Ultrafast optical and terahertz properties of

1D van der Waals heterostructures

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Concept: ID Van der Waals heterostructure











MAIN QUESTIONS

Questions addressed:

- 1. Are 1D different from 2D heterostructures? Do noncarbon nanotubes have a unique band structure?
- 2. Do they show curvature induced effects in comparison to 2D materials?
- 3. Does properties localized along the nanotubes?
- 4. Does 1D show individual properties of each nanotubes or combined (coupled properties) of a whole heterostructure?
- 5. What is a distribution of free charges and excitons in the system?

Microscopy: ID heterostructure



Transmission electron microscopy

Scanning transmission electron microscopy (STEM) confirmed the ID nature of the radial van der Waals heterostructures.



Electron Energy Loss Spectroscopy

C, C@BN and C@BN@MoS2established the CNT bundles to have mean diameter 7.9 nm, increasing to 11.1 nm afterBN overgrowth (average 4-5 walls of BN), and 14.1 nm after MoS2growth (2-3 walls),

Absorption: ID heterostructure



Absorption



The heterostructure consists of CNT, BNNT and MoS2NTs and hence can be viewed as exhibiting the combined optical absorption of each constituent, as long as each additional layer does not alter the optoelectronic properties of each encapsulated structure.

Raman spectroscopy: ID heterostructure







BN acts as a good tunnel barrier even for a monolayer, and hence reduces intertube conductivity, while the intratube (plasmonic) conductivity remains comparable.

The extra conductivity added by MoS2 NT may be associated with free charges in the outer MoS2NT if the as-grown material is doped: indeed MoS_2 monolayers are often n-doped as a result of S-vacancies. Alternatively, the MoS_2NTs may have altered the conductivity of the encapsulated CNTs, either as a result of a strain-induced change in their band structure, or by a small but finite charge transfer from the outer MoS2NTs to the inner CNTs.

OPTP: Excitons and free chargers



$$\sigma_{ph}d = \sigma_{before}d/(1 - i\omega\tau_{before}) - \sigma_{after}d/(1 - i\omega\tau_{after})$$





OPOP: Excitons and free chargers



Pump - probe delay time (ps)

Optical pump - optical probe

The free charges are observed to have a significantly distinguished signature from excitons, as expected from the large exciton binding energy of this material. While excitons trapped on a picosecond time scale free chargers are long lived. Many-body interactions between free charges are evident at high photoexcited carrier densities, resulting in bandgap renormalization effects.

3

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