



Experimental Study of Spatial Self-Phase Modulation (SSPM) Based on Laser Beam and Hybrid Functionalized Carbon Nanotubes/Silver Nanoparticles (F-Mwcnts/Ag-Nps) Acetone Suspensions

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Abstract: Focusing of Gaussian laser beam through nonlinear media can induce spatial self-phase modulation which forms a far field intensity pattern of concentric rings. The nonlinear refractive index change of material depends on the number of pattern rings. In this paper, a formation of tunable nonlinear refractive index change of hybrid functionalized carbon nanotubes/silver nanoparticles acetone suspensions (F-MWCNTs/Ag-NPs) at weight mixing ratio of 1:3 and volume fraction of 6×10^{-6} , 9×10^{-6} , and 18×10^{-6} using laser beam at wavelength of 473nm was investigated experimentally. The results showed that tunable nonlinear refractive indices were obtained and increasing of incident laser power density led to increase the nonlinear refractive index changes of suspension at each volume fraction. Moreover, the nonlinear refractive index changes ($\Delta n_{nl,max}$) at volume fraction of 18×10^{-6} irradiated by various laser intensities (137.31, 200, 228, 268, and 366.61 W/cm²) have higher value than others.

Key words: Functionalization of carbon nanotube, Nonlinear optics, nonlinear diffraction.

Introduction

A nonlinear optical phenomenon has been widely observed in various material such as optical materials and nanomaterials [1,2]. Nanomaterials carbon nanotubes (CNTs) possess high nonlinearity, unique electronic, chemical, and mechanical properties which can be used as nonlinear material in a numerous of potential applications[3]. Although, the outer wall of raw CNTs has to be visualized chemically inactive and the faces have been restricted in term of dispersion feasibility and caused bundling between carbon nanotubes tubules, that affected by the attractive van der Waals interaction among themselves [4]. One of the most hopeful ways to overcome those difficulties is the functionalization of CNTs.

High aspect ratios of CNTs is due to their smaller diameters and longer lengths makes it feasible to functionalize and to be used as a templates for nanoparticles (NPs) assemblies as well as improvement of their thermal properties[10]. Moreover, silver nanoparticles (Ag-NPs) also has ability to increase the nonlinear optical properties of the nano-fluid. This ability can be initiated from the presence of intense absorption peak in the visible range due to the surface plasmon resonance of electron[6]. Therefore, the hybrid (F-MWCNTs/Ag-NPs) acetone suspensions have to be used as a nonlinear media in the nonlinear diffraction technique. Hence, a large number of spatial effects can be revealed when an intense of a Gaussian laser beam passing through nonlinear media with an intensity

dependent refractive index[7-10]. Moreover, this process can produce a far field intensity pattern of concentric rings[11], and the number of rings indicates the maximum nonlinear refractive index change of material.

Experimental Work

Pristine-MWCNTs were covalently functionalized by utilizing a mixture of concentrated sulfuric acid and nitric acid. Foremost, 0.2 g of (MWCNTs) was treated with 100 mL of (98% H₂SO₄) and (63.01% HNO₃) with volume mixing ratio (3:1) in flask of 500ml. The flask was put in ultrasonic bath at temperature of 45°C during a time of 60 minute. The mixture was diluted with 600 mL of deionized water (D.I) and vacuum-filtered through a 0.22 μm polycarbonate membrane. Also that the filtered product was washed with D.I water until it's PH value became neutral. Moreover, the solid carbon was put in the laboratory oven at 90°C overnight .After one day the steps have been repeated by substituting the solvent with H₂O₂ (35%) as reduction reagent and the process procedure was identically repeated. The subsequent treatment with H₂O₂ was done in order to accomplish the oxidative process which started by H₂SO₄ and HNO₃, as a second step with a gentler manner. Figure (1) shows the MWNT with carboxylic groups covalently attached to the side walls .

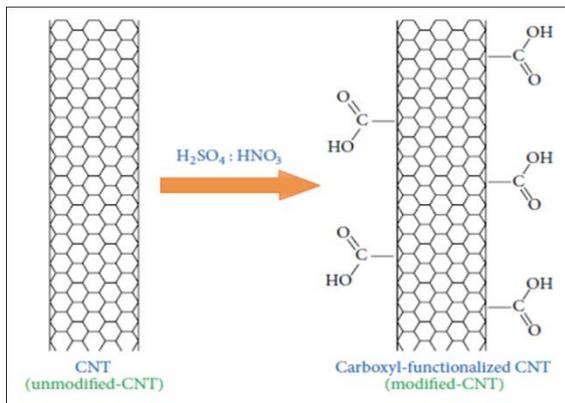


Fig. 1: Scheme of the chemical functionalization of a MWNT.

Hybrid nano-fluid was prepared in two-steps method. The weight of the nanoparticles was determined each one alone, and mixing them with weight mixing ratio of 1:3. Then, putting the previous weight of F-MWCNTs/Ag-NPs in a certain volume of acetone. Therefore, volume fraction (V) of nanoparticles suspended in the

solution has been calculated using equation1[12].

$$V = \frac{vs}{vs+VL} \tag{1}$$

Where *vs* is the volume of the particles and *V_L* is volume of acetone and *vs* = *m*/*ρ* where *ρ* is the mass density of the nanoparticles and *m* is the particles mass dispersed in the acetone.

In this work the diffraction patterns were obtained using Gaussian beam of CW diode pumped solid state laser (model MBL-FN-473nm-299mW-15050466), attenuator, Lens, CCD Camera, Laser Power-Meter and PC, as it is shown in Fig.(2).

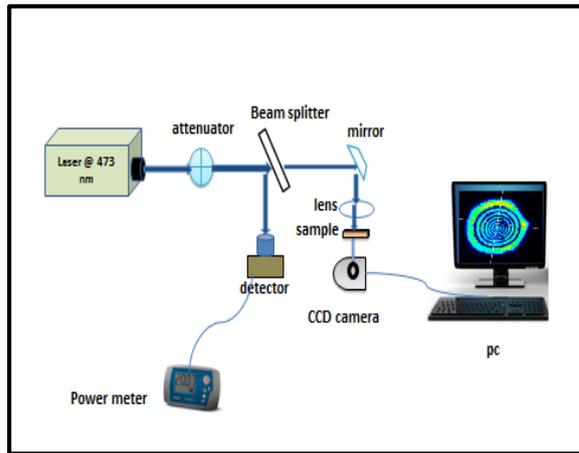


Fig. 2: Schematic diagram of the experimental set up.

Result and discusses

Fourier Transform Infra-Red (FTIR) is mainly used for the evaluation of the hydroxyl (-OH) and carboxyl (-COOH) groups that connected onto the surface of MWCNTs. Figure (3) shows FTIR Spectrum of F-MWCNTs using acid treatment. A small peak at 1571 cm⁻¹ was observed, corresponding to C=C stretching vibration bound of the CNTs which agreement with[13]. Broad absorption band has been noticed around 3439 and 3736 cm⁻¹ due to the O-H of the hydroxyl group stretching which agreement with[14]. The spectra of F-MWCNTs showed peak at 1703 cm⁻¹ which can be attributed to the C=O stretching of carboxylic acid group which agreement with[15]. Hence, it is conjecture that after treating MWCNTs by acid. The hydrophilic carboxyl (-COOH) groups became cohered onto MWCNTs surface.

The linear absorption spectra of the hybrid materials (F-MWCNTs /Ag- NPs) acetone suspension at volume fraction 9×10^{-6} was recorded by a Shimadzu UV-1800 spectrometer as shown in Fig.4. The spectrum showed absorption peaks at 475 nm which indicates that the utilizing laser source (473nm) was suitable for this process.

Due to the nonlinear spatial self-phase modulation (SSPM) effect, a series of concentric rings have been observed after focusing CW laser beam through a cuvette containing the previous hybrid nano-fluid (F-MWCNTs/Ag-

NPs acetone suspension) as it shown in Fig.(5). Thus, the increasing in the diffraction rings was due to the physical concept, that the hybrid metal nanoparticles (F-MWCNTs/Ag-NPs) spread in fluid lead to more absorption of the light energy due to Surface Plasmonic Resonance (SPR) of the free electrons excitation on the metal surface.

This process leads to transport the absorbed energy to particles and then to the encircling liquid results in suspensions temperature increasing which leads to change the refractive index of the nano-suspension.

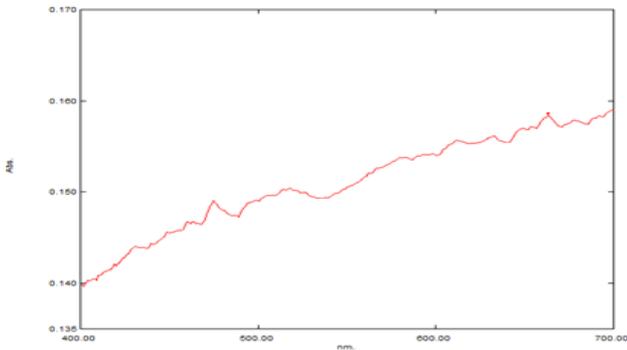


Fig. 3 : FTIR spectra of functionalized F-MWCNTs with acid treatment.

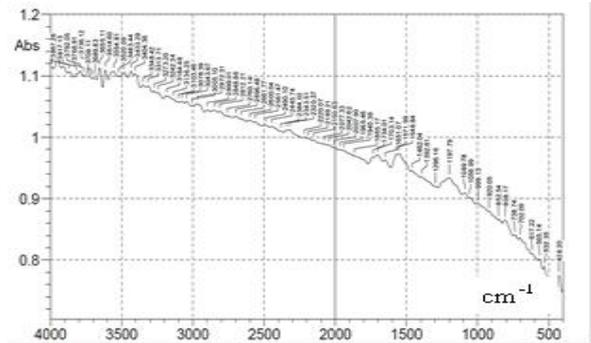


Fig. 4 : The UV-VIS absorption spectrum of MWCNTs/Ag-NPs acetone suspension at volume fraction 9×10^{-6}

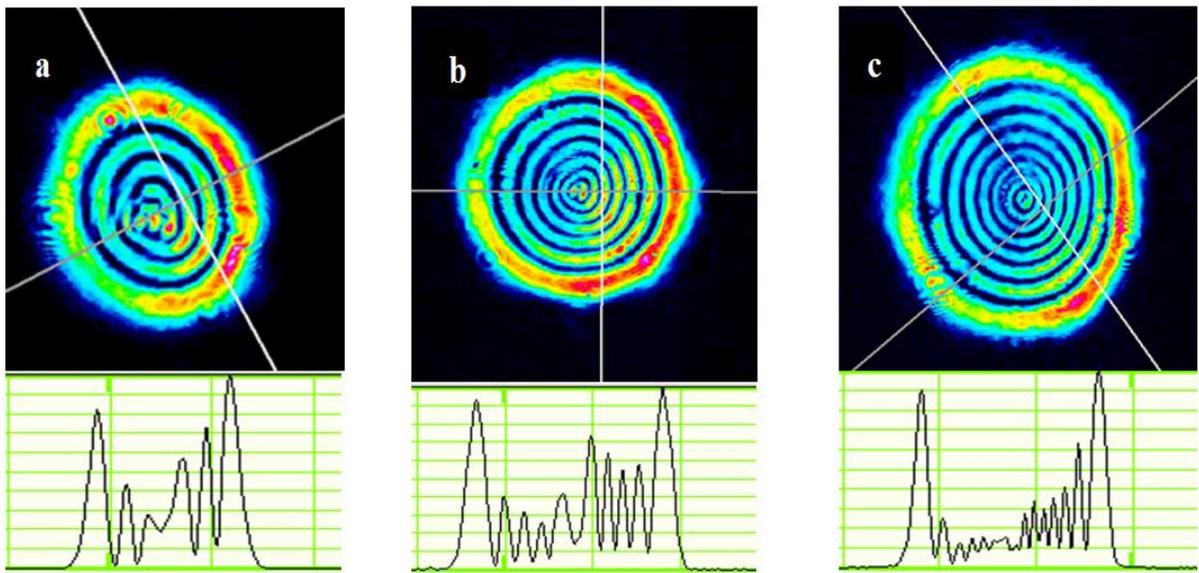


Fig 5: Diffraction patterns rings observed at volume fraction value of 6×10^{-6} irradiated by various laser intensity a) 137.31 W/cm^2 b) 200 W/cm^2 c) 228 W/cm^2 respectively.

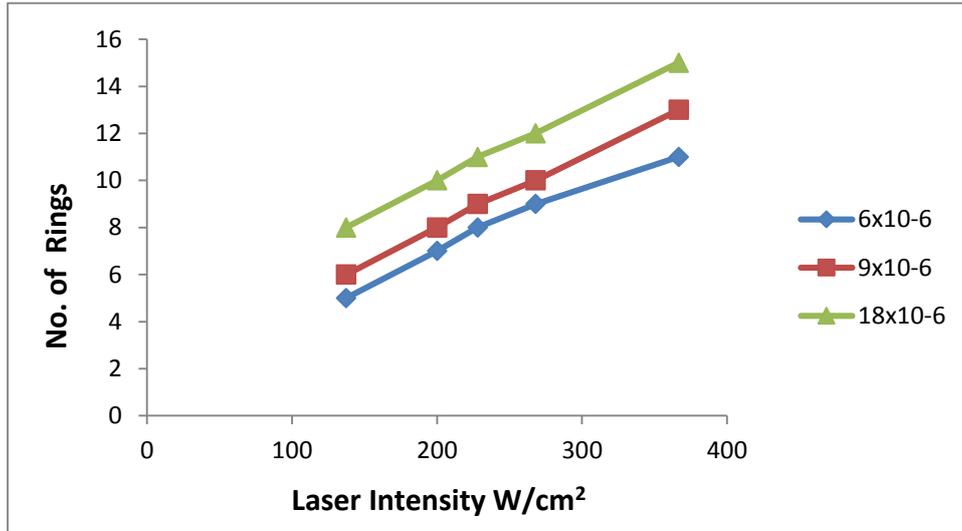


Fig. 6: The number of diffraction rings formed using (F-MWCNTs/Ag NPs) acetone suspension at various volume fraction irradiated by various laser intensities.

Figure 6 shows that the number of pattern rings increases with increasing of the laser intensity incident on hybrid nano-fluid (F-MWCNTs/Ag-NPs).

In order to know the effective nonlinear refractive change the laser power density was

fixed at a constant value (228 W/cm^2) and the highest number of rings was obtained at volume fraction of 18×10^{-6} as it is shown in the Figures 7. Therefore, the highest nonlinear refractive index change was achieved at the same volume fraction (Figure 8).

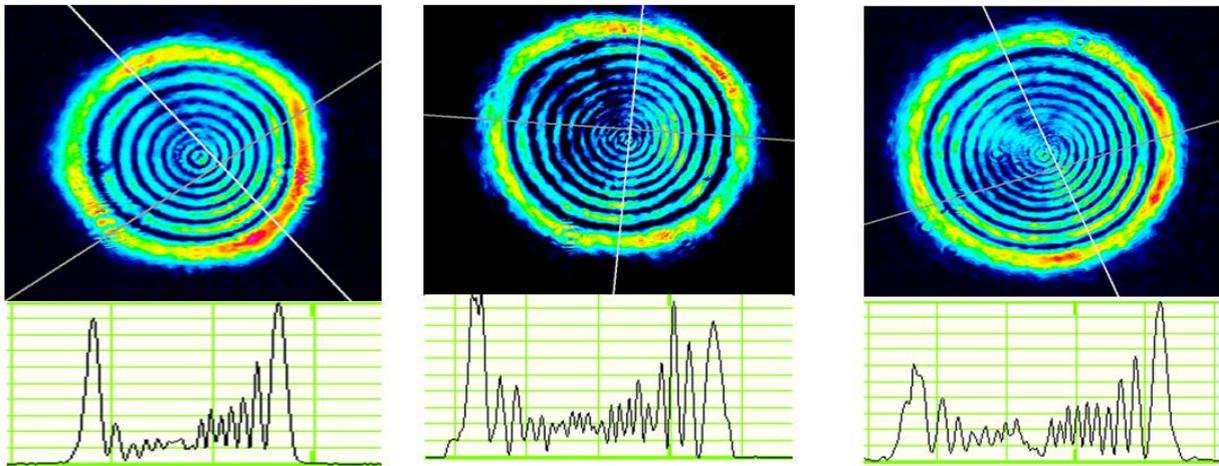


Fig.7: The self-diffraction pattern rings generated by using F-MWCNTs /Ag-NPs acetone hybrid nano-fluid at volume fraction of 6×10^{-6} , 9×10^{-6} and 18×10^{-6} respectively at laser power density 228 W/cm^2 .

Where the maximum change of nonlinear refractive indices ($\Delta n_{nl,max}$) for the previous suspensions have been calculated utilizing the following formula[16];

$$\Delta n_{nl, \max} = \frac{\lambda_{\text{beam}}}{L_{\text{material}}} N_{\text{rings}} \quad (2)$$

Where λ , L and N_{ring} are the laser wavelength, the material thickness, and the number of formed diffraction pattern rings respectively. It has been noticed that the value of $\Delta n_{nl, \max}$ of the used hybrid nano-fluid at volume fraction of 18×10^{-6} was higher than that of others as shown in Fig.(8).

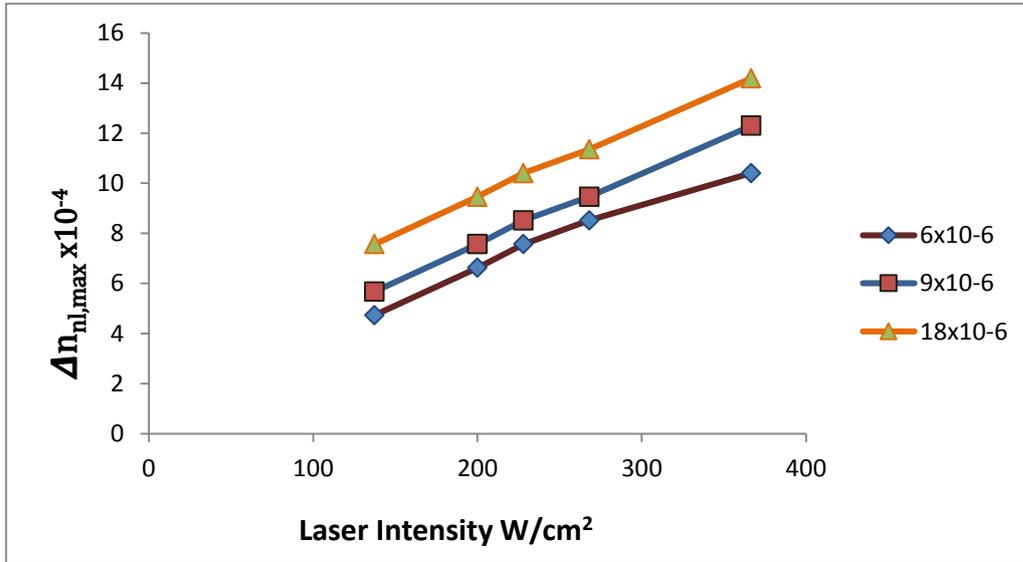


Fig. 8: The maximum change of F-MWCNTs/Ag-NPs acetone suspension at volume fraction value of 6×10^{-6} , 9×10^{-6} and 18×10^{-6} .

For each volume fraction value of F-MWCNTs/Ag-NPs with different applied intensities(I), the nonlinear refractive index (n_2)

was calculated employing equation 3 [17].

$$n_2 = \Delta n_{nl,max} / I \quad (3)$$

Table (1): The nonlinear refractive index of hybrid F-MWVNT_s/Ag-NPs acetone nano-fluid at various volume fraction and various laser intensity.

| Laser Intensity (W/cm ²) | $n_2 \times 10^{-6} \text{ cm}^2/\text{W}$ at volume fraction of | | |
|--------------------------------------|--|--------------------|---------------------|
| | 6×10^{-6} | 9×10^{-6} | 18×10^{-6} |
| 137.31 | 3.78 | 4.13 | 5.511 |
| 200 | 3.31 | 3.78 | 4.73 |
| 228 | 3.31 | 3.73 | 4.56 |
| 268 | 3.17 | 3.52 | 4.23 |
| 366.61 | 2.83 | 3.35 | 3.87 |

Table (1) shows that the nonlinear refractive index (n_2) increased by increasing the volume fraction of nano-fluids and decreased by increasing of incident laser intensity.

fraction value of hybrid nano-fluids also caused to increase both of $\Delta n_{nl,max}$ and n_2 of material. Consequently, a tunable maximum nonlinear refractive index changes were obtained by changing either the laser intensity or the volume fraction value .

Conclusions

In summary, a higher thermal conductivity and a good stability of MWCNTs suspended in acetone with a low aggregation were achieved by the functionalization process. As SSPM considered a thermal process, therefore, it was noticed that increasing of the incident laser intensity caused increasing the number of the diffraction pattern rings which lead to increase $\Delta n_{nl,max}$ of material (F-MWCNTs/Ag-NPs acetone). Moreover, increasing of the volume

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دراسة عملية لتنميط الطور الذاتي المكاني اعتمادا على حزمة ليزر ومعلق استون لهجين موظف للكربون الانبوبي النانوي متعدد الجدران/ جسيمات الفضة النانوية

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الخلاصة: ان حصر حزمة كاوسية لليزر خلال مادة لا خطية يمكن ان تحفز تنميط طور ذاتي مكاني والذي يكون نمط لدوائر متعددة المركز للاضاءة بعيدة المجال. أن تغير معامل الانكسار اللاخطي للمادة يعتمد على عدد حلقات النمط. في هذه الورقة تم التحري عمليا على تكوين منغيرات لمعاملات انكسار لا خطية متناغمة لمعلق استون وهجين موظف الكربون الانبوبي النانوي متعدد الجدران/جسيمات الفضة النانوية وبنسبة خلط وزنية 1:3 وباحجام كسرية $6 \cdot 10^{-6}$, $9 \cdot 10^{-6}$ و $18 \cdot 10^{-6}$ باستخدام حزمة ليزر نو طول موجي 473 نانومتر. رات النتائج انه تم الحصول على معاملات لاخطية متناغمة للمعلق وأن ازدياد كثافة القدرة الساقطة لليزر ادت الى زيادة تغير معامل الانكسار اللاخطي للمعلق ولكل حجم كسري. بالإضافة الى ان تغير معامل الانكسار اللاخطي عند الحجم الكسري $18 \cdot 10^{-6}$ كان اعلى من الآخرين عندما تم التشعيع بواسطة شدة ليزرية متنوعة (137,31، 200، 228، 268، 366,61 واط/سم²).