

# SCGSR final report: additional information

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## 1 Non-equilibrium temperature

We measured diffuse scattering from the same sample in-situ at multiple currents at 100°C. Specifically, we measured at 40 mA, 300 mA, 500 mA, and then with no current after flashing. The goal is to investigate the role of “flash” on the structure and dynamics (i.e. temperature). To do this, we compare the normalized difference in intensities. Define  $I_1 \equiv I_1(Q)$  as the intensity of some dataset and  $I_2 \equiv I_2(Q)$  as the intensity in some other dataset. In general, the intensity depends on wave vector  $\mathbf{Q}$ . For simplicity, we plot as a function of length  $|\mathbf{Q}| \equiv Q$ . The normalized difference in intensities is  $(I_1 - I_2)/(I_1 + I_2)$ . The reason to normalize is, if total intensity is small (i.e.  $I_1 + I_2$  is small), the difference  $I_1 - I_2$  will also be small even if it is a large fraction of the total intensity. Normalizing puts even low intensity peaks on the same footing as high intensity ones.

We plot the normalized difference in intensities in fig. 1.

We separate the physics into temperature effects and structural effects and argue that we can ignore the structural effects. Then, we fit the difference in intensities by assuming the only dependence on current is via temperature.

### 1.1 Methods

The Bragg scattering intensity measured by neutrons is

$$I(\mathbf{G}, T) \sim A \left| \sum_a b_a \exp(i\mathbf{G} \cdot \boldsymbol{\tau}_a) \exp(W_a(\mathbf{G}, T)) \right|^2 \quad (1)$$

with  $\mathbf{G}$  a Bragg peak,  $A$  a constant prefactor that depends on sample-size, incident-flux, etc. but not on  $\mathbf{G}$  or  $T$ ,  $a$  labels basis atoms and  $\boldsymbol{\tau}_a$  label basis positions.  $T$  is the sample temperature. We want to study the normalized difference in intensities between different measurements  $[I_1(\mathbf{G}, T_1) - I_2(\mathbf{G}, T_2)]/[I_1(\mathbf{G}, T_1) + I_2(\mathbf{G}, T_2)]$  where  $I(\mathbf{G}, T)$  is the intensity measured at Bragg peak  $\mathbf{G}$  for the same material but under different conditions; we assume

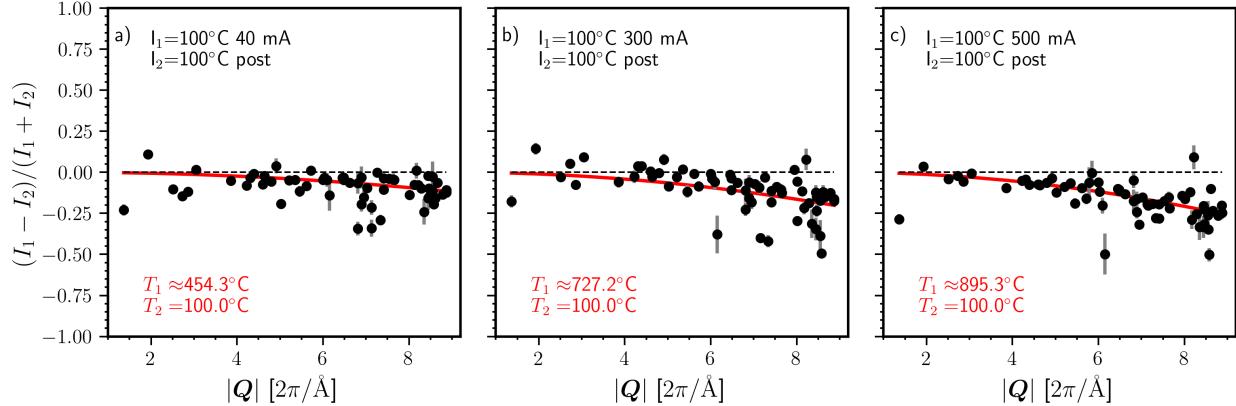


Fig. 1: Normalized difference in intensities. The datasets are labeled in the panels. The difference should be 0 for scattering from identical materials. Deviations from 0 signal structural and temperature differences. Since the flashed samples are approximately isostructural, we assume the leading deviation is due to temperature and estimate the in-situ temperature by fitting. The post-flash sample is ex-situ, so we fix  $T_2 = 100^\circ\text{C}$  (the furnace temperature). The estimated temperatures of the in-situ samples are shown in the panels.

the change in experimental conditions is captured by the *sample* temperature  $T$  (which is not necessarily equal to the furnace temperature).

As a simple approximation, we assume an effective Bravais unit-cell (single-atom basis) such that  $I(\mathbf{Q}, T) \approx AF(\mathbf{G}) \exp(2W(\mathbf{G}, T))$  with  $F(\mathbf{G})$  the form-factor. In a true Bravais lattice,  $F(\mathbf{G}) = b^2$  is independent of  $\mathbf{G}$ . However, to include the non-Bravaisness, we let  $F(\mathbf{G}) \sim |\sum_a b_a \exp(i\mathbf{G} \cdot \boldsymbol{\tau}_a)|^2$  depend on  $\mathbf{G}$  via the basis positions. Moreover, we assume that  $F(\mathbf{G})$  is independent of experimental conditions. This is a reasonable approximation for identical materials and even under different experimental conditions unless we cross a structural phase transition such that the unitcell changes. In our case, we are only interested in comparing different currents through a structure that we assume is already saturated with defects. Then the leading temperature dependence comes from the Debye-Waller factor  $\exp(W(\mathbf{G}, T))$ . Then (dropping explicit  $\mathbf{G}$  and  $T$  dependence for now)

$$\Delta I \equiv \frac{I_1 - I_2}{I_1 + I_2} = \frac{A_1 \exp(2W_1) - A_2 \exp(2W_2)}{A_1 \exp(2W_1) + A_2 \exp(2W_2)} \approx \frac{1 - \exp(2\Delta W)}{1 + \exp(2\Delta W)} \quad (2)$$

with  $\Delta W = W_2 - W_1$ . Since we only consider data from identical samples, we assumed the constant prefactor  $A$  is the same for all datasets. In general

$$W \equiv W_a(\mathbf{Q}, T) = -\frac{1}{2} \langle (\mathbf{Q} \cdot \hat{\mathbf{u}}_a)^2 \rangle \quad (3)$$

depends on the details of the phonon dispersions, displacements, and temperature. However, in line with our effective Bravais lattice approximation above, we approximate the unitcell as independent isotropic harmonic oscillator (i.e. the crystal is an Einstein solid). Then

$\langle (\mathbf{Q} \cdot \hat{\mathbf{u}}_a)^2 \rangle \approx Q^2 \langle u^2 \rangle$  with  $\langle u^2 \rangle$  the 1d mean-squared displacement:

$$W \approx -\frac{Q^2}{2} \frac{\hbar}{2m\omega} \left[ 2n_{BE} \left( \frac{\hbar\omega}{k_B T} \right) + 1 \right]. \quad (4)$$

The quantity  $[\hbar/2m\omega] = L^2$  has dimensions of length squared; we call  $\sqrt{\hbar/2m\omega} \equiv l_0$  an effective length scale. The amplitude of displacement is given by the thermal occupation  $\sim n_{BE}$ . We assume that  $\omega$  is independent of experimental conditions and all that changes is  $T$ . Define  $\Delta n = n_{BE}(\hbar\omega/k_B T_2) - n_{BE}(\hbar\omega/k_B T_1)$ . Then  $\Delta W = -Q^2 l_0^2 \Delta n / 3$  and

$$\Delta I \approx \frac{1 - \exp(-Q^2 l_0^2 \Delta n)}{1 + \exp(-Q^2 l_0^2 \Delta n)} \quad (5)$$

If we measure  $\Delta I$  and can estimate  $\omega$  and  $m$ , we can fit  $T_1$  and  $T_2$ . Moreover, if one of our datasets is at known temperature, we only have one temperature to fit.

In fig. 1 we use eq. 5 to estimate the temperature of the in-situ flashing datasets by comparing to the post-flash dataset. The samples have identical volume and are measured with the same flux etc. so volume and normalization factors are irrelevant.

To estimate the effective energy  $\hbar\omega$  and length scale  $l_0$ , we fit the ratios between an ex-situ sample at 20°C and the post-flash dataset 100°C since the temperatures are known and we can isolate the dependence on  $m$  and  $\omega$ . The results lead to  $\hbar\omega \approx 9.87$  meV and  $l_0 = 0.053$  Å, which are sensible:  $\sim 10$  meV is a typical phonon energy and  $l_0$  is similar to the displacement parameter calculated by refinement in JANA2020. Note, we made some drastic approximations to get to eq. 5 above. Still, the error in these approximations is somewhat compensated by fitting the effective  $\omega$  and  $m$  to the experimental data.

## 1.2 Temperature effects

The difference intensities,  $\Delta I$ , between different datasets in figs. 1 tell us about structural differences and, in the case of in-situ measurements, temperature differences between the samples. For identical structures at the same temperature, the ratios should be constant with the constant related to the ratio of sample volumes. For different structures, the Bragg intensities are different due to different form-factors. For identical structures at different temperatures, the Debye-Waller factor suppresses the intensity; if  $I_1$  is from a hotter sample,  $\Delta I < 1$ .

Comparison between different flashed/flashing samples [figs. 1] shows a weaker structural effect, but pronounced temperature effects. The temperature effect is due to Joule heating: the post-flash sample “100°C post” was flashed in-situ and then held at 100°C ex-situ. We estimate the temperature of the flashing samples by fitting the peak ratios ignoring structural effects; the temperature estimates are shown in the figure.

There is a clear trend of heating and, in the case of the 500 mA dataset, the sample is at nearly 900°C! We note that these estimates are likely not quantitatively accurate but serve as a solid proof of concept that we can measure the temperature difference by carefully analyzing neutron scattering data.

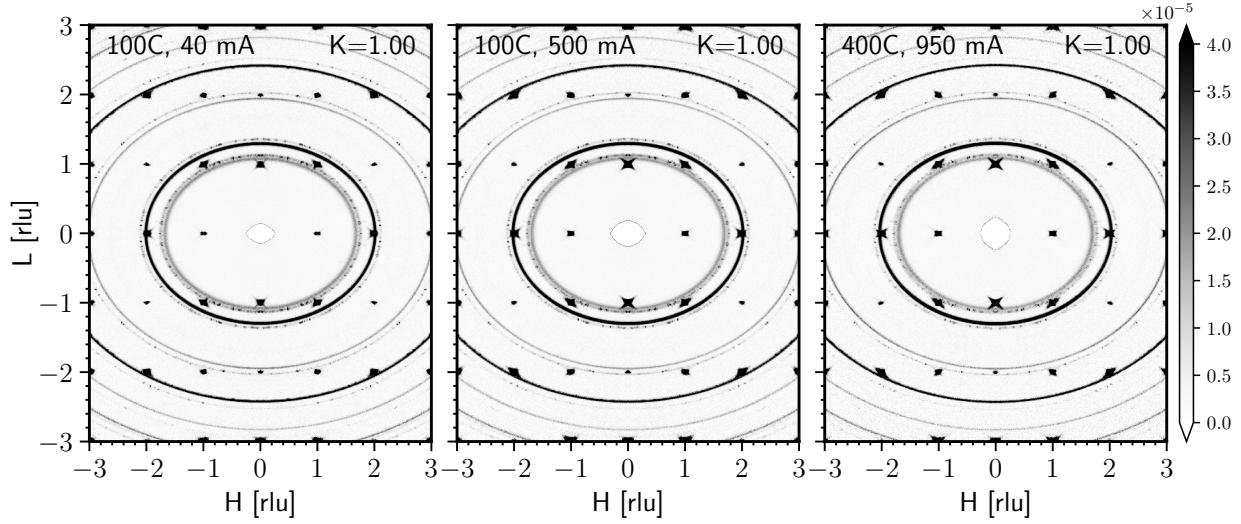


Fig. 2:  $\mathbf{Q}$ -dependence near  $K = 1$  of diffuse scattering from the ex-situ S1 dataset, with current along  $\mathbf{k}$ . The dark regions show pronounced diffuse scattering.

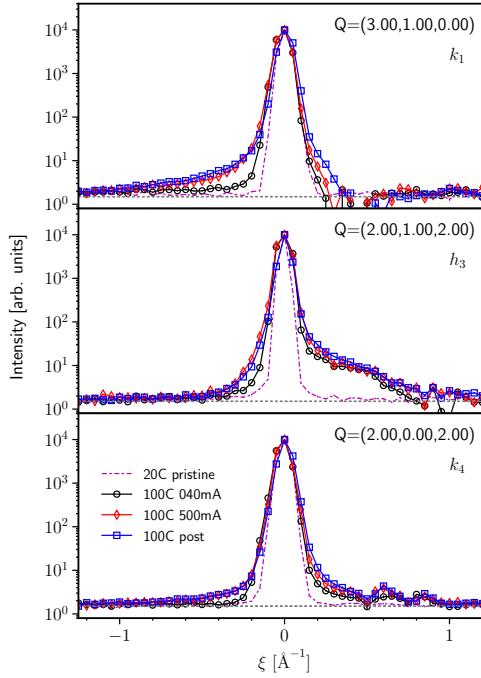


Fig. 3: Current and temperature dependence of the diffuse scattering features along different needles thru different Bragg peaks labeled in the figure. Background determined by taking cuts through nearby regions with no diffuse scattering was subtracted. Sample “100°C post” is the in-situ flashed crystal after the field was turned off and the sample cooled back to the furnace temperature 100°C. Background due to the furnace and electrodes was subtracted. Since the needles aren’t oriented along a high-symmetry direction, we plot data in  $\text{\AA}^{-1}$ .



Fig. 4:



Fig. 5:

## 2 Current dependence of diffuse scattering

In fig. 2, we show that there is no qualitative difference in the diffuse scattering while in fig. 3, we show that there is a quantitative difference. With increasing current density at fixed temperature, diffuse scattering is enhanced.

## 3 Sample-stick

I oversaw the procurement of a vacuum tube furnace by our lab at the University of Colorado Boulder (CU). I designed modifications to the furnace that enabled using a custom flash stick (also designed by me!) at CU. My lab mates are currently making heavy use of this to explore the role of sample environment on the physics of flash.

The modifications and fabrication were done by the machine shop at CU and parts assembled by an undergraduate research assistant in our group. The modifications to the furnace involved boring and tapping the furnace door to accept the sample stick. The sample stick itself is stainless steel with a vacuum feed-thru, kanthal electrodes (to withstand high temperature) and an alumina tube to (electrically) insulate the kanthal wires. See figs. 4-8.



Fig. 6:



Fig. 7:



Fig. 8: