Symmetry in Full Counting Statistics, Fluctuation Theorem, and Relations among Nonlinear Transport Coefficients in the Presence of a Magnetic Field

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(Dated: February 2, 2008)

We study full counting statistics of coherent electron transport through multi-terminal interacting quantum-dots under a finite magnetic field. Microscopic reversibility leads to the symmetry of the cumulant generating function, which generalizes the fluctuation theorem in the context of quantum transport. Using this symmetry, we derive the Onsager-Casimir relation in the linear transport regime and universal relations among nonlinear transport coefficients.

PACS numbers: 73.23.Hk, 72.70.+m

Full counting statistics (FCS) has become an active topic in the mesoscopic physics [1, 2, 3, 4, 5, 6, 7]. Recently, experiments have been conducted to measure third current cumulants [10] and the distributions [11, 12]. However, FCS has never been applied to exploration of general aspects, such as nonequilibrium thermodynamic structures in coherent electron transport. In this paper, we discuss these general aspects by studying symmetries in FCS, which is valid beyond a linear response regime.

Our argument is based on the microscopic reversibility, and is related to the steady state fluctuation theorem (FT) in nonequilibrium statistical mechanics [14, 15, 16]. FT is an important theory that holds even in the far-from-equilibrium regime. It is written as \(\lim_{\tau \to \infty} \ln[P(\Delta S)/P(-\Delta S)]/\tau = I_F\), where \(P(\Delta S)\) is the probability of entropy \(\Delta S\) = \(I_F\tau\), produced during time \(\tau\). This expression quantifies the probability of negative entropy, which can be finite for a short interval of time in small systems, as demonstrated in the context of the second law violation in colloidal particle experiments [14]. Remarkably, FT can reproduce Onsager’s reciprocal relations and the Kubo formula [12, 17] and predicts properties in the far-from-equilibrium regime [14].

Recently FT was studied with regard to classical mesoscopic electron transport, i.e., in the sequential tunneling regime in the quantum-dots, with Markovian approximations [17, 18]. These works highlighted the relation between FT and FCS. In this paper, we study the general relation between FT and FCS with respect to coherent electron transports in generic situations. We consider a multi-terminal interacting electron cavity under a finite magnetic field. The validity of FT has not yet been established in this regime. In electron transport, two thermodynamic forces—the thermal gradient and the bias voltage—produce entropy. Thus, we introduce a cumulant generating function to obtain heat and charge transfer, and exactly derive its new symmetry. The symmetry leads to a quantum version of FT and measurable universal relations among nonlinear transport coefficients. They are extensions of the Onsager-Casimir relation [19].

Model. We consider a mesoscopic cavity connected to \(m\) electron reservoirs. The total Hamiltonian consists of the reservoirs \(H_r\) (\(r = 1, \cdots, m\)), cavity \(H_d\), interaction \(H_{\text{int}}\), and tunneling \(H_T\)

\[
H = \sum_{r=1}^{m} H_r + H_d + H_{\text{int}} + H_T. \tag{1}
\]

The cavity is described as \(H_d = \sum_{ij\sigma} t_{ij} d_{i\sigma}^\dagger d_{j\sigma}\) where \(d_{i\sigma}\) annihilates an electron with spin \(\sigma\) at site \(i\). We assume the Coulomb interaction in the cavity \(H_{\text{int}} = \sum_{i\sigma \neq j\sigma} U_{i\sigma j\sigma} d_{i\sigma}^\dagger d_{j\sigma} d_{j\sigma}^\dagger d_{i\sigma}\)/2. The Hamiltonian for the reservoir \(r\) is \(H_r = \sum_{k\sigma} \varepsilon_{rk} a_{rk\sigma}^\dagger a_{rk\sigma}\), where \(a_{rk\sigma}\) annihilates an electron with spin \(\sigma\) and the wave vector \(k\).

The tunneling between the reservoirs and the cavity is described as \(H_T = \sum_{rki\sigma} t_{rki} a_{r\sigma}^\dagger a_{k\sigma}\). When a magnetic field \(B\) is applied, the elements of the hopping and tunneling matrix acquire the phases \(t_{ij} = |t_{ij}| \exp(i \phi_{ij})\) and \(t_{rki} = |t_{rki}| \exp(i \phi_{rki})\). The phases are odd functions of the magnetic field \(\phi(-B) = -\phi(B)\).

Our calculations follow the standard procedure with a perturbation series for \(H_{\text{int}}\). Throughout this paper, we use \(h = k_B = e = 1\). The density matrix at the initial time \(t = -\tau/2\) is assumed to be of the product form \(\rho_0 = \prod_s \rho_s\), (\(s = 1, \cdots, m, d\)), where \(\rho_s\) is the equilibrium distribution at temperature \(T_s = 1/\beta_s\) and chemical potential \(\mu_s\); \(\rho_s = \exp[-\beta_s(H_s - \mu_s N_s)]/\text{Tr} \exp[-\beta_s(H_s - \mu_s N_s)]\). The operator \(N_s\) is the number operator in reservoir \(N_s = \sum_{k\sigma} a_{r\sigma}^\dagger a_{r\sigma}\), or the cavity \(N_d = \sum_{i\sigma} d_{i\sigma}^\dagger d_{i\sigma}\). The charge and heat current operators are defined as \(I_{\text{cs}} = i [N_s, H_T]\) and \(I_{\text{hs}} = i [H_s, H_T]\). The expression for currents \(I_{\text{cr}}\) and \(I_{hr}\) are given by

\[
I_{\text{cr}} = -\sum_{i\sigma \neq j\sigma} i t_{rki} d_{i\sigma}^\dagger a_{k\sigma} + \text{H.c.}, \quad I_{hr} = -\sum_{i\sigma} i \varepsilon_{rk} t_{rki} d_{i\sigma}^\dagger a_{r\sigma} + \text{H.c.}.
\]
Cumulant generating function.— We introduce the characteristic function (CF) for the transmitted charge and heat during time $\tau$, $q_{a s} = \int_{-\tau/2}^{\tau/2} dt I_{a s}(t) \; (a = c, h)$:

$$Z\{\{\chi_{\alpha r}\}; B\} = \langle V^\dagger e^{iH \tau} V^2 e^{-iH \tau} V^\dagger \rangle, \quad V = \prod V_s,$$

where $V_s = \exp[-i(\chi_h H_s + \chi_c N_s)/2]$. This contains the counting fields for charge and heat current $\chi_{cs}$ and $\chi_{hs}$, respectively. The symbol $\langle \ldots \rangle$ denotes an average over the initial state. Equation (2) can be rewritten in the familiar form, where the counting fields play roles of fictitious gauge fields $[1, 2, 8]$, since $V_s^\dagger H_T V_s$ makes the gauge fields in the elements of the tunneling matrix. Note that Eq. (2) satisfies the normalization condition $Z\{\{0\}; B\} = 1$.

We use the CF and introduce the cumulant generating function (CGF) defined at the stationary state as

$$\mathcal{F}\{\{\chi_{\alpha r}\}; B\} = \lim_{\tau \to -\infty} \ln Z\{\{\chi_{\alpha r}\}; B\}/\tau. \quad (3)$$

The CGF generates cumulants from the derivatives with respect to the counting fields. The first and second derivatives generate the average current between the terminal and the cavity, and a symmetrized current correlation expressed as

$$\langle I_{\alpha 1} \rangle = \frac{\partial \mathcal{F}\{\{0\}; B\}}{\partial \chi_{\alpha 1}} \bigg|_{\tau = 0} = \lim_{\tau \to -\infty} \frac{\langle q_{11} \rangle}{\tau},$$

$$\langle I_{\alpha 1} I_{\alpha 2} \rangle = \frac{\partial^2 \mathcal{F}\{\{0\}; B\}}{\partial \chi_{\alpha 1} \partial \chi_{\alpha 2}} \bigg|_{\tau = 0} = \lim_{\tau \to -\infty} \frac{\langle q_{11} q_{22} \rangle - 2 \langle q_{12} \rangle \langle q_{22} \rangle}{2 \tau}.$$

Microscopic reversibility.— So far several symmetries in CGF are known [9]. For instance, CGF is a $2\pi$-periodic function of $\chi_{cs}$, which is a consequence of the discreteness of the charge. We take the time reversal symmetry in CGF into consideration. Let $\Theta$ be the time reversal operator, which evaluates the complex conjugate for complex numbers and reverses the spin operator [21]. This time reversal operator satisfies the equations: $\Theta i \Theta^{-1} = -i$ and $\langle n| \Theta O | n' \rangle = \langle n'| \Theta^\dagger O \Theta^{-1} | n \rangle$ [22]. Here $\Theta$ is an operator and $| n \rangle = \Theta | n \rangle$. Calculations with these equations and use of the definition of operator $V$ lead to the relation

$$Z\{\{\chi_{\alpha r}\}; B\} = Z\{-\chi_{\alpha r} + i A_{\alpha r}; -B\}, \quad (4)$$

where $A_{cs} = \beta_s \mu_s$ and $A_{hs} = -\beta_s$. This equality is critical in obtaining our central result [9].

When stationary, no extra charge and heat accumulate inside the cavity. This implies that CGF depends only on the differences between the reservoirs’ counting fields and is independent of the cavity’s counting field and initial state. Below, we present its proof using the Schwinger-Keldysh approach [1, 2, 21].

The entire CGF expression consists of a noninteracting part $\mathcal{F}_0$ and an interacting part $\mathcal{F}_{\text{int}}$. We first consider the noninteracting part. After a straightforward calculation, the analytical expression of $\mathcal{F}_0$ is obtained as

$$\mathcal{F}_0\{\{\chi_{\alpha s}\}; B\} = -\frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \; \ln \det \hat{g}(\omega) + \text{const.},$$

where $\hat{g}(\omega)$ is the Green function which has site, spin, and Keldysh indices. The normalization condition $\mathcal{F}_0\{\{0\}; 0\} = 0$ must be satisfied. The matrix elements at $(i\alpha, j\sigma')$ for the inverse of the Green function are given in the Keldysh space as

$$\hat{g}^{-1}(i\alpha, j\sigma') = \omega \delta_{ij} \hat{\tau}^3 - t_{ij} \hat{\tau}^3 - \sum_r \hat{\tau}^3 \hat{\Sigma}_{rij}(\omega) \hat{\tau}^3 - \hat{\eta}_d,$$

for $\sigma = \sigma'$ and 0 for other cases. The self-energy and the Pauli matrix $\hat{\tau}^3$ are

$$\hat{\Sigma}_{rij}(\omega) = \left( \begin{array}{cc} \Sigma^{++}_{rij}(\omega) & \Sigma^{+-}_{rij}(\omega) \\ \Sigma^{-+}_{rij}(\omega) & \Sigma^{--}_{rij}(\omega) \end{array} \right),$$

$$\hat{\tau}^3 = \left( \begin{array}{cc} 1 & 0 \\ 0 & -1 \end{array} \right).$$

In the wide-band limit $\Gamma_{rij} = 2\pi \sum_k t_{rki}^* t_{rki} \delta(\omega - \varepsilon_k)$ [22], the causal and anti-causal components are $\Sigma^{\pm\pm}_{rij}(\omega) = -i \Gamma_{rij} [1/2 - f_\pm(\omega)]$ with the Fermi/hole distribution function $f_\pm(\omega) = 1/(\exp[\pm \beta_\sigma (\omega - \mu_\sigma)] + 1)$. The lesser and greater components depend on reservoir’s counting fields as $\Sigma^{\pm\pm}_{rij}(\omega) = \pm i \Gamma_{rij} f_\pm(\omega) \exp[\pm (\chi_{s r} \omega + \chi_{c r})]$. The wide-band limit is not critical to our argument as long as the reservoirs form continuum energy spectra, which cover the entire energy range of the cavity. The term $\hat{\eta}_d$ has an infinitesimal contribution, depending on the counting fields and the initial state of the cavity. This term is crucial in the causality [21], but negligible compared to the self-energy terms of the reservoirs. This is plausible because the steady state should not depend on the initial state of the cavity. In addition, we consider the rotation of the Green function in the Keldysh space $R^\dagger g_{\text{int}}(\omega) R$, where the operator $R$ is defined as

$$R = \exp[i(\chi_{hs} \omega + \chi_{cs}) \hat{\tau}^3/2]. \quad (6)$$

This transformation does not change Eq. (4), instead it only replaces $\chi_{cs}$ by $\chi_{cs} - \chi_{hs}$ in the Green function. Therefore, we conclude that $\mathcal{F}_0$ depends only on the differences between the reservoirs’ counting fields. Thus, $\chi_{cs}$ by $\chi_{cs} - \chi_{hs}$ in the Green function.

We now consider the interaction part $\mathcal{F}_{\text{int}}$ based on the linked cluster expansions. It is formally written as

$$\mathcal{F}_{\text{int}} = \lim_{\tau \to -\infty} \ln \left[ e^{i S_{\text{int}}^+ - i S_{\text{int}}^-} e^{i S_J} | I_{\alpha s} = I^*_{\alpha s} = 0 \rangle / \tau \right] \quad (7)$$

Here the functions $S_J$ and $S_{\text{int}}^\pm$ are given as

$$S_J = - \sum_{i\alpha r} \int_{-\tau/2}^{\tau/2} dt dt' J_{i\alpha s}(t') \hat{\tau}^3 g_{i\alpha s}(t', t') \hat{\tau}^3 J_{i\alpha r}(t'),$$

$$S_{\text{int}}^\pm = - \int_{-\tau/2}^{\tau/2} dt H_{\text{int}} \left[ \frac{\delta}{\delta J_{\alpha s}^+(t)} \frac{\delta}{\delta J_{\alpha s}^-(t)} \right],$$

where the Grassmann source field in the Keldysh space $J_{i\alpha s} = \langle J_{i\alpha s}, J_{i\alpha s} \rangle$ is used. The function $H_{\text{int}}[\ldots]$ is obtained by substituting the derivatives of the Grassmann numbers for the fermion operators in the Hamiltonian $H_{\text{int}}$. We consider these functions in the Fourier space, and transform the Green function and Grassmann fields
using the rotation operator $\hat{R}$. Then we readily find that $\mathcal{F}_{\text{int}}$ is expressed by the simple replacement of the Green function in $S_{\text{f1}}$ by the inverse Fourier transform of $\hat{R}^\dagger \hat{y}_{\text{rf}r}\sigma^\dagger(\omega)\hat{R}$. In this mathematical structure, the charge and energy conservation in the interaction process is crucial. To check it, let us consider the calculation for one diagram that contributes to the second order in $\mathcal{F}_{\text{int}}$

$$\frac{1}{4} \sum_{s\sigma} \frac{\hat{s}^3}{s^3} \frac{\hat{s}^3}{s^3} U_{\sigma,s,j\sigma} U_{\sigma,j',\sigma'} \frac{d\nu}{2\pi} \frac{d\nu'}{2\pi} g_{s\sigma}^{\dagger}(\nu) g_{s\sigma}^{\dagger}(\nu') g_{s\sigma}^{\dagger}(\nu') g_{s\sigma}^{\dagger}(\nu'), \quad (8)$$

where the summations are performed over the site, spin, and Keldysh indices $s, s' = \pm$. Eq. (8) does not change on the rotation $\hat{R}^\dagger \hat{y}_{\text{rf}r}\sigma^\dagger(\omega)\hat{R}$, since diagrammatically the energy and charge are conserved. Therefore we find that $\mathcal{F}_{\text{int}}$ depends only on the difference of the reservoirs' counting fields. From Eq. (9), we obtain the symmetry in the interacting electron transport as

$$\mathcal{F}(\{\chi_{\text{r}}\}; B) = \mathcal{F}(\{-\chi_{\text{r}} + i\mathcal{A}_{\text{r}}\}; -B), \quad (9)$$

where $\chi_{\text{r}} = \chi_{\text{r}} - \chi_{\text{m}}$ and $\mathcal{A}_{\text{r}}$ represents the affinity $\mathcal{A}_{\text{r}} = -\chi_{\text{r}} - \mathcal{A}_{\text{m}}$. We can check that CGFs in interacting quantum-dots in Refs. [2, 3, 4] satisfy Eq. (9).

**Fluctuation theorem.**— Eq. (9) can be regarded as a quantum fluctuation theorem (FT). Let us consider a two-terminal setup and define the entropy produced as

$$\Delta S = \mathcal{A}_{\text{r}} q_{\text{r}} + \mathcal{A}_{\text{h}} q_{\text{h}}. \quad (10)$$

We fix the counting fields as $\chi_{\text{c1}} A_{\text{c1}}^{-1} = \chi_{\text{h1}} A_{\text{h1}}^{-1} = \chi$. The entropy production is obtained from the derivative of CGF with respect to $\chi$. Asymptotic form of the probability distribution can be obtained from the inverse Fourier transform of the CF with the variable $\chi$. Saddle-point analysis with the symmetry (9) yields the relation

$$\lim_{\tau \to \infty} \ln \frac{P(\Delta S; B)}{P(-\Delta S; -B)} / \tau = I_E. \quad (11)$$

This formula generalizes FT in the quantum regime under a finite magnetic field. At a uniform temperature, the entropy production is proportional to the charge current. In this case, Eq. (11) quantifies the probability of back flow of charge currents.

**General relations in nonlinear transport regime.**— We show that the symmetry (9) predicts general relations among nonlinear transport coefficients. We consider the situation that only the chemical potentials $\mu_1$ and $\mu_2$ are varied at a uniform temperature $\beta^{-1}$, and the charge currents are measured at terminals 1 and 2. We then compute cumulants of the currents, $\langle\{I_{\text{c1}}, I_{\text{c2}}\}\rangle = \partial^{k_1-k_2} \mathcal{F}(\{1\}; B) / \partial \{\chi_{\text{c1}}\}^{k_1} \partial \{\chi_{\text{c2}}\}^{k_2}$. The nonlinear transport coefficient is defined in the expansion of the cumulants with respect to the affinities as

$$L_{\ell_1\ell_2}^{k_1k_2}(B) = \left. \frac{\partial^{\ell_1+k_2} \langle I_{\text{c1}}, I_{\text{c2}}\rangle}{\partial A_{\text{c1}}^{\ell_1} \partial A_{\text{c2}}^{k_2}} \right|_{A_{\text{r}} = A_{\text{m}} = 0}. \quad (12)$$

The affinities $\mathcal{A}_{\text{c}j} (j = 1, 2)$ are written as $\mathcal{A}_{\text{c}j} = \beta (\mu_j - \mu)$ with a definite chemical potential $\mu$ for terminals 3 to $m$. We symmetrize the coefficients and CGF as

$$L_{\ell_1\ell_2}^{k_1k_2} = L_{\ell_2\ell_1}^{k_2k_1} (B) \pm L_{\ell_1\ell_2}^{k_1k_2} (-B),$$

$$\mathcal{F}^{\pm}(\{\chi_{\text{c}}\}) = \mathcal{F}(\{\chi_{\text{r}}\}; B) \pm \mathcal{F}(\{\chi_{\text{r}}\}; -B).$$

From Eq. (9), we obtain the equality $\mathcal{F}^{\pm}(\{\chi_{\text{c}}\}) = \pm \mathcal{F}^{\pm}(\{-\chi_{\text{c}} + i\mathcal{A}_{\text{c}}\})$. Note that CGF is a function of $\mathcal{A}_{\text{c}}$ as well as $\chi_{\text{c}}$ [17]. By taking the derivatives with respect to the affinities and counting fields for both sides of the equality of $\mathcal{F}^{\pm}$, we obtain the general relations among the coefficients as

$$L_{\ell_1\ell_2}^{k_1k_2} = \pm \sum_{n_1=0}^{\ell_1} \sum_{n_2=0}^{\ell_2} {\ell_1 \choose n_1} {\ell_2 \choose n_2} (-1)^n L_{\ell_1-n_1, \ell_2-n_2}^{k_1+n_1, k_2+n_2}(B), \quad (13)$$

where $n = n_1 + n_2 + k_1 + k_2$. With the equalities $L_{01}^{00} = 0$, the relations (13) can be further simplified. We consider equations (13) which satisfy $N = k_1 + k_2 + \ell_1 + \ell_2$. Equations for $N = 2$ reproduce the linear response results such as Kubo formula and Onsager-Casimir relations [23]

$$L_{01}^{00}(B) = L_{10}^{01}(B), \quad L_{10}^{01}(B) = L_{10}^{00}(B), \quad (14)$$

and $L_{01}^{11}(B) = L_{10}^{10}(B)$. Relations beyond linear response regime can be obtained for $N \geq 3$. We list some of the relations for $N = 3$ as

$$L_{20}^{10} = L_{10}^{20}, \quad (15)$$

$$L_{11}^{01} = L_{02}^{10} = 2 L_{11}^{01} - L_{01}^{22}, \quad (16)$$

$$L_{20}^{01} = L_{20}^{11} = 2 L_{10}^{10}, \quad (17)$$

$$L_{20}^{00} = L_{02}^{10} - 3 L_{00}^{30} / 6, \quad L_{00}^{30} = 0. \quad (18)$$

In mesoscopic experiments, large bias voltages can easily produce finite higher order coefficients, and the Onsager-Casimir relations can be violated [23, 26, 27]. Eqs. (15)-(18) demonstrate that beyond the Onsager relation, universal relations exist in the nonlinear transport regime.
These nontrivial relations rely solely on the microscopic reversibility and thus are insensitive to the setup details.

Three-terminal Aharonov-Bohm interferometer.— The simplest setup to demonstrate these relations would be a three-terminal Aharonov-Bohm (AB) ring with a threefold symmetry. We consider a ring consisting of three noninteracting quantum-dots, each of which connects to a reservoir, as shown in the inset in Fig. 1. The Hamiltonian of the ring is given as \( H_d = \sum_{\sigma} \sum_{i=1}^{3} \varepsilon d_{i\sigma}^\dagger d_{i\sigma} - t e^{i\phi/3} d_{1+}^\dagger d_{2+} + H.c. \) (\( d_{1\sigma} = d_{1\sigma}^\dagger \)). The tunnel coupling is \( \Gamma_{rij} = \Gamma \delta_{ij} \delta_{ri} \). The explicit form of Eq. (13) is given by

\[
\mathcal{F}_0 = \frac{1}{\pi} \int_{-\infty}^{\infty} d\omega \ln \left\{ 1 + \sum_{j,k=1}^{3} f_j^+ (\omega) f_k^- (\omega) (\epsilon (\chi_j - \chi_k) - 1) \right\}
\]

where \( \epsilon_{jkl} \) is the totally antisymmetric tensor. The transmission probability \( T_{even/odd} \) are written as

\[
T_{even} = \frac{1}{z^2 (1/4 + t^2 + z^2 - 2 t z \cos \phi/\Delta)} \quad \text{and} \quad T_{odd} = \frac{t^2 \sin \phi/\Delta}{z^2 (3/8 - 6z^2) + t^2 z (4z^2 - 3) - 12 t^2 \cos \phi + 9 t^4 (2z^2 + 1/4) + 4 \bar{T}^4 \cos^2 \phi}
\]

Figure (1) shows the linear conductance multiplied by \( 6/\pi \), as well as the nonlinear coefficients in Eqs. (15) and (17). It is clear that the relations (15) and (17) are satisfied. The overall structures depend on the temperature regions.

Two-terminal case.— Two-terminal geometry is a common setup used in experiments. Eqs. (15)–(18) are still satisfied, but the double script notations for the transport coefficients are replaced by single script: \( F_{k_1 k_2} \rightarrow (-1)^{(k_1 + k_2)}/F_{k_1 + k_2} \). In general, all cumulants of charge currents are symmetric in the magnetic field in noninteracting systems. Hence, Eqs. (17)–(18) cannot be measured. However, in noncentrosymmetric interacting systems, the coefficient \( L_{2,-} \) is generally finite. In this case, Eq. (18) predicts a finite third current cumulant even in the equilibrium. We confirmed a finite value of the coefficient \( L_{2,-} \) and \( L_{0,-} \) which is asymmetric in the magnetic field, in the model of a double dot AB interferometer with noncentrosymmetric geometry where one of the dots is capacitively coupled to an additional gate electrode. We observe that the relation (18) can be observed in the setup of the experiments.

Summary.— We derived the new symmetry in full counting statistics from the microscopic reversibility. This symmetry can be regarded as the quantum version of the fluctuation theorem under a magnetic filed. When there is no magnetic field, this reproduces the usual fluctuation theorem without quantum corrections. The symmetry leads to relations among nonlinear transport coefficients. Eqs. (15)–(18) would be measurable in present-day experiments. We hope this work will motivate further experimental and theoretical work on universal relations among nonlinear transport coefficients.

KS was supported by MEXT (No. 19740232). YU was supported by Special Postdoctoral Researchers Program of RIKEN.

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