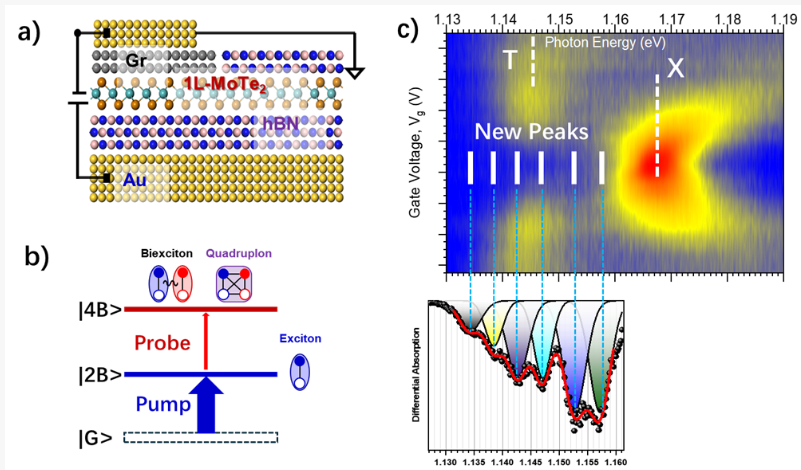
Scientists discovered evidence for a new four-body quasi-particle in 2D semiconductors

GA, UNITED STATES, March 28, 2025 /EINPresswire.com/ -- Quasi-particles are collective quantum elements of condensed matter. They are foundationally important for understanding physics of materials and for technological applications of semiconductors. Using ultrafast optical pump-probe technique, scientists discovered a series of new spectral features that are not attributable to existing quasi-particles. By using advanced quantum many-body theories, these spectral features could be uniquely explained by the existence of a new quasi-particle called quadruplon, a genuine four-body composite particle, different from the known bi-exciton.



a), Device structure: a monolayer of MoTe2 is sandwiched between two thin boron nitride layers. A thin graphene layer is used for contacting MoTe2 from the top. Gate voltage is applied below the bottom boron nitride layer. b), Schematic energy scheme show

One of the central tasks of physics is to understand how various materials possess certain properties such as electrical, optical, thermal, or magnetic properties. These properties underlie all technological applications of materials. Modern quantum theory explains such properties in terms of various “elementary excitations” or so-called quasi-particles. Well-known examples of such quasi-particles include phonons in a crystal, electrons and holes modified by the lattice in a semiconductor, or those further “dressed” by phonons called polarons. More complex quasi-particles include excitons of “two-body” type that are hydrogen-like electron-hole pairs. Two excitons could bind together forming an exciton-molecule, called bi-exciton, similar to a hydrogen molecule. Even though a bi-exciton contains a total of four particles (2 electrons and 2 holes), they are considered reducible to particles of two-body type (two excitons). Scientists have theorized in various contexts of physics that there are genuine four-body entities that are not reducible to two-body ones. But experimental evidence has been few and rare, and none has been found so far in semiconductors.

In a new paper published in eLight, a team of scientist led by Professor Cun-Zheng Ning from College of Integrated Circuits and Optoelectronic Chips, Shenzhen Technology University and Electronic Engineering Department, Tsinghua University, China, reported experimental results and supporting theory that show first evidence of the existence of a genuine four-body quasi-particle called quadruplon in a semiconductor of a mono-molecular thickness, the so-called two-dimensional material, Molybdenum Ditelluride.

The team used a monolayer of Molybdenum Ditelluride sandwiched by thin layers of boron nitride from the top and bottom after the top is contacted by a metal electrode. The bottom boron nitride layer serves as a dielectric layer under which “gate” electrode is used (see Figure). The device structure allows the team to tune the charges inside the monolayer semiconductor to study how the spectral responses will change with gate voltage. To study the spectral properties, the team then used an optical pump-probe technique: a strong short pulse of hundreds of femtosecond in length is sent to pump the electrons in the monolayer and a weak probe pulse is then used to see how the pump-excited monolayer absorbs photons of probe laser after a short delay. The rationale of such experiments is as follows: the strong pump excites electrons from the crystals to form various quasi-particles or multi-body entities such as excitons, or trions involving one electron and two holes (or two electrons and one hole), or bi-excitons mentioned above. The photons of the probe pulse are then absorbed by these newly created quasi-particles to jump to various higher new states, called final states, which typically involve now four-particles. By measuring the reflected or transmitted light of the probe pulse, one can gain important information about the final states, or the four-body states. Since these quasi-particles often have very short lifetime on the orders of picoseconds, a probe pulse must come soon enough before they die out. To get more systematic information from this pump-probe technique, the team varied gate voltage, strength of pump pulse, delay time between the pump and probe, the energy and polarization of the probe pulses, and the temperature of the samples etc. In a typical such experiment, various quasi-particles appear in terms of characteristic spectral peaks.

Scientists so far have only observed quasi-particles such as excitons, trions, bi-excitons, or entities that involve weakly couplings of these known entities. They often appear as a spectral peak or two below the exciton peak in an absorption spectrum.

“To our surprise, our measured absorption spectrum showed at least 6 peaks below that of exciton”, said Cun-Zheng Ning, the leader of the team. “The first thing one has to rule out in this situation is effects of defects. Our group has prepared and tested many hundreds of samples of this type over the last ten years. We knew how to tell the difference between a good sample with almost no defect peaks and a bad one with. We repeated experiments on five of our best samples with little defects and these peaks are repeatable and robust.” Continued Ning.

“To further establish the intrinsic nature of these peaks, we performed systematic temperature and pump-power dependence of these new spectral peaks. We tried every experiment we could think of and were able to do. All experimental evidence showed intrinsic nature and robustness of the new spectral features." Said Dr. Jiacheng Tang, who was a PhD student in Ning's group and the first author of the paper.

Once the new spectral features were established, the team tried to explain them in terms of existing theories that include excitons, trions, and bi-excitons. It turned out that these known quasi-particles could not explain all these new features. The team had to go beyond traditional approximations by including all the possible terms of Coulomb interaction between two electrons and two holes. To their pleasant amazement, the new theory could now produce all the key features of the experiment.

"Normally, one might consider the problem solved, since we got very good agreement between experiment and theory", said Ning. "But since the four-body interaction involves so many different terms, it is curious to ask which of these terms caused the new spectral peaks. Furthermore, we lacked an intuitive picture of understanding. So I challenged my student if we could work out an alternative theory that is more intuitive," continued Ning.

The alternative theory Ning had in mind is so-called cluster expansion. It is a technique to make successive approximation to the full Coulomb interaction which is not exactly solvable. The advantage of the cluster expansion is its intuitive nature.

"To our pleasant surprise and satisfaction, we were able to successfully establish the correspondence between the two theoretical approaches. We were able to show that the experimental results could be explained by both approaches," Ning further explained and he continued "Now the additional advantage of the cluster expansion approach is that one can artificially turn on and off a particular cluster to see which of these clusters is responsible for the new spectral features."

Eventually, the authors were able to show that there was a crucial cluster, the so-called fourth order cluster that involved two electrons and two holes which cannot be reduced to combinations of lower orders. Once this cluster is included, all the experimental features were reproduced. Thus they were able to finally pin point the unique role of this fourth order cluster, which corresponds to a new composite entity, or quasi-particle, involving two electrons and two holes. The authors call this new particle quadruplon, in accordance with some theoretical literature.

As for the future, the authors believe that there are a lot to be explored. They plant to measure similar spectral features in other material systems. In addition, the authors believe that the light emission properties of the newly discovered quadruplon are highly interesting, as they might reveal new quantum nature of the new highly correlated many-body complex.

References

DOI
10.1186/s43593-025-00081-1

Original Source URL
https://doi.org/10.1186/s43593-025-00081-1

Funding information
This work was supported by the National Natural Science Foundation of China (Grant No. 91750206, No. 61861136006); Pingshan Innovation Platform Project of Shenzhen Hi-tech Zone Development Special Plan in 2022(29853M-KCJ-2023-002-01); Universities Engineering Technology Center of Guangdong (2023GCZX005); Key Programs Development Project of Guangdong (2022ZDJS111); Natural Science Foundation of Top Talent at SZTU (GDRC202301), Tsinghua University Initiative Scientific Research Program.

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