

REVIEW

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Optical waveguides based on one-dimensional organic crystals

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Abstract

Optical waveguide of organic micro/nanocrystals is one of crucial elements in miniaturized integrated photonics. One-dimensional (1D) organic crystals with various optical features have attracted increasing interests towards promising photonic devices, such as multichannel signal converter, organic field-effect optical waveguide, sensitive detector, and optical logic gate. Therefore, a summary about the 1D organic micro/nanocrystals based optical waveguide is important for the rational design and fabrication of novel optical devices towards optoelectronics applications. Herein, recent advances of optical waveguide based on 1D organic micro/nanocrystals with solid, flexible, hollow, uniformly doped, core-shell, multiblock and branched structures are summarized from the aspects of the waveguide properties and applications in photonic devices. Furthermore, we presented our personal view about the expectation of future development in 1D organic optical waveguide for the photonic applications.

Keywords: Organic semiconductor molecules, Self-assembly, Organic micro/nanostructures, Optical waveguide, Organic photonics

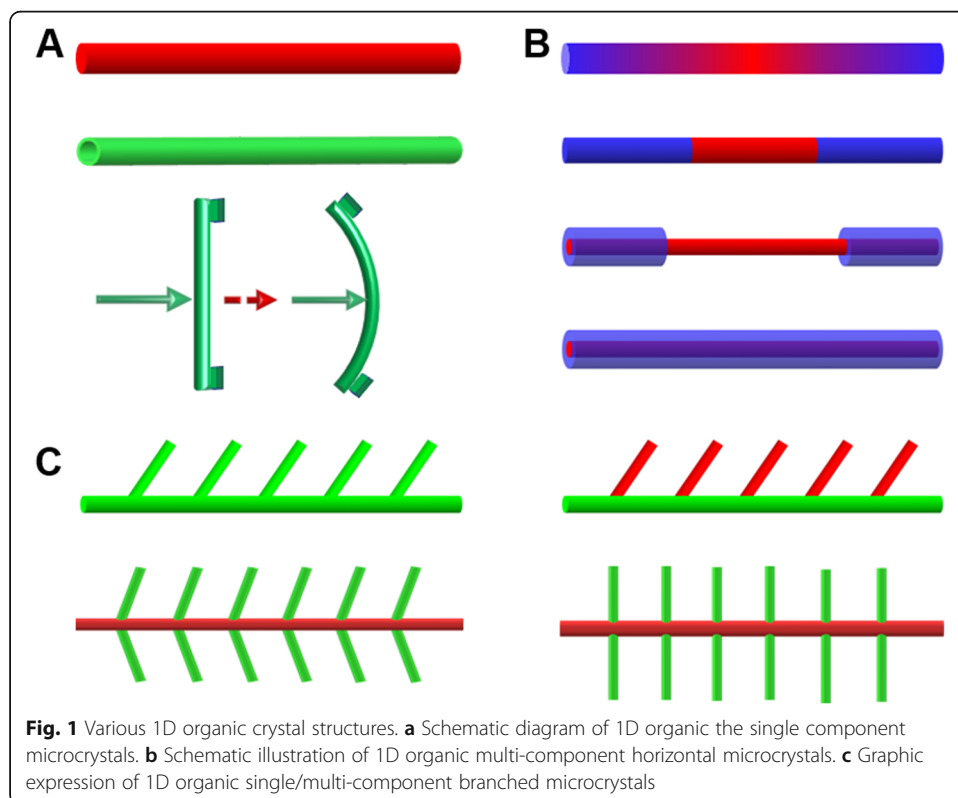
Introduction

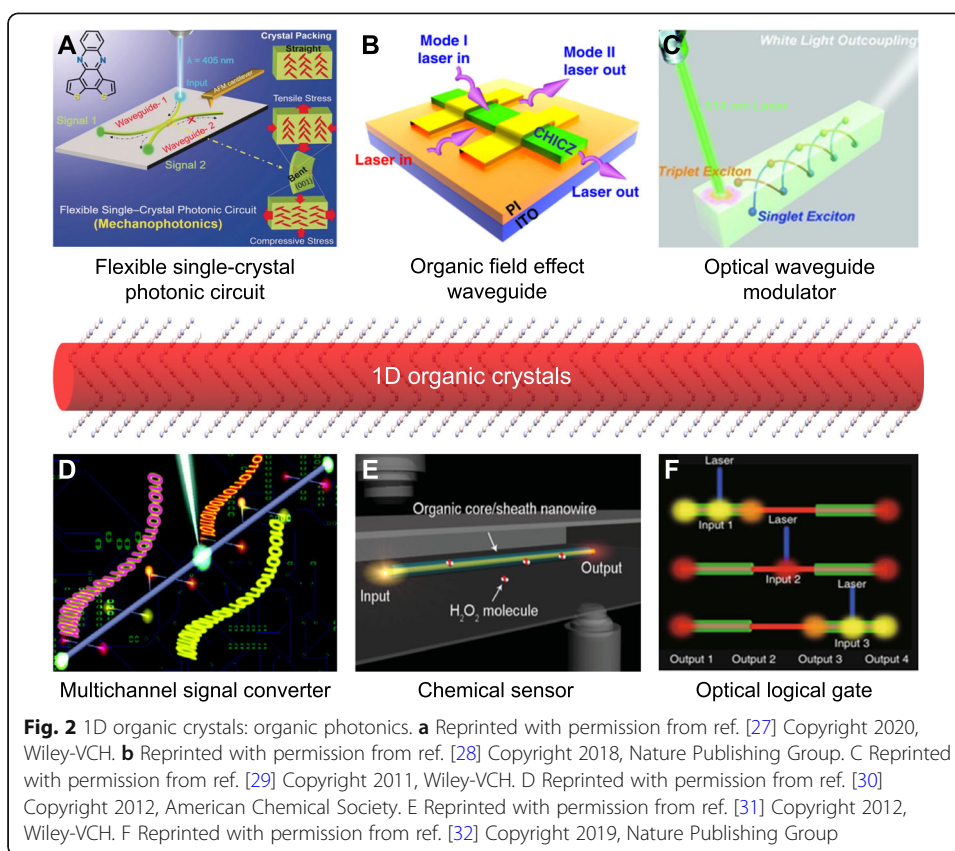
Optical waveguide, with light transmitting internally by total reflection at the interface of an optical medium, is a crucial element in many miniaturized integrated nanophotonics devices. Prof. Kao K, the father of optical fiber communications, was awarded the Nobel Prize 2009 in physics to praise his contribution in the transmission of light in fibers for optical communication. His achievements started a new era of human technology in information interaction. Up to date, micro/nanoscale optical waveguide based on various solid-state materials is one of the essential components to guide light. Remarkably, organic semiconductor materials with the excellent optoelectronic performance [1] have been proved to be promising candidates as the building blocks of photonic devices. Specifically, one-dimensional (1D) organic semiconductor crystals are generally suitable for the transport of photons, electrons, and excitons. Moreover, the 1D crystal with π -conjugated organic molecules can enhance charge-transporting mobility [2, 3]. These materials possess fundamental advantages including tailoring capability [4], good processability [5], few defects [2], uniform morphology [6], good thermal stability [7], solution processing [8] and high photoluminescence (PL)

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efficiency [9–11]. These organic molecular materials typically have weak intermolecular interactions between molecules, including hydrogen bonds, halogen bonds, π - π interactions, and van der Waals interactions. Optical properties of these organic molecules not only can be tuned via adjusting their molecular structures and packing mode, but also can be modified by a rational doping strategy. Therefore, these features allow such 1D crystals being used as “efficient optical waveguide” materials to significantly improve the wave propagation in media.

Additionally, the self-assembled 1D crystals from organic emissive molecules exhibit extensive optical-related applications, such as organic field-effect transistors (OFETs) [12–15], organic light-emitting transistors (OLETs) [16], chemical sensors [8, 17] and tunable color displays [18–20], which serve as the effective building blocks for organic integrated photonics. The photon propagation in the organic crystals has two different ways. The active optical waveguide refers to the optical output from the intrinsic emission of the sample by exciting it with an external energy source, whereas the passive optical waveguide mode refers to the light propagation of optical source [21–25]. But the single crystals cannot transport the multi-waveguide mode [26]. To address this issue, researchers pay more attention to multi-level micro/nanostructure. The organic heterostructures demonstrate the superior physicochemical features and the mixed passive/active waveguide mode, which is favorable for the development of multifunctional optoelectronic devices in the future. As is shown in Fig. 1d crystals have various morphologies including solid state, flexible, electrically controlled, hollow, uniformly doped, core/shell, multiblock, and branched structure. And their applications are illustrated in Fig. 2, including OFET, optical waveguide modulator, multichannel signal





converter, chemical sensor, optical logical gate and optical router. Table 1 also lists the representative examples of optical waveguides, elaborating the materials, crystal morphologies, emission colors and waveguide performances. In this review, after introducing the fundamentals of optical waveguides, we will focus on recent advances in optical waveguides of 1D crystals with various morphologies at micro/nanoscale as well as their diverse optoelectronic performance. Then we will discuss the challenges remained and provide our perspectives in this field.

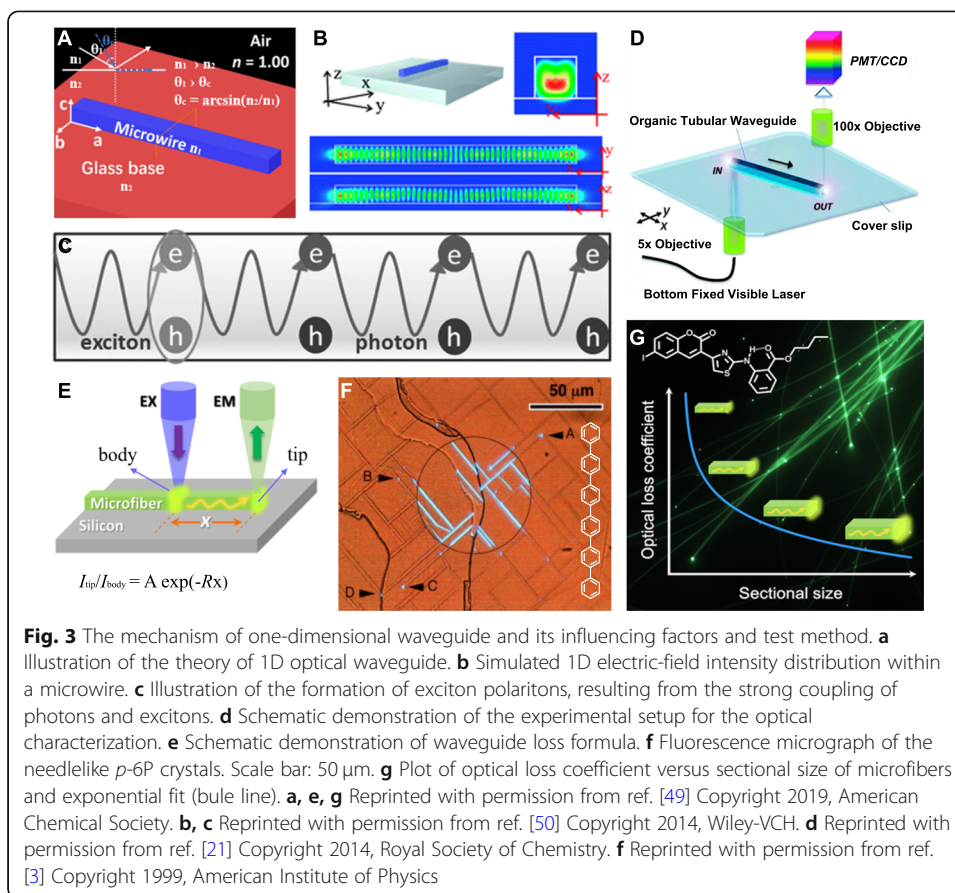
The fundamental mechanism of optical waveguide

Optical waveguide is an optical medium for wave transmission [1], which has attracted more and more researcher's attention. In this part, the optical waveguide of 1D micro/nanostructures based on organic small molecules is mainly described. Two factors are required to form an optical waveguide: one is a medium with a higher refractive index than that of the external environment [2], the other is total internal reflection. The optical waveguide mechanism is illustrated in Fig. 3a [49], where the transparent medium n_1 has higher refractive index than that of the environment (n_2 and n). The total internal reflection occurs when the light propagates from the denser medium (n_1) to the less dense medium (n_2 or n) and the incident angle θ_1 is greater than the critical angle (the formula is $\theta_c = \arcsin(n_2/n_1)$). Thereby, the optical field can be well-confined in 1D micro/nanostructures (Fig. 3b) [51].

As we have already known, organic crystals based on small organic molecules have outstanding crystallinity and elimination of crystal boundaries, which is beneficial to

Table 1 Representative examples of organic optical waveguides

| Morphology | Materials | Emission Colors | Loss Coefficient (dB/ μ m) | Refs |
|------------------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------------------|-----------------------------------------------------------------------|------|
| Microrod | Perylene diimide (PDI) | Red | 0.13 | [33] |
| | Diphenylalanine (FF) | White | | [34] |
| | 9,10-bis (phenylethynyl) anthracene (BPEA) | Orange | | [35] |
| Microribbon | 2,8-dichloro-5,11-dihexyl-indolo(3,2-b) carbazole (CHICZ) | Blue | 0.01 | [28] |
| | 1,4-bis((E)-4-(1,2,2-triphenylvinyl)-styryl)-2,5-dimethoxybenzene (TPDSB) | Green | 0.0175 | [36] |
| 1D flexible microstructure | Tetrafluoropyridyl derivative 1 | Orange | $R_{\text{bending}} = 0.00047$, $R_{\text{straight}} = 0.00043$ | [37] |
| | Pentafluorophenyl derivative 2 | Green | $R_{\text{bending}} = 0.001003$, $R_{\text{straight}} = 0.000703$ | [37] |
| | (E)-1-(4-(dimethylamino)phenyl)iminomethyl-2-hydroxyl-naphthalene (DPIN) | red | $R_{\text{bending}} = 0.000274$, $R_{\text{straight}} = 0.000270$ | [38] |
| | 1,4-bis(2-cyanophenylethynyl)benzene | Blue | $R_{\text{straight}} = 0.4967$ | [39] |
| 1D microtube | 9,10-bis (phenylethynyl) anthracene (BPEA) | Yellow | | [40] |
| | Polydiacetylene (PDA) | Red | 0.18 | [41] |
| | Cocrystal: 4,4'-((1E,1'E) - (2,5-dimethoxy-1,4-phenylene) bis (ethene-2,1-diyl)) dipyridine (DPEpe) and 1,4-diiodotetrafluorobenzene (F ₄ DIB) | Green | 0.0145 | [42] |
| | Perylene doped 2,4,5-triphenylimidazole (TPI) | UV/green | 0.097 | [43] |
| 1D uniformly doped microwire | Iridium (III) bis (2-phenyl benzothiazolato-N, C2') acetylacetonate ((BT) ₂ Ir(acac) doped 9,10-diphenylanthracene (DPA) | Yellow | 0.13 | [29] |
| | Perylene doped TPI | UV/green | 0.155 | [43] |
| 1D core/shell microrod | 9,10-bis (phenylethynyl) anthracene (BPEA)/ chemo-reactive bis(2,4,5-trichloro-6-carbopentoxo-phenyl) oxalate (CPPO) | Yellow/green | | [31] |
| | 4,4'-((1E,1'E) - (2,5-dimethoxy-1,4-phenylene) bis (ethene-2,1-diyl)) dipyridine (DPEpe)/DPEpe-HCl | Green/red | 0.0338 | [44] |
| | DPEpe-HCl/DPEpe | Red/green | 0.054 | [44] |
| | Cocrystal: DPEpe and 4-bromo-2,3,5,6-tetrafluorobenzoic acid (BrFTA)/DPEpe and 1,4-diiodotetra-fluorobenzene (F ₄ DIB) | Red/yellow | | [32] |
| 1D multiblock microstructure | <i>trans</i> -2,2'-((1E,1'E)-1,4-phenylenebis (ethene-2,1-diyl))-dibenzonitrile (<i>trans</i> -o-BCB)/ <i>Cis</i> -2,2'-((1E,1'E)-1,4-phenylenebis (ethene-2,1-diyl))-dibenzonitrile (<i>Cis</i> -o-BCB) | Blue/red | 0.035 | [45] |
| | [Ru (bpy) ₃] ²⁺ (bpy = 2,2'-bipyridine) doped [Ir (ppy) ₂ (pzpy)] ⁺ (ppy = 2 phenyl pyridine, pzpy = 2-(1H-pyrazol-1-yl) pyridine) | Green/red/orange | | [46] |
| | Closed-state (L-c) | Blue | | [22] |
| | Open-state (L-o) | Blue | | [22] |
| 1D branched microstructure | Aluminum tris(8-Hydroxyquinoline) (Alq ₃) and 1,5-diaminoanthraquinone (DAAQ) | Red/green | | [30] |
| | 1,4-dimethoxy-2,5-di[4'-(cyano) styryl] benzene (COPV) and 2,4,5 triphenyl imidazole (TPI) | Blue/green | | [47] |
| | 2,2'-((1E,1'E)-1,4-phenylenebis (ethene-2,1-diyl))-dibenzonitrile (o-BCB) | Green | | [48] |



optical transmission. In addition, 1D nanowires based on π -conjugated organic molecular aggregates can act as the high-performance optical waveguides (Fig. 3f) [3]. Further investigation on 1D organic crystal reveals that electrons and holes can be easier to form Frenkel exciton (Fig. 3c) [50–52]. When the cavity provides the exciton with adequate energy, photons and excitons in the cavity can strongly couple to form the exciton polaritons (EPs) [53]. Compared with uncoupled light, the refractive index of 1D organic crystal has enhanced by coupling light [29, 51, 54, 55], showing remarkable propagation properties [56]. This can help to break the diffraction limit so that 1D organic micro/nanostructure with subwavelength diameters is conducive to the further building blocks for miniaturized optoelectronic devices. However, because the high substrate effect caused the higher optical loss, many efforts have tried to overcome this drawback. For example, Huang et al. [57] and our group [42] fabricated hollow vertical organic nanoarrays and hollow microtubes whose optical waveguide performance have improved.

The waveguide capability of micro/nanostructure can be expressed by the loss coefficient (R). As shown in Fig. 3d [21] and Fig. 3e [49], the PL intensity at the excitation and emission points of the sample was measured. The loss coefficient (R) was calculated by single-exponential fitting $I_{\text{tip}}/I_{\text{body}} = A \exp(-RX)$, where X is the distance between the excited site and the emitting tip. And the lower value of R indicates the better propagation. However, scale effect should not be neglected

in waveguide losses. For example, Song et al. [49] fabricated 2-((5-(6-iodo-2-oxo-2H-chromen-3-yl) thiazol-2-yl) amino)-Benzoate (ICTAB) microfiber to prove the size-dependent effect of the waveguide (Fig. 3g), which indicates that the value R increased rapidly with decreasing the cross-sectional size of the microfibers.

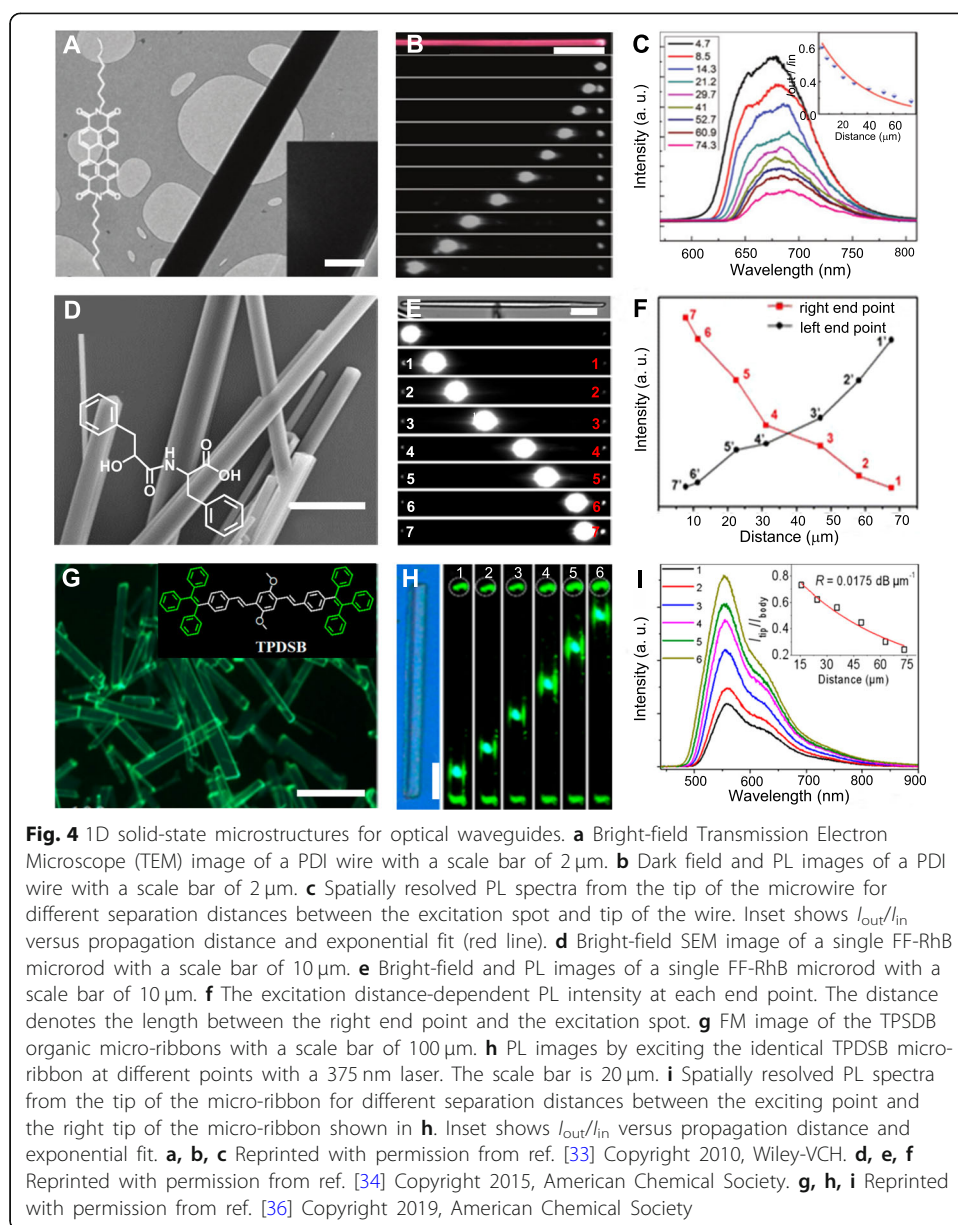
In this section, we briefly described the fundamental mechanism of optical waveguide, which will be helpful to understand 1D waveguide behaviors and the optical properties to be discussed in the following section.

1D waveguide behaviors and optical properties based on organic crystals

Scientific researchers have contributed to the rise of 1D organic micro/nanostructure materials in the past decade, prompting the potential applications of the materials as building blocks for miniaturized optoelectronic devices, such as logic gates [58], chemical sensors [59], photodetectors [60], organic solid-state lasers (OSSLs) [61–63], organic field-effect transistors (OFETs) [64], organic light-emitting transistors (OLETs) [65, 66] and optical waveguides [67]. Especially, optical waveguide based on 1D organic crystals has attracted enormous attention in the last few years due to their unique waveguide behaviors and optical properties. In the organic crystals, molecule stacking mode, molecule structure, and molecule interaction can contribute substantially to the charge-transporting mobility. Unlike inorganic crystals, 1D organic crystals are aggregated by weak intermolecular forces [20], such as hydrogen bonds, halogen bonds, π - π interactions, and van der Waals interactions [68]. In this regard, we will introduce 1D crystals with solid-state, solid electrically controlled, flexible, hollow, uniformly doped, core-shell, multiblock and branch structures. Their waveguide behaviors and optical properties are also discussed.

1D solid-state optical waveguides

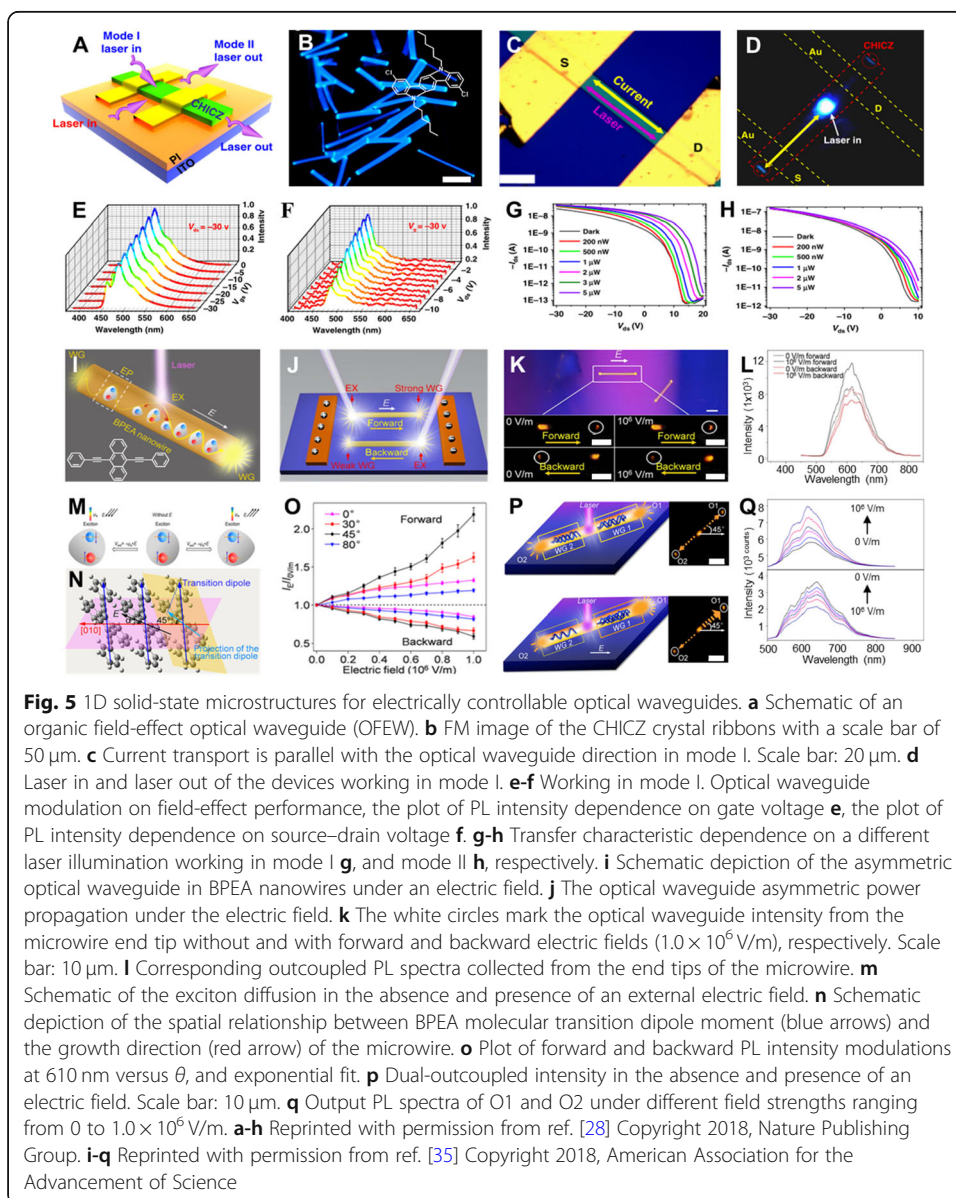
In the past decades, researchers are devoted to studying the 1D inorganic solid micro/nanoscale waveguides [69, 70], and the preparation method of the 1D structure is relatively mature. In comparison with 1D inorganic solid waveguide at micro/nanoscale, 1D organic materials possess many advantages, such as tailor-made structure [62, 71] and intense fluorescence [72, 73]. For example, Bao et al. [33] fabricated high-quality microwires of perylene diimide (PDI) molecules (Fig. 4a) through template-assisted self-assembly method on the surface of graphene oxide. The nanowire crystals are a high-quality medium for optical waveguides. And when the laser excites the different positions of the 1D microwire from the left to the right (Fig. 4b), the corresponding PL signals can be detected from the right terminus of the microwire (Fig. 4c), indicating that the intensity of the PL signals at the right tip of the microwire increases with the gradual decrease of the propagation distance. Li et al. [34] obtained rhodamine B (RhB) doped Diphenylalanine (FF) microrods with smooth surfaces (Fig. 4d). When the laser excites the different positions of the 1D microrod from the left to the right, the out-coupled intensity of the right tip of the microrod is gradually enhanced compared to the out-coupled intensity of the left tip of the microrod (Fig. 4e, and f). In terms of the microtubes with regular morphology and bright spot at microrod tips, suggesting that the microrod have good optical waveguide performance. According to the previous work, our group [36] designed and synthesized 1,4-bis((*E*)-4-(1,2,2-



triphenylvinyl)styryl)-2,5-dimethoxybenzene (TPDSB) with aggregation-induced emission (AIE) phenomenon (Fig. 4g). Through the solution-processing approach, these AIE molecules form the 1D green-emissive solid-state microstructures (Fig. 4h), which possess superior optical properties (Fig. 4i). In addition to the simple transmission of optical waveguide in 1D solid crystals, we can achieve the chiral waveguides. Prof. Chandrasekar et al. reported a chiral waveguide based on chiral structure, which shows circular dichroism effects in the nonlinear optical emission [74]. The chiral structure with the smaller optical waveguide loss suggests the better optical waveguide performance. Therefore, the chiral waveguide based on chiral structure contributes to the development of nonlinear optical nanophotonic devices with chirality information encoded.

1D electrically controlled optical waveguides

Electrically controlled optical waveguides have become critically important for optical communication, information processing, and high-density connection of optoelectronic devices [28, 35, 75–77]. However, it is a great challenge to efficiently achieve electrically controllable light propagation for practical applications in high-density photonic devices because of the non-interactive nature of photons. Herein, we give examples to demonstrate novel and effective strategies for 1D micro/nanostructures to manipulate photon transport. Hu et al. [28] controllably synthesized the single-crystalline 2,8-dichloro-5,11-dihexyl-indolo (3,2-b) carbazole (CHICZ) microribbons (Fig. 5b) for high performance optoelectronic device. It is shown in Fig. 5a that an individual CHICZ crystal ribbon with gold stripes as source and drain electrodes has two modes for optical waveguide directions, which are along the conducting channel (mode I) and perpendicular to the conducting channel (mode II), respectively. In terms of mode I



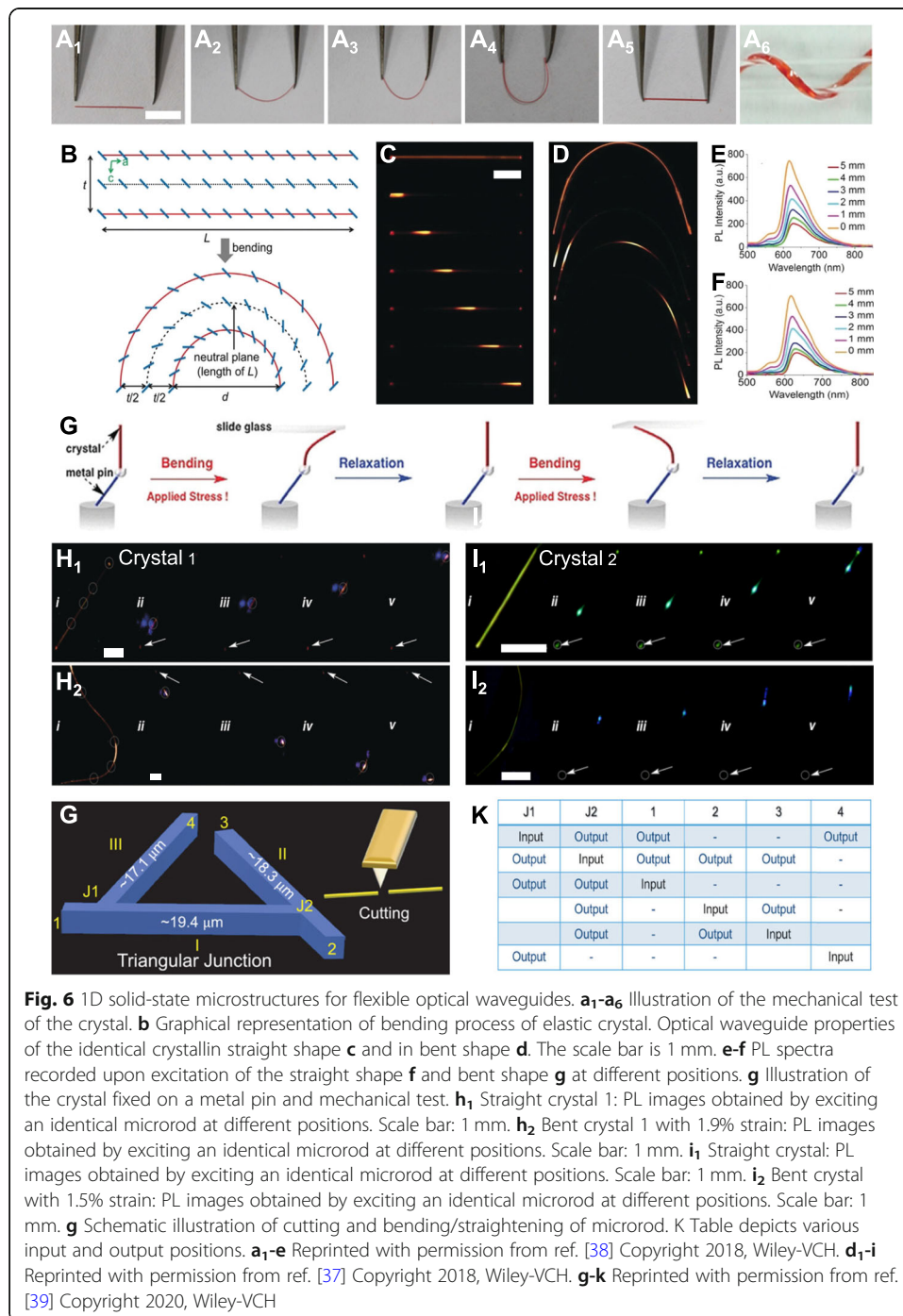
(Fig. 5c-d), the direction of the current transport is perpendicular to that of the laser. Under different source–drain voltage (V_{ds}), PL intensity gradually becomes smaller when V_{ds} ranges from 0 to -30 V with a constant V_g (gate voltage) = -30 V, and its magnitude of the decrease can be explained by the modulation-degree M ($M = (1 - (I_{on}/I_{off}) \times 100\%)$), under which case M is over 25% (Fig. 5e). The M value is over 75% when V_g is shifting from 0 to -30 V with a constant $V_{ds} = -30$ V (Fig. 5f). This indicates the modulation of field effect optical waveguide. At the same time, under different laser intensities, with V_{ds} scanning from 20 to -30 V and $V_g = -30$ V in mode I (Fig. 5g), the current modulation ratio R ($R = (I_{laser} - I_{dark})/I_{dark}$) is significantly increased up to 14,800 under $5 \mu\text{W}$ laser illumination. However, when other conditions remain unchanged, the current modulation ratio R is only increased up to 100 under $5 \mu\text{W}$ in mode II (Fig. 5h), suggesting an effective modulation on the performance of the optical waveguide transistor.

Additionally, Zhao et al. [35] realized the controllable self-assembly of the 9,10-bis(phenylethynyl) anthracene microwires with asymmetric photon transport. It is well known that the weak interaction between photons is little affected by the external electric field because photons are chargeless and massless gauge bosons. But photons can be coupled with excitons to form a new type of hybrid state known as EPs. The formation of EPs is shown in Fig. 5i, where the external electric field can control the exciton diffusion, leading to the directional flow of photons and forming the asymmetric optical waveguide. As shown in Fig. 5m, the exciton diffusion can be changed by the interaction potential V_{ext} ($V_{ext} = -\mu_e E$, where μ_e is the transition moment of the exciton and E is the external field.). V_{ext} can be modulated by two factors which are the electric field strength and the electric field direction. This process is shown in Fig. 5j-l, when the electrical field with certain direction is exerted, the waveguide output along the electrical field will be enhanced, whereas the waveguide output reverse to the electrical field will be weakened. According to the previous work on polarized emission of organic single crystal [33, 78–81], the PL intensity has the maximum value when the direction of incident light is parallel to the transition dipole moment of the organic crystal. The calculated result shows that the polarized angle θ between the transition dipole moment of the nanowire and the axis of the nanowire is 45° . By making the electric field and light propagation direction co-planar (Fig. 5n) and adjusting the direction and strength of the electric field, higher propagation can be achieved when the EPs propagate along the field direction (Fig. 5o). Moreover, when the laser excites the middle of the nanowire, waveguide intensities at wire tips are obviously different with electric field compared to waveguide intensity of wire tips without electric field, which is the asymmetric waveguide (Fig. 5p). The variable asymmetric out-coupled spectra and the modulation ratios spectra ($(I_E - I_0)/I_0$) of two wire tips are also obtained by changing the input electrical field (Fig. 5q). All the above results successfully demonstrate the electrically controlled optical waveguides in 1D solid-state.

1D flexible optical waveguides

Organic crystals with characteristics of brittle and crack are difficult to create flexible waveguides under the external stimuli, which makes it high requirement for the nature of material. Thus, the investigation of elastic organic crystals is one of the currently

developing trends in flexible photonics. Elastic organic crystals under the external stimuli usually have various outstanding mechanical behaviors, e.g., bending [37, 38, 82–85] and twisting [86, 87]. As a pioneer work, Prof. Zhang et al. firstly demonstrated the flexible microrods of (*E*)-1-(4-(dimethylamino)phenyl)iminomethyl-2-hydroxyl-naphthalene (DPIN) crystals (Fig. 6a₁–a₆), which is owing to their planar molecular conformation and periodic monoclinic packing pattern [38]. Moreover, the stretching of outer layer molecular distance and the compression of internal molecular distance by applied external force in the DPIN organic crystal are shown in Fig. 6b. The stretching of



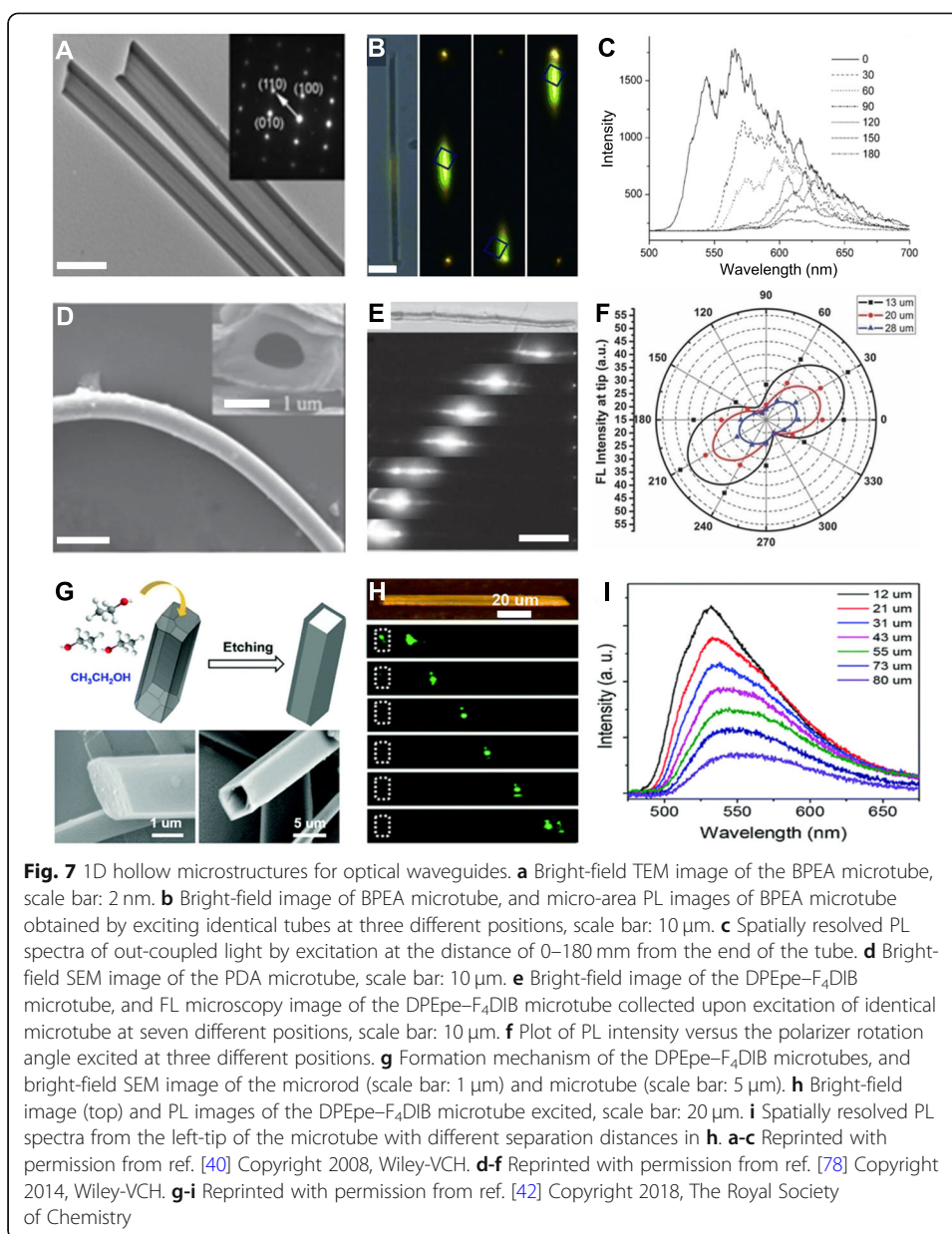
molecular out-layer structure and the compression of the internal-molecular structure can be realized by the $\pi \dots \pi$ interactions between molecules. In addition, the C-H ... O hydrogen bonds and the close packing features also are key factors for the achievement of elastic DPIN crystal. And the optical waveguides of the straight crystal and bent crystal were displayed in Fig. 6c and d. The optical loss values of the straight crystal (0.270 dB/mm) and bent crystal (0.274 dB/mm) have the close value (Fig. 6e-f), suggesting the highly elastic property and waveguide performance of DPIN organic crystal.

Additionally, Hayashi et al. [37] designed and synthesized a structure of tetrafluoropyridyl derivative 1 and pentafluorophenyl derivative 2 with π -conjugated structure and they made great efforts to prepare crystal 1 (orange emission) and crystal 2 (green emission) through the vapor diffusion process. Moreover, they calculated the interatomic distance and the intermolecular torsion angle of crystal 1 and crystal 2, indicating that the molecular structures in both crystal 1 and crystal 2 have a highly planar and rigid conformation. Consequently, crystal 1 and crystal 2 under repeatedly bending-relaxation test show good elasticity (Fig. 6a), and testing bent and straight optical waveguides of both crystal 1 and crystal 2 by exciting different positions of crystal 1 and crystal 2 with a laser beam (Fig. 6b₁, b₂, c₁, and c₂). It suggests that the flexible optical waveguide performance of crystal 1 ($R_{\text{Bent}} = 0.047$ dB/mm and $R_{\text{Straight}} = 0.043$ dB/mm, with no bending mechanofluorochromism) is better than that of crystal 2 ($R_{\text{Bent}} = 1.003$ dB/mm and $R_{\text{Straight}} = 0.703$ dB/mm, with bending mechanofluorochromism) owing to the lower molecular planarity of crystal 2.

The precise microscale operation technique of flexible crystal contributes to realize the flexible photonic devices. At present, Prof. Chandrasekar et al. fabricated 1,4-bis (2-cyanophenylethynyl) benzene elastic crystal through the vapor diffusion process [39]. In the waveguide's experiment, the 1,4-bis (2-cyanophenylethynyl) benzene elastic crystal with blue emission possesses lower optical waveguide loss, indicating that the micro-rods have the better optical waveguide performance. Using atomic force microscopy manipulation technique, the geometrical shape of elastic crystal can be tuned from bent crystal to straight crystal, and the length of crystal can be modified via the precise cutting of atomic force microscopy cantilever. As is shown in Fig. 6g, the geometrical shape of elastic crystals can become triangle geometries through the precise cutting of atomic force microscopy cantilever. The lengths of elastic crystals with triangle geometries are 19.4 μm , 18.3 μm and 17.1 μm , respectively. The elastic crystals with triangle geometries have two junctions (J1 and J2) and four output/input tips (1, 2, 3 and 4), respectively. When the input signal is respectively injected into J1, J2, 1, 2, 3 and 4, respectively, the elastic crystals with triangle geometries are selectively output emission. The Fig. 6k summarizes the various output and input optical signals, which can be used in the optical logic gate operation.

1D hollow optical waveguides

In comparison with 1D solid structures, 1D hollow tubular structures display the outstanding waveguide behaviors owing to reduced optical loss in the otherwise solid center and improved optical propagation inside the remaining outer shell [23–25, 40, 41, 88–90]. As shown in Fig. 7a, researchers [40] prepared a hollow tubular microrod

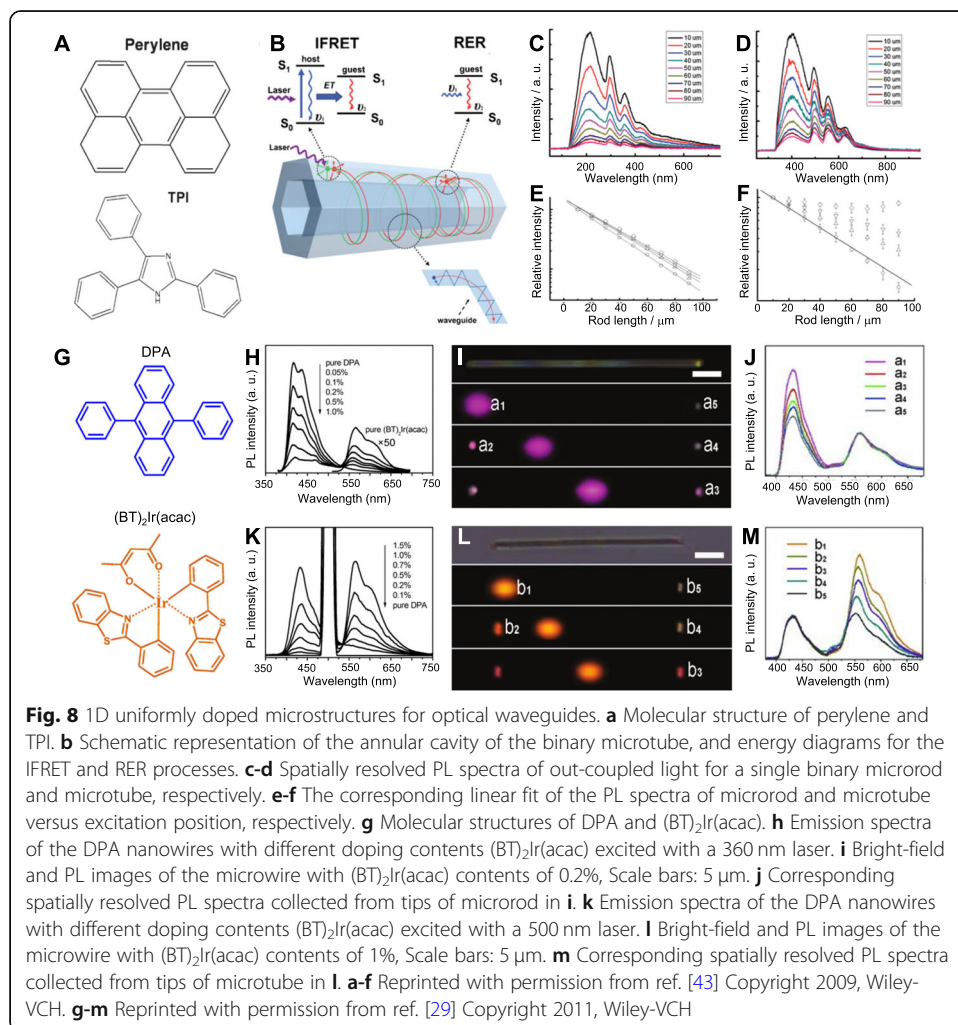


crystal based on the small organic molecule of 9,10-bis (phenylethynyl) anthracene, which possess glossy surface. And the tips of BPEA microtube have bright luminescence spots by exciting the different positions of this microtube via a laser beam (Fig. 7b) and the corresponding distance dependent PL spectra are shown in Fig. 7c, signifying the better waveguide performance. Moreover, Ming et al. [41] put great efforts to prepare 1D Polydiacetylene (PDA) microtubes via the hierarchical self-assembly method. Microtubes with homogenously smooth surface are shown in the scanning electron microscopy (SEM) image (Fig. 7d). It is indicated that the microtubes display the bright emission at tips (Fig. 7e–f) when the microtubes at center are excited by a laser beam. Furthermore, according to these pioneering works, our group [42] fabricated microrods and microtubes by the solution-processing approach. The 1D organic cocrystal DPEpe–F₄DIB microrods can be changed into microtubes after ethanol

etching, because the surface energy at the center of the microrods is relatively high compared to that of the outer surfaces. As shown in Fig. 7g, the microtubes have regular morphology with smooth surface, leading to the good optical waveguide performance with a low loss coefficient of 0.0145 dB/ μm (Fig. 7h, and i).

1D uniformly doped optical waveguides

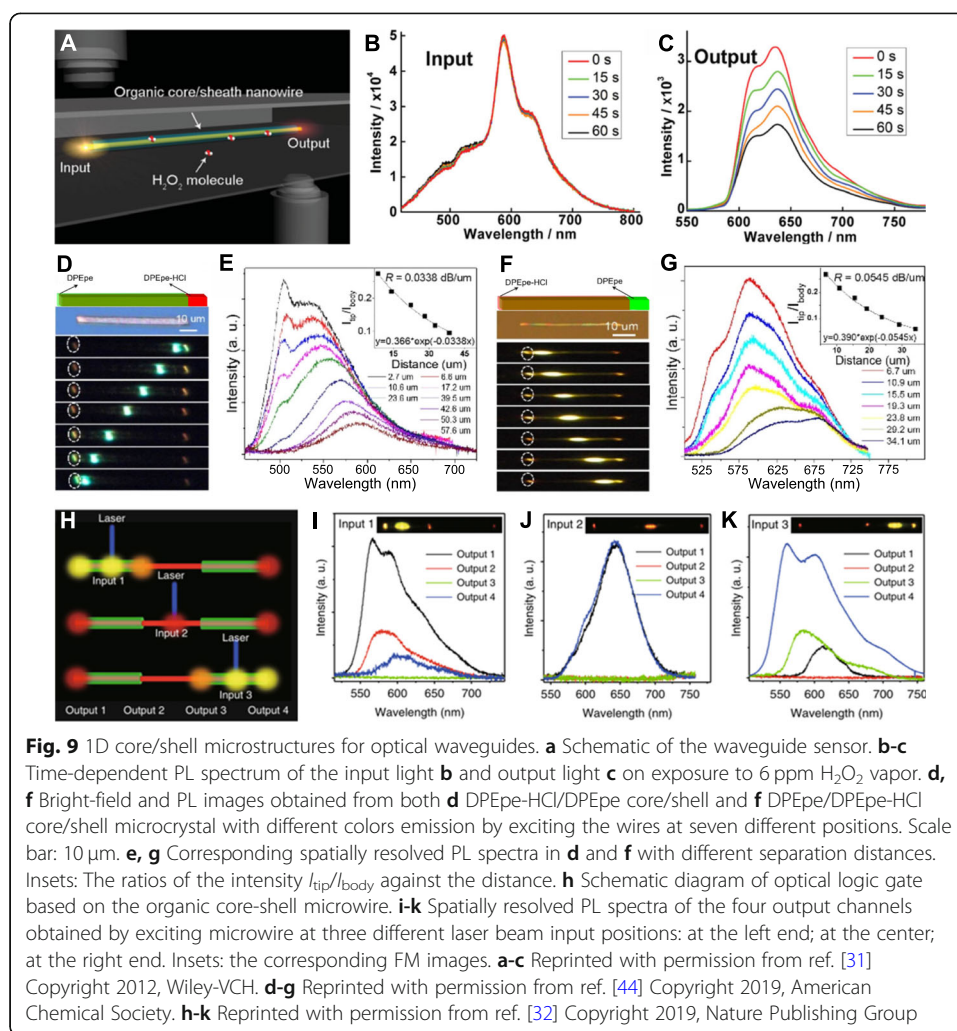
There have been many reports about 1D organic uniformly doped microstructures [10, 29, 43, 91, 92]. Their applications are mainly in optical modulation and full-color display for integrated optoelectronic devices. Herein, Yao et al. [43] fabricated 1D binary microtubes and microrods, respectively. The molecular structures of perylene and 2,4,5-triphenylimidazole (TPI) are shown in Fig. 8a. The perylene molecules, serving as an assistant, are evenly dispersed in the TPI matrix to etch the solid rod into microtube with the doping ratio of 1.25% in the host-guest system. Moreover, the etched hollow center of the microtubes leads to lower waveguide loss relative to microrods, resulting in a much longer propagation length (Fig. 8c-f). The schematic of both intermolecular fluorescence resonance energy transfer (IFRET) and remote energy relay (RER) process



is shown in Fig. 8b. 1D microstructure excited by a laser beam can conduct not only the UV emission but also the green emission because of the energy transfer between TPI and perylene components. The remote energy relay process refers to waveguide modulate in the binary microtube, suggesting that the doped perylene constituent in the binary microtube will reabsorb the ultraviolet photon of the TPI component and subsequently re-emit the photons in the green-emissive bands. Yao et al. [29] also reported the wavelength modulation in the Iridium (III) bis (2-phenyl benzothiazolato-N, C^{2'})acetylacetonate((BT)₂Ir(acac)) doped 9,10-di-phenylanthracene (DPA) organic nanowire via the fluctuations of singlet and triplet excitons during propagation (Fig. 8g). Using different doping ratio, the spectra of the doped nanowires upon a 350 nm laser excitation indicate that the orange emission is significantly enhanced (Fig. 8h), suggesting efficient singlet exciton energy transfer from DPA molecules to (BT)₂Ir(acac) molecules. Furthermore, the optical signal of the spectra of the doped nanowires upon a 500 nm laser excitation demonstrates the blue emission, showing efficient triplet exciton energy back transfer from (BT)₂Ir(acac) molecules to DPA molecules (Fig. 8k). The micro-area PL images obtained from two identical (BT)₂Ir(acac) doped nanowires with the doping concentration of 0.2% and 1.0% (Fig. 8i, l) display the blue emission and the orange emission respectively because of the exciton fluctuations, and their corresponding PL spectra are shown in Fig. 8j and m. In summary, this novel type of organic waveguide, acting as an optical regulator, is helpful for developing novel multi-component waveguide working as building blocks in integrated nanoscale devices [93].

1D core-shell optical waveguides

The core/shell micro/nanostructures can be divided into zero-dimension (0D) particles [94–96], 1D wires [31, 97], and two dimensional (2D) plates [98, 99] from the dimensionality. Great progress in the organic core/shell structures has been made after two decades' development in both fundamental research and applications. 1D organic core-shell heterostructures are superior in physicochemical features, such as evanescent coupling and carrier transport [31], and energy transfer [44]. There have been numerous reports about optical waveguides based on 1D organic core-shell heterostructures and their potential applications are mainly in chemical gas sensor [19, 31], multi-color optical waveguide [44] and optical logic gate [32, 100]. Figure 9a shows an example of the organic core/shell nanowires with waveguiding core and chemiluminogenic cladding that can be applied to the chemical gas sensor [31]. The organic core/shell nanowires consisting of the single-crystalline 9,10-bis (phenylethynyl) anthracene (BPEA) and bis (2,4,5-trichloro-6-carbopen-toxy-phenyl) oxalate (CPPO) molecules have great mechanical and optical properties. However, CPPO crystal itself is poor in mechanical and optical properties. This is because that the evanescent coupling between the core and shell resulted from the introduction of BPEA molecules is a key factor to improve optical property. The optical waveguide response of the nanowires exposed to H₂O₂ vapors is remarkably fast (35 ms) due to the high sensitivity and selectivity of CPPO molecules to H₂O₂ vapors. Therefore, to prove the chemiluminescence sensitivity and intensity of a single core/shell nanowire to H₂O₂ vapors, the incident light is inputted from one end of the core-shell nanowires and the output emission is measured at the other end in a sealed glass chamber with a 6 ppm H₂O₂ gas flowing system. The



corresponding PL spectra of the input and output light at different time are shown in Fig. 9b and c, suggesting that the core-shell nanowires with rapid and high selectivity of optical sensor of a trace of H_2O_2 gas.

Furthermore, our group [44] reported a 1D core-shell micro/nanostructures based on the π -conjugated organic molecules of DPEpe and DPEpe-HCl. The formation of DPEpe crystal and DPEpe-HCl crystal is reversible protonation/deprotonation process. As shown in Fig. 9d and f, self-assembled green-emissive single-crystal DPEpe micro-rods and red-emissive single-crystal DPEpe-HCl microrods can both act as the core or the shell. And the PL spectra of DPEpe/DPEpe-HCl core-shell microrods (Fig. 9g) has a significant red shift compared to those of DPEpe-HCl/DPEpe core-shell microrods (Fig. 9e), which implies the remarkable optical tunability of the core-shell system.

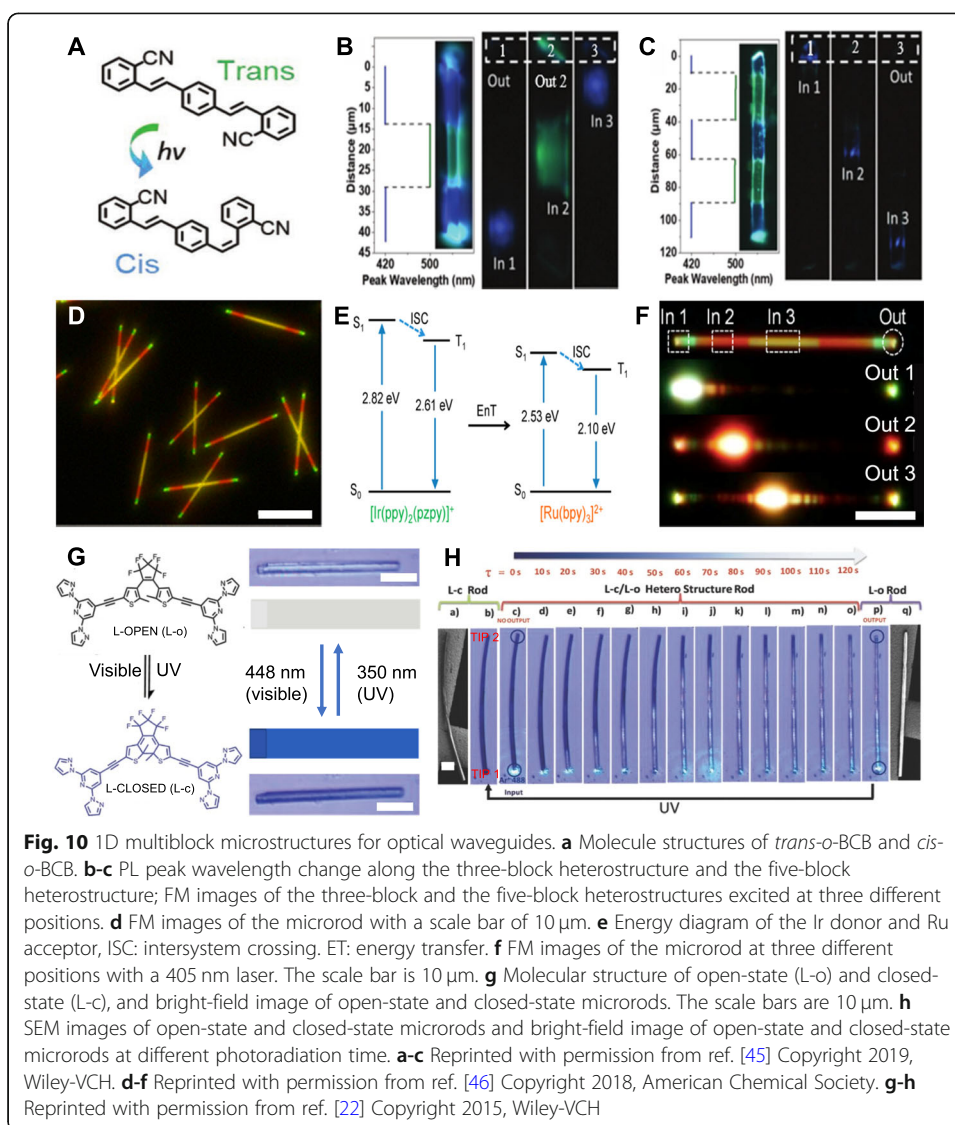
Recently, our group [32] also reported the horizontal epitaxial growth process via the hierarchical self-assembly of 1D core-shell micro/nanostructures. The DPEpe- F_4DIB cocrystal based on the hydrogen bond between DPEpe molecule and F_4DIB molecule displays the green emission and the DPEpe-BrFTA cocrystal based on the halogen bonds between DPEpe molecule and BrFTA molecule shows the red emission. 1D DPEpe-BrFTA/DPEpe- F_4DIB core-shell nanowires with a low lattice mismatching rate

between the two cocrystal are controllably synthesized by the precise manipulation of noncovalent interactions: hydrogen bonds, halogen bonds and π - π interaction. The multicolor emission properties of the heterostructure nanowire at Input 1, Input 2 and Input 3 excitation positions are shown in the plot of Fig. 9h and the output spectra of Output 1, Output 2, Output 3 and Output 4 channels were collected (Fig. 9i-k). When Input 1 and Input 3 positions of the core-shell nanowire were excited by a laser beam, the photon propagation process from excitation position to the two ends corresponds to the active and passive modes, respectively (Fig. 9i, and k). When excited at Input 2 of the core-shell nanowire, the photon propagation to the two ends is passive (Fig. 9j). This feature of 1D core-shell heterostructure waveguides can be applied in the optical logic gate operation at microscale.

1D multiblock optical waveguides

The optical waveguide of 1D multiblock micro/nanostructures consisting of the binary components make them serve as excellent candidates for optical applications [45, 46, 101–104]. Here, we previously reported 1D in-series organic multiblock heterostructures based on the *trans-o*-BCB and *cis-o*-BCB (Fig. 10a), whose construction employs a highly controllable photochromic method [45]. As shown in Fig. 10b and c, the 3-block heterostructure and the 5-block heterostructure can be realized by selectively covering the sections of the microrods, resulting in the molecular structure transformation from *trans-o*-BCB to the *cis-o*-BCB. The FL microscope images of *cis-o*-BCB structures and *trans-o*-BCB microcrystals exhibits the blue and green emission, respectively. The optical signal of the Out 1, where radiative energy transfer occurred, is blue/green-emissive, which indicates the passive/active mixed waveguide mode resulted from the energy transfer between *cis-o*-BCB structure and *trans-o*-BCB structure (Fig. 10b). When the input signal is injected in the In 1, In 2 and In 3, separately, The optical signal of the all output channels are blue-emissive, which denotes the passive waveguide mode (Fig. 10c). The efficient energy transfer in the 1D multiblock micro/nanostructures is a universal mechanism, it can realize the selective output wavelength and the multi-color emission. Furthermore, Yao et al. [46] reported the multi-color phosphorescence of 1D multiblock microstructures (Fig. 10d) with the emission from green to red based on efficient triplet energy transfer between the donor ($[\text{Ir}(\text{ppy})_2(\text{pzpy})]^+$) and acceptor ($[\text{Ru}(\text{bpy})_3]^{2+}$). Figure 10e displays the energy diagram of the Ir donor and Ru acceptor. The triplet state energy (T_1) of Ir donor (2.61 eV) is higher than the S_1 (2.53 eV) and T_1 (2.1 eV) level of Ru acceptor. It is suggested that the energy of Ir donor can transfer to the Ru acceptor. The FL microscopy image of a heterojunction nanorod illustrates the multi-color phosphorescence (Fig. 10f). When the input laser is injected into In 1, In 2 and In 3, the corresponding output ports Out 1, Out 2 and Out 3 show green, red and green/red mixed emission, respectively.

Additionally, Prof. Chandrasekar et al. reported a photoresponsive microrod based on dithienylethene molecule unit [22]. As shown in the Fig. 10g, the closed-state (L-c) and the open-state (L-o) microrods can be formed by their molecules. Moreover, the formation of closed-state (L-c) and open-state (L-o) microrods is reversible process under exposure to UV (355 nm) and visible light (448 nm). The open-state (L-o) microrod was excited by a laser beam (448 nm), which exhibits a

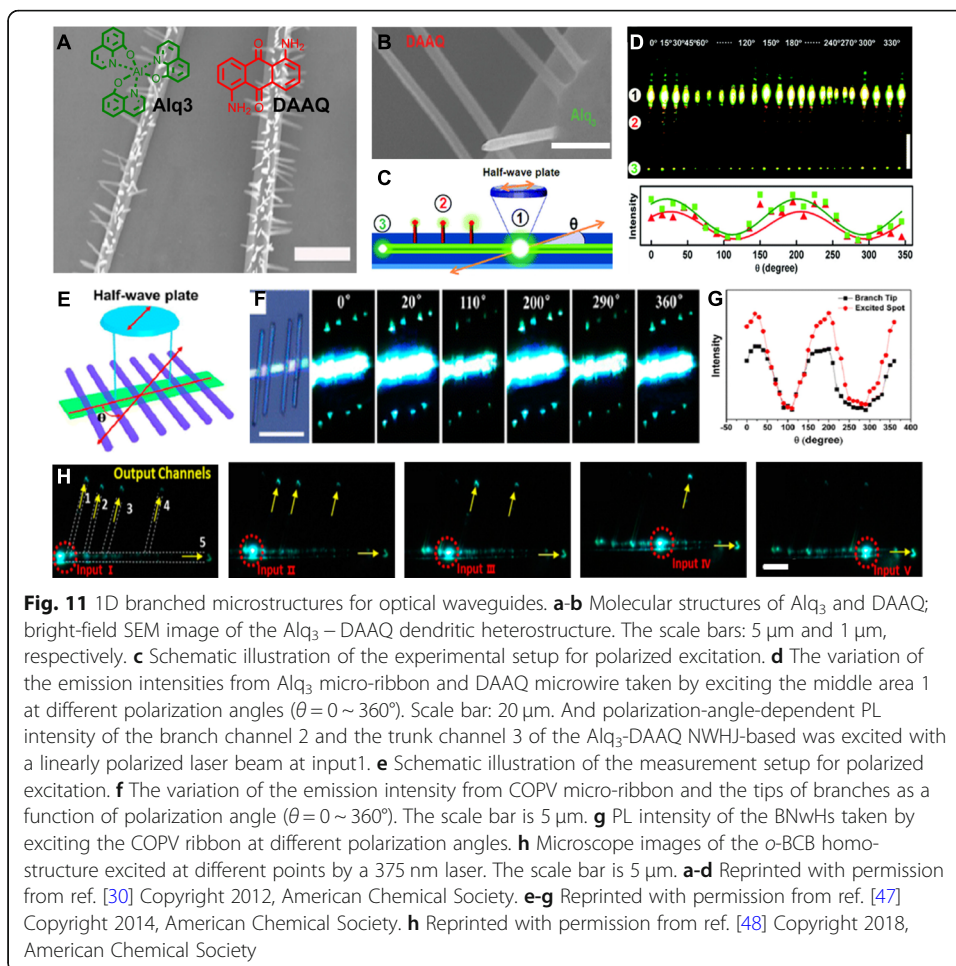


passive waveguide. However, the closed-state (L-c) microrod can absorb the energy of a laser beam (448 nm), converting to the open-state (L-o) microrod by a time-dependent solid state cyclo-reversion reaction, which can realize the output of transmission light at the end part. According to this photoresponsive feature, 1D multiblock heterostructure is composed of the closed-state (L-c) and the open-state (L-o) structure. The formation of 1D multiblock heterostructure is illustrated in Fig. 10h. the Tip 1 at the closed-state (L-c) microrod was excited by a 448 nm laser beam. At $t \approx 0$ s, input laser signal cannot propagate optical signal to Tip 2 of the closed-state (L-c) microrod. When the photoradiation time changes from 0 to 120 s, the length of open-state (L-o) microrod in 1D multiblock heterostructure gradually increases from 0 to 153 μm . Finally, At $t \approx 120$ s, the closed-state (L-c) microrod completely converts to the open-state (L-o) microrod, suggesting that input laser signal at the Tip 1 of open-state (L-o) microrod can transmission optical signal to Tip 2. Therefore, the photoresponsive 1D organic heterostructures with high controllability have the potential application for waveguide modulator.

1D branched optical waveguides

1D branched heterostructures and homostructures at micro/nanoscale can be prepared by various methods, including solvent slow evaporation method [48, 105, 106], two-step solution and vapor deposition process [30] and one-pot solution method [47]. 1D branched micro/nanostructure with fine photonic properties can be used in optical routers [30, 47, 107, 108], asymmetric optical waveguide [48, 106] and multichannel signal converter [109, 110]. Dendritic organic heterojunctions with aluminum tris (8-hydroxyquinoline) (Alq_3) microwire trunks and 1,5-diaminoanthraquinone (DAAQ) nanowire branches (Fig. 11a, and b) were successfully prepared by Zhao et al. through the two-step solution and vapor deposition process [30]. Dendritic organic heterojunctions emit multicolor light with a focused laser beam excited at the trunk. This phenomenon is attributed to both waveguide propagation and energy transfer from trunk to branches in 1D dendritic heterojunctions. Moreover, as shown in Fig. 11c and d, the PL intensities at the trunk tip and branch tip periodically change as the polarization angle increases from 0 to 360 degrees, implying the potential to act as the optical routers.

Fu et al. [47] also reported a 1D organic heterojunction with COPV microribbon trunks and TPI branches synthesized via a one-pot solution method for nanoscale optical router. This formation mechanism of this heterostructure is mainly caused by the



multiple hydrogen-bonding interactions between TPI and COPV constituents. The schematic illustration of optical channeling property [47] (Fig. 11e) exhibits a measurement setup for the polarized excitation. Furthermore, the polarization-angle-dependent PL intensities of excited spot and branch tip can also be controllably changed by rotating the incident polarization ($\theta = 0 \sim 360^\circ$) (Fig. 11f-g).

Recently, our group [48] successfully synthesized the branched nanowire homostructures consisting of the single component of *o*-BCB with asymmetric optical waveguide via a simple solvent slow evaporation method. As clearly seen in Fig. 11h, when the excitation position moves from Input I to Input V, only the output signal at the output ports that are at the right side of the input position can be detected, suggesting the asymmetric optical waveguide performance of the branched homostructure.

Advantages and disadvantages of 1D organic micro/nanostructures

1D organic solid crystals have inspired increasing interests due to their inherent features, such as weak molecular interaction, high purity and minimized defects, leading to good optical waveguide performance of 1D solid crystal. Therefore, these optical waveguides based on various structures made of 1D crystals demonstrate the low optical waveguide loss, the multi-functional optical properties, and so on. Compared with 1D solid structures, 1D hollow structures are promising for the high-performance optical waveguides on account of the reduced optical loss in the center of 1D hollow structures and the improved optical propagation inside the remaining outer shell. Therefore, 1D hollow optical waveguides are the excellent candidates for the practical applications. Generally, the 1D organic crystals with characteristics of brittle and crack, the optical waveguide based on 1D organic crystals cannot be applied in flexible photonics devices. 1D organic crystals with elastic feature are the good candidates for flexible photonics devices [27]. 1D crystals always have the single-color emission. Thus, this is not conducive to realize the multi-optical signal propagation of 1D crystals of one component. Doping in the organic crystals is an efficient strategy to realize the multicolor emissions, which can be applied in waveguide modulator [43]. At present, optical waveguides based on 1D organic crystals are symmetric, leading to the simple waveguide performance of 1D organic crystals. The symmetric waveguides based on organic crystals can realize the asymmetric optical transmission via the external electric field. The asymmetric waveguides of 1D organic crystals expand opportunities for creating scalable integration of photonics in a chip [28].

In comparison with 1D single structures, the multi-level structures, combined the advantages of single component materials, make up for the intrinsic deficiencies of single component materials. These fascinating organic topological heterostructures are superior in physicochemical features, which have been proved to be promising candidates as the building blocks of photonic devices. For example, the branched structures realize the photon transmission characteristics of multiple input/output channels, which is an ideal platform for optical router [47]. The multiblock structures demonstrate the modulation of active optical waveguide and passive optical waveguide, which can be applied in the optical logic gate operation at microscale [45]. The core-shell structures can improve stability of core layer and reduce its optical waveguide loss. When the shell layer of the core-shell structures is sensitive to gas molecules, the core/shell structures realize the application of chemical sensor [31].

Conclusion and outlook

As the indispensable component of optical circuits, optical waveguides have undergone tremendous development during the past decade. The most typical example is that Prof. Kao K have used optical fibers made by quartz glass for optical communication. Commercial optical fibers today, being composed of either silica or transparent polymers, have perfect data-carrying capacities. The organic optical waveguides are, relatively, a new area. Impressively, organic micro/nanostructures, especially 1D organic micro/nanostructures based on small molecules have shown unique advantages, such as tunable optical property, toiling molecule structure, high-thermal stability and controllable supramolecular self-assembly process by the solution-processing approach. These distinguishing features of 1D micro/nanoscale structures are expected to work as a promising optical waveguide in miniaturized photonic applications. Herein, we have summarized 8 different types of 1D organic micro/nanostructures developed recently for optical waveguides with specific properties, specific properties. Which have been applied for optical logic gate, optical router, multichannel signal converter, OFEW, waveguide modulator, and chemical sensor, etc. Therefore, these 1D organic micro/nanostructures can function as the outstanding optical elements in organic integrated photonics.

Notably, it highlights that the investigations of these 1D organic micro/nanostructures are in the early stage of research, which have plenty of room for further innovations. Indeed, there are still many challenges existing, which needs tremendous efforts for the exploration in this new field. Firstly, the main interaction in organic micro/nanostructure corresponds to the non-covalent interaction between molecules, such as van der Waals, hydrogen bonds, halogen bonds and π - π interactions. The 1D organic micro/nanocrystals with high-quality through a facile approach remain a huge challenge. Its nucleation growth process is very susceptible to the external environment such as solvent type, temperature, and solution concentration. The manipulation of the non-covalent interaction between molecules is an efficient strategy to fabricate the organic micro/nanocrystals. When the strong non-covalent interaction between molecules exists, the self-assembled 1D micro-nano structures can be designed and synthesized according to the molecular packing mode. It is easy to form the 1D organic structure. Although the optical waveguide based on organic materials has achieved great development, the optical performances in organic molecular crystals are still a significant drawback compared to commercial optical fibers composed of either silica or transparent polymers. This is a long way to realize practical application based on the optical waveguide of organic crystals. But in the past two decades, breakthroughs in optical waveguides based on 1D organic crystals have enabled us to have a deeper understanding of the tuning optical properties of 1D organic crystals and the construction of optical building blocks based on organic crystals. It is meaningful that these photonic components can be applied in chemical sensor, flexible single-crystal photonic circuit, organic field effect waveguide, multichannel signal converter optical logical gate, and optical waveguide modulator. Therefore, it is believed that the multi-functional organic micro/nanostructures with stability are great importance for photonic devices. In the future, scientific researchers will have efforts to design and synthesize the organic micro/nanostructure with multi-function and good stability to realize the practical photonic devices based on optical waveguides. Furthermore, the 1D organic

heterostructures comprising of multi-components remain a considerable challenge in lattice matching and surface-interface energy balance. This issue hinders the development of 1D organic heterostructures for the optical building blocks. Therefore, it is essential to develop 1D organic heterostructure with the facile fabrication methods.

In summary, photonic applications especially the optical waveguides based on 1D organic crystals have great prospect and high value. Scientists around the world have strong interests for the exploration in this field. Despite some existing challenges, we believe that developing a series of multi-functional waveguide devices will be the most disruptive progress in future.

Abbreviations

1D: One-dimensional; OFEW: Organic field-effect optical waveguide; IFRET: Intermolecular fluorescence resonance energy transfer; RER: Remote energy relay; 0D: Zero-dimension; 2D: Two dimensional; OFETs: Organic field effect transistors; OLETs: Organic light-emitting transistors; EPs: Exciton polaritons; PL: Photoluminescence; FL: Fluorescence; SEM: Scanning electron microscopy; TEM: Transmission electron microscope; UV: Ultraviolet

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Authors' contributions

These authors contributed equally: S. Chen, M.-P. Zhuo. Methodology, L.-S. Liao; writing—original draft preparation, S. Chen, M.-P. Zhuo, X.-D. Wang; writing—review and editing, G.-Q. Wei, X.-D. Wang, L.-S. Liao; supervision, L.-S. Liao. All authors read and approved the final manuscript.

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Availability of data and materials

All data needed to evaluate the conclusions in the manuscript are presented herein and/or the Supplementary Materials. Additional data related to this study may be requested from the authors.

Competing interests

The authors declare no competing financial interests.

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