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Origin of optical second-harmonic generation in spherical gold nanoparticles: Local surface and nonlocal bulk contributions

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The second-harmonic generation of 150 nm spherical gold nanoparticles is investigated both experimentally and theoretically. We demonstrate that the interference effects between dipolar and octupolar plasmons can be used as a fingerprint to discriminate the local surface and non-local bulk contributions to the second-harmonic generation. By fitting the experimental data with the electric fields computed with finite-element method (FEM) simulations, the Rudnick and Stern parameters weighting the relative nonlinear sources efficiencies are evaluated and the validity of the hydrodynamic model and the local density approximation approaches are discussed.

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