

# Distributed feedback organic lasing in photonic crystals

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**Abstract** Considerable research efforts have been devoted to the investigation of distributed feedback (DFB) organic lasing in photonic crystals in recent decades. It is still a big challenge to realize DFB lasing in complex photonic crystals. This review discusses the recent progress on the DFB organic laser based on one-, two-, and three-dimensional photonic crystals. The photo-physics of gain materials and the fabrication of laser cavities are also introduced. At last, future development trends of the lasers are prospected.

**Keywords** photonic crystals, microcavity lasers, distributed feedback (DFB)

## 1 Introduction

Photonic crystals, proposed by Yablonovitch [1] and John [2], have shown great potential for developing different photonic devices [3–5]. Great interest is focused on realizing the photonic band gap and the photonic localization of photonic crystals [6–8]. In the photonic band gap, the propagation of electromagnetic waves inside the photonic crystals is forbidden in all directions. It provides new possibilities for us to control the behavior of electromagnetic waves. Especially, the controllability of the optical density of modes helps us realize the enhancement of emission at the photonic band edge [9–11]. Based on this characteristic, much work has so far focused on potential applications in the fields of nanoscale lasers [12–14], optical switching [15–17], optical logic gates [18–20], gap solitons [21–23], sensors [24–26], and so on.

Since the pioneering work of Painter et al. [27], a significant effort has been devoted to the development of nanoscale lasers based on photonic crystals. The nanoscale lasers can be divided into two types: photonic band-gap defect mode lasers [27–29] and photonic band edge lasers

[30–32]. The former has a resonant cavity with a defect and laser oscillations origin from the resonant modes of the cavity. The latter has a resonant cavity without defects and lasing actions are enhanced by the optical density of modes at the band edge of photonic crystals.

A low threshold is an intrinsic feature of photonic band edge lasers [33]. It can be attributed to the low loss and high gain of the laser system. For photonic band edge lasers, the loss includes from the propagation loss and the radiation loss [30]. Generally, the propagation loss is very small for the photonic band edge lasers due to the extremely slow group velocity near the photonic band edges. The radiation loss mainly exists in one- (1D) and two- (2D) dimensional photonic crystals due to the poor confinement of light in a certain dimension. In theory, the radiation loss is quite small in the three-dimensional (3D) case because the 3D photonic bandgap enables a 3D confinement of light. The gain in the photonic band edge lasers will be discussed in detail later.

Photonic band edge lasers can be divided into two categories: lasers based on the guided modes and lasers based on the waveguide modes. For lasers based on the guided modes, the guided mode is related to the photonic band gap of photonic crystals. A photonic crystal acts as the laser cavity [34,35]. For lasers based on the waveguide modes, the waveguide mode is determined by a combination of a waveguide and a “photonic crystal” with weak modulation [36–38]. The latter is often referred to as the distributed feedback (DFB) lasers [39–41]. For 1D and 2D cases, the “photonic crystal” with weak modulation is known as “gratings”. The laser cavity consists of a grating and a waveguide [42–44]. The nature of the photonic band edge lasers can be explained by the dispersion relations in the laser cavity [36,45].

Furthermore, the development of gain materials has a great influence on the development of lasers. On the one hand, it can improve the performance of lasers; on the other hand, it can hasten new lasers. The gain materials involved in DFB lasers include organic semiconductors, inorganic semiconductors, dyes, quantum dots, perovskite, carbon dots, and so on. Due to the rapid development of the gain

materials, it is impossible to review completely the most relevant advances. This review focuses on characteristics of organic semiconductors, organic dyes, and semiconductor quantum dots.

In this paper, we briefly review some advances in DFB lasers in 1D, 2D, and 3D photonic crystals. We discuss photophysics of gain materials, design and fabrication of laser cavity, principles of modeling feedback mechanisms, and progress toward applications. The trends and challenges for DFB polymer lasers have also been discussed.

## 2 Gain materials

As one of the most important components of the laser system, gain materials play a crucial role in the laser performance. Some gain materials open up the prospect of high-performance lasers suitable for real applications. Inorganic semiconductor lasers dominate the laser applications for several decades [46–48]. However, the common inorganic semiconductor lasers cannot cover the whole visible spectral region. Dye lasers are investigated almost simultaneously with the discovery of the laser, which operates using dye molecules [49]. In addition, as a gain material with a zero-dimensional density of states, quantum dots were applied to lasers successfully in the 1990s [50–52]. There are some differences between organic semiconductors, organic dyes, and semiconductor quantum dots, including the film-forming property, electrical conductivity, and the difficulty of manufacturing. Recently, some new materials have been developed and introduced to lasers, such as perovskites [53–55], carbon nanodots [56,57]. We will not address these breathtaking advances in this paper.

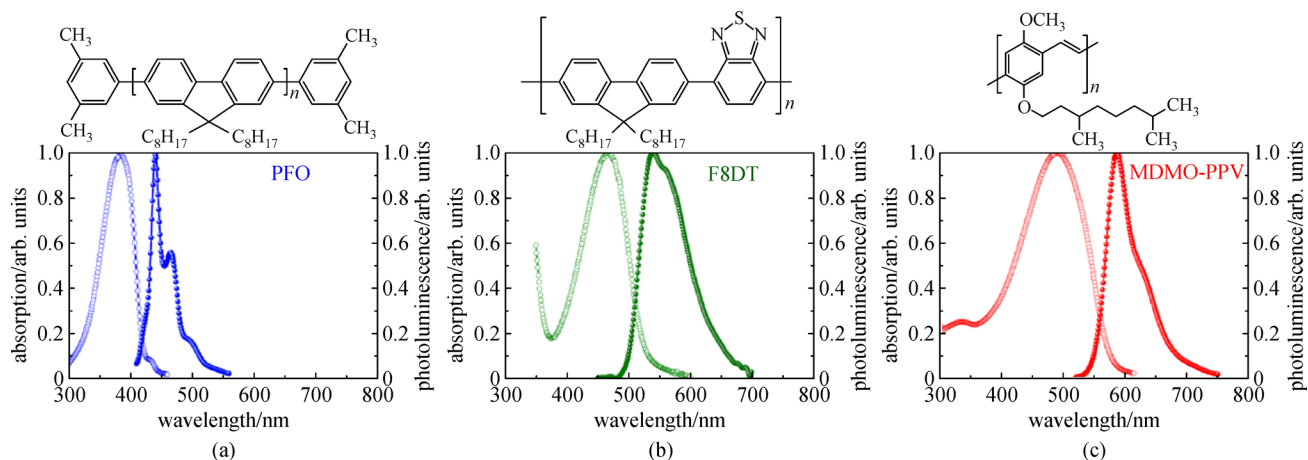
### 2.1 Organic semiconductors

Organic semiconductors are usually chain-like molecules,

which can be regarded as arrays of randomly oriented chromophores consisted of conjugated segments. The segment comprises a huge number of fundamental repeat units. The photophysical property of the material origins from the overlap of the molecular orbitals, which can be revealed by the time-resolved measurements. According to the molecular structure, organic semiconductors can be classified into small molecules (molecular weight  $< 10^3$ ) [58–60], macromolecules [61–63], and polymers (molecular weight  $< 10^4$ ) [64–66]. Small molecules include conjugated and non-conjugated molecules, organic metal complexes, and so on. Macromolecules include oligomers, starburst molecules, and dendrimers. Polymers include the poly(phenylenevinylene)s [67–69], the ladder-type poly(para-phenylene) [70–72], the polyfluorenes [73–75], and so on.

As an attractive gain material, organic semiconductors show rich and broad emission spectra from the near ultraviolet to infrared, large Stokes shift, strong absorption coefficients ( $\sim 10^5 \text{ cm}^{-1}$ ), low quenching rate at high concentrations, high fluorescence quantum efficiencies, and perfect charge transport properties. Rich and broad emission spectra enable the possibility of multi-wavelength emissions and tunable lasers. Large Stokes shift avoids the absorption of emission lights. Strong absorption coefficients imply a strong amplification of emission lights. Low quenching rate at high concentrations facilitates the easy fabrication of neat solid films. High fluorescence quantum efficiencies bring about excellent performances including low thresholds and high slope efficiencies. Perfect charge transport properties provide the potential to realize electrically pumped laser devices. Most of the features are derived from the enormous range of customizable structures. Moreover, the simple fabrication and flexibility of organic semiconductors provide more opportunities for electronics and optoelectronics.

The stimulated emission is firstly observed in a conjugated polymer film of poly(p-phenylene vinylene,



**Fig. 1** Absorption and PL spectra of (a) PFO, (b) F8BT, and (c) MDMO-PPV. The upper panel presents the corresponding molecular structure. Reproduced with permission [80]. Copyright 2015, RSC Publishing

PPV) [76]. Nowadays, there are plenty of polymers that are widely used in lasing applications. In this paper, we mainly focus on three types of polymers, poly[9,9-dioctylfluorenyl-2,7-diyl]-end capped with DMP (PFO, American Dye Source), poly[(9,9-dioctylfluorenyl-2,7-diyl)-alt-co-(1,4-benzo-(2,1',3')-thiadiazole)] (F8BT, American Dye Source), and poly[2-methoxy-5-(3',7'-dimethyloctyloxy)-1,4-phenylenevinylene] (MDMO-PPV, Sigma-Aldrich). The absorption (open circles) and photoluminescence (PL, close circles) spectra of PFO, F8BT, and MDMO-PPV are plotted in Fig. 1, respectively. Note that the Stokes shift (the deviation between the absorption and PL spectra) is large enough to avoid the absorption of emission lights. The upper panels of Fig. 1 show the molecular structures. The net gain coefficient of PFO, F8BT, MDMO-PPV are about 74, 26, 50  $\text{cm}^{-1}$ , respectively [77–79].

## 2.2 Organic dyes

Almost with the invention of the laser, the organic dyes came to people's attention. The dye laser action was firstly reported in 1966 [81]. After years of rapid development, the organic dye laser becomes a powerful tool for the development in the areas of physics, chemistry, and materials. Dyes are a class of colored materials which can impart color to other materials. Later organic compounds are included in the dyes, such as rhodamine 6G. Generally, the molecular weight of dyes is about several hundreds. Spectral narrowing effect can be observed in hundreds of organic dyes under pumping conditions. The emission wavelength varies from 190 to 1850 nm. Dyes include cyanine dyes [82–84], oxazine dyes [85–87], coumarin dyes [88–90], rhodamine dyes [91–93], and so on.

The advantages of dyes are strong absorptions, near unity quantum efficiency, broad spectra, excellent tunability, and easy fabrications. However, the dye is non-conductive, which is regarded as the main obstacle for realizing electrically pumping laser devices. Most optical behaviors of dyes can be understood by a quasi-four-level model [49]. For laser applications, the state of dyes can be solid, liquid, and gas. Figure 2 presents the absorption

(dotted curves) and PL (solid curves) spectra of three common laser dyes, coumarin 440 (C440), coumarin 153 (C153), and rhodamine 6G (R6G).

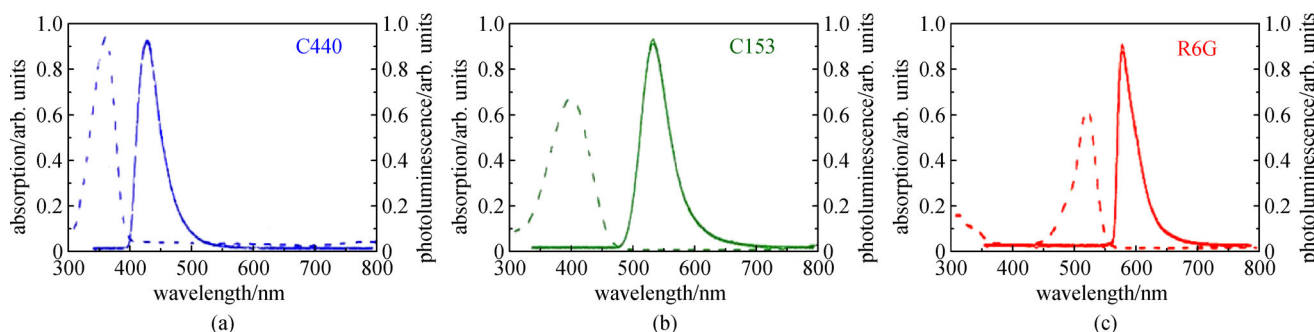
## 2.3 Semiconductor quantum dots

Quantum dots (QDs) are tiny clusters of semiconductors with dimensions of only several nanometers. The great potential of semiconductor QDs as gain materials for laser applications has been recognized since the appearance of QDs laser [94–96]. Nowadays, semiconductor QDs lasers are regarded as highly efficient and compact light sources. The direct electrical control of QDs lasers has also been realized. Two classes of QDs are very promising for laser devices [52]. One is III-V QDs, such as InGaAs/InAs QDs. The other is semiconductor nanoparticles, such as PbS and CdTe. Usually, the former is fabricated on a semiconductor substrate. The latter is incorporated with transparent dielectric matrices.

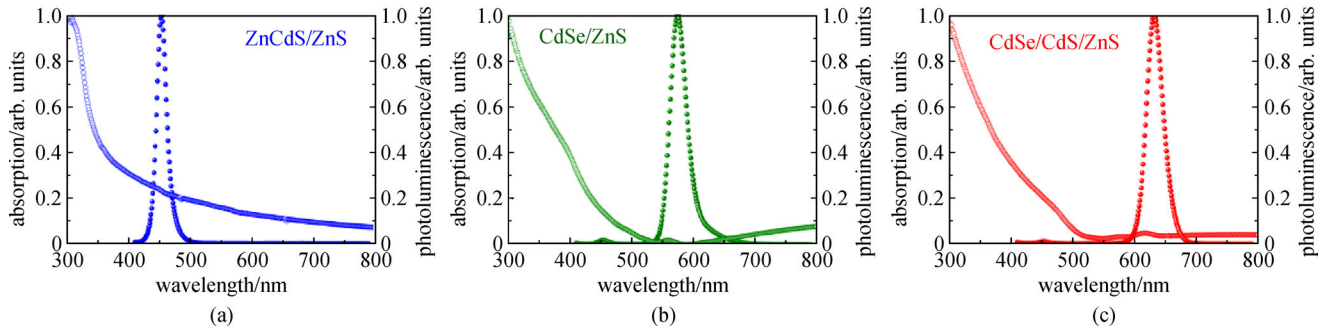
The advantages of semiconductor QDs are ultrafast carrier dynamics, low threshold current density, broadband gain and absorption, and high PL quantum yield. Such device designs have opened up new possibilities in ultrafast science and technology. The semiconductor QDs is sensitive to the temperature due to the high mobility, and the fabrication method of QDs devices is complicated compared with that of its counterparts mentioned above. Figure 3 demonstrates the absorption (open circles) and PL (solid circles) spectra of three common QDs, ZnCdS/ZnS CQDs, CdSe/ZnS CQDs, and CdSe/CdS/ZnS CQDs.

## 3 Laser cavities

The principal parts of a laser are the pump, the gain material, and the cavity. The pump supplies energy for the laser to operate, which includes optical pumping and electrical pumping. Prospects for the pump will be discussed later. The gain materials mentioned above amplify the light by simulated emission, which affects the temporal characteristics and the power characteristics of the laser. The cavity provides feedback of the light,



**Fig. 2** Absorption and PL spectra of (a) coumarin 440, (b) coumarin 153, and (c) rhodamine 6G. Reproduced with permission [90]. Copyright 2014, OSA Publishing



**Fig. 3** Absorption and PL spectra of (a) blue QDs, (b) green QDs, and (c) red QDs

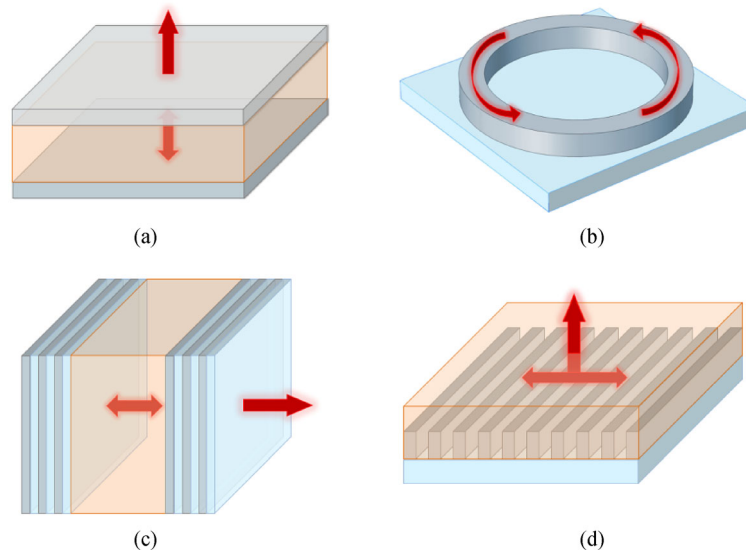
which effectively increases the optical path of the light through the gain materials to build up the laser oscillation. The cavity defines the frequency characteristics, the spatial characteristics, and the power characteristics of the laser. The frequency characteristics include the longitudinal, or axial, modes of the cavity, and the linewidth. The spatial characteristics include the pattern, polarization, and beam divergence of the laser. The power characteristics include the laser threshold and output efficiency. Generally speaking, the main parameters of the cavity contain the type, the material, the quality, and the size.

The most common cavity types can be divided into four categories: Fabry-Perot (FP) cavity [97–99], whispering-gallery-mode (WGM) cavity [100–102], distributed-Bragg-reflector (DBR) cavity [103–105], and DFB cavity [106,107], as shown in Fig. 4. There are periodic structures in the DBR cavity and DFB cavity. So, the lasing action in the DBR cavity and DFB cavity can be explained by the theory of photonic crystals. This review will focus on the DFB cavity. Recently, the compound cavity has begun to receive research attention, which can be regarded as a combination of several common cavity types [108–110].

The cavity supports a discrete set of wavelengths, which are also called the resonant wavelengths (frequencies). The relationship between the optical path of the cavity ( $P$ ) and the resonant wavelength ( $\lambda$ ) is described as  $P = k\lambda/2$ , where  $k$  is an integer. The discreteness of resonant wavelength origins from the boundary condition of the light in the cavity. The phase of light must be exactly the same after a round-trip propagation in the cavity. This is the major reason that most of the characteristics depend on the cavity. Moreover, the allowed resonant frequencies of the laser must be within the PL spectrum of the gain material. More strictly, to build up a stable oscillation of the laser mode, the gain should not be smaller than the loss in one round-trip of the cavity. Thus, in order to achieve lasing, the cavity must be designed carefully. The mode of the cavity should match the gain spectra of the material.

### 3.1 Types of laser cavities

As mentioned above, various cavity configurations are proposed to design the laser devices. Among them, the DFB cavity is regarded as the most promising solution for



**Fig. 4** Schematics of various cavity types. (a) FP cavity; (b) WGM cavity; (c) DBR cavity; (d) DFB cavity

realizing electrically pumped polymer lasers. Therefore, in the rest of this section, we will focus on the DFB cavity and summarize progress in design, fabrication, and feedback mechanism of the DFB cavity type.

According to the spatial structure, DFB cavities can be divided into 1D [111], 2D, and 3D structures. According to the transnational symmetry, DFB cavities can be divided into periodic, quasi-periodic, and aperiodic structures. Moreover, the DFB cavities can be divided into dielectric and metallic structures. Overall, the basic motivation for developing different laser cavities is to achieve a rich variety of temporal, spatial, spectral, and power properties. Figure 5 demonstrates the photonic crystals which can be employed as the DFB cavity. Theoretically, the random structure in Fig. 5(i) is not a DFB structure, which is usually used as a feedback cavity of random lasers [93]. Many irregular closed-loop paths can be excited in the cavity, which may support certain oscillation modes. Since the feedback mechanism of random lasers is quite different from that of DFB lasers [112,113], we will not address the random structure in detail.

1D–3D structures include gratings/complex lattices [114,115], quasi-crystals [116–118], chirped grating/gradual periodic structures [119], circular structure [120,121], spiral structure [122]. Note that 3D structures can be

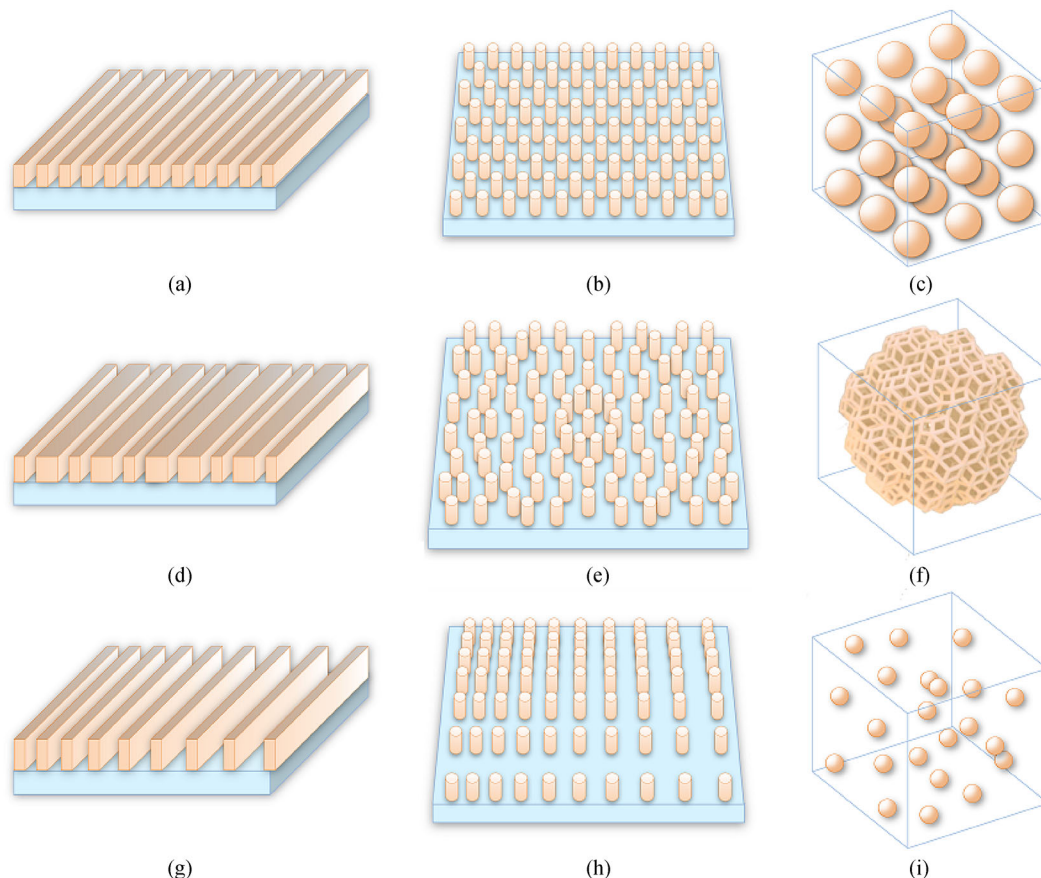
composed of several 1D/2D structures. All these structures can be employed as DFB cavities.

### 3.2 Design of laser cavities

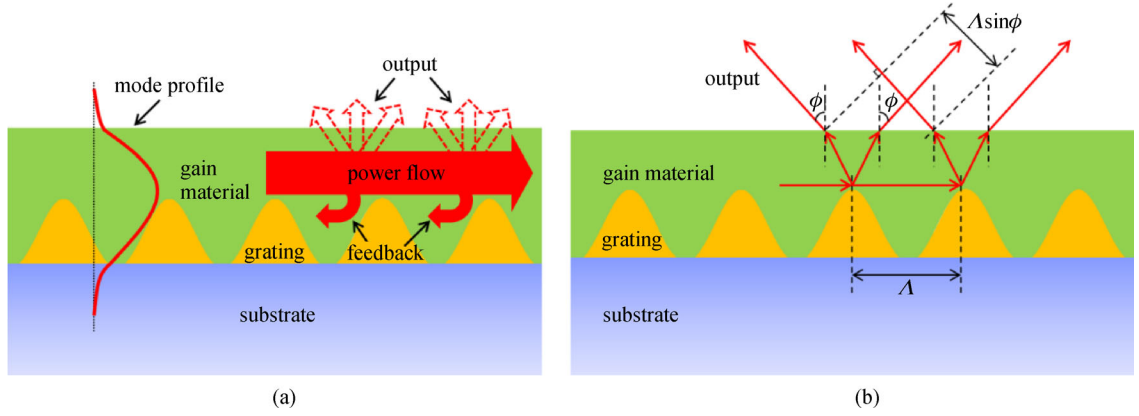
The main objectives of the design of laser cavities are to match the gain materials and to control the output characteristics. Several theories are developed to explore the property of DFB lasers, such as the diffraction theory, the couple wave theory, and the photonic bandgap theory. These theories provide a top-down approach to design the laser cavities.

According to the diffraction theory, there are three main roles of cavities, the feedback, the output coupling, and the waveguide. Some characteristics of DFB lasers can be obtained by employing the diffraction theory, such as output directions, output wavelengths, and mode numbers.

As shown in Fig. 6, a typical 1D DFB cavity consists of a grating and a waveguide. The waveguide plays two roles, guiding wave and providing gain. In Fig. 6(a), the red curve denotes the profile of the waveguide mode. Note that there exists a propagating mode and its counter propagating waveguide mode due to the diffraction of the grating [124]. The solid and dashed arrows indicate the feedback and the output direction, respectively. In Fig. 6(b), the red



**Fig. 5** Photonic crystals for DFB cavities. (a) 1D gratings; (b) 2D periodic structure; (c) 3D periodic structure; (d) Fibonacci quasi-crystals; (e) 2D quasi-crystals; (f) 3D quasi-crystals; (g) Chirped gratings; (h) 2D gradual periodic structure; (i) 3D random structure



**Fig. 6** (a) Schematic of the feedback and the outcoupling of the waveguide mode; (b) diffraction theory of DFB lasers. Reproduced with permission [123]. Copyright 2019, MDPI

arrows present the ray tracing of the propagating and emitting light. It is a simple diffraction picture.

The wavelengths of the waveguide mode must satisfy the Bragg condition in the cavity.

$$2n_{\text{eff}}\Lambda = m\lambda. \quad (1)$$

Here,  $n_{\text{eff}}$  is the effective refractive index of the waveguide mode,  $\Lambda$  is the grating period,  $m$  is a positive integer representing the number of standing wave nodes formed by the propagating and counterpropagating waveguide modes, and  $\lambda$  is the wavelength of the waveguide mode. The guided wave is diffracted by the grating at an angle  $\phi$ , forming the laser output as shown in Fig. 6. The emitted light should satisfy the condition of constructive interference:

$$\frac{2\pi n_{\text{eff}}}{\lambda}\Lambda + \frac{2\pi}{\lambda}\Lambda \sin\phi = 2\pi l, \quad (2)$$

where  $l$  is an integer that represents the diffraction order. By substituting Eq. (1) into Eq. (2), the relationship between the output direction of light and the diffraction order is obtained as

$$\frac{\sin\phi}{n_{\text{eff}}} = \frac{2l}{m} - 1, \quad l \in [0, m]. \quad (3)$$

Take the case of 1D gratings, the feedback is established by  $m$ th order diffraction, whereas the output coupling is supported by different diffraction with order numbers below or equal to  $m$ .

The mode number is decided by the parameter of the waveguide. For a given waveguide, there exist the critical thicknesses for the transverse electric mode (TE) and the transverse magnetic mode (TM), respectively. For the  $m$ th order TE mode ( $\text{TE}_m$ ), the critical thickness of the waveguide  $d_{\text{TE}}$  is given by

$$d_{\text{TE}} = \frac{(2m-1)\lambda}{4\sqrt{\varepsilon-1}}, \quad (4)$$

for the  $m$ th order TM mode ( $\text{TM}_m$ ), the critical thickness of the waveguide  $d_{\text{TM}}$  is given by

$$d_{\text{TM}} = \frac{m\lambda}{2\sqrt{\varepsilon-1}}, \quad (5)$$

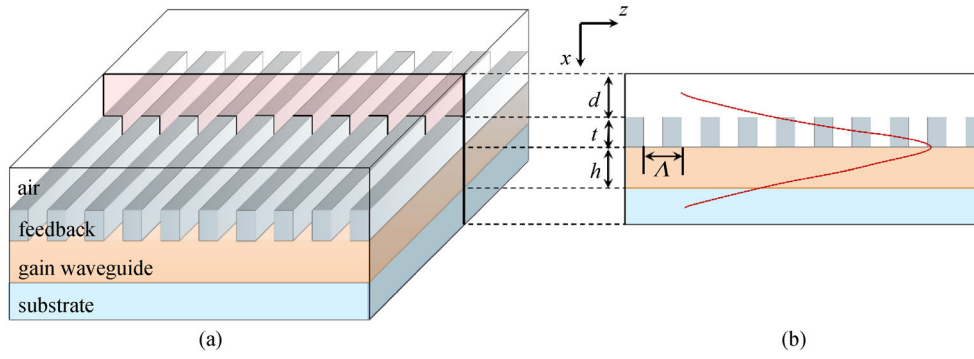
where  $\varepsilon$  is the effective refractive dielectric constant. According to the relationship of the waveguide mode with the waveguide thickness in Eqs. (4) and (5), if the waveguide thickness is larger than or equal to the critical thicknesses ( $d_{\text{TE}}$  or  $d_{\text{TM}}$ ), additional modes can be excited.

Under uncoupling conditions, 2D and 3D DFB cavities can be considered as a linear combination of 1D DFB cavities. Therefore, the diffraction theory is applicable to high dimensional cases.

The coupled wave theory reveals most of the physical mechanisms of DFB lasers, such as resonant mode patterns, mode selectivity, differential quantum efficiency, threshold conditions, and effects of end reflections [125–127]. Even under approximate conditions, the couple wave theory can be used to investigate the mode intensity distribution, the lasing wavelength, and the effective refractive index [128–130]. Here, takes an analytical approach for example, it is a combination of the coupled wave theory with the waveguide theory. The physical picture of this method is the resonant mode should meet both the Bragg condition and the waveguide condition.

Figure 7(a) presents a typical cavity, which consists of a grating and a gain waveguide. The cavity is reduced to a four-layered waveguide structure in Fig. 7(b). The electric field distribution in four waveguides in Fig. 7(b) can be defined as [129]

$$E_y(x,z) = e^{-ik_z z} \begin{cases} E_1 e^{a_1 x}, & x \in (-\infty, d], \\ E_2 e^{-a_2 x} + E_2' e^{a_2 x}, & x \in (d, d+t], \\ E_3 \cos(a_3 x + \Psi), & x \in (d+t, d+t+h), \\ E_4 e^{-a_4 x}, & x \in (d+t+h, +\infty), \end{cases} \quad (6)$$



**Fig. 7** (a) Schematic of DFB lasers; (b) reduced multi-layered model.  $\Lambda$  is the grating period;  $d$  is the thickness of air;  $t$  is the grating depth;  $h$  is the gain waveguide thickness. The red curve indicates the mode profile

here,  $k_z$  is the wave number in the  $z$ -direction.  $E_j$  and  $E_2'$  are the electric field amplitudes;  $a_j$  is the transverse wave number, and  $a_j = \frac{2\pi}{\lambda} \sqrt{n_{\text{eff}}^2 - n_j^2}$ ,  $j = 1, 2, 3, 4$ ;  $\Psi$  is a phase shift which is related with  $a_j$ .  $\Psi$  can be specified as  $m\pi + \tan^{-1} \frac{a_4}{a_3} - (d + t + h)a_3$ .  $m$  is a positive integer.

All electric field components can be calculated by applying the boundary condition. Therefore, the field distribution in each layer can be obtained [129,130]. By considering Eq. (1), the output wavelength can be also obtained.

Besides the couple wave theory, the photonic bandgap theory is also a full theory of DFB lasers [37,39,41]. The motivation comes from the fact that the resonant wavelength in Eq. (1) cannot propagate in the cavity due to the photonic bandgap. For easy understanding, a simplified model of 1D DFB lasers is derived using the coupled mode theory as follows. As shown in Fig. 7, the dielectric function of the cavity can be described by  $\varepsilon(x,z) = \varepsilon^{(0)}(x) + \Delta\varepsilon(x,z)$ . Here,  $\varepsilon^{(0)}(x)$  is the dielectric function of the cavity without considering the grating;  $\Delta\varepsilon(x,z)$  represents the periodic change of the dielectric function caused by the grating. Thus, the Fourier series of  $\Delta\varepsilon(x,z)$  is described by

$$\Delta\varepsilon(x,z) = \varepsilon_0 \sum_{m \neq 0} \Delta\varepsilon_m(x) e^{jm \frac{2\pi}{\Lambda} z}, \quad (7)$$

here  $\varepsilon_0$  is the dielectric constant in a vacuum.  $\Delta\varepsilon_m(x)$  is the  $m$ th Fourier coefficients. The wave equation of TE modes ( $E_y$  component) can be derived as

$$\left[ \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial z^2} + \omega^2 \mu_0 \varepsilon^{(0)}(x) \right] E_y = -\omega^2 \mu_0 \Delta\varepsilon(x,z) E_y, \quad (8)$$

where  $\omega$  is the angular frequency, and  $\mu_0$  is the permeability in a vacuum. As mentioned above, there is a propagating waveguide mode ( $A_+(z)$ ) and its counter-propagating waveguide mode ( $A_-(z)$ ) in the cavity. The electric field distribution in the cavity is described as

$[A_+(z)e^{-j\beta_z z} + A_-(z)e^{j\beta_z z}]E_y(x)$ , where  $\beta_z$  denotes the wavevector in the  $z$ -direction. When  $\beta_z = mG - \beta_z$ , the two waveguide modes strongly coupled with each other. Here the grating vector is defined as  $G = 2\pi/\Lambda$ . Considering  $\beta_z = 2\pi n_{\text{eff}}/\lambda_0$ , the Bragg condition in Eq. (1) is obtained.

If we define the two waveguide modes as  $a_+(z) = A_+(z)e^{-j\Delta\beta z}$  and  $a_-(z) = A_-(z)e^{j\Delta\beta z}$ , the coupled mode equation can be described as follows:

$$\frac{\partial}{\partial z} \begin{pmatrix} a_+(z) \\ a_-(z) \end{pmatrix} = -j \begin{pmatrix} \Delta\beta & \kappa \\ -\kappa^* & -\Delta\beta \end{pmatrix} \begin{pmatrix} a_+(z) \\ a_-(z) \end{pmatrix}, \quad (9)$$

here  $\beta_B = mG/2$ ,  $\Delta\beta = \beta_z - \beta_B$ , and  $\kappa$  is the coupling coefficient. By solving the eigenvalues of Eq. (9), the dispersion relationship of the resonant mode in the cavity is obtained as

$$\kappa = \beta_B \pm \sqrt{\Delta\beta^2 - |\kappa|^2}. \quad (10)$$

As shown in Eq. (10), the resonant wavelength satisfying the Bragg condition corresponds to the location of the photonic bandgap. Therefore, the photonic bandgap theory can predict the behavior of DFB lasers. For 2D and 3D cases, each photonic bandgap will affect the feedback due to the extended degree of freedom [36–38].

Special materials introduced in DFB lasers can also enrich the features, such as metallic materials [70,71,131], flexible materials [114,132,133], and fiber tips [134]. Take metallic materials as an example, plasmonics will improve the laser performance significantly by carefully designing [135–137]. Correspondingly, the related physical effect must be considered in the theoretical model [138,139].

### 3.3 Fabrication of laser cavities

One of the attractive advantages of DFB lasers based on organic materials is easy fabrication. A variety of fabrication schemes are used to introduce the organic materials in the DFB lasers, such as spin coating [140],

nanoimprint [141–143], nanograting transfer [144], thermal evaporation [144,145], horizontal dipping [146,147], ink-jet printing [148], and drop casting [149]. Note that the non-uniform film thickness should be considered in the last three methods. Recently, a versatile transfer coating method is proposed to assemble the DFB laser on arbitrary surfaces [80,134,150].

DFB cavities can be constructed by many approaches, such as interference lithography [44], nanoimprint lithography [151–153], photolithography [154], holographic interference [155–157], interference ablation [158,159], interference crosslinking [160,161], soft lithography [162], micromolding [147,163,164], electron beam lithography [165], and reactive ion etching [39].

The relative positions of the organic material, the DFB cavity, and the substrate are classified into three types, gain/cavity/substrate, cavity/gain/substrate, and active cavity/substrate, as shown in Fig. 8. There are some interesting differences in the laser performance of three configurations [44,107,159]. Based on the three configurations, complex cavities are designed to enrich the performance of DFB lasers, such as multilayer structures [130,140,166].

## 4 Advances in DFB laser based on organic materials

There are many excellent reviews dealing with the advances of DFB lasers based on organic materials [167–169]. In this paper, we will focus on some typical DFB lasers and the latest progress. These include new configurations, new fabrication methods, and performance improvements.

### 4.1 Lasing in 1D DFB cavities

As the most intuitive configuration, 1D DFB cavities has been investigated extensively. A variety of 1D structures are employed as DFB cavities, such as regular gratings [170], Fibonacci quasi-crystals [116], chirp gratings [35,171], beat gratings [172], and compound structures [108,109,173].

For 1D gratings, the output direction of the laser is related to the diffraction order followed Eq. (3). Therefore,

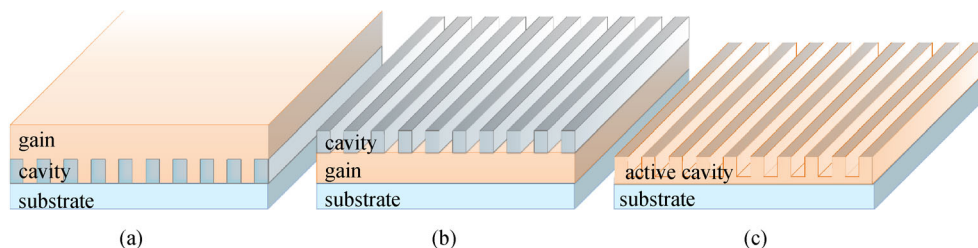
edge-emitting lasers and surface-emitting lasers are achieved for the 1<sup>st</sup> order laser and the 2<sup>nd</sup> laser, respectively [174]. For high-order lasers, oblique emitting can be observed as shown in Fig. 9.

For DFB cavities based on Fibonacci quasi-crystals, the lasers exhibit some intriguing features, such as directional output independent of the emission frequency and multi-wavelength operation [116]. All the features can be controlled by engineering the self-similar spectrum of the grating structure. The multi-wavelength operation is a very attractive topic in the field of lasers, which can also be realized in 1D DFB cavities. The main features of DFB lasers with chirped gratings are the single mode operation and excellent tunability [171]. The laser pattern and number of wavelengths can be flexibly adjusted by the beat structures consisting of several parallel gratings [172]. The case of compound structures is subtly different, in such cavities, ultralow thresholds can be achieved by controlling over the balance between feedback and output coupling [108,173].

### 4.2 Lasing in 2D DFB cavities

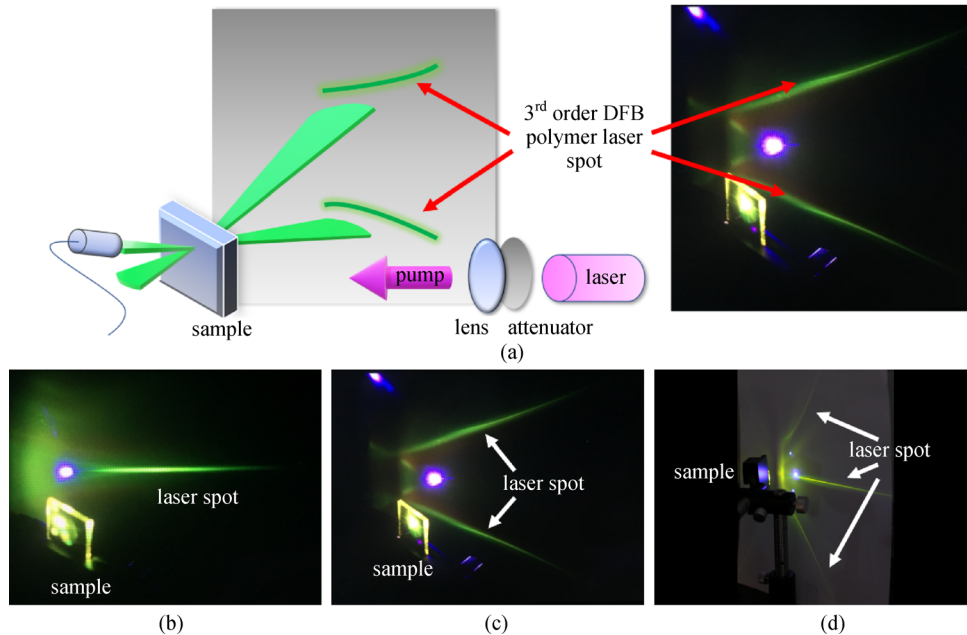
For laser cavities based on 2D DFB structures, the feedback is more effective. Thus, the laser performance of 2D DFB lasers is much better than that of 1D cases, such as thresholds, wavelength numbers, laser modes/patterns, phase distributions, polarization, and beam divergence. Most 2D photonic crystals are used to realized DFB lasing, such as square lattices [111], rectangular lattices [175], triangular lattices [114], hexagonal lattices [38], quasi-crystals [176], fan-shaped gratings [119], circular structures [121], and spiral gratings [122].

Generally, the 2D DFB cavity provides complete 2D feedback due to the 2<sup>nd</sup> Bragg diffraction and acts simultaneously as an output coupler by the 1<sup>st</sup> Bragg diffraction. Similar to the 1D cases, the balance between feedback and coupling can be controlled by adjusting the cavity parameters. So, the laser performance is affected by the strength of the cavity coupling [110]. There are numerous intriguing features in lasers with 2D DFB cavities. The radial/azimuthal polarization of the output beam is controlled by the parameter of square lattices [177,178]. Multi-wavelength emissions can be easily realized in rectangular lattices and triangular lattices



**Fig. 8** Schematic of DFB lasers with different configurations. (a) Gain/cavity/substrate; (b) cavity/gain/substrate; (c) active cavity/substrate





**Fig. 9** (a) Illustration of the experimental setup and formation mechanism of the pattern of a 3<sup>rd</sup> order DFB polymer laser; the purple spots shown in the right photograph are the reflection and diffraction of the pumping laser; (b) 2<sup>nd</sup> order laser pattern; (c) 3<sup>rd</sup> order laser pattern; (d) 4<sup>th</sup> order laser pattern. Reproduced with permission [123]. Copyright 2019, MDPI

[114]. Even the continuously tunability over a wide spectral range is achieved in fan-shaped gratings [119]. For circular cavities, the beam divergence is very small (~10 mrad) due to the symmetry of the cavity [179].

From a wavefront manipulation point of view, the DFB cavity can modulate the phase distribution of the emission light. For a spiral grating as a DFB cavity, vortex lasers with desired topological charge can be obtained by completely controlling the phase, handedness, and degree of helicity of the emitted beam [122]. Figure 10 demonstrates the profiles of vortex lasers generated by spiral gratings.

#### 4.3 Lasing in 3D DFB cavities

To date, relatively few studies have exploited the lasers based on 3D DFB cavities. The main reason is that most 3D photonic crystals are very difficult to be realized by micro-/nano-fabrication techniques. In this review, the 3D DFB cavities include 3D photonic crystals and stacked structures.

For 3D photonic crystals, lasing has been observed in holographic photonic crystals [154], liquid crystals [180], and opals photonic crystals [181,182]. In 3D photonic crystals, there exist many independent laser cavities which support multi-wavelength lasing emitted in different directions. Note that the symmetry of quasi-crystals is higher than that of periodic structures, which is easy to format photonic bandgaps. Therefore, the feedback for lasing is very efficient in quasi-crystals. Lasing has been observed in a 3D icosahedral quasicrystal fabricated by

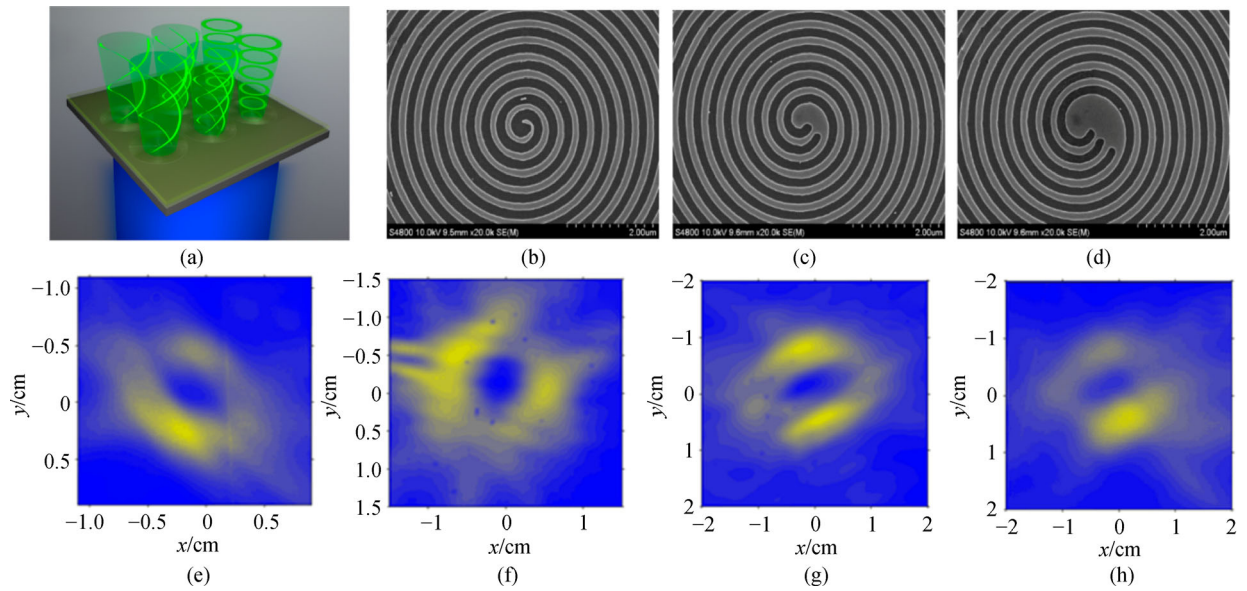
interference holography [183]. Multi-directional lasing is obtained due to the symmetry of quasi-crystals, as shown in Fig. 11.

The stacked structure consists of several 1D or 2D laser cavities [77,130,184]. Therefore, the laser properties of the stacked cavity are dependent on each component. For stacked structures, there are no 3D photonic bandgaps even considering the coupling effect.

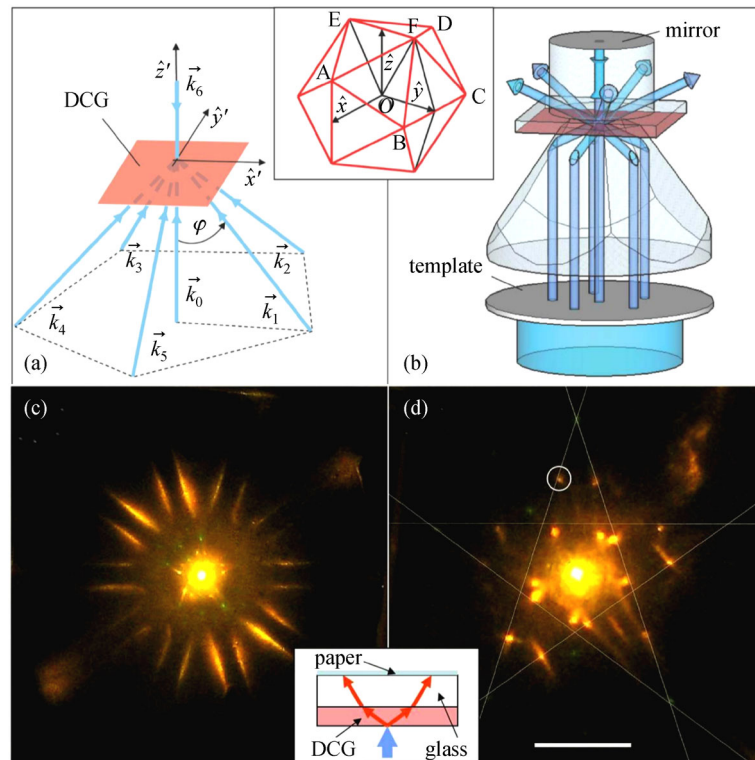
#### 4.4 Applications

As mentioned above, DFB cavities are the versatile building blocks for fundamental studies in nanoscale and potential applications. So far, many practical applications of organic DFB lasers have been proposed. One of the most straightforward applications is the visible light source integrated into spectroscopic systems. In particular, the lasers can be pumped by light-emitting diodes (LEDs) or laser diodes (LDs) [185,186], which accords with the trend of miniaturization of laser devices. It presents a versatile and powerful platform for various applications.

The broadly tunable emission throughout the visible range enables some applications in sensing [187–189], biomarker [190,191], high-performance light sources [192,193], on-chip communications [194,195], and optical circuits [196,197]. For example, label-free sensing can be achieved by a polymer DFB laser. The laser emission wavelength shifts with the variance of the effective refractive index modulated by the specific binding of the analyte [187]. Moreover, a lab-on-a-chip platform is constructed by integrating a 1<sup>st</sup> order organic DFB laser,



**Fig. 10** (a) Schematic of organic vortex laser arrays based on spiral gratings. SEM images of the center of the (b) one-arm spiral, (c) two-arm spiral, and (d) three-arm spiral gratings. Beam profiles recorded for the beams generated using (e) circular, (f) one-arm, (g) two-arm, and (h) three-arm spiral gratings. Reproduced with permission [122]. Copyright 2018, ACS Publishing



**Fig. 11** (a) 7-beam configuration for the icosahedral quasicrystal. The upper inset denotes an icosahedral quasicrystal lattice; (b) actual 7-beam arrangement using a truncated pentagonal pyramid; (c) icosahedral quasicrystal lasing pattern projected on the back side of the glass substrate (see lower inset). DCG is the abbreviation of the dichromate gelatin emulsions; (d) higher resolution projection of the icosahedral quasicrystal lasing for inner region. The lines are guides to the eyes. Reproduced with permission [183]. Copyright 2009, OSA Publishing

deep ultraviolet induced waveguides, and a nanostructured microfluidic channel into a poly (methyl methacrylate) substrate.

## 5 Summary and outlook

In summary, DFB lasing in photonic crystals has been extensively investigated in the past three decades. Numerous exciting developments have taken place in the field of DFB lasers based on organic materials. However, from the applications-based research point of view, there exist two limitations which baffle the marketization of such laser devices.

The first limitation is miniaturization. One the one hand, the size of the optical pump source is too large to integrate; on the other hand, the electrical pumping laser remains one of the major challenges. The challenges to be overcome include the excited-state triplet absorption, the absorption of metal contacts, and current densities required. In fact, organic semiconductors have some intrinsic drawbacks, such as low mobility and accumulated triplet states. The settlements may require significant innovations in materials science and engineering. Two feasible strategies for a trade-off between optical pumping and electrical pumping are indirect electrical pumping and fiber-based design. For indirect electrical pumping, the electrically driven light source (LEDs or LDs) is used to pump the organic semiconductor DFB laser optically. For the fiber-based design, the organic semiconductor DFB laser is fabricated on the fiber facet, removing the restriction of the electrical pumping.

The second limitation is the performance problems. Compared with commercial lasers, the output energy of the organic semiconductor DFB laser is relatively low, which is attributed to the small excitation volume. The continuous wave lasing is difficult to achieve in regular configurations. In most cases, pulsed wave lasing is obtained due to the long-lived triplet states. Moreover, some significant issues remain largely unexplored, such as the frequency repetition, pulse width, stability of materials, and lifetime of devices. Up to now, there are few researches involving the laser modulated techniques of organic semiconductor DFB lasers, including property manipulations and loading information. These techniques relate to amplitude modulation, intensity modulation, and phase modulation. Specific means include the  $Q$ -switching, mode locking, and so on. Further investigation is required to overcome the limitations to significantly enhance the laser performance. New opportunities and further progress can be expected from developing materials and techniques specifically for organic semiconductor DFB lasers.

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