ISSN 1674-1056
http://cpb.iphy.ac.cn
http://iopscience.iop.org/cpb
中国物理 B
Chinese Physics
中国物理 Chinese Physics B

Synthesis of free-standing Ga_2O_3 films for flexible devices by water etching of $Sr_3Al_2O_6$ sacrificial layers

Xia Wang(王霞), Zhen-Ping Wu(吴真平), Wei Cui(崔尉), Yu-Song Zhi(支钰崧), Zhi-Peng Li(李志鹏), Pei-Gang Li(李培刚), Dao-You Guo(郭道友), Wei-Hua Tang(唐为华)

Citation: Chin. Phys. B . 2019, 28(1): 017305. **doi:** 10.1088/1674-1056/28/1/017305

Journal homepage: http://cpb.iphy.ac.cn; http://iopscience.iop.org/cpb

What follows is a list of articles you may be interested in

One-dimensional method of investigating the localized states in armchair graphene-like nanoribbons with defects

Yang Xie(谢阳), Zhi-Jian Hu(胡智健), Wen-Hao Ding(丁文浩), Xiao-Long Lü(吕小龙), Hang Xie(谢航)
\Chin. Phys. B . 2017, 26(12): 127310. **doi:** 10.1088/1674-1056/26/12/127310

Band structure and edge states of star-like zigzag graphene nanoribbons

Hong Liu(刘红)
Chin. Phys. B . 2017, 26(11): 117301. **doi:** 10.1088/1674-1056/26/11/117301

Electronic properties of SnTe-class topological crystalline insulator materials

Jianfeng Wang(王建峰), Na Wang(王娜), Huaqing Huang(黄华卿), Wenhui Duan(段文晖)
Chin. Phys. B . 2016, 25(11): 117313. **doi:** 10.1088/1674-1056/25/11/117313

Band gap anomaly and topological properties in lead chalcogenides

Simin Nie(聂思敏), Xiao Yan Xu(许霄琰), Gang Xu(徐刚), Zhong Fang(方忠)
Chin. Phys. B . 2016, 25(3): 037311. **doi:** 10.1088/1674-1056/25/3/037311

Dislocation-mediated creep process in nanocrystalline Cu

Mu Jun-Wei, Sun Shi-Cheng, Jiang Zhong-Hao, Lian Jian-She, Jiang Qing
Chin. Phys. B . 2013, 22(3): 037303. **doi:** 10.1088/1674-1056/22/3/037303

SPECIAL TOPIC — Photodetector: Materials, physics, and applications

Synthesis of free-standing Ga_2O_3 films for flexible devices by water etching of $Sr_3Al_2O_6$ sacrificial layers*

Xia Wang(王霞)[1,2], Zhen-Ping Wu(吴真平)[1,2], Wei Cui(崔尉)[1,2], Yu-Song Zhi(支钰崧)[1,2], Zhi-Peng Li(李志鹏)[3], Pei-Gang Li(李培刚)[1,2], Dao-You Guo(郭道友)[4], and Wei-Hua Tang(唐为华)[1,2,†]

[1] *Laboratory of Information Functional Materials and Devices, School of Science, Beijing University of Posts and Telecommunications, Beijing* 100876, *China*
[2] *State Key Laboratory of Information Photonics and Optical Communications, Beijing University of Posts and Telecommunications, Beijing* 100876, *China*
[3] *School of Materials Science and Engineering, University of Science and Technology Beijing, Beijing* 100876, *China*
[4] *Center for Optoelectronics Materials and Devices, Department of Physics, Zhejiang Sci-Tech University, Hangzhou* 310018, *China*

(Received 8 October 2018; revised manuscript received 7 November 2018; published online 30 December 2018)

Flexible electronic devices have attracted much attention due to their practical and commercial value. Integration of thin films with soft substrate is an effective way to fabricate flexible electronic devices. Ga_2O_3 thin films deposited directly on soft substrates would be amorphous mostly. However, the thickness of the thin film obtained by mechanical exfoliation method is difficult to control and the edge of the film is fragile and easy to be damaged. In this work, we fabricated free-standing Ga_2O_3 thin films using the water-soluble perovskite $Sr_3Al_2O_6$ as a sacrificial buffer layer. The obtained Ga_2O_3 thin films were polycrystalline. The thickness and dimension of the films were controllable. A flexible Ga_2O_3 solar-blind UV photodetector was fabricated by transferring the free-standing Ga_2O_3 film on a flexible polyethylene terephthalate substrate. The results displayed that the photoelectric performances of the flexible Ga_2O_3 photodetector were not sensitive to bending of the device. The free-standing Ga_2O_3 thin films synthesized through the method described here can be transferred to any substrates or integrated with other thin films to fabricate electronic devices.

Keywords: free-standing Ga_2O_3 thin film, crystalline, $Sr_3Al_2O_6$, flexible photodetector

PACS: 73.22.Pr, 67.30.hr, 71.20.Nr, 85.60.Gz **DOI:** 10.1088/1674-1056/28/1/017305

1. Introduction

Ga_2O_3, with a wide band gap of 4.2–5.3 eV, exhibits large breakdown field, high dielectric constant and Baliga's figure of merit.[1–4] Meanwhile, Ga_2O_3 can work steadily in extreme environment, such as high temperature, high electric field, and high energy radiation. Ga_2O_3 with such properties has a wide range of potential applications in high-power electronic devices, deep ultraviolet (DUV) solar-blind photodetectors, gas sensors, and transparent electrodes on optoelectronic devices.[5–13] Ga_2O_3 Schottky barrier diodes have a high breakdown voltage.[14] The β-Ga_2O_3 field-effect transistors (FETs) exhibits a large on/off current rations ($\sim 10^{10}$).[15] Rectifiers and enhancement-mode FETs based on Ga_2O_3 can be used under higher electric field than either SiC or GaN based devices.[16] DUV solar blind photodetectors have important civil and military applications, such as missile tracking, short-range secure communication, ultraviolet astronomy, ozone holes monitoring, corona detection, and so on.[17–20]

Bendable, lightweight, flexible, sensitive, and wearable, electronic devices are urgently needed in emerging technological applications.[21–24] Flexible and transparent two-dimensional (2D) materials have been employed and developed in electronic technologies, including wearable energy-harvesting systems, folding electronic devices, curved screen electronic devices, soft portable devices, and rollup displays flexible Ga_2O_3 thin film photodetectors.[17] The Ga_2O_3 thin film layer deposited directly on the flexible polymer materials was non-crystalline, and the amorphous films were unstable for long-term device applications.[25–30] The Ga_2O_3 thin film layer can be fabricated by integrating exfoliated Ga_2O_3 microlayers,[31] but the thicknesses and dimensions of the Ga_2O_3 film layer were uncontrollable with the mechanical exfoliation method. In addition, the edge of the film is not intact.

In this paper, a millimeter sized (5 mm × 8 mm) free-standing Ga_2O_3 thin film was synthesized, and the flexible Ga_2O_3 solar-blind UV photodetector was fabricated. The Ga_2O_3 thin film layer transferred to the flexible substrates in this research was crystalline and its thickness was controllable. In addition, the edge of the film was intact. The experimental results demonstrated that the bending of the film had no obvious influence on its electrical performance. This provides an effective transfer method to fabricate crystalline Ga_2O_3 thin

*Project supported by the National Natural Science Foundation of China (Grant Nos. 51572033, 51572241, 61774019, 61704153, and 11404029), the Fund of State Key Laboratory of IPOC (BUPT), and the Open Fund of IPOC (BUPT), Beijing Municipal Commission of Science and Technology, China (Grant No. SX2018-04).

†Corresponding author. E-mail: whtang@bupt.edu.cn

© 2019 Chinese Physical Society and IOP Publishing Ltd

film onto flexible substrates for applications in flexible and wearable electronics.

2. Experimental

The SAO buffer layer (BL) was deposited on silicon substrates by radio frequency (RF) magnetron sputtering, and then Ga_2O_3 thin film was grown on the SAO BL. The base pressure in chamber was 1×10^{-4} Pa, and the distance between SAO target (99.99% purity) and substrates was 6 cm. The substrates temperatures were 750 °C. The $Sr_3Al_2O_6$ (SAO)[32,33] BL was deposited under a working pressure of 0.4 Pa and RF power of 120 W. The Ga_2O_3 thin film was deposited under a working pressure of 0.8 Pa and RF power of 70 W. Then the Ga_2O_3/SAO bilayer thin films on the Si substrates was immersed into deionized water. The crystal Ga_2O_3 thin film separated from substrates when SAO film was etched in water.[31,33] Finally, the free-standing Ga_2O_3 thin film was transferred to the flexible substrates polyethylene terephthalate (PET). The crystal structure of the as-grown film was investigated by transmission electron microscopy (TEM). UV-visible (UV-vis) absorption spectrum was taken using Hitachi U-3900 UV-vis spectrophotometer. The current–voltage (I–V) of the Ga_2O_3 film-based detectors were measured by Keithely 2450 source meter. To test the electric properties of the flexible Ga_2O_3 film, interdigital Ti/Au electrodes were deposited on the film by radio frequency magnetron sputtering using shadow mask. The electrode fingers of shadow mask were 200 μm wide and 2800 μm long with a 200 μm spacing gap.

3. Results and discussion

Figure 1(a) is the flow chart of the process to fabricate free-standing Ga_2O_3 films on PET. Figure 1(b) shows the photograph of free-standing Ga_2O_3 thin film floating on the surface of the deionized water. A millimeter sized (5 mm × 8 mm) free-standing Ga_2O_3 thin film was obtained. Figure 1(c) is the optical microscope image of free-standing Ga_2O_3 thin film in the water, and it appeared that the edge of film is intact. The free-standing Ga_2O_3 thin film can be transferred to any substrates. A photograph of Ga2O membrane transferred onto the flexible PET is shown in Fig. 1(d).

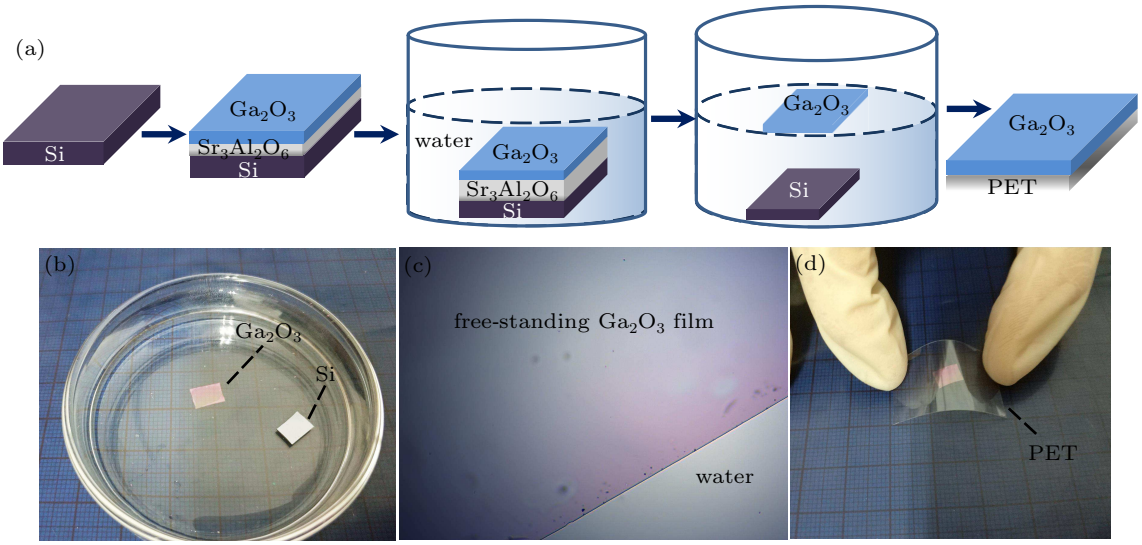


Fig. 1. (a) Schematics of the free-standing Ga2O thin film fabrication process. (b) Photograph of the free-standing thin film on the surface of the water. (c) An optical microscope image of the edge of free-standing Ga2O3 thin film. (d) An image of the device fabricated by transferring free-standing Ga2O3 film onto the PET substrates.

Figure 2(a) shows the structure and morphology of the Ga_2O_3 thin film characterized by transmission electron microscopy (TEM). The high-resolution TEM (HRTEM) image shows that the Ga_2O_3 thin film was polycrystalline. The polycrystalline structure was further confirmed by selected area electron diffraction (SAED) pattern in the illustration. Figure 2(b) shows the top SEM morphologies of the Ga_2O_3 thin film. It is obvious that the film was dense. The grain size of film was about dozens of nanometers. Figure 2(c) gives the ultraviolet–visible (UV) absorbance spectrum of the free-standing Ga_2O_3 thin film. The spectrum of the measured sample exhibits a sharp absorption edge at wavelength of ∼ 260 nm. The band gap of β-Ga_2O_3 thin film is about 4.75 eV obtained by fitting the extrapolating linear region. The plot $(ahv)^2$ versus hv is shown in the inset of Fig. 2(c), where h is Planck's constant, a is the absorption coefficient, and v is the frequency of the incident photon. Figure 2(d) shows the XPS spectrum with the binding energy in range from 1111 eV to

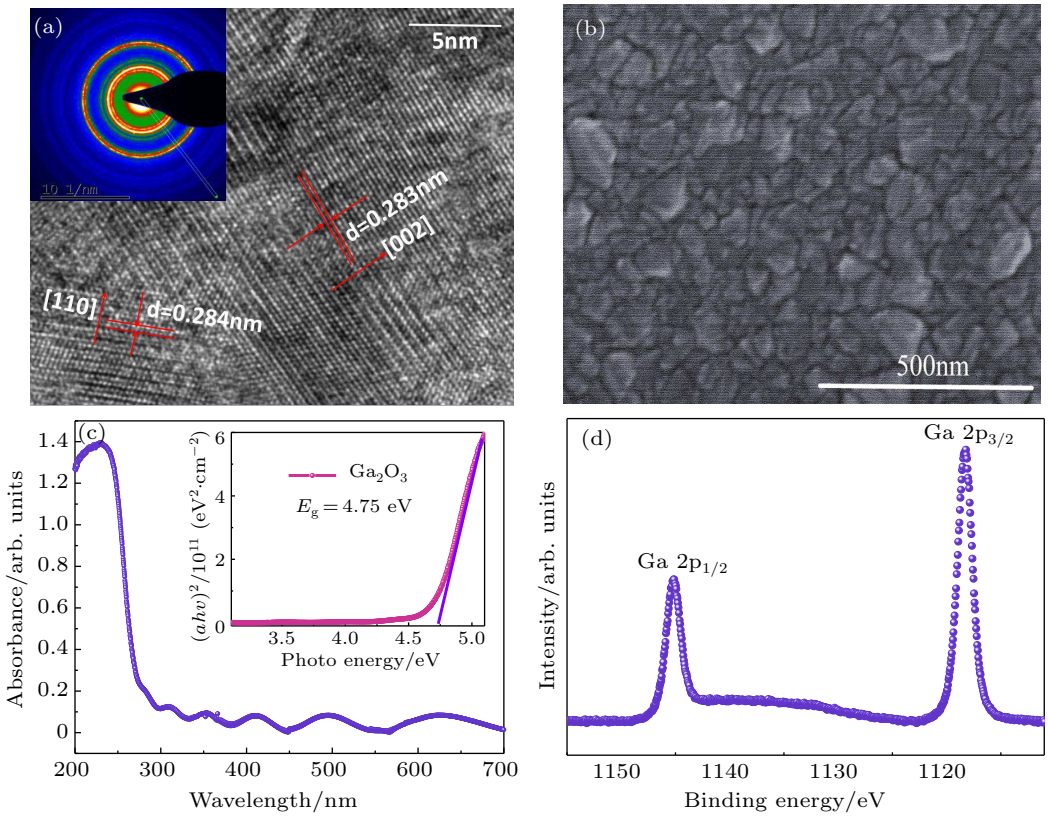


Fig. 2. (a) The HRTEM image of Ga_2O_3 thin film; inset: the corresponding SAED pattern. (b) The top surface view SEM image. (c) The UV absorbance spectrum of Ga_2O_3 thin film. (d) XPS spectra of Ga 2p core levels.

1155 eV. The energy peak of Ga $2p_{1/2}$ and Ga $2p_{3/2}$ are centered, which is attributed to the presence of gallium oxide and not gallium metal or others.

To investigate photoelectric properties of the film, flexible Ga_2O_3 based solar-blind UV photodetector was fabricated and a three-pair interdigital Ti/Au electrodes were deposited on top of the β-Ga_2O_3 thin film to construct a MSM structure device. The thickness of Ti layer and Au layer were 30 nm and 100 nm, respectively. The electrode fingers were 100 μm wide, 2800 μm long, 100 μm spacing gap, and 3 pair fingers. The 365 nm and 254 nm wavelength were used as light sources, respectively. The effective irradiated area was ~ 0.03 cm^2. A lamp with a center wavelength of 254 nm and 365 nm was used as a UV illumination source. In Fig. 3(a), *I*–*V* curve measured under the 365 nm light (0.5 mW/cm^2 intensity level on the sample surface) does not show significant difference to the *I*–*V* curve measured in dark, which suggests that the Ga_2O_3 thin film was not sensitive to 365 nm light. In contrast, the current shown a sharp jump when the device was exposed to 254 nm light. Figure 3(b) displays the *I*–*V* curves of the Ga_2O_3 film under rigid, bending upward and bending downward states. The curved Ga_2O_3 film was considered to be on a circumference of radius r, whose value implies the degree of bending. The curvature degree of Ga_2O_3 film were $r = \infty$, $r = 5$ mm, and $r = -5$ mm, respectively. Seen from Fig. 3(b), the Ga_2O_3 thin film exhibits almost the same electrical performance on the flexible PET, whether or not the film was bending. This implies that bending stress and bending direction had no influence on electrical performance of the films. The ignorable difference in these curves may be ascribed to the contact conditions between the Ga_2O_3 thin film and the electrodes. The insets shows the photographs of the flexible Ga_2O_3 thin film in the case of rigidity, bending upward and bending downward, respectively.

Figure 3(c) shows the time-dependent photoresponse performance of flexible Ga_2O_3 thin film under the case of rigid. The Ga_2O_3 thin film was tested under 254 nm light by on/off switching and with an applied bias of 5 V. The intensity level of 254 nm illumination source on the sample surface was 1 mW/cm^2. The results displayed a good repeatability, lower dark current and high photo-conductivity of the free-standing Ga_2O_3 film based device. The flexible Ga_2O_3 film based device yields a high level of signal-to-noise ratio under DUV illumination. Moreover, the device shows a short rise time and fall time under 5 V bias voltage. The quantitative analysis of the current rise and decay process involves the fitting of the photo-response curve with a exponential relaxation equation:[33]

$$I = I_0 + Ae^{-t/\tau}, \tag{1}$$

where I_0 is the steady state photocurrent, t is the time, A is constant, and τ is a relaxation time constant. In Fig. 3(c), the photoresponse processes are well fitted. τ_r and τ_d are the time constants for the rise edge and decay edges, respectively. For

the device, the current rise and fall time are estimated to be $\tau_r \approx 137$ ms and $\tau_d \approx 83$ ms. There are many reported results about Ga_2O_3 film based devices, and the response speed was in the range of 94.83 s[1]–1.5 ms.[19] The response speed of the device based on free-standing Ga_2O_3 film was faster than some reported results,[34–36] demonstrating its potential applications in optoelectronic device fabrication. Figure 3(d) shows the *I*–*t* photoresponse stability under illuminations of 254 nm light. With an incident light wavelength of 254 nm and an applied bias of 5 V, we found that the photoresponsivity of the photoelectric detector was 5×10^{-4} A/W. The light/dark current ratio of the photoelectric detector was approximately 100 with the 254 nm light intensity of 1 mW/cm^2. With the same growth parameters, the Ga_2O_3 film prepared directly on the Al_2O_3 substrate has better performance. The photoresponse of the prepared photodetector is 0.3 A/W[37] and dark current ratio is 16000.[38] There are some possible reasons that affect the performance of the device, e.g., the quality of film, the interface state of film/substrate, and the surface state of film. However, further study is necessary to improve the performance of free-standing Ga_2O_3 thin films based optoelectronic device.

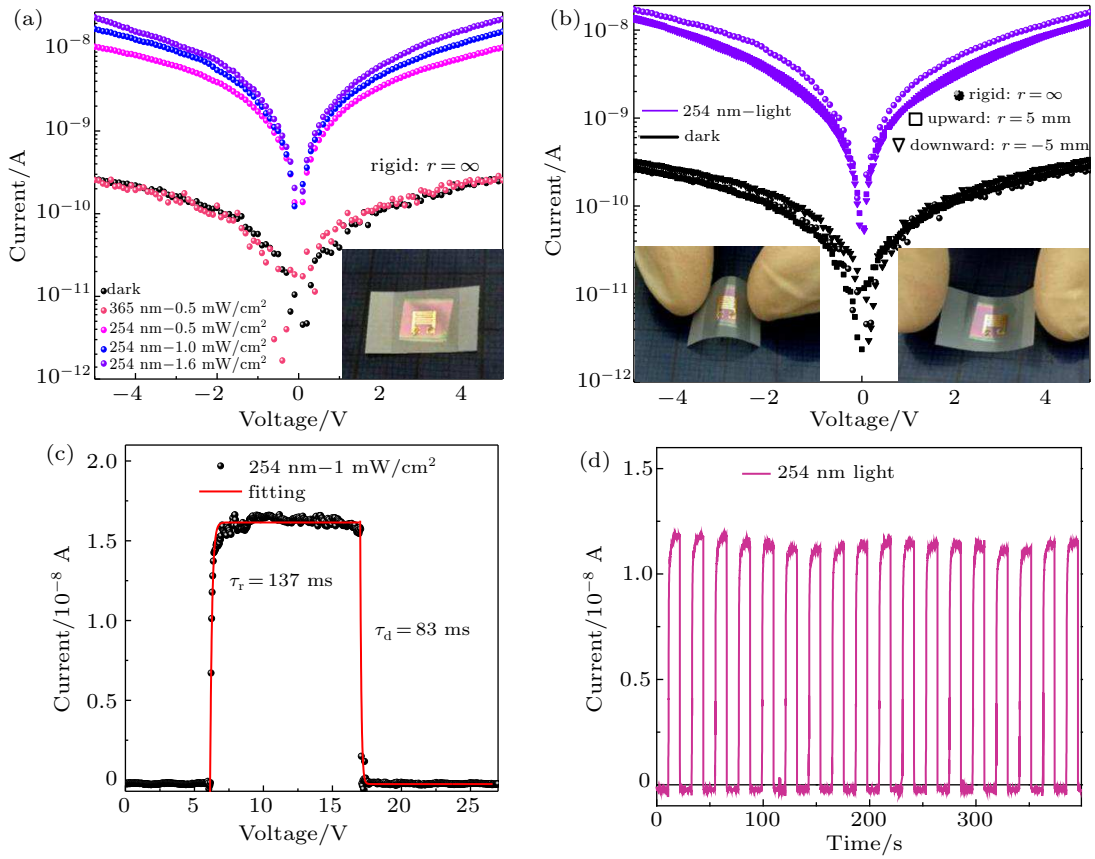


Fig. 3. *I*–*V* characteristics curves: (a) at different light intensity; (b) under the states of the rigid, bend upward, and bend downward. The insets show the photographs of the flexible Ga_2O_3 thin film at the rigid, bend upward, and bend downward. Time-dependent photoresponse: (c) experimental curve and fitted curve of the current rise and decay process to 254 nm illuminations under the state of rigid. (d) The *I*–*t* curve under 254 nm light illumination.

4. Conclusion

In summary, a free-standing Ga_2O_3 thin film has been fabricated using $Sr_3Al_2O_6$ as a sacrificial layer. The free-standing Ga_2O_3 thin film was polycrystalline, thickness was controllable, and the film edge was intact. Additionally, the photoelectric performances testing of the flexible Ga_2O_3 photodetector showed that the free-standing Ga_2O_3 thin film had a similar electrical performance whether bending or not, meaning no degradation after bending. The experimental results demonstrated that it was a practical way to fabricate free-standing Ga_2O_3 thin films by using $Sr_3Al_2O_6$ as a sacrificial layer. The free-standing Ga_2O_3 film can be used to fabricate flexible and wearable electronic devices by transferring it to flexible substrates for application.

References

[1] Kong W Y, Wu G A, Wang K Y, Zhang T F, Zou Y F, Wang D D and Luo L B 2016 *Ad. Mater.* **28** 10725

[2] Zhao B, Wang F, Chen H, Wang Y, Jiang M, Fang X and Zhao D 2015 *Nano Let.* **15** 3988

[3] Kokubun Y, Miura K, Endo F and Nakagomi S 2007 *Appl. Phy. Let.* **90** 031912

[4] Bilodeau R A, Zemlyanov D Y and Kramer R K 2017 *Adv. Mater. Interfaces* **4** 1600913

[5] Cui S J, Mei Z X, Hou Y N, Chen Q S, Liang H L, Zhang Y H, Huo W X and Du X L 2018 *Chin. Phys. B* **27** 067301

[6] Bhachu D S, Scanlon D O, Sankar G, Veal T D, Egdell R G, Cibin G, Dent A J, Knapp C E, Carmalt C J and Parkin I P 2015 *Chem. Mater.* **27** 2788

[7] Manandhar S and Ramana C V 2017 *Appl. Phys. Lett.* **110** 61902

[8] Guo D Y, Wu Z P, An Y H, Guo X C, Chu X L, Sun C L, Li L, Li P G and Tang W H 2014 *Appl. Phys. Lett.* **105** 023507

[9] Guo D Y, Wu Z P, Li P G, An Y H, Liu H, Guo X C, Yan H, Wang G F, Sun C L, Li L H snd Tang W H 2014 *Opt. Mater. Express* **4** 1067

[10] Higashiwaki M, Sasaki K, Murakami H, Kumagai Y, Koukitu A, Kuramata A, Masui T and Yamakosh S 2016 *Semicond. Sci. Technol.* **31** 034001

[11] An Y H, Zhi Y S, Cui W, Zhao X L, Wu Z P, Guo D Y, Li P G and Tang W H 2016 *Appl. Phs. A-mater.* **122** 1036

[12] Kim M, Seo J H, Singisetti U and Ma Z 2017 *J. Mater. Chem. C* **5** 8338

[13] Higashiwaki M, Kuramata A, Murakami H and Kumagai Y 2017 *J. Phys. D: Appl. Phys.* **50** 333002

[14] Konishi K, Goto K, Murakami H, Kumagai Y, Kuramata A, Yamakoshi S and Higashiwaki M 2017 *Appl. Phys. Lett.* **110** 103506

[15] Zhou H, Si M, Alghamdi S, Qiu G, Yang L and Ye P D 2017 *IEEE Electron Device Lett.* **38** 103

[16] Zhou H, Maize K, Qiu G, Shakouri A and Ye P D 2017 *Appl. Phys. Lett.* **111** 092102

[17] Hou X, Liu B, Wang X, Wang Z, Wang Q, Chen D and Shen G 2013 *Nanoscale* **5** 7831

[18] Oh S, Kim C K and Kim J 2018 *ACS Photonics* **5** 1123

[19] Lin R C, Zheng W, Zhang D Zhang Z J, Liao Q X, Yang L and Huang H 2018 *ACS Appl. Mater. & Inter* **10** 22419

[20] Liu X Z, Yue C, Xia C T and Zhang W L 2016 *Chin. Phys. B* **25** 017201

[21] Xiong Z N, Xiu X Q, Li Y W, Hua X M, Xie Z L, Chen P, Liu B, Han P, Zhang R and Zheng Y D 2018 *Chin. Phys. Lett.* **35** 058101

[22] Zhu H L, Fang Z Q, Preston C, Li Y Y and Hu L B 2014 *Energy & Environmental Science* **7** 269

[23] Yao B, Huang L, Zhang J, Gao X, Wu J, Cheng Y, Xiao X, Wang B, Li Y and Zhou J 2016 *Adv. Mater.* **28** 6353

[24] Jin J, Lee D, Im H G, Han Y C, Jeong E G, Rolandi M, Choi K C and Bae B S 2016 *Adv. Mater.* **28** 5141

[25] Zhang Y, Mei Z, Cui S, Liang H, Liu Y and Du X 2016 *Adv. Electron. Mater.* **2** 1500486

[26] Cui S J, Mei X, Zhang Y H, Liang L and Du L 2017 *Adv. Opt. Mater.* **5** 1700454

[27] Lu S, Qi J, Liu S, Zhang Z, Wang Z, Lin P, Liao Q, Liang Q and Zhang Y 2014 *ACS Appl. Mater. Interfaces* **6** 14116

[28] Hu P, Wang L, Yoon M, Zhang J, Feng W, Wang X, Wen Z, Idrobo J C, Miyamoto Y, Geohegan D B and Xiao K 2013 *Nano Lett.* **13** 1649

[29] Shrestha N K, Bui H T, Lee T and Noh Y Y 2018 *Langmuir* **34** 4575

[30] Liu C P, Ho C Y, Dosreis R, Foo Y, Guo P F, Zapien J A, Walukiewicz W and Yu K M 2018 *ACS Appl. Mater. Interfaces* **10** 7239

[31] Liu N, Fang G, Zeng W, Zhou H, Cheng F, Zheng Q, Yuan L, Zou X and Zhao X 2010 *ACS Appl. Mater. Interfaces* **2** 1973

[32] Lu, Baek D, Hong S, Kourkoutis L, Hikita and Hwan H 2016 *Nat. Mater.* **15** 1255

[33] Baek D, L, Hikita, Hwan H and Kourkoutis L F 2017 *ACS Appl. Mater. Interfaces* **9** 54

[34] Yang C, Liang H W, Zhang Z Z, Xia X C, Tao P C, Chen Y P, Zhang H Q, Shen R S, Luo Y M and Du G T 2018 *RSC Adv.* **8** 6341

[35] Alema F, Hertog B, Ledyaev O, Volovik D, Thoma G, Miller R, Osinsky A, Mukhopadhyay P, Bakhshi S, Ali H and Schoenfeld W V 2017 *Phys. Status Solidi* **214** 1600688

[36] Kong W Y, Wu G A, Wang K Y, Zhang T F, Zou Y F, Wang D D and Luo L B 2016 *Application. Adv. Mater.* **28** 10725

[37] Peng Y K, Zhang Y, Chen Z W, Guo D Y, Zhang X, Li P G, Wu Z P and Tang W H 2018 *IEEE Photon. Technol. Lett.* **30** 993

[38] Wang X, Chen Z W, Guo D Y, Zhang X, Wu Z P, Li P G and Tang W H 2018 *Opt. Mater. Express* **8** 2918