



STUDY OF SPECTRAL PARAMETERS OF LIGHT SCATTERING LINES IN SURFACTANTS

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Annotation

Nano and microparticles have optical, structural, and chemical properties that differ from both their building blocks and the bulk materials themselves. These different physical and chemical properties are induced by the high surface-to-volume ratio. As a logical consequence, to understand the properties of nano- and microparticles, it is of fundamental importance to characterize the particle surfaces and their interactions with the surrounding medium. Recent developments of nonlinear light scattering techniques have resulted in a deeper insight of the underlying light-matter interactions. They have shed new light on the molecular mechanism of surface kinetics in solution, properties of interfacial water in contact with hydrophilic and hydrophobic particles and droplets, molecular orientation distribution of molecules at particle surfaces in solution, interfacial structure of surfactants at droplet interfaces, acid-base chemistry on particles in solution, and vesicle structure and transport properties.

Keywords: SFG: sum frequency generation, SHG: second harmonic generation, NLS: nonlinear light scattering, HRS: hyper-Rayleigh scattering, λ : wavelength, MG: malachite green, PS: polystyrene

Introduction

Light scattering occurs when photons encounter impurities or inhomogeneities. Such impurities can consist of density fluctuations, molecules, or particles. The observation that SHG could occur in a centrosymmetric medium in a non-phasmomatched direction was explained by noting that on the molecular time and length scale, there are local density fluctuations in any medium: The incoming laser beam generates a small dipole contribution in every molecule. On average, in an isotropic material, these contributions vanish, but when enough photons are present, a small residual signal resulting from the fluctuations in the distribution in the medium remains. Non-linear elastic and inelastic light scattering have been termed hyper-Rayleigh scattering (HRS)





and hyper-Raman scattering, respectively (2). The intensity in both types of scattering depends linearly on the number of source molecules as it is incoherent scattering.

Main Part

When the size of the inhomogeneity becomes an appreciable fraction of the scattered wave-length (λ_{SF}), detectable coherent effects begin to emerge. NLS from particles with sizes $\sim \lambda_{SF}/50$ can already display the effects of correlated emission (coherence). These coherent processes can originate from either the surface of the particles or the bulk. The use of a nonlinear, second-order scattering process to obtain information about surface chemistry of particles in liquids was demonstrated in 1996 by the Eisenthal group (3). They reported the emission of SH signal from malachite green (MG) adsorbed to the surface of micron-sized centrosymmetric polystyrene (PS) particles. The SH scattered intensity was observed to depend linearly on the number of particles in solution and quadratically on the MG concentration.

The quadratic dependence proves that the signal that originates from MG molecules adsorbed on the surface of each particle adds up coherently, whereas the linear dependence proves that the signal from different particles adds up incoherently.

Vibrational sum frequency scattering (SFS) was demonstrated more than a decade later (4) and can also be used to probe the interfacial properties of particles in liquid and solid media. The difference with second harmonic scattering (SHS) is that the incoming beams are of different frequency, and one of the frequencies can be tuned to the energy of vibrational transitions. The resulting effect is that the vibrational spectrum of a molecular layer with a thickness of typically one to two molecular dimensions at the particle interface can be measured.

The aim of this review is to give an introduction to NLS methods and to discuss recent advances in the field. More specifically, the connecting framework between the different methods is presented. Figure 1 displays objects of different sizes that have been or could be studied with NLS: molecules, micelles, nanoparticles, vesicles, droplets (emulsions), cells. The size of the object with respect to the wavelength in combination with the type of light-matter interaction determines the dominant scattering process. For excitation with visible light, typically parametric light scattering, such as HRS and hyper-Raman scattering, occur for objects < 10 nm, SHS, SFS, and random quasi phase matching occur for objects < 20 μm . Larger objects produce reflected or refracted beams, which can be measured with SHG or SFG in reflection or transmission mode.



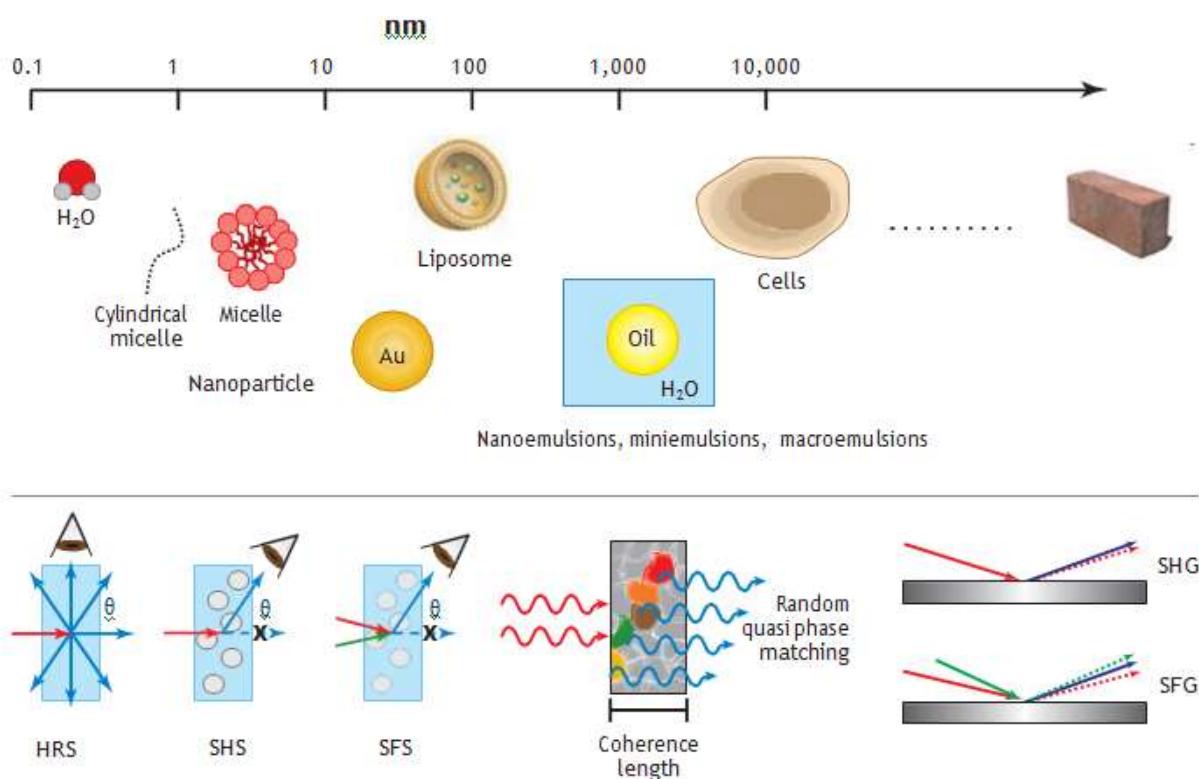


Figure 1. Display of objects, from small to large, from which light can induce nonlinear light scattering phenomena: molecules, micelles, nanoparticles, vesicles, droplets (emulsions), and cells. For excitation with visible light, HRS and hyper-Raman scattering typically occur for objects <10 nm; SHS, SFS, and random quasi phase matching occur for objects <20 μ m. Larger objects produce reflected or refracted beams, which can be measured with SHG and SFG in reflection or transmission mode. Abbreviations: HRS, hyper-Rayleigh scattering; SFG, sum frequency generation; SFS, sum frequency scattering; SHG, second harmonic generation; SHS, second harmonic scattering. In Section 2, we give an overview of the fundamentals behind nonlinear light scattering. Section 3 is devoted to a description of experimental parameters. Section 4 picks up where Section 2 stopped, discussing the models and studies done to understand the optical properties of nanoparticles and microparticles. In Section 5, we discuss recent studies to understand the interfacial chemistry found in solution, and Section 6 offers a short introduction to biologically relevant topics. Finally, we end with conclusions and outlook. Due to space constraints, we do not treat all topics in depth. We focus our attention on offering an introduction aimed at pointing out the cohesion between the different studies, and we confine ourselves mostly to work done in liquid media. Recently, three reviews have appeared: one by Eisenthal (5) discussing the topic of the SHS of nanoparticles, microparticles, and liposomes in water; one by Ray

(6) discussing the non-linear optical properties of nanoparticles with a focus on HRS; and one by Brevet (7) on SHG in nanostructures.

Fundamental concepts of nonlinear light scattering

In this section, we describe the fundamentals needed to understand the principles of NLS. We use the generalized case for light scattering from a particle induced by two beams having wave vectors \mathbf{k}_1 and \mathbf{k}_2 .

Where necessary we consider plane waves of the form $\mathbf{E}_i = \mathbf{E}_0 e^{-i\omega_i t} e^{-i\mathbf{k}_i \cdot \mathbf{r}}$. The beam waist is always much bigger than the particle. **Figure 2** shows a representative geometry,

where the two incoming beams lie in the same (horizontal) plane as the outgoing scattered beam.

For light scattering to occur, a discontinuity or an object for a photon to scatter on must be present. An electromagnetic field \mathbf{E} that interacts with a molecule can induce a molecular dipole \mathbf{p} of the form (2, 8)

$$\mathbf{p} = \alpha^{(1)} \cdot \mathbf{E} + \frac{1}{2} \beta^{(2)} : \mathbf{E} \mathbf{E} + \frac{1}{3} \beta^{(3)} : \mathbf{E} \mathbf{E} \mathbf{E} + \dots, \quad (1)$$

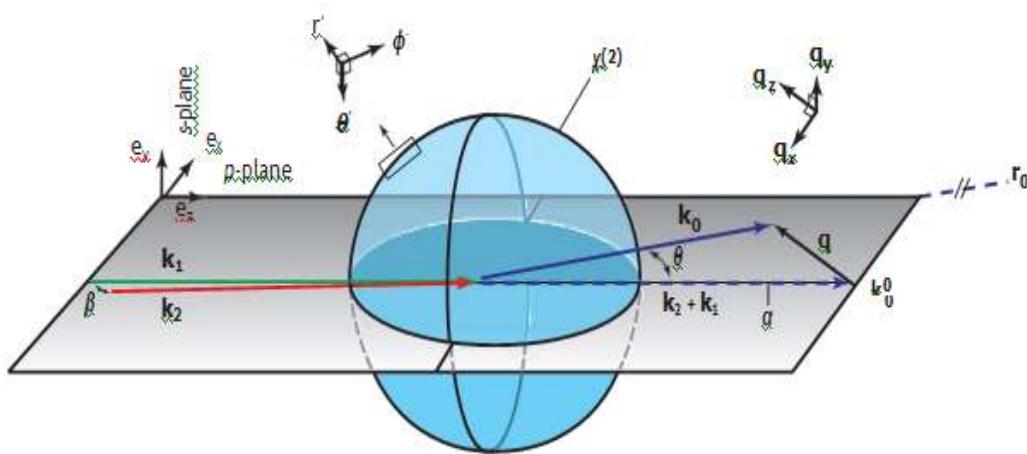


Figure. 2. In-plane scattering geometry for nonlinear light scattering experiments, indicating the \mathbf{k} -vectors of the incoming beams (\mathbf{k}_1 and \mathbf{k}_2) and the scattered sum frequency beam (\mathbf{k}_0), scattering wave vector (\mathbf{q}), and in-plane scattering angle (θ). β is the angle between \mathbf{k}_2 and \mathbf{k}_1 , and α is the angle between \mathbf{k}_2 and \mathbf{k}_1 .

Beams polarized parallel to the plane of incidence are defined as p-polarized, whereas beams with an oscillating field in the y-direction are indicated as s-polarized. The direction $\mathbf{k}_1 + \mathbf{k}_2$ is called the forward direction ($\theta = 0$).



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