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Quantum size effects in the intrinsic third order nonlinear optical susceptibility of metal clusters: Ag nanospheres-silica glass composites

Univ. of Tsukuba [1], Hokkaido Univ. [2], National Inst. for Materials Science [3], ○Rodrigo Sato[1], Masato Ohnuma[2], Keiji Oyoshi[3], Yoshihiko Takeda[1,3]

E-mail: SATO.Rodrigo@nims.go.jp

Metal nanoparticle (NP) materials show characteristic and interesting optical properties [1]. A great number of studies of linear and nonlinear optical properties were carried out in recent years. For Ag NPs, a substantial divergence of the linear properties from the classical predictions was found for particles smaller than 20 nanometers [2]. However, the role of quantum size effects in the nonlinear optical properties of metal clusters is poorly understood. From a theoretical study [3], it is expected that nonlinear optical properties of metal clusters are sensitive to quantum size effects. Materials exhibiting large nonlinear susceptibility are key components for nanophotonics applications [4]. In this context, the nonlinearity of Ag nanospheres fabricated by ion implantation into silica glass with various sizes has been investigated.

Dispersion of the third-order optical susceptibility $\chi^{(3)}$ was evaluated from linear optical properties measured by spectroscopic ellipsometry (Fig. 1a) and nonlinear properties measured by a spectroscopic femtosecond white light continuum pump-probe [5]. Pump pulse duration and repetition rate were 130 fs and 1 kHz, respectively. Figure 1b and 1c show the effective $|\chi^{(3)}{}_{eff}|$ and intrinsic $|\chi^{(3)}{}_{m}|$ at E = 4.6 x 10^8 V/m. The nonlinearity of Ag NPs composite reflects the $\chi^{(3)}{}_{m}$ of the Ag NP itself and the dielectric confinement (local field enhancement). The $\chi^{(3)}{}_{m}$ of Ag NP presents a substantial intensity increase as the NP size decreases most probably due to discretization of the conduction electrons (quantum size effect). The results demonstrate the possibility of controlling the metal optical nonlinearity by quantum size effects.

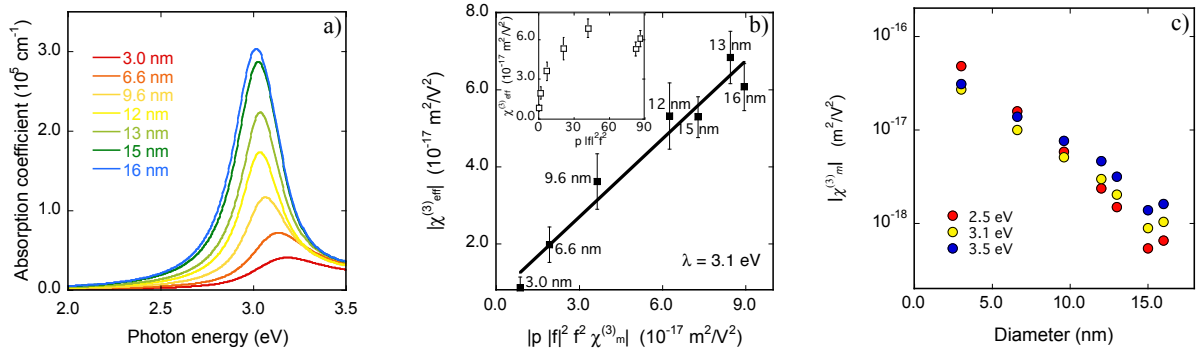

Fig. 1 a) Absorption coefficient, b) effective and c) intrinsic third order susceptibility of Ag NPs embedded in silica glass with NP sizes ranging from 3.0 to 16 nm.

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