

## Optical Nonlinearity and Transient Response of Fullerene Compounds

Kejun LI<sup>1,2</sup>, Rodrigo SATO<sup>1</sup>, Hiroaki MAMIYA<sup>1</sup>, Takatsugu WAKAHARA<sup>1</sup>, Yoshihiko TAKEDA<sup>1,2</sup>

<sup>1</sup>National Institute for Materials Science, <sup>2</sup>University of Tsukuba

Fullerenes have large optical nonlinearity due to the delocalization of  $\pi$ -electron conjugated systems [1] and have been applied to optical limiting [2]. Experimental data for nonlinearity have been reported by several papers, however, these data is fragmental and the whole picture has not been fully understood. The aims of research are to figure out third-order optical nonlinearities of Fullerene materials experimentally and to improve the function for device applications. Here we show experimental evaluation of third-order optical susceptibility spectra and transient response of C<sub>60</sub>, C<sub>70</sub> and Sc<sub>3</sub>N@C<sub>80</sub> solid thin films and discuss the mechanism.

Thin film samples were fabricated by vapor deposition. Femtosecond time-resolved pump-probe spectroscopy was utilized to obtain nonlinear optical response and power modulation. The pulse width and pumping wavelength were 146 fs and 400 nm, respectively. Spectroscopic ellipsometry was applied for the linear optical characterization. Third-order optical susceptibility spectra were evaluated with the formula below by combining linear response from spectroscopic ellipsometry with nonlinear optical response data from pump-probe spectroscopy,

$$\delta\epsilon_{eff}(\omega_{probe}) = D\chi_{eff}^{(3)}(-\omega_{probe}; -\omega_{pump}, \omega_{pump}, \omega_{probe}) |E_0(\omega_{pump})|^2$$

Figure 1 shows experimentally evaluated third-order optical susceptibility spectra of C<sub>60</sub> thin film and C<sub>70</sub> thin film. The amplitude indicates both of them are of the order of  $1 \times 10^{-18} \text{ m}^2 / \text{V}^2$ . For C<sub>60</sub>, the real part of the third-order susceptibility is positive and much larger than the imaginary, suggesting that nonlinear refraction is dominant in the nonlinear optical process. For C<sub>70</sub>, the real part is positive while the imaginary part is negative, suggesting that nonlinear refraction enhances and absorption weakens.

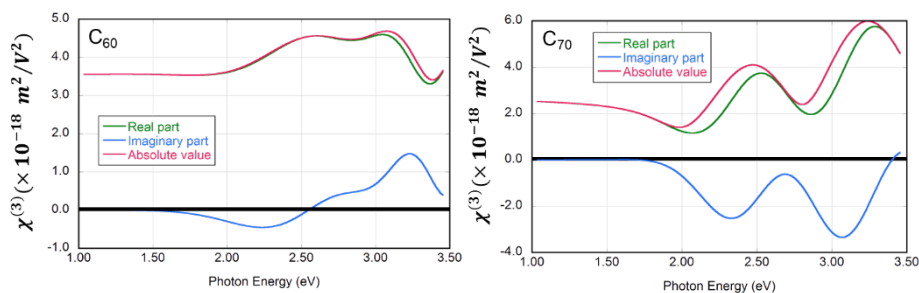


Fig.1 the third-order susceptibility of C<sub>60</sub> and C<sub>70</sub> thin films

- [1] G. P. Zhang, X. Sun, and T. F. George, *J. Phys. Chem. A* **113**, 1175–1188 (2009).  
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