Near diffraction-limited laser emission from a polymer in a high finesse planar cavity

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We report near diffraction-limited laser emission from the conjugated polymer BEH:PPV in a cavity made with two dielectric mirrors providing a high finesse planar cavity. The laser has a sharp intensity threshold, a strong directionality, and a high degree of polarization. These emission characteristics are compared with those of a single polymer layer without optical feedback. © *1998 American Institute of Physics.* [S0003-6951(98)00603-2]

Electroluminescence from conjugated polymers[1] has opened new doors for the application of organic materials in electrically pumped light-emitting devices. Structural flexibility, easy processing, wavelength tunability, and low cost of the organic materials make polymeric light-emitting diodes very attractive. Photoluminescence line narrowing and stimulated emission in a variety of conjugated polymers have been reported recently[2–7] and make the prospects of electrically pumped polymer lasers exciting. For laser applications, the polymer as active medium has to be combined with an optical feedback structure that is suitable for electrical carrier injection. The most straightforward way to provide feedback is to design a cavity that uses two plane mirrors. Emission properties from planar polymer cavities under optical excitation have been investigated recently.[2,6] In each case, a highly reflective distributed Bragg reflector and a silver mirror formed the cavity. The observed emission is characterized by a threshold like behavior, a nonlinear increase of one mode associated with the maximum of the optical gain as well as an increasing degree of directionality with excitation intensity. This behavior has been interpreted as lasing.

Whether or not lasing has been achieved is difficult to decide, in particular, since (i) a threshold like change of the emission spectra and intensity has also been observed from polymer layers without any optical feedback,[3–7] and (ii) mode selection and enhanced directionality has also been reported for photoluminescence and electroluminescence from resonant cavity organic diode structures.[8,9] In this letter, we report a detailed study of lasing from a cavity and compare directly the emission features from polymer layers inside and outside a high finesse planar resonator, i.e., with and without optical feedback, under identical conditions. We show that it is possible to achieve laser action from the cavity made with two dielectric mirrors. To the best of our knowledge, we report for the first time unambiguous features of laser emission such as high degree of polarization (>50:1 parallel to the optical excitation) and high directionality close to the diffraction limit.

As the active absorbing and emitting material, we use the semiconducting conjugated polymer BEH:PPV [Poly(2,5-bis(2′-ethyl-hexyl-oxy)-1,4-phenylenevenylene].[10] We have previously investigated optical gain and stimulated emission in this material.[7] In BEH:PPV, strong exciton-phonon interaction forms a multilevel system well suited for laser applications. Inversion of the exciton-one phonon transition can be achieved leading to gain coefficients of up to 10^4 cm^{-1} at very high excitation (the highest γ value reported so far for a conjugated polymer). Photoluminescence line narrowing, stimulated emission, and a corresponding increase in emission efficiency has been observed for exciton densities above 10^{18} cm^{-3}.

The inset of Fig. 1 shows the laser structure under investigation. To get a high quality planar cavity, we produced dielectric mirrors by electron beam deposition that contain a stack of 21 alternating quarter-wave layers of low (SiO_2) and high (TiO_2) refractive index materials. These mirrors are characterized by low losses and extremely high reflectivities (>99%) in the gain region of the polymer, e.g., 99.7% at 626 nm. An 80% transmission at the excitation wavelength (555 nm) allows efficient optical pumping. A 200 nm film of

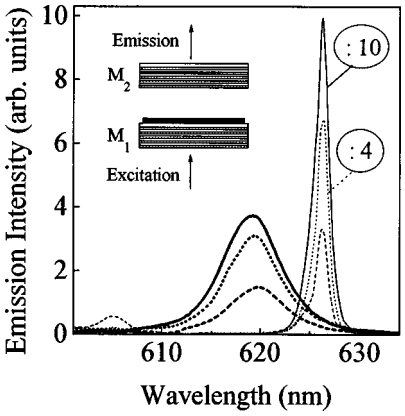

FIG. 1. Emission spectra of the 9 μm polymer cavity (M_1 and M_2, thin lines) and the polymer layer (only M_1, thick lines) under 40 (dashed lines), 63 (dotted lines), and 100 μJ/cm^2 (solid lines) excitation, respectively. Inset: Investigated structure.

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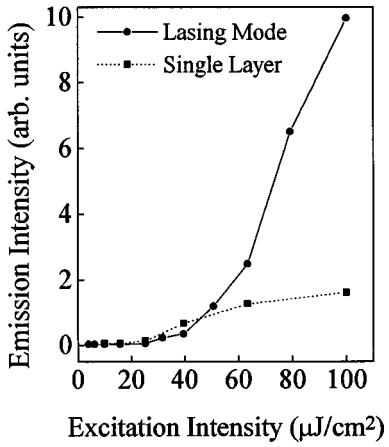

FIG. 2. Excitation intensity dependence of the lasing cavity mode emission and the spectrally integrated single layer emission.

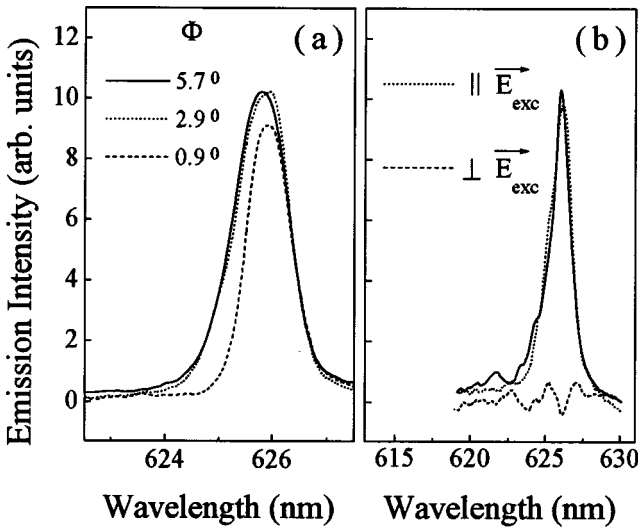

FIG. 3. Polymer cavity output above lasing threshold. (a) Directionality: Φ gives the half-apex angle of the detection cone. (b) Polarization: Detection without analyzer (solid line, normalized to parallel detection), parallel to the excitation (dotted line), and perpendicular to the excitation (dashed line).

BEH:PPV is spin-coated from solution onto one of the dielectric mirrors (M_1). We compare the emission of this single layer with that of a high quality planar resonator made simply by adding a second mirror M_2 under identical excitation and detection conditions. The optical thickness of the resonator is 9 μm determined from the cavity modes observed in transmission. We excite through the mirror M_1 with 100 fs optical pulses generated by an amplified CPM laser system. The central wavelength of the excitation pulses of 555 nm is in resonance with the $\pi-\pi^*$ absorption band yielding 80% absorption in the 200 nm polymer layer. The excitation laser was focused to a diameter of 100 μm on the polymer film.

The photoluminescence spectrum of a single BEH:PPV layer undergoes a dramatic change at a threshold excitation intensity of 25 μJ/cm^2.[7] The initially broad emission spectrum displaying several vibronic structures and extending over more than 100 nm collapses into a narrow emission band of 8 nm width centered at 620 nm as shown by the thick curves in Fig. 1. A soft threshold in emission intensity accompanies that spectral narrowing as seen by the solid squares in Fig. 2 where the spectrally integrated emission intensity is plotted as a function of excitation intensity. We attribute the threshold to an increase of the photoluminescence efficiency above an intensity for which strong stimulated emission suppresses nonradiative recombination. Consequently, the small photoluminescence efficiency ($\eta \approx 1\%$) measured with an integrating sphere at low excitation on BEH:PPV films reached almost 100% at an excitation intensity of 40 μJ/cm^2. A similar behavior has been observed for single layers of various conjugated polymers.[3–5] However, spectral narrowing of the emission and a threshold in emission intensity are insufficient to prove laser action.

In the resonator configuration, the situation changes dramatically. Two longitudinal cavity modes at 605 and 626 nm overlap with the polymer emission. For excitation below 25 μJ/cm^2, emission can only be detected around 605 nm. Even extremely small material losses do not allow the weak emission at 626 nm to leave the high Q cavity ($R=99.7\%$). On the other hand, the lower mirror reflectivities at 605 nm ($R=95\%$) allows a cavity output even in the presence of small losses. As the excitation increases above 25 μJ/cm^2, the threshold for stimulated emission is reached inside the cavity causing the radiative emission probability to increase. Correspondingly, a larger number of photons are emitted into the cavity mode at 626 nm. With the simultaneous increase of the gain (which has been shown to grow proportional with the excitation intensity),[7] this results in a gradual increase of the output at 626 nm with a soft intensity threshold comparable to the one observed in the single layer. This leads to a drastic change of the intensity ratio between the 626 and 605 nm modes similar to the mode ratio change observed in Refs. 2 and 6. However, the lasing threshold has not been reached yet and the cavity emission remains smaller than the single layer emission. The cavity losses are still larger than the optical gain in the polymer.

By further increasing the excitation intensity, a real laser threshold can be reached at 50 μJ/cm^2 as can be clearly seen in Fig. 2. Now, the emission of the cavity into the detection cone (10° half-apex angle) is clearly larger than the single layer emission. If we assume that we collect approximately 1/9 of the total emission of the single layer (90° half-apex angle) in our detection cone, we expect the directional laser emission (see next paragraph) to be nine times higher for the same efficiency. That nine times increase is approximately observed at 100 μJ/cm^2 as shown in Fig. 2, emphasizing the feedback effect of the cavity and very efficient laser action. A full width of 1.5 nm is observed for the 626 nm laser emission, to be compared with the 8 nm width of the spectrally narrow single layer emission. For this high quality cavity, the laser threshold is about twice the threshold for stimulated emission in the single layer. Above the laser threshold, the cavity output grows approximately linear with the excitation as expected for any laser action.

A striking evidence for laser action is the directionality of the laser emission. Figure 3(a) shows that the whole laser emission is concentrated in a cone of smaller 3° half-apex angle. Here, we collected the emission at 63 μJ/cm^2 excitation through apertures of 1.5, 5, and 10 mm diameters placed 50 mm from the sample, giving half-apex angles of 0.9°,

2.9°, and 5.7°, respectively. Even for 0.9° collection, the measured intensity is almost the same as for an open aperture. This value is close to the diffraction limit, which is about 0.8° for a plane wave and an excitation diameter of 100 μm. For the single layer, the collected light intensity decreases by a factor of about 6 when reducing the aperture from 10 to 1.5 mm.

As can be seen in Fig. 3(a), the intensity of the high-energy part of the emission spectrum is reduced for the smallest collection angle. This phenomenon is well known and has also been observed, e.g., in GaAs vertical cavity surface emitting layers. It is the result of the presence of higher order transverse modes. A slight deviation from the ideal plane cavity leads to partial removal of the degeneracy of the transverse modes. Higher order modes occur at shorter wavelengths. Since these modes have a larger spatial extension they get suppressed in closing the aperture. Additionally, this observation indicates that the linewidth of a single cavity mode is smaller than 1.5 nm. Figure 3(b) shows the polarization of the emission above the lasing threshold (55 μJ/cm^2 excitation). The polarization degree is better than 50:1 parallel to the exciting laser. On the contrary, neither the cavity output at 605 nm nor the stimulated emission from the single layer has a noticeable degree of polarization.

We would like to emphasize the differences of our observations to previous works claiming lasing from conjugated polymers in a planar cavity configuration.[2,6] First, neither of these reports compared the cavity output with single layer emission to show increased emission in a given direction. Second, both letters report a relative reduction of the main mode emission intensity for detection at a small angle with respect to normal. Tessler *et al.*[2] observed about 50% reduction whereas Diaz-Garcia *et al.*[6] saw a 35% reduction detecting at 7° and 3° to normal, respectively. In Ref. 6, the observed decrease in intensity is also influenced by the simultaneous shift of the emission wavelength away from the gain maximum. To the contrary, we can not detect any emission under an angle larger than 3° when we excite the cavity above lasing threshold, a clear evidence for laser action. Finally, strong polarization of the cavity output serves as another hint for coherent laser oscillation and has also not been demonstrated in Refs. 2 and 6. We believe that clear laser performance could be obtained because of our high quality cavity made with highly reflective, low loss dielectric mirrors. In this way, we were able to reduce the losses of the cavity by more than two orders of magnitude compared to the silver mirrors used in Refs. 2 and 6.

In summary, we have shown clear evidence for laser action using a semiconducting polymer as the active material in a high finesse planar resonator configuration. The laser threshold is about twice the threshold for stimulated emission in a single layer. The output is nearly diffraction-limited and has a polarization ratio larger than 50:1.

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