Experiments confirm light-squeezing 2-D exciton-polaritons can exist

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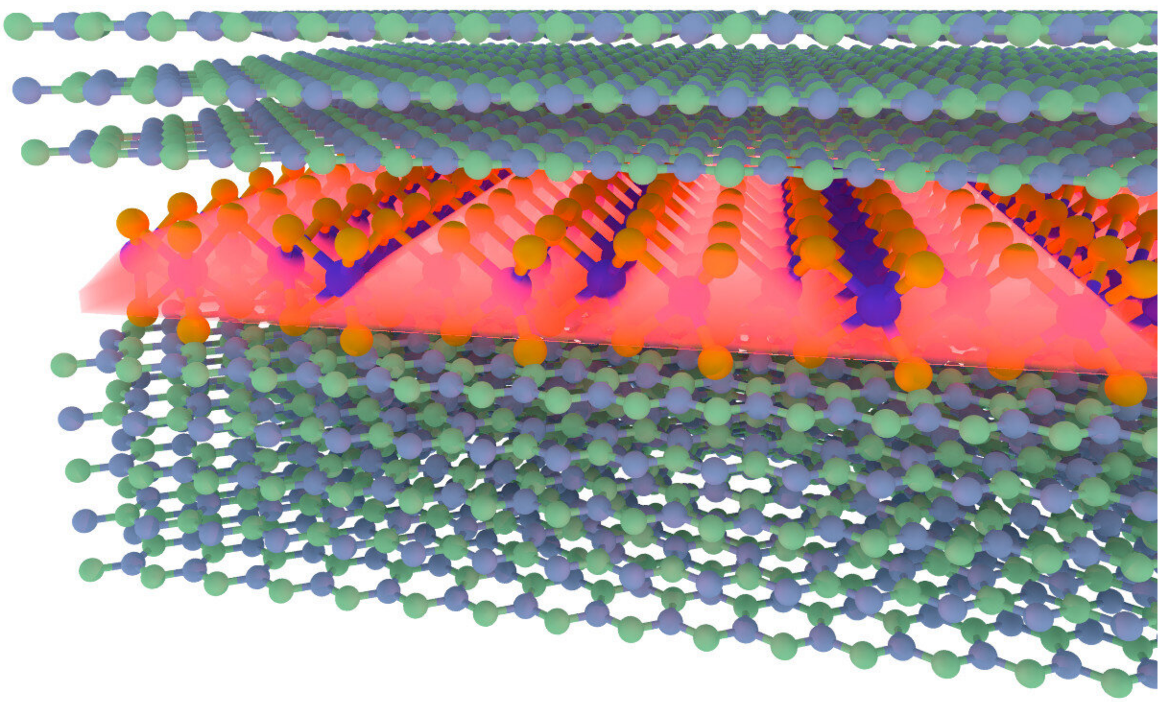

Researchers pinpoint the conditions for forming unusual 2D exciton-polaritons in TMDs. Credit: Fabien Vialla

Measurements of the optical response of 2-D transition-metal dichalcogenides have now pinpointed real material systems in which a hypothesized light-squeezing quasiparticle can form. The 2-D exciton-polariton, which couples light to bound electron-hole pairs in the form of excitons in an unusual way, can confine light to dimensions orders of magnitude below the diffraction limit. Confining light to such a high degree may affect more than the resolving power of imaging devices and detector sensitivity. Recent studies of cavity modes have suggested that highly confined light could also alter the inherent properties of materials.

Polaritons describe a wide range of quasiparticles that are half light and half matter. As a result, it is possible to manipulate one aspect using the other. Polaritons in 2-D materials in particular have attracted much interest in this respect, because the light confinement they exhibit can be particularly extreme, and can be manipulated through the matter aspect of the quasiparticle. This has already attracted interest in graphene (monolayers of hexagonal crystalline carbon), in which light coupling with resonant electrons—plasmon-polaritons—may lead to more convenient devices for cheaper, broader-wavelength, high-performance infrared detectors.

2-D forms of transition-metal-dichalcogenides (TMDs) semiconductors such as MoS_2, $MoSe_2$, WS_2 and WSe_2 have also attracted interest over the past eight years, but these materials behave quite differently. Far more prone to defects than graphene, TMDs do not support plasmons. However, excitons have been observed owing to the semiconducting nature of TMDs, even at room temperatures. Itai Epstein and group leader Frank Koppens, both researchers at Institut de Ciencies Fotoniques (ICFO) in Spain, led an international team of collaborators to shed light on a particular type of exciton polariton in 2-D TMDs that no one has so far observed.

A new kind of polariton

The exciton polaritons observed so far couple to light perpendicular to the plane of the monolayer, but theories suggest that light could couple to excitons of a monolayer TMD in a way that more closely resembles the coupling to plasmons. "It couples to the exciton in such a manner that both are then bound to the monolayer itself and propagate along it as a special kind of wave," explains Epstein, as he describes what distinguishes these 2-D exciton-polaritons from the exciton-polaritons that have been observed before.

However, it was not clear if TMD monolayers can actually provide the required material response to support such 2-D exciton-polaritons, as previous observations suggested that they might not. "It was important for us to show experimentally that this is not some idea that is not related to reality," Epstein adds. "We showed that if one can control the properties of the TMD excitons, the conditions required for the 2-D exciton-polaritons are, indeed, achievable to obtain from a real TMD."

What the quasiparticle needs

The excitons in 2-D TMDs have already proved to be a font of fascinating phenomena. In fact, Koppens and Epstein had recently reported measurements of excitons in 2-D TMDs that absorb close to 100% of the light that falls on them. Coming from a background in plasmonics, Epstein was interested in how the resonant conditions for this 100% absorption resembled the conditions needed for the existence of 2-D exciton-polaritons.

One of the first things people do when trying to observe interesting effects in 2-D materials is encapsulate it in 2-D hexagonal boron nitride (hBN). Sometimes described as the real "wonder material" in 2-D materials research, hBN is very flat and clean, which helps it not only to preserve, but to improve the characteristics of 2-D materials. For example, it has already been shown that excitons in a 2-D TMD encapsulated in hBN resemble the characteristics of excitons in a monolayer that is completely defect-free.

The second trick is to suppress the lattice vibrations that dampen the excitons, making it nigh on impossible to observe the elusive 2-D exciton polaritons. These lattice vibrations can be suppressed by lowering the temperature. The damping processes are expressed as an imaginary term in the complex value of a material's permittivity (its polarizability in response to the electromagnetic field of incident light). However, for the plasmon-like 2-D exciton-polaritons to exist, as well as low damping, the real part of the permittivity needs to be negative. By measuring optical characteristics like the reflection contrast and complex permittivity of hBN encapsulated 2-D TMDs at cryogenic temperatures, Epstein, Koppens and their collaborators were able to identify the frequency range and conditions where the real part of the permittivity was negative while the damping was low. They could also calculate and compare the light confinement of the 2-D exciton-polariton versus a surface-plasmon-polariton at the interface of an hBN monolayer on a gold substrate. The confinement of the 2-D exciton-polariton was over 100 times greater than the surface-plasmon-polariton.

In the report, Epstein, Koppens and their collaborators describe the structures needed to observe the 2-D exciton polaritons themselves, either TMD patterned into nanoribbons or hBN-encapsulated 2-D TMD placed on a thin metallic grating. While using a grating would get around the losses incurred from rough edges when patterning the TMD itself, both approaches require formidably precise nanofabrication. Epstein considers these structures "definitely feasible," although their construction will be challenging. "We are now focusing efforts on achieving the capabilities to fabricate the required patterned structures in a reliable and consistent manner by using cutting-edge nano-fabrication facilities," he adds.

Koppens highlights how the developments may feed into the emerging field of cavity mode photonics, which looks at how virtual photons that pop in and out of existence affect the behaviour of a system, even in a vacuum and in the absence of light. Experiments have shown that the products of chemical reactions can be different in an optical cavity and changes to materials properties such as the onset of superconductivity have been predicted. Extreme light confinement can act on systems in the same way as an optical cavity. "The effect works best when light is strongly compressed—the more compressed, the stronger the interaction with the material," says Koppens. Research along these lines may point toward interesting effects on the material properties of the TMD when conditions are met for these 2-D exciton polaritons to form.

More information: Itai Epstein et al. Highly confined in-plane propagating exciton-polaritons on monolayer semiconductors, *2D Materials* (2020). DOI: 10.1088/2053-1583/ab8dd4