

Ultra-low thermal conductivity and acoustic dynamics of Si nanostructured metalattices probed using ultrafast high harmonic beams

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Abstract. We extend optical nanometrology capabilities to smaller dimensions by using tabletop coherent extreme ultraviolet (EUV) beams. Specifically, we characterize thermal transport and acoustic wave propagation in 3D periodic silicon inverse metalattices with <15nm characteristic dimensions. Measurements of the thermal transport demonstrate that metalattices may significantly impede heat flow, making them promising candidates for thermoelectric applications. Extraction of the acoustic wave dispersion down to ≈ 100 nm shows good agreement with finite element predictions, confirming that these semiconductor metalattices were fabricated with a very high-quality. These results demonstrate that EUV nanometrology is capable of extracting both dispersion relations, and thermal properties of 3D complex nano-systems, with applications including informed design and process control of nanoscale devices.

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1 Introduction

Nanoscale metamaterials make it possible to engineer the transport, electronic and magnetic properties of materials, which is critical for nanoelectronics, thermoelectric and data storage devices [1]. Nanoscale inverse opal metalattices combine precise nanostructuring with long range order by infiltrating various materials into nanosphere opal templates (see Fig. 1). This enables unique properties not accessible otherwise [2].

While modern nanofabrication techniques can synthesize these complex systems at nanometer length scales, most current characterization techniques struggle to probe them. Charge, spin and phonon dynamics can be extremely fast on nanoscale dimensions and most widely-used optical characterization techniques suffer from the diffraction limit of visible light. Therefore, better functional nanometrology techniques must have fs to ps temporal resolution, as well as nm spatial resolution.

In this work, we extend ultrafast short wavelength high harmonic nanometrology to probe thermal and acoustic dynamics in novel silicon metalattices - which are polycrystalline silicon infiltrated into a self-assembled 14-30nm face-centered cubic silica nanosphere templates to form a nanoscale inverse opal (Fig. 1). Our data indicate an extremely low thermal conductivity in the metalattice due to impeded phonon transport. The short wavelength (≈ 30 nm) and pulse duration (≈ 10 fs) of the tabletop high harmonic generation source match the intrinsic length- and time-scales of the relevant nanoscale dynamics, making it an ideal probe of nanostructured systems [3, 4].

2 Experimental section: EUV nanometrology

We first deposit arrays of periodic nickel nanolines on top of the Si metalattices with a grating periodicity, P , ranging from 4000nm to 120nm and grating linewidths, L , from 1000nm down to 30nm. A ≈ 25 fs laser pump pulse then heats the Ni gratings, launching thermal and acoustic dynamics in the silicon metalattice. After a controlled delay, a 30nm wavelength extreme ultraviolet (EUV) beam diffracts from the nanogratings to simultaneously probe thermal transport into the metalattice, and surface acoustic waves (SAWs) confined in the metalattice [5]. In our EUV nanometrology (Fig. 1), we monitor the change in diffraction efficiency as a function of pump-probe time delay to extract these surface dynamics.

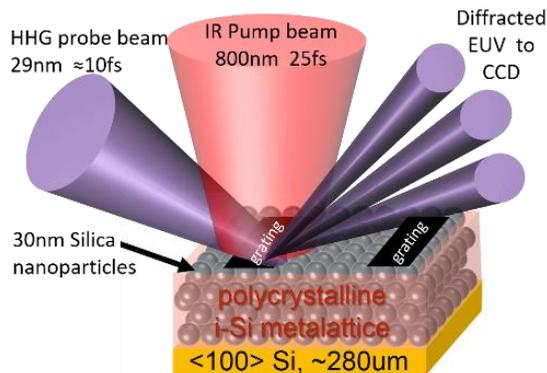


Fig. 1. EUV nanometrology of a silicon metalattice with silica spheres of 14–30nm. An IR laser excites the sample, and an ultrafast HHG beam then probes the thermal and acoustic response.

3 Thermal transport and acoustic dynamics of Si metalattices

The change in the EUV diffraction efficiency as a function of the pump-probe time delay (Fig. 2A), shows the thermal relaxation through the metalattice together with the superimposed acoustic oscillations. From Fig. 2A, we can clearly see that none of the heated gratings can fully cool during the time window of our measurement, which was up to 8ns. This is in contrast to the case when Ni gratings are fabricated on bulk Si substrates with a similar intrinsic thermal boundary resistivity, which instead cool within 1ns [4]. This suggests poor thermal conduction through the Si metalattice and therefore an extremely low thermal conductivity. Additionally, by using equilibrium Green–Kubo atomistic simulations (see Fig.2B) we observe that silicon metalattices reduce thermal conductivity by two orders of magnitude below the prediction of continuum calculations. Although these simulations were performed for metalattices with no silica spheres inside and with a cavity size of 2nm, the calculated values also support our experimental findings. This suggests that nanostructured metalattices may impede heat flow even more than had been initially thought. The ability to impede the flow of phonons, while allowing current to flow, can dramatically impact applications such as optimized thermoelectric materials.

An analysis of the acoustic oscillations from Fig. 2A also enabled a unique capability to probe the dispersion relation of this nanosystem, which is shown in Fig. 2C. From a finite element analysis (FEA), we extracted the phonon dispersion via continuum mechanics and observe that the predicted SAW dispersion is in excellent agreement with the experimentally measured values. This finding not only confirms that EUV nanometrology is capable of extracting the dispersion relation of complex materials, but also shows that these silicon metalattices were fabricated with very high-quality.

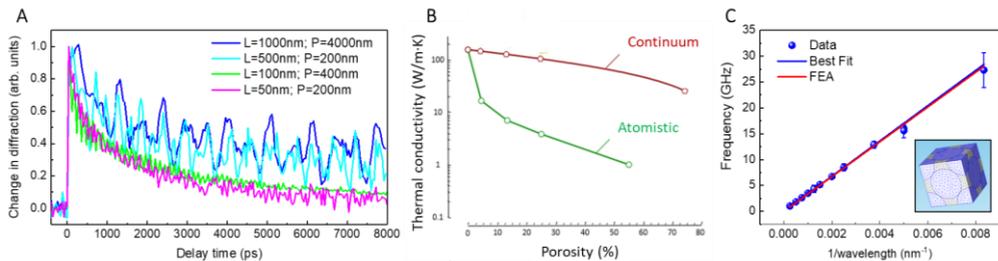


Fig. 2. Thermal transport and acoustic dynamics of a 30nm Si metalattice A) Change in diffraction as a function of pump-probe delay for gratings with linewidths of 1000-50nm. B) Equilibrium Green–Kubo atomistic simulations of Si metalattices. The thermal conductivity is highly reduced as the porosity increases compared to continuum mechanics. C) From the SAWs launched, we extract a dispersion relation which is in excellent agreement with theory. Inset shows the simulated structure.

References

1. M. Maldovan, *Nature* **503**, 209–217 (2013).
2. J.E. Han, V.H. Crespi, *Phys. Rev. Lett.* **86**(4), 696-699 (2001).
3. M. E. Siemens, Q. Li, R. Yang, K.A. Nelson, E.H. Anderson, M.M. Murnane, H.C. Kapteyn, *Nat. Mater.*, **9**(1), 26–30 (2010).
4. K. M. Hoogeboom-Pot, J.N. Hernandez-Charpak, X. Gu, T.D. Frazer, E.H. Anderson, W. Chao, R.W. Falcone, R. Yang, M.M. Murnane, H.C. Kapteyn, D. Nardi, *Proc. Natl. Acad. Sci.*, **112**(16), 4846–4851 (2015).
5. A. Rundquist, C.G. Durfee, Z. Chang, C. Herne, S. Backus, M. Murnane, H. Kapteyn, *Science* **280**(5368), 1412–1415 (1998).