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RPPR Final Report
as of 19-Apr-2021

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DUNS Number: 001425594 EIN: 042103594
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Final Report for Period Beginning 01-May-2015 and Ending 31-Dec-2020
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Begin Performance Period: 01-May-2015 **End Performance Period:** 31-Dec-2020
Report Term: 0-Other
Submitted By: Nuh Gedik Email: gedik@mit.edu
Phone: (617) 253-3420
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STEM Degrees: **STEM Participants:**

Major Goals: In this project, we investigated controlling the properties of materials by simply shining light onto them. This is based on the fact that, if the energy of the incident light is below the absorption threshold, one will not have direct absorption making it possible to observe coherent phenomenon. In this case, electrons and photons can coherently hybridize forming so-called Floquet-Bloch states. Our goal is to use these Floquet-Bloch states to engineer materials with distinct properties. By tuning the properties of the incident light (such as fluence, frequency or polarization), one can change the resultant material properties.

Our goal was to establish Floquet-Bloch states in systems other than topological insulators and study their formation and decay dynamics in these different material systems. The plan was then to investigate using Floquet-Bloch states to both engineer new phases of matter and to change the properties of existing materials. Examples are light induced quantum anomalous phase in graphene and optical modification of the exchange interaction in Mott insulators using the Floquet-Bloch states.

Accomplishments: During the grant period, we have made significant advances in multiple directions. First, we have greatly improved capabilities of time and angle resolved photoemission spectroscopy (tr-ARPES). We can now reliably perform tr-ARPES using 11 eV, 18, 26 or 31 eV at a very high repetition rate of 300 kHz. Our energy resolution is also one of the best in the world (16 meV at 11 eV and 30meV at 30 eV). These development are absolutely key in detecting the electronic band dispersion of light induced exotic phases. Secondly, we achieved distinguishing Floquet-Bloch (FB) and Volkov states in a systematic manner in Bi2Se3. This will enable selective study of FB states in tr-ARPES in different systems. Finally, we used our advanced techniques to investigate the properties of light induced novel phases and excitations in different systems such as charge density waves or excitonic insulator candidates. I will detail our achievements in these three directions by referencing the papers we published.

1. Development of state of the art tr-ARPES based on high harmonic generation

a. Time-resolved XUV ARPES with tunable 24–33 eV laser pulses at 30 meV resolution (Nat Commun 10, 3535 (2019).)

High harmonic generation of ultrafast laser pulses can be used to perform angle-resolved photoemission spectroscopy (ARPES) to map the electronic band structure of materials with femtosecond time resolution. However, currently it is difficult to reach high momenta with narrow energy resolution. Here, we combine a gas phase extreme ultraviolet (XUV) femtosecond light source, an XUV monochromator, and a time-of-flight electron analyzer to develop XUV-based time-resolved ARPES. Our technique can produce tunable photon energy between 24–33 eV with an unprecedented energy resolution of 30 meV and time resolution of 200 fs. This technique enables time-, energy- and momentum-resolved investigation of the nonequilibrium dynamics of electrons in materials with a full access to their first Brillouin zone. We evaluate the performance of this setup through exemplary measurements on various quantum materials, including WTe_2, WSe_2, $TiSe_2$, and $Bi_2Sr_2CaCu_2O_{8+\delta}$.

b. High resolution time- and angle-resolved photoemission spectroscopy with 11 eV laser pulses (Review of Scientific Instruments 91, 043102 (2020))

Performing time- and angle-resolved photoemission (tr-ARPES) spectroscopy at high momenta necessitates extreme ultraviolet laser pulses, which are typically produced via high harmonic generation (HHG). Despite recent advances, HHG-based setups still require large pulse energies (from hundreds of μJ to mJ) and their energy resolution is limited to tens of meV. Here, we present a novel 11 eV tr-ARPES setup that generates a flux of 5×10^{10} photons/s and achieves an unprecedented energy resolution of 16 meV. It can be operated at high repetition rates (up to 250 kHz) while using input pulse energies down to 3 μJ. We demonstrate these unique capabilities by simultaneously capturing the energy and momentum resolved dynamics in two well-separated momentum space regions of a charge density wave material $ErTe_3$. This novel setup offers the opportunity to study the non-equilibrium band structure of solids with exceptional energy and time resolutions at high repetition rates.

2. Developing a methodology to sperate Floquet-Bloch and Volkov States

Selective scattering between Floquet–Bloch and Volkov states in a topological insulator
(Nature Physics 12, 306–310 (2016))

The coherent optical manipulation of solids is emerging as a promising way to engineer novel quantum states of matter. The strong time-periodic potential of intense laser light can be used to generate hybrid photon–electron states. Interaction of light with Bloch states leads to Floquet–Bloch states, which are essential in realizing new photo-induced quantum phases. Similarly, dressing of free-electron states near the surface of a solid generates Volkov states, which are used to study nonlinear optics in atoms and semiconductors9. The interaction of these two dynamic states with each other remains an open experimental problem. Here we use time- and angle-resolved photoemission spectroscopy (Tr-ARPES) to selectively study the transition between these two states on the surface of the topological insulator Bi_2Se_3. We find that the coupling between the two strongly depends on the electron momentum, providing a route to enhance or inhibit it. Moreover, by controlling the light polarization we can negate Volkov states to generate pure Floquet–Bloch states. This work establishes a systematic path for the coherent manipulation of solids via light–matter interaction.

3. Studying light induced novel phases and excitations in different systems

a. The Spontaneous Symmetry Breaking in Ta_2NiSe_5 is Structural in Nature (submitted to PRX (2021))
The excitonic insulator is an electronically-driven phase of matter that emerges upon the spontaneous formation and Bose condensation of excitons. Detecting this exotic order in candidate materials is a subject of paramount importance, as the size of the excitonic gap in the band structure establishes the potential of this collective state for superfluid energy transport. However, the identification of this phase in real solids is hindered by the coexistence of a structural order parameter with the same symmetry as the excitonic order. Only a few materials are currently believed to host a dominant excitonic phase, Ta_2NiSe_5 being the most promising. Here, we test this scenario by using an ultrashort laser pulse to quench the broken-symmetry phase of this transition metal chalcogenide. Tracking the dynamics of the materials electronic and crystal structure after light excitation reveals surprising spectroscopic fingerprints that are only compatible with a primary order parameter of phononic nature. We rationalize our findings through state-of-the-art calculations, confirming that the structural order accounts for most of the gap opening. Not only do our results uncover the long-sought mechanism driving the phase transition of Ta_2NiSe_5, but they also conclusively rule out
any substantial excitonic character in this instability.

b. Evidence for topological defects in a photoinduced phase transition (Nature Physics 15, 27–31(2019))

Upon excitation with an intense laser pulse, a symmetry-broken ground state can undergo a non-equilibrium phase transition through pathways different from those in thermal equilibrium. The mechanism underlying these photoinduced phase transitions has long been researched in the study of condensed matter systems1, but many details in this ultrafast, non-adiabatic regime still remain to be clarified. To this end, we investigate the light-induced melting of a unidirectional charge density wave (CDW) in LaTe3. Using a suite of time-resolved probes, we independently track the amplitude and phase dynamics of the CDW. We find that a fast (approximately 1 picosecond) recovery of the CDW amplitude is followed by a slower re-establishment of phase coherence. This longer timescale is dictated by the presence of topological defects: long-range order is inhibited and is only restored when the defects annihilate. Our results provide a framework for understanding other photoinduced phase transitions by identifying the generation of defects as a governing mechanism.

c. Origin of the exciton mass in the frustrated Mott insulator Na2IrO3 (PHYSICAL REVIEW B 96, 235141 (2017))

We use a three-pulse ultrafast optical spectroscopy to study the relaxation processes in a frustrated Mott insulator Na2IrO3. By being able to independently produce the out-of-equilibrium bound states (excitons) of doublons and holons with the first pulse and suppress the underlying antiferromagnetic order with the second one, we were able to elucidate the relaxation mechanism of quasiparticles in this system. By observing the difference in the exciton dynamics in the magnetically ordered and disordered phases we found t

Training Opportunities: Graduate students and postdocs are trained in time-resolved optical and electron spectroscopies. These techniques are very powerful yet they are underrepresented in the physics departments. The trained personnel will form the future generation of scientists that will apply these techniques to different problems in physics.

Results Dissemination: Several invited talks were given and multiple papers were published in top journals.

Honors and Awards: Ludwig-Genzel-Prize (2020)
Moore Experimental Investigator in Quantum Materials (2015 and 2020)
Miller Visiting Professorship at UC Berkeley (2019)

Protocol Activity Status:

Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI
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Person Months Worked: 3.00 **Funding Support:**
Project Contribution:
National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
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Person Months Worked: 1.00 **Funding Support:**
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National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Timm Rohwer
Person Months Worked: 10.00 **Funding Support:**
Project Contribution:
National Academy Member: N

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

Participant: Doron Azoury
Person Months Worked: 3.00 **Funding Support:**
Project Contribution:
National Academy Member: N

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Person Months Worked: 3.00 **Funding Support:**
Project Contribution:
National Academy Member: N

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Project Contribution:
National Academy Member: N

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Project Contribution:
National Academy Member: N

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Project Contribution:
National Academy Member: N

Participant Type: Graduate Student (research assistant)
Participant: Ilkem Ozge Ozel
Person Months Worked: 6.00 **Funding Support:**
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Article Title: Selective scattering between Floquet–Bloch and Volkov states in a topological insulator
Authors: Fahad Mahmood, Ching-Kit Chan, Zhanybek Alpichshev, Dillon Gardner, Young Lee, Patrick A. Lee, Nu
Keywords: Ultrafast, ARPES, Floquet
Abstract: The coherent optical manipulation of solids is emerging as a promising way to engineer novel quantum states of matter. The strong time-periodic potential of intense laser light can be used to generate hybrid photon–electron states. Interaction of light with Bloch states leads to Floquet–Bloch states, which are essential in realizing new photo-induced quantum phases. Similarly, dressing of free-electron states near the surface of a solid generates Volkov states, which are used to study nonlinear optics in atoms and semiconductors9. The interaction of these two dynamic states with each other remains an open experimental problem. Here we use time- and angle-resolved photoemission spectroscopy (Tr-ARPES) to selectively study the transition between these two states on the surface of the topological insulator Bi2Se3. We find that the coupling between the two strongly depends on the electron momentum, providing a route to enhance or inhibit it. Moreover, by controlling the
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Article Title: Origin of the exciton mass in the frustrated Mott insulator
Authors: Zhanybek Alpichshev, Edbert J. Sie, Fahad Mahmood, Gang Cao, Nuh Gedik
Keywords: Mott insulators, dynamics, ultrafast
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Article Title: High resolution time- and angle-resolved photoemission spectroscopy with 11 eV laser pulses
Authors: Changmin Lee, Timm Rohwer, Edbert J. Sie, Alfred Zong, Edoardo Baldini, Joshua Straquadine, Philip \
Keywords: ARPES, instrumentation, high harmonic generation
Abstract: Performing time- and angle-resolved photoemission (tr-ARPES) spectroscopy at high momenta necessitates extreme ultraviolet laser pulses, which are typically produced via high harmonic generation (HHG). Despite recent advances, HHG-based setups still require large pulse energies (from hundreds of ?J to mJ) and their energy resolution is limited to tens of meV. Here, we present a novel 11 eV tr-ARPES setup that generates a flux of 5 × 1010 photons/s and achieves an unprecedented energy resolution of 16 meV. It can be operated at high repetition rates (up to 250 kHz) while using input pulse energies down to 3 μJ. We demonstrate these unique capabilities by simultaneously capturing the energy and momentum resolved dynamics in two well-separated momentum space regions of a charge density wave material ErTe3. This novel setup offers the opportunity to study the non-equilibrium band structure of solids with exceptional energy and time resolutions at high repetition rates.
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Acknowledged Federal Support: **Y**

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Journal: Physical Review X
Publication Identifier Type: Publication Identifier:
Volume: Issue: First Page #:
Date Submitted: 3/29/21 12:00AM Date Published:
Publication Location:
Article Title: The spontaneous symmetry breaking in Ta2NiSe5 is structural in nature
Authors: Edoardo Baldini, Alfred Zong, Dongsung Choi, Changmin Lee, Marios H. Michael, Lukas Windgaetter, Ig
Keywords: Excitonic insulator, ARPES
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Acknowledged Federal Support: **Y**

RPPR Final Report
as of 19-Apr-2021

Partners

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

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    b. **High resolution time- and angle-resolved photoemission spectroscopy with 11 eV laser pulses (Review of Scientific Instruments 91, 043102 (2020))**

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