

Supplementary Information: Tailoring Ultra-fast Charge Transfer in MoS₂

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1 Characterisation of MoS₂

XRD was recorded with CuK α radiation ($\hbar\omega = 8048$ eV, $\lambda = 1.54$ Å) with a Bruker TWIN-TWIN instrument. The Raman measurements were acquired using a Renishaw micro Raman system with a 532 nm argon-ion laser.

The left panel in Fig. 1 shows a general view of the morphology of the rGO-doped MoS₂ and reveals a uniform sheet-structure. The sheet-structures also indicates a large surface area. The image also indicates that the individual sheets are very thin.

The XRD spectra in Fig. 1 indicates that the MoS₂ in the rGO-doped sample is crystalline. The broad peaks indicate a small crystallite size of the MoS₂ in the sample *i.e.* the MoS₂ is layered very thin or poly-crystalline on the rGO sheets. Raman-spectroscopy shows that the rGO-doped MoS₂ consists of pure MoS₂ and reduced graphene oxide, as can be seen in Fig. 1. The Raman spectra of the doped material shows the characteristic E_{2g}¹ and A_{1g} peaks of the in and out of plane vibrations from MoS₂, with a peak split of ≈ 26.4 cm⁻¹ indicates that the MoS₂ is bulk-like¹. The characteristic D and G peaks of graphene oxide are also present, with a higher intensity of the G peak compared to the D peak.

As can be seen in Figure 2 and Fig. 4 there is one component of molybdenum and one of sulphur, in all three samples. This indicates high phase-purity of both systems. The shoulder on the high binding energy side of the Mo 3p peaks and the broad background under the 3p_{1/2} component are attributed to bulk and surface plasmons². Fig. 3 shows the results from a least squares fit of the C 1s spectra from the reduced graphene oxide, Fig. 4 shows the O 1s spectra. The O 1s result is similar to that of reduced graphene oxide from reference 3, there is however a shift to higher binding energies compared to their results, these could be from the addition of MoS₂. Shifts in the O 1s binding energy as function of reduction is also reported elsewhere⁴. The O 1s spectrum in Figure 4 together with C 1s indicates reduced graphene oxide since neither S 1s nor Mo 3p show any oxide contribution.

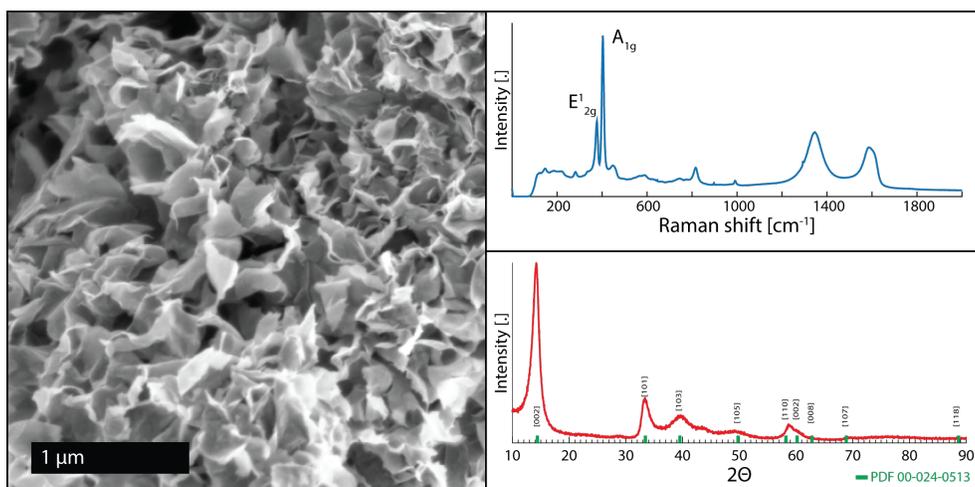


Fig. 1 Left; SEM image of the rGO-doped MoS₂ top right; Raman spectra acquired using a 532 nm laser at 2 mW bottom right; XRD spectra of the rGO-doped MoS₂.

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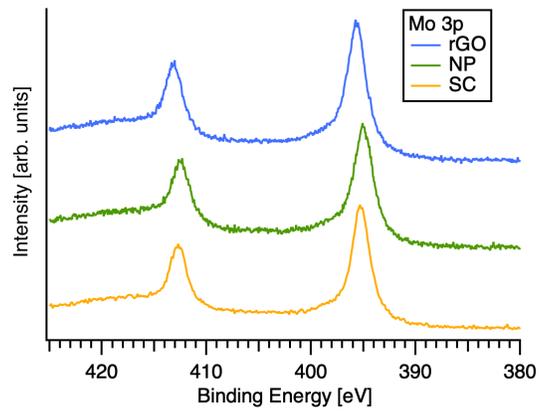


Fig. 2 Mo 3p spectra recorded at a photon energy of 2005 eV for the three different samples.

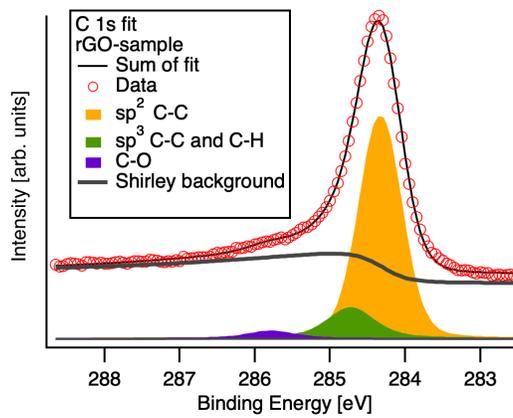


Fig. 3 Least square fit of the C 1s photoline for the rGO sample recorded at 2005 eV photon energy.

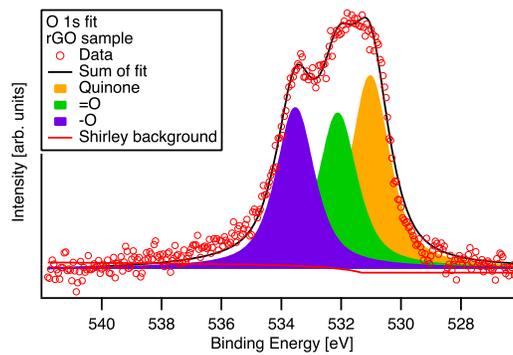


Fig. 4 Least square fit of the O 1s spectra recorded at a photon energy of 2005 eV for the rGO sample.

2 The life time width: S 1s

The finite life time of the core ionised state is reflected in a Lorentzian broadening of the spectral profile.

The S 1s X-ray photoelectron spectra were taken at a photon energy of 6015 eV, far above the ionisation threshold. The binding energy scale were calibrated against the Au 4f_{7/2} binding energy. The combined experimental broadening from the monochromatic photon bandwidth and the electron energy analyser is modelled with a Gaussian.

The spectra were modelled with a least squares fit of a Voigt profile (combining the Lorentzian and Gaussian contributions to the spectral imprint) and a Shirley background function.

The relation between to the Lorentzian width and the core-hole life time is given by Eq. 1.

3 Fitting of the Auger spectra

The Auger spectra were fitted using a least squares method from the SPANCF Igor package⁵. A Shirley type background⁶, asymmetric PCI-type for the charge transfer Auger line and Voigt line for the spectator Auger line. First the normal Auger spectra was fitted above the ionisation threshold to get a fixed energy position for the main peak. The individual spectra were then fitted from high photon energy towards lower photon energy using the kinetic energy position from the Normal Auger spectra and knowing the photon energy step the position of the Raman peak could be determined.

$$\tau_c = \frac{\hbar}{\Gamma} \quad (1)$$

The theoretical framework has been described by Ohno⁷.

References

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