New Quantum Materials: 2D Topological Thermoelectrics and Superconductors

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 electrical and thermal transport characterization of 2D crystalline solids with emergent quantum phases.** My philosophy is centered around finding connections between different subfields in quantum materials research to identify new ground for exploration that will motivate future fundamental research with relevance to technology. My background working in transport measurements and 2D materials matches the current interests of the van der Waals-Zeeman Institute with its long history.

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 in 2D materials [1, 2], and thermoelectric performance can be enhanced with short-range magnetic order [3]. 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. The overlap of topologically non-trivial states with superconductivity and thermoelectricity is of particular interest.

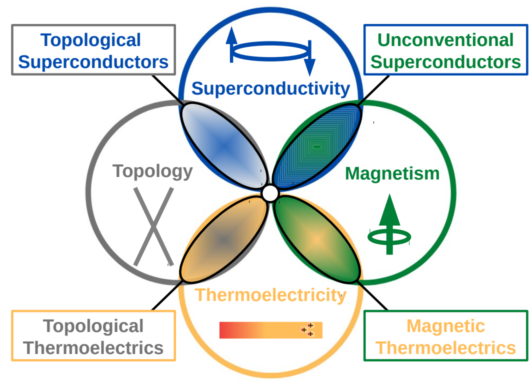

Figure 1: Various properties of quantum materials, and emergent quantum states at the overlap.

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 Bi_2Se_3 [7]. Tuning of carrier concentration through atomic substitution clarifies the contribution of surface 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]. A path towards topological superconductivity is doping topological insulators, such as the case for $Cu_xBi_2Se_3$ [11]. Unfortunately, the material suffers from low superconducting volume fraction due to inhomogeneity of the intercalated Cu [11]. To overcome these challenges we need new families of topological materials where we can grow high-quality single crystals and study their properties using low temperature electrical and thermal transport measurements.

I have worked on synthesis and low temperature transport of many materials, including chalcogenides, antiperovskite oxides, 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. In these materials I have measured the electrical transport under high magnetic field, Seebeck coefficient, and thermal conductivity. Besides transport measurements, I developed a deep understanding of the materials by utilizing heat capacity, magnetization, muon spin-relaxation (μSR), and various types of spectrsocopy, including X-ray photoelectron spectroscoy, X-ray absorption, resonant x-ray scattering, and Mößbauer. I identified a new phase of $AgSbO_3$ with strong Sb-O hybridization [12] and identified a new topological Weyl semimetal Ag_2BiO_3 [13]. At UBC, I have grown **high-quality superconducting cuprate and Fe(Se,Te) crystals for collaborative work**, and measured x-ray absorption of superconducting (Ba,K)SbO_3 for collaborators in Germany [14]. In Kyoto, I found superconductivity in the highly air-sensitive **$Sr_{3-x}SnO$** [15] containing the unusual Sn^{4-} [16]. We observed in bulk $Sr_{3-x}SnO$ evidence for **Dirac electrons** using NMR [17] and signs of **unconventional superconductivity** in muon spin relaxation (μSR) [18], then studied thin-films of Sr_3SnO using β-NMR in search of chiral surface states [19].

Topological properties have been realized in the class of materials with **2D square-nets**, and we have demonstrated a non-trivial drumhead surface state in ZrSiTe [20]. I now work on the quasi-2D **metal antimonides MSb$_2$** ($M = \mathrm{Ca, Sr, Eu, Yb}$ with distorted square-nets. We have characterized the semimetallicity and superconductivity of

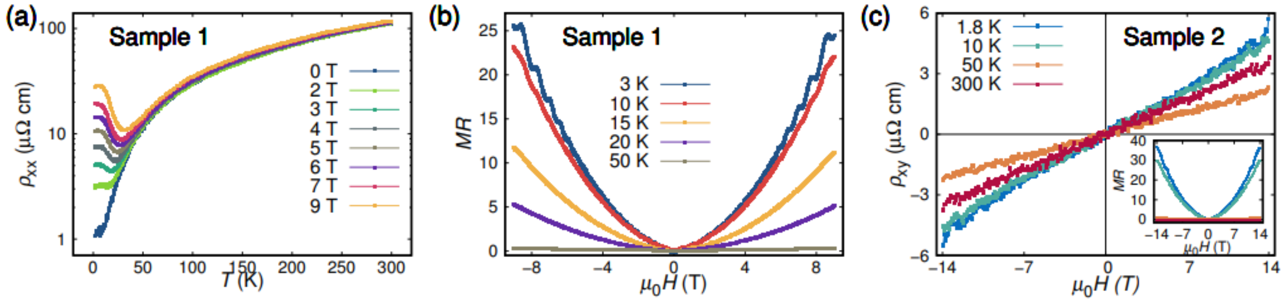

Figure 2: (a) Temperature dependence of the longitudinal resistivity ρ_{xx} for different magnetic fields $\mathrm{H} \parallel c*$ with $\mathrm{I} \perp c*$ of $CaSb_2$ (Sample 1). (b) Magnetic field dependence of the resistance (MR) with $\mathrm{H} \parallel c*$ of $CaSb_2$ (Sample 1). (c) MR and Hall resistivity with $\mathrm{H} \parallel c*$ of another $CaSb_2$ crystal (Sample 2).

$CaSb_2$ [21] (data shown in Fig. 2), and studied the superconductivity using μSR [22]. Antimonides have promising thermoelectric properties [23] and host topological states [24]. I plan to study the MSb$_2$ family of materials to elucidate their potential for **topological thermoelectricity**. For collaborative work on spectroscopy and tunneling experiments I have grown other 2D materials, including $NbIrTe_4$, WTe_2, WSe_2, and MoS_2.

I discovered superconductivity and electron-phonon drag, through low temperature transport and Raman spectrosocpy measurements, in non-centrosymmetric semimetal $LaRhGe_3$ [25]. We have also been working on high-entropy materials, studying the properties of magnetic spinel oxides [26]. This emergent field offers a new perspective on the tunability of stability and properties of materials using entropy [27, 28, 29]. My most recent work demonstrates a novel concept of charge-entropy stabilization in the selenide (Ag,Sn)Se, with 2+/4+ fluctuations on Sn, by utilizing Mößbauer spectroscopy and X-ray photoelectron spectroscopy [30]. We have expanded our work to include high-entropy van der Waals materials, where various electronic properties can be realized [31]. I develop a holistic skill-set by studying many quantum materials, which will enable me to find new emergent quantum phases in 2D materials.

At Amsterdam, I will explore the superconducting 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 with various combination of elements [32, 33] 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. We will further expand the work to include the twisting of exfoliated layers, which has proven a powerful knob for tuning the properties of 2D materials by varying the twist angle the resulting Moiré lattice [34, 35].

I will investigate topological superconductivity and thermoelectricity in the 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 MSb$_2$, then collaboratively study the scattering mechanisms of quasiparticles [36]. Although quasi-2D with strong bonding between the layers, these materials can be exfoliated [37].

I look forward to collaborations with members of the Quantum Materials Cluster at the University of Amsterdam (UofA) and the wider network in the Netherlands. Bulk crystal growth techniques and transport measurement capabilities I propose to build will enable new collaborations with faculty in the area of k-space spectroscopy, magnetism, interfaces, and AC conductivity. My research will benefit from the available facilities at UofA for bulk synthesis and low temperature transport. I will complement the activity at the department with my expertise in the measurement and analysis of heat capacity, magnetic susceptibility μSR, neutron diffraction, and X-ray spectroscopy experiments.

My funding strategy will include applying for funds from the Dutch Research Council (NWO) and Royan Netherlands Academy of Arts and Sciences (KNAW) for instrumentation and salaries. I will apply for funding by the European Commission that supports quantum science, particularly ones that support exchanges with Canada and Japan. I will consult experienced members of the department for help in identifying other funding sources and for collaboration when applying to larger grants. To complement the work at UofA, 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 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 2D quantum materials research at UofA and develop new strong collaborations.

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