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Original Research Article

Theory of nonlinear scattering and second harmonic generation in powder

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ABSTRACT
Using the nonlinear polarisation term from Mie theory, this study aims to create a theoretical model to explain the experimental data of scattered second harmonic output intensity in powder sample. Experimental research on the scattered second harmonic signal from the urea sample was conducted using the Kurtz powder method. Theoretical justifications for the Kurtz technique's findings presuppose that the powder's particles I are submerged in a transparent liquid medium with a nearly same refractive index and (ii) are planar parallel slabs of a single crystal. According to us, if we first take into account the cooperative scattering that results from single particle nonlinear scattering occurring in the medium, the general treatment for second harmonic generation analysis from powders may be studied more effectively. We have created a theoretical model in this research to estimate second harmonic generation in powder samples. The particles must be significantly larger than the wavelength of light and be assumed to be spherical in order for the model to work.

1. Introduction

It has been proven that second harmonic production is a very effective spectroscopic instrument for studying a variety of physical and chemical phenomena. It is applied for both surface research and verifying the homogeneity of waveguide samples [2, 3] as well as for the creation of blue light laser sources that can be used in opto-electronic devices [1, 2]. A significant area of scientific interest is the theoretical growth of the second harmonic generation in small particles and randomised disordered media. It is important to note that second harmonic generation (SHG) theory in metallic and semiconducting systems is well researched [4–9].

Restrictive models of the intrinsic bulk or surface [10–13] nonlinear response of the sphere are used in investigations on the second harmonic behaviour of tiny particles. A tiny sphere of centrosymmetric material irradiated by linearly polarised light was the subject of a recent calculation by Dadap et al. [14] to determine second harmonic Rayleigh scattering. The second harmonic generation (SHG) electromagnetic theory from the surface of a sphere with a radius smaller than the wavelength of light is the subject of their research. They postulated that the second harmonic generation in tiny spheres is caused by locally excited electric quadrupoles and nonlocally excited electric dipoles. Small spherical particle SHG caused by an inhomogeneous longitudinal field was investigated by Brudny et al. in their study published in 2015 [15]. They have taken into consideration particles with a radius smaller than a light wave's wavelength. SHG by two-dimensional particles has recently been numerically analysed by Valencia et al [16]. They came to the conclusion that when symmetrical particles are lit along its axis of symmetry, SHG does not occur. Even so, a combination of s and p polarised light was discovered to cause s polarised SHG.

The goal of the current research is to create a theoretical model that uses the nonlinear polarisation factor from Mie theory to account for experimental observations of scattered SH output intensity in powder samples [17]. Experimental research on the scattered second harmonic signal from the urea sample was conducted using the Kurtz powder method [18]. Theoretical justifications for the Kurtz technique's findings presuppose that the powder's particles I are submerged in a transparent liquid medium with a nearly same refractive index and (ii) are planar parallel slabs of a single crystal. We believe that if we first take into consideration the single particle nonlinear scattering that results from the medium before accounting for the cooperative scattering, the general approach for SHG analysis from powders can be evaluated more effectively. The Mie scattering hypothesis [17] can be used to explain the scattering caused by organic powders because their particles are typically significantly larger than the wavelength of light. We have created a theoretical model in the current study to estimate SHG in powder sample. The particles must be significantly larger than the wavelength of light and be assumed to be spherical in order for the model to work.

2. Theoretical formulation

Due to the nonlinear interaction of a plane monochromatic wave with the spherical particle, we have been able to find the solution to Maxwell's equations that describe the electric field at second harmonic frequency. We have assumed that the characteristics of the medium drastically shift across the spherical particle. When the particles don't show second-order nonlinearity, the normal Mie scattering results apply. Second harmonic generation can occur in molecules with high first-order hyperpolarizability values. A greater number of scattering centres will result in more harmonic production. The scattered second harmonic light from the sample's powder will appear as a result of the coherent superposition of the dispersed second harmonic. The nonlinear term takes the place of the source term in this scenario as there isn't a source wave at second harmonic that has been scattered from the sample.

The Maxwell's field equations in terms of the Hertz vector are written as follows in spherical polar coordinates:

$$\frac{1}{r}\frac{\partial^2(r\Pi)}{\partial r^2}+\frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\Pi}{\partial\theta}\right)+\frac{1}{r^2\sin\theta}\frac{\partial^2\Pi}{\partial\phi^2}+k^2\Pi=0 \tag{1}$$

Here, r, θ, ϕ are polar coordinates and k is the wave vector of the electromagnetic wave. The wave's electric field amplitude E has the following relationships to the Hertz vector:

$$E_r=\frac{\partial^2(r\Pi)}{\partial r^2}+kr\Pi\,, \tag{2}$$

$$E_\theta=\frac{1}{r}\frac{\partial^2(r\Pi)}{\partial r\partial\theta}\,, \tag{3}$$

$$E_\phi=\frac{1}{r\sin\theta}\frac{\partial^2(r\Pi)}{\partial r\partial\phi}\,. \tag{4}$$

Ref. [17] provides the accepted solution to Equation (1) for the potential of the incident wave.

$$\Pi=\frac{1}{rk^2}\sum_{l=1}^{\infty}i^{l-1}\frac{2l+1}{l(l+1)}\psi_l(rk)P_l^{(1)}(\cos\theta)\cos\phi\,. \tag{5}$$

Here, $\psi_l(rk)=\sqrt{\frac{\pi kr}{2}}J_{l+1/2}(kr)$.

Every finite domain of the kr plane has the function $\psi_l(rk)$, which is regular. The second harmonic production may result from the blending of two fundamental waves in the spherical particle. We can think of the nonlinear term as a source term in the case of the second harmonic. The second-order nonlinear optical susceptibility is represented by $P_N=\chi^{(2)}E_1E_2$. E_1 and E_2 are the electric fields of the mixing fundamental waves, and the nonlinear term is caused by the medium's finite second-order susceptibility. When we include the nonlinear factor in Eq. (1) in terms of Hertz vectors, we obtain

$$\frac{1}{r}\frac{\partial^2(r\Pi)}{\partial r^2}+\frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial\Pi}{\partial\theta}\right). \\ +\frac{1}{r^2\sin^2\theta}\frac{\partial^2\Pi}{\partial\phi^2}+k_2^2\Pi=4\chi^{(2)}\Pi_0\Pi_e r^2k_{lm}^2\,. \tag{6}$$

Here, k_2 and k_{lm} stand for the fundamental wave in a medium and the second harmonic wave in a vacuum, respectively. The interacting fundamental waves are represented by Π_e and Π_0. For the scattered second harmonic wave, the aforementioned equation has been formulated, and the following formula has been used to get its solution:

$$S(w)=\Pi(w)+PI\,. \tag{7}$$

Here, w is the Hertz vector for the second harmonic wave in the medium and $\Pi(w)$ stands for the wave within the medium. $\Pi(w)$ is the complementary result of leaving out the nonlinear term, and PI is the specific integral discovered as:

$$PI=\frac{4\chi^{(2)}r^2k_{lm}^2}{k_{2m}^4}\frac{\partial^2}{\partial r^2}(\Pi_0\Pi_e)+\frac{4\chi^{(2)}(\Pi_0\Pi_e)r^2k_{lm}^2}{k_{2m}^2}\,. \tag{8}$$

For $\Pi(w)$, we use the solution similar to Eq. (5) as:

$$\Pi(w)=\frac{1}{rk_{2m}^2}\sum_{l=1}^{\infty}A_l\psi_1(k_2r)P_l^{(1)}(\cos\theta)\cos\phi\,. \tag{9}$$

Calculated from the boundary conditions, A_1 is the perturbative term resulting from the nonlinearity. k_{2m} is the second harmonic wave's internal wave vector.

Similar representations of the Hertz vector for scattered output can be found in Ref. [17].

$$\Pi(s)=\frac{1}{rk_2^2}\sum_{l=1}^{\infty}B_1\xi_l^1(k_2r)P_l^{(1)}(\cos\theta)\cos\phi\,, \tag{10}$$

where $\xi_l^1(k_2r)=\sqrt{\pi k_2r/2}H_{l+1/2}^{(1)}(kr)$ and the tangential and radial components of the field should be continuous across the surface of the sphere, which is one of the usual boundary requirements that determines the value of the constant B_1. We have introduced the boundary conditions mathematically as:

$$\frac{\partial}{\partial r}\left(\Pi(s)\right)=\frac{\partial}{\partial r}\left(\Pi(w)+PI\right), \tag{11}$$

and

$$k_2r\Pi(s)=k_{2m}r\Pi(w)+k_{2m}PI\,. \tag{12}$$

Knowing the scattered field, which may be connected to the Hertz vector $\Pi(s)$ via Eqs. (2) - (4) allows one to calculate the scattered output intensity. (2) - (4). The solution to Eq. (6) has been found by applying Eq. (5) for $\Pi(s)$; the procedures to solve for $\Pi(w)$ are as follows. Therefore, the expression of $\Pi(w)$ can be used to get $\Pi(s)$ in Eqs. (11) and (12). These are a lot of mathematically complex steps, and the only answers one can find are numerical. You can write the expression for the scattered output intensity as follows:

$$I(s)=\left|\frac{\partial^2\left(r\Pi(s)\right)}{\partial r^2}+k_2r\Pi(s)\right|_{r=a}^2+\left|\frac{1}{r}\frac{\partial^2\left(r\Pi(s)\right)}{\partial r\partial\theta}\right|_{r=a}^2 \\ +\left|\frac{1}{r\sin\theta}\frac{\partial^2\left(r\Pi(s)\right)}{\partial r\partial\phi}\right|_{r=a}^2\,. \tag{13}$$

3. Experimental study of second harmonic generation

The Kurtz powder method has been used to measure SHG. The sample was exposed to a Q-switched 1064 nm 15 ns pulsed Nd:YAG laser with a peak output of 5 MW for this. A 98:2 beam splitter was used to create a reference beam by normalising the variations in the primary laser light. A second harmonic separator (SHS) was used to filter out the fundamental input radiation from the second harmonic (SH) radiation at 532 nm that was received at the output. The generated SH was seen on a Tektronics 100 MHz digital storage oscilloscope after being detected by a photo multiplier tube (PMT) (DSO). 250V was used as the PMT's biassing voltage. The PMT responds linearly to this biassing voltage. For two different ranges of particle size r, namely viz; $60 < r < 75$ μm and $75 < r < 90$ μm, we have investigated the variation of SH signal intensity as function of intensity of fundamental.

4. Results and discussions

We used our study on a sample of urea that had been exposed to 1.064 μm Nd-YAG laser. The material parameters are taken as $k_1 = 5.96\times10^6$ m^{-1}, $k_2 = 1.18\times10^7$ m^{-1}, $k_{1m} = 9.38\times10^6$ m^{-1}, $k_{2m} = 1.77\times10^7$ m^{-1}, and $\chi^{(2)} = 2.8\times10^{-12}$ mV^{-1}.

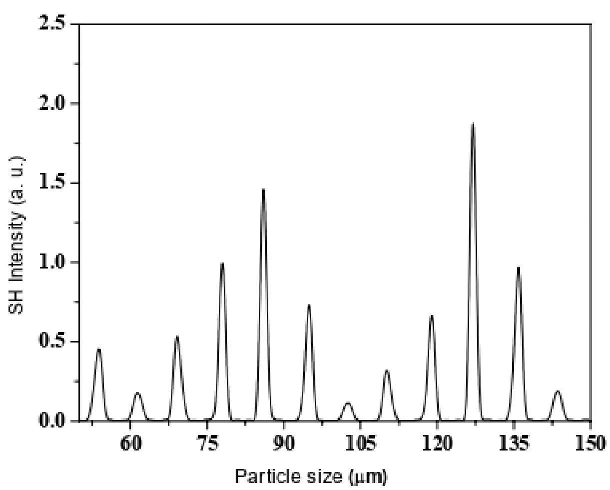

Figure 1. Variation of second harmonic intensity of Urea as a function of particle size in single particle scattering (theory).

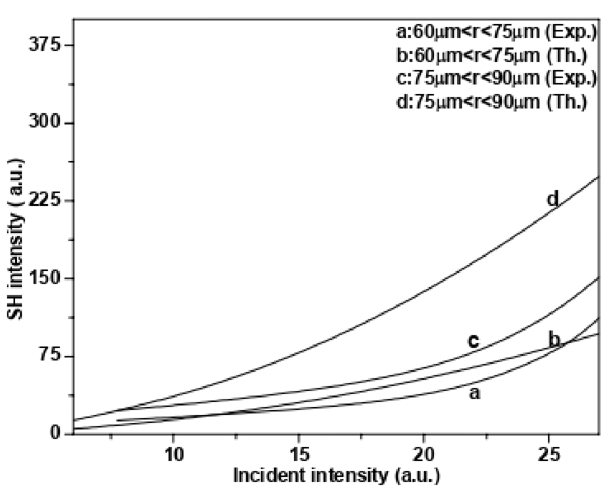

Figure 2. Variation of output second harmonic intensity of Urea as a function of intensity of fundamental in different particle size ranges.

For $l = 1$, the numerical solutions have been discovered. In single particle scattering, Figure 1 shows how second harmonic intensity varies with particle size. The graphic shows that the second harmonic intensity exhibits oscillatory behaviour rather than a monotonous increase with increasing particle size. For all intents and purposes, we chose a powder made up of several particles, and although though this result cannot be scientifically verified, it provides insight into the relationship between second harmonic intensity and particle size.

We have got the numerical findings for the scattered second harmonic intensity from a powder of certain particle size range in order to make the results consistent with the experiment. We have taken into account the powder's Gaussian distribution of particle sizes. For two different ranges of particle size, we looked at how the SH signal's intensity changed in relation to the basic signal's intensity. We've presented the variance in second harmonic intensity in figure 2 as a function of fundamental intensity. The experimental results are shown by curves (a) and (c), whereas the theoretical results are shown by curves (b) and (d). We discover that the basic intensity has a parabolic relationship with the dispersed output intensity. The same graph also demonstrates that when average particle size increases, output intensity rises. All four of the curves display a parabolic shape, which is a sign of second-order nonlinearity. The same figure also demonstrates that the output intensity rises as average particle size rises, and the theoretical findings are in good agreement with the experimental findings.

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