Thermal transport in nanoporous holey silicon membranes investigated with optically-induced transient thermal gratings: Supplementary Material

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LONG-TIME, APPROXIMATELY CONSTANT CONTRIBUTION TO THE SIGNAL DURING TTG MEASUREMENTS

For some of the measurements performed in this work a contribution to the measured signal at much longer timescales than the normal TTG decay was observed. The raw TTG traces for all regions investigated at all grating periods are plotted in Fig. S1, where it can be seen that the signal in some of the measurements of the patterned regions does not decay to the pre-pump baseline by the end of the TTG decay. We see that this contribution is present in the holey silicon regions at the larger grating periods studied—i.e., grating periods of 6.6 μm and 7.5 μm for region A, and 7.5 μm alone for region B. Interestingly, this long-time signal contribution does not appear in any of the TTG measurements of the unpatterned membrane. The timescales of these very slow transients (10s - 100s of μs—much longer than the acquisition time window used to capture the entirety of the "true" TTG signal but shorter than the time between pump pulses) are roughly consistent with thermal diffusion of the deposited heat at a diffusivity of α_{eff} out of the 100 μm-diameter pump spot. However, it is not clear why this contribution would be present at some grating periods while not in others, nor is it clear why such signal would be present in the heterodyned TTG signal at all. This very slow contribution to the signal is well-separated in terms of timescale from the faster decay on 10s-100s ns timescales (which we take to be the "true" TTG signal corresponding to thermal transport from the peaks to the nulls of the thermal grating), and we choose to treat it as a constant offset when fitting the faster decay to determine α_{eff}. Fig. 2(c) is recreated in Fig. S2, where the measurements corresponding to TTG traces which did not decay to the baseline over the acquisition time window are indicated with arrows. We see that the presence of this long-time signal does not have any appreciable effect on the α_{eff} values obtained, which indicates that it is a separate and independent contribution to the signal that has no impact on the signal arising from thermal transport from the peaks to the troughs of the transient grating.

DETAILS ON DENSITY FUNCTIONAL THEORY CALCULATIONS

Parameters for the lattice dynamical calculations were obtained from DFT calculations as implemented in VASP[1–4]. The calculations were performed on the primitive unit cell, with

a 5 x 5 x 5 supercell, a 500 eV plane wave energy cutoff, and a choice of AM05 function for treating exchange correlation[5,6]. The phonon mean free paths were calculated at $T = 300$ K with the TDEP[7] package in the relaxation time approximation on a 70 x 70 x 70 q-point grid, assuming natural isotope distribution. The mean free paths is given by $|\mathbf{v}\tau|$, where $\mathbf{v}$ is the group velocity and τ is the scattering time. The latter is computed based on third-order force constants and isotope disorder scattering, while the group velocity is computed from the phonon dispersions. Details can be found in Ref.[8].

UNCERTAINTES IN COMPUTED THERMAL CONDUCTIVITY VALUES

We have investigated the uncertainty in the κ_{eff} values obtained from the OpenBTE and MC-BTE methods. OpenBTE features both spatial and angular discretization in solving the BTE, resulting in numerical errors. To mitigate this effect, we checked convergence in both spatial and angular domains up to error less than 10^{-6} in κ_{eff} between successive iterations, and estimated the uncertainty due to discretization to be less than 3%. For MC-BTE, the standard error due to the stochastic nature of the technique was less than 1%, and the systematic error was no greater than 5%.

The uncertainty in computed κ_{eff} values also contains a component that is propagated from the inputs of the DFT calculations described above. We have estimated the computational uncertainties associated with discretization of momentum space in the DFT calculations on the calculated thermal conductivity values to be $\sim$10%. We note that some uncertainties, e.g., associated with choices of computational parameters in the DFT calculations[9] (pseudopotential, exchange correlation functional, etc.) remain outside the scope of our analysis.

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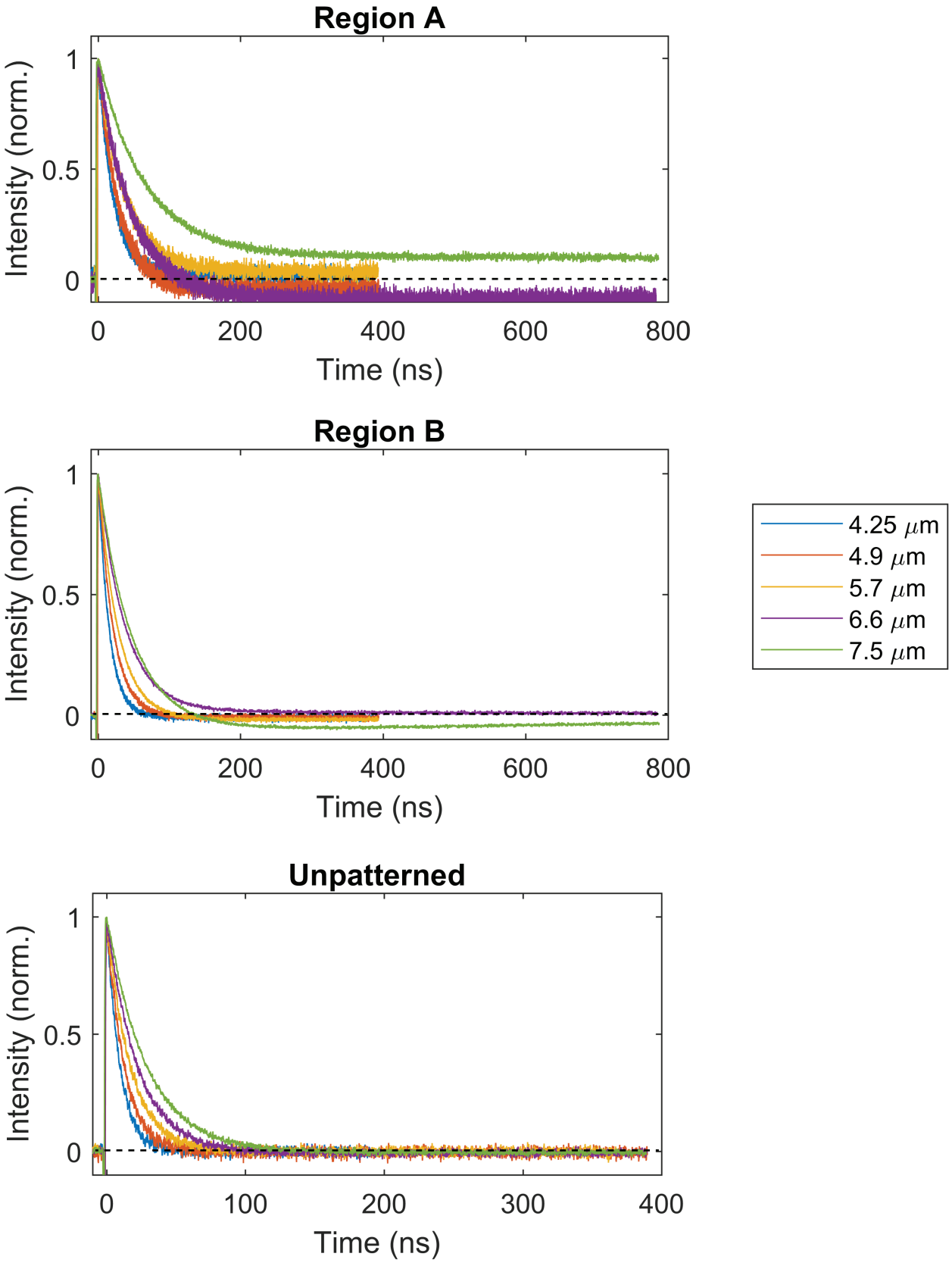

FIG. S1. Normalized TTG traces obtained for all regions at every grating period measured, with baselines set to the pre-pump values.

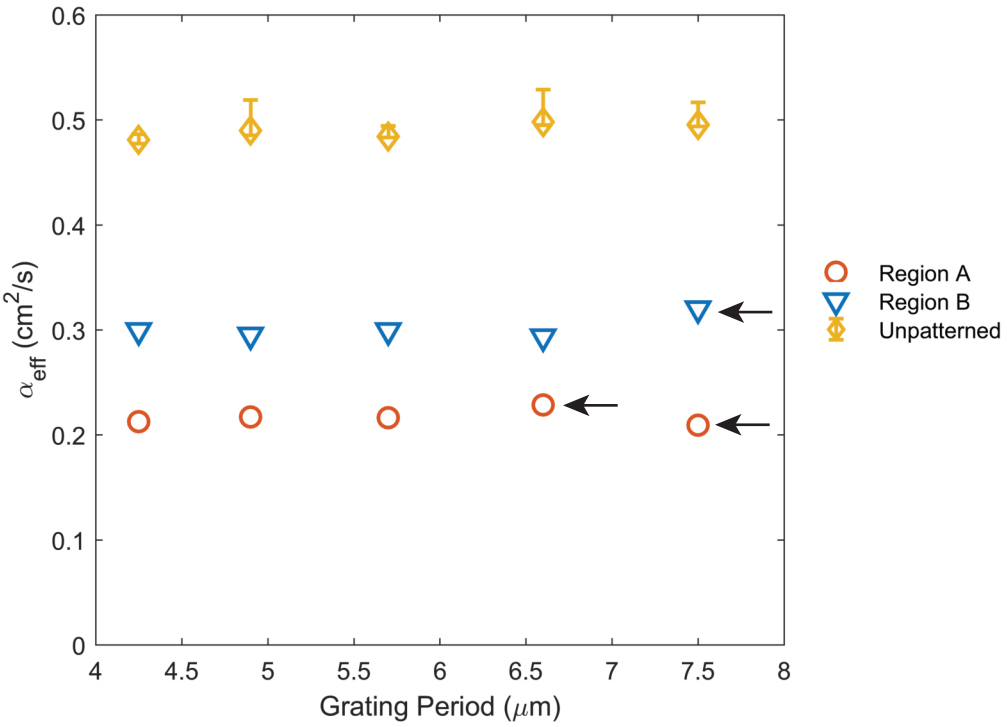

FIG. S2. α_{eff} values determined from the data in Fig. S1, where arrows correspond to measurements where the signal at the end of the acquisition time window remains $> 5\%$ maximum amplitude away from the pre-pump baseline values. We see that our determined values of α_{eff} are independent of the presence of this long-time contribution to the signal for all regions.