Identification of stacking configurations in a few-layered-MoS_2 sample by low-frequency Raman spectroscopy

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Two-dimensional (2D) layered materials, such as MoS_2, are often used in electronic and optoelectronic devices as active media in the form of a few layers. The orientations of different layers and their stacking configurations alter the crystal symmetry, thereby modifying the band structure and the electronic properties of the device. Experimental studies have found the onset of a gate-tunable superconducting and insulating phase in bilayer graphene at a specific layer orientation angle [1]. Since MoS_2 also has layer stacking, and layer number dependent properties, a thorough understanding of the stacking sequences and the number of layers is essential for a full exploration of their potential. Raman spectroscopy serves as a non-destructive method for the identification of various configurations of layer stacking and for the determination of the number of layers in MoS_2.

Widely studied Raman modes of MoS_2 are the high-frequency (HF) modes, A_{1g} and E_{2g}, which arise due to the out-of-plane and the in-plane vibrations of atoms within each layer, respectively, as shown in Fig.1(a). These two modes shift in opposite directions with an increase in the number of layers. However, since the HF modes are primarily dominated by the chemical covalent bonds between the atoms, van der Waals force between the layers have little effect on them, making them insensitive to interlayer coupling, and hence, to the stacking configuration of the layers. On the other hand, the low-frequency modes (LF) that solely originate from the interlayer oscillations can be used for the identification of the stacking order and for probing interfacial qualities. In addition, they show a better response than HF modes to the number of layers. As shown in Fig. 1(b), shear and breathing modes correspond to the in-plane and the out-of-plane vibrations of entire layers, respectively. These modes originate from the weak interlayer van der Waals interactions, hence, they appear in the extreme low-frequency range in the Raman spectrum. Since they lie very close to the Rayleigh scattered light, it is difficult to detect them using conventional Raman spectroscopic techniques. In order to observe the LF Raman modes, we utilized a pair of Bragg notch filters (Fig.2) that effectively filters out the laser wavelength down to a few wavenumbers. An optical setup with Bragg notch filters and a piezo scanning stage makes it possible to obtain LF Raman images. In this research, we identified different stacking configurations, the number of layers, and the interfacial qualities in a few-layered-MoS_2 sample using low-frequency Raman spectroscopy. When incorporated with Tip-enhanced Raman spectroscopy (TERS), the low-frequency TERS can image MoS_2 at very high spatial resolution. This combined technique would probe different stacking registries over nanoscale dimensions of the sample and unveil the changes in stacking defects and non-uniformity of the MoS_2 layers on a nanoscale [2]. Our technique to investigate interlayer interactions would help improving the quality of MoS_2 based devices for their better performances.

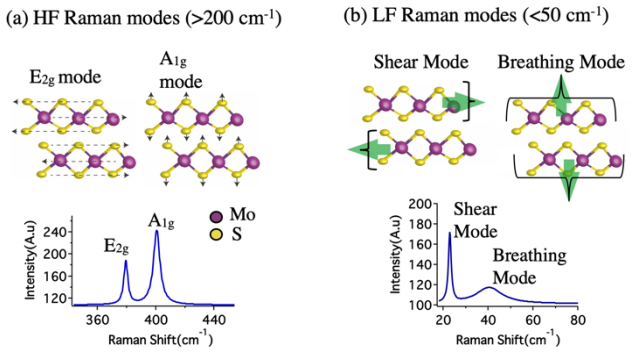

Fig. 1:(a) Schematic of the HF E_{2g} and A_{1g} modes in a two layered MoS_2 sample and a corresponding Raman spectrum. **(b)** Schematic of the LF shear and breathing modes in a two-layered MoS_2 sample and a corresponding Raman spectrum.

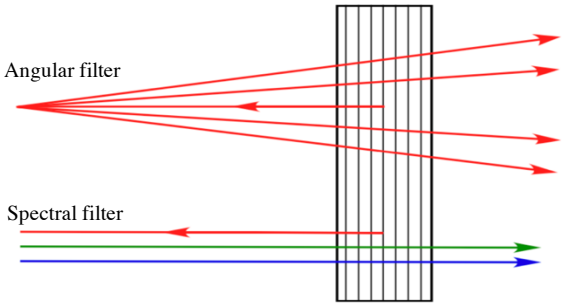

Fig. 2: Schematics of a Bragg notch filter. It is a narrow band spectral and angular filter that allows selection of a narrow spectral region of light within an acceptance angle determined by the Bragg conditions.

References

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Acknowledgements

This research was supported by JSPS Core-to-Core Program of Osaka University.