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Theory of unidirectional spin Hall magnetoresistance in heavy-metal/ferromagnetic-metal bilayers

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Recent experiments have revealed nonlinear features of the magnetoresistance in metallic bilayers consisting of a heavy metal (HM) and a ferromagnetic metal (FM). A small change in the longitudinal resistance of the bilayer has been observed when reversing the direction of either the applied in-plane current or the magnetization. We attribute such nonlinear transport behavior to the spin-polarization dependence of the electron *mobility* in the FM layer acting in concert with the spin accumulation induced in that layer by the spin Hall current originating in the bulk of the HM layer. An explicit expression for the nonlinear magnetoresistance is derived based on a simple drift-diffusion model, which shows that the nonlinear magnetoresistance appears at the first order of the spin Hall angle, and changes sign when the current is reversed, in agreement with the experimental observations. We also discuss possible ways to control sign of the nonlinear magnetoresistance and to enhance the magnitude of the effect.

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Recently, nonlinear magnetoresistance has been observed experimentally in several heavy-metal (HM)/ferromagnetic-metal (FM) bilayers [1–3]. In these experiments, both the current and the magnetization of the FM layer lie in the plane of the layers and are mutually perpendicular, as shown in Fig. 1. For a given magnitude of the current density (in the range of 10^7–10^8 A/cm^2), it has been found that the longitudinal resistance changes when the current direction is reversed. Furthermore, by injecting an ac current, it has been observed that the second harmonic component of the longitudinal resistance changes sign as the magnetization direction is reversed [1]: This shows that, different from the familiar linear transport, the magnetoresistance has a linear dependence on the current density.

A definitive interpretation of these experimental observations has not yet emerged. Avci *et al.* associated the nonlinear magnetoresistance with the modulation of interface scattering potential induced by the spin Hall effect and the ensuing interfacial resistance change, similar to the interfacial contribution of giant magnetoresistance (GMR) [4–7]. Another interpretation of the effect invokes magnon excitation in the FM layer due to electron spin-flip scattering at the interface [2,3]. While this process has recently been shown to play a key role in the spin-charge conversion in HM/ferromagnetic-insulator (FI) layered structures [8–13], it is usually neglected in metallic systems, for which it is a good approximation to assume that the spin current is continuous at the interface [4].

In this Rapid Communication, we present a simple analytical theory of the nonlinear magnetoresistance in HM/FM bilayers. We propose that the effect arises from the combined action of spin accumulation induced by the spin Hall effect in the HM layer and the spin-polarization dependence of the electron mobility in the FM layer. As schematically shown in Fig. 1, when an in-plane current is driven in a HM/FM bilayer, a spin Hall current flowing perpendicular to the layers is generated in the bulk of the HM layer and subsequently creates spin accumulation on both sides of the interface. Spin accumulation is known as a local quantity that characterizes an excess density of electrons with one specific spin orientation and a corresponding depletion of electrons with the opposite spin orientation, so that no charge accumulation is created. Although such local spin-dependent density variation in the HM layer would not alter the conductivity of the layer in which the mobility of electrons is spin independent, the conductivity of the FM layer is indeed modified by the spin accumulation inside the layer. This may be best understood by thinking of the spin accumulation near the interface as an artificial ferromagnetic layer. Based on our understanding of the current-in-plane (CIP) GMR [14–17], we would anticipate a change in longitudinal resistance when the "magnetization" of the artificial FM layer (i.e., the direction of the spin accumulation) switches from parallel to antiparallel (or vice versa) to that of the "natural" FM layer. The only difference from CIP-GMR lies in the fact that "magnetization" of the artificial FM layer is generated by the electric current itself via the spin transport perpendicular to the layers. This simple analogy immediately demonstrates the nonlinear character of the corresponding magnetoresistance effect.

When an in-plane current is applied to a HM/FM bilayer, a spin current propagating perpendicular to the layers is generated by the spin Hall effect [18–21] in the HM. This transverse spin current affects the linear in-plane resistivity via the inverse spin Hall effect, a phenomenon that has been intensively studied [22–28] and goes under the name of "spin Hall magnetoresistance" [29,30]. In addition, the modulation of the electron spin density in the ferromagnet generates a nonlinear resistivity, as discussed above and shown in detail below. Here, we treat both linear and nonlinear contributions on equal footing through a set of equations that couple the spin and charge transport in directions parallel and perpendicular to the plane of the layers.

To be specific, let us assume the external electric field is applied in the x direction, i.e., $\mathbf{E}_{\text{ext}} = E_x\hat{\mathbf{x}}$ (E_x could be either positive or negative), and fix the magnetization vector of the FM layer in the positive $\hat{\mathbf{y}}$ direction, which is also taken as the quantization axis of the electron spin. The HM/FM interface is located at $z = 0$. The general drift-diffusion equation for

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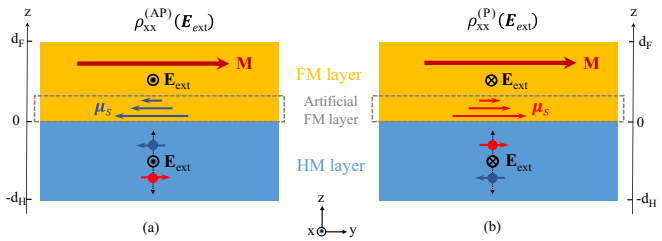

FIG. 1. Schematic diagrams showing the mechanism of nonlinear longitudinal resistivity in HM/FM bilayers due to spin accumulation in the FM layer induced by the spin Hall effect in the HM layer. The external electric field $\mathbf{E}_{\rm ext}$ is applied in the positive x direction in (a) and in the negative x direction in (b). The dotted arrows in the HM layer denote the directions of the spin Hall currents. The solid arrows in the gray dashed boxes describe the magnitude and direction of the spin accumulation $\boldsymbol{\mu}_s$ which may be regarded as an artificial FM layer. The difference in longitudinal resistivity of the bilayer in (a) antiparallel and (b) parallel configurations arises from the spin dependence of the mobility in the FM layer, in analogy with CIP-GMR.

electrons with spin orientation α [$\alpha = \pm$ or $\uparrow$ ($\downarrow$) denoting the spin orientation parallel or antiparallel to the magnetization] can be written as follows,

$$j_x^\alpha(z) = \sigma^\alpha(z)E_x - \alpha\theta j_z^\alpha(z) \tag{1}$$

and

$$j_z^\alpha(z) = \sigma^\alpha(z)\frac{d}{dz}\mu^\alpha(z) + \alpha\theta j_x^\alpha(z), \tag{2}$$

where j_i^α is the current density carried by spin-α electrons with $i = x$ or z denoting the spatial direction of flow, θ is the bulk spin Hall angle (SHA), and $\mu^\alpha(z)$ is the spin-dependent chemical potential, which is related to the nonequilibrium part of the electron density $n^\alpha(z)$ as follows,

$$\mu^\alpha(z) = [N^\alpha(\varepsilon_F)]^{-1}n^\alpha(z) - \phi(z), \tag{3}$$

with $N^\alpha(\varepsilon_F)$ being the density of states of spin-α electrons at the Fermi level, and $\phi(z)$ being the spin-independent part of the chemical potential. Notice that in Eqs. (1) and (2) we have assumed a spatially dependent local conductivity controlled by the electron spin density as follows,

$$\sigma^\alpha(z) = \nu^\alpha\big[n_0^\alpha + n^\alpha(z)\big], \tag{4}$$

where n_0^α and ν^α are the equilibrium density and the mobility of spin-α electrons, respectively. The charge and y-spin current densities are defined as $j_i(z) \equiv j_i^\uparrow(z) + j_i^\downarrow(z)$ and $Q_i^y(z) \equiv j_i^\uparrow(z) - j_i^\downarrow(z)$.

We also assume that charge neutrality is locally maintained, i.e.,

$$n^\uparrow(z) + n^\downarrow(z) = 0. \tag{5}$$

In metals, this is justified by the observation that the integrated space charge vanishes beyond a very short screening length, of the order of angstroms. This supplemental condition links the transport in the two spin channels. In what follows, we shall discuss the transport in each layer separately.

In the HM layer, the equilibrium conduction electron density is spin independent and therefore transport coefficients such as the mobility and the diffusion constant can be taken to be spin independent up to first order in the current-induced spin polarization. Equations (1) and (2) reduce to

$$j_x^\alpha(z) = \sigma_H^\alpha(z)E_x - \alpha\theta_H j_z^\alpha(z) \tag{6}$$

and

$$j_z^\alpha(z) = \frac{1}{2}\alpha\theta_H\sigma_{0,H}E_x + \sigma_{0,H}\frac{d}{dz}\mu^\alpha(z), \tag{7}$$

where $\sigma_{0,H} = \nu_H n_{0,H}$ is the bulk Drude conductivity of the HM. Notice that in the second of these equations we are keeping only terms up to first order in θ_H.

In a steady state, the spin-dependent current density satisfies the generalized continuity equation

$$\frac{d}{dz}j_z^\alpha(z) = \sigma_{0,H}\frac{d^2}{dz^2}\mu^\alpha(z) = \frac{n^\alpha(z) - n^{-\alpha}(z)}{\tau_{{\rm sf},H}}, \tag{8}$$

where $\tau_{{\rm sf},H}$ is the spin-flip relaxation time. With Eq. (3), we may express the right-hand side (rhs) of this equation in terms of the chemical potentials which are found to satisfy the following differential equations,

$$\frac{d^2}{dz^2}\mu_c(z) = 0 \tag{9}$$

and

$$\frac{d^2}{dz^2}\mu_s(z) - \frac{\mu_s(z)}{L_H^2} = 0, \tag{10}$$

where we have defined the sum and difference of the chemical potentials as $\mu_c(z) \equiv [\mu^\uparrow(z) + \mu^\downarrow(z)]/2$ and $\mu_s(z) \equiv [\mu^\uparrow(z) - \mu^\downarrow(z)]/2$, respectively, and $L_H \equiv \sqrt{\sigma_{0,H}\tau_{{\rm sf},H}/2N_H(\varepsilon_F)}$ as the spin diffusion length.

For the transport in the FM layer, we neglect the anomalous Hall effect since the SHA is usually an order of magnitude smaller than that in the HM layer. This assumption allows us to simplify the equations for current densities in FM layer as

$$j_x^\alpha(z) = \nu_F^\alpha\big[n_{0,F}^\alpha + n^\alpha(z)\big]E_x \tag{11}$$

and

$$j_z^\alpha(z) = \sigma_{0,F}^\alpha\frac{d}{dz}\mu^\alpha(z), \tag{12}$$

with $\sigma_{0,F}^\alpha$ being the bulk conductivity of the spin-α channel in the FM layer. In a steady state, the continuity equation reads

$$\frac{d}{dz}j_z^\alpha(z) = \sigma_{0,F}^\alpha\frac{d^2}{dz^2}\mu^\alpha(z) = \frac{n^\alpha(z) - n^{-\alpha}(z)}{\tau_{{\rm sf},F}}. \tag{13}$$

Making use of Eqs. (3) and (5) to express $n^\alpha - n^{-\alpha}$ in terms of $\mu^\alpha - \mu^{-\alpha}$, we find that the equation for μ_c takes the form

$$\frac{d^2}{dz^2}\mu_c(z) + p_\sigma\frac{d^2}{dz^2}\mu_s(z) = 0, \tag{14}$$

where $p_\sigma \equiv (\sigma_{0,F}^\uparrow - \sigma_{0,F}^\downarrow)/(\sigma_{0,F}^\uparrow + \sigma_{0,F}^\downarrow)$ is the conductivity spin asymmetry. On the other hand, the equation for μ_s remains of the same form as in the HM layer [Eq. (10)], except for replacing L_H by the ferromagnetic spin diffusion length $L_F = \sqrt{\sigma_{0,F}(1-p_\sigma^2)\tau_{{\rm sf},F}/2N_F(\varepsilon_F)(1-p_N^2)}$, where

$p_N \equiv (N_F^{\uparrow} - N_F^{\downarrow})/(N_F^{\uparrow} + N_F^{\downarrow})$ is the spin asymmetry in the density of states at the Fermi level.

For the boundary conditions at the interface ($z = 0$), neglecting interfacial spin-flip scattering and the small interfacial resistance [4,31], we assume that both the spin current density flowing in the z direction and the chemical potentials are continuous, i.e., $Q_z^y(0^-) = Q_z^y(0^+)$ and $\mu^{\alpha}(0^-) = \mu^{\alpha}(0^+)$. At the same time, since there is no charge flow in the z direction, we set $j_z(z) = 0$ everywhere. Also, we take Q_z^y to vanish at the two outer surfaces, i.e., at $z = -d_H$ for the HM layer and $z = d_F$ for the FM layer, with d_H and d_F being the thicknesses of the HM and FM layers, respectively.

By inserting the general solutions of the chemical potentials and the spin current densities into the boundary conditions for each interface, we can now determine all transport quantities of interests. For example, up to first order in θ_H, the in-plane charge current density in the FM layer is given by

$$j_x(z) = \sigma_{0,F} E_x + \tfrac{1}{2}(\nu_F^{\uparrow} - \nu_F^{\downarrow})[n^{\uparrow}(z) - n^{\downarrow}(z)]E_x, \tag{15}$$

where $\sigma_{0,F} = n_{0,F}^{\uparrow}\nu_F^{\uparrow} + n_{0,F}^{\downarrow}\nu_F^{\downarrow}$ is the total bulk conductivity of the FM and the spin accumulation is given by

$$n^{\uparrow}(z) - n^{\downarrow}(z) = -\frac{2(\theta_H L_H)N_F(\varepsilon_F)\left(1 - p_N^2\right)\tanh\left(\frac{d_H}{2L_H}\right)\cosh\left(\frac{d_F - z}{L_F}\right)}{\cosh\left(\frac{d_F}{L_F}\right) + \left(1 - p_\sigma^2\right)\left(\frac{\sigma_{0,F}L_H}{\sigma_{0,H}L_F}\right)\sinh\left(\frac{d_F}{L_F}\right)\coth\left(\frac{d_H}{L_H}\right)} E_x. \tag{16}$$

Note that the negative sign in front of the expression on the rhs of Eq. (16) implies that minority electrons are accumulated near the interface when both θ_H and E_x are positive.

Equations (15) and (16) are quite remarkable. First, we observe that the correction to the in-plane charge current density [i.e., the second term on the rhs of Eq. (15)] is proportional to E_x^2, since $n^{\uparrow}(z) - n^{\downarrow}(z)$ is the linear response of the spin density to the external electric field. Second, the nonlinear contribution appears at the first order in the SHA, in contrast to the linear spin Hall magnetoresistance which is known to be of second order in the SHA [30]. The above features qualitatively agree with recent experimental observations [1,32].

Equation (15) makes clear that, in our interpretation, the spin dependence of the electron *mobility*, i.e., the nonzero value of $(\nu_F^{\uparrow} - \nu_F^{\downarrow})$, is essential to the appearance of a nonlinear magnetoresistance. Indeed, if the mobilities were not spin dependent, the total in-plane conductivity $\sigma^{\uparrow} + \sigma^{\downarrow}$ would remain unchanged by virtue of the charge neutrality condition (5). This is exactly what happens in the HM layer, where the in-plane charge current remains unchanged up to $O(\theta_H)$. The underlying physics is rather transparent: If majority electrons in the FM layer exhibit higher mobility than minority electrons [i.e., $\nu_F^{\uparrow} > \nu_F^{\downarrow}$], then accumulation of majority electrons will lead to an increase in the conductivity, and vice versa. The crucial role of spin asymmetry in the electron mobility of the FM is also consistent with the absence of nonlinear magnetoresistance effect in HM/FI bilayers [such as Pt/yttrium iron garnet (YIG)] measured in recent experiments [32].

The total longitudinal resistivity of the bilayer can be calculated as $\rho_{xx} = (d_H + d_F)E_x/\int dz j_x(z)$, where the current density is integrated over the thickness of the bilayer. Similar to GMR, the amplitude of the unidirectional spin Hall magnetoresistance (USMR) is characterized by the ratio

$$\text{USMR} = \frac{\rho_{xx}(E_x) - \rho_{xx}(-E_x)}{\rho_{xx}(E_x)}. \tag{17}$$

Up to first order in θ_H, we obtain

$$\text{USMR} \simeq 6\left(\frac{\sigma_{0,F}L_F}{\sigma_{0,H}d_H + \sigma_{0,F}d_F}\right)\frac{(p_\sigma - p_N)(\theta_H E_x L_H/\varepsilon_F)\tanh\left(\frac{d_H}{2L_H}\right)\tanh\left(\frac{d_F}{L_F}\right)}{1 + \left(1 - p_\sigma^2\right)\left(\frac{\sigma_{0,F}L_H}{\sigma_{0,H}L_F}\right)\tanh\left(\frac{d_F}{L_F}\right)\coth\left(\frac{d_H}{L_H}\right)}, \tag{18}$$

where we have used the relations $\nu_F^{\alpha} = \sigma_{0,F}^{\alpha}/n_{0,F}^{\alpha}$ and $N_F^{\alpha} = 3n_{0,F}^{\alpha}/2\varepsilon_F$ for the free-electron model with ε_F being the Fermi energy of the FM. Note that $\nu_F^{\uparrow} - \nu_F^{\downarrow}$ is proportional to the difference of the spin polarization of conductivity p_σ and of density of states at Fermi energy p_N.

In Fig. 2, we plot the USMR as a function of the thickness of one layer while the thickness of the other is fixed. As we have pointed out previously, although the leading order nonlinear correction to the in-plane current density only occurs in the FM layer, the HM layer also plays an essential role by inducing the spin accumulation in the FM layer via the spin Hall effect. Therefore, when the thickness of either layer becomes much smaller than the corresponding spin diffusion length, the USMR diminishes. On the other hand, when the thickness of either layer is much larger than the spin diffusion length, more current is shunted into the bulk of the layers and hence the interfacial effect of USMR gets diluted, as indicated by the prefactor on the rhs of Eq. (18). Not surprisingly, the USMR peaks around the respective spin diffusion length of each layer. The dependence of the USMR on the thickness of the HM layer agrees qualitatively with experiments, whereas the dependence on the thickness of the FM layer has not yet been measured.

With Eq. (18), we can also make quantitative comparisons of the calculated magnitude of USMR with experimentally observed values. For a Pt (6 nm)/Co (3 nm) bilayer

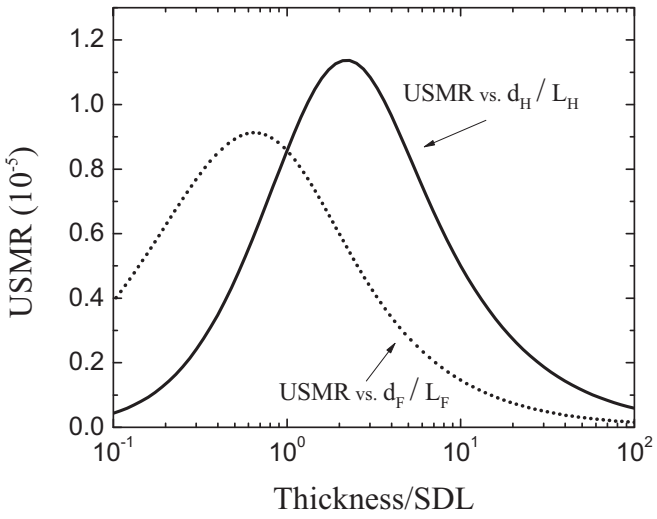

FIG. 2. USMR as a function of the thickness of the HM layer [scaled with the spin diffusion length (SDL) L_H] for fixed thickness of the FM layer of $d_F = L_F = 10$ nm (solid line) and as a function of the thickness of the FM layer (scaled with L_F) for fixed thickness of the HM layer of $d_H = L_H = 5$ nm (dotted line). Other parameters assumed in the numerical calculation are $\theta_H = 0.1$, $|E_x| = 10^{-4}$ V/nm, $p_\sigma - p_N = 0.5$, $\epsilon_F = 5$ eV, and $\sigma_{0,H} = \sigma_{0,F} = 0.033\ (\mu\Omega\,\mathrm{cm})^{-1}$. The calculations are done to first order in SHA.

with $E_x = 10^{-4}$ V/nm and the following material parameters, $\theta_H = 0.1$ [1,33,34], $\sigma_H = 0.02\ (\mu\Omega\,\mathrm{cm})^{-1}$, $L_H = 5$ nm, $\sigma_F = 0.05\ (\mu\Omega\,\mathrm{cm})^{-1}$, $L_F = 40$ nm [31], $\varepsilon_F = 5$ eV, and $p_\sigma - p_N = 0.5$, we find USMR $\simeq 0.9 \times 10^{-5}$, which is only a factor of 2 smaller than the experimental value [1,32]. We have checked that the USMR is negligibly reduced in the presence of an interfacial resistance $r_I \sim 1$ fΩ m [35].

We notice that the magnitude of the USMR may be underestimated due to several simplifying assumptions we adopted in our model calculation. First, in the derivation of Eq. (18), we assumed spherical Fermi surfaces and constant density of states at the Fermi energy. Strong energy dependence of the density of states near the Fermi surface (e.g., in Ni [36]) may enhance the effect just as it enhances the spin accumulation-induced nonlinear GMR effect observed in dual spin valves [36]. Second, we neglected the indirect influence of the spin accumulation on transport parameters such as the bulk and interfacial resistances due to electron-electron correlation or shift of the scattering potential.

In the presence of interfacial spin-flip scattering, there would be a partial loss of spin current across the interface (known as spin memory loss [31,37–40]). This effect can be easily incorporated in our treatment through a simple change in the boundary conditions for the spin current [41]. Spin memory loss, treated in this way, results in a reduction of the USMR given in Eq. (18) by a factor of order unity. However, the absorbed spin current may in turn lead to additional contribution to the USMR via interfacial spin-dependent scattering [42].

An interesting observation based on Eq. (18) is that the USMR depends linearly on the difference of $p_\sigma - p_N$, which suggests that the sign of the USMR also depends on the overall sign of $p_\sigma - p_N$. In Ref. [43], Fert and Campbell showed that the signs of p_σ for various binary alloys of transition metals may change depending on the relative position of the d bands of the host and the impurity. For example, they showed that p_σ of NiFe is positive whereas that of FeCr is negative [43]. By making use of this property, a "reversed" CIP-GMR, that is to say, a CIP-GMR in which the antiparallel alignment of magnetizations has *lower* resistance than the parallel arrangement, could be explained in a Fe/Cu superlattice with half of the Fe layers being intercalated with thin Cr layers [44]. Similar experiments can be carried out to test our theory of USMR. For example, we predict that the USMR in Pt/FeCr should have an opposite sign to that in the Pt/NiCr bilayer.

Our model calculation also suggests several ways to enhance the USMR. For metallic systems, the effect would be amplified in an asymmetric trilayer structure of the form $HM_1/FM/HM_2$, with HM_1 and HM_2 having opposite signs of θ_H (for example, $HM_1 = Pt$ and $HM_2 = Ta$). In such a structure, the orientations of the spin accumulations on opposite sides of the FM layer will be identical, hence the contributions of the two interfaces to the USMR will add constructively. Our theory also suggests that an enhanced USMR may be found in paramagnetic and ferromagnetic semiconductor bilayers, which have much lower carrier densities than their metallic counterparts. As shown by Eq. (18), the USMR is inversely proportional to the Fermi energy which scales with the equilibrium free-electron density as $\varepsilon_F \propto n_e^{2/3}$. Very recently, Olejník *et al.* found that the USMR in ferromagnetic-(Ga,As)Mn/paramagnetic-(Ga,As)Mn bilayers is larger than that in metallic bilayers by several orders of magnitudes [45], and they attributed the big enhancement to the low carrier densities in their semiconducting systems.

In summary, we have developed a drift-diffusion theory for HM/FM bilayers with an in-plane electric current. The theory is self-consistent in the sense that it takes into account the effect of the current-induced spin accumulation on the longitudinal resistance. The unidirectional magnetoresistance is an effect of first order in the spin Hall angle of the HM layer, in contrast to the linear spin Hall magnetoresistance which is an effect of second order in the spin Hall angle. We have suggested ways to control the sign of the nonlinear magnetoresistance and to amplify the magnitude of the effect by a judicious choice of materials and/or nanostructure engineering. It appears that conducting bilayers consisting of a ferromagnet and a paramagnetic metal with a large spin Hall angle have considerable potential to work as reversible diodes that may be controlled by the magnetic direction of the ferromagnetic layer.

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