

## Giant gate-tunable bandgap renormalization and excitonic effects in a 2-D semiconductor

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Identify the thickness of monolayer ReSe2. (A) The optical image of monolayer ReSe2 (inside the dashed rectangle) transferred on G/h-BN. The inset image is the dark field optical image for the ReSe2 flake. (B) The AFM image of monolayer ReSe2. Inset: the step height of exfoliated ReSe2 flake is measured to be ~0.8  $\pm$  0.1 nm, suggesting a monolayer thickness. Credit: Science Advances, doi: 10.1126/sciadv.aaw2347

Investigating the remarkable <u>excitonic</u> effects in two-dimensional (2-D) semiconductors and controlling their <u>exciton binding energies</u> can unlock the full potential of 2-D materials for future applications in <u>photonic</u> and



optoelectronic devices. In a recent study, Zhizhan Qiu and colleagues at the interdisciplinary departments of chemistry, engineering, advanced 2-D materials, physics and materials science in Singapore, Japan and the U.S. demonstrated large excitonic effects and gate-tunable exciton binding energies in single-layer <u>rhenium diselenide</u> (ReSe<sub>2</sub>) on a backgated graphene device. They used <u>scanning tunneling spectroscopy</u> (STS) and <u>differential reflectance spectroscopy</u> to measure the quasiparticle (QP) electronic and optical bandgap (Eopt) of single-layer ReSe<sub>2</sub> to yield a large exciton binding energy of 520 meV.

The scientists achieved continuous tuning of the electronic bandgap and exciton binding energy of monolayer  $\text{ReSe}_2$  by hundreds of millielectron volts via electrostatic gating. Qiu et al. credited the phenomenon to tunable Coulomb interactions arising from the gate-controlled free carriers in graphene. The new findings are now published on *Science Advances* and will open a new avenue to control bandgap renormalization and exciton binding energies in 2-D semiconductors for a variety of technical applications.

Atomically thin two-dimensional (2-D) semiconductors usually display large <u>bandgap renormalization</u> (shifts in physical qualities) and extraordinary excitonic effects due to <u>quantum confinement</u> and reduced <u>dielectric screening</u>. Light-matter interactions in these systems are governed by enhanced excitonic effects, which physicists have studied to develop <u>exciton-based devices at room temperature</u>. A unique feature of 2-D semiconductors is their unprecedented tunability relative to both electric and <u>optical properties</u> due to <u>doping and environmental</u> <u>screening</u>.





STM images of moiré pattern in monolayer ReSe2/graphene. (A to C) Representative moiré patterns observed in the experiment. (D to F) Calculated moiré patterns obtained from the geometrical analysis.  $\theta$  is the stacking angle between ReSe2 and graphene. Credit: *Science Advances*, doi: 10.1126/sciadv.aaw2347

Researchers can engineer theoretically predicted and experimentally demonstrated Coulomb interactions in 2-D semiconductors to tune the



quasiparticle bandgap (Eg) and exciton binding energies (Eb) of samples, with methods such as <u>chemical doping</u>, <u>electrostatic gating</u> and <u>engineering environmental screening</u>. Among the reported techniques, electrostatic gating offers additional advantages such as continuous tunability and excellent compatibility for integration in modern devices. However, an overlap of the band-edge absorption step with strong excitonic resonances makes it challenging to accurately determine the Eg of 2-D semiconductors from their optical absorption spectrum alone.

Scientists had therefore used scanning tunneling spectroscopy and optical spectroscopy to directly probe the Eb of 2-D semiconductors and measure Eg and the optical bandgap (Eopt). In the present work, Qiu et al. similarly used this approach to demonstrate gate-tunable Eg and excitonic effects in monolayer ReSe<sub>2</sub> on a back-gated graphene field-effect transistor (FET) device. They observed a large Eb of 520 meV for monolayer ReSe<sub>2</sub> at zero gate voltage, followed by continuously tuning from 460 to 680 meV via electrostatic gating due to gate-controlled free carriers in graphene. The ability to precisely tune the bandgap and excitonic effects of 2-D graphene semiconductors will provide a new route to optimize interfacial charge transport or light-harvesting efficiency. Qui et al. expect the present findings to profoundly impact new electronic and optoelectronic devices based on artificially engineered van der Waals heterostructures.

Qui et al. first imaged the monolayer  $\text{ReSe}_2$  to show a distorted 1T structure with <u>triclinic</u> symmetry. The four Re atoms slipped from their regular octahedral sites due to charge decoupling to form a 1D chain-like structure with interconnected diamond-shaped units. Due to the topological features, the monolayer  $\text{ReSe}_2$  exhibited unique in-plane anisotropic electronic and optical properties useful for near-infrared polarization-sensitive optoelectronic applications.





Gate-dependent dI/dV and differential reflectance spectra of a monolayer ReSe2 on graphene. (A) dI/dV spectrum of monolayer ReSe2 (blue line) at Vg = 0 V together with the calculated LDOS (dashed red line). (B) Energy position of VB maximum (VBM; red points) and CB minimum (CBM; dark blue points) as a function of the gate voltage. (C) Gate-dependent dI/dV spectra of the monolayer ReSe2 on graphene/h-BN measured at 4.5 K. As-applied gate voltage is indicated above each STS curve. The VBM and CBM were indicated by light red and light blue points, respectively. (D) Gate-dependent differential reflectance spectra of the monolayer ReSe2 on graphene/h-BN measured at 5 K. The corresponding gate voltage is indicated on the side of each differential reflectance spectrum. Note: The original differential reflectance spectra after background subtraction (circles); fitted curves using the Lorentzian function (solid lines). a.u., arbitrary units. Credit: *Science Advances*, doi: 10.1126/sciadv.aaw2347.



To probe carrier-dependent excitonic effects, the scientists first transferred a monolayer  $\text{ReSe}_2$  flake on to a clean back-gated graphene FET (field effect transistor) device. The device constituted of several components according to a previously established recipe to include a SiO<sub>2</sub> substrate, which contrasted with the constituent <u>atomic flatness of hexagonal boron nitride (hBN)</u> that markedly reduced surface roughness and charge inhomogeneity in graphene. The use of graphene allowed direct scanning tunneling microscopy (STM) measurements of the gated single-layer  $\text{ReSe}_2$  while improving the electrical contact to monolayer  $\text{ReSe}_2$ .

After STM imaging the atomically resolved image revealed a diamond chain-like structure as expected for monolayer  $\text{ReSe}_2$  with a distorted 1Tatomic structure. The scientists observed the stacking alignment of the material along two crystallographic orientations as <u>moiré patterns</u>, where monolayer  $\text{ReSe}_2$  containing a triclinic lattice symmetry lay on graphene with a honeycomb lattice.





Gate-tunable bandgap renormalization and exciton binding energy of monolayer ReSe2 on graphene. (A) A plot of QP bandgap Eg (black points), optical bandgap Eopt (red points), and exciton binding energy Eb (blue points) as a function of gate voltage. Note: The Eopt =  $1.47 \pm 0.01$  eV remains constant when the gate voltage increases from -40 to 40 V. Note: The same Eopt is used for the calculation of Eb at the gate voltage of -63, -60, -50, and +45 V. The solid blue line refers to the theoretically predicted Eb as a function of the gate voltage (refer to section S8 for more details). (B) Illustration of the screening of electron-hole interactions in monolayer ReSe2 by the gate-controlled free carriers in graphene. (C) Schematic illustration of gate-tunable Eg and Eb of monolayer ReSe2 at the gate voltage of -63 and +45 V, respectively. Credit: Science Advances, doi: 10.1126/sciadv.aaw2347.

When they probed the local electronic properties of ReSe<sub>2</sub> using STS (scanning tunneling spectroscopy) the scientists observed differential



conductance (dI/dV) spectra in several moiré regions to exhibit similar features. As a unique feature of the study, Qiu et al. probed the quasiparticle (QP) band structures as a function of gate voltage.

The optical bandgap (Eopt) remained nearly constant at all gate voltages in contrast to the monotonic reduction of Eg, in agreement with <u>previous</u> <u>experimental studies</u>. To verify this, they performed photoluminescence measurements of the monolayer ReSe<sub>2</sub>/graphene/h-BN sample at different gate voltages at room temperature (RT). The gate-dependent photoluminescence spectra revealed a nearly constant Eopt of monolayer ReSe<sub>2</sub>.

The scientists then determined the exciton binding energy and derived a large, gate-tunable bandgap renormalization for ReSe<sub>2</sub> in the hybrid device. They sought the physical origins of the gate-tunable QP bandgap renormalization and exciton binding energy in the monolayer ReSe<sub>2</sub> by excluding contributions from the out-of-plane field-induced polarization wave functions and substantiated their origin from gate-induced free carriers in graphene. Theoretical results of the study also showed that moderate doping in graphene could substantially reduce exciton binding energy (Eb) by hundreds of milli-electron volts as the free-carrier concentration in graphene increased. In addition, Qiu et al. directly compared the theory with their experimental results.

In this way, Zhizhan Qiu and co-workers successfully tailored the QP bandgap and exciton binding energy in a 2-D semiconductor by controlling doping of the underlying graphene with electrostatic gating. The results showed that screening from a graphene substrate had profound impact on Coulomb interactions that lead to broad tunability of the electronic band gap and exciton binding energy. The findings revealed <u>many-electron physics</u> in hybrid 2-D semiconductors or graphene systems. The work will pave the way to control excitonic effects and precisely tune the exciton binding energies in 2-D



semiconductors for a variety of technical applications.



Calculation of Eb in monolayer ReSe2 as a function of the carrier density in graphene substrate. Exciton binding energy (Eb) and Thomas-Fermi screening radius (rs) as a function of electron concentration (n) in graphene. (A) The carrier-dependent Eb for

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