Giant Gap-Plasmon Tip-Enhanced Raman Scattering of MoS_2 Monolayers on Au Nanocluster Arrays

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Abstract

We present the results on a gap-plasmon tip-enhanced Raman scattering study of MoS_2 monolayers deposited on a periodic array of Au nanostructures on a silicon substrate forming a two dimensional (2D) crystal / plasmonic heterostructure. We observe a giant Raman enhancement of the phonon modes of the MoS_2 monolayer located in a plasmonic gap between Au tip apex and Au nanoclusters. Tip-enhanced Raman (TER) mapping allowed us to determine the gap-plasmon field distribution responsible for the formation of hot spots. These hot spots provided an unprecedented giant Raman enhancement of $5.6 \cdot 10^8$ and a spatial resolution as small as 2.3 nm at ambient conditions. Moreover, due to strong hot electron doping in the order of $1.8 \cdot 10^{13}$ cm^{-2}, we observed a structural change of MoS_2 from 2H to 1T phase. Thanks to the very good spatial resolution, we were able to spatially resolve those doping sites. To the best our knowledge this is the first report of such phenomenon with nm spatial resolution. Our results open the perspectives of optical diagnostics with nanometer resolution for many other 2D materials.

Figures

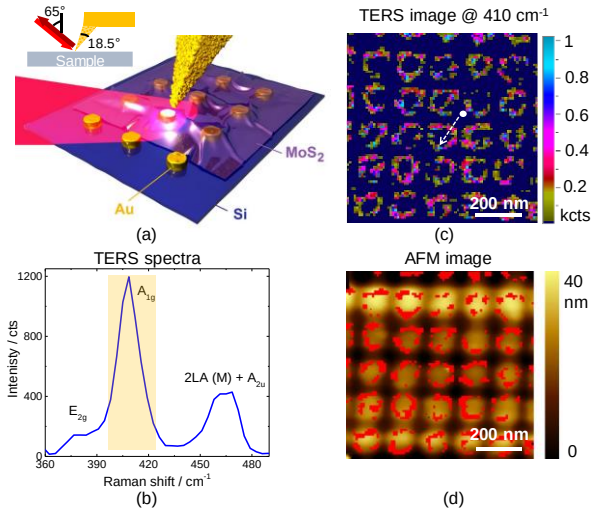

Figure 1: (a) Schematic of TERS experiment. The incident and collection geometry is shown in the inset. (b) A representative TERS spectra of 1L-MoS_2 on Au nanocylinders array using 785 nm excitation and homemade Au coated silicon tip. (c) A spatial map of Raman intensity of A_{1g} mode. The white dot with arrow line indicates the pixel from where TERS spectra is acquired in (b). TERS map is acquired within the area marked by a shaded rectangular box around A_{1g} mode in (b). (d) The corresponding AFM topography of the TERS map. The topography is superimposed by the maximal intensity map shown in (c) for better understanding.

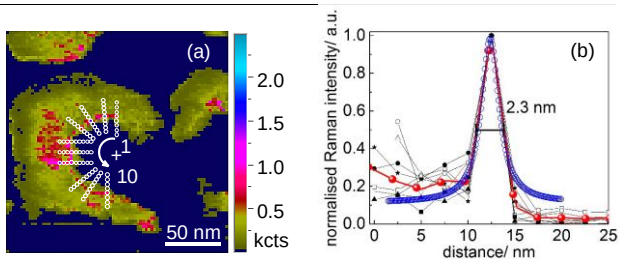

Figure 2: (a) High resolution TERS map of A_{1g} mode 1L-MoS_2 on Au nanocylinder. The step size is 2 nm. Ten cross sections are taken along the hot spots as shown in figure (a). (b) The cross sections and their average with the Gauss fit.