Strain tuning excitons in 2D semiconductors and probing strain in thin films with Raman spectroscopy

We present two novel experimental approaches at the intersection between optical spectroscopies and strain engineering of 2D materials and thin films.

First, we use mechanical strain to characterize and manipulate excitons in 2D semiconductors, tuning their emission energies to bring them into resonance and induce a hybridization between excitonic species. To achieve this experimentally, we develop an electrostatic technique to apply controllable, biaxial strain of up to 3% on suspended 2D membranes while probing their photoluminescence in-situ at cryogenic temperatures. We fingerprint valley character of excitonic states using their strain-induced energy shifts. We exploit their different strain responses to bring various defect excitons into energetic resonance such that they form new hybrid states that inherit the properties of the constituent species. This mechanism allows us to identify multiple previously inaccessible excitons with wavefunctions residing in K, Γ, and Q valleys in the momentum space [1]. Moreover, the hybridization between dark and bright excitons reported here may play a critical role in the operation of single quantum emitters based on WSe_2 [2].

In the second part of the talk we introduce surface-sensitive and bulk-suppressed Raman spectroscopy to reveal and quantify strain in thin films. We realize this using a nanoporous plasmonic gold membrane (PAuM) recently introduced by us [3] that enables optical access to thin materials by increasing the surface-to-bulk Raman signal ratio by three orders of magnitude. This membrane simultaneously blocks the otherwise dominant bulk signal and enhances Raman scattering at the surface of the sample. The irregular slot-shaped nanopores in these easy-to-produce, transferable membranes act as plasmonic antennas and enhance the Raman response of the surface or thin film underneath. We show that 90 % of the Raman enhancement occurs within the top 2.5 nm of the material, proving surface sensitivity. We use this approach to quantify strain in a 12.5 nm thin Silicon quantum well layer used in quantum computing technologies [4].

[1] A. Kumar, P.H.L. et al., accepted at Nat. Comms. (2024)

[2] P. Hernández López et al. Nat. Comms. 13, 7691 (2022)

[3] R. Wyss et al. ACS Applied Materials & Interfaces, 14(14), 16558-16567 (2022)

[4] R. Wyss et al. Nat. Comms. 15, 5236 (2024)