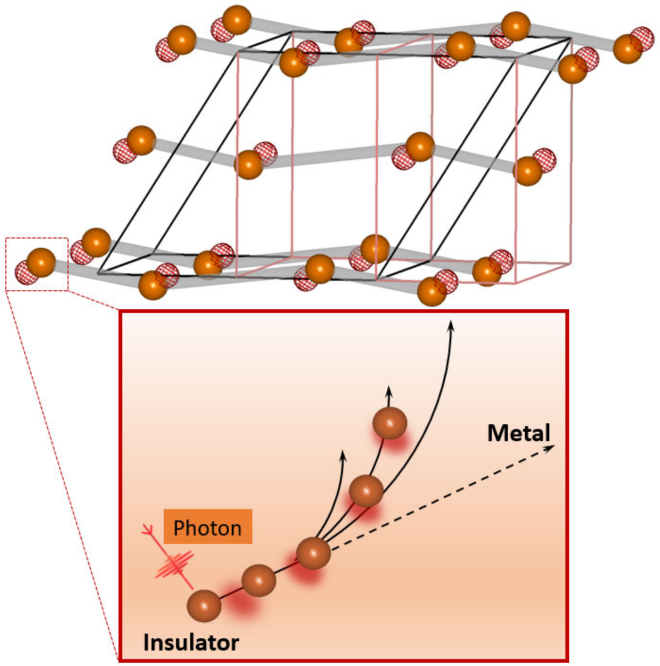
Capture ultrafast atom dimerization and rotation dynamic pathways



Top: Atomic structure of VO_2 in its steady states showing the positions of V-atoms in the insulator phase (orange balls) and the metallic phase (hollow spheres). Bottom: light-pulse triggered insulator to metal transition, where the motion of V-atoms in the first stage is linear toward the metallic state, then curved in the second stage.

Scientific Achievement

We discover unexpected nonlinear atomic motion trajectories during the early stage of the insulator-metal transition (IMT) in vanadium dioxide.

Significance and Impact

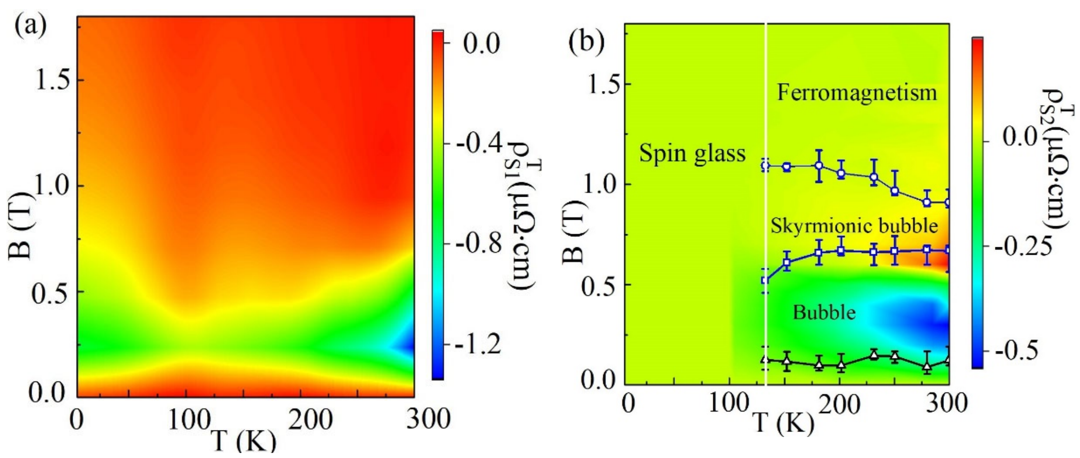
The gigantic resistivity change accompanying the IMT can be used to emulate neurons or synapses in human brains. Understanding IMT dynamics allows us to develop devices for high speed, high accuracy and low energy-consumption neuromorphic computing.

Research Details

- Using MeV-UED we accurately map the atomic motion trajectories through quantitative analysis of diffraction intensity and structure refinements.
- Ab-initio molecular dynamics calculations support the experimental findings and identify the phonons coupling to photoexcitation.

J. Li, L. Wu, S. Yang, X. Jin, W. Wang, J. Tao, L. Boatner, M. Babzien, M. Fedurin, M. Palmer, W. Yin, O. Delaire, and Y. Zhu, "Direct detection of V-V atom dimerization and rotation dynamic pathways upon ultrafast photoexcitation in VO2", Phys. Rev. X 12, 021032 (2022).

Topological Hall Effect Anisotropy in Kagome Bilayer Metal Fe_3Sn_2



Topological Hall anisotropy in Fe_3Sn_2: Topological Hall effect with (a) current along c axis and magnetic field inside Kagome plane and (b) current inside Kagome plane and magnetic field along c axis.

*Qianheng Du, Zhixiang Hu, Myung-Geun Han, Fernando Camino, Yimei Zhu and C. Petrovic, Physical Review Letters **129**, 236601 (2022).*

Scientific Achievement

Large topological Hall effect and detection of skyrmions for interlayer current and in plane magnetic field.

Significance and Impact

The effect is present at high temperarures in micron size crystals typically used in devices.

Research Details

- Single crystals of Fe_3Sn_2 were grown by the high temperature self-flux method at CMPMSD and fabricated using focused ion beam at CFN in mesoscopic size for transport and Lorentz transition electron microscopy (TEM) studies.
- Topological Hall effect is linked to field-induced anisotropy and reorientation in magnetic structure that influences noncollinear spin texture with nonzero scalar spin chirality.
- Spin reorientation and skyrmionics bubbles are confirmed by Lorentz TEM, for the first time when magnetic field is in Kagome planes.

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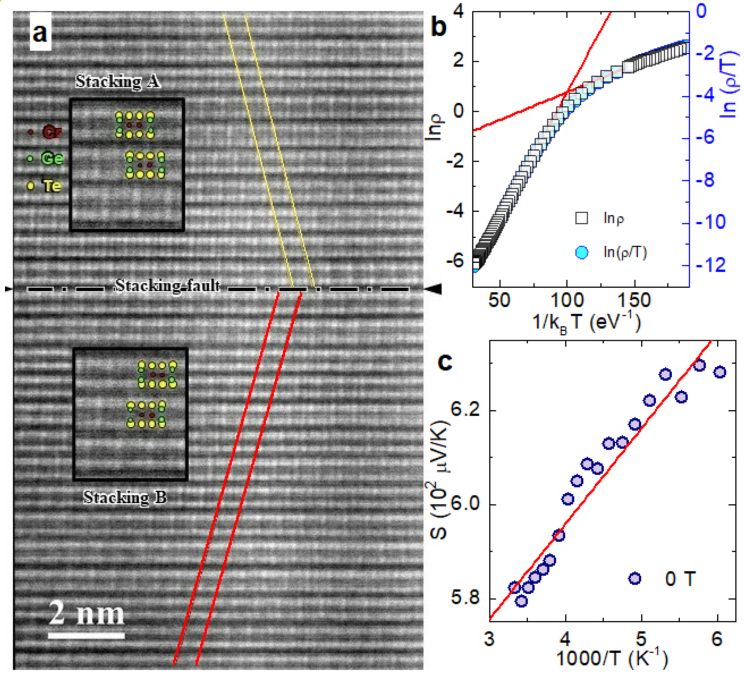
NATIONAL HIGH MAGNETIC FIELD LABORATORY

Brookhaven National Laboratory

Polaronic Conductivity in $Cr_2Ge_2Te_6$



Atomic resolution TEM image a) of selected area in $Cr_2Ge_2Te_6$ crystal. Two insets present the enlarged area showing the two different stackings separated by a stacking fault, indicated with a dotted line. Two inclined yellow and red lines show the stacking sequence of the Te/Ge columns along the c – axis. Temperature-dependent resistivity b) and thermopower c) indicate polaronic transport .

Scientific Achievement

Conduction in magnetic two-dimensional van der Waals (2D vdW) $Cr_2Ge_2Te_6$ crystals is enhanced by stacking fault crystal structure distortion.

Significance and Impact

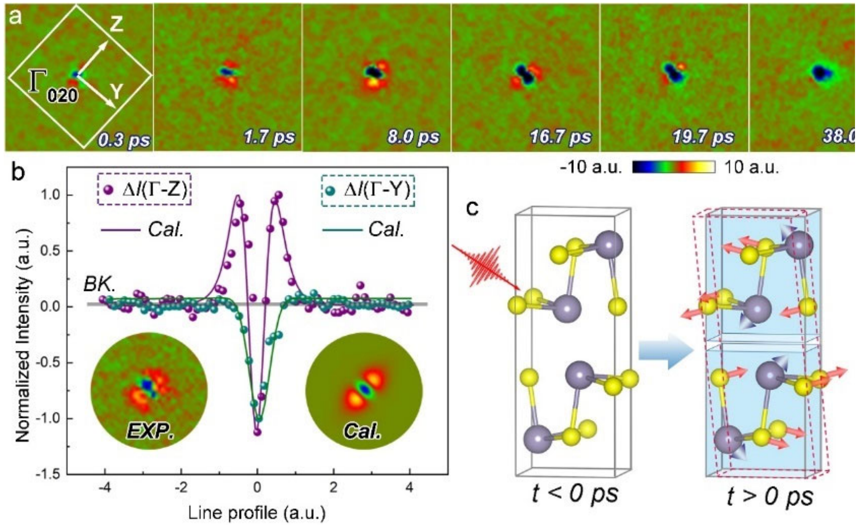
This opens possibility for manipulation of charge transport by electron-phonon and spin-orbit coupling-based tailoring of polaron properties.

Research Details

- Transmission electron microscopy (TEM) imaging reveals stacking faults of vdW layers with about 0.027 nm^{-1} density.
- Conduction states are involved in polaronic transport via hopping and are sensitive on details of magnetocrystalline anisotropy.

Yu Liu, Myung-Geun Han, Yongbin Lee, Michael O. Ogunbunmi, Qianheng Du, Christie Nelson, Zhixiang Hu, Eli Stavitski, David Graf, Klaus Attenkofer, Svilen Bobev, Liqin Ke, Yimei Zhu, and Cedomir Petrovic, "Polaronic conductivity in $Cr_2Ge_2Te_6$ single crystals" Advanced Functional Materials 32, 2105111 (2022).

Photoinduced transient state in thermoelectric SnSe



(a) Time-resolved snapshots of (020) Bragg peak at 90K showing the evolution of its intensity distribution at Γ point of the Brillouin zone. (b) Line profiles of the intensity along the two orthogonal directions (dots are experiment and lines are simulation). (c) Schematic of the photoinduced atomic displacement (yellow balls represent Sn and purple balls Se). Transverse optical phonons (marked by the arrows) are responsible for the observed atomic motion and transient domain state. The domains and their boundaries can be scattering centers for phonons to improve thermoelectric performance of the material.

Scientific Achievement

We reveal photoinduced anharmonic lattice distortion that leads to a transient domain state only surviving for ~20ps at 90K.

Significance and Impact

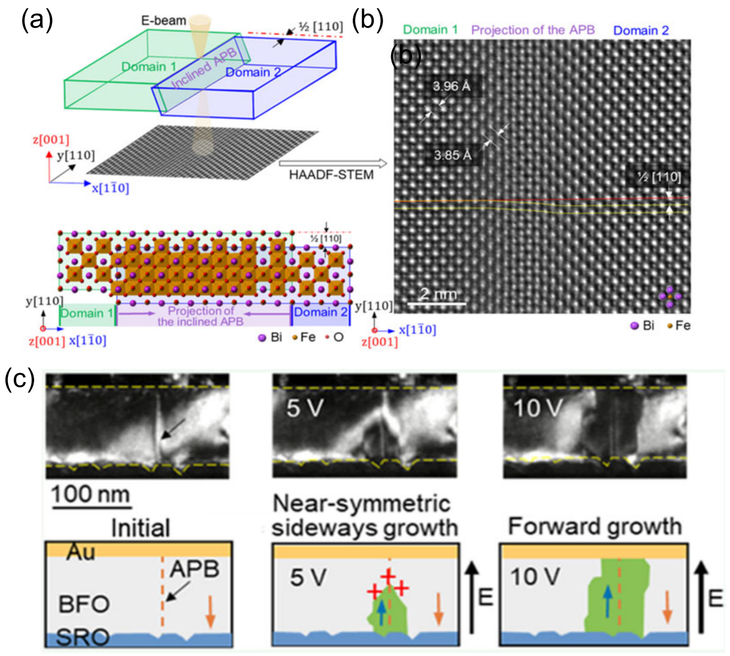
Lattice manipulation via ultrafast laser pulses can introduce novel phonon scattering centers that reduce thermal conductivity and improve the performance of thermoelectric materials.

Research Details

- Femtosecond laser and MeV-UED were used to induce lattice distortion and probe the lattice response dynamics.
- A Monte Carlo based multislice diffraction simulation method as developed to interpret the observed asymmetrical diffuse Huang scattering near the Bragg peaks.
- DFT calculations suggest the distortions correspond to a superposition of a few special phonon modes with strong anharmonicity and phonon scattering.

W. Wang, L. Wu, J. Li, N. Aryal, X. Jin, Y. Liu, M. Fedurin, M. Babzien, R. Kupfer, M. Palmer, C. Petrovic, W. Yin, M.P. M. Dean, I.K. Robinson, J. Tao, Y. Zhu; "Photoinduced anisotropic lattice dynamic response and domain formation in thermoelectric SnSe", npj Quantum Materials 6:97 (2021).

Defect-Engineered Domain Switching in $BiFeO_3$



(a) Schematic showing the relative orientations of the imaging direction and the antiphase boundary (APB). An atomic model in the imaging direction also shown. (b) Atomic resolution scanning transmission electron micrograph of the APB. Lattice parameter measurements show a reduced lattice parameter at the APB. (c) Dark-field TEM images and corresponding domain schematics under external biases showing the domain switching behavior at the APB in a sequence of nucleation, symmetric sideway growth, and forward growth.

Scientific Achievement

We reveal a functional role played by planar-defect in the ferroeletric domain switching.

Significance and Impact

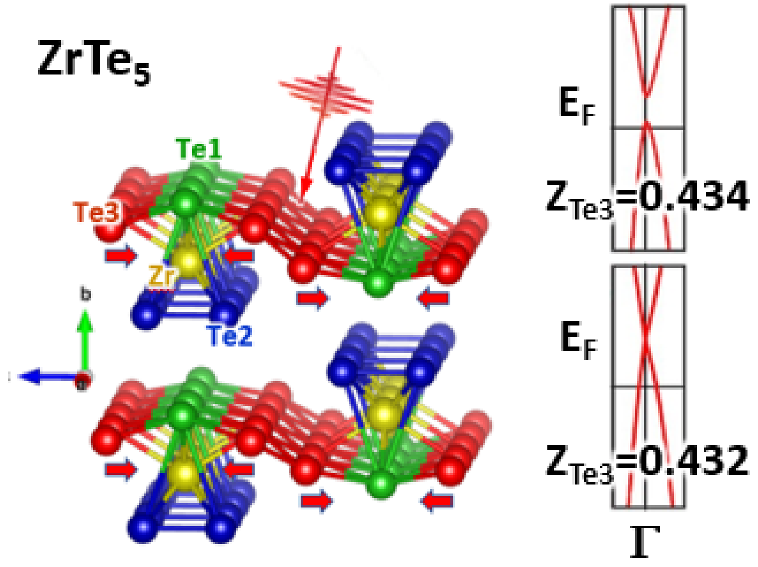
In situ TEM experiments revealed shear-strain-induced nucleation and symmetric domain switching at the structural antiphase boundary in ferroelectric thin films. This study shows that an extended structural defect can be used as an active switching element in reprogrammable domain-wall-based electronic devices.

Research Details

- *In situ* electric biasing in TEM was utilized to extensively study the influence of an antiphase boundary on the switching behavior in a (110)-oriented $BiFeO_3$ thin film.
- Atomic-scale structural analysis including unit-cell shift, lattice parameter, and Fe-displacement are carried out to reveal the structural origins for the symmetric switching.

Y. Zhang, M.-G. Han, D. Sando, L. Wu, N. Valanoor, Y. Zhu;, "Antiphase-Boundary-Engineered Domain Switching in a (110)-Oriented $BiFeO_3$ Film", ACS Applied Electrnonic Materials 3, 3226-3233 (2021).

Capture photon-induced transition form topological insulator to Dirac semimetal



(left panel) Schematic of the $ZrTe_5$ crystal structure showing Te3 lattice displacement (red arrows) along the c-axis in its photoinduced transient state. (right panel) The direct-gaps at Γ point for two Te3 positions derived by DFT. There is a small gap before photoexcitation (top, insulating state). The gap is closed in the presence of strong spin-orbit coupling after photoexcitation (bottom, Semimetal state).

Scientific Achievement

Using photoexcitation to manipulate and control material's emerging behavior and functionality.

Significance and Impact

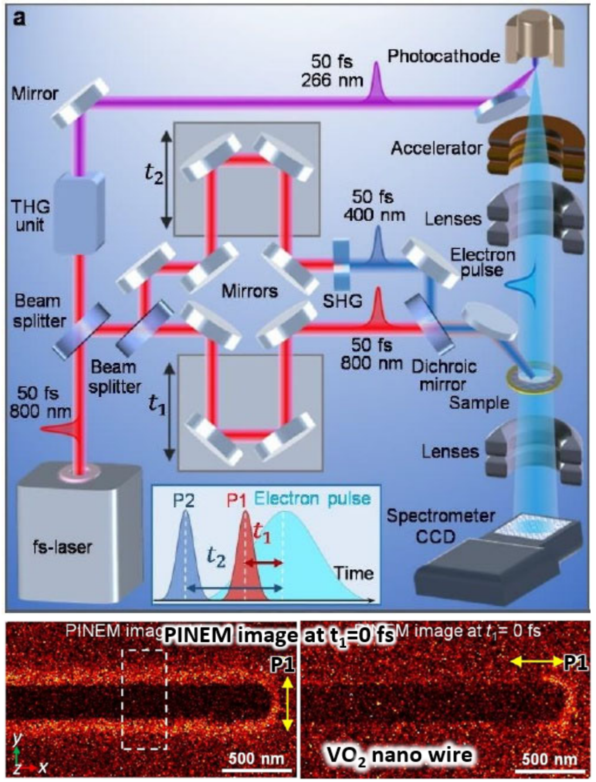
Revealing the femtosecond photon-induced changes in local atomic bond length of $ZrTe_5$ leads to electronic topological phase transition.

Research Details

- Quantitative ultrafast electron diffraction analysis via structure refinement to determine atom positions of the photoexcited transient state without structural symmetry breaking
- With experimentally determined lattice displacements DFT calculations reveal how a transient Dirac semimetal state can be induced by photoexcitation in a topological insulator
- Unraveling two timescales related to electron, spin and lattice coupling and correlations.

Konstantinova, T.; Wu, L.; Yin, W.-G.; Tao, T.; Gu, G. D.; Wang, X.J.; Yang, J.; Zaliznyak, I. A.; Zhu, Y. "Photoinduced chiral Dirac semimetal in ZrTe5", npj Quantum Materials, 5:80, 1-8 (Nov. 2020).

Two-color near-field UEM at nm-fs resolution



(top) Setup of two-color near-field ultrafast electron microscopy. Optical pulse P1 is used for gating and P2 for pumping to improve temporal resolution via PINEM. (bottom) Mapping photoinduced ultrafast dielectric response in VO_2 nanowire with polarization direction perpendicular and parallel to the wire.

Scientific Achievement

Using two-color photoinduced nearfield electron microscopy (PINEM) to achieve 10^{-21} m·s spatiotemporal resolution for ultrafast electron microscopy (UEM)

Significance and Impact

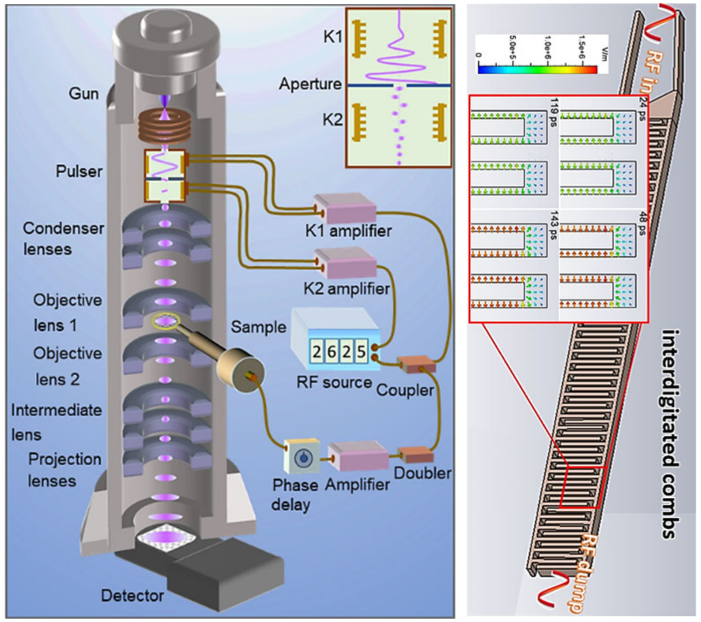
The novel approach allows a spatiotemporal resolution several orders of magnitude better than the conventional optical probes and static imaging, enabling direct visualization of ultrafast dielectric response in Mott insulators and I-M transitions in quantum materials.

Research Details

- The 50fs optical pulses split into two, one for gating via PINEM and the other for pumping to improve resolution.
- Spatially mapping nearfield dynamics of single VO_2 nanowires was compared with theory, revealing ultrafast photo-doping drives the system into a metallic state on a timescale of ~150 fs without yet perturbing the crystalline lattice.

Fu, X.; Barantani, F.; Gargiulo, S.; Madan, I.; Berruto, G.; Lagrange, T.; Jin, L.; Wu, J.; Vanacore, G.M.; Carbone, F.; and Zhu, Y.; "Nanoscale-femtosecond dielectric response of Mott insulators captured by two-color near-field ultrafast electron microscopy", Nat. Comm., 11:5770 (Nov. 2020).

Direct visualization of electromagnetic wave dynamics



(left) Schematic of the laser-free UEM showing the integration of the RF-driven pulser and the frequency-double, delay-control RF circuit for sample excitation. The inset is a schematic of the pulser, consisting of two traveling-wave metallic comb stripline elements ($K_{1,2}$). (right) The interdigitated comb structure used to observe the propagation and dynamics of the EM waves. Snapshots of the 5.25GHz electric field distributions where color and arrow correspond to the amplitude and direction of the waves are also included.

Scientific Achievement

Development of the laser-free frequency-tunable electron pulser and its first application of time-resolved direct visualization of electromagnetic (EM) wave dynamics.

Significance and Impact

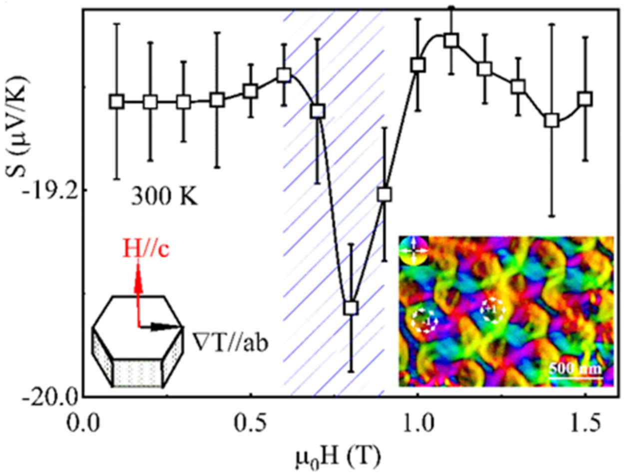
The electron pulser can be retrofit into any conventional microscope for ultrafast electron microscopy (UEM) without compromising its original performance and requiring expensive fs lasers. The ability of visualizing GHz EM wave propagation allows the study of electrodynamics in devices for information transmission/processing technology.

Research Details

- The pulser consists of a modulator and a demodulator (K_1, K_2) with an aperture between them to chop a continuous beam into RF pulses with a tunable repetition rate.
- A protype sample was fabricated to reveal the transient oscillating EM field around the tines of the combs, time-resolved polarization and field distributions. Numerical simulations of the wave dynamics were also incorporated.

Fu, X., Wang, E., Zhao, Y., Liu, A., Montgomery, E., Gokhale, V.J., Gorman, J.J., Jing, C., Lau, J.W., and Zhu, Y., "Direct visualization of electromagnetic wave dynamics by laser-free ultrafast electron microscopy", Science Advances, ***6*** *abc3456 (Oct. 2020).*

Room temperature skyrmion thermopower in Fe_3Sn_2



Magnetic field-dependence of thermopower at 300 K upon the application of magnetic field H orthogonal to thermal gradient ∇T (left inset). When the skyrmion bubbles form, there is a drop in the value of thermopower. The right inset shows the skyrmion bubbles in color-contour composite image. Skyrmions are induced by rapidly changing magnetic field from 1000 to 11.7 mT. Based on the in-plane spin rotation sense, the topological charge of skyrmionic bubble is determined as ±1.

Scientific Achievement

We demonstrate the first room-temperature thermoelectric signature of skyrmion bubbles in a simple thermal gradient. This is observed in Fe_3Sn_2, a Kagome Dirac crystal with massive Dirac fermions.

Significance and Impact

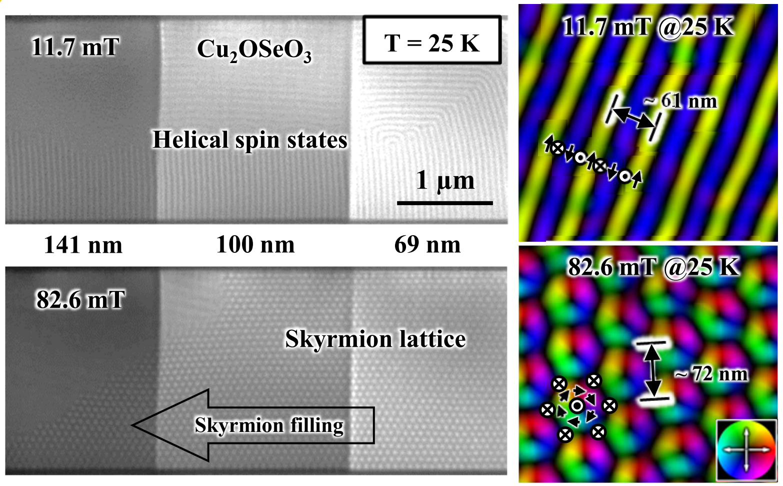
Nanoscale magnetic skyrmions are quintessential embodiment of strong electronic correlations and topology. The results pave the way for future information storage and spin caloritronic devices using skyrmion manipulation by thermal gradients.

Research Details

- Phonon- or magnon-drag contribute at low temperatures but room-temperature thermopower is dominated by electronic diffusion, allowing for the skyrmionics bubble detection.
- Transmission electron microscopy imaging of the magnetic spin textures at room temperature confirms that skyrmionics bubbles are induced in external magnetic field at (600 – 800) mT where thermoelectric anomaly is observed.

Qianheng Du, Muyng-Geun Han, Yu Liu, Weijun Ren, Yimei Zhu and C. Petrovic "Room-Temperature Skyrmion Thermopower in Fe_3Sn_2" Advanced Quantum Technologies, 2000058, 1-6 (2020).

Controlled evolution of skyrmions in multiferroic Cu_2OSeO_3



Magnetic spin textures and their evolution under various magnetic field and temperature in multiferroic Cu_2OSeO_3. Lorentz cryo-electron-microscopy images (left) showing stripe helical spin states and hexagonal skyrmion lattices, respectively. Magnetic induction maps reconstructed by the phase retrieval method (right) showing spin orientation and arrangement (color legend) of helical spin states and skyrmion lattices.

M.-G. Han, et al., Science Advances ***6****, eaax2138 (2020).*

Scientific Achievement

Systematic study on topological chiral spin textures and their transitions reveals two novel phenomena: anisotropic scaling in helix and skyrmion channeling.

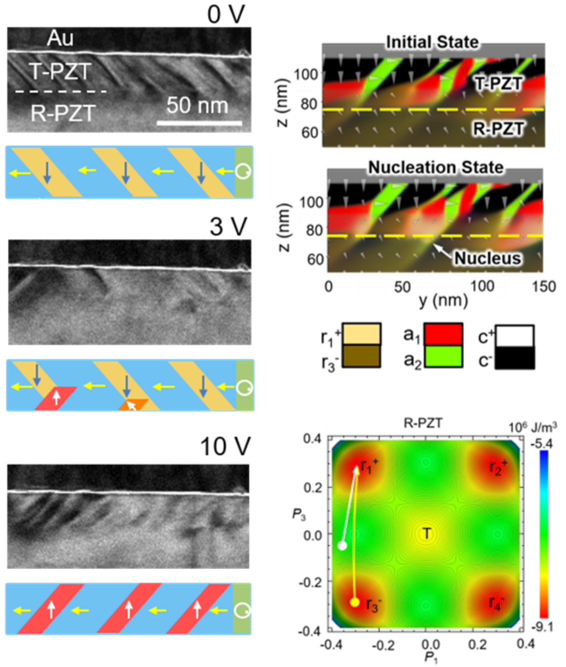
Significance and Impact

Novel spin behaviors provide a viable way towards controlled manipulation of helices and skyrmion lattices, envisaging chirality-controlled skyrmion-based spintronic devices.

Research Details

- Topological spin textures, helices and skyrmions, and their transitions in multiferroic Cu_2OSeO_3 are manipulated and controlled by varying magnetic field, temperature, material thickness and chemical doping. Their spin structures are characterized by cryogenic Lorentz phase microscopy.
- Rotation of spin helix (scaling) and gradual filling of skyrmions (channeling) from thinner to thicker sections are observed with increasing magnetic field.

Programmable ferroelastic domain walls



(Left panel) TEM images showing reproducible 90° rotation of ferroelastic domain walls in the tetragonal $PbZr_{0.2}Ti_{0.8}O_3$ (T-PZT)/rhombohedral $PbZr_{0.55}Ti_{0.45}O_3$ (R-PZT) bilayer, with *c/a* polydomains under external biases. At 3 V, nucleation occurs at the triple junction where the T/R-PZT interface meets the *c/a* domain wall, confirmed by our phase field simulation (Right top). (Right bottom) Free-energy landscape highlights two switching paths: yellow arrow for ordinary rhombohedral switching and white arrow for our experimental case at the triple junction.

Scientific Achievement

We report a deterministic ferroelastic domain switching in ferroelectric bilayers and revealed its underlying switching mechanisms and physical origins for dramatically enhanced electromechanical responses (d_{33} ~ 300 pm/V).

Significance and Impact

Beyond simple observation of electric-field-induced ferroelastic domain walls, reproducible reorientation of ferroelastic domain walls reported in this study provides a viable pathway toward designing electromechanical devices based on programmable ferroelastic domain walls.

Research Details

- Using *in situ* electrical biasing in transmission electron microscope ferroelastic domain switching process are revealed in ferroelectric bilayers.
- Switching pathways including nucleation sites and polarization rotations are elucidated by atomic resolution imaging and confirmed by phase field simulation.

Zhang, Y., Han, M-G., Garlow, JA., Tan, Y., Xue, F., Chen, L-Q., Munroe, P., Nagarajan, V., and Zhu, Y., "Deterministic Ferroelastic Domain Switching Using Ferroelectric Bilayers", Nano Letters, 19, 5319-5326 (2019).