Off-resonance 영역 Au:SiO_2 나노복합물질의 3차 비선형 광특성 분석

Studies on third-order optical nonlinearities of Au:SiO_2 nanocomposite films at off-resonant region

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Metal nanocomposite materials turned out to be potential candidates for applications in all-optical photonic devices due to their large third-order optical nonlinearities[1,2]. In this work, metal nanocomposites containing Au nanoparticles embedded in SiO_2 were deposited on fused silica substrates by alternating sputtering of SiO_2 and Au at a substrate temperature of 300° C and a constant volume fraction of 1% [3]. The nanocomposite films were prepared with different Au mean diameters between 2.3 nm and 5.7 nm, while the overall thickness was kept constant at 600 nm. The linear absorption peaks induced by surface plasmon resonance were shifted toward longer wavelengths from 500 nm to 560 nm with increasing Au particle sizes.

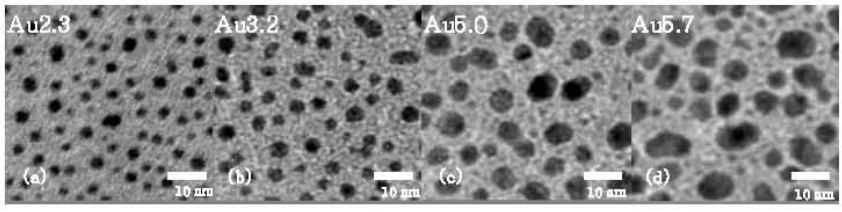

Fig. 1. TEM images of the SiO_2/Au/SiO_2 nanocomposite films with Au mean diameters of (a) 2.3nm, (b) 3.2nm, (c) 5.0nm and (d) 5.7nm.

To investigate third-order optical nonlinearities of the Au:SiO_2 nanocomposite films such as nonlinear refraction and nonlinear absorption at off-resonant wavelengths, z-scan technique was used[4]. The excitation source was a Kerr lens mode-locked femtosecond Ti:sapphire laser, which delivers 85fs pulses at 92MHz. For estimation of third-order

nonlinear optical properties at off-resonant region near 520nm, laser pulses near 780 nm and 820 nm and its second harmonics at 410 nm was used. Nonlinear refractive indices and nonlinear absorption coefficients at these off-resonant wavelengths were determined by closed- and open- aperture measurements of transmittance, respectively.

In the NIR wavelengths, far from SPR absorption band, the estimated values of $\mathrm{Re}[\chi^{(3)}]$ exceeded the theoretical results approximately three orders of magnitude. The sign change of $\mathrm{Im}[\chi^{(3)}]$ from positive to negative was observed for Au mean diameters of $\leq$ 5.0nm. At the UV region near the absorption maxima, $\chi^{(3)}$ was mainly given by nonlinear absorption, showing negative values of $\mathrm{Im}[\chi^{(3)}]$. As expected from the theoretical analysis, the size-dependent variation of the third-order nonlinearity could be qualitatively confirmed for $\mathrm{Re}[\chi^{(3)}]$ at 780nm and 820nm and for $\mathrm{Im}[\chi^{(3)}]$ at 410nm, respectively. The distinct discrepancy between the experimental and theoretical values of $\mathrm{Re}[\chi^{(3)}]$ and $\mathrm{Im}[\chi^{(3)}]$ in magnitude and sign can be understood with neglecting the spatial field variations and local field enhancements near the surface of Au nanoparticles as well as by the inter-particle interactions in the theoretical model. Detailed consideration of the local field distributions and enhancements is required to clarify our experimental results.

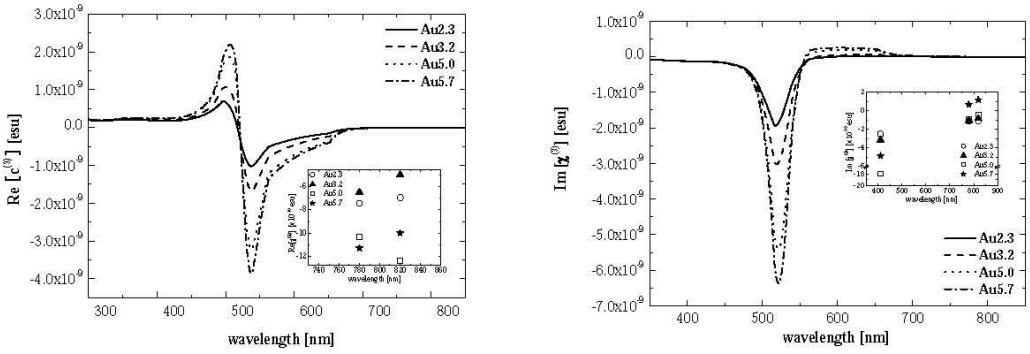

Fig. 2. The theoretical and experimental(curves) $\mathrm{Re}[\chi^{(3)}]$ & $\mathrm{Im}[\chi^{(3)}]$ values(inset). The off-resonant measurements are performed at 410nm, 780nm and 820nm, respectively.

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