Femtosecond Thin-Film Laser Amplifiers Using Chirped Gratings
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Supporting Information

ABSTRACT: Ultrafast injection-locked amplification is achieved by sending femtosecond supercontinuum pulses into a polymeric thin film coated on a distributed feedback (DFB) microcavity consisting of chirped gratings. The spatial variation of the grating period led to the resonance of the DFB microcavity at different wavelengths for injection at different locations. This enables convenient and continuous tuning of the amplification spectrum by displacing the grating structures. The large area of the grating structures enabled large tuning range. The amplification spectrum can be continuously tuned from 545 to 580 nm through sliding the grating structures by about 3.5 mm. Sub-1 ps lifetime has been measured for the amplification process with a net amplification factor as large as 33. Injection locking enabled high-quality control of the divergence and transverse mode of the output laser beam.

1. INTRODUCTION
Organic semiconductors exhibit multifold advantages in their high efficiency of emission, solution processibility, high flexibility, integration of thin-film structures into high compact devices, and broad-band tunability in the emission spectrum. Optically pumped organic lasers based on distributed feedback (DFB) resonators have been extensively reported.1–5 Strong optical confinement in the DFB microcavities enabled low-threshold and high-efficiency operation of the laser device.6–10 However, low output power and large divergence of the output laser beams make the thin-film organic lasers very limited in practical applications.

Laser amplification technique is an effective approach to solve the above problems. Injection locking cannot only enable achievement of high-power lasers but also significantly improve the divergence, the transverse mode, the bandwidth, and the spectroscopic tunability of the amplification output.11–14 We have recently demonstrated injection-locked amplification (ILA) of femtosecond laser pulses using DFB microcavities.15,16

In our previous work, efficient amplification of the injected pulses was achieved in a femtosecond time scale with the divergence and far-field transverse mode determined by the injection laser beam.15 However, the tuning range of the amplifier is very limited due to the strict requirements by the DFB resonance. Using slant gratings, we have largely extended the tuning range of the injection-locked amplifier through breaking the strict confinement in the DFB microcavities based on vertical gratings.16 Slanting the grating lines has not only broadened the tuning range of the amplifier but also broadened bandwidth of the amplified spectrum due to the reduction in the confinement strength of the microcavities.

In this work, we demonstrate injection-locked organic thin-film amplifiers based on DFB microcavities constructed by chirped gratings with vertically arranged grating lines. In such a design, the optical confinement strength was not reduced, as compared with an unchirped DFB grating with a fixed period. However, the tuning range of the amplifier can be largely extended by spatial variation of the injection location on the chirped gratings.

2. DESIGN AND FABRICATION OF CHIRPED DFB GRATINGS
Although chirped gratings have been employed in thin-film laser devices17,18 based on DFB microcavities, these devices are all laser oscillators. Femtosecond laser amplifiers using chirped gratings have not been reported, so far. The amplifiers supply laser beams with much enhanced power or pulse energy, much reduced laser beam divergence, and much improved transverse mode quality. Thus, the laser beam can be conveniently controlled for further application. In particular, the output spectrum of the amplifier can also be dominantly controlled by the injection, where the matching between the resonance of the DFB microcavity, the spatially distributed grating periods, and the color of the injection laser beam are crucial for the efficient operation of the amplifier. These performances show advantages of the amplifiers over the oscillator and the purpose for developing laser amplifiers. Furthermore, the dynamics of the amplification process in the femtosecond time scale may reveal more physics for the amplification processes.

As has been mentioned above, using chirped gratings will not weaken the total confinement strength of the DFB microcavities, as compared with unchirped configurations. This can be confirmed by our theoretical simulation results on the intensity of the resonance mode of DFB microcavities based on a normal grating with a fixed period and a chirped grating with varied periods, as demonstrated in Figure S1. The chirped grating shows a resonance spectrum with its peak intensity...
reduced to about 58% with respect to that for a normal grating. However, the bandwidth is broadened from about 1.6 nm to about 3.6 nm at full width at half-maximum (FWHM), and the integration of the emission spectrum is almost the same for both configurations, implying nearly no reduction in the total efficiency for the energy confinement.

Figure 1a shows the design and fabrication method of the chirped grating structures. A cylindrical lens was inserted into the two-beam interference lithography scheme, which induces a spatial distribution of the incident angle ($\theta_1 < \theta < \theta_2$). Thus, the separation angle between the two incident UV laser beams reduces with increasing the horizontal coordinate value ($x$). The overlap between the two laser beams on the surface of the photoresist (PR) layer is shown in the lower panel of Figure 1a. We used a smaller diameter for laser beam B with a circularly shaped transverse mode and a larger one for beam A with an elliptical shape, thus chirped gratings were produced along $Y$-axis within a overlapped area, as shown in the lower panel of Figure 1a. A photograph of the finished device is also included in the lower panel of Figure 1a, which also demonstrates a chirped diffraction pattern. The scanning electron microscope (SEM) images are shown in Figure 1b, which were measured on three different sites with a separation distance of 1 mm using JSM-6510 from JEOL Ltd. The corresponding grating period ($\Lambda$) was increased from 330 to 350 nm, implying a spatial chirp of about 10 nm/mm. In the interference lithography, a UV laser at 360 nm with a total power of 30 mW was used to perform the exposure, a cylindrical lens with a focal length of 30 mm was used to converge beam A, a photoresist AR-P3170 from Allresist GmbH was used as the recording medium, and a separation angle of $\theta = 31^\circ$ was employed in the interference lithography scheme. The diameter of the effective area of the grating structures is about 10 mm. The modulation depth of the grating is about 160 nm, which is basically constant over the whole structure area.

### 3. INJECTION-LOCKED ORGANIC AMPLIFIER FOR FEMTOSECOND LASER PULSES

#### 3.1. Microcavity Laser Resonator/Amplifier Based on Chirped DFB Gratings

For the thin-film laser oscillators and amplifiers, the second-order Bragg condition supplies optical feedback for the intracavity resonance and the first-order supports the surface-emitting output of the laser resonator or the injection/output of the amplifier, as has been demonstrated in detail in refs 19–21. The chirped gratings actually supply a series of DFB microcavities arranged spatially with continuously changing grating periods. After being coated with an active layer of the laser medium, each microcavity is a DFB laser oscillator with its intrinsic resonance defined by the local grating period. This implies a series of DFB lasers with a continuously tunable emission spectrum.

However, for the design of amplifiers, the injection laser beam is coupled into the DFB microcavity through a Bragg diffraction process by $n_{eff}\Lambda(x) = \lambda$. $\Lambda(x)$ is the period of the chirped grating, which is varied by changing the position ($x$) and expressed as $\Lambda(x) = \Lambda_0 + \gamma x$ with $\Lambda_0$ and $\gamma$ denoting the smallest grating period on one end of the structures and the chirping rate, respectively. $n_{eff}$ is the effective refractive index of the active medium-coated grating system. The injection was then confined into the DFB microcavity through a second-order Bragg process by $2n_{eff}\Lambda(x) = 2\lambda$, resulting in a multipass oscillation of the injection in the microcavity and its efficient interaction with the excited molecules in the active medium. These mechanisms constitute the amplification processes, which is dominantly locked by the injection laser pulses in their divergences, transverse modes, and spectra. Apparently, the resonance wavelength ($\lambda$) can be continuously tuned by changing the period of the grating ($\Lambda$) through adjusting the injection position ($x$).

In the construction of the organic amplifier, a light-emitting polymer semiconductor poly(9,9-dioctylfluorenyl-2,7-diyl-alt-co-(1,4-benzo-[2′,3′]-thiadiazole) (F8BT, from American Dye Sources) was dissolved in chloroform with a concentration of 23 mg/mL, which was spin-coated onto the chirped grating structures shown in Figure 1. A rotation speed of 2000 rpm and a duration of 30 s were employed in the spin-coating process, so that F8BT thin film with a thickness of about 120 nm (measured by a spectroscopic ellipsometer from Ellitop) was produced on the top surface of the chirped grating, as shown in Figure 2a. F8BT is a typical light-emitting polymer extensively applied in optically pumped lasers.22–25 The absorption and photoluminescence (PL) spectra of F8BT are peaked at about 460 and 540 nm, respectively. In principle, the physics, the mechanisms, and the design of the amplifiers are not limited by the conjugated polymer materials. Provided that the polymer is an efficient emission material, and the emission spectrum covers the resonance of the DFB microcavity, thin-film amplifiers can be achieved with an excellent efficiency. This enables a large extension of the tuning range of the organic thin-film amplifiers by utilizing different polymer materials with the grating period adjusted broadly.

Before characterizing the injection-locked laser amplification performance, we first investigated the device illustrated schematically in Figure 2a as a laser oscillator. We pumped...
Figure 2. (a) Construction of the organic laser amplifier by spin coating the chirped PR grating with F8BT. (b) The optically pump lasing oscillation performance with spatial tunability based on the chirped DFB-grating microcavities.

3.2. Injection-Locked Femtosecond Amplification with Spatial Tunability. A femtosecond pump-probe system was employed in the investigation on the injection-locked amplification (ILA) process. Femtosecond laser pulses at 800 nm that were produced from a Ti:sapphire amplifier (Legend Elite from Coherent Inc.) were frequency doubled by a BBO crystal to produce 150 fs pulses at 400 nm with a repetition rate of 1 kHz. A portion of the 800 nm laser pulses was sent to a heavy-water cell with a thickness of about 3 mm to produce supercontinuum pulses extending from about 340–1200 nm, which were used as the injection pulses. It needs to be noted that the supercontinuum simply supplies a broad band light source, which can be easily used to replace a tunable light source. Meanwhile, for the pump-probe system, the supercontinuum is naturally available, making all experiments much easier. The matching between the grating period and the injection wavelength can be conveniently achieved, where a single supercontinuum beam contains all required spectral components for the injection.

The geometry for the measurements on the amplification process is presented in Figure 3a, where the injection locking scheme was introduced, as compared with the laser oscillator in Figure 2a. Injection into the microcavities at different locations on the chirped DFB gratings leads to the amplification at different wavelengths. Similar to the laser oscillator, the amplification at longer wavelengths can be achieved at a larger grating period. The relationship between all spectra involved in the amplification process was plotted in Figure 3b by the solid curves. The pump spectrum (blue) centered at about 400 nm is within a mild absorption region by F8BT (red). The injection (black) is overlapped with a PL spectrum (dark yellow) in a broad band within the visible spectrum (450–700 nm), implying an excellent matching between the material properties and the laser spectra.

In the experimental studies on ILA, the thin-film amplifier was mounted on a linear translation stage. The injection laser beam was incident along the normal of the substrate of the chirped gratings. Meanwhile, the pump beam was incident at an angle of about 30°, which was optimized to achieve the highest amplification and to avoid coupling of the pump laser into the detection head. Figure 4 shows the amplification dynamics of the injection supercontinuum pulses at different locations on the chirped DFB grating. A pump fluence of about 12 μJ/cm² was employed for all of the measurements. The injection beam was focused by a curved mirror with a focal length of about 200 mm, so that the spot size of the injection is within the area covered by the injection beam. The solid green circles in Figure 3b plot the peak wavelengths of all spectra involved in the amplification process and the ILA spectrum (solid green circles) constructed by measuring the peak wavelengths of all of the amplified spectra.
pump and the injection pulses for a relative injection location changed from \( X = 0 \) (\( \Lambda \approx 320 \text{ nm} \)) to 3.2 mm (\( \Lambda \approx 352 \text{ nm} \)), where corresponding center wavelength was tuned from 551 to 561, 564, 567, 570, and 577 nm. The delay between the pump and the injection was changed from 0 to 1 ps. The open circles show the emission spectrum without injection at each injection location. Obviously, nearly all of the emission spectra without injection exhibit lasing peaks, implying that the pump fluence was slightly above the lasing threshold, and the ILA process is the most efficient when it is overlapped with the resonance of the DFB microcavity. This also reveals the regenerative-amplification nature of such an amplification process.

Furthermore, we need to stress that polarization dependence of both the laser oscillation and amplification output has been observed. The most efficient laser output has been observed for TE pump and TE injection, where both the pump and the injection laser beams were polarized along the grating lines. This is the same as what we have presented in ref 15 for unchirped gratings. All of the presented spectroscopic data in this work have been measured using TE pump and TE injection configurations.

One of the most important advantages of the injection-locked amplifiers is the control and large improvement of the divergence and transverse mode of the output laser beam. These performances are dominantly determined by the injection laser beam. As commonly observed for the thin-film DFB laser oscillators, the output has a very large divergence and laser beam rapidly spreads into the space, so that it is...
The divergence of the injection-locked amplifier output is completely defined by that of the injection beam. The injection can be an excellently recollimated laser beam with an extremely small divergence. As shown in Figure S2, the injection beam diverges very slightly as compared with the output of the laser emission after propagating in a same distance from the device.

The amplification factor can be evaluated by calculating $\eta = I_{\text{ILA}}(\lambda)/I_{\text{in}}(\lambda)$, where $I_{\text{ILA}}$ and $I_{\text{in}}$ are the intensities of the amplified and injected spectra at a peak wavelength of $\lambda$, respectively, as shown by the yellow-filled circles in Figure 4g. The amplification factor varies roughly in a same rule as the amplification intensity. The intensity spectrum agrees with the plot by the solid green circles in Figure 3b labeled with “ILA”, which is located just beside the peak and on the beginning of the falling edge of the PL spectrum. In Figure 4g, we also present the amplification factor $\eta = I_{\text{ILA}}(\lambda)/I_{\text{OSC}}(\lambda)$, as plotted by the empty circles, where $I_{\text{OSC}}$ is the intensity of the output spectrum of lasing oscillation without injection at a same pump fluence, which is peaked at $\lambda$. This actually demonstrates how much the output of the device is enhanced through injection-locked amplification with respect to that of the laser oscillator. The smallest value of $\eta$ is 7.14 at 564 nm and the largest is 33.8 at 570 nm. Obviously, stronger lasing intensity above the threshold before the injection led to a reduction in the amplification factor. The lasing oscillation processes consume a large amount of the population on the excited states due to strong stimulated emission processes, thus reducing the amount of excited molecules that interact with the injection laser pulses for the amplification processes. The competition between the amplification and the lasing oscillation processes, the saturation of the amplification processes, and the mismatch between the injection and the required in-coupling angle are responsible for such a reduction.

Figure 5 shows the amplification dynamics at 551, 557, 562, 568, and 574 nm, when the temporal overlap between the pump and the injection pulses was adjusted in the time range of about 5 ps. Fitting the tail of the dynamic curves shows a lifetime of about 478 fs for the amplification processes at 551, 557, and 562 nm. However, the lifetime was increased to about 783 fs for 568 and 574 nm. A straightforward explanation for such a difference can be the influence by the length of the DFB microcavities. Longer wavelength corresponds to larger grating period and longer microcavity or penetration depth into the gratings, resulting in longer dwelling time inside the microcavity, thus extending the lifetime of the ILA process. For understanding the dwelling time, we need to consider again that the injection pulses were coupled into the microcavity through a first-order Bragg diffraction, which were confined in the microcavity in the form of oscillation supported by a second-order Bragg process. The oscillating pulse traveled back and forth inside the microcavity for a number of rounds before being coupled out. We define the time of the round trips of the injection pulse inside the microcavity as its dwelling time.

As shown in Figure 5, the efficient amplification process was observed only within the first 1 ps. This can be understood by considering that the injection pulse induced strong interaction between the large amount of the injection photons and the excited molecules during the amplification process, consuming a large amount of population on the excited states through stimulated emission and resulting in a rapid reduction in the amplification efficiency. Therefore, although F8BT has a PL lifetime in nanoseconds, the most efficient injection-locked amplification takes place only within the first 1 ps.

We need to stress that the amplification performance has been optimized through adjusting the matching between the DFB resonance modes and emission efficiency of the active medium. In the fabrication of the chirped gratings, we have produced a much larger variation range of the grating period than that has been used and demonstrated. The fabricated structures cover a total area with a diameter of 10 mm, as shown in Figure 1. However, we have used a small part of the grating with a diameter of only about 3 mm. This area has been chosen after the optimization of the period range of the chirped gratings to obtain the most efficient amplification spectrum, as shown by the green-filled circles in Figure 3b. We have moved the local area of the amplifier over the whole structures and fixed the position of pump-probe scheme when the highest amplification was obtained at its peak intensity. Although we have optimized the performance of the amplifiers through finding the most efficient area of the chirped grating, we can still extend the tuning range by adjusting the variation range and variation rate of the grating period. Thus, each spectral component of the PL spectrum can find its matching grating period, and the tuning range can be extended to cover the whole PL spectrum. Furthermore, since there is no limitation to the organic active materials that can be applied in such thin-film amplifiers, we can easily tune the amplification spectrum in the visible or even to the near infrared spectral range by utilizing different polymers. Additionally, making use of the large-area structures of the chirped gratings, we can still extend the tuning range by using different organic semiconductors and different areas of the chirped gratings can be fabricated on different locations. Thus, using a single device, multiple amplifiers supply potentials for a large tuning range of the amplification.

The amplification factor can be improved by optimizing the geometric parameters of the gratings, smoothness of the surface of the spin-coated F8BT layer, and the spatially chirping rate, enhancing the Q factor of the microcavities and consequently the interaction length between the injected seed pulses and the excited molecules. The polymer semiconductors have much lower damage threshold than laser crystals, we used a very mild excitation in the pump-probe measurement to ensure the validness of the measurement data. This implies that we still have much space to enhance the amplification factor by increasing the pump fluence. Balancing the
confinement and output of the microcavities should also be optimized to achieve a larger amplification.

4. CONCLUSIONS

We report the injection-locked amplification of femtosecond laser pulses using DFB microcavities consisting of chirped gratings spin-coated with a thin layer of polymeric semiconductor. A large-range tuning of the amplification spectrum from 545 to 580 nm was achieved through varying the spatial position of the injection. An amplification factor ranging from 7.14 to about 33.8 was achieved at different wavelengths. The divergence and transverse mode of the output of the amplifier were determined by the injection laser beam. This is potentially important for the practical applications of organic laser devices.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsomega.9b00232.

Calculated resonance modes of the DFB microcavities based on chirped and unchirped gratings, photograph of the laser spots of the injection beam and the output of the laser oscillator emission; AFM images of the chirped gratings measured at two different locations (PDF)

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Notes

The authors declare no competing financial interest.

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