Unveiling the Mechanism of Phonon-Polariton Damping in α-MoO_3

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Phonon polaritons (PhPs) – light coupled to lattice vibrations – in the highly anisotropic polar van der Waals material molybdenum trioxide (α-MoO_3) have recently been a subject of intense research due to their extreme subwavelength field confinement[1], directional propagation[2–4] and unprecedented low losses[1,5,6]. However, most previous studies were focused on exploiting the squeezing and steering capabilities of α-MoO_3 PhPs for controlling light at the nanoscale, without inquiring much into the dominant microscopic mechanism that determines their long lifetimes, key for their implementation in nanophotonic applications. In this work we explore the fundamental mechanisms of PhP damping in α-MoO_3 by combining *ab initio* density functional perturbation theory (DFPT) calculations with experimental scattering-type scanning near-field optical microscopy (s-SNOM) and conventional Fourier-transform infrared (FTIR) spectroscopy measurements over a wide temperature range (8 – 300 K). The excellent agreement between the experiment and the theory in reproducing the polaritonic lifetime, achieved without involving any adjustable parameters, allows us to identify third-order anharmonic phonon-phonon scattering as the main damping mechanism of α-MoO_3 PhPs. These results thus unveil the fundamental limits of low-loss PhPs, critical for validating their implementation into nanophotonic devices.

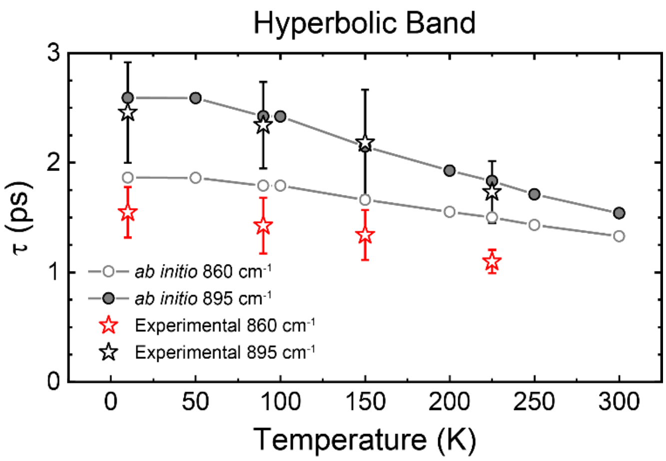

Figure 1. Temperature Dependence of the PhPs Lifetimes in α-MoO_3. Theoretical (circles) and experimental (star symbols) PhPs lifetimes for a 104nm-thick α-MoO_3 flake as a function of temperature for the hyperbolic RB (ω_0 = 860 cm^{-1} and ω_0 = 895 cm^{-1}). Gray straight lines are guides to the eye.

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