A 2DCC Story: Phonons and Excitons in ZrS2 – ZrSe2 Alloys

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Zirconium disulfide (ZrS_2) and zirconium diselenide ($ZrSe_2$) are promising 2D materials for future optoelectronics due to their indirect band gaps and predicted strain-dependent optical properties. Alloying these materials to produce ZrS_xSe_{2-x} ($x = 0...2$) enables continuous tuning of band offsets and the optical absorption spectrum, both of which are necessary for device engineering. Prior to the 2DCC, the ZrS_xSe_{2-x} alloy series had not been fully synthesized or explored. Here we report on the synthesis [1], properties [2], and oxidation [3] characteristics of ZrS_xSe_{2-x}. Studies of the spontaneous oxidation of ZrS_xSe_{2-x} shows the process is rapid and increases with Se content, resulting in a collapse of the van der Waals gap. Low-temperature Raman spectroscopy and room-temperature spectroscopic ellipsometry (SE) measurements provide information about the role of disorder and substitutional doping on ZrS_xSe_{2-x} phonons and excitons. By comparing with density functional theory (DFT) calculations, we find that the Raman spectrum for large values of x is dominated by infrared active phonons. SE measurements of the complex refractive index show a red-shift of the exciton transitions with increasing Se and strong light–matter interactions with low optical loss in the near infrared. These results are promising for future integration of ZrS_xSe_{2-x} in optoelectronic devices.

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[1] J. J. Fox, S. Bachu, R. L. Cavalero, R. M. Lavelle, S. M. Oliver, S. Yee, P. M. Vora, N. Alem, and D. W. Snyder, *Chemical Vapor Transport Synthesis, Characterization and Compositional Tuning of ZrS_xSe_{2-x} for Optoelectronic Applications*, Journal of Crystal Growth **542**, 125609 (2020).

[2] S. M. Oliver, J. J. Fox, A. Hashemi, A. Singh, R. L. Cavalero, S. Yee, D. W. Snyder, R. Jaramillo, H.-P. Komsa, and P. M. Vora, *Phonons and Excitons in $ZrSe_2$–ZrS_2 Alloys*, J. Mater. Chem. C **8**, 5732 (2020).

[3] S. S. Jo et al., *Growth Kinetics and Atomistic Mechanisms of Native Oxidation of ZrS_xSe_{2-x} and MoS_2 Crystals*, Nano Lett. **20**, 8592 (2020).