New Quantum Materials:
The Search for 2-Dimensional 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 condensed matter 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.

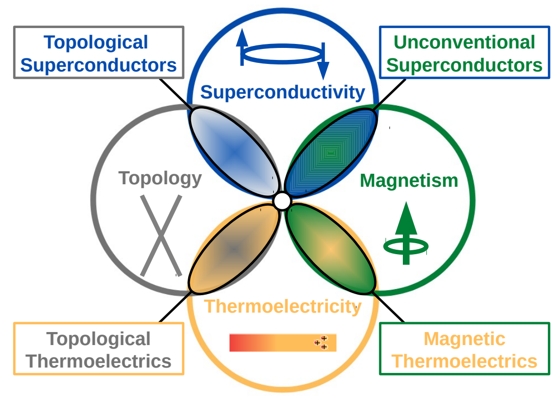

Figure 1: Various properties of quantum materials, and emergent quantum states at the overlap.

Topological thermoelectrics are those materials with topologically non-trivial electronic 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 Bi_2Se_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 $Cu_xBi_2Se_3$ [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.

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 identified a new topological Weyl semimetal $\mathbf{Ag_2BiO_3}$ using high-pressure high-temperature methods [12]. We highlight the similarity between Ag_2BiO_3 (space group $Pnn2$) and the perovskite type bismuthate $BaBiO_3$ (space group $I2/m$), both with nominal "Bi^{4+}" state that disproportionates into two distinct sites. Suppression of the disproportionation in Ag_2BiO_3 (space group $Pnna$) will stabilize a Dirac semimetal state and can even lead to superconductivity. At UBC, we have grown **high-quality superconducting cuprate and Fe(Se,Te) crystals using flux method and (Ba,K)BiO_3 crystals using electrochemical growth**, and helped my collaborators measure x-ray absorption of superconducting (Ba,K)SbO_3 synthesized at high-pressure [13].

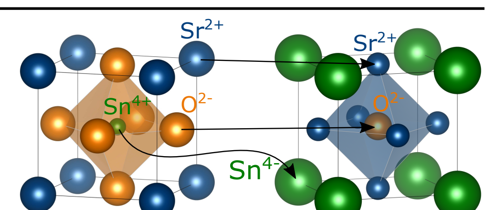

Figure 2: Cyrstal structures of $SrSnO_3$ (perovskite oxide, left) and Sr_3SnO (antiperovskite oxide, right), highlighting ionic states of atoms. The Sn^{4-} state makes Sr_3SnO a topological Dirac-semimetal.

In Kyoto, I found superconductivity in the highly air-sensitive $\mathbf{Sr_{3-x}SnO}$ [14] containing the unusual Sn^{4-} state revealed in Mößbauer spectroscopy [15], crystal structure of the antiperovskite oxide Sr_3SnO and typical perovskite oxide $SrSnO_3$ are shown in Fig. 2. We observed in bulk $Sr_{3-x}SnO$ evidence for **Dirac electrons** using NMR [16] and signs of **unconventional superconductivity** in muon spin relaxation (μSR) [17], then studied thin-films of Sr_3SnO using β-NMR in search of chiral surface states [18]. Recently, we have shown using Mößbauer spectroscopy an unusual "Sn^{3+}" state at low temperatures in superconducting selenide $AgSnSe_2$ (space group $Fm3m$), which arises from 2+/4+ fluctuations on Sn [19] as revealed by X-ray photoelectron spectroscopy (XPS).

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 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** $M\mathbf{Sb_2}$ ($M = \mathrm{Ca, Sr, Eu, Yb}$, crystal structure shown in Fig. 3) with distorted square-nets. We have characterized the semimetallicity and superconductivity of $CaSb_2$ [22], and studied the superconductivity using μSR [23]. Antimonides have promising thermoelectric properties [24] and host topological states [25]. I will study the $M\mathrm{Sb_2}$ family of materials with the aim of achieving **topological thermoelectricity**.

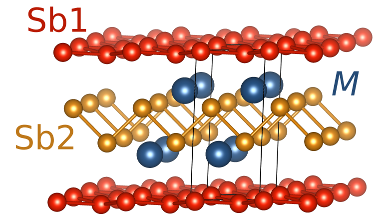

Figure 3: Crystal structure of $M\mathrm{Sb_2}$ highlighting the two different Sb chains. Sb1 chains form a distorted square-net.

Recently, I discovered superconductivity and electron-phonon drag in non-centrosymmetric semimetal $LaRhGe_3$ [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], where I consider the role of charge-entropy in the stability of rocksalt selenides [19]. Furthermore, we work on materials with high-mobility of electrons, where electron-electron interactions are dominant, resulting in viscous/ballistic electron flow, including recent work on the oxides ReO_3 and $PdCoO_2$ [30]. 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 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 inlcude $M = \mathrm{Sn, Pb, Sb, Bi, La, Ce, Sm, Tb}$, $T = \mathrm{Ti, V, Nb, Ta, Cr}$, and $X = \mathrm{S, Se}$ [31, 32], 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. We will expand on the possible combination of elements on the M and T sites by utilizing high-pressure synthesis.

I will investigate the thickness dependence of thermoelectric and superconducting properties in 2D chalcogenides. Specifically, I will explore how the superconducting transition temperature and phonon energies of $NbSe_2$, grown by Metal-Organic Chemical Vapor Deposition (MOCVD), vary with thickness. These results will be compared to those of exfoliated $NbSe_2$ single crystals, which are synthesized in my lab. Additionally, I will study the doping and thickness dependence of the thermoelectric properties of Sb_2Te_3, also grown by MOCVD, with the goal of identifying methods to tune the electron-phonon coupling for optimized thermoelectric efficiency.

I will investigate topolgical superconductivity and thermoelectricity in the $M\mathrm{Sb_2}$ family, focusing on their topological properties in relation to Sb-Sb distortions. In these antimonides, the distortion of Sb-Sb bonds into zigzag chains leads to non-symmorphic space group in the crystal structure, and the emergence of nodal-line states. My work will focus on the superconducting pairing mechanism and identifying any connection to topological properties in $M\mathrm{Sb_2}$. Additionally, I will examine how the presence of these nodal-line states contribute to enhanced thermoelectric efficiency. The effect of high-pressure synthesis on the symmetry of these antimonides will be studied.

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, μSR, 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, and utilize MOCVD for growth of 2D materials.**

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 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 Ba_2CuO_4 [33] and $KBiO_3$ [34]. **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 limited our understanding of such high-pressure materials.

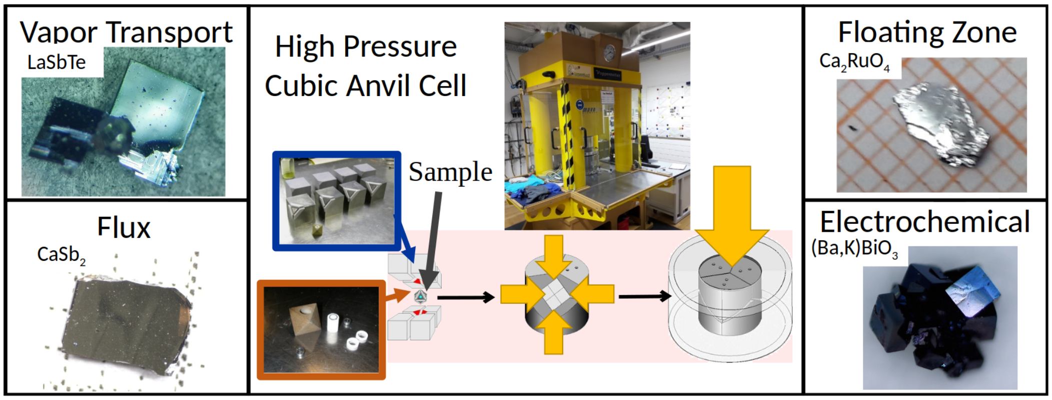

Figure 4: Crystals I have grown: Topological crystalline insulator, LaSbTe, vapor transport (top left), Superconducting Dirac nodal-line semimetal, $CaSb_2$, self-flux (bottom left), Mott Insulator, Ca_2RuO_4, floating zone (top right), unconventional superconductor, $(Ba,K)BiO_3$, electrochemical (bottom right), and high-pressure synthesis setup with cubic-anvil cell at MPI-Stuttgart (center).

However, recently some progress was made in integrating crystal growth techniques into high-pressure synthesis, and this is demonstrated with work on hexagonal boron nitride crystals [35]. Utilizing recent technical advancements in anvil cell design [35], 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. 4) 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^3. 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, highlighted in Fig. 4, and utilize MOCVD for 2D materials.

Synergy with the University of Ottawa (UOttawa)

I look forward to collaborations with members of the Department of Physics at UOttawa working on theory and experiments involving quantum materials and devices. Bulk crystal growth techniques and high-pressure apparatus I propose to build will enable new collaborations with faculty in the area of materials and photonics. In particular, I envision fruitful projects with with faculty working on materials under extreme pressure, topological materials, sensing devices, and non-linear optics. Besides funding allocated for the Tier 2 Chair position, I plan to apply for Canada Foundation for Innovation and NSERC funding to support my research.

My research will expand on the activities at the Department of Physics, and I will benefit from the available facilities at the UOttawa, including its Nexus for Quantum Technologies Institute. For bulk synthesis, I will utilize synthesis equipment like box furnaces, tube furnaces, quartz sealing station, and glovebox. As my lab expands, it will include a floating zone furnace, arcmelter, and high-pressure synthesis machine; 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 μSR and X-ray spectroscopy experiments. My lab will provide research groups at the UOttawa with high-quality single crystal samples.

To complement the work at the UOttawa, I plan to maintain my collaborations at CLS (XPS, XAS), TRIUMF (μSR, β-NMR), University of British Columbia (STM, ARPES, Theory), Kyoto University (NMR, Mößbauer spectroscopy), University of Tokyo (high-pressure/high-field measurements), Stanford University (scanning SQUID), ETH Zurich (Theory), ESPCI Paris (thermal transport) and MPI-Stuttgart/Dresden (Raman and high-energy X-ray Spectroscopy). Finally, by building on my robust background in quantum materials, I hope to expand on the experimental quantum materials research at UOttawa and bring it to new heights.

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 $BaCu_{5.9}STe_6$ and $BaCu_{5.9}SeTe_6$. *Inorg. Chem.*, 54:845–849, 2015.

[6] P. Jafarzadeh, **M. Oudah**, A. Assoud, N. Farahi, E. Müller, and H. Kleinke. High thermoelectric performance of $Ba_3Cu_{16-x}(S,Te)_{11}$. *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 Bi_2Se_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 $Cu_xBi_2Se_3$ and its implications for pairing in the undoped topological insulator. *Phys. Rev. Lett.*, 104:057001, 2010.

[12] **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 Ag_2BiO_3. *Phys. Rev. Mat.*, 5:064202, 2021.

[13] 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 $(Ba,K)SbO_3$. *Nat. Mater.*, 21(6):627–633, 2022.

[14] **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 $Sr_{3-x}SnO$. *Nat. Commun.*, 7:13617, 2016.

[15] **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 $Sr_{3-x}SnO$. *Sci. Rep.*, 9:1–9, 2019.

[16] S. Kitagawa, K. Ishida, **M. Oudah**, J. N. Hausmann, A. Ikeda, S. Yonezawa, and Y. Maeno. Normal-state properties of the antiperovskite oxide $Sr_{3-x}SnO$ revealed by ^{119}Sn-NMR. *Phys. Rev. B*, 98:100503, 2018.

[17] 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 $Sr_{3-x}SnO$ revealed by μSR. *Phys. Rev. B*, 101:174503, 2020.

[18] W. MacFarlane, **M. Oudah**, R. McFadden, D. Huang, A. Chatzichristos, D. Fujimoto, V. Karner, R. Kiefl, C. Levy, R. Li, et al. ^{8}Li β-NMR studies of epitaxial thin films of the 3d topological dirac semimetal Sr_3SnO. In *Journal of Physics: Conference Series*, volume 2462, page 012057. IOP Publishing, 2023.

[19] **M. Oudah**, D. Takegami, S. Kitao, J. Lado, A. Meléndez-Sans, D. S. Christovam, M. Yoshimura, K.-D. Tsuei, G. McNally, M. Isobe, K. Küster, M. Seto, B. Keimer, D. Bonn, L. H. Tjeng, G. Sawatzky, and H. Takagi. Future material demand for automotive lithium-based batteries. *Commun. Mater.*, 6:58, 2025.

[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. Quasiparticle 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 $CaSb_2$. *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 $CaSb_2$. *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 $Bi_{1-x}Sb_x$. *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 $LaRhGe_3$. *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, G. H. Johnstone, S. Mugiraneza, **M. Oudah**, J. Rottler, and A. M. Hallas. Phase stability of entropy stabilized oxides with the α-PbO_2 structure. *Commun. Mater.*, 4(1):45, 2023.

[29] 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.

[30] 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 $PdCoO_2$. *Phys. Rev. X*, 14(1):011018, 2024.

[31] G. A. Wiegers. Misfit layer compounds: Structures and physical properties. *Prog. Solid State Chem.*, 24:1–139, 1996.

[32] 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.

[33] 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.

[34] 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.

[35] M. Zastrow. Meet the crystal growers who sparked a revolution in graphene electronics. *Nature*, 572(7768):429–433, 2019.