Asymmetric composition dependence of lattice dynamics in MoS$_{x}$Se$_{2-x}$ layers

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The mixed layered crystals allow continuous tunability of the optical band gap, rendering 2D transition metal dichalcogenide alloys appealing materials for electronic and optoelectronic device applications. Hence, characterization of these systems including understanding of the arrangement of substituting atoms in one or more layers has become fundamentally important.

We report on comparative Raman scattering studies of monolayer and bulk MoS$_{x}$Se$_{2-x}$.

Contrasting reconstruction of the phonon spectrum upon dilution of one chalcogenide by the other ($x=0^{+}$ vs $2^{-}$) as well as apparently discontinuous features around $x=1$ (MoSSe) are found.

Our single- and few-layer flakes of MoS$_{x}$Se$_{2-x}$ were obtained by mechanical exfoliation from bulk crystals grown by chemical vapor transport (CVT). Flakes with single-layer regions were identified using atomic force microscopy (AFM) and subsequently by observation of strong photoluminescence (PL).

The results for bulk MoS$_{x}$Se$_{2-x}$ reveal two distinct sets of features related to the $E_{2g}^{1}$ and $A_{1g}$ modes of pure members of series [1]. As composition $x$ changes, the in-plane $E_{2g}^{1}$ mode shows a two-mode behavior, whereas the out-of-plane $A_{1g}$ mode presents more complex evolution (Fig. 1a). When $x$ slightly increases we can identify four Raman peaks in the range 200-280 cm$^{-1}$. They evolve in different manner as composition changes, and we divided them into two types related to different distribution of the chalcogenide atoms within the MoS$_{x}$Se$_{2-x}$ layers (Fig. 1b). The first type corresponds to the Se-Se case and originates at the frequency of MoSe$_{2}$-like $A_{1g}$ mode. The second type at higher frequencies with regard to $A_{1g}$ mode is related to the Se-S case. Among these we can identify at least three components, shifting to higher frequency upon increasing $x$. They are well resolved for $0.2 < x < 0.4$, when the 2Se$_{2}$ + 1SeS, 2Se$_{2}$ + 1S$_{2}$ or 2Se$_{2}$ + 2SeS arrangements are the most probable. The frequencies of these modes are not significantly affected by reducing thickness down to a monolayer, suggesting that series of the Se-S modes are related to intralayer (rather than interlayer) interaction. However, the frequency of the Se-Se $A_{1g}$ peak increases with increasing layer number by about 1.5 cm$^{-1}$. In contrast, the $E_{2g}^{1}$ peak red-shifts with increasing the number of layers (Fig 1c).

Fig.1 (a) The polarized Raman spectra of MoS$_{x}$Se$_{2-x}$, (b) The top and edge views of atomic structure, (c) The Raman spectra on monolayer and bulk structures for $x=0.4$.