Optical nonlinearity and quantization of Au nanomaterials

Boyi ZHANG[1,2], Rodrigo SATO[1], Hiroaki MAMIYA[1], Keiji OYOSHI[1], Mykhailo CHUNDAK[1], Masato OHNUMA[3], Yoshihiko TAKEDA[1,2]

[1]National Institute for Materials Science, [2]University of Tsukuba,[3]Hokkaido University

(ZHANG.Boyi@nims.go.jp)

Abstract: Gold is one of the most commonly used material in nonlinear plasmonics, but its nonlinear properties including size quantization keep unclear. Here the nonlinear response and third-order optical susceptibility of Au nano sphere and thin film samples are evaluated. The contribution of electron transitions to nonlinear response are discussed.

1. Introduction

Metal nanostructured materials have generated considerable interest owing to ultrafast response and large nonlinearity in plasmonics. The promising properties will be widely applied to nanophotonics, such as harmonic generations, ultrafast switching and so on. Gold is widely used in these fields due to its stability and workability. To make active use of the optical nonlinearity, the complete understanding of the origin is needed. In previous researches, the third order susceptibility $\chi^{(3)}$ can be measured only at one single wavelength and think the main contribution is from the interband transitions for gold. These fragmental results strongly limit the understanding of the nonlinear behaviors. This research is aimed to analyze the frequency dependent spectra and clarify the different size behavior of two different electron transitions.

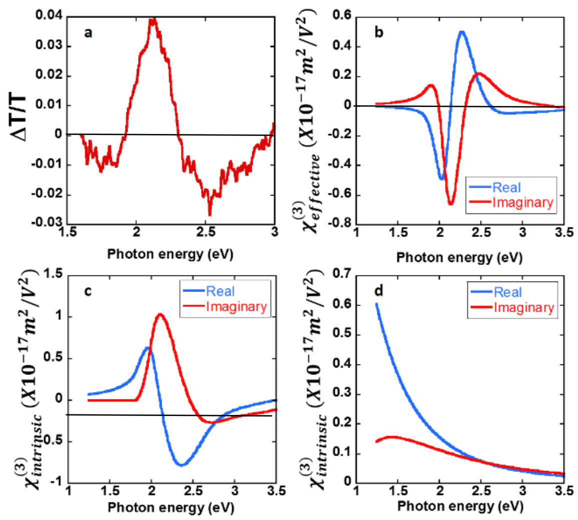
2. Experimental

Gold nanoparticles are fabricated by negative ion implantation with 60 keV and the particle size is controlled by ion fluence. The particle size is evaluated by small angle X-ray scattering(SAXS). Gold thin films are fabricated by Magnetron sputtering. Spectroscopic ellipsometry is applied to analyze the linear optical structure. Femtosecond Pump and probe spectroscopy is used to obtain the nonlinear response. From these combined analysis, we evaluate the spectra of third-order optical susceptibility for various Au nanomaterials.

3. Results and discussion

These graphs give the transient modulation caused by nonlinearity of Au particles with a size of 4.5 nm. Figure 1(a) shows the transient transmission change by pumping at 400 nm. Figure 1(b) shows $\chi^{(3)}$ of Au/Silica glass composite. The relationship between effective $\chi^{(3)}$ and intrinsic one is described as the following equation:

$$\chi^{(3)}_{effective} = p f_l^2 |f_l|^2 \chi^{(3)}_{intrinsic}, \quad f_l = \frac{3\varepsilon_d}{\varepsilon_m + 2\varepsilon_d}.$$



Figures 1(c) and 1(d) show the intrinsic $\chi^{(3)}$ originated from two electron transitions. The transient absorption around 500 nm (2.48 eV) is caused by interband transition, while the bleaching part around 600 nm mainly reflects the intraband transition. The latter contribution is not negligible small.

In this research, we experimentally evaluate the nonlinear response of several Au nanomaterials with spectra and discuss the contribution from two electron transitions with size effects in detail.

Fig.1 Nonlinear optical response of Au nanoparticles: (a)The transient transmission change by femtosecond laser pulse excitation, (b)Effective $\chi^{(3)}$ of Au/Silica glass composite, (c)Intrinsic $\chi^{(3)}$ originated from interband transition and (d)from intraband transition.