Resonant exciton scattering reveals Raman forbidden phonon modes in layered GeS

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Germanium monosulfide with an anisotropic puckered crystalline structure has recently attracted much attention due to its unique optical and electronic properties [1]; however, exciton–phonon interactions were only superficially elucidated.

Here, we study the resonant Raman scattering and the photoluminescence of the optically active Γ-exciton in layered GeS flakes and evaluate the exciton and phonon responses on variations in the excitation energy, laser-light and emission polarizations, temperature, and laser power. A double-resonance mechanism allows for observing Raman forbidden (dark) first- and second-order longitudinal-optical phonon modes [2] whose symmetries and energies are moreover calculated by density functional perturbation theory. For (quasi)-resonant exciton excitation, the selection rules become relaxed so that a fourth-order Fröhlich intraband process is mediated by the scattering of the electron with a longitudinal-optical and an acoustic phonon. Our results demonstrate considerable coupling between phonons and photogenerated carriers in GeS flakes and the high efficiency of multiorder scattering in optical processes.

[1] C.H. Ho et al., *Adv. Optical Mater* **5**, 1600814 (2017).
[1] J. Jadczak et al., *The Journal of Physical Chemistry Letters* **14** (17), 3986-3994 (2023).