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

Year 1 Narrative Report – February 2022
Nebraska EPSCoR

The EQUATE team--comprised of engaged researchers, collaborators, advisors, partners, administrators and education and outreach (E/O) coordinators--rolled up their sleeves and began their work in accordance with the project's NSF-approved Strategic Plan.

The progress made within the first year showcases EQUATE's place at the forefront of quantum materials science and technologies by advancing the second quantum revolution through discoveries, innovation, and workforce development. The Year 1 (Y1) results exemplify the way that all three Focused Research Groups (FRG) of the center leverage an interdisciplinary multi-campus interdepartmental approach. Much of the experimental work done in FRG 1 includes complex oxide quantum materials and heterostructures. Their strong electron-electron correlation accompanies potentially useful functionalities such as voltage-controlled Néel vector switching and tunable phase transitions.

In addition to magnetoelectric thin films, molecular magnetoelectrics have been synthesized and characterized in an interdisciplinary approach involving chemistry (synthesis) and physics (characterization). Also, two-dimensional (2D) van der Waals quantum materials and heterostructures of complex oxide/2D materials have been fabricated and investigated with emphasis on their unique electronic properties useful for spintronic devices and applications in information technology.

A hallmark of EQUATE's work has been its collaborative approach. It includes theory guided experimentation on one hand, and interdisciplinary experimental work at the interface between physics, chemistry, and engineering realized in cross-FRG collaborations on the other hand. Theoretical work in FRG 1 resulted in the prediction of a persistent spin texture in a ferroelectric complex oxide, a new form of a micromagnetic topological Hall effect and topological phase transitions in magnetic nanostructures, and micromagnetic simulations of non-trivial spin textures which include skyrmions with potential application in future information technological applications such as memory.

There has been vivid interaction between FRG1 and FRG2 already in this early stage of EQUATE. For instance, magnetic thin film growth optimized in FRG1 has been applied in FRG2 for Nitrogen-Vacancy (NV)-center spectroscopy which ultimately enables entanglement of spin qubits via magnons. Another successful start into interaction between FRGs with involvement from engineering, physics and chemistry concerns the work on quantum sensors for low field magnetic resonance spectroscopy. Here an NV setup for low field magnetic resonance has been developed and employed to investigate iron in biomolecules. This work has important implications for understanding the electron transport chain in mitochondria.

To advance the characterization abilities of solid-state qubits, FRG2 senior investigators (SIs) developed and successfully tested the first THz-EPR (Electron Paramagnetic Resonance) ellipsometer, paving the way for spectroscopy in the virtually uncharted THz domain. Theoretical breakthroughs have been achieved in the creation of entangled flying qubits via parametric down conversion in nanophotonic structures. Entangled photons are a critical component in the realization of quantum communication applications.

FRG3 has taken tactical steps towards quantum emulation and simulation, with major equipment purchases allowed to upgrade the magneto-optical potassium traps paving the way for quantum emulation in Bose-Einstein condensation of ultracold atom gases. At the same time, FRG3 demonstrated significant progress in a complementary solid state experiments emulating quantum systems in a Bose-Einstein condensate of exciton-polariton quasiparticles. These impressive results were the scientific basis for the NSF early CAREER award for EQUATE FRG3 SI Bao. A poster child example for cross-institutional support and sustainability has been the no-cost loan of a leak detector from the Nebraska Center for Materials and Nanoscience at UNL to the FRG3 leader at Creighton University.

New faculty hires at the University of Nebraska-Lincoln (UNL) and the University of Nebraska at Kearney (UNK), fostering synergy between the FRGs, are in full swing with in-person interviews and colloquia starting as early as February 2022. EQUATE has provided seed grant funding for two rising stars of quantum science. In addition, a monthly seminar series attended by all SIs, graduate students, postdocs, and undergraduate students has been successfully launched early this year and continues on a monthly basis. Currently it takes place in a hybrid format with about 50% in-person participation and 50% online participation.

EQUATE's web-presence is now fully established (https://equate.unl.edu/welcome) and the project's Education and Outreach (E/O) and workforce development are progressing as proposed. Prominent among E/O accomplishments, the first EQUATE-organized Nebraska Research and Innovation Conference (NRIC) is in preparation and will take place April 14, 2022. The conference addresses "Commercializing quantum technologies in Nebraska: From Research to Licensing" with contributions from a diverse group of renowned quantum scientists and entrepreneurs, NUtech Ventures, the technology commercialization affiliate of the University of Nebraska, and presenters from the Nebraska Business Development Center.

Furthermore, tangible steps towards education in quantum information science and workforce development have been initiated by development of a new class MATL 492/892: Introduction to Quantum Materials and Technologies. This class will be given to students every spring semester the next 5 years, including Spring semester of 2022 (January-May). In addition to the many successful E/O projects there is already a tradition of vibrant exchange between the various EQUATE campuses. Examples include UNL interacting with Creighton University through equipment exchange; with University of Nebraska at Omaha (UNO) via an in-person presentation on "Science and Technology Harnessed for National Security" for a group of congressional staff; and with UNK via an in-person Science Café organized by the local Sigma Xi chapter and UNK's Department of Physics on "Emergent Quantum Materials and Technologies: What are they and why should I care?" and through the monthly seminar series.

The project's first year has seen the initiation of all important tasks without substantial delay. EQUATE expects to continue on this path.

FRG1 – Quantum Materials

For **Thrust 1, Objective 1a**, SI **Binek** deposited Cr_2O_3 and boron-doped Cr_2O_3 ($B:Cr_2O_3$) thin films on V_2O_3 electrodes and using these heterostructures to fabricate Pt-based Hall devices via photo-lithography. The team successfully reproduced previously published switching results in B-doped Cr_2O_3 and confirmed that the polarity of the applied voltage determines the switching characteristic. These positive results enabled the investigation of boron-diffusion via temperature-dependent magnetotransport measurements. Meanwhile, collaborators at NIST provided data on neutron depth profiling on the B-doped films for various temperature protocols. Their data confirmed the hypothesis that Boron tends to diffuse towards the film surface for temperatures significantly higher than room temperature. Binek group also fabricated Cr_2O_3 and $B:Cr_2O_3$ films for collaboration with other SIs. SI **Laraoui** (FRG2) group utilized NV-center microscopy to confirm surface magnetic properties unique to magnetoelectric antiferromagnets. SI **Guo** group probed the phonon modes of $B:Cr_2O_3$ and observed pronounced changes in the spectral response upon B doping and compared with that of pristine Cr_2O_3. In collaboration with SI **Dowben**, the team also studied the effects of chromia substrates on the magnetism of Pd. (meets metrics)

Two-dimensional (2D) antiferromagnetic van der Waals (vdW) materials such as $CrCl_3$ are promising material candidates for developing antiferromagnetic AFM spintronics. SI **Hong** group fabricated for the first-time high quality $CrCl_3$ flakes on mica using physical vapor transport approach. **Figure 1** shows the Raman spectrum taken on a typical $CrCl_3$ flake, which reveals four E_g peaks (115.5 cm^{-1}, 207. 1 cm^{-1}, 247.1 cm^{-1}, and 345.2 cm^{-1}) and two A_{1g} peaks (164. 9 cm^{-1} and 299.8 cm^{-1}), consistent with the reported results for monoclinic $CrCl_3$. Raman studies performed after three months shows that thick samples are stable in the ambient condition. High resolution transmission electron microscopy (HRTEM) studies conducted by Jeffrey Shield group at University of Nebraska-Lincoln further confirms the high crystallinity of these samples. Our ability to fabricate large $CrCl_3$ thin films can facilitate their implementation in spintronics devices. (meets metrics)

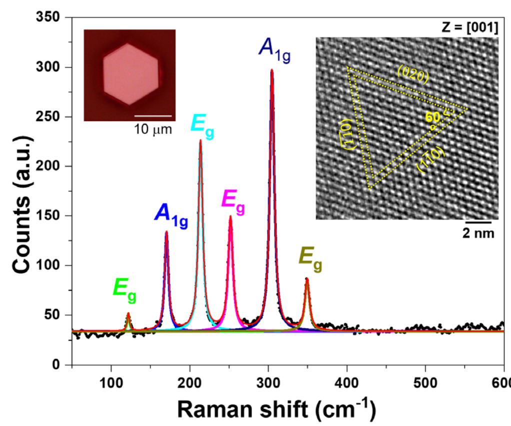

Figure 1. *Raman spectrum taken on a $CrCl_3$ flake with fits. Insets: (left) Optical image and (right) HRTEM image of $CrCl_3$ flakes.*

For **Thrust 1, Objective 1b**, SI **Tsymbal** predicted that orthorhombic $LiTeO_3$ with *Pnn*2 (No. 34) structure can host the persistent spin texture (PST) based on the general symmetry considerations and using the Material Project. The lattice constants and atomic coordinates of bulk $LiTeO_3$ are obtained using full structural relaxation. **Table 1** summarizes the lattice constants and atomic positions of this compound. For the orthorhombic crystal system with two perpendicular mirror reflections M_x and M_y, polarization along the x or y direction is forbidden by symmetry. The calculated polarization along the z direction is 33.0 μC/cm^2, comparable to that of the conventional ferroelectric oxide $BaTiO_3$.

Figure 2a shows the calculated band structure of $LiTeO_3$, which reveals a band gap of about 1.96 eV. The conduction band minimum is located near the X point. **Figures 2b-c** show the calculated spin textures around the X and Y points, which clearly indicate the appearance of the PST. The theoretical prediction can guide the experimental material search, and the modeling results will be compared with the measured material properties.

Compound	Atom	Wyckoff position	*x*	*y*	*z*
$LiTeO_3$ *a* =5.102 Å *b* = 5.293 Å *c* = 8.988 Å	Li1	2a	0.00000	0.00000	0.31812
	Li2	2b	0.00000	0.50000	0.66325
	Te1	2a	0.00000	0.00000	0.89667
	Te2	2b	0.00000	0.50000	0.08939
	O1	4c	0.21217	0.65593	0.92734
	O2	4c	0.21455	0.18574	0.06889
	O3	4c	0.20975	0.68100	0.23479

Table 1. *Lattice constants and atomic positions of $LiTeO_3$.*

$BiInO_3$ is a potentially polar oxide with distinct optical properties, whose origin could result from the surface that has not been well characterized until now. Using angle-resolved x-ray photoemission spectroscopy (ARXPS), SI **Dowben**'s team identified a large surface to bulk core-level binding energy shift for the In $3d_{5/2}$ core-level, indicating surface properties of $BiInO_3$ thin films very different from the bulk. $BiInO_3$ terminates in indium at room temperature and loses bismuth from the surface of the film at T = 573 K. The Debye–Waller plots suggest effective Debye temperatures of 263 ± 10 K and 556 ± 27 K, for the surface and bulk components of In 3d core-level, respectively.

SIs **Hong** and **Xu** have deposited epitaxial $NiCo_2O_4$ (001) thin films of various thicknesses on $MgAl_2O_4$ (001) substrates with different conditions and carried out the magnetic characterizations. **Figure 3a** shows the x-ray diffraction and reflection measurements taken on a 30 nm thick $NiCo_2O_4$ (001) film, from which the film structure and thickness have been characterized, respectively. The dependence of background oxygen pressure during the epitaxial growth of $NiCo_2O_4$ films have been studied. With high growth pressure, the magnetic transition temperature saturates at a value above 400 K with a square-shaped magnetic hysteresis. With low growth pressure, in addition to the reduction of the magnetic ordering temperature, a change in the magnetic hysteresis indicating multiple magnetic components is observed. (meets metrics)

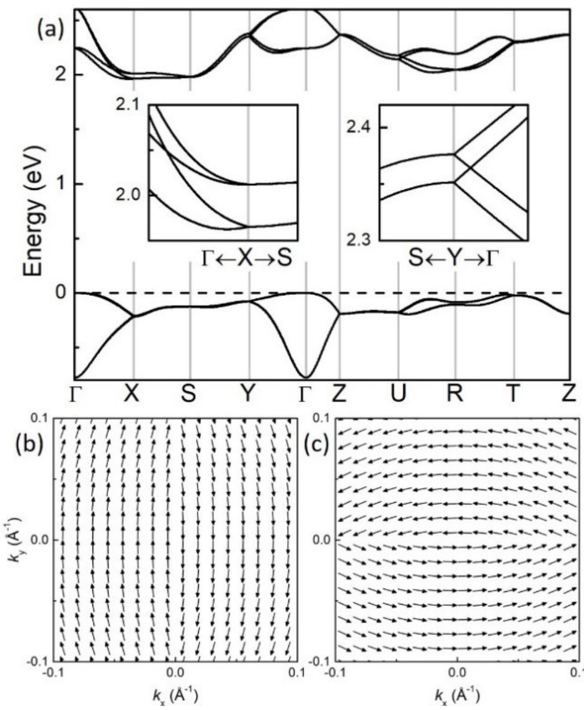

Figure 2. *(a) Band structure of bulk $LiTeO_3$. (b-c) Spin textures in the k_z = 0 plane around the X (b) and Y (c) points for the lowest conduction band.*

Using the optimal oxygen pressure, the **Xu** group studied the thickness dependence of the magnetism of $NiCo_2O_4$. **Figure 3b** shows the temperature dependence of magnetization. The magnetic transition temperature reduces from 400 K for thick films (consistent with the spin polarized inverse photoemission) to about 120 K for the film of 1.6 nm. In addition, the low-temperature magnetization reduces from above 2 μ_B/f.u. for thick films to about 0.75 μ_B/f.u. for the film of 1.6 nm. These results suggest that the surface of the $NiCo_2O_4$ films has a significantly reduced magnetic transition temperature and smaller saturation magnetization than bulk. Using spin-polarized photoemission spectroscopy, **Dowben**'s group established that the surface spin polarization of $NiCo_2O_4$ is canted, contributing to the suppressed net magnetization. Hong group showed that strong perpendicular magnetic anisotropy can be sustained even for ultrathin $NiCo_2O_4$ films down to 1.5 unit cell thickness. Studies of anomalous Hall effect reveal the competition between intrinsic Berry phase effect and dirty metal spin scattering.

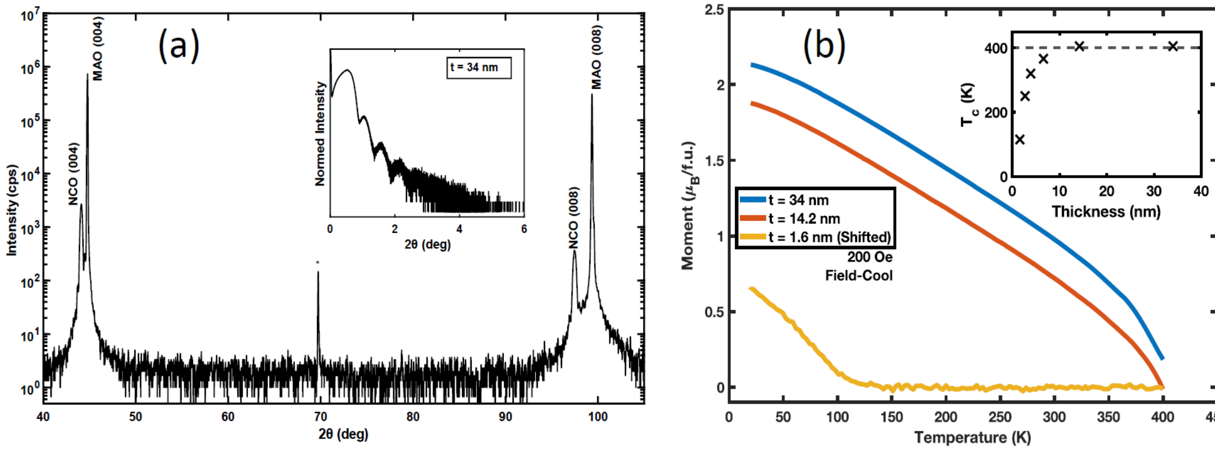

Figure 3. *(a) X-ray diffraction and (inset) x-ray reflection taken on 34 nm $NiCo_2O_4$ on $MgAl_2O_4$ (001). (b) Magnetic moment per formula unit of $NiCo_2O_4$ measured in 200 Oe upon warming after cooling in the magnetic field. Inset: magnetic transition temperature as a function of film thickness.*

Despite an absence of significant segregation of either cobalt oxide or iron oxide, the core level photoemission binding energy shifts tend to indicate that the surface for $CoFe_2O_4$(111) thin films grown on Al_2O_3(0001) is significantly different from the bulk. $CoFe_2O_4$(111) thin films show a surface-to-bulk core level shift in both the Co 2p and Fe 2p core level photoemission spectra. Surface weighted components in the core level photoemission spectra of both Co $2p_{3/2}$ and Fe $2p_{3/2}$ can be distinguished from the bulk components, by angle resolved X-ray photoemission spectroscopy (ARXPS), for $CoFe_2O_4$(111) thin films. The surface termination of $CoFe_2O_4$(111) contains both Co and Fe with no evidence of strong preferential surface termination of either an iron or cobalt oxide, except for $CoFe_2O_4$(111) in the thin film limit. With extensive annealing above room temperature, the cobalt oxide component of very thin $CoFe_2O_4$(111) films, grown on Al_2O_3 (0001), will lose oxygen. Similarly, **Xu** group also carried out the growth of $Tm_3Fe_5O_{12}$ epitaxial thin films on $Gd_3Ga_5O_{12}$ substrates. The successful fabrication of high-quality thin film samples is critical for achieving electric-control of the magnetic and topological states.

SI **Skomski** investigated various aspects of the micromagnetic topological Hall effect (THE), including the fundamental quantum-mechanical implications of the *B*-field and of magnetic anisotropy, and micromagnetic topological phase transitions in nanostructures. A paper in preparation establishes a relationship between the Lifshitz transition in metals (**Fig. 4a**) and the topological behavior of thin-film domains (**Fig. 4b-g**). The domain walls in the micromagnetic analogy (yellow) correspond to the Fermi surface in the Lifshitz analogy, which separates occupied (gray) electron levels from unoccupied levels (white). The topology, characterized by the topological number *Q*, does not depend on the shape and size of the domains and leads, in thin-film experiments, to a THE proportional to the magnetic flux quantum times *Q*. The transition from (f) to (g) is a topological phase transition (TPT), where a trivially small magnetization change translate into a big jump in *Q*. This bump is accompanied by a phenomenon newly discovered in the present EQUATE, namely a topological hysteresis. The effect is difficult to measure in ordinary thin films, where the large micron-size size *L* of the domains dilutes the effect of *Q* on the THE, which scales as Q/L^2. However, it is observable in thin films made from cluster-deposited Co nanoparticles.

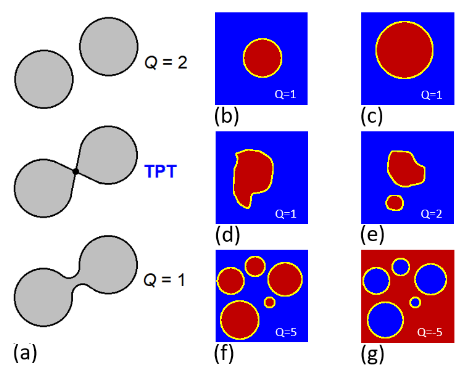

Figure 4. *(a) Lifshitz transition involving a metal. (b-g) Topological phase transition involving thin-film domains.*

Figure 5 shows the simulated magnetic and topological hysteresis loops (**Fig. 5a**), experimental THE hysteresis (**Fig. 5b**), and the simulated spin structure (**Fig. 5c**) of cluster-deposited thin films of Co nanoclusters. The topological hysteresis loops (red and blue curves in Fig. 5a) are very different from the magnetic hysteresis loop $m(H) = M_z(H)/M_s$ (black curve in **Fig. 5a**). The noncoplanar spin structure in **Fig. 5c**, which creates a Berry curvature for conduction electrons and consequently the THE. Coplanar noncollinear spin structures, for example planar domain walls, do not create a THE, because they lack the topological domain-wall closure characteristic of a nonzero *Q*, as in **Fig. 4b-d**. This result yields important insight into the analysis of the THE effect caused by the curling-type vortex mode in magnetic nanosparticles and the spin structure of Co-Si magnets.

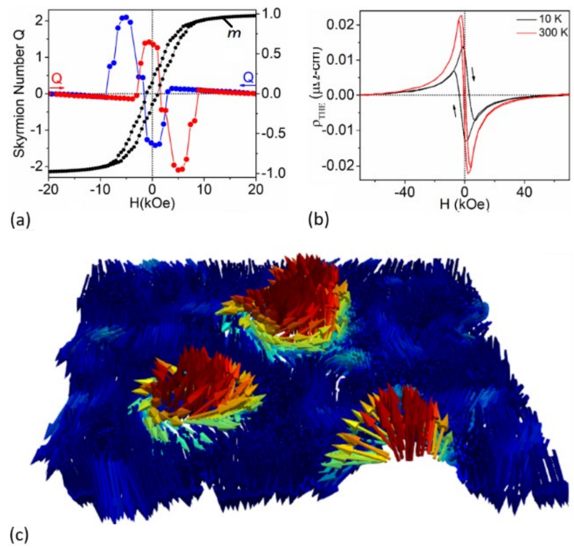

Figure 5. *Simulated spin structure and skyrmion properties: (a) magnetic hysteresis loop and field dependence of skyrmion number Q, (b) THE hysteresis, and (d) 3D spin structure at 7 kOe.*

SI **Kovalev** team performed micromagnetic simulations of topological spin textures such as spirals and skyrmions in rare-earth iron garnets. The simulations used parameters in the ballpark of what could be achievable in REIG, considering in-plane and perpendicular anisotropies with the energy scale given by $K_U = 1\,kJm^{-3}$, DMI of $D = 50\,\mu J\,m^{-2}$, exchange stiffness of $A = 0.84\,pJ\,m^{-1}$, and saturation magnetization of $M_S = 50\,kA\,m^{-1}$. By tuning the magnetic field strength and type of anisotropy, *i.e.,* in-plane and perpendicular, it is possible to tune systems between different phases (**Figure 6**). In particular, the magnetic field can drive the systems from the spiral phase to the skyrmion phase. Furthermore, anisotropy can be tuned by strain via magneto-elastic coupling. As a result, it is conceivable to realize transitions to in-plane phases, such as square lattice of vortices and antivortices (**Fig. 6**). The strain can be tuned by the substrate as well as by changing the temperature. The team also studied the realizations of skyrmions in three sublattice antiferromagnets using multiscale approaches, combining micromagnetics, *ab initio*, and Monte Carlo [*Phys. Rev. Materials* **5**, 054401 (2021)]. Micromagnetics simulations have been used to study generation of skyrmions and bimerons from boundary instabilities [*Phys. Rev. B* **104**, 064417 (2021)].

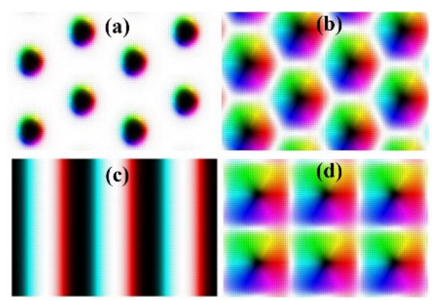

Figure 6. *Micromagnetic simulation of rare-earth iron garnets: Perpendicular anisotropy ($1\,kJm^{-3}$) (a) with and (b) without 6 mT magnetic field; in-plane anisotropies with strength of (c) $1.5\,kJm^{-3}$ and (d) $2\,kJm^{-3}$, with 15 mT magnetic field and simulated area of 600 x 400 nm^2.*

For **<u>FRG1's Thrust 1, Objective 1c</u>**, SI **Skomski** carried out modeling of exchange interactions in rare earth (RE) ferromagnets (FM) and antiferromagnets, which are scientifically interesting and important in various areas of technology, such as permanent magnetism, quantum computing, and magnetocalorics. **Figure 7** is a schematic of a solid containing one type of iron-series transition metal atoms (blue) and two rare-earth species (red and yellow). An intriguing picture emerges in the case of two or more rare-earth sublattice (red and yellow) interacting with an iron-series transition metal sublattice (blue). The atomic structure of the magnet is generally noncubic and leads to a hierarchy of interactions, as evidenced from the dependence of the Curie temperature T_c. The transition-metal sublattice (*TT*) exchange normally yields the main contribution, the RE transition metal (RT) exchange comes next, and the RE-RE exchange interaction (RR and RR') are often small. However, even when quantum-mechanical and/or statistical mean-field theory is used, an exact solution is possible only for two sublattices (T, R) and, with some labor, three sublattices (T, R, R'). However, systems with four or more sublattices (T, R, R', R'', ...) exist and are technologically important, as exemplified by misch metal magnets. In this case, the restriction to the leading TM-TM and RR-TM interactions and the resulting bond topology yields an exact solution.

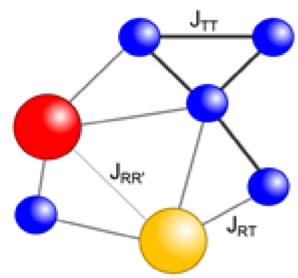

***Fig. 7**. Schematic atomic structure of a rare-earth transition-metal solid.*

For FRG1's Thrust 2, Objective 2a, together with SI **Dowben** (FRG1) and collaborators from University at Buffalo, SI **Binek** group investigated electron transport in graphene on chromia films. Small magnetoelectric susceptibility response was detected by implementing an AC superconducting quantum interference magnetometry technique. Using this newly established technique, the team studied chromia powder samples for fingerprints of axion electrodynamics and used the high sensitivity of the susceptometer to investigate magnetoelectric response in thin films.

The team further developed a Magneto-Electric FET (MEFET)-RAM, a nonvolatile cache memory design based on 2-Transistor-1-MEFET (2T1M) memory bit-cell with separate read and write paths. We have shown that MeF-RAM is a promising candidate for fast non-volatile cache memory (NVM). The MeF-RAM design has been benchmarked against other memory technologies, including both volatile memory (i.e., SRAM, eDRAM) and other popular non-volatile emerging memory (i.e., ReRAM, STT-MRAM, and SOT-MRAM). The MeF-RAM reduces Energy Area Latency (EAT) product on average by ∼98% and ∼70% compared with typical 6T-SRAM and 2T1R SOT-MRAM counterparts, respectively.

For **FRG1's Thrust 2, Objective 2b**, SI **Guo** group built a confocal microscope based optical setup for low frequency Raman spectroscopy and micro-reflectance spectroscopy and demonstrated its performance using a number of calibration samples and samples under study. High quality lead halide perovskite crystals were synthesized for use as substrates in studies of dynamic emission Stokes shift. Utilizing the home-built microscopic imaging setups, Guo group directly visualized general and consecutive ferroelastic phase transitions in lead halide perovskite crystals. As shown in **Figure 8**, the ferroelastic twin domain walls have sufficient optical contrast under simple non-polarized light illumination. The team elucidated the wavelength and polarization dependence behavior of the twin domain walls and investigated the mechanisms that generates such optical contrast.

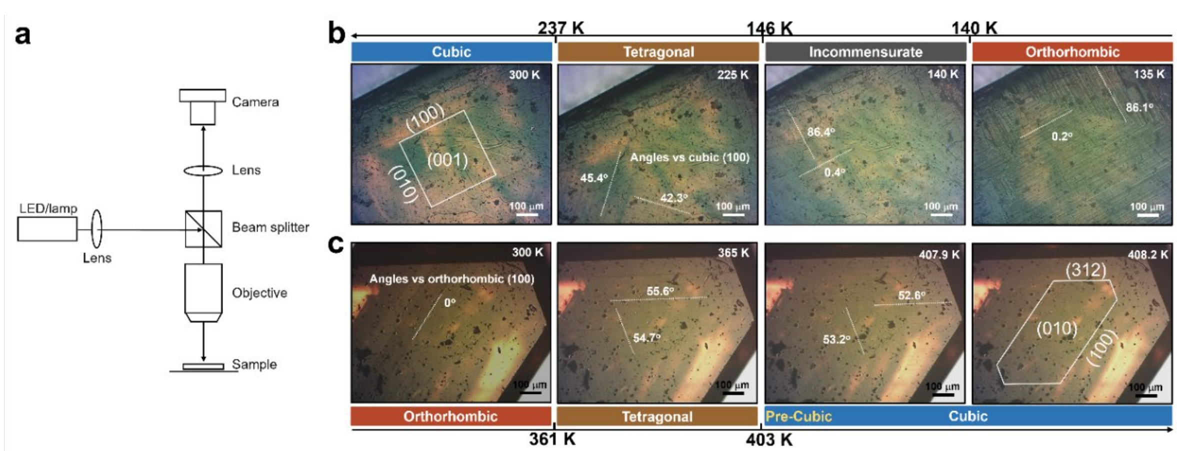

***Figure 8.** Direct visualization of ferroelastic domain walls and their switching behaviors using optical path in (a), through consecutive phase transitions in (b) $MAPbBr_3$ and (c) $CsPbBr_3$.*

Benchmarking our low-frequency Raman spectroscopy measurements, **Guo**'s team was able to measure collective lattice phonon modes in molecular crystals and framework phonon modes in lead halide perovskites, which reproduced and improved upon previously published work. An asymmetrical spectral continuum background was observed in Raman scattering in the low frequency (THz) region in metallic and dielectric systems and its origin and implications were analyzed. Micro-reflectance spectroscopy was used to investigate the band edge electronic transitions in lead halide perovskites and low dimensional materials.

For FRG1's **Thrust 2, Objective 3**, SI **Xu**'s group successfully fabricated spin crossover molecular (SCO) crystal thin films on dielectric substrates. The characterizations of these samples are currently underway, and the metrics will be met on schedule. SI **Dowben**'s group Built Fe(trz)2-based light polarization phototransistors, and met the metrics by demonstrating its electric and optoelectronic performance.

SI **Lai**'s group focused on developing a layer-by-layer (LBL) assembly method to fabricate SCO thin films with near room temperature transition. Lai group chose [Fe-(pyrazine){$Pd(CN)_4$}] as the model system since a similar system with Pt instead of Pd was investigated previously. The team first synthesized the starting material, $(TBA)_2Pd(CN)_4$. This compound was then purified and systematically characterized using ^1H NMR, ^{13}C NMR, IR, and XRD. These results confirm the successful synthesis of the Pd-containing starting material. Next, we used the proposed LBL method to deposit the [Fe-(pyrazine){$Pd(CN)_4$}] thin film on a polycrystalline gold substrate. In brief, a clean gold substrate was first placed in a solution containing 1 mM mercaptopyridine for ~19 hr to generate a well-packed self-assembled monolayer. The mercaptopyridine-modified substrate was soaked alternately (total of 5 cycles) in 100 mM $Fe(BF_4)_2$, 100 mM $(TBA)_2Pd(CN)_4$, and 100 mM 4,4'-azopyridine for 1 min at room temperature. Next, the substrate was soaked alternately (total of 20 cycles) in 100 mM $Fe(BF_4)_2$, 100 mM $(TBA)_2Pd(CN)_4$, and 100 mM pyrazine for 1 min at -60 °C. Last, the film was dried with N_2 and stored in the glove box.

The resultant 25-layer film was analyzed using both SEM/EDX and SQUID magnetometer. The SEM/EDX analysis confirms the presence of a thin film that contains Pd and Fe, and the SQUID magnetometer data allude to the presence of a weak hysteresis loop. The lack of abrupt spin transitions could be a result of residual solvent content and the extent of crystallinity of the film. For most LBL deposited thin films, film crystallinity is greatly affected by the presence of excess unreacted reagents and other contaminants. Thus, additional rinsing steps might be required to improve the overall film quality.

X-ray Absorption Spectra (XAS) was also employed to provide additional support of the magnetometry results. The SCO behavior is observable in the Fe L_3 ($2p_{3/2}$) X-ray absorption spectra as a major e_g peak at ~708 eV, decreasing with increasing temperature. In the high spin (HS) state, e_g orbitals are partly filled, leaving the t_{2g} subsequently partly depopulated, and thus accessible in XAS. The HS state corresponds to the Fe L_3 ($2p_{3/2}$) edge to a decrease of the peak intensity at 709 eV and an increase of the corresponding t_{2g} peak at ~706 and 708 eV. A HS state fraction, using the spectra from other Fe(II) SCO complexes as the benchmark of the spin states, was established. The HS state fraction *versus* temperature plot generated from the XAS data shows an increase in HS occupancy with increasing temperature across the SCO transition (**Figure 9**). Complete HS state occupancy,

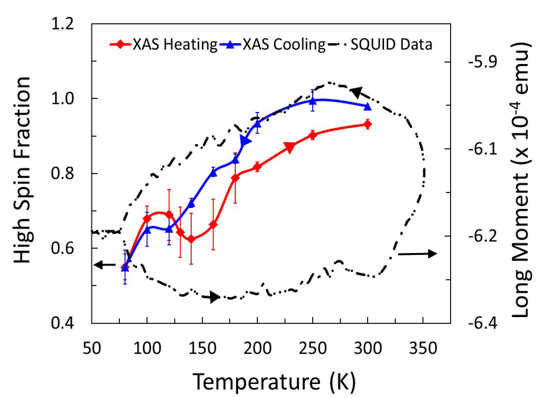

***Figure 9**. Fraction of HS state occupancy, as abstracted from the Fe L_3 ($2p_{3/2}$) edge X-ray absorption spectra, for both heating and cooling sequences, overlaid with the long moment data from SQUID for a 25-layer [Fe-(pyrazine){$Pd(CN)_4$}] film.*

however, could not be achieved until above 300 K, despite the evidence of hysteresis based on the HS fraction plot.

In summary, **Lai**'s group developed a viable method to fabricate an SCO thin film, and met the metrics by characterizing the properties using SEM/EDX, SQUID, and XAS. A manuscript based on these results is currently under review in *Physica E: Low-dimensional Systems and Nanostructures*. Next, this team will focus on optimizing the film fabrication procedure and developing new SCO systems suitable for thin film-based devices. SI **Guo** group is in the process of setting up low frequency Raman spectroscopy for measuring collective framework phonon modes, and will meet the metrics on schedule.

FRG2 – Quantum Technology

The goals of FRG 2 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 1, FRG 2 investigators collaborated extensively with investigators from other thrusts in FRG 2 and FRG 1 (**Binek**, **Lai**, **Kovalev**). The scientific and educational achievements are detailed below.

In **Thrust 1: Quantum Sensing and Metrology,** the FRG2 group advanced in its efforts to **Probe Spin-Magnon Interactions in Ferromagnetic (FM) Waveguides.**

For Objective 1.a (Probing Spin-Magnon Interactions in Ferromagnetic [FM] Waveguides): in collaborating with **Binek** (FRG1), FRG2's **Laraoui** group grew CoFe and CoFeB thin films on sapphire with different thicknesses ranging from 5 nm to 30 nm. Initial X-ray diffraction (XRD) measurements on Ti(3 nm)/CoFe(30 nm)/Al_2O_3 (0.5 mm) revealed FeCo (Fe_7Co_3) peaks **(Figure 10a)** and VSM magnetometry measurements on 10-nm and 30-nm thick CoFe films revealed perpendicular magnetic anisotropy with square hysteresis loops, **(Figure 10b)**. Ferromagnetic resonance (FMR) experiments are under progress to measure CoFe and CoFeB magnon spectra. In Year 1, we performed room temperature (RT) FMR and NV microscopy characterization of magnon modes in high quality single crystal YIG thin films, purchased commercially from Matesy Gmbh. We made Au electrodes on top of YIG 200 nm thick YIG films to control the magnon modes. To perform NV measurements on YIG under variable temperatures and magnetic field we purchased a cryostat (3.5-350 K) from Montana Instruments, electromagnet (up to 3 T) with chiller from GMW, and high frequency microwave equipment (frequencies up to 26 GHz). The cryogenic NV setup is under construction. **Kovalev** (FRG1) performed theoretical

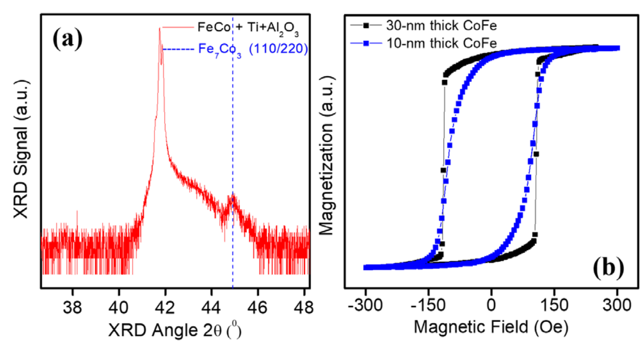

***Figure 10. (a)** XRD measurements of Ti(3 nm)/CoFe(30 nm)/Al_2O_3 (0.5 mm), revealing FeCo peaks (Fe_7Co_3). **(b)** Magnetization curves measured with VSM magnetometer, showing square hysteresis loops with perpendicular magnetic antitropy.*

calculation and modeling of magnon and skyrmion modes in magnetic insulators YIG and TmIG (see the FRG1 section of this narrative report).

Significant Results: We characterized the magnetic and structural properties of CoFe and CoFeB using VSM magnetometry, and XRD spectroscopy, and we confirmed high perpendicular magnetic anisotropy (square hysteresis loops with high saturation magnetization), **Figure 10**. The CoFe and CoFeB thin films were made by magnetron sputtering. We studied spin wave (magnon) modes in YIG, purchased commercially, and connected with AU electrodes, by using FMR and NV quantum microscopy and confirmed surface magnon modes with frequency range from 2 GHz to 8 GHz depending on applied magnetic field.

Key outcomes or other achievements: Laraoui's group in collaboration with **Binek** (FRG 1) developed a growth recipe for CoFe and CoFeB thin films, reproducible with different growth conditions. The **Laraoui** group built an NV setup that works at RT conditions and magnetic field up to 0.3 T in confocal and far-field configurations. **Laraoui** developed a new University of Nebraska-Lincoln class, MATL- 492/892: Introduction to Quantum Materials and Technologies. This class will be given to students every spring semester the next 5 years, including Spring semester of 2022 (January-May). (meets metrics)

For Objective 1.b (Quantum Sensors for Low Field Magnetic Resonance Spectroscopy): in EQUATE's Year 1, the **Laraoui** group optimized the NV setup for low field magnetic resonance (LFMR) spectroscopy. We made diamond chips doped (implanted with 15N) with a thin (10-200 nm) NV layer for imaging iron-based radicals and spin-crossover SCO: [Fe(Htrz)2 (trz)](BF4), (Htrz = 1H-1,2,4,-triazole) molecule made by **Lai** (FRG 1). The **Liou** group developed a temperature heater (RT-100 C) and integrated it to the NV-LFMR setup in **Laraoui**'s lab. The **Liou** group fabricated grating on NV doped-diamond substrates (4 mm x 4 mm x 0.5 mm) for T1 imaging of Fe contained biomolecules (hemoglobin, cytochrome C).

Significant Results: FRG2 performed NV based T1 relaxometry imaging technique to map iron in cytochrome C (Cyt C) proteins. Cyt C plays in important role in the electron transport chain of mitochondria and it is in the Fe(III) paramagnetic state under ambient conditions **(Figure 11b)**. To functionalize the biomolecules to the diamond surface, **Lai** group (FRG 1) carboxylated the 10 nm NV doped diamond substrate (**Fig. 11a**). We measured on Cyt C under different concentrations and locations of the 10 nm NV doped diamond chip. We showed a significant reduction of the NV T1 from few milliseconds to hundreds of microseconds **(Figures 11c, 11d)**. We are integrating the nanograting made by **Liou** and T1 imaging in under progress.

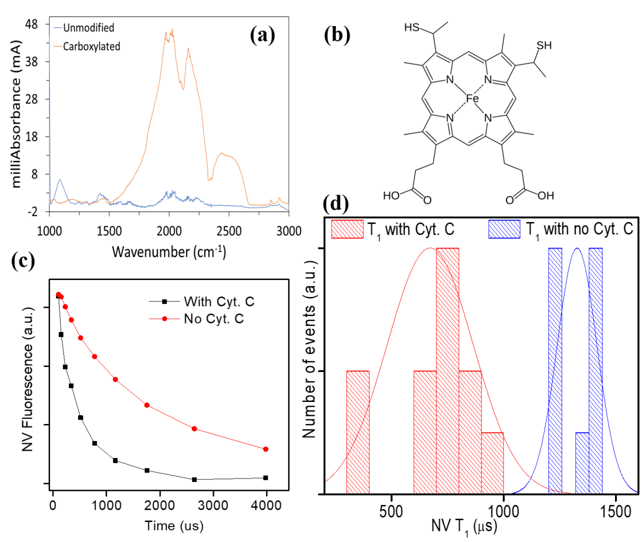

***Figure 11. (a)** 4 mm x 4 mm 10nm- NV doped diamond chip is carboxylated to attach Cyt.C molecules. **(b)** structure of one of the hemes in Cyt.C. **(c)** T1 measurements done with and without Cyt. C molecules. **(d)** histogram of T1 variation with and without Cyt.* C

Key outcomes or other achievements: Laraoui in collaboration with **Liou** (FRG 2) and **Lai** (FRG 1), FRG2 submitted two abstracts on NV sensing of iron in biomolecules for oral presentations by graduate students at APS March Meeting to be held in March 2022 in Chicago, IL, and at MRS Spring Meeting to be held in Honolulu, HI in May 2022.

In the areas of training and professional development: Laraoui trained a female graduate (second-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 1 undergraduate) and postdoc learn new aspects of quantum sensing technologies every week. FRG2 also joined live events relevant to our research from Argonne National Lab. (CNM). For example, we attended a seminar by world-renowned researcher Ronald L. Walsworth, Jr. (Department of Physics University of Maryland) on diamond quantum sensing.

For **Objective 1.c (Characterizing new solid-state Qubits in Ultrawide Band Gap semiconductors)**, the **M. Schubert** group designed and built a prototype THz-EPR ellipsometer setup and developed a new method for calibration. The setup was used to measure EPR signatures with nuclear magnetic resonance of the nitrogen defect in 4H silicon carbide (SiC) single crystals as a function of magnetic field and THz frequency (**Figure 12)**. This team also measured THz-EPR ellipsometry in iron-doped monoclinic symmetry gallium oxide single crystals, and measured phonon mode properties, optical, and electronic properties in high-quality alpha and beta aluminum gallium oxide samples from collaborators in Cornell and UCSB. We used the free electron laser facilities at BESSY II in Berlin and measured hyperbolic polariton properties in beta gallium oxide. We further developed the strain stress relationships for phonon modes and electronic band to band transitions for monoclinic symmetry gallium oxide. The **M. Schubert** group further collaborated with Lincoln, NE-based industry partner J.A. Woollam Co., Inc., for development of THz-EPR ellipsometry.

Significant Results: Marking a milestone for Year 1, FRG2 successfully calibrated and tested the prototype THz-EPR ellipsometer setup and received external funding support from collaborator J.A. Woollam Co.,

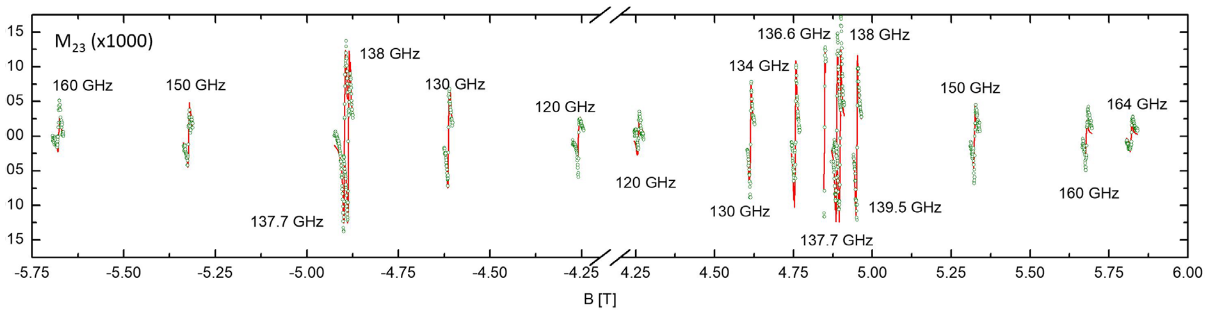

Figure 12. *First ever THz-EPR ellipsometry data (here Mueller matrix element M23 which is directly proportional to the circular dichroisms in the magnetic susceptibility). Examples show N-doped 4H-SiC revealing the spin-½ triplets split by hyperfine interaction caused by N nuclear spin I=1, for large ranges of frequency versus field, marking 2 milestones for Year 1. [Accepted in APL January 2022].*

Inc. to purchase critical components for the THz-EPR ellipsometer setup. We note that measuring the NV centers in diamond, originally planned for Y1 will be performed in Y2. We correctly modeled the polarization properties of the nuclear magnetic resonance of the nitrogen defect in 4H SiC single crystals in agreement with expectation from previous conventional low-frequency EPR measurements. Our frequency-dependent magnetic susceptibility model for the polarization response of the magnetic moments associated with the spin excitation is also valid to describe spin transitions observed in gallium oxide, and we identified two sets of EPR spin S=5/2 signatures in iron-doped beta-gallium oxide single crystals whose transitions are observed for the first time at very high frequencies, marking a milestone already for Year 2. These transitions are superimposed by zero-field splitting effects, whose Hamiltonian appears to be unknown for monoclinic symmetry. We then determined the complex phonon mode behaviors in alpha and beta aluminum gallium oxide alloys, and that hyperbolic polaritons in beta gallium oxide belong to a new class: hyperbolic shear polaritons. We determined the complex optical and electronic properties of alpha and beta aluminum gallium oxide and the strain stress relationships for longitudinal optical phonon modes in beta gallium oxide, and for electronic band to band transitions at the Brillouin zone center for monoclinic symmetry gallium oxide.

Key outcomes or other achievements: Our prototype THz-EPR ellipsometer setup is ready to be used for analysis of new quantum qubits. **M. Schubert** received a new award from Air Force Office of Scientific Research for further application of THz EPR ellipsometry. Additionally, FRG2 published our first EPR results in the open access 2022 Roadmap paper on Magnetooptics in *Journal of Physics D: Applied Physics*.

For FRG2's **Thrust 2, Objective 2.a**, in Year 1, the SIs 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 enantiomers of highly ordered nanostructure L shapes (nano boomerangs) using heterostructure iGLAD from silicon and silver as precursors. We measured optical properties of Si-Ag nano boomerangs over broad spectral range. This FRG2 team fabricated zirconium oxide nano helices and nanocolumnar structures using iGLAD, and measured their optical properties from near infrared to deep ultraviolet. We performed computational analyses for the 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 also performed computational analyses of the nonlinear optical properties for all-dielectric silicon helical nanostructures 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.

The **Argyropoulos** group collaborated with **E. Schubert** and **M. Schubert** (FRG2) 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 Y1. Samples of chiral nanostructures were sent to University of Memphis collaborator to perform nonlinear spectroscopy and compute the polarized second and third harmonic generation from these nanostructures. Simulations of the chiral nonlinear harmonic generation were performed in **Argyropoulos** group to verify the experimental results. Our iGLAD synthesis of enanthiomer chiral structures for nonlinear applications marked another milestone for Year 1 **(Figure 13)**.

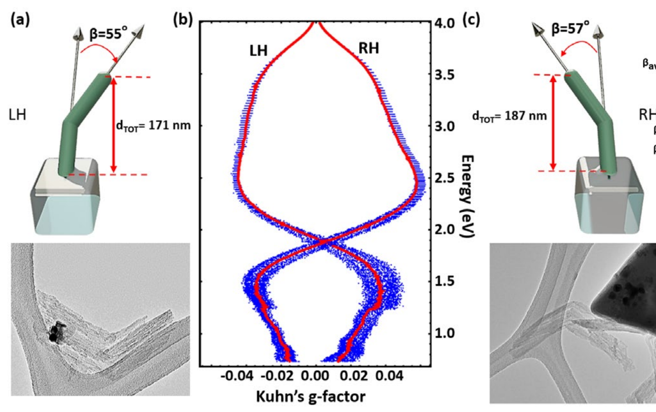

***Figure 13. (a)**, **(c)** Enantiomers of highly ordered nanostructure L shapes (nano boomerangs) using heterostructure iGLAD from silicon and silver as precursors, which reveal bisignate chiral response ("Kuhn's g-factor"). The spectra in **(b)** are nearly symmetric reflecting the excellent structural properties which are*

E. Schubert and **M. Schubert** measured a strong broadband spectral optical bisignate (positive and negative) circular dichroism from the near infrared to the ultraviolet spectral range which is controlled by nano boomerang structure parameters. They detected a strong circular dichroism in the ultraviolet spectral rang 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. **Argyropoulos** and **Bao** were awarded a grant from the Nebraska Center for Energy, entitled "Robust Topologically Protected Energy -Efficient On-Chip Microlaser for Secure Data Center Communication Systems." **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.

For FRG2's <u>Thrust 2, Objective 2.b</u>, in Year 1 the **Argyropoulos** group modeled spontaneous parametric down conversion and multiqubit entanglement in various nanophotonic platforms. **Bao** group is developing an experimental set-up to produce and measure SPDC two-photon entangled pairs. **Bao** purchased all equipment required for the experiment including tunable femtosecond laser, single photon detectors, microwave, and optical equipment.

Argyropoulos' group demonstrated multiqubit quantum entanglement in elongated distances, extended time periods, and, even more importantly, independent of the emitters' positions by using epsilon-near-zero (ENZ) nanostructures (**Figure 14**) that can maximize the coherence of light–matter interactions at room temperature. They compared the multiqubit entanglement between various plasmonic systems by computing the negativity and genuine multipartite entanglement metrics. They presented a novel high fidelity two-qubit quantum phase gate based on the super-radiance collective quantum spontaneous emission response at epsilon-near-zero (ENZ) nanowaveguides. The quantum phase gate design along with the computed fidelity is shown in Fig. 14b. These results were published in *Applied Physics Letters* and highlighted as an Editor's Pick.

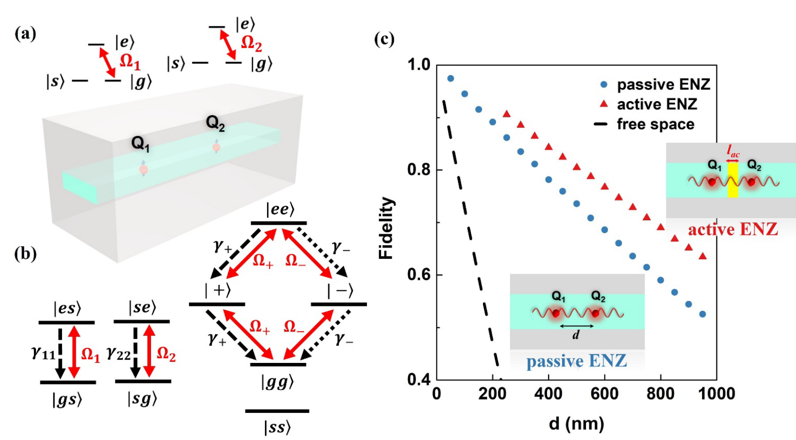

Figure 14. ***(a)*** *Two L atoms (Q_1 and Q_2) embedded inside the ENZ nanochannel. The $|e\rangle-|g\rangle$ transitions resonantly couple to the ENZ mode.* ***(b)*** *Diagram of the two L atoms energy levels.* ***(c)*** *Gate fidelity as a function of inter-qubit separation distance when passive and active ENZ plasmonic waveguides or free space are used.*

The **Argyropoulos** group designed and demonstrated a new scalable, ultrathin, and efficient entangled single-photon pair nanophotonic source based on spontaneous parametric down-conversion (SPDC) working at room temperature. The design was based on a plasmonic metasurface made of silver nanostripes combined with a bulk lithium niobate ($LiNbO_3$) crystal where localized gap plasmons were generated to realize a SPDC source. This work was published in the high-impact journal *Nanoscale* and our work was featured in the inside front cover of this journal, as shown in **Figure 16**. To realize a SPDC source, the Bao group purchased major equipment to build up the SPDC set-up.

The **Argyropoulos** group modeled nonlinear and quantum optical effects and applied the modeling to various nanophotonic plasmonic structures. We were awarded a grant from the Nebraska Center for Energy Sciences Research with **Bao** (FRG2) to study robust topologically protected energy-efficient on-chip microlasers for secure data center communication systems.

For FRG2's **Objective 2.c**, in Year 1 **Laraoui** purchased equipment (tunable pulsed picosecond laser, single photon detectors, and optical equipment) and by the middle of Year 1 it was integrated into the optical setup for single-photon characterization.

Laraoui built single photon characterization setup and used it to characterize single photon properties of NV centers in diamond, **Figure 15**. In particular we imaged single NVs (**Figure 15a**), measured single NV spectrum (**Figure 15b**), revealing zero-phonon-line (ZPL) proporties, measured antibunching $g^{(2)}$ signal, revealing single photon properties of the defects ($g^{(2)}$ signal < 0.5), **Figure 15c**. We also measured the fluorescence lifetime of single NV center found it to be ~ 12 ns, **Figure 15d**.

Argyropoulos collaborated with **Laraoui** to simulate nanopatch plasmonic nanoantenna configurations to be combined with NV-centers in diamond and defects in hBN to efficiently generate bright single-photons.

Laraoui built single photon characterization setup and tested it on single NV centers in diamond. **Argyropoulos** was awarded a Seed Grant from EQUATE with the new ECE faculty female hire in quantum materials Dr. **Yanan (Laura) Wang**. The project aims to dynamically control quantum emitters in 2D materials (e.g., hBN) via strain engineering. This project fits well with **Objective 2.c** in controlling the quantum emission of new defect system for scalable quantum networks.

In FRG2 aspects of EQUATE's training and professional development, **Laraoui** trained an undergraduate (junior) student from mechanical engineering on aspects for doing scientific research in the lab from basic design of instrumentation/setup to fully run experiments. For example, the student designed and built a PC controlled magnetic field setup which is now integrated with NV microscopy of magnon modes in FM waveguides. A first-year PhD student was trained on nanofabrication (EBL, Optical lithography, DW, FIB) and characterization (SQUID, VSM, XRD, SEM) facilities at NCMN and NERCF. During weekly group meetings, FRG2 shared knowledge in quantum optical processes with emphasis on single-photon emission and quantum nonlinear optical effects to students (graduate and undergraduate) and postdocs. One undergraduate student (Rochak Rijal) from UNL's ECE department was trained in modeling of plasmonic nanostructures during Summer 2021.

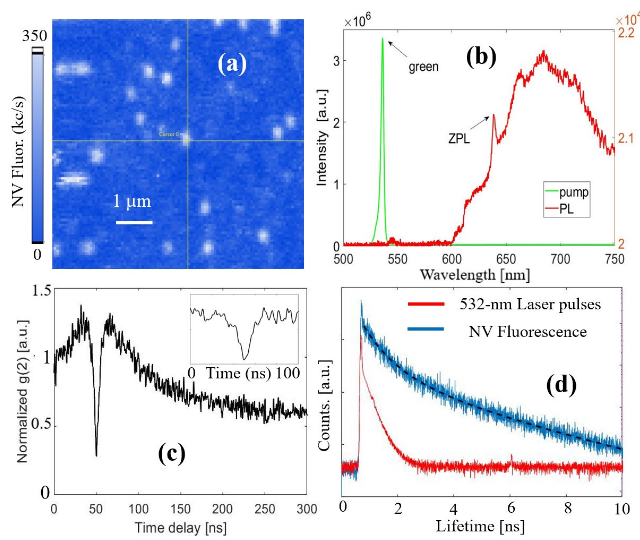

Figure 15. (a) *fluorescence image of single NVs in diamond.* ***(b)*** *Spectra of single NV (red) with zero-phonon line (ZPL) and green excitation laser (532-nm).* ***(c)*** *Antibunching curve of single NV, demonstrating single photon properties* ***(d)*** *Lifetime measurements of NV (blue) and reference pulsed picosecond laser (red). Lifetime of the NV center is 12 ns.*

Laraoui also taught the concepts of single photon generation in wide range of materials including wide-band gap semiconductors and 2D materials in his MATL492/892 class (Introduction to Quantum Materials and Technologies).

Kovalev trained **Laraoui**'s first-year PhD student on using MuMax software to model FRM and magnon modes in ferromagnetic insulators (work under progress).

Argyropoulos educated his postdoc and graduate students in the theoretical concepts to model quantum entanglement and spontaneous parametric down conversion. He led an undergraduate class (ECEN306) relevant to electromagnetic theory where he introduced to the undergraduate students to research opportunities within EQUATE and briefly presented EQUATE-related research done by FRG2. He also trained **Laraoui**'s postdoc on using COMSOL to simulate nanopatch plasmonic nanoantenna configurations that will be combined with NV-centers in diamond and defects in hBN to efficiently generate bright single-photons.

M. Schubert visited Linkoping University in Sweden and gave a lecture series on ellipsometry in materials science. **E. Schubert** visited the Leibniz-Institute for Surface Engineering in Germany and trained local collaborators in ion beam assisted glancing angle deposition.

For disseminating results to communities of interest, **Laraoui**, in collaboration with **Liou** (FRG 2) and **Lai** (FRG 1), submitted two abstracts on NV sensing of iron in biomolecules for oral presentations by graduate students at APS March Meeting to be held in March 2022 in Chicago, IL, and at MRS Spring Meeting to be held in Honolulu, HI in May 2022 (for details, see Y1 products).

Laraoui and his students gave multiple talks at the APS and MRS meetings (for details, see Y1 products). A manuscript was submitted for an invited APL perspective paper, titled "Opportunities for Nitrogen-Vacancy-Assisted Magnetometry to investigate 2D magnetic materials."

FRG2 members gave multiple talks and poster presentations including invited talks at 2022 SPIE Photonics West, 2021 URSI General Assembly and Scientific Symposium (GASS), 2021 International Conference on Electromagnetics in Advanced Applications (ICEAA), and 2021 IEEE AP-S/URSI National Radio Science Meeting (for details, see Y1 products).

The **Argyropoulos** group published the spontaneous parametric down-conversion results in the high-impact journal *Nanoscale* (see **Figure 16**) and this work was featured in the inside front cover of this journal. He also published the multiqubit entanglement results to *Applied Physics Letters* and this work was selected as Editor's pick. **Argyropoulos**' group also gave multiple talks and poster presentations including invited talks at 2022 SPIE Photonics West, 2021 URSI General Assembly and Scientific Symposium (GASS), 2021 International Conference on Electromagnetics in Advanced Applications (ICEAA), and 2021 IEEE AP-S/URSI National Radio Science Meeting (for details see Y1 products).

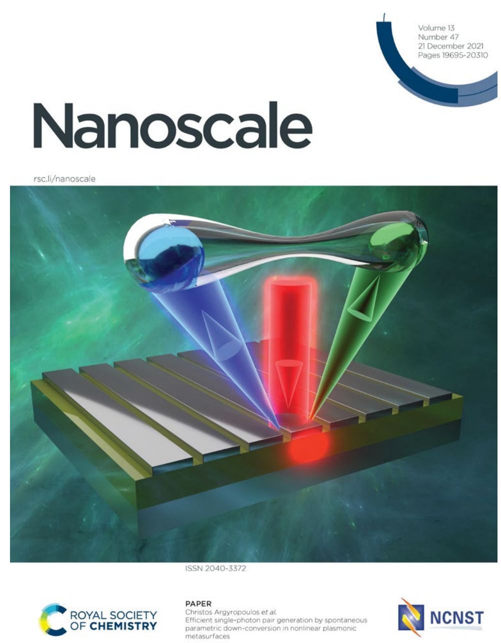

Figure 16. *Entangled single-photon pair nanophotonic source based on spontaneous parametric down-conversion selected as cover to the journal Nanoscale.*

E. Schubert and **M. Schubert** submitted and published a total of 17 publications: five are published and four are accepted and awaiting publication (see EQUATE Year 1 products). Of particular note, one of these papers is aligned to publish in the journal *Nature* (2020 Impact Factor 49.962), titled: "Hyperbolic Shear Polaritons in Low-Symmetry Crystals"; the editors are deciding whether to use their image for the weekly cover issue of *Nature* upon publication. This group published an invited review paper in *Applied Physics Review* (2020 Impact Factor 19.162), titled "A Review of Band Structure and Material Properties of Transparent Conducting and Semiconducting Oxides: Ga_2O_3, Al_2O_3, In_2O_3, ZnO, SnO_2, CdO, NiO, CuO, and Sc_2O_3"--highlighted as an "Editor's Pick." This FRG2 team also presented 9 scientific talks and in various in-person and online events, including one invited talk at 2022 SPIE Photonics West.

In Year 1, FRG 2 SIs made significant impact 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 an industry partner, 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**'s paper in Nature will appear likely with its cover picture referring to the hyperbolic shear polariton discovery in monoclinic symmetry gallium oxide.
- **Argyropoulos**' work on generation of entanglement of single-photon pair nanophotonic source based on spontaneous parametric down-conversion was published and selected as cover to the high-impact journal *Nanoscale*.
- **Laraoui** shared the syllabus of a new class MATL 492/892: Introduction to Quantum Materials and Technologies with science and engineering departments at UNL. For Spring 2022 (January - May), so far 26 students (8 female) registered for the class with 18 graduate students and 7 undergraduate students from Physics, Chemistry, Electrical & Computer Engineering, and Materials & Mechanical Engineering departments.

In terms of **Changes/Problems,** FRG2 noted:

- Due to the supply chain issues caused by the COVID-19 pandemic, there was a significant delay in shipping the equipment purchased in Year 1 in FRG 2. For example, the lead time to get MW switches was 36 weeks.
- Facility moves for the labs of some FRG2 members at the University of Nebraska-Lincoln (**Laraoui**, **Bao**) to a new Engineering Research Center in the months of December 2019-February 2022 has delayed the building of the cryogenic NV and SPDC setups by three months. This move will give these FRG2 members the opportunity to optimize the other experimental setups in our labs (improve the stability and noise).

FRG3 – Quantum Information Processing

As outlined in EQUATE's strategic plan, Year 1 (Y1) for FRG3 is dominated by building personnel and capability in quantum information processing. Major successes include the purchase of capital equipment, development of experimental capability and the recruitment of three postdoctoral researchers and two graduate students. These accomplishments lay the foundation for the main scientific progress expected in subsequent years.

Key personnel are FRG-3 leader **Wrubel** (Creighton University), **Bao** (UNL), **Armstrong** (UNK), and **Mei** and **Sabirianov** (UNO). As expected, SI **Wong** (Creighton University) was on leave to serve at the National Quantum Coordination Office (NQCO) in Washington, DC. FRG3 research is enhanced by collegial collaboration with EQUATE's other FRG representatives (see **Figure 17**).

For **Thrust 1. Quantum Emulation** under **Objective 1.a: Surpassing the Standard Quantum Limit in a ^{41}K Bose-Einstein Condensate (BEC),** the FRG3 group focused on building new capability in our experimental apparatus and training in new postdoctoral, graduate and undergraduate researchers during this first year of the EQUATE grant. Two capital equipment purchases arrived: a cooling laser for potassium and an optical trapping laser. A third and final capital equipment purchase for an optical lattice laser is progressing through the approval process.

Figure 17. *EQUATE researchers Christian **Binek** (left) with FRG1 and Jonathan **Wrubel** with FRG3 share project updates as they interact during an equipment exchange.*

FRG3 made several major steps towards achieving a Bose-Einstein condensate including the purchase of an optical trapping laser and necessary hardware, rebuilding the 2D magneto-optical trap to take advantage of an isotopically enriched potassium-41 source, installation of a high-numerical aperture imaging system, and installation of the RF Feshbach resonance system. FRG3 expects to achieve Bose-Einstein condensation before the end of Year 1, to meet that milestone. Beyond these significant results, other key FRG3 outcomes and achievements included:

- Recruited a postdoctoral candidate
- Recruited two graduate students and five undergraduate students
- Installed critical hardware for imaging cold atoms and producing the RF Feshbach resonance
- Developed a four-coil magnetic field system for uniform fields
- Determined the dominant sources of error in the laser analysis hardware

Under **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 took place as planned. This laser will provide the capability to do quantum emulation of many condensed matter systems including systems important for quantum computation.

Beyond these significant results, achievements, and outcomes, **Objective 1.c: Quantum Emulation with an Exciton-Polariton BEC** seeks to demonstrate quantum emulation in the novel halide-perovskite exciton-polariton system, which could yield a room-temperature Bose-Einstein condensate. The first step in that direction was achieved with the production of halide-perovskite samples, and demonstration of photoluminescence capability in exciton polaritons. Fruitful collaboration between SI **Bao** (UNL) and SI **Wrubel** (Creighton) was established to speed progress regarding the emulation of supersolids in cold-atom and exciton-polariton systems. (See **Figure 18**.) In addition, halide-perovskite samples were made in collaboration with UC-Berkeley and successfully measured. In terms of sharing with audiences, one paper is under minor revision for the journal *Nature Materials*.

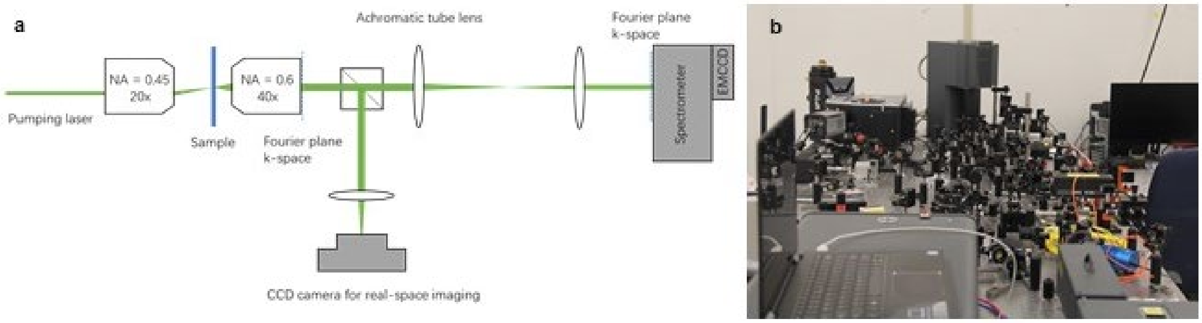

Figure 18. *Schematic diagram (a) and actual photograph (b) of the optical setup in transmission geometry via FRG3's Bao lab.*

For **Objective 1.d: Theory of Polarons in Dipolar and Spinor Gases**, the main Y1 activity has been to successfully adapt computational codes needed for simulation of polarons with the standard and dipolar Gross-Pitaevskii equation. This is a foundational step towards understanding polarons in dipolar and spinor gases. While a postdoctoral researcher was successfully recruited for more of this work, arrival was delayed by the VISA process.

Under **Thrust 2. Quantum Computation**, several advances merit mention:

In **Objective 2.a: Quantum Walk Algorithms**, due to SI **Wong**'s temporary duty with the NQCO, no activities were planned in this area for Y1. Nonetheless, SI **Wrubel** was able to recruit a theory graduate student who has already begun work towards the subsequent year's activities for the Quantum Walk Algorithm objective.

In **Objective 2.b: Crosswire Quantum Dots for Quantum Computing**, FRG3 has created a model quantum dot (QD) in cross-wire geometry. The ground state of this work 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. (meets metrics) Time evolution of the electron states will be analyzed by time-dependent solutions.

Additionally in Y1, SIs **Mei** and **Sabirianov** have been collaborating with SI **Dowben** (FRG1). Their work together 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. This collaboration has also resulted in progress on the development of lithium tetraborate as a scintillation detector for neutrons.

In FRG3's **training and professional development**, Creighton University's SI **Wrubel** is training one postdoctoral researcher, two graduate students and five undergraduate students in experimental ultracold atom techniques, atomic physics theory, Bose-Einstein condensate theory, and quantum computing. He also serves as an advisor to the University of Nebraska at Kearney (UNK) faculty search committee, in a hiring that is supported by EQUATE funding.

At UNK, SI **Armstrong** is recruiting a postdoctoral researcher, and at the University of Nebraska at Omaha (UNO), **Mei** and **Sabirianov** are recruiting a postdoctoral researcher and an undergraduate researcher.

For **dissemination of results**, SI **Wrubel** will give presentations at APS DAMOP meeting (May/June); he also gave a tour of his lab to undergraduate students from UNO. **Armstrong** plans a presentation at either APS March meeting or APS DAMOP meeting (May/June 2022). SI **Bao** has one paper under minor revision in Nature Materials. **Mei** and **Sabirianov** have submitted manuscripts toward two journal articles.

In terms of **Changes/Problems**, for several FRG3 SIs (Wrubel, Armstrong, Mei, and Sabirianov) the recruitment of postdoctoral researchers was slow and in some cases delayed by VISA issues. Despite the challenges presented by the current environment, all of these SIs have successfully recruited candidates.

- **Wrubel** notes: a vacuum leak in part of our apparatus paused our measurements and necessitated rebuilding a part of the vacuum system. This became an opportunity to improve the system and install a potassium-41 enriched source, which is an important step towards Bose-Einstein condensation.
- **Armstrong** notes: VISA/COVID-19 issues have delayed the hiring of a post-doc, who may not be able to start until April (was budgeted to start in October 2021).

Outreach and Workforce Development

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.

For Objective 4.1 (Equip Nebraska students (grades 6-12) with resources for success in STEM workforce):

YNS Summer Camps. Year 1 was considered a developmental year for future EQUATE summer camps. Significant effort was expended to develop content and activities related to quantum topics that would be used for future outreach activities and incorporated into existing YNS camps. Additionally, a request for proposals for new YNS summer camps was distributed across the state including to academic institutions and non-profit organizations. Seven proposals were submitted for consideration spanning the state with all except 1 camp listed as residential. **(meets metric)**

YNS High School Researchers. The Nebraska Center for Materials and Nanoscience (NCMN) hosted 11 high school researchers during the summer of 2021, with financial support from the JA Woollam Foundation. Additionally, two students were YNS High School Researchers with the Wong Laboratory on the University of Nebraska at Omaha (UNO) campus. **(meets metric)**

Mobile Labs. During Year 1, EQUATE Outreach efforts were focused on the development of quantum-themed mobile labs to provide hands-on activities and education materials to teachers throughout the state of Nebraska. Course materials and activities were created and used with a variety of middle and high school students in Lincoln. Additionally, the activities were evaluated by participants from Girls Inc who visited the NCMN during an in-service activity. Students were asked to fill out an evaluation form after each event which has allowed for improvements to be made to the materials. It is expected quantum mobile labs will be deployed for teacher use earlier than expected. We plan to continue using student and teacher evaluations to improve the kits throughout the course of the grant. **(meets metric)**

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 2021-2022 school year, MBSC labs were used in more than 20 Nebraska classrooms reaching over 960 students with 48.6% URM and 49% female. The schools include Omaha

Westside, Minden, McCook, Lincoln, Kearney, and Hemingford High Schools. Additionally, faculty at Central Community College used the molecular biology mobile labs to help with course development of a new biotechnology offering. **(On track to meet metric)**

Remotely Accessible Instrument (RAIN) Program. With Year 1 as a development year for the RAIN program, EQUATE outreach staff actively participated in project planning meetings with 4-H representatives to create a strategic plan for project implementation and develop student curriculum. EQUATE outreach staff also worked to repair necessary equipment and train on use of associated instrumentation. **(meets 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). The EQUATE grant was not active in time to recruit nor hire RET participants for summer timing in Year 1 of the grant cycle. Efforts for this program have been focused on participant recruitment activities. EQUATE representatives have attended several educator conferences including Nebraska Association of Teachers of Science (NATS), Nebraska Alliance for Conservation and Environment Education (NACEE), and the Nebraska Summit on Math and Science Education. **Metrics: Yrs 1-5, 5 participants annually in EQUATE labs.**

Summer Institute for Middle School Teachers (SIMST). During this first year, outreach staff met with other participating institutions to determine the extent of collaboration between sites. Locally, curriculum was developed that includes both nanoscience and quantum related topics and activities teachers could implement in their classrooms. **(meets metric)**

University of Nebraska-Lincoln chemistry professor **Mark Griep**, along with an outside collaborator, presented at two events focused on helping educators incorporate culturally relevant STEM curriculum for indigenous students. The first event, sponsored by Nebraska EPSCoR, was a virtual workshop titled "Sharing Cycle of Science Learning," as part of the **Indigenous Pedagogy Virtual Academy (IPVA)**. There were 94 registrants, the largest number at any IPVA workshop, but there were 32 zoom links to the workshop, several of them with multiple people. Additionally, Griep presented a virtual symposium "Indigenizing your Chemistry (or any) Curriculum" as part of the **2021 Colorado Science Conference**.

Objective 4.2: Continue influencing the STEM pipeline through programs for undergraduate and graduate students, and postdoctoral researchers.

Postdoctoral Scientist Mentoring Program. There were 7 postdoctoral fellows working on the project in the first year. **(meets metric)**

Graduate and Undergraduate Student Mentoring. Twenty-nine graduate students and 10 undergraduate students were mentored by EQUATE Science Investigators (SIs) in Year 1. Metrics: 16 grad, 18 undergrad (cumulative 90 Undergrad and 80 grad).

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. It aims at informing participants of the ongoing research within each FRG, as well as stimulating inter- and cross-FRG collaboration.

The first meeting was held on January 26, 2022, both in-person in Jorgensen Hall and via Zoom. The first session's speaker was Kun Wang, a graduate student from Hong's group in FRG1. **Metrics: 12 meetings per year.**

Nanoscience/Quantum Class, Minicourses, and Training. Introduction to Quantum Materials and Technologies (MATL 492/892) was a new course offering for both undergraduate and graduate students for spring 2022 with recruitment across several departments including Physics, Chemistry, Mathematics, Electrical & computer engineering, and Materials & Mechanical engineering departments. This course was developed by FRG2 leader Abdelghani **Laraoui** and has 18 students registered so far. Additionally, EQUATE investigators, Argyropolous and Binek, introduced EQUATE topics, research, and opportunities to ungergraduate and graduate students in Electrical and Computer Engineering (ECEN 306) and Physics (PHY 201 and PHY 988). Metric: Cumulative 35 grad students, 250 participants.

Research Experiences for Undergraduate (REU) Students. The EQUATE grant was not active before student recruitment materials and application deadlines were scheduled through UNL Graduate Studies, administrators of the centralized REU management platform. However, One REU student was able to research EQUATE topics due to an additional grant obtained by Guo. Guo's student, Jade Casasant, evaluated ferroelastic phase transition behaviors in lead halide perovskites. Casasant presented her research in a poster presentation at the Chemistry REU Poster Competition (won Chemistry travel award) and the UNL Summer Research Symposium (won first place among all 98 posters). She has been accepted to present her poster in the ACS Spring 2022 meeting. **Metric: Yrs 1-5, 3 students per year in EQUATE laboratories. Metric: Yrs 1-5, 1 student annually at Central Community College.**

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 2021, Nebraska EPSCoR awarded 7 recipients (exceeds metric). The 2021 proposals were reviewed on November 10 by Nebraska's State Committee for final selection of funding. **Metrics: 6 awards annually.**

***Faculty Mentoring through EQUATE Seed Grants*.** This program solicits seed proposals requesting one year of funding through a UNL-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), which includes support for one GRA and $2k for materials and supplies. **Metric: Yr 1: 2 awards, Yrs 2 -5: 1 award annually.**

Four proposals were submitted for consideration, and two were selected for funding in October 2021. The proposals chosen were:

- **Siamak Nejati** of UNL Chemical and Biomolecular Engineering proposed "Heterostructures of 2D Materials and Porphyrin-Based Covalent Organic Frameworks as a Tunable Device Platform for Quantum Technologies." Nejati will work with UNL Chemistry's **Alex Sinitskii** on this research.
- **Yanan "Laura" Wang** of UNL Electrical and Computer Engineering proposed "Dynamic Control of 2D Single-Photon Quantum Emitters via Strain Engineering." She will work with UNL ECE colleague Christos **Argyropoulos**, who is an investigator on the EQUATE project. Metrics: 5 early-career faculty.

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

In summer of 2021, Nebraska EPSCoR sponsored 20 scholarships for students (55% female, 13% URM) in the **Bright Lights** Summer Learning Adventures program. Students in this program are exposed to a variety of STEM topics over the summer.

The **2022 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**

All Girls All Math (AGAM) Camp, a weeklong virtual camp, was attended by 20 students (90% female, 55% URM) from all over the United States. Students learned about cryptography and coding, attended virtual tours of both national and local museums, and participated in small group activities such as virtual escape rooms which prompted use of mathematical reasoning, problem solving skill, and communication.

Nebraska EPSCoR provides support for 5 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**

During fall of 2021, the first **Tribal Career Fair** was held in South Sioux City, NE. The event was developed and led by **Griep**. Between 35- 40 people (approx. 32% female and 79% URM) attended, including 13 exhibitors who ran eight booths. The students were energized, and the exhibitors said they would return for future years' events (several guests suggested locating the event at tribal community high schools).

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

A new relationship was developed between Girls Inc. Eureka! Program managers in Omaha, Nebraska, Nebraska EPSCoR, and NCMN in Lincoln, Nebraska. This allowed 12 Girls Inc. participants to attend a facility tour with quantum related activities at NCMN hosted by EQUATE Outreach team members: Jodi **Sangster** (Nebraska EPSCoR Outreach Coordinator), Steve **Wignall** (EQUATE and NCMN Outreach Coordinator), and Han **Pham** (NCMN Outreach Specialist). Additionally, Sangster hosted 12 Girls Inc. participants on UNO's campus for engineering-related activities.

Wignall, Sangster, and Pham participated in a repeating afterschool program held at 5 Title 1 middle schools and 2 high schools in Lincoln, Nebraska. Every 2 months, EQUATE staff spends time with students, teaching quantum topics and doing hands-on activities. 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, outreach staff impacts 138 students every 2 months, 93 middle school (65 female and 56 URM) and 45 high school (26 female and 31 URM) students.

During Summer 2021, **Janyce Woodard** at Little Priest Tribal College (LPTC) Extension led more than 30 students through the Indigenous Plant Watercolors hands-on activity developed by Griep. The ability to host more students was hampered by the COVID-19 pandemic that resulted in a community lockdown.

The University of Nebraska-Lincoln (UNL) College of Engineering conducts **Discover Engineering Days (DED)** to introduce middle school students (grades 6-8) and teachers to the various fields in engineering, physics and the science programs offered at UNL. The college's outreach coordinates hands-on activities that emphasize engineering habits such as collaboration, problem solving, creative, and critical thinking skills. The DED offers 7 workshops over the course of the year, averaging approximately 130-150 students at each meeting. EQUATE outreach has been involved with this program throughout the year, teaching the students about Triboelectric Energy and how it relates to Quantum. Students involved in this program come from both rural and urban schools, with strong representation of URM and female attendees.

Objective 4.5 Increase public awareness of EQUATE progress

EQUATE Management Team (MT) Meetings took place monthly in Year 1; the MT has become a driving force in each FRG's coordination. The MT is collaboratively planning the 2022 **Nebraska Research and Innovation Conference**—with the first-year topic of "Commercializing Quantum Technologies in

Nebraska: From Research to Licensing." The event is set for Thursday, April 14 in downtown Lincoln, with the University of Nebraska technology commercialization organization, "NUtech Ventures," providing several speakers. Members of EQUATE's Industrial Advisory Board are also slated as presenters and/or panelists, along with a poster session including EQUATE scientists and engineers with faculty, students and postdoctoral researchers. Approximately 60 guests are expected to attend. (meets metrics)

The milestone of four **journal papers/presentations** per EQUATE investigator is on track for being met by the end of Year 1 (Y1). A total of 54 publications and presentations was achieved as of February 2022.

EQUATE was a top story in the **2021 Annual PR Report** by Nebraska EPSCoR. 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)

The EQUATE **website** (equate.unl.edu) was launched in Summer 2021 and the number of site visitors was 578 as of February 2022; this six-month tally should serve as baseline for 10% year over year visits in project years ahead. EQUATE's **social media** presence, tagged by #NebEQUATE, had an active first year of 50 mentions – from the May 2021 announcement's online celebration to the journal publications, awards and other achievements--including podcast interviews—earned by EQUATE participants. In addition, an overview video (https://youtu.be/JMP5e701xpA) was produced about the NSF EQUATE award and collaboration. (meets metrics)

Figure 19. *Guests from Omaha's Girls Inc. create spectroscopes after touring EQUATE labs at the University of Nebraska-Lincoln.*

Tours, museum exhibits and public event participation were somewhat curtailed by COVID-19 pandemic precautions. The museum exhibit was impacted by an extended closure of the host facility. Museum staff were unable to collect detailed attendance data, but they estimate approximately 200 - 250 people interacted with the display (metric not met). The display is in the process of being transferred to a new location. EQUATE participants were still able to successfully increase public awareness through facility tours (meets metric) and attendance at public events including Nebraska Alliance of Conservation and Environment Education Conference and Nebraska Association of Teachers of Science (meets metric).

Data Management

Just as the research activities of EQUATE are taking off with notable productivity, so does the generation of different types of data. As of today, EQUATE has primarily generated data by computer-controlled setups in FRG1, FRG2 and FRG 3. This includes imaging techniques, most notably the NV-center spectroscopy performed in FRG 2, computational data associated with density functional theory calculations (FRG1 and FRG3) and micromagnetic simulations of non-trivial topological spin textures (FRG2). In addition, our E/O activities created data from surveys and assessments. All data, including metadata, fall into the categories of decimal floating-point numbers, ASCII sequences, txt format, tagged image file format (TIFF), and joint photographic expert group format (JPEG).

In accordance with the insights gained at EQUATE's strategic planning meeting (August 17-19, 2021), the SIs have the freedom to determine some details of the format of the metadata and exchange of raw data, which has not been requested as of today. That said, the harmonizing of data storage on UNL's cloud-based data storage system OneDrive is in progress, making data and metadata accessible for all participating institutions. Cloud-based storage is accompanied and backed up by data storage in physical media (desktop computer hard disk drives in the labs of each individual researcher). Evaluation of the data management system is scheduled to be addressed by the EQUATE Management Team (MT) before the end of Year 1, at the MT's May meeting. The evaluation process will focus on reliability and practicality of the implemented data management and includes a request for exchange of all processed data between each SI and the management team.

Sustainability

Despite the short time period of EQUATE's existence, there have been remarkable strides towards sustainability. As anticipated in the strategic plan, results of EQUATE research and collaboration has served as a foundation for new proposals already extending work beyond the EQUATE lifetime.

The most notable success story is Dr. Bao's (FRG 3) winning of an NSF CAREER Award. EQUATE results obtained on Bose-Einstein condensates of quasiparticles in solids with applications in quantum emulation served as critical foundation of Dr. Bao's winning CAREER proposal.

Moreover, an NSF major research instrumentation proposal entitled "Acquiring optical access for quantum sensing capabilities in a cryogenic scanning probe microscope" has been resubmitted. The lead PI is EQUATE's scientific director with major contributions from the leaders of FRG2 and FRG1. If funded, this instrument will significantly contribute to the work done in both FRGs 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. The instrument will be housed and maintained by the Nebraska Center for Materials and Nanoscience and be part of UNL's materials science core facilities.

A major step towards sustainability was achieved when UNL Chancellor Ronnie Green selected and announced a set of eight grand challenge themes for the University of Nebraska-Lincoln, which will obtain over the next five years institutional support of $40 million (twice the budget of EQUATE). EQUATE has been instrumental in motivating UNL leadership to select Quantum Science and Engineering as one of the grand challenge themes. EQUATE's scientific director, Dr. 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. Currently Dr. Binek is in the process of assembling a team to develop a catalyst proposal in the framework of the grand challenges. The tentative title of the proposal is "Quantum Approaches Addressing Global Threats". It involves about 30 UNL faculty, several of whom are EQUATE SIs--including the leaders of FRG1 and FRG2, and early CAREER recipient Dr. Bao.

Arguably the most profound impacts on sustainability are the two tenure-track faculty hires currently taking place at UNL and UNK; both hires are anticipated to be in place in spring 2022. They will foster the synergy between the FRGs and help to continue advances in quantum materials science and technology long after the lifetime of EQUATE.