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(G*) A Theory of Fourth and Fifth Harmonic Generations in Metallic Nanohybrids

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The enhancement of the light-matter interaction through localized surface plasmon resonances (LSPRs) by the heterostructure of noble metal and copper sulfide nanoparticles has aroused wide concern. The higher-order nonlinear process has also gained considerable interest for its efficient enhancement of the harmonic generation process in harmonically resonant heterostructures. In this work, a theory of fourth harmonics generations (4HG) and fifth harmonics generations (5HG) is developed for metallic nanohybrids. Theoretical calculations were performed for a triple-layer nanohybrid in an ensemble of Au, Al and CuS metallic nanoparticles. When a probe field is applied to the nanohybrids, the photons would be coupled to the surface charges, and forming the surface plasmon polaritons (SPPs). The applied field would also induce dipoles, and these dipoles interact with each other which causes dipole-dipole interaction (DDI). With the produced SPP and DDI fields, the intensities of the output 4HG and 5HG fields are calculated by using the coupled mode formalism based on Maxwell's equations. The susceptibilities of different metallic nanoparticles are determined by the density matrix method under their localized SPP resonance frequencies. It is found that the 4HG and 5HG intensities depend on the fourth and fifth-order magnetic susceptibility. In the presence of SPP and DDI, the light-matter interaction is significantly enhanced by the coupling of their LSPRs. The output 4HG and 5HG intensities of the Al/Au/CuS triple-layer nanohybrids formed by the coupled LSPRs are calculated and compared with the experimental data, which showed the consistency with the theoretical model. The findings illustrate the effectiveness of producing higher harmonic generations within resonant plasmonic structures. This hybrid system can be also applied to manufacturing optical nano-switching devices.

Keyword-1

harmonic generation

Keyword-2

triple-layer nanostructures

Keyword-3

metallic nanoparticles

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