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Momentum-Dependent Oscillator Strength Crossover of Excitons and Plasmons in Two-Dimensional PtSe2

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1 Abstract

The 1T-phase layered PtX2 chalcogenides has attracted widespread interest due to its thickness dependent metal-semiconductor transition driven by strong interlayer coupling. However, its fundamental excitation spectrum remains poorly understood. Here we combine first principles calculations with momentum (q) resolved electron energy loss spectroscopy (q-EELS) to study the collective excitations in 1T-PtSe2 from the monolayer limit to the bulk. Interestingly, the absence of long-range screening in the two-dimensional (2D) limit, inhibits the formation of long wavelength plasmons. Our work unravels the excited state spectrum of layered 1T-PtSe2 and establishes the qualitatively different momentum dependence of excitons and plasmons in 2D materials.

2 Basic electronic properties

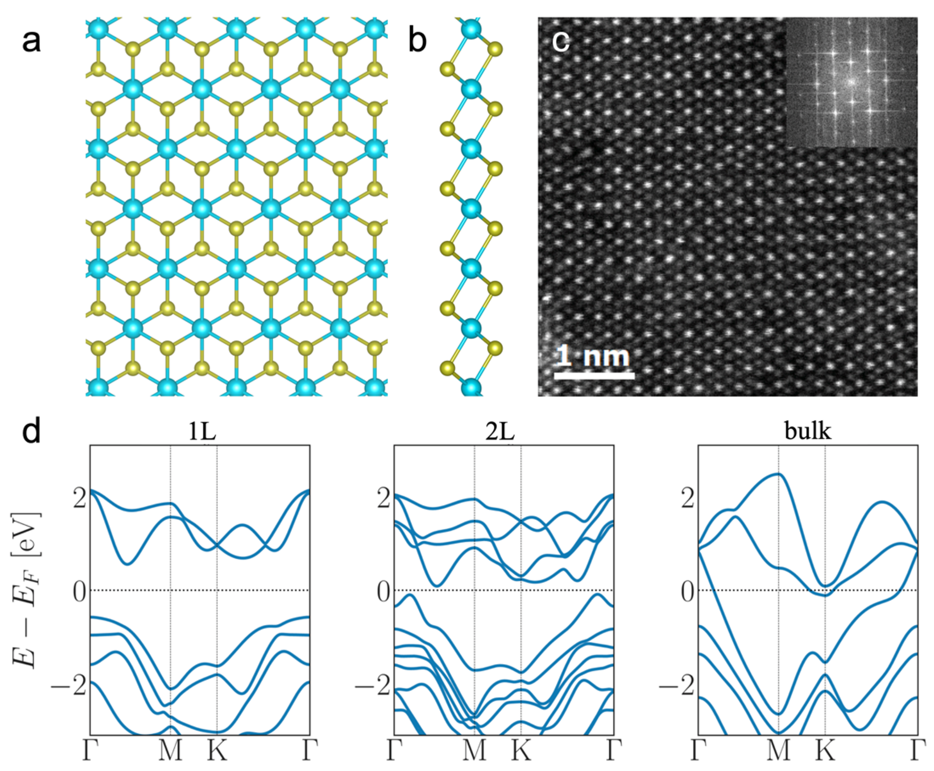

Figure 1: Atomic and electronic structures of PtSe2. (a-b) Top- and side-view structure models of monolayer 1TPtSe2. Cyan balls are Pt and yellow are Se atoms. (c) ADF-STEM image of monolayer PtSe2. (d) Thickness-dependent electronic band structure of PtSe2 in the 2D Brillouin zone. One can clearly see the characteristic insulator to metal transition as the thickness increases.

3 Momentum resolved EELS spectrum

The Coulomb interaction works fundamentally different in 2D and 3D materials due to reduced screening in 2D,

$$v_q^{3D} = \frac{1}{q^2}, \quad v_q^{2D} = \frac{1}{q}$$

This difference leads to the suppression of the exchange contribution to the Coulomb interaction in the optical limit, which results in the absence of plasmon formation in the optical limit in 2D. This leads us to conclude that the B and C peaks are plasmonic in nature.

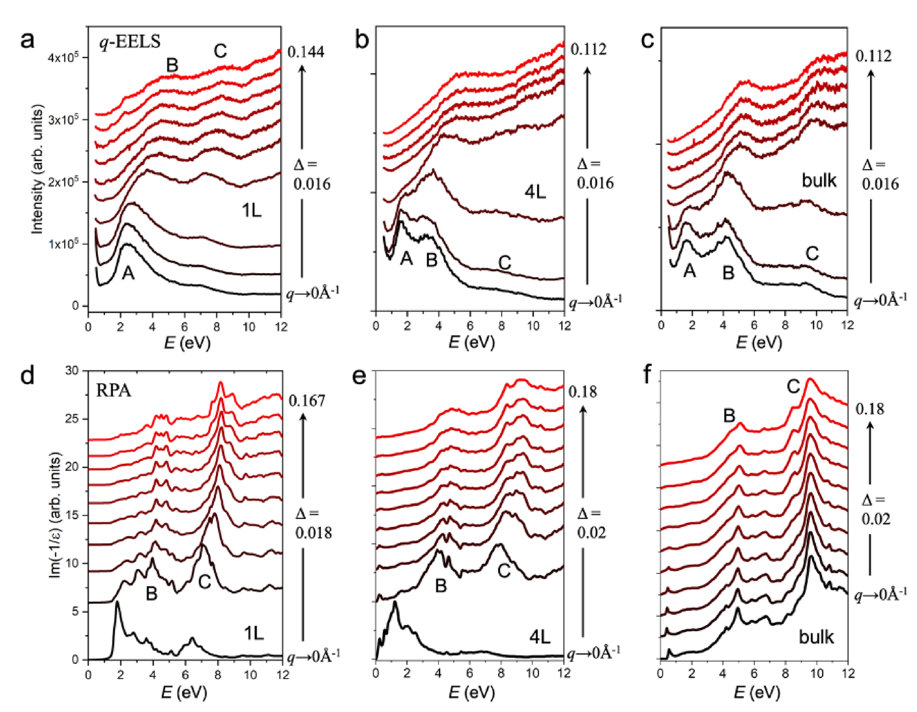

Figure 2: Momentum-dependent loss function of PtSe2. (a-c) Experimental q-EELS of 1L, 4L and bulk PtSe2, respectively. (d-f) Calculated q-dependent loss function in the random phase approximation (RPA) of 1L, 4L and bulk PtSe2, respectively. The low energy excitonic peak A, and the intermediate and high energy plasmonic peaks B and C are indicated.

4 Plasmon formation

We can study the plasmon formation as a function of q via a mode-decomposition of the RPA dielectric function,

$$\epsilon(\boldsymbol{r}, \boldsymbol{r}', \omega) = \sum_n \epsilon_n(\omega)\phi_n(\boldsymbol{r}, \omega)\rho_n(\boldsymbol{r}', \omega),$$
$$\nabla^2\phi_n(\boldsymbol{r}, \omega) = 4\pi\rho_n(\boldsymbol{r}, \omega).$$

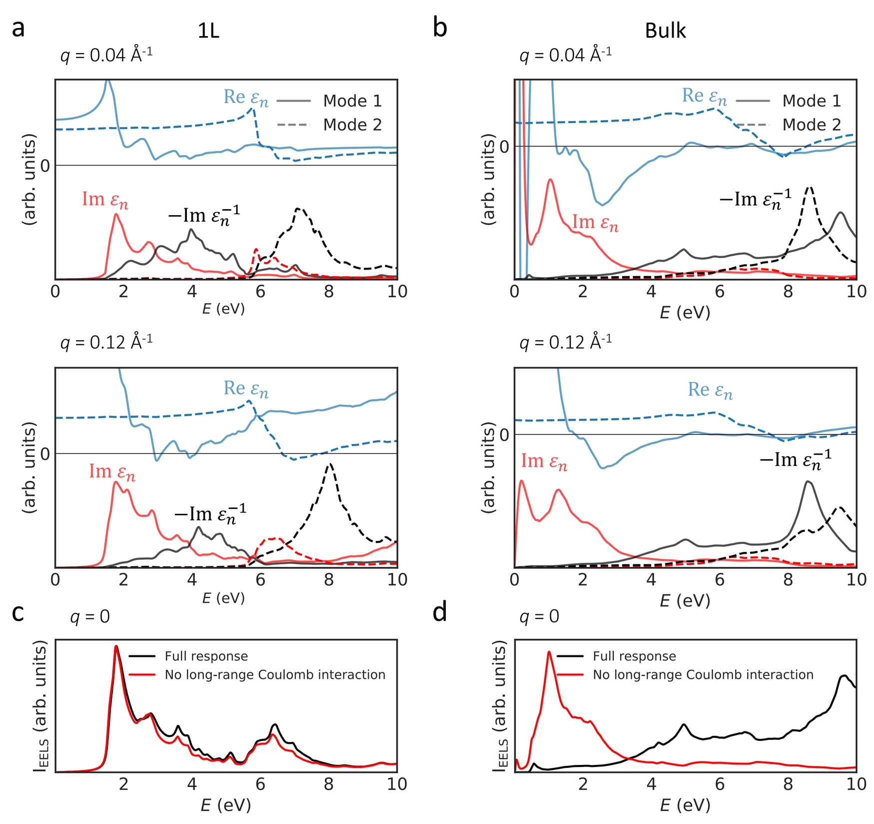

Figure 3: Plasmon formation in 2D vs 3D. (a-b) Evolution of the eigenvalues obtained from the spectral decomposition of the RPA dielectric function for 1L and bulk PtSe2 as a function of q. The loss spectra of the 1L system are more sensitive to q than those of the bulk because of the gradual turning on of the long-range screening in 2D with increasing q. (c-d) The macroscopic RPA loss function in the q=0 limit for 1L and bulk. Inclusion of the long-range component of the Coulomb interaction has essentially no effect on the 1L spectrum while the bulk spectrum is affected dramatically due to the formation of plasmons.

5 Studying the excitonic A-peak with the Bethe-Salpeter Equation

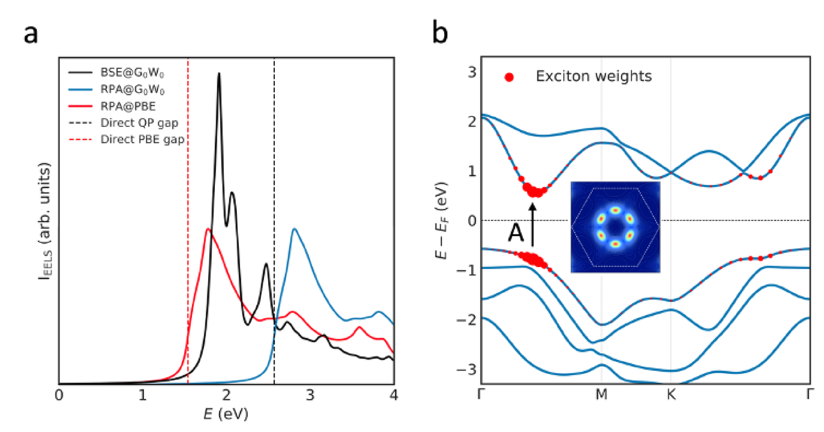

Figure 4: Excitonic origin of the A-peak. (a) Comparison of BSE and RPA calculated loss functions of monolayer PtSe2 in the q→0 limit. The BSE spectrum is calculated on top of the G0W0 band structure. For comparison, the RPA spectrum is calculated on top of the PBE band structure and the PBE band structure corrected to match the G0W0 band gap, respectively. (b) The BSE calculated excitonic weights of the A peak. The colored inset shows the 2D projected exciton wavefunction distribution in k space. Both the excitonic weights and k-space wavefunction show that the A exciton originates from direct transitions at the midpoint of ΓM.

6 Conclusion

We have unraveled the elementary electronic excitations in layered PtSe2 using a combination of q-EELS measurements and theoretical calculations. Further, our work advances the understanding of the connection between dielectric screening and the formation of collective excitations in solids, and establishes the fundamental basis for photonic and optoelectronic applications of low-dimensional PtSe2.