Visualizing Ultrasmall Silica-CTAB Hybrid Nanoparticle for Generating High Photoluminescence

Yongju He[1], Chuan-Cun Shu[2*], Yu Guo[3] , Mengqiu Long[2*] , Hui Xu[2]

[1] School of Material Science and Engineering, Central South University, Changsha, Hunan 410083, China

[2] Institute of Super-microstructure and Ultrafast Process in Advanced Materials, School of Physics and Electronics, Central South University, Changsha, Hunan 410083, China

[3] Hunan Provincial Key Laboratory of Flexible Electronic Materials Genome Engineering, School of Physics and Electronic Science, Changsha University of Science and Technology, Changsha 410114, China

Corresponding authors (∗):cc.shu@csu.edu.cn&mqlong@csu.edu.cn

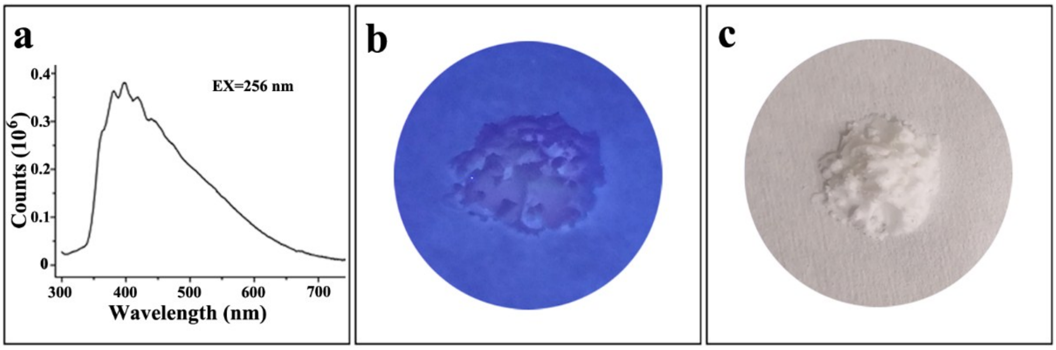

Figure S1 (a) Photoluminescence emission spectra of SCHN under 256 nm excitation, photographs of SCHN-12 in their solid state under (b) 256 nm UV lamp and (c) day light.

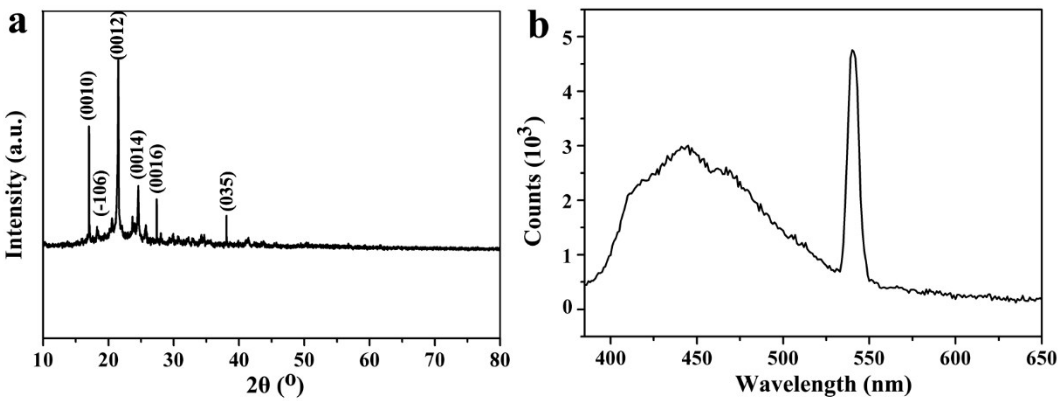

Figure S2 (a) The powder XRD patterns of the partial CTAB-removing SCHN-12, (b) photoluminescence emission spectra of the partial CTAB-removing SCHN-12 under 365 nm excitation.

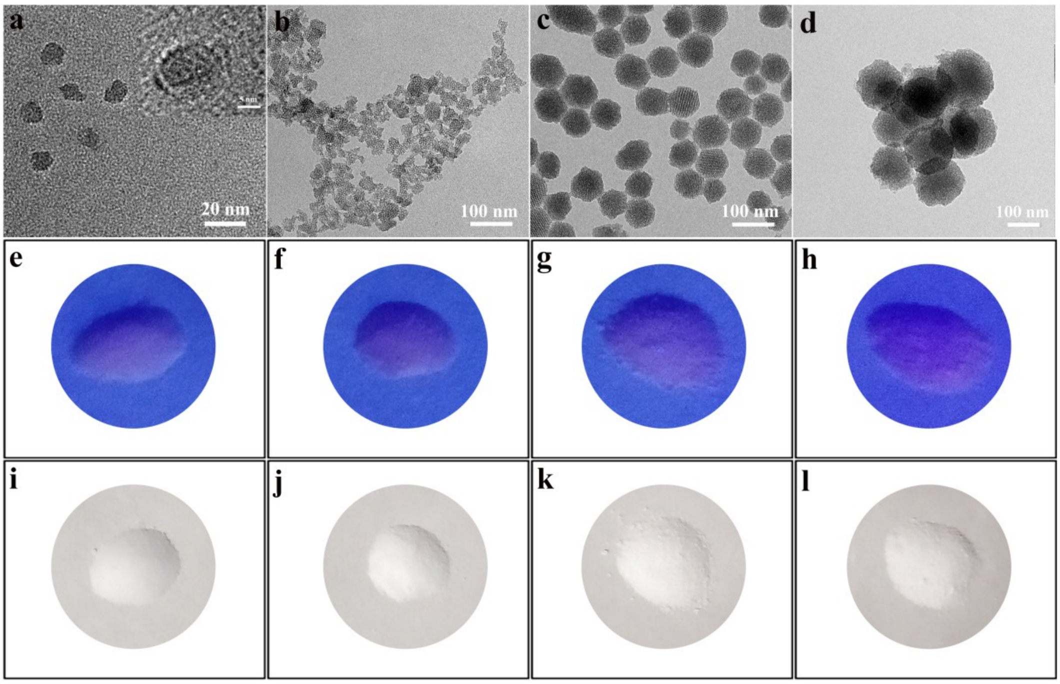

Figure S3 TEM images and photographs of PSN with different particle sizes. TEM images of (a) PSN-12, (b) PSN-23, (c) PSN-80 and (d) PSN-150. (b) Photographs of (e, i) PSN-12, (f, j) PSN-23, (g, k) PSN-80 and (h, l) PSN-150 in their solid state under 365 nm UV lamp (upper) and day light (down).

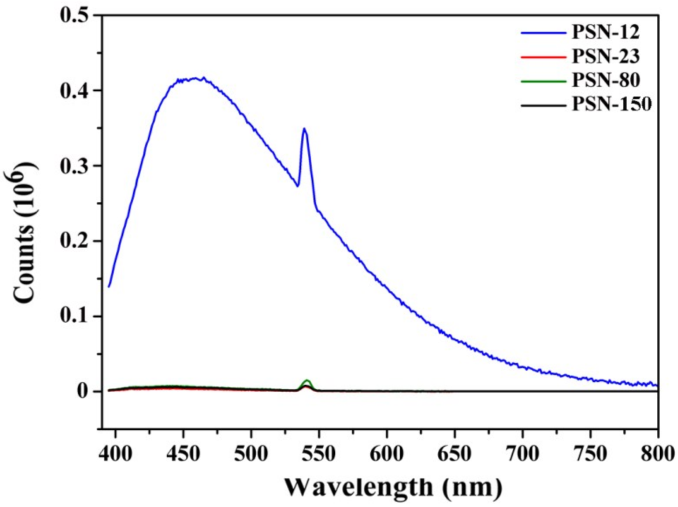

Figure S4 Photoluminescence emission spectra of PSN-12, PSN-23, PSN-80 and PSN-150 under 365 nm excitation.

Table S1 The hydrodynamic particle sizes of SCHN samples.

Sample	Size (nm)	PDI
SCHN-12	14.1±1.2	0.128±0.029
SCHN-23	26.7±2.9	0.198±0.013
SCHN-80	95.6±3.2	0.208±0.035
SCHN-150	180.3±8.6	0.226±0.020

Table S2 The hydrodynamic particle sizes of PSN samples.

Sample	Size (nm)	PDI
PSN-12	13.9±2.2	0.134±0.022
PSN-23	26.4±6.1	0.181±0.006
PSN-80	91.9±3.7	0.239±0.018
PSN-150	175.2±4.9	0.242±0.027

Table S3 The quantum yields of PSN with different particle sizes.

Sample	PSN-12	PSN-23	PSN-80	PSN-150
Quantum yield	1.03 %	1.07 %	1.38 %	0.93 %

Table S4 The fluorescence lifetimes of SCHN with different particle sizes.

Sample	**Fluorescence lifetimes**	
	τ_1	τ_2
SCHN-12	1.14	9.18
SCHN-23	1.15	9.26
SCHN-80	1.07	9.37
SCHN-150	1.15	9.47

Theoretical model

We consider two different two-state molecules $|S_{c0}\rangle$ and $|S_{c1}\rangle$ with a transition frequency ω_1 for CTAB, and $|S_{p0}\rangle$ and $|S_{p1}\rangle$ with a transition frequency ω_2 for PSN, and the two molecules are coupled through the dipole-dipole interaction in a strength of β. A four-state quantum system is formed with states $|S_{c0}S_{p0}\rangle$, $|S_{c0}S_{p1}\rangle$, $|S_{c1}S_{p0}\rangle$, and $|S_{c1}S_{p1}\rangle$, and the Hamiltonian H_0 with the dipole-dipole interaction can be written as

$$H_0 = \begin{pmatrix} -\omega_0 & 0 & 0 & 0 \\ 0 & -\Delta & \beta & 0 \\ 0 & \beta & \Delta & 0 \\ 0 & 0 & 0 & \omega_0 \end{pmatrix} \tag{1}$$

Where $\omega_0 = (\omega_1 + \omega_2)/2$ and $\Delta = (\omega_2 - \omega_1)/2$. By diagonalizing the nondiagonal Hamiltonian H_0, four collective (Dicke) states can be given by [1,2]

$$\begin{aligned} |G\rangle &= |S_{c0}S_{p0}\rangle \\ |S\rangle &= \alpha|S_{c0}S_{p1}\rangle + \gamma|S_{c1}S_{p0}\rangle \\ |A\rangle &= \alpha|S_{c1}S_{p0}\rangle - \gamma|S_{c0}S_{p1}\rangle \, |E\rangle = |S_{c1}S_{p1}\rangle \end{aligned} \tag{2}$$

Where the corresponding eigenvalues are $E_G = -\omega_0$, $E_S = w$, $E_A = -w$, and $E_E = \omega_0$, where $\alpha = \frac{r}{\sqrt{r^2 + \beta^2}}$, $\gamma = \frac{r}{\sqrt{r^2 + \beta^2}}$, $w = \sqrt{\Delta^2 + \beta^2}$, and $r = \Delta + \sqrt{\Delta^2 + \beta^2}$. From Eq. (2), we can obtain

$$\begin{aligned} |S_{c0}S_{p0}\rangle &= |G\rangle \\ |S_{c1}S_{p0}\rangle &= \alpha|A\rangle + \gamma|S\rangle \\ |S_{c0}S_{p1}\rangle &= \alpha|S\rangle - \gamma|A\rangle \, |S_{c1}S_{p1}\rangle = |E\rangle \end{aligned} \tag{3}$$

The interaction Hamiltonian between molecules and field reads

$$H_c(t) = -\varepsilon(t)\mu_c\left(|S_{c1}S_{p0}\rangle\langle S_{c0}S_{p0}| + |S_{c0}S_{p1}\rangle|S_{c1}S_{p1}\rangle\right) - \varepsilon(t)\mu_p\left(|S_{c0}S_{p1}\rangle\langle S_{c0}S_{p0}| + |S_{c1}S_{p0}\rangle|S_{c1}S_{p1}\rangle\right) + h.c. \tag{4}$$

By inserting Eq. (3) into Eq. (4), we can rewrite the interaction Hamiltonian in the basis of the four collective states,

$$H_c(t) = -\varepsilon(t)\begin{pmatrix} 0 & \mu_c\alpha - \mu_p\gamma & \mu_c\gamma + \mu_p\alpha & 0 \\ \mu_c\alpha - \mu_p\gamma & 0 & 0 & \mu_p\alpha - \mu_c\gamma \\ \mu_c\gamma + \mu_p\alpha & 0 & 0 & \mu_c\alpha + \mu_p\gamma \\ 0 & \mu_p\alpha - \mu_c\gamma & \mu_c\alpha + \mu_p\gamma & 0 \end{pmatrix} \tag{5}$$

From Eq. (5), we can see that the effective transition dipole moments of the transition between states $|G\rangle(|E\rangle)$ and $|S\rangle(|A\rangle)$ are

$$\mu_{GS} = \mu_c\gamma + \mu_p\alpha$$
$$\mu_{GA} = \mu_c\alpha - \mu_p\gamma$$
$$\mu_{ES} = \mu_c\alpha + \mu_p\gamma$$
$$\mu_{ES} = \mu_p\alpha - \mu_c\gamma$$

References

[1] Z. Ficek, R. Tanas, Entangled states and collective nonclassical effects in two-atom systems, *Phys. Rep.*, **2002**，372, 369-443.

[2] Y. Guo, X. B. Luo, S. Ma, C.-C. Shu, All-optical generation of quantum entangled states with strictly constrained ultrafast laser pulses, *Phys. Rev. A*, **2019**, 100, 023409.