Optical Properties of Janus Transition Metal Dichalcogenide Monolayers WSSe and MoSSe

Marko M. Petrić[1]*
Matteo Barbone[1], Malte Kremser[2], Viviana Villafañe[1], Amine Ben Mhenni[2], Ying Qin[3], Yasir Sayyad[3], Yuxia Shen[3], Sefaattin Tongay[3], Giancarlo Soavi[5], Jonathan J. Finley[2], Andrés R. Botello-Méndez[4], and Kai Müller[1]
[1]*Walter Schottky Institut and Department of Electrical and Computer Engineering and MCQST, Technische Universität München, Am Coulombwall 4, 85748 Garching, Germany*
[2]*Walter Schottky Institut and Physik-Department, Technische Universität München and MCQST, Am Coulombwall 4, 85748 Garching, Germany*
[3]*Materials Science and Engineering, School for Engineering of Matter, Transport and Energy, Arizona State University, Tempe, Arizona 85287, USA*
[4]*Universidad Nacional Autónoma de México, Institute of Physics, 20-364, 01000 México, D.F., México*
[5]*Institute of Solid State Physics, Friedrich Schiller University Jena, Jena 07743, Germany*
*Marko.Petric@wsi.tum.de

Janus transition metal dichalcogenides (TMDs) lose the horizontal mirror symmetry of ordinary TMDs, leading to the emergence of additional features, such as native piezoelectricity, Rashba effect, and enhanced catalytic activity [1]. Raman, photoluminescence (PL) and second-harmonic generation (SHG) spectroscopy are essential nondestructive, phase- and composition-sensitive tools used to monitor the synthesis of materials, get insights into their excitonic properties, investigate nonlinear optical response, etc. However, comprehensive and complete study of the fundamental optical properties of Janus monolayers is still missing. Here, we discuss the Raman spectra of WSSe and MoSSe measured at room and cryogenic temperatures, near and off resonance [2]. By combining polarization-resolved Raman data with calculations of the phonon dispersion and using symmetry considerations, we identify the four first-order Raman modes and higher-order two-phonon modes (Figure 1). Moreover, we observe defect-activated phonon processes, which provide a route toward a quantitative assessment of the defect concentration and, thus, the crystal quality of the materials. Using PL spectroscopy, we study the excitonic properties of these materials. We also confirm that monolayers of WSSe and MoSSe inherit nonlinear optical response of their parent material when excited by in-plane polarized light. Furthermore, widely tuneable pulsed excitation reveals their rich electronic structure. Our work establishes a solid background for future research on material synthesis, study, and application of Janus TMD monolayers.

References

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Figures

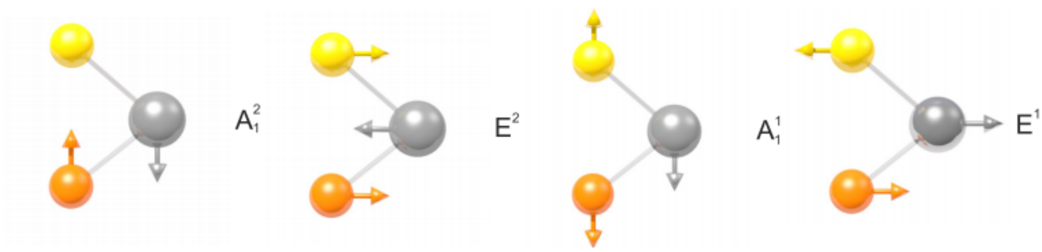

Figure 1: Schematic representation of the atomic vibrations at the center of the Brillouin Zone. Transition metal, selenium, and sulfur atoms are identified by gray, orange, and yellow, respectively.