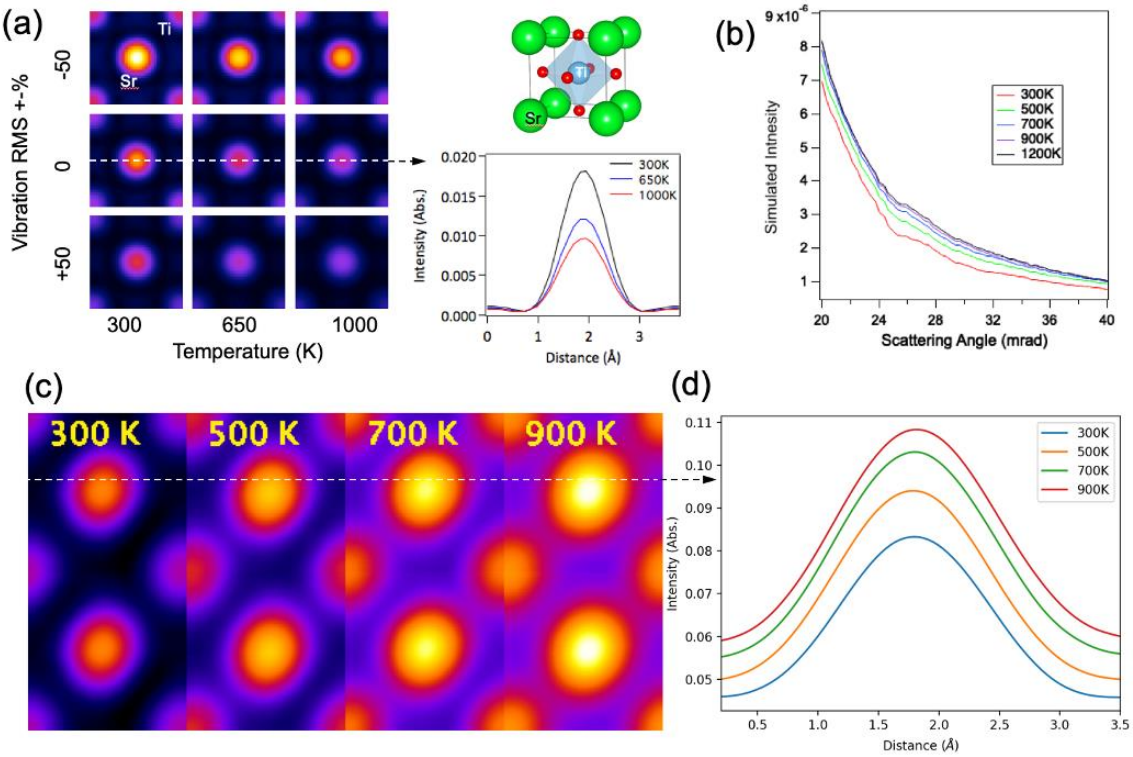

Figure 1. (a) Simulated HAADF images of a SrTiO3 unit cell, as a function of temperature. (b) Simulated annular averaged scattering intensity at the range between 20 and 40 mrad, showing increase in intensity as a function of temperature. (c) Experimental STEM image of SrTiO3 collected with the angle range of 20 to 40 mrad, and (d) the line profile across Sr columns.

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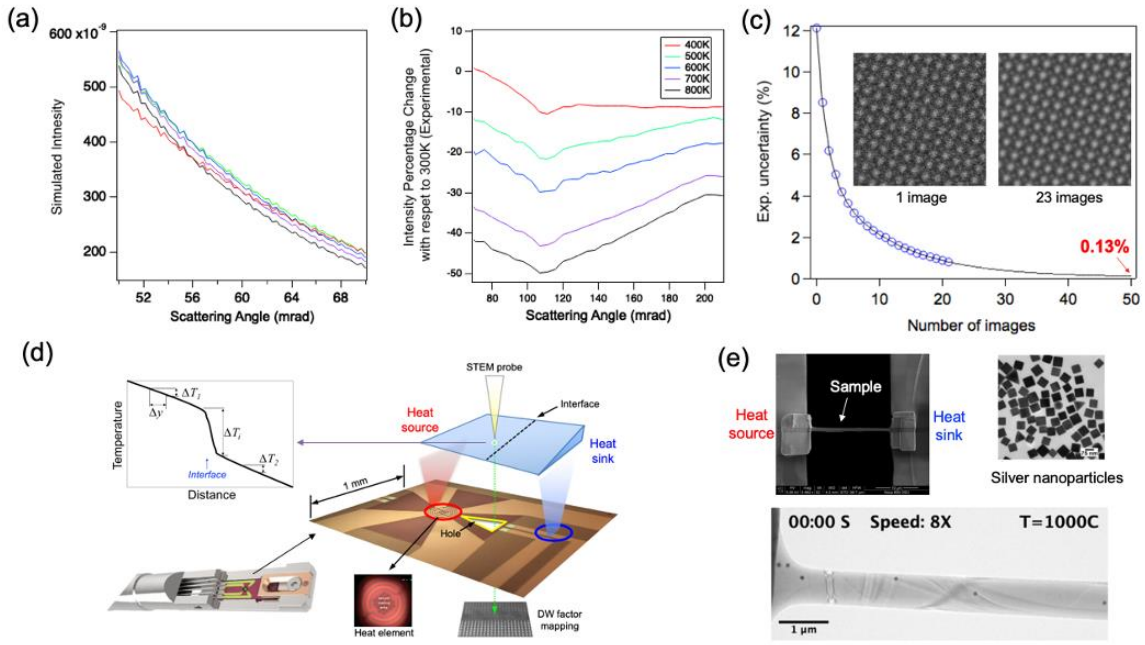

Figure 2. (a) Simulated intensity of annular averaged scattering signal between 50 and 70 mrad. The temperature dependence of intensity changes its sign (negative to positive) within this range. (b) Experimental intensity percentage change as a function of scattering angle. (d) In situ setup for the thermal gradient across the TEM sample. (e) Temperature calibration using silver nanoparticles.

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