Influence of defects, strain, and charged impurities on excitons in natural and synthetic MoS_2 sources

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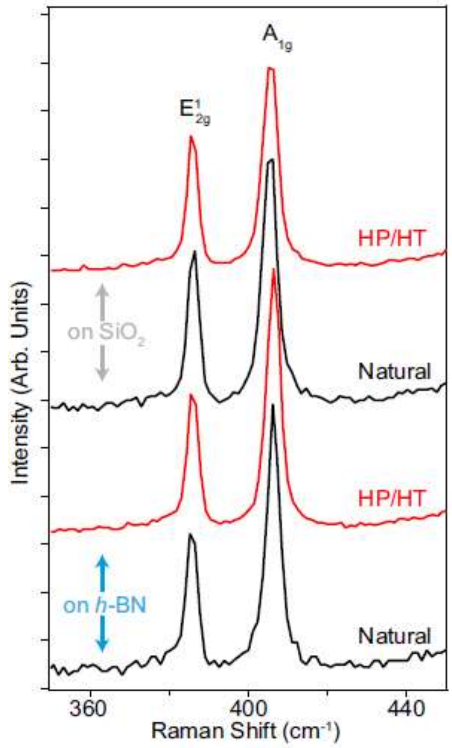
Abstract
Few- and single-layer MoS_2 host substantial densities of defects. A chemical treatment eliminating defects has allowed to demonstrate a photoluminescence quantum yield close to unity and long lifetimes [1]. Nevertheless, the nature of defects in non-treated samples and their role in radiative and non-radiative recombination remain as open questions. They are thought to influence the doping level, the crystal structure, and the binding of electron-hole pairs. We disentangle the concomitant spectroscopic expression of all three effects, and identify to which extent they are intrinsic to the material or extrinsic to it, i.e. related to its local environment. We do so by using different sources of MoS_2 – a natural one and a novel one prepared at high pressure and high temperature(HP/HT) – and different substrates bringing varying amounts of charged impurities, and by separating the contributions of internal strain and doping in Raman spectra (Fig.1). Photoluminescence unveils various optically-active excitonic complexes. We discover a defect-bound state having a low binding energy of 20 meV, that does not appear sensitive to strain and doping, unlike charged excitons. Conversely, the defect does not significantly dope or strain MoS_2. Scanning tunneling microscopy and density functional theory simulations point to substitutional atoms, presumably individual nitrogen atoms at the sulfur site.
Our work shows the way to a systematic understanding of the effect of external and internal fields on the optical properties of two-dimensional materials [2].

Figures



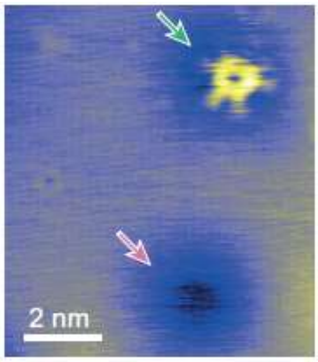


Figure 1: (Top)Raman spectra (532 nm-wavelength laser) for MoS2 single-layers exfoliated from a natural crystal (black) and from a HP/HT source (red), on SiO2 and *h*-BN. (Bottom) STM topograph close-up view on new kind of defects(green) in HP/HT source.

References

[1] M. Amani *et al*. *Science*, 350 (2015), pp. 1065–1068.
[2] S. Dubey *et al*., *ACS Nano, 11.11 (2017): 11206-11216.*