3^{rd} ORDER NONLINEAR OPTICAL PROPERTIES OF $Au:SiO_2$ NANOCOMPOSITE FILMS WITH VARYING Au PARTICLES SIZE

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Extensive studies on metal nanoparticle dispersed composite (MNCC) systems have been made during the past decade to explore the potential applications using their characteristic optical properties, which are strongly related with the excitation of a so called "surface plasmon (SP)" due to the dielectric confinement effect of free electrons in the metal particles[1,2].

In this study, dependence of the 3rd order nonlinear coefficients on the size of the Au nano-particles in the $Au:SiO_2$ composite films together with on the wavelength have been examined experimentally. Composite films with various size of the metal nano-particles at constant Au volume fraction were fabricated by the use of alternating deposition technique using a multi-target RF magnetron sputtering at 300 ^oC. The composite films of about 600nm thickness were prepared on fused silica substrates with varying nominal thickness of Au layer, h=0.5, 1.0, 1.5 and 2.0 nm. The Au volume fraction, p, was adjusted at 1% by controlling the ratio of the norminal thickness of metal layer to that of the matrix layer.

Linear optical properties were measured for wavelengths from 250 to 1100 nm using a spectrophotometer. The third order nonlinear coefficients were measured using a Z-scan method. The system consisted of a mode-locked Nd:YAG laser operating at 10 Hz with pulse width of 30 ps and an optical parametric generator(OPG) having tunable scan range of 420~2300nm. The refractive and absorptive contributions to the nonlinearity were resolved from the closed and open aperture Z-scan, respectively.

The SP resonance wavelengthes of the composite films were in the range of 500 ~ 560 nm, increasing with increasing Au nominal thickness, i.e. increasing Au particle size (Fig. 1). The frequency dependence of the third-order nonlinear refractive index of $Au:SiO_2$ film of a given Au particle size showed a clear crossover behavior from self-focusing (positive real part of susceptibility) at high frequency to self-defocusing (negative real part of susceptibility) at low frequency, which is similar to the report obtained for $Ag:SiO_2$ nanocomposite [3]. The magnitudes of imaginary part of the third-order nonlinear susceptibilities measured at the SP wavelength increased as the Au particle size increased, while the sign exhibiting characteristics of saturable absorption (Fig. 2). For a given nanocomposite film, the values of the effective third-order nonlinear susceptibility remained at relatively constant level with variation of focused laser intensities on the film in the range of 0.5 ~ 3.5 GW/cm^2.

This research was supported by a grant (code #: 05K1501-02110) from 'Center for Nanostructured Materials Technology' under '21st Century Frontier R&D Programs' of the Ministry of Science and Technology, Korea.

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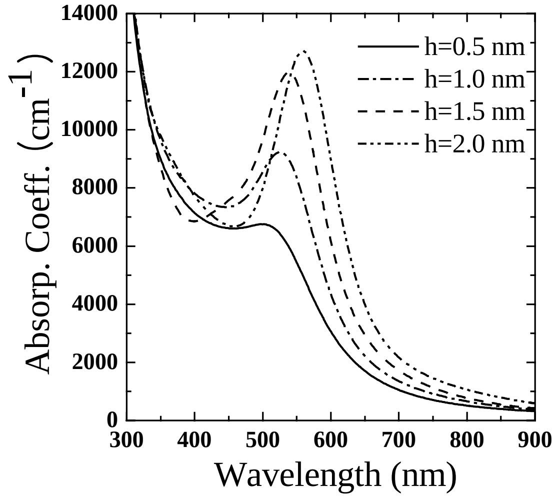
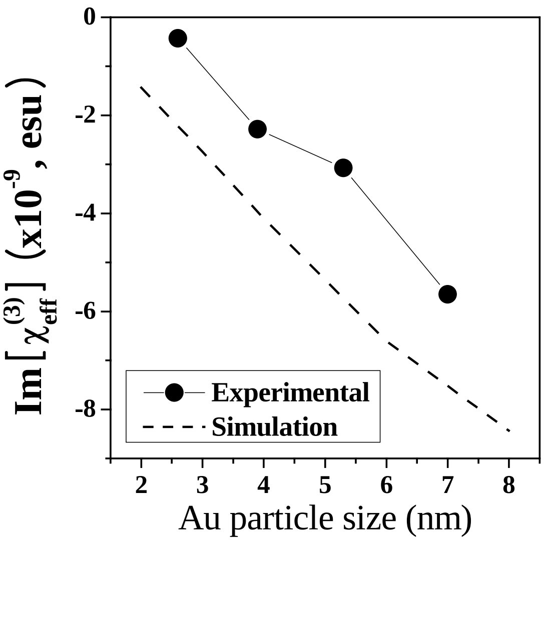
Figures :



[Fig. 1] Optical absorption spectrum of Au-SiO2 composite films with varying Au particle sizes.



[Fig. 2] Measured(closed symbol) and calculated(dashed line) imaginary $\chi^{(3)}$ value at the SP frequency.