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Observation of anomalous Stokes versus anti-Stokes ratio in MoTe2 atomic layers[1] THOMAS GOLDSTEIN, SHAO-YU CHEN, Department of Physics, University of Massachusetts Amherst, DI XIAO, Department of Physics, Carnegie Mellon University, ASHWIN RAMASUBRAMANIAM, Department of Mechanical Industrial Engineering, University of Massachusetts Amherst, JUN YAN, Department of Physics, University of Massachusetts Amherst — We grow hexagonal molybdenum ditelluride (MoTe2), a prototypical transition metal dichalcogenide (TMDC) semiconductor, with chemical vapor transport methods and investigate its atomic layers with Stokes and anti-Stokes Raman scattering. We report observation of all six types of zone center optical phonons. Quite remarkably, the anti-Stokes Raman intensity of the low energy layer-breathing mode becomes more intense than the Stokes peak under certain experimental conditions, creating an illusion of 'negative temperature'. This effect is tunable, and can be switched from anti-Stokes enhancement to suppression by varying the excitation wavelength. We interpret this observation to be a result of resonance effects arising from the C excitons in the vicinity of the Brillouin zone center, which are robust even for multiple layers of MoTe2. The intense anti-Stokes Raman scattering provides a cooling channel for the crystal and opens up opportunities for laser cooling of atomically thin TMDC semiconductor devices.

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