Nonreciprocal on the nanoscale: nonlinear generation via multipole interference

Ekaterina Poutrina[1,2] **and Augustine Urbas**[1]

[1]*Air Force Research Laboratory, Materials and Manufacturing Directorate, Wright Patterson, Ohio 45433,*[2]*UES, Inc., 4401 Dayton Xenia Rd, Beavercreek, OH 45432*

Abstract: We demonstrate that non-reciprocal directionality of nonlinear generation is inherent to and realistically observable in the nonlinear multipolar response of nanostructures. Alternatively, nonlinear multipolar interference can ensure a directionally selective inhibition of the nonlinear process.

OCIS codes: 164.4236, 290.4230, 160.4330

Probing the nonlinear response in light-matter interactions offers the advantage of lifting some fundamental limitations existing within the scope of linear operations. One of the most striking examples is the ability of breaking the reciprocity of light transmission. Light propagation through a medium characterized by a symmetric, time-independent, and linear permittivity tensor stays the same when reversing the direction of incidence, or interchanging the source and the detector [1]. The above reciprocity theorem persists even in a lossy system, where time-reversal symmetry does not hold. While serving as an innate foundation in any optical design, it presents a fundamental challenge for applications requiring a non-reciprocal directionality of light propagation.

A common technique in utilizing a nonlinear response for achieving a non-reciprocal light transmission relies on directionality-dependent local field patterns in structures possessing an asymmetry along the propagation direction, either in their geometry or in material composition [2, 3]. Placing a nonlinear element within the region of the highest contrast between the local field patterns induced by forward versus backward-propagating incident beams results in the variation of the efficiency of the transmitted light, for the excitation intensities strong enough to invoke the nonlinear response.

Here, we show that the option of non-reciprocity in the directionality and the efficiency of nonlinear generation is inherently incorporated in the response of nanoelements exhibiting magnetic dipolar (or higher-order multipolar) Mie resonance(s) in their linear response. In particular, we show that nonlinear generation from a nanoelement, where it regularly occurs nearly isotropically in all directions (due to the lack of phase matching constraints within nanoscale interaction volumes), can be made both unidirectional and non-reciprocal, such that the generation occurs *predominantly in a single direction* which, additionally, *remains unchanged* with respect to a fixed laboratory coordinate system when reversing the direction of the fundamental beam(s) [4]. In contrast with the previous studies, the proposed approach does not require asymmetry in either the geometry or the material composition of the nanoelement. Rather, it relies on the multiplicative nature of the nonlinear response which, in turn, allows a *simultaneous* change in phase of *both* the electric *and* the magnetic, *nonlinearly induced*, dipolar modes, when switching the phase of *a single* (*either* electric *or* magnetic) vector of the fundamental field. This phenomena is also qualitatively different from a directional scattering achieved via linear multipole interference at Kerker [5] condition, where the enhanced and the suppressed sides always switch when reversing the excitation beam. We discuss the limitations, brought by group symmetry [6] restrictions, on the number fundamental beams than can be decoupled in such a manner from the resulting direction of nonlinearly generated beam. Furthermore, the interference can occur between various pathways *within* the electric and magnetic (*nonlinearly* produced) dipolar modes. As a result, non-reciprocity in terms of just a change in the *efficiency* of nonlinear generation when reversing the direction of any subset of the fundamental beams is inherent to and expected in the nonlinear response of most nanoelements, even the symmetric ones, and for most of the nonlinear processes. Targeted engineering of the relative strengths of various pathways within each (nonlinearly produced) multipolar mode may then alow an interferometric cancellation of the generated field for a given nonlinear process, for certain respective directions of the fundamental beams.

We further consider a physical implementation of the suggested approach using purely symmetric dimer structures made of non-magnetic materials. We develop the retrieval procedure identifying the effective magneto-electric hyperpolarizabilities and use it to predict the parameters allowing a non-reciprocal directionality of nonlinear generation in such geometry. We confirm these predictions numerically using COMSOL Multiphysics [www.comsol.com] software package, demonstrating an excellent agreement for the theoretically-predicted optimal combinations of fundamental frequencies allowing the manifestation of the phenomena.

Reliance on multipolar interference in the suggested approach inherently assumes the manifestation of the described phenomena on the nanoscale, through the response of subwavelength-scale elements. These nanoelements can thus be used as building blocks to construct a metasurface or a medium with similar unique features in its nonlinear response. Here, we present an example of such a metasurface operating as as one-way nonlinear mirror (Fig. 1).

As follows from the above discussion, in addition to the manifestation of a magnetic dipolar (or higher-order multipolar) mode in the linear response, both non-reciprocal directionality of nonlinear generation and inhibition of the nonlinear response require a careful engineering of the respective strengths of various pathways *within* each (electric or magnetic) type of the nonlinearly produced multipolar partial wave. As such, these phenomena are not expected to manifest in natural nonlinear materials, even those possessing natural magnetic dipolar transitions. They, however, can be achieved via a tailored design of the effective magneto-electric nonlinear polarizabilities of a nanoelement.

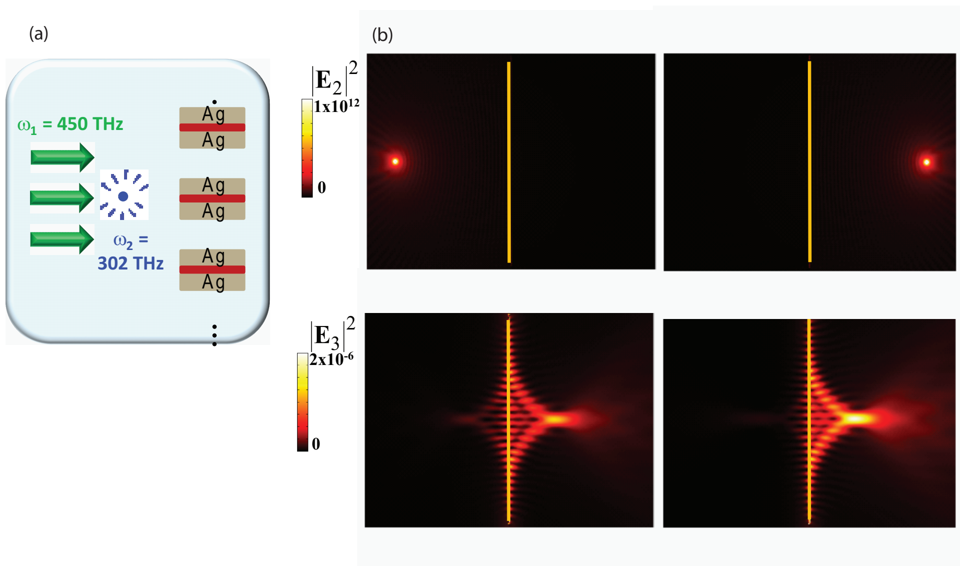

Fig. 1. One-way nonlinear mirror. (a) Metasurface geometry and excitation configuration. (b) An image of the line source at frequency ω_2 is produced via DFG at frequency $\omega_3 = 148$ THz on the right side of the metasurface, independently of the source location on the left or right. The yellow line shows schematically the metasurface location.)

References

1. L. D. Landau, E. M. Lifshits, and L. P. Pitaevskii, *Electrodynamics of continuous media (2nd Edition)* (Pergamon, 1984), p.308.
2. Y. Fan, J. Han, Z. Wei, C. Wu, Y. Cao, X. Yu, and H. Li, “Subwavelength electromagnetic diode: One-way response of cascading nonlinear meta-atoms,” Appl. Phys. Lett. **98**, 151903 (2011).
3. A. M. Mahmoud, A. R. Davoyan, and N. Engheta, “All-passive nonreciprocal metastructure,” Nature Commun. **6**, 8359 (2015).
4. E. Poutrina abd A. Urbaas, “Multipolar interference for nonreciprocal nonlinear generation,” Sci. Rep. 6, 25113 (2016).
5. M. Kerker, D. S. Wang, and C. L. Giles, “Electromagnetic scattering by magnetic spheres,” J. Opt. Soc. Am. 73, 765 (1983).
6. R. Birss, “Symmetry and magnetism”, in *Selected topics in solid state physics* (North-Holland Pub. Co., 1964).