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Photovoltaic effect and tribovoltaic effect at liquid-semiconductor interface

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ABSTRACT

Contact electrification involving semiconductors has attracted attention for that it generates direct current. But its mechanism is still under debate, especially for the liquid-semiconductor cases. Here, the tribo-current is generated by sliding a DI water droplet on a semiconductor wafer, such as Si and TiO_2, under the light irradiation. It is revealed that the photoexcited electron-hole pairs at the interface will contribute to the tribo-current, and the enhanced tribo-current increases with the increased light intensity or the decreased light wavelength. The results suggest that the tribo-current at the DI water-semiconductor interfaces is induced by the tribovoltaic effect, in which electron-hole pairs are excited during contact owing to the energy released by the newly formed bonds, which can be named as “bindington”. The electron-hole pairs are further driven by the built-in electric field to move from one side to the other side at the interfaces, generating a direct current. The findings imply that the electron transfer exist at the liquid-solid interface in the CE, and support the “two-step” model for the formation of the electric-double layer, which was first proposed by Wang.

1. Introduction

Contact electrification (CE) is one of the most mysterious physical phenomena, which occurs in our daily life. The mechanism of the CE involving metals and insulators has been widely discussed for more than 2600 years [1,2], while the semiconductors were neglected. Recently, the CE involving semiconductors has attracted attention for that it produces direct current [3–6]. In contact, the bonding interaction between the two surfaces will release energy. Here, the energy released by the newly formed bond is defined as “bindington”. It was proposed by Wang that the tribovoltaic effect is responsible for the CE involving semiconductors, in which electrons are excited the “bindington” at the P-N junction or the Schottky junction during friction, and the excited electrons are further driven by the built-in electric field to move from one side to the other side at the interfaces, generating a direct current [7]. The tribovoltaic effect provides an explanation for the CE involving semiconductors, while more experimental evidence is needed to further support this point, especially for the liquid-semiconductor cases.

For the solid-solid cases, such as a solid metal sliding on a semiconductor surface, it is not controversial that the direct current is generated by the electron transfer [8–12]. And several experiments support that the electron transfer behavior at the solid metal/-semiconductor interface follows the tribovoltaic effect [3,6,10]. It was recently reported that the direct tribo-current can also be generated by sliding a water droplet over a semiconductor surface [13]. When the water is involved in the sliding, it is suspected that the CE may be due to ion transfer, simply because ions are often present in aqueous solutions, such as H^+ and OH^- [14]. The ion transfer contradicts to the tribovoltaic effect that is based on electron transfer. Therefore, it is urgent to determine the identity of the charge carriers in the liquid-semiconductor CE, and verify the tribovoltaic effect at liquid-semiconductor interfaces. The tribovoltaic effect is usually analogous to the photovoltaic effect, which can also occur at liquid-semiconductor interfaces [13,15,16], the only difference is that the electron-hole pairs in the triboelectric effect are excited by the “bindington”, instead of the light irradiation. Hence, the tribovoltaic current at the liquid-semiconductor interfaces should be in the same direction with the photo-current, both of which depend on the direction of the built-in electric field at the interfaces. It is also expected that the tribovoltaic current can be affected by the light irradiation, since that the light irradiation can increase the electron-hole pair concentration at the liquid-semiconductor interfaces.

In this paper, a water droplet is dragged by a quartz tube to slide on

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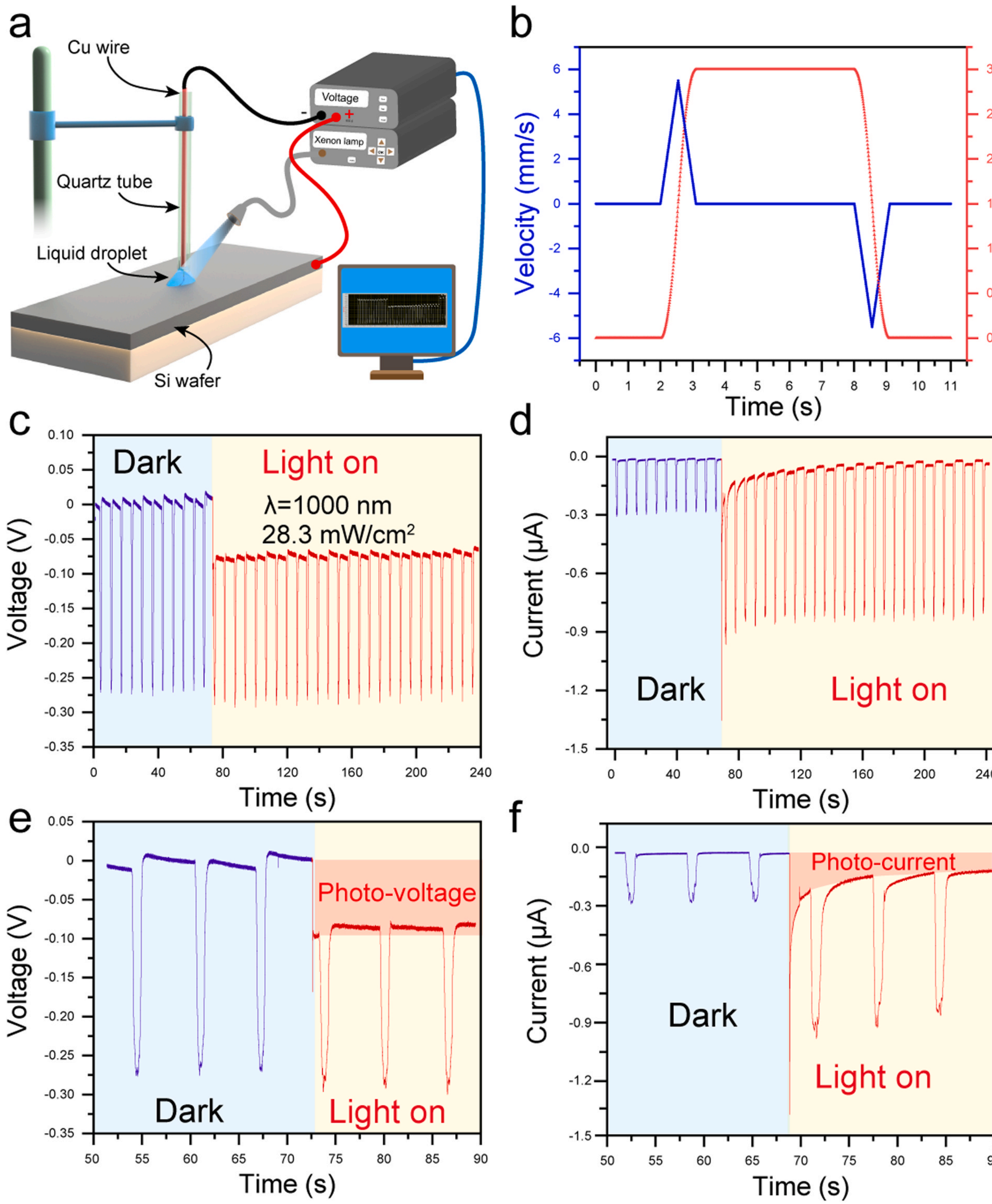

Fig. 1. The coupling of photovoltaic effect and tribovoltaic effect at the DI water and Si wafer interface. (a) The setup of the experiments. (b) The relative position and velocity between the Si wafer and the DI water droplet. The oscillogram of the (c) open-circuit voltage and the (d) short-circuit current when a DI water droplet slides on a N-type Si surface with and without light irradiation (1000 nm, 28.3 mW/cm^2). Six cycles of the (e) open-circuit voltage and (f) short-circuit current signals before and after the light was switched on, which are extracted from (c) and (d).

both N-type and P-type Si surfaces, with and without light irradiation. We focus on the effects of the light wavelength and intensity on the photo-current and the tribo-current. The results turn out that the tribo-current has the same direction as the photo-current, and the light irradiation is found to enhance the tribo-current at the liquid-semiconductor interface, which suggests that the tribo-current is induced by the tribovoltaic effect, and the charge carriers are determined to be electrons. Moreover, the coupling interaction between the tribovoltaic effect and the photovoltaic effect is also studied at the water-TiO_2 interfaces, to verify the universality of the tribovoltaic effect.

2. Results and discussion

The effects of the light irradiation on the tribo-current and the tribo-

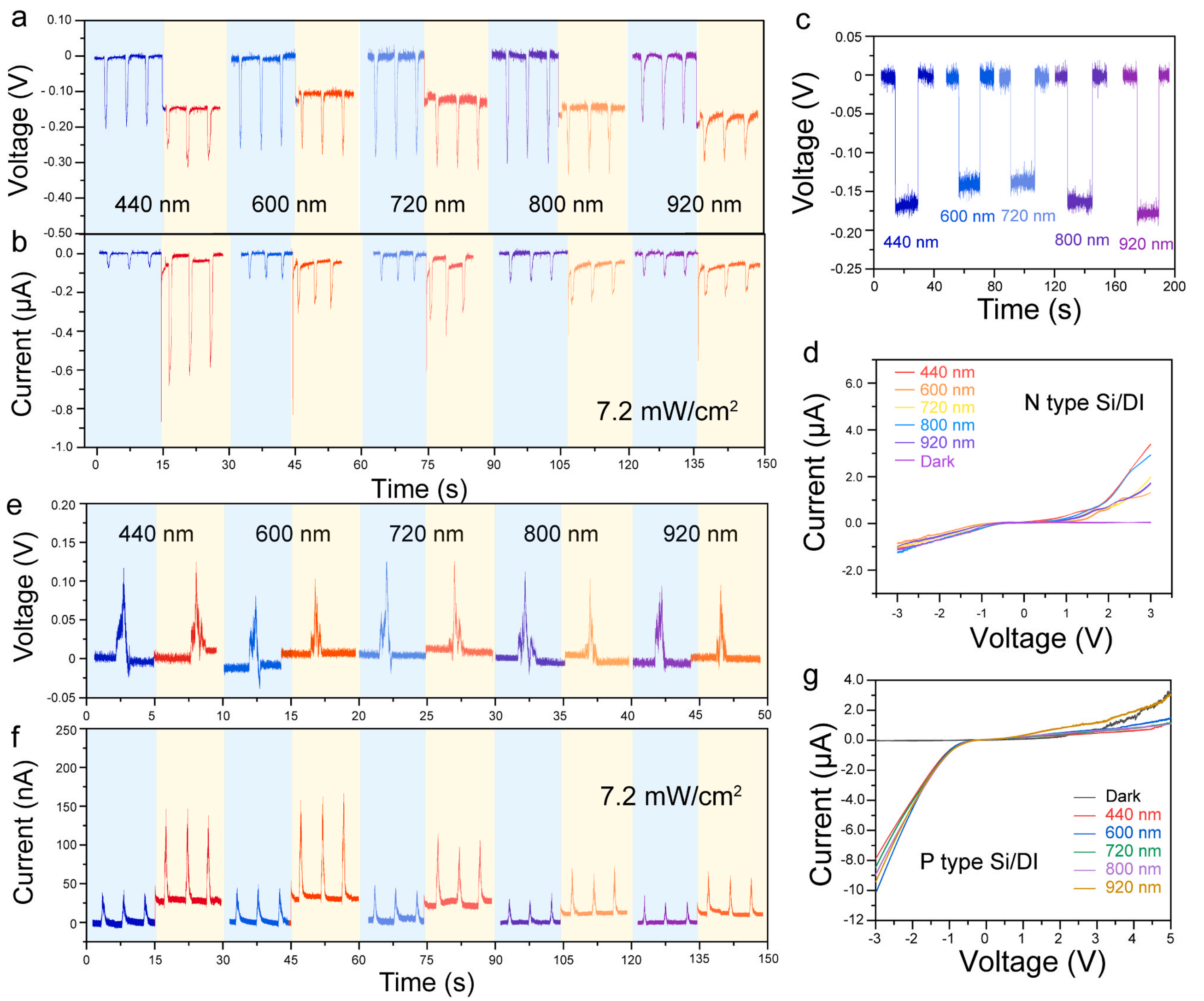

Fig. 2. Effect of light wavelength on the coupling interaction between photovoltaic effect and tribovoltaic effect. The effect of light wavelengh on the (a) tribo-voltage, (b) tribo-current between the DI water and the N-type Si. (c) The effect of light wavelengh on the photo-voltage. (d) The current-voltage curves between the DI water and the N-type Si. The effect of light wavelengh on the (e) tribo-voltage, (f) tribo-current between the DI water and the P-type Si. (g) The current-voltage curves between the DI water and the P-type Si.

voltage at the deionized water (DI water) and the Si wafer interface was investigated. The Si wafer and a quartz tube were mounted on a mobile platform and an immobile holder, respectively, as shown in Fig. 1a, and a photo of the device is shown in Fig. S1. The DI water droplet was dragged by the quartz tube to slide on the Si wafer surface, as shown in Movie S1,S2. And one cycle of the relative position and velocity between the DI water droplet and the Si wafer are given in Fig. 1b, as a function of time. In the experiments, a Cu wire was fixed in the tube as an electrode contacted with the DI water, and the short-circuit current and the open-circuit voltage were measured by using electrometer (Keithley 6514, The external voltage and current from the Si wafer to the DI water droplet are defined as positive). The distance between the quartz tube and the Si wafer was about 0.3 mm during sliding. All that make sure that the joint of the quartz tube and the DI water is transparent and the light generated by the xenon lamp can reach the interface between the DI water and the Si wafer.

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Fig. 1c,d give the tribo-voltage and the tribo-current when the DI water droplet sliding over a Si wafer (N-type with 1 Ωcm resistivity) in the dark condition, and under light irradiation (1000 nm, 28.3 mW/cm^2 light intensity). The experimental details are shown in Movie S3. In the dark condition, the tribo-current and the tribo-voltage were about −0.3 μA and −0.3 V, which means that the direction of the tribo-current was from the N-type Si side to the DI water side at the interface. We measured the current-voltage curves at the N-type Si and the DI water interface, and the results are shown in Fig. S2a. The current-voltage curve in the dark condition shows the rectification performance of the junction. It suggests that the built-in electric field was formed at the interface when the DI water contacts the N-type Si wafer, and the direction of the built-in electric field at the interface also points from the N-type Si side to the DI water side. (It should be noticed that the direction from the Si wafer to the DI water at the interface is defined as positive at the current-voltage test.) When the lamp was switched on, the light irradiated on the Si and the DI water interface, and the tribo-voltage and tribo-current were affected by the light irradiation (Fig. 1c,d). We extract six cycles of the voltage and current signals before and after the light was switched on, as shown in Fig. 1e,f. The photo-voltage and photo-current are about −0.08 V and −0.3 μA (the red region), respectively, which is found to be the same as that without sliding (Fig. S2b). For the tribo-signal component, the tribo-voltage is found to slightly decrease, while the tribo-current significantly increases. These

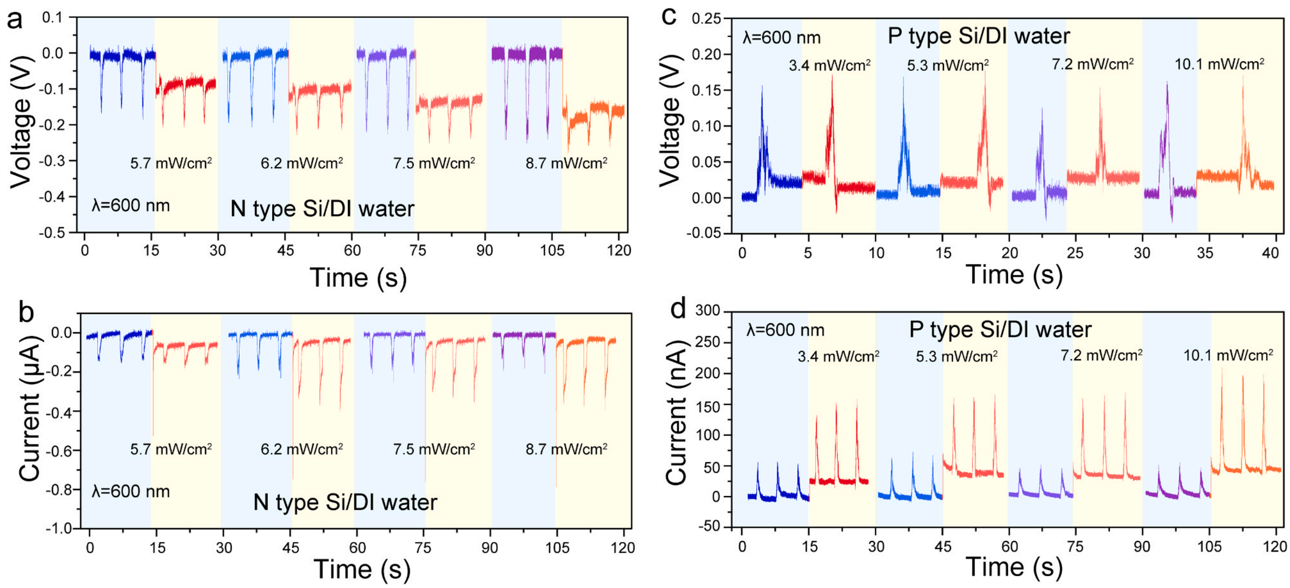

Fig. 3. Effect of light intensity on the coupling interaction between photovoltaic effect and tribovoltaic effect. The effect of light intensity on the (a) tribo-voltage, (b) tribo-current between the DI water and the N-type Si. The effect of light intensity on the (c) tribo-voltage, (d) tribo-current between the DI water and the P-type Si.

results cannot be explained by the desorption/adsorption of the ions on the Si surfaces. Because the voltage and current generated by the ion desorption/adsorption both depend on the rate of the desorption/adsorption. A decreasing of the ion transfer induced voltage should lead to a decreasing of the ion transfer induced current. Therefore, the tribo-voltage and tribo-current should be caused by the electron transfer.

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We notice that the photo-voltage and photo-current were of the same direction as the tribo-voltage and tribo-current, which is consistent with the physics mechanism of the tribovoltaic effect. And the effect of the light irradiation on the tribo-current was not caused by the change of the temperature, which was measured to be less than 1 ℃ after 10 min irradiation (Figs. S2c,d). As shown in Fig. S1a, the rectification performance of the junction is weakened when the light irradiated on the interface. This is caused by the increasing of the electron-hole pairs concentration at the interface excited by the light irradiation, which may also be responsible for the change of the tribo-voltage and the tribo-current. It becomes harder for the energy released by the electron transition during friction to excite more electron-hole pairs, since its concentration is already high under light irradiation. Hence, the tribo-voltage decreases when the photo-voltage is generated. But for the tribo-current, it increases due to the high electron-hole pairs concentration at the interface, which can be considered as a reduction in the internal resistance. Based on the observation and explanation, the generation of the tribo-voltage and tribo-current are induced by electron transfer, rather than ion desorption/adsorption.

In order to further verify the tribovoltaic effect, the effect of the light wavelength on the tribo-voltage and the tribo-current was investigated, and both N-type Si and P-type Si were used in the experiments. As shown in Fig. 2a,b, the photo-voltage during sliding (or without sliding, Fig. 2c) remains almost unchanged when the light wavelength various from 440 nm to 920 nm (the light intensity remains on 7.2 mW/cm^2), and the tribo-voltages are slightly decrease under light irradiation. The light-enhanced tribo-current is observed, and it is found that the shorter the light wavelength, the larger the enhanced tribo-current (Fig. 2b). The tribo-current ups to −0.6 μA, when a light with 440 nm wavelength irradiate on the interface. And the enhanced tribo-current decreases to be less than −0.2 μA, when the light wavelength is 920 nm. These results also consistent with the tribovoltaic effect. As shown in Fig. 2d, the current-voltage curves suggest that the rectification performance of the junction becomes weaker under light irradiation, due to the decreasing of the internal resistance. A shorter wavelength light has a higher energy, which can generate more electron-hole pairs at the interface. When a tribovoltaic voltage is generated by the electron transition or the bonding interaction between the water droplet and the Si wafer, the electron-hole pairs generated by the light irradiation are also driven by the tribovoltaic voltage and contribute to the tribovoltaic current. Therefore, a shorter light wavelength will produce more electron-hole pairs, and further results in a larger light-enhanced tribovoltaic current.

When the water droplet sliding on a P-type Si with 1 Ωcm resistivity, the direction of the tribo-voltage and the photo-voltage reversed to be from the water side to the P-type Si side at the interface (Fig. 2e,f). This is caused by the reversion of the built-in electric field direction, which is suggested by the current-voltage curves, as shown in Fig. 2g. And the direction of the tribo-voltage and tribo-current is consistent with the tribovoltaic effect. Similar to that at the DI water and the N-type Si interface, the tribo-voltage at the DI water and the P-type Si interface slightly decreases, while the tribo-current increases under light irradiation. Also, the increasing of the tribo-current depends on the light wavelength, a shorter light wavelength will result in a larger tribo-current. These results support the point that the tribo-voltage and tribo-current at the liquid-semiconductor interface are induced by the tribovoltaic effect, and reveal the coupling interaction between the tribovoltaic effect and the photovoltaic effect.

The effect of the light intensity on the coupling interaction between the tribovoltaic effect and the photovoltaic effect was also investigated. It is found that the tribo-voltage between the N-type Si and the DI water will not be significantly affected by the light intensity as well as the photo-voltage (Fig. S3a), while the tribo-current increases with the light intensity when the light intensity various from 5.7 mW/cm^2 to 8.7 mW/cm^2, as shown in Fig. 3a,b. There is an approximate logarithmic relationship between the photo-voltage and the light intensity, which suggests that the effect of the light intensity on the photo-voltage is limited when the light intensity is high enough [17]. The tribovoltaic effect is analogous to the photovoltaic effect, it is reasonable that the light intensity effect on the tribovoltaic voltage is not significant. For the tribovoltaic current, it is similar to the effects of the light wavelength. A higher light intensity will also excite more electron-hole pairs at the interface and results in a lower internal resistance, which is implied in the current-voltage curves at the N-type Si and the DI water interface under the light irradiation with different intensity (Fig. S3b). And the

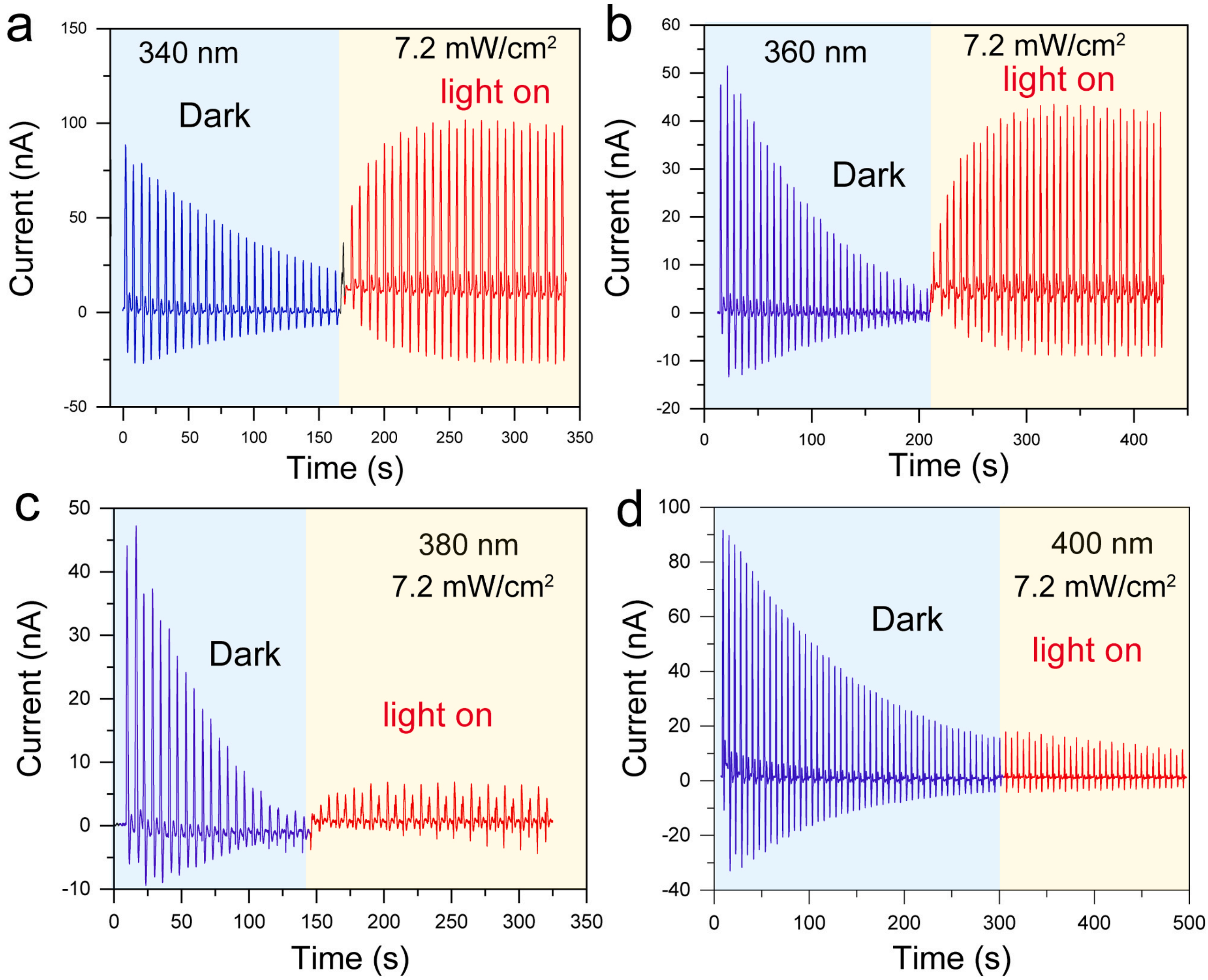

Fig. 4. The coupling of photovoltaic effect and tribovoltaic effect at the DI water and the TiO_2 interface. The oscillogram of the short-circuit current when a DI water droplet slides on a TiO_2 surface in the dark condition and under light irradiation with (a) 340 nm, (b) 360 nm, (c) 380 nm, (d) 400 nm light wavelengh, 7.2 mW/cm^2 light intensity.

photoexcited electron-hole pairs will contribute to the tribovoltaic current, lead to an enhancement on the tribo-current. It is worth noting that the tribo-current under 5.7 mW/cm^2 light irradiation is slightly lower than that in the dark condition. This may be caused by that the light excited electron-hole pairs concentration is too low to contribute the tribovoltaic current in this situation, and the reduction of the tribovoltaic voltage under light irradiation lead to the decreasing of the tribovoltaic current. When the DI water sliding over the P-type Si surface, the tribo-voltage and tribo-current reverse to be positive as expected (Fig. 3c,d). And the light-enhanced tribo-current also increase with the light intensity. The light intensity effect also suggests that the tribo-voltage and tribo-current at the water-Si interface are generated due to the electron-hole pairs at the interface, which are excited by the CE and further driven by the built-in electric field, generating direct current, so called as tribovoltaic effect.

In previous studies, the semiconductors used in the CE were usually narrow gap semiconductors, such as Si and MoS_2 [8,13]. In order to verify the universality of the tribovoltaic effect and the coupling interaction between the tribovoltaic effect and the photovoltaic effect, the wide gap semiconductor (TiO_2) was used as the semiconductor contact side in the experiments. As shown in Fig. 4a, the tribovoltaic current can also be generated at the DI water and TiO_2 interface, and the light irradiation is also found to enhance the tribovoltaic current, just like that occurs at the DI water and the Si interfaces. The different is that the tribovoltaic current will decay when the semiconductor contact side is TiO_2 in dark condition. When the light is switched on (340 nm, 7.2 mW/cm^2), the tribovoltaic current start to increase. This is because that the H^+ and OH^- will adsorb on the TiO_2 surface, occupy the active sites, and inhibit the electron transition and the release of the "bindington" at the DI water and the TiO_2 interface [16,18,19], which further reduces the excited electron-hole pairs and the tribovoltaic current. Under the light irradiation, the ions desorbed from the surface, and the tribovoltaic current increases. In this point of view, the ion transfer can inhibit rather than generate the tribo-current.

The results of the light wavelength effect are shown in Fig. 4b–d. The photo-current at the DI water and the TiO_2 interface is found to decrease with the increasing of the light wavelength. As expected, the tribovoltaic current under light irradiation also decreases when the light wavelength increases. When the light wavelength ups to 400 nm (3.1 eV), the photo-current is close to 0 nA, and the tribovoltaic current cannot be enhanced (Fig. 4d). These results suggest that the enhancement of the tribovoltaic current induced by the light irradiation can also occur at the water and the wide gap semiconductor surface (TiO_2), but a higher photon energy is required, comparing to that on narrow gap semiconductor surface (Si).

The mechanism of the tribovoltaic effect at the liquid-semiconductor interface has been described in previous work [13]. Here, we focus on

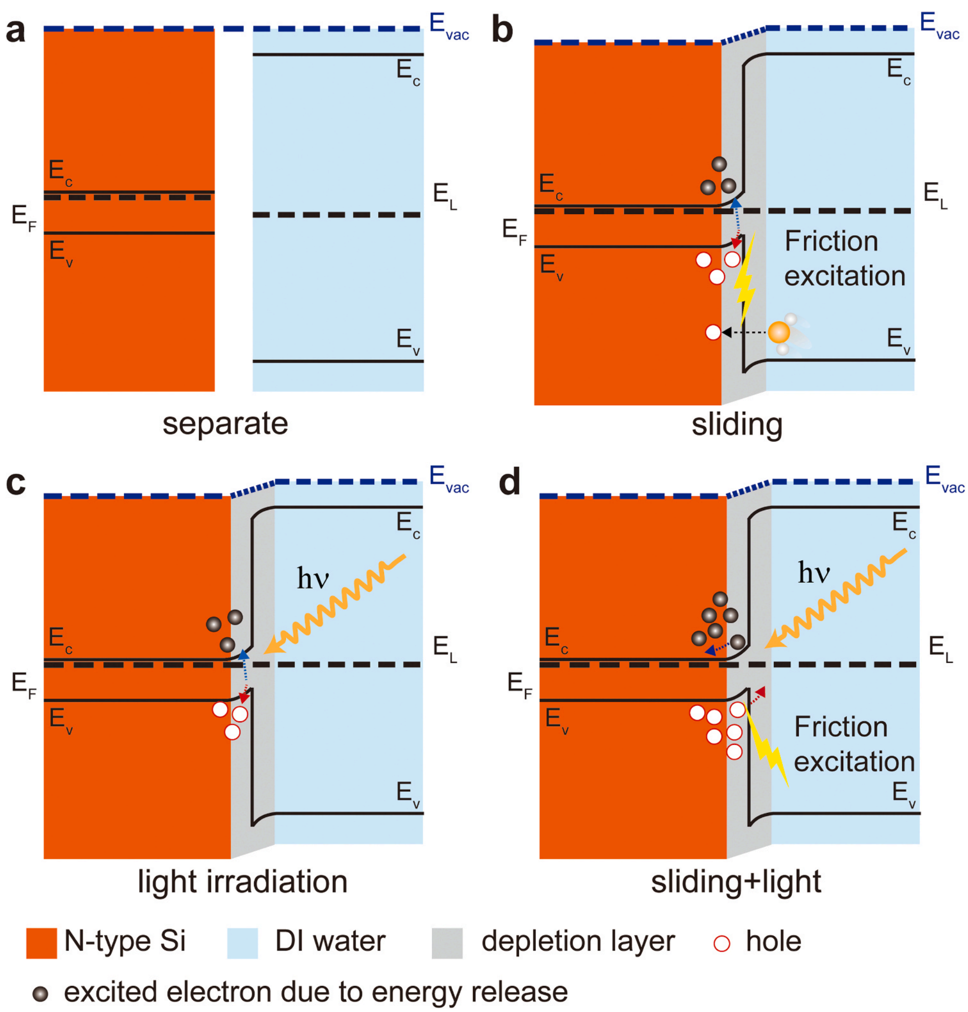

Fig. 5. Energy band diagram of the coupling interaction between photovoltaic effect and tribovoltaic effect. Energy band diagram of the N-type Si and the DI water (a) before sliding and (b) in sliding. (c) The photon excitation at the N-type Si and the DI water interface. (d) The coupling of photovoltaic effect and tribovoltaic effect at the DI water and N-type Si interface. (E_c is the bottom of the conduction band, E_f is the Fermi level of the N-type silicon, E_L is the 'Fermi level' of the DI water and E_v is the top of the valence band.).

the mechanism of the light effects on the tribo-current and a band structure model is proposed, in which the coupling of the tribovoltaic effect and photovoltaic effect in the experiments is simply explained by the increasing of the electron-hole pairs concentration at the liquid-semiconductor interface. Fig. 5a gives the band structure of the N-type Si and the DI water before sliding. When the DI water droplet slides on the N-type Si, the built-in electric field will be established. Meanwhile, the "bindington" will be released at the DI water and the Si interface, and excite electron-hole pairs (Fig. 5b). The photon will also excite the electron-hole pairs at the water-semiconductor interface, as shown in Fig. 5c. When the photon excitation and friction excitation occur simultaneously at the interfaces, the electron-hole pairs concentration will increase significantly (Fig. 5d). The excited voltage at the heterojunction mainly depends on the strength of the built-in electric field at the interface, especially when the excitation intensity is high enough [17,20]. Therefore, the increase of the total excited voltage is very limited when the light irradiates on the sliding DI water-Si interfaces, and the tribovoltaic voltage component even slightly decease. While the excited current heterojunction usually increases super-linearly with the increasing concentration of the electron-hole pairs, which may be caused by the decreasing internal resistance at the interface [17]. It means that the excited current is much more sensitive to the electron-hole pairs concentration comparing to the excited voltage. Hence, the tribovoltaic current will be significantly enhanced by the light irradiation.

The light enhanced tribovoltaic current should has implications in both science and technology. It provides a strong evidence for the point that the electron transfer can occur that the liquid-solid interface in the CE. And the results are consistent with the "two-step" model for the formation of electric-double layer (EDL), in which the electron transfer is considered to be the first step [7,21]. On the other hand, the discoveries in this work give a method to optimize the output performance of the liquid-solid nanogenerators [22,23].

Based on this study and previous works [5,6,10,13], we can give a clear definition of tribovoltaic effect here. The tribovoltaic effect refers to a phenomenon in which an energy "quantum" is released once an atom-atom bond is formed at the interface of two contacting materials, such released "binding" energy would excites electron-hole pairs at a metal-semiconductor interface or semiconductor-semiconductor PN junction. The electrons are separated from the holes due to the built inner electric field at the interface. The tribovoltaic effect office occurs even in the contacting of liquid and a semiconductor. This definition is analogous to that of the photovoltaic effect, in which the electron-hole pairs at a metal-semiconductor or semiconductor-semiconductor interfaces are excited by the incident photon, and further separated by the built inner electric field.

In conclusion, the tribo-voltage and tribo-current at the DI water-semiconductor interfaces were generated under light irradiation. It is revealed that the tribo-current and the photo-current are of the same direction and both depend on the built-in electric field at the interface between the DI water and the semiconductors. Moreover, the tribo-current was found to be enhanced by the light irradiation, and the enhanced tribo-current increased with the increasing light intensity and the decreasing light wavelength. These results support that the tribo-current was induced by the tribovoltaic effect, in which the electron-hole pairs at the interface are excited by the “bindington”, and driven by the built-in electric field at the heterojunction. An energy band model was further proposed to explain the coupling interaction between the tribovoltaic effect and the photovoltaic effect, in which the increasing of the electron-hole pairs concentration under light irradiation is responsible for the enhancement of the tribovoltaic current. Our findings suggest that the electron transfer exist at the liquid-solid interface in the CE, which is consistent with the “two-step” model for the formation of EDL.

3. Experimental section

3.1. Sample preparation

The P-type silicon is boron doped with 1 Ωcm resistivity and the N-type silicon is phosphorus doped with 1 Ωcm resistivity. The DI water was produced by a deionizer (Hitech, China), and the resistivity of the DI water used here was 18.2 MΩcm.

3.2. Current-voltage measurements

The current-voltage curves were measured by an electrochemical workstation (CHI600E, China). The scan rate was set to be 0.1 V/s, the sample interval was 0.001 V.

3.3. Light generation

The light was generated by a Xenon lamp (XBO-150 W, USA) in a Xenon lamp house (NEWPORT-67005, USA). The optical filters with the cut off wavelengh from 200 nm to 2500 nm were used to generate the light with a certain wavelengh. And the bandwidth of the optical filters was 10 nm. A optical power meter was used to measure the intensity of the light. (PM100D, THORLABS, USA).

CRediT authorship contribution statement

Mingli Zheng: Conceptualization, Methodology, Visualization, Data curation, Writing - original draft. **Shiquan Lin:** Conceptualization, Methodology, Visualization, Data curation, Writing - original draft. **Zhen Tang:** Resources, Writing - review & editing. **Yawei Feng:** Resources, Writing - review & editing. **Zhong Lin Wang:** Conceptualization, Methodology, Supervision, Writing - review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.nanoen.2021.105810.

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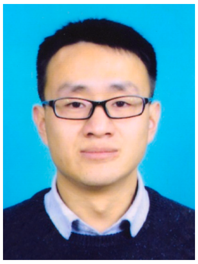

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