Experimental Investigation of Third-order Susceptibility: Effect of Geometries and Materials

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Metal nanostructured materials have generated considerable interest owing to ultrafast response and large nonlinearity in plasmonics. This research is aimed to clarify the spectral dependence of $\chi^{(3)}$, which is strongly influenced by the geometries and materials. Thin films and spherical nanoparticles of Au, Ag and TiN were fabricated. Particularly, Ag nanoparticles were fabricated with sizes ranging from 3 to 16 nm for size quantization. Triangular plates, nanorods and nanobipyramids of Au nanoparticles were synthesized for geometry influence. All the nanoparticles were embedded in PVA (Au, TiN) or SiO_2 substrate for further characterization. $\chi^{(3)}$ was evaluated by a combined analysis of spectroscopic ellipsometry and pump probe spectroscopy.

The results are discussed in three parts. (1) Interband and intraband contributions of $\chi^{(3)}$:Through the comparison between $\chi^{(3)}$ of Au thin film (only interband transitions excited) and Au nanoparticles (both interband and intraband transitions contributed), we separated the two components of $\chi^{(3)}$. (2) Geometry factors: The investigation of Ag nanoparticles with different sizes and Au nanoparticles with different shapes results that $\chi^{(3)}$ is strongly influenced by geometry factors. The quantum size effect and local filed effect are discussed and compared. (3) Broadband SPR response of TiN: Traditional plasmonic materials suffer from low optical thresholds that limit their usages. Titanium nitride has shown superior properties such as thermal stability, low cost and CMOS-compatible fabrication process. We experimentally obtained $\chi^{(3)}$ of TiN thin film and nanoparticles in an unprecedented wide bandwidth range with a broadband response.

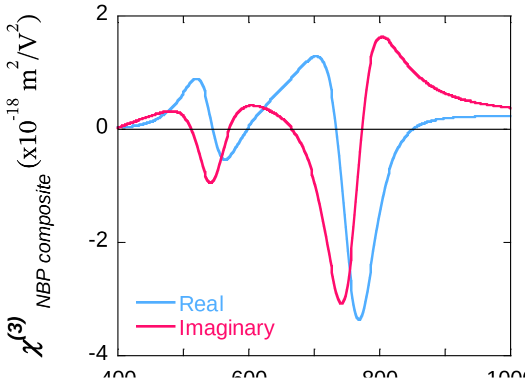

Fig. 1 The spectral dependence of $\chi^{(3)}$ of Au nanobipyramids/PVA composite: The LSPR of longitudinal mode (750 nm) shows stronger enhancement than the transverse mode (532 nm).