Self-assembled MoS$_2$/rGO Nanocomposites with Tunable UV-IR Absorption

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Figure S1 shows the SEM images of MoS$_2$/rGO composites on a SiO$_2$/Si substrate after annealing for the different TAA/GO ratios: (a) 1:0, (b) 1:1, (c) 1:2 and (d) 1:3. rGO deriving from GO acts as a dispersing platform, efficiently reducing the aggregation of MoS$_2$ with a decrease of the TAA/GO ratio and forming self-assembled layered MoS$_2$/rGO structures during the hydrothermal process. The morphology of the annealed structure is slightly different from the morphology of as-grown structure (Fig. 1).

Figure S1. SEM images of MoS$_2$/rGO composites on a SiO$_2$/Si substrate after annealing for the different TAA/GO ratios: (a) 1:0, (b) 1:1, (c) 1:2 and (d) 1:3.
Figure S2. Raman spectra of MoS$_2$ and MoS$_2$/rGO composites with different GO content before and after annealing (fitted with Lorentzian functions).

The Raman spectra of MoS$_2$ (Fig. S2) show the weak peak at 820 cm$^{-1}$ which corresponds to the M=O bending stretch vibrations for MoO$_{3-x}$ and can be ascribed to the formation of molybdenum oxide during annealing process [1-2]. This peak disappears in the spectra of MoS$_2$/GO composites, indicating a stronger binding of oxygen to carbon. The origin of the main peaks of the MoS$_2$/r-GO composite ($E_{2g}^1$, $A_{1g}$ and G, D) is discussed in the description of Fig. 2b. The 520 cm$^{-1}$ peak and the peaks at ~2700 and ~2930 cm$^{-1}$ correspond to the Si substrate [3] and the second-order 2D and D+G bands of graphene [4-5], respectively.

Figure S3. X-ray photoelectron spectra of MoS$_2$ and MoS$_2$/rGO composites corresponding to (a) survey, and (b) C 1s core levels.
The chemical composition and binding energies of elements in MoS$_2$/rGO composites were investigated using XPS. Figure S3 shows the X-ray photoelectron spectra of MoS$_2$ and MoS$_2$/rGO composites corresponding to (a) survey, and (b) C 1s core levels. To fit the spectra of composites with MoS$_2$/GO ratios 1:2 and 1:3, two doublets and splittings of 1.2 and 3.1 eV for S 2p and Mo 3d, respectively, were used. The stronger C 1s signal of the MoS$_2$/GO (1:2) compared to MoS$_2$ (Fig. S3a) clearly indicates a significant hydrothermal GO reduction during the synthesis. The low intensity of the Mo 3d signals is apparently due to the formation of Mo$_2$S$_x$O$_{1-x}$ and Mo$_2$S$_5$, which shows an intermediate product in the MoS$_3$-to-MoS$_2$ transition reacting with oxygen from GO deoxygenation. The spectra of C 1s can be decomposed into six components. Peaks at 284.6, 285.3, 285.8, 286.6, 287.5, 288.99 correspond to C=C (sp$^2$) of 48.1 %, C-C (sp$^3$) of 20.4 %, C-OH of 16.4 %, C-O-C of 9.5 %, C=O of 1.2 %, and O=C-OH of 4.4 %, respectively, further confirming the existence of rGO in the composites (Fig. S3b).

**Table S1.** Detailed characteristics of Raman scattering of MoS$_2$ and MoS$_2$/rGO composites.

| Specimen | As-grown | Annealed |
|----------|----------|----------|
| MoS$_2$  | E$_{12g}$ | A$_{1g}$ | E$_{12g}$ | A$_{1g}$ | D | G |
| As-grown | 381.2 | 407.6 | 35.1 | 12.8 | / | / |
| Annealed | 381.2 | 407.4 | 26.96 | 12.7 | / | / |
| MoS$_2$/rGO (1:1) | As-grown | 381.9 | 406.6 | 30.6 | 19.9 | 1342.3 | 1576.4 |
| Annealed | 382.1 | 405.3 | 28.4 | 17.6 | 1341.7 | 1573.98 |
| MoS$_2$/rGO (1:2) | As-grown | 382.1 | 404.6 | 22.0 | 13.6 | 1345.9 | 1578.6 |
| Annealed | 382.6 | 404.5 | 21.3 | 15.7 | 1348.7 | 1581.4 |
| MoS$_2$/rGO (1:3) | As-grown | 382.6 | 404.4 | 17.9 | 12.1 | 1347.6 | 1581.7 |
| Annealed | 382.7 | 404.1 | 20.5 | 15.2 | 1349.1 | 1581.9 |
The chemical reaction for the hydrothermal synthesis can be described as:

$$4MoO_4^{2-} + 9CSNH_2^+ + 12H_2O = 4MoS_2 + SO_4^{2-} + 9COOH^+ + 6OH^- + 9NH_3$$

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