Efficient light generation in TMDC with high spatial resolution using Cathodoluminescence

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Probing disorder in VdW heterostructures



Intrinsic disorder



STM image of synthetic MoS₂ (Dubey et al, ACS Nano, 2017)



Extrinsic disorder (interfaces)



AFM image of BN encapsulated MoS₂ (showing bubbles, cracks and wrinkles)

TEM image of graphene on hBN (Kretinin et al Nano Lett., 2014)

How do extrinsic sources of disorder affect the optoelectronic properties of TMD monolayer heterostructures ?

Interlayer coupling affecting PL



Fang, Hui, et al. PNAS 2014

Can we go below the diffraction limit in probing disorder by luminescence ?

What happens when the bubbles that are as small as 100 to 200nm??

A relevant Probe : Cathodoluminescence (CL)





Electron beam excitation

High spatial resolution ~ 10nm.

A relevant Probe : Cathodoluminescence





Electron beam excitation

High spatial resolution ~ 10nm.

Deterministic transfer method



Manual (micromanipulated) stamping





e/h recombination requires no phonons! (direct band-gap)



Castellanos – gomez et al. 2D Mater. 2014

Cathodoluminescence with charge transfer



h-BN is playing two roles : protection, buffer layer AND Generation of e/h pairs (excitons)

Good interlayer coupling mandatory!!

Nayak et al in preparation (Zheng et al Nanoletters 2017)

CL mapping of the Heterostructure



CL spectrum is sharper because the probing area is reduced and not affected by convolution with defective area

Why CL is appearing only in selective subregions of the heterostructure ?

Photoluminescence mapping



PL Intensity 10µm

PL map before cathodoluminescence

PL map after cathodoluminescence

Interestingly the PL is quenched after SEM e-beam irradiation exactly at the zones where CL was not seen.

So CL is invasive only at specific (dirty) places only



SEM has induced further irreversible defects in the "dirty" uncoupled areas

Are these new defects intrinsic or extrinsic to MoS2 ?

To explore, let us perform Raman spectroscopy mappings

Raman mapping before CL



Clear correlation between coupling and doping.

Dirty areas experience high doping by the contaminants





Defects in PL quenched region

Broadening of Raman peaks is linked to structural defects (Mignuzzi et al PRB 2015)



Why defects are generated by SEM in uncoupled region?

- Threshold for knock-on > 10keV >> 1 keV
- Poor contacts $MoS_2/h-BN \rightarrow$ water vapour/air
- Volmer reaction is promoted by electron in aqueous environment

 \rightarrow possible adsorption of Hydrogen on MoS₂

Xie, Junfeng, et al. Advanced materials 2013

Chemical reaction in uncoupled regions

Conclusions and Outlook

- Interlayer coupling is very important in heterostructures to harness the intrinsic property of the 2D material.
- Efficient light generation under e-beam irradiation in a VdW heterostructures with high spatial resolution (beyond diffraction limit) and also to probe disorder at nm scale level.
- Heterostructures using pickup techniques based on VdW attraction gives good homogeneous electronic coupling.

Refs:

S.Dubey et al ACS Nano (2017)G. Nayak et al, in preparationT. Jakubczyk et al, in preparation



Thanks for your attention.

