グローバル COE 物質科学イノベーション講演会

演 題：**Nonlinear optical properties of molecules and nanoparticles**

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場 所：創成科学研究棟 4F セミナー室 B・C（04－214・213）

要 旨：

When light of wavelength λ falls on molecules in solution, a part of it is scattered by the molecules which is known as Rayleigh scattering. A small part of light in interaction with molecules is scattered at half the incident wavelength (λ/2) or twice the frequency. This incoherent second harmonic Rayleigh (SHR) or hyper-Rayleigh scattering (HRS) light is directly proportional to the square of the molecular second order polarizability (β) which is an intrinsic property of a molecule and is zero if the molecule has a center of symmetry. Exploiting the relationship between molecular nonlinearity with the density of the molecular scatterers, symmetry at the microscopic as well as macroscopic levels and anisotropy of the second order polarization tensor, we have demonstrated many new chemical applications of this HRS technique.

As a demonstration of the powerfulness of this technique two examples are chosen for this talk. In the first example,[1] I will present how the geometry of a charge transfer complex in solution is determined by this technique which is much superior to the NMR technique widely used for the purpose.

In the second example,[2] I will show how the technique has been used to probe the origin of large quadratic nonlinearity (β) in noble metal nanoparticles. We find that the second order nonlinearity has contributions from both the surface and the bulk of the nanoparticle. Insofar as the size (r) of the particle remains small compared to the wavelength of light used in the measurement, the origin of SHG is purely dipolar and the β scales as r^2 (where r is the radius of the particle). When the size to wavelength ratio increases, the retardation of light becomes important and higher order multipolar contribution to SHG becomes prominent. The scaling of β in such a case goes as $r^{2.3}$. Polarization resolved SH scattering measurements provide clear evidence to this transition from purely dipolar mechanism to a multipolar mechanism as a function of d/λ (where d is the diameter of the particle and λ is the wavelength of light). Resonance enhancement by the presence of the SPR band is the reason for the large β, however, when measurements are made far away from resonance, the value of β remains two orders of magnitude greater than those of highly efficient NLO molecules indicating that the noble metal nanoparticles have intrinsically highly polarizable structures.

REFERENCES

1. Sampa Ghosh et al. Proceedings of SPIE,Vol. 7413, 74130N1-8, in Linear and Nonlinear Optics of Organic Materials IX, T. D. Goodson III (ed.) (2009); J. Chem. Phys. (2010) in press

2. M. Chandra et al. J. Phys. Chem. C 111, 10652-10656 (2007); Chem. Phys. 358, 203-208 (2009); International J. of Green Nanotech.: Phys. and Chem., 1, 10-25 (2009)

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