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RPPR Final Report
as of 13-Feb-2023

Agency Code: 21XD

Proposal Number: 68990EMMUR
INVESTIGATOR(S):

Agreement Number: W911NF-16-1-0361

Name: David Hsieh
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EIN: 951643307

Report Date: 31-Jan-2023

Date Received: 29-Jan-2023

Final Report for Period Beginning 01-Jul-2016 and Ending 31-Oct-2022

Title: Quantum Materials by Design with Electromagnetic Excitation

Begin Performance Period: 01-Jul-2016

End Performance Period: 31-Oct-2022

Report Term: 0-Other

Submitted By: David Hsieh

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Phone: (626) 395-4758

Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 1

STEM Participants: 4

Major Goals: The major goals of our MURI program were to create new out-of-equilibrium quantum phases of matter in correlated electron systems via strong electromagnetic field excitation, and to realize strategies to control their properties on-demand. Starting in year 2, our efforts were organized into 4 thrusts identified as the most promising areas of investigation within the scope of our program, and they did not change from that point onwards. The 4 thrusts were: (1) Floquet engineering, (2) Coherent mode control, (3) Photo-doping, and (4) THz control.

Accomplishments: Please see uploaded PDF file.

Training Opportunities: Students and postdocs were given the following training opportunities: mentorship from both their research advisor and other PIs on the MURI team; frequent interactions with members of both their own and other groups on the MURI team; opportunities to present their findings at international conferences and workshops; and technical training across interdisciplinary fields of the MURI through collaboration. These training opportunities provide them with theoretical and lab-based skills, data interpretation and analysis skills, scientific presentation and teaching skills, and professional development pathways.

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Results Dissemination: Results from this project were disseminated via talks and presentations at professional conferences, workshops and universities, publication in scientific journals and outreach initiatives. A summary of talks and presentations on our MURI results is provided below in chronological order. See “Products” section for the publications list.

2016-17

Wilson group gave three contributed talks at the APS March Meeting. Wilson delivered two invited talks at the CHESS annual user meeting and the Oak Ridge Young Investigator meeting.

Stemmer presented at the DARPA workshop “Exotic Materials Achieved by Non-Equilibrium Activation”.

Refael group gave a contributed talk at the APS March Meeting.

Averitt presented a talk to UCSD Undergraduate Women in Physics (UWIP) on ultrafast optical spectroscopy of materials. He also presented a talk to the UCSD Society of Physics Students (SPS) on ultrafast optical spectroscopy of materials.

Hsieh group presented a talk at the APS March Meeting, the conference on Strongly Correlated Electron Systems (SCES) in Prague, Czech Republic, and the Stanford & SLAC Ultrafast Materials Science workshop.

2017-18

Stemmer group gave a contributed talk at the APS March Meeting and also presented a poster at the Workshop on “Correlated Electron Systems – Novel Developments” in Minneapolis, Minnesota.

Wilson group delivered invited talks at the following conferences and universities: CIMTEC 14th Materials Congress; NIST Center for Neutron Research 50th Anniversary Symposium; Chez Pierre MIT Seminar; Physics Department Colloquium, University of Houston.

Refael group presented contributed talks at the APS March Meeting and invited talks at the following conferences and universities: Physics Department Colloquium, University of Washington; KITP Workshop on Dynamical Systems; The Abdus Salam International Centre for Theoretical Physics; Gordon Conference on Non-Equilibrium Quantum Matter and Scalable Quantum Computing.

Averitt group presented talks to the UCSD Undergraduate Women in Physics (UWIP) and to the UCSD Society of Physics Students (SPS) on ultrafast dynamics and control in materials and gave presentations at the APS March Meeting and the Gordon Research Conference on Ultrafast Phenomena in Cooperative Systems.

Hsieh group gave contributed talks at the APS March Meeting.

2018-19

Averitt group gave four invited talks and one contributed talk. Additionally, talks were presented to undergraduate scientists at UCSD through the Society of Physics Students (SPS). Further, lab tours and introductory discussions on laser-based studies of materials were presented to middle school students through the UCSD Young Physicist Program (YPP).

Balents gave talks at the following meetings: Low dimensional emergent phenomena in correlated systems and topological quantum matter, Georgian National Academy of Sciences, Tbilisi, Georgia; CEMS symposium on emergent quantum materials, U. Tokyo.

Hsieh gave talks at the KITP Quantum workshop, UCSB, the Light on the Quantum Leap workshop, Arizona State University, and university seminars at Harvard and UCLA. His group presented this work at the APS March Meeting. They also provided lab tours and discussions on the subject of dynamical engineering of electronic materials for the FUTURE workshop at Caltech for undergraduate women in physics from across the nation.

Refael gave colloquia at Temple University, Johns Hopkins University, and ETH Zurich. Refael's student working on this project presented the work at conferences, including the APS March Meeting.

Stemmer group presented talks at the APS March Meeting.

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Wilson gave a talk and his graduate students delivered contributed talks at the APS March Meeting.

2019-20

Averitt group gave contributed and invited talks at national and international conferences. This includes three invited talks. Additionally, talks on these results were presented to undergraduate scientists at UCSD through the Society of Physics Students (SPS). Further, lab tours and introductory discussions on laser-based studies of materials have been presented to middle school students through the UCSD Young Physicist Program (YPP). Unfortunately, 5 invited talks on the results of this research had to be canceled due to the pandemic.

Balents gave a talk titled “Dynamics of quantum magnets” at the IBSPCS-KIAS International Workshop on Frustrated Magnetism, IBS PCS, Daejeon, South Korea.

Hsieh gave talks at the Nonequilibrium Superconductivity conference (CCQ, Flatiron institute), Nature conference on 2D Materials: Visions of Future Research and Applications (China), and at Stanford University, CU Boulder, Tsinghua University and CSU Long Beach. His group was scheduled to present this work at the cancelled APS March Meeting.

Refael gave a talk on “Floquet higher-order topological insulators: principles and path towards realizations” at the Nonequilibrium Superconductivity conference (CCQ, Flatiron institute), and university seminars at Stanford University and U. Penn.

Stemmer gave an invited talk at the Second International Symposium on Advanced Electron Microscopy and Spectroscopy (SAEMS) and a Plenary talk at the International Symposium on Integrated Functionalities (ISIF 2019), Dublin, Ireland “Intertwined Orders in SrTiO₃: Ferroelectricity and Superconductivity”.

Wilson gave an invited talk at the SSRL Users meeting at Stanford. Additionally, graduate students Eli Zoghlin and Zach Porter planned to deliver contributed talks at the APS March meeting; however this meeting was canceled due to COVID-19.

Young gave a talk on the subject of Pomeranchuk instabilities at an online KITP workshop entitled “Correlated Phases in Moire Materials: One Year Later”.

2020-21

Averitt group presented contributed and invited talks at national and international conferences. This includes five invited talks. Additionally, an introductory talk on photo-induced dynamics in quantum materials was presented at the UC San Diego Undergraduate Women in Physics (UWIP) Winter 2021 Virtual Coffee Hour Series. Balents presented results in several seminars. His students and postdocs also gave talks at the APS March Meeting.

Hsieh group presented at various conferences and workshops (including the APS March meeting) as well as university seminars and colloquia. Hsieh gave invited talks at the MRS Spring/Fall meeting, the Spectroscopy and Strongly Correlated Electron Systems meeting, and the Chinese Physical Society meeting.

Refael presented colloquia on Floquet control at several universities. These included: University of Lisbon, University of Ohio, Monash University, Cal-State Northridge. In addition, Refael presented a popular physics lecture at the Sigma Camp for youth aged 12-16 on a popular physics topic: From energy storage and production to powering space travel - an order of magnitude approach. (<https://sigmacamp.org/2020/lectures>).

Stemmer group presented their work at conferences and workshops. Stemmer gave an invited talk at the (virtual) Fourth International Symposium on Advanced Electron Microscopy and Spectroscopy (SAEMS).

Members of the Wilson group presented growth results at the virtual spring meeting of the Materials Research Society.

2021-22

Averitt group gave a talk at the National Society of Physics Students (SPS) Zone 18 meeting held at University of San Diego (USD). The title of the presentation was “Photons across space, time, and matter”.

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Balents presented results in several seminars and colloquia.

Hsieh group presented at various conferences and workshops (including the APS March Meeting) as well as university seminars and colloquia. Hsieh gave an invited talk at PIPT7: Photoinduced Phase Transitions and Cooperative Phenomena.

Refael presented his results at several conferences.

Stemmer group presented their work at conferences and workshops. Stemmer gave an invited talk in an invited session at the American Physical Society March meeting held in Chicago.

Honors and Awards: 2016-17

Leon Balents received the Yzurdiaga Chair in theoretical physics

Richard Averitt was elected as Fellow of the American Physical Society

Andrea Young was the recipient of both Packard and Sloan Fellowships

David Hsieh received a PECASE award

2017-18

Andrea Young received the New Horizons Prize

2018-19

Leon Balents was elected a member of the National Academy of Sciences

Nuh Gedik was awarded a Miller Visiting Professorship at UC Berkeley

Gil Refael was named a Simons Investigator

Susanne Stemmer received a UCSB Graduate Mentor Award

Andrea Young was awarded a PECASE

2019-20

Nuh Gedik was awarded the 2020 Ludwig-Genzel-Prize and a Moore Experimental Investigator in Quantum Materials

Andrea Young was awarded the IUPAP Young Scientist Prize

2020-21

David Hsieh received the Brown Investigator Award

Susanne Stemmer was recognized as a 2021 Outstanding Referee by the American Physical Society

Andrea Young was a Blavatnik Award Finalist

2021-22

Stephen Wilson was elected as a fellow of the Neutron Scattering Society of America in 2022.

Eli Zoghlin, graduate student of Wilson, supported under the award was also awarded a prestigious Miller postdoctoral fellowship at Johns Hopkins University and will begin that position in September 2022. Eli Zoglin also received a highly competitive DOE SCGSR fellowship to perform work at the Spallation Neutron Source from January 2022 until April 2022.

David Hsieh was named a Moore Experimental Physics Investigator

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI

Participant: David Hsieh

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

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Participant Type: Co PD/PI

Participant: Gil Refael

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Hanbyeol Jeong

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Wangzhou Wu

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Andrea Capa Salinas

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Eli Zoghlin

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Reina Gomez

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Alexander Potts

Person Months Worked: 2.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

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Participant: Abhay Nayak

Person Months Worked: 3.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Leon Balents

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Urban Seifert

Person Months Worked: 2.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Kelson Kaj

Person Months Worked: 3.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Mustafa Ali

Person Months Worked: 3.00

Project Contribution:

National Academy Member: N

Funding Support:

ARTICLES:

RPPR Final Report
as of 13-Feb-2023

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Journal of Crystal Growth
Publication Identifier Type: DOI **Publication Identifier:** 10.1016/j.jcrysgro.2016.12.010
Volume: 459 **Issue:** **First Page #:** 203
Date Submitted: 8/29/17 12:00AM **Date Published:** 2/2/17 12:00AM
Publication Location:

Article Title: Floating zone growth of λ -Na_{0.90}MnO₂ single crystals

Authors: Rebecca Dally, Raphaële J. Clément, Robin Chisnell, Stephanie Taylor, Megan Butala, Vicky Doan-Ngu

Keywords: characterization, floating zone technique, single crystal growth, manganites

Abstract: Single crystal growth of λ -Na_xMnO₂ ($x=0.90$) is reported via the floating zone technique. The conditions required for stable growth and intergrowth-free crystals are described along with the results of trials under alternate growth atmospheres. Chemical and structural characterizations of the resulting λ -Na_{0.90}MnO₂ crystals are performed using ICP-AES NMR, XANES, XPS, and neutron diffraction measurements. As a layered transition metal oxide with large ionic mobility and strong correlation effects, λ -Na_xMnO₂ is of interest to many communities, and the implications of large volume, high purity, single crystal growth are discussed.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevLett.119.186803
Volume: 119 **Issue:** **First Page #:** 186803
Date Submitted: 10/16/18 12:00AM **Date Published:** 11/2/17 2:00PM
Publication Location:

Article Title: Response of the lattice across a filling-controlled Mott metal-insulator transition

Authors: Honggyu Kim, Patrick B. Marshall, Kaveh Ahadi, Thomas E. Mates, Evgeny Mikheev, Susanne Stemme

Keywords: Mott insulator transition, titanates

Abstract: The lattice response of a prototype Mott insulator, SmTiO₃, to hole doping is investigated with atomic-scale spatial resolution. SmTiO₃ films are doped with Sr on the Sm site with concentrations that span the insulating and metallic sides of the filling-controlled Mott metal-insulator transition (MIT). The GdFeO₃-type distortions are investigated using an atomic resolution scanning transmission electron microscopy technique that can resolve small lattice distortions with picometer precision. We show that these distortions are gradually and uniformly reduced as the Sr concentration is increased without any phase separation. Significant distortions persist into the metallic state. The results present a new picture of the physics of this prototype filling-controlled MIT, which is discussed.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: APL Materials
Publication Identifier Type: DOI **Publication Identifier:** 10.1063/1.4998772
Volume: 5 **Issue:** **First Page #:** 096101
Date Submitted: 9/7/17 12:00AM **Date Published:**
Publication Location:

Article Title: Growth of strontium ruthenate films by hybrid molecular beam epitaxy

Authors: Patrick Marshall, Honggyu Kim, Kaveh Ahadi, Susanne Stemmer

Keywords: Ruthenates, thin film MBE

Abstract: We report on the growth of epitaxial Sr₂RuO₄ films using a hybrid molecular beam epitaxy approach in which a volatile precursor containing RuO₄ is used to supply ruthenium and oxygen. The use of the precursor overcomes a number of issues encountered in traditional MBE that uses elemental metal sources. Phase-pure, epitaxial thin films of Sr₂RuO₄ are obtained. At high substrate temperatures, growth proceeds in a layer-by-layer mode with intensity oscillations observed in reflection high-energy electron diffraction. Films are of high structural quality, as documented by x-ray diffraction, atomic force microscopy, and transmission electron microscopy. The method should be suitable for the growth of other complex oxides containing ruthenium, opening up opportunities to investigate thin films that host rich exotic ground states.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Applied Physics Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1063/1.5041282
Volume: 113 **Issue:** 6 **First Page #:** 061113
Date Submitted: 10/16/18 12:00AM **Date Published:** 8/1/18 7:00AM
Publication Location:

Article Title: Terahertz metamaterial perfect absorber with continuously tunable air spacer layer

Authors: Jacob Schalch, Guangwu Duan, Xiaoguang Zhao, Xin Zhang, Richard D. Averitt

Keywords: Far infrared spectroscopy, Absorption spectroscopy, Dielectric properties, Metamaterials

Abstract: We present a comprehensive investigation of a continuously tunable metamaterial perfect absorber operating at terahertz frequencies. In particular, we investigate a three-layer absorber structure consisting of a layer of split ring resonators and a metallic ground plane, with a central layer consisting of a mechanically tunable air-spaced layer. The absorber was characterized using terahertz time-domain spectroscopy in reflection (at normal incidence) as a function of spacer thickness from 0 to 1000??m. Our experimental measurements reveal the detailed evolution of the absorption bands as a function of spacing, in excellent agreement with analysis using interference theory and simulation. Our Fabry-Pérot-like structure provides an avenue for achieving massive tunability in metamaterial absorber devices.

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Journal: Advanced Materials

Publication Identifier Type: DOI

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Volume: 30

Issue: 22

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Date Submitted: 10/16/18 12:00AM

Date Published: 5/1/18 12:00AM

Publication Location:

Article Title: Phototunable Dielectric Huygens' Metasurfaces

Authors: Kebin Fan, Jingdi Zhang, Xinyu Liu, Gu-Feng Zhang, Richard D. Averitt, Willie J. Padilla

Keywords: coupled mode theory, dielectric metasurfaces, eigenmodes, Huygens' metasurfaces, tunable materials

Abstract: Conventional dielectric metasurfaces achieve their properties through geometrical tuning and consequently are static. Although some unique properties are demonstrated, the usefulness for realistic applications is thus inherently limited. Here, control of the resonant eigenmodes supported by Huygens' metasurface (HMS) absorbers through optical excitation is proposed and demonstrated. An intensity transmission modulation depth of 99.93% is demonstrated at 1.03 THz, with an associated phase change of greater than $\pi/2$ rad. Coupled mode theory and S-parameter simulations are used to elucidate the mechanism underlying the dynamics of the metasurface and it is found that the tuning is primarily governed by modification of the magnetic dipole-like odd eigenmode, which both lifts the degeneracy, and eliminates critical coupling. The dynamic HMS demonstrates wide tunability and versatility which is not limited to the spectral range demonstrated, offering a new path for reconfigurable metasurface

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Optica

Publication Identifier Type: DOI

Publication Identifier: 10.1364/OPTICA.5.000303

Volume: 5

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Date Submitted: 10/16/18 12:00AM

Date Published: 3/1/18 8:00AM

Publication Location:

Article Title: Electromechanically tunable metasurface transmission waveplate at terahertz frequencies

Authors: Xiaoguang Zhao, Jacob Schalch, Jingdi Zhang, Huseyin R. Seren, Guangwu Duan, Richard D. Averitt,)

Keywords: Metamaterials; Artificially engineered materials; Polarization-selective devices; Anisotropic optical materials; Optical microelectromechanical devices.

Abstract: Dynamic polarization control of light is essential for numerous applications ranging from enhanced imaging to material characterization and identification. We present a reconfigurable terahertz metasurface quarter-wave plate consisting of electromechanically actuated microcantilever arrays. Our anisotropic metasurface enables tunable polarization conversion through cantilever actuation. Specifically, voltage-based actuation provides mode-selective control of the resonance frequency, enabling real-time tuning of the polarization state of the transmitted light. The polarization tunable metasurface has been fabricated using surface micromachining and characterized using terahertz time domain spectroscopy. We observe a ~ 230 GHz cantilever actuated frequency shift of the resonance mode, sufficient to modulate the transmitted wave from pure circular polarization to linear polarization.

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Journal: Optics Express

Publication Identifier Type: DOI

Publication Identifier: 10.1364/OE.26.002242

Volume: 26

Issue: 3

First Page #: 2242

Date Submitted: 10/16/18 12:00AM

Date Published: 1/1/18 12:00AM

Publication Location:

Article Title: Analysis of the thickness dependence of metamaterial absorbers at terahertz frequencies

Authors: Guangwu Duan, Jacob Schalch, Xiaoguang Zhao, Jingdi Zhang, Richard D. Averitt, Xin Zhang

Keywords: Spectroscopy, Absorption, Spectroscopy, terahertz, metamaterials

Abstract: Metamaterial absorbers typically consist of a metamaterial layer, a dielectric spacer layer, and a metallic ground plane. We have investigated the dependence of the metamaterial absorption maxima on the spacer layer thickness and the reflection coefficient of the metamaterial layer obtained in the absence of the ground plane layer. Specifically, we employ interference theory to obtain an analytical expression for the spacer thickness needed to maximize the absorption at a given frequency. The efficacy of this simple expression is experimentally verified at terahertz frequencies through detailed measurements of the absorption spectra of a series of metamaterials structures with different spacer thicknesses. Using an array of split-ring resonators (SRRs) as the metamaterial layer and SU8 as the spacer material we observe that the absorption peaks redshift as the spacer thickness is increased, in excellent agreement with our analysis.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

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Volume: 97

Issue: 3

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Date Submitted: 10/16/18 12:00AM

Date Published: 1/1/18 12:00AM

Publication Location:

Article Title: Identifying the perfect absorption of metamaterial absorbers

Authors: G. Duan, J. Schalch, X. Zhao, J. Zhang, R. D. Averitt, X. Zhang

Keywords: Electromagnetism, Light propagation, transmission & absorption, Metamaterials

Abstract: We present a detailed analysis of the conditions that result in unity absorption in metamaterial absorbers to guide the design and optimization of this important class of functional electromagnetic composites. Multilayer absorbers consisting of a metamaterial layer, dielectric spacer, and ground plane are specifically considered. Using interference theory, the dielectric spacer thickness and resonant frequency for unity absorption can be numerically determined from the functional dependence of the relative phase shift of the total reflection. Further, using transmission line theory in combination with interference theory we obtain analytical expressions for the unity absorption resonance frequency and corresponding spacer layer thickness in terms of the bare resonant frequency of the metamaterial layer and metallic and dielectric losses within the absorber structure.

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Acknowledged Federal Support: Y

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Volume: 16

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Date Submitted: 10/16/18 12:00AM

Date Published: 10/1/17 12:00AM

Publication Location:

Article Title: Towards properties on demand in quantum materials

Authors: D. N. Basov, R. D. Averitt, D. Hsieh

Keywords: quantum materials

Abstract: The past decade has witnessed an explosion in the field of quantum materials, headlined by the predictions and discoveries of novel Landau-symmetry-broken phases in correlated electron systems, topological phases in systems with strong spin-orbit coupling, and ultra-manipulable materials platforms based on two-dimensional van der Waals crystals. Discovering pathways to experimentally realize quantum phases of matter and exert control over their properties is a central goal of modern condensed-matter physics, which holds promise for a new generation of electronic/photonic devices with currently inaccessible and likely unimaginable functionalities. In this Review, we describe emerging strategies for selectively perturbing microscopic interaction parameters, which can be used to transform materials into a desired quantum state.

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review Letters

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevLett.121.107201

Volume: 121

Issue: 10

First Page #:

Date Submitted: 10/16/18 12:00AM

Date Published: 9/1/18 12:00AM

Publication Location:

Article Title: Floquet Engineering of Multiorbital Mott Insulators: Applications to Orthorhombic Titanates

Authors: Jianpeng Liu, Kasra Hejazi, Leon Balents

Keywords: Floquet systems, Magnetism, Mott insulators, Oxides

Abstract: We consider driving multiorbital Mott insulators using laser radiation. We derive general expressions for periodically driven spin-orbital models using time-dependent perturbation theory in the strong interaction limit. We show that the effective exchange interactions of the Floquet spin-orbital Hamiltonian are highly tunable via variations of the frequency, amplitude, and polarization of the laser. We also take the effect of a finite bandwidth of excitations into account and study possible heating effects. We further apply our formalism to orthorhombic titanates YTiO₃ and LaTiO₃ based on first-principles calculations, and we find that the spin exchange interactions in these compounds can be engineered to a large extent by tuning the frequency and electric-field amplitude of the laser.

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Acknowledged Federal Support: Y

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Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.97.155160
Volume: 97 **Issue:** 15 **First Page #:**
Date Submitted: 10/16/18 12:00AM **Date Published:** 4/1/18 12:00AM
Publication Location:

Article Title: Electron nematic fluid in a strained

Authors: Patrick B. Marshall, Kaveh Ahadi, Honggyu Kim, Susanne Stemmer

Keywords: Ferromagnetism, Magnetotransport, Metamagnetism, Strongly correlated systems, thin films, 4-terminal techniques

Abstract: Sr₃Ru₂O₇ belongs to the family of layered strontium ruthenates and exhibits a range of unusual emergent properties, such as electron nematic behavior and metamagnetism. Here, we show that epitaxial film strain significantly modifies these phenomena. In particular, we observe enhanced magnetic interactions and an electron nematic phase that extends to much higher temperatures and over a larger magnetic-field range than in bulk single crystals. Furthermore, the films show an unusual anisotropic non-Fermi-liquid behavior that is controlled by the direction of the applied magnetic field. At high magnetic fields, the metamagnetic transition to a ferromagnetic phase recovers isotropic Fermi-liquid behavior. The results support the interpretation that these phenomena are linked to the special features of the Fermi surface, which can be tuned by both film strain and an applied magnetic field.

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.98.064310
Volume: 98 **Issue:** 6 **First Page #:**
Date Submitted: 10/16/18 12:00AM **Date Published:** 8/1/18 12:00AM
Publication Location:

Article Title: Berry electrodynamics: Anomalous drift and pumping from a time-dependent Berry connection

Authors: Swati Chaudhary, Manuel Endres, Gil Refael

Keywords: Cold gases in optical lattices, Geometric & topological phases, Topological phases of matter, Transport properties, Ultracold gases

Abstract: The Berry curvature of a Bloch band can be interpreted as a local magnetic field in reciprocal space. This analogy can be extended by defining an electric field analog in reciprocal space which arises from the time-dependent Berry connection. We explore the term in the semiclassical equation of motion that gives rise to this phenomenon, and show that it can lead to anomalous drift in wave-packet motion. A similar effect arises from changes in the band population due to periodic driving, where the resulting drift depends on the nature of the drive and can be expressed in terms of a shift vector. Finally, these effects can be combined to build a pump with a net anomalous drift during a cyclic evolution in momentum space.

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Journal: SciPost Physics

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Publication Identifier: 10.21468/SciPostPhys

Volume: 5

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Date Submitted: 10/16/18 12:00AM

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Publication Location:

Article Title: From dynamical localization to bunching in interacting Floquet systems

Authors: Yuval Baum, Evert P. L. van Nieuwenburg, Gil Refael

Keywords: Dynamical localization, Floquet engineering

Abstract: We show that a quantum many-body system may be controlled by means of Floquet engineering, i.e., their properties may be controlled and manipulated by employing periodic driving. We present a concrete driving scheme that allows control over the nature of mobile units and the amount of diffusion in generic many-body systems. We demonstrate these ideas for the Fermi-Hubbard model, where the drive renders doubly occupied sites (doublons) the mobile excitations in the system. In particular, we show that the amount of diffusion in the system and the level of fermion-pairing may be controlled and understood solely in terms of the doublon dynamics. We find that under certain circumstances the diffusion in the system may be eliminated completely. We conclude our work by generalizing these ideas to generic many-body systems.

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Volume: 97

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Date Submitted: 10/17/18 12:00AM

Date Published: 4/1/18 12:00AM

Publication Location:

Article Title: Topological energy conversion through the bulk or the boundary of driven systems

Authors: Yang Peng, Gil Refael

Keywords: Floquet systems, Spin-orbit coupling, Surface states, Synthetic gauge fields, Topological phases of matter

Abstract: Combining physical and synthetic dimensions allows a controllable realization and manipulation of high-dimensional topological states. In our work, we introduce two quasiperiodically driven one-dimensional systems which enable tunable topological energy conversion between different driving sources. Using three drives, we realize a four-dimensional quantum Hall state which allows energy conversion between two of the drives within the bulk of the one-dimensional system. With only two drives, we achieve energy conversion between the two at the edge of the chain. Both effects are a manifestation of the effective axion electrodynamics in a three-dimensional time-reversal-invariant topological insulator. Furthermore, we explore the effects of disorder and commensurability of the driving frequencies, and show the phenomena are robust.

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Publication Location:

Article Title: Absence of thermalization in finite isolated interacting Floquet systems

Authors: Karthik Seetharam, Paraj Titum, Michael Kolodrubetz, Gil Refael

Keywords: Bloch-Floquet theorem, Hubbard model, Numerical techniques

Abstract: Conventional wisdom suggests that the long-time behavior of isolated interacting periodically driven (Floquet) systems is a featureless maximal-entropy state characterized by an infinite temperature. Efforts to thwart this uninteresting fixed point include adding sufficient disorder to realize a Floquet many-body localized phase or working in a narrow region of drive frequencies to achieve glassy nonthermal behavior at long time. Here we show that in clean systems the Floquet eigenstates can exhibit nonthermal behavior due to finite system size. We consider a one-dimensional system of spinless fermions with nearest-neighbor interactions where the interaction term is driven. Interestingly, even with no static component of the interaction, the quasienergy spectrum contains gaps and a significant fraction of the Floquet eigenstates, at all quasienergies, have nonthermal average doublon densities.

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Date Submitted: 10/17/18 12:00AM

Date Published: 6/1/18 12:00AM

Publication Location:

Article Title: Quantized transport and steady states of Floquet topological insulators

Authors: Iliya Esin, Mark S. Rudner, Gil Refael, Netanel H. Lindner

Keywords: Edge states, Floquet systems, Semiconductors, Topological insulators, Transport properties, Bloch-Floquet theorem

Abstract: Robust electronic edge or surface modes play key roles in the fascinating quantized responses exhibited by topological materials. Even in trivial materials, topological bands and edge states can be induced dynamically by a time-periodic drive. Such Floquet topological insulators (FTIs) inherently exist out of equilibrium; the extent to which they can host quantized transport, which depends on the steady-state population of their dynamically induced edge states, remains a crucial question. In this work, we obtain the steady states of two-dimensional FTIs in the presence of the natural dissipation mechanisms present in solid state systems. We give conditions under which the steady-state distribution resembles that of a topological insulator in the Floquet basis. In this state, the distribution in the Floquet edge modes exhibits a sharp feature akin to a Fermi level, while the bulk hosts a small density of excitations.

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Date Submitted: 10/17/18 12:00AM

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Publication Location:

Article Title: Setting Boundaries with Memory: Generation of Topological Boundary States in Floquet-Induced Synthetic Crystals

Authors: Yuval Baum, Gil Refael

Keywords: Photonics, Topological materials

Abstract: When a d-dimensional quantum system is subjected to a periodic drive, it may be treated as a (d+1)-dimensional system, where the extra dimension is a synthetic one. This approach, however, affords only a limited level of control of the effective potential along the synthetic direction. In this work, we introduce a new mean for controlling the Floquet synthetic dimension. We show that arbitrary potentials, as well as edges in the synthetic dimension, could be introduced using a memory component in the system's dynamics. We demonstrate this principle by exploring topological edge states propagating normal to synthetic dimensions. Such systems may act as an optical isolator which allows the transmission of light in a directional way. Also, we suggest an experimental realization of the memory effect in spins coupled to nanofabricated Weyl semimetal surface states.

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Volume: 9 Issue: 1

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Date Submitted: 10/17/18 12:00AM

Date Published: 1/1/18 4:00PM

Publication Location:

Article Title: Unidirectional spin density wave state in metallic $(\text{Sr}_{1-x}\text{La}_x)\text{TiO}_4$

Authors: Xiang Chen, Julian L. Schmeh, Zahirul Islam, Zach Porter, Eli Zoghlin, Kenneth Finkelstein, Jacob P. C

Keywords: strongly correlated electrons, spin-orbit coupling

Abstract: Materials that exhibit both strong spin-orbit coupling and electron correlation effects are predicted to host numerous new electronic states. One prominent example is the $\nu=1/2$ Mott state in SrTiO_4 , where introducing carriers is predicted to manifest high temperature superconductivity analogous to the $\nu=1/2$ Mott state of La_2CuO_4 . While bulk superconductivity currently remains elusive, anomalous quasiparticle behaviors paralleling those in the cuprates such as pseudogap formation and the formation of a d-wave gap are observed upon electron-doping SrTiO_4 . Here we establish a magnetic parallel between electron-doped SrTiO_4 and hole-doped La_2CuO_4 by unveiling a spin density wave state in electron-doped SrTiO_4 . Our magnetic resonant X-ray scattering data reveal the presence of an incommensurate magnetic state reminiscent of the diagonal spin density wave state observed in the monolayer cuprate $(\text{La}_{1-x}\text{Sr}_x)\text{CuO}_4$.

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Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevLett.120.127601
Volume: 120 **Issue:** 12 **First Page #:**
Date Submitted: 10/17/18 12:00AM **Date Published:** 3/1/18 8:00AM
Publication Location:

Article Title: Floquet Engineering in Quantum Chains

Authors: D.M. Kennes, A. de la Torre, A. Ron, D. Hsieh, A.J. Millis

Keywords: Order parameters, Quantum statistical mechanics, 1-dimensional systems, Quantum spin models, Density matrix renormalization group, Exact solutions for many-body systems, Lattice models in condensed matter

Abstract: We consider a one-dimensional interacting spinless fermion model, which displays the well-known Luttinger liquid (LL) to charge density wave (CDW) transition as a function of the ratio between the strength of the interaction U and the hopping J . We subject this system to a spatially uniform drive which is ramped up over a finite time interval and becomes time periodic in the long-time limit. We show that by using a density matrix renormalization group approach formulated for infinite system sizes, we can access the large-time limit even when the drive induces finite heating. When both the initial and long-time states are in the gapless (LL) phase, the final state has power-law correlations for all ramp speeds. However, when the initial and final state are gapped (CDW phase), we find a pseudothermal state with an effective temperature that depends on the ramp rate.

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Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.98.155128
Volume: 98 **Issue:** **First Page #:** 155128
Date Submitted: 10/17/18 12:00AM **Date Published:** 10/16/18 7:00AM
Publication Location:

Article Title: Sr₃Ir₂O₇F₂: Topochemical conversion of a relativistic Mott state into a spin-orbit driven band insulator

Authors: Christi Peterson, Michael W. Swift, Zach Porter, Raphaële J. Clément, Guang Wu, G. H. Ahn, S. J. Moo

Keywords: Iridates, Magnetic insulators, Mott insulators, Single crystal materials, Density functional theory, Infrared spectroscopy, Magnetization measurements, Neutron diffraction, Nuclear magnetic resonance, X-ray diffraction

Abstract: The topochemical transformation of single crystals of Sr₃Ir₂O₇ into Sr₃Ir₂O₇F₂ is reported via fluorine insertion. Characterization of the newly formed Sr₃Ir₂O₇F₂ phase shows a nearly complete oxidation of Ir⁴⁺ cations into Ir⁵⁺ that in turn drives the system from an antiferromagnetic Mott insulator with a half-filled $J_{\text{eff}}=1/2$ band into a nonmagnetic $J=0$ band insulator. First principles calculations reveal a remarkably flat insertion energy that locally drives the fluorination process to completion. Band structure calculations support the formation of a band insulator whose charge gap relies on the strong spin-orbit coupling inherent in the Ir metal ions of this compound.

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Date Submitted: 9/2/19 12:00AM

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Publication Location:

Article Title: Dimensional crossover in a layered ferromagnet detected by spin correlation driven distortions

Authors: A. Ron, E. Zoghlin, L. Balents, S. D. Wilson, D. Hsieh

Keywords: 2D magnets, second harmonic generation

Abstract: Magneto-elastic distortions are commonly detected across magnetic long-range ordering (LRO) transitions. In principle, they are also induced by the magnetic short-range ordering (SRO) that precedes a LRO transition. However these distortions are difficult to resolve because the associated atomic displacements are exceedingly small and do not break symmetry. Here we demonstrate high-multipole nonlinear optical polarimetry as a sensitive and mode selective probe of SRO induced distortions using CrSiTe₃ as a testbed. This compound is composed of weakly bonded sheets of nearly isotropic ferromagnetically interacting spins that, in the Heisenberg limit, would individually be impeded from LRO by the Mermin-Wagner theorem. Our results show that CrSiTe₃ evades this law via a two-step crossover from two- to three-dimensional magnetic SRO, manifested through two successive and previously undetected totally symmetric distortions above its Curie temperature.

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Date Submitted: 8/29/21 12:00AM

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Publication Location:

Article Title: A linear magneto-electric phase in ultrathin MnPS₃ probed by optical second harmonic generation

Authors: Hao Chu, Chang Jae Roh, Joshua O. Island, Chen Li, Sungmin Lee, Jingjing Chen, Je-Geun Park, Andri

Keywords: second harmonic generation, 2D antiferromagnet, magneto-electric effect

Abstract: The transition metal thiophosphates MPS₃ (M = Mn, Fe, Ni) are a class of van der Waals stacked insulating antiferromagnets that can be exfoliated down to the ultrathin limit. MnPS₃ is particularly interesting because its Neel ordered state breaks both spatial-inversion and time-reversal symmetries, allowing for a linear magneto-electric that is rare among van der Waals materials. However, it is unknown whether this unique magnetic structure of bulk MnPS₃ remains stable in the ultrathin limit. Using optical second harmonic generation rotational anisotropy, we show that long-range linear magneto-electric type Neel order in MnPS₃ persists down to at least 5.3 nm thickness. However an unusual mirror symmetry breaking develops in ultrathin samples on SiO₂ substrates that is absent in bulk materials, which is likely related to substrate induced strain.

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Publication Location:

Article Title: Decoupling of static and dynamic antiferromagnetic critical behavior in an optically pumped Mott insulator

Authors: A. de la Torre, M. Buchhold, Y. Baum, G. Zhang, N. J. Laurita, J. W. Harter, L. Zhao, I. Phinney, X. Chei

Keywords: Mott insulators, critical phenomena, photo-doping

Abstract: Dynamically driven interacting quantum many-body systems have the potential to exhibit exotic properties that defy the laws of equilibrium statistical mechanics. A paradigm for strongly interacting quantum particles is the half-filled Fermi-Hubbard model, which hosts an antiferromagnetic Mott insulating ground state. It has theoretically been shown that following a sudden perturbation, a plethora of thermally inaccessible transient magnetic phenomena can emerge. Here we demonstrate the capability to impulsively suppress antiferromagnetism in the Mott insulator Sr₂IrO₄ using ultrafast n-type photo-doping, and to interrogate its ensuing spin dynamics using a combination of time-resolved second harmonic optical polarimetry and coherent magnon spectroscopy. In striking contrast to the equilibrium theory of critical phenomena, the divergences of the antiferromagnetic correlation length and relaxation time decouple when driven out-of-equilibrium.

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Date Submitted: 9/2/19 12:00AM

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Publication Location:

Article Title: Quasiparticle relaxation dynamics in URu₂xFe_xSi₂ single crystals

Authors: Peter Kissin, Sheng Ran, Dylan Lovinger, Verner K. Thorsmølle, Noravee Kanchanavatee, Kevin Huang

Keywords: heavy fermions, ultrafast spectroscopy

Abstract: We investigate quasiparticle relaxation dynamics in URu₂xFe_xSi₂ single crystals using ultrafast optical-pump optical-probe (OPOP) spectroscopy as a function of temperature and Fe substitution (x), crossing from the hidden-order (HO) phase (x=0) to the large-moment antiferromagnet (LMAFM) phase (x=0.12). At low temperature, the dynamics for x=0 and x=0.12 are consistent with the low-energy electronic structure of the HO and LMAFM phases that emerge from the high-temperature paramagnetic (PM) phase. In contrast, near the bicritical point separating HO and LMAFM (x=0.1), two transitions occur over a narrow temperature range (from 15.5–17.5 K). A PM to HO transition occurs at an intermediate temperature followed by a transition to the LMAFM phase at lower temperature.

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Publication Location:

Article Title: Floquet spin and spin-orbital Hamiltonians and doublon-holon generations in periodically driven Mott insulators

Authors: Kasra Hejazi, Jianpeng Liu, Leon Balents

Keywords: Floquet engineering, spin order

Abstract: We consider Mott insulators driven by periodic coherent laser radiation, using both single-orbital and multiorbital models, noting that the latter is of more interest in solid-state systems. We derive general expressions for the resulting periodically driven spin models and spin-orbital models using time-dependent perturbation theory. First, we show that the effective exchange interactions of the Floquet Hamiltonians are highly tunable by the frequency, amplitude, and polarization of the laser. Second, we take the effect of finite bandwidth of excitations into account and study possible heating effects. Using the same formalism with a slight modification we also consider the small-frequency regime and study the dielectric breakdown of Mott insulators.

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Publication Location:

Article Title: Optical excitation of magnons in an easy-plane antiferromagnet; application to Sr₂IrO₄

Authors: U. F. P. Seifert, L. Balents

Keywords: iridates, coherent magnons

Abstract: In condensed matter systems, higher temperature typically disfavors ordered phases leading to an upper critical temperature for magnetism, superconductivity, and other phenomena. A notable exception is the Pomeranchuk effect in ³He, in which the liquid ground state freezes upon increasing the temperature. The anomalous transition arises due to the large spin entropy of the paramagnetic solid phase, which can overcome the ground state energy difference at finite temperature. Here we show that a similar mechanism describes the finite temperature dynamics of spin- and valley- isospins in magic-angle twisted bilayer graphene. In the vicinity of one hole per superlattice unit cell, quantum oscillations show the ground state to be an isospin unpolarized Fermi liquid. Tilted field magnetotransport measurements show that increasing the Zeeman energy, E_Z , drives a transition from the Fermi liquid to a spin- and valley- polarized isospin ferromagnetic state. The same transition can be actuated

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Date Submitted: 8/29/21 12:00AM

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Publication Location:

Article Title: Orbital Floquet Engineering of Exchange Interactions in Magnetic Materials

Authors: Swati Chaudhary, David Hsieh, Gil Refael

Keywords: Floquet engineering, AC Stark effect

Abstract: The transition metal thiophosphates MPS3 (M = Mn, Fe, Ni) are a class of van der Waals stacked insulating antiferromagnets that can be exfoliated down to the ultrathin limit. MnPS3 is particularly interesting because its Neel ordered state breaks both spatial-inversion and time-reversal symmetries, allowing for a linear magneto-electric that is rare among van der Waals materials. However, it is unknown whether this unique magnetic structure of bulk MnPS3 remains stable in the ultrathin limit. Using optical second harmonic generation rotational anisotropy, we show that long-range linear magneto-electric type Neel order in MnPS3 persists down to at least 5.3 nm thickness. However an unusual mirror symmetry breaking develops in ultrathin samples on SiO2 substrates that is absent in bulk materials, which is likely related to substrate induced strain.

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Publication Location:

Article Title: Cooling by photo-doping -- Light-induced symmetry breaking in the Hubbard model

Authors: Philipp Werner, Martin Eckstein, Markus Müller, Gil Refael

Keywords: Photo-doping, symmetry breaking

Abstract: An elusive goal in the field of driven quantum matter is the induction of long-range order. Here, we propose a mechanism based on light-induced evaporative cooling of holes in a correlated fermionic system. Since the entropy of a filled narrow band grows rapidly with hole doping, the isentropic transfer of holes from a doped Mott insulator to such a band results in a drop of temperature. Strongly correlated Fermi liquids and symmetry-broken states could thus be produced by dipolar excitations. Using nonequilibrium dynamical mean field theory, we show that suitably designed chirped pulses may realize this cooling effect. In particular, we demonstrate the emergence of antiferromagnetic order in a system which is initially in a weakly correlated state above the maximum Néel temperature. Our work suggests a general strategy for inducing strong correlation phenomena in periodically modulated atomic gases in optical lattices or light-driven materials.

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Volume: 123 **Issue:** 1 **First Page #:**
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Publication Location:

Article Title: Floquet Second-Order Topological Insulators from Nonsymmorphic Space-Time Symmetries

Authors: Yang Peng, Gil Refael

Keywords: Topological insulators, Floquet engineering

Abstract: We propose a systematic way of constructing Floquet second-order topological insulators (SOTIs) based on time-glide symmetry, a nonsymmorphic space-time symmetry that is unique in Floquet systems. In particular, we are able to show that the static enlarged Hamiltonian in the frequency domain acquires the reflection symmetry, which is inherited from the time-glide symmetry of the original system. As a consequence, one can construct a variety of time-glide symmetric Floquet SOTIs using the knowledge of static SOTIs. Moreover, the time-glide symmetry only needs to be implemented approximately in practice, enhancing the prospects of experimental realizations. We consider two examples, a 2D system in class AIII and a 3D system in class A, to illustrate our ideas, and then present a general recipe for constructing Floquet SOTIs in all symmetry classes.

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Date Submitted: 9/2/19 12:00AM **Date Published:** 3/1/19 8:00AM
Publication Location:

Article Title: Topological frequency conversion in a driven dissipative quantum cavity

Authors: Frederik Nathan, Ivar Martin, Gil Refael

Keywords: Quantized pumping, entangled states

Abstract: Recent work [Martin et al., Phys. Rev. X 7, 041008 (2017)] shows that a spin coupled to two externally supplied circularly polarized electromagnetic modes can effectuate a topological, quantized transfer of photons from one mode to the other. Here, we study the effect in the case when only one of the modes is externally provided, while the other is a dynamical quantum mechanical cavity mode. Focusing on the signatures and stability under experimentally accessible conditions, we show that the effect persists down to the few-photon quantum limit and that it can be used to generate highly entangled “cat states” of cavity and spin. By tuning the strength of the external drive to a “sweet spot,” the quantized pumping can arise starting from an empty (zero-photon) cavity state. We also find that inclusion of external noise and dissipation does not suppress but rather stabilizes the conversion effect, even after multiple cavity modes are taken into account.

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Date Submitted: 9/2/19 12:00AM

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Publication Location:

Article Title: Anisotropic magnetoresistance in the itinerant antiferromagnetic EuTiO₃

Authors: Kaveh Ahadi, Xuezeng Lu, Salva Salmani-Rezaie, Patrick B. Marshall, James M. Rondinelli, Susanne S

Keywords: magnetoresistance, titanates

Abstract: We report on measurements of the anisotropic magnetoresistance (AMR) of doped EuTiO₃. It is shown that the primary contribution to the AMR is the crystalline component, which depends on the relative orientation between the magnetic moments and the crystal axes. With increasing magnetic field, a fourfold crystalline AMR undergoes a change in its alignment with respect to the crystal axes. The results are discussed in the context of the coupling between spin canting, electronic structure, and transport. We discuss the potential role of Weyl points in the band structure. At high fields, the AMR transitions to uniaxial symmetry, which is lower than that of the lattice, along with a crossover from positive to negative magnetoresistance.

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Article Title: Evolution of structure and magnetism across the metal-insulator transition in the pyrochlore iridate (Nd_{1-x}Ca_x)₂Ir₂O₇

Authors: Zach Porter, Eli Zoghlin, Samuel Britner, Samra Husremovic, Jacob P. C. Ruff, Yongseong Choi, Daniel

Keywords: iridates,

Abstract: We report on the evolution of the thermal metal-insulator transition in polycrystalline samples of Nd₂Ir₂O₇ upon hole doping via substitution of Ca²⁺ for Nd³⁺. Ca substitution mediates a filling-controlled Mott-like transition with minimal resolvable structural changes and without altering site symmetry. Local structure confirms that Ca substitution does not result in local chemical phase separation, and absorption spectroscopy establishes that Ir cations maintain a spin-orbit entangled electronic configuration. The metal-insulator transition coincides with antiferromagnetic ordering on the Ir sublattice for all measured samples, and both decrease in onset temperature with Ca content. Weak low-temperature upturns in susceptibility and resistivity for samples with high Ca content suggest that Nd sublattice antiferromagnetism continues to couple to carriers in the metallic regime.

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Volume: 122 Issue: 15

First Page #:

Date Submitted: 9/2/19 12:00AM

Date Published: 4/1/19 7:00AM

Publication Location:

Article Title: Overdamped Antiferromagnetic Strange Metal State in Sr₃IrRuO₇

Authors: Julian L. Schmeh, Thomas R. Mion, Zach Porter, Michael Aling, Huibo Cao, Mary H. Upton, Zahirul Isla

Keywords: iridates, resonant x-ray scattering

Abstract: The unconventional electronic ground state of Sr₃IrRuO₇ is explored via resonant x-ray scattering techniques and angle-resolved photoemission measurements. As the Ru content approaches $x=0.5$ in Sr₃(Ir_{1-x}Ru_x)₂O₇, intermediate to the Jeff=1/2 Mott state in Sr₃Ir₂O₇ and the quantum critical metal in Sr₃Ru₂O₇, a thermodynamically distinct metallic state emerges. The electronic structure of this intermediate phase lacks coherent quasiparticles, and charge transport exhibits a linear temperature dependence over a wide range of temperatures. Spin dynamics associated with the long-range antiferromagnetism of this phase show nearly local, overdamped magnetic excitations and an anomalously large energy scale of 200 meV—an energy far in excess of exchange energies present within either the Sr₃Ir₂O₇ or Sr₃Ru₂O₇ solid-solution end points. Overdamped quasiparticle dynamics driven by strong spin-charge coupling are proposed to explain the incoherent spectral features of the strange metal state.

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Volume: 31 Issue: 24

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Publication Location:

Article Title: Preferential quenching of 5d antiferromagnetic order in Sr₃(Ir_{1-x}Mn_x)₂O₇.

Authors: Julian L Schmeh, Eli Zoghlin, Zach Porter, Xiaoping Wang, Jacob P C Ruff, Wei Tian, Zahirul Islam, St

Keywords: antiferromagnetism, iridates

Abstract: The breakdown of J=1/2 antiferromagnetism in the limit of strong disorder is studied in Sr₃(Ir_{1-x}Mn_x)₂O₇. Upon Mn-substitution, antiferromagnetic ordering of the Ir cations becomes increasingly two-dimensional, resulting in the complete suppression of long-range Ir magnetic order above $x = 0.25$. Long-range antiferromagnetism however persists on the Mn sites to higher Mn concentrations ($x > 0.25$) and is necessarily mediated via a random network of majority Ir sites. Our data suggest a shift in the Mn valence from Mn⁴⁺ to Mn³⁺ at intermediate doping levels, which in turn generates nonmagnetic Ir⁵⁺ sites and suppresses long-range order within the Ir network. The collapse of long-range J=1/2 antiferromagnetism and the survival of percolating antiferromagnetic order on Mn-sites demonstrates a complex 3d–5d exchange process that surprisingly enables minority Mn spins to order far below the conventional percolation threshold for a bilayer square lattice.

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Volume: 99

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Publication Location:

Article Title: Polarized Raman spectroscopy study of metallic

Authors: Wencan Jin, Siwen Li, Jianpeng Liu, Qiang Han, Zach Porter, Christi Peterson, Julian Schmeh, Ibrahim

Keywords: Raman scattering, iridates

Abstract: We have used rotational anisotropic polarized Raman spectroscopy to study the symmetries, the temperature, and the doping dependence of the charge ordered state in metallic $(\text{Sr}_{1-x}\text{La}_x)\text{Ir}_2\text{O}_7$. Despite the fact that the Raman probe size is greater than the charge ordering length, we establish that the charge ordering breaks the fourfold rotational symmetry of the underlying tetragonal crystal lattice into twofold, as well as the translational symmetry, and forms short-range domains with 90° rotated charge order wave vectors, as soon as the charge order sets in below $T_{\text{CO}} \approx 200\text{K}$ and across the doping-induced insulator metal transition. We observe that this charge order mode frequency remains nearly constant over a wide temperature range and up to the highest doping level. These features above are highly reminiscent of the ubiquitous unidirectional charge order in underdoped high-TC copper-oxide-based superconductors (cuprates).

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Journal: Nature

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Publication Identifier: 10.1038/s41586-019-1304-2

Volume: 571

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Date Submitted: 9/2/19 12:00AM

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Publication Location:

Article Title: Spin-orbit-driven band inversion in bilayer graphene by the van der Waals proximity effect

Authors: J. O. Island, X. Cui, C. Lewandowski, J. Y. Khoo, E. M. Spanton, H. Zhou, D. Rhodes, J. C. Hone, T. Ta

Keywords: graphene, quantum spin Hall effect

Abstract: Spin-orbit coupling (SOC) is the key to realizing time-reversal-invariant topological phases of matter. SOC was predicted by Kane and Mele to stabilize a quantum spin Hall insulator; however, the weak intrinsic SOC in monolayer graphene has precluded experimental observation in this material. Here we exploit a layer-selective proximity effect—achieved via a van der Waals contact with a semiconducting transition-metal dichalcogenide—to engineer Kane–Mele SOC in ultra clean bilayer graphene.

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Date Submitted: 8/29/21 12:00AM

Date Published: 11/4/20 8:00AM

Publication Location:

Article Title: Ultrafast enhancement of ferromagnetic spin exchange induced by ligand-to-metal charge transfer

Authors: A. Ron, S. Chaudhary, G. Zhang, H. Ning, E. Zoghlin, S. D. Wilson, R. D. Averitt, G. Refael, D. Hsieh

Keywords: Ultrafast magnetism

Abstract: We theoretically predict and experimentally demonstrate a non-thermal pathway to optically enhance superexchange interaction energies in a material based on exciting ligand-to-metal charge transfer transitions, which introduces lower-order virtual hopping contributions that are absent in the ground state. We demonstrate this effect in the layered ferromagnetic insulator CrSiTe₃ by exciting Te-to-Cr charge-transfer transitions using ultrashort laser pulses, and detecting coherent phonon oscillations that are impulsively generated by superexchange enhancement via magnetoelastic coupling. This mechanism onsets below the temperature scale where short-range in-plane spin correlations begin to develop, and disappears when the excitation energy is tuned away from the charge-transfer resonance, consistent with our predictions.

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Volume: 372

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Date Submitted: 8/29/21 12:00AM

Date Published: 6/18/21 7:00AM

Publication Location:

Article Title: Imaging orbital ferromagnetism in a moiré Chern insulator

Authors: C. L. Tschirhart, M. Serlin, H. Polshyn, A. Shragai, Z. Xia, J. Zhu, Y. Zhang, K. Watanabe, T. Taniguchi,

Keywords: Moire materials

Abstract: Electrons in moiré flat band systems can spontaneously break time reversal symmetry, giving rise to a quantized anomalous Hall effect. Here we use a superconducting quantum interference device to image stray magnetic fields in one such system composed of twisted bilayer graphene aligned to hexagonal boron nitride. We find a magnetization of several Bohr magnetons per charge carrier, demonstrating that the magnetism is primarily orbital in nature. Our measurements reveal a large change in the magnetization as the chemical potential is swept across the quantum anomalous Hall gap consistent with the expected contribution of chiral edge states to the magnetization of an orbital Chern insulator. Mapping the spatial evolution of field-driven magnetic reversal, we find a series of reproducible micron scale domains whose boundaries host chiral edge states.

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Journal: Applied Physics Letters

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Date Submitted: 8/28/20 12:00AM

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Publication Location:

Article Title: On-chip terahertz modulation and emission with integrated graphene junctions

Authors: Joshua O. Island, Peter Kissin, Jacob Schalch, Xiaomeng Cui, Sheikh Rubaiat Ul Haque, Alex Potts, Ta

Keywords: On-chip THz spectroscopy

Abstract: Electrons in moiré flat band systems can spontaneously break time reversal symmetry, giving rise to a quantized anomalous Hall effect. Here we use a superconducting quantum interference device to image stray magnetic fields in one such system composed of twisted bilayer graphene aligned to hexagonal boron nitride. We find a magnetization of several Bohr magnetons per charge carrier, demonstrating that the magnetism is primarily orbital in nature. Our measurements reveal a large change in the magnetization as the chemical potential is swept across the quantum anomalous Hall gap consistent with the expected contribution of chiral edge states to the magnetization of an orbital Chern insulator. Mapping the spatial evolution of field-driven magnetic reversal, we find a series of reproducible micron scale domains whose boundaries host chiral edge states.

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Journal: Nature

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Date Submitted: 8/29/21 12:00AM

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Publication Location:

Article Title: Isospin Pomeranchuk effect and the entropy of collective excitations in twisted bilayer graphene

Authors: Yu Saito, Jingyuan Ge, Kenji Watanabe, Takashi Taniguchi, Erez Berg, Andrea F. Young

Keywords: Graphene

Abstract: In condensed matter systems, higher temperature typically disfavors ordered phases leading to an upper critical temperature for magnetism, superconductivity, and other phenomena. A notable exception is the Pomeranchuk effect in ^3He , in which the liquid ground state freezes upon increasing the temperature. The anomalous transition arises due to the large spin entropy of the paramagnetic solid phase, which can overcome the ground state energy difference at finite temperature. Here we show that a similar mechanism describes the finite temperature dynamics of spin- and valley- isospins in magic-angle twisted bilayer graphene. In the vicinity of one hole per superlattice unit cell, quantum oscillations show the ground state to be an isospin unpolarized Fermi liquid. Tilted field magnetotransport measurements show that increasing the Zeeman energy, E_Z , drives a transition from the Fermi liquid to a spin- and valley- polarized isospin ferromagnetic state. The same transition can be actuated

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Volume: 2

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Date Submitted: 8/28/20 12:00AM

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Publication Location:

Article Title: Phonon dynamics in the Kitaev spin liquid

Authors: Mengxing Ye, Rafael M. Fernandes, Natalia B. Perkins

Keywords: Quantum spin liquid

Abstract: The search for fractionalization in quantum spin liquids largely relies on their decoupling with the environment. However, the spin-lattice interaction is inevitable in a real setting. While the Majorana fermion evades a strong decay due to the gradient form of spin-lattice coupling, the study of the phonon dynamics may serve as an indirect probe of fractionalization of spin degrees of freedom. Here we propose that the signatures of fractionalization can be seen in the sound attenuation and the Hall viscosity. Despite the fact that both quantities can be related to the imaginary part of the phonon self-energy, their origins are quite different, and the time-reversal symmetry breaking is required for the Hall viscosity. First, we compute the sound attenuation due to a phonon decay by scattering with a pair of Majorana fermions and show that it is linear in temperature (T). We argue that it has a particular angular dependence providing the information about the spin-lattice coupling and

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Publication Location:

Article Title: Mapping the structural, magnetic and electronic behaviour of $(\text{Eu}_{1-x}\text{Ca}_x)\text{2Ir2O7}$ across a metal-insulator transition

Authors: Eli Zoghlin, Zach Porter, Samuel Britner, Samra Husremovic, Yongseong Choi, Daniel Haskel, Geneva I

Keywords: iridates, metal insulator transition

Abstract: In this study, we employ bulk electronic properties characterization and x-ray scattering/spectroscopy techniques to map the structural, magnetic and electronic properties of $(\text{Eu}_{1-x}\text{Ca}_x)\text{2Ir2O7}$ as a function of Ca-doping. As expected, the metal-insulator transition temperature, TMIT, decreases with Ca-doping until a metallic state is realized down to 2 K. In contrast, TAFM becomes decoupled from the MIT and (likely short-range) AFM order persists into the metallic regime. This decoupling is understood as a result of the onset of an electronically phase separated state, the occurrence of which seemingly depends on both synthesis method and rare earth site magnetism. PDF analysis suggests that electronic phase separation occurs without accompanying chemical phase segregation or changes in the short-range crystallographic symmetry while synchrotron x-ray diffraction confirms that there is no change in the long-range crystallographic symmetry. X-ray absorption measurements confirm the Jeff

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Date Submitted: 8/29/21 12:00AM

Date Published: 10/16/20 7:00AM

Publication Location:

Article Title: Optically induced flat bands in twisted bilayer graphene

Authors: Or Katz, Gil Refael, Netanel H. Lindner

Keywords: graphene, Floquet

Abstract: Twisted bilayer graphene at the magic twist angle features flat energy bands, which lead to superconductivity and strong correlation physics. These unique properties are typically limited to a narrow range of twist angles around the magic angle with a small allowed tolerance. Here, we report on a mechanism that enables flattening of the band structure using coherent optical illumination, leading to emergence of flat isolated Floquet-Bloch bands. We show that the effect can be realized with relatively weak optical beams at the visible-infrared range (below the material bandwidth) and persist for a wide range of small twist angles, increasing the allowed twist tolerance by an order of magnitude. We discuss the conditions under which these bands exhibit a nonzero Chern number. These optically induced flat bands could potentially host strongly correlated nonequilibrium electronic states of matter.

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Date Submitted: 8/29/21 12:00AM

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Publication Location:

Article Title: Phonon-induced Floquet second-order topological phases protected by space-time symmetries

Authors: Swati Chaudhary, Arbel Haim, Yang Peng, Gil Refael

Keywords: phonon, topological

Abstract: For systems with spatial and nonspatial symmetries, the topological classification depends not only on these symmetries but also on the commutation/anticommutation relations between spatial and nonspatial symmetries. The coexistence of spatial and nonspatial symmetries together with appropriate commutation/anticommutation relations between them can give rise to crystalline and higher-order topological phases, which host gapless boundary modes. Alternatively, space-time symmetries in a Floquet system can take the role of spatial symmetries in deciding the topological classification. Promoting a spatial symmetry to a space-time symmetry can alter the commutation relations, which in turn can modify the topological properties of the system. We show how a coherently excited phonon mode can be used to promote a spatial symmetry with which the static system is always trivial to a space-time symmetry which supports a nontrivial Floquet topological phase. We demonstrate this effect by considering

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Journal: Physical Review Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevLett.125.087601
Volume: 125 **Issue:** **First Page #:** 087601
Date Submitted: 8/29/21 12:00AM **Date Published:** 8/20/20 7:00AM
Publication Location:

Article Title: Order-Disorder Ferroelectric Transition of Strained SrTiO₃

Authors: Salva Salmani-Rezaie, Kaveh Ahadi, William M. Strickland, Susanne Stemmer

Keywords: ferroelectric, transmission electron microscopy

Abstract: SrTiO₃ is an incipient ferroelectric that is believed to exhibit a prototype displacive, soft mode ferroelectric transition when subjected to mechanical stress or alloying. We use high-angle annular dark-field imaging in scanning transmission electron microscopy to reveal local polar regions in the room-temperature, paraelectric phase of strained SrTiO₃ films, which undergo a ferroelectric transition at low temperatures. These films contain nanometer-sized domains in which the Ti columns are displaced. In contrast, these nanodomains are absent in unstrained films, which do not become ferroelectric. The results show that the ferroelectric transition of strained SrTiO₃ is an order-disorder transition. We discuss the impact of the results on the nature of the ferroelectric transition of SrTiO₃.

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Journal: Nanoletters
Publication Identifier Type: DOI **Publication Identifier:** 10.1021/acs.nanolett.0c02285
Volume: 20 **Issue:** **First Page #:** 6542
Date Submitted: 8/29/21 12:00AM **Date Published:** 8/6/20 7:00AM
Publication Location:

Article Title: Polar nanodomains in a ferroelectric superconductor

Authors: Salva Salmani-Rezaie, Kaveh Ahadi, Susanne Stemmer

Keywords: ferroelectric, domains

Abstract: The mechanisms by which itinerant carriers compete with polar crystal distortions are a key unresolved issue for polar superconductors, which offer new routes to unconventional Cooper pairing. Strained, doped SrTiO₃ films undergo successive ferroelectric and superconducting transitions, making them ideal candidates to elucidate the nature of this competition. Here, we reveal these interactions using scanning transmission electron microscopy studies of the evolution of polar nanodomains as a function of doping. These nanodomains are a precursor to the ferroelectric phase and a measure of long-range Coulomb interactions. With increasing doping, the magnitude of the polar displacements, the nanodomain size, and the Curie temperature are systematically suppressed. In addition, we show that disorder caused by the dopant atoms themselves presents a second contribution to the destabilization of the ferroelectric state. The results provide evidence for two distinct mechanisms that suppress the

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Volume: 102

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Date Submitted: 8/31/20 12:00AM

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Publication Location:

Article Title: Magnetoelastic coupling to coherent acoustic phonon modes in the ferrimagnetic insulator

Authors: D. J. Lovinger, E. Zoghlin, P. Kissin, G. Ahn, K. Ahadi, P. Kim, M. Poore, S. Stemmer, S. J. Moon, S. D.

Keywords: ultrafast magnetism, titanates

Abstract: In this work we investigate single crystal GdTlO₃, a promising candidate material for Floquet engineering and magnetic control, using ultrafast optical pump-probe reflectivity and magneto-optical Kerr spectroscopy. GdTlO₃ is a Mott-Hubbard insulator with a ferrimagnetic and orbitally ordered ground state (TC = 32 K). We observe multiple signatures of the magnetic phase transition in the photoinduced reflectivity signal, in response to above band-gap 660-nm excitation. Magnetic dynamics measured via Kerr spectroscopy reveal optical perturbation of the ferrimagnetic order on spin-lattice coupling timescales, highlighting the competition between the Gd³⁺ and Ti³⁺ magnetic sub-lattices. Furthermore, a strong coherent oscillation is present in the reflection and Kerr dynamics, attributable to an acoustic strain wave launched by the pump pulse. The amplitude of this acoustic mode is highly dependent on the magnetic order of the system, growing sharply in magnitude at TC, indicative of strong

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Date Submitted: 8/29/21 12:00AM

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Publication Location:

Article Title: Ultrathin terahertz triple band metamaterial absorbers: consideration of interlayer coupling

Authors: Chunxu Chen, Sultan Can, Jacob Schalch, Xiaoguang Zhao, Guangwu Duan, Richard D. Averitt and Xir

Keywords: metamaterials, terahertz

Abstract: We present a polarization insensitive air spaced triple band metamaterial perfect absorber (MMPA) consisting of a metamaterial layer and metallic ground plane operating at terahertz frequencies. Three near-unity absorption peaks can be individually determined by the geometry of the ring resonators within one-unit cell since the inter-unit cell coupling is negligible. However, for sufficiently small interlayer spacing (~ 25 nm), coupling between the metamaterial layer and the ground plane is non-negligible. Therefore, near-field interactions must be taken into account for a full understanding of the electromagnetic response (EMR). Interference theory is often used to model the EMR of MMPAs analytically, in which interlayer coupling between the metamaterial and ground plane is usually neglected, resulting in a predicted blueshift of the absorption peaks in comparison to experiment. To account for near-field coupling, we incorporate correction terms into the analytical interference model

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Journal: Annual Review of Condensed Matter Physics
Publication Identifier Type: DOI **Publication Identifier:** 10.1146/annurev-conmatphys-031218-01371
Volume: 12 **Issue:** 1 **First Page #:** 247
Date Submitted: 8/29/21 12:00AM **Date Published:** 3/1/21 8:00AM
Publication Location:
Article Title: Topology and Symmetry of Quantum Materials via Nonlinear Optical Responses
Authors: J. Orenstein, J.E. Moore, T. Morimoto, D.H. Torchinsky, J.W. Harter, D. Hsieh
Keywords: nonlinear response, second harmonic generation
Abstract: The transition metal thiophosphates MPS3 (M = Mn, Fe, Ni) are a class of van der Waals stacked insulating antiferromagnets that can be exfoliated down to the ultrathin limit. MnPS3 is particularly interesting because its Neel ordered state breaks both spatial-inversion and time-reversal symmetries, allowing for a linear magneto-electric that is rare among van der Waals materials. However, it is unknown whether this unique magnetic structure of bulk MnPS3 remains stable in the ultrathin limit. Using optical second harmonic generation rotational anisotropy, we show that long-range linear magneto-electric type Neel order in MnPS3 persists down to at least 5.3 nm thickness. However an unusual mirror symmetry breaking develops in ultrathin samples on SiO2 substrates that is absent in bulk materials, which is likely related to substrate induced strain.
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Journal: Nature
Publication Identifier Type: DOI **Publication Identifier:** 10.1038/s41586-021-04051-8
Volume: 600 **Issue:** **First Page #:** 235
Date Submitted: 8/23/22 12:00AM **Date Published:** 12/8/21 4:00PM
Publication Location:
Article Title: Giant modulation of optical nonlinearity by Floquet engineering
Authors: Jun-Yi Shan, M. Ye, H. Chu, Sungmin Lee, Je-Geun Park, L. Balents and D. Hsieh
Keywords: Floquet engineering, 2D magnets
Abstract: Strong periodic driving with light offers the potential to coherently manipulate the properties of quantum materials on ultrafast timescales. Recently, strategies have emerged to drastically alter electronic and magnetic properties by optically inducing non-trivial band topologies, emergent spin interactions and even superconductivity. However, the prospects and methods of coherently engineering optical properties on demand are far less understood. Here we demonstrate coherent control and giant modulation of optical nonlinearity in a van der Waals layered magnetic insulator MnPS3. By driving far off-resonance from the lowest on-site Mn d-d transition, we observe a coherent on-off switching of its optical second harmonic generation efficiency on the timescale of 100 femtoseconds with no measurable dissipation.
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Date Published: 12/1/20 8:00AM

Publication Location:

Article Title: Hyperbolic Cooper-Pair Polaritons in Planar Graphene/Cuprate Plasmonic Cavities

Authors: Michael E. Berkowitz, Brian S. Y. Kim, Guangxin Ni, Alexander S. McLeod, Chiu Fan Bowen Lo, Zhiyuan

Keywords: Cuprate superconductors graphene hyperbolic Cooper-pair polaritons plasmonic cavities van der Waals heterostructures

Abstract: Hyperbolic Cooper-pair polaritons (HCP) in cuprate superconductors are of fundamental interest due to their potential for providing insights into the nature of unconventional superconductivity. Here, we critically assess an experimental approach using near-field imaging to probe HCP in Bi₂Sr₂CaCu₂O_{8+x} (Bi-2212) in the presence of graphene surface plasmon polaritons (SPP). Our simulations show that inherently weak HCP features in the near-field can be strongly enhanced when coupled to graphene SPP in layered graphene/hexagonal boron nitride (hBN)/Bi-2212 heterostructures. This enhancement arises from our multilayered structures effectively acting as plasmonic cavities capable of altering collective modes of a layered superconductor by modifying its electromagnetic environment. The degree of enhancement can be selectively controlled by tuning the insulating spacer thickness with atomic precision. Finally, we verify the expected renormalization of room-temperature graphene SPP using near-

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Date Published: 11/1/20 7:00AM

Publication Location:

Article Title: Spin-orbit-enhanced magnetic surface second-harmonic generation in Sr₂IrO₄

Authors: K. L. Seyler, A. de la Torre, Z. Porter, E. Zoghlin, R. Polski, M. Nguyen, S. Nadj-Perge, S. D. Wilson, D.

Keywords: Antiferromagnetism, second harmonic generation

Abstract: An anomalous optical second-harmonic generation (SHG) signal was previously reported in Sr₂IrO₄ and attributed to a hidden odd-parity bulk magnetic state. Here we investigate the origin of this SHG signal using a combination of bulk magnetic susceptibility, magnetic-field-dependent SHG rotational anisotropy, and overlapping wide-field SHG imaging and atomic force microscopy measurements. We find that the anomalous SHG signal exhibits a twofold rotational symmetry as a function of in-plane magnetic field orientation that is associated with a crystallographic distortion. We also show a change in SHG signal across step edges that tracks the bulk antiferromagnetic stacking pattern. While we do not rule out the existence of hidden order in Sr₂IrO₄, our results altogether show that the anomalous SHG signal in parent Sr₂IrO₄ originates instead from a surface-magnetization-induced electric-dipole process that is enhanced by strong spin-orbit coupling.

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Journal: Physical Review B

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Date Submitted: 8/29/21 12:00AM

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Publication Location:

Article Title: Controlling ligand-mediated exchange interactions in periodically driven magnetic materials

Authors: Swati Chaudhary, Alon Ron, David Hsieh, Gil Refael

Keywords: Floquet engineering, magnetic exchange, van der Waals materials

Abstract: A periodic drive could alter the effective exchange interactions in magnetic materials. Here, we explore how exchange pathways affect the effective interactions of periodically driven magnetic materials. Aiming to apply Floquet engineering methods to two-dimensional magnetic materials, we consider realistic models and discuss the effect of a periodic drive on ligand-mediated exchange interactions. We show that depending on bond angles and the number of ligand ions involved in the exchange process, drive-induced changes can be very different from those calculated from direct-hopping models considered earlier. We study these effects and find that the presence of ligand ions must be taken into account, especially for TMTCs where ligand ion mediated next-neighbor interactions play a crucial role in determining the magnetic ground state of the system.

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Date Submitted: 8/23/22 12:00AM

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Publication Location:

Article Title: On-demand terahertz surface wave generation with microelectromechanical-system-based metasurface

Authors: Chunxu Chen, Kelson Kaj, Xiaoguang Zhao, Yuwei Huang, Richard D. Averitt, Xin Zhang

Keywords: metamaterials, terahertz

Abstract: During the past decade, metasurfaces have shown great potential to complement standard optics, providing novel pathways to control the phase, amplitude, and polarization of electromagnetic waves utilizing arrays of subwavelength resonators. We present dynamic surface wave switching at terahertz frequencies utilizing a mechanically reconfigurable metasurface. Our metasurface is based on a microelectromechanical system consisting of an array of micro-cantilever structures, enabling dynamic tuning between a plane wave and a SW for normal incidence terahertz radiation. This is realized using line-by-line voltage control of the cantilever displacements to achieve full-span phase control. Full-wave electromagnetic simulations and terahertz time-domain spectroscopy agree with coupled mode theory, which was employed to design the metasurface device. A conversion efficiency of nearly 60 percent has been achieved upon switching between the PW and SW configurations.

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Journal: Nature Reviews Physics

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Volume: 4

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Date Submitted: 8/23/22 12:00AM

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Publication Location:

Article Title: Imaging with metamaterials

Authors: Willie J. Padilla, Richard D. Averitt

Keywords: metamaterials, terahertz

Abstract: Metamaterials enable precise tailoring of light–matter interactions at the subwavelength scale, permitting access to the full range of electromagnetic responses encoded in Maxwell’s equations and operating across a huge swath of the electromagnetic spectrum. These salient features have, during the past two decades, fuelled fundamental metamaterials research which, in turn, has unveiled an impressive assemblage of potential applications. Imaging is at the forefront of these developments, leveraging the ability of metamaterials to achieve arbitrary, specific and tunable scattering responses that are poised for integration with a host of materials and devices. We review the impact of metamaterials and metasurface on imaging science and technology from microwave to optical frequencies. Our focus is on metamaterial-based imaging components and the associated imaging modalities that benefit from these advances.

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Date Submitted: 8/23/22 12:00AM

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Publication Location:

Article Title: Broadband Terahertz Silicon Membrane Metasurface Absorber

Authors: Yuwei Huang, Kelson Kaj, Chunxu Chen, Zhiwei Yang, Sheikh Rubaiat Ul Haque, Yuan Zhang, Xiaoguo

Keywords: terahertz, metasurface

Abstract: Metasurface absorbers are of particular interest in numerous photonic applications including detectors, photovoltaic cells, and emissivity coatings. We introduce a thin membrane silicon metasurface absorber with periodic elliptical holes that, as demonstrated theoretically and experimentally, achieves very high absorption over a 500 GHz bandwidth at normal incidence. Based on the analysis of the effective medium theory, the broadband absorption is attributed to proximal electric and magnetic dipole resonances. The absorption amplitude can also be tuned by 20 percent with above-gap photoexcitation. Due to the unit cell geometry, the carrier density on the top surface and sidewalls of the membrane must be taken into account. Our dynamic membrane silicon metasurface absorber is notably thin and CMOS-compatible, providing a promising platform to realize compact terahertz devices including detectors, modulators, and switches.

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Journal: Advanced Optical Materials
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Volume: 9 **Issue:** 22 **First Page #:** 2101215
Date Submitted: 8/23/22 12:00AM **Date Published:** 9/1/21 7:00AM
Publication Location:

Article Title: Tunable Toroidal Response in a Reconfigurable Terahertz Metamaterial

Authors: Chunxu Chen, Kelson Kaj, Yuwei Huang, Xiaoguang Zhao, Richard D. Averitt, Xin Zhang

Keywords: terahertz, metamaterials

Abstract: Compared with the traditional electric and magnetic multipoles, the existence of a dynamic toroidal moment has received increasing interest in recent years. This is due to its novel electromagnetic response, including dynamic non-radiating charge-current configurations and non-reciprocal interactions. Reconfigurable terahertz metamaterials where artificial toroidal metamolecules and traditional microelectromechanical systems bi-material cantilever structures are integrated within the same unit cell are presented. Through modification of the bending angle by thermal actuation, the toroidal dipole intensity increases by five orders of magnitude in the out-of-plane direction with an overall increase in the toroidal intensity of nearly an order of magnitude. Terahertz time-domain spectroscopy is used to determine the evolution of the transmission as a function of the bending angle.

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Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Nanophotonics
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Volume: 11 **Issue:** 17 **First Page #:** 3769
Date Submitted: 8/23/22 12:00AM **Date Published:** 8/17/22 2:00PM
Publication Location:

Article Title: Active and tunable nanophotonic metamaterials

Authors: Kebin Fan, Richard D. Averitt, Willie J. Padilla

Keywords: metamaterials, subwavelength resonators

Abstract: Metamaterials enable subwavelength tailoring of light-matter interactions, driving fundamental discoveries which fuel novel applications in areas ranging from compressed sensing to quantum engineering. Importantly, the metallic and dielectric resonators from which static metamaterials are comprised present an open architecture amenable to materials integration. Thus, incorporating responsive materials such as semiconductors, liquid crystals, phase-change materials, or quantum materials (e.g., superconductors, 2D materials, etc.) imbue metamaterials with dynamic properties, facilitating the development of active and tunable devices harboring enhanced or even entirely novel electromagnetic functionality. Ultimately, active control derives from the ability to craft the local electromagnetic fields; accomplished using a host of external stimuli to modify the electronic or optical properties of the responsive materials embedded into the active regions of the subwavelength resonators.

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Volume: 105

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Date Submitted: 8/23/22 12:00AM

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Publication Location:

Article Title: Ultrafast optical excitation of magnetic dynamics in van der Waals magnets: Coherent magnons and BKT dynamics in

Authors: Urban F. P. Seifert, Mengxing Ye, Leon Balents

Keywords: correlated electrons, van der Waals magnets, ultrafast

Abstract: Optical pump-probe experiments carried out in the time domain reveal both the intrinsic low-energy dynamics and its connections to higher-energy excitations in correlated electron systems. In this work, we propose two microscopic mechanisms for the optical generation of coherent magnetic modes in van der Waals magnets, and derive the corresponding effective light-spin interactions: either through pumping atomic orbital excitations resonantly or via a light-induced Floquet spin Hamiltonian, the ground state of the system is driven out of equilibrium. The subsequent long-time relaxational dynamics can then be probed using, e.g., the magneto-optical Kerr effect or transient grating spectroscopy. As an example, we apply our framework to NiPS₃, which is magnetically ordered in the bulk, and is conjectured to realize the XY model in the monolayer limit.

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Journal: arxiv

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Volume:

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Date Submitted: 8/23/22 12:00AM

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Article Title: Generating coherent phonon waves in narrow-band materials: a twisted bilayer graphene phaser

Authors: Iliya Esin, Ilya Esterlis, Eugene Demler, Gil Refael

Keywords: graphene, phonon, twistronics

Abstract: Twisted bilayer graphene (TBG) exhibits extremely low Fermi velocities for electrons, with the speed of sound surpassing the Fermi velocity. This regime enables the use of TBG for amplifying vibrational waves of the lattice through stimulated emission, following the same principles of operation of free-electron lasers. Our work proposes a lasing mechanism relying on the slow-electron bands to produce a coherent beam of acoustic phonons. We propose a device based on undulated electrons in TBG, which we dub the phaser. The device generates phonon beams in a terahertz (THz) frequency range, which can then be used to produce THz electromagnetic radiation. The ability to generate coherent phonons in solids breaks new ground in controlling quantum memories, probing quantum states, realizing non-equilibrium phases of matter, and designing new types of THz optical devices.

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Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevResearch.4.043060
Volume: **Issue:** **First Page #:**
Date Submitted: 1/24/23 12:00AM **Date Published:** 2/15/22 8:00AM
Publication Location:

Article Title: Topological frequency conversion in Weyl semimetals

Authors: Frederik Nathan, Ivar Martin, Gil Refael

Keywords: Weyl semimetals, nonlinear optics

Abstract: Doped charge carriers in an antiferromagnetic (AFM) Mott insulator are unstable to spin fluctuation-mediated Cooper pairing as well as various unconventional types of charge, spin, and orbital current ordering. Whether photodoped carriers, in the form of empty (holons) and doubly occupied (doublons) sites propagating in an AFM background, can also exhibit interesting charge instabilities remains an open question. There is numerical and spectroscopic evidence that AFM correlations enhance attractive holon-doublon (HD) interactions, suggesting that a conducting HD plasma might spontaneously self-organize into an insulating fluid of neutral bound pairs, dubbed Hubbard excitons (HEs). However, this out-of-equilibrium phase transition has not been detected experimentally. Here, we report the transient stabilization of a HE fluid in the AFM Mott insulator Sr₂IrO₄ using ultrafast terahertz conductivity.

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Journal: Applied Physics Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1063/5.0067681
Volume: 119 **Issue:** 16 **First Page #:** 162601
Date Submitted: 8/23/22 12:00AM **Date Published:** 10/18/21 2:00PM
Publication Location:

Article Title: Reducing surface depletion of superconducting SrTiO₃ films with EuTiO₃ capping layers

Authors: Hanbyeol Jeong, Nicholas G. Combs, Simon Munyan, Arman Rashidi, Susanne Stemmer

Keywords: superconductivity, ferroelectrics

Abstract: Doped SrTiO₃ thin films are of significant scientific interest for their superconducting, ferroelectric, and thermoelectric properties. Air-exposed thin films of doped SrTiO₃ suffer from surface carrier depletion that makes it difficult to obtain thin, conducting films, especially at low doping densities and temperatures. Here, we show that thin (10 nm) EuTiO₃ capping layers can prevent surface depletion and allow for conducting heterostructures with SrTiO₃ films as thin as 10 nm. We also investigate the superconducting properties of EuTiO₃ capped SrTiO₃ films. We show that the superconducting transition temperature decreases systematically as the SrTiO₃ film thickness is reduced, which is likely due to proximity effects.

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Journal: Applied Physics Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1063/5.0095172
Volume: 121 **Issue:** 1 **First Page #:** 012601
Date Submitted: 8/23/22 12:00AM **Date Published:** 7/7/22 7:00AM
Publication Location:

Article Title: Similarity in the critical thicknesses for superconductivity and ferroelectricity in strained SrTiO

Authors: Hanbyeol Jeong, Ryan Russell, Nicholas G. Combs, Tyler N. Pardue, John W. Harter, Susanne Stemmer

Keywords: superconductivity, ferroelectrics

Abstract: The possible connection between superconductivity and polar order in SrTiO₃ has been discussed extensively in the recent literature. Here, the thickness dependence of the superconducting and ferroelectric transitions in strained, epitaxial films of SrTiO₃ films is studied. Both superconductivity and ferroelectricity are absent in thinner films (25 nm and below). We discuss the possible origins of the thickness dependence of both phenomena.

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Article Title: Role of locally polar regions in the superconductivity of

Authors: Salva Salmani-Rezaie, Hanbyeol Jeong, Ryan Russell, John W. Harter, Susanne Stemmer

Keywords: superconductivity, ferroelectrics

Abstract: Understanding the interaction between polar and superconducting order parameters may hold the key to several classes of superconductors that remain poorly understood, including SrTiO₃ and several tellurides. Here we show that doped, strained SrTiO₃ films can exhibit both global or local polar order, respectively, depending on the amount of epitaxial mismatch strain, thereby providing a platform to understand how inversion symmetry breaking affects superconductivity. We find that the superconducting critical temperature correlates with the length scale of polar order. In particular, the transition temperature is enhanced when polar nanodomains are sufficiently large or, in the extreme limit, films are globally ferroelectric. In these cases, the Cooper pairs reside in a noncentrosymmetric environment. Conversely, low transition temperatures are found when the nanodomains are small.

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Volume: 119

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Publication Location:

Article Title: Molecular beam epitaxy of phase-pure antiperovskite Sr

Authors: Wangzhou Wu, Nicholas G. Combs, Susanne Stemmer

Keywords: antiperovskite, molecular beam epitaxy, topological materials

Abstract: The antiperovskite oxide Sr₃SnO has attracted substantial interest due to its topologically non-trivial band structure. Sr-deficient Sr_{3-x}SnO can become superconducting, making it a candidate intrinsic topological superconductor. Here, we show that epitaxial, phase-pure Sr_{3-x}SnO films can be synthesized by molecular beam epitaxy (MBE) using solid Sr and SnO₂ sources. We show that Sn-rich growth conditions result in a large amount of a Sn-rich impurity phase, which is challenging to detect in x-ray diffraction. Carrier densities and the amount of the impurity phase change systematically with the growth conditions, indicating that MBE provides excellent control over the films' stoichiometry. We discuss the electrical properties, including quantum interference phenomena, which support the topological nature of the films.

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Volume: 18 **Issue:** 5 **First Page #:**
Date Submitted: 1/24/23 12:00AM **Date Published:** 11/1/22 7:00AM
Publication Location:

Article Title: Complementary Vanadium Dioxide Metamaterial with Enhanced Modulation Amplitude at Terahertz Frequencies

Authors: Yuwei Huang, Xuefei Wu, Jacob Schalch, Guangwu Duan, Chunxu Chen, Xiaoguang Zhao, Kelson Kaj,

Keywords: Metamaterials, permittivity, photonics

Abstract: One route to create tunable metamaterials is through integration with “on-demand” dynamic quantum materials, such as vanadium dioxide (VO₂). This enables modalities to create high-performance devices for historically challenging applications. Indeed, dynamic materials have often been integrated with metamaterials to imbue artificial structures with some degree of tunability. Conversely, metamaterials can be used to enhance and extend the natural tuning range of dynamic materials. Utilizing a complementary split-ring resonator array deposited on a VO₂ film, we demonstrate enhanced terahertz transmission modulation upon traversing the insulator-to-metal transition (IMT) at approximately 340 K. Our complementary metamaterial increases the modulation amplitude of the original VO₂ film from 0.42 to 0.68 at 0.47 THz upon crossing the IMT, corresponding to an enhancement of 62%. Our results highlight that symbiotic integration of metamaterial arrays with quantum materials provides a powerful

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Journal: Nature Physics
Publication Identifier Type: **Publication Identifier:**
Volume: **Issue:** **First Page #:**
Date Submitted: 1/24/23 12:00AM **Date Published:**
Publication Location:

Article Title: A Hubbard exciton fluid in a photo-doped antiferromagnetic Mott insulator

Authors: Omar Mehio, Xinwei Li, Honglie Ning, Zala Lenarcic, Yuchen Han, Michael Buchhold, Zach Porter, Nich

Keywords: Excitons, Mott insulators, Antiferromagnetism

Abstract: Doped charge carriers in an antiferromagnetic (AFM) Mott insulator are unstable to spin fluctuation-mediated Cooper pairing as well as various unconventional types of charge, spin, and orbital current ordering. Whether photodoped carriers, in the form of empty (holons) and doubly occupied (doublons) sites propagating in an AFM background, can also exhibit interesting charge instabilities remains an open question. There is numerical and spectroscopic evidence that AFM correlations enhance attractive holon-doublon (HD) interactions, suggesting that a conducting HD plasma might spontaneously self-organize into an insulating fluid of neutral bound pairs, dubbed Hubbard excitons (HEs). However, this out-of-equilibrium phase transition has not been detected experimentally. Here, we report the transient stabilization of a HE fluid in the AFM Mott insulator Sr₂IrO₄ using ultrafast terahertz conductivity.

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Partners

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I certify that the information in the report is complete and accurate:

Signature: David Hsieh

Signature Date: 1/29/23 12:38AM

Summary of Accomplishments (Jul. 1 2016 – Oct. 31 2022)

In this final progress report, we highlight significant theoretical and experimental advancements made during the entire performance period of the award. Although we organize our presentation into four major thrusts, the boundaries between thrusts are highly porous. Many theoretical ideas, experimental approaches and materials were shared between the thrusts. Often, results from one thrust seeded new ideas and breakthroughs in other thrusts. Therefore, these accomplishments reflect a single underlying synergistic effort.

Thrust 1. Floquet Engineering

One of our major goals at the outset of the project was to study the possibility of coherently tuning the hopping (t) and magnetic exchange (J) in correlated magnetic insulators by intensely driving them at optical frequencies tuned below their bandgap to avoid heating – the vision being to eventually use this approach to induce magnetic phase transitions. The **Balents** group focused on the problem of realistic modeling of dynamical tuning of t and J in a Mott insulator in the intermediate frequency range $\Delta > \hbar\omega > W$ where Δ is the Mott gap and W is the bandwidth.

They developed a many-body approach based on large Hubbard interactions U to address this situation, which gives explicit formulae for spin and orbital exchange under optical illumination, for a multi-orbital Hubbard model with full Kanamori interactions.

Balents then applied this to the rare-earth titanate family, $RTiO_3$, by carrying out *ab initio* density functional theory calculations for several titanates and then extracting

tight-binding parameters for the t_{2g} bands from these, as well as physically relevant Hubbard U and Hund's coupling J_H , and crystal field splittings that are important in removing t_{2g} orbital degeneracy. Their results [1] showed that the exchange parameters of $RTiO_3$ could in principle be strongly renormalized, and even reversed in sign, via Floquet engineering using realistic driving amplitudes and frequencies (Fig. 1).

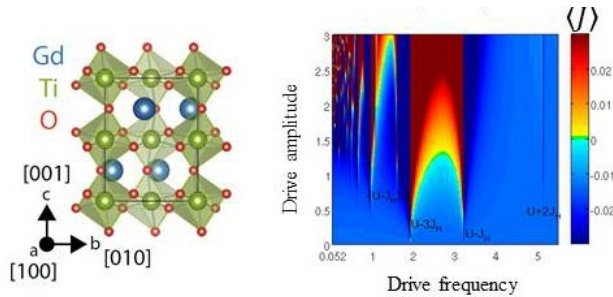


Fig. 1: Calculated in-plane Floquet-modified exchange interaction strengths in $RTiO_3$.

In parallel, **Refael** and **Hsieh** theoretically investigated the prospects for using periodic driving to coherently modify ligand-mediated superexchange interactions in ABX_3 type van der Waals magnets. Considering realistic models and the effects of a periodic drive on different ligand-mediated exchange pathways (Fig. 2), **Refael** and **Hsieh** showed that depending on bond angles and the number of ligand ions involved in the exchange process, drive-induced changes can be

very different from those calculated from direct-hopping models considered earlier. For ABX_3 materials, the ratio of nearest, next-nearest and next-next-nearest neighbor superexchange play a crucial role in determining the magnetic ground state of the system. **Refael** and **Hsieh** showed that this ratio can be controlled by changing the frequency, polarization or intensity of the driving laser field [2]. By sweeping these parameters in their calculations, they then determined the necessary experimental parameter values needed to impart magnetic phase transitions for $MnPS_3$. In addition to direct light modification of hopping, **Refael** and **Hsieh** also presented a new scheme to control spin exchange between two transition metal ions that makes use of a periodic drive to induce an AC Stark shift of the ligand orbital energies that mediate the superexchange [3].

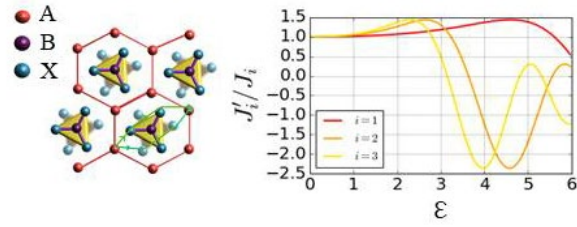


Fig. 2: Left: crystal structure of ABX_3 highlighting different superexchange pathways. Right: renormalization of first, second and third nearest-neighbor exchange strength in ABX_3 as a function of the Floquet parameter ϵ , for circular polarization and a sub-gap driving frequency.

To test these theoretical predictions, **Hsieh** first demonstrated the use of rotational anisotropy second harmonic generation (RA-SHG) as a means to directly sense the antiferromagnetic (AFM) order parameter in $MnPS_3$ single crystals with light (Fig. 3). Together with **Young**, who fabricated clean exfoliated few-layer $MnPS_3$ devices on pure SiO_2 substrates, they showed the bulk magneto-electric type AFM order parameter persists down to 7-8 monolayer thickness [4].

Balents and **Hsieh** followed up the preceding work by developing a single-ion model of the AFM insulator $MnPS_3$ [Fig. 4(a)] to explain the large SHG intensity observed below its AFM ordering temperature of 78 K. Then they calculated the out-of-equilibrium band gap and RA-SHG response of $MnPS_3$ under strong periodic driving conditions over a range of photon energies. They demonstrated that by driving $MnPS_3$ using a photon energy below both the charge transfer gap (~ 3 eV) and $d-d$ transition (~ 2 eV), the bandgap can be coherently enhanced by over 200 meV and that the SHG signal can be suppressed by greater than 90 % for a drive electric field of 10^9 V/m. To test these predictions, **Hsieh** performed pump-wavelength dependent time-resolved RA-SHG and time-resolved SHG spectroscopy measurements on $MnPS_3$ single crystals. An ultrafast coherent suppression of the SHG intensity was demonstrated to be in quantitative agreement with theoretical predictions [Fig. 4(b)]. In contrast, the SHG dynamics following pumping on resonance with the $d-d$ transition shows a slow recovery typical of heating [Fig.

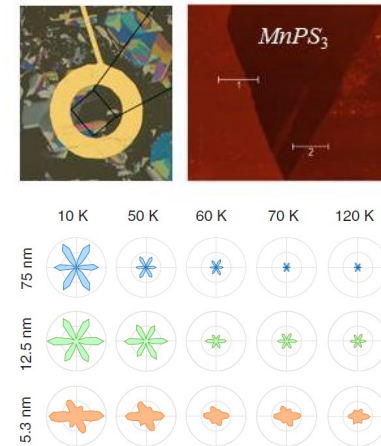


Fig. 3: Top: Optical and atomic force microscope images of $MnPS_3$ nanoflakes. Bottom: RA-SHG results on $MnPS_3$ nanoflakes versus temperature.

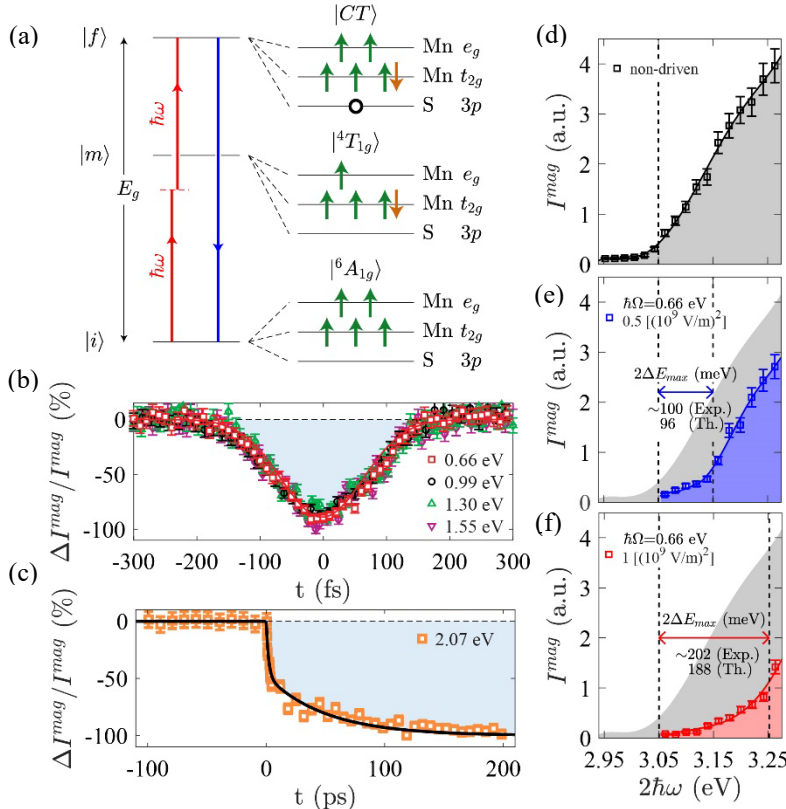


Fig. 4: (a) Left: Black horizontal lines denote the initial, intermediate, and final multi-electron states involved in the SHG process in MnPS₃ (red and blue arrows). Right: Orbital and spin configurations of the states. (b) SHG transients for different sub-gap pump photon energies for a fixed pump amplitude of 10^9 V/m. The overlaid red curve shows the theoretically predicted SHG response for 0.66 eV drive convolved with the probe pulse profile. (c) SHG transient upon pumping on-resonance with the $d-d$ transition. Black curve is a guide to the eye. (d) Non-driven SHG spectroscopy curve. (e) and (f), Transient $t = 0$ SHG spectra at two different pump amplitudes. Solid curves in (d) – (f) are guides to the eye. Vertical dashed lines mark the intensity upturns at the band edge. Values of the observed and theoretically calculated energy shifts are indicated.

4(c)]. Transient SHG spectroscopy data confirm that the band edge, marked by the intensity onset in the SHG spectrum [Fig. 4(d) – (f)], shifts to higher energy with increasing drive amplitude, in quantitative agreement with theory. Remarkably, this is opposite to typical ultrafast experiments where electronic gaps are suppressed upon photo-excitation. Overall, this work [5] demonstrated sub-gap driving as a viable route to Floquet engineering van der Waals magnets.

Averitt explored routes to dynamically engineer magnetic exchange interactions in GdTlO₃ (GTO) inspired by **Balents'** theoretical work. Briefly, GTO has an orthorhombic perovskite-type unit cell and relatively large GdFeO₃-type distortion. What we determined makes GTO particularly interesting is that it lies very close to a ferromagnetic-to-antiferromagnetic (FM-AFM) phase boundary, just on the FM side. This makes GTO magnetism particularly sensitive to structural changes. Below the critical temperature $T_c = 32$ K it is ferrimagnetically (fM) ordered, with the Ti³⁺ spins aligned ferromagnetically along the c -axis, coupled antiferromagnetically to the Gd sublattice. The magnetism saturates at $6 \mu_B$ ($7 \mu_B$ Gd – $1 \mu_B$ Ti) in a relatively small field of ~ 0.1 T, with no discernable hysteresis.

By performing time-resolved magneto-optical Kerr effect (MOKE) measurements on high quality GTO thin films and bulk crystals synthesized by **Stemmer** and **Wilson** respectively, **Averitt** observed distinct magnetic field dependent dynamics in GTO. At low fields (~ 0.1 T), following photoexcitation, there is an overall initial increase in the magnetization due to a

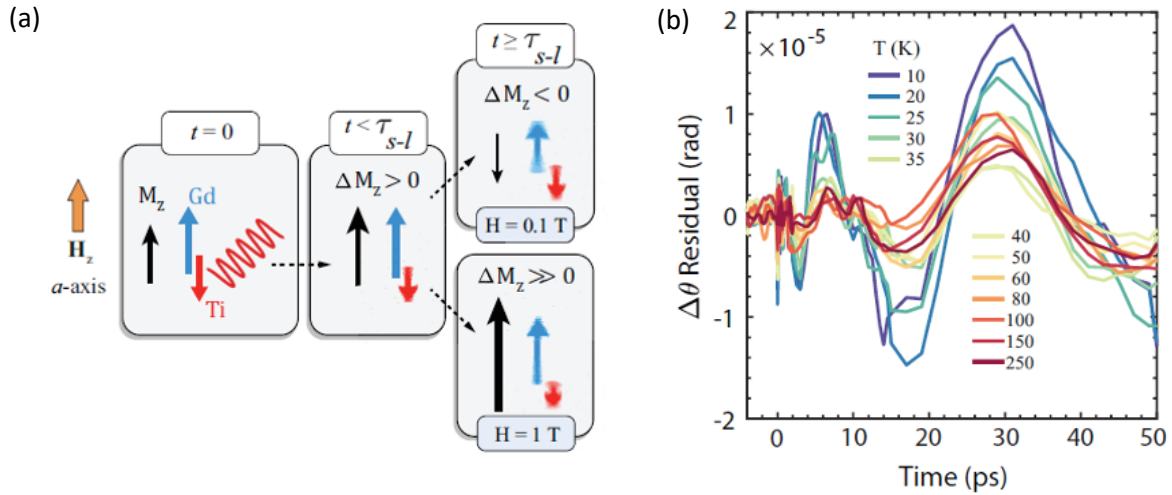


Fig. 5: (a) Schematic depiction of the spin dynamics in GTO. Photoexcitation directly perturbs the Ti spins, which fluctuate and decrease their projection along the applied field H (parallel to a -axis, $+z$) in $t < \sim 100$ ps, increasing the Kerr signal. At longer times: if H and/or magnetic order is weak, induced spin fluctuations and the AFM exchange coupling lowers the projection of Gd spins along z . Conversely, if H is larger than the exchange field, at 1 T, Gd does not reorient and no negative component of the signal is observed. (b) GTO coherent acoustic phonon response, isolated by subtracting the exponential fits from the time-resolved Kerr data (at 1 T applied field). The appearance of the oscillations in the Kerr response indicates coherent acoustic phonon manipulation of the magnetic order, presumably from exchange modulation.

decrease in the magnetic order of the Ti sublattice, crossing over to an eventual decrease in the magnetization on a longer timescale (~ 100 ps) where spin-lattice relaxation results in a decrease in the Gd sublattice magnetization. However, for higher fields (~ 1 T), only an increase in the magnetization is observed, resulting from the perturbation of the Ti sublattice, with the Gd spins locked into place. Figure 5(a) presents a summary of the dynamics. Surprisingly, in both the photoinduced reflectivity dynamics and transient Kerr rotation [Fig. 5(b)], coherent acoustic phonon oscillations are observed, with a large increase in the amplitude below T_c , revealing strong magneto-elastic coupling. This data is consistent with the interpretation of an acoustic strain wave with strong magneto-elastic coupling. This mechanism involves elastic stress modifying the magnetic anisotropy, which exerts a torque on the spins and alters the net magnetization. This, in turn, implies transient exchange modification on an ultrafast timescale. Therefore, this work represents a method of using light to indirectly alter the magnetic degrees of freedom on ultrafast timescales, through coupling to an acoustic phonon mode [6].

Thrust 2. Coherent Mode Control

Transfer of energy from pump photons into coherent structural and coherent magnetic modes of a material offers a potential route for dynamically controlling quantum materials. To this end, **Hsieh** and **Averitt** found that coherent phonons can be impulsively stimulated in bulk crystals of the van der Waals ferromagnet CrSiTe_3 (grown by **Wilson**) both above and below the Curie

temperature ($T_c \sim 31$ K). What is particularly interesting however is the fact that when the pump photon energy is tuned on-resonance with the Te-to-Cr charge transfer energy, the phase of the A_{1g} phonon oscillation flips by 180° across a characteristic temperature of around 100 K (Fig. 6) [7]. On the other hand, for off-resonant pumping, no change in the phonon

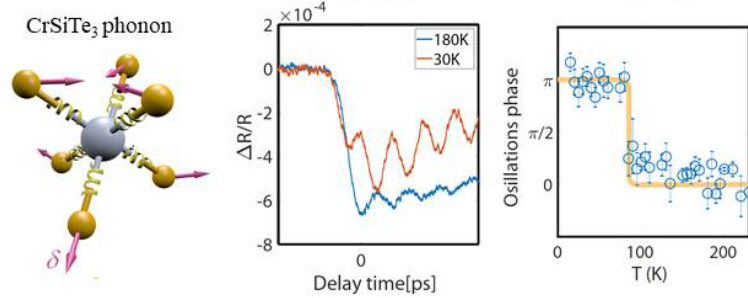


Fig. 6: Coherent A_{1g} phonon oscillations in CrSiTe_3 excited on resonance with the charge transfer transition. A clear flip of phase is evident across ~ 100 K.

oscillation phase is observed across all temperatures. To understand the significance of this 100 K temperature scale, **Hsieh** and **Balents** used RA-SHG studies combined with classical Heisenberg model calculations to show that the rise of in-plane spin correlations in CrSiTe_3 occurs right below 100 K [8]. Using this information, **Refael** developed a theory that explains the complete set of coherent phonon data, which is based on a new pathway for optically modifying the ligand mediated super-exchange interaction between neighboring Cr sites through charge transfer induced Pauli blocking [7]. This work opens a new avenue for ultrafast optical manipulation of magnetic exchange beyond Floquet engineering and has demonstrated coherent phonon spectroscopy as a new method to measure this effect.

Averitt discovered that coherent magnons with B_{2g} symmetry could be impulsively generated by light in the spin-orbit coupled Mott insulator Sr_2IrO_4 . By conducting time-resolved MOKE measurements on single crystals synthesized by **Wilson**, **Averitt** showed that depending on whether the pump photon energy is below or above the Mott gap, the phase of the coherent magnon oscillations either flips or does not flip, respectively, upon reversing the photon helicity [9].

Balents theoretically tackled this problem by studying the non-equilibrium effects of laser excitation on the uniform and staggered magnetization of AFM ordered Sr_2IrO_4 [10]. In particular, **Balents** showed that coherent magnon generation occurs via an inverse Faraday effect, with the likely channel for this process being through direct two-magnon generation where the effective field that initiates the coherent magnons is “beyond Floquet” in the sense that both the incident electric field and the magnon-

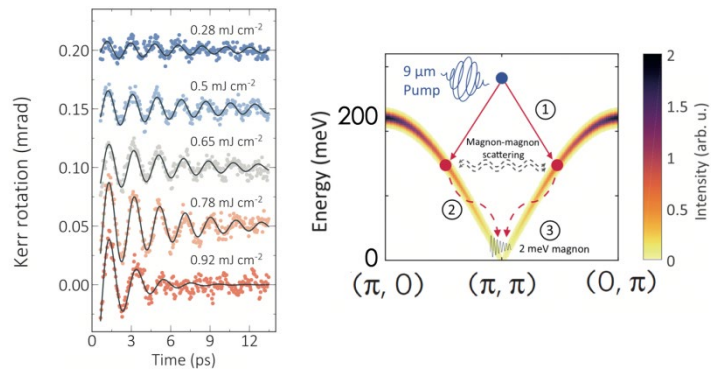


Fig. 7: (Left) Coherent magnons initiated with $9 \mu\text{m}$ circularly polarized pulses (non-resonant with Mott gap). Data taken at 15 K, as a function of fluence, revealing a decrease in the coherence lifetime, presumably related to increased magnon-magnon scattering. (Right) Depiction of the initial 2-magnon process which generated the $q = 0$ coherent magnons.

magnon scattering are required to initiate the observed dynamics (Fig. 7). The response has both resonant and non-resonant components and is predicted to be strongest close to the upper edge of the two-magnon continuum, yielding specific dependencies on pump frequency, polarization and fluence, which were fully verified by **Averitt** using time-resolved MOKE. These results provide a mechanism for mode-selective excitation in the spin channel of magnetically ordered quantum materials.

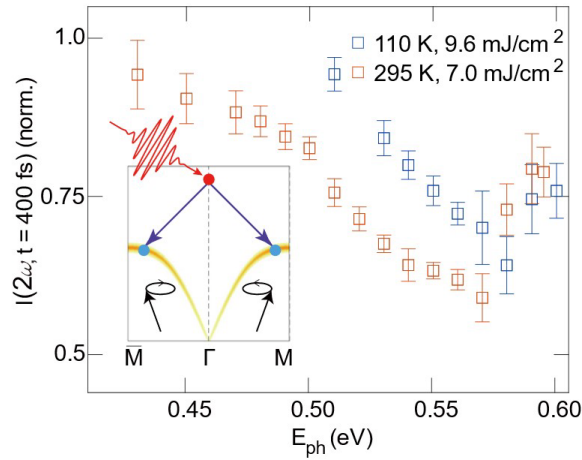


Fig. 8: Pump photon energy dependence of SHG intensity at the fixed pump-probe delay time $t = 400$ fs at 295 K and at 110 K from $\text{Sr}_2\text{Cu}_3\text{O}_4\text{Cl}_2$, whose Néel temperature is 380 K. Inset: Schematic of the two-magnon generation process.

Building on the previous work by **Balents** and **Averitt**, **Hsieh** studied the extreme possibility of using two-magnon pumping to induce magnetic phase transitions. Using time-resolved RA-SHG, **Hsieh** showed that by driving $\text{Sr}_2\text{Cu}_3\text{O}_4\text{Cl}_2$ – a model Mott insulating cuprate – below its band gap (~ 1.8 eV), one can suppress charge excitations even for electric fields up to 10^9 V/m. In this sub-gap pumping regime, **Hsieh** observed a collapse of the AFM order parameter (Fig. 8), which is most efficient when the driving frequency is resonant with twice the Brillouin zone boundary magnon energy (~ 0.57 eV), consistent with two-magnon pumping [11]. This work presents a novel out-of-equilibrium pathway for controlling magnetism through direct magnon bath engineering.

Thrust 3. Photo-doping

Chemically doped Mott insulators exhibit a plethora of interesting electronic instabilities, begging the question of whether photo-doping, realized through above-gap optical excitation, can provide a means to transiently simulate the effects of chemical doping. We were particularly interested in the possibility of simulating the effects of electron doping in Sr_2IrO_4 with light because chemical n -doping has been shown to give rise to both pseudogap and possibly d -wave high- T_c superconducting behavior in this compound. Our approach to optical n -doping was to pump with a photon energy tuned to the $J_{\text{eff}} = 3/2$ to upper Hubbard band transition. To study the consequences of pumping, **Hsieh** used time-resolved RA-SHG to track the ultrafast magnetization dynamics of Sr_2IrO_4 crystals provided by **Wilson**. By coupling these results with time-resolved MOKE experiments from **Averitt**, an out-of-equilibrium phase diagram immediately after pumping was mapped (Fig. 9). This strongly resembles the known equilibrium temperature versus lanthanum (electron) doping phase diagram of Sr_2IrO_4 and suggests that photo-doping instantaneously acts like chemical doping [12]. By studying the recover dynamics of the magnetic order parameter, **Hsieh** also found a critical slowing down behavior upon

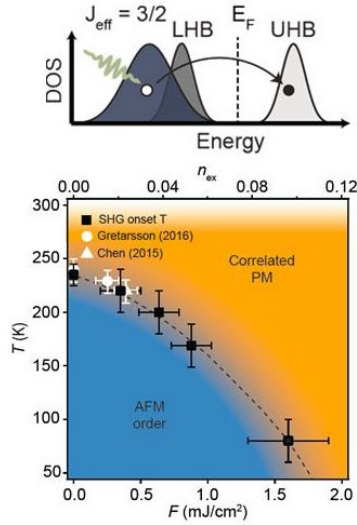


Fig. 9: (Top) Illustration of the n -type photo-doping strategy used in Sr_2IrO_4 . (Bottom) Out-of-equilibrium phase diagram mapped at 600 fs delay between pump and probe pulses.

traversing a phase boundary in the out-of-equilibrium phase diagram. To understand this, **Refael** showed that energy in the magnon subsystem is trapped after pump excitation due to the rapid sub-picosecond recovery of the charge excitations. This means that recovery of the magnetic order occurs primarily through magnon thermalization via magnon-magnon scattering. **Refael** developed a field theoretic model to understand this regime and showed that magnon thermalization indeed undergoes a critical slowing down upon quenching of the antiferromagnetic order [12]. Going beyond collapsing magnetic states by photo-doping, **Refael** demonstrated a mechanism to induce long-range magnetic order starting from a paramagnetic state based on light-induced evaporative cooling of holes. Since the entropy of a filled narrow band grows rapidly with hole doping, the isentropic transfer of holes from a doped Mott insulator to such a band can result in a drop of temperature. Using non-equilibrium dynamical mean field theory, **Refael** showed that suitably designed chirped pulses allow one to realize this cooling effect. In particular,

Refael demonstrated the emergence of AFM order in a system that is initially in a weakly correlated state above the maximum Néel temperature [13]. These works suggest a general strategy for inducing strong correlation phenomena and electronic orders in photo-doped materials.

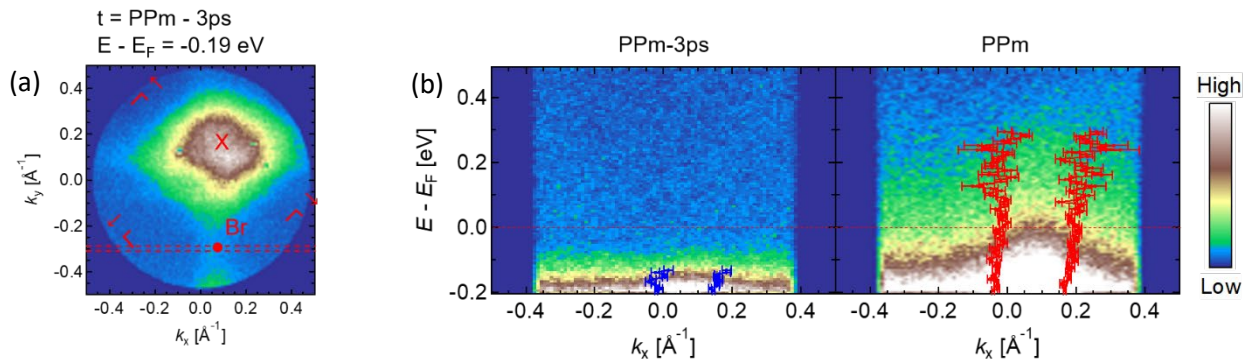


Fig. 10: Electronic band feature of the IMT at the branch point (Br). (a) Constant energy surface at PPm - 3 ps. Red dotted lines show the slicing location along with the momentum integration range for the E-k cuts in panel (b). (b) E-k cuts at PPm - 3 ps and PPm. Note that the color scale is saturated to clearly show the weak signals at high energies. Overlaid lines are the mean $\pm$ FWHM of Gaussian fits to the MDCs at each energy.

Add-on funding to the MURI supported efforts by **Gedik** to perform time- and angle-resolved photoemission spectroscopy (tr-ARPES) on Sr_2IrO_4 using XUV probe photon energies, allowing a wide region in the Brillouin zone ($\sim 1 \text{ \AA}^{-1}$ in size) to be accessed. The effects of photo-doping were studied by using linearly polarized pump pulses with 1.2 eV photon energy. Upon excitation, **Gedik** found that spectral weight appears within the Mott gap across momentum

space and relaxes back on the picosecond timescale, suggestive of a transient insulator-to-metal transition (IMT) [14]. To investigate the change of spectral features in detail, **Gedik** performed Gaussian fitting on momentum distribution curves (MDC) at different binding energies at the “branch point” in the Brillouin zone [Fig. 10(a)]. As shown in Figure 10(b), lines of the mean momentum ($\pm$ FWHM) extracted from Gaussian fits to MDCs at each energy capture the detailed shape of the electronic band structure. At the time delay where the photo-induced signal is maximum (dubbed “PPm”), the electronic band crosses the Fermi level and extends higher up in energy. This is in clear contrast to the electronic band structure acquired at PPm – 3 ps. In fact, the pillar like band shape looks similar to the one reported from the metallic state at the M point in chemically doped Sr_2IrO_4 . The origin of this photo-doping induced IMT remains an open question.

The development of large Sr_2IrO_4 single crystals from **Wilson** enabled their study using time-resolved time-domain THz spectroscopy (TDTS). Using this technique, **Hsieh** discovered evidence for the formation of an exciton fluid in photo-doped Sr_2IrO_4 . Time-resolved optical conductivity data extracted from the TDTS data, acquired using a 0.6 eV drive that is resonant with the lower-to-upper Hubbard band transition, are shown in Figure 11. The broad exciton mode can clearly be seen evolving as a function of time. By examining the temporal evolution of the exciton mode and its dependence on both temperature and pump fluence, **Hsieh** found evidence that these are Hubbard excitons that are bound via antiferromagnetic exchange interactions, unlike conventional excitons that are bound via Coulomb forces [15]. This interpretation was further corroborated by performing exact diagonalization calculations of an extended Hubbard model appropriate for Sr_2IrO_4 , showing internal Hubbard exciton energy levels consistent with the energy scale observed in TDTS measurements. This work constitutes the first direct observation of a Hubbard exciton fluid in a 2D AFM Mott insulator.

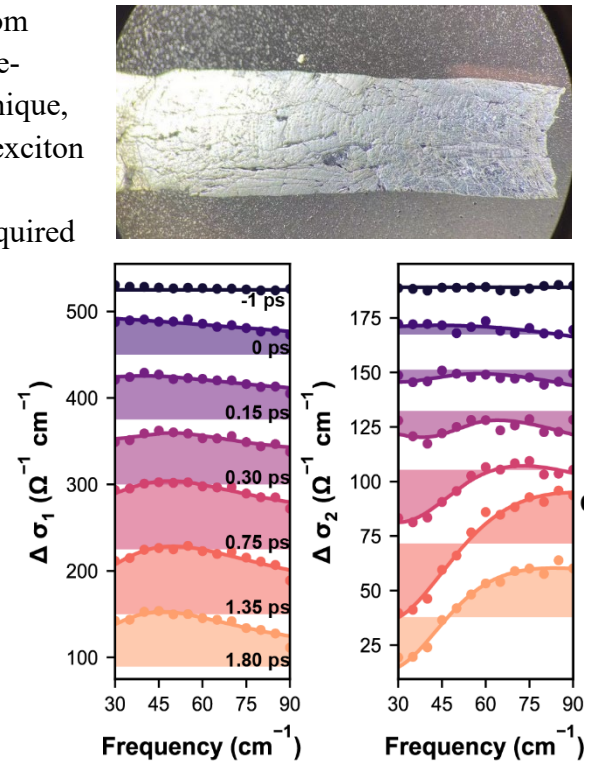


Fig. 11: Top: Image of Sr_2IrO_4 crystal synthesized via the traveling solvent floating zone method. Bottom: The real (left) and imaginary (right) parts of the transient optical conductivity of Sr_2IrO_4 as a function of time. Drude-Lorentz fitting shown as solid lines.

We also studied the effects of photo-doping in ABX_3 titanates, which are known to exhibit highly tunable magnetic and ferroelectric phases upon rare earth doping and epitaxial strain. In equilibrium, **Stemmer** successfully stabilized metallic ferromagnetic films of $\text{Eu}_{0.985}\text{Sm}_{0.015}\text{TiO}_3$ on DyScO_3 substrates. The film remains metallic down to the lowest temperatures measured and

static RA-SHG measurements by **Hsieh** verified that it has a polar point group $mm2$, thus realizing a rare material platform where ferromagnetism, ferroelectric (FE) order and metallicity can all be controlled by the film growth conditions. To understand whether a similarly rich phenomenology occurs away from equilibrium, **Hsieh** performed time-resolved RA-SHG measurements on pure EuTiO_3 grown on DyScO_3 substrates (supplied by **Stemmer**), which is a FE polar insulator. Pump photon energy dependent time-resolved SHG revealed a clear onset of photo-doping as soon as the photon energy exceeds the $\text{Eu } 4f$ to $\text{Ti } 3d$ charge gap (Fig. 12). In the transient photo-doped phase, time-resolved RA-SHG studies by **Hsieh** show that the FE polar structure is retained, indicating that a transient polar metallic phase is realized, which is well known to be difficult to achieve in equilibrium. These results [16] provided evidence for the presence of two mechanisms that contribute to FE order in pure EuTiO_3 grown on DyScO_3 substrates, namely short-range and long-range dipolar interactions. While the long-range interactions were found to be easily screened by photo-carriers excited across the $\text{Eu } 4f$ valence to $\text{Ti } 3d$ conduction bands, short-range interactions turned out to be robust against photo-doping. Therefore, FE order in EuTiO_3 is only slightly diminished even at the highest photo-doping levels achievable. This suggests that strained EuTiO_3 is similar to CaTiO_3 in the sense that the FE order is predominantly driven by short-range interactions. More generally, this work establishes time-resolved RA-SHG as a method to identify transient polar metals and to separate the contribution of short- and long-range interactions in FE materials.

Stemmer also showed that doping and magnetic fields result in clear changes in the magneto-transport of EuTiO_3 films grown under biaxial tensile epitaxial stresses. To elucidate the effects on crystalline symmetry, **Stemmer** performed measurements of the anisotropic magnetoresistance (AMR) of doped EuTiO_3 films [17]. They showed that the primary contribution to the AMR is the crystalline component, which depends on the relative orientation between the magnetic moments and the crystal axes. With increasing magnetic field, a four-fold crystalline AMR undergoes a change in its alignment with respect to the crystal axes. The results show that EuTiO_3 exhibits a strong coupling between spin canting (which is influenced by the

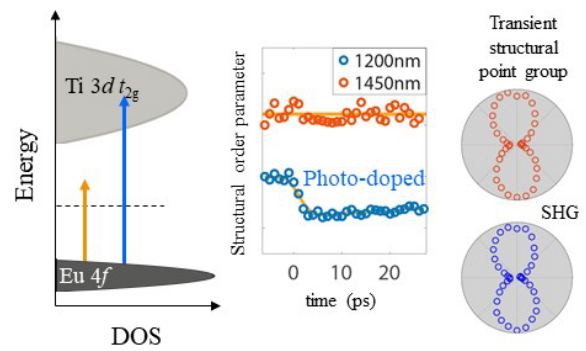


Fig. 12: Time-resolved SHG intensity for pump photon energy below (orange) and above (blue) the charge gap of $\text{EuTiO}_3/\text{DSO}$ showing a clear response only in the latter case. Time-resolved RA SHG data collected after pumping show persistence of the ferroelectric structure.

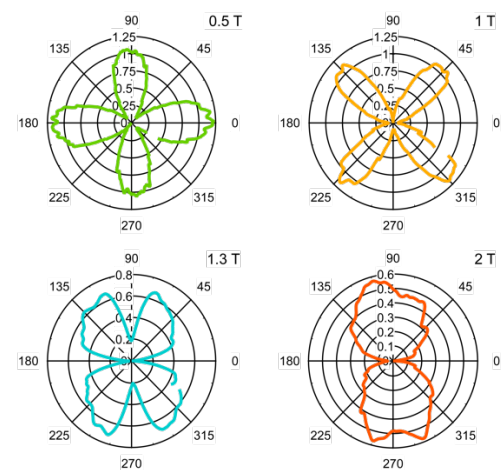


Fig. 13: AMR of doped EuTiO_3 films. Shown is the longitudinal resistance for different in-plane applied magnetic fields (see labels).

magnetic field) and the electronic structure, as evidenced by the transport. This can be attributed to Weyl points in the band structure. At high fields, the AMR transitions to uniaxial symmetry, which is lower than the symmetry of the lattice, along with a cross-over from positive to negative magnetoresistance (Fig. 13). This points to an electronic nematic phase, whose origin has yet to be fully understood. These results show that doped EuTiO_3 is a tunable system for which there is a strong coupling of spin orientation and lattice symmetry, which should be conducive to dynamical control and detection.

Thrust 4. THz control

Two-dimensional van der Waals materials such as graphene provide a platform for highly tunable correlated phases that may be conducive to optical manipulation. We were interested in developing a theoretical understanding of possible dynamical phenomena in graphene-based heterostructures and in developing techniques to interrogate their ultrafast properties on-chip.

Refael theoretically explored optical Floquet engineering in twisted bilayer graphene (TBG). At the magic twist angle, this system features flat energy bands, which leads to superconductivity and strong correlation physics. In equilibrium, these unique properties are typically limited to a narrow range of twist angles around the magic angle with a small tolerance. **Refael** reported on a mechanism that enables flattening of the band-structure using coherent optical illumination, leading to emergence of flat isolated Floquet-Bloch bands. It was shown that the effect can be realized with relatively weak optical beams in the visible-infrared range (below the material bandwidth) and persist for a wide range of small twist angles, increasing the allowed twist tolerance by an order of magnitude [18]. **Refael** also worked out the conditions under which these bands exhibit a non-zero Chern number, opening the way to search for novel strongly correlated, non-equilibrium electronic states of matter in optically induced flat band materials.

Refael also proposed a mechanism by which a strong beam of coherent phonons can be generated in TBG [19]. This system exhibits extremely low Fermi velocities for electrons, with the speed of sound surpassing the Fermi velocity. This regime enables the use of TBG for amplifying vibrational waves of the lattice through stimulated emission, following the same principles of operation as free-electron lasers. **Refael** theoretically demonstrated a lasing mechanism relying on the slow-electron bands to produce a coherent beam of acoustic phonons. Moreover, they proposed a device based on undulated electrons in TBG, dubbed the “phaser” (Fig. 14). The device generates phonon beams in a terahertz (THz) frequency range, which can then be used to produce THz electromagnetic

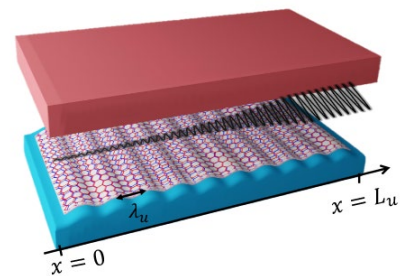


Fig. 14: A layer of TBG is encapsulated between two gates of doped semiconductors. A nano-undulator is realized by a modulated-in-space uniaxial strain. The resonant frequency is controlled by the wavelength of the periodic structure, λ_u .

radiation. The ability to generate coherent phonons in solids breaks new ground in controlling quantum memories, probing quantum states, realizing non-equilibrium phases of matter, and designing new types of THz optical devices.

Since the bandwidth of TBG is on the scale of meV, it is natural to drive and probe them using THz radiation. Because free space THz probing cannot be applied to nanomaterials, **Young** and **Averitt** collaboratively developed picosecond-resolved transport measurements using on-chip THz generation based on Si-on-sapphire Auston switch technology. They demonstrated gate modulated THz transmission through a graphene nano device and succeeded in observing optical pump induced changes in its THz transmission (Fig. 15) [20]. Throughout the course of the award, **Young** continually updated the design of the on-chip THz spectrometer for improved signal-to-noise, bandwidth, sample throughput and ease of sourcing parts. The most recent iteration uses a commercial (Protemics) on-chip THz device with a motherboard (MB) – daughterboard (DB) architecture, allowing for quick THz spectroscopy measurements of the samples fabricated on the DB while keeping the photoconductive (PC) switches on the MB unchanged [Fig. 16(a)]. This device architecture can measure the reflected and transmitted THz pulse. The MB is fabricated on an ultrathin cyclic olefin polymer (COP) to minimize radiation loss. However, the COP substrate is not suitable for integration with van der Waals device fabrication methods due to its low glass transition temperature ($\sim 150^\circ\text{C}$). Instead, **Young** developed lithographic techniques to fabricate devices on an ultrathin (50 μm) quartz substrate, which has excellent thermal stability and THz loss properties.

A thin film of a superconductor (NbN) was sputtered on the quartz DB along with transmission lines and aligned to the MB. **Young** measured the THz reflection at room temperature as shown in Figure 16(b), showing a bandwidth of about 0.7 THz. Three main features were observed in the reflection: (i) incident THz pulse, (ii) partial reflection at the MB-DB interface caused by the impedance mismatch due to the change in dielectric environment, and (iii) reflected THz pulse from the NbN sample. The high magnitude of the THz pulse reflected from the sample (40% of the incident THz pulse) indicates a good coupling of the transmission lines at the MB-DB interface and at the sample. Owing to the low thermal conductivity of polymers, the THz board cannot be efficiently cooled down by conduction. Therefore, **Young** used an exchange gas cryostat to cool down to cryogenic temperatures. Small thermal drift can misalign the laser spot

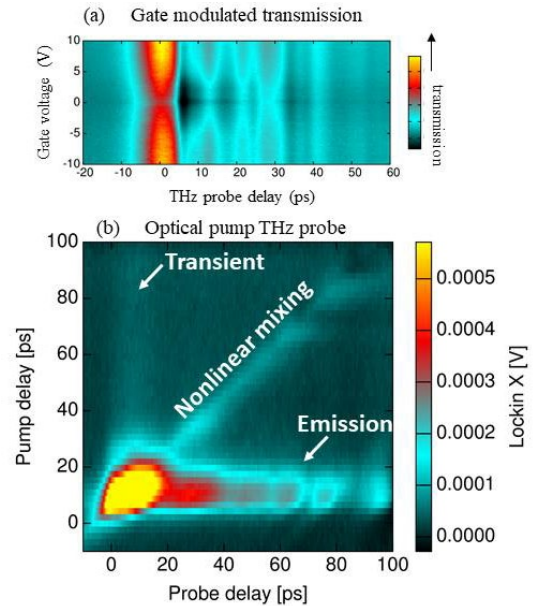


Fig. 15: (a) Transmission of a THz transient through graphene modulated by a gate. (b) Optical pump induced change in THz transmission observed as a nonlinear mixing channel.

on the PC switches and cause unwanted phase shifts. Thus, every THz transient was measured carefully after auto-aligning the laser on the emitter (E) and the receiver (A) switch. The THz reflection from a test NbN sample was measured while varying the temperature across the superconducting transition ($T_C \sim 10.6$ K) as shown in Figure 16(c). The THz transients remain the same in the normal state but show a robust ~ 100 fs phase shift in the superconducting state across multiple cooldowns. This abrupt phase shift is indicative of a transition from a resistive (normal) to an inductive (superconducting) response. Work continues to optimize the system and develop protocols to extract the complex optical conductivity from the time-domain data. But these efforts have brought reliable on-chip THz spectroscopy of 2D materials much closer to reality.

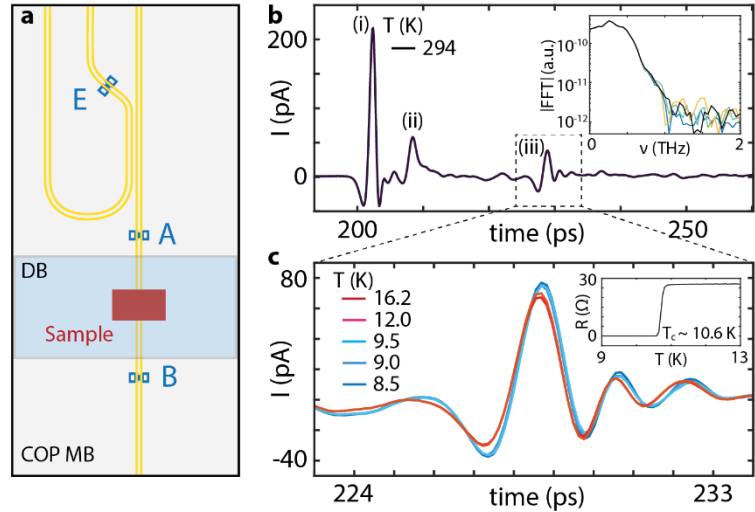


Fig. 16: On-chip THz time domain spectroscopy (THz-TDS) of a superconducting thin film. (a) Illustration of an on-chip THz measurement setup. THz pulses are emitted by photoconductive (PC) switch E. The reflected and the transmitted THz pulse is measured at PC switch A and B, respectively. (b) Time domain THz reflection of thin film NbN measured at room temperature, showing the (i) incident pulse, (ii) reflected pulse from the MB-DB interface, and (iii) reflected pulse from the sample. Inset, Fourier analysis of the reflected THz signal for multiple sweeps yielding a bandwidth of 0.7 THz. (c) Time domain THz reflection of NbN above and below the superconducting transition (inset, $T_c \sim 10.6$ K) showing a clear phase shift.

As part of the effort to prepare suitable van der Waals materials for study with the on-chip THz technology, **Young** successfully developed interlayer proximity effects for engineering new ground states in graphene that may be amenable to dynamical engineering [21]. Of particular note is the successful effort to induce Kane-Mele spin-orbit coupling in graphene using proximal layers of transition metal dichalcogenides. Using ultra-clean graphene heterostructures composed of graphite gates and high quality hBN and WSe₂ dielectrics [Fig. 17(a)], they observed reconstruction of the bilayer graphene band structure arising from spin-orbit coupling which itself emerges from the proximity of the graphene to the strong spin-orbit semiconductor. Two effects are of note: first, **Young** observed spin splitting of valence and conduction bands [Fig. 17(b)]. Second, they found a new phase at charge neutrality when no electric field is applied to

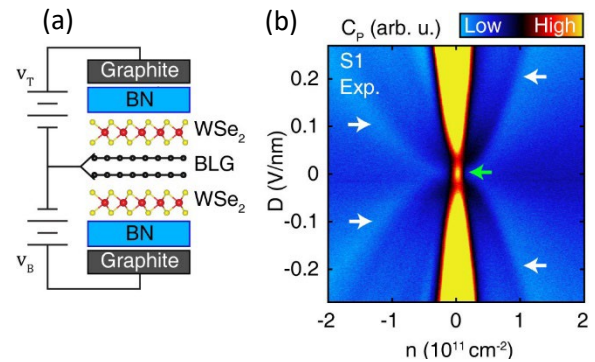


Fig. 17: (a) Device schematic for realizing spin-orbit proximity effect in graphene. (b) Penetration field capacitance, sensitive to inverse density of states. White arrows indicate features associated with spin splitting of bands; green arrow shows new topological phase.

the graphene bilayer. Additional analysis indicated that this phase is protected by fragile topology, and hosts spin symmetry protected helical edge states. It is expected that control of spin-orbit coupling in 2D layered materials at the meV scale will provide a useful platform for THz drive induced non-equilibrium phases.

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