Electronic Supplementary Information

Composition-Tunable Nonlinear Optical Properties of Alloy Ternary $CdSe_xS_{1-x}$（x = 0–1）Quantum Dots

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1. The Z-scan experimental results obtained with an open aperture

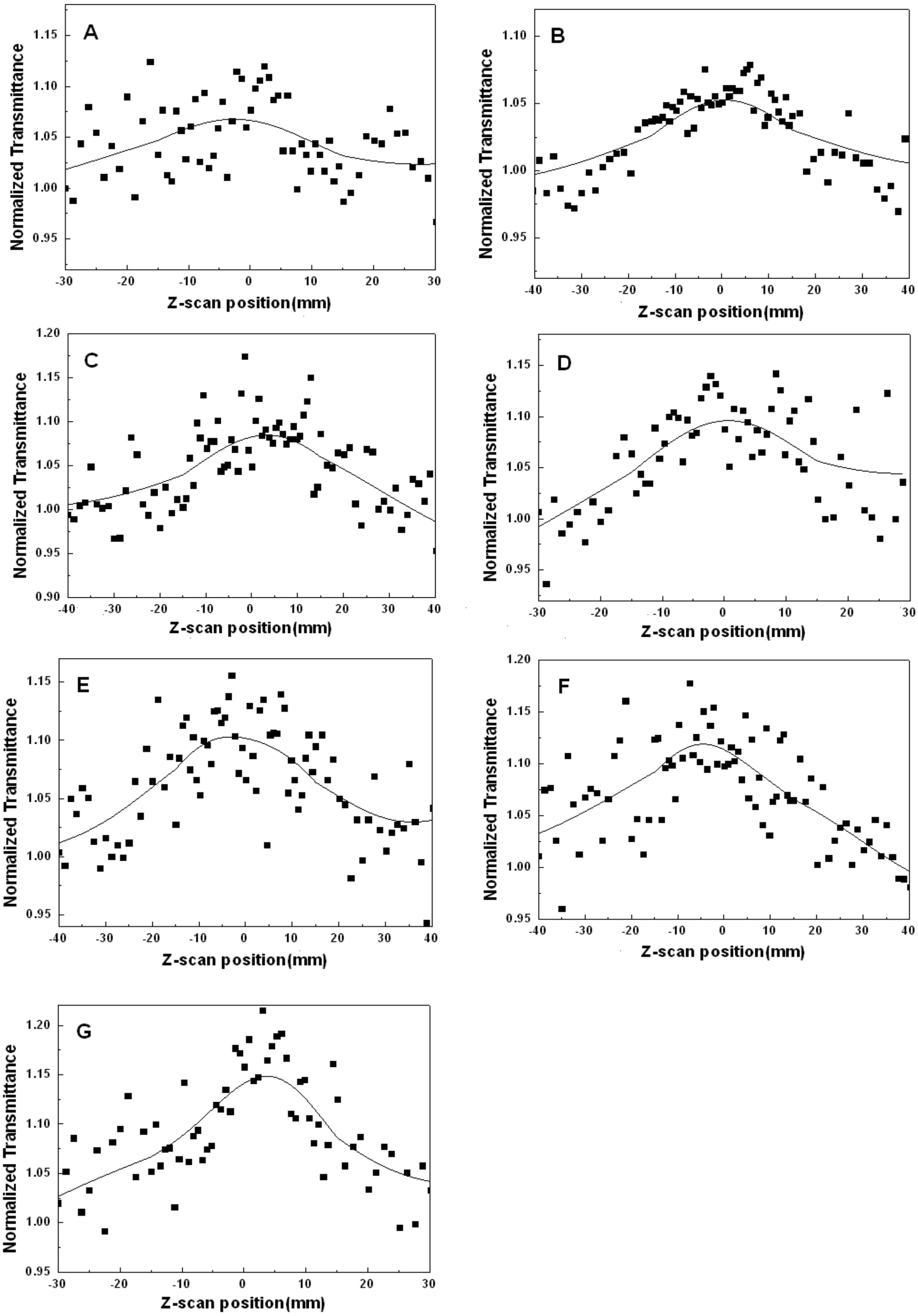

Fig. ESI1 The open-aperture Z-scan curves of the $CdSe_xS_{1-x}$ QDs with different compositions: (A) CdS, (B) $CdSe_{0.14}S_{0.86}$, (C) $CdSe_{0.33}S_{0.67}$, (D) $CdSe_{0.43}S_{0.57}$, (E) $CdSe_{0.56}S_{0.44}$, (F) $CdSe_{0.76}S_{0.24}$ and (G) CdSe.

2. The Z-scan experimental results obtained with an closed aperture

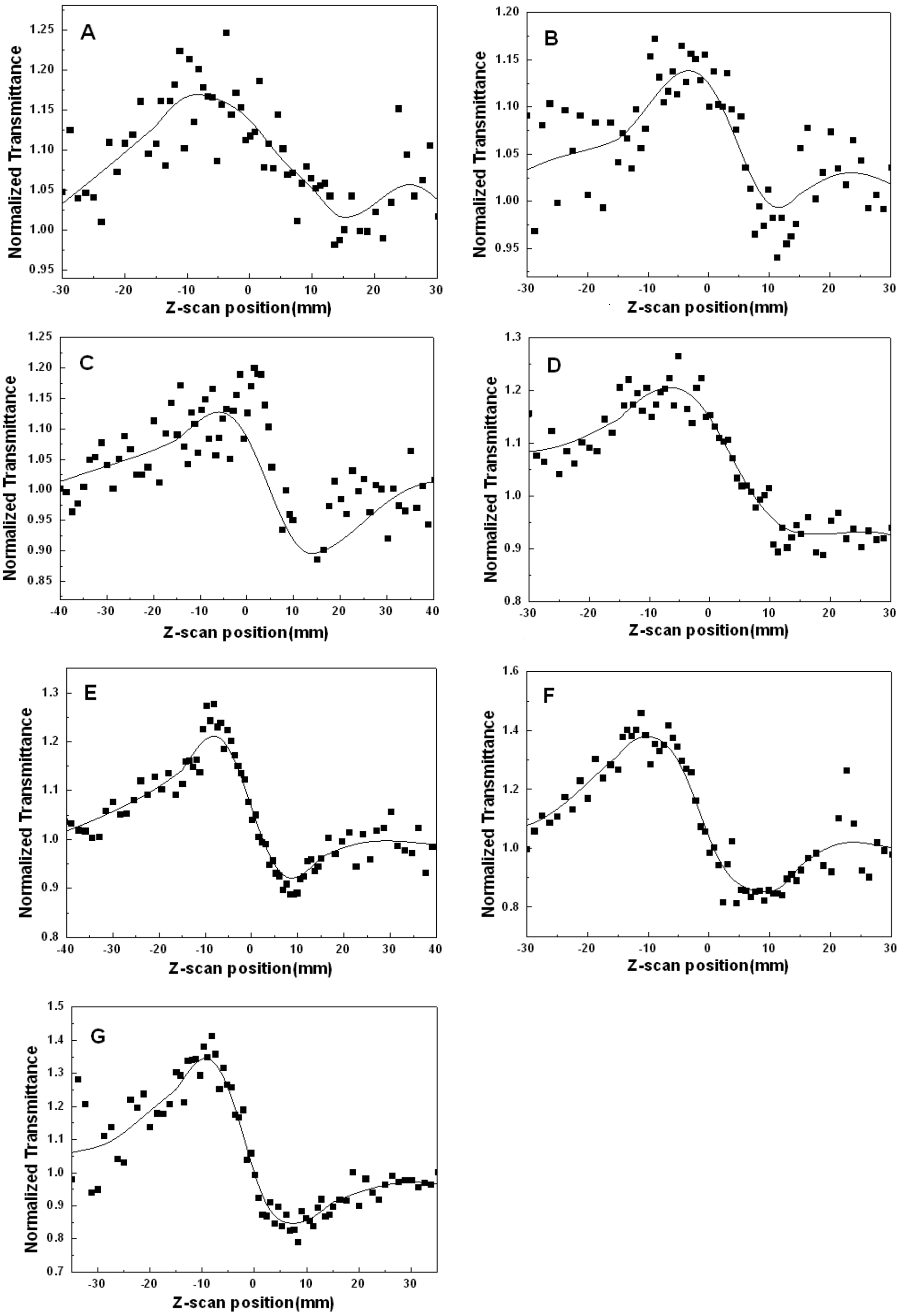

Fig. ESI2 The closed-aperture (S=0.054) Z-scan curves of the $CdSe_xS_{1-x}$ QDs with different compositions: (A) CdS, (B) $CdSe_{0.14}S_{0.86}$, (C) $CdSe_{0.33}S_{0.67}$, (D) $CdSe_{0.43}S_{0.57}$, (E) $CdSe_{0.56}S_{0.44}$, (F) $CdSe_{0.76}S_{0.24}$ and (G) CdSe.

3. Calculation of the nonlinearity of $CdSe_xS_{1-x}$ QDs

The purely nonlinear refraction Z-scan data are obtained by dividing the closed-aperture data with the corresponding open aperture data. $\Delta\Phi_0$ can be obtained from the purely nonlinear refraction Z-scan data by fitting the open aperture data using Eq.（1） :

$$\mathrm{T}=1+\frac{4x}{(x^2+9)(x^2+1)}\Delta\Phi_0 \qquad (1)$$

where T is the normalized transmittance of the sample with, $\Delta\Phi_0$ is the phase shift at the focus, $\mathrm{x}=\mathrm{z}/z_0$, $z_0 = pw_0{}^2/l$ is the laser diffraction length.

The nonlinear refractive index g of the $CdSe_xS_{1-x}$ QDs is determined according to Eq.（2）and (3):

$$|\Delta\Phi_0| = (2p/l)g\,I_0L_{eff} \qquad (2)$$

$$n_2(esu) = (cn_0/40p)g\,(m^2/\mathrm{W}) \qquad (3)$$

where I_0 is on-axis pulse peak intensity on sample in the form of $\mathrm{I}_0=\mathrm{E}/pw_0^2t$ where E is the pulse energy, w_0 is the radius of the beam waist, t is the pulse width, l is the laser wavelength, L_{eff} is the effective length of the sample, which is related to the sample thickness L and the linear absorption coefficient α, in the form of $\mathrm{L}_{eff} = [1-\exp(-a\mathrm{L})]/a$, c is the speed of light, and n_o is the linear index of refraction of the quantum dots.

The nonlinear absorption , β of the $CdSe_xS_{1-x}$ quantum dots can be obtained from Z-scan data by fitting the open aperture data using Eq.(4):

$$\mathrm{T(z,S=1)}=\sum_{m=0}^{\infty}\frac{[-q_0(z,0)]^m}{(m+1)^{3/2}} \qquad (4)$$

Where $q_0 = b\,\mathrm{I}_0\mathrm{L}_{eff}\big/(1+\mathrm{z}^2/\mathrm{z}_0^2)$, $z_0 = pw_0{}^2\big/l$ is the laser diffraction length.

The nonlinear susceptibility is a result of interaction between light wave field and medium. It can be evaluated through measuring n_2 or γ . To the sample having the nonlinear absorption, the nonlinear susceptibility is considered to be a complex number. For $CdSe_xS_{1-x}$ quantum dots, the value of third–order nonlinear susceptibility $c^{(3)}$ is calculated by Eq.(5):

$$c^{(3)} = c_{\mathrm{R}}{}^{(3)} + i\,c_{\mathrm{I}}{}^{(3)} \qquad (5)$$

The real and imaginary parts of the third-order nonlinear susceptibility were deduced by using the values γ and β ,as shown in Eqs.(6), (7) and (8).

$$\mathrm{Re}\,c^{(3)} = 2n_0{}^2 e_0 c g \qquad (6)$$

$$\mathrm{Im}\,c^{(3)} = n_0{}^2 e_0 c\, l\, b\big/2p \qquad (7)$$

$$c^{(3)} = \left\{\left(\mathrm{Re}\,c^{(3)}\right)^2 + \left(\mathrm{Im}\,c^{(3)}\right)^2\right\}^{\frac{1}{2}} \qquad (8)$$

Where n_o is the linear refractive index of the quantum dots, e_0 is the permittivity of free space.

4. Nonlinearity of CdSe0.43S0.57 with different sizes

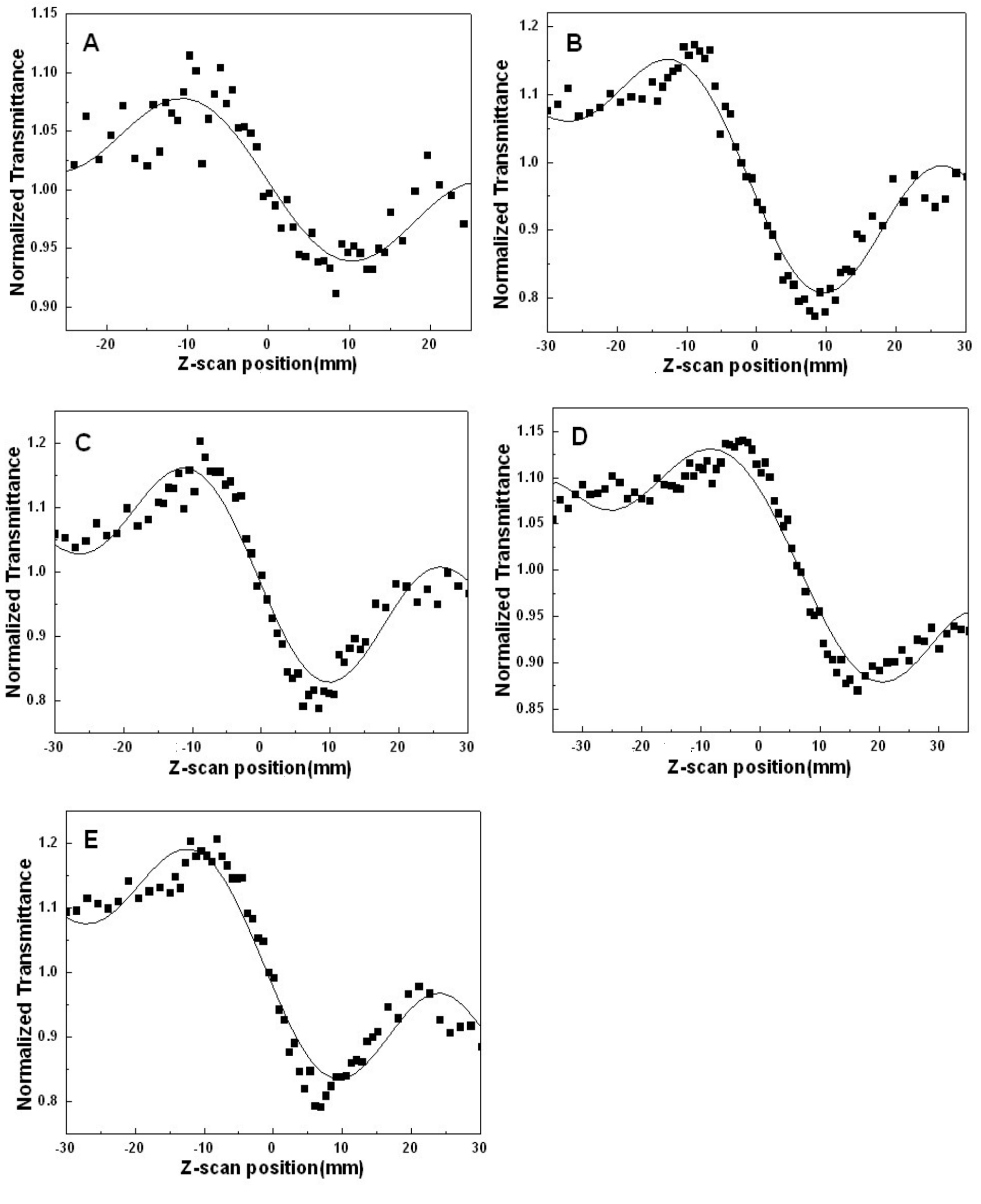

Fig. ESI3 Z-scan curves of $CdSe_{0.43}S_{0.57}$ with different sizes: (A) 2.0 nm, (B) 2.3 nm, (C) 2.6 nm, (D) 2.9 nm, (E) 3.1 nm.

Table. ESI1 Calculated values of the nonlinearity of $CdSe_{0.43}S_{0.57}$ with different sizes.

Average size (nm)	$CdSe_{0.43}S_{0.57}$	β(m/W)	n_2 (esu)	$Im(\chi^{(3)})^a$	$Re(\chi^{(3)})^b$	$\chi^{(3)}$ (esu)
2.0	170 °C	-3.35×10^{-11}	-5.15×10^{-11}	-1.69×10^{-19}	-1.76×10^{-19}	2.44×10^{-19}
2.3	190 °C	-3.60×10^{-11}	-5.38×10^{-11}	-1.82×10^{-19}	-1.84×10^{-19}	2.59×10^{-19}
2.6	210 °C	-3.52×10^{-11}	-5.28×10^{-11}	-1.78×10^{-19}	-1.80×10^{-19}	2.52×10^{-19}
2.9	230 °C	-3.47×10^{-11}	-5.35×10^{-11}	-1.76×10^{-19}	-1.83×10^{-19}	2.54×10^{-19}
3.1	250 °C	-3.63×10^{-11}	-5.45×10^{-11}	-1.83×10^{-19}	-1.86×10^{-19}	2.61×10^{-19}