Identification and behavior of GaSe Raman modes as a function of polarization and thickness down to the monolayer

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Abstract

Gallium selenide is a layered metal chalcogenide compound with peculiar but attractive optical properties that could prove interesting for various optoelectronic applications. Its very high d_{22} coefficient, along with its optical transparency from 0.65 to 18 µm and high optical damage threshold, makes it an ideal material for non-linear optical applications. The band structure of bulk GaSe presents a pseudo-direct bandgap, with the direct transition sitting 20 meV above the indirect one, allowing for efficient photoconductivity and photoluminescence. This bandgap transition also presents highly anisotropic selection rules, forbidding light emission perpendicularly to the basal plane, which could allow optical control of charge polarization.

In this study, we have analyzed the angular and polarization dependence of the Raman signal from 2D GaSe flakes of varying thicknesses, down to the monolayer. These results are examined in the light of the few-layer structure symmetry and associated Raman tensors, clarifying the nature of all Raman-allowed vibrational modes. We confirmed the nature of the three major Raman peaks A_1^1, E'_{TO} and A_1^4, and further studied the two LO modes near 240 cm^{-1}, which present a peculiar polarization and *k*-vector depedency that is not compatible with their commonly assigned nature. We also present a rapid means of determining the crystalline quality and oxidation degree of a sample via Raman measurements, and we clearly identify raman modes from three different oxydation products of GaSe. This proves essential as thin flakes tend to photo-oxidize rapidly [1].

References

[1] T. Beechem, B Kowalski, M. Brumbach et al., Applied Physics Letters, **107** (2015) 173103.

Figures