Rapid Communication

Amplified spontaneous emission from metal-backed poly[2-methoxy-5-(2'-ethylhexyloxy)-1, 4-phenylenevinylene] film*

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We investigate the amplified spontaneous emission (ASE) from an Ag-backed poly[2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylenevinylene] (MEH-PPV) film with different film thicknesses. The ASE characteristics of Ag-backed MEH-PPV films with different thicknesses show that increasing the film thickness can reduce the influence of the Ag cladding. The threshold, the gain, and the loss of the device with a thickness of 170 nm are comparable to those of a metal-free device. The lasing threshold of this device is about 7.5 times that of a metal-free device. Our findings demonstrate that Ag-backed MEH-PPV film with an appropriate thickness can still be a good polymer gain material for the fabrication of solid-state lasers.

Keywords: polymer semiconductor, amplified spontaneous emission, metal-backed film

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1. Introduction

Conjugated polymers are attractive gain media for novel optoelectronic device due to their broad spectral emission ranges, good luminescence qualities, and easy processing from solution, such as in the field of the polymer semiconductor laser (PSL).[1−5] These characteristics suggest that it should be possible to construct compact electrically pumped lasers by using polymers materials.[1−4] At present, optically pumped amplified spontaneous emission (ASE) has been achieved from various polymer materials.[6−9] However, electrically pumped polymer solid-state lasers have been so difficult to achieve.[6,8] Though de Villers and Schwartz[10] and Andrew *et al.*[11] reported that the Ag electrode has less effect on emission quenching than other metal electrodes, previous attempts of an electrode layer have shown a substantial rise of the lasing threshold compared with that in free-metal lasing mode.[12,13] Therefore, most PSLs are used to be fabricated with non-conducting materials, hence excluding electrically pumped operation.[14] In order to achieve an electrically pumped PSL, it is important to demonstrate that an optically pumped laser can operate in the presence of a metal.[14] In addition, it is essential to optimize the film thickness, which will influence the performance of the metal-backed PSL.

In this work, we fabricate poly[2-methoxy-5-(2'-ethylhexyloxy)-1, 4-phenylenevinylene] (MEH-PPV) directly on metallic Ag substrates, demonstrating that ASE is possible in the presence of a metal electrode in slab geometry. MEH-PPV is a well-studied conjugated polymer that has good optical and electronic properties.[7,11,15−17] We present a detailed study of the ASE and gain characteristics of the glass/Ag/MEH-PPV/air asymmetric slab optical waveguide. Furthermore, we obtain a lower threshold and higher gain by optimizing the film thickness.

2. Experiment

The spin-coating method was used to prepare the polymer films. The film thickness depends on

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the spin-coating speed. MEH-PPV was dissolved in toluene (the concentration of the MEH-PPV solution was 7 mg/mL). A 100-nm thick Ag was fabricated by thermal evaporation in a high-vacuum system with a base pressure of less than 3×10^{-4} Pa without breaking the vacuum chamber. Then, the solution was spin coated onto an Ag-covered glass substrate. Thus, a glass/Ag/MEH-PPV/air configuration formed an asymmetric slab optical waveguide. After spin coating, the spin-coated thin films were dried in a vacuum at 70 °C for 1 h. The thickness of the MEH-PPV layer was measured by Ambios Technology XP-2 Surface Profilometer. Three samples with different thicknesses were obtained: 130, 150, and 170 nm MEH-PPV films spin-coated on Ag-backed glass substrates, separately. In comparison, MEH-PPV film with a thickness of 170 nm was fabricated by spin coating on an Ag-free glass substrate.

The sample was optically pumped at 500 nm by an optical parametric oscillator (OPO), and pumped by a Q-switched neodymium ion-doped yttrium aluminum garnate (Nd^{3+}:YAG) laser. The OPO delivered 5-ns pulses at a repetition rate of 10 Hz. The pulse energy incident on the sample was adjusted by inserting a neutral density filter into the beam path. A narrow excitation stripe of 0.5 mm×3 mm was focused on the sample with an adjustable slit and a cylindrical lens. The emitted light was collected from the edge of the MEH-PPV film and focused into an ACTTON 150 Spectrograph through a cut-off filter.[7] All measurements were carried out under ambient conditions at room temperature.

3. Results and discussion

Figure 1(a) illustrates the normalized emission spectra of the Ag-backed MEH-PPV film (170 nm) at various pump energy intensities. When the pump intensity exceeds 0.48 μJ per pulse, a spectral narrowing occurs at a wavelength of 622 nm. The dependences of the output light intensity and the full width at half maximum (FWHM) of the emission spectrum on the pump energy intensity are shown in Fig. 1(b). With the increase of the pump intensity, the output light intensity increases while the FWHM decreases and shows an abrupt drop at the threshold of 0.5 μJ per pulse. The FWHM of the emission spectrum reduces to about 8.9 nm at 2 μJ per pulse. It can be seen that amplified spontaneous emission can be obtained from Ag-backed polymer films.

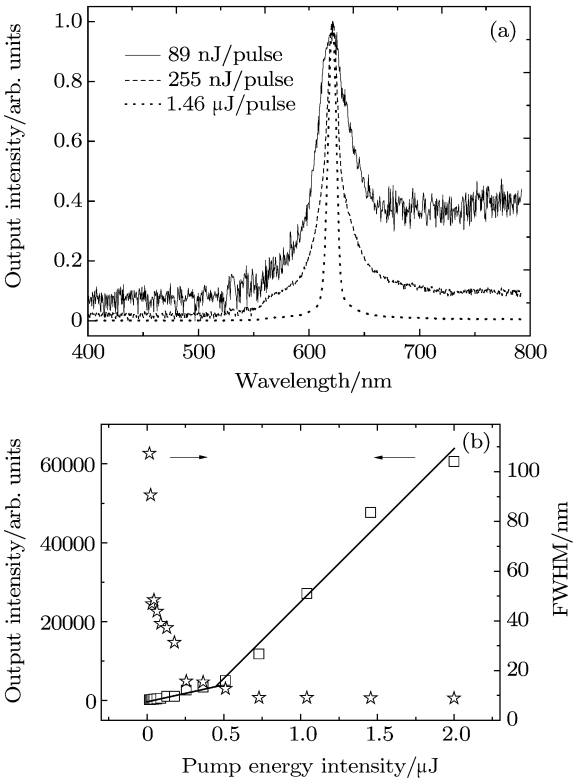


Fig. 1. (a) Normalized emission spectra of the Ag-backed MEH-PPV film at various pump energy intensities, (b) dependences of the output light intensity (open squares) and the FWHM (open stars) of the emission spectrum on pump energy intensity.

The output emission intensities of four samples, each as a function of the pump intensity, are shown in Fig. 2. It can be seen that the metal Ag layer affects the emission spectrum of the MEH-PPV film strongly. When the film thickness increases further, to 170 nm, a stronger ASE peak is observed, which is comparable to that of Ag-free MEH-PPV film. Obviously, the emission spectrum of the Ag-backed MEH-PPV film depends on the film thickness. An appropriate film thickness can reduce the influence of the Ag layer. To investigate the influence of a metal Ag layer on ASE from MEH-PPV films with different thicknesses, we compare the thresholds, the gains, and the losses of Ag-backed MEH-PPV films of different thicknesses with those of the Ag-free MEH-PPV film. The emission intensities increase slowly with the increase of the pump intensity for the thinner Ag-backed MEH-PPV films (130 and 150 nm). However, the thicker film (170 nm) shows an abrupt change in slope, followed by a linear increase in the output signal as the excitation energy is further increased. Therefore, the thresholds of pump energy for ASE can be easily discerned from each other at 1.54 and 1.06 μJ per pulse, which correspond to Ag-backed MEH-PPV films with thicknesses of 130 and 150 nm, respectively. It is found

that when the Ag-backed film is thicker, the threshold (0.48 μJ per pulse) is more comparable to that of the Ag-free MEH-PPV film (0.06 μJ per pulse). The lasing threshold of this device is about 7.5 times that of a metal-free device.

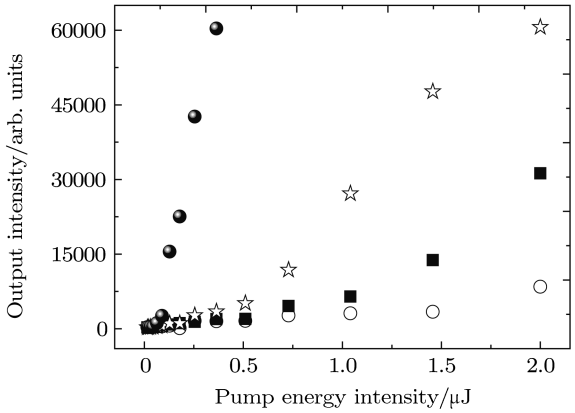


Fig. 2. Output emission intensities integrated over all wavelengths, each as a function of pump intensity, for Ag-free MEH-PPV film (solid circles) and Ag-backed MEH-PPV films in thicknesses of 130 (open circles), 150 (solid squares), and 170 nm (open stars).

The net gain was measured by using the usually variable-stripe-length method, which was employed to characterize the gains of Ag-backed MEH-PPV films with thicknesses of 130, 150, and 170 nm and the Ag-free MEH-PPV film. This method involves the detection of ASE emission from the film edge as a function of excitation length. For the case of ASE, the output emission $I(\lambda)$ should obey the following equation:[1,2]

$$I(\lambda) = \frac{A(\lambda) I_{\mathrm{p}}}{g(\lambda)} \left(\mathrm{e}^{g(\lambda)L} - 1 \right), \tag{1}$$

where $A(\lambda)$ is a constant related to the cross section for spontaneous emission, I_{p} is the pump intensity, g is the net gain coefficient, and L is the length of the pumped stripe. Figure 3 shows the ASE intensities of three samples, each as a function of excitation length at 1.5 μJ per pulse pump influence. The solid curves in Fig. 3 are the fitting of the experimental data to Eq. (1), giving net gains of 1.65, 2.51, and 12.82 cm^{-1}, at a pump energy of 1.5 μJ per pulse for the Ag-backed MEH-PPV films with thicknesses of 130, 150, and 170 nm, and 30 cm^{-1} for the Ag-free MEH-PPV film, respectively. It can be seen that the gain of Ag-backed MEH-PPV film with the thicknesses of 170 nm is close to that of Ag-free MEH-PPV film. This indicates that an Ag-backed MEH-PPV film with an appropriate thickness is still a good polymer gain medium material for the fabrication of solid-state lasers.

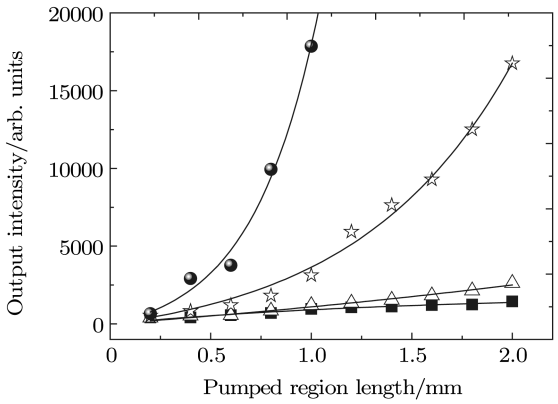


Fig. 3. Dependences of emission intensity at the peak wavelength on the excitation length at indicated pump intensities for Ag-free MEH-PPV film (solid circles) and for Ag-backed MEH-PPV films in thicknesses of 130 (solid squares), 150 (open triangles) and 170 nm (open stars).

The loss coefficient of the waveguide was measured by using a method in which the pumped length was kept constant ($L = 3$ mm) and the pumped region was moved away from the edge of the device. Since the emission from the end of the pump stripe remains constant, the detected signal from the edge of the sample should decrease as[1,14]

$$I = I_0 \, \mathrm{e}^{-ax}, \tag{2}$$

where a is the waveguide loss coefficient and x is the length of the unpumped region from the end of the pump region to the edge of the sample. Thus, the loss coefficient can be estimated. Figure 4 shows the emission intensities, each as a function of distance from the film edge for the Ag-backed MEH-PPV films with thicknesses of 130, 150, and 170 nm

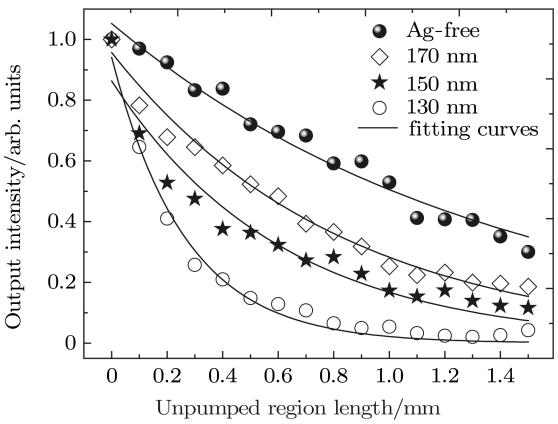


Fig. 4. Intensities of light emitted from the edge of a waveguide, each as a function of the distance between the pump stripe and the edge of the MEH-PPV film.

and for the Ag-free MEH-PPV film, respectively. The solid curve is the fitting to Eq. (2), giving loss coefficients of 37.67, 16.39, and 12.19 cm^{-1} for the Ag-backed MEH-PPV films with thicknesses of 130, 150, and 170 nm, and 7.34 cm^{-1} for the Ag-free MEH-PPV film under the same experiment conditions, respectively. The larger loss of the Ag-backed MEH-PPV film is mainly from the optical loss arising from metallic contacts. However, increasing film thickness can reduce the influence of the metal layer.

The ASE parameters of the Ag-backed MEH-PPV films with different thicknesses and the Ag-free MEH-PPV film are summarized in Table 1. We clearly observe that introduction of a metal Ag layer indeed influences the ASE performance of the MEH-PPV film. One reason for this result is that the metal layer is prone to quench a polymer fluorescence, hence increasing the ASE thresholds.[10,13,14] Another reason is that the light propagation loss from metallic contact weakens the optical gain.[2] However, it is noted that the effect of the Ag cladding on the gain medium can be reduced by increasing the thickness of the gain medium layer. When the Ag-backed film becomes thicker, the ASE performance is more comparable to that of the Ag-free MEH-PPV film. These values are much better than ASE characteristics of Ag-backed DCJTB: PS films.[14] Thus, the Ag-backed MEH-PPV film is still a good organic gain medium material for the fabrication of solid-state lasers, when the thickness of the MEH-PPV film increases to an appropriate value (170 nm).

Table 1. Amplified spontaneous emission parameters of 170 nm Ag-free MEH-PPV film and Ag-backed MEH-PPV films with thicknesses of 130, 150, and 170 nm.

Parameters	Ag-backed films			Ag-free film 170 nm
	130 nm	150 nm	170 nm	
Peak/nm	620	621	622	622
FWHM/nm	9.76	9.30	8.95	11.1
Threshold/μJ	1.543	1.062	0.482	0.064
Gain/cm^{-1}	1.65	2.51	12.82	30
Loss/cm^{-1}	37.67	16.38	12.19	7.34

4. Conclusion

We demonstrate a detailed study of ASE characteristic of Ag-backed MEH-PPV films with different thicknesses, showing that the introduction of the metal Ag layer indeed affects the ASE performance of the MEH-PPV film. It is found that the increase of film thickness can reduce the influence of the Ag layer. Compared with the Ag-free MEH-PPV films, the film of 170 nm is better in the sense of the ASE performance. The lasing threshold is about 7.5 times that of an Ag-free device operating at the same wavelength and fabricated on the same substrate. Our studies are promising results toward the realization of future polymer injection lasers.

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