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Polarized Raman study of CVD-grown MoS_2 layered structures

We report polarized Raman spectroscopy of a few layer MoS_2. The MoS_2 layered structures were grown using a chemical vapor deposition (CVD) method and transferred to a SiO_2 substrate. Optical and atomic force microscope images showed that small multilayer region was formed at the center area of a triangular-shaped basal monolayer. The multilayer region consisted of deformed-triangular-shaped bilayer and trilayer MoS_2 stacked one by one with decreasing their sizes. Bulk MoS_2 with a thickness of ~ 40 nm was formed at the center region. Representative Raman spectrum obtained from monolayer MoS_2 revealed the in-plane and out-of-plane E_{2g}^1 and A_{1g} phonon modes at approximately 385 cm^{-1} and 405 cm^{-1}, respectively. In addition, the 2LA(M) phonon mode was observed at 450 cm^{-1}. To investigate polarization anisotropies of the individual optical phonons with increasing the number of layers, polarized Raman scattering measurements were performed on the monolayer, bilayer, and trilayer MoS_2 films. Polarized Raman results of monolayer MoS_2 showed that the A_{1g} intensity exhibited strong scattering anisotropy when the angle of the incident laser polarization varied with respect to the scattered polarization. The intensity of the A_{1g} phonon mode was a maximum (minimum) when the incident polarization was parallel (perpendicular) to the scattered polarization. The 2LA(M) phonon mode showed the similar polarization-angle dependent behavior. The E_{2g}^1 phonon mode in monolayer MoS_2 did not show any noticeable changes in its intensity as a function of the incident polarization angle. The angle-resolved intensity profiles of the A_{1g} and E_{2g}^1 phonon modes were in excellent agreement with the calculations based on the Raman polarization selection rules. Interestingly, however, the angle-resolved Raman intensities of the E_{2g}^1 phonon mode for bilayer and trilayer MoS_2 showed a deviation from the polarization selection rules. For example, the polarization anisotropy of the E_{2g}^1 phonon mode was increased systematically with increasing the number of layers. The 2LA(M) phonon mode also showed a deviation from the polarization selection rules. It is noted that the deformed-triangular-shaped MoS_2 multilayer edges were not in parallel to the basal monolayer edge, indicating the presence of a slight interlayer twist. Therefore, we suggest that these anomalous polarized Raman results of multilayer MoS_2 are due to the presence of a stacking misalignment between MoS_2 layers. Our study suggests that the polarized Raman spectroscopy can provide valuable information on the stacking disorder in two-dimensional transition-metal dichalcogenide semiconductors.

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