Supplementary Information (SI) for Journal of Materials Chemistry C.

Supporting Information

Ultrasensitive SERS Detection Using UV-Ozone Treated Nb_2O_{5-x} Nanosheets Coupled with Plasmonic Nanoparticles: An Integrated Experimental and Theoretical Study

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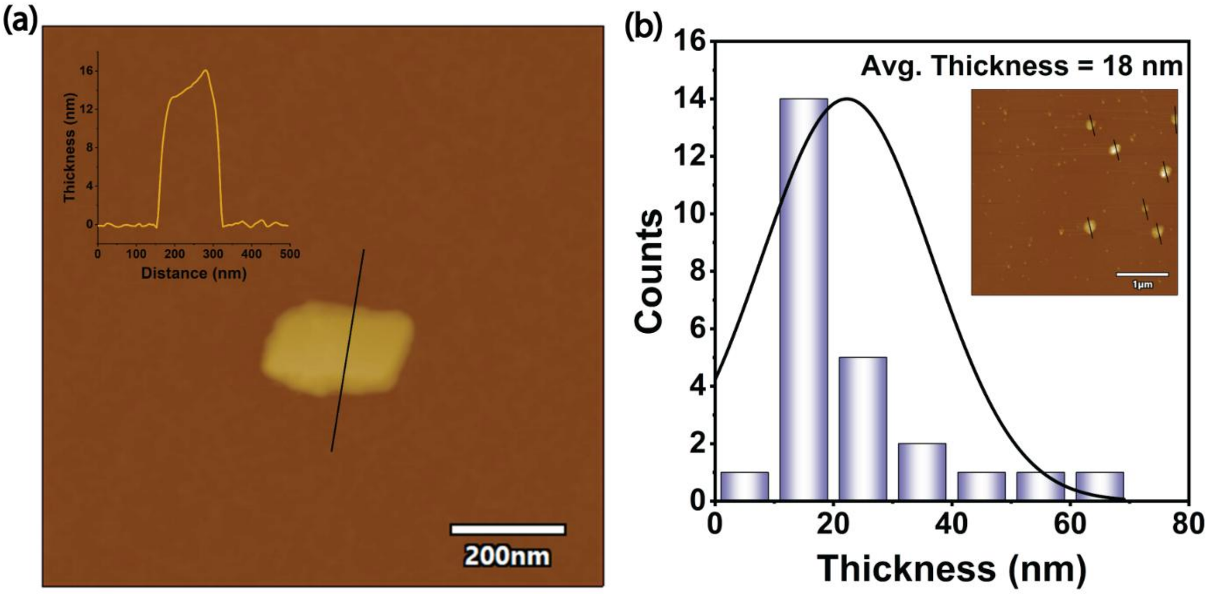
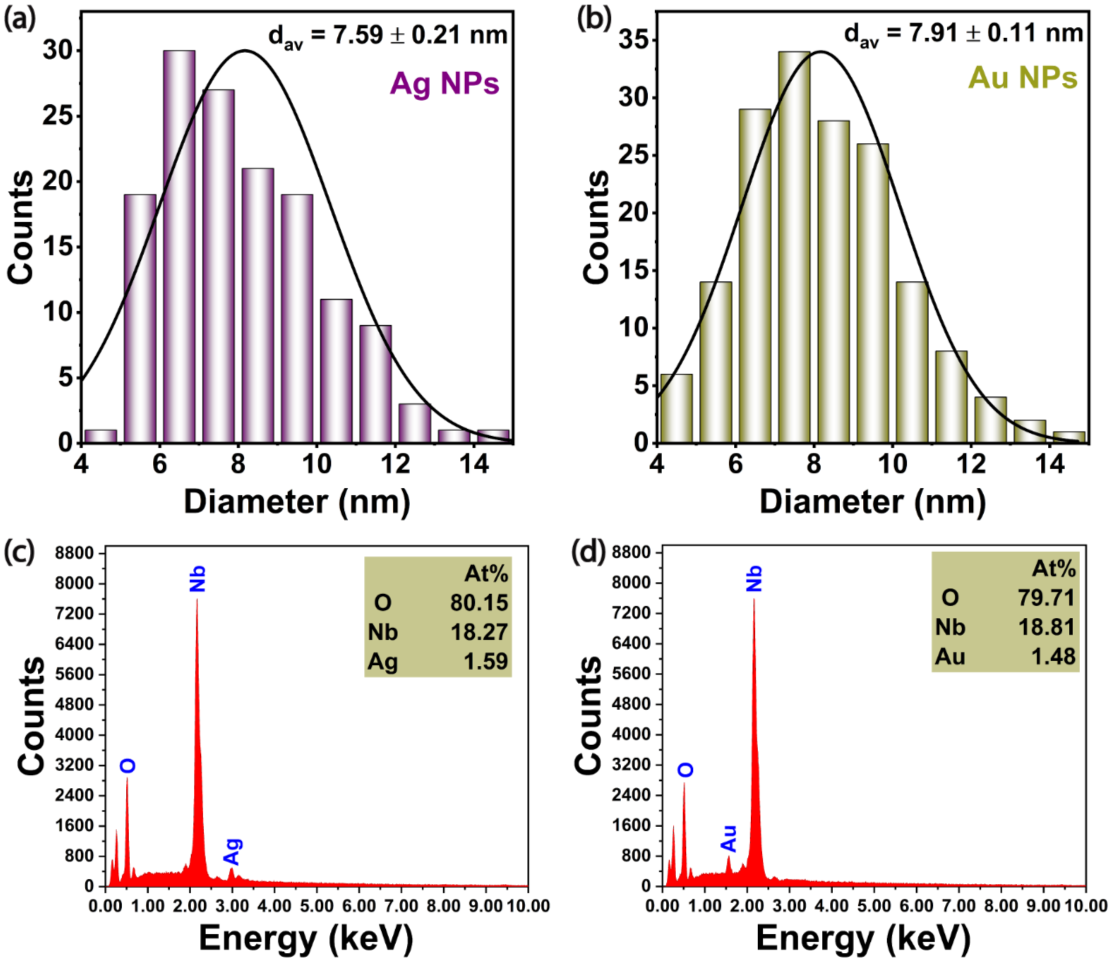


Fig. S1 (a) AFM image of an individual Nb_2O_5 nanosheet showing a thickness of approximately 16 nm. (b) Thickness distribution of multiple nanosheets, indicating an average thickness of about 18 nm. The inset shows an optical micrograph depicting the distribution of several Nb_2O_5 nanosheets obtained from a selected region of the substrate.



	At%
O	80.15
Nb	18.27
Ag	1.59

	At%
O	79.71
Nb	18.81
Au	1.48

Fig. S2: Size distribution of the (a) Ag NPs decorated on the Nb_2O_5 nanosheets, and (b) Au NPs decorated on the Nb_2O_5 nanosheets. EDX spectra showing the elemental composition acquired from the surface of (c) Ag-NbNS, and (d) Au-NbNS.

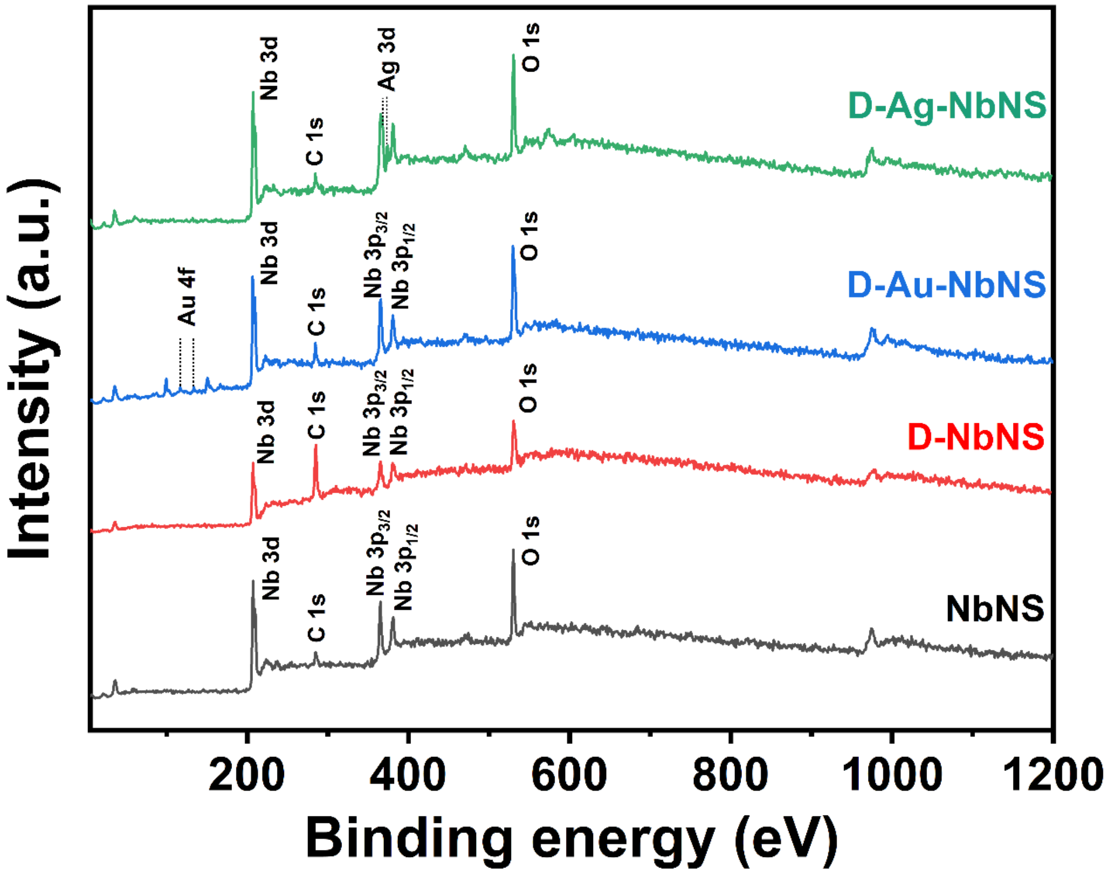


Fig. S3: The XPS survey spectra of NbNS, D-NbNS, D-Au-NbNS, and D-Ag-NbNS.

S1. Calculation of SERS EF

Calculation of SERS EF: The SERS enhancement factor (EF) was calculated using the widely adopted expression

$$EF = \frac{I_{SERS}}{N_{SERS}} \times \frac{N_{Bulk}}{I_{Bulk}} \tag{1}$$

where I_{SERS} is the maximum Raman intensity of the probe molecule adsorbed on the SERS substrate, and I_{Bulk} is the maximum Raman intensity of the same molecule measured without the substrate. N_{SERS} and N_{Bulk} denote the number of probe molecules within the laser-illuminated volume for the SERS and bulk measurements, respectively. These two quantities are critical for an accurate evaluation of EF, as they depend on several experimentally measured parameters.

The number of molecules contributing to the SERS and bulk signals can be expressed as:

$$N_{SERS} = CVN_A A_{Raman}/A_{Sub} \tag{2}$$

$$N_{Bulk} = \rho h A_{Raman} N_A/M \tag{3}$$

Here, C is the molar concentration of the dye, V is the drop-cast solution volume, and N_A is Avogadro's number. A_{Raman} is the laser-illuminated area, while A_{Sub} is the effective substrate area estimated from the circular spot formed after solvent evaporation. M and ρ are the molecular weight and density of the dye molecules, respectively. The confocal depth of the excitation laser, h, is determined from the laser spot diameter W_0:

$$W_0 = \frac{1.22\lambda}{NA} \tag{4}$$

$$h = \left(\frac{2\pi}{\lambda}\right) W_0^2 \tag{5}$$

where λ is the excitation wavelength, and NA is the numerical aperture of the objective lens.

In this study, SERS spectra were recorded using an Ar+ laser with excitation wavelengths of 488 nm and 514 nm, employing a 100x objective lens (Olympus, NA = 0.90). The corresponding laser spot diameters were calculated to be 661.5 nm and 697.43 nm, yielding confocal depths of 5.63 μm and 5.94 μm, respectively.

By combining Eqs. (2) and (3), the ratio of bulk to SERS molecules is obtained as:

$$\frac{N_{Bulk}}{N_{SERS}} = \frac{\rho h A_{subs}}{MCV} \qquad (6)$$

For Malachite Green (MG) molecules with molar mass M = 364.91 $g.mol^{-1}$ and density ρ=1.03 $g.cm^{-3}$, while for ciprofloxacin the molar mass M = 331.34 $g.mol^{-1}$.

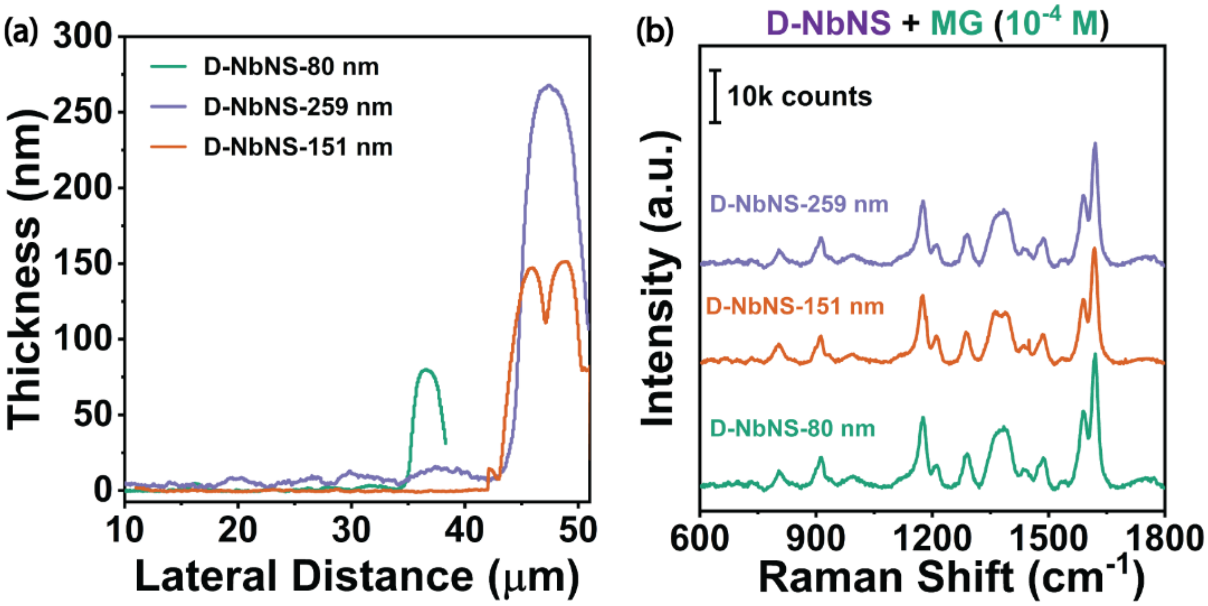


Fig. S4: (a) Profilometer measurements showing the thickness of D-NbNS layers obtained at different spin-coating conditions. (b) Corresponding SERS spectra of MG (10^{-4} M) recorded from D-NbNS substrates with varying thicknesses.

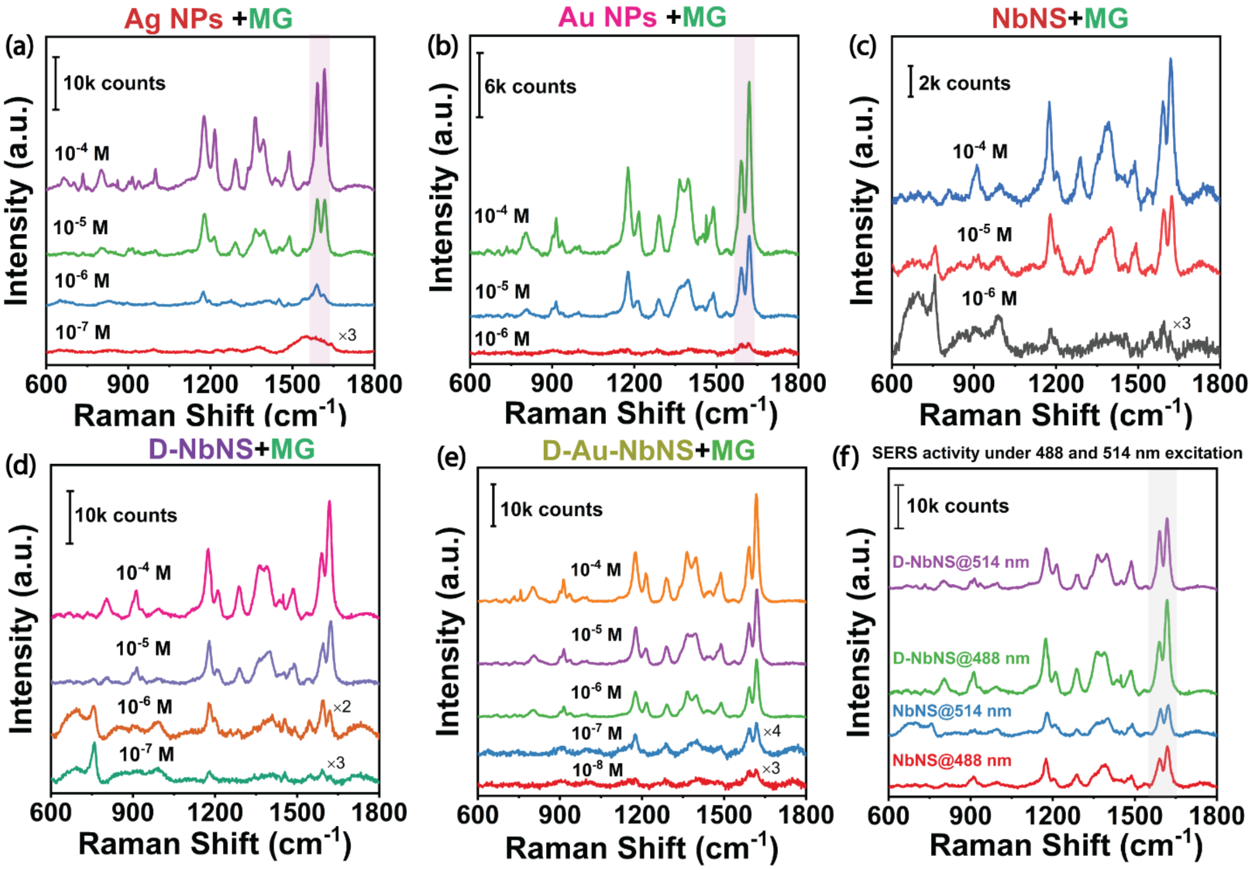


Fig. S5: Concentration-dependent SERS spectra of: (a) Ag NPs acquired under 488 nm excitation, (b) Au NPs under 514 nm excitation, (c) NbNS and (d) D-NbNS recorded with 488 nm excitation, and (e) D-Au-NbNS measured under 514 nm excitation. (f) Difference in SERS intensity from NbNS and D-NbNS substrates under different laser excitations (488 nm and 514 nm).

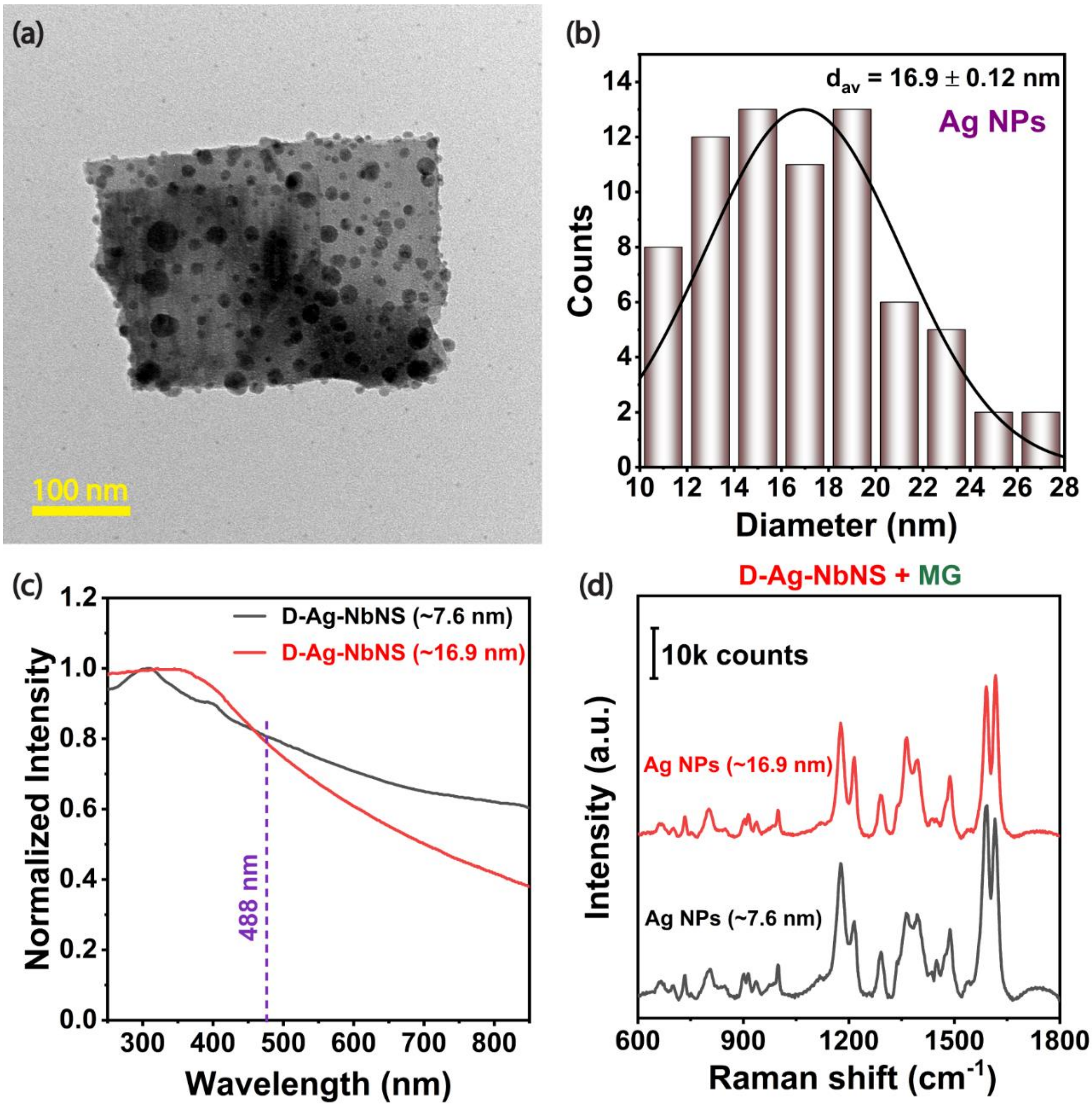


Fig. S6. (a) FETEM image of an Nb_2O_5 nanosheet decorated with Ag NPs, showing a uniform distribution of nanoparticles with an average size of ~16.9 nm. (b) Statistical size distribution of the synthesized larger Ag NPs. (c) UV-Vis absorption spectra of the two hybrid substrates (D-Ag-NbNS) containing Ag NPs of two different sizes. (d) Comparison of the corresponding SERS spectra obtained from these substrates.

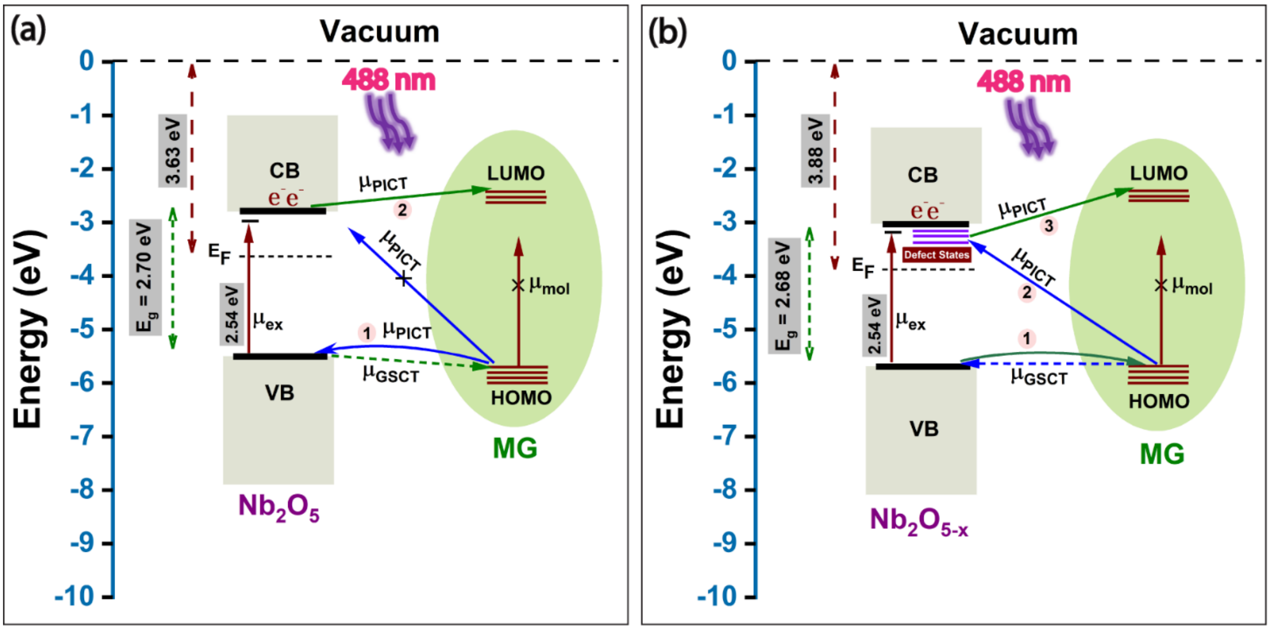


Fig. S7: Schematic illustration of the energy band alignment and potential charge-transfer pathways in (a) the NbNS-MG system and (b) the D-NbNS-MG system under 488 nm. excitation.

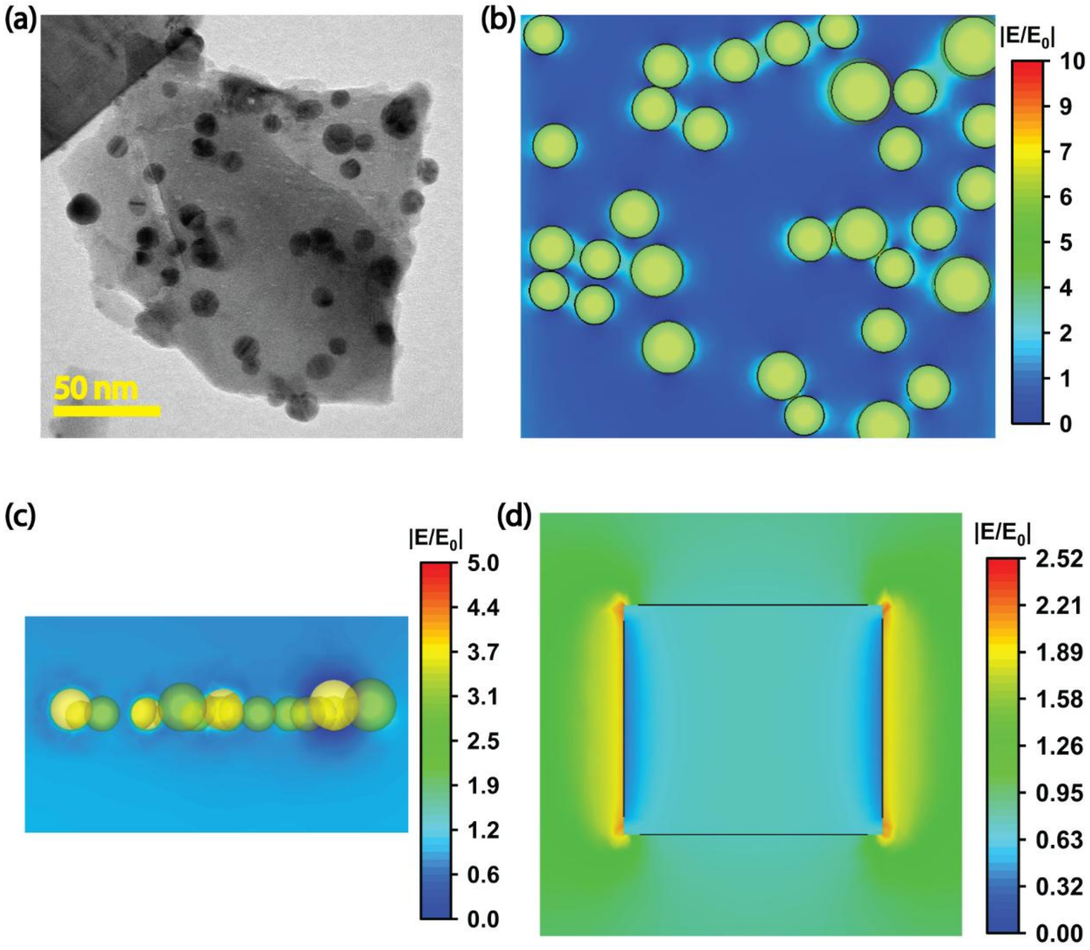


Fig. S8: (a) FETEM image of Ag NP-decorated Nb_2O_5 nanosheet. (b) FEM simulation of E-field enhancement in Au nanoparticle-decorated Nb_2O_5 nanosheet. (c) Electric field (E) distribution in the yz-plane for bare Ag NPs. (d) Mie scattering-induced E-field enhancement in a bare Nb_2O_5 nanosheet.