High performance, self-powered photodetectors based on a graphene/silicon Schottky junction diode†

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Electron–hole pair separation and photocurrent conversion at two-dimensional (2D) and three-dimensional (3D) hybrid interfaces are important for achieving high performance, self-powered optoelectronic devices such as photodetectors. In this regard, herein, we designed and demonstrated a graphene/silicon (Gr/Si) (2D/3D) van der Waals (vdW) heterostructure for high-performance photodetectors, where graphene acts as an efficient carrier collector and Si as a photon absorption layer. The Gr/Si heterojunction exhibits superior Schottky diode characteristics with a barrier height of 0.76 eV and shows good performance as a self-powered detector, responding to 532 nm at zero bias. The self-powered photodetector functions under the mechanism of photovoltaic effect and exhibits responsivity as high as 510 mA W⁻¹ with a photo switching ratio of 10⁵ and a response time of 130 μs. The high-performance vdW heterostructure photodetector demonstrated herein is attributed to the Schottky barrier that effectively prolongs the lifetime of photo-excited carriers, resulting in fast separation and transport of photoexcited carriers. The self-powered photodetector with superior light harvesting and carrier transport behaviour is expected to open a window for the technological implementation of Si-based monolithic optoelectronic devices.

1. Introduction

Semiconductor photodetectors, which are optoelectronic devices that harvest photon energy through distinct electronic processes, with ultra-fast response and high responsivity are of tremendous societal importance in many applications including optical communication, sensing, motion detection, missile warning and biomedical imaging. Particularly, there is a surge of interest towards developing self-driven or self-powered photodetectors that can operate unconventionally without consuming external power. In general, a Schottky photodiode can function under both photovoltaic mode (at zero bias or self-powered) and photoconductive mode (under reverse bias). Schottky junction-based self-powered photodetectors inherit numerous merits such as an easy and inexpensive fabrication process, simple device architecture, straightforward integration with CMOS technologies, self-powered operation and broadband photodetection.

Graphene possesses several outstanding electronic and optical properties because of its linear dispersion relation near the K point in the reciprocal lattice. In particular, the prospective combination of ultra-high carrier mobility, broadband absorption from the entire ultraviolet to the far infrared region, and ultra-fast luminescence make graphene an appealing material for photonic and optoelectronic devices. However, the major challenges for the development of graphene-based photodetectors are associated with the intrinsic properties of graphene, such as relatively low absorption cross-section, zero-band gap, lack of gain mechanisms, and ultra-fast carrier recombination on a time scale of picoseconds. Such factors restrict the photoresponsivity of pure graphene-based photodetectors to a very low value of 10⁻⁶ A W⁻¹. However, a unique feature of graphene is its ability to form a Schottky junction when coupled with a more strategic semiconducting material (such as Si, SiC, GaN, ZnO, GaAs, Ge, MoS₂ and some of their nanostructures). In such heterostructures, a strong vdW contact offers a Schottky barrier (SB), which can overcome the flaws associated with conventional graphene photodetectors, allowing efficient photodetection with demonstrated applications such as barrirors, solar cells, broadband photodetectors and chemical sensors.
Using these vdW heterojunctions, we can not only take advantages of the atomically thin nature of graphene (excellent tunability of the Fermi level by a single anode–cathode bias, high current carrying density (10^4 A cm^-2) and broadband light absorption), but also adopt the merits of SB (efficient photoexcited carrier separation under zero bias). Consequently, we can realize Gr/Si vdW heterostructures as self-powered and high-performance photodetectors.

Due to an appropriate design of band alignment and uniqueness of different materials, graphene–semiconductor contacts are pervasive and promising in semiconductor technologies and are projected to potentially reinstate ultra-shallow doped regions in state-of-the-art CMOS technologies. Moreover, such heterostructures are used as a key component of high-performance nanoelectronics devices, which unambiguously determine the performance of the device in areas such as switching speed, open circuit voltages in solar cells and the transistor ON-state current and ON/OFF ratios. Importantly, utilizing graphene as a transparent electrode would help to achieve high efficiency tunable solar cells and photodetectors. In other words, we can tune the potential barrier at the Gr/Si interface via the single anode-cathode bias or external gate voltage to graphene since the Fermi energy of graphene is directly linked with the external voltage.\(^\text{14,19,20}\)

Among the graphene–semiconductor devices developed in recent times, the Gr/Si vdW heterojunction has fascinating advantages in the desired production of cost-efficient, straightforward and high throughput solar cells and photodetectors. The Gr/Si heterostructure devices could act as a hybrid system by converting photons into electric current by utilizing graphene as an efficient carrier collective and Si as a high photon absorption layer to realize a highly efficient photodetector. Clearly, the transformation of photoexcited carriers into electric current at a Gr/Si heterojunction can be accomplished by exploiting the built-in field that accompanies SB to achieve fast separation and transportation of the photogenerated electron–hole pairs. Furthermore, an intrinsically designed SB between graphene and silicon compromises low dark current and limited short noise, which renders effective photoswitching ability (\(>10^3\)).

In the light of these findings, numerous studies have been conducted to fabricate Gr/Si-based Schottky junction photodetectors and considerable findings have been realized in the past few years. Chen et al.\(^\text{21}\) revealed the Schottky diode behaviour of the Gr/Si junction and subsequently, Tongay et al. improved the transport characteristics of the junction.\(^\text{19}\) An et al.\(^\text{22}\) demonstrated tunable photosresponsivity up to 0.435 A W\(^{-1}\) for 850 nm irradiation. Zhu et al.\(^\text{23}\) employed a vertical structure photodetector based on reduced graphene oxide and Si with a maximum responsivity of 63 mA W\(^{-1}\) and a photo-to-dark current ratio of 6.25 \(\times\) 10^4 for 445 nm excitation. Bartolomeo et al.\(^\text{20}\) demonstrated a novel SB device utilizing graphene on a matrix of nanotips patterned on the n-Si substrate. The textured device structure improved light absorption and the electric field at the apex of nanotips due to enhanced charge separation and transport, resulting in a high responsivity of 3 A W\(^{-1}\) for white LEDs.\(^\text{20}\) It has been shown that coupling the Si quantum dots with graphene can improve the responsivity of the bulk Si-based Schottky junction up to 0.495 A W\(^{-1}\).\(^\text{24,25}\)

In spite of many reports, most of the above-mentioned devices lack one or more of the following abilities: photodetection in the visible-light zone with high responsivity and photo-to-dark current ratio, fast response and decay time, self-powered operation without any external driving force, and a simple device fabrication process. Hence, the goal of the present study was to fabricate a hybrid system based on the combination of graphene and silicon that can work as a well-defined Schottky junction diode. In addition, we show that the Gr/Si Schottky junction diode performs well as a self-powered detector that responds to 532 nm illumination without any bias owing to the realization of SB. Furthermore, we extensively focused on the realization and characterization of the Gr/Si interface toward the proper understanding of the device physics at the junction interface, which governs the photocurrent mechanism. Thus, the present study advances Gr/Si-based self-powered photodetectors with the potential advantage of impressive consistency, long-term repeatability and robustness for future optoelectronic device applications.

2. Experimental procedure and device fabrication

A self-limiting growth of BLG with a high surface coverage of 90% was grown by APCVD at 970 °C using a C\(_2\)H\(_2\) precursor on a copper foil. The graphene film was transferred directly onto the n-Si substrate after removing the native surface oxide layers with HF to make direct contact with Si. The wafer used in this study was lightly doped n-Si with a resistivity of 1–10 Ω cm. The surface features of the transferred graphene were examined using FESEM-Carl Zeiss-Sigma. The features and stacking dimension of the graphene layer was confirmed by confocal Raman spectroscopy at an excitation wavelength of 532 nm. The electrical properties were measured at room temperature and ambient conditions using an Everbeing probe station interfaced with a Keithley 4200 source meter. A good Ohmic contact to the graphene was achieved by depositing Ti (10 nm)/Au (85 nm) using e-beam evaporation. The active area of the device is 2 mm \(\times\) 2 mm; graphene is in intimate contact with Si via a vdW contact. Rear contact with a thickness of Ag (85 nm) was deposited on the back surface of Si. The photodetector properties of the Gr/Si heterojunction were characterized by the laser at a wavelength of 532 nm. A continuous-wave 532 nm semiconductor diode laser source was used as an excitation source and the incident power of the laser was calibrated using a laser power meter. Transient photoswitching measurements were performed using a chopper to modulate the 532 nm-laser and the signal was collected using a digital storage oscilloscope.

3. Results and discussion

3.1. Graphene/n-Si vdW heterojunction as Schottky diode

First, we examined graphene before and after transfer onto Si substrates using FESEM. Fig. S1 (ESI†) shows the FESEM...
images of as-grown graphene on a Cu foil, revealing the well-optimized growth of high-quality graphene via a self-limiting approach. The schematic structure of the Gr/Si device is shown in Fig. 1a. Fig. 1(b) shows the FESEM images of transferred graphene without any cracks and apertures over the transferred area, revealing the excellent transfer process. However, the observed random wrinkle features were formed during the transfer process and did not seem to affect the photoresponse. Using Raman spectroscopy, we resolved three major bands, namely, D band at 1350 cm⁻¹, G band at 1585 cm⁻¹ and 2D band at 2700 cm⁻¹, which are attributed to sp² bonded C–C, confirming graphene formation, as shown in Fig. 1(c). The relatively small intensity of the defect band suggests a high crystalline quality of BLG even after the transfer process. 26 BLG with Bernal stacking was validated from the asymmetric nature of 2D band, which is well fitted with four Lorentzian peaks and a 2D/G band ratio of >1.1, as shown in Fig. 1(d).

The current density–voltage (J–V) characteristics of the Gr/Si heterostructure exhibits the strong rectifying character of a well-defined Schottky diode, as shown in Fig. 2(a). In a Schottky diode configuration, the SB, the depletion width, and the built-in field are regarded as characteristics of the diode and the key factors in achieving high-performance devices. 27 Therefore, we extensively studied the electrical characteristics of the Gr/Si interface to understand the fundamental charge carrier transport mechanism across the junction interface. Due to the distinct work functions of graphene and Si, the electrons in Si lower their energy by flowing to graphene. This is because the Fermi energy of Si must equilibrate with the Fermi energy of graphene, leaving behind the positively charged states in the depletion region of the Si side. As a result, an upward bending of Si bands occurs to form a built-in electric field in the vicinity of the Gr/Si heterojunction interface. The specific band structure of the Gr/Si Schottky diode at thermal equilibrium is schematically shown in Fig. 2(b). In forward (reverse) bias, the potential barrier for majority carriers to flow from Si to graphene (from graphene to Si) is reduced (increased), resulting in high (reduced) dark current.

The rectification behavior at the vdW contact can be explained by the influence of SB between graphene and Si, which may be described using thermionic emission theory, where the large forward current is a consequence of lowering the band bending and is expressed as

\[
I = A^* T^2 \exp \left( \frac{q\phi_b}{k_B T} \right) \left[ \exp \left( \frac{qV}{n k_B T} \right) - 1 \right]
\]

where \(A^*\) is the effective junction area (~4 mm²), \(A^*\) is the Richardson constant (112 A cm⁻² K⁻²), \(T\) is the temperature (300 K), \(\phi_b\) is the zero-bias Schottky barrier height (SBH) at the junction between G and Si, \(k_B\) is the Boltzmann constant, \(q\) is the electronic charge, and \(n\) is the ideality factor. Using exponential fitting, the zero-bias barrier height of the Schottky junction in the dark was deduced to be 0.76 eV.

It should be noted that unintentional formation of interface native impurities will result in a high density of states at a given energy in Si, which could lead to Fermi level pinning (FLP) and result in a significant discrepancy in barrier height. 28 However, we exclude the prospect of FLP, considering the ideal, defects-free and good quality interface with saturated dangling bonds. Graphene grown in a typical CVD system is more vulnerable to the absorbance of water or oxygen molecules in the air and impurities during the transfer process, which could shift the Fermi level \(\left( E_F \right)\) below the Dirac point \(\left( E_D \right)\), forming an unintentionally hole-doped (p-type) graphene. Assuming the work function of graphene (4.8 eV) and the electron affinity of Si (4.05 eV), the measured SBH is identical with the theoretical prediction based on the Schottky–Mott relation. 19

**Fig. 1** (a) Schematic diagram of the Gr/Si vdW heterojunction device. (b) FESEM image of large area graphene transferred onto Si, which demonstrates the maximum uniformity over the active area. (c) Raman spectrum of graphene transferred onto a Si substrate. (d) Evidence of AB-stacked bilayer graphene via band splitting.

**Fig. 2** (a) J–V characteristics of the Gr/Si vdW heterojunction; inset shows the non-saturation reverse current leakage as a function of applied bias. (b–d) Schematic energy band diagram of the Gr/Si interface under (b) zero bias, (c) reverse bias and (d) forward bias. The tunable Fermi level with respect to the applied bias is clearly depicted and the green arrow shows the direction of the current flow.
In addition, under reverse bias conditions, i.e., when graphene is negatively charged, our experimental results reveal that the current increases monotonously with the increase in magnitude of the bias and does not reach complete saturation behaviour, as shown in the inset of Fig. 2(a). The observed non-saturation reverse current characteristics can be understood via the tuning of the graphene Fermi level with the application of a single anode–cathode bias. For instance, application of a reverse bias to graphene raises its Fermi energy and also reduces the work function, which become more pronounced as the reverse bias increases, thus causing the variation in SBH, as schematically shown in Fig. 2(c) and (d). This indicates that the barrier for electrons to flow from graphene to Si is reduced as the bias voltage is increased, thus preventing saturation of the reverse current.\(^{19,29}\) However, this behaviour cannot be observed in a conventional metal-based semiconductor Schottky junction owing to their fixed Fermi levels and SB.\(^{30}\) Moreover, the ideality factor (\(\eta\)), which is a figure-of-merit, indicates the quality of the diode and measures how defects and/or other factors can mediate the current transport at the Gr/Si Schottky junction. \(\eta\) is extrapolated to be 6. An ideality factor greater than 1 (\(\eta > 1\)) in graphene-based devices is predominantly ascribed to the bias dependent work functions of graphene and also to barrier inhomogeneities.

### 3.2. Self-powered and photodiode characteristics of the Gr/Si vdW heterojunction

Following the primary diode characterizations, we turned our attention to the photodiode characteristics of the Gr/Si Schottky junction by illuminating a 532 nm laser over a range of various powers on the graphene layer. In general, the prospective combination of the extraordinary charge carrier properties and the wide bandwidth absorption of graphene allows for the Gr/Si Schottky diode to work in both the energy gap excitation mode (i.e., when the incident photon energy is greater than the semiconductor band gap energy) and the internal photoemission mode (i.e., when the incident energy is greater than the SB energy and less than the semiconductor band gap energy).\(^{11}\) The absorbed photons having energy gap excitation mode could create electron–hole pairs in both the graphene and the Si side. However, due to the relatively low absorption coefficient for visible light and the ultrafast recombination rate of photogenerated carriers, graphene does not act as an absorption layer to separate the carriers and generate the photocurrent. Thus, the spectral response of the Schottky junction is limited to above the band gap of Si where the maximum absorption can be seen in Si in the visible range. Hence, when a laser at 532 nm is exposed to the Gr/Si Schottky junction, most photons are likely to penetrate graphene and absorb in Si. As a result, incident photons create an electron–hole pair in the vicinity of the Si depletion width. The vdW contact of graphene and Si is favorable for delivering a relatively large built-in field (\(V_{bi} \sim 0.5–0.7 \text{ eV}\)) along with a depletion region, which effectively precludes the charge recombination mechanism as the photo generated carriers must exit from the generation region before they recombine. As a result, the holes and electrons can spatially be separated and pulled toward graphene and Si, respectively, by \(V_{bi}\), yielding an unprecedented photoresponsivity over a broad range.

On the contrary, if \(V_{bi}\) in the Schottky junction is low, the photogenerated carriers would easily recombine in the depletion region itself without contributing to external photocurrent in the circuit. As a consequence, there is no net increase in photocurrent in the forward region due to the deficient \(V_{bi}\) at the interface and changes in the polarity of bias, leading to inefficient separation of the exciton.\(^{32}\) Therefore, the forward photocurrent remains almost constant regardless of the illumination intensity. Clearly, the larger \(V_{bi}\) broadens the depletion region (\(W_{dr} \sim \sqrt{V_{bi}}\)), which is expected to effectively prolong the lifetime of the photoexcited carriers and favors more efficient charge separation and transport. Owing to sufficient \(V_{bi}\) at the interface, no external bias is needed to promote efficient and ultra-fast carrier separation, allowing the device to operate in a self-powered condition. The use of graphene is beneficial since it not only transports the incident photons to the active area, but also satisfies the basic need of transporting the photoexcited carriers to the external circuit under self-powered conditions. Moreover, no significant photocurrent could be generated from the internal photoemission mode due to the high energy of the incident photons. This implies that absorption in graphene may become apparent at low energies beyond \(\sim 1150 \text{ nm}\).

A remarkable photocurrent from the Gr/Si vdW heterostructure was observed, as shown in Fig. 3, indicating that the absorbed photons are efficiently converted into photocurrent. The influence of the photoresponse on various irradiation intensities was also investigated and plotted in Fig. 3(b). The photocurrent exhibits a fast and wide-range response to visible light illumination. The photocurrent increases with an increase in laser power, which is well consistent with the fact that the
amount of photogenerated carriers is nearly proportional to the absorbed photon flux. As the light intensity varied from several μW cm⁻² to 5.7 mW cm⁻², the short circuit current density increased monotonically from 31 μA cm⁻² to 1 mA cm⁻² and the open circuit voltage also drastically increased from 4 mV to 200 mV, indicating that a greater number of photo-excited carriers gets separated and collected as photocurrent (Fig. 3(c)). The conduction mechanism of the Gr/Si vdW heterostructure photodetector can be explained with the help of an energy band diagram, as shown in Fig. 3(d). Careful analysis of our findings indicates that the Gr/Si heterojunction functions under the physical mechanism of the photovoltaic effect (at zero bias conditions). It is clear that the photovoltaic effect utilizes the $V_{hd}$, induced by the SB in the interface region, to sweep away the band to band excited electron–hole pairs, thus giving rise to the photocurrent.

An important feature of a photodetector is its photoresponsivity, which is a measure of the electrical response of the device to incident photons, which is defined as

$$R = \frac{I_p}{P_{laser}}$$  

(2)

where $I_p$ is the measured photocurrent and $P_{laser}$ is the incident optical power. The responsivity at 532 nm was found to be $\sim 510$ mA W⁻¹, given by its $P_{laser}$ of 60 μW cm⁻² and $I_p$ of 31 μA cm⁻² (as shown in Fig. 4(a)), which is much higher than that of the previously reported vertical heterostructure device based on reduced graphene oxide and Si at 445 nm. There are several factors that can lead to a marked increase in the responsivity of our device. At first, the manifestation of SBH and the non-uniform coverage of graphene with a significant number of apertures yield an inhomogeneous heterojunction, which could promote bulk exciton recombination and hinder photocarrier transport, thus reducing the responsivity of the device. Furthermore, the reduced low trap states at the surface or interface, as a result of the high-quality and defect-free vdW heterojunction, would also influence the device performance.

When the illumination was ON, the photocurrent rose to a high value and then increased gradually to reach a maximum and stable value, as shown in Fig. 4(b) and Fig. S2 (ESI†). Such an increasing trend in current after reaching a high value is due to the lack of a charge recombination mechanism. However, when the illumination was OFF, the current returned to its low value very rapidly and reached saturation. The visible ON/OFF ratio of the Gr/Si vdW device reaches a maximum value exceeding $10^3$ at a light intensity of 5.7 mW cm⁻² under self-powered conditions and is nearly identical regardless of the switching cycles. The near zero variation in the ON/OFF photocurrent implies that there is no severe defects-assisted charge transfer process to promote the recombination process; thus, the number of separated electron–hole pairs remains identical over multiple cycles. As shown in Fig. 4(a), we plot the ON/OFF ratio of the Gr/Si vdW heterojunction as a function of incident optical power intensity under self-powered conditions, which shows a strong linear dependence on the light intensity and is expected to surge as the illumination intensity is increased beyond 5.7 mW cm⁻². Upon illumination under self-powered conditions, the responsivity of our device decreases with the increase in laser power, as shown in Fig. 4(a). This might be due to the fact that the density of the effective photoinduced state decreases as the light intensity increases owing to the presence of a small number of traps at the interface. The $J$–$V$ characteristics of the photodiode under illumination of 660 nm is shown in Fig. S2 (ESI†) and the responsivity was found to be slightly higher than that of 532 nm. Thus, the device is expected to demonstrate higher responsivity value at higher wavelength (but below the energy gap of Si) and low power regime if we consider that the quantum efficiency ($\eta_{eff}$) is constant, and the photoresponsivity follows the following equation:

$$R = \frac{\eta_{eff} \cdot A \cdot W}{hc}$$  

(3)

Briefly, the number of electrons (holes) generated and collected as photocurrent by photons is higher for longer wavelengths, thus leading to wavelength dependent photoresponsivity.

Furthermore, unlike in self-powered conditions, the visible ON/OFF ratio has a great impact on the bias voltage, where it reduces as the negative bias increases while keeping close to two orders of magnitude (ON/OFF = $\sim 10^2$). This might be due to the fact that as discussed earlier, the induced external bias controls the position of the Fermi level, which is proficient at modulating the Fermi level of graphene and does not allow the dark current to reach saturation under reverse bias. As a result, the dark current density significantly increased under negative
bias voltages. Such a non-saturating dark current or a reverse leakage current is likely to cause a drastic change in the ON/OFF photoswitch as the negative bias voltage is increased, as shown in Fig. 3(b).22 Furthermore, the responsivity of the device grows monotonically with the reverse bias voltage, but it is mostly driven by the position of the quasi-Fermi level of graphene and its relative position with respect to the quasi-Fermi level of holes in Si.22 This implies that the device can also work under a photoconductive mode (reverse bias mode) with high responsivity.

Another very important feature of the device is the response and decay time, that is, how quickly the device responds when an incident photon is turned ON or OFF. To ascertain this, transient photoswitching of the devices was performed with a chopper to modulate the 532 nm laser. The fast-varying photocurrent signal was collected using a digital storage oscilloscope. As shown in Fig. S4 (ESI†), the Gr/Si vdW photodetector exhibits a stable and repeatable photoresponse to 20 Hz incident photons. From the single standardised modulation cycle, the dynamic time response of the device can be calculated from 10% to 90% of the saturated value of the photoresponse, while the dynamic decay time can be calculated from 90% to 10% of the peak value (Fig. 4(c) and (d)). The transient photoresponse of the Gr/Si vdW photodetector shows up over the sub-millisecond scale with high precision with a response time and decay time of 130 µs and 135 µs, respectively. Moreover, the device exhibits a relatively fast OFF transient similar to the ON transient, implying that a very fast recombination occurs without any charge transfer effect.33 The observed response and decay time is significantly better than the previously reported photodetector based on graphene and Si heterostructure upon illumination with a 890 nm laser.28 Such a vast enhancement in the photoresponse could be associated with the built-in field, which can facilitate an effective and a very fast separation of a large amount of photoexcited carriers.34 The ON–OFF switching of the device is very fast and reversible over numerous cycles, thus illustrating the impressive consistency, long-term repeatability and robustness of the device. In addition, the photo-stability analysis of the device was performed, as shown in ESI† Fig. S5. A persistent trend with near zero change in photoresponse was observed for more than 500 s, which clearly reveals the long-term stability of the Gr/Si heterojunction device. Eventually, considering the responsivity, reproducibility and speed of the device, we anticipate that our device could strongly meet the maximum requirements of a photodetector for practical applications such as high response speed, self-driven operation, high ON/OFF ratio, excellent stability, low cost fabrication and good reproducibility.

4. Conclusions

A self-powered photodetector with improved responsivity, improved photo-to-dark current ratio and fast response to visible light (532 nm) was designed and demonstrated using a CVD-grown Gr/Si vdW heterostructure. The Gr/Si vdW Schottky junction delivers a superior SBH of 0.76 eV, which is strong enough to separate the photoexcited carriers, resulting in unprecedented device performance. A maximum photoresponsivity of 510 mA W⁻¹ was observed for the low incident light intensity of 60 µW cm⁻² under self-powered conditions. A photocurrent rectification ratio of 10⁵ with a very fast response time (as short as 130 µs) was observed. Our Schottky junction satisfies the maximum requirements of a photodetector for practical applications such as high response speed, self-driven operation, high ON/OFF ratio, excellent stability, low cost fabrication and good reproducibility.

Conflicts of interest

There are no conflicts to declare.

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Notes and references

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