

## New Quantum Materials: The Search for Topological Thermoelectrics and Topological Superconductors

### Overview

Quantum materials have properties that can revolutionize sustainable energy technologies and quantum computing. Every material has a band structure and is made up of atoms described by quantum mechanics. However, quantum materials specifically refers to those materials whose properties cannot be approximated by a classical description; such materials can host various entangled electronic phases including superconductivity, magnetism, thermoelectricity, and topological phases. **The focus of my group will be on the design, synthesis, and low temperature characterization of crystalline solids with emergent quantum phases.** My philosophy is centered around finding connections between different subfields in materials research to identify new ground for exploration that will motivate future fundamental research with relevance to technology.

The foundation of chemical engineering is built on the ideas of material balance and energy balance. On this foundation gained in my early training, I have built expertise in the chemistry and physics of quantum materials. My search for novel materials will be guided by the overlap of electronic properties, shown in Fig. 1. **Unconventional superconductivity** appears in the vicinity of magnetism [1, 2], and thermoelectric performance can be enhanced with short-range magnetic order [3]. We will study **materials with a known magnetic ground state and tune their properties in search of a high thermoelectric figure-of-merit  $zT$  and unconventional superconductivity.** The topology of the electronic band structures is important, and topologically non-trivial electronic states, containing knots or twists, result in topological materials, such as topological insulators and Dirac/Weyl semimetals.

**Topological thermoelectrics** are those materials with topologically non-trivial states that contribute to their high thermoelectric efficiency. A key challenge in the field of thermoelectrics is to decouple the electrical conductivity from the thermal conductivity, with the goal of achieving a “phonon-glass electron-crystal” (PGEC) [4]. The brute-force “classical” methods to improve thermoelectric efficiency include introducing ionic disorder to scatter phonons without scattering electrons, and some success was achieved in my work on copper chalcogenides [5, 6]. However, a more elegant and inherently “quantum” path towards achieving PGEC in a material may be found in topological insulators such as  $\text{Bi}_2\text{Se}_3$  [7], where the electronic conductivity is robust against disorder due to topologically-protected electronic states [8]. Some topological materials have been demonstrated to violate the Lorenz number, and this should be utilized in the search for efficient thermoelectrics [9]. However, despite tremendous efforts, these materials seem to have hit an intrinsic limit in terms of thermoelectric efficiency [10]. Also, doped topological insulators such as  $\text{Cu}_x\text{Bi}_2\text{Se}_3$  [11] are some of the leading candidates for realizing **topological superconductivity**, but these doped crystals suffer from low superconducting volume fraction [11], likely due to inhomogeneity of the intercalated Cu atoms. To overcome these challenges we need new families of topological materials where we can grow high-quality single crystals with new techniques, such as high-pressure synthesis.

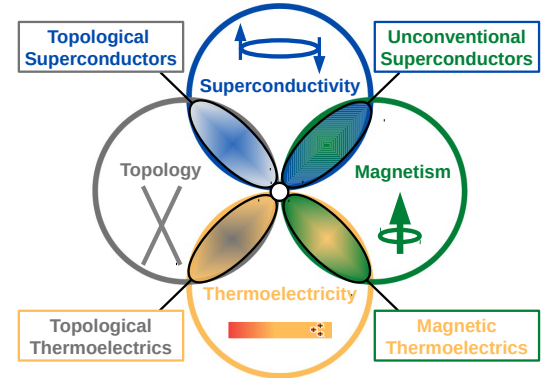


Figure 1: Various properties of quantum materials, and emergent quantum states at the overlap.

### Previous Work

I have worked on synthesis of many materials, including chalcogenides, antiperovskite oxides (Fig. 2), bismuthates, irridates, ruthenates, cuprates, silver oxides, high-entropy oxides, and pnictides, using a wide variety of synthesis methods, including flux, vapor transport, floating zone, electrochemical, and high-pressure. We synthesized a new phase of  $\text{AgSbO}_3$  with strong Sb-O hybridization [12] and identified a new topological Weyl semimetal  $\text{Ag}_2\text{BiO}_3$  using high-pressure high-temperature methods [13]. We highlight the similarity between  $\text{Ag}_2\text{BiO}_3$  (space group  $Pnn2$ ) and the perovskite type bismuthate  $\text{BaBiO}_3$  (space group  $I2/m$ ), both with nominal “ $\text{Bi}^{4+}$ ” state that disproportionates into two distinct sites. Suppression of the disproportionation in  $\text{Ag}_2\text{BiO}_3$  (space group  $Pnna$ ) will stabilize a Dirac semimetal state and can even lead to superconductivity. At UBC, I have grown **high-quality superconducting cuprate and  $\text{Fe}(\text{Se},\text{Te})$  crystals using flux method and collaborated on the electrochemical growth of  $(\text{Ba},\text{K})\text{BiO}_3$  crystals**, and measured x-ray absorption of superconducting  $(\text{Ba},\text{K})\text{SbO}_3$  synthesized at high-pressure by collaborators in Germany [14].

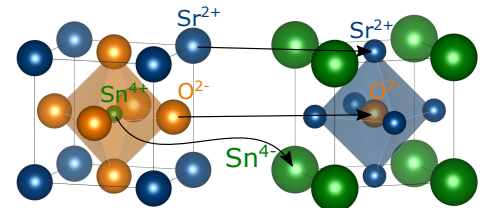


Figure 2: Crystal structures of  $\text{SrSnO}_3$  (perovskite oxide, left) and  $\text{Sr}_3\text{SnO}$  (antiperovskite oxide, right), highlighting ionic states of atoms. The  $\text{Sn}^{4+}$  state makes  $\text{Sr}_3\text{SnO}$  a topological Dirac-semimetal.

In Kyoto, I found superconductivity in the highly air-sensitive  $\text{Sr}_{3-x}\text{SnO}$  [15] containing the unusual  $\text{Sn}^{4-}$  state revealed in Mößbauer spectroscopy [16], crystal structure of the antiperovskite oxide  $\text{Sr}_3\text{SnO}$  and typical perovskite oxide  $\text{SrSnO}_3$  are shown in Fig. 2. We observed in bulk  $\text{Sr}_{3-x}\text{SnO}$  evidence for **Dirac electrons** using NMR [17] and signs of **unconventional superconductivity** in muon spin relaxation ( $\mu\text{SR}$ ) [18], then studied thin-films of  $\text{Sr}_3\text{SnO}$  using  $\beta\text{-NMR}$  in search of chiral surface states [19].

Topological properties have been realized in the class of materials with **2D square-nets** in their structure, where these properties can be predicted through simple chemical considerations [20], and recently we have demonstrated a non-trivial drumhead surface state in  $\text{ZrSiTe}$  [21]. At UBC, I have expanded the work on the square-net family of compounds to further elucidate the origin of their topologically non-trivial states. Specifically, I synthesized **metal antimonides**  $\text{MSb}_2$  ( $M = \text{Ca}, \text{Sr}, \text{Eu}, \text{Yb}$ , crystal structure shown in Fig. 3) with distorted square-nets. We have characterized the semimetallicity and superconductivity of  $\text{CaSb}_2$  [22], and studied the superconductivity using  $\mu\text{SR}$  [23]. Antimonides have promising thermoelectric properties [24] and host topological states [25]. I plan to study the  $\text{MSb}_2$  family of materials to elucidate their potential as **topological thermoelectricity**.

Recently, I discovered superconductivity and electron-phonon drag in non-centrosymmetric semimetal  $\text{LaRhGe}_3$  [26]. We have also been working on high-entropy materials, studying the properties of magnetic spinel oxides [27]. This emergent field offers a new perspective on the tunability of stability and properties of materials using entropy [28, 29, 30]. My most recent work demonstrates a novel concept of charge-entropy stabilization in the selenide  $(\text{Ag}, \text{Sn})\text{Se}$ , with  $2+/4+$  fluctuations on Sn, by utilizing Mößbauer spectroscopy and X-ray photoelectron spectroscopy (XPS) [31]. Furthermore, I have worked on materials with high-mobility of electrons, where electron-electron interactions are dominant, resulting in viscous/ballistic electron flow. To this end we study the high-mobility oxides  $\text{ReO}_3$  and  $\text{PdCoO}_2$  [32], where we apply fluid mechanics concepts to electron flow in quantum materials. Having studied many quantum materials has allowed me to develop a holistic skill-set, which will help me in searching for new emergent quantum phases.

### Future Projects

**I will search for superconductivity in Ag-based materials using high-pressure synthesis, including the Ag oxides/fluorides.** One target is superconducting perovskite oxides containing silver, where under high-pressure silver can replace alkali metals in oxides. The silver oxides are more stable than their alkali metal analogues and some are Dirac/Weyl semimetals. Another target is superconducting silver fluorides. The proposed family of high-pressure phases containing  $\text{AgF}_2$  (shown in Fig. 4) can host superconductivity, analogous to superconductivity in the  $\text{CuO}_2$  based materials [33]

**I will explore the magnetic and thermoelectric properties of natural superlattices, such as misfit structures**  $[(MX)_{1+\delta}]_m(TX_2)_n$ . These misfit structures are layered compounds containing two building blocks,  $MX$  rocksalt layers and  $TX_2$  dichalcogenide layers. Compounds reported include  $M = \text{Sn}, \text{Pb}, \text{Sb}, \text{Bi}, \text{La}, \text{Ce}, \text{Sm}, \text{Tb}$ ,  $T = \text{Ti}, \text{V}, \text{Nb}, \text{Ta}, \text{Cr}$ , and  $X = \text{S}, \text{Se}$  [34, 35], which offer a vast map for exploration and expansion to include new elements. I will explore magnetic order in relation to the 2D limit in isolated  $TX_2$ , and enhancement of thermoelectric efficiency due to phonon scattering at the superlattice interface. This will bridge together the work on exfoliated and bulk  $TX_2$  materials.

**I will investigate topological superconductivity and thermoelectricity in the  $\text{MSb}_2$  family, and explore their topological properties in light of Sb-Sb distortions in these materials.** In this family of antimonides, the distortion of Sb-Sb bonds into zigzag chains gives rise to non-symmorphic space group of the crystal structure, and the emergence of nodal-line state in their electronic structure. My work will focus on the superconducting pairing mechanism and identifying any connection to topology in  $\text{MSb}_2$ .

### Rationale for Experimental Techniques

In my lab, we will study the interplay between crystal structure, electronic structure, and physical properties in new topological thermoelectrics and superconductors. To successfully study various materials one needs a variety of synthesis and characterization tools. We will utilize electrical and thermal transport,  $\mu\text{SR}$ , and X-ray spectroscopy to study materials in my lab. On the synthesis front, **I plan to combine state-of-the-art crystal growth methods with high-pressure synthesis.**

Having the niche of high-pressure synthesis techniques in my lab will enable exploration of new range of parameter space for synthesis. While it is common knowledge that most materials show a wide variety of distinct phases in their

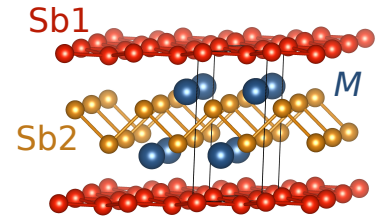


Figure 3: Crystal structure of  $\text{MSb}_2$  highlighting the two different Sb chains. Sb1 chains form a distorted square-net.

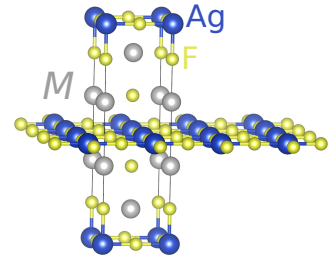


Figure 4: Crystal structure of layered silver fluoride,  $\text{M}_2\text{AgF}_4$ , a proposed new family of superconductors.

temperature-pressure phase diagram, conventional synthesis routes cannot make effective use of the pressure dimension and solely rely on temperature, deeply limiting the possible phases. This is especially important for superconductors, because the phase with the highest transition temperature for a specific ratio of elements is sometimes only stabilized at high pressures. However, high-pressure synthesis allows us to **add another dimension to the phase diagrams** and expands the number of crystalline materials we can make. By applying pressure to the constituent elements and by providing energy, in the form of heat, we can form high-pressure phases distinct from those formed at ambient conditions. These metastable phases can be “quenched” before removing the pressure from the reaction vessel and retrieving the sample. Quantum materials discovered using high-pressure synthesis include  $\text{Ba}_2\text{CuO}_4$  [36] and  $\text{KBiO}_3$  [37]. **While the need for high pressure is widely recognized, a key challenge in the field has been to grow single crystals of high-pressure materials that are large enough for devices and physics experiments**, such as angle-resolved photoemission spectroscopy (ARPES) and scanning tunneling microscope (STM). The lack of experimentally synthesized crystals has kept our understanding of such high-pressure materials limited despite the novel properties they host.

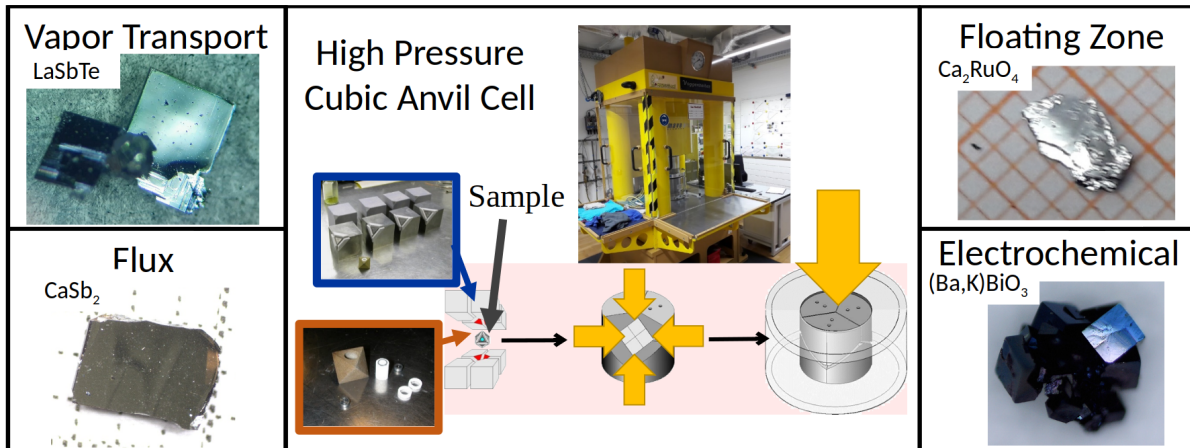


Figure 5: Crystals I have grown: Topological crystalline insulator,  $\text{LaSbTe}$ , vapor transport (top left), Superconducting Dirac nodal-line semimetal,  $\text{CaSb}_2$ , self-flux (bottom left), Mott Insulator,  $\text{Ca}_2\text{RuO}_4$ , floating zone (top right), unconventional superconductor,  $(\text{Ba},\text{K})\text{BiO}_3$ , electrochemical (bottom right), and high-pressure synthesis setup with cubic-anvil cell at MPI-Stuttgart (center).

However, recent progress has been made in integrating crystal growth techniques into high-pressure synthesis, and this is demonstrated with work on hexagonal boron nitride crystals [38]. Utilizing recent technical advancements in anvil cell design [38], I propose elevating the conventional growth methods of flux growth to work at high, GPa-level pressure. In particular, I will incorporate an **octahedral anvil cell with a Walker-type press** (center of Fig. 5) and a belt press. These type of presses can achieve pressures up to 20 GPa and temperatures of up to 1400 °C, and, most importantly, have a large sample space of up to 100 mm<sup>3</sup>. With the large sample space I will be able to synthesize large, high-pressure grown samples **suitable for everything from device-level transport to neutron and muon experiments**. Besides searching for new, high-pressure stabilized phases of materials, my lab will utilize different methods to synthesize crystals of new quantum materials, including those shown in Fig. 5.

### Synergy with University of California, Santa Barbara (UCSB)

I look forward to collaborations with members of the Materials Department at UCSB working on theory and experiments involving quantum and electronic materials. Bulk crystal growth techniques and high-pressure apparatus I propose to build will enable new collaborations with faculty in the area of energy materials and materials for quantum information. I look forward to collaboration with faculty working on superconducting, thermoelectric, magnetic, and disordered materials.

My research will benefit from the available facilities at UCSB. For bulk synthesis, I will utilize synthesis equipment such as box furnaces, tube furnaces, quartz sealing station, and glovebox. As my lab expands, it will include other crystal growth techniques and high-pressure synthesis equipment; I will evaluate the properties of materials synthesized in my lab with a broad range of techniques, including heat capacity, magnetic susceptibility, thermal transport, and electrical transport, with visits to user facilities for  $\mu\text{SR}$ , neutron diffraction, and X-ray spectroscopy experiments. My lab will provide research groups at UCSB with high-quality single crystal samples.

To complement the work at UCSB, I plan to maintain my collaborations at CLS (XPS, XAS), TRIUMF ( $\mu\text{SR}$ ,  $\beta\text{-NMR}$ ), University of British Columbia (STM, ARPES, Theory), Kyoto University (NMR, Mößbauer spectroscopy), University of Tokyo (high-pressure measurements), ETH Zurich (Theory), and MPI-Stuttgart (Theory and X-ray Spectroscopy). Finally, by building on my robust background in quantum materials, I hope to expand on the quantum materials research at UCSB and develop new strong collaborations.

## References

- [1] B. Keimer, S. Kivelson, M. Norman, S. Uchida, and J. Zaanen. From quantum matter to high-temperature superconductivity in copper oxides. *Nature*, 518(7538):179–186, 2015.
- [2] G. Stewart. Superconductivity in iron compounds. *Rev. Mod. Phys.*, 83(4):1589, 2011.
- [3] J. Christensen and B. Frandsen. Understanding the short-range magnetic correlations in mnte through magnetic pair distribution function analysis. *Acta Crystallogr.*, 77:305, 2021.
- [4] G. S. Nolas, G. A. Slack, and S. B. Schujman. Semiconductor clathrates: A phonon glass electron crystal material with potential for thermoelectric applications. In *Semicond. Semimet.*, volume 69, pages 255–300. Elsevier, 2001.
- [5] **M. Oudah**, K. M. Kleinke, and H. Kleinke. Thermoelectric properties of the quaternary chalcogenides  $\text{BaCu}_{5.9}\text{STe}_6$  and  $\text{BaCu}_{5.9}\text{SeTe}_6$ . *Inorg. Chem.*, 54:845–849, 2015.
- [6] P. Jafarzadeh, **M. Oudah**, A. Assoud, N. Farahi, E. Müller, and H. Kleinke. High thermoelectric performance of  $\text{Ba}_3\text{Cu}_{16-x}(\text{S}, \text{Te})_{11}$ . *J. Mater. Chem. C*, 6:13043–13048, 2018.
- [7] J. Gooth, G. Schierning, C. Felser, and K. Nielsch. Quantum materials for thermoelectricity. *MRS Bull.*, 43:187–192, 2018.
- [8] M. Neupane, S.-Y. Xu, L. A. Wray, A. Petersen, R. Shankar, N. Alidoust, C. Liu, A. Fedorov, H. Ji, J. M. Allred, et al. Topological surface states and dirac point tuning in ternary topological insulators. *Phys. Rev. B*, 85:235406, 2012.
- [9] C. Fu, Y. Sun, and C. Felser. Topological thermoelectrics. *APL Mater.*, 8:040913, 2020.
- [10] A. Adam, A. Elshafaie, A. E.-M. A. Mohamed, P. Petkov, and E. Ibrahim. Thermoelectric properties of te doped bulk  $\text{Bi}_2\text{Se}_3$  system. *Mater. Res. Express*, 5:035514, 2018.
- [11] Y. S. Hor, A. J. Williams, J. G. Checkelsky, P. Roushan, J. Seo, Q. Xu, H. W. Zandbergen, A. Yazdani, N. P. Ong, and R. J. Cava. Superconductivity in  $\text{Cu}_x\text{Bi}_2\text{Se}_3$  and its implications for pairing in the undoped topological insulator. *Phys. Rev. Lett.*, 104:057001, 2010.
- [12] **M. Oudah**, M. Kim, R. Dinnebier, G. McNally, K. Foyevtsova, D. Bonn, and H. Takagi. A new high-pressure high-temperature phase of silver antimonate  $\text{AgSbO}_3$  with strong Ag–O hybridization. *Inorg. Chem.*, 2024.
- [13] **M. Oudah**, M. Kim, K. Rabinovich, K. Foyevtsova, G. McNally, K. Berkay, K. Küster, R. Green, V. A. Boris, G. A. Sawatzky, A. Schnyder, D. A. Bonn, B. Keimer, and H. Takagi. Electronic structure of the bond disproportionated bismuthate  $\text{Ag}_2\text{BiO}_3$ . *Phys. Rev. Mat.*, 5:064202, 2021.
- [14] M. Kim, G. M. McNally, H.-H. Kim, **M. Oudah**, A. S. Gibbs, P. Manuel, R. J. Green, R. Sutarto, T. Takayama, A. Yaresko, et al. Superconductivity in  $(\text{Ba}, \text{K})\text{SbO}_3$ . *Nat. Mater.*, 21(6):627–633, 2022.
- [15] **M. Oudah**, A. Ikeda, J. N. Hausmann, S. Yonezawa, T. Fukumoto, S. Kobayashi, M. Sato, and Y. Maeno. Superconductivity in the antiperovskite dirac-metal oxide  $\text{Sr}_{3-x}\text{SnO}$ . *Nat. Commun.*, 7:13617, 2016.
- [16] **M. Oudah**, J. N. Hausmann, S. Kitao, A. Ikeda, S. Yonezawa, M. Seto, and Y. Maeno. Evolution of superconductivity with Sr-deficiency in antiperovskite oxide  $\text{Sr}_{3-x}\text{SnO}$ . *Sci. Rep.*, 9:1–9, 2019.
- [17] S. Kitagawa, K. Ishida, **M. Oudah**, J. N. Hausmann, A. Ikeda, S. Yonezawa, and Y. Maeno. Normal-state properties of the antiperovskite oxide  $\text{Sr}_{3-x}\text{SnO}$  revealed by  $^{119}\text{Sn}$ -NMR. *Phys. Rev. B*, 98:100503, 2018.
- [18] A. Ikeda, Z. Guguchia, **M. Oudah**, S. Koibuchi, S. Yonezawa, D. Das, T. Shiroka, H. Luetkens, and Y. Maeno. Penetration depth and gap structure in the antiperovskite oxide superconductor  $\text{Sr}_{3-x}\text{SnO}$  revealed by  $\mu\text{SR}$ . *Phys. Rev. B*, 101:174503, 2020.
- [19] W. MacFarlane, **M. Oudah**, R. McFadden, D. Huang, A. Chatzichristos, D. Fujimoto, V. Karner, R. Kiefl, C. Levy, R. Li, et al.  $^8\text{Li}$   $\beta$ -NMR studies of epitaxial thin films of the 3d topological dirac semimetal  $\text{Sr}_3\text{SnO}$ . In *Journal of Physics: Conference Series*, volume 2462, page 012057. IOP Publishing, 2023.



- [20] S. Klemenz, S. Lei, and L. M. Schoop. Topological Semimetals in Square-Net Materials. *Annu. Rev. Mater. Res.*, 49:185–206, 2019.
- [21] B. Stuart, S. Choi, J. Kim, L. Muechler, R. Queiroz, **M. Oudah**, L. Schoop, D. Bonn, and S. Burke. Quasi-particle interference observation of the topologically nontrivial drumhead surface state in ZrSiTe. *Phys. Rev. B*, 105(12):L121111, 2022.
- [22] **M. Oudah**, J. Bannies, D. Bonn, and M. Aronson. Superconductivity and quantum oscillations in single crystals of the compensated semimetal CaSb<sub>2</sub>. *Phys. Rev. B*, 105(18):184504, 2022.
- [23] **M. Oudah**, Y. Cai, M. D. T. Sanchez, J. Bannies, M. Aronson, K. Kojima, and D. Bonn. Time-reversal symmetry breaking superconductivity in CaSb<sub>2</sub>. *Phys. Rev. B*, 110(13):134524, 2024.
- [24] B. Sales, D. Mandrus, and R. K. Williams. Filled skutterudite antimonides: a new class of thermoelectric materials. *Science*, 272:1325–1328, 1996.
- [25] J. C. Teo, L. Fu, and C. Kane. Surface states and topological invariants in three-dimensional topological insulators: Application to Bi<sub>1-x</sub>Sb<sub>x</sub>. *Phys. Rev. B*, 78:045426, 2008.
- [26] **M. Oudah**, H.-H. Kung, S. Sahu, N. Heinsdorf, A. Schulz, K. Philippi, M.-V. De Toro Sanchez, Y. Cai, K. Kojima, A. P. Schnyder, et al. Discovery of superconductivity and electron-phonon drag in the non-centrosymmetric weyl semimetal LaRhGe<sub>3</sub>. *npj Quantum Mater.*, 9(1):88, 2024.
- [27] G. H. Johnstone, M. U. González-Rivas, K. M. Taddei, R. Sutarto, G. A. Sawatzky, R. J. Green, **M. Oudah**, and A. M. Hallas. Entropy engineering and tunable magnetic order in the spinel high-entropy oxide. *J. Am. Chem. Soc.*, 144(45):20590, 2022.
- [28] S. S. Aamlid, **M. Oudah**, J. Rottler, and A. M. Hallas. Understanding the role of entropy in high entropy oxides. *J. Am. Chem. Soc.*, 145(11):5991–6006, 2023.
- [29] S. S. Aamlid, G. H. Johnstone, S. Mugiraneza, **M. Oudah**, J. Rottler, and A. M. Hallas. Phase stability of entropy stabilized oxides with the  $\alpha$ -PbO<sub>2</sub> structure. *Commun. Mater.*, 4(1):45, 2023.
- [30] S. S. Aamlid, M. Kim, M. U. González-Rivas, **M. Oudah**, H. Takagi, and A. M. Hallas. Effect of high pressure synthesis conditions on the formation of high entropy oxides. *Appl. Phys. Lett.*, 125(2), 2024.
- [31] **M. Oudah**, D. Takegami, S. Kitao, J. Lado, A. Meléndez-Sans, D. Christovam, M. Yoshimura, K. Tsuei, G. McNally, M. Isobe, K. Küster, M. Seto, B. Keimer, D. Bonn, H. Tjeng, G. Sawatzky, and H. Takagi. Charge-entropy-stabilized selenide Ag<sub>x</sub>Sn<sub>1-x</sub>Se. *Submitted to Commun. Mater.*, 2024.
- [32] G. Baker, T. W. Branch, J. Bobowski, J. Day, D. Valentinis, **M. Oudah**, P. McGuinness, S. Khim, P. Surówka, Y. Maeno, et al. Nonlocal electrodynamics in ultrapure PdCoO<sub>2</sub>. *Phys. Rev. X*, 14(1):011018, 2024.
- [33] W. Grochala. Silverland: the realm of compounds of divalent silver-and why they are interesting. *J. Supercond. Nov. Magn.*, 31(3):737–752, 2018.
- [34] G. A. Wieggers. Misfit layer compounds: Structures and physical properties. *Prog. Solid State Chem.*, 24:1–139, 1996.
- [35] D. R. Merrill, D. B. Moore, S. R. Bauers, M. Falmbigl, and D. C. Johnson. Misfit layer compounds and ferecrystals: Model systems for thermoelectric nanocomposites. *Materials*, 8(4):2000–2029, 2015.
- [36] W. Li, L. Cao, J. Zhao, R. Yu, J. Zhang, Y. Liu, Q. Liu, G. Zhao, X. Wang, Z. Hu, et al. A new superconductor of cuprates with unique features. *arXiv:1808.09425*, 2018.
- [37] N. Khasanova, A. Yamamoto, S. Tajima, X.-J. Wu, and K. Tanabe. Superconductivity at 10.2 K in the K–Bi–O system. *Phys. C: Supercond. Appl.*, 305:275–280, 1998.
- [38] M. Zastrow. Meet the crystal growers who sparked a revolution in graphene electronics. *Nature*, 572(7768):429–433, 2019.