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Polariton-Mediated High Harmonic Generation with Co-Optimized Metasurfaces

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Abstract: We present a co-optimization strategy using nonlinear polaritonic metasurfaces coupled to few-layer hBN to enable polariton-mediated fifth-harmonic generation, where hexagonally packed silicon-on-sapphire resonators yield simultaneous mid-IR and near-IR resonances and predict ~1100-fold efficiency enhancement. © 2026 The Authors.

1. Introduction

High-harmonic generation (HHG) is an extreme nonlinear optical (NLO) process when intense, ultrashort laser pulses interact with an NLO material and produce photons at integer multiples of the excitation laser frequency.[1,2] HHG has produced light sources spanning across far-infrared to ultraviolet (UV) for their applications in attosecond light pulses, extreme UV and near-IR (NIR) imaging and spectroscopy, investigations of quantum phase transitions and observing fundamental charge dynamics in matter with high-photon-energy radiation.[3] HHG in traditional NLO crystals and solid-state materials observes lower generation efficiency for various reasons, including lack of broadband phase-matching, as well as low local fields and damage thresholds. Hexagonal boron nitride (hBN), which is a 2D van der Waals (vdW) material, is a promising material platform for HHG[4] with a large bandgap of 6 eV, allowing for generation of a large range of harmonics and high thermal conductivity, making the material more resistant to laser damage. Moreover, hBN's Reststrahlen bands (RBs) are in the 7–8 μm range,[5,6] facilitating MIR-driven HHG.

In this work, we report a novel co-optimization methodology based on hBN coupled to silicon-on-sapphire (SOS) nonlinear photonic metasurfaces (MSs) for generation of enhanced high harmonics. Our methodology tailors a single metasurface to simultaneously resonate to both MIR (pump) and NIR (HHG) frequencies at well-overlapped dipole-like modes. This results in boosting the HHG efficiency through generation of phase-matched localized fields which greatly enhances the emission of the fifth harmonic (FHG) by three orders of magnitude, enabling a novel polariton-mediated HHG mechanism. These results open opportunities towards developing first-of-its-kind on-chip and efficient UV and NIR light sources.

2. Results and discussion

Figure 1a illustrates polariton-mediated HHG process whereby an intense femtosecond MIR laser pulse excitation interacts with a few-layer film (14 nm) of hBN draped over a dielectric MS based on SOS. For fabrication, electron beam lithography and reactive ion etching are used to create hexagonally packed silicon nanoresonators with 900 nm height and 408 nm diameter, as shown in close-up and survey SEM images and Fig. 1b and c, respectively. We use a wet-chemical assisted transfer[7] to drape and cover large 150 μm^2 MS devices with hBN. Figure 1d shows an atomic force microscope image of the hBN-draped SOS MS, suggesting the draping of large area hBN in contact with the MS without any visible damage. This uniform integrity of the draped hBN is also shown in Fig. 1e with an optical image recorded during Fourier transform infrared spectroscopy (FTIR). We show the NIR characterization in Fig. 1f where the calculated and experimentally recorded a mode is captured at λ_{FHG} = 1.42 μm. Fig. 1f shows a good match of the experimentally recorded NIR resonance with finite-difference time-domain (FDTD) calculations, with the exception of the resonance linewidth, which can be explained with the sample imperfection and microscope's finite numerical amplitude. The inset shows a dipolar electric-field distribution along the Z-axis with 6-fold enhanced localization. In Fig. 1g-h, we show calculated and FTIR-recorded MIR phonon-polaritonic (PhP) modes from different hBN-draped MSs with varying inter-resonator gaps (g). Figure 1g, shows the FDTD-calculated PhP mode at λ_{pump} = 7.1 μm, so that the PhP mode is spectrally co-optimized to be at the fifth harmonic of the NIR photonic mode, along

with hBN's transverse-optical (TO) phonon mode centered around 7.3 μm. Figure 1h shows FTIR-recorded MIR spectra from five different devices with varying *g*, showing a systematic emergence of the PhP mode for the gap varied from 142 nm to 150 nm. The solid line in black shows a control measurement suggesting that in the absence of MS, the mode at 7.1 μm disappears and only the intrinsic TO-phonon mode remains.

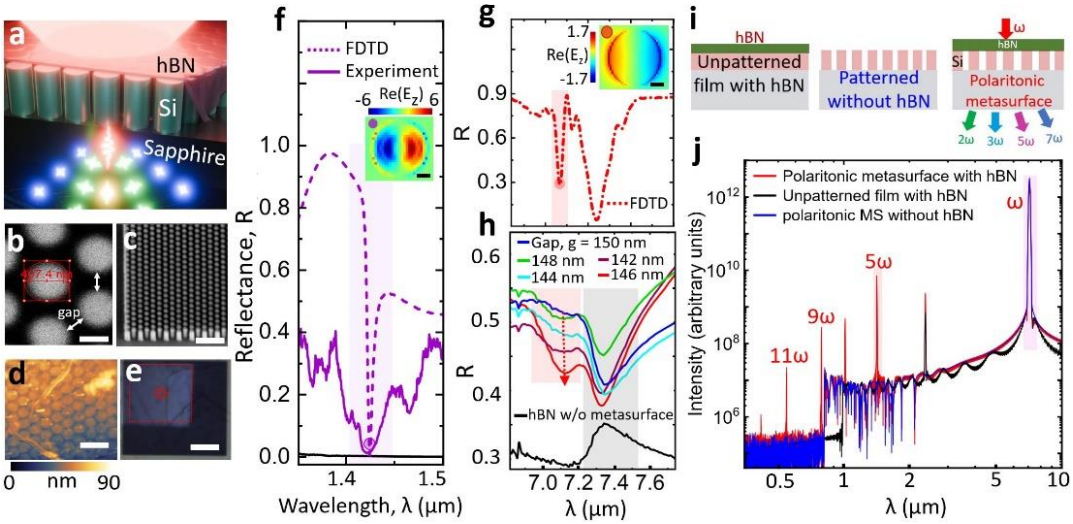


Fig. 1: (**a**) Artistic rendering of polariton-mediated high harmonic generation (HHG) with polaritonic metasurface (MS)-coupled hBN. (b-c) SEM image of the fabricated silicon-on-sapphire (SOS) MS without hBN that show the (**b**) top-view and (**c**) height of the hexagonally-packed circular silicon nanoresonators (SNRs). (**d**) Atomic force microscopy shows topography and (**e**) optical image of multilayered 14 nm of hBN draped on SOS MS. Scale bars in (b), (c), (d) and (e) are 400 nm, 3 μm, 1 μm and 40 μm, respectively. (**f**) Calculated and experimentally recorded near-IR photonic resonances at 1.42 μm (purple circle). Inset shows real part of electric fields along the Z-axis suggesting a dipolar mode. (**g**) Calculated phonon-polaritonic (PhP) mode at 7.1 μm. Inset shows Re (E_z) suggesting a dipolar nature of the PhP mode. (**h**) Experimentally recorded PhP modes systematically emerge by controlling the gap between the SNRs that matches with calculated spectrum shown in (g). Black solid line represents spectrum from few-layer hBN on unpatterned SOS substrate. (**i-j**) Calculations considering (i) three different cases suggest that (j) only by adding a thin, 14 nm layer of hBN onto MSs can lead to ~1100-fold enhancement in the fifth harmonic generation.

The FTIR measurements from the MSs provides the basis for high-harmonic experiments. For understanding the prospects of HHG enhancement, we performed nonlinear HHG calculations by employing the structural parameters from the fabricated MSs. For three configurations in Fig. 1i, we show results of HHG calculations in Fig. 1j, suggesting that without the co-optimized MSs, the hBN multilayer alone would not provide FHG enhancement. It also suggests that the SOS MS by itself without any hBN cannot generate any HHG either. Interestingly, only for the case when the co-optimized SOS MS are coupled with a multilayered hBN, the enhancement in FHG reaches a ~1100-fold improvement in HHG efficiency. This hints that upon intense MIR pump excitation, the PhP mode at 7.1 μm generates its' fifth harmonic at 1.42 μm, which overlaps and gets phase-matched with the NIR photonic mode, boosting the FHG efficiency.

Overall, these results establish polariton-mediated FHG as a viable mechanism for dramatically enhancing nonlinear optical emission in hybrid metasurface–vdW systems. The demonstrated three-orders-of-magnitude efficiency boost highlights the impact of simultaneously tailoring MIR and NIR resonances within a unified structure. These insights lay the groundwork for scalable devices that leverage phonon–polariton physics for advanced spectroscopy and integrated photonics.

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