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RPPR Final Report
as of 23-Apr-2021

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STEM Degrees: **STEM Participants:**

Major Goals: (in 2010) Strongly coupled multiferroic materials that operate near room temperature do not exist today. Most of the present day material design approaches are top-down strategies that focus on combining “properties.” Electric-magnetic cross-coupled responses are then assumed to follow from the mere presence of different microscopic degrees of freedom. Understanding the fundamental mechanisms and key materials parameters that facilitate a strong cross coupling between the various “functions” in order to increase their functionality remains a daunting task. Considering the many talented scientists working on this problem and their combined inability to date to coax the desired functionality out of materials with their top-down approach, it is apparent that a new approach is needed to make the desired leap in progress. Given the complexity of the problem it would not be surprising if rationalized design criteria lead to structures that do not currently exist in nature and in fact cannot be synthesized by conventional bulk synthesis routes.

To address this issue and to further advance our bottom-up rational design strategy the proposed research will explore new mechanisms and their material embodiments along two themes:
(1) revealing hidden phases in bulk materials
(2) novel heterotructure/superlattice combinations of known bulk materials.

In either case a typical plan of work involves
- Perform first-principles calculations of known materials
- Development of detailed analytical models and simple crystal chemistry design rules
- Apply rules to search for new materials realizations.

Search for and developing new mechanisms -

Rational design starts with the identification of a microscopic mechanism that leads to the desired macroscopic phenomenon. What if the mechanism is currently not known? A key step in the pursuit of new mechanisms is to understand structure-property relationships in well-defined examples. The power of a theory-driven, first-principles approach in this regard is the ability to explore a multitude of phases that simply are not accessible experimentally. Our approach to guide the development of new models is to perform first-principles simulations on known bulk A-B-O materials in structures and two dimensional layered hetrostructures that have either not yet been found to form in nature – yet possibly could be synthesized via epitaxial stabilization or high pressure routes – or have been formed only under extreme conditions such as high pressure and/or high temperatures making a systematic study of their structure-property relationships unfeasible. Here the focus is not on discovering new materials per se. Instead the intellectual focus is on understanding new ways spin–lattice interactions – the basic materials physics behind strongly coupled multiferroics – can emerge from real materials and how to control it.

Revealing hidden phases: Novel 3d arrangement of atoms -
The macroscopic properties of materials depend critically on the spatial arraignment of the constituent atoms chosen by nature. Entirely new phenomena may emerge from the same basic ingredients, e.g., A-B-O atoms, depending on crystal structure. Epitaxial stabilization and high- pressure synthesis of new phases of materials opens the door to entirely new family of strongly- coupled multiferroics. Case in point, the mineral Ilmenite FeTiO3 is a hard paraelectric (i.e., a paraelectric not close to a ferroelectric transition), collinear antiferromagnetic insulator when formed in the ground state ordered corundum structure, yet ferroelectrically-induced weak- ferromagnetism, which is the basic mechanism leading towards electric field switching of magnetic domains, occurs in the metastable LiNbO3 polymorph of FeTiO3. This unique multifunctional property of the LiNbO3 polymorph was discovered through rational design as discussed in previous sections, yet the phase itself had been previously synthesized in a high pressure experiment – its magnetic and ferroelectric properties simply never measured. A systematic study of the structural property relationships would have undoubtedly uncovered it as well. A second example is the recent work of Rondinelli1 et al who have studied the non- perovskite, antiferromagnetic–paraelectric, hexagonal BaMnO3 in the cubic perovskite phase, showing not only that it has a ferroelectric instability, as shown in Fig. 9 (right), and that the lowest energy phase within this manifold of perovskite states is ferroelectric, but that the ferroelectric mechanism itself is driven by the magnetic ion, a result previously though not to occur. The ramifications of spin-lattice coupling in this novel system for which the magnetic and ferroelectric ions are the same are still unclear.

Proposed Work: In order to guide the development of new models we will perform a systematic search for metastable perovskite polymorphs of known magnetic non-perovskite ABO3 materials. We are particularly interested in the case where the polymorph has previously not been observed or if it had been observed, only at high pressure. We will study the structural, dielectric, and magnetic properties of selected polymorphs to develop models of how the symmetry and the topology of the crystalline motif influences how lattice degrees of freedom interact with the spins. We will also explore metastable phases of known pervoskites. For example, the ground state of CaTiO3 is a highly distorted orthorhombic perovskite. It is not ferroelectric despite the fact that BaTiO3 is the prototype perovskite ferroelectric and that SrTiO3 is a material close to a ferroelectric instability (SrTiO3 in fact has been driven ferroelectric with strain and applied electric fields). A systematic investigation of the lattice instabilities of the cubic perovskite CaTiO3 however, reveals that indeed a ferroelectric instability does occur, as shown in Fig. 9 (left), even though experimentally it never forms in a polar structure. In fact the PI's recent first-principles work2 (in collaboration of Karin Rabe at Rutgers) has reveled a low- lying rhombohedral ferroelectric phase, close in energy to the ground state, that is a prime candidate for epitaxial stabilization. More relevant to the problem at hand is the system FeAlO3, which like CaTiO3 forms in the orthorhombic perovskite structure. Is there a low-lying metastable phase of FeAlO3 that is ferroelectric? Can this "hidden ferroelectricity" be revealed? If so, how does the ferroelectric distortion couple to the magnetism?

Hard-soft Heterostructures – what other "exchange bias" devices await at the nanoscale?
Proposed Work: We seek to identify the multiferroic analog of the exchange bias mechanism. We will interleave two or more epitaxially matched bulk materials to form a superlattice structure. In this case one layer will be a "hard" ferroelectric material while the other a "soft" ferroelectric material. By "hard/soft" ferroelectric we mean one where the correlation length of the ferroelectric distortion is long or short respectively. At least one of the materials will also be magnetic. For example, the correlation length of the ferroelectric distortion in BaTiO3 is relatively short (only a few unit cells) as in CaTiO3, which can be seen by the relatively flat dispersion of the unstable (imaginary) phonon branch emanating from the ?-point in Fig. 9. On the other hand, the correlation length of the ferroelectric instability in the perovskite BaMnO3 is relatively short in real space, as seen in Fig. 9 (right) by the highly dispersive unstable mode emanating from the ?-point. Can making a "ferroelectric-spring" system, i.e., a heterostructure of perovskite BaTiO3/BaMnO3, increase the coupling between the ferroelectric instability and the spin system?

Other potential system that we will investigate is the room temperature 3,4
ferroelectric/ferromagnet PbTi1/2Fe1/2O3. Here we will create (PbTiO3)n/(PbFeO3)m
superlattices/

Accomplishments: "What I can not create, I do not understand"- Richard Feymann

RPPR Final Report
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Written on his “Last Blackboards” when he died, I have always found Feynman’s quote surprising when considering the usual approach in materials physics, which is loosely: discover new material, measure properties, and if those properties are exciting, race to write down the Hamiltonian, at the end of which it is usually declared mission accomplished, the fundamental problem is over.

Prior to this PECASE award, I had been asking myself, “in the spirit of Feynman, what is being created in this process?” It has been my strong believe that until we can start with a Hamiltonian and create a realization, we truly do not understand the fundamentals of the problem. I am talking equally about Hamiltonians that describe a known phenomenon in a known material and about those Hamiltonians/phenomena where a material realization is currently unknown. The goal of this PECASE was the start of my long-term goal to develop an ab initio strategy towards Materials Discovery & Designer Hamiltonians.

It may seem strange today, 2021, that this goal for the community at large is not obvious. But when I wrote this proposal, "materials design" was still something that most theorists/physicists had not fully realized accepted as science, A year or two after being awards the PECASE, I was also received the honor of being named a MacArthur "genius" Fellow from the John D. & Catherine T. MacArthur Foundation.

Although we are far from realizing this dream, This PECASE award ha facilitated a few successesl on the pathway towards Designer Hamiltonians in complex materials. One example funded by this PECASE was collaboration with Prof Darrell Schlom in MSE at Cornell --Prof Schlom is an expert in the atomic-layer MBE growth of novel complex oxide materials -- entitled

Exploiting dimensionality and defect mitigation to create tunable microwave dielectrics, Nature 502, (2013)

This was the experimental realization of a highly tunable ground state arising from the emergence of a local ferroelectric instability in biaxially strained Srn+1TinO3n+1 phases with n???3 at frequencies up to 125?GHz.

It was my groups work that suggested - from our materials-by-design approach - that in contrast to traditional methods of modifying ferroelectrics—doping or strain—in this unique system an increase in the separation between the (SrO)2 planes, which can be achieved by changing n, bolsters the local ferroelectric instability. This was based on a prediction by us, that the distribution of phonon instabilities throughout Brillouin Zone in SrTiO3 could be "shaped" by adding "digital" interfaces within SrTiO3 to "disrupt" the coherence of phonon instability. Srn+1TinO3n+1 phases naturally SrO interfaces within SrTiO3.

The (theory) challenge was then to understand how this novel phonon confinement-like effect could be controlled with "n", the separation between interfaces. It tuned out - and the Beauty of a theory-driven experimental pursuit of new materials (rather than simply having experimentalist "chasing" theory predictions) we (theory+experiments) discovered something new and not anticipated by the theory -- that this new control parameter, n, can be exploited to achieve a figure of merit at room temperature that rivals all known tunable microwave dielectrics.

Another success of this PECASE concerns the Major Goal of

(1) revealing hidden phases in bulk materials

in 2011, Nicole Benedek and I published a PRL entitled "Hybrid improper ferroelectricity: a mechanism for controllable polarization-magnetization coupling" (this was not PECASE funded), which had ushered in a new era to the field of ferroelectrics and the field of mutlitferroics. It was then a radical idea that cross-coupling of functionalities and hence "electric-field control of ..." could be achieved if we could understand how ferroelectricity could be induced by the same kind of lattice distortions that controls electronic conductivity & magnetism, namely octahedral rotations (which has been subsequently experimentally confirmed and extended by the community). Without exaggeration the subfield of what became know as hybrid improper ferroelectricity has been exciting for both theorists, experimentalist, and device physicists.

the was however a question which needed to be understood to facilitate practical devices. How does the polarization switch from Up to down in an hybrid improper ferroelectric? Decades of work showed how this happens in perovskite ferroelectrics like BaTiO3, but nothing was known in this new class of multifunctional materials.

In PECASE funded work, Elizabeth Nowadnick and I worked this out and published as an Editors Suggestion in PRB, 20016 entitled "Domains and ferroelectric switching pathways in Ca3Ti2O7 from first principles". Not only did this work show how switching occurs, it revealed that the switching barriers (which are related in a nontrivial way to coercive fields) are much smaller than the "critics" suggested. Turns out, that key to the switching is the fact that a "hidden phase" exists at the domain wall itself!!! This non-bulk phase facilitates ferrroelectric switching via a non-bulk pathway.Notably, in a series of experimental papers (PNAS, nature materials, and Advanced Materials) by the group of Sang-Wok Cheong at Rutgers our idea was confirmed.

Last key result of this PECASE (I'm hesitant to call it a success until experiments realizes the idea) was again published with (now Prof) Nowadnick and again addresses the major goal of

(1) revealing hidden phases in bulk materials

"Coupled structural distortions, domains, and control of phase competition in polar SmBaMn2O6" published as an Editors Suggestion in PRB in 2019. This work looked at an "old" system, the manganites. What we discovered - building upon the "switching" work discussed above - is that domains in this family of compounds are "interface materials' in their own right. While single-phase SmBaMn2O6 could be a new electric-field controllable CMR-like system if made in thin films and epitaxially trained, we discovered that anti-ferroelectric like distortions stabilize a network of domain wall vortices, in which ferroelectricity, ferromagnetism, and metallicity coexist, and postulated how to control these multi functionalities by controlling the domain structure. Furthermore, we show that the crystal structure provides a knob to control competing electronic and magnetic phases at structural domain walls

Training Opportunities: A major goal of this PECASE award was the training of students and post doctorate fellows.In particular, the PI has placed an emphases two aspects of their training: 1) to be successful in "Materials Design", a cross-disciplinary training in physics, chemistry, and materials chemistry is required (which when I wrote the PECASE proposal was had yet to be "fully embraced" by the physics community) and 2) on the "non-science" professional skills that most students have the most trouble realizing are equally important.

the result: of the 5 post docs who participated in PECASE research, 4 have faculty positions, including 2 women* scientists
*Prof Hena Das, Tokyo Institute of Technology,
*Prof Beth Nowadnick, UC-Merced (dept of Materials Science)
Prof Saurabh Ghosh, Department of Physics and Nanotechnology & SRM Research Institute (Chennai)
Prof Sung Gu Kang, University of Ulsan, dept of chemical engineering (Korea)

Results Dissemination: In addition to the > 50 Invited talks and seminars that the PI gave during the time period of this award (at least a 1/3 of which included significant accomplishments from this program), the PI has developed a general audience talk to reach High school Students from disadvantaged backgrounds, similar to his own.

Although I was asked to put together this talk after being awarded a MacArthur "genius" Fellowship in 2013, the section of the talk on "what I do now" is about the research and approach to that research that was supported by the PECASE. The talk
“From throwing rocks and punk rock to designing rocks atom by atom: how a wrong-way kid from Philly became a MacArthur Fellow,”
was presented at:

1. Archbishop Ryan High School, Philadelphia, PA, May 2014
2. The TANMS Center, UCLA, Los Angles CA, June 2014
3. Cornell University CATALYST Academy (~50 URM, juniors and seniors in High School), Ithaca NY, every year since July 2016 (minus obviously 2020).

Honors and Awards: Elected a Fellow of the American Physical Society, via the Division of Materials Physics, in 2015

Received Tenure, School of Applied & Engineering Physics, Cornell University, in 2014.

Elected a MacArthur "genius" Fellow, The John D. and Catherine T. MacArthur Foundation in 2013

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PARTICIPANTS:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Elizabeth Ashley Nowadnick
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Hena Das
Person Months Worked: 5.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Saurabh Ghosh
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Sung Gu Kang
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Brian Michael Abbett
Person Months Worked: 12.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Kuntal Roy
Person Months Worked: 12.00 **Funding Support:**
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Abstract: Pb2MnTeO6, a new double perovskite, was synthesized. Its crystal structure was determined by synchrotron X-ray and powder neutron diffraction. Pb2MnTeO6 is monoclinic (I2/m) at room temperature with a regular arrangement of all the cations in their polyhedra. However, when the temperature is lowered to similar to 120 K it undergoes a phase transition from I2/m to C2/c structure. This transition is accompanied by a displacement of the Pb atoms from the center of their polyhedra due to the 6s2 lone-pair electrons, together with a surprising off-centering of Mn2+ (d5) magnetic cations. This strong first-order phase transition is also evidenced by specific heat, dielectric, Raman, and infrared spectroscopy measurements. The magnetic characterizations indicate an anti-ferromagnetic (AFM) order below TN approximate to 20 K; analysis of powder neutron diffraction data confirms the magnetic structure with propagation vector k = (0 1 0) and collinear AFM spins. The observed jump in dielectric p
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I certify that the information in the report is complete and accurate:
Signature: Craig J Fennie
Signature Date: 4/14/21 4:24PM

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