

# Giant Gap-plasmon Tip-Enhanced Raman Scattering of MoS<sub>2</sub> Monolayers on Au Nanocluster Arrays

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## S-1: Photoluminescence of monolayer MoS<sub>2</sub> on Au nanoclusters

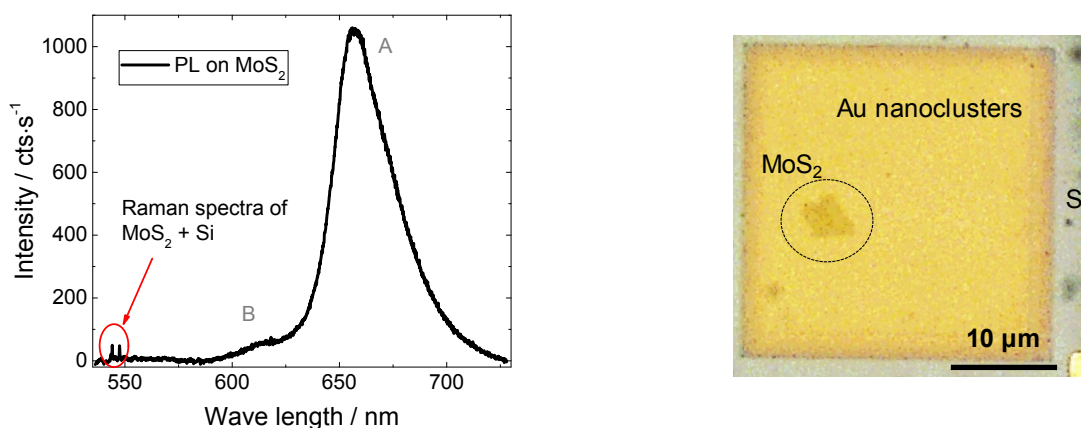


Fig. 1s: Photoluminescence spectrum (left) of MoS<sub>2</sub> monolayer on Au nanoclusters (right). The spectrum was acquired using a Horiba Xplora Raman system coupled with an electron multiplying charge-coupled detector. A monochromatic laser source of 532 nm excitation was used with an excitation power of 105 μW at the sample surface to avoid any photoinduced damage of the sample. Monolayer MoS<sub>2</sub> is a direct band gap semiconductor with an optical band gap around 1.9 eV. In our spectra the PL peak is centered around 660 nm equivalent to 1.87 eV. We also observed both A and B peak of MoS<sub>2</sub> monolayer originated from spin-orbit coupling.

## S-2: Determination of spatial resolution

The spatial resolution was determined from the analysis of the TERS map for the A<sub>1g</sub> mode shown in Fig. 1s a). TERS spectra were measured from pixel 1 to 10 along the radial lines indicated in Fig. 1s a). The A<sub>1g</sub> mode TERS profiles taken along the lines along with the profile averaged over 10 radial lines are presented in Fig. 1s b). The TERS profiles for the A<sub>1g</sub> mode reveals a sharp maximum with the FWHM of (2.3±0.3) nm determined from the best Gaussian fit of the TERS profile. The FWHM value corresponds to the TERS resolution.

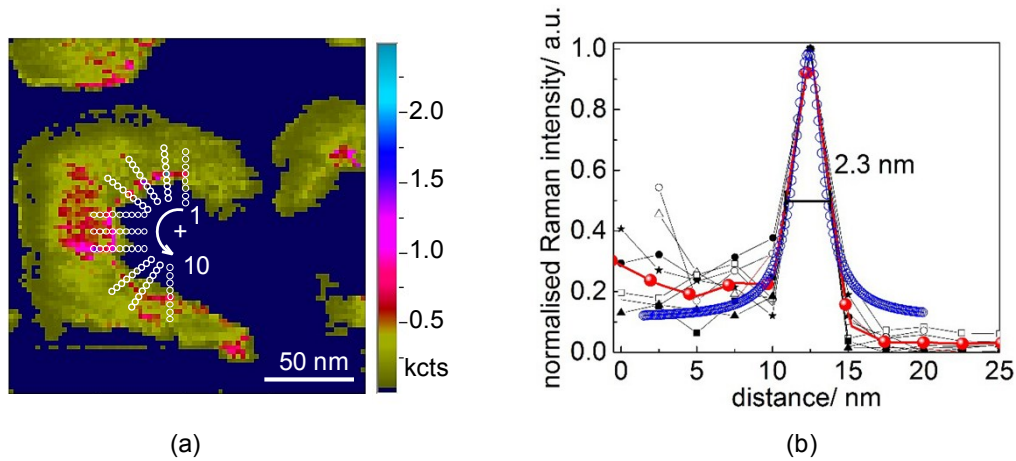


Fig. 2s: Determination of spatial resolution. a) High resolution TERS image of a single Au nanocluster covered with a MoS<sub>2</sub> monolayer. b) The A<sub>1g</sub> mode TERS profiles derived from the TERS spectra measured from pixel 1 to 10 along the radial lines indicated in image b). The averaged profile of the A<sub>1g</sub> mode intensity over 10 profiles is indicated by large solid spheres. Open circles show the best Gaussian fit of the averaged intensity profile.

### S-3: Site dependent peak positions of MoS<sub>2</sub>

As it is discussed in the main text, the peak position of Raman modes is site dependent. There are three factors (strain, heating, and doping) influencing the Raman spectra in our TERS measurement (see discussion in the main text). In order to check if the spectral shifting of the Raman modes is identical, we took several line profiles at the edges of the rings in TERS image. In Fig. 3s two of such line profiles are presented. Raman spectra of 6 pixels along the lines were taken and presented. As can be seen, the positions of both A<sub>1g</sub> and A<sub>2u</sub> modes are shifted towards lower frequencies while going through the bright pixels (hot spots).

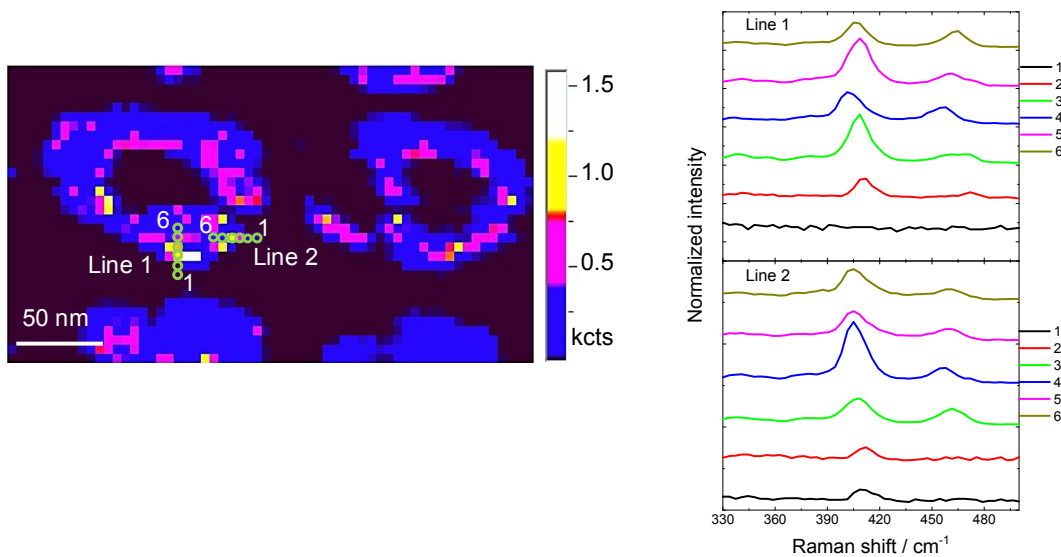


Fig. 3s: Site dependent TERS spectra. Two line profiles (denoted by Line 1 and Line 2) along the edge of the ring were taken (left). Raman spectra of six pixels in each line shown by green open circles (left image) are presented in the right.