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Support information

A novel sensitive ACNTs-MoO2 SERS substrate boosted by synergistic enhancement effect

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The static adsorption experiments of R6G

The samples of the same quality were mixed with 4 mL R6G solution (10^{-5} M), then let it stand for 30 seconds after ultrasonic dispersion, and took the upper liquid after centrifugation.

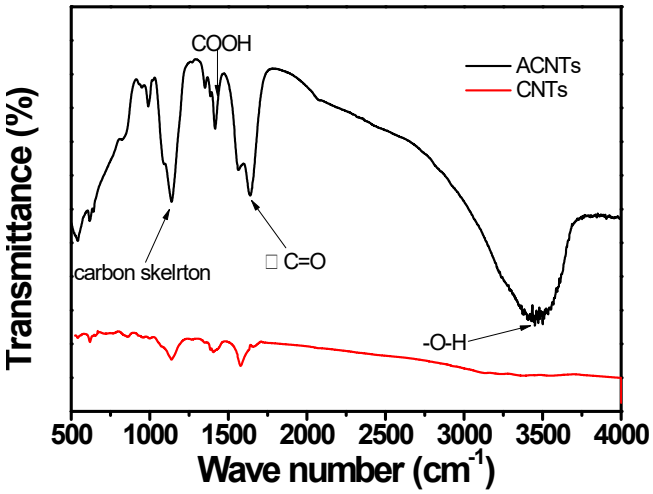

Fig. S1 FT-IR spectra of CNTs and ACNTs.

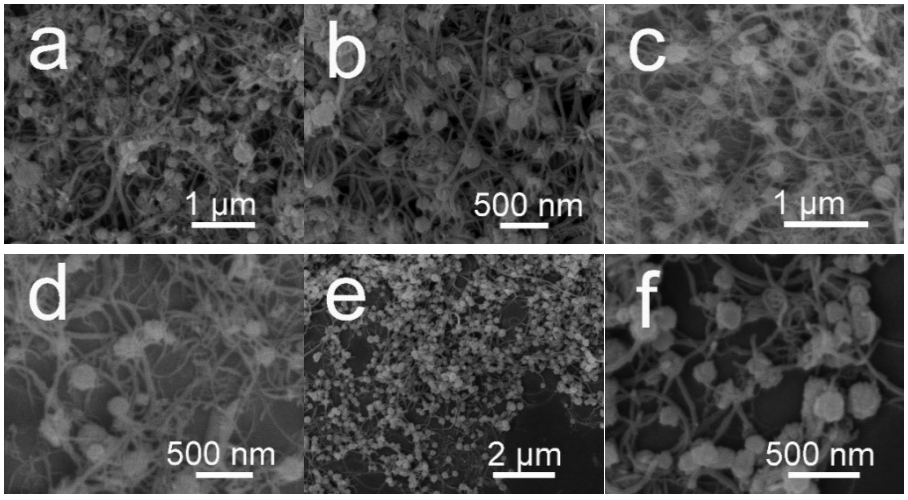

Fig. S2 SEM images of (a-b) ACNT-M0.75 (without addition of CTAB in raw materials), (c-d) ACNT-M0.375 and (e-f) ACNT-M1.5.

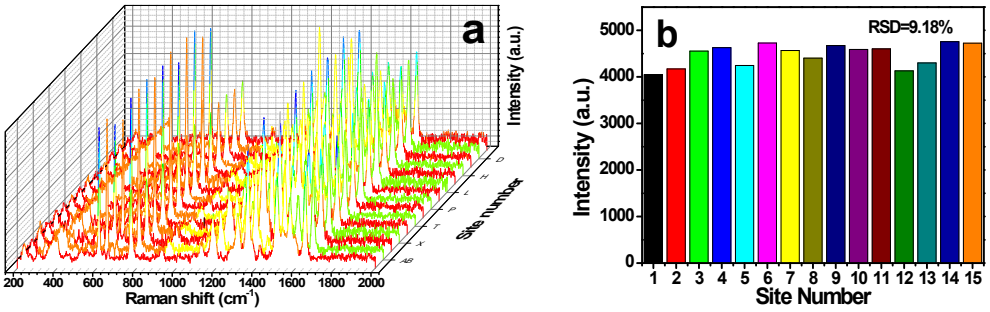

Fig. S3 (a) SERS spectra of R6G collected randomly from 15 spots of ACNTs-M0.75 substrate, (b) SERS intensity distribution at 612 cm^{-1} corresponding to part (a).

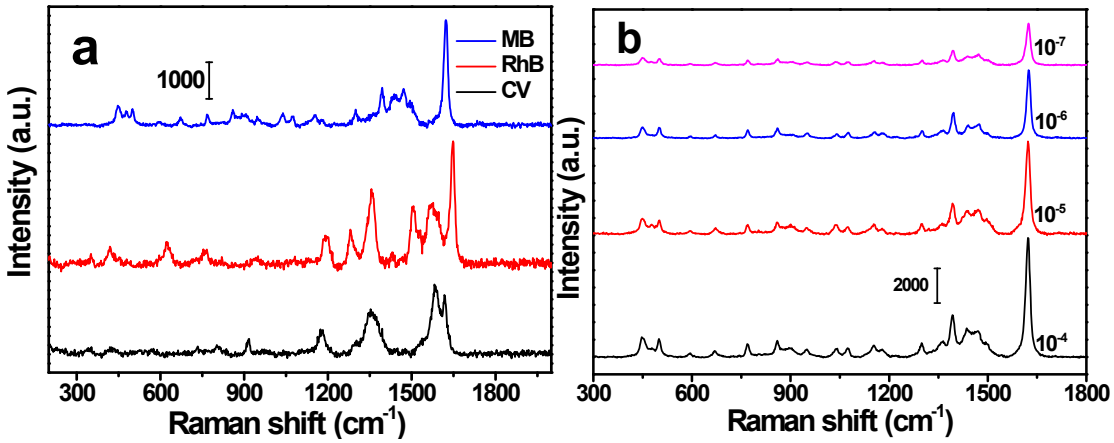

Fig. S4 (a) SERS spectra of a series of organic dyestuff with concentration of 10^{-6} M (MB, RhB and CV), (b) Raman spectra of MB with different concentrations on ACNTs-M0.5.

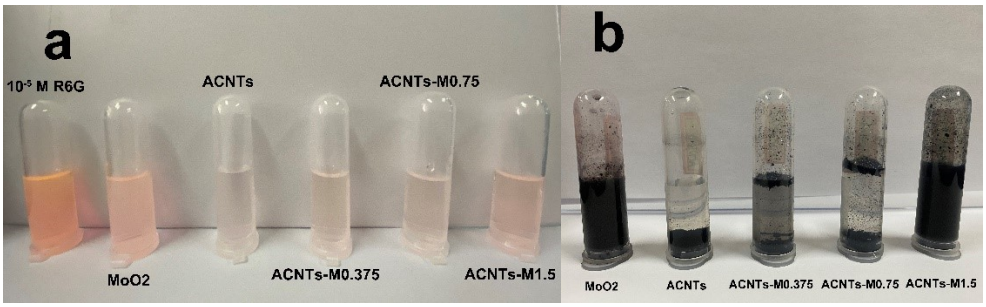

Fig. S5 the adsorption performance of R6G of different samples, (a) the upper layer of liquid after centrifugation after mixing for 30 seconds and (b) static settlement for 5 minutes.

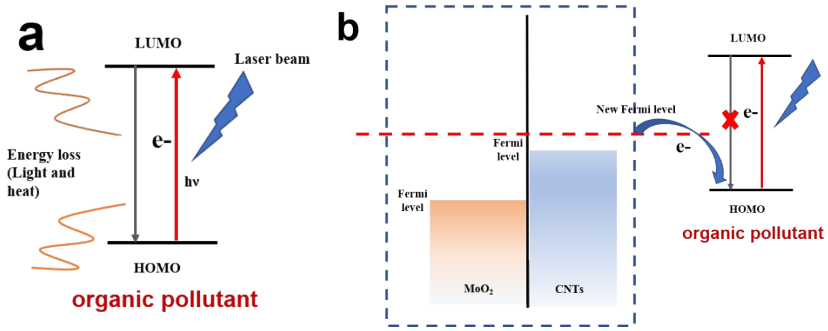

Fig. S6 Schematic illustration of (a) electron transition under excitation conditions, and (b) charge transfer between the molecule and the substrate.

Conductivity is a macroscopic concept related to free electrons in the metal. According to the DFT calculation results, the ACNTs-M0.75 presents a higher DOS density, which indicates that the composite system possesses a better conductivity. According to previous reports, the Fermi level of metal oxides and CNTs align themselves to have a new single Fermi level[1, 2]. When there is no SERS substrate, the fallback of excited state electrons will cause energy loss and thus cannot obtain obvious Raman signals. As shown in Fig.S4b, when organic molecules are adsorbed on the substrate surface, charge transfer can occur between the new Fermi level and the electron orbitals of molecules. The good electrical conductivity of the system promotes the charge transfer between the molecule and the substrate, thus enhances the Raman signals of organic molecules.

Table S1 The calculation results of I_D/I_G of different samples.

	CNT	ACNT	ACNT-$M_{0.25}$	ACNT-$M_{0.5}$	ACNT-M_1
I_D	1091.08	1258.69	1630.91	757.81	2358.61
I_G	1475.16	1678.44	2450.04	1138.88	3534.07
I_D/I_G	0.740	0.750	0.666	0.665	0.667

Table S2 Comparison of the SERS performance based on carbon materials and MoO_2 in the literature.

Sample	Probe molecule	LOD	EF	Excitation wavelength	Reference
Oxide graphene	R6G	1×10^{-5} M	10^4	514 nm	3[3]
Ag nanocubes/GO	4-chlorobiphenyl	1×10^{-6} M	3.9×10^8	532 nm	4[4]
N doped graphene quantum dots	RhB	1×10^{-10} M	-	514 nm	5[5]
Porous carbon nanowire	R6G	1×10^{-6} M -	1×10^6	532/785 nm	6[6]
CNT	Cd^{2+}	1 ppd	G=10	514 nm	7[7]
CNT/AgNPs	R6G	1×10^{-15} M	5.35×10^{11}-	532 nm	8[8]
MoO_2/GO	**R6G**	**1×10^{-9} M**	**1.05×10^7**	**532 nm**	**Our previous work[9]**
ACNTs/MoO_2	**R6G**	**1×10^{-10} M**	**5.13×10^7**	**532 nm**	**This work**

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