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Abstract
Presentation of two methods based on Raman spectroscopy for determining the number of layers of MoS$_2$.

Key words
MoS$_2$, Molybdenum disulfide, nanomaterials analysis, Raman spectroscopy, Raman imaging

Layered low-dimensional structures

There are two types of vibrational modes in MoS$_2$ (as in all layered structures): vibrational modes inside layers (intralayer) and modes resulting from the movements of complete layers (interlayer). The intralayer modes are primary related to the chemical composition of a layer (or layers) and are their fingerprint; however these can be slightly influenced by the number of layers. The interlayer modes, due to the high mass of the layers, are observed at very low frequencies and their position depends on the number of layers. As a result, both types of modes can be used to determine the number of layers.

The optical image of the sample of MoS$_2$ investigated here is shown on the left (courtesy of A. Shukla IMPMC, Pierre and Marie Curie University, France). The same sample was investigated already in [1].

Method 1: Analysis of fingerprint modes (intralayer)

Figure 2a: A Raman map of MoS$_2$ with different number of single layers based on the fingerprint modes analysis.

The number of layers can be found by determining the distance between two fingerprint peaks (observed at ~383 cm$^{-1}$ and ~408 cm$^{-1}$).

Method 2: Analysis of low-frequency modes (interlayer)

Figure 3a: A Raman map of MoS$_2$ with different number of single layers based on low frequency modes analysis.

The analysis is very straightforward and consists of mapping individual peaks corresponding to different number of single layers (the spectra...
in bulk MoS$_2$ \footnote{2}). With increasing number of single layers the mode at \textasciitilde383 cm$^{-1}$ shifts to lower frequencies and the mode at \textasciitilde408 cm$^{-1}$ shifts to higher frequencies (figure 2b).

To obtain the image presented at the figure 2a, first, the fitting of peaks were done to find exact peak positions, than the difference between the peak positions were calculated and imaged. When the number of layers is higher than four the frequency of both peaks converge to the bulk values, which results in decreasing capacity of differentiating between the layers of different thickness.

Conclusions

The two methods give complementary results and allow the determination of the number of layers. Method 2 (using low frequency modes) gives excellent contrast; however it does not show single layer regions (which is related to the nature of the modes, rising from interaction between at least two layers). Method 1 (using fingerprint modes) shows all the layers, but the contrast is poorer, particularly for higher numbers of layers. The best result can be obtained combining the two methods:

Experimental details

All the measurements (low-frequency and fingerprint) were done using ultra-low frequency ULF$^\text{TM}$ filters which allow a high throughput measurement in a full Raman range, down to <10 cm$^{-1}$ (http://www.horiba.com/scientific/products/raman-spectroscopy/accessories/ultra-low-frequency-raman-module/).

Further reading