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Thermoelectric Effects in Spin Valves Based on Layered Magnetic Structures

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General aspects of thermoelectric effects in spin valves consisting of two magnetic layers separated by a non-magnetic spacing layer are considered, with the main focus on the spin Seebeck effects. The Seebeck and spin Seebeck effects are considered in both current-in-plane and current-perpendicular-to-plane geometries. The corresponding thermopower and spin thermopower in the macroscopic limit of electronic transport are also considered. Physical origin of the spin effects is discussed in detail.

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1. Introduction

One can observe recently an increasing interest in thermoelectric properties of nanomaterials, which appears due to the following two reasons. The first one is of applied character and concerns expected applications of thermoelectric phenomena for converting thermal energy to electric energy at nanoscale [1, 2]. The second reason is of both fundamental and applied characters and concerns various spin related effects in thermoelectricity, which were discovered in recent years. These effects can be considered as spin counterparts of already well known thermoelectric phenomena related to electron charge [3–6].

Conversion of thermal to electric energy is an essence of the conventional Seebeck effect, where an electrical voltage ΔV is crated by a temperature difference ΔT between the two ends of a system. This effect is described quantitatively by the thermopower (or the Seebeck coefficient) S, defined as $S = -\Delta V/\Delta T$ in the absence of charge current flowing in the system, I=0 (e.g. in an open circuit system) [7]. Advantage of nanoscale systems over bulk materials follows from the fact that thermoelectric efficiency in nanoscale systems can be enhanced owing to violation of the Wiedemann–Franz law due to energy quantization and Coulomb correlations [8–13].

Magnetic materials with relatively long spin relaxation time, when transport in the two spin channels can be considered as independent, can exhibit additionally spin thermoelectric properties [3–5, 14–16]. Similarly to the conventional Seebeck effect, one can observe in these materials the so-called spin Seebeck effect, which is described by the corresponding spin thermopower or spin Seebeck coefficient $S_{\rm s}$. This effect consists in generation of a spin voltage $\Delta V_{\rm s}$ by a temperature gradient,

i.e. $S_{\rm s} = -\Delta V_{\rm s}/\Delta T$ [5, 15]. The spin Seebeck effect results from interplay of charge and spin transport, and can be important in various spintronic devices [4]. When the spin relaxation processes are absent, a spin accumulation builds up at the ends of a system with open boundary conditions, which suppresses the spin current, similarly as charge accumulation suppresses charge current. As a result, there is an electrical voltage as well as spin voltage between the two ends of a system when these ends have different temperatures. However, when spin relaxation is fast, there is no spin accumulation, even if spin current is induced by a temperature gradient, and only an electric voltage appears between the two ends of the system. Interestingly, the electrical voltage in these two limiting situations, i.e. with and without spin accumulation (spin voltage), may be remarkably different due to the interplay of the charge and spin transport and charge and spin accumulations.

Thermoelectric phenomena are of practical use when the thermoelectric efficiency described by the figure of merit Z obeys the inequality ZT>1, where T denotes temperature. In the case of conventional thermoelectricity, the thermoelectric efficiency is given by the relation $ZT=S^2GT/\kappa$ [17], where G and κ stand for the electrical and thermal conductance, respectively. Additionally, to describe spin thermoelectricity in magnetic materials one can introduce the spin figure of merit described by $Z_sT=S_s^2|G_s|T/\kappa$, where G_s is the spin conductance defined as $G_s=G_{\uparrow}-G_{\downarrow}$, with G_{σ} ($\sigma=\uparrow,\downarrow$) denoting electrical conductance in the well-defined spin- σ channel [5].

In this paper we consider thermoelectric phenomena in typical current-in-plane (CIP) and current-perpendicular-to-plane (CPP) spin valves consisting of two magnetic layers separated by a nonmagnetic spacing layer. Such devices are known to exhibit giant magnetoresistance (GMR) effect due to rotation of the films' magnetic moments from parallel to antiparallel alignment, which occurs as a result of spin-dependent transport. Our main interest, however, is in spin thermoelectric effects which may occur in these devices.

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2. General aspects of thermoelectricity in spin valves

Let us consider a typical spin valve consisting of two magnetic layers separated by a thin nonmagnetic layer, and assume that the magnetic moments of the layers are oriented in their planes. Resistance of the system in the CIP or CPP geometry depends then on the angle ϕ between magnetic moments of the two magnetic films [18–23] and can be written approximately as

$$R(\phi) = R_P + \Delta R \sin^2(\phi/2),\tag{1}$$

where R_P is the resistance in the parallel magnetic configuration ($\phi = 0$), while ΔR is the difference in resistances of antiparallel, R_{AP} , and parallel configurations, $\Delta R = R_{AP} - R_P$. Alternatively, one may write the corresponding conductance $G(\phi)$ as

$$G(\phi) = G_P - \Delta G \sin^2(\phi/2), \tag{2}$$

where G_P is the conductance in the parallel configuration and $\Delta G = G_P - G_{AP}$. Usually, $\Delta R > 0$, but in some systems $\Delta R < 0$ may also occur.

Let us assume the lateral size (total thickness) of the spin valve in the CIP (CPP) geometry is longer than the spin diffusion length, so the spin effects in thermoelectricity are negligible. In metallic systems the Seebeck coefficient S obeys the Mott formula [24, 25], i.e. S is proportional to the temperature T and to the derivative of the conductivity with respect to the chemical potential μ . In our case we write this relation as

$$S = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln G \Big|_{\epsilon = E_{\rm F}}, \tag{3}$$

where k_B is the Boltzmann constant, e is the electron charge (e < 0), while the derivative of the corresponding (CIP or CPP) conductance with respect to energy e is taken at the Fermi level e = E_F . Note that the thermopower e vanishes when the conductance is constant near the Fermi level.

Taking into account the above formula and Eq. (2), one may write the thermopower S for arbitrary magnetic configuration in the form

$$S(\phi) = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln G(\phi) \Big|_{\epsilon = E_F}$$

$$= -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln (G_P - \Delta G \sin^2(\phi/2)) \Big|_{\epsilon = E_F}. \tag{4}$$

In the collinear (i.e. parallel and antiparallel) configurations, the above formula gives

$$S^{P(AP)} = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln G^{P(AP)} \Big|_{\epsilon = E_F}.$$
 (5)

Thus, to find the thermopower S one needs to know how the conductance depends on energy around the Fermi level. In the case of spin valves, the conductance depends on the materials used to fabricate the spin valve structure and in general is a rather complex function of the conductances of individual layers.

What follows from the above is that the thermopower in spin valves depends on magnetic configuration, provided the GMR is nonzero. Thus, one may define the

magnetothermopower (MTP) as a difference in the thermopowers in parallel and antiparallel configurations [15], MTP = $S(\phi = 0) - S(\phi = \pi)$, that is

$$MTP = \frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln \left(\frac{G_{AP}}{G_P} \right) \Big|_{\epsilon = E_F}.$$
 (6)

Note that generally the Fermi energy $E_{\rm F}$ may depend on magnetic configuration due to size effects, which for notation simplicity is not indicated explicitly in the above formula.

Consider now thermoelectric effects in the opposite limit, i.e. when the corresponding length of the system is smaller than the spin diffusion length, so transport in one spin channel is independent of transport in the second spin channel. For simplicity, we restrict further considerations in this section to collinear, i.e. parallel and antiparallel alignments of the magnetic moments of the two ferromagnetic films. Total electrical conductance in both configurations can be presented as a sum of the individual conductances of the spin-↑ and spin-↓ channels

$$G^P = G^P_{\uparrow} + G^P_{\downarrow},\tag{7}$$

$$G^{AP} = G_{\uparrow}^{AP} + G_{\downarrow}^{AP}. \tag{8}$$

In turn, the corresponding spin conductances are

$$G_s^P = G_{\uparrow}^P - G_{\downarrow}^P, \tag{9}$$

$$G_s^{AP} = G_{\uparrow}^{AP} - G_{\downarrow}^{AP}. \tag{10}$$

For each spin channel one can define the corresponding thermopower:

$$S_{\sigma} = -\frac{\delta V_{\sigma}}{\Delta T} = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln G_{\sigma} \Big|_{\epsilon = E_F}$$
 (11)

for $\sigma = \uparrow, \downarrow$. Now, the voltages generated in the two spin channels can be different. Thus, one can write this voltage as

$$V_{\sigma} = V + \bar{\sigma}V_{s},\tag{12}$$

where $\bar{\sigma} = +(-)$ for $\sigma = \uparrow (\downarrow)$. Following this, one may introduce the conventional electrical thermopower as

$$S = S_{\uparrow} + S_{\downarrow},\tag{13}$$

and spin thermopower as

$$S_s = S_{\uparrow} - S_{\downarrow}. \tag{14}$$

Accordingly, the electrical thermopower may be written in the form

$$S = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln(G_{\uparrow} G_{\downarrow}) \Big|_{\epsilon = E_F}, \tag{15}$$

while the spin thermopower as

$$S_s = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln \frac{G_{\uparrow}}{G_{\downarrow}} \Big|_{\epsilon = E_F}.$$
 (16)

The above formulae are general, i.e. valid for arbitrary spin valve and for both collinear configurations. First, one can easily conclude that the electric thermopower S given by Eq. (15) is different from that given by Eq. (5), and this takes place in both parallel and antiparallel magnetic configurations. As mentioned in Introduction, this difference appears as a result of spin accumulation at the ends of the system. Second, assuming a symmetrical spin valve, when both magnetic layers are equivalent, one has

 $G_{\uparrow} \neq G_{\downarrow}$ in the parallel configuration, and consequently also $S_{\uparrow}^{P} \neq S_{\downarrow}^{P}$. As a result, the corresponding spin thermopower is generally nonzero, $S_{s}^{P} \neq 0$, i.e. thermal gradient may cause not only an electric voltage V but also a spin voltage V_{s} . In the antiparallel configuration, in turn, $G_{\uparrow} = G_{\downarrow}$ for a symmetrical spin valve, and thus also $S_{\uparrow}^{AP} = S_{\downarrow}^{AP}$. The electrical thermopower S_{s}^{AP} is then nonzero, but the spin thermopower S_{s}^{AP} vanishes, $S_{s}^{AP} = 0$. Note that when the spin valve is asymmetrical, the spin thermopower may occur also in the antiparallel configuration.

Similarly as in the absence of spin thermoelectricity, one can define magnetothermopower:

$$\text{MTP} = \frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln \left(\frac{G_{\uparrow}^{AP} G_{\downarrow}^{AP}}{G_{\uparrow}^{P} G_{\downarrow}^{P}} \right) \Big|_{\epsilon = E_F}. \tag{17}$$

and spin magnetothermopower, MTP_s , defined as the difference in spin thermopowers in the parallel and antiparallel configurations,

$$\mathrm{MTP}_{s} = \frac{\pi^{2}k_{B}^{2}T}{3|e|} \frac{\partial}{\partial \epsilon} \ln \left(\frac{G_{\uparrow}^{AP}}{G_{\downarrow}^{AP}} \frac{G_{\downarrow}^{P}}{G_{\uparrow}^{P}} \right) \Big|_{\epsilon = E_{\mathrm{F}}}. \tag{18}$$

When the spin valve is symmetrical, one should put $G_{\uparrow}^{AP} = G_{\downarrow}^{AP}$ in the above formulae, so spin magnetother-mopower is then determined only by the second term of the logarithm argument.

3. CIP configuration: macroscopic limit

Consider now a spin valve in the CIP geometry and assume the macroscopic limit. It is known that GMR effect disappears in this limit, which corresponds to the situation when thickness of the spacing layer is larger than the corresponding electron mean free path. We will consider in this section only collinear alignment of the magnetic moments of both magnetic layers. Assume first the case when spin effects and spin accumulation in the magnetic layers are absent due to spin-flip scattering. The in-plane conductance can be then written as

$$G \cong G^1 + G^2 + G^0, \tag{19}$$

where G^1 , G^2 and G^0 denote the conductances of the two magnetic layers and of the nonmagnetic layer, respectively. The corresponding thermopower is then

$$S = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln \left(G^1 + G^2 + G^0 \right) \Big|_{\epsilon = E_F}. \tag{20}$$

Note that the thermopower in the antiparallel configuration is then the same as in the parallel one, so the magnetothermopower is equal to zero (similarly as GMR).

Assume now the spin accumulation and spin effects play a role. The in-plane conductance G^P_{σ} in the spin channel σ can be then written as

$$G_{\sigma} \cong G_{\sigma}^1 + G_{\sigma}^2 + G^0/2, \tag{21}$$

for $\sigma = \uparrow$ and $\sigma = \downarrow$. Here, G_{σ}^{1} and G_{σ}^{2} denote the spin-dependent conductances of the two magnetic layers in the spin valve. Assuming the two spin channels as independent, one can define the spin-dependent thermopower:

$$S_{\sigma} = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln \left(G_{\sigma}^1 + G_{\sigma}^2 + G^0/2 \right) \Big|_{\epsilon = E_F}. \tag{22}$$

As above, we can define the conventional electrical thermopower

$$S = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln\left(\left(G_{\uparrow}^1 + G_{\uparrow}^2 + G^0/2\right)\right) \times \left(G_{\downarrow}^1 + G_{\downarrow}^2 + G^0/2\right)\Big|_{\epsilon = E_F}, \tag{23}$$

and spin thermopower as

$$S_s = -\frac{\pi^2 k_B^2 T}{3|e|} \frac{\partial}{\partial \epsilon} \ln \left(\frac{G_{\uparrow}^1 + G_{\uparrow}^2 + G^0/2}{G_{\downarrow}^1 + G_{\downarrow}^2 + G^0/2} \right) \Big|_{\epsilon = E_F}.$$
 (24)

When magnetic configuration is parallel, then in both magnetic layers spin- \uparrow corresponds to majority (M) electrons, while spin- \downarrow to spin minority (m) ones. The conventional thermopower S can be then written as

$$S^{P} = -\frac{\pi^{2}k_{B}^{2}T}{3|e|} \frac{\partial}{\partial \epsilon} \ln\left(\left(G_{M}^{1} + G_{M}^{2} + G^{0}/2\right)\right) \times \left(G_{m}^{1} + G_{m}^{2} + G^{0}/2\right)\Big|_{\epsilon = E_{F}},$$
(25)

while the spin thermopower S_s^P takes the form

$$S_{s}^{P} = -\frac{\pi^{2}k_{B}^{2}T}{3|e|}$$

$$\times \frac{\partial}{\partial \epsilon} \ln \left(\frac{G_{M}^{1} + G_{M}^{2} + G^{0}/2}{G_{m}^{1} + G_{m}^{2} + G^{0}/2} \right) \Big|_{\epsilon = E_{F}}.$$
(26)

In the antiparallel configuration, in turn, \uparrow and \downarrow correspond to M and m in the left (1) layer, and to m and M in the right (2) magnetic layer. Thus, the conventional thermopower takes the form

$$S^{AP} = -\frac{\pi^2 k_B^2 T}{3|e|}$$

$$\times \frac{\partial}{\partial \epsilon} \ln \left(\left(G_M^1 + G_m^2 + G^0/2 \right) \times \left(G_m^1 + G_M^2 + G^0/2 \right) \right) \Big|_{\epsilon = E_F},$$

$$(27)$$

while the spin thermopower is given by the formula

$$S_s^{AP} = -\frac{\pi^2 k_B^2 T}{3|e|}$$

$$\times \frac{\partial}{\partial \epsilon} \ln \left(\frac{G_M^1 + G_m^2 + G^0/2}{G_m^1 + G_M^2 + G^0/2} \right) \Big|_{\epsilon = E_F}.$$
(28)

When the spin valve is symmetrical, $G_M^1=G_M^2$ and $G_m^1=G_m^2$, the spin thermopower in the antiparallel configuration vanishes, $S_s^{AP}=0$.

One point requires some comments. In the macroscopic limit the electrical conductance is independent of the magnetic configuration, i.e. the resistance in both parallel and antiparallel configurations is the same, so the GMR vanishes. Despite of this, when spin relaxation in the magnetic layers is slow, one can observe spin thermopower, which may be different in the two magnetic configurations. Moreover, the conventional thermopowers in both configurations are also different, and also differ from the thermopower in the absence of spin effects

(see Eq. (18)), which follows from the spin accumulation as already discussed in the previous section. To understand the difference in behavior of the resistance and spin thermopower, one should mention that spin thermopower also exists in a single magnetic slab when its length is smaller than the spin diffusion length [4]. However, the contributions from both magnetic layers in spin valve cancel each other in the antiparallel configuration. This means that local spin currents flow between the two magnetic layers, though the total spin current is equal to zero.

Following the preceding section, one can introduce the magnetothermopower MTP and spin magnetothermopower MTP_s . The corresponding formulae can be easily obtained from the formulae (25) to (28) and the appropriate definitions, so we will not present them explicitly. Note, the magnetothermopower does not vanish.

4. CPP configuration: macroscopic limit

Consider now a spin valve in the CPP configuration, and again assume the macroscopic limit. There is some difference between the CPP and CIP configurations as GMR in the macroscopic limit disappears in the CIP configuration, while it still remains quite substantial in the CPP geometry. This difference follows from the role of electrons which assure spin contact between both magnetic layers in the spin valve structure. In the case of CIP geometry, such electrons travel roughly perpendicular to the electric field, and thus do not contribute to current. In the CPP case, in turn, such electrons propagate parallel to electric field, and therefore contribute to current. This is the reason why the GMR disappears with the spacer thickness faster in the CIP geometry than in the CPP one, and in the latter case remains finite in the macroscopic limit.

In the absence of spin effects, the effective conductance of the spin valve in the CPP geometry may be written in the macroscopic limit as,

$$G\cong \frac{(G^1+G^2)G^0+G^1G^2}{G^1G^2G^0}. \tag{29}$$
 Thus, the corresponding thermopower takes the form

$$S = -\frac{\pi^2 k_B^2 T}{3|e|}$$

$$\times \frac{\partial}{\partial \epsilon} \ln \left(\frac{(G^1 + G^2)G^0 + G^1 G^2}{G^1 G^2 G^0} \right) \Big|_{\epsilon = E_F}.$$
(30)

When the spin diffusion length is long, the spin accumulation and spin effects may occur in the thermoelectricity. In this limit, conductance in the spin- σ channel

$$G_{\sigma} \cong \frac{(G_{\sigma}^{1} + G_{\sigma}^{2})G^{0} + 2G_{\sigma}^{1}G_{\sigma}^{2}}{G_{\sigma}^{1}G_{\sigma}^{2}G^{0}}.$$
 (31)

Accordingly, the corresponding thermopower S_{σ} takes the form

$$S_{\sigma} = -\frac{\pi^2 k_B^2 T}{3|e|}$$

$$\times \frac{\partial}{\partial \epsilon} \ln \left(\frac{(G_{\sigma}^1 + G_{\sigma}^2) G^0 + 2G_{\sigma}^1 G_{\sigma}^2}{G_{\sigma}^1 G_{\sigma}^2 G^0} \right) \Big|_{\epsilon = E_F}.$$
(32)

Thus, in the parallel magnetic configuration one finds

$$S^{P} = -\frac{\pi^{2}k_{B}^{2}T}{3|e|}$$

$$\times \frac{\partial}{\partial \epsilon} \ln \left(\frac{(G_{M}^{1} + G_{M}^{2})G^{0} + 2G_{M}^{1}G_{M}^{2}}{G_{M}^{1}G_{M}^{2}G^{0}} \right.$$

$$\times \frac{(G_{m}^{1} + G_{m}^{2})G^{0} + 2G_{m}^{1}G_{m}^{2}}{G_{m}^{1}G_{m}^{2}G^{0}} \Big|_{\epsilon = E_{F}}$$
(33)

for the conventional thermopower, and

$$\begin{split} S_{s}^{P} &= -\frac{\pi^{2}k_{B}^{2}T}{3|e|} \\ &\times \frac{\partial}{\partial \epsilon} \ln \left(\frac{(G_{M}^{1} + G_{M}^{2})G^{0} + 2G_{M}^{1}G_{M}^{2}}{G_{M}^{1}G_{M}^{2}G^{0}} \right. \\ &\left. \times \frac{G_{m}^{1}G_{m}^{2}G^{0}}{(G_{m}^{1} + G_{m}^{2})G^{0} + 2G_{m}^{1}G_{m}^{2}} \right) \Big|_{\epsilon = E_{F}} \end{split}$$
(34)

for the spin thermopower.

In turn, in the antiparallel magnetic configuration we

$$S^{AP} = -\frac{\pi^2 k_B^2 T}{3|e|}$$

$$\times \frac{\partial}{\partial \epsilon} \ln \left(\frac{(G_M^1 + G_m^2) G^0 + 2G_M^1 G_m^2}{G_M^1 G_m^2 G^0} \right)$$

$$\times \frac{(G_m^1 + G_M^2) G^0 + 2G_m^1 G_M^2}{G_m^1 G_M^2 G^0} \Big|_{\epsilon = E_F}$$
(35)

for the conventional thermopower, and

$$S_{s}^{AP} = -\frac{\pi^{2}k_{B}^{2}T}{3|e|}$$

$$\times \frac{\partial}{\partial \epsilon} \ln \left(\frac{(G_{M}^{1} + G_{m}^{2})G^{0} + 2G_{M}^{1}G_{m}^{2}}{G_{M}^{1}G_{m}^{2}G^{0}} \right)$$

$$\times \frac{G_{m}^{1}G_{M}^{2}G^{0}}{(G_{m}^{1} + G_{M}^{2})G^{0} + 2G_{m}^{1}G_{M}^{2}} \Big|_{\epsilon = E_{F}}$$
(36)

for the spin thermopower. As in the CIP case, when spin relaxation is slow, one can observe spin thermopower, which may be different in the two magnetic configurations. Moreover, the conventional thermopowers in both configurations are also different, and also differ from the thermopower in the absence of spin effects (see Eq. (28)). Again, when the spin valve is symmetrical, $G_M^1 = G_M^2$ and $G_m^1 = G_m^2$, the spin thermopower in the antiparallel configuration vanishes, $S_s^{AP} = 0$. Similarly as in the preceding section, one can introduce the magnetothermopower MTP and spin magnetothermopower MTP_s, and the corresponding formulae can be easily obtained from the formulae (33) to (36) and the appropriate definitions, so we will not present them here explicitly.

5. Conclusions

We have described general aspects of the conventional and spin thermoelectric effects in metallic spin valves consisting of two magnetic layers separated by a nonmagnetic layer. Thermoelectric properties in the macroscopic limit of electronic transport are also considered. We have distinguished between situations of long and short spin diffusion length. In the latter case the spin thermoelectric effect vanishes, so one may observe only the conventional Seebeck effect. In the former case, in turn, one can observe both conventional and spin Seebeck effects.

Interestingly, the conventional thermopower (or equivalently the Seebeck coefficient) in both limiting situation may be different. This difference appears due to the absence of spin accumulation (spin voltage) when the spin diffusion length is short. When the spin diffusion length is long (of the order of or longer than the system length), then both electrical voltage and spin voltage are generated in the system under open circuit boundary conditions. However, when the spin valve is symmetric, i.e. both magnetic films have equal parameters and thickness, the spin thermopower in the antiparallel configuration vanishes, while it appears only in the parallel configuration.

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