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MoS₂/CNT Core-Shell Nanocomposites for Enhanced Nonlinear Optical Performances

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Background





- Molecular-scale thickness;
- Indirect-to-direct bandgap transition as the decreasing of the number of monolayer;
- Ultrafast carrier dynamics (intraband ~fs, interband~ps);
- Sizable and layer-dependent bandgap, typically in the 1-2 eV;
- Fairly good photoluminence and electroluminescence properties;
- High optical gain, e.g., 2D nanolasers;
- Remarkable exciton effects, i.e., high binding energy, large oscillator strength and long lifetime.

Background



Carbon nanotubes (CNTs) with highly delocalized π -electrons have attracted great interests with superior electric and optical properties owning to their unique structure.

MoS₂/CNT nanocomposites



electrocatalysts for hydrogen evolution Youn, et al., ACS Nano, 8, 5164 (2014)



electrocatalysts for hydrogen evolution Zhang, et al., ACS Appl. Mater. Interfaces 7, 12193 (2015)



CNT–MoS₂ composite as anode material in lithium ion batteries Chem. Commun., 50, 3338 (2014).



What is the nonlinear optical performances (NLO) of MoS₂/CNTs heterostructures?

How does the charge transfer between MoS₂ and CNTs affect the NLO performances of the nanocomposites?

What is the direction of charge transfer in MoS₂/CNTs heterostructures?





Background 1



2 Synthesis and Characterization





Synthesis method:

solvothermal method (NMP as solvent)



Pure CNTs

MoS₂/CNTs nanocomposites

Zhang et al. Chem. Eur. J., 2017, 23, 3321

TEM and EDS results

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Core-shell structured MoS₂/CNTs nanocomposites

Zhang et al. Chem. Eur. J., 2017, 23, 3321

Raman and absorption results



Zhang et al. Chem. Eur. J., 2017, 23, 3321













Nonlinear optical measurements: open-aperture Z-scan technology

Set-up: PD0 Samp

 Laser
 PD1

 Beam Splitter
 Lens
 $-Z \leftrightarrow +Z$ Detector

Dispersions and PVA polymers







Dispersions prepared by LPE method

PVA films

Zhang et al. Chem. Eur. J., 2017, 23, 3321

NLO, fs, 1030 nm



Zhang et al. Chem. Eur. J., 2017, 23, 3321

NLO, fs, Polymer films

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NLO parameters of the samples measured using the Z-scan technique for fs at 1030 nm. [a]= NMP dispersions; [b]= PVA films.

Sample	T [%]	α ₀ [cm ⁻¹]	NLO response	β _{eff} [cm GW ⁻¹]	<i>I_{sat}</i> [GW cm ⁻²]
CNTs ^[a]	97	0.30	SA+OL	(0.99±0.15)×10 ⁻²	11.0±2.1
MoS ₂ -L/ CNTs ^[a]	96	0.40	SA+OL	(0.93±0.17)×10 ⁻²	15.8±3.5
CNTs ^[a]	63	4.62	SA+OL	(5.50±0.23)×10 ⁻²	48.2±4.9
MoS ₂ -L/ CNTs ^[a]	62	4.78	SA+OL	(6.25±0.26)×10 ⁻²	31.8±4.2
CNTs ^[b]	80	26.3	SA+OL	(25.5±3.5)×10 ⁻²	58.3±5.1
MoS ₂ -L/ CNTs ^[b]	80	28.3	SA+OL	(30.9±4.5)×10 ⁻²	52.1±4.9

NLO, ns, Dispersions



Normalized transmission as functions of input laser fluence for MoS_2 , CNTs, and MoS_2 /CNTs dispersions with the transmission of ~80% for ns, respectively.

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Nonlinear extinctive coefficient \beta_{\text{eff}}: ~ 33.9 for 532 nm,
~ 7.2 for 1064 nm;
Imaginary third-order susceptibility Im\chi^{(3)}: ~ 2.7 for 532 nm,
~ 3.5 for 1064 nm.
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Zhang et al. Chem. Eur. J., 2017, 23, 3321



NLO coefficients of the nanocomposites dispersions in NMP for ns at 532 nm and 1064 nm.

Sample	λ [nm]	Ι ₀ [μJ]	T [%]	α ₀ [cm ⁻¹]	β _{eff} [cm GW ⁻¹]	$Im\chi^{(3)}$ [× 10 ⁻¹³ , esu]
MoS ₂	532	200	74	0.30	0.045 ± 0.02	0.16 ± 0.07
MoS ₂ -L/CNTs			80	0.22	1.51±0.2	5.49±0.73
MoS ₂ -H/CNTs			81	0.21	1.31 ± 0.1	4.76±0.36
CNTs			78	0.25	0.56±0.02	2.03±0.07
MoS_2	1064	400	82	0.20	0.054 ± 0.01	0.39 ± 0.07
MoS ₂ -L/CNTs			81	0.21	0.39 ± 0.03	2.83 ± 0.22
MoS ₂ -H/CNTs			80	0.22	0.27 ± 0.02	1.96 ± 0.15
CNTs			80	0.22	0.11 ± 0.01	0.80 ± 0.07













- Few-layer MoS₂ and CNTs core-shell heterostuctures were successfully synthesized.
- Higher SA performance than nanotubes alone under fs pulses at 1030 nm were observed.
- ➢ For ns pulses, improved OL performances were revealed relative to those of single MoS₂ and CNTs.
- Photo induced electron transfer and the effective coupling between CNTs and MoS₂ contribute to the enhanced nonlinear modulation behavior towards different temporal pulses.

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