The 27th Iranian Conference on Optics and Photonics (ICOP 2021), and the 13th Iranian Conference on Photonics Engineering and Technology (ICPET 2021).University of Sistan and Baluchestan, Zahedan, Iran, Feb. 2-4, 2021

کد مقاله : ۳-۱۶۹۴-۱۰-A

بررسی لیزرهای کاتوره‌ای شامل ذرات پراکننده با اثرات غیرخطی

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چکیده- تاثیر خواص نوری غیرخطی در لیزرهای کاتوره‌ای، نتایج ارزشمندی را به همراه داشته است. در این پژوهش، اثر ضریب شکست غیرخطی ذرات پراکننده را بر گسیل لیزر کاتوره‌ای بررسی می‌کنیم. به این منظور، میکروساختارهای اکسید مولیبدن با استفاده از روش لایه‌نشانی الکتروشیمی سنتز شده‌اند. ضریب شکست غیرخطی این ساختارها با انجام روش جاروب z بررسی شده است. سپس با افزودن این ساختارها به محیط یک لیزر کاتوره‌ای معمولی، تغییراتی در تابش آن‌ها مشاهده شده است. نتایج نشان داده‌اند که به ازای انرژی‌های دمش بالاتر از آستانه، علامت ضریب شکست غیرخطی می‌تواند بر شدت گسیلی تاثیر بگذارد. به این صورت که، نمونه‌ای با علامت مثبت ضریب شکست غیرخطی منجر به افزایش در شدت گسیلی شده است.

کلید واژه- اکسید مولیبدن، روش جاروب z، ضریب شکست غیرخطی، لیزر کاتوره‌ای.

Investigation of Random Lasers Consisting of Scattering Particles with Nonlinear Effects

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Abstract-The effect of nonlinear optical properties in random lasers (RLs) has brought valuable results. In this study, we present the influence of the nonlinear refractive index of scattering particles on RL emission. In such regard, microstructures of Molybdenum Oxide were synthesized by the electrochemical deposition method. The nonlinear refractive index of the structures was investigated by performing the Z-scan technique. Next, variations in their emission were observed by adding the structures to a conventional RL. The results have demonstrated that the sign of the nonlinear refractive index can affect the emitted intensity for the pump energies above the threshold. In such a way that the sample with a positive sign of the nonlinear refractive index has led to an increase in the emitted intensity.

Keywords: Molybdenum Oxide, Nonlinear refractive index, random laser, Z-scan technique.

The 27thIranian Conference on Optics and Photonics (ICOP 2021)
The 12thIranian Conference on Photonics Engineering and Technology (ICPET 2021)
University of Sistan and Baluchestan, Zahedan, Iran, Feb. 2-4, 2021.

1. Introduction

In the past several decades, random lasers (RLs) have attracted much interest due to their significant features and applications [1]. In such lasers, multiple scattering due to a disordered medium has played an important role to provide laser feedback. However, the amplification process due to a gain medium works on the same principles as in conventional lasers. In other words, the RLs mechanism leads the light to be frequently scattered within the gain medium and be amplified along some random paths. Based on the scattering regimes, RLs are classified into non-resonant and resonant random lasers. In the first type of such lasers, the scattering is not strong enough. As a result, a single broad peak formed in the emission spectrum. However, for the resonant RLs, the emission spectrum consists of several narrow lasing peaks owing to strong scattering. In this case, light can return to the first scatterer and be trapped in a closed-loop path.

The nonlinear optical response of materials has been studied with several methods including the Z-scan technique, which relies on the variations of beam irradiance [2]. In this method, a nonlinear medium is translated across the focal point and along the direction of beam propagation. Therefore, the transmittance changes due to light-matter interactions. It is known as the open aperture Z-scan, while by placing an aperture in the far-field region of the beam, we can have the closed aperture Z-scan. Thereby, both the nonlinear absorption and the nonlinear refractive coefficients can be measured with the above setups respectively.

Transition metal oxides such as Molybdenum Oxide are attracting widespread interest due to their wide range of optical and electrical properties. In particular, Molybdenum trioxide exhibits pronounced opportunities for applications due to its wide optical bandgap [3]. Moreover, it can be prepared by a variety of techniques, which include electrochemical deposition [4]. The advantages of this method are low growth temperature, low-cost, etc.

Few studies have been published on the nonlinear effects in rando. m lasersIn 2019, Prizia et al. studied the nonlinear absorption of graphene oxide (GO) using the Z-scan experiment. They observed the reverse saturable absorption behaviour of GO. Next, they added GO to a standard random laser based on titanium dioxide powder and rhodamine B. Finally, it was shown that the RSA of the GO caused a sharp decrease in the RL emission [5].

This study aims to investigate the effects of the nonlinear refractive index of scattering particles on random laser emission. In this context, we briefly present the synthesis and measurement of the nonlinear optical properties of Molybdenum Oxide structures using the Z-scan technique. Finally, we demonstrate the influence of adding them into a standard laser and its emission.

2. Experiments

The electrochemical deposition experiments were carried out using a three-electrode glass cell. Two fluorine-doped tin oxide (FTO) glasses were used as working and counter electrodes. Also, an Ag/AgCl electrode is used as the reference electrode. In addition, an aqueous solution of Na_2MoO_4 (0.3 M and 0.6 M) and Na_2SO_4 (1.2 M and 0.6 M) were prepared as the electrolyte and supporting electrolyte respectively. Besides, the above cell and the electrolyte were immersed in a water bath at 70˚C. The experiment was performed in a constant voltage of -1.1 V.

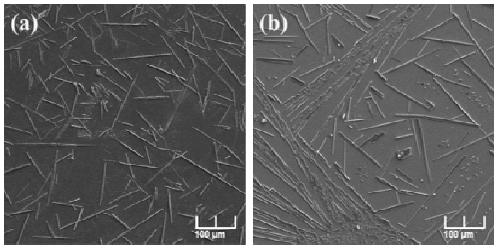

Fig. 1: The scanning electron microscopy images (SEM) of (a) Sample 1 and (b) Sample 2 in 100 μm scale.

Finally, the thin films were annealed at 500°C for 2 h in an oven. Above all, two samples with different electrolyte concentrations were synthesized.The morphologies of the synthesized structures were analyzed by using scanning electron microscopy (SEM) which its images in 100 µm scale are shown in Fig. 1.As can be seen, the structures are rod in nature and on the scale of the micrometer. Also, they were found to be randomly distributed.

3. Results and Discussion

In order to investigate the nonlinear response of the samples, the Z-scan experiment has been carried out with a continuous wave Nd: YAG laser at 532 nm. A Gaussian beam with an input power of 18 mW was focused on the sample using a 2 cm focal length lens. The transmittance variations were detected and Fig. 2 demonstrates the closed aperture Z-scan curves.

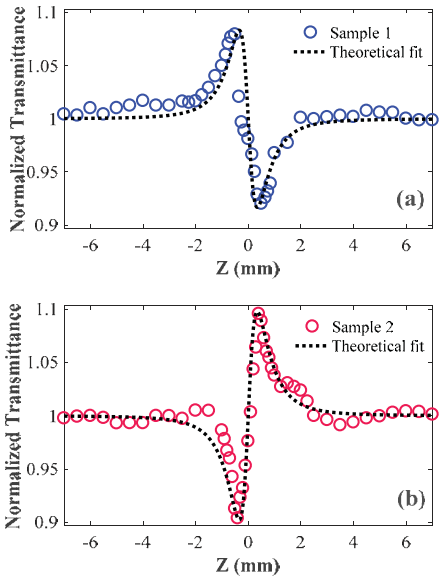

Fig. 2: Normalized closed aperture Z-scan curves for (a) Sample 1 and (b) Sample 2.

As can be seen in Fig. 2(a), which corresponds to sample 1, a pre-focal peak is followed by a post-focal valley. It clarifies the self-defocusing property and the negative sign of the nonlinear refractive index. However, for sample 2 the opposite phenomenon has occurred which states the positive sign of the nonlinear refractive index and it is shown in Fig. 2(b). Generally, the change in sign of the nonlinear refractive index might be due to the changing of the electrolytes concentration and the existence of the sulfur element with different concentrations. In addition,the resulting curves were fitted with the theoretical equation [2] and the nonlinear refractive coefficients were measured and presented in Table 1. Furthermore, the open Z-scan experiment found evidence for a negligible amount of the nonlinear absorption coefficients as Illyaskutty et al. have already claimed for the pure Molybdenum Oxide [6].

Table I. The nonlinear refractive index of Molybdenum Oxide structures.

Sample No	n_2 $\times 10^{-8}$ (cm^2/w)
1	-2.07
2	2.42

Since we were interested to study the influence of nonlinear effects on random lasers, we decided to add the synthesized Molybdenum Oxide structures as scatterer particles to a standard random laser based on titanium dioxide (TiO_2) powder and rhodamine B dye. With the purpose of this RL fabrication, we drop coated the surface of the above thin films with a solution consisting of 0.06 M TiO_2 and 0.007 M rhodamine B in the ethylene glycol. Next, the samples were pumped with a 532 nm frequency-doubled Nd-YAG pulsed laser, which was focused using a 15 cm focal length cylindrical lens. Fig. 3 shows the emitted intensity and Full width at half maximum (FWHM) of the samples versus different pump energies. It should be noted that a pure sample without Molybdenum Oxide Structures was also prepared for a better comparison. As can be seen in Fig. 3(a) and (b), there is a sudden change in the intensity growth slope above a specific value of pump energy, which indicates the laser threshold behavior. It is also consistent with the value from FWHM. The results show that there are no significant differences between the thresholds corresponding to both samples. However, the data show significant variations in emitted intensity for the samples including Molybdenum Oxide compared to pure one.

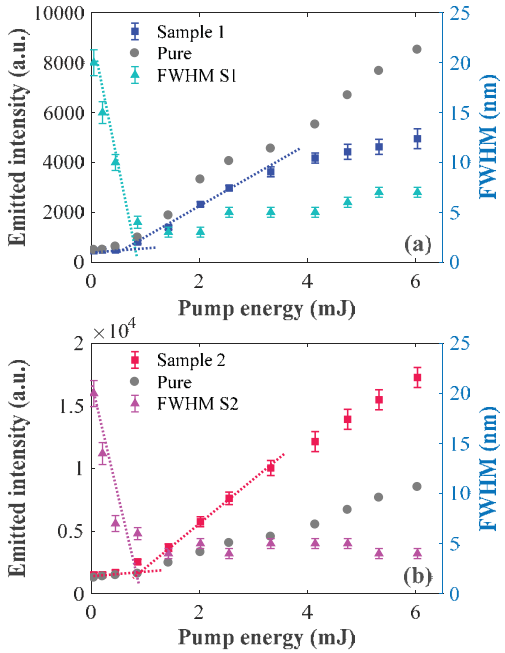

Fig. 3: Maximum emitted intensity and Full width at half maximum versus different pump energies corresponding to (a) Sample 1 and (b) Sample 2 compared to the pure Sample.

As indicated in Fig .3(a), the emitted intensity of S1 decreases compared to the pure one. While Fig .3(b) demonstrates an increase in the emitted intensity of S2. It is worth mentioning that both these variations occur above the threshold and for the higher pump energies. With respect to the above considerations, we speculate that these variations might be due to the nonlinear optical properties of Molybdenum Oxide. For S1, which has the negative nonlinear refractive index, the self-defocusing effect might have led to such behaviour. However, the Self-focusing effect might be the reason for behaviour of S2 in Fig. 3(b). Furthermore, the results in Fig. 3(b) might be due to the fact that the refractive index contrast and the multiple scattering have increased according to the Kerr nonlinearity and positive sign of nonlinear refractive index of S2. Finally, these findings confirm that random lasers can be influenced by nonlinear effects.

4. Conclusions

In conclusion, the microrods of Molybdenum Oxide were synthesized using the Electrodeposition method. Their nonlinear refractive index was measured by performing the Z-scan technique. The structures were used as scattering particles in a random laser based on TiO_2 and rhB. The differences in emission intensity for pump energies above the thresholdhaveshown compared to the conventional RL emission. We have observed an enhancement in the emission intensity corresponding to the structure with a positive sign of the nonlinear refractive index, which was due to the self-focusing effect and the enhancement of refractive index contrast. However, the emitted intensity has reduced for the sample with a negative sign of the nonlinear refractive index.

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