

Hybrid Metasurface for Efficient & Broadband Second Order Nonlinear Processes

R. Sarma¹, D. de Ceglia², N. Nookala³, M. A. Vincenti⁴, S. Campione¹, O. Wolf¹, M. Scalora⁵, M. Belkin³, and I. Brener¹

¹Sandia National Laboratories, Albuquerque, NM, USA

²Department of Information Engineering, University of Padova, Padua, Italy

³University of Texas at Austin, Austin, TX, USA

⁴Department of Information Engineering, University of Brescia, Brescia, Italy

⁵Charles M. Bowden Research Laboratory, US Army AMRDEC, Redstone Arsenal, AL, USA

Abstract: Second harmonic generation (SHG) is one of the most studied nonlinear optical phenomenon thanks to its extensive practical applications. Efficient SHG can be achieved by either improving light-matter interaction or searching for materials with higher nonlinear susceptibility values. Together with nonlinear process efficiency, low losses and damage thresholds are desirable, as well as large operating bandwidth, scalability, integrability and ease of fabrication. We demonstrate that a hybrid approach that combines dielectric metasurfaces with semiconductor quantum well can satisfy all the aforementioned conditions, paving the way for novel designs of low loss, broadband and efficient ultrathin nonlinear optical devices.

Since the advent of laser, SHG has been extensively studied for numerous applications [1]. Because of the typically weak nonlinear response of materials, efficient SHG can be achieved only under phase matching conditions and using high power lasers. However, there is still a need to achieve efficient SHG at moderate pump intensities using ultrathin nonlinear optical devices [2]. Several approaches have been proposed, such as the use of two-dimensional transition metal dichalcogenides [3-6] and multi-quantum-well (multi-QW) semiconductor heterostructures [7-9]. In fact, ISTs allow to engineer extremely large $\chi^{(2)}$ values [10] and therefore can be used as nonlinear media for SHG in ultrathin nonlinear devices where phase matching conditions are not required. Nonlinear processes can be also boosted increasing light-matter interactions, i.e. confining electromagnetic fields to subwavelength volumes using resonators, metasurfaces or photonic crystals [11-18]. So far, the highest nonlinear response for SHG (normalized by length) by a nanostructure has been obtained at mid-infrared wavelengths using a hybrid approach which couples plasmonic metasurfaces to ISTs of semiconductor QWs [10, 19-21]. Although efficient, this approach suffers from the use of plasmonic nanostructures for coupling the pump light to the ISTs and out-coupling the second-harmonic (SH) light, which have high dissipative losses that induce heating effects and lowering of damage thresholds. Another possible strategy involves the use of Mie-like resonances in all-dielectric metasurfaces fabricated from nonlinear crystals such as GaAs [13-16]. This method solves the problems of metal absorption but they have intrinsically low $\chi^{(2)}$ and sustain only modest field enhancement. High field enhancement in all-dielectric metasurfaces can be attained by exploiting resonances with higher quality factors (Q) such as Fano resonances and bound states in the continuum. However, a higher Q significantly reduces the spectral bandwidth of the nonlinear device. The inverse relation between Q and spectral bandwidth is a fundamental limitation that dictates the bandwidth of SHG in a nonlinear device.

Here we experimentally demonstrate a new hybrid approach for high efficiency SHG that overcomes the limitations described above by combining the advantages of IST nonlinear metasurfaces and all-dielectric nonlinear metasurfaces. Our method uses high- Q leaky mode resonances (LMRs) [22-24] in dielectric nanostructures coupled to ISTs of semiconductor QWs. Our all-dielectric device has small dissipative losses and therefore a high damage threshold. Furthermore, since the LMR wavelength can be varied by changing the period of the grating, the high Q resonances can be tuned to fit the wavelength

needs of a wide range of applications. Moreover, our approach allows us to achieve high Q resonances over a large bandwidth by tuning the incident angle of the pump light. The combination of a giant χ^2 within the IST band and high Q photonic resonances far from the IST band allows to achieve a large bandwidth of SHG. Fig. 1(a) shows a sketch of the fabricated structure, while Fig. 1 (b) and (c) show the efficiency of the reflected SH simulated and measured, respectively. Although demonstrated for mid-infrared wavelengths, our approach of using LMRs coupled to ISTs can be scaled to both longer and shorter wavelengths. Finally, our device can operate in both reflection and transmission mode, is monolithically integrable, and easy to fabricate. Thus, this new system is well-suited for practical applications.

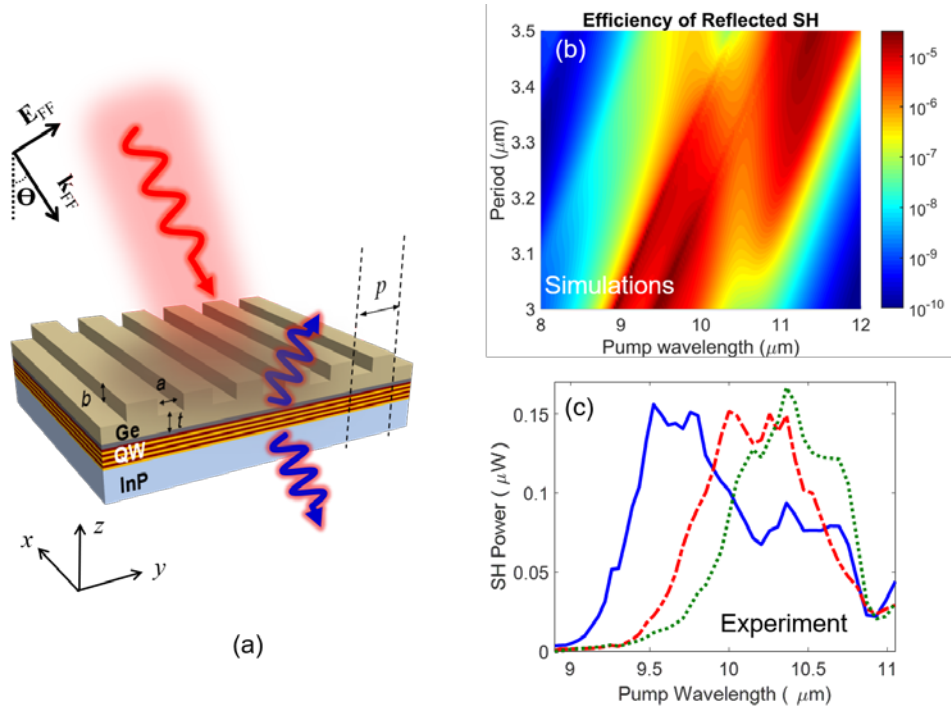


Fig. 1 (a) Sketch of the device and illumination scheme. The geometrical parameters of the fabricated devices are $a = 1070$ nm, $b = 1200$ nm, $t = 500$ nm, and $p = 3150, 3250,$ and 3350 nm (for different structures tested). The thickness of multi-QW layer is 656 nm; (b) Efficiency of reflected SH (in log 10 scale) as a function of pump wavelength and period p . Incident pump intensity is 10 kW/cm²; (c) Measured reflected SH power at normal incidence as a function of pump wavelength from structures with $p = 3.15$ μm (blue solid line), 3.25 μm (red dashed line), and 3.35 μm (green dotted line) at the incident pump intensity of approximately 3.3 kW/cm².

References

- [1] Franken, P. A.; Hill, A. E.; Peters, C. W.; Weinreich, G. Generation of Optical Harmonics. *Phys. Rev. Lett.* **1961**, 7, 118.
- [2] Krasnok, A.; Tymchenko, M.; Alu, A. Nonlinear Metasurfaces: A Paradigm Shift in Nonlinear Optics. *Materials Today* **2018**, 21(1), 8-21.
- [3] Malard, L. M.; Alencar, T. V.; Barboza, A. P. M.; Mak, K. F.; Paula, A. M. Observation of Intense Second Harmonic Generation from MoS₂ Atomic Crystals. *Phys. Rev. B* **2013**, 87, 201401(R).
- [4] Le, C. T.; Clark, D. J.; Ullah, F.; Senthilkumar, V.; Jang, J. I.; Sim, Y.; Seong, M. J.; Chung, K. H.; Park, H.; Kim, Y. S. Nonlinear Optical Characteristics of Monolayer MoSe₂. *Ann. Phys.* **2016**, 528, 551-559.
- [5] Li, Y.; Rao, Y.; Mak, K. F.; You, Y.; Wang, S.; Dean, C. R.; Heinz, T. F. Probing Symmetry Properties of Few-Layer MoS₂ and h-BN by Optical Second-Harmonic Generation. *Nano Lett.* **2013**, 13(7), 3329-3333.

- [6] Lin, K.; Weng, S.; Lyu, P.; Tsai, T.; Su, W. Observation of Optical Second Harmonic Generation from Suspended Single-Layer and Bi-Layer Graphene. *Appl. Phys. Lett.* **2014**, 105, 151605.
- [7] Fejer, M. M.; Yoo, S. J. B.; Byer, R. L.; Harwit, A.; Harris Jr., J. S. Observation of Extremely Large Quadratic Susceptibility at 9.6-10.8 μm in Electric-Field-Biased AlGaAs Quantum Wells. *Phys. Rev. Lett.* **1989**, 62, 1041.
- [8] Rosencher, E.; Bois, P.; Nagle, J.; Delattre, S. Second Harmonic Generation by Intersub-band Transitions in Compositionally Asymmetrical MQWs. *Electron. Lett.* **1989**, 25 (17), 1063-1065.
- [9] Capasso, F.; Sirtori, C.; Cho, A. Coupled Quantum Well Semiconductors with Giant Electric Field Tunable Nonlinear Optical Properties in the Infrared. *IEEE J. Quantum Electron.* **1994**, 30, 1313-1326.
- [10] Lee, J.; Tymchenko M.; Argyropoulos C.; Chen, P. Y.; Lu, F.; Demmerle, F.; Boehm, G.; Amann, M. C.; Alu, A.; Belkin, M. A. Giant Nonlinear Response from Plasmonic Metasurfaces Coupled to Intersubband Transitions. *Nature* **2014**, 511, 65.
- [11] Butet, J.; Brevet, P. F.; Martin, O. J. F. Optical Second Harmonic Generation in Plasmonic Nanostructures: From Fundamental Principles to Advanced Applications. *ACS Nano* **2015**, 9(11), 10545-10562.
- [12] Kauranen, M. Nonlinear Plasmonics. *Nat. Photonics* **2012**, 6, 737-748.
- [13] Liu, S.; Sinclair, M. B.; Saravi, S.; Keeler, G. A.; Yang, Y.; Reno, J.; Peake, G. M.; Setzpfandt, F.; Staude, I.; Pertsch, T.; Brener, I. Resonantly Enhanced Second-Harmonic Generation Using III-V Semiconductor All-Dielectric Metasurfaces. *Nano Lett.* **2016**, 16(9), 5426-5432.
- [14] Camacho-Morales, R.; Rahmani, M.; Kruk, S.; Wang, L.; Xu, L.; Smirnova, D. A.; Solntsev, A. S.; Miroschnichenko, A.; Tan, H. H.; Karouta, F.; Naureen, S.; Vora, K.; Carletti, Luca; Angelis, C. D.; Jagadish, C.; Kivshar, Y. S.; Neshev, D. N. Nonlinear Generation of Vector Beams from AlGaAs Nanoantennas. *Nano Lett.* **2016**, 16(11), 7191-7197.
- [15] Liu, S.; Keeler, G. A.; Reno, J. L.; Sinclair, M. B.; Brener, I. III-V Semiconductor Nanoresonators – A New Strategy for Passive, Active, and Nonlinear All-Dielectric Metamaterials. *Adv. Opt. Mater.* **2016**, 4(10), 1457-1462.
- [16] Gili, V. F.; Carletti, L.; Locatelli, A.; Rocco, D.; Finazzi, M.; Ghirardini, L.; Favero, I.; Gomez, C.; Lemaitre, A.; Celebrano, M.; Angelis, C. D.; Leo, G. Monolithic AlGaAs Second-Harmonic Nanoantennas. *Optics Express* **2016**, 24(14), 15965-15971.
- [17] Buckley, S.; Radulaski, M.; Biermann, K.; Vuckovic, J. Second Harmonic Generation in Photonic Crystal Cavities in (111)-Oriented GaAs. *Appl. Phys. Lett.* **2013**, 103, 211117.
- [18] Buckley, S.; Radulaski, M.; Petykiewicz, J.; Lagoudakis, K. G.; Kang, J. H.; Brongersma, M.; Biermann, K.; Vuckovic, J. Second Harmonic Generation in GaAs Photonic Crystal Cavities in (111)B and (001) Crystal Orientations. *ACS Photonics* **2014**, 1(6), 516-523.
- [19] Wolf, O.; Campione, S.; Benz, A.; Ravikumar, A. P.; Liu, S.; Luk, T. S.; Kadlec, E. A. Shaner, E. A.; Klem, J. F.; Sinclair, M. B.; Brener, I. Phased-Array Sources Based on Nonlinear Metamaterial Nanocavities. *Nature Communication* **2015**, 6, 7667.
- [20] Lee, J.; Nookala, N.; Gomez-Diaz, J. S.; Tymchenko, M.; Demmerle, F.; Boehm, G.; Amann, M. C.; Alu, A.; Belkin, M. A. Ultrathin Second-Harmonic Metasurfaces with Record-High Nonlinear Optical Response. *Adv. Optical Mater.* **2016**, 4 (5), 664-670.
- [21] Nookala, N.; Xu, J.; Wolf, O.; March, S.; Sarma, R.; Bank, S.; Klem, J.; Brener, I.; Belkin, M. Mid-Infrared Second-Harmonic Generation in Ultra-thin Plasmonic Metasurfaces without a Full-Metal Backplane. *Appl. Phys. B* **2018**, 124 (7), 124-132.
- [22] Magnusson, R.; Wang, S. S. New Principle for Optical Filters. *Appl. Phys. Lett.* **1992**, 61, 1022-1024.
- [23] Hessel, A.; Oliner, A. A. A New Theory of Wood's Anomalies on Optical Gratings. *Appl. Opt.* **1965**, 4, 1275-1297.
- [24] Fan, S.; Suh, W.; Joannopoulos, J. D. Temporal Coupled-Mode Theory for the Fano Resonance in Optical Resonators. *J. Opt. Soc. Am A* **2003**, 20, 569-572.