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Theory of Non-Linear 2D Spectroscopy for Topological Quantum Matter

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14. ABSTRACT
During the project, we have focused on diagnosing the nature of exotic many-electron topological states in theory and experiment and exploring novel experimental setups of realizing them, which have potential uses in quantum technology. For the detection of the states in theory, guided by the linear/non-linear responses of topological states, we have developed novel many-body topological invariants for Chern insulator, chiral hinge insulators, and also several 2D higher-order topological insulators, which are calculable in numerical simulations. Furthermore, we have designed the detection scheme for the useful quantum states in experiments, i.e. non-Fermi liquids and topological Weyl semimetals from the linear response, crystalline topological band insulators from their resonant inelastic x-ray scattering intensity. These all provide valuable contributions to the detection of the exotic quantum states which receive a huge attention from the community. Not only this, our team also explored the new experimental setups, which may be helpful in realizing interesting quantum states. For instance, we have demonstrated the realization of the steady Floquet states in low-dimensional Andreev qubit setups, which opens up a new route to explore the non-equilibrium many-body quantum states. This may be very useful in controlling the qubits via the microwaves. Another interesting setup was the twisted Josephson junctions between the two high-Tc superconductors, which was speculated to realize the time-reversal invariant topological superconductors. However, we have disproved this claim and shown that it is actually a regular d-wave superconductor. In the submitted drafts, we have investigated high temperature excitonic condensate in the quantum Hall setups of large-angle twisted bilayer graphene, and non-linear optical response to diagnose the Dirac fermion in graphene. Finally, we have found the effective theory for far-more exotic quantum states, known as fracton, which may be used for quantum memory. This work, although purely theoretical, could be useful in classification of fractons and generating new fractonic phases.

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Section 2. Final Technical Report

Grant No. FA2386-20-1-4029

PI: Prof. Dr. Gil Young Cho

Project Abstract: We will theoretically develop a new general diagnosis method for topological quantum states, which can be experimentally implemented in the lab. The topological quantum states include quantum Hall liquids and spin liquids, which exhibit the non-local fractional particles. Because of the fractionalized particles, information can be stored non-locally in space and processed without decoherence. This makes the topological quantum states an ideal platform for quantum computation. However, how these states can be unambiguously identified in experiments is still unclear. Solid state physics of the last fifty years is founded upon the framework of the linear response theory and associated experimental measurement such as spin susceptibility. The linear response made great success in diagnosing usual phases of matter such as magnetism. However, it cannot be applied to detect fractional excitations of topological quantum states. Hence, one needs to go beyond the standard realm of the linear response theory. Indeed, it has been previously suggested in a few simple models that certain non-linear responses can detect the fractional excitation and thus the non-local entanglement. However, the systematic understanding of the non-linear responses for generic topological quantum states is absent. Motivated from this, we will systematically investigate the non-linear 2D spectroscopy of various topological quantum states. We believe that our proposal will provide a guiding principle for characterizing topological quantum states in the lab and this will in turn greatly help the realization of quantum computers based on topological quantum states. Our proposal will also broaden the realm of modern condensed matter physics beyond "linear response theory".

Accomplishments

Instruction: The information provided in this section allows AFOSR to assess whether satisfactory progress has been made during this reporting period.

Research Objectives of the Project

We plan to build a generic theory on the non-linear 2D spectroscopy of topological quantum matters by computing the responses for several topological quantum states. This will clarify the structures of the non-linear 2D spectroscopy in relation with fractional excitations. Furthermore, we will attempt to excavate a new ability of the 2d spectroscopy, i.e., detection of the fractional statistics of the excitations. Ultimately, we would like to prove that the non-linear 2D spectroscopy is a versatile, long-sought tool to diagnose the topological quantum states. To achieve this goal, we will perform the plans as outlined below.

1. We plan to investigate the generic structure of non-linear 2D spectroscopy of fractional excitations by calculating the responses for several paradigmatic topological quantum states. The target systems include the spin liquids and quantum Hall liquids.
2. We will investigate how the fractional statistics of fractional excitations can be manifested in the non-linear 2D spectroscopy. The target systems include the spin liquids and quantum Hall liquids.

List of Accomplishments

We first list the accomplishments achieved during this project. We published the following list of the papers (in peer-reviewed journals) and also preprints (in the Arxiv):

1. Many-Body Invariants for Chern and Chiral Hinge Insulators, B. Kang et al, Physical Review Letters (2021)
2. Twisted van der Waals Josephson Junction Based on a High-Tc Superconductor, J. Lee et al, Nano Letters (2021)
3. Non-Fermi Liquids in Conducting Two-Dimensional Networks, J. Lee et al, Physical Review Letters (2021)
4. Chiral Anomaly in Non-centrosymmetric systems induced by spin-orbit coupling, S. Cheon et al, Physical Review B (2022)
5. Many-Body Quadrupolar Sum Rule for Higher-Order Topological Insulators, W. Lee et al, Physical Review B (2022)
6. Steady Floquet-Andreev states in graphene Josephson junctions, S. Park et al, Nature (2022)
7. Metrology of Band Topology via Resonant Inelastic X-ray Scattering, S. Lee et al, ARXIV:2108.02211 (2021) – Under Review at Physical Review Letters
8. A Symmetry Principles for Gauge Theories with Fractons, Y. Hirono et al, ARXIV:2207.00854 – Under Review at Sci-post

The above eight manuscripts were reported in AFOSR survey website. There are two more manuscripts, which are neither published nor uploaded (as preprints) in ARXIV, but are submitted to the journals. They are:

9. Robust Interlayer-Coherent Quantum Hall states in Twisted Bilayer Graphene, D. Kim et al – Under Review at Nature Materials
10. Gate-Tunable Quantum Pathways of Massless Dirac Fermions in High Harmonic Generation, S. Cha et al - Under Review at Nature Communications

In total, we had ***6 manuscripts published in journals and 4 manuscripts under review*** at journals during the two years of the support from AOARD/AFOSR. We will explain the details of these accomplishments below one by one.

1: Many-Body Invariants for Chern and Chiral Hinge Insulators, B. Kang et al, Physical Review Letters (2021)

This work allows the extension of the higher order topology (which was previously defined only for clean, non-interacting electronic systems) to the disordered, interacting systems and shows how they can be efficiently identified in theory/numerical calculations.

The paper was published in one of the most impactful physics journals, Physical Review Letters.

2: Twisted van der Waals Josephson Junction Based on a High-Tc Superconductor, J. Lee et al, Nano Letters (2021)

Stacking two-dimensional van der Waals (vdW) materials rotated with respect to each other show versatility for the study of exotic quantum phenomena. Especially, anisotropic layered materials have great potential for such twistronics applications, providing high tunability. This work reports anisotropic superconducting order parameters in twisted Bi2Sr2CaCu2O8+x (Bi-2212) vdW junctions with an atomically clean vdW interface, achieved using the microcleave-and-stack technique. The vdW Josephson junctions with twist angles of 0° and 90° showed the maximum Josephson coupling, which was comparable to that of intrinsic Josephson (IJ) junctions in the bulk crystal. As the twist angle approaches 45°, Josephson coupling is suppressed, and eventually disappears at 45°. The observed twist angle dependence of the Josephson coupling can be explained quantitatively by theoretical calculation with the d-wave superconducting order parameter of Bi-2212 and finite tunneling incoherence of the junction. The results reported in this work reveals the anisotropic nature of Bi-2212 and provided a novel fabrication technique for vdW-based twistronics platforms compatible with air-sensitive vdW materials.

The paper was published in one of the most impactful applied physics journals, Nano Letters. This paper was also **highlighted in Journal Club for Condensed Matter Physics in 2022 August**.

3: Non-Fermi Liquids in Conducting Two-Dimensional Networks, J. Lee et al, Physical Review Letters (2021)

This work shows the possibility of emergence of the novel 2D non-Fermi liquids in various classes of moiré materials, which contain 2D networks of conducting 1D wires. We demonstrated how their non-Fermi nature can be experimentally manifested by investigating the non-linear temperature dependences of their linear responses. This work points to the novel ways of realizing non-Fermi liquids (more than 100s of new non-Fermi liquid states) as well as verifying their characters.

The paper was published in one of the most impactful physics journals, Physical Review Letters.

4: Chiral Anomaly in Non-centrosymmetric systems induced by spin-orbit coupling, S. Cheon et al, Physical Review B (2022)

The chiral anomaly may be realized in condensed matter systems with pairs of Weyl points. This work shows that the chiral anomaly can be realized in diverse non-centrosymmetric systems even without Weyl point pairs when spin-orbit coupling (SOC) induces nonzero Berry curvature flux through Fermi surfaces (FS). This motivates the condensed matter chiral anomaly to be interpreted as a FS property rather than a Weyl point property. The SOC-induced anomaly reproduces the well-known charge transport properties of the chiral anomaly such as the negative longitudinal magnetoresistance and the planar Hall effect in Weyl semimetals. Since it is of SOC origin, it also affects the spin transport and gives rise to anomaly induced longitudinal spin currents and the magnetic spin Hall effect, which are absent in conventional Weyl semimetals.

This work has been published in Physical Review B.

5: Many-Body Quadrupolar Sum Rule for Higher-Order Topological Insulators, W. Lee et al, Physical Review B (2022)

The modern theory of polarization establishes the bulk-boundary correspondence for the bulk polarization. This work extends it to a sum rule of the bulk quadrupole moment by employing a many-body operator introduced in Kang et al. [B. Kang, K. Shiozaki, and G. Y. Cho, Phys. Rev. B 100, 245134 (2019)] and Wheeler et al. [W. A. Wheeler, L. K. Wagner, and T. L. Hughes, Phys. Rev. B 100, 245135 (2019)]. The sum rule that we propose consists of the alternating sum of four observables, which are the phase factors of the many-body operator in different boundary conditions. This work clearly demonstrates its validity through extensive numerical computations for various noninteracting tight-binding models. We also observed that individual terms in the sum rule correspond to the bulk quadrupole moment, the edge-localized polarizations, and the corner charge in the thermodynamic limit on some models.

This paper was published in Physical Review B.

6: Steady Floquet-Andreev states in graphene Josephson junctions, S. Park et al, Nature (2022)

This work shows the first ever realization of the steady Floquet states in solid state systems, whose nature is thoroughly investigated via superconducting tunnel probe. The experimental data is compared with extensive theoretical calculations on linear responses and thus supports unambiguously the emergence of the steady states. It may serve as the fundamentals for the experimental studies of steady far-from-equilibrium states, including topological and/or non-topological, periodic and/or quasi-periodic non-equilibrium states.

The paper was **published in one of the most impactful science journals, Nature**.

7: Metrology of Band Topology via Resonant Inelastic X-ray Scattering, S. Lee et al, ARXIV:2108.02211 (2021) – Under Review at Physical Review Letters

This work investigated the possibility of exploiting RIXS for probing the topology of the electronic band structure, which is classified by a topological band index. This work shows, by calculating the linear responses of several topological phases, that the RIXS spectral intensity is shown to depend on the band indices, which are mathematically defined "mod 1". Hence, it represents a breakthrough in theoretical understandings of RIXS by revealing its new ability, namely probing the band topology.

The paper is currently at the last stage of review at Physical Review Letters.

8: A Symmetry Principles for Gauge Theories with Fractons, Y. Hirono et al, ARXIV:2207.00854 – Under Review at Sci-post

Fractonic phases are new phases of matter that host excitations with restricted mobility. This work shows that a certain class of gapless fractonic phases are realized as a result of spontaneous breaking of continuous higher-form symmetries whose conserved charges do not commute with spatial translations. We refer to such symmetries as nonuniform higher-form symmetries. These symmetries fall within the standard definition of higher-form symmetries in quantum field theory, and the corresponding symmetry generators are topological. Worldlines of particles are regarded as the charged objects of 1-form symmetries, and mobility restrictions can be implemented by introducing additional 1-form symmetries whose generators do not commute with spatial translations. These features are realized by effective field theories associated with spontaneously broken nonuniform 1-form symmetries. At low energies, the theories reduce to known higher-rank gauge theories such as scalar/vector charge gauge theories, and the gapless excitations in these theories are interpreted as Nambu-Goldstone modes for higher-form symmetries. Due to the nonuniformity of the symmetry, some of the modes acquire a gap, which is the higher-form analogue of the inverse Higgs mechanism of spacetime symmetries. The gauge theories have emergent nonuniform magnetic symmetries, and some of the magnetic monopoles become fractonic. We identify the 't Hooft anomalies of the nonuniform higher-form symmetries and the corresponding bulk symmetry-protected topological phases. By this method, the mobility restrictions are fully determined by the choice of the commutation relations of charges with translations. This approach allows us to view existing (gapless) fracton models such as the scalar/vector charge gauge theories and their variants from a unified perspective and enables us to engineer theories with desired mobility restrictions.

The paper is currently under review at the peer-reviewed journal, Sci-post.

9: Robust Interlayer-Coherent Quantum Hall states in Twisted Bilayer Graphene, D. Kim et al – Under Review at Nature Materials

Bilayer quantum Hall systems host quantum phases which cannot be realised in single-layer systems. Bose–Einstein condensation is the prototypical example where electron and hole excitations from different layers form exciton condensates owing to interlayer Coulomb interactions. However, such an exotic quantum state has only been realised at sub-Kelvin temperatures in conventional GaAs systems, mainly because of weak interlayer couplings. This work introduces a novel two-dimensional electronic system with ultrastrong interlayer interactions, namely twisted bilayer graphene with a large twist angle, as an ideal ground for realising interlayer-coherent excitonic condensates. In these systems, subnanometre atomic separation between the layers allows significant interlayer interactions, while interlayer electron tunnelling is geometrically suppressed due to the large twist angle. By fully exploiting these two features we demonstrate that a sequence of odd-integer quantum Hall states with interlayer coherence appears at the second Landau level (N = 1). Notably the energy gaps for these states are of order 1 K, which is several orders of magnitude greater than those in GaAs. Furthermore, a variety of quantum Hall phase transitions are observed experimentally. All the experimental observations are largely consistent with our phenomenological model calculations. Hence, we establish that a large twist angle system is an excellent platform for high-temperature excitonic condensation.

This paper is currently under review at Nature Materials.

10: Gate-Tunable Quantum Pathways of Massless Dirac Fermions in High Harmonic Generation, S. Cha et al - Under Review at Nature Communications

Under strong laser field, electrons in solids radiate high-order harmonics by travelling quantum pathways along Bloch bands in sub-laser-cycle timescale. Imaging these pathways through high harmonic radiation can enable an ultrafast and all-optical tomography to measure band structure and electronic properties as recently demonstrated for semiconductors. However, tomography of semimetals has been challenging because pathways in semi-metallic bands remain largely elusive yet. In this work, we visualize widely tunable quantum pathways of massless Dirac fermions in graphene by controlling chemical potential. High harmonic generation (HHG) measurement reveals multiple interband excitation channels under destructive interference which can be systematically modulated by chemical potential. Furthermore, in accordance with our theoretical calculation, we identify a unique mechanism for massless Dirac fermions where elliptically polarized excitation efficiently drives carrier motion via intricate coupling between interband and intraband transitions. Our work provides microscopic understandings on HHG to visualize sub-cycle dynamics of massless quasiparticles in various quantum semimetals.

This paper is currently under review at Nature Communications.

Impacts

<u>**Instruction:**</u> This component is used to describe ways in which the work, findings, and specific products of the project have had an impact during this reporting period. Describe distinctive

contributions, major accomplishments, innovations, successes, or any change in practice or behavior that has come about as a result of the project. You can report on the following impact categories, but you are not required to report on all categories.

On Principal Disciplines of the Project

During the last two years, our group's work has **published several key papers in the field of topological phases and quantum materials**. They answer the most important questions on the quantum many-body systems: theoretical, experimental diagnosis of them, and also the search for the novel platforms to find them.

(1) Efficient diagnosis of strongly-correlated topological phases in Theory

A. Many-Body Invariants for Chern and Chiral Hinge Insulators, B. Kang et al, Physical Review Letters (2021)

B. Many-Body Quadrupolar Sum Rule for Higher-Order Topological Insulators, W. Lee et al, Physical Review B (2022)

(2) Devising **novel experimental approaches to diagnose the nature of the correlated quantum materials**

A. Twisted van der Waals Josephson Junction Based on a High-Tc Superconductor, J. Lee et al, Nano Letters (2021)

B. Non-Fermi Liquids in Conducting Two-Dimensional Networks, J. Lee et al, Physical Review Letters (2021)

C. Metrology of Band Topology via Resonant Inelastic X-ray Scattering, S. Lee et al, ARXIV:2108.02211 (2021)

D. Gate-Tunable Quantum Pathways of Massless Dirac Fermions in High Harmonic Generation, S. Cha et al - Under Review at Nature Communications

(3) Novel **platforms to find the topological phenomena**

A. Chiral Anomaly in Non-centrosymmetric systems induced by spin-orbit coupling, S. Cheon et al, Physical Review B (2022)

B. Steady Floquet-Andreev states in graphene Josephson junctions, S. Park et al, Nature (2022)

C. A Symmetry Principles for Gauge Theories with Fractons, Y. Hirono et al, ARXIV:2207.00854

D. Robust Interlayer-Coherent Quantum Hall states in Twisted Bilayer Graphene, D. Kim et al – Under Review at Nature Materials

The above works will serve as an important step to the quest of searching for topological phases, which can **boost up the development of the topological quantum computations, as the project originally targeted**.

On Development of Human Resources

PI of this project has received ***2 major awards*** during the project.

- "***Rising Physicist Award*" from Korean Physics Society (2021)**: This award selects only a few physicists every year among the level of assistant/associate professors. It is the most prestigious award that the junior-level professors can receive from Korean Physics Society (KPS).
- "***This month's Science, Technology, and Researcher Award*" from Ministry of Science and ICT/National Research Foundation of Korea (2022)**: This award selects only one researcher in every fields of the science and technology. It is one of the highest honors that the researcher in science and engineering can receive from the government of Korea.

Both of them are highly selective and **the awards acknowledge that the PI of this project has been successful in research**.

Changes

Instruction: In this section, please incorporate any and all changes that you would like your program officers to know about the grant. The principal investigator is reminded that the recipient organization is required to obtain prior written approval from program officers whenever there are significant changes in the project or its direction.

Changes in approach: Originally, PI thought that the project would be purely theoretical. However, PI found the chances to collaborate with the experimental researchers and made more research papers produced (than originally expected) from the collaborations with experimentalists.

Significant changes in the use of human subjects, vertebrate animals, and/or biohazards: It doesn't apply to the current project. It doesn't involve any of the human subjects, vertebrate animals, and/or biohazards.

Technical Updates

Instruction: This section will include any and all technical updates that you would like to provide to your program officer. You are encouraged to upload graphs and other visualizations that highlight the work done during this reporting period.

As you can see from the above, the project produced ***10 manuscripts during the 2 years***, among which 4 papers were already published in the top-notched journals: 2 Physical Review Letters, 1 Nano Letter, and 1 Nature. **This has been an extremely successful project**.