

# Highly Stable On-Chip Embedded Organic Whispering Gallery Mode Lasers

Shi-Yang Lu, Hong-Hua Fang, Jing Feng, Hong Xia, Tie-Qiang Zhang, Qi-Dai Chen,  
and Hong-Bo Sun, *Member, IEEE*

**Abstract**—Chip-embedded organic resonator is fabricated with 2,5-Bis(4-biphenyl)thiophene (BP1T) crystals encapsulated with polydimethylsiloxane (PDMS). Whispering gallery mode lasing is demonstrated in these on-chip embedded crystalline microresonators, without decline in the spectral properties, and performance in comparison to the unencapsulated devices. The encapsulated lasers exhibit quality factor ( $Q$ ) as high as 1066, and lasing threshold is about 120 nJ ( $1.5 \text{ mJ/cm}^2$ ). Meanwhile, with protection of the PDMS, operation lifetime of the laser is increased from  $2.1 \times 10^6$  to  $4.6 \times 10^6$  pulses.

**Index Terms**—Microchip lasers, optical resonators, organic semiconductors,  $Q$ -factor, whispering gallery modes (WGMs).

## I. INTRODUCTION

WHISPERING gallery mode (WGM) lasers have been considered as important building blocks in large-scale photonic integrated circuits (PICs), because they possess small footprint, low mode volumes, as well as high quality factors [1]–[3]. Organic semiconductors have shown great potential for such kind of applications, because of their features such as low cost, large gain cross sections [4]–[6]. Over the past decade, organic WGM lasers in the form of microtoroids [7], [8], microspheres [9], [10], microdisks [11], [12] or microring [13], have been well demonstrated with high quality factor when isolated in air.

To integrate these organic microlaser into a PIC chip, such resonators have to be coupled with passive components like waveguide, or buried in a chip to construct complex photonic circuits [2], [14]–[17]. As a light source in the chip, it requires that the lasing performance of microlasers are stable and not affected by the surroundings. The dilemma is that the properties of WGM laser is highly sensitive to the environmental change. When integrated on chip, the organic laser often suffers from serious decline in performance, and even the lasing modes disappear, because of the reduction in refractive index contrast ( $\Delta n$ ) to environmental medium. Thus, the development of robust microlasers with adequate stability in spectral properties and performance is important for practical applications in different working environment.

Manuscript received February 19, 2014; revised May 9, 2014; accepted April 14, 2014. Date of publication May 21, 2014; date of current version June 20, 2014.

S.-Y. Lu, H.-H. Fang, J. Feng, H. Xia, Q.-D. Chen, and H.-B. Sun are with the State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun 130012, China (e-mail: honghua.fang@gmail.com; hbsun@jlu.edu.cn).

T.-Q. Zhang is with the College of Physics, Jilin University, Changchun 130023, China.

Color version of one or more of the figures in this paper are available online at <http://ieeexplore.ieee.org>

Digital Object Identifier 10.1109/JLT.2014.2326429

Additionally, another important issue related to application is the degradation of organic lasers in presence of oxygen and water [18]–[20]. It is known that when exposed to air, organic active materials would lead to rapid degradation under either optical or electrical excitation. For these reasons, an appropriate encapsulation configuration for organic device protection is needed to ensure practicable lifetimes.

In this paper, we present chip-embedded organic WGM optical microresonators, and successfully achieve highly stable lasing emission with high performance. The comparison of lasing properties demonstrate that the spectral properties of microlasers are not deformed when they are encapsulated in polydimethylsiloxane (PDMS). Furthermore, the operational lifetime of lasers is doubly increased in comparison to the unencapsulated microlasers.

## II. PROCEDURE FOR PAPER SUBMISSION

### A. Device Fabrication

Shown in Fig. 1 is embedded microresonator configuration. The optical gain material used in the experiment is the organic single crystal of 2,5-Bis(4-biphenyl)thiophene (BP1T), a representative of thiophene/phenylene co-oligomers (TPCOs) [21], [22]. The circle microdisk resonator was fabricated by top-down method combined with photolithography and reactive ion etching (RIE). Firstly, the slice-like BP1T single crystals were grown in physical vapor transport quartz tube [23], and transferred onto a cleaned silica substrate, which was spin-coated with photoresist (NOA 61) of 100 nm. Secondly, a thickness of 100 nm polyvinyl alcohol (PVA) was spin-coated on the crystals, following a SU-8 2025 photoresist of  $2 \mu\text{m}$  thickness. Thirdly, desired patterns were transferred to the SU-8 photoresist layer by a standard photolithography, and then the cured SU-8 photoresist served as mask during the RIE process. Finally, the remaining PVA and SU-8 were removed from the crystal surface and PDMS with thickness of  $20 \mu\text{m}$  is placed on the crystal microdisk and cured in a nitrogen atmosphere on a hotplate at  $90^\circ\text{C}$  for 4 h.

### B. Laser Characterization

Micro-luminescence spectra were resolved by a home-build micro-photoluminescence ( $\mu\text{-PL}$ ) system, with which individual BP1T microdisk resonators were investigated, as shown in Fig. 1(b). The second harmonic of a regenerative amplifier (Spitfire, Spectra Physics), which produced laser wavelength of 400 nm with a frequency of 1 kHz, was used as pump source. The pump beam was focused onto the sample with a spot size

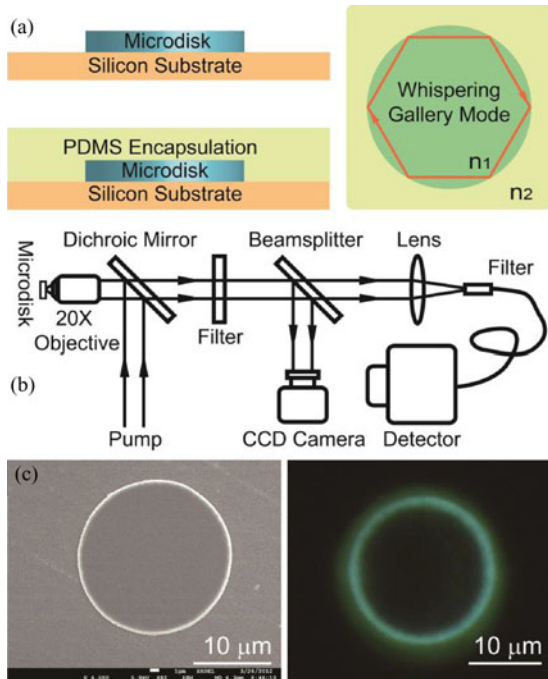


Fig. 1. (a) Schematic diagram of fabricated BPIT crystal microdisk without and with PDMS encapsulation in a nitrogen atmosphere. Light is reflected at the boundary between the high ( $n_1$ ) and low ( $n_2$ ) refractive index media due to total internal reflection in WGM resonators. (b) Home-built  $\mu$ -PL system. (c) SEM image and fluorescence photograph of one fabricated BPIT crystal microdisk.

around 100  $\mu\text{m}$ , through an objective lens, to fully excite the crystal microdisks. The emission light from the single crystal was collected by the same objective, and then split into two parts: one was detected by a spectrograph (grating: 1200/mm, Shamrock 303, Andor) and the other was imaged by a CCD camera. The output power of the pump was controlled by neutral density filter.

### III. LASER CHARACTERISTICS

#### A. Laser Performance Comparison

The micro-photoluminescence spectra was firstly investigated in embedded crystalline microresonators. The power-dependent PL emission spectra for an embedded microresonator is displayed in Fig. 2. For excitation intensity below 100 nJ ( $1.2 \text{ mJ/cm}^2$ ), the emission from the central region of the device is peaked at about 496 nm with a broad spectrum. As the excitation intensity increases, sharp peaks located at around  $\approx 500 \text{ nm}$  emerge from the emission spectrum, with rapid increase in the emission intensity. A plot of intensity of peaks with excitation energy is shown in inset of Fig. 2. The threshold is extracted from the change of slope in the pump intensity dependence, and it is found to be  $\approx 120 \text{ nJ}$  ( $1.5 \text{ mJ/cm}^2$ ), which is comparable to the reported dye-doped microdisk laser operated in air [11]. Such a non-linear increase in emission intensity with regular comb-like spectrum confirm that the lasing action with strong optical resonate modes occurs in the embedded microcavity. When the lasing spectrum at different pump pulse energy was

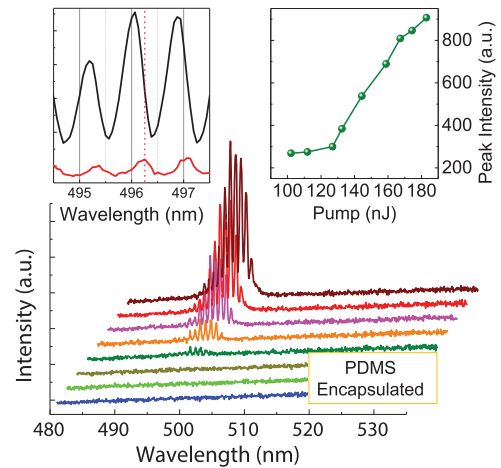


Fig. 2. Measured spectra of the lasing emission of the laser at different pump intensity. The inset (left) shows the enlarged spectrum at 130 and 175 nJ for comparison, and the right illustrate nonlinear dependence of the emitted intensity on the pump intensity, indicating a threshold of 120 nJ.

compared, it is interesting to note that the lasing modes slightly blue-shifted ( $\sim 0.2 \text{ nm}$ ), as shown in Fig. 2. The possible reason may be attributed to the change of cavity size and the radiation induced photo-oxidation [10].

In a WGM resonator, light inside the cavity is trapped due to total internal reflection, and forms a closed light-propagating loop in it. Thus, it demands that refractive index of resonator materials must be higher than that of environmental medium. Variation of refractive index contrast may impose a dramatic effect on the Q-factors and lasing performance. High refractive index contrast of the interface allows for strong feedback, while low index contrast with the surrounding medium causes heavy radiation loss. In order to evaluate the effect of package polymer (PDMS) on the lasing performance, the lasing properties of bared microlasers were studied in comparison with the encapsulated one. Fig. 3 displays the collected laser spectra from two different microlasers with same size. One is operated in air, while the other is embedded into PDMS, whose refractive index is about 1.42 [24]. Similar spectrum can be observed in these two devices. In WGM, lasing peaks are separated from each other, and the spectral spacing  $\Delta\lambda$  between two adjacent peaks is given by the equation:

$$\Delta\lambda = \lambda^2 / \pi n_{\text{eff}} D \quad (1)$$

where  $n_{\text{eff}}$  is the effective refractive index and  $D$  is the diameter of the microdisk. From the experiments, it is easily found that there exists slight difference in the spectral spacing. The  $\Delta\lambda$  for the disk in the air is 0.95 nm, while for the encapsulated, it is 1.11 nm. According to Eq. (1), the calculated effective refractive index for microdisk in air is about 4.92, which is higher than that of most of the organic semiconductors. The effective index decreases to 4.21 when it is embedded into PDMS, a value that is still relatively high, which indicates why the lasing emission still occurs though the environmental index is increased. Quality factor ( $Q$ ), an important parameter as a measure of energy losses in optical resonators, is further studied. From the experiment, the

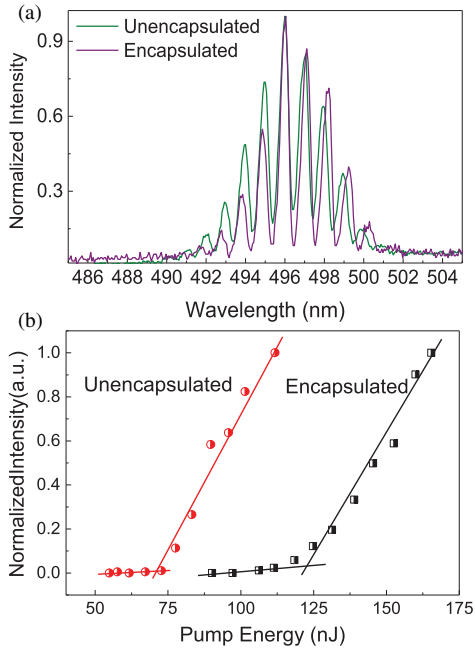


Fig. 3. (a) Laser spectra from two different microlasers (in air and encapsulated with PDMS) with same size. (b) Emission intensity *versus* pump energy for the unencapsulated and encapsulated laser.

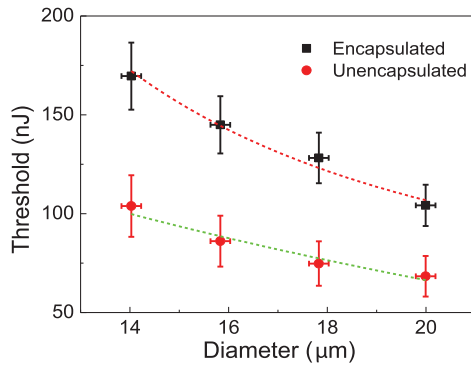


Fig. 4. Threshold comparison before (red circles) and after embedding (black squares) for different size crystal lasers.

$Q$  factor is estimated to be  $\approx 1171$  for microdisk in air, according to the definition  $Q = \lambda/\delta\lambda$ , where  $\lambda$  is the peak wavelength and  $\delta\lambda$  is the linewidth of the peak. The  $Q$  factor remains as high as 1066 after the encapsulation. It means that the light can be effectively trapped in single crystal cavity, and the WGM is well supported in cavity, though the index contrast is lowered. Meanwhile, there is no evidence of the difference in slope efficiency between the two lasers, as shown in Fig. 3(b).

In order to obtain more information about the lasing characteristics from the WGM cavity, a size-dependent lasing measurement was carried out on individual microdisks embedded and non-embedded, as summarized in Fig. 4. It is noted that, in both cases, the lasing threshold decreases with the increase in diameter of the microdisk, and the experimental data can be approximately fitted to a  $1/D^2$  function [25]. The increase

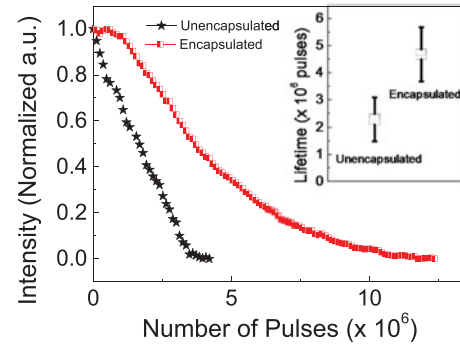


Fig. 5. Evolution of the peak intensity with the repeated pulses number of bare (black pentagram) and encapsulated (red squares) microdisk lasers. The inset is the operational lifetime of bare and encapsulated microdisk lasers.

in lasing threshold can be attributed to the smaller microdisk cavity volume and the roughness of the microdisk. A smaller diameter provides less spatial overlap between the WGM and the BPIT gain medium, resulting in more light leakage (optical loss) from the cavity. Similarly, the surface roughness of the microdisk is another important factor affecting the lasing threshold. The increase in the environmental index lead to decrease index contrast. It is normal to expect that the lasing threshold for the embedded lasers is higher than that of unencapsulated, because of increased light loss due to lowered index contrast. Fortunately, the lasing threshold is within reasonable range. As shown in Figs. 3(b) and 4, it is about 1.5 times higher for a microdisk with  $D \approx 14 \mu\text{m}$  in threshold.

### B. Device Photostability

As important photonic chip materials, the coverage of PDMS is expected to isolate the crystal resonator from the corrosion of oxygen and water. In order to assess the performance of the role of PDMS encapsulation, the operational lifetime of the BPIT microdisks was further tested, operating above the lasing threshold at about  $1.5 P_{th}$ . The emission was recorded as a function of time, while keeping the pump energy constant. Fig. 5 shows a typical evolution of the peak intensity with the repeated pulses number (1000 pulses in one second) from  $18 \mu\text{m}$  microdisks. The unencapsulated microdisk lasers operated in air over  $2 \times 10^6$  excitation pulses in average, corresponding to 0.5 h at 1 KHz, before decaying to  $1/e$  of their initial emission intensity. Upon encapsulating, the laser operation time was increased to  $4.6 \times 10^6$  excitation pulses, corresponding to more than 1.25 h, a value two times longer than that of bared lasers. For the microdisk with  $14\text{--}20 \mu\text{m}$ , we have observed similar improvement (around 2 times) in operational lifetime. This demonstrates the effectiveness of the encapsulation provided by the cover of PDMS. Taking into account the intensity of the employed excitation intensity, the estimated overall device operational lifetime could be further prolonged by optimizing the pump power to close to laser threshold.



#### IV. CONCLUSION

In summary, we have microfabricated on-chip microresonators of single crystals embedded in PDMS, and successfully achieved WGM lasing. The lasing performance of microdisk was very stable and not heavily affected by the cover medium of PDMS. The operational lifetime of encapsulated lasers increase by two times upon continuous pumping at excitation intensity above lasing threshold. These results suggest that the crystal microlasers show great potential as application for light sources in photonic chip.

#### ACKNOWLEDGMENTS

The authors gratefully acknowledge support from National Basic Research Program of China (973 Program) under Grants #2014CB921302 and #2011CB013003, and from Natural Science Foundation of China under Grant #51373064, #51335008, and #61378053. H. H. Fang is grateful for discussion with Sampson Adjokatse.

#### REFERENCES

- [1] V. S. Ilchenko and A. B. Matsko, "Optical resonators with whispering-gallery modes-part II: applications," *IEEE J. Sel. Top. Quantum. Electron.*, vol. 12, no. 1, pp. 15–32, Jan./Feb. 2006.
- [2] K. Kosma, G. Zito, K. Schuster, and S. Pissadakis, "Whispering gallery mode microsphere resonator integrated inside a microstructured optical fiber," *Opt. Lett.*, vol. 38, no. 8, pp. 1301–1303, Apr. 2013.
- [3] L. He, Ş. K. Özdemir, and L. Yang, "Whispering gallery microcavity lasers," *Laser Photon. Rev.*, vol. 7, no. 1, pp. 60–82, Feb. 2013.
- [4] H. H. Fang, R. Ding, S. Y. Lu, J. Yang, X. L. Zhang, R. Yang, J. Feng, Q. D. Chen, J. F. Song, and H. B. Sun, "Distributed Feedback Lasers Based on Thiophene/Phenylene Co-Oligomer Single Crystals," *Adv. Funct. Mater.*, vol. 22, no. 1, pp. 33–38, Jan. 2012.
- [5] H.-H. Fang, R. Ding, S.-Y. Lu, X.-L. Zhang, J. Feng, Q.-D. Chen, and H.-B. Sun, "Flexible lasers based on the microstructured single-crystalline ultrathin films," *J. Mater. Chem.*, vol. 22, no. 45, pp. 24139–24144, Sep. 2012.
- [6] H. H. Fang, J. Yang, J. Feng, T. Yamao, S. Hotta, and H. B. Sun, "Functional organic single crystals for solid-state laser applications," in *Lasers and Photonics Reviews*. New York, NY, USA: Wiley, 2014.
- [7] T. Grossmann, M. Hauser, T. Beck, C. Gohn-Kreuz, M. Karl, H. Kalt, C. Vannahme, and T. Mappes, "High-Q conical polymeric microcavities," *Appl. Phys. Lett.*, vol. 96, no. 1, pp. 013303–013303–3, Jan. 2010.
- [8] A. M. Armani, A. Srinivasan, and K. J. Vahala, "Soft lithographic fabrication of high  $Q$  polymer microcavity arrays," *Nano Lett.*, vol. 7, no. 6, pp. 1823–1826, May 2007.
- [9] V. D. Ta, R. Chen, and H. D. Sun, "Self-Assembled Flexible Microlasers," *Adv. Mater.*, vol. 24, no. 10, pp. OP60–OP64, 2012.
- [10] R. Chen and H. D. Sun, "Single mode lasing from hybrid hemispherical microresonators," *Sci. Rep.*, vol. 2, p. 224, 2012.
- [11] J.-F. Ku, Q.-D. Chen, R. Zhang, and H.-B. Sun, "Whispering-gallery-mode microdisk lasers produced by femtosecond laser direct writing," *Opt. Lett.*, vol. 36, no. 15, pp. 2871–2873, Jul. 2011.
- [12] H. H. Fang, R. Ding, S. Y. Lu, Y. D. Yang, Q. D. Chen, J. Feng *et al.*, "Whispering-gallery mode lasing from patterned molecular single-crystalline microcavity array," *Laser Photon. Rev.*, vol. 7, no. 2, pp. 281–288, Mar. 2013.
- [13] F. Sasaki, S. Kobayashi, S. Haraichi, S. Fujiwara, K. Bando, Y. Masumoto, and S. Hotta, "Microdisk and Microring Lasers of Thiophene-Phenylene Co-Oligomers Embedded in Si/SiO<sub>2</sub> Substrates," *Adv. Mater.*, vol. 19, no. 21, pp. 3653–3655, Nov. 2007.
- [14] L. Eldada and L. W. Shacklette, "Advances in polymer integrated optics," *IEEE J. Sel. Top. Quantum. Electron.*, vol. 6, no. 1, pp. 54–68, Jan./Feb. 2000.
- [15] M.-C. Estevez, M. Alvarez, and L. M. Lechuga, "Integrated optical devices for lab-on-a-chip biosensing applications," *Laser Photon. Rev.*, vol. 6, no. 4, pp. 463–487, Jul. 2012.
- [16] D. V. Plant and A. G. Kirk, "Optical interconnects at the chip and board level: challenges and solutions," *Proc. IEEE*, vol. 88, no. 6, pp. 806–818, Jun. 2000.
- [17] V. D. Ta, R. Chen, L. Ma, Y. J. Ying, and H. D. Sun, "Whispering gallery mode microlasers and refractive index sensing based on single polymer fiber," *Laser Photon. Rev.*, vol. 7, no. 1, pp. 133–139, 2013.
- [18] C. Vannahme, S. Klinkhammer, M. B. Christiansen, A. Kolew, A. Kristensen, U. Lemmer, and T. Mappes, "All-polymer organic semiconductor laser chips: Parallel fabrication and encapsulation," *Opt. Exp.*, vol. 18, no. 24, pp. 24881–24887, Nov. 2010.
- [19] P. Reusswig, S. Nechayev, M. Tomes, T. Carmon, M. Baldo, and C. Rotschild, "Efficient incoherent pumping for high-Q micro-lasers," in *Proc. Conf. Lasers Electro-Opt.*, San Jose, CA, USA, 2012, p. CTh4M.7.
- [20] L. Persano, A. Camposo, P. Del Carro, P. Solaro, R. Cingolani, P. Boffi, and D. Pisignano, "Rapid prototyping encapsulation for polymer light-emitting lasers," *Appl. Phys. Lett.*, vol. 94, no. 12, pp. 123305–1–123305–3, Mar. 2009.
- [21] S. Hotta, T. Yamao, S. Z. Bisri, T. Takenobu, and Y. Iwasa, "Organic single-crystal light-emitting field-effect transistors," *J. Mater. Chem. C*, vol. 2, no. 6, pp. 965–980, Jan. 2014.
- [22] S. Hotta and T. Yamao, "The thiophene/phenylene co-oligomers: exotic molecular semiconductors integrating high-performance electronic and optical functionalities," *J. Mater. Chem.*, vol. 21, no. 5, pp. 1295–1304, Oct. 2011.
- [23] J. Yang, H.-H. Fang, R. Ding, S.-Y. Lu, Y.-L. Zhang, Q.-D. Chen, and H.-B. Sun, "High-quality large-size organic crystals prepared by improved physical vapor growth technique and their optical gain properties," *J. Phys. Chem. C*, vol. 115, no. 18, pp. 9171–9175, Apr. 2011.
- [24] R. Chen, V. D. Ta, and H. Sun, "Bending-Induced Bidirectional Tuning of whispering gallery mode lasing from flexible polymer fibers," *ACS Photon.*, vol. 1, no. 1, pp. 11–16, 2014.
- [25] D. J. Gargas, M. C. Moore, A. Ni, S.-W. Chang, Z. Zhang, S.-L. Chuang, and P. Yang, "Whispering gallery mode lasing from zinc oxide hexagonal nanodisks," *ACS Nano*, vol. 4, no. 6, pp. 3270–3276, Apr. 2010.

**Shi-Yang Lu** received the B.S. degree in physics from the College of physics, Jilin University, Changchun, China, in 2011. He is currently working toward the Master's degree in State Key Laboratory on Integrated Optoelectronics. His current research interests include fabrication of organic single crystal laser devices.

**Hong-Hua Fang** obtained the B.Sc degree in electronic science and technology from Jilin University, Changchun, China, in 2006. He received the Master's degree from the laboratory of Prof. Hong-Bo Sun at Jilin University in 2009. His thesis work was focused on ultrafast dynamics and nonlinear optics of novel semiconductors. He received the Ph.D. degree in the same group in 2013. His thesis work mainly focused on optoelectronics of organic semiconductor and their applications in laser devices. After that, he joined the Photophysics and OptoElectronics group of the Zernike Institute for Advanced Materials, University of Groningen, Groningen, The Netherlands. His research interests include physics and devices based on emerging semiconductors materials.

**Jing Feng** received the B.S. and Ph. D. degrees in microelectronics and solid-state electronics from Jilin University, Changchun, China, in 1997 and 2003, respectively. She worked as a Postdoctoral Researcher at RIKEN, Japan, from 2003 to 2006. She joined Jilin University in 2006, where she is currently a Professor of College of Electronic Science and Engineering and State Key Laboratory on Integrated Optoelectronics. Her research interests include organic optoelectronic devices. She has published over 50 scientific papers in academic journals.

**Hong Xia** received the B.S. and Ph.D. degrees in chemistry from Jilin University, Changchun, China, in 2001 and 2006, respectively. She joined the faculty at Jilin University in 2006, where she is currently an Associate Professor of photoelectronics. Her research interest includes micro-nanofabrication by laser based on functional resins and polymers.

**Qi-Dai Chen** received the B.S. degree in physics from the University of Science and Technology of China, Hefei, China, in 1998, and the Ph.D. degree in the Institute of Physics, Chinese Academy of Sciences, Beijing, China, in 2004. He was doing Postdoctorate research at Osaka City University from 2005 to 2006. He is currently an Associate Professor with Jilin University. His current research interests include laser microfabrication, ultrafast laser spectroscopy, photochemistry, and photophysics.

**Tie-Qiang Zhang** received the Graduation degree from Department of Physics, Jilin University, Changshua, China, where he is currently a Professor of optical engineering. His research is focused on new technology research for optical imaging and detection.

**Hong-Bo Sun** (M'xx) received the Ph.D. degree from Jilin University, Changchun, China, in 1996. He worked as a Postdoctoral Researcher at the University of Tokushima, Tokushima, Japan, from 1996 to 2000, and then as an Assistant Professor in the Department of Applied Physics, Osaka University, Osaka, Japan. He became a Project Leader in 2001. In 2005, he became a Professor (Changjiang Scholar) at Jilin University. His research is focused on laser micronanofabrication, and its application in microoptics, micromachines, microfluids, and microsensors. Dr. Sun was awarded by the Optical Science and Technology Society in 2002, and won an Outstanding Young Scientist Award issued by the minister of MEXT (Japan) in 2006.