Coupled Modes Enhance Random Lasing in Plasmonic Thin Etalons

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Research Article

Keywords: Plasmonic random laser, Light-emitting polymers, etalon nanostructure, Multiple scattering.

Posted Date: January 21st, 2022

DOI: <https://doi.org/10.21203/rs.3.rs-1234492/v1>

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Abstract

In this work, we experimentally and theoretically proved a flexible fine random laser from a two-face etalon plasmonic structure based on PDMS. Accordingly, PDMS was fabricated using the nanoimprint lithography method and coated by a thin gold layer with a thickness of 35 nm using a PVD device and light-emitting polymer (F8PT) to enhance the scattering and efficiency of the random laser. Using a plasmonic gold grating as a substrate, the simulation results compared the upside and downside of the plasmonic etalon structure. Moreover, an enhancement was observed in light transmission, and it is common to predict high efficiency in random lasing properties. The experimental results showed a comparison between normal plasmonic etalon samples and symmetric and asymmetric etalon-based nanostructures with thicknesses of 200, 400 and 600 μm and reported that random lasing properties had better results for samples with thinner spaces based on coupled mode effects. Correspondingly, this was done by increasing the intensity and decreasing the lasing threshold from 22 μJ in normal etalon to 16 μJ in the thinnest etalon structure.

I. Introduction:

Recently, random plasmonic lasers with high efficiencies based on different nanostructures have attracted much attention due to their flexibility, ease of manufacture, and wide application [1–5]. Of the motives, plasmonic random laser beams could also be obtained by scattering in both liquid and thin film media, but solid-state media are considered more efficient, which solves many problems in practical applications [6, 7]. Multiple scattering and optical gain can be accessed by two kinds of materials, which can lead to amplification in random media [8]. Nanoparticles play an important role as scattering centers in random media, depending on the type of nanoparticles. Additionally, optical gain can be provided with light-emitting polymers [9], dyes [10], semiconductors [11], etc.

Plasmonic etalon nanostructures are used to prepare random lasers in thin films by using light-emitting polymers, which have properties different from those of conventional lasers. For example, they are easy to fabricate and cost-effective and have small mode volumes [12–14]. The emission spectrum can be enhanced in the plasmon-assisted random laser by coupling the light-emitting polymer and the localized LSPR of the gold layer. Under the condition of enough overlap between the emission spectra of light-emitting polymers and the LSPR spectrum of the gold layer, the gold layer could usually absorb the incident photons and transfer them to the obtained medium (light-emitting polymer), leading to low threshold random laser action [15]. In our previous work, the Au NPs had a larger scattering cross-section due to enhanced scattering in the green region due to LSPR [16]. Therefore, the present study aimed to utilize one- and two-face etalon samples to achieve a lasing green region. For this purpose, we studied the lasing properties for both one- and two-face etalon samples coated with a gold layer and light-emitting polymer (F8PT). As a result, it was shown that the emission spectrum for the sample with two faces has better results than the sample with one face. Accordingly, this was done by the apparent increase in the intensity, a lower lasing threshold, and a full-width at half-maximum (FWHM) of approximately 1 nm at the same pumping energy.

II. Experimental Part:

In this research, double 2D grating (or Etalon) nanostructures based on polydimethylsiloxane (PDMS) were fabricated using the nanoimprint lithography method, and the random lasing properties of the proposed samples were investigated.

In this regard, a schematic layout of the fabrication process of the etalon-based nanostructure is shown in Fig. 1(a). For this purpose, two charge-coupled devices (CCDs) have been used with two-dimensional grating patterns. First, the first CCD was placed on a glass substrate, and a plexiglass frame was then placed around it. After that, the second CCD was placed on the top of the frame in front of the first CCD and then fixed by thermal adhesive. Additionally, a mixture of polydimethylsiloxane (PDMS) and curing agent (at a weight ratio of 10:1) was prepared and injected into the frame. The cell was placed inside a metal clamp to achieve good fixation of the CCDs on both sides of the frame.

Subsequently, for drying and fixing the 2D pattern onto the PDMS, the cell was placed on the heater at temperatures ranging from 50 to 100°C for 1 hour. Finally, the CCDs were removed from the sample after good drying of the PDMS composite, which took place for approximately one day at room temperature.

In this way, a double 2D grating nanostructure with a thickness of approximately 1.4 mm (normal etalon) was prepared to be coated on both sides by a thin gold layer with a thickness of 35 nm using a PVD device. Afterward, the sample was left for 24 hours before being coated with a layer of light-emitting polymer poly [(9,9-dioctylfluorenyl-2,7-diyl)-alt-co-(1,4-benzo-(2,1', 3)-thiadiazole)] (F8BT, American Dye Source) at a concentration of 8 mg/mL in xylene solutions. Next, these layers were coated onto one and two sides of the etalon sample using the drop weight method, and the actual image is shown in Fig. 1(b). Scanning electron microscopy (SEM) images of the two main stamps used as up- and downbuilding blocks of the etalon structure are shown in Figs. 1(c)-(d).

In the next step, symmetric and asymmetric etalon-based nanostructures with thicknesses of 200, 400, and 600 μm were fabricated according to the process mentioned above, and the random lasing properties of the fabricated samples were investigated. Asymmetric etalon-based nanostructures were produced by 45° rotating CCDs. Asymmetric plasmonic etalons can support several extra modes, which come from breaking the symmetry of the structure and the coupling between plasmonic and cavity (etalon) modes.

The random lasing properties of the etalon sample were studied by a second-harmonic generation Nd:YAG laser with a repetition rate of 10 Hz and pulse width of 5 ns. The required data were then collected by ocean spectrometry at 45° to facilitate the actual test. The pumped energy of the laser beam was tuned continuously using an optical attenuator, as shown in Fig. 1(e).

III. Results And Discussions:

Figure. 2 shows the light emitting polymer absorption spectrum, PL spectrum (F8PT) and electric field distribution of our normal etalon structure in three parts. As indicated, the broad absorption peak is at 456 nm with a Au layer (blue line) and without a Au layer (green line), and the absorption peak of Au NPs is observed at 526 nm (black line), corresponding to a broad localized surface plasmon resonance (LSPR) spectrum. Of note, the absorption spectrum of light-emitting polymer (F8PT) with a Au NP layer appears higher than that without Au NPs, which may be due to the (LSPR) Au NPs. In addition, the red line in Fig. 2(c) shows the PL spectra of the light-emitting polymer (F8PT), which was observed at 542 nm and measured by the spectrometer.

The proposed structure was simulated using the mode solutions module of the Lumerical software, and the optical electric field distribution was also investigated for our proposed structure. The simulated structure consisted of arrays of unit cells with lattice constants of $a_1 = 3040$ nm and $a_2 = 5150$ nm for both up- and downgrading, respectively, which were confirmed by scanning electron microscopy (SEM) images (Figs. 1(c)-(d)). The thicknesses of the PDMS, Au, and F8PT layers were estimated as $t_{\text{PDMS}} = 8000$, $t_{\text{Au}} = 35$ nm, and $t_{\text{F8PT}} = 250$ nm, respectively. In addition, a mesh size of 3.5 nm was considered in both the x and y directions. The refractive index values of PDMS and Au were obtained from the data presented in the study by Schneider et al. and Rakić et al., respectively [17].

The optical electric field distribution of the first three modes for the upper side of the etalon sample at a wavelength of 532 nm is shown in Figs. 2(a) and (b). In this figure, the light propagating inside the etalon structure in the x-direction after passing through the F8PT layer is observable. Furthermore, electric field localization around the gold grating was observed only for the left edge of the etalon structure (or upside). In addition, the optical electric field distribution of the first three modes for the downside of the etalon sample at a wavelength of 532 nm is shown in Fig. 2(b). As seen, the light propagated inside the structure, which enhanced the transmission of the input light and the electric field localization around the gold grating for the left edge of the etalon structure.

Correspondingly, this decreased the transmission of the central SLR wavelength, common in any plasmonic media. In addition, for the sample with a strong plasmonic substrate on the right edge of the etalon structure, we enhanced the transmission of input light. These two enhancements in the transmission of light from the sample with the plasmonic substrate make it common to predict high efficiency in random lasing by the sample with the plasmonic gold grating used as a substrate. Furthermore, we used a gold grating with a light-emitting polymer medium to enhance the scattering and efficiency, which are reported in the rest of this work.

Furthermore, reflection spectra of normal etalon and symmetric and asymmetric plasmonic etalons with thicknesses of 200, 400 and 600 μm were measured at incidence angles of 58 degrees, as shown in Figs. 3(a), (b) and (c), respectively. The excitation of the surface lattice resonance (SLR) of these 2D grating structures typically occurs at incidence angles ranging between 50 and 60 degrees [18, 19]. In this research, sweeping of the incidence angle showed that a sharp and strong SLR response occurs at an incidence angle of 58 degrees. As shown, SLR dips were achieved for the fabricated plasmonic etalon-

based nanostructure, corresponding to the coupling between localized surface plasmon resonances (LSPRs) of nanorods at the corner of each unit cell and diffracted order waves in the 2D periodic structure. As seen, the depth and width of the SLR mode increase with increasing thickness of the plasmonic etalon nanostructure. The plasmonic SLR mode is sharper for etalon with a thickness of 400 μm , and the broader SLR mode was measured for etalon with a thickness of 600 μm . In addition, several extra modes were measured for asymmetric plasmonic etalons (Fig. 3(b)), which come from breaking the symmetry of the structure and the coupling between plasmonic and cavity (etalon) modes.

To investigate the effects of these modes on the random lasing efficiency, we recorded random lasing of normal, symmetric, and asymmetric etalon-based nanostructures with different thicknesses of 1.4 mm, 200, 400, and 600 μm .

At first glance, Fig. 4(a) shows the emission spectrum of the normal etalon plasmonic sample random laser at different pump energies. The results were analyzed based on the changes in the slope efficiency and threshold lasing of the etalon plasmonic random laser by comparison with the face-coating samples.

In normal etalon samples, spontaneous emission was observed with low pumping energy below the lasing threshold. With an increase in pumping energy above the threshold power, the emission of the gain medium was strongly scattered, which resulted in a rapid enhancement in the spontaneous emission due to an increase in the optical path and confinement of emitted light by multiple scattering of these samples. Thus, a very narrow emission peak emerged at 541 nm, with a full width at half-maximum (FWHM) less than 1 nm, which implies the occurrence of coherent plasmonic random lasing (Fig. 4(b)).

In addition, the threshold power of the random lasing appearance reaches 22 μJ , and the maximum intensity reaches 30874 a.u. at a high pumping energy of 46 μJ due to strong confinement provided by the two faces of the etalon sample (Fig. 4 (c)).

At second glance, as mentioned before, we changed the etalons into three different thicknesses to obtain more useful modes and additional efficient random lasing. Thus, the emission spectra of the symmetric plasmonic etalon samples at 200, 400, and 600 μm were recorded (Fig. 5). It is observed that the emission spectrum of the symmetric etalon sample with a thickness of 200 μm is higher than that for the symmetric etalon samples 400 and 600 μm due to the effect of backscattering, which is featured here and plays a prominent role in improving the emission from the thinner etalon sample in comparison with the standard etalon sample, in agreement with the simulation results in Fig. (2).

To more evaluate the performance of the random plasmonic laser under the influence of the thickness of the plasmonic etalon samples, asymmetric etalon samples were used while maintaining the same thickness for the symmetric etalon samples used as shown in Fig. 5, by these samples, it is noticed that it offers better results than the first sample through the apparent increase in the intensity of the emission at the same pumping power, due to this style, the confirming losses between the asymmetric etalon samples were overcome, the possibility of forming a confined sharp peak with high intensity has attitudinized

easier because the trapping of photon and forcing it to change its path has become more likely than in the symmetric etalon samples.

The emission spectrum of the thin symmetric etalon sample is higher than that of the thicker symmetric etalon samples (60009, 44038, and 35678 a.u.), respectively.

In addition, as shown in Figure 6(a), we can compare the laser emission of thin samples in the higher pumping environment with normal samples to confirm the higher laser emission intensity and fine lasing modes in asymmetric structures in comparison with symmetric ones. Likewise, modes overlapping in symmetric etalons separate into two main regions exactly in the same position of the normal etalon sample's lasing wavelength according to Fig. 6(b).

Furthermore, by asymmetric etalons, we can adjust lasing wavelengths by mode controlling in the thickness from 200 to 600 micrometers and decrease in the unit cell of up and down faces of etalon into two main green and red useful wavelengths.

Furthermore, the FWHM of the thinner cell symmetric etalon sample affected by the backscattering phenomenon is less than the FWHM of the thicker symmetric etalon samples. The laser threshold of the thinner etalon sample indicated by the black line is shifted to a lower pumping energy, set as 16 μJ . In comparison, the thicker etalon sample green line has a laser threshold of approximately 26 μJ (Fig. 6 (c)).

Moreover, the figure shows a clear improvement in the value of the slope efficiency of the thinner etalon sample thickness compared to the thicker etalon sample thickness. All these results were obtained at an angle of 45° degrees. By changing this angle and the emission spectrum obtained directly from another side, the plasmonic multiwavelength caused strong confinement of the electric field and a high Purcell factor was observed to increase the plasmonic multiwavelength emission spectrum of the gain etalon samples.

IV. Conclusion:

In sum, etalon plasmonic samples were proposed to achieve a very narrow emission peak emerging at 541 nm, 532 and 632 nm by normal, asymmetric and symmetric structures, respectively. Notably, the simulation results showed an enhancement in light transmission by comparing the upside and downside of the plasmonic etalon structure. The experimental results reported comparing normal and thinner etalon plasmonic samples and observed fine and more efficient random lasing properties with mode overlapping in thin samples. Moreover, the lasing threshold, 26 μJ , and FWHM, 1 nm, of normal etalon changed to 16, 22 and 26 μJ in 200, 400 and 600 μm thin etalon samples, respectively. The peak intensity of the thin symmetric etalon sample is higher than that of the thicker symmetric etalon samples (60000, 44000, and 36000 a.u.), respectively, and the peak intensities for the asymmetric etalon samples are higher than those for the symmetric etalon samples. A twofold enhancement in the laser intensity and fine lasing in asymmetric thinner structures can open new insight into the new generation of random laser structures.

Declarations

Disclosures

The authors declare no conflicts of interest.

Data availability statement:

All data generated or analysed during this study are included in this published article.

Authors contributions:

S. Haddawi did the lasing measurement and writing laser parts, N. Roostaei prepare the main samples and S. M. Hamidi supervise the work and writing process.

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Figures

(a)

(b)

(c)

(d)

(e)

Figure 1

(a) Schematic diagram of the normal plasmonic etalon nanostructure fabrication process, (b) a real image of the etalon plasmonic nanostructure coating by (F8PT) light emitting polymer, (c) and (d) scanning electron microscopy of Up and Down building block of the etalon sample. (e) Schematic diagram of the experimental setup used for a random laser under green light pumping.

Figure 2

Electric field distribution of the first three modes and transmission at the main resonance wavelength (for the (a) upside and (b) down sides of the normal etalon structure coating by F8PT. (c) Absorption spectrum of Au NPs (black line), light-emitting polymer (F8PT) without a gold layer (green line) and with Au NPs (blue line), and PL of a light-emitting polymer (F8PT) (red line).

Figure 3

Reflection spectra of (a) symmetric, (b) asymmetric plasmonic etalons with thicknesses of 200, 400 and 600 m and (c) norma etalon under p-polarization at an incidence angle of 58 degrees.

Figure 4

Lasing action and emission spectra of a normal etalon sample at (a) different pump energies, (b) 46 μJ and (c) an emission intensity as a function of pumping energy.

Figure 5

Lasing action of the symmetric and asymmetric etalon structures by 200, 400 and 600 micrometers.

Figure 6

(a) Lasing action of the symmetric and asymmetric etalon structures at 48 μJ , (b) Lasing action of the symmetric ones in the comparison with normal ones at 48 μJ and (c) the emission intensity as a function of pumping energy in symmetric structures.