

Probing Confined Water Monolayer via Excitonic Sensor in Transition Metal Dichalcogenides

Yinan Liu

Adrián Dewambrechies¹, Ayan Halder¹, Sviatoslav Kovalchuk¹, Charlotte Berrezueta¹, Rafael Nadas², Andrey Chaves³, Nikolai Severin², Kirill I. Bolotin¹

¹Freie Universität Berlin, Arnimallee 14, 14195 Berlin, Germany

²Humboldt Universität zu Berlin, Newtonstraße 15, 12489 Berlin, Germany

³Universidade Federal do Ceará

yinan.liu@fu-berlin.de

Water under confinement exhibits phase dynamics, vibrational, and dielectric properties that differ significantly from bulk water [1], yet at the nanoscale and in the single layer limit, it remains challenging to characterize experimentally. In this work, we investigate a monolayer of water intercalated between muscovite mica and 2-dimensional (2D) materials [2], specifically transition metal dichalcogenides (TMDC), which are excellent hosts for excitonic physics. Through tracking the optical response of excitons that are sensitive probes of its surrounding dielectric environment [3], we are able to measure the effective dielectric constant of 2D water from 4K to 295K. Moreover, when a water monolayer is introduced to the TMD-mica interface, there exhibits two characteristic screening strength at cryogenic temperatures and elevated temperatures, hinting at a phase transition like crossover. We further compliment this result with conventional Raman, as well as tip enhanced Raman spectroscopy to probe the hydrogen-bonding networks of confined water monolayer across the temperature range. This study fingerprints the highly coveted experimental characterization of nano-water in the true 2D limit, offering insights into its rich phase and structural properties under confinement.

References

- [1] Kapil, V. et al. *Nature*, 609 (2022) 512-516
- [2] Xu, K. et al. *Science*, 5996 (2010) 1188-1191
- [3] Xu, Y. et al. *Nature*, 581 (2020) 241-218

Figures

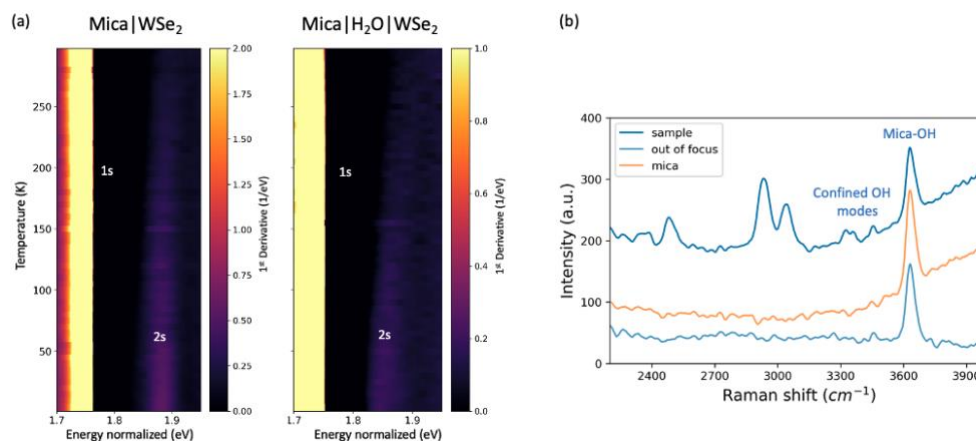


Figure 1: (a) Colormap of differential reflectivity first derivative with the first excited Rydberg state (1s) normalized to its position at 4K on a WSe₂|mica device before the introduction of water (left), and after (right). The non-monotonic change of 1s and 2s spacing across temperature when a proximal monolayer of water is induced indicates at least two distinct dielectric regimes of confined water at the interface. (b) Raman spectra of the Mica|H₂O|WSe₂ device at 4K.