Microwave surface transport in narrow-bandgap PdSe₂-MOSFETs

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Abstract

Group-10 transition-metal dichalcogenides are puckered van der Waals semiconductors, with a narrow bandgap, envisioned for ultra-broadband infrared (IR) detection. To assess their dynamical transport properties we investigate PdSe₂ n-MOSFETs by using microwave admittance spectroscopy. We report on surface channel inversion-depletion-accumulation behaviors with a depletion length of 15 nm, a mobility of 110 cm² V⁻¹ s⁻¹, and a bulk bandgap of 0.15 eV. Our 10 µm long devices have an electronic cut-off frequency in the GHz range promising a large gain-bandwidth product, competitive with that of III–V (InAs) and II–VI (HgCdTe) devices. The integration of bulk absorption and surface readout in PdSe₂-MOSFETs is a monolithic geometry suitable for fast IR detection in the application-rich 1–10 µm range, which includes molecular spectroscopy, atmospheric communications and thermal sensing.

1. Introduction

Beyond the mostly investigated group-6 transition-metal dichalcogenides (TMDs; MoS₂, MoSe₂, WSe₂) [1, 2], noble-metal-based group-10 TMDs (PtSe₂, PdSe₂, PdT₂) have recently attracted attention due to their high carrier mobility and thickness-dependent bandgap, inherited from their puckered 2D pentagonal structure [3]. As a representing member, PdSe₂ is air-stable, easily exfoliated, and theoretically possesses an indirect bandgap of 1.3 eV in single layers that vanishes in few-layers and bulk crystals [4–6]. The transport band gap of bulk PdSe₂ is largely unknown experimentally: recent studies have estimated an optical band gap on the order of 0.5 eV [7], contrasting with ab initio predictions for the electronic band gap smaller or on the order of 0.2 eV [4, 6]. With respect to 3D semiconductors used for IR detection, the predicted bandgap is smaller than that of InAs (0.354 eV) and comparable with that of HgCdTe ternary alloys (0.1–0.3 eV). Among van der Waals materials, PdSe₂ photo-transistors can be regarded as semiconducting alternatives to semimetallic graphene photodetectors [8], including mid-infrared (IR) photo-transistors [9] and near-IR photo-mixers [10]. The predicted PdSe₂ bandgap is demarcated by a valence band whose maximum lies at the Γ point, and a conduction band whose minimum lies in the vicinity of the S point, both bands are characterized by an effective mass \( m^* \sim 0.2m_0 \) [11]. Alike PtSe₂ [12] or PdT₂ [13] phototransistors, few-layer PdSe₂ field-effect transistors (FETs) have been investigated, showing a large mobility \( \mu > 100 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1} \) [4, 5], and a broadband photoresponse from visible to the near infrared (NIR) range [7, 14]. Remarkably, the photoresponse can be extended to the long-wavelength mid-infrared (MIR) range in quasi-bulk samples [15, 16]. PdSe₂ mobility remains impurity limited and likely to improve with progress in material growth up to phonon-limited values [11]. Large among TMDs, it cannot surpass that of narrow bandgap semiconductors (~30 000 cm² V⁻¹ s⁻¹ in [17]). We show here that it is sufficient to secure a GHZ electronic response. The ultra broadband optical
gain, in the wavelength range 0.4–8 µm \[15, 16\], and the large NIR gain $G_{\text{NIR}} \sim 700 \text{ A W}^{-1}$ \[15\], when combined with a large electronic bandwidth, are assets of bulk PdSe$_2$ photo-transistors \[18\]. In this work, we provide a comprehensive study of electronic transport of PdSe$_2$ transistors.

Specifications for a fast and broadband IR phototransistor include: a narrow band gap material of sufficient thickness, wavelength compatible device dimensions, and a fast photo carrier collection and detection. High-mobility MOSFETs fulfill these constraints in combining a quasi-intrinsic bulk absorption with an efficient charge collection in the surface layer followed by a fast diffusion to the contacts. The latter mechanism limits their electronic cutoff frequency and photo-detection speed. We study Metal-hBN-PdSe$_2$ transistors. Due to bulk screening, DC transport in thick PdSe$_2$ (thickness $t_{\text{PdSe}} \gtrsim 100$ nm) is qualitatively different from that of few layers conventional (unscreened) PdSe$_2$ 2D-FET devices ($t_{\text{PdSe}} \lesssim 20$ nm) with a conductivity that vanishes in the bandgap. The thick PdSe$_2$ transistors are candidate MOSFETs\(^6\) which are van der Waals variants of conventional 3D semiconductor MOSFETs with a surface-layer dominated transport \[17\], and a semiconducting bulk. Compared to conventional few layer PdSe$_2$ 2D-FETs, PdSe$_2$ MOSFETs benefit both from the more efficient absorption of incoming light by the bulk material, and from the natural protection of the conducting buried surface layer from the environment. In this letter we focus on the low-bias electronic characterization of bulk PdSe$_2$-MOSFETs, equipped with low Schottky-barrier (SB) Pd contacts \[19\], and compare them with few-layer transistors. PdSe$_2$-MOSFETs exhibit ambipolar transport, ensured by either p-type or n-type electrostatic doping regimes, separated by a channel depletion regime; that we define in the following with respect to an estimated residual bulk n-type doping as the characteristic surface inversion-depletion-accumulation regimes. This ambipolarity is partially warped towards low temperatures with the emergence of Schottky-contacts in the (n-doped) accumulation regime. From the thermal-activation analysis of the bulk conductance at surface depletion and of the SB resistance, we obtain two consistent estimates, $\Delta_{\text{bulk}} = 0.14 \pm 0.01 \text{ eV}$ and $\Delta_{\text{SB}} = 0.15 \pm 0.01 \text{ eV}$, of PdSe$_2$-MOSFET transport bandgap. These estimates contrast with the band gap value reported by literature ($\Delta_{\text{bulk}} \simeq 0.5 \text{ eV}$) \[5, 7\], essentially based on optical techniques, preserving dominant photon absorption mechanisms different from the indirect fundamental electronic band gap absorption. Dynamical transport properties are characterized by microwave admittance spectroscopy, which is a broad-band (100 kHz to 10 GHz) variant of standard semiconductor gate capacitance techniques \[19\]. Note that high-frequency characterization of PdSe$_2$ 2D-FETs is precluded by the large channel resistance in depletion regime. Thanks to efficient SB capacitive coupling, microwave measurements allow overcoming SB-resistance and revealing the ambipolar channel conductance and field-effect gate capacitance of PdSe$_2$-MOSFETs as function of doping and temperature. The resistance-peak gate voltage $V_{\text{CNP}}$ indicates a small bulk n-doping, corresponding to a donor density $N_D \lesssim 10^{17} \text{ cm}^{-3}$, presumably due to residual Se vacancies, which is a general trend in the literature on dichalcogenides \[20\]. However, in the series described in supplementary informations (available online at stacks.iop.org/2DM/8/035035/mmedia) section 2 (figure S2, panel c), three samples show no noticeable bulk doping and two samples shows a small bulk p-doping. The large electron and hole mobilities, $\mu_e \simeq 110 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and $\mu_h \simeq 40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, secure a GHz bandwidth in our ten-micrometer-long n-MOSFETs suited for IR sensing. The capacitance-voltage $C_E(V_G)$ dependence confirms the MOSFET inversion-depletion-accumulation regimes and allows measuring the depletion-layer thickness $\delta_{\text{dep}} \simeq 15 \text{ nm}$. This value puts on quantitative basis the above distinction between 2D-FETs and MOSFETs based on DC transport. Finally the potential of PdSe$_2$-MOSFETs for fast IR detection is discussed.

2. Device fabrication and setup

PdSe$_2$ transistors are fabricated by exfoliating high-quality PdSe$_2$ crystals \[4\] grown by a self-flux method as described in \[4\], and transferring them onto large hBN flakes, exfoliated from high-quality hBN crystals grown under high pressure-high temperature as described in \[21\] acting as bottom-gate dielectric. Incipient to the stamping technique, the hBN thickness varies from device to device as specified in the supplementary informations (table S1 in section S12). Transistors are deposited on a gold bottom gate itself evaporated on SiO$_2$ substrate. They are embedded in 50 Ω coplanar wave guides (CPW) used for DC and RF characterization. Note that this RF-device geometry precludes 4-terminal DC transport measurement and direct separation of channel and contact resistance. PdSe$_2$ samples are chemically etched, when needed, to the final rectangular dimensions (length $L \lesssim 10 \mu$m and width $W \gtrsim 10 \mu$m) and equipped with low-resistance palladium (Pd) contacts using laser lithography and e-beam evaporation. The bottom gate is made of gold, which has a work function ($W_{\text{Au}} = 5.3 \pm 0.2 \text{ eV}$) close to that of Pd contacts ($W_{\text{Pd}} = 5.4 \pm 0.2 \text{ eV}$) \[23\] and of PdSe$_2$ valence band edge ($W_{\text{PdSe}} = 5.4 \text{ eV}$) \[3, 24\]. Devices show a year-long stability, in spite of the absence of top capping. PdSe$_2$ structural quality is assessed by Raman spectroscopy, exemplified in figure S11-a, with peaks

\(^6\) Literally MISFETs as the hBN gate insulator is not an oxide.
associated with optical phonon inelastic scattering at \( \hbar \omega_{\text{OP}} \approx 145 \text{ cm}^{-1} = 18 \text{ meV} \) \cite{4, 5, 25}.

We have investigated two series of transistors qualified as thin (\( t_{\text{PdSe}_2} \lesssim 20 \text{ nm} \)) and thick (\( t_{\text{PdSe}_2} \gtrsim 90 \text{ nm} \)). We focus here on one representative thick device (sample S43) shown in the inset of figure 1(a), with \( t_{\text{PdSe}_2} = 90 \text{ nm} \) (\( t_{\text{hBN}} = 34 \text{ nm} \)). Statistical relevance of S43 is illustrated in figures SI2(a–d) according to the room-temperature transfer characteristics and typical RF admittance spectra of the thick device series. For comparison, we show in the inset of figure 1(b) one representative thin device (sample S02) with \( t_{\text{PdSe}_2} = 9 \text{ nm} \) (\( t_{\text{hBN}} = 47 \text{ nm} \)). Their micrometer sizes, specified in the caption of figure 1, match IR wavelengths. Devices are characterized in a cryogenic DC-40 GHz probe station, their RF scattering parameters being measured over a 5-decade (70 kHz to 40 GHz) bandwidth with a vector network analyzer (Anritsu-MS4644B). Thru-lines and dummy structures are used for CPW and stray capacitance de-embedding purposes. From the full set of scattering parameters we extract the device admittance matrix, including the zero-bias gate-source contribution of interest for the physical characterization of MOSFETs \cite{22}.

3. Direct current characterization

As seen in figures 1(a) and (b), S43 and S02 devices show ambipolar transport at room temperature with similar resistances at large (positive and negative) doping, and a resistance maximum at a charge neutrality point (CNP) \( V_{\text{CNP}} \approx -1 \text{ V} \). The similarity of resistances in thin (S02) and thick (S43) doped \( \text{PdSe}_2 \)-transistors already suggests prominent screening effects in bulk samples. The temperature-independent CNP shift indicates a small (n-type) bulk doping. The resistance of thin S02 in figure 1(b) eventually saturates at \( 10 \text{ M}\Omega \) at channel depletion, due to the limited resolution of our setup.

The overall shapes of the transfer characteristics of S43 are qualitatively different. They show plateaus at positive gate voltage and low temperature
(figure 1(a)) that we attribute to the build up of a SB at the PdSe2-Pd interface due to work function mismatch between the Pd contact and the conduction band of PdSe2. This barrier is elusive at negative gate voltage, confirming the good alignment between the work functions of metallic Pd and bulk PdSe2. The transfer characteristic of S43 also shows a maximum at $V_g \simeq -1.6$ V for $T \gtrsim 150$ K, signaling the suppression of surface transport at surface charge neutrality; its temperature dependence is used below to estimate the bulk bandgap of PdSe2. Below 150 K the Schottky contact resistance dominates over bulk resistance (see the 130 K curve in figure 1(a)), blurring the resistance maximum and precluding its quantitative analysis in terms of bulk thermal activation.

To quantify SB effects in S43, we show in figure 1(c) an activation plot together with high-temperature fits (dashed lines) with the Richardson law [26]:

$$
R \times T = \frac{4\pi^2 e^4}{3k_B m^* AD} \exp \left( \frac{\Phi_{AC}}{k_B T} \right)
$$

where $\Phi$ the SB height, $A$ an effective contact area, and $D < 1$ an electronic transmission factor accounting for wave-function mismatch between metallic Pd and PdSe2. Note that a factor 2 has been introduced for the additive source and drain contributions. As seen in figure 1(c), the resistance obeys equation (1) above 100 K and saturates below presumably due to quantum tunneling. The high-temperature extrapolate, $\ln (R_{[40]} T_{[50]}) \sim 13$ (at $V_c = +10$ V), agrees with the prefactor in equation (1) taking $A \sim 0.1 \mu m^2$ and $D \sim 0.01$. From the slope we extract $\Phi(V_g)$, plotted in the inset of figure 1(c), which shows a stepwise increase from $\Phi_{INV} \sim -0.02$ eV in the inversion regime to $\Phi_{ACC} \sim +0.15 \pm 0.01$ eV in the accumulation regime (blue squares). The small negative SB extracted in the inversion regime is physically irrelevant and discloses the absence of SB between metallic Pd and PdSe2. From the sum $\Phi_{ACC} + \Phi_{INV}$ we infer a first estimate of PdSe2 bandgap: $\Delta_{SB} \sim (\Phi_{ACC} + \Phi_{INV}) \sim 0.15 \pm 0.01$ eV.

We have also performed an analysis of activated transport at depletion ($V_g \simeq -1.6$ V) using the standard law for intrinsic bulk semiconductors, $RT^{1/2} = A \exp (\Delta_{bulk}/2k_B T)$. The analysis is limited to a reduced temperature range $T = 150–300$ K where the resistance peak at surface charge neutrality is visible, securing a negligible contribution of contact SB effects. We deduce an estimate of the bandgap $\Delta_{bulk} = 0.15$ eV. A similar analysis performed on S02 yields a somewhat larger $\Delta_{bulk} \simeq 0.3$ eV. This blueshift of the electronic band gap is in line with the band gap value of 0.57 eV for a slightly thinner PdSe2 flake (6.8 nm-thick) reported in Ref. [7], also based on an Arrhenius plot. These values remain below the ultimate theoretical expectation $\Delta \simeq 1.3$ eV for single layer PdSe2. Based on these two independent determinations, we conclude that transport bandgap of semi-bulk PdSe2 does not completely vanish but rather saturates at $\Delta_{PdSe2} = 0.15$ eV, consistently with theoretical prediction in [4]. Note that while the SB is used here for bandgap characterization purpose, it can be minimized by appropriate contact engineering [27] for applications.

4. High-frequency admittance spectroscopy

Field-effect capacitance is a well established characterization technique of semiconductor devices [19]. It can be performed in capacitors (MOSCAPs) or transistors (MOSFETs). MOSCAPs are commonly made of silicon [28], or narrow-bandgap InAs [29] semiconductors, with a top gate and a thin oxide insulator, distinctively called MOS devices. The gate capacitance is generally measured between the top gate and the (weakly) doped bulk. Here we use an in-plane variant of admittance spectroscopy for a long channel transistor geometry, taking into account the Schottky contact impedance. This variant is similar to previous capacitor modeling that has been introduced for gate capacitance spectroscopy to study graphene electronic compressibility [22] and used to characterize high-mobility hBN-encapsulated graphene devices (including plasma-resonant capacitors) [30], the topological Dirac states at the surface of Bi2Se3-tetrasydnmites [33, 34], the interface states of topological heterojunctions between normal and inverted HgTe/CdHgTe semiconductors [31], as well as the edge states of strained HgTe quantum wells [32]. The complex gate-source admittance is measured over a broad frequency range allowing for a combined in-situ characterization of gate capacitance $C_G(V_g)$ and channel conductivity $\sigma(V_g)$. For ideal carrier injection in the channel, the admittance spectrum reflects a crossover from an elastic (1/$C_G$) dominated regime at low frequency to a lattice-friction dominated regime at high frequency. Such an RF crossover from elastic-dominated to friction-dominated response is quite general and has been previously reported in the context of vortex matter dynamics in superconductors [35–37]. In atomically thin 2D materials, screening is negligible and one can readily extract from $C_G$ the quantum compressibility contribution $C_Q = e^2/DOS$ in series with the geometrical capacitance $C_{geo}$. This provides in turn a determination of the chemical potential $E_F = \int (C_G/C_{geo} - 1) dV_g$ and the diffusion constant $D = \sigma/C_{geo}$, yielding a genuine scattering spectroscopy encoded in the $D(E_F)$ dependence [31]. For massive PdSe2 carriers, the 2D density of states in the surface layer, $DOS = m^* / \pi \hbar^2 \sim 10^{14}$ cm$^{-2}$ eV$^{-1}$, is large such that $C_Q \gg C_{geo}$, and the gate-voltage dependence of $C_G$ mostly stems from series dielectric channel contributions. These contributions read
The depletion layer depth at neutrality, and δ_{inv} that of the p⁺-n junction in the inversion regime.

Figure 2 shows typical RF admittance spectra of the PdSe₂ n-MOSFET S43. The gate admittance \( Y_{GS}(\omega) \) is reported both at room temperature \( T = 294 \text{ K} \) (d)–(f) and low temperature \( T = 176 \text{ K} \) (a,c). Negative and positive gate voltages correspond to (hole) inversion and (electron) accumulation regimes. A room-temperature depletion spectrum, for \( V_G \sim -1 \text{ V} \), is shown in panel (e). Also shown in the figures are theoretical fits with a 1D diffusion model [22, 32] based on a lumped element description (sketched in figure 2(b)):

\[
Y_{GS}(\omega) = \frac{1 - e^{-\gamma L}}{Z_D + Z_0} + \frac{1}{1 - \Gamma_D \Gamma_S} \exp(-\gamma L),
\]

where \( Z_0 = \sqrt{\frac{1}{r_{ew} f}} \) and \( \gamma = \sqrt{\frac{r_{ew} f}{\sigma_s}} \) are the line impedance and propagation constant, respectively, with \( r = R/L \) and \( c_l = C_{GS}/L \) the resistance and gate capacitance per unit length. \( Z_D \) and \( Z_0 \) are the source and drain contact impedance, and \( \Gamma_D = \frac{Z_D - Z_0}{Z_D + Z_0} \Gamma_S = \frac{Z_D Z_S - Z_0^2}{Z_D + Z_0} \) are the reflection coefficients at the drain and source contact respectively. The channel conductivity is modeled as \( Z_{SB} = (R_{SB}^{-1} + j C_{SB} f) \) and the channel admittance is \( Y_{GS}(\omega) \). The SB contribution appears as a low-frequency crossover at \( f_\text{inv} \sim 1/R_{SB} C_{SB} \sim 10 \text{ MHz} \) in (c).

Using a model fitting for each gate bias point (see the supplementary videos for T=176K and T=300K), we perform somehow a "radiography" of
in the depletion regime ($V_g \approx -0.7$ V), and drops down to $\sigma^{-1} \approx 5$ kΩ in the accumulation regime ($V_g = +10$ V), and $\sigma^{-1} \approx 50$ kΩ in the inversion regime ($V_g = -10$ V). Carrier mobility is fair, with $\mu_{acc} \approx 110$ cm$^2$ V$^{-1}$ s$^{-1}$ and $\mu_{inv} \approx 40$ cm$^2$ V$^{-1}$ s$^{-1}$, approaching the $\mu \approx 250$ cm$^2$ V$^{-1}$ s$^{-1}$ value reported in PdSe$_2$ FETs after annealing [4, 5]. Gate elastance (blue circles, note that data are shifted by $-0.25$ m$^2$ F$^{-1}$ for clarity) is characteristic of n-MOSFETs [19, 28, 29]: it saturates in the accumulation regime at the (nominal) gate insulator value $C_G \approx C_{ABN} \approx 0.74$ mF m$^{-2}$, takes a minimum $C_G \approx 0.52$ mF m$^{-2}$ at depletion, and increases to a plateau $C_G \approx 0.56$ mF m$^{-2}$ characteristic of an inversion regime [19]. From the additivity of insulator ($1/C_{ABN}$) and semiconductor ($1/C_{PdSe_2}$) elastance contributions, we deduce $C_{PdSe_2} \approx 1.75$ mF m$^{-2}$ at depletion and $C_{PdSe_2} \approx 2$ mF m$^{-2}$ in the inversion regime. Relying of the theoretical estimate $\epsilon_{PdSe_2} = \epsilon_0$ [38], we infer $\delta_{depi} \approx 15$ nm and $\delta_{inv} \approx 13$ nm. These small screening lengths, which are consistent with the Debye length $L_D = \sqrt{k_B T \epsilon_{PdSe_2}/e^2 N_D} \gtrsim 7$ nm for a bulk doping $N_D \lesssim 10^{12}$ cm$^{-3}$, secure the MOSFET interpretation of our 90 nm-thick 543 device, and justify the qualitative difference between our thin and thick transistor series.

The difference between 294 and 176 K data in figure 3 illustrates thermal effects on mobility and carrier density. As expected the capacitance gate modulation is more contrasted at 176 K due to a smaller contribution of the thermal carriers to screening. To quantify the first effect we cannot rely on direct measurement of $C_G$ at depletion due to the above-mentioned poor quality of the fits. Instead, we have used a three-parameters empirical interpolation formula, $C'(V_g) = C_{G}^{-1}(+10\text{ V}) - a\tanh\left[\left(V_g - V_G}\right)/b\right] + c\sqrt{b^2 + \left(V_g - V_G\right)^2}$ where $a$, $b$ and $c$ encode respectively for the capacitance step between depletion and accumulation, the amplitude and width of the capacitance dip at neutrality. Catching the room temperature $C_G(V_G)$ data (dotted blue line), it is used to fit 176 K data over the inversion/accumulation regimes (dashed blue line). With this procedure we infer $\delta_{inv} \approx 15$ nm and $\delta_{depi} \approx 20$ nm at 176 K. Temperature effect on $\sigma(V_G)$ is also instructive: room-temperature data are mainly upshifted by a constant amount $\Delta \sigma \approx 3$ µS with respect to 176 K data, implying that: (1) the mobility is essentially $T$-independent as expected for impurity scattering, and (2) the bulk contribution, which is suppressed at 176 K, is thermally activated at room temperature with a bulk resistivity $\rho_{bulk} = \Delta \sigma \approx 0.15$ $\Omega$. This value is consistent with a thermal carrier density $n_0(300\text{ K}) \approx 1 \times 10^{11}$ cm$^{-3}$ for a bandgap $\Delta_{bulk} = 0.15$ eV, and a bulk mobility $\mu_{bulk} \approx 20$ cm$^2$ V$^{-1}$ s$^{-1}$. The finite bulk mobility augers well of a fast bulk-surface dynamics of photo-carriers under illumination as anticipated in section [1].
5. Conclusion

In this letter we have reported on low-bias DC transport and RF gate capacitance spectroscopy in high-quality PdSe₂ crystals providing qualitative and quantitative evidence that semi-bulk van der Waals PdSe₂ transistors behave as genuine narrow-bandgap MOSFETs. This extensive electronic characterization allows down-pinning relevant material parameters such as PdSe₂ work function, transport bandgap, mobility, and thermal carrier density. In particular, we have deduced a fundamental electronic bandgap of 0.15 eV in agreement with the ab initio studies [4, 6]. Our work suggests that the optical gap of 0.5 eV for bulk PdSe₂ reported in the literature [7] originates from higher energy transitions and offers guidance for further IR optoelectronic applications, in particular for MIR detection. Finite bias properties (not reported here) confirm the quasi-intrinsic quality of our PdSe₂ transistors. However they call for a different out-of-equilibrium physics (phonon-induced velocity saturation, self heating, etc) which is beyond the scope of the present paper.

The demonstration of PdSe₂-MOSFETs opens new perspectives for applications of 2D materials in high-speed optoelectronics including ultra broadband optical detectors covering the MIR range. The present demonstration of GHz electronic bandwidth at the MIR-relevant 10 μm length scale, put in perspective with literature photo-detection measurements of an optical gain G = 700 AW⁻¹ [15], opens an avenue to PdSe₂ MOSFETs for room-temperature monolithic broadband and high-speed sensing, competing the extensively used CdHgTe detectors [39].

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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