Emergent QUAntum materials and TEchnologies (EQUATE)
NSF EPSCoR RII Track-1 Project #OIA-2044049

Year 2 Narrative Report – February 2023
Nebraska EPSCoR

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EXECUTIVE SUMMARY

The EQUATE team of researchers, collaborators, advisors, partners, administrators, and coordinators for education and outreach, has been working diligently to advance in accordance with the project's NSF-approved strategic plan. The NSF-approved update of the strategic plan incorporates changes in the scientific personnel which occurred due to the untimely passing of senior investigator Ralph Skomski and the successful hire of the new senior investigator Aleksander Wysocki at the University of Nebraska at Kearney (UNK). Skomski's vacant position within Focused Research Group (FRG) 1 has been filled by quantum material scientist and UNL assistant professor Robert Streubel, after a consensus vote by the EQUATE Management Team. This report highlights major achievements of Year 2 including how the new science investigators became integrated and contributed to EQUATE since they joined the project.

Building on the results of the first year, the progress made by EQUATE within Year 2 strengthened its position as a leading center in quantum materials science and technologies. EQUATE drives the second quantum revolution with increasing intensity through discoveries, innovation, and workforce development. The Year 2 results demonstrate the effectiveness of the center's interdisciplinary multi-campus-based collaborative approach. The team strives for constant improvement by implementing critical feedback.

Competent criticism was obtained from EQUATE's External Review Panel (ERP), among other sources. The ERP assembled August 17-18, 2022 for an in-person review at the University of Nebraska-Lincoln (UNL) with active participation of the entire EQUATE team from the director to undergraduate students. ERP members included panel chair Dr. Axel Hoffmann (panel chair), University of Illinois at Urbana-Champaign; Dr. Chris J. Palmstrøm, University of California Santa Barbara; Dr. Jeanie (ChunNing) Lau, Ohio State University; and Admiral (USN ret.) Osie Combs, President, Pacific Engineering. As a result of the interaction with the review panel, the EQUATE team intensified the interaction between FRGs on all levels. Tangible measures include a biweekly inter-FRG meeting which includes all EQUATE researchers. The inter-FRG meetings together with EQUATE's monthly seminars are archived and documented on the EQUATE website (https://equate.unl.edu/welcome). The disseminated Year 2 collaborative outcomes, and ongoing projects with collaborations across FRGs, are a testimony to the success of this approach.

FRG1's focus on quantum materials reached multiple milestones in Year 2 and brought new breakthroughs in fundamental quantum materials science with applications in quantum and spintronic devices based on quantum materials. Achievements include highly collaborative work within and across FRGs and between experimentalists and theorists, e.g., on magnetoelectric antiferromagentic oxides where Néel vector rotation by pure electric means, i.e., in the absence of magnetic fields and dissipative electric currents has been demonstrated. In collaboration with NIST, the team was able to demonstrate that temperature treatment can vastly amplify the possibilities for ultra-low power and ultra-fast voltage-controlled antiferromagnetic spintronics. The same thin film materials also serve as functional platforms for voltage-controlled proximity effects. Theoretical modeling supports the notion of quantum interference phenomena of spin currents in proximate graphene with possible applications in new transistor architectures. Two dimensional materials are not only investigated in proximity geometry but also as standalone antiferromagnetic van der Waals materials. The emergent properties of two simultaneously present quantum phenomena are theoretically investigated in systems where switchable polarization of a ferroelectric material controls the topologically protected spin arrangement of magnetic skyrmions. New development of instrumentation by EQUATE scientists led to new findings at the experimental forefront. Examples include the FRG 1 development of an optical method to measure the elastic hysteresis curve of stress-induced domain wall movement which, together with the center's theory support, allowed to unravel universality in the behavior of ferroelastic domain walls.

In FRG2, the implementation of a new solid-state driven fast-frequency sweeping THz source helps advancing the investigation of solid-state qubits in ultra-wide band gap semiconductors. The nitrogen vacancy (NV) based magnetometry of FRG2 impacts multiple research thrusts within FRG2 and across the FRGs. For example, in Year 2, the NV quantum sensor setup for low field magnetic resonance spectroscopy developed in FRG2 was utilized to investigate individual spin-crossover molecule rods fabricated in FRG1. NV microscopy was used to image boundary magnetization in antiferromagnetic magnetoelectric thin films grown via pulsed laser deposition in FRG1. Collaboration between experiment and theory within FRG2 enabled new ultrathin nanocomposite nonlinear nanostructures substantially enhancing the efficiency in their nonlinear optical properties including up and down conversion. A setup to measure spontaneous parametric down-conversion of two-photon entangled pairs complements the fabrication of nonlinear nanostructures. In addition, plasmonic nanopatch antennas are designed and modeled which enable enhanced second-harmonic generation from individual antennas. These nanophotonics achievements have impact on quantum communication technologies based on photons as flying qubits.
FRG3 saw multiple breakthroughs in Year 2 as well.

Among the top achievements of FRG3, and also EQUATE as a center, is the work on solid state quantum emulation aided by halide perovskites. It allows for Bose-Einstein condensates in exciton-polariton systems with application in quantum emulation and the study of quantum fluids. The condensed matter Bose-Einstein condensates are complemented by experimental cold-atom efforts and successful operation of a stable sub-Doppler magneto-optical trap reaching temperatures < 80 micro Kelvin. The cold atom experiments are accompanied by theoretical work on static impurities placed into trapped dipolar Bose gases. Collaborative work between theory and experiment takes place across the FRGs in the context of cross-road quantum dot systems for potential high-temperature qubit networks. Similarly, new qubit platforms from magnetic molecules and magnetic adatom systems which represent an efficient quantum spin-light interface are investigated by EQUATE's newest senior investigator, Aleksander Wysocki: a faculty hire as part of UNK's institutional commitment to this EPSCoR project. A second search, at UNL, for an experimental quantum materials scientist bridging work in FRG1 and FRG2 started in October 2021 but no candidate could be hired. The search continues via the UNL Department of Physics and Astronomy, with the help of a new search committee until the position is filled with the ideal candidate.

Education and Outreach (E/O) as well as workforce development are arguably EQUATE's flagship achievements. Of key interest are the multiple all-EQUATE encompassing events. After Year 1's first EQUATE-organized Nebraska Research and Innovation Conference (NRIC) on April 14, 2022-- "Commercializing quantum technologies in Nebraska: From Research to Licensing"-- prepared EQUATE SIs and researchers for potential technology commercialization of the quantum technologies they invent during the center's tenure. The 2023 NRIC conference on "Topology and Valley-Driven Quantum Phenomena" (https://nric.nebraska.edu/) is set for March 17, with leadership from FRG1; this event features seven internationally-renowned experts and participation of all EQUATE researchers. EQUATE's seminar series (https://equate.unl.edu/seminars) is quite successful with a mix of EQUATE presenters and high-profile guest speakers such as quantum ambassadors from IBM, a quantum computing expert from Google, and guest faculty presenters on applied machine learning and Density Functional Theory (DFT) approaches incorporating machine learning. The seminar series is complemented by the inter-FRG meetings (https://equate.unl.edu/inter-frg) where informal exchange of ideas between FRGs happens.

The EQUATE effort for education in quantum information science and workforce development is continuously ramped up and currently builds on two courses taught. One is a continuation of MATL 492/892: Introduction to Quantum Materials and Technologies, taught by FRG2 leader Abdelghani Laraoui, and the second course (ECEN 491/891-001: Quantum Communication and Quantum Computation) was developed and taught by EQUATE's female seed grant awardee Yannan Laura Wang.

EQUATE's second year saw significant progress in all FRGs in line with the strategic plan. This was achieved despite significant obstacles which include the death of a productive science investigator and financial challenges associated with the ongoing 40-year high in inflationary pressure affecting competitive salaries and stressing the budget allocated to perform the proposed work. Strategies to cope with these challenges include EQUATE's original SWOT analysis which, prior to this center's funding, identified the risks of inflation and its detrimental effects on purchasing power as a potential non-transitory reality.

FRG1 – Quantum Materials (2022-2023)

In Year 2, FRG1 continued to gain understanding and control of a range of emerging quantum phenomena driven by correlation, topology, spin-orbit coupling (SOC), and ferroic switching by combining sample synthesis (SIs **Binek, Hong, Lai, Xu**) and material characterization (**Binek, Dowben, Guo, Hong, Xu**) with theoretical studies (**Kovalev, Streubel, Tsymbal**). The goal of FRG1's Thrust 1 is to realize a range of emergent topology, spin, and correlation phenomena in novel ferroic materials, including topological antiferromagnets, two-dimensional (2D) van der Waals (vdW) magnets, and correlated oxides. Thrust 2 aims at achieving magnetoelectric (ME) and valley control of layered 2D vdW materials. The goal of Thrust 3 is to explore the material design of ferroic molecules and metal-organic frameworks (MOFs) as new platforms for constructing spin-qubit.

For **Thrust 1**, **Objective 1a**, SI Binek collaborated with FRG2 leader and SI Abdelghani Laraoui to apply NV spectroscopy to image boundary magnetization in Cr_2O_3 and boron-doped chromia ($B:Cr_2O_3$). Evidence hinting voltage-induced changes of the domains was observed. This work is recently published in *RSC Advances* (2023). SI **Binek** investigated the effect of B-doping on the properties of Cr_2O_3 thin films in collaboration with NIST. The team showed the effect of annealing on the B-profile within a homogeneously doped film. The cold neutron depth profiling (cNDP) data (Fig. 1) shows, remarkable, annealing increases rather than decreases the B-concentration gradient. This phenomenon is not explainable by mere diffusion, but rather reflecting a thermodynamically driven surface segregation process where homogeneity of the chemical potential is a consequence of a B-concentration gradient. The effect of B-segregation was investigated via the spin Hall magnetoresistance measurements with the help of an adjacent Pt Hall bar, which shows that the increased B-concentration near the surface increases the surface Néel temperature. (meets metrics) This means the operation temperature for $B:Cr_2O_3$ based-spintronic devices can be tuned towards complementary metal-oxide simulator (CMOS) compatibility. The most prominent change for B-doped chromia compared to its pure counterpart is the ability to show non-volatile rotation of the Néel vector in response to an applied electric field without a magnetic field. The finding of boron surface segregation is unexpected but vastly amplifies the possibilities for implementing $B:Cr_2O_3$ as a voltage-controlled antiferromagnetic constituent for constructing heterostructures with voltage-controllable emergent spin-dependent quantum transport.

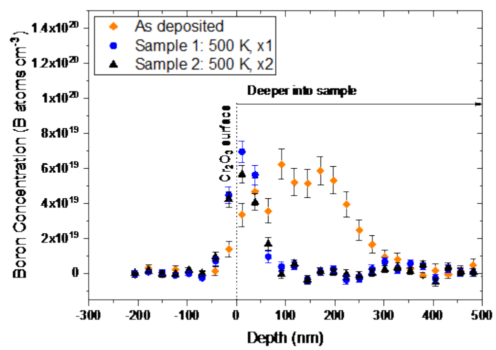

Fig. 1. *B-concentration depth profile via cNDP of the as-deposited (diamonds) and annealed samples (circles and triangles). Depth = 0 nm: $B:Cr_2O_3$/vacuum interface. Depth ≈ 225 nm: $B:Cr_2O_3(0001)/Al_2O_3$ (0001)-substrate interface.*

SIs **Dowben** and **Binek** identified experimentally ferromagnetic spin polarization in the conduction band of palladium (Pd) thin film overlayers induced by interfacial $Cr_2O_3(0001)$ single crystal, especially in the thin limit of Pd (thickness of around 1-4 nm). The spin polarized unoccupied band structure has been measured by spin-polarized inverse photoemission spectroscopy (SPIPES), and the occupied band structure has been mapped via angle-resolved photoemission (Fig. 2b). Spin polarized inverse photoemission spectroscopy studies reveal a significant spin polarization in 10 Å thick Pd films on $Cr_2O_3(0001)$ at 310 K, which is above the Néel temperature of bulk Cr_2O_3. While $Cr_2O_3(0001)$ has surface moments that tend to align along the surface normal, the spin polarization for Pd on Cr_2O_3 contains an in-plane component of about 5% (Fig. 2a). Even though induced polarization in the Pd overlayer on chromia is possible because of the boundary polarization at the chromia in the single domain state, the Pd thin film appears to be ferromagnetic on its own, with the induced spin polarization correlated to strain.

Further evidence for magnetization of Pd on Cr_2O_3 is provided by measuring the Cr_2O_3/Pd(buffer)/[Co/Pd]$_n$ exchange bias systems. The magnitude of the exchange bias field is virtually unaffected by the Pd thickness varing between 1 and 2 nm over a wide temperature range. The extracted Curie temperature for Pd on chromia is above 470 K. (meets metrics) A manuscript based on this work has been submitted to *Journal of Physics Condensed Matter*.

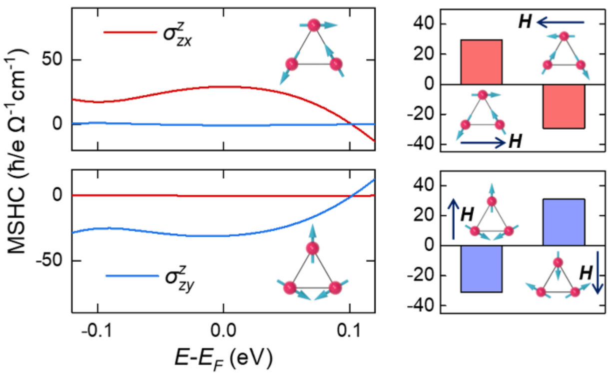

Fig. 2. *(a) Room temperature (RT) spin polarized photoemission spectra of out-of-plane spin polarization and (b) SPIPES for 5 Å Pd on Cr_2O_3, with (bottom) corresponding spin polarization. The blue (red) triangle indicates spin up (down) components. (c) RT occupied band structure for 5 Å Pd on Cr_2O_3. Top: Fermi surface. Bottom: wave vector versus binding energy. Dashed curves: a guide to eye. The arrows indicate the Rashba like spin-orbit splitting of the Pd surface state.*

Two-dimensional (2D) antiferromagnetic van der Waals (vdW) materials such as $CrCl_3$ and $MnBi_2Te_4$ (MBT) are promising material candidates for developing spintronics. SI **Hong** group deposited high quality $CrCl_3$ flakes via the physical vapor transport technique and characterized their magnetotransport properties using tunnel junction device geometry. Figure 3a shows the temperature-dependence of tunneling current taken on a graphite/6-layer $CrCl_3$/graphite tunnel junction, which exhibits pronounced negative tunneling magnetoresistance at low temperature. A kink has been observed at around 17 K in the zero-field curve, indicating the Néel temperature. The results are consistent with the literature reports for mechanically exfoliated $CrCl_3$ samples. Hong group also synthesized MBT flakes on mica substrates via the chemical vapor deposition method using Bi_2Te_3 and MnTe powder as precursors. Figure 3b shows the optical image of as-grown MBT flakes, confirming that the samples prefer horizontal growth and form thin flakes with triangle shapes. Atomic force microscopy (AFM) measurements reveal MBT flakes down to 1.6 nm thickness (Fig. 1c). Raman spectroscopy studies, however, show shifts of the Raman peaks compared with those of exfoliated MBT (composition 2:4), suggesting variation in sample composition. Future studies are required for optimizing the sample stoichiometry.

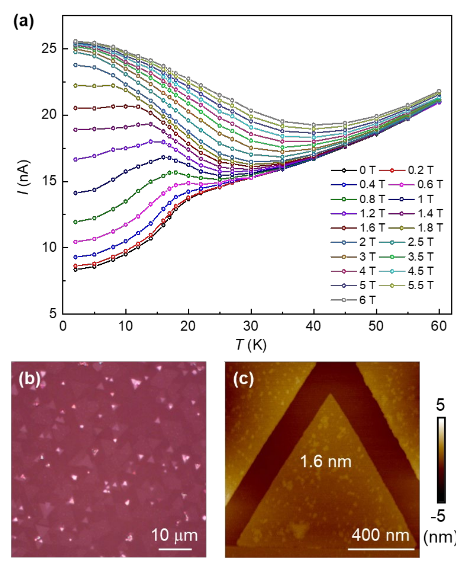

Fig. 3. *(a) Temperature-dependence of tunneling current taken on a $CrCl_3$ tunnel junction at different magnetic fields. (b) Optical and (c) AFM images of MBT flakes on mica substrate.*

Current induced spin-orbit torques driven by the conventional spin Hall effect are widely used to manipulate the magnetization. This approach, however, is nondeterministic and inefficient for the switching of magnets with perpendicular magnetic anisotropy that are demanded by the high-density magnetic storage and memory devices. In collaboration with the experimental group at Tongji University, SI **Tsymbal** demonstrated that efficient perpendicular magnetization switching can be achieved by exploiting a magnetic spin Hall effect in noncollinear antiferromagnets, such as Mn_3Sn. The magnetic group symmetry of Mn_3Sn allows generation of the out-of-plane spin current carrying spin polarization collinear to its direction induced by an in-plane charge current. This spin current drives an out-of-plane anti-damping torque, providing deterministic switching of the perpendicular magnetization of an adjacent Ni/Co multilayer. Due to being odd with respect to time reversal symmetry, the observed magnetic spin Hall effect and the resulting spin-orbit torque can be reversed with reversal of the antiferromagnetic order (Fig. 4). Contrary to the conventional spin-orbit torque devices, the demonstrated magnetization switching does not need an external magnetic field and requires much lower current density. The predicted and experimentally observed magnetic spin Hall effect allows its use as the spin-torque source to engineer novel energy-efficient spintronic devices.

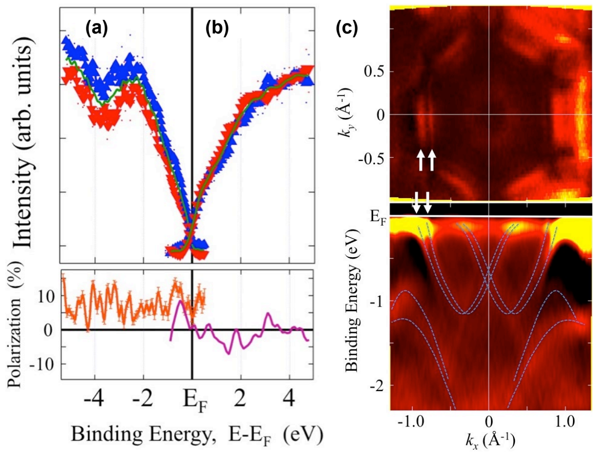

Fig. 4. *Calculated magnetic spin Hall conductivity σ_{zx}^{z} and σ_{zy}^{z} in Mn_3Sn. Left panel shows the σ_{zx}^{z} and σ_{zy}^{z} as a function of energy for AFM1 and AFM2. Right panel shows the sign change of σ_{zx}^{z} and σ_{zy}^{z} when the magnetic moments in the two antiferromagnetic layers are reversed by in-plane magnetic fields.*

For **Thrust 1**, **Objective 1b**, SIs **Dowben, Hong**, and **Streubel**, in collaboration with V. Nagarajan group at University of New South Wales, characterized epitaxial $BiInO_3$ thin films deposited on (Ba,Sr)RuO_3/$SrTiO_3$ (Fig. 5a) using pulsed laser deposition (PVD) and studied the magnetocapacitance in Ni on $BiInO_3$. AFM topography image shows smooth surface of the $BiInO_3$ film (Fig. 5b). (meets metrics) Kelvin probe force microscopy (KPFM) measurements show that the surface potential of the $BiInO_3$ film is typically 0.30–0.38 V higher than that of the Pt–Ir tip with small variations at different locations (Fig. 5c). As shown in Fig. 6, the Ni/$BiInO_3$ interface is a p-type Schottky contact with an obvious asymmetry and pronounced rectification depending on the initial bias voltage. The remanent magnetization in the Ni film leads to a magnetic field-dependent electronic transport and the corresponding magnetocapacitance. A manuscript on this work is currently in preparation.

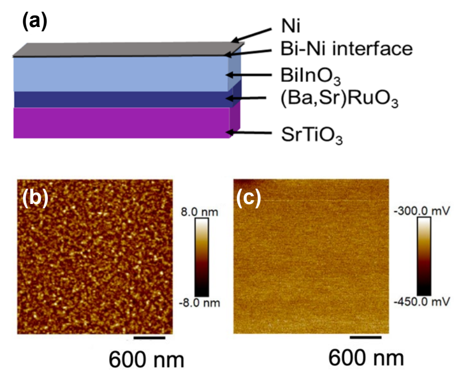

Fig. 5. *(a) Schematic of $BiInO_3$ heterostructure with top Ni adlayer. (b) AFM and (c) KPFM images taken on $BiInO_3$ heterostructures.*

SI **Hong** group developed the process flow for fabricating epitaxial ferroelectric tunnel junctions (FTJs) based on high quality oxide heterostructures composed of $PbZr_{0.2}Ti_{0.8}O_3$ (PZT) tunnel barriers and $LaNiO_3$ (LNO) correlated electrodes, which are deposited using off-axis RF magnetron sputtering.

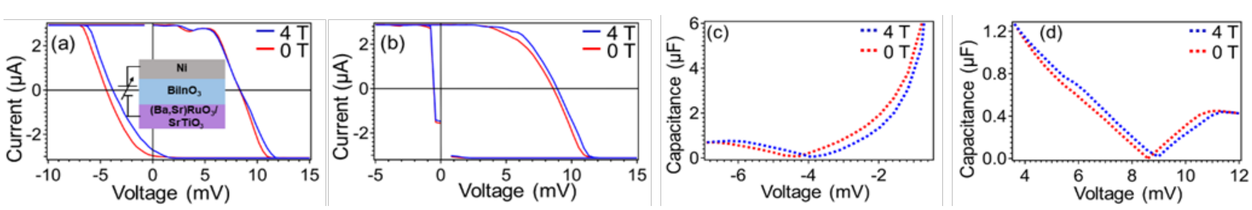

Fig. 6. *(a-b) RT I–V characteristics of a Ni(3 nm)/$BiInO_3$/(Ba,Sr)RuO_3/$SrTiO_3$ heterostructure with (a) increase in voltage (positive loop), and (b) decrease in voltage (negative loop) taken at 0 T and 4 T magnetic field. Inset of (a): device schematic. (c-d) Calculated RT C–V characteristics with (c) increase in voltage (positive loop), and (d) decrease in voltage (negative loop) at 0 T and 4 T.*

As shown in Fig. 7a-f, the heterostructure is first patterned via photolithography followed by Ar plasma etching. After etching, an insulating TiO_2 layer is deposited by RF magnetron sputtering to encapsulate the bottom oxide electrode area. The tunnel junction areas are then defined into the cross-stripe geometry using photolithography followed by Au contact deposition. Figure 7g shows the optical image of 4 FTJ devices fabricated on a 9 nm LNO/4 nm PZT/12 nm LNO heterostructure deposited on (001) $SrTiO_3$ substrate. (meets metrics)

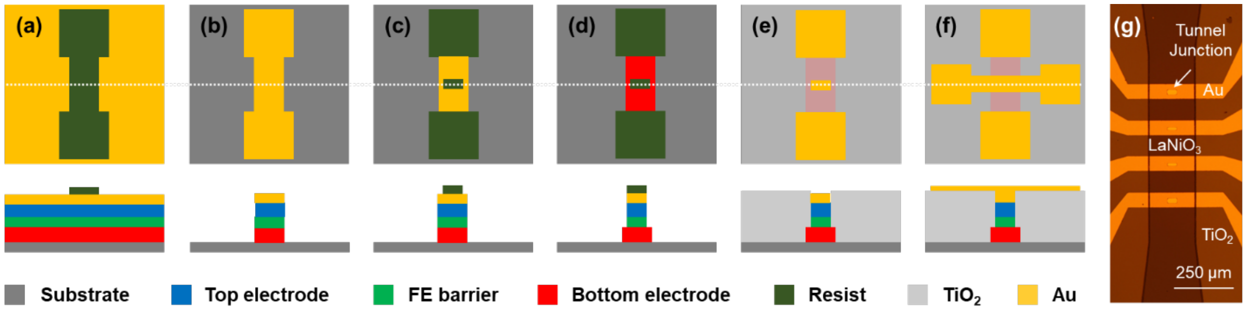

***Fig. 7. (a)-(f)** Process flow for fabricating FTJ. **(g)** Optical image of 4 FTJ devices fabricated on an LNO/PZT/ LNO heterostructure.*

SI **Tsymbal** implemented the linear response methods based on the Kubo formula to compute anomalous and spin Hall conductivities from first principles. First, density functional theory (DFT) calculations are performed using a plane-wave pseudopotential method with the fully relativistic ultrasoft pseudopotentials implemented in Quantum-ESPRESSO. The tight-binding Hamiltonians are then obtained from the maximally localized Wannier functions using the Wannier90 code. Anomalous and spin Hall conductivities were calculated based on the tight-binding parametrization of the band structure. The developed first-principles approach for calculating the anomalous and spin Hall effects will allow the exploration of the transport properties of a broad range of materials relevant for spintronic applications.

SIs **Hong** and **Xu** deposited magnetic oxide thin films using off axis RF magnetron sputtering and PLD to study the magnetic state and spin structure, investigating their dependence on temperature, thickness, and strain. Thin (001) $NiCo_2O_4$ films (1.2 - 50 nm) were grown on (001) $MgAl_2O_4$ (MAO) substrates. Spinel phase and crystallinity were verified using x-ray diffraction (XRD); film thickness was extracted from XRD Laue oscillation around the main Bragg peaks and x-ray reflectivity (XRR) measurements. Magnetometry and anomalous Hall measurements confirmed the perpendicular magnetic anisotropy and revealed the Curie temperature of $NiCo_2O_4$ films. The Curie temperature is about 170 K for 1.5 unit cell films and saturates at about 420 K for films thicker than 30 nm. A paper reporting the magnetotransport properties of ultrathin $NiCo_2O_4$ (001) films is published in *Applied Physics Letters* (2022).

SI **Xu** deposited (111)-oriented $CoFe_2O_4$ thin films of thicknesses from 1.7 to 50 nm on α-Al_2O_3 (0001) substrates. XRD (Fig. 8a) and XRR confirmed the spinel structure and smooth surface and interfaces.

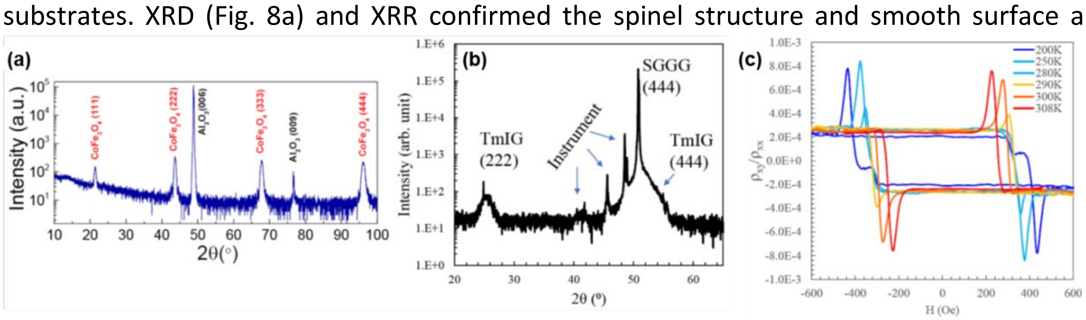

***Fig. 8**. (a) XRD pattern of $CoFe_2O_4$ (111) grown on Al_2O_3 (0001) substrate. (b) XRD pattern of $Tm_3Fe_5O_{12}$ (001) grown on (001) $Gd_{2.6}Ca_{0.4}Ga_{4.1}Mg_{0.25}Zr_{0.65}O_{12}$ (SGGG) substrate. (c) Anomalous Hall effect and topological-Hall-like effect taken on Pt (5 nm)/$NiCo_2O_4$ (15 nm) bilayer structures.*

The in-plane crystal structure was analyzed using the thickness-resolved reflection high energy electron diffraction (RHEED). Interfacial layer of about 1 nm shows larger lattice constants due to epitaxial strain. Also, (001) oriented $Tm_3Fe_5O_{12}$ (TmIG) films 5 – 10 nm were grown on $Gd_3Ga_5O_{12}$ **(**GGG) and $Gd_{2.6}Ca_{0.4}Ga_{4.1}Mg_{0.25}Zr_{0.65}O_{12}$ (SGGG) substrates. XRD (Fig. 8b) pattern confirms the epitaxial growth and shows the garnet structure of the films. The (222) peak is forbidden for the cubic garnet structure and appears in the TmIG films, suggesting that the epitaxial strain lower the cubic symmetry to the tetragonal symmetry. Magnetometry studies indicate perpendicular magnetic anisotropy, potentially due to the tetragonal symmetry.

SI **Xu** studied the effect of the interface between ferrimagnetic $NiCo_2O_4$ and strongly spin-orbit coupled metal Platinum (Pt) on the spin structure and texture of $NiCo_2O_4$. Pt/$NiCo_2O_4$ bilayer structures were prepared on $MgAl_2O_4$ (001) substrates using PLD. As shown in Fig. 8c, the anomalous Hall resistance exhibits square loops expected from $NiCo_2O_4$. In addition, near the coercive field, an asymmetric topological-Hall-like effects (humps) is observed. The magnitude of the humps and the magnetic field where the humps appear change dramatically with temperature. These results suggest complex spin interactions at the $NiCo_2O_4$/Pt interface which may give rise to topological spin texture. (meets metrics)

In collaboration with the industry partner (Vida Products Inc., Rohnert Park, CA 94928), SI **Streubel** synthesized yttrium iron garnet (YIG, $Y_3Fe_5O_{12}$) films using metal-organic decomposition epitaxy. The resulting films are soft-magnetic, low-spin-damping materials that can function as ferromagnetic resonators up to 40 GHz in both in-plane and out-of-plane geometry. The saturation magnetization and magneto-crystalline anisotropy coincide with values for single-crystals and epitaxial films grown with liquid phase epitaxy, PLD, and sputtering.

Theoretical calculations indicate that $NiCo_2O_4$ is a half-metal with fully spin polarized carriers. It is important to ascertain whether $NiCo_2O_4$ is a high polarization material and whether the spin polarization has a projection in the plane of the thin film. SI **Dowben** probed the electronic structure and magnetic properties of the surface layer of a 12 nm $NiCo_2O_4$ film via SPIPES, which is an extremely surface sensitive technique for probing spin majority, spin minority, and spin integrated density of states. The spin majority (minority) density of states is obtained when the spin polarization of incident electron in plane of the sample is parallel (opposite) to the in-plane applied magnetic field. A noticeable difference in spin majority and spin minority intensity was observed in the unoccupied states above Fermi level indicating spin polarized unoccupied states of the $NiCo_2O_4$ thin films. Although the in-plane polarization is estimated to be small, the value is non-zero, which indicates that the magnetic moments have a surface component, and the moments therefore are slightly canted in surface region. $NiCo_2O_4$ strained on $MgAl_2O_4$ possesses perpendicular magnetic anisotropy, and the presence of in-plane spin-resolved unoccupied states indicates the influence of the surface on magnetic ordering. The surface to bulk core level shift of $NiCo_2O_4$ thin films was measured and sample to sample variations were determined.

For **Thrust 1**, **Objective 1c**, SIs **Binek** and Abdelghani **Laraoui** (FRG2) applied NV spectroscopy to image the magnetization switching dynamics in Cr_2O_3 and B:Cr_2O_3. NV imaging of 90 degree switching of surface spin of B:Cr_2O_3 is currently underway, and the metrics will be met on schedule.

SI **Kovalev** studied magnon realizations of bosonic quantum error correcting codes and identified systems that can potentially lead to realizations of quantum states of magnons protected against decoherence. A YIG sphere coupled to photons system is currently being studied. Other candidate systems will also be considered. The metrics will be met on schedule.

For **Thrust 2**, **Objective 2a**, SIs **Binek** and **Dowben**, in collaboration with Jon Bird at the University of Buffalo, observed the signature of quantum interference effects in the nonlocal current below 40 K in graphene on chromia. (meets metrics) It has been shown experimentally that graphene on a magnetic substrate can show the spin Hall effect at room temperature. However, the impurities in the graphene heterostructure affect the transport signatures and the formation of puddles contributes to resistance at the Dirac point. SI **Kovalev** studied theoretically transport in proximized graphene (Fig. 9a) and showed how the local and nonlocal signatures can reveal information about the existence of quantum effects, *i.e.*, the crossover from diffusive to ballistic transport fully accounting for quantum effects. For modeling the disorders, random Gaussian potential with finite correlation length was considered, which allows tuning of the system between different transport regimes encountered in experiments. The Landauer-Buttiker approach was adopted implementing the freely available kwant code, which was modified to include the correlated Gaussian disorder and various proximity effects. The modeling shows that local transport response in the diffusive regime exhibits the universal conductance fluctuations,

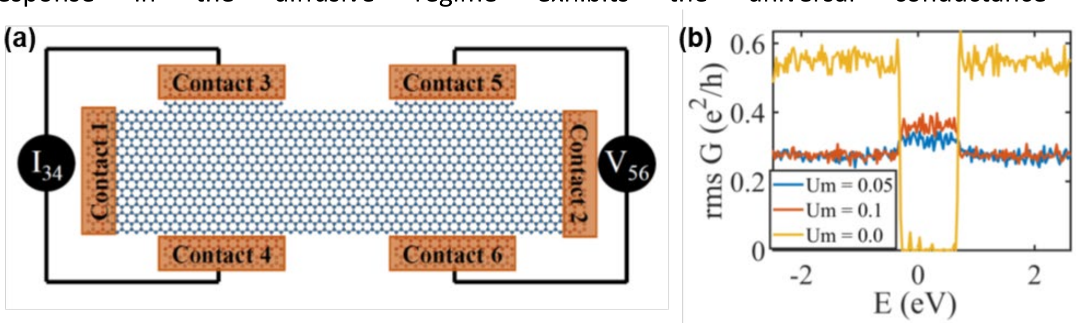

Fig. 9. *(a) Schematic for measuring both local and nonlocal transport in proximized graphene. (b) Simulated magnitude of local conductance fluctuations at different magnetic disorder levels.*

The magnitude of which is affected by the universality of the system (Fig. 9b). In particular, the Rashba spin-orbit interaction and the presence of magnetic disorder can modify the universality of the system and affect the universal conductance fluctuations. A manuscript comparing the modeled nonlocal transport to experimental results for graphene on Chromia is under preparation.

SI **Streubel** established the characterization capability for ferromagnetic resonances in YIG films. While the quality factor for in-plane resonances shows a temperature dependence due to two-magnon scattering, the out-of-plane quality factor is virtually temperature-independent and approaches 1,000 at 40 GHz. (meets metrics) The investigated YIG films possess a > 100 times larger quality factor compared with commercial devices based on CMOS voltage-controlled oscillators. This is equivalent to a 40 dB phase noise advantage for any realistic operation and/or outdoor temperature, *e.g.*, (−75 ~ 100) ºC, at comparable production costs. The inhomogeneous line broadening and, hence, quality factor can be further enhanced using more expensive GGG(111) substrates with superior crystallographic order. This work helps establish broadband ferromagnetic resonance spectroscopy (experimental and numerical) and the paper is currently under peer-review.

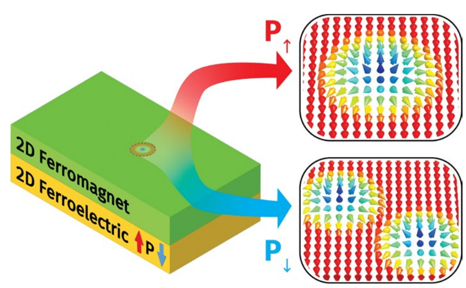

Fig. 10. *Schematic view of ferroelectric control of magnetic skyrmions. Reversal of electric polarization of an adjacent ferroelectric material changes the DMI interaction and alters the appearance of skyrmions in a 2D ferromagnet.*

SI **Tsymbal** investigated theoretically ferroelectric control of magnetic skyrmions in 2D vdW heterostructures and predicted that the DMI and the related skyrmion behaviors in a Fe_3GeTe_2 monolayer can be controlled by ferroelectric polarization of an adjacent 2D vdW ferroelectric In_2Se_3. First-principles density functional theory calculations and micromagnetic modeling showed that ferroelectric polarization of In_2Se_3 produces a sizable DMI in a Fe_3GeTe_2/In_2Se_3 van der Waals heterostructure. The magnitude of DMI can be controlled by the switching of ferroelectric polarization, leading to notable changes in the skyrmion formation (Fig. 10). (meets metrics) Furthermore, the sign of DMI in a $In_2Se_3/Fe_3GeTe_2/In_2Se_3$ multilayer changes with ferroelectric switching, reversing the chirality of the magnetic skyrmions. The predicted electrically controlled skyrmions can be useful for spintronic devices based on the proposed vdW heterostructures.

For **Thrust 2**, **Objective 2b**, SIs **Dowben** and **Tsymbal** investigated the electronic structure of transition metal trichalcogenides (TMTs), the 2D systems with quasi-one-dimensional chains. The elemental contributions to the conduction bands of the TMT TiS_3 and ZrS_3 were examined using X-ray absorption spectroscopy (XAS) at the Ti and S 2*p* edges and the Zr 3*p* edges. DFT theory calculations of TiS_3 and ZrS_3 were carried out to identify the orbital contributions to the density of states. These calculations were correlated with the XAS results, showing that the bottom of the conduction band, for both TiS_3 and ZrS_3, is comprised mainly of hybridized transition-metal-sulfur orbitals, either Ti 3*d* and S 3*p* orbitals or Zr 4*d* and S 3*p* orbitals. Although weak, the experimental results indicate that the S weighted contribution to the conduction band minimum for ZrS_3 is greater than in the case of TiS_3. This work is published in *Journal of Physical Chemistry C* (2022).

The TMTs are less susceptible to undesirable edge defects, which enhances their promise for low-dimensional optical and electronic device applications. Yet the performance of 2D devices based on TMTs have been hampered by contact-related issues. SI **Dowben** group made a diligent effort to both elucidate and summarize the interfacial interactions between gold and various TMTs. X-ray photoemission spectroscopy data, supported by electrical transport measurements, provided insights into the nature of interactions at the Au/In_4Se_3, Au/TiS_3, Au/ZrS_3, Au/HfS_3, and $Au/HfSe_3$ interfaces. This work is published in *Journal of Materials Research* (2022).

It was theoretically predicted that the bilayer 2H-phase MoS_2 and WSe_2 are promising material candidates for realizing the valley spin valve (VSV) effect. SI **Hong** group fabricated graphene/WSe_2 heterostructures via the dry transfer technique for constructing in-plane junctions and VSVs. Ultra large vdW flakes up to mm size were prepared via mechanical exfoliation on plasma treated SiO_2 substrates. Nano-gap patterns were fabricated onto the graphene flakes using e-beam lithography following by reactive ion etching, with gap width down to 100 nm achieved. The nano-fabrication process developed will be adopted to fabricate more complex vdW heterostructures with dual gates for constructing the VSV devices. Theoretical search of 2D materials for realizing nonvolatile VSV is currently underway.

SI **Guo** set up low temperature confocal micro/spectroscopy and visualized the optical contrast of ferroelastic twin domain walls in lead halide perovskites (Fig. 11a). (meets metrics) The study of the optical contrast at the domain wall reveals the wavelength and polarization dependence, linear and nonlinear optical response, as well as universal appearance. Physical models were developed to both understand the optical contrast origin and to produce insight on the microscopic structural basis of the universality of ferroelasticity. An analytical/numerical optical model combining input from DFT calculated

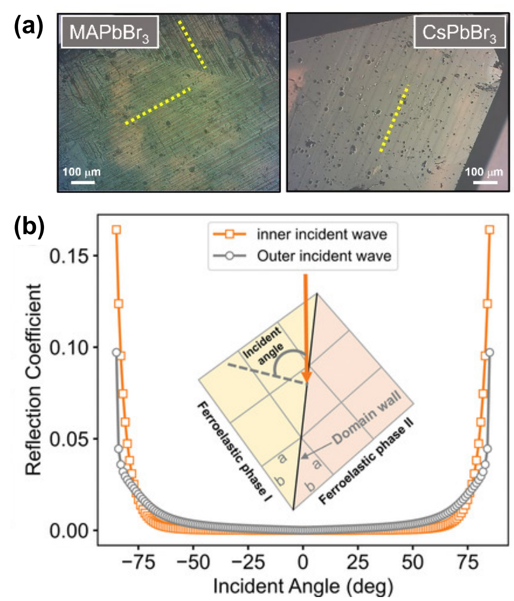

Fig. 11. *(a) Imaging of universal ferroelastic domain walls under non-polarized light. (b) Optical modeling of the reflection coefficients at the twin domain walls as internal optical reflectors. Inset: diagram of the high incident angle imaging condition*.

dielectric functions was implemented to elucidate the direct optical contrast originates from finite optical reflections at the wall interface between two compositionally identical, orientationally different, optically anisotropic domains inside the material bulk (Fig. 11b). Fundamental understanding of the universality of ferroelastic domain walls was gained by correlating experiments with first-principles calculations. Based on the basic knowledge gained, SI Guo also demonstrated an optical method to measure the elastic hysteresis curve of domain wall movement in response to external stress, which can serve as a basis for using the domain walls as nonvolatile, on-off switchable and position-tunable optical reflectors in these emergent semiconductor materials. This work is published in *Advanced Materials* (2022).

SIs **Guo** and **Binek** carried out optical investigations of the structural and magnetic properties of Cr_2O_3 and B:Cr_2O_3 upon voltage poling. The photoluminescence from chromia films showed pronounced spectral changes upon boron doping and voltage poling. The two-magnon mode was identified in boron doped chromia, where voltage poling induced pronounced changes in the position and line-shape of the magnon spectrum upon. (meets metrics) In addition, the laser annealing behavior of B:Cr_2O_3 films potentially provides a foundation of optical patterning of antiferromagnetic devices. In collaboration with SIs **Wysocki** (FRG3), **Mei** (FRG3), and **Sabirianov** (FRG3), the team is developing numerical models and performing first-principles calculations to extract quantitative information on the spin-spin coupling in B:Cr_2O_3.

In spin crossover (SCO) molecular materials, transition metal centers exhibit spin-state transitions that are driven by entropy and structural changes. The SCO transition is promising as a working principle for molecular switches. For **Thrust 3**, **Objective 3**, SIs **Xu** and **Dowben** fabricated SCO molecular crystal thin films on dielectric substrates and performed AFM, electric, and spectroscopy characterizations. Xu deposited polycrystalline $[Fe\{H_2B(pz)_2\}_2(bipy)]$ films (100 - 1000 nm) on 5×5 mm^2 Al_2O_3 substrates (at 300 K) using physical vapor deposition in high vacuum (1.0×10^{-7} Torr), with a growth rate of 0.1 Å/s. The film morphology and thickness calibration was done using AFM; the root mean square roughness was found to be about 50 nm. The C2/c crystal structure for the bulk $[Fe\{H_2B(pz)_2\}_2(bipy)]$ were confirmed in the films using the powder XRD patterns. The subtle structural differences between the high-spin and the low-spin states were also probed by the XRD. SI **Dowben** investigated modifications driven by 7,7,8,8-tetracyanoquinodimethane (TCNQ) to the spin state configuration of $[Fe(3\text{-}bpp)_2](TCNQ)_2$ co-crystal and both spin state and electric conductivity of the mixture of $[Fe\{H_2B(pz)_2\}_2(bipy)]$ and TCNQ. The Fe^{2+} site in the $[Fe(3\text{-}bpp)_2](TCNQ)_2$ co-crystal has a sizable orbital moment. During XAS measurements, the iron ion is partially excited to the high spin state and strong surface effects are indicated. Mixing TCNQ with the $[Fe\{H_2B(pz)_2\}_2(bipy)]$ spin crossover complex leads to a molecular combination with increased conductivity and drift carrier lifetimes. $[Fe\{H_2B(pz)_2\}_2(bipy)]$ thin films with TCNQ, grown using dimethylformamide (DMF), are to great extent locked mainly in the low spin state across a broad temperature range and exhibit drift carrier lifetimes approaching 0.5 s. When deposited onto a ferroelectric polyvinylidene fluoride thin film substrate, $[Fe\{H_2B(pz)_2\}_2(bipy)]$ shows enhanced transistor carrier mobility, likely associated with the increasing cationic character of $[Fe\{H_2B(pz)_2\}_2(bipy)]$ thin films with TCNQ. (meets metrics) The manuscript of this work has been submitted to *Materials Chemistry and Physics*.

Future molecular microelectronics require the electronic conductivity of the device to be tunable without impairing the voltage control electronic properties of the molecules. SIs **Dowben**, **Lai**, and **Streubel**, in collaboration with Talat S. Rahman at the University of Central Florida, investigated the electronic structures and orbital levels of two cobalt-based valence tautomeric systems, $[Co(SQ)(Cat)(4\text{-}CN\text{-}py)_2]/[Co(SQ)_2(4\text{-}CN\text{-}py)_2]$ and $[Co(SQ)(Cat)(3\text{-}tpp)_2]/[Co(SQ)_2(3\text{-}tpp)_2]$, and their dependence on the local environment by interfacing with a semiconducting polyaniline polymer or a polar poly-D-lysine molecular thin film. A metal-ligand unoccupied state has been identified in $[Co(SQ)(Cat)(3\text{-}tpp)_2]/[Co(SQ)_2(3\text{-}tpp)_2]$ through density functional theory and the combination of X-ray photoemission spectroscopy, XAS, and inverse photoemission spectroscopy experiments. This metal-ligand unoccupied state is largely insensitive to the interface between the polymer and spin crossover complexes when the latter is in the high spin state. Alteration of the electronic structure are limited to slight modifications to binding energy. No sizable change in the Co 2*p* absorption spectra is observed corroborating an electronic structure of the central Co atom that is unaffected by the environment. The $[Co(SQ)_2(4\text{-CN-py})_2]$ complex exhibits dynamical effects over a wide range of temperature. The orbital moment, determined by X-ray magnetic circular dichroism (XMCD) with decreasing applied magnetic field, indicates a nonzero critical field for net alignment of magnetic moments, an effect not seen with the spin moment of $[Co(SQ)_2(4\text{-CN-py})_2]$. This work is published in *Chemical Communications* (2022). Comparing DFT calculations with the experimental data reveals a remarkable agreement and indicates the existence of an optically active ligand-to-metal charge transfer (LMCT) state in Co-tpp that are absent near the Fermi level in high-spin state Co-py. The spin-polarized, delocalized LMCT state originates from the hybridization of 3-tpp and Co orbitals and bears great potential for magnetoresistive applications. This work is published in *Nanoscale* as part of the 2023 *Emerging Investigators special issue* (invited contribution).

Applications of quantum information science (QIS) generally rely on the generation and manipulation of qubits. Still, there are ways to envision a device with a continuous readout, but without the entangled states. SI **Dowben** provided an alternative to the qubit, namely the solid-state version of the Mach-Zehnder interferometer, in which the local moments and spin polarization replace light polarization. This work provides some insights into the mathematics that dictates the fundamental working principles of quantum information processes that involve molecular systems with large magnetic anisotropy. (meets metrics) This work is published in *J. Phys. Cond. Matter* (2022). Transistors based on such systems lead to the possibility of fabricating logic gates that do not require entangled states. It further illustrates that novel approaches exist to address the issues pertaining to the scalability of quantum devices, but may face the challenge of finding suitable materials for desired functionality that resemble what is sought from QIS devices.

SI **Lai** carried out the fabrication and characterization of $[Fe(Htrz)_2(trz)](BF_4)$ and $[Fe(Htrz)_2(trz)](BF_4)$/Polyaniline composites. Several triazole-based Fe(II) polymeric complexes have been developed for their applications in SCO devices in the past decades. $[Fe(Htrz)_2(trz)](BF_4)$ is more commonly studied as it shows a well-defined and reversible low spin to high spin transition with hysteresis at temperatures slightly above room temperature (*i.e.*, 330 -380 K) (Fig. 12a-b).

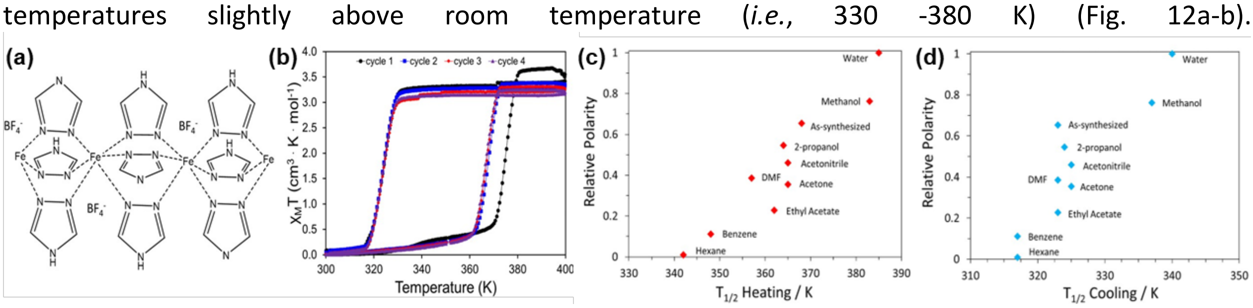

Fig. 12. *(a) Structure of $[Fe(Htrz)_2(trz)](BF_4)$. (b) VSM 4-cycle heating and cooling measurements of $[Fe(Htrz)_2(trz)](BF_4)$ at 2 T from 300 K to 400 K. (c-d) Effects of relative polarity of the introduced solvent on $T_{1/2}$ during VSM (c) heating and (d) cooling cycles.*

This SCO polymer is also sensitive to the presence of solvents, which may result in changes in both the hysteresis width and position. Although researchers have alluded to the effects of solvents on the spin transition temperature ($T_{1/2}$) of $[Fe(Htrz)_2(trz)](BF_4)$, a comprehensive study and more thorough analysis are required to elucidate the effects of solvents of different polarity. Unlike previous investigations focusing on the effects of solvents used during the synthesis of the SCO, **Lai**'s group utilized solvents of different polarity as a post-synthesis "treatment" of $[Fe(Htrz)_2(trz)](BF_4)$ polymer to alter the intermolecular interactions, crystal structure, and packing and compared the SCO behavior of the solvent-exposed samples with the as-synthesized sample. This approach is more convenient and versatile since the polymer can be synthesized under one experimental condition, such as from anhydrous ethanol in this case, and further manipulated by simply exposing the as-synthesized sample to a specific solvent.

Solvents including hexane, benzene, dimethylformamide (DMF), ethyl acetate, acetone, 2-propanol, methanol, and water were selected based on their relative polarity, suitability for device fabrication, and being capable of dispersing $[Fe(Htrz)_2(trz)](BF_4)$ in the presence of a conductive polymer such as polyaniline (PANI). A small amount of the selected solvent was mixed with the as-synthesized sample, and the sample mixture was allowed to dry in the fume hood for up to 3 days. The as-synthesized sample and solvent-exposed samples were then analyzed using the vibrating sample magnetometry (VSM), and $T_{1/2}$ was recorded and compared. Upon analysis of the VSM results for both the heating and cooling cycles, samples treated with higher polarity solvents, such as water and methanol, show higher $T_{1/2}$, whereas samples treated with lower polarity solvents, such as benzene and hexane, show lower $T_{1/2}$ (Fig. 12c-d). These results clearly highlight the effects of post-synthesis solvent treatment, which can change the crystal structure and packing of the SCO polymer and thus the $T_{1/2}$. It is a simple yet effective method to tune spin transitions and can potentially be used with other SCO polymers.

Owing to the favorable SCO properties of $[Fe(Htrz)_2(trz)](BF_4)$, researchers are interested in employing this polymer in the fabrication of nonvolatile voltage controlled spin state switching memory devices. The high resistance of most SCO thin films remains a key impediment to creating a competitive memory device, while adding a conducting polymer such as PANI or polypyrrole (PPy) may lower the on state resistance to below 1 Ω cm. SIs **Lai** and **Dowben** explored the design and fabrication of an $Fe(Htrz)_2(trz)](BF_4)$/PANI composite suitable for device applications. To determine the majority carrier and drift carrier lifetimes in this SCO/conducting polymer composite, the current voltage and capacitance voltage characteristics of two $Fe(Htrz)_2(trz)](BF_4)$/PANI composites were studied at both the low spin state at T = 320 K and the high spin state at T = 380 K. The first composite was fabricated with PANI-1, and the second composite was prepared with PANI-2. PANI-1 was directly synthesized into the emeraldine salt form using hydrochloric acid, whereas PANI-2 was synthesized from a purchased PANI emeraldine base and further doped with p-toluenesulfonic acid to create the emeraldine salt.

For the $Fe(Htrz)_2(trz)](BF_4)$/PANI-1 composite, drift carrier lifetimes are found to be in excess of microseconds, with indications that the majority carriers are holes. While the drift carrier lifetime is much lower when the $Fe(Htrz)_2(trz)](BF_4)$/PANI composite is in the low spin state, there are indications that for at least one PANI composite (*i.e.*, the $Fe(Htrz)_2(trz)](BF_4)$/PANI-1 composite), carrier mobility is much higher in the low spin state, so mobility is consistent with higher conductivity of low spin state (Fig. 13a).

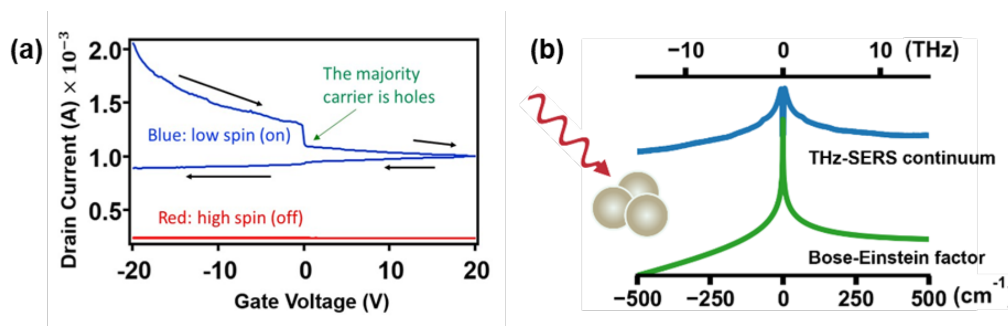

Fig. 13 *The source to drain current transfer characteristics of the $Fe(Htrz)_2(trz)](BF_4)$/PANI-1 composite. Source to drain currents are plotted for gate voltages ranging from -20 V to +20 V at 15 V source voltage. The blue and red curves indicate the drain current at T = 320 K and T = 380 K, respectively. (b) Asymmetrical spectral continuum background in LF-SERS in the terahertz range.*

These results show that the conductance changes expected with $[Fe(Htrz)_2(trz)](BF_4)$ are retained with the addition of PANI and that the composite can act as a thermal or optical switch. They also allude to the importance of the type of PANI used in the fabrication of these devices. The $Fe(Htrz)_2(trz)](BF_4)$/PANI-1 composite has lower on state resistance than the $Fe(Htrz)_2(trz)](BF_4)$/PANI-2 composite. The effects of conductive polymers properties, $Fe(Htrz)_2(trz)](BF_4)$ to PANI ratio, and thin film deposition solvent on the resultant device's performance will be further explored in the coming year. The effects of this will also be examined. Composites with other conducting polymers such as PPy will also be fabricated, and the results will be compared to those from the current $Fe(Htrz)_2(trz)](BF_4)$/PANI composites.

SI **Guo** established the low-frequency surface-enhanced Raman spectroscopy (LF-SERS) to measure the collective framework phonon modes. An asymmetrical spectral continuum background was observed in LF-SERS and an understanding of the electronic Raman scattering process and the inherent asymmetrical Bose-Einstein factor was developed. This work produced a numerical model to treat the background and extract quantitative information. This work is published in *Journal of Physical Chemistry C* (2022). In collaboration with SIs **Laraoui** (FRG2), **Liou** (FRG2), and **Lai** (FRG1), SI **Guo** carried out optical control and Raman spectroscopy measurements on iron triazole SCO molecules. Samples were prepared on diamond substrates with well-defined spin states. Raman response of high spin and low spin states of the SCO molecules were measured. The study shows spin crossovers induced by high power excitation laser, followed by *in situ* Raman observations.

SI **Binek** worked on magnetic nanoparticles for magnetocaloric applications and quantum dots in collaboration with NCAT colleague and center director Dhananjay Kumar. This work is published in *Journal of Applied Physics* (2022, Editor's choice). **Binek** also collaborated with FRG3 SIs **Sabirianov** and **Mei** to explore crossroad structures for qubit applications. The device fabrication is currently underway.

FRG1 SIs contributed greatly not only to research but also Education and Outreach. In Year 2, three SIs, **Guo**, **Hong**, and **Xu**, mentored EQUATE REU students, and many of them presented their research at the Nebraska Summer Research Program Symposium and other local research symposiums. SI Guo's REU student, Madeleine Brown, presented her research (not EQUATE-funded but relevant to EQUATE) at the UNL Chemistry REU Poster Competition and won the Chemistry Travel Award. SI **Hong**'s REU student, Savanna Richardson, also received a best poster award. SIs **Guo**, **Lai**, and **Streubel** hosted NCMN/EQUATE high school students, and those students presented their posters at several symposiums, including the EQUATE External Review Panel meeting on August 17. EQUATE graduate students Jia Wang and Thilini Ekanayaka (from **Hong** and **Dowben** groups, respectively) presented posters at the 27th NSF EPSCoR National Conference in November 2022. In addition to training graduate, undergraduate, and high school students, EQUATE SIs also reached out to middle school students. FRG1 SIs **Binek** and **Lai** led hands-on activities with 75 middle school students in the Education Talent Search summer camp (June 2022).

In terms of Changes/Problems, FRG1's transition from SI **Skomski** to SI **Streubel** has been documented in EQUATE's strategic plan. Despite an intensive international search, which hosted five on-campus candidate interviews, the Department of Physics at UNL was unsuccessful in hiring a new EQUATE assistant professor. UNL has committed to redouble its efforts to complete EQUATE's new hire recruitment in Year 3.

FRG 2 – Quantum Technologies

The goals of FRG2 explore different quantum technologies based on solid-state spin qubits for quantum sensing and metrology, and photons for ultrafast, compact, and low-power quantum communication nanophotonic devices. The FRG is divided into two research thrusts. Thrust 1 (**Laraoui**, **Liou**, **M. Schubert**) focuses on studying spin-magnon interactions in magnonic waveguides, exploring hyperpolarization using hybrid diamond quantum sensors with ferromagnetic nanoparticles (FMN) for low-field (LF) magnetic resonance spectroscopy, and investigating new quantum defects in ultra-wide bandgap (UWBG) semiconductors. Thrust 2 (**Argyropoulos**, **E. Schubert**, **Bao**) explores hybrid nanoscale optical nanostructures for generating single photon sources and entangled photon pairs. During Year 2, FRG 2 investigators collaborated extensively with investigators from other thrusts in FRG 2 and FRG 1 (**Binek**, **Xu**, **Hong**, **Lai**, **Guo**, and **Kovalev**). The scientific and educational achievements are detailed below.

For **Thrust 1: Quantum Sensing and Metrology, Objective 1.a - Probing Spin-Magnon Interactions in Ferromagnetic (FM) Waveguides:** In Year 2, with collaboration with **Xu** (FRG1), we grew TmIG thin films on garnet substrates GGG (111) and SGGG (111), using pulsed laser deposition (PLD) monitored by in-situ reflection high energy electron diffraction (RHEED), where GGG and SGGG stand for $Gd_3Ga_5O_{12}$ and $Gd_{2.6}Ca_{0.4}Ga_{4.1}Mg_{0.25}Zr_{0.65}O_{12}$ respectively. As shown in Fig. 14(a), the x-ray diffraction (XRD) spectra indicate epitaxial growth with a smooth surface. Moreover, the observation of the (222) peaks indicate a lattice distortion of the TmIG films caused by the epitaxial strain, because the (222) diffraction peaks are forbidden for the bulk garnet body center cubic (bcc) crystal structure. The positions of the (222) peaks correspond to a 0.349 nm spacing between the (222) planes, which is smaller than the 0.355 nm bulk value, indicating an in-plane tensile strain. The thickness of fully strained film (critical thickness) is estimated as 5 nm from the width of the (222) peak, while the rest of the film is relaxed. This allows generation of a strain gradient by controlling the film thickness. The magnetic properties of the films have been examined using magneto optical Kerr effect (MOKE). As shown in Fig. 14(b) with a polar configuration, i.e., normal incidence of light and magnetic field along the out-of-plane (111) direction, near-square-shaped hysteresis loops were observed, indicating approximately perpendicular magnetic anisotropy (PMA). This PMA is consistent with the broken cubic symmetry by epitaxial strain, which indicates a spin-lattice coupling. We have used Ferromagnetic Resonance (FMR) setup built in Year 1 to characterize the YIG and TmIG films. In Fig. 14 (c) we plot the FMR measurements on a 30-nm thick TmIG/GGG film. The FMR linewidth is in the range of 100-400 MHz depending on the applied magnetic field and the FRM frequency (Fig. 14 (d)), giving a damping value ~ 0.001. We have also used nitrogen vacancy (NV) based magnetometry to measure the surface propagating spin waves (SW) at the sub-micron scale, seen by the amplification of the local microwave magnetic field due to coupling of NV spin with stray-field produced by the spin waves in 3-um thick YIG/GGG (Fig. 14(a)) and 35-nm thick TmIG/GGG (Fig. 14(c)). We imaged the propagating surface SWs (Fig. 14(a)) in YIG where an interference of the SWs with MW AC field is clearly seen, Fig. 14(b). For TmIG we monitored the NV rabi frequency ON and OFF the film and saw an amplification of the rabi

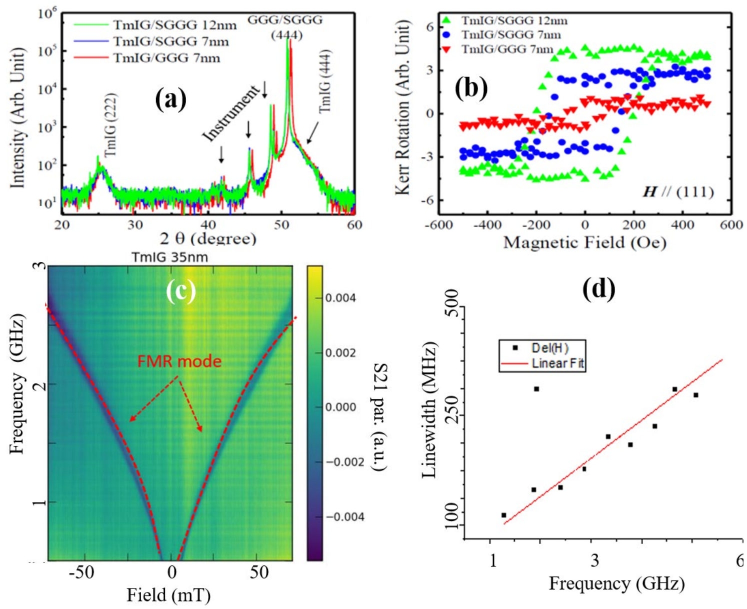

Fig. 14. ***(a)*** *XRD data on TmIG films grown on (111) GGG and SGGG.* ***(b)*** *Polar MOKE measurements of TmIG films grown on sGGG.* ***(c)*** *FMR measurement on TmIG (30 nm)/GGG showing the main FRM mode.* ***(d)*** *linewidth of the FMR mode vs the FMR frequency.*

frequency at periodic field regions (10-20 mT and 40-50 mT), indicating a strong coupling regime of the NV spin qubits with the magnon modes of TmIG, Fig. 14(d). We are studying the effects of thickness and substrate (sGGGs vs GGG) on the SW properties and will perform experiments on the transport of spin waves and the mechanisms of NV-magnon coupling in TmIG nanostructures, relevant for quantum magnonic applications.

Laraoui group in collaboration with **Xu** (FRG 1) developed a growth recipe for TmIG films, reproducible with different growth conditions. **Laraoui** implemented microwave (MW) frequency modulation that allowed to speed up the NV imaging acquisition from > 10 hours to few hours depending on the scanned region size and resolution.

Laraoui trained a high school student from Lincoln, NE on wire bonding of MW stripelines on pcb chips for NV and FMR experiments. **Laraoui** also trained an REU student from Ohio State University in the summer of 2022 on automating the FMR measurements in Fig. 15. A second-year PhD student was trained on nanofabrication (EBL, Optical lithography, DW, FIB), characterization (SQUID, VSM, XRD, SEM) facilities at NCMN and NERCF, and micromagnetic modeling (MuMax).

Laraoui and his students (PhD, high-school and undergraduate) gave multiple talks and posters at local workshops, national meetings such as the 2023 APS and MRS Spring meeting. **Laraoui** is invited to give a seminar on probing Nanoscale Magnetic Phenomena Using Diamond Quantum Sensing Microscopy at the Virtual Lecture Series on Nanodiamond. **Laraoui** published a perspective paper in Applied Physics Letters, entitled "Opportunities for nitrogen-vacancy-assisted magnetometry to study magnetism in 2D van der Waals magnets." A third-year PhD student from FRG2 in collaboration with a PhD student and postdoc (of FRG1's **Binek**) published a paper in *RSC Advances*, titled "Nanoscale imaging of antiferromagnetic domains in epitaxial films of Cr_2O_3 via scanning diamond magnetic probe microscopy."

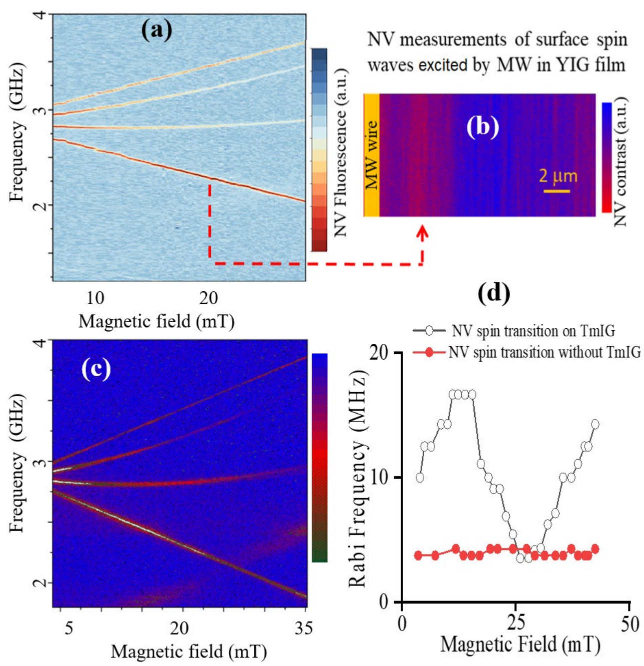

***Fig. 15. (a)** NV-ODMR spectra map taken at different applied magnetic fields on 3 mm thick YIG/GGG. **(b)** Propagating surface SW mapped with NV at an optimized NV-SW coupling field of 20 mT (2.2 GHz). **(c)** NV-ODMR spectra map taken at different applied magnetic fields on 30 nm thick TmIG/GGG. **(d)** Rabi frequency vs applied magnetic field showing regions of a strong coupling between magnons and NV spin qubits.*

For **Objective 1.b - Quantum Sensors for Low Field Magnetic Resonance Spectroscopy**, in Year 2 **Laraoui** and **Liou** used NV-low field magnetic resonance (LFMR) setup (built in Year 1) to study spin-crossover (SCO): [Fe(Htrz)2 (trz)](BF4), (Htrz = 1H-1,2,4,-triazole) molecule rods made by **Lai** (FRG 1). While the bulk magnetic properties of Fe triazole molecules are widely studied by magnetometry techniques their properties at the individual level are missing. Here we used NV-LFMR to image individual Fe triazole SCO nano-rods of size varying from 10 to 1000 nm (See. Fig. 16(a) for 1 mm rods). We performed scanning electron microscopy (SEM) and Raman spectroscopy in collaboration with Guo (FRG1) to find out the size of the nano-rods and to confirm the spin state of the SCO molecule respectively. We drop-casted 1 mm SCO rods on top of a diamond substrate doped with

200-nm thick NV layer. The stray magnetic fields produced by individual nano-rods (Fig. 15(d)) are imaged as a function of the applied magnetic field (up to 3500 G) and correlated with SEM (Fig. 15(b)) and Raman (not shown here). We found that most of the low-spin (LS) states are paramagnetic in contrary to prediction of a diamagnetic behavior, Fig. 15(c). Some of the clustered rods have a superparamagnetic behavior (e.g., P4 in Fig. 15(c)) and we explain this by the formation of iron oxide layer at the surface of the SOC rods. We are integrating the nanograting made by Liou (in Year 1) and perform temperature studies and T1 imaging of cytochrome C (in progress).

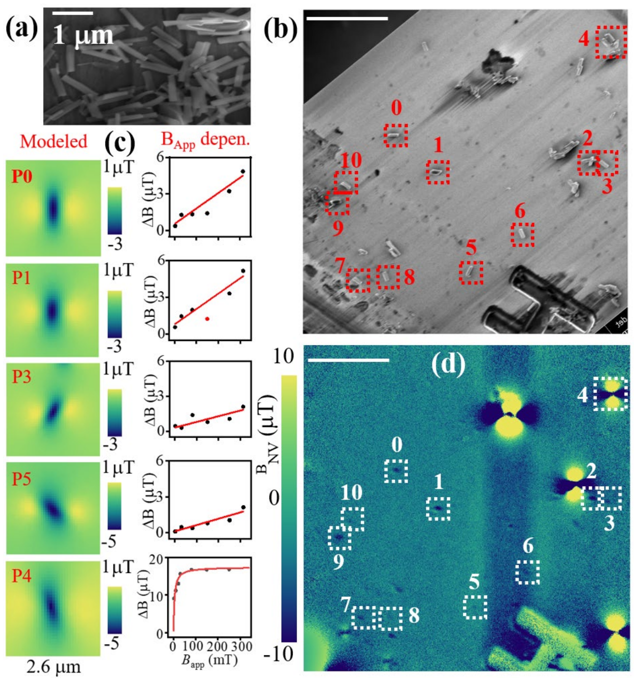

***Fig. 16. (a)** SEM image of 1 mm Fe triazole rods. **(b)** SEM image of isolated 1 mm Fe triazole rods drop casted on top of the diamond substrate. **(c)** modeled (left) and magnetic field dependence (right) of selected rods in (**b**). **(d)** ODMR image of the molecules in **(b)** showing weak paramagnetic and strong (super paramagnetic) diploes, confirmed by field dependence.*

Laraoui, in collaboration with **Liou** (FRG 2) and **Guo** and **Lai** (FRG 1), submitted an abstract on NV sensing of Fe-Triazole nanorods for oral presentation (accepted) to be presented by a female graduate student at the APS 2023 March Meeting.

Further, **Laraoui** trained a female graduate (third-year PhD) student, supervised by **Liou**, on aspects of quantum biosensing using NV-LFMR microscopy. **Laraoui** held weekly group meetings jointly with **Liou** group where students (3 graduates and 2 undergraduates) and postdoc learn new aspects of quantum sensing technologies every week. We also joined live events relevant to our research from Adamas Nanotechnologies, Argonne National Lab, and Ames National Lab.

In addition, **Laraoui** in collaboration with **Liou** (FRG 2) and **Guo** and **Lai** (FRG 1) will present two oral talks (one invited) on NV imaging of Fe-triazole SCO rods (Fig. 16) for oral presentations by graduate students at APS March 2023 Meeting in Las Vegas, NV, and at the Adamas Virtual Lecture Series on Nanodiamond in January 2023. A paper titled "Studying spin states of single nano-rods of Fe (II) triazole spin-crossover molecules using diamond quantum sensing microscopy" was submitted to the high impact journal *Nanoscale*.

For **Objective 1.c Characterizing new solid-state Qubits in Ultrawide Band Gap semiconductors**, the **Schubert** group implemented a new solid-state driven fast-frequency sweeping THz source into the prototype THz-EPR ellipsometer setup and developed fast methods for calibration across wide frequency spectrum. THz-EPR Ellipsometry data acquired in Year 1 on Fe-doped monoclinic gallium oxide were analyzed using a 4^{th} order spin-spin interaction Hamiltonian approaches. Resulting experimental and model for imaging part of the magnetic susceptibility are shown in Fig. 17. Significant changes with frequency of spin levels reveal two sets of zero-field split s=5/2 systems due to Fe in Ga_i and Ga_ii sites. Quantitative analysis of the Hamiltonian parameters and identification of their monoclinic distortion is reported for the first time. Measurements are ongoing on Cr, Cu, Co, Cs, SI, and Al doped and co-doped Gallium Oxide and Sn and Ge doped AlGaO alloys in epitaxial layers. We further developed the strain stress relationships for phonon modes and electronic band to band transitions for monoclinic symmetry

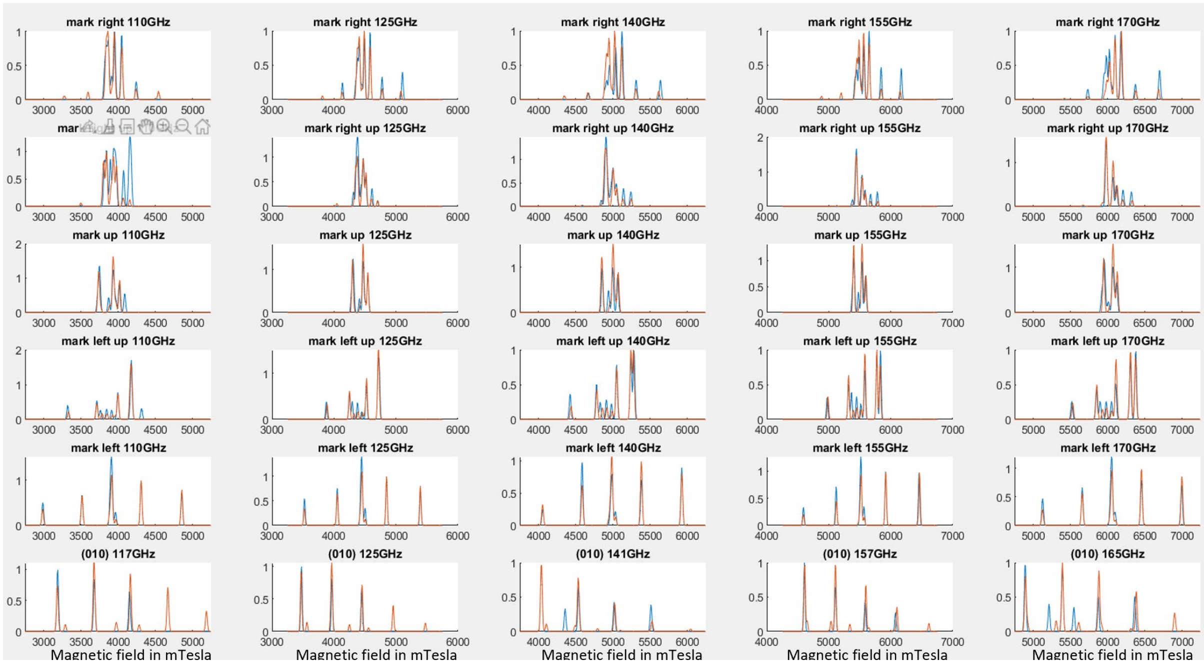

Fig. 17. *THz-EPR Ellipsometry measured imaginary part of the magnetic susceptibility for Fe-doped Gallium Oxide single crystals with different crystal cuts and orientation relative to magnetic field. Signatures are caused by tetrahedral and octahedral site Fe3+ with zero-field splitting within the monoclinic crystal field environment. Experiment (brown) is matched well by model (blue) using 4th order spin-spin interaction Hamiltonian parameters. Model analysis reveals the monoclinic distortion of the spin levels within the highly anisotropic defect sites. Comparison with theoretical approaches in collaboration with Prof. Chris van de Walle (UCSB) are under way in Year 2.*

aluminum gallium oxide as a function of aluminum composition. The **Schubert** group further collaborated with Lincoln, NE-based J.A.Woollam Co., Inc., for development of THz-EPR ellipsometry. We also developed a numerical approach to predict best orientations for measurements of monoclinic materials in ellipsometry reducing necessary measurement time.

Marking our milestone for Year 2, we successfully calibrated and tested the prototype THz-EPR ellipsometer setup with solid state synthesizer source. NV centers in diamond are measured in Year 2. Marking another milestone for Year 2, we demonstrated THz-EPR-E for detection and characterization of single defect spin systems in gallium oxide bulk crystals. We used our eigenpolarization model developed for SiC in Year 1, and extracted the magnetic susceptibility as a function of frequency and field. From this function we identified spin level transitions due to two different s=5/2 systems established by Fe located in two different Ga sites. We determined Hamiltonian parameters which agree well with theoretical predictions. We demonstrated the usage of THz-EPR E for broad characterization of defects in solids. We report on our findings in a forthcoming publication in the Nature publishing group.

Our prototype THz-EPR ellipsometer setup can be explored for use in frequency-scanning rather than field scanning EPR measurements. New potential quantum qubits are investigated including doped AlN and AlGaN secured from collaborators at Linkoping University, Sweden. M. **Schubert** received a supplement from the National Science Foundation investigating doping and alloying related properties in monoclinic aluminum gallium oxide.

In addition **M. Schubert** visited Lund and Linkoping Universities in Sweden, the Fritz Haber Institute of the Max Planck Society and the Leibniz Institute for Crystal Growth both in Berlin, Germany and presented seminars on oxide based high power electronic materials research for the future of green energy and on THz EPR methods for quantum defect characterization. The **Schubert** group trained new female graduate student Teresa Gramer and new female undergraduate student Jenny Olander together with undergraduate students Grant Ross (REU student) and Preston Sorenson (ECE, University of Nebraska-Lincoln) in ellipsometry basics, basic instrumentation design, and fundamentals of quantum material engineering. **M. Schubert** accepted a new position in 2023 as visiting guest professor within the Wallenberg Centre for Quantum Technology at Chalmers University, Sweden.

The **Schubert** group published one paper in the journal Physical Review Applied (2021, Impact Factor 4.931), titled: "Strain and composition dependencies of the near bandgap optical transitions in monoclinic $(AlxGa1\text{-}x)_2O_3$ alloys with coherent biaxial in-plane strain on (010) Ga_2O_3." This group also presented 40 scientific talks in various in-person and online events at national and international conferences, including 13 invited talks and one plenary talk at the International Conference on Spectroscopic Ellipsometry 9 in Beijing, China. As another highlight, female undergraduate student Megan Stokey won Best Student Award Paper of the Ellipsometry Group Focus Session during the AVS 68 International Symposium and Exhibition (AVS-68) at Pittsburgh, PA, in November 2022. The **Schubert** group also published a total of 25 peer-reviewed papers. Further, the **Schubert** group presented an invited talk on Terahertz Electron Paramagnetic Resonance ellipsometry for interrogating point defects in ultrawide bandgap semiconductors at the prestigious conference on defects in solids for quantum technologies (CECAM 2022) at Stockholm, Sweden in June 2022.

In FRG2's **Thrust 2: Quantum Communication: Transmitting Data with Single or Entangled Photons**, for **Objective 2.a Enhanced Nonlinear Optical Effects at the Nanoscale** in Year 2, we designed, modeled, and fabricated new ultrathin nanocomposite nonlinear nanostructures to substantially enhance the efficiency in their nonlinear optical properties. **Argyropoulos** modeled optical harmonic generation in various nanophotonic platforms. **E. Schubert** and **M. Schubert** fabricated and analyzed enantiomers of highly ordered nanostructure L shapes (nano boomerangs) with different distortion angles using heterostructure iGLAD from silicon and silver as precursors. We measured optical properties of Si-Ag nano boomerangs over a broad spectral range. We fabricated zirconium oxide nano helices and nanocolumnar structures using iGLAD. We measured the optical properties from near infrared to deep ultraviolet spectral region for nanocolumnar zirconia metamaterials and we performed chiral-optical characterization for zirconia nanohelices in the deep UV. We obtained the band-to-band transitions by critical point model dielectric function analysis. Computational studies have been performed to investigate the hybrid photonic behavior of zirconia-silver heterostructured helices. A dielectric nanogap of alumina was created by ALD between columnar subsections of two different materials to study interface properties in heterostructured metamaterials which can create strong light confinement for enhanced photon upconversion. We performed a systematic elemental and structural analysis of zirconia and silicon-silver metamaterials by TEM, SEM, selected area diffraction, EDX and XRD. We performed computational analyses for the

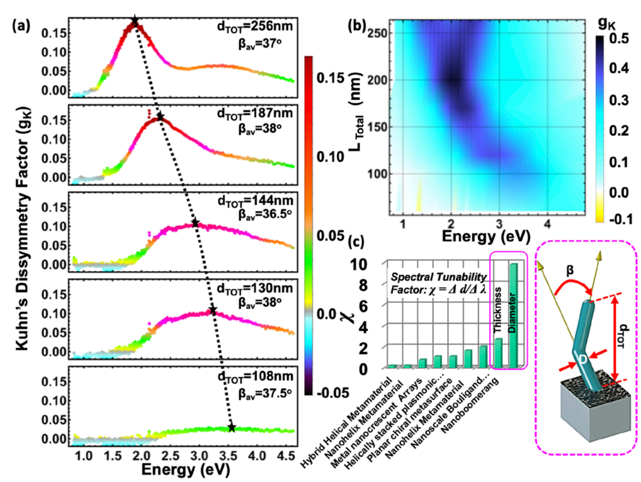

***Fig. 18. (a)** Experimentally measured and **(b)** theoretically computed spectral evolution of the Kuhn's dissymmetry factor for different total thickness (d_{TOT}) of nano-boomerang metamaterial design. **(c)** Comparison of tunability factor (c) values from literature with our work.*

effect of increasing the index of refraction surrounding Si-Ag chiral heterostructure column thin films using hypothetical silicon oxide and titanium oxide onto their chiral optical properties. We performed computational analyses of the nonlinear optical properties for silicon-silver helical nanoheterostructures where structural parameters were varied aiming to understand the effect onto the left and right polarized second harmonic coefficient over the spectral range from the near infrared to the deep ultraviolet. Measurements of the second harmonic generation for L shape and helical metamaterial platforms have been performed in collaboration with Dr. Hoang from University of Memphis.

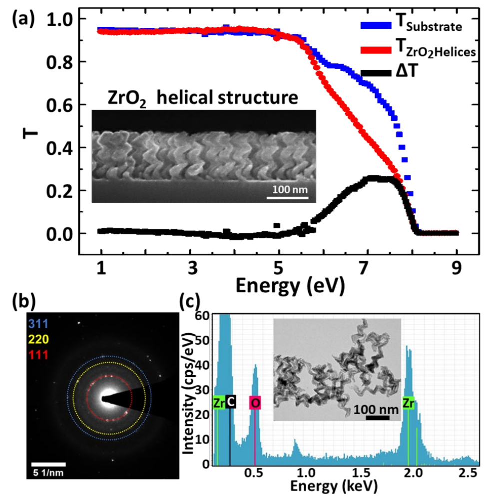

Fig. 19. ***(a)*** *Experimentally measured transmission spectrums from quartz substrate (blue symbols), ZrO_2 helices with three full turns (red symbols) and the difference between (black symbols).* ***(b)*** *diffraction pattern of dispersed helical structures* ***(c)*** *EDX spectrum of ZrO_2 helices. The inset is the scanning TEM image of ZrO_2 helices on a mesh grid.*

The **Argyropoulos** group collaborated with **E. Schubert** and **M. Schubert** (FRG 2) to model chiral metamaterials combined with topological nanostructures to enhance linear and nonlinear chirality with the goal to achieve this effect at the ultimate low radiation power level--i.e., the single-photon level--marking another milestone for Year 2. Various chiral nanostructure samples were sent to our University of Memphis collaborator for nonlinear optical measurements. We observe chiral second harmonic generation from helical and nanoboomerang structures (Fig. 18). Notably, depending on the distortion angle of nano-boomerang structure, the second harmonic nonlinear chirality response can also be tuned. Simulations of the chiral nonlinear harmonic generation were performed by **Argyropoulos** to verify the experimental results. The iGLAD synthesis of enantiomer chiral structures and their nonlinear optical characterization are marked as the prominent progress achievements for Year 2.

E. Schubert and **M. Schubert** measured a strong and extremely broadband transmissive bisignate (positive and negative) circular dichroism from the near infrared to the ultraviolet spectral range which is controlled by nano-boomerang structure parameters (distortion angle and the length of columnar subsegments). All-dielectric nano-boomerang structures offer alternative routes to high loss metals and can pioneer the use of such metamaterial design in myriads of photonic applications. The systematic fabrication and chiroptical characterization of all-dielectric Si nano-boomerangs with various lengths showed spectral tunability (with the highest tunability factor (19c)) of their chiral response (Fig. 19). Such capability of engineering the spectral location of chiral extrema has the uttermost importance for wide-variety applications including photonic integrated circuit designs, quantum sensing, broadband circular polarizers, nonlinear optical devices etc. Additionally, further theoretical efforts were undertaken to decompose total circular scattering dichroism ($\Delta Stot=S_{LCP}-S_{RCP}$) into its electro-magnetic modes. Indicated by such simulations, electric-dipoles seem to drive the substantial chirality response. Zirconia (ZrO_2), an ultra-wide band gap metal oxide material was fabricated in **E.**

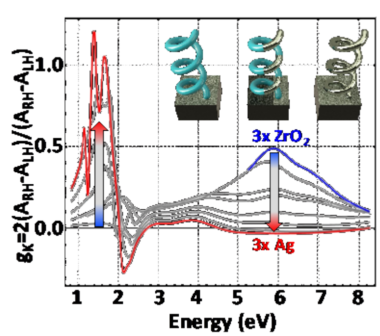

Fig. 20. *Theoretically computed spectral evolution of the Kuhn's dissymmetry factor for different ZrO_2 ratios per helix turn in the case of three turn RH ZrO_2-Ag hybrid helical metamaterials.*

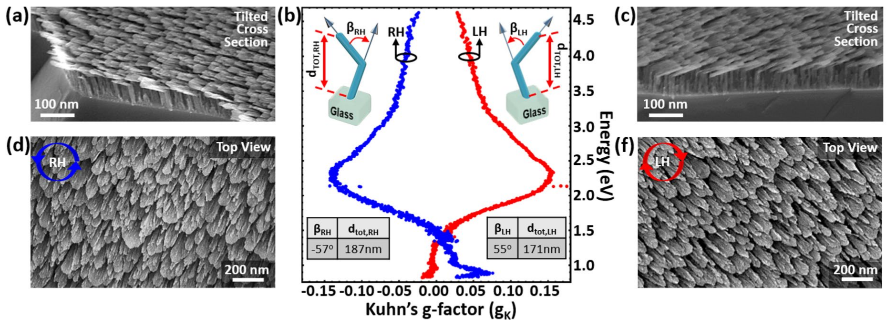

*Fig. 21. High resolution tilted cross section and top view SEM images of **(a), (d)** RH **(c), (f)** LH L-shaped chiral metamaterials. **(b)** The corresponding spectral evolutions of the LH (red line) and RH (blue line) metamaterial measured Kuhn's dissymmetry chiral factors.*

Schubert's lab, and **M. Schubert** detected a strong circular dichroism in the deep ultraviolet spectral range for Zirconia nano helices. The chiral response can be tailored in its magnitude and spectral position using structural parameters (Fig. 19). We envision a potential use of these chiral metamaterials in areas which include high power required chiroptic photonic/electric circuit designs or UV active topological insulators. Furthermore, finite element modeling showed that incorporations of plasmonic helical subsegments in zirconia helices leads towards an emergence of multiple chiral plasmonic resonances (see Fig. 20).

Simulations of the chiral linear response and nonlinear harmonic generation were performed in **Argyropoulos** group to verify the experimental results. **E. Schubert** and **M. Schubert** measured a strong spectral transmissive circular dichroism from the near infrared to the ultraviolet spectral range which is controlled by the L-shaped chiral metamaterial (Fig. 21) structure parameters. They detected a strong circular dichroism in the ultraviolet spectral range for the zirconium oxide nano helices, which can be tailored in its magnitude and spectral position using structural parameters. They also observed a strong chiral narrow band (notch) filter characteristics in Si-Ag chiral heterostructure column thin films with high index material backfilling, and measured a strong, frequency tunable enantiomeric response of the second harmonic generation in silicon helical nanostructures, with 10 times larger conversion efficiency than the best previously reported efficiency of comparative silicon nanopillar structures.

Argyropoulos modeled nonlinear and quantum optical effects and applied the modeling to various nanophotonic plasmonic structures. He is in the process of being awarded a grant from NSF DMR with FRG2's **E. Schubert** to study chiral metamaterials and their usage in quantum optical nanophotonic applications.

E. Schubert and **M. Schubert** demonstrated by computational modeling a helical nanostructure achievable by iGLAD with a chiral edge state with topological properties controllable by structure design with nearly 100% circular dichroism. This is a key achievement for the development of new hybrid composite photonic nanostructures. Our computation analyses for the second harmonic response of silicon chiral nanostructures open the door for incorporation of frequency tunable high efficiency chiral nonlinear photonic nanostructures.

Sharing these advances, **Argyropoulos** educated his postdoc and graduate students in the theoretical concepts to model quantum entanglement and spontaneous parametric down conversion. **E. Schubert** visited the Leibniz-Institute for Surface Engineering in Germany and trained local collaborators in ion beam assisted glancing angle deposition.

E. Schubert and **M. Schubert** submitted and published a total of 17 publications, of which 5 are published and 4 are accepted and awaiting publication. As one highlight, one paper in the journal *Nature* (2020 Impact Factor 49.962), is titled: "Hyperbolic Shear Polaritons in Low-Symmetry Crystals." The editors are in the process of deciding whether to use our cover image for the weekly cover issue of Nature upon publication. We have presented nine scientific talks in various in-person and online events. **Argyropoulos**, with **E. Schubert** and **M. Schubert**, is preparing a publication based on L-shaped chiral metamaterials which will be submitted soon in a high-impact journal.

Argyropoulos published five articles in high-profile journals, and gave multiple talks at conferences, including an invited talk at Metamaterials 2022.

For **Objective 2.b Robust Entangled Photon Generation from Nanoscale Structures**, in Year 2 the **Argyropoulos** group modeled the enhanced second-harmonic generation (SHG) from individual plasmonic nanopatch antennas (NPAs). **Bao** group is developing an experimental set-up to produce and measure spontaneous parametric down-conversion (SPDC) using different processes.

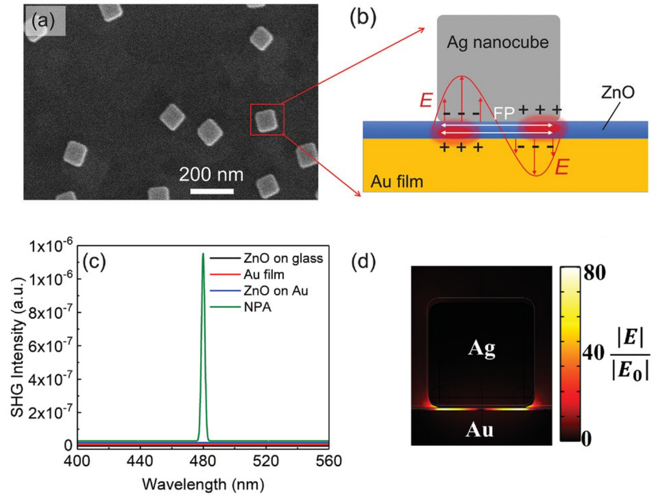

***Fig. 22. (a-b)** Nanopatch antennas and their field profile at the ZnO nanogap. **(c)** Enhanced SHG intensity from nanopatch antennas at their resonant frequency. **(d)** Simulated field enhanced in nanogap.*

Argyropoulos was able to enhance SHG from individual plasmonic NPAs which were formed by separating silver nanocubes from a smooth gold film using a sub-10 nm zinc oxide spacer layer, as demonstrated in Fig. 22(a),(b). When the NPAs were excited at their fundamental plasmon frequency, a 104-fold increase in the intensity of the SHG wave was measured which is depicted in Fig. 22(c). The computed field enhancement in the nanogap is shown in Fig. 22(d). Moreover by integrating quantum emitters that have an absorption energy at the fundamental frequency, a second-order nonlinear exciton–polariton strong coupling response was observed for the first time with a Rabi splitting energy of 19 meV. The nonlinear frequency conversion using NPAs thus provided an excellent platform for nonlinear control of the light−matter interactions in both weak and strong coupling regimes which is expected to have a great potential for applications in optical engineering and information processing. This work was published in the high-impact journal of Advanced Optical Materials. In addition, the presented ultrathin structure can achieve maximum nonlinearity enhancement and enhanced spontaneous parametric downconversion (SPDC) leading to an optimum entangled photon pair generation rate which is one of the main objectives of the FR2 thrust 2.

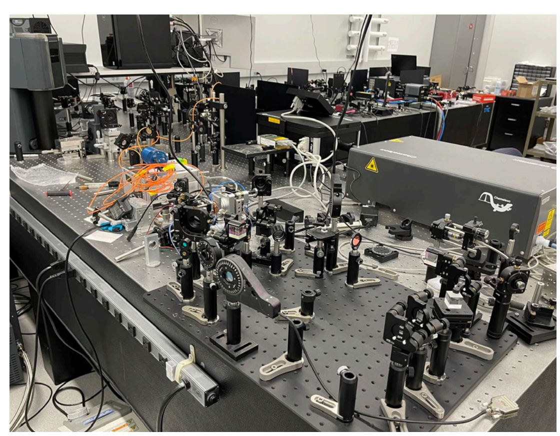

***Fig. 23.** Setup in **Bao**'s lab with newly installed coherent fs laser and SPDC setup being built.*

Argyropoulos studied the temperature dynamics of hot electrons generated at plasmonic nanopatch antennas and their transition into thermal carriers at various timescales from femto to nanoseconds. Additionally, the hot electron temperature and generation rate threshold values were investigated by using a hydrodynamic quantum nonlocal model approach. The derived temperature dependent material properties were used to study the ultrafast transient nonlinear modification in the absorption spectrum before plasmon-induced lattice heating was established leading to efficient tunable nanophotonic absorber designs. His results were published in the high-impact journal *Nanophotonics* and elucidated the role of hot electrons in the response of nanopatch antenna absorbers which can be used to design more efficient photocatalysis, photovoltaics, and photodetection devices.

Bao group is developing an experimental set-up to produce and measure SPDC two-photon entangled pairs (Fig. 23). **Bao** purchased all equipment required for the experiment including tunable femtosecond laser, single photon detectors, microwave, and optical equipment and built the setup shown in Fig. 23.

Also, **Argyropoulos** was awarded a DURIP Grant from ONR with another UNL faculty not involved in EQUATE (Prof. Craig Zuhlke), which will purchase a high-power ultrashort pulsed laser that can be used for future EQUATE projects relevant to SPDC or other nonlinear quantum optical processes. **Argyropoulos** was awarded Year 2 of a grant from the Nebraska Center for Energy Sciences Research with **Bao** (FRG 3) to study robust topologically protected energy-efficient on-chip microlasers for secure data center communication systems. Two works led by **Bao** group (FRG2/FRG3) were published in high-impact journals: *Nature Materials* and *Nature Communications*, respectively.

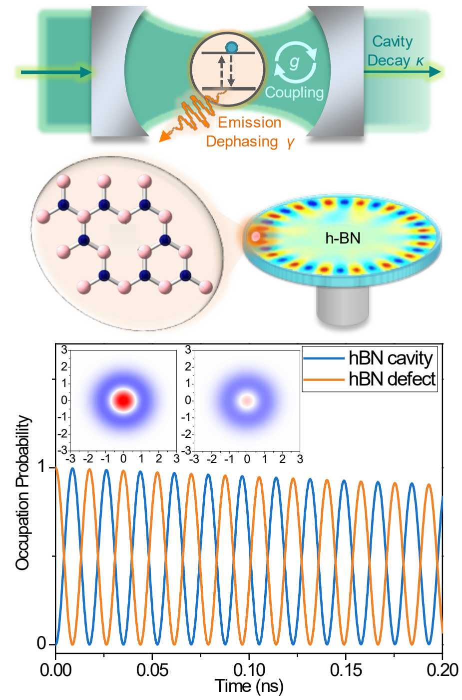

***Fig. 24. (a)** Schematic illustration of emitter-cavity coupled system based on hBN. **(b)** Simulated Rabi oscillations and Wigner functions of (a) in strong coupling region.*

Additionally, **Argyropoulos** educated his graduate students in the theoretical concepts to model quantum entanglement and spontaneous parametric down conversion. **Argyropoulos** taught an undergraduate class (ECEN306) relevant to electromagnetic theory; there he introduced to the undergraduate students the research opportunities within EQUATE and briefly presented EQUATE-related research done in his group.

Argyropoulos published a paper on efficient nonlinear process and strong coupling based on plasmonic nanocubes (Fig. 24) in the high impact journal *Advanced Optical Materials*. **Argyropoulos** also published a paper in *Nanophotonics* relevant to plasmonic absorbers that excite quantum hot-electrons.

For **Objective 2.c Efficient Single Photon Generation from Nanoscale Structures**, in Year 2 the **Laraoui** group optimized the optical setup to characterize single-photon emitters (SPEs) in two-dimensional (2D) layers and flakes. **Argyropoulos** collaborated with **Laraoui** (Thrust 1, FRG2) to simulate nanopatch plasmonic nanoantenna configuration similar to in Fig. 24 that is combined with hBN defects to efficiently generate single photons (the outcome of this work will be reported in Year 3 of EQUATE's strategic plan document).

Under the SEED Grant "Dynamic Control of 2D Single-Photon Quantum Emitters via Strain Engineering," in Year 2 **Wang** group investigated the quantum electrodynamics (QED) of emitter-cavity coupled systems based on 2D hBN Crystals. A defect-embedded microdisk cavity was proposed, as shown in Fig. 24(a) and analyzed utilizing the Quantum Toolbox in Python (QuTip). Energy transfer dynamics and photon statistics of the coupled system can be visualized as occupation probability and Wigner function plots (Fig. 24(b)). Clear Rabi oscillation and negative phase terms can be observed when the cavity is designed with a radius of 2.5 μm and a thickness of 250 nm, the corresponding cavity decay rate of 0.1 GHz and a coupling rate of 28.5 GHz. Strong coupling and coherent energy/state transfer can be predicted in the designed systems. The methodology developed in this study provides valuable guidelines for designing emitter-cavity coupled systems, which are the key building blocks towards realizing functional quantum photonic circuits.

Moreover, **Wang** group explored a novel family of 2D crystals, cesium lead halide $CsPb_2X_5$ (X = Cl, Br) perovskites. Distinct from the hallmark 2D materials (such as graphene, hexagonal boron nitride, and transition metal dichalcogenides) usually synthesized by chemical vapor deposition or mechanical/liquid exfoliation from bulk crystals, $CsPb_2X_5$ perovskites can be directly synthesized via hydrothermal reactions. **Wang** group developed a hydrothermal synthesis protocol that can precisely control the phase, dimension, and thickness of $CsPb_2X_5$ crystals and systematically characterized the thickness-dependent optical and mechanical properties. Utilizing the wide-bandgap attributes of $CsPb_2X_5$ (3.5 eV to 4.8 eV, depending on the choice of halide elements and their mixture ratio) and the controllable transformation into the luminescent $CsPbX_3$ phase, the **Wang** group will further investigate this naturally forming quantum dot-cavity coupled systems. These research activities will facilitate a low-cost, scalable, and versatile platform for integrated quantum photonics.

During weekly group meetings, **Wang**, **Laraoui**, **Bao**, and **Argyropoulos** shared their knowledge in quantum optical processes with emphasis on single-photon emission and quantum nonlinear optical effects to students (graduate and undergraduate) and postdocs. **Laraoui** extended the concepts of single photon generation in range of materials including wide-band gap semiconductors and 2D materials in his MATL492/892 class (Introduction to Quantum Materials and Technologies)--including non-UNL students at UNO, UNK, and Creighton University.

Sharing these developments in Year 2, FRG2 members gave multiple talks and poster presentations including contributed and invited talks at the MRS Spring meeting (**Argyropoulos**, **Laraoui**) and the 2023 APS March Meeting (**Laraoui**, **Argyropoulos**). **Wang** gave an invited talk at the American Vacuum Society (AVS) conference, "Photonic Crystals based on Two-Dimensional Materials."

<u>Impacts</u>

In year 2 FRG 2 SIs made significant impacts in many aspects of research and education, including:

- The THz EPR system developed by **M. Schubert** group has garnered serious interest for commercial replication by industry partner, the J.A. Woollam Co., Inc. If this instrument can make it to market, a very large number--possibly thousands--of modern EPR instruments may operate under the principles developed in this award.
- **M. Schubert** group published a paper in the journal *Physical Review Applied* (2021, Impact Factor 4.931) titled: "Strain and composition dependencies of the near bandgap optical transitions in monoclinic."
- **Argyropoulos** was invited to give a talk at 16th International Congress on Artificial Materials for Novel Wave Phenomena - Metamaterials 2022 in Sienna, Italy on September 12. This is the premier event for metamaterial research. The talk was about hot electron generation in nanocubes and the paper was published in the high-impact (8.45) *Nanophotonics* journal.
- **Bao** group published two high impacts journals of Nature Materials and Nature Communications, respectively study robust topologically protected energy-efficient on-chip microlasers for secure data center communication systems.
- **Laraoui** shared the syllabus of a new elective MATL- 492/892: Introduction to Quantum Materials and Technologies with science and engineering departments at non-UNL campuses (UNO, UNK, and Creighton University). For Spring 2023 (January- May), so far 13 students (4 female) registered for the class with 9 graduate students and 4 undergraduate students from Physics, Chemistry, Electrical & computer engineering, and Materials & Mechanical engineering departments.

- **Wang** developed a new syllabus for course ECEN-491/891: Quantum Communication and Quantum Computation and introduced a quantum programming module that exposes the students to real-world quantum computation systems, leveraging the IBM quantum resources. From Spring 2022 to Fall 2022, 16 students (2 undergraduates and 14 graduates) enrolled in this course, and four of them were female students.

Changes/Problems

Unfortunately, FRG2's **Argyropoulos** accepted a position at Pennsylvania State University, starting January 2023. EQUATE's Management Team successfully filled this vacancy with his mentee, **Dr. Ufuk Kilic**, who will serve as an SI in FRG2 for the remainder of the EQUATE award period.

FRG3 – Quantum Information Processing

As outlined in EQUATE's strategic plan, Year 2 for FRG3 builds upon the personnel investments in Year 1 to achieve the team's first significant results in quantum information processing. Major successes include the demonstration of room-temperature Bose-Einstein condensation in exciton-polariton system, stable sub-Doppler ultracold atoms, demonstration of computational techniques for ultracold polarons, characterization of computing efficiency of nonlinear quantum walk algorithms, collaboration on crossroad quantum dot realization, and theoretical simulations of magnetic molecule and adatom systems.

Key personnel are FRG3 leader **Wrubel** (Creighton University), and Science Investigators (SIs) **Bao** (UNL), **Armstrong** (UNK), **Mei** and **Sabirianov** (UNO), and **Wysocki** (UNK). As expected, SI **Wong** (Creighton University) continues his leave to serve at the National Quantum Coordination Office (NQCO) in Washington DC until the end of 2023.

In terms of Outreach/Dissemination, FRG3 currently mentors one postdoc, three master of science graduate students, and six undergraduate students; two in this group are female, one is black, and one is Hispanic/Latino. Our main outreach activity was working with a summer high-school research student through the Creighton Haddix STEM corridor program. Two students gave poster presentations at a national conference: Division of Atomic, Molecular and Optical Physics (DAMOP).

FRG3 expects to generate at least two publications by the end of Year 2: one on the quantum computing theory project and one on our sub-Doppler octupole MOT. Also, two masters' students will defend their theses by May 2023. FRG3 researchers have given four presentations at regional and national conferences.

For **Thrust 1. Quantum Emulation**'s **Objective 1.a: Surpassing the Standard Quantum Limit in a ^{41}K BEC**, our experimental cold-atom effort has focused on completing the apparatus needed to achieve Bose-Einstein condensation. The key to achieving Bose-Einstein condensation is loading a sufficient number of atoms into an optical dipole trap. The number of atoms is determined by their temperature and the time they have to cool into the dipole trap. We implemented techniques for measuring the temperature of the atoms in the magneto-optical trap (MOT) via time-of-flight fluorescence imaging and used this technique to maximize the number of atoms and minimize their temperature with precise computer control of the laser fields.

We were able to demonstrate the successful operation of the first stable sub-Doppler magneto-optical trap with temperatures less than 80 μK, and atoms stable for more than 0.5 seconds. This is sufficiently cold we will be able to load unprecedented numbers of atoms into our optical dipole trap.

The key to this innovation was the use of a magnetic field with octupole symmetry. Fig. 25 shows the radial size of the potassium atoms in the magneto-optical trap as a function of the time after release or time-of flight. The atoms expand because of their Maxwell-Boltzmann distribution of velocities at a given temperature. By fitting the expansion versus time we can extract the initial size of the cloud and its temperature. As the quadrupole trap is smoothly changed into an octupole trap, we find that the cloud gets bigger and colder. This is expected because of the octupole magnetic field giving a weaker confining potential, plus the onset of sub-Doppler cooling forces in the octupole field.

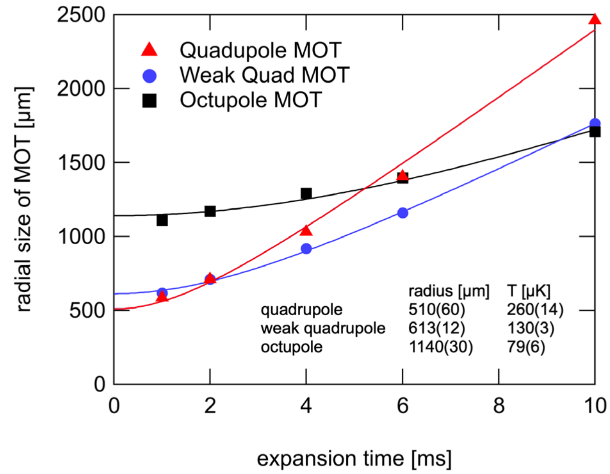

Fig. 25. Measured radial size of the cloud of potassium atoms in the magneto-optical trap versus expansion time, with 2-parameter fits. Red triangles are a quadrupole magnetic field, blue circles, a weak quadrupole, and black squares are the unique octupole magnetic field arrangement.

Typically sub-Doppler temperatures can only be reached with untrapped atoms for 20 ms or less, but here we demonstrate stable sub-Doppler temperatures for 500 ms. This is expected to dramatically improve the loading of our far-off resonance optical dipole trap. We have designed and tested optics and mounts for the 1550 nm, 30 W, optical dipole trap laser. Before the end of this project year, we expect to have loaded the dipole trap and achieved Bose-Einstein condensation, by taking advantage of our unique octupole MOT.

Other significant experimental projects include preparing for magnetic field shielding of the experiment, receiving quotes for scientific imaging cameras which will be purchased before the end of Year 2. We are also prepared to begin measurement of the lifetime of atoms in the presence of the radio-frequency Feshbach resonance.

Overall progress in this area is slightly behind schedule; alignment of the octupolar MOT took extra time, yet is also a major scientific achievement. Going forward, we expect to catch up by the end of Year 3.

In **Objective 1.b: Emulation of Novel Spin Systems**, the main activity for emulation of novel spin systems will take place in subsequent years, but the capital equipment purchase of the main 2D lattice laser was completed. This laser will provide the capability to do quantum emulation of many condensed matter systems including systems important for quantum computation. We have also begun layout of the optical system for the 2D lattice laser.

For **Objective 1.c: Quantum Emulation with an Exciton-Polariton BEC**

Quantum fluids exhibit quantum mechanical effects at the macroscopic level, which contrast strongly with classical fluids. Gain-dissipative solid-state exciton-polaritons systems are promising emulation platforms for complex quantum fluid studies at elevated temperatures. Recently, halide perovskite polariton systems have emerged as materials with distinctive advantages over other room-temperature systems for future studies of topological physics, non-Abelian gauge fields, and spin-orbit interactions. However, the demonstration of nonlinear quantum hydrodynamics, such as superfluidity and Čerenkov flow, which is a consequence of the renormalized elementary excitation spectrum, remains elusive in halide perovskites. Here, using homogenous halide perovskite single crystals, we report, in both one- and two-dimensional cases, the complete set of quantum fluid phase transitions from normal classical fluids to scatterless polariton superfluids and supersonic fluids—all at room temperature, clear consequences of the Landau criterion. Specifically, the supersonic Čerenkov wave pattern was observed at room temperature. The experimental results are also in quantitative agreement with theoretical predictions from the dissipative Gross-Pitaevskii equation. (Fig. 26) Our results set the stage for exploring the rich non-equilibrium quantum fluid many-body physics at room temperature and pave the way for important polaritonic device applications.

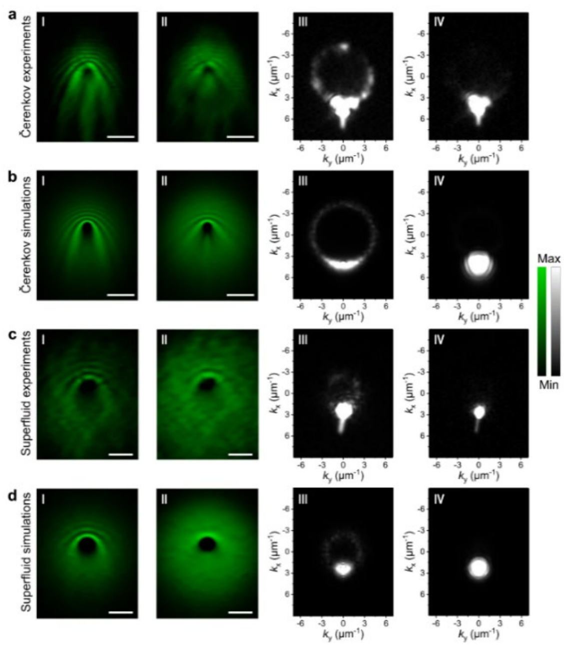

Fig. 26. ***a**-I-II, Experimental real-space images of polariton flowing with injection momentum of $k_{//}$ = 4.2 μm^{-1} across a defect with resonance pumping power in the cavity of 0.07 (I) and 6 (II) μJ cm^{-2}, respectively. **a**-III-IV, The experimental saturated images of momentum distributions corresponding to **a**-I and **a**-II, respectively. At low polariton density (**a**-I, III), the scattering by the defect generated parabolic interference wavefronts and Rayleigh scattering ring. When polariton density increased, Čerenkov behavior in the supersonic regime (c_s<v_p) was shown as the linear wavefronts and the increased aperture angle. The scattering ring gets modified at high polariton density (**a**-IV). **b**-I-II, the time-averaged simulation in real-space before and after the transition, respectively. Evenly distributed 0-5 meV exciton energy random variations are added to only 0.3% of the spatial pixels to better fit the experimental results. **b**-III-IV, The time-averaged simulation images of the momentum space corresponding to **b**-I and **b**-II, respectively. **c**-I-II (**c**-III-IV), Experimental real-space (momentum space) images of polariton flowing with injection momentum of $k_{//}$ = 2.3 μm^{-1} with resonance pumping power in the cavity of 0.06 (I) and 5.6 (II) μJ cm^{-2}, respectively. At high polariton density (**c**-II, IV), the polariton fluid entered the frictionless superfluid regime and the interference fringes and scattering ring vanished. **d**-I-II (III-IV), The simulated time-averaged real-space (momentum space) simulations before and after the transition. Evenly distributed 0-5 meV exciton energy random variations are added to only 1% of the spatial pixels to better fit the experimental results. Compared to simulations, the additional short white lines at the bottom of the **a**-III-IV and **c**-III-IV images are streak artifacts caused by the scanning of the CCD camera.*

One significant impact from this work was the **Bao** group's research on solid state of quantum emulation with halide perovskites, published in two very high-impact journals -- *Nature Materials* and *Nature Communications*; these articles have been read/accessed over 8,000 times in the last six months by the scientific community. (UNL is the lead institution for both works.) In terms of Outreach/Dissemination, **Bao** presented these results at the International Conference on Spontaneous Coherence in Excitonic Systems (ICSCE-11) as an invited speaker. Bao also shared the results at the MRS Fall 2022 meeting in Boston, and in the Nature series journal, *Light: science and applications*, with results viewed by more than 1,000 people.

For **Objective 1.d: Theory of Polarons in Dipolar and Spinor Gases**. **Armstrong** has been working on static impurities placed into trapped dipolar Bose gases. In this system, the background particles (or bath particles) are dipolar with their dipole moments aligned by an external field along the *z*-axis. We modified standard dipolar Gross-Pitaesvskii (GPE) codes to include a dipolar impurity in two dimensions (2D). This impurity is a particle with some different property from the bath particles. Chiefly of interest for our calculations is that this impurity's dipole moment magnitude and orientation are different and controllable in a potential experiment. We have codes for two different geometries: where the gas is trapped in the *xy*-plane and then where they are confined in the *xz*-plane. In the former geometry, the dipoles are polarized perpendicular to the particles' confinement, the dipolar interaction is purely repulsive and is isotropic. In the latter case, there is both attraction and repulsion in the system and the system is anisotropic. We calculated the impurity self-energy as well as the number density for these systems. The self-energy is the difference in energy between a system without an impurity and the system with an impurity. The results examined so far produced mostly expected results. In the purely attractive case, the impurity repels particles from the center of the trap and towards the wall, with the reverse happening in the anisotropic case (Fig. 27). Another interesting feature that occurs in the *xz*-plane system is that the system can collapse if the impurity has too strong a dipole moment.

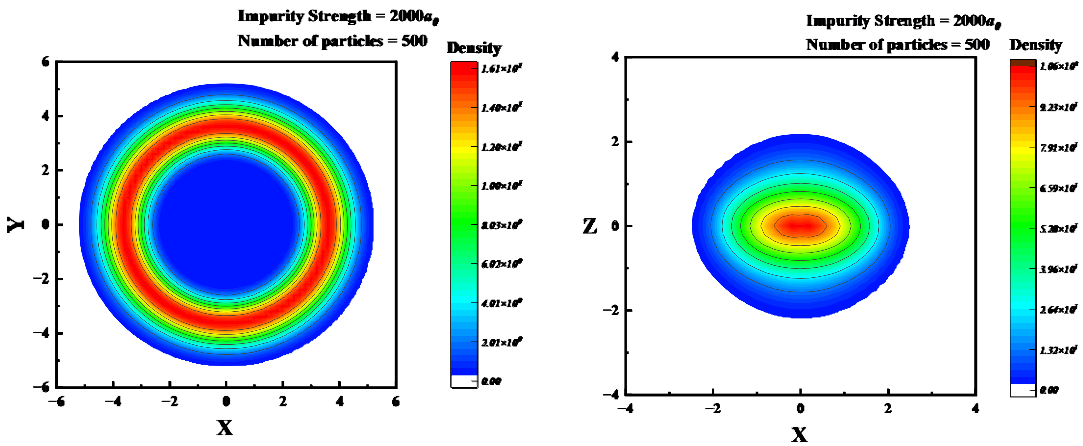

Fig. 27. *These panels show the density profiles of a 2D dipolar gas with an impurity implanted at the origin. The dipole polarization is always along the z-axis (left is perpendicular and right is parallel polarization). Note the anisotropy in the right-hand figure.*

After getting a good understanding of these codes, we put in a second impurity in our system and then calculated the energy of the system as a function of the distance between these impurities. The goal was to get an idea of the impurity-impurity interaction as well as any screening that the bath provides so we can compare this energy to the bare interaction. So far, the results for these calculations have shown overall very small energies, so we do not think these calculations are completely understood at this time. We have recently started similar calculations in 3 dimensions (3D), but also looking at time dependence. Here, we introduce an impurity and see how the system evolves in response to the introduced impurity. These were done in response to a new collaboration that started in October 2022, discussed below. Some early results are shown in Fig. 28, where one can see how the density evolves over time. Most recently, we started exploring making the 3D trap anisotropic. The limits are compressing the *z*-axis to make almost a 2D pancake geometry, and stretching the *z*-axis to make almost a quasi-1D system (cigar configuration).

The impacts of our work thus far have been fairly limited, as it is a new project. However, in Fall 2022, after a visit from a collaborator, we started collaborating with a researcher at the University of Hannover in Germany. They were looking at impurities in 3D dipolar Bose systems, but were approaching the problem with a different method than the GPE-based approach that we were developing. We thought our calculations would be complimentary and are working together on these 3D systems.

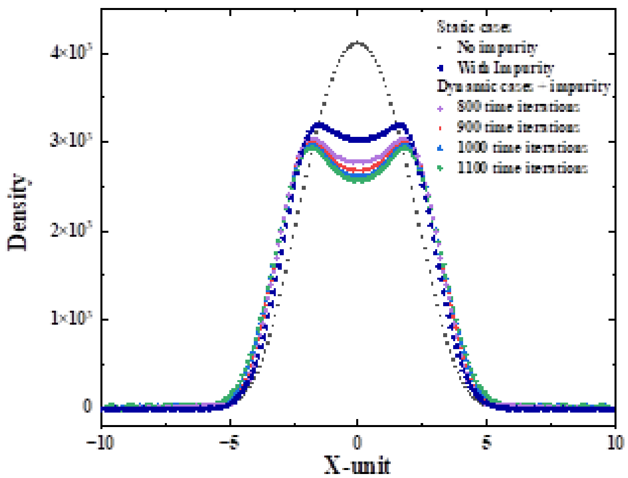

Fig. 28 *Number density of bosons in a 3D isotropic trap. Densities include the Gaussian-shaped density without an impurity, the static result with a repulsive impurity implanted at the origin, and the density evolution for several different time iterations. Note the ripples in the in the density towards the edges in response to the implantation of the impurity.*

In workforce development, FRG3 supported sending SI **Armstrong**'s postdoc, Neelam Shukla, to the ITEC conference in San Diego in July 2022. This is nuclear fusion research, which was a part of her PhD, and it is important for post-docs to stay in touch with their research community. Shukla also gave a poster there, which recognized EQUATE as funding source for her travel there. She will attend an IAEA conference (same subject, May 2023 in Vienna), and visit one of our research collaborators there.

The challenge has been the delayed arrival of this post-doc, six months later than planned. Progress is slower than we would like, as we have not produced a publication yet. However, this work will be shared in a presentation at the APS March 2023 meeting. A presentation was also given just at the end of the Year 1 period which was not previously reported; it took place at the 6th Conference on Nuclei and Mesoscopic Physics at Michigan State University (May 26, 2022). We are optimistic in submitting an article for publication by the end of Year2 with our new collaborators in Hannover.

Also, in outreach, **Armstrong** plans to be a judge for the Central Nebraska Science Fair (March 2023) and participate in the UNK APEs (Astronomy, Physics, and Engineering) summer camp in May 2023, for which EQUATE is contributing some funding.

In **Thrust 2. Quantum Computation,** under **Objective 2.a: Quantum Walk Algorithms**, for theoretical quantum computing, FRG3 has completed a manuscript titled "Quantum Search with a General Cubic-Quintic Nonlinearity," which is nearly ready for submission to *Physical Review A*. Continuing that work, we are looking at how entanglement evolves during quantum search algorithms. This work is led by SI **Wrubel** in collaboration with SI **Wong**, who remains on leave with the National Quantum Coordination Office until the end of 2023. In addition, one student gave a related poster presentation at a regional conference (SQUINT).

For **Objective 2.b: Crosswire Quantum Dots for Quantum Computing**, **Mei and Sabirianov** created a model quantum dot (QD) in cross-wire geometry. The ground state of this consists of two states controllable by the geometric parameters such as size of the QD. The numeric simulation code has been developed (Matlab) that solves time-independent Schrödinger equation for qubits made of single (or double) QD as well as 1D and 2D lattices of these. Time evolution of the electron states analyzed by time-dependent solutions.

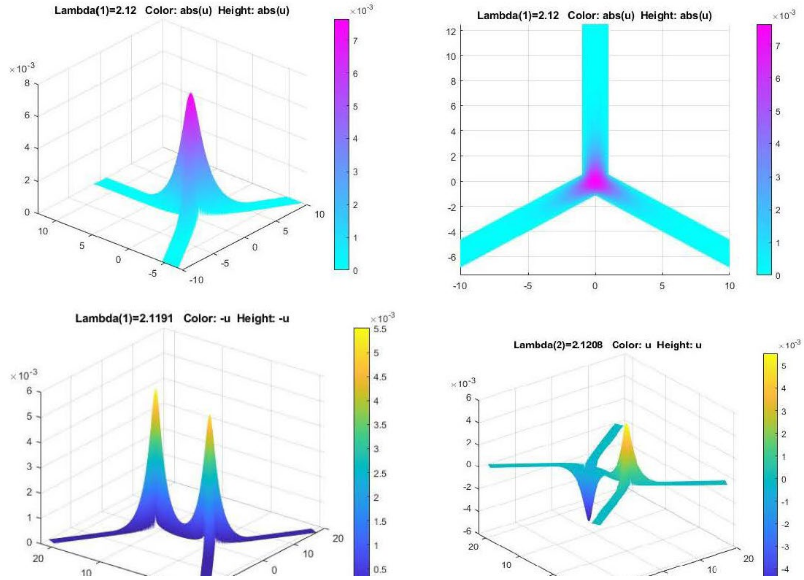

Fig. 29. *(top) Wave functions of the ground state of Y – junction (only quantized state) and eigenvalues (Lambda). There is strong localization of the state at the QD. (bottom) Wave functions of two quantized states of double wave Y – junction.*

SIs **Sabirianov** and **Mei** at UNO are working on the cross-road quantum dot systems, *i. e.* finite size 1-2 dimensional clusters in various geometries: first we computed their electronic properties, then proceed to study their interactions with external electromagnetic waves. (Fig. 29) On the experimental side, with interest from SI **Binek** of FRG1, we have had several research meetings and will continue this collaboration on mutual interest.

The graphene nanoflakes are optically active in the visible. An alternative is the isoelectronic hexagonal-BCN, where a nanoflake should be optically active in the near UV. Hexagonal BCN (h-BCN) has a larger band gap although the band gap should be smaller than the 5.6 eV of hexagonal BN. While h-BCN is isoelectronic to graphene, a nanoflake with a hole in the middle can be chiral and will be chiral relative to most substrates. The first principles density functional theory have been used to explore the structure h-BCN nanoflakes on Rh(111) and other face-centered cubic metals like Ir(111) and Ni(111) with the edges hydrogen terminated. From this, chirality is evident and is enhanced as the nanoflakes of h-BCN do not lay flat on the surfaces of all metals further breaking inversion symmetry significantly. We anticipate that such h-BCN nanoflakes should exhibit dichroism to circularly polarized light in the near VUV (Fig. 30).

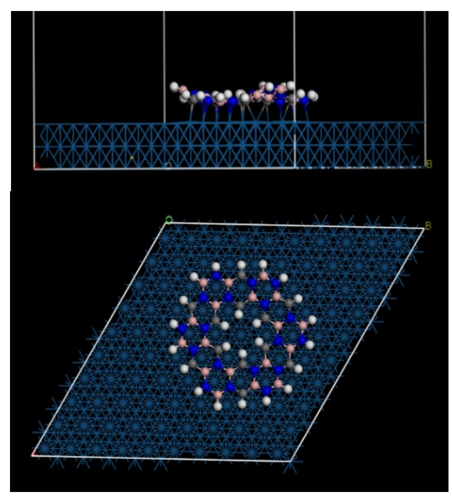

Fig. 30. *Side view and top view of Ir(111)-10x10-HCBN optimized structure. Three of six ends of the HCBN flake are attached to the Ir surface and three are tilted, showing the size of HCBN is a slightly larger than Ir lattice. But the adsorption is stronger than 2layer-Ir surface with 20 N-Ir or C-Ir bonds.*

Moiré superlattices have become promising platforms for studying emergent phenomena, such as strongly correlated physics and non-trivial topology in quantum materials. However, Moiré superlattices obtained by exfoliation and restacking via aligning/twisting van der Waals layers are typically small in size and accompanied by gradual spatial modulation or local domain formation. We analyzed the epitaxial growth of a hybrid covalent-van der Waals system Cr_5Te_8/WSe_2, with a thickness of Cr_5Te_8 down to a single unit cell and yet a size as large as 50 µm, by chemical vapor deposition (Fig. 31). Different from conventional Moiré systems, a fully commensurate, single-crystalline 3 × 3 (Cr_5Te_8)/7 × 7 (WSe_2) moiré supercrystal over the entire superlattice is achieved, through dative bond formation. This work has established a distinct epitaxial growth concept termed “dative epitaxy”, which could not only provide unrealized two-dimensional superlattices for exploring emergent physics, but also address the long-standing challenge of growing two-dimensional covalent materials and heterostructures with high crystal quality for semiconductor and other industrial applications.

Sabirianov, **Mei** and research associates at UNO work with Prof. Carolina Ilie and her colleagues at State University of New York at Oswego, and SI **Dowben** of UNL on the quantum dot nanoflakes of h-BCN on (111) surfaces of metals, such as Ni, Ir, and Rh. We performed structural optimization of all the adsorption structures and are in the processes of collecting their HOMO-LUMO gaps to write up the draft “The h-BCN Monolayers: Optically Active Graphenic Nanoflakes Graphenic Two-Dimensional BCN.” Also the group of FRG1’s **Binek** is currently in the process of fabricating cross-wire junction using 2D graphene or metal systems.

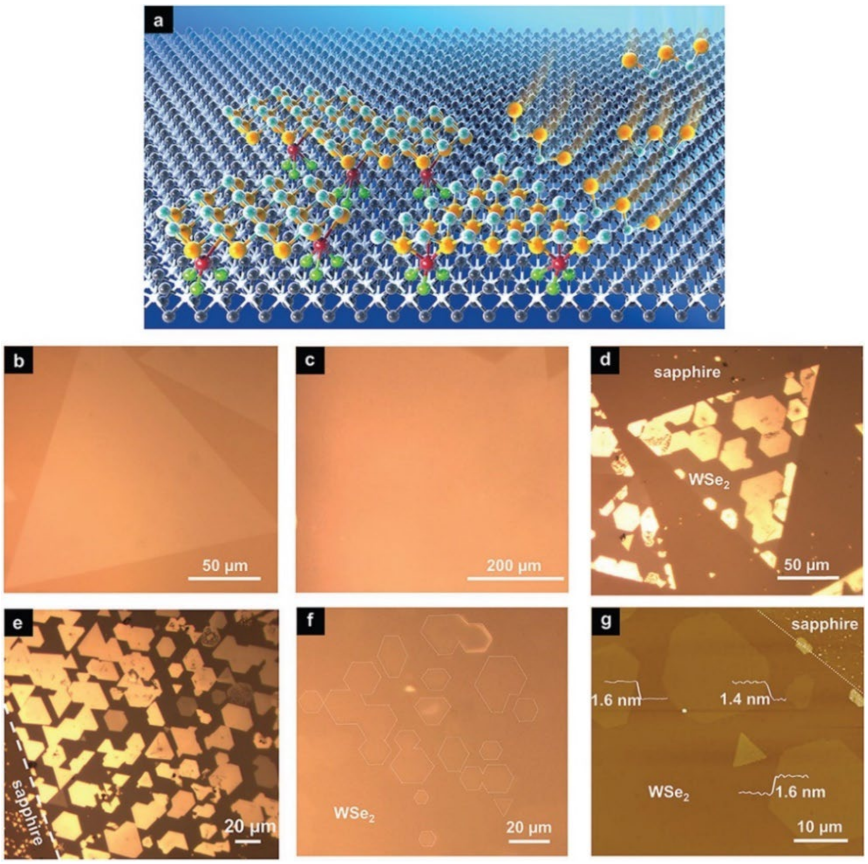

Figure 31. *Epitaxial growth process of Cr5Te8/WSe2 heterostructures, optical and atomic force microscopy images of WSe2 and Cr5Te8/WSe2 heterostructures. a) A schematic of the epitaxial growth processes of 2D Cr5Te8/WSe2 heterostructures, showing monomer adsorption, desorption, and diffusion. It also shows chemical bonding between interfacial Cr (red) and Se atoms (green). b,c) Optical microscopy images of monolayer WSe2 of sizes of ≈200 µm (b) and ≈1 mm (c); d) 2D Cr5Te8/WSe2 heterostructures; and e,f) highly aligned Cr5Te8 crystals with a thickness of ≈10 nm (e) and 1.4–2.8 nm (the dashed lines serve as visual aids to discern the boundaries of 2D crystals) (f) on a single monolayer WSe2. g) An AFM image of an area of one unit cell thick Cr5Te8 crystals; the dashed line shows the boundary between monolayer WSe2 and sapphire substrate.*

These Year 2 collaborations have impacted FRG3 with new ideas and research directions. **Mei** and **Sabirianov** plan international collaborations with researchers from Jordan, Germany, France, and Algeria with these collaborators visiting UNO in Spring 2023.

Grants from the Nebraska Research Collaboration Initiative are internal to the University of Nebraska, and only for one year; **Mei** and **Sabirianov** plan to recruit a research associate to work on DFT calculations and molecular dynamics simulation to investigate pathways and mechanisms of the catalytic processes, then develop preliminary results required to pursue outside funding; while preparing proposals to NSF, DOE, and NASA, **Mei** and **Sabirianov** are attending workshops organized by those funding agencies.

This FRG3 area is on schedule and exploring the feasibility of making novel qubit systems towards quantum computations. The novel geometries were motivated by possibility of using 2D systems with three-fold symmetry. Geometry strongly affected the electronic structure; for example, the change from cross-wire geometry (4-fold symmetry) to Y-shaped junction reduces the number of quantized states

In terms of Outreach/Dissemination, FRG3's UNO group actively involved six undergraduate and one graduate student in EQUATE research; two postdoctoral research associates also worked on the EQUATE project. They hosted a visiting research associate (Ahmad Alsaad) in Summer-Fall of 2023; he worked on several EQUATE-related projects. FRG3 also presented results from their EQUATE research at the March 2023 meeting of American Physical Society. Four presentations were given in 2022, and five more were submitted for 2023. Also, **Sabirianov** included EQUATE-related topics into UNO's Quantum Mechanics elective course.

Objective 2.c: Computational design of new spin-qubit materials

SI Aleksander **Wysocki** joined the EQUATE collaboration as a new hire, assistant professor at the University of Nebraska at Kearney in August 2022. Since then, his research activities were primarily focused on the computational design of new spin-qubit materials project. In particular, Wysocki has explored magnetic molecules and magnetic adatom systems for which an efficient quantum spin-light interface can be realized. Achieving a coherent spin-photon interface in spin-qubit systems is one the most important challenges in quantum information science technologies. The work has focused on the anionic $Eu(BA)_4$ molecule (see Fig. 32, left) that consists of an Eu^{3+} ion inside an approximate biaugmented triangular prism of oxygen ions. The oxygen ions are bonded to aromatic hydrocarbon ligands. The electronic structure of the complex was calculated using *ab initio* multireference quantum chemistry methods including spin-orbit coupling (CASSCF+CASPT2+RASSI-SOC). The work was done in collaboration with Professor Kyungwha Park from Virginia Tech. The calculated ground state is the Eu $4f^6$ $^7F_0$ multiplet. Using light, the molecule can be optically excited into the $^5D_0$ multiplet (see Fig. 32, right). The calculated excitation energy is 2.29 eV which is in excellent agreement with experimental measurements. While both $^7F_0$ and $^5D_0$ electronic states are nondegenerate, the Eu nuclear spin (I=5/2) can be used to encode spin-qubit states.

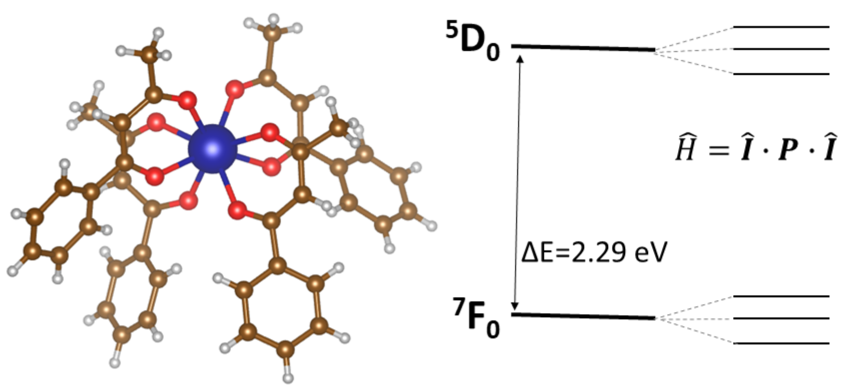

Fig. 32 *(Left) atomic structure of the $Eu(BA)_4$ molecule. Blue, red brown, and, grey spheres represent Eu, O, C, and H atoms, respectively. (Right) ground $^7F_0$ and excited $^5D_0$ multiplets calculated for the $Eu(BA)_4$ molecule. The nuclear quadrupole interaction split the electronic states into electronic-nuclear states.*

Importantly, since $^7F_0$ and $^5D_0$ multiplets have a zero electronic magnetic moment, the magnetic hyperfine interaction does not contribute to decoherence. One can then expect long coherence time qubit states that can be detected and coherently manipulated by optical means. In future, the nuclear quadrupole coupling parameters will be calculated for the molecule from *ab initio* and an effective spin Hamiltonian will be diagonalized to evaluate the electronic-nuclear spectrum. The oscillator strengths for the transitions between the $^7F_0$ -$^5D_0$ electronic nuclear states will also be calculated. Further, the possibility of tuning the molecule properties by an external electric field will be explored.

Wysocki has also established a collaboration with EQUATE members **Guo** (FRG 1) and **Mei** (FRG 3) to study spin waves and Raman spectra in Boron-doped Cr_2O_3. Wysocki and other EQUATE researchers have submitted a proposal to NSF on a topic of quantum sensing which is relevant to EQUATE. The proposal is titled, "QuSeC-TAQS: Probing quantum entanglement in correlated spin systems via qubit spectrometry and spin correlation measurements." EQUATE Science Director **Binek** is the lead PI, with **Hong**, **Kovalev**, **Laraoui**, and **Wysocki** as co-PIs. In addition, on November 7 **Wysocki** gave a Sigma Xi Science Café talk in the Kearney, Nebraska community on next-generation materials for quantum information science, including discussion of his EQUATE research.

For FRG3's notable <u>Changes</u>, <u>Accomplishments</u>, <u>Impacts</u>, and <u>Outreach/Dissemination</u> for Year 2, the addition of SI Wysocki factored positively in each of these categories.

Education, Outreach and Workforce Development (2022-2023)

The goal of the EQUATE Workforce Development Portfolio is to prepare the next generation of highly qualified and motivated students, educators, and researchers through comprehensive programs, activities, and mentoring through the duration of the grant. Among this area's advances in Year 2, the EQUATE team established a new logo to more clearly and energetically convey the quantum research addressed. The new logo appears on the cover of this report, as well as the EQUATE website and other program info.

<u>**Objective 4.1: Equip Nebraska students (grades 6-12) with resources for success in STEM workforce.**</u>

YNS Summer Camps. Year 2 was an exciting time for Nebraska EPSCoR's Young Nebraska Scientists (YNS) summer camps, with six faculty-led camps planned for the season. Two new camps were offered directly through YNS including Discovering Food Science: How the Cookie Crumbles (a residential high school camp), and Engineering Plant Cell Walls (a middle school day camp). Additionally, Renewable Energy Technologies, originally planned as a virtual high school camp, was redesigned with the goal of creating a mobile camp for middle school students. This inaugural test run of "Camps on the Go" was held in Columbus, NE – a hub for many rural Nebraskans. YNS summer camps were attended by 128 students (46% female and 34% URG). Ninety-one campers were exposed to EQUATE-specific topics. **Metrics: Yr 1 development, Yrs 2-5, 85 participants (30 at EQUATE-specific summer camps) annually. (meets metric)**

YNS High School Researchers. YNS, in conjunction with the Nebraska Center for Materials and Nanoscience (NCMN) and the JA Woollam Foundation, were able to host 15 students in laboratories at UNL. The students were hosted by EQUATE researchers including **Lai, Laraoui, Streubel, Bao, Guo**. Additionally, three students were mentored in Omaha; 2 students participated in the program as part of the Wong Laboratory on the UNO campus and 1 student was hosted by **Wrubel** at Creighton University as part of the Haddix STEM Corridor program. Sixteen of the students presented posters at the UNL Summer research Fair; while 4 HSRs presented at EQUATE's external review panel--impressing researchers, graduate students, and panel members alike. Overall, 59% of HSRs were female and 41% were from underrepresented groups in STEM. **Metrics: Yrs 1-5, 8 students annually. (meets metric)**

Mobile Labs. During the second year, three quantum-themed mobile labs were deployed providing hands-on activities and education materials to teachers throughout the state of Nebraska. The EQUATE kits were created to work at both the middle and high school level and include topics such as spectral analysis, tribo-electric series, and particle wave duality. Additionally, a module was developed using the RAIN program for a citizen science-based mobile lab. The molecular biology for secondary classrooms (MBSC) mobile labs (pGlo Bacterial Transformation, Restriction Enzyme Digestion of Lambda DNA, and PV92 PCR) continued to enhance secondary educational curriculum. In the 2022-2023 school year, mobile labs have reached approximately 800 students so far with 31% URG and 52% female. The schools include Omaha

Westside, Minden, Lincoln, Kearney, Louisville, and Oxford High Schools. **Metric: 1,000 students/yr with 3 EQUATE-specific kits offered during Yrs 2-5. (meets metric)**

Remotely Accessible Instrument (RAIN) Program. The RAIN program was developed as a collaboration between 4-H representatives and the NCMN. Staff turnover through the 4-H program has been extreme during the past 2 years. The original plans and relationship needed to implement previously planned RAIN activities will need to be rebuilt from scratch. EQUATE outreach has repackaged the RAIN program as an easily implemented mobile lab to collect data about metals in soil and vegetables throughout Nebraska using X-ray Fluorescent (XRF) spectrometry. The XRF has been repaired and staff are trained in anticipation of the new activities. **Metrics: Yr 1 development; Yrs 2-5, 5 events annually. (on track to meet metric)**

Teacher Programs

Teacher development is an important part of equipping Nebraska's students for STEM success. Beyond the mobile labs program, additional opportunities are included within EQUATE to provide teachers with the knowledge and skills necessary to prepare the next generation of STEM professionals.

Research Experience for Teachers (RET). Two area physics educators participated in the RET program through the NCMN: Kendra Sibbernsen from Metropolitan Community College and Chris Deeter from Lincoln Lutheran High School. The participant numbers were lower than expected even after investing significant time and effort into recruiting activities at events such as Nebraska Association of Teachers of Science (NATS), Nebraska Alliance for Conservation and Environment Education (NACEE), the Nebraska Summit on Math and Science Education, and the Lincoln Public Schools Teacher Connector. The low number of participants seemed to be a common issue within EPSCoR jurisdictions and beyond, as educators have been facing many challenges stemming from the COVID pandemic and associated staffing shortages. **Metrics: Yrs 1-5, 5 participants annually in EQUATE labs. (revamping efforts to meet metric)**

Summer Institute for Middle School Teachers (SIMST). Nine teachers participated in the 2022 SIMST. The program was held virtually so teachers could participate from the comfort of their own homes. Boxes of supplies for all activities were shipped to the participants beforehand along with instructional materials. The teachers worked together with direction from **Wignall** and **Phan** to perform quantum-related activities designed specifically for middle school students. The virtual format was popular for this program and allowed us to reach teachers outside of the Lincoln metropolitan area. **Metrics: Yr 1 development; Yrs 2-5 10 participants annually. (meets metric)**

<u>**Objective 4.2: Continue influencing the STEM pipeline through programs for undergraduate and graduate students, and postdoctoral researchers.**</u>

Postdoctoral Scientist Mentoring Program. Eight postdoctoral fellows (1 female) worked for EQUATE in Year 2. **Metrics: 35 postdoctoral scientists, (7 per year). (meets metric)**

Graduate and Undergraduate Student Mentoring. Twenty-nine graduate students (34% female) and 18 undergraduate students (46% female) worked in EQUATE laboratories in Year 2. **Metrics: 16 grad, 18 undergrad (cumulative 90 Undergrad and 80 grad). (meets metric)**

Student Seminar Series. This monthly seminar series brings together all EQUATE participants to hear short, informal presentations from postdoctoral researchers, graduate, and undergraduate students. It provides an opportunity for EQUATE trainees to improve their presentation and science communication skills. The series shares ongoing research within each FRG, and stimulates inter- and cross-FRG collaboration.

Fig. 33 – *A digital ad / flyer promoted the 2022 talk by UNL alumnus Rob Buckley, who works on Quantum Computing with Google.*

Year 2 of EQUATE brought some exciting presenters to the seminar series. UNL physics alum Rob Buckley (Fig. 33) met with students and faculty to discuss his quantum computing work at Google. EQUATE's **Bao** presented his work using novel optical material platforms. EQUATE collaborators from India, Korea, and Purdue presented about machine learning techniques for quantum material discovery and metamaterials. Several graduate students are slated for the spring session of presentations. **Metrics: 12 meetings per year. (meets metric)**

Nanoscience/Quantum Class, Minicourses, and Training. Introduction to Quantum Materials and Technologies (MATL 492/892) was held for the second time for both undergraduate and graduate students for spring 2023 with recruitment across several departments including Physics, Chemistry, Mathematics, Electrical & Computer Engineering, and Mechanical & Materials Engineering departments. This course was developed by **Laraoui** and has 12 students registered (2 female). EQUATE Seed Grant awardee **Wang** developed a new syllabus for course ECEN-491/891: Quantum Communication and Quantum Computation and introduced a quantum programming module that exposes the students to real-world quantum computation systems, leveraging the IBM quantum resources. The class was designed to run alternate semesters from the introductory course with 16 students (two undergraduates and 14 graduates) enrolled in this course, and four of them were female students. Additionally, **Argyropoulos** introduced EQUATE topics, research, and opportunities to undergraduate and graduate students in Electrical and Computer Engineering (ECEN 306). **Metric: Cumulative 35 grad students, 250 participants. (on track to meet metric)**

Research Experiences for Undergraduate (REU) Students. EQUATE funding supported four 4 REU students (25% female) during the summer of 2023 with SIs **Hong**, **Xu**, and **Laraoui**. One student, Jackson Savage, had previously attended a YNS summer camp as a middle school student (that 2015 camp on microscopy was led by EQUATE investigators **M. and E. Schubert** during Nebraska EPSCoR's 2010-2016 Track-1 project). It was rewarding that a former YNS camper is pursuing nanoscience and quantum research as a young adult. Central Community College was unable to recruit a student for summer research this year. However, two students were funded to perform summer research in the Department of Chemistry at the University of Nebraska Kearney through the Small Colleges Research Experiences program. **Metric: Yrs 1-5, 3 students per year in EQUATE laboratories. (meets metric) Metric: Yrs 1-5, 1 student annually at Central Community College. (revamping efforts to meet metric)**

Teaching and Research Equipment (TRE) Grant. The TRE was created to assist with the replacement of outdated equipment and/or acquisition of new equipment to enhance education and research activities at small colleges throughout Nebraska. The grant covers up to $50,000 with a 20% institutional match. The inaugural year began with a request for proposals during fall of 2021, with equipment in place by May 2022. The Nebraska EPSCoR State Committee approved five applications at institutions including Chadron State College, Midland University, Southeast Community College, Doane University, and Nebraska Wesleyan University. This program is expected to impact approx. 968 students and 25 faculty members during Year 2 of EQUATE.

Objective 4.3 Provide professional development opportunities for faculty across Nebraska's colleges.

FIRST Award Program. Nebraska EPSCoR's annual **FIRST Awards** program provides assistance to this state's early-career, tenure-track faculty in the amount of $25,000 (with an equal match required from each recipient's academic department). The grants are designed to help early career faculty initiate their research programs and compete more effectively for NSF CAREER Awards. In 2022, Nebraska EPSCoR awarded 7 recipients (exceeds metric). The 2022 proposals were reviewed on November 2nd by Nebraska's State Committee for final selection of funding. **Metrics: 6 awards annually. (meets metric)**

Faculty Mentoring through EQUATE Seed Grants. This program solicits seed proposals requesting one year of funding through a university-wide call for proposals. Seed proposals should be tightly focused on a particular research topic in the area of quantum science and technologies, and typically involve one or two PIs at the tenure-track assistant professor level. Projects that address quantum phenomena relevant to the current EQUATE FRG research themes are especially welcome. Funding for seed projects is limited to $56K/year (total cost). **Metric: Yr 1: 2 awards, Yrs 2 -5: 1 award annually. (meets metric)**

Four proposals were submitted for consideration, and two were selected for funding in November 2022 (exceeds metric). The proposals chosen were:

- **Mohammed Ghashami** of UNL Mechanical and Materials Engineering proposed "Quantum Size Effect on Radiative and Electronic Transport." Mohammed will work with UNL Mechanical and Materials Engineering's Abdelghani Laraoui, EQUATE SI and FRG2 leader, on this research.
- **Martin Centurion** of UNL Physics and Astronomy proposed "Characterization of the Ultrafast Structural Response in Photoexcited Materials." He will work with various EQUATE SIs on this project.

Objective 4.4: Increase the number of underrepresented minorities in STEM fields.

In summer of 2022, Nebraska EPSCoR supported 12 participants in the **Munroe Myer Institute's High School Career Explorations** summer camp. This camp was designed to expose Native American high school students to health science professions such as Recreation Therapy, Physical Therapy, Occupational Therapy, Severe Behaviors, Psychology, Pediatric Feeding Disorders, Genetics and Genetic Counseling, Speech Pathology, Neurodevelopmental Disorders, and Developmental Disabilities. The camp was held as two one-week sessions. The first was for commuter students in the Omaha area and the second was a residential camp for tribal students from outlying regions.

The **2023 Women in Science Conference** is designed to expose high school students to a wide variety of science-related fields and encourage them to pursue majors and careers in science, technology, engineering, and mathematics, through the work of women currently working in the sciences. The conference will take place in April. **Metric: 90 students and 10 teachers (on track to meet metric)**

All Girls All Math (AGAM) Camp, a weeklong virtual camp, was attended by 17 students (94% female, 71% URG) from all over the United States. Students were able to learn about cryptography and coding, experience virtual tours of both national and local museums, and participate in small group activities such as virtual escape rooms which challenged participants to use mathematical reasoning, problem solving skill, and communication ability. The camp ended with a virtual scavenger hunt designed by the undergraduate chaperones that led students through UNL campus landmarks.

Education Services Unit (ESU) 11's **Summer Honors Program** was designed to mitigate the disparities in education and activities available to rural students as compared to their urban counterparts. Over 80 rural high school students (56% female and 36% URG) were exposed to advanced topics in biology, physics, engineering, and mathematics during the two-week summer experience. Additionally, Nebraska EPSCoR is partnering to implement **Family Math Game Nights** in Chadron, Nebraska to increase math readiness of rural elementary students and **Professor Sko's Zoom Labs** to teach rural and underserved elementary students about physics.

Nebraska EPSCoR provides support for five student researchers from the state tribal colleges to attend the **Nebraska Academy of Sciences**. The meeting is held yearly in April and is expected to take place as usual. **Metric: 5 students at NAS (on track to meet metric)**

College prep groups (NCPA, Upward Bound, Girls Inc, etc). Metric: 100 URM/yr: low income, first gen

Ten young women from Lincoln's **Girls Inc Eureka!** Program visited the NCMN for a tour and hands-on quantum activities led by **Wignall** and **Pham**. An additional 10 students, all female, attended a similar activity as part of the Big Red 4-H summer camp. **Sangster** along with Civil and Environmental Engineering faculty hosted 17 women from Omaha's Girls Inc Eureka! Program on the UNO campus to learn about chemistry and water quality. **Wignall** and **Sangster** shared quantum related topics and hands-on activities with 11 **Upward Bound** students (64% female) during a three-day minicourse held at UNL.

Wignall, **Sangster**, and **Pham** were able to participate in a repeating afterschool program held at five Title 1 middle schools in Lincoln, Nebraska. Each semester, EQUATE staff spends time with students teaching them about quantum topics and doing hands-on activities (**Fig. 34**). This program is organized by the Office of TRIO at UNL which strives to support low-income, first generation, and/or disabled students so they may reach their full potential. Through this program, EQUATE outreach staff impacts over 350 students; 86% are first generation students and 67% are from low-income families. During summer of 2022, 65 students from the program who had completed middle school were invited to UNL for a summer camp and visited the NCMN for tours and activities led by EQUATE researchers. Students learned about quantum mechanics using cat memes and videos (**Bao**), chemistry and DNA (**Lai**), and experienced sound waves as dancing flames (**Binek**).

The College of Engineering started **Discover Engineering Days (DED)** to introduce middle school students (grades 6-8) and their teachers to fields in engineering, physics and the science programs University of Nebraska-Lincoln offers. Hosted by the college's outreach organization the events are filled with hands-on activities that utilize engineering habits such as collaboration, problem solving, creative, and critical thinking skills. DED offers seven workshops over the course of the year, averaging approximately 130-150 students at each meeting. EQUATE has been involved with DED throughout the year, teaching the students about Triboelectric Energy and how it relates to Quantum. Students involved in DED come from both rural and urban schools, with strong representation of URM and female attendees.

Figure 34. *EQUATE's Summer 2022 activities, clockwise from top left: Middle school students learn nanoscience in Columbus, Nebraska; Upward Bound students gain light frequency knowledge from EQUATE outreach representatives; YNS High School Researchers visit Lincoln's JA Woollam Co.; REU students (minus one, who had COVID-19) enjoy orientation at UNL.*

__Objective 4.5 Increase public awareness of EQUATE progress__

After success with EQUATE's first annual conference (on Technology Commercialization) in Year 1, the Year 2 event is planned for March 17 with the topic: Topology and Valley-Driven Quantum Phenomena (planned by FRG1's Hong). Eighty attendees and 30 posters are expected for the Year 2 event. Future conferences will be led by other FRGs in turn, including EQUATE's Outreach team. **(meets metric)**

Management Team (MT) Meetings continued in each month of EQUATE's Year 2; seminars have added renewed energy with inter-FRG discussions as a focus, along with presenters.

In Year 2 the number of **journal papers and presentations** (four per EQUATE investigator) metric was exceeded with 61 published journal papers and 52 conference presentations, as of February 2022.

Nebraska EPSCoR's **Annual PR Report** (**Fig. 36**) for 2022 featured EQUATE research on its cover. More than 1,000 copies of this publication were printed, with distribution to a mailing list of 800 recipients, plus a PDF version is posted at NE EPSCoR's website, epscor.nebraska.edu. **(meets metric)**

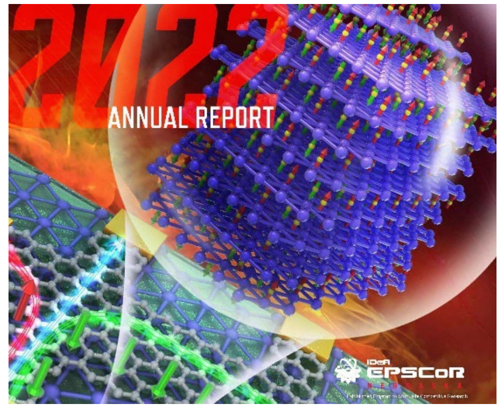

Fig. 35. The cover image from Nebraska EPSCoR's annual report publication--depicts research by EQUATE FRG1 that was featured in the journal, Advanced Materials (reproduced with permission). This collaboration studied spin-dependent electronic transport in graphene, specifically magnetoelectric antiferromagnetic thin film. They observed a large spin relaxation length, which is promising for developing new transistors — attracting international attention.

The EQUATE **website** (equate.unl.edu) continued to grow, with 2,216 users making 6,223 pageviews from Feb. 1, 2022 - Jan. 31, 2023. This exceeds the metric of 10% growth year over year, compared with Year 1 (578 users). EQUATE's **social media** presence, tagged by #NebEQUATE, had an active year of >55 mentions, above the Year 1 baseline of 50 mentions – celebrating journal publications, outreach opportunities, and other achievements via EQUATE. **(meets metric)**

In EQUATE's Year 2, the Outreach Team recalibrated its metrics re: **Tours, museum exhibits and public event participation** in light of the pandemic. The focus became two museum placements of the Nano exhibit (rather than attendance numbers, which may not be tracked at some placements), and the revised metric was met. EQUATE facility tours reached 152 guests **(meets metric)** and EQUATE continued to have a presence at public events, including the Nebraska Association of Teachers of Science **(meets metric).**

DATA MANAGEMENT

As EQUATE's research productivity is ramping up in Year 2, a variety of data was generated, disseminated, exchanged, and managed. Data are primarily produced by computer-controlled systems in the labs of all FRGs. This includes imaging techniques, like NV-center spectroscopy in FRG2, computational data from density functional theory calculations in FRG1 and FRG3, and micromagnetic simulations of non-trivial topological spin textures in FRG2. Additionally, data is also obtained through surveys and assessments carried out by the E/O activities. The data generated have various formats, including decimal floating-point numbers, ASCII sequences, txt format, TIFF and JPEG, along with metadata. The SIs have the flexibility to decide on certain aspects of metadata format and raw data exchange. As of today, EQUATE is at a point where storage of data and metadata on UNL's OneDrive, a cloud-based storage system, with data access to all participating institutions is virtually achieved.

However, a new challenge emerged in 2022. Over the summer of 2022, the president of the University of Nebraska released the executive memorandum No.16 (https://nebraska.edu/-/media/unca/docs/offices-and-policies/policies/executive-memorandum/policy-for-responsible-use-of-university-computers-and-information-systems.pdf) dealing with the responsible use of university computers and information systems. The long-term goals expressed in Memorandum 16 affect the ease by which data can be accessed, exchanged, and stored—in particular with more stringent rules for the individual labs of the SIs, and especially with regard to data storage and exchange on systems other than university networks. Remote control of experiments needs to be revisited, as well as the way in which data are stored on individual computers in labs and transferred from there. This is an ongoing process, which is expected to take a couple of years to complete. The argument that EQUATE, as an NSF-funded center, is paid for by the taxpayer and thus generates data which belong to the public, will not overturn the stringent safety measures which will be implemented by the University of Nebraska's Information Technology Services (ITS). Changes, which involve data management of EQUATE, may be requested by ITS and are not negotiable by EQUATE members. EQUATE members will engage with ITS representatives about the practicality of their measures but strictly follow all requests made by ITS. As a result, significant change in the data management should be anticipated. The details of such measures (and possible costs associated) will become clear in subsequent years in collaboration with ITS.

SUSTAINABILITY

EQUATE continues to make significant progress towards sustainability. As outlined in the strategic plan, the research and collaboration carried out by EQUATE serves as a foundation for new proposals that extend beyond the lifetime of the organization. A notable success story has been Wei **Bao** (FRG 3) earning an NSF CAREER Award, based on his research conducted on Bose-Einstein condensates of quasiparticles in solids with applications in quantum emulation. His work in this field turned into one of the major highlights of EQUATE achievements in 2022. In last year's report, an NSF major research instrumentation (MRI) proposal was submitted and under review. The MRI proposal, led by EQUATE's scientific director (**Binek**), with significant contributions from the leaders of FRG2 and FRG1 is now funded and will equip EQUATE with an NV center scanning probe microscope accessible to all EQUATE researchers and housed and maintained by the Nebraska Center for Materials and Nanoscience. This instrument will greatly enhance the work done in EQUATE FRGs by using high-resolution quantum sensing to image quantum states of matter such as persistent current edge stages in topological insulators, spin textures, single molecules, magnetic domain walls, and more. An NSF pre-proposal entitled "QuSeC-TAQS: Probing quantum entanglement in correlated spin systems via qubit spectrometry and spin correlation measurements" has been developed by EQUATE SIs from all three FRGs including the new hire and FRG3 theorist SI Aleksander **Wysocki**, EQUATE scientific director and FRG1 SI **Binek** as well as FRG 1 leader Xia **Hong**, FRG1 theorist SI Alexey **Kovalev**, and FRG 2 leader Abdelghani **Laraoui**. If the team is invited to write a full proposal and is funded, the work of EQUATE on the topic of quantum sensing can be extended in scope and duration beyond the lifetime of EQUATE.

A sustainability opportunity remains to be provided by UNL's Grand Challenges, a set of eight themes for the University of Nebraska-Lincoln, providing institutional support of $40 million over a five-year period. EQUATE has been instrumental in motivating UNL leadership to select Quantum Science and Engineering as one of the Grand Challenges themes. EQUATE's scientific director, Christian **Binek**, was not only instrumental in formulating this grand challenge theme but also became a member of the steering committee approved by the chancellor's executive leadership team. Under leadership of Dr. **Binek** and Dr. Susan Hermiller (Mathematics UNL), a team of 30 UNL faculty was assembled that developed and submitted a catalyst proposal titled "Quantum Approaches Addressing Global Threats" in the framework of the UNL Grand Challenges. Many EQUATE SIs--including the leaders of FRG1 and FRG2, and CAREER Award recipient SI **Bao**, had a leading role in this proposal. The proposal got excellent and very good ratings but was not funded in the first round of proposal. However, the team plans to resubmit a revised proposal in the 2023 competition. In fact, funding of the grand challenge proposal later in the lifetime of EQUATE is beneficial to better fulfill the sustainability role as a grant which continues and extends the work on quantum materials and technologies initiated by EQUATE.