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**Thermal Phonon Diffraction from Atomically Rough Surfaces** NAVANEETHA K RAVICHANDRAN, HANG ZHANG, AUSTIN MINNICH, California Institute of Technology — Reflection of thermal phonons from free boundaries strongly influences the thermal resistance of thin films. Despite much effort, the specular parameter, which is the probability of specular phonon reflection, has not been experimentally measured while theory is often based on Zimans model introduced over 50 years ago. Here we report the first direct measurement of the phonon wavelength-dependent specular parameter at a free surface with atomic-scale roughness. Using the transient grating experiment on free-standing silicon membranes over temperatures from 80 - 450 K, we probe different parts of the thermal phonon spectrum by varying the grating period over length scales commensurate with phonon mean free paths. We extract the specular parameter from the measured data by using Bayesian inference to invert a transfer function based on the Boltzmann equation with ab-initio bulk phonon properties as input. We find that thermal phonons with wavelength longer and even comparable to the atomic surface roughness amplitude are frequently specularly reflected, far above the rate predicted by Zimans theory. Our work provides direct experimental insights into the interaction of phonons at rough surfaces that will impact the performance of thermoelectrics and LEDs.

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