Quantum advantage in a spintronic engine with coherently coupled ultrafast strokes using molecular superexchange

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Abstract

Recent theory and experiments have showcased how to harness quantum mechanics to assemble heat/information engines with efficiencies that surpass the classical Carnot limit. So far, implementing work-producing quantum resources has required atomic engines driven by external laser and microwave energy sources. We propose a spin electronic implementation that operates autonomously. Our concept heuristically deploys several known quantum resources upon placing a quantum-entangled chain of spin qubits formed by the Co centers of phthalocyanine (Pc) molecules between electron-spin selecting Fe/C₆₀ interfaces. Density functional calculations reveal that transport fluctuation strokes across the interfaces can stabilize spin coherence on the Co paramagnetic centers, which host spin swap engine strokes. Across solid-state vertical molecular nanojunctions, we measure large enduring dc current generation, sizeable output power above room temperature, and two quantum thermodynamical signatures. The Fe/C₆₀ interface’s record 89% spin polarization also enables a spintronic feedback and control over the flow and direction of charge current. Beyond these first results, further research into spintronic quantum engines, and retooling the spintronic-based information technology chain, could help accelerate the transition to clean energy.

Main Text

Classical engines convert heat into work by transferring heat from hot to cold thermal baths using a working substance that is sequentially thermalized to each bath. The engine’s entropy increases, and the temperature ratio of these baths determines the engine’s maximum efficiency at zero power output. This Carnot limit embodies the 2nd law of thermodynamics.

Quantum engines can surpass this limit by retooling its underlying concepts, using engine strokes that emulate Nature’s principle of least action. A stroke’s action is characterized by its duration and the rate at which it couples the WS to the baths. If the engine’s baths and WS are endowed with quantum properties, they can constitute so-called ‘quantum resources’ (QRs) that provide an additional work capacity of quantum origin called ‘ergotropy’.
So far, only engines with an atomic WS have demonstrated this quantum advantage using 1-10MHz electromagnetic strokes \(^3,^5,^6\), but required cumbersome equipment (microwave/laser sources), i.e. aren’t autonomous. Conversely, due to a mesoscopic WS that is much more prone to decoherence, on-chip electronic engines\(^{15,16}\) have thus far revealed sub-Carnot efficiencies despite much faster (2-10GHz electronic) strokes.

![Figure 1: a molecule-based spintronic quantum engine.](image)

(a) Spintronic implementation of the transport fluctuation (TF) stroke between the device electrode in its FM ground state and the spin states of the working substance (WS) ’s nearest PM center, mediated by a spintereface with full transport spin polarization. Quantum coherence and decoherence processes on the WS are shown. This stroke appears in the (b) overall engine schematic that also shows the spin swap (SS) strokes on a PM center’s spin states, and between the PM centers forming the WS, against thermal fluctuations for \(k_B T > \Delta\). Quantum resources are color-coded in green in panels a-b. See main text for details. (c) Spatial charge transfer maps across Fe/C\(_{60}\)/CoPc reveal sizeable hybridization on C\(_{60}\) (i.e. the spintereface) and electron tunneling between C\(_{60}\) and CoPc across an antibonding state. Green/cyan isocontours depict charge gain/loss of 0.0007 e/Å\(^3\). The AFM case is shown. (e) The antibonding state’s DOS around \(E_F\) reveals how the Co \(d_\uparrow\) and C \(p_z\) orbitals share a spectral feature that appears only in the spin \(\uparrow\) band. This illustrates the high spin polarization and bandwidth of the TF stroke (see panel a).

We propose spintronics\(^7\) as a potent scientific and industrial platform to achieve an electronic autonomous engine with a quantum advantage, and augment our prior spintronics-only description\(^17\) with QR concepts\(^1-^6,^8-^11\). These QRs heuristically originate from a tailored electronic interaction between a ferromagnetic (FM) thin film and the discrete spin states of paramagnetic (PM) centers, which form the engine’s working substance (WS). It is continuously driven out of equilibrium through quantum fluctuations that are autonomously rectified, thereby generating a spin-polarized current. We use this novel intersection between spintronics and quantum thermodynamics, alongside ab-initio calculations, to explain experimental spin-based energy output involving several quantum thermodynamical signatures.
Referring to Fig. 1(a), the (+M,0,0) magnetic orientation of the Fe layer sets a spin referential for other electronic interactions, starting with the spin-polarized charge transfer toward adjacent C_{60} molecules, which acquire a low density of spectrally narrow states with high spin polarization \( P \) at the Fermi level \( E_F \). These states, also termed a ‘spininterface’\(^{14}\), spintronically interact as a non-thermal bath with the WS’s PM centers\(^{2,18,19}\). Prior to electronic interaction, the S=1/2 spins borne by the Co \( d_z^2 \) orbital \( (x,y,z)=(0,0,\pm 1) \) on the Pauli sphere, see Fig. 1a) of Co phthalocyanine molecules are energetically degenerate. The electronic interactions mediated by transport fluctuations (TF) between the FM electrode and PM centers across the spinterface describe the engine’s TF stroke. The spinterface’s full spin polarization leads in the forward TF stroke (i.e. FM electrode \( \rightarrow \) PM center) to the injection into the WS of coherent spins along \((-1,0,0)\) (i.e. a QR\(^{2,19}\); coded in blue in Fig. 1b), against the WS’s orthogonal \((0,\pm 1,\pm 1)\) decoherence processes (in purple in Fig. 1a-b).

The reverse TF stroke, which also requires that the spin on the WS be \((-1,0,0)\), is a reduced channel of spin decoherence for the WS’s spins (Fig. 1b), and constitutes an autonomous measurement of the WS state, i.e. a QR\(^6\): the spinterface acts as an autonomous Maxwell demon\(^{20}\) that gains information on the WS and uses it as feedback to control the electron tunneling rate across the barrier. Overall, these TFs generate resistive losses due to spin scattering between the spinterface and FM film with very different \( P \). Synergistically, though, the Fe FM thin film sinks the resulting spin-based entropy (and thus heat) by returning to the FM ground state\(^{21}\).

To characterize our experiment’s TF stroke, we calculated (see Methods) the \( bcc \) Fe(110)/C\(_{60}\)(1ML)/CoPc(1ML) system. We observe a \( 1.231e^+ \) transfer from Fe that is delocalized onto C\(_{60}\) (Fig. 1c). A much weaker \( 0.03e^- \) transfer from C\(_{60}\) to CoPc occurs across an antibonding state. This tunneling-mediated electronic interaction results in spectral features across \( E_z \) only for spin \( \uparrow \) electrons with a minimum 45.3meV width that are shared both by the Co \( d_{z^2} \) orbitals of CoPc and the \( p_z \) orbitals of the neighboring C sites of C\(_{60}\) (Fig. 1d).

The resulting TF stroke, with a bandwidth-inferred frequency \( f_{TF} = 1.38 \times 10^{14} \) Hz, is thus fully spin-polarized as required (see Fig. 1a), and mediates a quantum correlation between the non-thermal bath and the WS. This non-Markovian regime of interactions, as a QR\(^6\), controls and optimizes the WS’s quantum thermodynamic state (i.e. a QR\(^3\)) by lifting the Co spin degeneracy by \( \Delta = 0.7 \) meV (Figs. 1b-c). This effective magnetic field \( H = 6T \), generated by spintronic anisotropy\(^{17}\), is much stronger than that for a noble metal spacer\(^{22}\) because it originates from C\(_{60}\)-mediated Fe-Co antiferromagnetic superexchange\(^{21}\).

The TF strokes quantum correlate with endmembers of the WS’s spin qubits borne by 1-D molecular chains with magnetic exchange energy \( J \)\(^{12}\). When \( k_BT < J \), the qubits become entangled and the WS’s Co paramagnetic fluctuations become antiferromagnetically (AF) coupled. Having the WS operate close to its magnetic phase transition\(^{18}\), and exhibit quantum entanglement\(^{4,11}\) below the transition, both represent QRs.

The spin swap (SS) strokes are the thermal fluctuations on and between the spin-split PM centers, and the ergotropic return from a non-passive (i.e. excited) state to the ground state\(^{20}\). Although stochastic in origin, these fluctuations can yield QRs in the WS by inducing coherence\(^4\) and by energizing the superposition of quantum states\(^3\). SS operations between qubits\(^4,5\) can also invert the spin population of the PM center involved in the TF stroke, i.e. generate a \((1,0,0)\) state. This non-passive state, and more generally other orthogonal decoherence processes in the WS along \((0,\pm 1,\pm 1)\) (in purple in Fig. 1a-b), are subjected to TF stroke-induced passivation (at \( \sim 10^{13} \) Hz, see above) to the \((-1,0,0)\) state (i.e. a QR\(^{20}\)). Other TF stroke paths are forbidden due to spin-conserved tunneling (see grey crosses in Figs. 1a-b). For \( k_BT \geq 0.7 \) meV, the SS stroke cutoff \( f_{SS} = 169.5 \) GHz, against spin-lattice and spin-spin decoherence processes\(^{24}\) (respectively labelled \( f_1 \) and \( f_2 \) in Fig. 1a). Since the faster \( f_2 = 1 \) MHz for CoPc at \( 7K \)\(^{24}\), and can increase \( \sim 100x \) at \( 300K \)\(^{25}\), we infer that \( f_2 \ll f_{TF}, f_{SS} \) throughout our experiments at \( 40 < T \) (K) < \( 360 \).
Thus, in our concept, which shares similarities with existing theory\textsuperscript{4,8,10,11}, the engine’s ultrafast TF and SS strokes are quantum-correlated and are much faster than the WS decoherence, i.e. implement a regime of small action required\textsuperscript{3} to attain a quantum advantage. The asymmetry in the TF stroke on either side of the WS (thick vs. thin transmission arrows of Fig. 1a) serves to 1) mitigate any cancellation effects on the WS of TF strokes from the two electrodes (see Fig. 1b), and 2) defines different electronic temperatures on either side of the WS (i.e. a QR\textsuperscript{10}). Overall, our engine’s operation involving several QRs differs from the spin-based classical thermoelectric effects of spin caloritronics\textsuperscript{26}.

Figure 2: Long-lived spontaneous electrical signals. Data on metallic nanojunction A: (a) time dependence of \(I_{sp}\) at H=0 (grey) and upon applying a H field orthogonal to the electrode magnetizations. (b) I(V) data at 240K. The magenta crosspoint represents the experimental (V,I) error. The top insets to panel (d) show the time dependence of \(I_{sp}\) and \(V_{Off}\), while the zoom around V=0 (lower inset) reveals an I(V) hysteresis that contains features with a sub-\(k_B T\) spectral resolution in (c) the current derivative dI/dV. Forward (black) and return (red) traces are shown. (d) Return dI/dV traces for 40K, 60K, 85K and 240K reveal essentially identical features with a sub-\(k_B T\) spectral resolution, i.e. the thermodynamical signature of a quantum resource.

Turning to experiments, our nanoscale vertical devices (see Methods) exhibit a large persistent non-zero spontaneous current \(I_{sp}\)~\(\sim\)10\(\mu\)A (see Fig. 2a for nanojunction A). Its amplitude is not strongly affected by intermittent sweeps of an external magnetic field up to 2T applied perpendicularly to the electrode magnetizations. This confirms that, in our implementation, the engine’s primary energy source is not the external applied magnetic field\textsuperscript{27}. We present in Fig. 1(b) repeated I(V) sweeps at 240K, from which we infer a slope resistance \(R_s=157\Omega\). From \(I_{sp}\) (t) (see top inset), we find at V=0 that the offset current \(I_{Off}=-26\mu\)A = \(I_{sp}\). This suggests that, once a bias voltage V has been applied, and on the timescale of hours, applying V=0 does not confer energy to the device. \(I_{Off}\) and the bias offset \(V_{Off}=4.05\)mV at I=0 (see top inset), are respectively 230x and 100x larger than the experimental offset errors observed for a 100\(\Omega\) calibrated resistance (magenta crosspoint in Fig. 2b).
The lower inset to Fig. 2(b) reveals a slight, hysteretic deviation from a linear response that depends on the sweep direction (red and black arrows). Within this 1.4mV bias window, the numerical derivative (see Fig. 2c) reveals features with an energy extent as low as 0.3meV despite an expected thermal smearing of 2-3k_BT upon transport, with k_BT=20.7meV here. This sub-k_BT spectral resolution is mostly unchanged upon reducing thermal fluctuations by a factor of 6, as are the main spectral features (denoted A, B and C in Fig. 2d).

These sub-k_BT spectral features, which reflect feedback-induced noise reduction, constitute a quantum thermodynamical signature of the non-thermal bath properties of the spinterface within the quantum engine’s spintronic operation. The 1.4meV energy window for deviations from linear behavior pegs a limit for the energy extent of all the spin states along the WS’s 3-spin long chain(s), with a spin splitting on each PM fluctuator of 0.7meV (c.f. theory).

**Figure 3:** Thermally activated, large output electrical power across the nanoharvester. (a) I(V) data from nanojunction B at H=0T within 40 < T(K) < 360. Top inset: zoom at low bias. Lower inset: P(V) data showing P_max = 450nW at 40K. The magenta crosspoint is the experimental error (see Methods). (b) ln vs 1/T plots of (top) R_s, (middle) V_off and (bottom) P_max. P_max decreases from 370nW at 40K to 24nW at 360K per two thermal activation regimes, with a 120K crossover temperature. The activation energy E_a is given for each regime.

Due to discrete states within the barrier, spintronic regimes involving multiple metallic and semiconducting nanochannels may coexist in a device. The slight decrease in junction conductance with increasing temperature (see Fig. 2d) confirms the metallic nature of nanojunction A. This, and several other metallic nanojunctions, exhibit 17 < P_max(nW) < 55 for 150 < R_s(Ω) < 800 at 40K (not shown). We now turn to semiconducting nanojunction B, for which we observe (see Fig. 3a) a mostly linear I(V) at 360K (R_s=1.05kΩ) that becomes increasingly non-linear as T is lowered to 40K (R_s=25.9kΩ). The apparent common crosspoint of I(V,T) data (inset of Fig. 3a) is orders of magnitude beyond measurement error (see magenta crosspoint) and can be explained as a bias-induced compensation point of transport across the spin potential landscape.

As seen in the ln vs. 1/T plots of Fig. 3(b), the R_s data follows R_s = R_0 e^{E_a/k_BT} with a single thermal activation energy E_a over 40 < T(K) < 360. In contrast, V_off and the maximum power P_max both exhibit two activation regimes with a crossover at 120K, i.e. at k_BT=J for CoPc (see Refs. 12 and Fig. 1b). This constitutes a quantum thermodynamical signature of the
WS’s phase transition as a QR. When the qubits are no longer entangled for T=120K, T more strongly impacts performance through an increase in $E_\text{s}$ that may reflect hopping transport\textsuperscript{31} between spins.

It is possible that our QR-enabled spintronic engine can address poorly explained prior experiments\textsuperscript{17,27,32}. Here, we observe $P_{\text{Max}} = 450$ nW at 40K (Fig. 3a inset) — a 450x increase over the previous record measured at 3K and $H=1$ T\textsuperscript{27}, and at 295K a 270x improvement\textsuperscript{17}. At 360K, $P_{\text{Max}} = 24$ nW, which is promising for applications. Our results thus not only dwarf those from possibly similar experiments\textsuperscript{17,27,32}, but also those from mesoscopic quantum heat engines\textsuperscript{15}.

![Image](image_url)

**Figure 4:** Spintronic features of the thermal energy harvester. (a) $I(H)$ data acquired on nanojunction C at 40K. (b) $I(V)$ data at $H=0$ T and $H=-0.5$ T, revealing a linear behavior with $R_s=63\Omega$ and $R_0=550\Omega$ around $V_{\text{Off}}=2.13$ mV and $V_{\text{Off}}=2.76$ mV, respectively. The blue crosses reflect the $I(H)$ data from panel a. (c) $R_S(H)$ calculated from two $I(H)$ datasets from panel (a). The blue crosses indicate $R_s$ inferred from panel b. The two spintronic $V_{\text{Off}}$ lead to extremal values of -$100 < MC (%) < \infty$ in $I(H)$ data, as do the two spintronic $R_s$ regarding -$100 < MR (%) < \infty$.

To confirm these devices’ spintronic underpinnings, the $I(H)$ data at 40K from nanojunction C in Fig. 4a reveal a strong in-plane $H$ dependence of junction current, which saturates for $|H|>0.5$ T (not shown) upon achieving the parallel orientation of FM electrode magnetization. The current can be suppressed at $H=0$ and $H=-0.5$ T for $V_{\text{Off}}=2.76$ mV and $V_{\text{Off}}=2.13$ mV, respectively. Thus, the junction’s two magnetic states promote differing $V_{\text{Off}}$. They also drive a sign change in current at $V=2.67$ mV, with $I=\pm 1\mu A$. These ($V,I$) pairs are confirmed by $I(V)$ data (Fig. 4b), and lie well beyond possible experimental offsets (see Methods). Using $I(H)$ data from panel a, we plot $R_S(H)$ in Fig. 4c. Consistency between these three data panels is visualized by blue crosses in panels b and c. The ‘optimistic’ $MR' = \frac{R_S(-0.5T)}{R_S(0T)} - 1=770\%$ implies\textsuperscript{7} an average transport spin polarization $P = 89.1\%$ of the two Fe/C\textsubscript{60} spintronic selectors\textsuperscript{14}. The magnetocurrent $MC = \frac{I(-0.5T)}{I(0T)} - 1=-100\%$ and 1470\% at each spintronic $V_{\text{Off}}$. This showcases this device class as a spintronically controlled switch of current direction/flow.

Our results introduce spintronics onto the quantum technologies roadmap as a very promising platform to implement autonomous quantum engines, and raise interesting questions for further research. A current can drive the electronic properties of paramagnetic centers: temperature, exchange coupling and even entropy production at a phase transition without a temperature gradient\textsuperscript{17,33,34}. Does this engine require electrical priming to operate, i.e. is it a quantum battery\textsuperscript{6,9}? Can precise thermometry confirm the entropy sinking\textsuperscript{21} by the FM state of the electrodes? Can certain aspects (e.g. TF stroke asymmetry\textsuperscript{10}, exchange coupling\textsuperscript{12}, number/parity of qubits) be optimized, e.g. using molecular engineering? Does zero-point energy play a role? Answering these questions, e.g. using in operando electron spin resonance, would shed insight into the autonomous engine’s multiple interlocking quantum resources, and help to determine its efficiency. Looking ahead, the most promising vector to industrialize this quantum technology is MgO.
spintronics, which so far has mostly targeted information storage/processing needs. To transform it into a dual-use information/energy technology workhorse will require mastering the insertion of paramagnetic centers, e.g. by controlling oxygen vacancies. Harvesting/storing this most basic form of energy --- ambient thermal energy, could help to alter our nomadic energy needs and accelerate the transition to clean energy.

Methods

Device preparation

Si/SiOx//Cr(5)/Fe(50)/CoOx(n ML)/CoPc(3ML)/C60/(5ML)/Fe(10)/Cr(50) heterostructure stacks were grown in-situ and at room temperature in an ultra-high vacuum multichamber cluster by dc sputtering (metals) and thermal evaporation (CoPc). All numbers are in nm; 1ML C60=0.9nm. 1ML CoPc=0.4nm. The SiOx substrate was annealed at 110°C and allowed to cool down prior to deposition. Metals were sputtered in a P=3.5E-3 mbar Ar atmosphere. Molecules were deposited at P=3E-9mbar. The C60 thickness n for nanojunctions A, B and C was 3ML, 1ML and 1ML, respectively. The Fe layers are in-plane magnetized. Nanojunctions were crafted using 300nm-diameter SiO2 nanobeads thanks to a recently developed resist- and solvent-free nanojunction process, and were wirebonded to a sample chip. The positive contact was connected to the junction’s top electrode. All data were acquired with the sample at a constant, nominally uniform temperature T. Since T≥40K in our experiment, the spin engine’s kBT=0.7meV condition is always satisfied.

Discussion of measurements artifacts

Out of 193 junctions processed, 10 were neither open-circuit or short-circuit, and 9 exhibited a combined current/voltage offset at least one order of magnitude larger than those observed on our measurement line using a calibrated resistance of similar amplitude. Control Fe/CoPc/Fe junctions did not exhibit such offsets. Measurements took place the dark using an electrically grounded cryostat and long metallic wires. An external microwave excitation is thus unrealistic. The change in sign of the current with H (see Fig. 4a) and the voltage amplitude (up to 74mV, see Fig. 3b) both point against a thermovoltage drop along the leads. The effect persists at T=40K when the sample heater is turned off, and decreases with increasing temperature. This casts aside a black-body radiation effect, as well as any stray T gradient between the cryostat’s cold finger and the sample. Reference nanojunctions containing only a CoPc spacer layer yielded current- and bias-offsets within the range found using calibrated resistances. This is also the case after permanent electrostatic degradation (see Junction B).

On junction B, current and bias offsets that are orders of magnitude beyond experimental error for those Rs are observed at all temperatures, and reach (84mV, -13.8μA) at 40K (see lower inset of Fig. 3a). Indeed, using calibrated resistances to determine experimental bias and current offsets, we obtain (1kΩ, 20μV, 30nA) and (10kΩ, 30μV, 0.8nA). These experiment offsets are dwarfed by the device offsets (compare magenta crosspoint in Fig. 3a and top inset with data). Since Rs decreases with increasing T, an experiment current offset would increase with increasing T, but loff follows the opposite trend (see Fig. 3a). Once permanent electrostatic degradation causes Rs to decrease from 25.9kΩ to 8.1kΩ at 40K, we recover offsets within these boundaries. The crossing of I(V,T) data within (-5.3<V(mV)<-2.9, -12.5<IL(μA)<-13.6) (see top inset of Fig. 3a), which also exceeds experimental error, suggests a bias-induced adjustment of the spin state potential landscape so as to compensate thermal broadening effects.

Ab-initio calculations

The structural and magnetic properties of the bcc Fe(110)/C60/CoPc system were calculated using density functional theory (DFT) by means of the Vienna ab-initio simulation package (VASP) and its built-in projector augmented wave (PAW) pseudopotentials. The exchange and correlation potentials are within the generalized gradient approximation (GGA) as

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parametrized by Perdew, Burke, and Ernzerhof. The van der Waals (vdW) weak interactions were computed within the GGA-D3 approach developed by Grimme and later implemented in the VASP package. A kinetic energy cutoff of 450 eV was used for the plane-wave basis set. To include the correlation effects of transition-metal 3d electrons, we adopted the DFT-GGA+U method, using a Hubbard U-term of 5 eV and an exchange parameter of 1 eV. See references in Ref. 13 for further details. The supercell geometry comprises the following three Bravais vectors: (22.96 Å, 0, 0), (14.35 Å, 20.29 Å, 0), and (0, 0, 37 Å), using 80 iron atoms per layer in the bcc structure along (110). The vacuum region separating the periodic images is 25 Å.

Atomic positions were relaxed by annulling the force on the atoms to within 10⁻⁴ meV/Å. We first relaxed the subsystem composed of 3 layers of Fe(110) and the C₆₀ to find the equilibrium distance, then optimized the CoPc-C₆₀ distance to find the ground state. We then used these configurations to study the magnetic state of the entire Fe(110)/C₆₀/CoPc system. To this end, we considered both ferromagnetic and antiferromagnetic couplings of the CoPc with Fe(110) substrate. After the atomic relaxations, we found that the antiferromagnetic configuration is more stable by about -0.7 meV. The C₆₀-Fe(110) distance is about 2.26 Å, and that between C₆₀ and CoPc is 2.6 Å. In both magnetic states, the magnetic moment of the iron atoms below the C₆₀ molecules is 2.29 μB, and 2.72 μB far from C₆₀. The magnetic moment of Co in CoPc is +1.34 μB (FM coupling) and -1.34 μB (AFM coupling). The magnetic moment on the CoPc ligands is -0.082 μB (FM coupling) and +0.089 μB (AFM coupling).

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Authors Contributions

M.B. conceived the experiment. B.C., C.K. and V.d.C. grew the samples, with help from G.V. and B.G.. L.K. made the nanojunctions, with help from D.M., A.B., R.B. and B.C. . B.C., M.B. and L.K. performed measurements with help from L. J., E.M., S.M., E.S, H. P.G., S.E., M.G., F.C. and P.O. . M.B., B.C. and L.K. analyzed the data, with input from M.H., F.M., B.V., D.L., W.W. and S.B. . F.N. performed the calculations, with input from M.A. and M.B. . M.B. wrote the manuscript, with help from M.L. and input from all authors.

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