Directional Control of Second-Harmonic Generation from a Metal Nanoparticle

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Due to a strong plasmonic enhancement and symmetry breaking at surface, second-harmonic generation (SHG) from centrosymmetric metal nanoparticles has been observed and becomes a topic of growing interest. While many studies focused on enhancing second-harmonic (SH) signals, few attentions have been paid to the directional control of SHG, which is full of challenges both in numerical modeling and physical design. In this work, unidirectional radiation of second harmonic waves from a gold nanosphere is realized with the help of passive dielectric nanospheres. The unidirectional radiation leads to both spectral and spatial separations between the incident wave and scattered SH field. The surface integral equation method is adopted to simulate the nonlinear sphere nanoantenna since centrosymmetric materials only have surface nonlinearity contributing to SHG under the electric dipole approximation. The mutual coupling between fundamental and SH fields is rigorously captured by a modified equivalent principle taking the depletion of fundamental field into account.

Considering a plane wave excitation, according to the selection rule, the SH radiation is strictly zero along the incident direction, i.e., there is neither forward nor backward SH radiation along the direction of the incident wave. Moreover, the SH radiation pattern for a plasmonic small sphere can be modeled by the emission from induced electric dipole and electric quadruple. When the incident wave is off the plasmonic resonance, fundamental and SH fields fall into the visible light regime simultaneously, which facilitates an experimental detection to a great extent. In this situation, the SH radiation from the equivalent dipole dominates over that from the equivalent quadruple. To realize the unidirectional radiation, the metal sphere is placed between two dielectric spheres functioned as director and reflector. By tuning the geometric sizes, locations and orientations of the metal sphere and two dielectric spheres, their far-field interferences and near-field couplings are manipulated to obtain the unidirectional SH radiation. The directional control of SHG is quite useful for far-field detection and sensing of nonlinear signals.