Electromagnetic Asymmetry and Magnetic Resonance Enhanced Second-Harmonic Generation in Plasmonic Nanocavities

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Second-harmonic generation (SHG) is not only crucial for nonlinear optical devices, but also a powerful tool for investigating symmetry-related physical phenomena. However, the efficiency of SHG in plasmonic nanostructures is often hindered by the centrosymmetric crystal structures of noble metals, limiting its potential in advanced nanophotonics. Here, we present two distinct strategies to significantly improve SHG efficiencies in plasmonic nanocavities: symmetry-breaking-induced mode hybridization in asymmetric nanosphere dimers and magnetic dipole (MD) resonance induced Lorentz contribution in ultra-compact dimer-on-film nanocavities.

In symmetric nanosphere dimers, SHG is suppressed by destructive interference caused by geometric symmetry. By varying the diameter of either nanosphere to induce asymmetry $\delta = |d_1 - d_2| / (d_1 + d_2)$, we achieve substantial far-field SHG enhancement[1]. We also show that geometric asymmetry primarily contributes to the SHG in the off-resonance excitation case since the near-field enhancement effect is lower in the off-resonance condition. Overall, in our system, SHG is mainly influenced by two factors: symmetry breaking and near-field enhancement. The first effect changes the effective second-order susceptibility, while the second changes $\mathbf{E}(\omega)$ (local field).

Subsequently, we demonstrate that plasmon-induced magnetic dipole resonance in strongly coupled inversion-symmetry-broken dimer-on-film nanocavities can significantly amplify the SHG intensity by an order of magnitude in experiment (two orders of magnitude in theory)[2]. Unlike many previous works that focus on the electric near-field enhancement mechanism, we show that our ultra-compact nanocavities, featuring strong magnetic field enhancement and significant spatial overlap of electric and magnetic fields, allow the hydrodynamic Lorentz contribution ($\mathbf{P}_1 \times \mathbf{H}_1$), to dominate in SHG. This leads to an SHG conversion efficiency of 6×10^{-8} W^{-1}. Through detailed polarization and symmetry analyses, we explore the interaction of the hydrodynamic Lorentz contribution with the electric dipole and electric quadrupole terms. By harnessing the magnetic dipole resonance, we demonstrate the critical role of Lorentz-driven second-order nonlinearity, paving the way toward highly efficient and compact nonlinear photonic devices.

These findings highlight electromagnetic asymmetry and MD resonance as effective strategies for optimizing SHG performance in plasmonic nanocavities. By providing a comprehensive framework for manipulating nonlinear optical interactions at the nanoscale, this work advances the development of high efficiency plasmonic nanophotonic devices for applications in optical sensing, biomedical imaging, and quantum information processing.

References

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2. Yaorong Wang, Ilya Razdolski, Shixuan Zhao, Fan Yang, Xiu Liang, Yuri Kivshar, Dangyuan Lei* Enhanced magnetic second-harmonic generation in ultra-compact plasmonic nanocavities. (under revision with Light Sci. Appl.)