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Topical Review

Recent progress in piezo-phototronics with extended materials, application areas and understanding

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Abstract

The coupling effect between different properties in a controllable manner is of great interest for getting full understanding of materials, exploring novel physical phenomena and developing new applications. Piezo-phototronics is an emerging field which explores the three-way coupling among mechanical, optical and electrical properties in materials with noncentrosymmetric crystal structure. It has demonstrated its capability to work on different kinds of optoelectronic devices and can be utilized in various applications. Here, we will give a brief review of the tremendous progress, focusing on extended materials, application areas and understanding in the last two years. New material systems including quantum wells, two-dimensional materials and alloys have been investigated in this field. Novel applications in sonophotocatalysis, flexible electronics, photoluminescence and plasmonics have emerged. Meanwhile, temperature dependence of the piezo-phototronic effect was studied for the first time to gain in-depth understanding of the fundamental physics behind the phenomenon. All of this progress demonstrates the power of this three-way coupling effect in extended fields and shows the growing broader interest in different research areas.

Keywords: piezo-phototronics, three-way coupling, controllable modulation

(Some figures may appear in colour only in the online journal)

1. Introduction

Since it was introduced in 2010 [1, 2], the field of piezo-phototronics has attracted more and more research interest and has very rapidly developed with great efforts in the past several years. Piezo-phototronics focuses on the three-way coupling of mechanical, optical and electrical properties in materials with noncentrosymmetric crystal structure. Previously it has been demonstrated that strain induced localized polarization charges can effectively modulate/control carrier generation, separation, transport, and/or recombination by modifying the local electrical field distribution in the vicinity of a metal–semiconductor (M–S) junction and homo-/heterojunction. Different processes have been investigated to show the capability of the three-way coupling effect, including photodetection [3–8], photovoltaics [9–12], electroluminescence (EL) [13–18], photoelectrochemistry [19, 20], etc. Previous results demonstrated that, by optimizing built-in electric field to promote separation of photogenerated carriers, the sensitivity/response of a photodetector, the conversion efficiency of a solar cell and the performance of photoelectrochemical photoanodes can be enhanced greatly by the piezo-phototronic effect. On the other hand, this effect can also be used to control EL properties via adjusted carrier injection efficiency or carrier recombination efficiency by modifying local electrical field at related interfaces. Its controllable nature makes the piezo-phototronic effect versatile in various mechanical, optical and electrical coupled processes. As the understanding of the effect has developed, researchers with diverse backgrounds have put great efforts into new material systems and novel application areas which have emerged in the last two years. In this article, these progresses will be reviewed from three aspects. First we will focus on progress which utilizes the piezo-phototronic effect to optimize carrier generation and the separation process, including the demonstrated enhancement of photoresponse in new material systems and applications in new areas. Then we move on to progress related to adjusting the carrier recombination process, including the EL, photoluminescence (PL) and plasmonic effect associated with new device platforms. Finally, the temperature dependence of the piezo-phototronic effect is reviewed for further understanding of the fundamental physics. At the end, perspectives of this three-way coupling effect are presented.

2. Optimizing carrier generation and separation process

In the general photoelectric process, electrons and holes are generated under light illumination when the energy of the incident photons is equal to or larger than the band gap of the semiconductor. Only carriers collected by electrodes are effective for photodetection or energy conversion. Similarly in the photoelectrochemical process, only the photoexcited carriers diffused onto the photoelectrode surface or photocatalyst surface can be used to perform electrochemical reduction reactions. Thus many attempts have been made to enhance the carrier separation efficiency in order to promote the performance of photodetectors/solar cells and the activity of photocatalysts [21–26]. Previously the piezo-phototronic effect has been demonstrated to take this responsibility to work on devices constructed with traditional piezoelectric material [3–12, 19, 20]. In the last two years, some new material combinations have been investigated, such as ZnO/ZnS core/shell nanowires [27, 28], Au–MgO–ZnO metal–insulator–semiconductor structures [29], $Mg_xZn_{1-x}O$/Si p-n junctions [30], ZnO/spiro-MeOTAD heterojunctions [31], etc. In the case of $Mg_xZn_{1-x}O$, ZnO thin film is alloyed with Mg to investigate how the alloying process and Mg content work on the piezoelectric coefficient and thus how the corresponding piezo-phototronic effect affects the performance improvement of photodetectors. Apart from these, new device platforms have been introduced by utilizing this effect. Optical-fiber-ZnO/CdS nanowire hybridized structure was adapted to show enhanced sensitivity and photoresponsivity in UV/visible photosensing with the piezo-phototronic effect [32]. In another case, asymmetric metal contacts were used on freestanding GaN membrane to construct metal–semiconductor–metal (M-S-M) structure with enhanced on/off ratio and sensitivity as a photoswitch by utilizing this effect [33]. However, the most interesting progress is the demonstration of the piezo-phototronic effect on single-atomic-layer MoS_2 [34].

In 2014, experimental results showed that thin MoS_2 flakes with an odd number of atomic layers had piezoelectric properties and a strong piezotronic effect was demonstrated in single-layer MoS_2 [35]. Recently, the piezoelectric polarization charges created at the metal–MoS_2 interface have been used to modulate the separation/transport of photogenerated carriers. In this work, mechanical exfoliated single-layer MoS_2 flakes were transferred onto a polyethylene terephthalate (PET) substrate, as shown in figure 1(a). Cr/Pd/Au electrodes were deposited to make the metal–MoS_2 interface parallel to the 'zigzag' direction of MoS_2. To investigate the piezo-phototronic process, a laser beam with a wavelength of 442 nm was illuminated on the device when strain was introduced in MoS_2 by a bending substrate. Photocurrent was measured under systematically tuned optical illuminations and mechanical strains, as shown in figure 1(b). It is clear that at a fixed strain condition photocurrent in MoS_2 increases as the illumination power density is increased. But if the illumination condition is fixed, the relationship between the photocurrent and the strain is not in such a monotonic form. Figure 1(c) shows three typical conditions. First, when there is no light illumination, the dark current in the device decreases with increasing compressive strain and increases with increasing tensile strain. While when laser illumination is on, maximum photocurrent is reached under different compressive strains for different illumination intensities. To discuss the working mechanism, the result obtained under illumination intensity of $4.29\ \mathrm{mW\ cm^{-2}}$ is enlarged in figure 1(d). Electron–hole pairs are generated under illumination in MoS_2. When there is no strain applied (1 in figure 1(d)), the built-in electric field at the Schottky contact area separates the electrons and holes. The efficiency is limited and highly dependent on the barrier characteristics. When a mechanical strain is applied to the device (2–4 in figure 1(d)), induced piezoelectric polarization charges at the zigzag edges can affect the metal–MoS_2 contacts, which modify the Schottky barriers' height and width, and thus tune the optoelectronic process. There should be an optimized local electrical field distribution to get the highest electron–hole separation efficiency [36, 37], at which the highest photocurrent is obtained (3 in figure 1(d)). Under different illumination intensities, this situation is satisfied with different strain conditions. An optimized photoresponsivity of $2.3 \times 10^4\ \mathrm{A\ W^{-1}}$ is obtained in flexible optoelectronics based on monolayer MoS_2, which is the highest number for monolayer MoS_2 phototransistors. This is the first demonstration of the piezo-phototronic effect on two-dimensional atomically thin materials. In another work, the photoresponsivity of a high performance MoS_2 field-effect transistor was enhanced by the piezo-phototronic effect via a bent GaN nanowire used as the local gate to introduce piezopotential for tuning [38]. These studies may enable the development of flexible nanooptoelectromechanical systems, adaptive biooptoelectronic probes and ultrathin optoelectronics.

Another important development that should be mentioned is the emerging application of sonophotocatalysis [39].

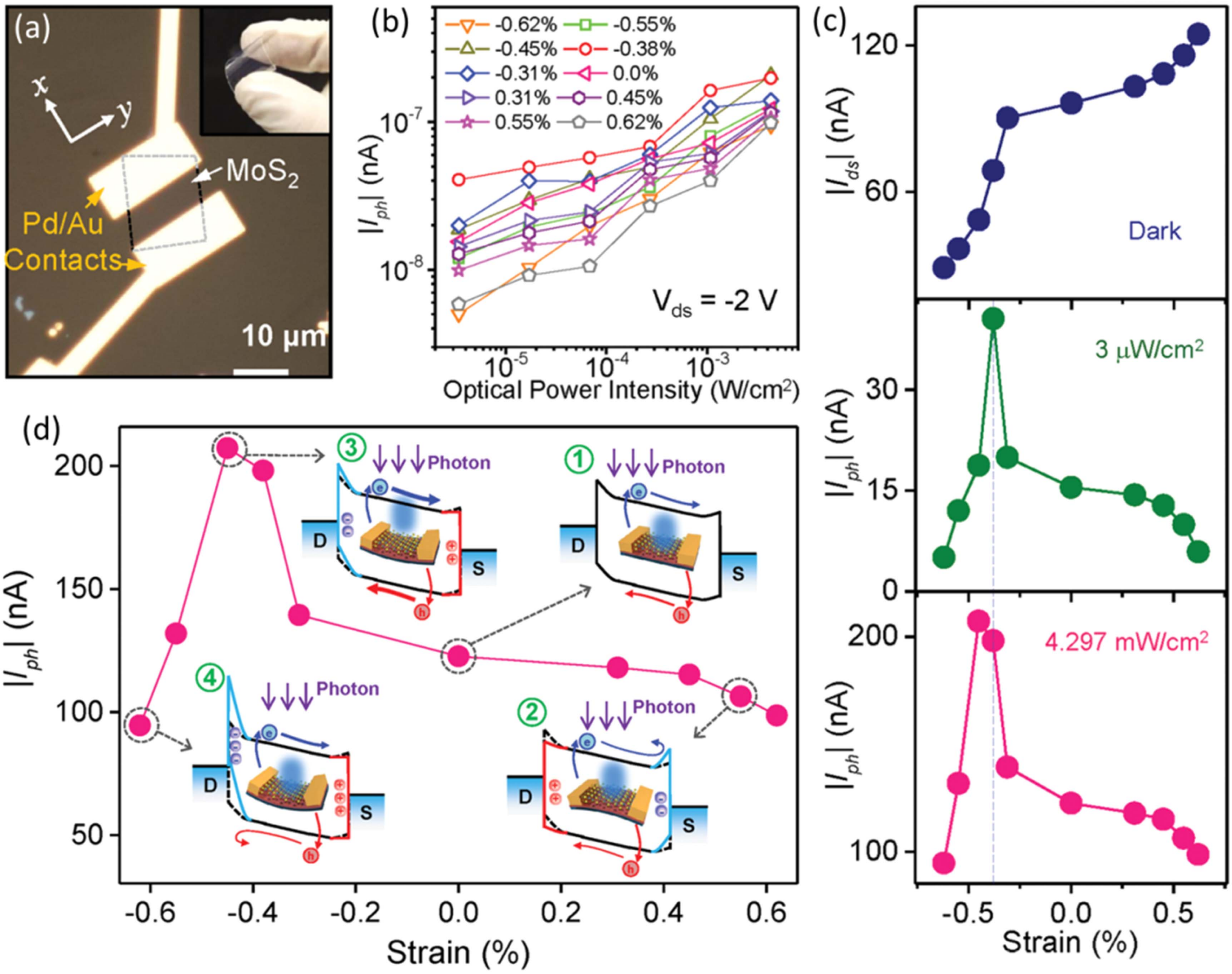

Figure 1. (a) Optical image of a flexible phototransistor constructed with monolayer MoS_2 on a PET substrate. The inset shows the flexibility of the device. (b) Change in photocurrent by strain under different illumination intensities. (c) Strain dependence of the dark current (top) and photocurrent (middle and bottom). (d) Working mechanism of the piezo-phototronic response in a single layer MoS_2 phototransistor. Reproduced with permission from [34] John Wiley & Sons. [© 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim].

Generally photoinduced carrier recombination in semiconductor photocatalysts is a big issue for low photocatalytic activity. Heterostructure can be used to introduce a built-in electric field in photocatalyst particles to promote photoinduced carrier separation [25, 26]. But a static field can be easily saturated by free carriers in the space. Thus photocatalysis enhancement will vanish [40, 41]. In this work, an Ag_2O-$BaTiO_3$ hybrid photocatalyst was synthesized as shown in figure 2(a). Ag_2O nanoparticles (NPs) were assembled on $BaTiO_3$ nanocubes by chemical precipitation under ultrasonic irradiation. Ag_2O NPs work as the photocatalyst. There is a built-in electric field in the ferroelectric $BaTiO_3$ nanocube because of its spontaneous polarization. This built-in field can be periodically altered through an ultrasonic wave utilizing the piezoelectric field generated in $BaTiO_3$ to avoid the saturation problem we mentioned above. The mechanism is illustrated in figure 2(b). Therefore, enhanced carrier separation by the built-in field can be preserved with the help of ultrasonic irradiation. Accordingly, the enhancement of photocatalytic performance of Ag_2O-$BaTiO_3$ hybrid photocatalyst can be maintained continuously. The photodegradation of Rh B under UV light irradiation with ultrasonic treatment was tested to compare the photocatalytic activity of Ag_2O and Ag_2O-$BaTiO_3$ hybrid nanostructure. Figures 2(c) and (d) are the absorption spectra of Rh B solution with different sonophotocatalysts. For Ag_2O, the sonophotocatalytic degradation rate of Rh B is only 90% in 2 h. However, for the $Ag_2O-BaTiO_3$ hybrid photocatalyst, the sonophotocatalytic degradation rate can reach 100% in the same time period. This shows that the utilization of the piezo-phototronic effect will provide a new strategy for high performance photocatalysis applications.

3. Adjusting the recombination process

Light-emitting diodes (LEDs) have attracted much scientific and commercial interest for their great application potential in

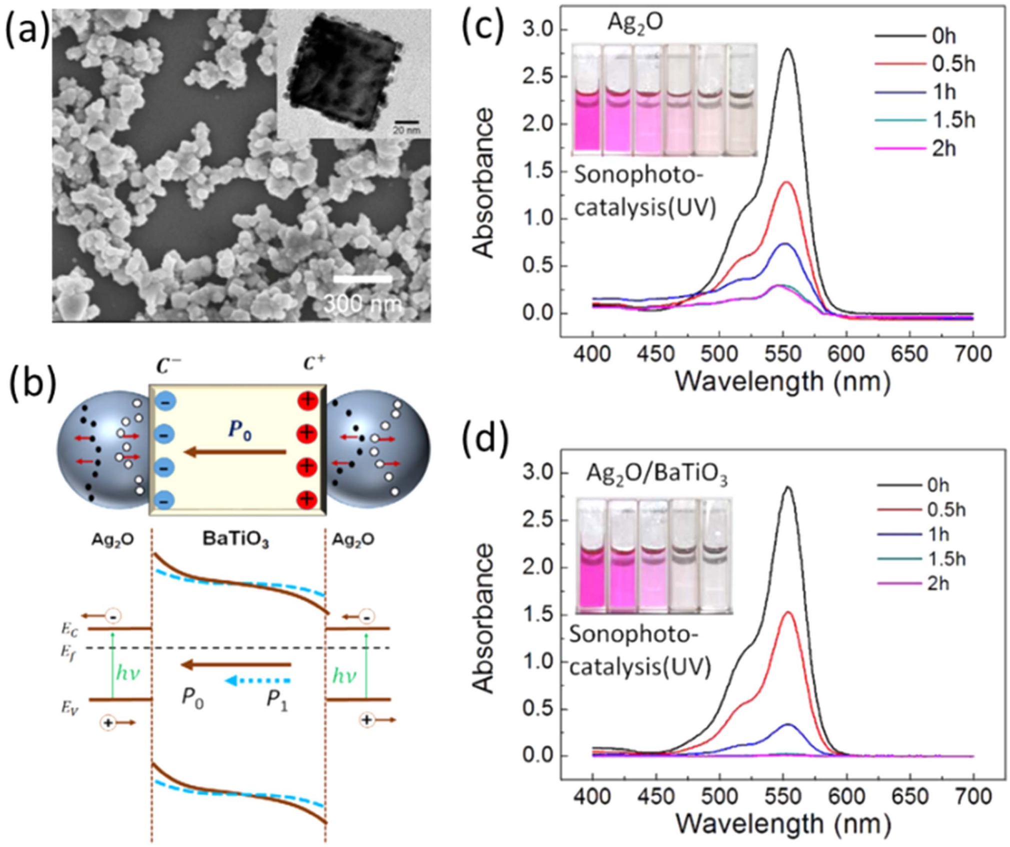

Figure 2. (a) Scanning electron microscopy (SEM) images of Ag_2O-$BaTiO_3$ hybrid nanocubes; the inset is a transmission electron microscopy (TEM) image. (b) Working mechanism of the piezo-phototronic effect on sonophotocatalysis of the hybrid structure. (c) The absorption spectra of Rh B solution with Ag_2O under UV light irradiation with ultrasonic treatment. (d) The absorption spectra of Rh B solution with Ag_2O-$BaTiO_3$ hybrid nanocubes under UV light irradiation with ultrasonic treatment. Reprinted with permission from [39]. Copyright 2015 American Chemical Society.

artificial lighting and displays. Many approaches have been made to enhance the light emission intensity, which currently is a big obstacle to real application [42–46]. Carrier recombination is a very important step in EL-related processes. The previously demonstrated piezo-phototronic effect on controlling EL properties by adjusting carrier injection or the recombination process has been further proved with different LED structures recently [47–49]. This three-way coupling among mechanical, optical and electrical properties also brings new LED applications in pressure mapping [16]. In recent progress, this application has been extended into a flexible platform [50]. An LED array composed of p-poly (3, 4-ethylenedioxythiophene)-polystyrenesulfonate (PEDOT: PSS) and ZnO nanowires (NWs) was fabricated on an ITO/PET substrate. Figure 3(a) is an optical image of a fabricated flexible device. As shown in figure 3(b), patterned ZnO NWs were first grown on the substrate. Then the inter-NW space was filled with SU8 to obtain good mechanical stability of the device. An Au film was deposited after spin-coating of PEDOT:PSS film to form a common top electrode with better conductivity. The pressure mapping performance was investigated by pressing an SU8 stamp with a convex character pattern of 'BINN' on the device. Figure 3(c) shows the EL images of the device under pressure of 0, 22, 44, 66 and 88 MPa, respectively. Crosstalk between adjacent pixels is hardly observed. An enhancement factor E of the LED intensity is introduced to evaluate how pressure works on the change of light emission intensity. It is defined as $E = I_p/I_0$, where I_0 and I_p are the intensities of the LED under zero and corresponding pressures, respectively. The relationship between enhancement factor and pressure of six devices is summarized in figure 3(d). It shows a linear dependence. The change of emission light intensity obtained from six adjacent NW-LEDs under different pressures is recorded in figure 3(e). It is clear that when the applied pressure increases the light emission intensity increases. The working mechanism is illustrated in figure 3(f) with the energy band diagram of the ZnO NW/p-polymer LED. The hole injection barrier at the interface is lowered under pressure which is induced by the inner-crystal piezo-potential. Therefore, the modified band structure increases the hole injection rate from PEDOT:PSS into ZnO and thus the recombination rate of carriers in ZnO NWs. This work also showed that the range of pressure measurement can be adjusted by varying the density and diameter of the ZnO NWs in each pixel via controlling the growth conditions of the ZnO NW arrays. Such devices are promising candidates for applications in electronic skins.

For pressure mapping, researchers have also demonstrated another approach by utilizing PL imaging with the

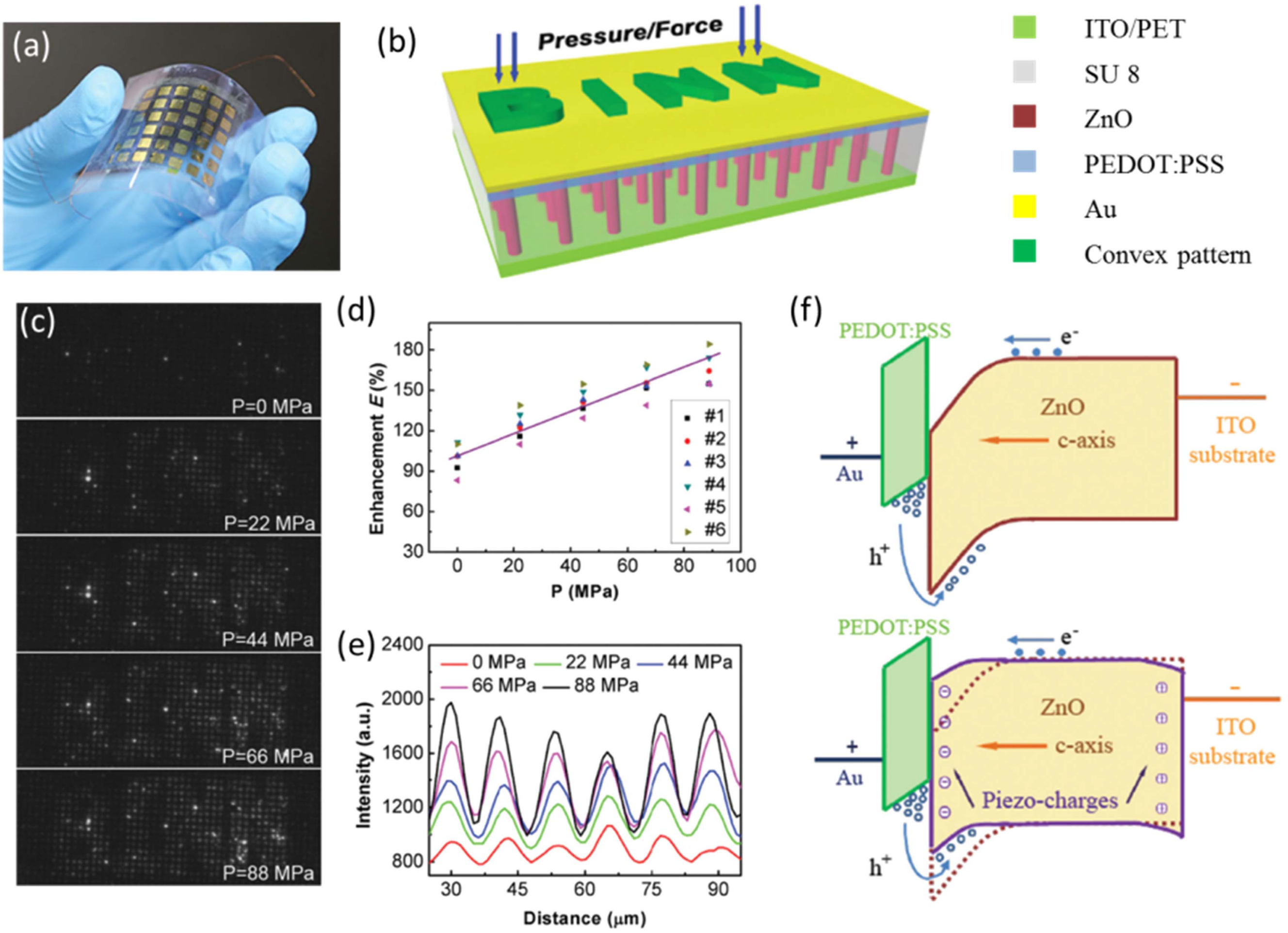

Figure 3. (a) Optical image of a fabricated flexible device with LED arrays for pressure mapping. (b) Schematic illustration of the NW-LED-based pressure sensor arrays. (c) EL images of the device under pressure of 0, 22, 44, 66, and 88 MPa, respectively. (d) Enhancement factor E of six NW-LEDs as a function of applied compressive pressure of up to 88 MPa. (e) The change of emission light intensity obtained from six adjacent NW-LEDs under different pressures. (f) Schematic band diagram of an n-ZnO/p-polymer p-n junction without (top) and with (solid line in the bottom) applied compressive strain. Reproduced with permission from [50]. John Wiley & Sons. [© 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim].

piezo-phototronic effect for the first time [51]. In this work, an InGaN/GaN multiple quantum well (MQW) pillar was realized by top-down fabrication from an epitaxial InGaN/GaN multilayer on a *c*-plane bipolished sapphire substrate, as shown in figure 4(a). The density of the nanopillar array is $6.25 \times 10^6\,\mathrm{cm}^{-2}$. The diameter, height of each pillar and distance between two adjacent pillars are 0.8, 1.2 and 4 μm, respectively. This corresponds to a pixel resolution of 6350 dpi. The uniformity is excellent. The inter-nanopillar space was filled with poly(methyl methacrylate) (PMMA), which is transparent in the visible light range. Then the tips of the nanopillar array were exposed by oxygen plasma etching, as shown in figure 4(b). The PL image of the InGaN/GaN MQW nanopillar array was excited by a laser with a wavelength of 405 nm. Figure 4(c) is the recorded signal. It is very uniform in a large area. Each pixel is a blue-light emitter, as shown in the insert of figure 4(c). A good spatial resolution for pressure mapping is guaranteed by no cross-talk between adjacent blue-light spots. PL spectra of the MQW nanopillar array at different excitation power densities are shown in figure 4(d). The PL intensity increased with increasing excitation optical power from 2 to 20 mW mm^{-2}. When pressure was applied to the structure, figure 4(e) presents the PL spectra recorded from a typical MQW nanopillar. It shows that the PL intensity has a very strong dependence on the magnitude of the applied pressure, while the PL peak wavelength is kept the same. The relative PL intensity change ($|L_\sigma - L_0|/L_0$) as a function of applied pressure σ_s is presented in figure 4(f), in which L_0 and L_σ denote the integral PL intensity between 420 and 513 nm under zero pressure and the applied pressure introduced by a stamp with a convex character pattern of 'BINN', respectively. For the nanopillar inside the stamp, it shows a linear relationship, while for the nanopillar outside the stamp, there is almost no response. This is because that the photogenerated electron–hole pairs in the MQW will be quickly separated by the increased built-in electric field under pressure, resulting in a decrease of radiative recombination probability and hence increased PL intensity. Figure 4(g) is the optical image of the stamp with 'BINN' character in contact with the MQW nanopillar array.

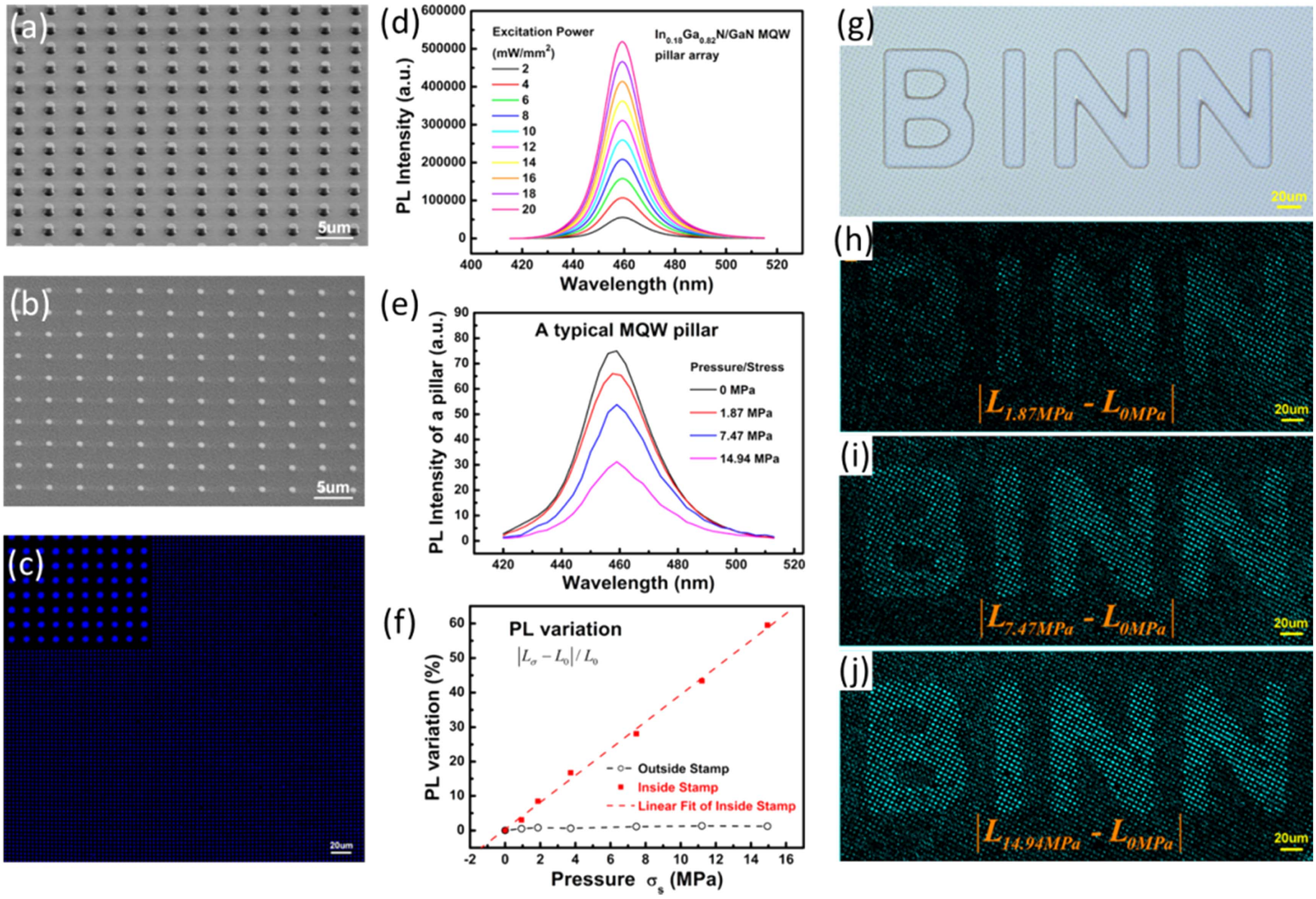

Figure 4. (a) SEM image of an InGaN/GaN MQW nanopillar. (b) SEM image of the pillar array after PMMA coating and oxygen plasma etching to expose tips. (c) PL image of the MQW pillar array. (d) PL spectra of the MQW pillar array at different excitation power densities. (e) PL spectra of a typical MQW pillar under different pressure. (f) Changes of PL intensity inside and outside the BINN stamp as a function of the applied pressure. (g) Optical image of the BINN stamp in contact with the MQW nanopillar array. (h)–(j) PL intensity differential images recorded under 1.87 MPa, 7.47 MPa and 14.94 MPa, respectively. Reprinted with permission from [51]. Copyright 2015 American Chemical Society.

Figures 4(h)–(j) are PL intensity differential images recorded under 1.87 MPa, 7.47 MPa and 14.94 MPa, respectively. A high-resolution pressure image can be obtained by this approach. The advantage of this approach by utilizing PL is that it is an all-optical method. The fabrication complexity will be reduced with no electrical interconnect layout required. It is suitable for large-area, highly uniform and high-resolution sensing applications.

Plasmonic enhancement by metal NPs has offered new opportunities to engineer and improve the performance of optoelectronic devices [52–55]. Recent work shows that localized surface plasmonic (LSP) resonance can be coupled with the piezo-phototronic effect to enhance the PL of InGaN/GaN MQWs coated with Ag NPs [56]. The structure under investigation is schematically shown in figure 5(a). InGaN/GaN MQWs were grown on (0001)-oriented sapphire substrates by metal–organic chemical vapor deposition. The active region is composed of five-period $In_{0.17}Ga_{0.83}N$/GaN MQW structures with a thickness of 2.2 nm. Ag thin film was deposited on the MQWs' top surface. Then Ag NPs with a mean size of 50 nm were obtained by annealing the sample at 300 °C for 5 min in N_2 atmosphere. The MQWs-LSPs resonance process is schematically shown in figure 5(b). In MQWs without coating, the photon-generated electron–hole pairs are terminated through radiative recombination k_{rad} or nonradiative recombination k_{non}. When Ag NPs coating is introduced, electron–hole pairs excited within the MQW couple to electron vibrations at the metal–semiconductor interface when the energies of electron–hole pairs in MQWs and of the metal LSPs are similar. Then the resulting LSPs can be efficiently scattered as light as a result of inherent surface roughness in the evaporated metal coatings. This new recombination path increases the spontaneous recombination rate, and leads to the enhancement of light emission by LSP-MQW coupling [57]. To introduce the piezo-phototronic effect, the sample is fixed by a jig on a plate, with a jackscrew pinning on the back to apply force at the center of the sample. Figure 5(c) shows the simulated tensile stress distribution when an external stress of 10 N is applied by using a finite-element analysis method. In order to facilitate direct comparison, Ag NPs only covered half of the MQW surface, and the other half was left bare. Figures 5(d) and (e) present the PL spectra of InGaN/GaN MQWs under straining condition without and with Ag NPs, respectively. The spectra are measured on six equidistant points on the sample, as

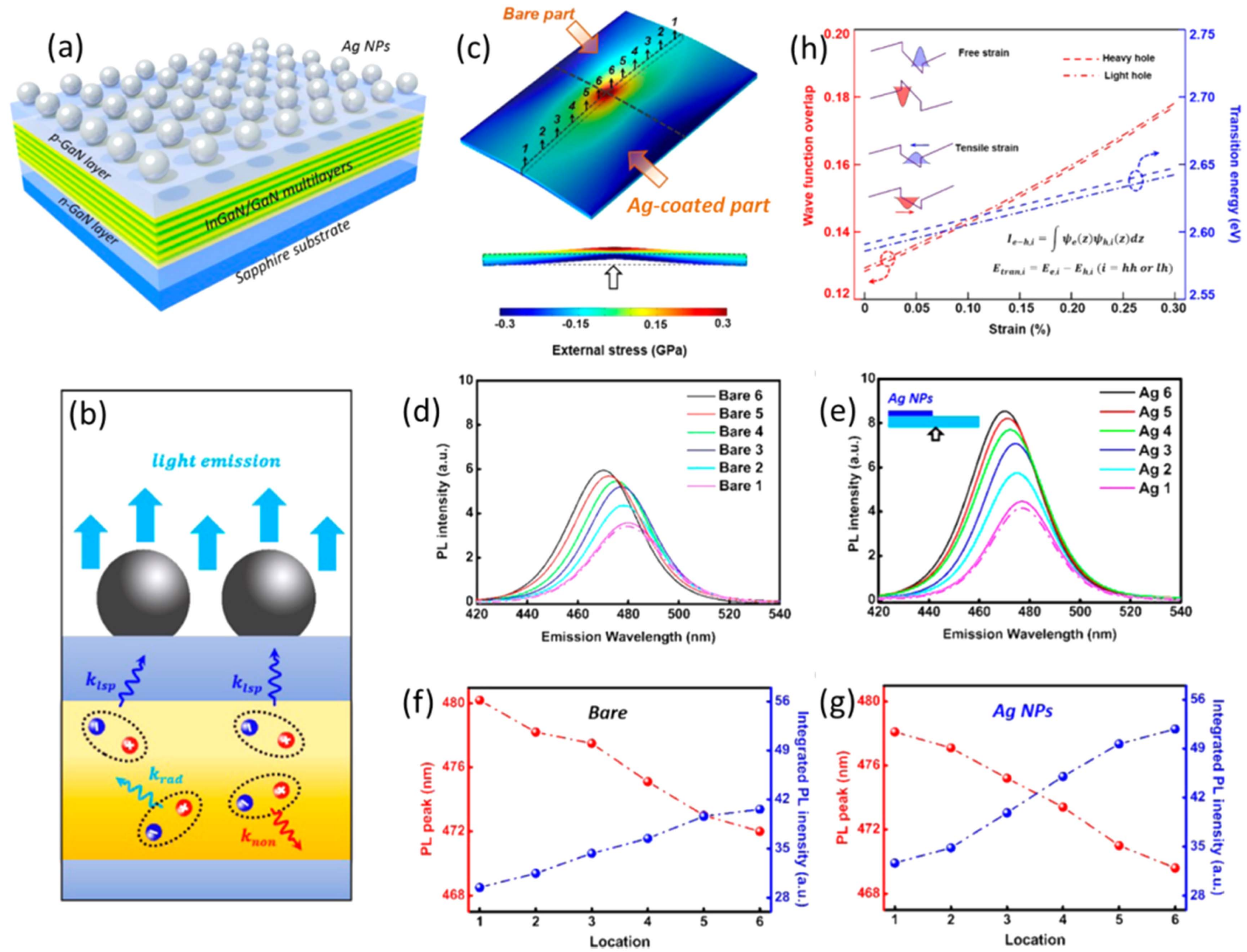

Figure 5. (a) Schematic view of Ag NP-coated InGaN/GaN MQWs. (b) Schematic diagram to show the mechanism of the LSP-MQWs resonance process. (c) Simulated distribution of tensile stress induced by an external applied stress of 10 N. PL performance under straining condition for (d) bare InGaN/GaN MQWs and (e) Ag NP-coated InGaN/GaN MQWs. The shift of PL emission peak and integrated PL intensity for (f) bare InGaN/GaN MQWs and (g) Ag NP-coated MQWs. Reprinted with permission from [56]. Copyright 2016 American Chemical Society.

marked in figure 5(c). Obviously, the PL intensity increases significantly from the edge to the center with increased stress and the peak wavelength gets blue shifted in both cases. The shift of emission peak and integrated PL intensity with different recording locations of the two cases is plotted in figures 5(f) and (g), respectively. The increasing magnitude of PL intensity is larger for Ag NP-coated MQWs than the bare MQWs. Theoretical calculation indicated that the spatial overlap of the electron-heavy hole and electron-light hole both increase with external strains and so does the effective band gap, as shown in figure 5(h). Thus the blue shift of the PL peak is ascribed to increased effective bandgap with reduced piezoelectric field, while the monotonic increase of PL intensity is attributed to the piezo-phototronic effect. This research provides an approach to utilizing plasmonics with the piezo-phototronic effect and brings widespread applications of piezo-phototronics to high-efficiency artificial lighting, on-chip integrated plasmonic circuits, etc.

4. Understanding the physics of the piezo-phototronic effect

Temperature dependence behavior is very useful for understanding the physics of a property or phenomenon [58–61]. Previously temperature dependence of the piezotronic effect has been investigated for proving the proposed fundamental mechanism [62]. Recently, the temperature dependence of the three-way coupling among mechanical, optical and electrical properties in CdS NWs has been systematically investigated by varying the temperature from 77 to 300 K, and the piezo-phototronic effect is significantly enhanced by over 550% when lowering the system temperature [63]. In this work, CdS NW devices were fabricated on aluminum foil covered with a layer of Kapton tape. The electric properties of the device were measured in a micromanipulation cryogenic probe system, as shown in the inset of figure 6(a). The device was bent with a micromanipulator and a beam of green laser

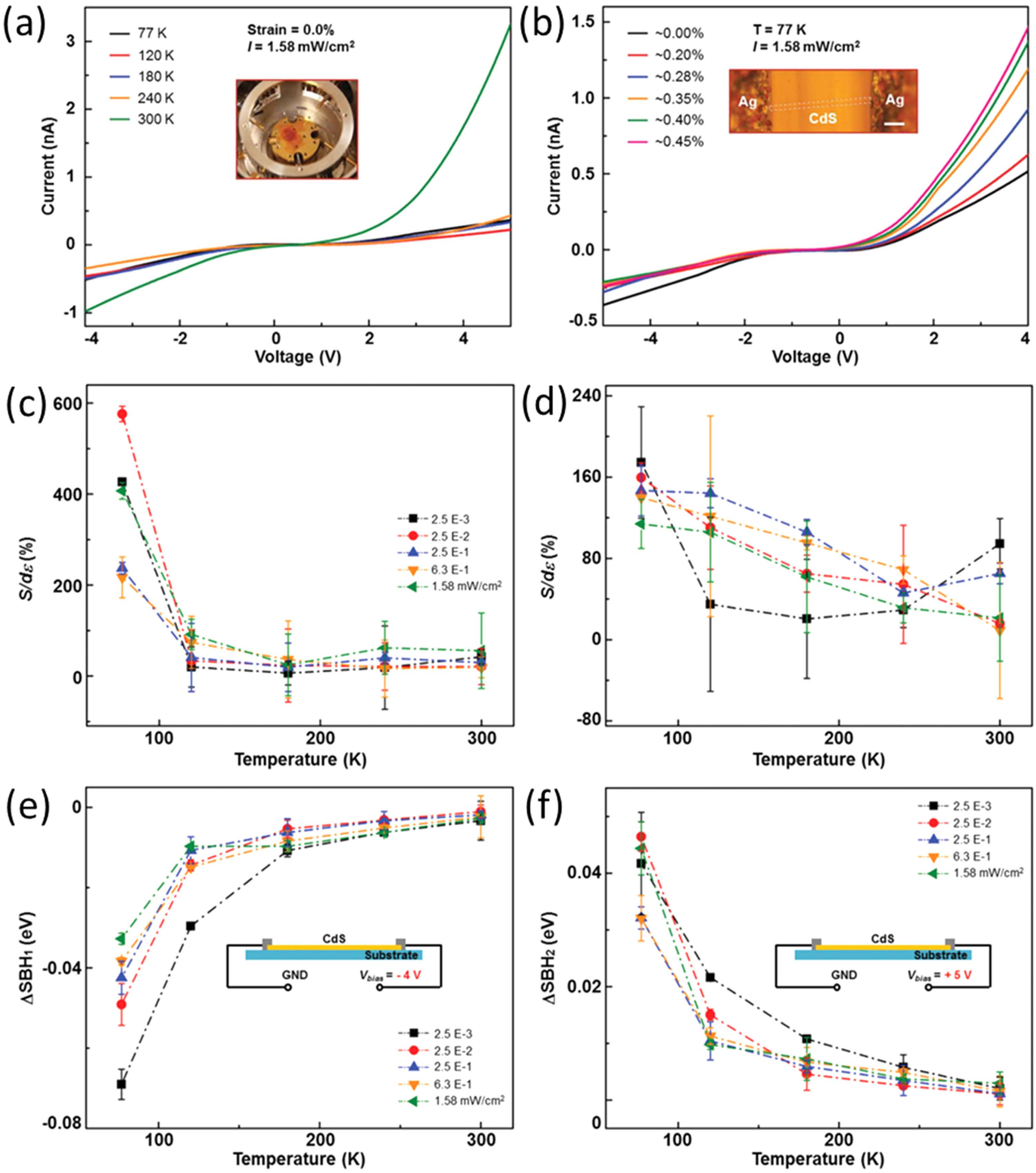

Figure 6. (a) Typical *I–V* characteristic of a CdS NW device at various temperatures under illumination. (b) Typical *I–V* characteristic of CdS NW devices at 77 K with applied strain under illumination. The piezo-phototronic factors as a function of temperature with different illumination intensities under (c) −4 and (d) +5 V biased voltages. Calculated changes of SBH as a function of temperature at the reversely biased Schottky contact under (e) −4 and (f) +5 V biased voltages. Reproduced with permission from [63] John Wiley & Sons. [© 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim].

(wavelength = 532 nm) was introduced to shine on the device. Figure 6(a) is the typical *I–V* characteristic to show the temperature dependence of the devices under illumination. When there is no strain, the current passing through the device decreases as the system temperature decreases under an optical illumination density of 1.58 mW cm^{-2}. At 77 k with the same illumination condition, when strain is introduced, the output currents increase at positive biased voltages while decrease at the negative side when the externally applied tensile strains are increased, as shown in figure 6(b). A physical parameter, piezo-phototronic factor, is defined as the response sensitivity per unit strain:

$$\frac{S}{d\varepsilon} = \frac{I_{\varepsilon_2} - I_{\varepsilon_1}}{I_{\varepsilon_1}(\varepsilon_2 - \varepsilon_1)}$$

where I_{ε_2} and I_{ε_1} correspond to the photocurrent under ε_2 and ε_1 straining conditions at the same illumination intensity, respectively. The piezo-phototronic factors under −4 and +5 V biased voltages are plotted as a function of temperatures with different illumination intensities in figures 6(c) and (d), respectively. A larger piezo-phototronic factor is observed at lower temperature for all the cases. To further understand this phenomenon, changes of Schottky barrier height (SBH) are simulated by a program PKUMSM based on an M-S-M model [64]. The calculated changes of SBH as a function of temperature at the reversely biased Schottky contact under −4 and +5 V bias voltages are shown in figures 6(e) and (f), respectively. The significantly enhanced piezo-phototronic effect at low temperatures is attributed to the effective polarization charges increase, which leads to more changes in the SBH. It results from the reduced screening effect caused by the decreased charge carriers mobility and density when the system is cooling down. In another work, temperature dependence of the piezo-phototronic effect in *a*-axis GaN nanobelts was investigated [65]. In this material system, there is a competition between the enhanced attraction of electrons to noncompletely screened positive piezocharges and the enhanced detrapping/activation of bounded electrons, which leads to a local minimum of output currents at a certain straining condition. Both of these works investigate the temperature dependence and the fundamental working mechanism of the piezo-phototronic effect, providing guidance for the design and fabrication of piezo-phototronic optoelectronic devices.

5. Perspectives

Piezo-phototronics provide a new platform for materials research and new optoelectronic device exploration. The three-way coupling makes this area attractive for all researchers with mechanical, optical and electrical backgrounds, and its development needs multidisciplinary contributions. The rapid progress of piezo-phototronics in the last several years has proved this. The controllable tuning characteristics make piezo-phototronics very powerful to introduce new vitality to traditional devices and explore devices with new working principles. The extended material systems, emerging application areas and further understanding of piezo-phototronics we reviewed here represent just a short summary of the recent progress in the last two years. More things are waiting to be explored to further release the huge potential of piezo-phototronics.

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