Filterless narrowband near-infrared Si photodetector

Guodan Wei (weiguodan@sz.tsinghua.edu.cn)
Tsinghua University https://orcid.org/0000-0003-2209-2789

Zhuhua Xu
Tsinghua-Berkeley Shenzhen Institute

Chuying Sun
University of Hong Kong

Miao He
Tsinghua-Berkeley Shenzhen Institute

Siyi Min
University of Hong Kong

Cong Zhao
Tsinghua University

Zhenghao Liu
Southern University of Science and Technology

Jingzhou Li

Wen-Di Li
University of Hong Kong https://orcid.org/0000-0002-7005-2784

Man-Chung Tang
Tsinghua University

Hongyan Fu
Tsinghua University https://orcid.org/0000-0002-4276-0011

Feiyu Kang
Tsinghua University

Jiangyu Li
Southern University of Science and Technology

Yang Shen
Tsinghua University

Article

Keywords: Si nanograting, organic thin film, photodetector, narrow-band, near-infrared, filter-less

Posted Date: March 28th, 2022

DOI: https://doi.org/10.21203/rs.3.rs-1480222/v1
Abstract

Spectrally selective narrowband Si photodetection is critical for near-infrared (NIR) medical imaging, optical communication, light detection and autonomous mobiles. For conventional inorganic semiconductors such as silicon with broadband absorption from visible until 1.1 µm, it remains challenging to achieve narrowband photodetection without integrating optical filters. We demonstrate the very first narrowest NIR silicon photodetectors without any bandpass filter, reaching the full-width-at-half-maximum of only 26 nm at 912 nm. The narrowband response is inherently attributed to the resonant enhancement effect of optical microcavities formed by patterned Si nanograting combined with the organic D18:Y6 blend film. The Conductive-Atomic Force Microscope (C-AFM), Kelvin Probe Force Microscope (KPFM) and transient absorption (TA) spectroscopy characterization indicate effective charge transfer between Si and organic layers, significantly contributing to ultrafast photoresponse of 74 µs. Therefore, this design concept opens up the new possibility of developing filter-less and effective low-cost NIR detection, enabling entirely new configurations of optical imaging devices with narrowband tunability for telecommunications and imaging applications.

Introduction

Narrowband photodetectors have attracted intensive attention for full-color imaging and visible-blind near-infrared (NIR) detection in image sensing\(^1\)–\(^3\), optical communication\(^4\)–\(^6\), robot vision\(^7\)–\(^9\) and autonomous mobiles for self-driving cars\(^10\)–\(^12\). There are commonly two possible strategies to realize high resolution and low-noise narrowband photodetection: the integration of additional optical components such as bandpass filter and optical cavity with the traditional broadband photodetectors; or the adoption of the truly narrowband photodetectors with very narrow NIR absorption active layers. These extra optical parts definitely increase the device complexity, leading to degradation of the color quality and constancy. In addition, the commercially available bandpass filters have very limited full-width at half-maximums (FWHMs) less than 100 nm in specific wavelength ranges.

It is growing interest to achieve narrowband NIR PDs on commonly used bulk semiconductor materials such as silicon which could further extend its applications in medical, industrial, communication as well as night-vision imaging and security. While it is possible to realize narrowband detection either by coupling selective optical filters\(^13\) to detect specific wavelength or adopting narrowband absorption materials\(^14\)–\(^17\), it still remains challenging to construct a narrow NIR imaging detector with traditional Si devices\(^12\),\(^18\). These filters could potentially increase the PD manufacture cost and architectural complexity, limiting pixel density and the quality of color sensing. Therefore, numerous efforts have been spent on exploring filterless narrowband PDs in recent years such as plasmonic enhanced absorption of particular wavelength, optical manipulation to control the sub-band enhanced light absorption and effective charge collection narrowing for internal quantum efficiency manipulation. For example, by sandwiching an organic BODIPY dye molecule with intrinsically versatile and narrowband absorption, a flexible filterless narrowband PDs with high gain has been obtained at 530 nm response peak with a 60
In addition, 2D layered perovskites were also as good candidate materials for narrowband photodetectors with sharp absorption attributed to natural 2D quantum-well structure. Another choice is to construct specials device structure such as optical microcavity to realize select-wavelength detection. The resonance microcavity was widely applied in the realization of very narrowband detection due to the resonance enhance absorption effect, which greatly enhance the optical field in the NIR range through cavity-enhanced charge transfer absorption. However, these types of microcavities were constructed by two reflective silver mirrors and the enhanced resonant NIR absorption is still limited by the photoactive donor and acceptor blend film. In contrast, based-on well-developed wet-etching technique, the optical microcavity could be directly fabricated on the silicon and other substrates, sensitive inorganic low-dimensional photodetectors have been demonstrated through improved light management and harvesting with SiO2 nanograting array or a Fabry-Pérot microcavity.

Herein, filterless narrowband NIR photodetector with ultrafast response of 74 μs successfully fabricated on nanograining silicon/organic (D18:Y6) bulk heterojunction devices. We demonstrate a 40-fold enhancement of the external quantum efficiency (EQE), with FWHM as narrow as 26 nm, reaching the narrowest peak to our best knowledge. The variation of the covered organic layer thicknesses could effectively tune the resonant peak from 892 nm to 953 nm. The calculation results simulated by COMOSL Physics demonstrate the response peak is strongly attributed to the resonance enhancement of the created optical microcavity inside the silicon nanograting channels covered with the organic blend thin film. The charge transfer analysis for the inside of D18:Y6 and interface of Si nanograting/D18:Y6 demonstrated the introducing of D18:Y6 film can promote carrier transfer and charge collection, contributing to enhanced photocurrent generation. We believe the integration of patterned Si/organic heterojunction could open a new window towards filter-less highly responsive narrowband photodetectors for NIR photodetection.

**Results And Discussion**

The device illustration shown in Fig. 1a consists of the Si substrate with nanograining and organic film including D18:Y6 mixture and BCP films. The film of D18:Y6 film and BCP are spin-coated on the Si nanograting to form vertical structure photodetector. Fig. S1 demonstrates the dark current can be decreased by 100 times after introducing of BCP, indicating efficiently suppressing leakage current. The size of nanograting is designed as period of 614 nm, width of intracavity and cavity wall of 246 nm and 368 nm and depth of 212 nm as shown in Fig. 1c. The reflection spectrum of Si nanograting has further verified the enhance absorption at 890 nm as shown in Fig. 1d, indicating strong optical interference effect resulted from periodic nanograting on Si surface. The etch processing in sequence has been described in Fig. S2. Figure 1b shows the illustration of energy level aligned with the transition path of photogenerated electrons and holes. The detailed process of device fabrication is shown in Fig. S3. Figure 1e shows the SEM images after D18:Y6/BCP coating on Si nanograting, and the morphology of patterned grooves of the thin organic layers. The corresponding cross section in the Fig. S4 exhibits the
closed microcavity which function as light-trapping structure to improve NIR photon harvesting, contributing to resonance enhancement effect at special wavelength for further narrow-band response. The Ag electrodes are thermal evaporation on BCP surface and Si nanograting substrate, respectively. Figure 1f exhibits the surface topography images of D18:Y6/BCP film with periodic grooves on the Si nanograting substrate by Atomic Force Microscope (AFM) testing which is consistent with SEM image in Fig. 1e, increasing light absorption area. The organic thin film of uniform and homogeneous composition is the key point for the photogenerated charge collection and migration. The surface of D18:Y6 film on Si substrate is characterized by AFM as shown in Fig. S5, indicating excellent film formation. The molecular structures of D18 and Y6 are shown in Fig. S6. The photoluminescence (PL) mapping of D18:Y6 film can be used to evaluate the uniform ratio of D18 and Y6, because the PL quenching of D18 will appear through the electrons of the excited state transferring from D18 to Y6, and different ratio of D18 and Y6 will exhibit PL quenching with different levels. The uniform PL intensity, position and FWHM of 645 nm after quenching as shown in Fig. S7, indicating the uniform distribution of D18 and Y6 for the organic bulk thin film.

Figure 2a shows the $I-V$ curves under dark and the incident illumination of 900 nm laser beam with 378 uW sweeping from −3 to 3 V. The $I-V$ curves exhibits the typical rectifying characteristic. Meanwhile, the control devices are fabricated and the $I-V$ curves are shown in Fig. S8a-c. To explore the performance advantage of Ag/Si-nanograting/D18:Y6/BCP/Ag structure device, the Ag/Si-nanograting/Ag, Ag/Si/D18:Y6/BCP/Ag and Ag/Si/Ag devices are conducted. The corresponding on/off ratios are shown in Fig. S8d, demonstrating the Ag/Si-nanograting/D18:Y6/BCP/Ag structure device exhibits prominent on/off ratio and higher than control devices by approximate 100 times at the same test condition. Figure 2b shows the current-time curve and response time including rise and decay time of 74/82 µs, demonstrating the ultrafast response speed. To further explore the photoresponse characteristic, the photocurrents dependence on wavelength with different thickness of D18:Y6 film are tested, demonstrating resonant sharp peak in Fig. 2c. The response peaks could be successfully modulated from 892 nm to 953 nm by reducing the organic layer thicknesses, indicating the facile tunability. To note, the FWHM can be further tuned down to 26 nm, reaching the narrowest response for Si-based NIR PDs as shown in Fig. 2d. The thickness of D18:Y6 film is tested by AFM as shown in Fig. S9. In addition, the EQE dependence on wavelength for the peak of 912 nm is taken out for comparison with the device of Ag/Si-nanograting/Ag and Ag/Si/D18:Y6/BCP/Ag as shown in Fig. 2e, indicating only Ag/Si-nanograting/D18:Y6/BCP/Ag photodetector exhibits the very narrowband detector with high EQE of response peak to 37%. The peak value is 40 times higher than the base line value for the pure Si nanograting device. For bare Ag/Si/D18:Y6 device without any nanograting, only weak photoresponse could be observed. Therefore, it is critical to construct the narrowest NIR PDs with the integration of the organic D18:Y6 layer with optimized thickness on well-patterned Si nanograting substrate, constructing a resonance enhancement platform for the excellent narrow-band detection. Meanwhile, the responsivity, and detectivity dependence on wavelength are calculated, and peak values are up to 275 mA/W and $6\times10^8$ Jones as shown in Fig. S10. The calculation methods of $EQE$, responsivity and detectivity are shown in Supporting section. And the detectivity, responsivity and $EQE$ of device with the D18:Y6
thickness of 53 nm and 256 nm are also shown in Fig. S11 and S12, which can be up to $9.5 \times 10^8$ Jones, 965 mA/W and 129%, respectively. The photocurrent mappings under bias voltage sweeping with different thickness of D18:Y6 are shown in Fig. S13, indicating the obvious narrowband response peak under different bias voltage. Figure 2f shows the photocurrent dependence on power of exciting light of 500 nm corresponding Ag/Si-nanograting/D18:Y6/BCP/Ag, Ag/Si-nanograting/Ag and Ag/Si/D18:Y6/Ag photodetector, respectively. The linear characteristic of Ag/Si-nanograting/D18:Y6/BCP/Ag photodetector with the highest linear dynamic region (LDR) of 94.2 compared to the control devices demonstrates efficient separation of electron-hole pairs in devices, further indicating high-performance photodetector.

To further analyze the situation of resonance enhancement and light trapping from optical microcavity, the $|E|^2$ intensity (electric field intensity) distributions in Si nanograting without and with the organic film are calculated through COMSOL. Figure 3a shows the illustration of light locating at Si nanograting surface and resonance. The calculation results suggests there is resonance enhance of 890 nm as shown in Fig. 3b. Then, the light trapping and resonance mode are analyzed according to the device structure and work situation. Figure 4a shows the illustration of light trapping and resonance situation in the microcavity, and the light is trapped and reflected repeatedly between the faces of bottom of intracavity and D18:Y6 film after penetrating the organic film. The calculated spatial profile of $|E|^2$ intensity corresponding to illustration of Fig. 4a is shown in Fig. 3c. The $|E|^2$ intensity distribution on the vertical section of intracavity area is stronger than that of the cavity wall, and the increased field intensities are mainly localized around the marginalized region of cavity wall, which exhibits the strong localization of light. These indicate the appearing of enhance absorption attributed to nanograting, and further exhibiting in the photodetection performance. The $|E|^2$ field intensity dependence on wavelength as shown in Fig. 3d indicates there is an enhancement peak located at 910 nm which corresponding to the response peak of device basically as shown in Fig. 2e. The analog calculation indicated the narrowband detector peak is attributed to the resonance enhancement of optical microcavity. In addition, the effect of resonance enhance is influenced by the compactness which means the distance of organic film and the bottom of the intracavity. The different thickness of D18:Y6 film will result the different compactness in intracavity, and further changing the size of microcavity. Through changing the size of microcavity, the wavelength of resonance enhance can be adjusted.29,40

To invenstigate the interface effect of Si/D18:Y6, the measured $I-V$ curve on the formed pure Si/D18:Y6 film device, exhibiting an apparent rectifying characteristic of P-N junction diodes as shown Fig. S14. To observe the micro zone state of periodic Si nanograting with D18:Y6 film, the Si nanograting/D18:Y6 thin film is scanned by AFM and corresponding KPFM as shown in Fig. 4b and c. The AFM mapping exhibits the white areas is the region of cavity wall (contacting with D18:Y6 film) and the dark region is intracavity combined with the SEM images in Fig. 1e. The corresponding KPFM mapping exhibits the difference of surface potential which can provide another perspective to analyze charge transfer at the interface of Si nanograting and D18:Y6 film. The Fermi level difference can be reflected by the potential difference, and the conversion process is shown in Supporting Information Section. The inserts show the height and corresponding potential dependence on distance which are extracted from the position of the white
dotted line in Fig. 4b and c, respectively. According to the scanning results and the calculation, the Fermi level of cavity wall region is the lower than that of intracavity region, indicating there are charge transfer at the area of cavity wall due to the lower conduction band energy level of p-type Si, and charge transfer results the formation of build-in electric field at the interface of cavity wall and organic film. The AFM and KPFM of control device with Ag/Si/D18:Y6/BCP/Ag show uniform topography and potential as shown in Fig. S15, indicating indispensable role of Si nanograting. According to the energy level marching, the electrons after exciting transfer from D18:Y6 film to Ag electrode through BCP, and the holes transfer from D18:Y6 film to Si nanograting, and to Ag electrode. The build-in electric field can facilitate photogenerated electron-hole pairs separation and decrease the recombination. On the other hand, the C-AFM of D18:Y6 film on Si nanograting is tested to investigate the photocurrent distribution under the bias voltage of -2 V with the light excitation. The AFM and corresponding light C-AFM mapping are shown in Fig. 4d and e, respectively. The position marked with green dotted line in AFM image locates at the region of intracavity, and the green dotted lines on C-AFM mappings refer corresponding positions with low current. Through the light C-AFM mapping combined with AFM mapping, the current from cavity wall region is higher than that of intracavity, demonstrating cavity wall region is more beneficial to the charge transfer to Ag electrode including dark and photocurrent due to the good contact between top of cavity wall and D18:Y6 film. So, the deposited D18:Y6 film could not only form a microcavity for optical resonant absorption, but also facilitating effective charge transfer generated from the Si nanograting substrate, followed by swift charge transport to Ag electrode. Due to the spatially separated electron-hole pairs at the interface between D18:Y6 and Si nanograting substrate, hole carriers could successfully transport through Si substrate to reach another Ag electrode, reaching effective photocurrent generation. The low current region is corresponding to the intracavity which can provide resonance enhancement, and the photogenerated carriers transfer through cavity wall to D18:Y6 film and reaches Ag electrode as shown the illustration of Fig. 4a

Figure 5a shows the absorption spectra of D18, Y6 and D18:Y6 film, indicating absorption of broad band from 500 to 870 nm. As the device structure in Fig. 4a, the light of 500 to 870 nm will be absorption by D18:Y6 film. The PL of neat D18, Y6 and their mixture are shown in Fig. 5b, exhibiting the PL quenching of D18 in the mixture film. The PL quenching demonstrates charge transfer, and the energy level marching further illustrates the photogenerated electron transfer from D18 to Y6. In addition, the transient absorption (TA) spectroscopy is used to investigate the charge transfer and recombination process in mixed film of D18:Y6. The 400 nm is selected as the wavelength of pump (1 KHz, 100 fs, around 1 µJ/cm²/pulse), and the probe range is 500 nm to 1200 nm as shown in Fig. 5c. The two absorption peaks (negative signal) in TA spectra located at 589 and 850 nm called ground state bleaching corresponding to the absorption of pristine D18 and Y6, respectively. The positive signal is from the emitting of high-energy state absorption from 830 to 1000 nm. The kinetic decay-associated spectra (DAS) process at 850 nm for Y6 and 589 nm for D18 are fitted to show the exciton lifetime of 22.9 ps for Y6 and 16.2 ps for D18, indicating the ultrafast charge transfer as shown in Fig. 5d.
Overall, the D18:Y6 film has played important roles to realize this ultrafast narrowest NIR photodetectors. The Si nanograting structure with periodic pattern has generated inherent optical diffraction effect, creating an obvious absorption peak at 890 nm whereas pure silicon has no such sharp absorption peak. But Si nanograting only device couldn't form an idealized device for narrow photoresponse at NIR peak, lacking of effective charge transfer layer. The construction of an optimized organic blend layer on top of the Si nanograting structure has provided an ideal platform, which it could not only form a microcavity for NIR optical resonant absorption, but also facilitating effective charge transfer generated from the Si nanograting substrate, followed by swift charge transport to Ag electrode. The ultrafast charge transfer at the interface between silicon and organic blend layer ensures the fast response of NIR.

**Conclusion**

In summary, the high-performance photodetector integrating on Si with narrowband with smallest FWHM of 26 nm is fabricated successfully, obtaining $EQE$ of 37% at response peak of 912 nm, and response time (rise and decay) is fast to 74/82 µs. Meanwhile, the NIR response peak could be successfully tuned from 890 nm to 950 nm by reducing organic thicknesses from 265 nm to 53 nm. The excellent performance is attributed to the spatial optical microcavity which could be geometrically formed between the intracavity of silicon nanograting and as-deposited organic blend layers. The well-formed heterojunction between silicon nanograting walls and organic layers could facilitate the charge separation upon excitation of the photogenerated NIR photons, and percolated charge transfer path of D18:Y6 organic blend layers for electron and hole collections, leading to forty fold enhancement of the photoresponse compared with silicon nanograting only devices. The well-designed microcavity structure realizes the narrowband detect by intracavity enhance and cavity wall transferring photogenerated carriers by blend organic film with ultrafast charge transfer. Our work will open up a promising road to realize filter-less narrowband NIR photodetector with ultrafast response based on silicon integrated optoelectronics.

**Experimental Section**

**Materials characterization**

The photoluminescence (PL) and Raman spectra were recorded on Microscopic confocal Raman spectrometer (Horiba Lab RAM HR800, America) at room temperature. The absorption spectra were taken from the HP 8453 spectrophotometer. The reflection spectrum is taken on the Ellipsometer (J.A.Woollam, M2000UI, USA). The atomic force microscopy (AFM) and Kelvin probe force microscopy (KPFM) was taken by Dimension Icon (Bruker Innova, Germany). The SEM images was taken from the field emission scanning electron microscope (JEOL-7401). The transient absorptions (TA) were measured in Helios pump-probe system (Ultrafast Systems LLC) combined with an amplified femtosecond laser system (Coherent), and under 400 nm excitation at 1 KHz, 100 fs, around 1 µJ/cm²/pulse.
**Si nanograting etching:** The silicon templates with uniform nano-gratings are fabricated through homemade interference lithography followed by inductively coupled plasma (ICP) etching, chrome (Cr) evaporation and lift-off process. Firstly, the silicon substrate is spin-coated with a 200 nm thick antirefection coating (ARC, AZ Barli II, Micro Chemicals) and baked at 200°C for 1 min. A layer of 200 nm thick positive photoresist (AZ MiR 701, Micro Chemicals) film is then spin-coated for interference lithography. After patterning the photoresist, the oxygen reactive ion etching (RIE) is used to etch the ARC layer and the photoresist pattern is then converted into Cr mask by depositing 30 nm-thick chrome using electron-beam evaporation. Then lift-off is performed with ultrasonic agitation in RCA-1 solution ($\text{NH}_3\cdot\text{H}_2\text{O}:\text{H}_2\text{O}_2:\text{H}_2\text{O} = 1:1:5$) at 75°C for 3 min. The Cr mask pattern is transferred into silicon with a 430 nm structure depth by CHF3/SF6 plasma in the ICP system with 68/10 sccm flow rates, a 5 mTorr pressure, a 150 W source power, and a 50 W bias power. Finally, after the removal of Cr mask, the fabrication of nano-structured silicon molds was completed. The silicon (p-type; electrical resistivity 7–9 $\Omega\cdot\text{cm}$; crystal orientation $<100>$) is purchased from Suzhou Jingsi Electronic Research Co., LTD. The size of nanograting is designed as period pattern of 614 nm (width of intracavity and cavity wall of 246 nm and 368 nm and depth of 212 nm.

**Device fabrication:** The D18 and Y6 as the mass ratio of 1:1.6 are dissolved in chloroform as 11 mg/ml. The mixed solution needs to be stirred and dissolved for 8 hours. The D18:Y6 film was spin-coated on the Si substrate with nanograting as 3000 rmp for 30 s. To get the different thickness of D18:Y6 film, the different concentration is used, and 5.5, 11, 22 mg/ml for 53, 123, 265 nm, respectively. the BCP is dissolved in isopropanol as 0.5 mg/ml and spin-coated on D18:Y6 film as 2000 rmp for 30 s. Finally, Ag electrodes were deposited by thermal evaporation of 300-nm-thick under $5\times10^{-4}$ Pa with deposition rate of 2 Å s$^{-1}$.

**Device test**

All I-V and current-time curves measurements are carried out with a home-build system at room temperature which consisted of the optical and electrical parts. The optical part includes SC-pro and AOTF-PRO produced by OYSL and can produce 430–1450 nm light which provide the illumination needed by photodetectors. The electrical part consists of probe and Keithley 2600B and can detect the photodetectors’ current. The fast current-time curves were measured using the semiconductor analyzing system and probe station at room temperature (Keithley 4200A-SCS, USA, Lake Shore, USA). Conductive-AFM-based measurement is performed on Asylum Research Cypher AFM with/without the built-in LED sheds light on the sample from the top under the bias voltage of -2 V.

**Declarations**

**Acknowledgements**

We thank funding support from National Natural Science Foundation of China (Grant Number: 52027817), Science and Technology Planning Project of Shenzhen Municipality (Grant number:...
JCYJ20200109144615514) and Scientific research startup funding (Grant number:01010600009).

**Author contributions**

G. D. Wei, Z.H. Xu, J. Z. Li, M. Tang, F.Y. Kang and W.D. Li conceived the ideas and coordinated the work. Z.H.Xu carried out the device fabrication and test. C.Y. Sun designed and fabricated silicon nanograting substrate. M. He conducted relative control experiments and tests. S. Min conducted the subsequent processing for the suitable Si nanograting. C. Zhao performed simulation optical field distribution of the PD. Thank for the help of Chuwei Liang, Kai Pu, Xubiao Li, Yuan Liu and Zijie Jiang for this research. G. D. Wei guided the experiment. All authors commented on the manuscript.

**Competing interests**

The authors declare no competing interests.

**References**


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Figures
Figure 1

(a) The illustration of device structure. (b) The illustration energy level marching. (c) The SEM of Si nanograting. Insert: The corresponding cross-section SEM image. (d) The reflection spectrum of nanograting Si substrate. (e) and (f) The SEM and AFM images of Si nanograting with D18:Y6 film, respectively.
Figure 2

(a) The I-V curves of Ag/Si-nanograting/D18:Y6 photodetector under exciting light of 900 nm and 378 µW. (b) The response time including rise and decay time. Insert: The current-time curve, The red region refers light on and another region refers light off. (c) The normalized photocurrent dependence on wavelength with different thickness of D18:Y6 film under the bias of -2 V. (d) Corresponding change of peak position and FWHM dependence on thickness of D18:Y6. (e) The EQE for the device of Ag/Si-nanograting/Ag, Ag/Si-nanograting/D18:Y6 (123 nm)/BCP/Ag and Ag/Si/D18:Y6 (123 nm)/BCP/Ag. (f) Corresponding photocurrent dependence on power of exciting light.
Figure 3

(a) and (b) The illustration and COMSOL calculation results of resonance enhancement in Si nanograting, respectively. (c) The view of analog calculation results by COMSOL for the resonance enhancement in the microcavity. (d) The resonance enhancement results dependence on wavelength.
Figure 4

(a) The illustration of resonance cavity and charges transport path. (b) and (c) The AFM and corresponding KPFM mapping of Ag/Si-nanograting/D18:Y6/BCP/Ag device, respectively. Inserts: The corresponding curves of height and potential dependence on distance extracted from the position marked with white dotted lines in (b) and (c), respectively. (d) and (e) The AFM, corresponding light C-AFM
mapping of Ag/Si-nanograting/D18:Y6/BCP/Ag device, respectively. The position marked with green dotted lines refer the same region in AFM and C-AFM images.

Figure 5

(a) and (b) The absorption and PL spectra of D18, Y6 and D18:Y6 film, respectively. (c) The TA spectra of D18:Y6 film. (f) The kinetic decay-associated spectra (DAS) process at 850 nm for Y6 and 589 nm for D18.

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