U.S. DEPARTMENT OF ENERGY | Office of Science

Office of Science Graduate Student Research

2022 Solicitation 2 - Final Report for: Tyler Chase Sterling

OVERVIEW

Awardee and Project Information

Awardee Name:	Tyler Chase Sterling
Graduate Institution:	University of Colorado Boulder
Discipline:	Condensed Matter
Doctoral Dissertation/Thesis Title:	Physics of Energy Materials Investigated by Neutron Scattering
Degree Objective:	Ph.D.
Expected Graduation Date:	12/2024
SCGSR Research Project Title:	Probing the Non-Equilibrium Process of Electroluminescence Via in situ Neutron Diffuse Scattering
SCGSR Research Project Abstract:	It was recently demonstrated that application of a moderate electric field at elevated temperatures modifies materials' structural, electrical, optical, and other properties turning them effectively into new materials. When the field is applied and the material is heated above room temperature very briefly, its electrical conductivity increases dramatically and it begins to glow in an electroluminescence (EL) like phenomenon described as "flash." The physical mechanisms behind this process and the properties of the new materials that it creates are not understood. Even more puzzling, it was recently shown that applying a magnetic field to a sample undergoing EL causes nearby materials to EL too. We propose to perform in situ neutron diffuse scattering (NDS) experiments under simultaneous electric and magnetic fields. An apparatus for in situ electric field experiments already exists and we have used it to prove that in situ NDS can be used to probe the EL state. We plan to modify the apparatus to work under simultaneous electric and magnetic fields. We will apply these new tools to a technologically important class of materials: relaxors. The first step will be to solve for the response of the local atomic order to applied fields. Then we will develop computational models for the non-equilibrium many body processes involved in EL.
Starting Date:	6/12/2023
Ending Date:	5/31/2024
Host DOE Laboratory:	Oak Ridge National Laboratory (ORNL)
Division at the Host DOE Laboratory:	Neutron Scattering Division
Collaborating DOE Laboratory Scientist Name:	Feng Ye
Collaborating DOE Laboratory Scientist Email:	yef1@ornl.gov
Supervisor for Collaborating DOE Laboratory Scientist:	N/A
Scientific user facility involved in the SCGSR research project:	Spallation Neutron Source (SNS)

ACCOMPLISHMENTS

Research Goals and Objectives

Research Goals and Objectives:	We proposed to study a recently discovered phenomenon called 'flash' whereby a crystal is placed under a moderate electric field at high temperature. At fixed temperature, increasing the E-field over a critical value results in a sudden drop in the resistivity of the crystal and a metallic current begins to flow. Alternatively, we can hold the E-field fixed and raise the temperature over a critical value. Once the resistivity drops, a metallic current begins to flow and the material emits light ("flashes") in a electroluminescence-like phenomenon. Flash occurs in many materials under a wide range of conditions. To date, flash is more-or-less completely enigmatic. We planned diffuse neutron scattering experiments to the examine the structure of a crystal during flash. Our primary goals were (i) to extend what is known about flash to a technologically relevant material, relaxor PMNPT, (ii) to study the effect of simultaneous E-field and B-field, and (iii) to develop non-equilibrium models for the physics of flash based on what is learned in steps (i) and (ii). To some extent, we have addressed or begun addressing each of these goals. For all of them, we have actionable plans for how to continue to study these problems in the future.

In the case of (i), we performed in-situ neutron diffuse scattering measurements not only for PMNPT, but also SrTiO3, additional TiO2 samples, and attempted an experiment on La2CuO4 (the crystal broke). These experiments were done on-schedule (July 25th-28th, 2023) and were completed successfully, though the data were more-or-less null (as discussed in the mid-project report). In short, for PMNPT and SrTiO3 there was no visible diffuse scattering suggesting either the absence of defects (unlikely), uncorrelated defects (possible), or that defects formed only near the surface of samples these samples. In TiO2, diffuse scattering suggested the formation of bulk Wadsley defects (these data support earlier results that we are publishing). A reasonable course of action to identify defects near the surface would be X-ray diffuse scattering.

My lab mates at the University of Colorado (CU) did preliminary testing on the affect of B-field during flash. The original experiment used an EM-coil; we attempted to replicate the experiment with a permanent magnet. We found no affect due to magnetic field. We supposed that the coil simply acts as a heating element rather than the B-field being significant. Currently, my lab mates are trying to replicate the experiment using a coil and I will participate in the work in the future. Based on our own inconclusive results with B-field, and considering the technical complexity of a simultaneous E- and B-field apparatus, we decided not to pursue such a device at this time. Rather, we used our funds to invest in a new in-situ E-field apparatus to use at the HFIR source at ORNL for triple-axis neutron scattering experiments. The sample environment team designed it and our group paid to have it manufactured (March 2024). The goal of this new sample stick is to study the non-equilibrium dynamics during flash. If we can independently prove that the B-field is significant, we still plan to commission a new stick.

Step (i) at least partially enabled us to make claims about non-equilibrium physics during flash. Specifically, we are able to estimate to the temperature of the in-situ flashing sample. See the additional material for details. Currently, the temperature of an in-situ sample is completely unknown and attempts to measure it are dubious: some groups have measured the light spectrum and fit it as a black body. This is a poor approximation (the crystals are not close to perfect black bodies and also exhibit absorption/emission lines). Due to data quality limitations, our estimate not better, but does serve as a good proof of concept for follow up experiments. We have submitted (accepted) proposals to ARCS at the SNS and TAX at HFIR to do inelastic scattering measurements (we built an in-situ apparatus for HFIR). The goals are (a) to look for significant changes in the phonon spectra during flash and (b) to measure the temperature via the phonon populations. Both of these experiments will contribute to more detailed theoretical understanding of the microscopic physics of flash. Considering this, we have at least partially (~50%) addressed goal (iii).

Note: I am graduating in Fall 2024 and staying on a post-doc to continue this work. We have built our own vacuum furnace (I lead this effort) at CU, enabling us to study flashed samples with more control over conditions than other groups currently can. This effort was, in large part, inspired by the discovery that flash occurs quite differently in vacuum while flashing in-situ in vacuum furnaces at ORNL. Moreover, I have plans to measure the light emissions spectrum and do in-situ Raman measurements in the vacuum furnace at CU, among other things. Thanks for enabling all of this work!

Project Accomplishments

Project Accomplishments: We proposed to study a recently discovered phenomenon called 'flash' whereby a crystal is placed under a moderate electric field at high temperature. At fixed temperature, increasing the E-field over a critical value results in a sudden drop in the resistivity of the crystal and a metallic current begins to flow. Alternatively, we can hold the E-field fixed and raise the temperature over a critical value. Once the resistivity drops, a metallic current begins to flow and the material emits light ("flashes") in a electroluminescence-like phenomenon. Flash occurs in many materials under a wide range of conditions. To date, flash is more-or-less completely enigmatic.

We planned diffuse neutron scattering experiments to the examine the structure of a crystal during flash. Our primary goals were (i) to extend what is known about flash to a technologically relevant material, relaxor PMNPT, (ii) to study the effect of simultaneous E-field and B-field, and (iii) to develop non-equilibrium models for the physics of flash based on what is learned in steps (i) and (ii). To some extent, we have addressed or begun addressing each of these goals. For all of them, we have actionable plans for how to continue to study these problems in the future.

In the case of (i), we performed in-situ neutron diffuse scattering measurements for PMNPT and several other samples: SrTiO3, TiO2 with different flash conditions, and La2CuO4 (which broke during the experiment). These results were discussed extensively in the mid-progress report, so I will only briefly summarize them here. These experiments were more-or-less null (as discussed in the mid-project report). In short, for PMNPT and SrTiO3 there was no visible diffuse scattering suggesting either the absence of defects (unlikely), uncorrelated defects (possible), or that defects formed only near the surface of samples these samples (likely). In TiO2, diffuse scattering suggested the formation of bulk Wadsley defects (these data support earlier results that we are publishing). A reasonable course of action to identify defects near the surface would be X-ray diffuse scattering, though this will be complicated by the low sensitivity of X-rays to oxygen. I may attempt this in the future.

Our diffuse scattering results on TiO2 are consistent with our hypothesis (that is shared by others in the field) that the formation of oxygen defects is relevant for flash. On the other hand, the absence of diffuse scattering in the other samples we measured doesn't support this. However, these null-results don't necessarily contradict this hypothesis either. As mentioned above, it may be possible that oxygen defects exist, but are randomly distributed or only present near the surface (in either case, we wouldn't see this with neutron scattering).

An interesting result (from data on TiO2) is that, while the shape of diffuse scattering isn't affected by the in-situ current, the magnitude of diffuse scattering is (see additional material). Moreover, Bragg peaks are correlated with current density. We were able to use the integrated Bragg peaks vs. current to estimate the temperature of in-situ flashing samples (see additional material and see below). This latter result serves a good proof of concept for follow up inelastic scattering experiments that we have planned later this year to more precisely measure the temperature. Moreover, it shows that we can use these in-situ measurements to understand the non-equilibrium physics of flash.

A side-effect of the experiments at ORNL is that we noticed a non-trivial change in the flash conditions when the sample is flashed in vacuum (the in-situ furnaces at ORNL at high vacuum). Inspired by this, 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 (also designed by me!) flash stick at CU. My lab mates are currently making heavy use of this to explore the role of sample environment on the physics of flash. Moreover, I have plans to measure the light emission spectrum vs. sample environment and to do in-situ Raman measurements in the vacuum furnace at CU, among other things. I will graduate this Fall (2024) and will re-join my group as post-doc to continue this work.

Regarding the simultaneous E- and B-field: my lab mates at the University of Colorado (CU) did preliminary testing on the affect of B-field during flash. The originally published experiment used an EM-coil; we attempted to replicate the experiment with a permanent magnet. We found no affect due to magnetic field. We supposed that the coil simply acts as a heating element rather than the B-field being significant. Currently, my lab mates are trying to replicate the experiment using a coil and I will participate in this work in the future. So far, the reported experimental set up doesn't agree with the claimed physics and we are skeptical of this effect. Based on our own inconclusive results with B-field, and considering the technical complexity of a simultaneous E- and B-field apparatus, we decided not to pursue such a device at this time. Rather, we used our funds to invest in a new in-situ E-field apparatus to use at the HFIR source at ORNL for triple-axis neutron scattering experiments. We commissioned the ORNL sample-environment team to designed the new sample stick and we paid to have it manufactured (completed in March 2024). The goal of this new sample stick is to study the non-equilibrium dynamics during flash. In the future, if we can independently prove that the B-field is significant (which my group is trying to do with our in-house vacuum furnace at CU), we will commission a new stick and will apply for beam time through the usual channels.

To some extent, we were able to make claims about non-equilibrium physics during flash. Specifically, we are able to estimate to the temperature of the in-situ flashing sample based on our neutron scattering measurements. See the additional material for details. Currently, the temperature of an in-situ sample is completely unknown and attempts to measure it are dubious: some groups have measured the light spectrum and fit it as a black body. This is a poor approximation (the crystals are not close to perfect black bodies and also exhibit absorption/emission lines). Due to data quality limitations, our estimate is not better, but does serve as a good proof of concept for follow up experiments.

To that end, we have submitted (accepted) proposals to ARCS at the SNS and TAX at HFIR to do inelastic scattering measurements using the in-situ apparatuses we commissioned. Being able to accurately measure the temperature and, more importantly, correlate it with the current density will provide valuable insight on flash. The specific goals are (a) to look for significant changes in the phonon spectra during flash and (b) to measure the temperature via the phonon populations. For (a) we are interested in either changes in phonon energies or phonon line-shapes as well the appearance of new peaks. Changes in phonon energies and widths are directly related to interactions between electrons and phonons, or electrons and photons (the E-field), enabling us to directly relate the experiment to non-equilibrium models for the physics. Most likely, the former is the most important effect since there is a large current flowing during flash. On the other hand, if new peaks show up in the spectrum, this implies a change in the structure (i.e. a change in the atomic displacement pattern). While not a direct probe of the non-equilibrium dynamics, this is an interesting result if observed.

For (b), we want a way to precisely measure the temperature. Phonons are Bosons, so follow the Bose-Einstein distribution. i.e., the intensity of peaks in the neutron spectrum is related to the temperature of the sample. While flashing, the temperature of the sample increases (Joule heating). We have (successfully) proposed to very carefully measure the peak heights on both ARCS and TAX at ORNL. These experiments will be completed this year. We mention that we attempted a related experiment on ARCS in June 2024 that was unsuccessful due to equipment failure (part of the spectrometer wouldn't move into place). This was discussed in detail in the mid-project report, so I won't belabor it here.

We have also attempted to use our diffuse scattering data to refine the structure of TiO_2 during flash and correlate any in-situ structural changes with flash. i.e. we want to 'fit' the real Bragg scattering intensities by letting atomic displacements in the unitcell be free parameters. Note, we haven't attempted this for PMNPT or $SrTiO_3$ yet since TiO_2 is proving more fruitful overall. I will look into the other datasets when we have finished with TiO_2. Refining the structure involves integrating Bragg peaks from the diffuse scattering data. Unfortunately, the Bragg peak intensities are too noisy to make any significant progress. The poor data quality is due to 'wobble'; it is very difficult to center a sample in the in-situ apparatus and, while rotating the sample during scattering measurements, the sample 'wobbles' around the axis of rotation. The resulting displacement in the sample position apparently has a significant effect on the Bragg peak intensities. Still, we are able to use the trend in peak intensities with current to estimate the temperature (see additional materials).

Uploading Additional Materials

Would you like to upload any additional materials?	Yes
Additional Materials:	scgsr_final.pdf

Use of DOE Laboratory Resources

DOE Laboratory Facilities and Research Capabilities:	I heavily used the CORELLI instrument at the Spallation Neutron Source (SNS) at Oak Ridge National Laboratory (ORNL). We had about 6 days beam time and measured 5 samples. We also attempted an experiment at ARCS at the SNS, but that experiment failed due to equipment failures. Note, we submitted proposals for follow up experiments in 2024 to redo the failed ARCS experiment and to do a new experiment at the reactor source (HFIR) at ORNL.

Reporting Changes or Issues

Were there any changes or issues in accomplishing the research goals and objectives?	No

PRODUCTS

Products as a Direct Result of the SCGSR Project

Do you have any completed products?	Yes
Publication Type:	Presentation - Conference, Workshop, or Symposium
Title:	Introduction of oxygen vacancy planes into rutile TiO2 via electric current revealed by neutron scattering
Author(s):	Tyler Sterling, Feng Ye, Seohyeon Jo, Anish Parulekar, Gang Cao, Rishi Raj, Dmitry Reznik
Conference, Workshop, or Symposium Name:	Aspen Winter Physics Conferences: Disorder and Quantum Phases of Matter
Presentation Type:	Poster/Lead Author
Conference, Workshop, or Symposium Location:	Aspen CO
Conference, Workshop, or Symposium Date:	12/14/2023

Planned Products and/or Products Under Development

Are you developing or planning to develop any type(s) of the above mentioned research products as a direct result of the SCGSR research project?	Yes
Publication Type:	Archival Publication - Peer Reviewed Scientific or Technical Journal Article
Working Article Title:	Introduction of oxygen vacancy planes into rutile TiO2 via electric current revealed by neutron scattering
Author(s):	Tyler Sterling, Feng Ye, Seohyeon Jo, Anish Parulekar, Gang Cao, Rishi Raj, Dmitry Reznik
Tentative Journal Name:	Physical Review X
Publication Volume:	
Publication Issue Number:	
Publication Page Numbers:	
Publication Year:	2024

Publication Type:	Presentation - Conference, Workshop, or Symposium
Working Title:	Introduction of oxygen vacancy planes into rutile TiO2 via electric current revealed by neutron scattering
Author(s):	Tyler Sterling, Feng Ye, Seohyeon Jo, Anish Parulekar, Gang Cao, Rishi Raj, Dmitry Reznik
Targeted Conference, Workshop, or Symposium Name:	American Conference on Neutron Scattering
Presentation Type:	Oral Presentation
Targeted Conference, Workshop, or Symposium Location:	Knoxville TN
Targeted Conference, Workshop, or Symposium Date:	6/23/2024

IMPACTS

Contribution to Thesis / Dissertation

Research Project Progress:	The majority of my the relevant part of my thesis project has been completed as a direct result of the fellowship. I planned diffuse scattering experiments at ORNL and completed them. The results were somewhat null, but informed a directly related experiment we already had data for. I developed some software tools for use on the new datasets as well as the old one and this has enabled us to finish a paper (soon to be submitted). The paper is the 'backbone' of my thesis. Moreover, we have planned more experiments in our lab at CU (I am staying as a post-doc) and follow up experiments at ORNL.

Graduate Training and Education

Knowledge and Skills: The main skills I learned (and hoped to learn!) were about the instruments at the SNS. I hope to be a professor with my own PhD students: learning the details of neutrons scattering enables me to teach these things to my students in the future! I worked alone on CORELLI for multiple days and learned about the instrument, neutrons, furnaces, etc. I also learned about data reduction, which is arguably more difficult to use/understand than the hardware.

Development of the Principle Discipline

Impacts of Principle Discipline(s): We have used neutron scattering to reveal ordering of defects (in TiO2) or lack-there-of (in PMNPT, SrTiO3). The nature of these defects and the response to current has important implications for understanding flash. We are also developing software tools to better enable understanding a certain type of diffuse scattering (total diffuse scattering): this will be of use to the scientific community at large, not just us. Moreover, we have commissioned a new sample-holder that enables in-situ experiments at HFIR and these tools will be of interest to many users in the future.

OTHER ACTIVITIES

Participation in Other Activities at the Host DOE Laboratory During the SCGSR Project Period

Participation in research Seminars/Workshops within the division you are currently: Occasionally

Participation in Research Seminars/Workshops in other divisions: None

Participation in Training Seminars/Workshops on Professional Development: None

Seminars/Workshops description: Many of the professional development workshops appeared to be targeted at folks who wanted to enter industry or national labs. I intend to pursue a professorship and a career in academia, so I felt like the workshops were irrelevant. There were other workshops on networking etc., but these aren't particularly relevant to me. I did attend some talks on research topics in my division (neutron scattering) because I am very interested in this material.

ABSTRACT

General Audience Abstract

General Audience Abstract: We proposed to study a recently discovered phenomenon called ‘flash’ where a crystal is placed in electric field at high temperature. At fixed temperature, increasing the E-field over a critical value results in a sudden drop in the resistivity. Alternatively, we can hold the E-field fixed and raise the temperature over a critical value. Once the resistivity drops, a metallic current begins to flow and the crystal emits light (“flashes”) in a electroluminescence-like phenomenon. To date, flash is more-or-less completely enigmatic.

We performed neutron scattering experiments to examine the structure of oxides PMNPT, SrTiO3, and TiO2. In TiO2, we found that extended defects form as oxygen leaves the crystal and that these defects are affected by the size of the current. For PMNPT and SrTiO3 there was no visible change in scattering, suggesting either the absence of defects (unlikely), random defects (possible), or defects only near the surface (likely). The current dependence of diffuse scattering in TiO2 is a new result! It is possible that defects in SrTiO3 and PMNPT (if present) are sensitive to current, but we can't say more.

We used our results of current dependence of scattering in TiO2 to estimate the temperature of flashing crystals. The temperature is related to the non-equilibrium dynamics, which is of interest DoE research areas in general. Our results are approximate, but serve as a valuable proof of concept that we can probe the non-equilibrium nature of flash with neutrons. To that end, we commissioned a new apparatus that enables in-situ scattering at the reactor at ORNL and have planned two follow up experiments this year to do a more detailed study the non-equilibrium physics!

All of this work was enabled by the DoE SCGSR fellowship, which funded me to work at ORNL. This experience introduced me to the culture and life at a DoE lab! I look forward to keeping up with all of the connections I have made.

COMMENTS

Reflection or Comments About Your Overall SCGSR Experience at the Host DOE Laboratory

Would you like to provide reflection or comments about your overall SCGSR experience at the Host DOE Laboratory? No

SUBMIT DATE

Submit Date:	5/23/2024 5:45 PM

SCGSR final report: additional information

Tyler C. Sterling

Department of Physics, University of Colorado Boulder, Boulder CO, 80309, USA

May 23, 2024

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 $\boldsymbol{Q}$. For simplicity, we plot as a function of length $|\boldsymbol{Q}| \equiv Q$. 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

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

with $\boldsymbol{G}$ a Bragg peak, A a constant prefactor that depends on sample-size, incident-flux, etc. but not on $\boldsymbol{G}$ 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 $[I_1(\boldsymbol{G}, T_1) - I_2(\boldsymbol{G}, T_2)]/[I_1(\boldsymbol{G}, T_1) + I_2(\boldsymbol{G}, T_2)]$ where $I(\boldsymbol{G}, T)$ is the intensity measured at Bragg peak $\boldsymbol{G}$ for the same material but under different conditions; we assume

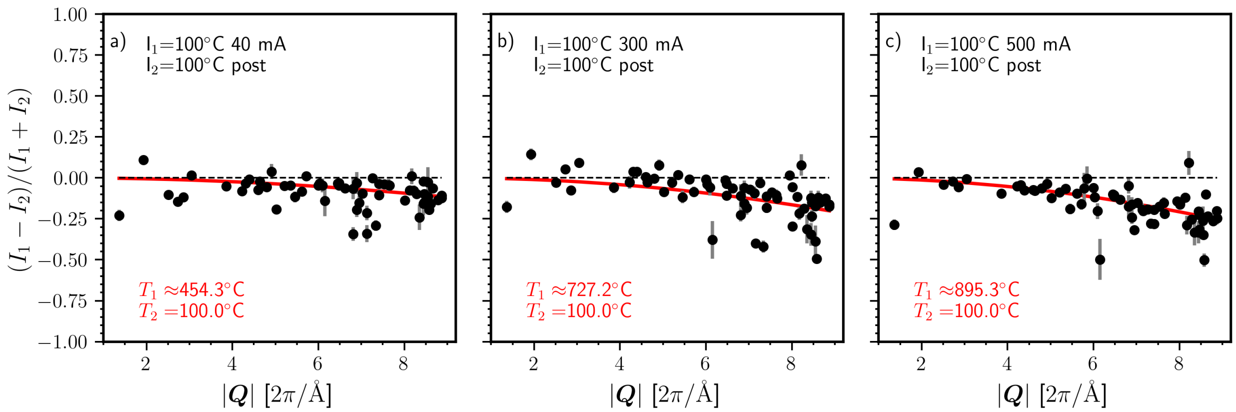

Fig. 1: Normalized difference in intensities. The datasets are labeled in the panels. The difference should be 0 for scattering from identical materials. Deviations from 0 signal structural and temperature differences. Since the flashed samples are approximately isostructural, we assume the leading deviation is due to temperature and estimate the in-situ temperature by fitting. The post-flash sample is ex-situ, so we fix $T_2 = 100°$ 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 $I(\boldsymbol{Q},T) \approx AF(\boldsymbol{G})\exp(2W(\boldsymbol{G},T))$ with $F(\boldsymbol{G})$ the form-factor. In a true Bravais lattice, $F(\boldsymbol{G}) = b^2$ is independent of $\boldsymbol{G}$. However, to include the non-Bravaisness, we let $F(\boldsymbol{G}) \sim |\sum_a b_a \exp(i\boldsymbol{G}\cdot\boldsymbol{\tau}_a)|^2$ depend on $\boldsymbol{G}$ via the basis positions. Moreover, we assume that $F(\boldsymbol{G})$ 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 $\exp(W(\boldsymbol{G},T))$. Then (dropping explicit $\boldsymbol{G}$ and T dependence for now)

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

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

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

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

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

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

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 ω is independent of experimental conditions and all that changes is T. Define $\Delta n = n_{BE}(\hbar\omega/k_BT_2) - n_{BE}(\hbar\omega/k_BT_1)$. Then $\Delta W = -Q^2 l_0^2\Delta n/3$ and

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

If we measure ΔI and can estimate ω 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 ω. The results lead to $\hbar\omega \approx 9.87$ meV and $l_0 = 0.053$ Å, which are sensible: ~ 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 ω and m to the experimental data.

1.2 Temperature effects

The difference intensities, ΔI, 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.

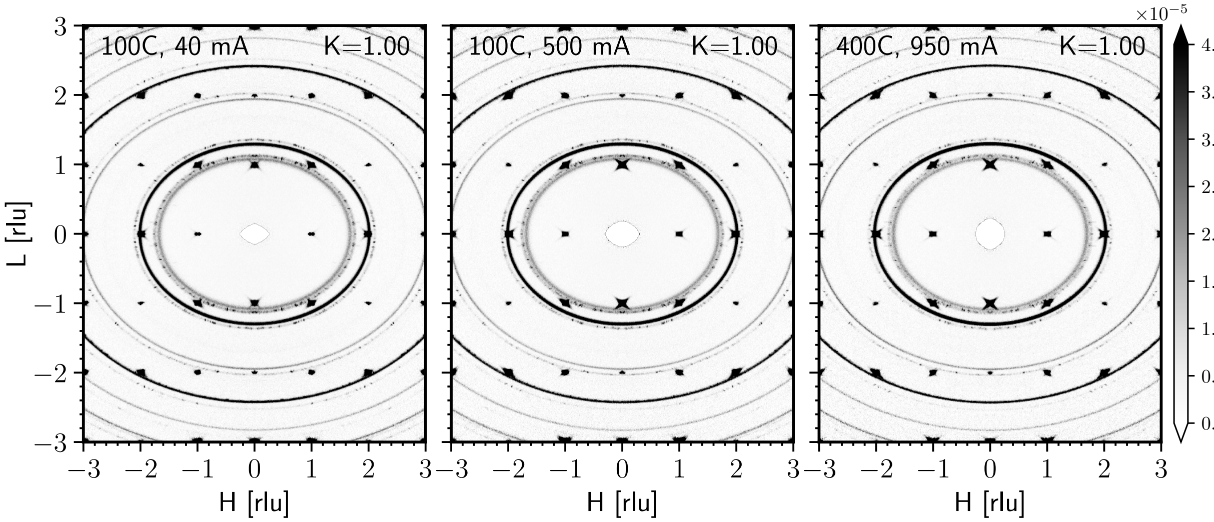

Fig. 2: $\boldsymbol{Q}$-dependence near $K = 1$ of diffuse scattering from the ex-situ S1 dataset, with current along $\boldsymbol{k}$. The dark regions show pronounced diffuse scattering.

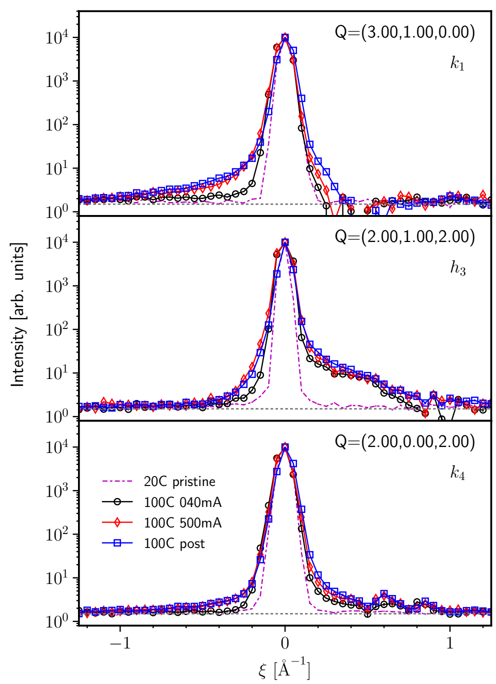

Fig. 3: Current and temperature dependence of the diffuse scattering features along different needles thru different Bragg peaks labeled in the figure. Background determined by taking cuts through nearby regions with no diffuse scattering was subtracted. Sample "100°C post" is the in-situ flashed crystal 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 Å^{-1}.

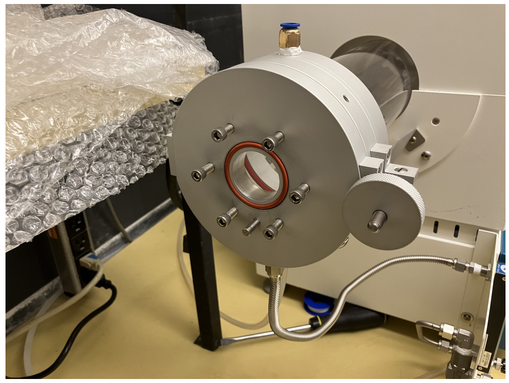

Fig. 4:

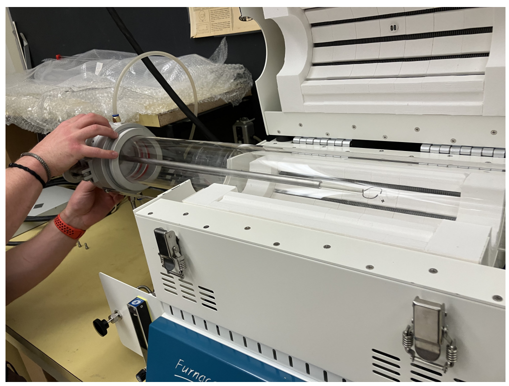

Fig. 5:

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 differnce. 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: