Evidence of excitonic insulators in moiré superlattices

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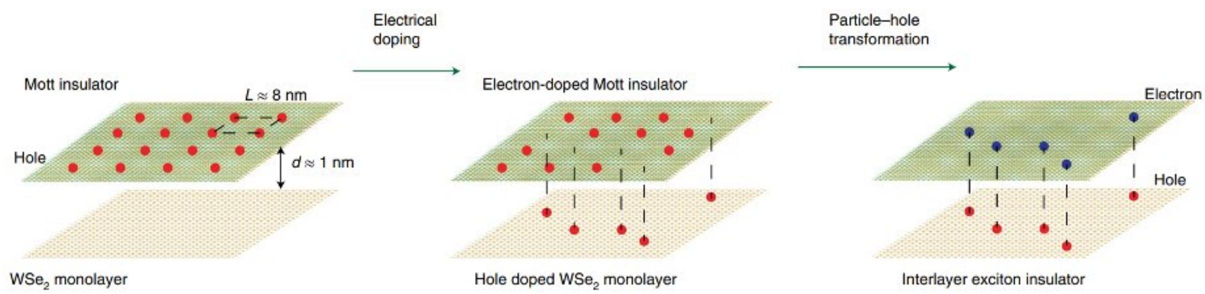


The double-layer system consists of the Mott insulator (top layer) and band insulator (the WSe_2 monolayer). The Mott insulator has one hole per moiré lattice site, and the band insulator is intrinsic. We add electrons to the Mott insulator and an equal number of holes to the WSe_2 monolayer. The holes in the WSe_2 monolayer will avoid positions below the moiré lattice site that are occupied by holes owing to the strong interlayer Coulomb interaction. After the particle-hole transformation relative to the Mott insulator, electrons doped into the Mott insulator can spontaneously bind the holes in the Mott insulator. Credit: Zhang et al

Excitons are quasiparticles that are formed in insulators or semiconductors when an electron is promoted to a higher energy band, leaving a positively charged hole behind.

At the presence of strong Coulomb interaction, electrons and holes (vacancies left by electron that is viewed positively charged quasiparticle) form tightly bound electron-hole pairs, which are called excitons.

This process causes the electron and hole to bind together, creating an exciton, which is essentially a mobile concentration of energy that behaves similarly to particles. Excitons are ubiquitous in optically excited semiconductors. However, in rare scenarios, they can spontaneously form in a small bandgap semiconductor or semimetal.

In the 1960s, physicist Nevill Mott put forward an interesting theoretical hypothesis, suggesting that if the band structure of a material was to be tuned in a specific way (i.e., with an upper energy level below the lower energy level at certain points), then the system's ground state would contain excitons. Excitons in such a system would be neutrally charged, thus the material could be classified as an insulator.

While many physicists built on Mott's interesting hypothesis, so far it had never been proven in an experimental setting. This was until last year, when two different research teams at Princeton University and at University of Washington gathered the first experimental evidence of an excitonic insulating state in monolayer tungsten ditelluride.

Recently, research by another two research groups demonstrated the creation of excitonic insulators, using what are known as moiré superlattices. Moiré superlattices are heterostructures characterized by 2D layers stacked on top of each other, with a twist angle or a lattice mismatch. The first of these studies, conducted by the team at UC Berkeley and published in *Nature Physics*, reported the observation of a correlated interlayer exciton insulating state in a heterostructure made up of a WSe_2 monolayer and a WS2/WSe_2 moiré bilayer.

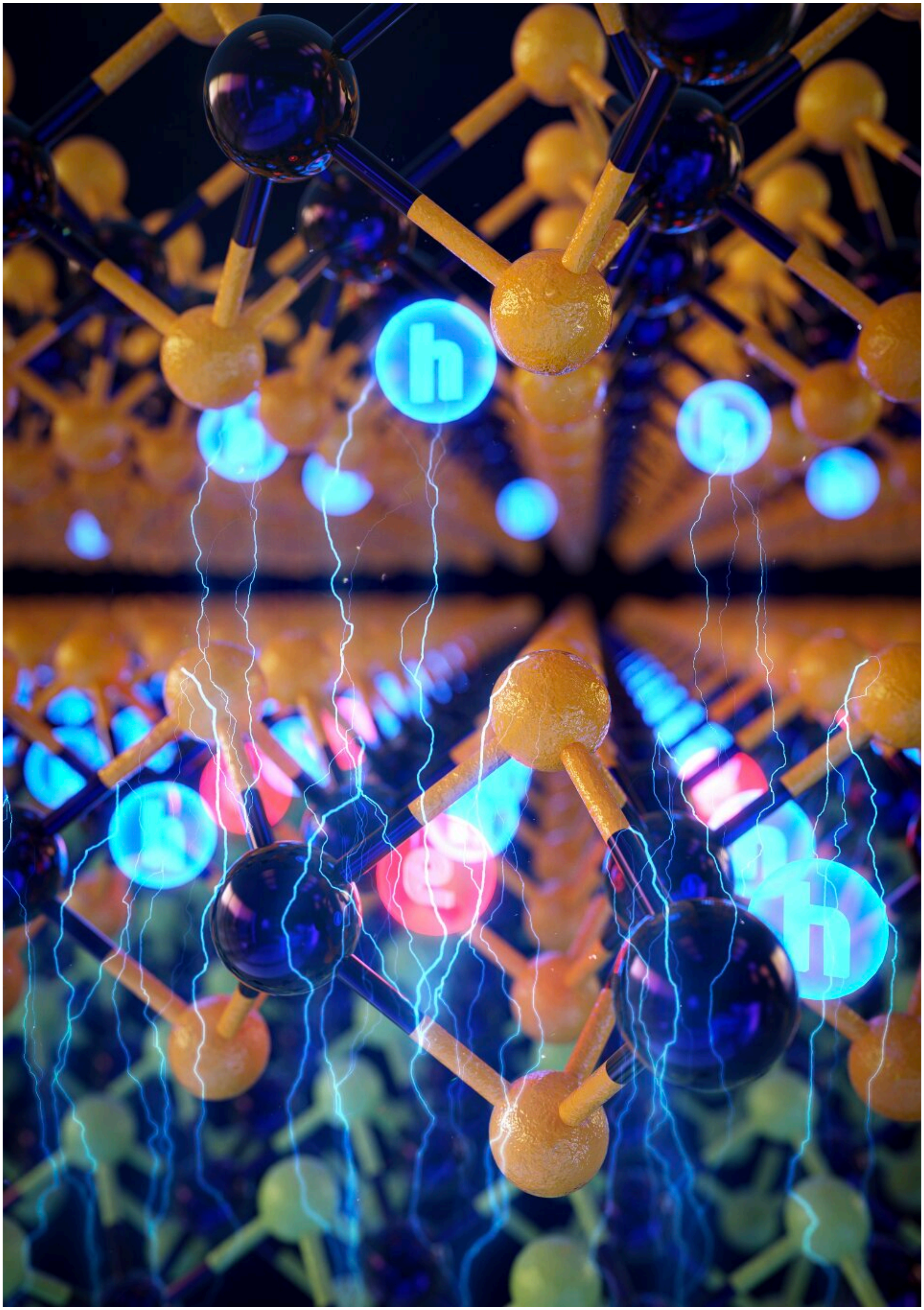
"Excitonic insulators, first proposed by N.F. Mott in 1961, had already been demonstrated in quantum Hall double-layer system, where Landau levels in a strong magnetic field are flat electronic bands that suppress the kinetic energy and enhance the electron-hole correlation,"

Zuocheng Zhang, one of the researchers at UC Berkeley who carried out this other study, told Phys.org. "We considered whether we could achieve the interlayer exciton insulator at zero magnetic field."

Moiré superlattices are widely investigated structures that are also known to host flat electronic bands. Zhang and his colleagues decided to integrate the moiré superlattice into a double-layer system and then looked for the excitonic insulating state at a magnetic field of zero.

"We realized a double-layer heterostructure composed of a WS_2/WSe_2 moiré bilayer and a WSe_2 monolayer," Zhang explained. "A 1-nm-thick hBN separates these two layers. We stack up the moiré bilayer, insulating hBN layer, and a WSe_2 monolayer by using the polymer-based try transfer technology."

The other group who observed an excitonic insulator in a moiré superlattice included researchers from different institutes in the US, China and Japan, including the Rensselaer Polytechnic Institute, the University of Electronic Science and Technology of China, University of California Riverside, University of Texas at Dallas, Arizona State University and the National Institute for Materials Science in Japan. This large research collaboration specifically used a natural bilayer WSe_2 and one monolayer WS2 to construct a three-layer excitonic insulator.



A schematic that shows the EI state, with the effective electrons and holes occupying different layers of WSe_2. Credit: Chen et al

"The goal of our study was to demonstrate a new insulating state, proposed more than 50 years ago by Leonid Keldysh and others," Sufei Shi, one of the researchers who carried out the study, told Phys.org. "It is predicted that, in a small bandgap semiconductor or a semimetal, coexisting electrons and holes will spontaneously bond when the Coulomb interaction is strong, forming an insulating ground state, excitonic insulator. This state is believed to share some similarity with the quasiparticles (BCS copper pair) that give rise to superconductivity and can lead to macroscopic coherent phenomena."

The key objective of the recent work by Shi and his colleagues was to create a robust excitonic insulator system using 2D materials. These materials were combined to form a new periodic structure, using band engineering techniques.

"We choose the combination of a natural bilayer WSe_2 and one monolayer WS2 to construct a three-layer excitonic insulator," Shi said. "Both of these materials were obtained through mechanical exfoliation (the same technique used to obtain graphene)."

After obtaining the materials for their system, the researchers assembled them to form a moiré superlattice, precisely controlling the twist angle in-between the layers (i.e., with 0 or 60 degrees). They then tried to engineer it so that it would have both electrons and holes, to enable the excitonic insulator state.

"In the moiré system, a flat energy band is formed at the interface between WSe_2 and WS2, which allows us to tune the carrier polarity, i.e., the carriers are hole-like near the top of the band and electron-like near the bottom of the band," Prof. Yong-Tao Cui from UC Riverside, a senior author of the second work, said.

"The additional layer of WSe_2 contributes a hole band. Therefore, by using an electric field, we can tune the flat moiré band to host electrons while the holes are in the second WSe_2 band. This creates the condition of coexisting electrons and holes, which interact strongly to form the excitonic insulator state. This hypothesis was also confirmed by the calculations ran by Prof. Chuanwei Zhang's group at UT Dallas."

The new correlated interlayer exciton insulator demonstrated by Zhang and his colleagues at UC Berkeley included the holes of a band insulator (in the WSe_2 monolayer) and electrons of a Mott insulator (in the WS2/WSe_2 moiré bilayer). The insulator state demonstrated by Shi and his colleagues, on the other hand, was based on a natural WSe_2 bilayer and a WS2 monolayer.

"Our study highlights the opportunities for exploring new quantum phenomena in double-layer moiré systems," Zhang added. "The interlayer excitons in our system can potentially form an exciton condensate at sufficiently low temperatures. We now plan to perform further experiments aiming at the demonstration of exciton superfluidity."

The recent studies by these two teams of researchers highlight the potential of double-layer moiré systems as platforms for realizing quantum phases. In the future, they could pave the way for more research using moiré superlattices to study 2D correlated many-body physics.

"We have constructed a robust excitonic insulator with transition temperature as high as 90 K," Shi added. "The system is also highly tunable with an electric field. This robust EI system enables the future study of EI, especially on the new quantum states and their macroscopic coherent effects. For example, we will explore the superfluidity of the excitons."

More information: Dongxue Chen et al, Excitonic insulator in a heterojunction moiré superlattice, *Nature Physics* (2022). DOI: 10.1038/s41567-022-01703-y

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