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 wave-length 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

3 Momentum resolved EELS spectrum

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

\[
\begin{align*}
\nu^{3\text{D}} &= \frac{1}{q^2}, \\
\nu^{2\text{D}} &= \frac{1}{q} 
\end{align*}
\]

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.

4 Plasmon formation
We can study the plasmon formation as a function of \(q\) via a mode-decomposition of the RPA dielectric function,

\[
e(r, r', \omega) = \sum_n \varepsilon_n(\omega) \phi_n(r, \omega) \rho_n(r', \omega),
\]

\[
\nabla^2 \phi_n(r, \omega) = 4\pi \rho_n(r, \omega).
\]

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

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.