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Investigation of Size-dependent Nonlinearity of Ag Nano-Fluids using Self-defocusing Technique

Sally A. Al-Ani and **Mazin M. Elias**

mazinelias2012@gmail.com

Institute of Laser for Postgraduate Studies, University of Baghdad, Baghdad, Iraq

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Abstract: In this work, the effect of different particle size on the nonlinear optical properties of silver nanoparticles in de-ionized water was studied. The experimental observation of the far field diffraction patterns by CCD camera in two and three dimensions. The maximum change of nonlinear refractive index and the relative phase shift were calculated. The self-defocusing technique was used with a continuous-wave radiation from DPSS Blue laser .The wavelength is 473 nm with an output power of 270 mW. All the Ag colloids samples containing the sizes 15, 30, 50, and 70 nm of silver nanoparticles used in the study were chemically prepared. It was found that the nonlinear refractive index is a particle size dependent and of the order of 10^{-7} cm^2/W, while its maximum change is in the order of 10^{-4}. The phase shift increased from 8π up to 14π with particle size increment. The increase in the particle size of the Ag nanoparticles leads to increasing the number of rings and to the material possessing more nonlinearity with the nonlinear refractive index increment.

Keywords: *particle size, refractive index, self-defocusing*

Introduction

Nonlinear optical properties of materials have become increasingly important since the invention of laser. The far-field diffraction ring pattern has gained more attention for its unique properties and potential applications. Since, it was first reported in 1967s [1]. Similar phenomena were observed in several systems, such as atomic vapors, liquid crystals, polymers, and nanostructured materials [2-5]. Nanoparticles exhibit many interesting nonlinear properties, which are mainly related to the increase in the ratio of surface area to volume, where the realm of quantum effects and the surface plasmons are predominate [6]. The nonlinear aspects have not been explored much so far. On the other hand, much effort has been put into the field of integrated nonlinear optics and a medium with large nonlinearity is desirable [7]. Due to their relatively large third-order nonlinearity and ultra- fast response time, silver nanoparticles have wide applications as nonlinear materials for optical switching, optical limiting and beam flattening [8]. Most of the reported nonlinear characterization of nanoparticles in dielectric materials was performed using lasers at wavelengths close to the absorption maximum of the surface plasmon resonance of nanoparticles [9]. In our experiment, we used DPSS blue laser with wavelength 473 nm focused on silver nano-fluids that had SPR peaks around 400 nm in order to investigate the effect of particle size increment on the nonlinear properties for these nano-fluids.

The Experimental Set-up

The used experimental setup is depicted in Figure (1). A Gaussian light beam was incident from a CW blue DPSS laser (model MBL-FN-473nm-200mW) with a wavelength of **473 nm**. We used an attenuator to control the incident power on the nano-fluid sample. The power was fixed to be at **270 mW**.

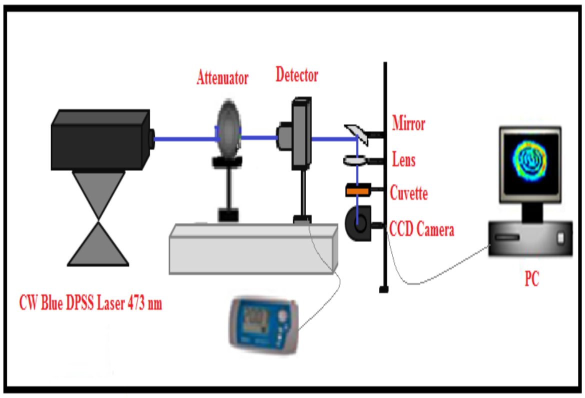

Fig. (1) Schematic diagram of the experimental set up

The beam was focused by a lens of **5 cm** focal length, propagated through a cell containing the prepared nano-fluid. The far-field diffraction patterns were collected by a CCD camera (model Beamage - CCD12, gentec-EO, Canada) in two-dimension and three-dimension images, and the results were recorded and analyzed by a computer. A colloidal solution of spherical shaped silver nanoparticles with different particle size was chemically prepared in de-ionized water using sodium borohydride $NaBH_4$ to reduce silver nitrate $AgNO_3$ in the presence of PVP as a stabilizer. The average nano-silver particle size increased from **15 nm** to **30, 50** and **70 nm,** respectively, and the concentration for all samples was kept constant at about **70 ppm.** In this work, the effect of the change in particle size on the emerged diffraction pattern, the maximum change of nonlinear refractive index, the phase shift and the nonlinear refractive index were studied.

Results and Discussion

The two-dimension images of the emerged diffraction patterns recorded by the CCD camera were analyzed in order to study the influence of the increment in the particle size on the observed profile.

As shown in Figure (2), at high laser intensity **784 W/cm^2**, the number of rings increased with increasing the particle size from **4** rings for the average particle size of **15** nm up to **5, 6** and **7** rings for the sizes of **30, 50** and **70 nm** respectively. A linear relation can be established by plotting the number of fringes as function of particle size as seen in Figure (3). The increasing in particle size affects and increases the nonlinear refractive index of the nano-fluid which in turn acts on the propagation of light inducing more diffraction indicated by the increment in the number of rings.

Sample A Sample B Sample C Sample D

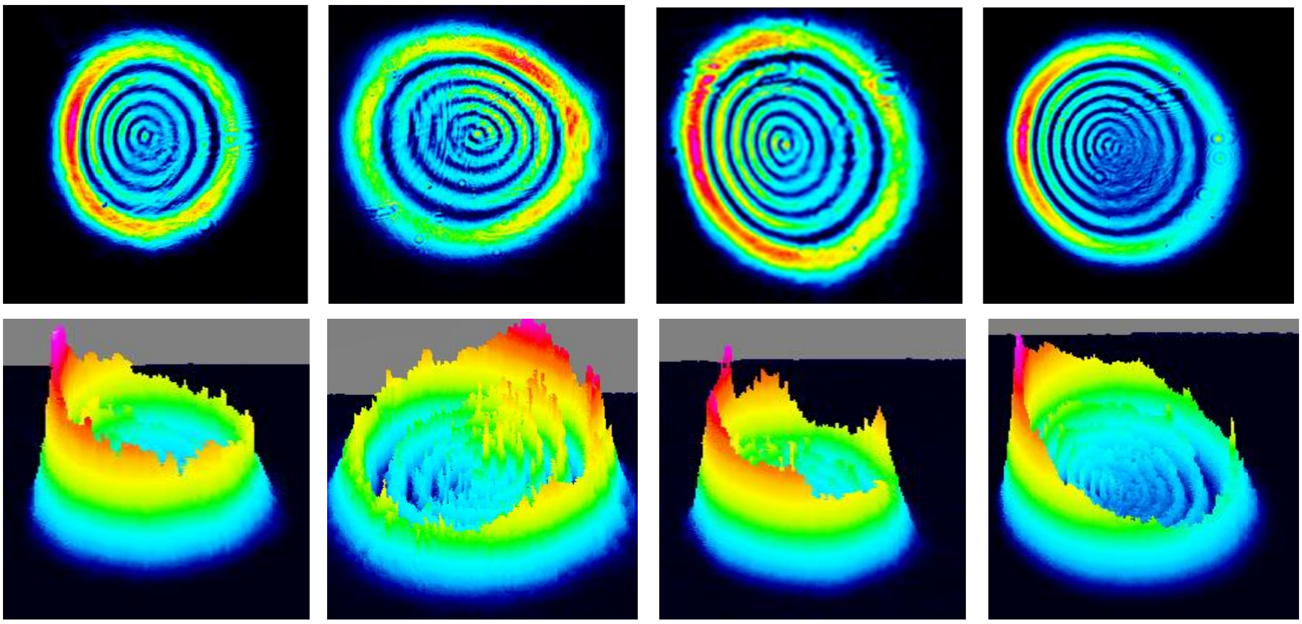

Fig. (2) CCD camera photographs for sample A, B, C and D with average particle sizes 15, 30, 50 and 70 nm respectively at laser intensity **784 W/cm^2** show: (1): The two- dimension image for the far-field diffraction ring patterns and (2): The three –dimension image for the far-field diffraction ring patterns.

colloids samples especially if it is mentioned that these samples never undergo the stirring process before they are applied in the experiment.

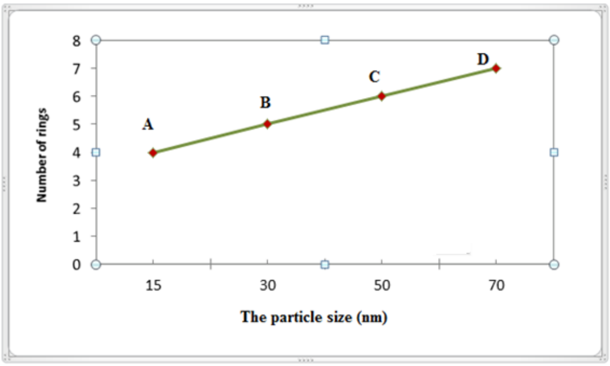

Fig. (3) :The increment in number of rings as the particle size increased in linear relationship for the nano-silver in DI samples A, B, C and D with average particle sizes 15, 30, 50 and 70 nm respectively.

The three- dimension images show how the intensity is high at the outer ring more than the inner ones (the red color refers to the higher intensity) which indicate the presence of third order nonlinear lens-like behavior of spatial self phase modulation SSPM (self-defocusing phenomenon) in the prepared silver nano-fluid.

The observed diffraction patterns are generated due to the interaction of a Gaussian beam with self-defocusing media. These patterns are so clear and their rings number is easy to count due to the homogeneity of the prepared nano-silver

It is well known that for Gaussian beam propagation through a lens-like medium, the maximum change of nonlinear refractive index $\mathbf{\Delta n_{nl,max}}$ is proportional to the number of fringes. The values of the $\Delta n_{nl,max}$ for all samples were determined and listed in Table (1) by Eq. 1 (10):

$$\Delta_{nl,max} = \frac{\lambda_{beam}}{L_{material}} N_{rings} \qquad (1)$$

where λ is the wavelength of the light, $L_{materiel}$ is the thickness of the sample and N is the number of rings. In this experiment, λ_{beam} equals **473 nm** and $L_{materiel}$ is **5 mm**.

Table (1) Number of diffraction rings, the maximum change of nonlinear refractive index, the nonlinear refractive index and the phase shift for the prepared silver nanoparticles in DI at laser intensity of **784 W/cm^2.**

Samples	Average Particle size (nm)	Number of rings	$\Delta n_{nl,max} \chi 10^{-4}$	n_2 $\chi 10^{-7}$ cm^2/W	(Δ φ)
A	15	4	3.78	4.8	8 π
B	30	5	4.73	6.0	10 π
C	50	6	5.67	7.2	12 π
D	70	7	6.62	8.4	14 π

At high laser intensity **784 W/cm^2** and with increasing the particle size, the maximum change of nonlinear refractive index is increased too from **3.78 x 10^{-4}** up to **6.62x 10^{-4}** as the particle size increased from **15 nm** to **70 nm,** respectively**.** We can estimate the effective nonlinear refractive index n_2 for all samples of silver nano-fluids from the relation (11):

$$\Delta n = n_2 I \qquad (2)$$

where Δn is the maximum change of nonlinear refractive index and I is the input intensity. The results illustrated in Table (1) show that the nonlinear refractive index for silver nano-fluids increases with the increment of the particle size from **4.8 x 10^{-7} cm^2/W** up to **8.4 x 10^{-7}** cm^2/W as the average particle size increased from **15 nm** to **70 nm**. This would indicate that by increasing the nanoparticle size, the nano-fluid would possess more nonlinearity; these results agree with reference [12]. The major factor influencing the diffraction ring pattern is the nonlinear refractive index of the material; i.e, diffraction ring patterns can be realized easily in the materials of high nonlinear refractive index [13]. Since the laser beam used in the experiment has a Gaussian distribution, the relative phase shift (Δφ) suffered by the beam while traversing the sample of thickness (L) can be written as (14):

$$\Delta\Phi = KL\Delta n \qquad (3)$$

where $k=2\pi/\lambda$ is the wave- vector in vacuum and $\boldsymbol{\lambda}$ is the laser beam wavelength. The phase shift ($\Delta\ \varphi$) at higher intensity **784 W/cm^2** was determined in Table (1) for all samples. We observed the phase shift tends to increase linearly with the increasing of particle sizes from **$\Delta\varphi = 8\ \pi$** up to **$14\ \pi$** for the AgNPs in DI that had average particle size increased from **15 nm** to **70 nm,** respectively. In the presence of phase distortion greater than π, the SSPM of single beam leads to self-diffraction rings around the propagation direction [15]. This can be observed by the expansion in the rings number in the self-diffraction patterns, while the particle size was increased.

Conclusions

In summary, the influence of the changing in particle size of Ag nanoparticles in DI on their nonlinear behavior was studied. The diffraction patterns were recorded in two-dimension and three-dimension images. The increase in the number of rings with particle size increment was experimentally verified. The maximum change of nonlinear refractive index **$\Delta n_{nl,max}$** , the nonlinear refractive index n_2 and phase shift $\Delta\varphi$ of Ag nano-fluids were successfully measured for a range of different particle sizes. It was found that by increasing the particle size, the material would possess more linearity.

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استقصاء اعتماد اللاخطية على الحجم لعالق الفضة النانوية باستخدام تقنية التشتت – الذاتي

سالي عبد الرزاق العاني مازن مانوئيل الياس

معهد الليزر للدراسات العليا، جامعة بغداد، بغداد، العراق

الخلاصة: في هذا العمل تم دراسة تأثير اختلاف الحجم على الصفات البصرية اللاخطية لعالق الفضة النانوية في الماء المنزوع الايونات. وقد تم تسجيل الملاحظات التجريبية للانماط البعيدة مدى الاستلام بواسطة كاميرا بصور ثنائية وثلاثية الابعاد. ان مقدار التغير الاعلى في معامل الانكسار اللاخطي قد تم حسابه مع فرق الطور المتعلق به وباستخدام تقنية التشتت الذاتي بواسطة موجة مستمرة من ليزرصلب ازرق مضخ بالدايود ليزر ذي طول موجي **473 نانومتر** وبطاقة **270 ملي واط** . حيث ان جميع نماذج عالق الفضة المحتوي على الاحجام **15 و 30 و50 و70** نانومتر المستخدم في هذه الدراسة قد تم تحضيرها كيميائياً. وجدنا ان معامل الانكسار اللاخطي يعتمد على حجم الجسيمات وضمن المرتبة $\mathbf{10^{-7}}$ **واط / سم 2**. في حين ان مقدار التغير الاعلى كان بالمرتبة 10^{-4}. لوحظ ان فرق الطور قد ازداد من 8π صعودا الى 14π مع زيادة حجم الجسيمات . ان الزيادة في حجم جسيمات الفضة النانوية يؤدي الى زيادة في عدد الحلقات وامتلاك المادة بذلك لا خطية اكبر مع ارتفاع معامل الانكسار اللاخطي.