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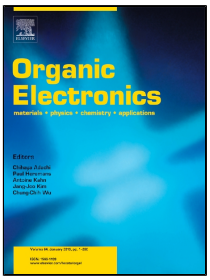

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Organic Photodetectors Based on Copper Phthalocyanine Films Prepared by a Multiple Drop Casting Method

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Abstract:

Organic Photodetectors have high potential for use in the next generation of information technology systems because of the mechanical flexibility, large-area detection and low production cost of organic materials for photodetectors. A red light photodetector based on phthalocyanine compounds is one of the representations. There are some disadvantages for the common methods to fabricate phthalocyanine films, such as expenses of high operation temperature, ultrahigh vacuum processing, large materials loss, complicated operation, large energy consumption and poor solubility. To solve these problems, a multiple drop casting method is proposed in this paper. This method could prepare phthalocyanine films from a relatively dilute solution, and give an alternative way to fabricate the phthalocyanine photodetectors especially for the phthalocyanine with poor solubility.

KEYWORDS: organic, photodetector, phthalocyanine, multiple drop casting

1. Introduction:

In recent years, photodetectors based on organic materials have received more and more attention due to their mechanical flexibility, large-area detection and low production cost, thus they have high potential for use in the next generation of information technology systems.[1-6] One of the representations is phthalocyanine compound, such as copper phthalocyanine (CuPc),[7-15] lead phthalocyanine (PbPc),[16-18] cobalt phthalocyanine (CoPc),[19] tin phthalocyanine (SnPc),[20] zinc phthalocyanine (ZnPc),[21-24] nickel phthalocyanine (NiPc),[25] titanyl-phthalocyanine (TiOPc),[26-29] vanadyl-phthalocyanine (VOPc)[30, 31] and other phthalocyanine[32, 33] which hold an outstanding chemical stability, thermal stability, low field-effect mobility, and environmental friendliness. In addition, the Q-band absorption (a good coverage typically in the range of 600–800 nm) makes them definitely appealing for red photons harvesting.

The phthalocyanine films of photodetectors could be fabricated via different methods such as spin casting[33], drop casting[9] and vacuum thermal deposition[14, 15]. The vacuum thermal deposition method is well developed by many researchers, but this process has some disadvantages such as expenses of high operation temperature, ultrahigh vacuum processing, large materials loss, complicated operation, and large energy consumption. As an alternative, the solution process based method (spin casting and drop casting) have the advantages of low cost and potential for the fabrication of the phthalocyanine photodetectors. However, phthalocyanine compounds are mostly not soluble well in either water or organic solvents. Due to the poor solubility of phthalocyanine materials, phthalocyanine films are hardly prepared via a traditional solution process. In this paper a multiple drop casting method is proposed, and a photodetector is fabricated based on a copper phthalocyanine (CuPc) film. CuPc solution was dropped onto a spinning substrate by a rate of 0.1 mL/minute till all the

CuPc solution was consumed. Moreover, the CuPc solution only consists of CuPc and solvent, and this method is a resin-free way which is different from traditional solution process. The multiple drop casting method could prepare phthalocyanine films from a relatively dilute solution, and give an alternative way to fabricate the phthalocyanine photodetectors especially for the phthalocyanine with poor solubility.

2. Experimental

2.1. Synthesis and characterization of CuPc films

CuPc films were deposited on the substrate of *c*-sapphire by a multiple drop casting method. Firstly, the CuPc solution (0.1 mmol·L^{-1}) was prepared by drop 14.4 mg CuPc (Aldrich, sublimed grade, dye content 99%) into 250 mL trichloromethane (Beijing Chemical Works, Chloroform A.R., 99%) followed by an ultrasonic treatment for 10 minutes. Secondly, the *c*-sapphire was fixed on the spin coater and the spin rate was kept at 2000 rpm. Thirdly, the CuPc solution was dropped onto the surface of the substrate by a rate of 0.1 mL/minute till all the CuPc solution (10 mL) was consumed. After that, the substrate was heated at 110 ^{o}C for 10 minutes. The films were characterized by using scanning electron microscopy (SEM) (HITACHI S-4800), X-ray diffraction (XRD) (Rigaku, D-Max 2550) using Cu Kα radiation (λ= 0.154 nm) and UV-Vis transmission spectrometry (Shimadzu UV-3101PC).

2.2. Fabrication and characterization of the photodetectors

After the deposition of the CuPc films on the *c*-sapphire, Au electrodes (~20 nm thick) were prepared on the films by a sputtering method at room temperature and interdigital electrodes were realized by a photolithography and a wet etching procedure. After that, the CuPc photodetectors were formed. The processes for the fabrication of the films and devices could be seen in **figure 1** schematically. Current–Voltage (*I–V*) properties and transient response

spectra of the photodetectors were measured by using a semiconductor device analyzer (Agilent B1500A) with a red laser (650 nm).

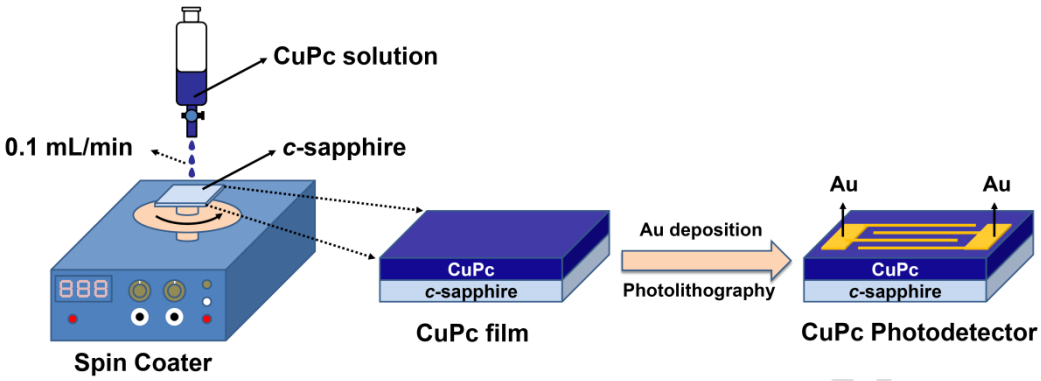

Figure 1. Schematic illustration of the fabrication of CuPc photodetectors.

3. Results and discussion:

The CuPc film was characterized by SEM, XRD and UV-Vis transmission spectra. **Figure 2a** shows the SEM image of the CuPc film. It can be seen that the surface of the film has a little of rod-like CuPc crystals probably due to the poor solubility of copper phthalocyanine. According to the cross section SEM images (**figure 2b**) of the film, the thickness of CuPc film is around 140 nm. **Figure 2c** shows the XRD pattern of the film. It can be seen that, two diffraction peaks located at 7.0° and 9.2° could be assigned as (−101) and (101) planes of monoclinic CuPc, respectively. The diffraction peaks are relatively weak, indicated that the crystallinity of CuPc is relatively low. The UV-Vis absorption spectrum of the film is shown in **figure 2d**. As is shown, CuPc film has a strong absorption in UV (389 nm) and visible (540–780 nm) spectrum region, and in the visible region there are two obvious absorption peaks located at 652 nm and 751 nm, respectively.

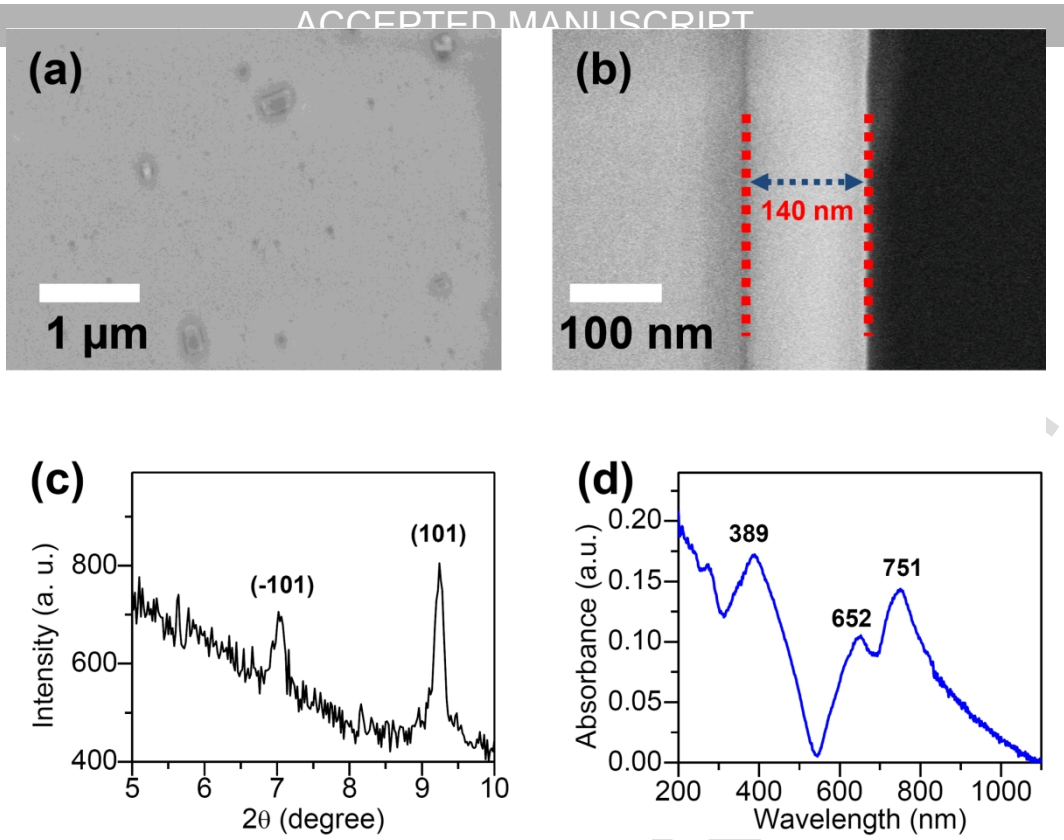

Figure 2. (a) and (b) SEM images of CuPc films; (c) XRD pattern of CuPc film. (d) UV-Vis transmission spectrum of CuPc film.

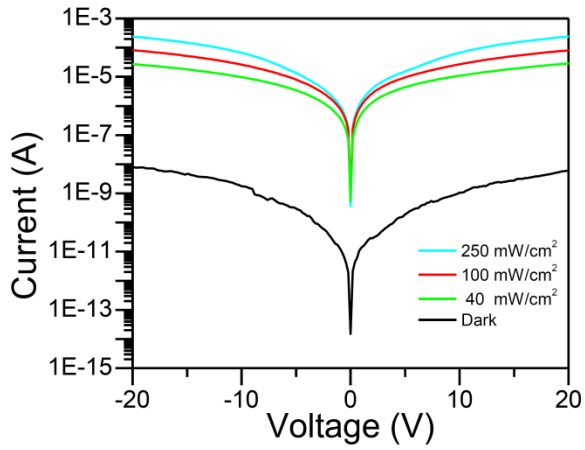

Figure 3. The *I–V* curves of CuPc photodetectors in dark and under 650 nm red laser illumination with different power density.

I–V characteristics of the CuPc photodetectors under dark and illumination conditions are shown in **figure 3**. The dark current (I_{dark}) of the device at a bias of 10 V is around 1 nA, and the absolute value of dark current increases with the increasing of the bias under both forward and reverse bias. Under the illumination of red laser (40 mW·cm^{-2}), the current

could be rise to 10.9 μA at a bias of 10 V. The absolute value of photocurrent also increases with the increasing of the bias in the similar way of dark condition. The *I–V* curves under different power density are also shown in figure 3. With the power density increasing, the current increases linearly. At a bias of 10 V, the photocurrent (I_{photo}) under a 650 nm red laser illumination are 10.9 μA, 26.8 μA and 66.8 μA for the power density of 40 mW·cm^{-2}, 100 mW·cm^{-2} and 250 mW·cm^{-2}, respectively.

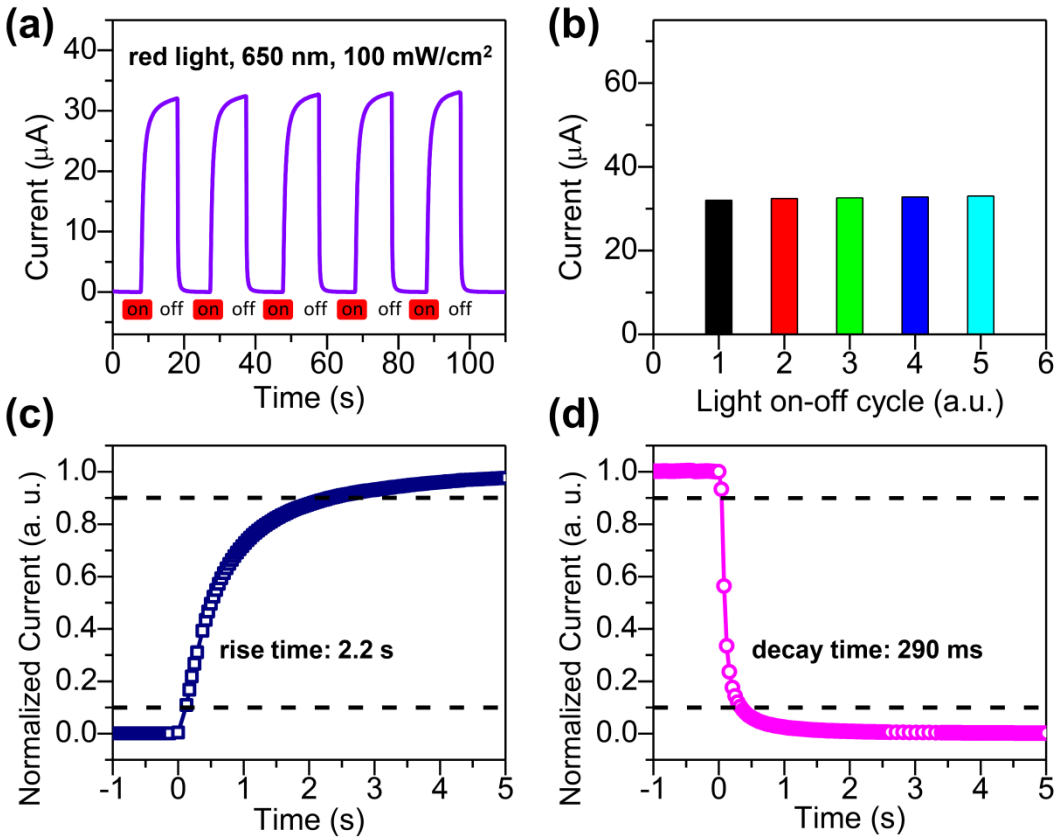

Figure 4 (a) Time-dependent photocurrent response of CuPc photodetector measured under illumination by 650 nm red light with the power density of 100 mW·cm^{-2} at 10 V; (b) Peak photocurrent of different light on-off cycles; (c) Rise edge of the current response; (d) Decay edge of the current response.

Figure 4a shows the time-dependent photocurrent response of CuPc photodetector measured under illumination by 650 nm red light (100 mW·cm^{-2}) at 10 V. Under the red light illumination, the current increases to around 32.4 μA. After turning off the light, the current quickly returns to its original value. As shown in **figure 4b**, the time-dependent response to

the light on–off cycles shows good stability and reproducibility, and the peak photocurrent is maintain at about 32.4 μA. **Figure 4c** and **4d** show the rise and decay edges of the current response, respectively. It can be seen that, the 10−90% rise time (defined as the time for the current rising from 10% to 90% of the peak value) of the CuPc photodetector is around 2.2 s. On the other hand, the 90−10% decay time (defined as the time for the current dropping from 90% to 10% of the peak value) of the device is around 290 ms.

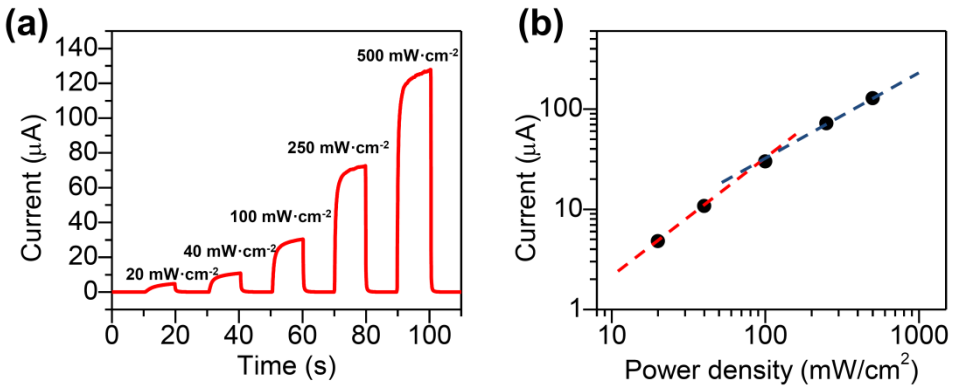

Figure 5 (a) Time-dependent photocurrent response of CuPc photodetector measured under illumination by 650 nm red light with different power density at 10 V; (b) Photocurrent as a function of power density of red light.

To investigate the ability of the device to respond to the different power density of the light, 650 nm light with the intensities ranging from 20 to 500 mW·cm^{-2} was irradiated on the CuPc photodetector at 10 V as shown in **Figure 5a**. With increasing of the light intensity, the device shows a corresponding response. **Figure 5b** shows the photocurrent as a function of light power density. It can be seen that, the photocurrent increases with the increasing of power density from 20 to 500 mW·cm^{-2}. It was reported that, the photocurrent (I_{ph}) follows the following power law:[12]

$$I_{\mathrm{ph}} \propto P_{\mathrm{ph}}^{\gamma} \qquad (1)$$

According to the line fitting data which shown in figure 5b, two values of γ could be obtained, and γ=1.14 and γ=0.90 for the relatively low and high power density, respectively. It is probably that, the smaller value at high power density is resulted either from the buildup

of space-charges, or bimolecular recombination of photo holes and photoelectrons.[12]

The active area (S) of our device is 0.5 mm^2, thus the power of incident light (P_{ph}) could be calculated according to the active area multiply by power density, and the responsivity (R) and external quantum efficiency (EQE) could be calculated according to the following equations:

$$R = \frac{I_{ph}}{P_{ph}} \tag{2}$$

$$EQE = \frac{hc}{q\lambda} * R \tag{3}$$

In the above equations, I_{ph} represents the photocurrent, P_{ph} represents the power of incident light, h, c, q and λ are Planck Constant, the velocity of light in vacuum, the electron charge and the wavelength of incident light, respectively. The values of R and EQE as a function of power density are shown in **figure 6**. The values of responsivity are range from 48−60 mA·W^{-1}, and the values of EQE are range from 9−11%. When the power density is 100 mW·cm^{-2}, the device has the highest responsivity and external quantum efficiency.

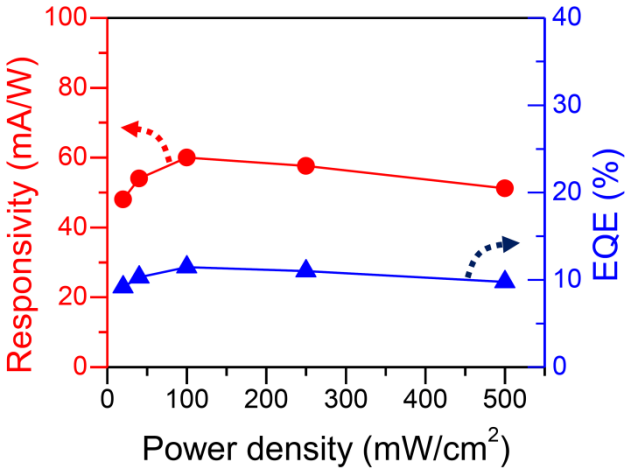

Figure 6. The values of the responsivity (R) and external quantum efficiency (EQE) as a function of power density

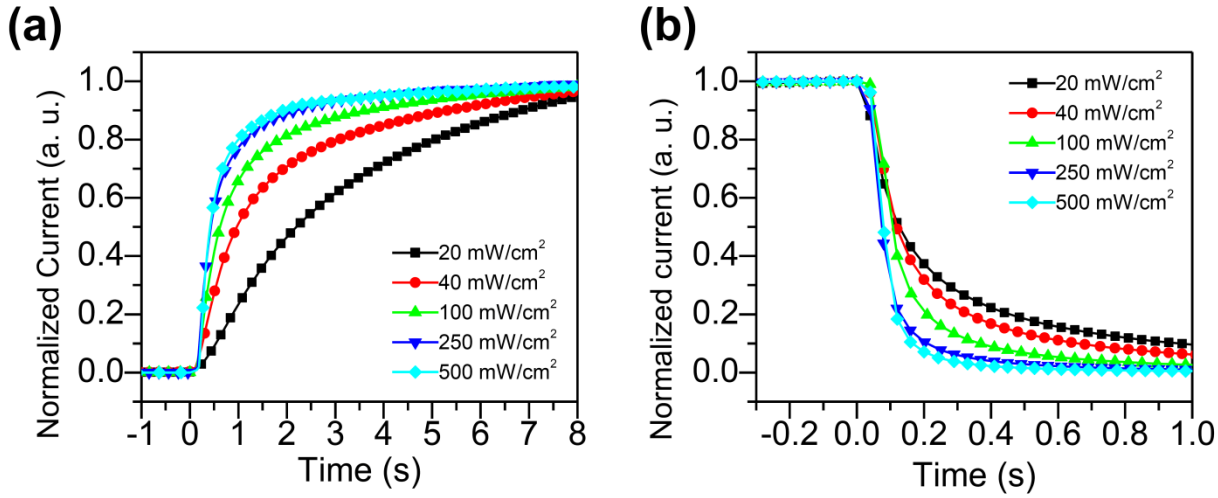

Figure 7. (a) the rise and (b) decay edges of the current response under different power density

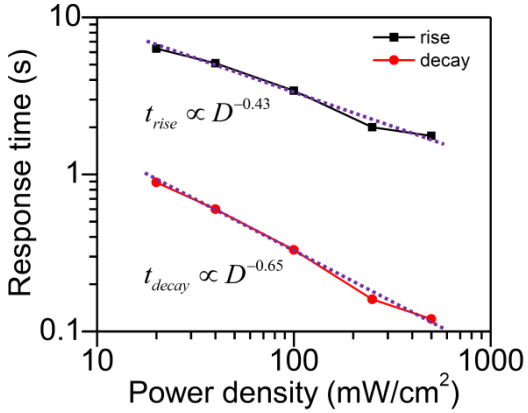

Figure 8. Response time as a function of power density

Figure 7a and **7b** show the rise and decay edges of the current response under different power density, respectively. The rise time and decay time decrease with the increasing of the power density. The same phenomenon could be found in a research of photoconduction properties of transition metal oxide semiconductors, and it probably due to the photocarrier collection efficiency which is very sensitive to the power of incident light.[34] The response time as a function of power density was shown in **figure 8**. According to the results, the response time decreases with an increase of power density, and an inverse power-law can also been found for the *t-D* relationship, i.e., $t \propto D^{-\alpha}$, where α=0.43 and 0.65 for the rise time and decay time, respectively. (*D* represents the power density).

Responsivity	Dark Current	rise time	decay time	Ref.

(mA•W^{-1})	(nA)	(s)	(s)	
12	100	–	–	[7]
0.3	60	–	–	[10]
33	40	8*10^{-8}	8*10^{-8}	[11]
350	10^{5}	5.7*10^{-4}	5.1*10^{-4}	[12]
60	600	–	–	[14]
30	100	4.5*10^{-8}	3.8*10^{-8}	[15]
60	1	2.2	0.29	This Work

Table 1 Comparison of the photoresponse parameters between the CuPc based photodetectors

Table 1 shows the comparison between our device and typical high performance CuPc based photodetectors. It can be seen that rare photodetectors could achieve both high responsivity, low dark current and fast response at the same time. In our research, a CuPc photodetector is prepared using a facile method, and it has a relatively high responsivity and low dark current. The poor response speed is probably due to the poor crystalline of CuPc film and the simple structure of the device. Nevertheless, our research gives an alternative way to fabricate the phthalocyanine photodetectors especially for the phthalocyanine with poor solubility.

4. Conclusion:

In this paper, a facile method is proposed to prepare CuPc film photodetector. CuPc films were deposited on the substrate of *c*-sapphire by a multiple drop casting method. This method could prepare phthalocyanine films from a relatively dilute solution, and give an alternative way to fabricate the phthalocyanine photodetectors especially for the phthalocyanine with poor solubility. Furthermore, our device has a relatively high responsivity (60 mA·W^{-1}) and low dark current (1 nA).

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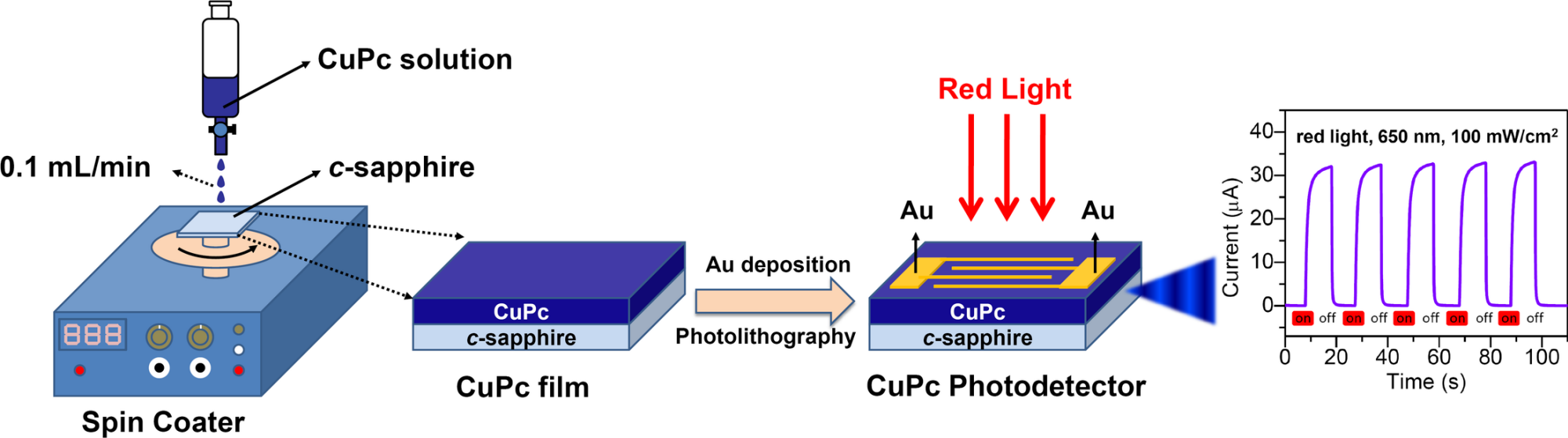
- Phthalocyanine based organic photodetectors have received more and more attention

- The common methods to fabricate phthalocyanine films have some disadvantages, such as expenses of high operation temperature, ultrahigh vacuum processing, large materials loss, complicated operation, large energy consumption and poor solubility.

- A multiple drop casting method could prepare phthalocyanine films from a relatively dilute solution, and it is a resin-free way which is different from traditional solution process

- Photodetector based on phthalocyanine film prepared by multiple drop casting method has a relatively high responsivity (60 $mA \cdot W^{-1}$) and low dark current (1 nA)


CuPc solution
0.1 mL/min
c-sapphire
Spin Coater
CuPc
c-sapphire
CuPc film
Au deposition
Photolithography
Red Light
Au
Au
CuPc
c-sapphire
CuPc Photodetector
red light, 650 nm, 100 mW/cm²
Current (μA)
Time (s)
on off on off on off on off on off