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Enhancement of amplified spontaneous emission in organic gain media by the metallic film

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ABSTRACT

Metallic films were widely used in many micro-cavities or as electrodes. However, the quenching of fluorescent molecules and the large absorption loss of metallic films are generally considered fatal for the lasing. We report the enhancement of amplified spontaneous emission (ASE) of organic gain media in planar waveguide structure with metallic film. Compared to the metal-free device, the ASE threshold of device with metallic film is reduced by 3.7 times by introducing the spacer layer between metallic film and organic gain media. It is found that the radiative decay rate, quantum yield of fluorescent molecules and the net gain of media are enhanced by half-cavity effect of Ag film, which lead to the enhanced ASE and lower lasing threshold.

1. Introduction

Organic semiconductors are attractive gain media for compact and versatile laser system because these gain media assured wide tenability of wavelength, ease of processing, flexibility, and high efficiency with potentially low cost [1–7]. In the past decades, although all kinds of optically pumped lasers based on organic solid materials were demonstrated, it had not been possible to obtain the stimulated emission under electrical pumping [8–10]. The metal electrodes are usually integrated into the electrical pumping. One of the major challenges for the electrically pumped organic solid lasers is the negative effect of metallic electrode to stimulate emission of organic dyes. The metallic electrodes generally lead to the quenching of the dyes and the large absorption loss, which will be fatal for the lasing [11–13].

In order to evaluate the effect of metallic electrodes on lasing of dyes, the optical pumped organic lasers were usually investigated in presence of metallic electrode. For the planar waveguide structure with metallic film, substantial efforts have been made to reduce the negative effect of metallic film and to restore the amplified spontaneous emission (ASE), such as distributed feedback metallic structure [14,15], low loss metal cladding [13,16–18], and thin spacer of polystyrene or oxidized Ca [18]. In these pervious published works, however, the lasing thresholds for optical pumped devices with metallic films are still higher than the one without metallic film. Therefore, the lasing of organic gain media in presence of metallic films with the lower pumped threshold is desired.

In this letter, we demonstrated an enhanced ASE based on the planar waveguide structure with metallic film. By introduction of a spacer layer between organic gain media and metallic film, the ASE threshold of the device is reduced by 3.7 times compared with that of the device without metallic film. In this works, the presence of the metallic film can provide a half-cavity structure that modified the photonic mode density (PMD). As the results, radiative decay rate and quantum yield (QY) are changed and enhanced, which lead to the improved ASE and lower lasing threshold. To the best of our knowledge, it is the first time to show the ASE of the device with metallic film that

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exhibits the lower pumped threshold than that of the metal-free device.

2. Experiment

In our experiment, the gain media were the blend of Polystyrene (PS), tris(8-hydroxyquinolinato)aluminum (Alq_3) and 4-(dicyanomethylene)-2-tert-butyl-6(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran (DCJTB), in which PS is the host inert material, Alq_3: DCJTB is the gain media which is a typical donor–acceptor reported in previous published [19,20], Alq_3 is used as donor and DCJTB is used as acceptor. There exists a large overlap between the photoluminescence (PL) spectrum of Alq_3 and the absorption spectrum of DCJTB which guarantees the perfect energy transfer between Alq_3 and DCJTB.

We then prepared the device with the configuration: Glass/Ag film/SiO_2/PS:Alq_3:DCJTB, which is shown in Fig. 1. On the glass substrate, the Ag film was thermally evaporated under a vacuum of 1×10^{-5} Pa at the rate of 0.3 nm/s, then SiO_2 layer was deposited onto the Ag film by vacuum sputtering. And then, the chloroform solution with the blends (PS:Alq_3:DCJTB = 200: 100: 3.5, wt%) was spin-coated on the SiO_2 layer with the speed of 3000 rpm under ambient conditions, the spin-coated films were annealed at 110 °C for 10 min.

In the experiment, as for the configuration of Glass/Ag film/SiO_2/PS:Alq_3:DCJTB, the thickness of the blended layer of PS:Alq_3:DCJTB was 300 nm, the thickness of Ag film was 100 nm, and the different thicknesses of SiO_2 layer were employed to show the different ASE behaviors. The thickness and refractive index of the films were measured with Ellipsometer (SE MF-1000, Korea). The PL lifetimes were measured by steak camera. PL quantum yields (PLQYs) were measured by an absolute photoluminescence quantum yield measurement system (Hamamatsu C11347). The devices were pumped by a Nd:YAG laser (355 nm/5.55 ns/10 Hz) (Surelite I, Continuum Corp., USA). Through a pinhole filter, a slit and a cylindrical lens, the laser beam was formed as a stripe with the size of 7 mm × 1 mm, and was perpendicular to the surface of the devices. Edge emission spectra were measured by Fiber Optic Spectrometer (Ocean Optics SpectraSuite, USB2000). The ASE threshold, peak intensity and Full Width at Half Maximum (FWHM) were measured. All measurements were carried out under ambient environment.

3. Results and discussion

For comparison, the device without Ag film, the structure of Glass/PS:Alq_3:DCJTB (300 nm), was prepared as the reference. Fig. 2 shows the edge-emission spectra and output intensity of reference device as a function of the pump energy intensity. As reported and in the previous reports [1,2], the neat gain media film exhibits an obvious ASE behavior. When it is pumped by laser pulses with low energy, it exhibits a broad spontaneous emission spectrum and the FWHM is 80 nm. Once the excitation energy becomes large enough, the emission spectrum collapses to a much narrower emission with FWHM of 10 nm. As shown in Fig. 2, the ASE threshold of 30.6 μJ/cm^2 is then determined.

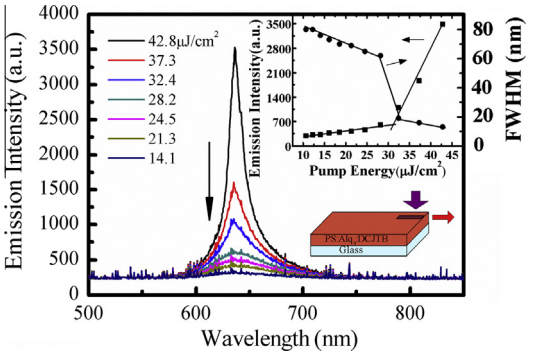

Fig. 2. Emission spectra of the device Glass/PS:Alq_3:DCJTB. The inset shows the dependence of the ASE intensity and the FWHM of the emission spectrum on the pump energy intensity. The ASE threshold is 30.6 μJ/cm^2.

To study the effect of Ag film on ASE, we prepared devices with the metallic film shown in Fig. 1, in which different thicknesses of SiO_2 layer were used as spacer layers, and deposited between the gain media film and the Ag film. Fig. 3(a)–(f) show the edge-emissions of the devices as Fig. 1, the thicknesses of SiO_2 layer are 0 nm, 20 nm, 50 nm, 80 nm, 110 nm and 150 nm, respectively. As the reference device, all the devices with Ag film present the characteristics of ASE. At low excitation energy, the devices exhibit the broad spontaneous emission spectra with FWHMs over 80 nm, and when the excitation energy reaches ASE threshold, the spectra suddenly become narrow, the FWHMs reduce to about 8 nm. In Fig. 3(g), we can find that the ASE thresholds change with different thicknesses of SiO_2 layer. It shows that with the increasing of the SiO_2 thickness, the ASE threshold reduces at first and then increases. The lowest ASE threshold is found when the SiO_2 thickness of device is 80 nm. For the corresponding device of Glass/Ag film (100 nm)/SiO_2 layer (80 nm)/PS:Alq_3:DCJTB (300 nm) (Device A), the ASE threshold decreases to 8.2 μJ/cm^2, which is reduced by 3.7 times

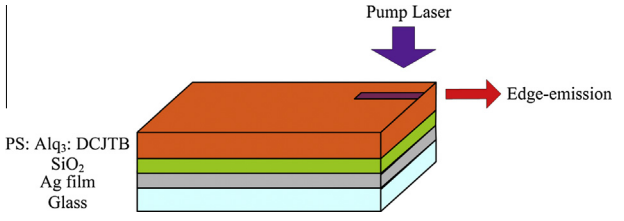

Fig. 1. Schematic illustration of device Glass/Ag film/SiO_2 layer/PS:Alq_3:DCJTB.