

Supplementary Material

One Order Enhancement of Charge Carrier Relaxation Rate by Tuning Structural and Optical Properties in Annealed Cobalt Doped MoS₂ Nanosheets

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S1. AFM Image Analysis

The surface topography and number of layers in 2% CoA MoS₂ nanoflakes are determined by atomic force microscopy (AFM) as shown in Fig. S1. The variation in AFM height profile as shown in Fig. S1 (b), is measured along the green solid line in Fig. S1 (a) indicates that the sample consists of 15 layers.

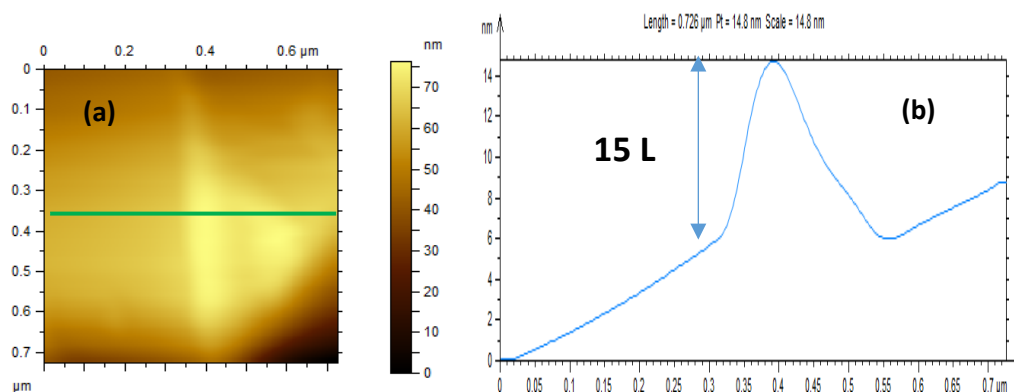


Figure S1. (a) AFM image of 2% CoA MoS₂ nanoflakes (b) Height profile of 2% CoA MoS₂ taken along the green solid line of (a).

S2. XPS study

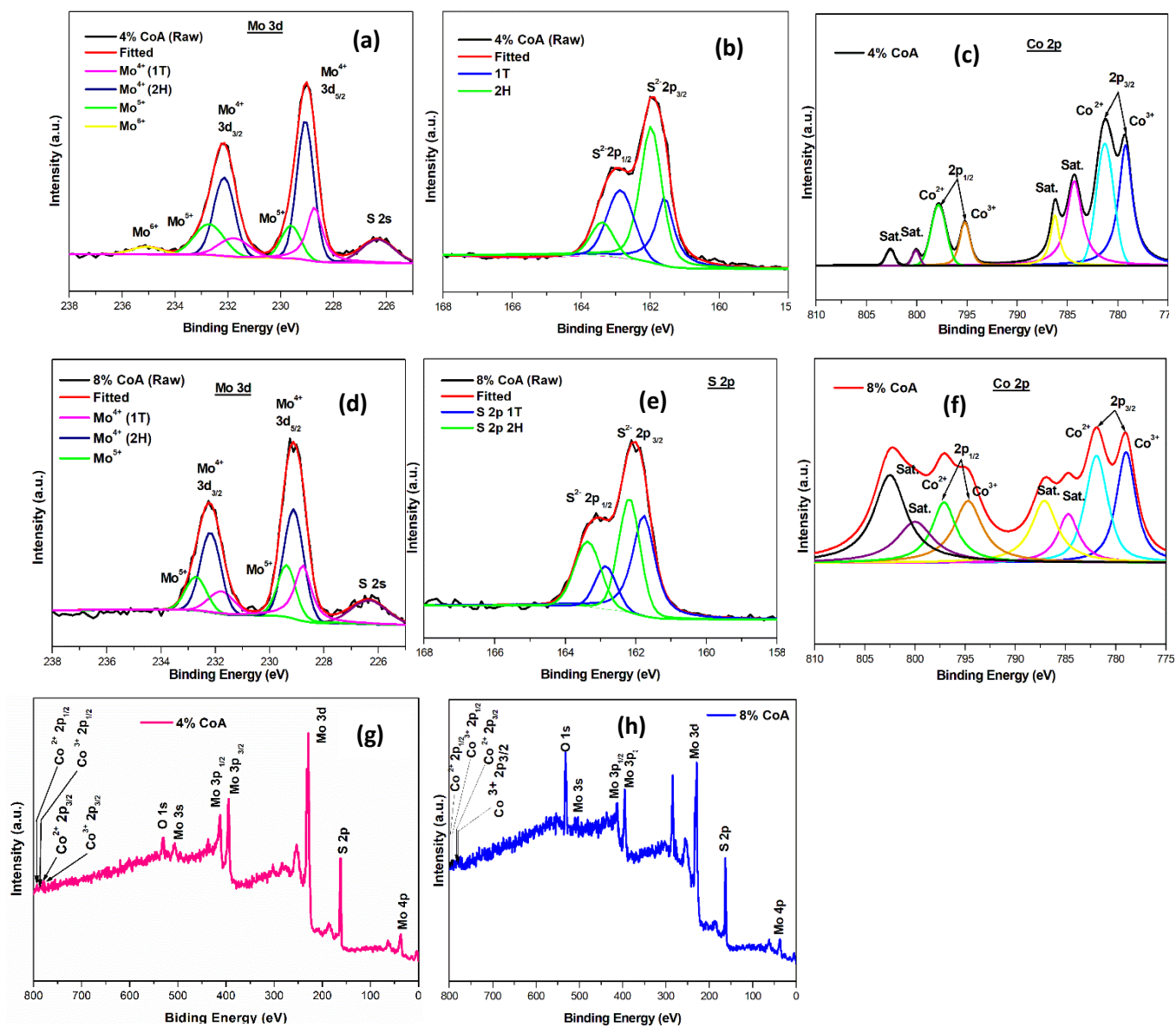


Figure S2. XPS narrow scan spectra of Mo 3d (a), (d), S 2p (b), (e) and Co 2p (c), (f) for 4% CoA, 8% CoA MoS_2 . Survey spectra of Mo (3d, 3p, 3s), S 2p, C1s, Co 2p are shown in (g) and (h) corresponding to 4% CoA, 8% CoA MoS_2 .

Table S1 Variation of oxidation peaks of Mo, S and Co in annealed MoS₂ with cobalt doping

Doping percentages	1T		2H		1T		2H		Co ³⁺		Co ²⁺	
	Mo 3d _{5/2} (eV)	Mo 3d _{3/2} (eV)	Mo 3d _{5/2} (eV)	Mo 3d _{3/2} (eV)	S 2p _{3/2} (eV)	S 2p _{1/2} (eV)	S 2p _{3/2} (eV)	S 2p _{1/2} (eV)	Co 2p _{3/2} (eV)	Co 2p _{1/2} (eV)	Co 2p _{3/2} (eV)	Co 2p _{1/2} (eV)
4% CoA	228.71	231.76	229.06	232.14	161.57	162.86	161.97	163.39	779.22	795.21	781.26	797.81
8% CoA	228.76	231.77	229.11	232.18	161.77	162.85	162.18	163.36	778.96	794.72	781.90	797.13

Table S2 Variation in position of oxidation peaks of Mo⁵⁺, Mo⁶⁺ and S2s in unannealed and annealed MoS₂ with cobalt doping

Doping percentages	Mo ⁵⁺		Mo ⁶⁺	S2s
	3d _{5/2} (eV)	3d _{3/2} (eV)	3d _{5/2} (eV)	(eV)
0% Co	229.72	232.41	234.42	226.38
0% CoA	229.66	232.73	-----	226.39
2% Co	229.75	232.99	235.64	226.36
2% CoA	229.72	232.86	-----	226.71
4% Co	229.61	233.37	235.57	226.38
4% CoA	229.6	232.71	235.09	226.32
8% CoA	229.37	232.66	-----	226.36

S3 Steady-state and Transient Absorption Spectroscopy

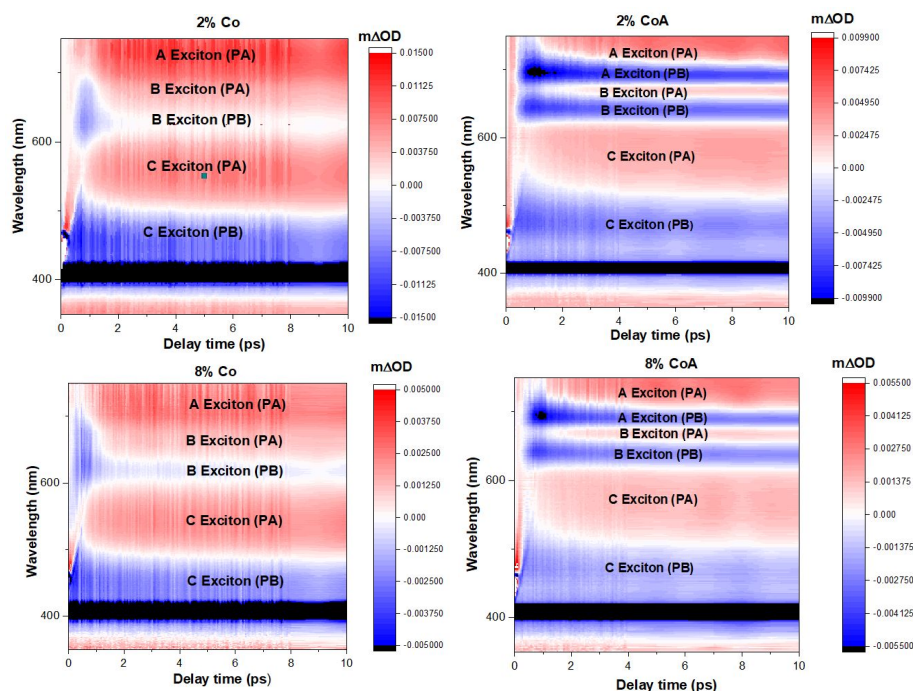


Figure S3. Differential absorption ($\Delta A/A_0$) map of (a) 2%, (b) 2% CoA (c) 8% Co (d) 8% CoA as the function of both delay time and probe photon energy with the pump photon energy of 3.1 eV at average pump fluence of 22 $\mu\text{J}/\text{cm}^2$ and 77 $\mu\text{J}/\text{cm}^2$ for unannealed and annealed sets respectively.

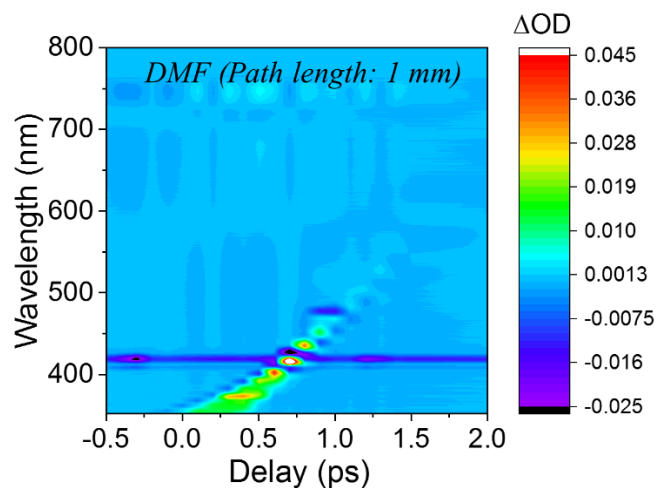


Figure S4. Transient absorption spectrum of DMF following 415 nm pump excitation.

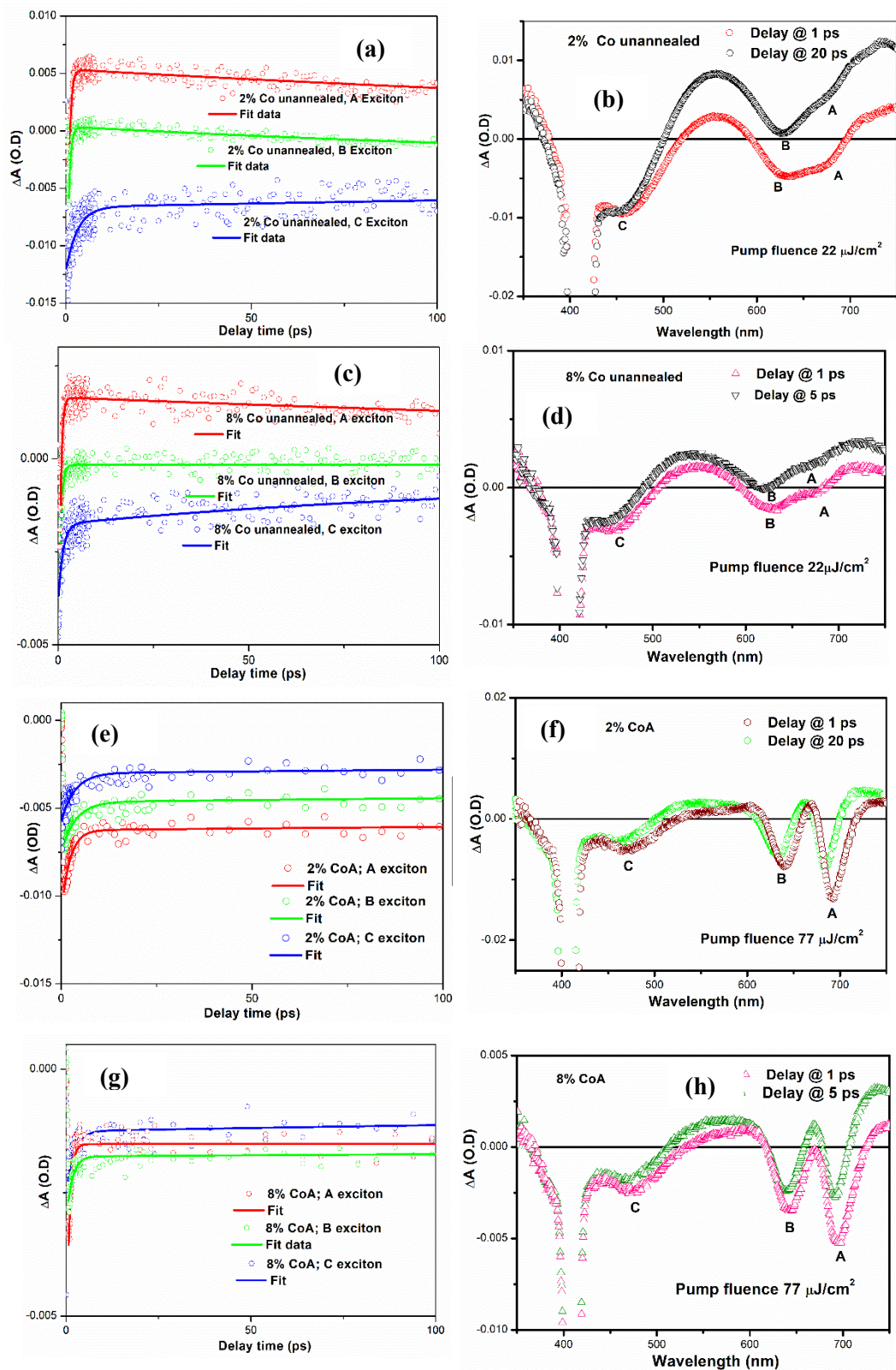


Figure S5. Transient absorption dynamics of (a) 2% Co unannealed, (c) 8% Co unannealed MoS₂ (e) 2% CoA, (g) 8% CoA MoS₂ having pump wavelength of 400 nm and probe wavelength of 460, 627, 681 nm (unannealed) and 462, 632 and 687 nm (annealed) respectively. The solid lines are fits of a biexponential decay function. Figs. 12. (b), (d), (f) and (h) depicts the transient absorption spectra of 2% Co unannealed, 8% Co unannealed, 2% CoA, 8% CoA MoS₂ respectively at 1 ps and longer time 5 ps (8% Co, 8% Co A) and 20 ps (2% Co, 2% Co A) delay.

