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Nonlinear graphene plasmonic waveguides: pulse propagation equation

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ABSTRACT

Treating surface-only nonlinear optical response of graphene as the nonlinear boundary condition in Maxwell equations and applying perturbation expansion, the pulse propagation equation for graphene plasmonic waveguides is derived. Effective nonlinear coefficient due to the graphene is derived and compared to bulk nonlinear response of dielectrics.

Keywords: Graphene plasmonics, active plasmonics, surface nonlinear waves, pulse propagation equation

1. INTRODUCTION

Applications of graphene in photonics and optoelectronics are being actively discussed in recent years.^{1,2} In particular, graphene plasmonics is considered as a promising alternative to conventional plasmonics with noble metals.^{3,4} Compared to conventional metals, graphene offers tunability, low losses and extremely high nonlinear optical response. The latter has been predicted theoretically to exceed typical response of dielectrics by at least eight order of magnitude.^{5,6} The particularly strong nonlinear response of graphene has been confirmed in several experiments, including direct measurements with optical Kerr gate⁷ and z-scan⁸ techniques, as well as observation of four-wave mixing with graphene flakes,⁹ a range of nonlinear effects in a graphene-coated photonic crystal nano-cavity,¹⁰ third harmonic generation from single- and multi-layered graphene flakes.¹¹

Being purely two-dimensional crystal, graphene is fundamentally different from any bulk material, and its nonlinear optical response requires adequate theoretical description. Recently, we developed a modification of the conventional perturbation expansion procedure of Maxwell equations to include graphene as the nonlinear boundary condition.¹² This method has been applied for analysis of CW nonlinear graphene plasmons in single- and multi-layered planar structures,^{12,13} and to investigate the enhancement of nonlinearity in a graphene-clad tapered dielectric fibre.¹⁴ In this work, we further develop the perturbation expansion procedure to consider frequency-mixing and nonlinear pulse propagation in a single-layer graphene plasmonic waveguide and graphene nanoribbon waveguides.

2. PERTURBATION EXPANSION OF MAXWELL EQUATIONS

Consider a waveguide with a fixed structure in the (x, y) -plane and homogeneous along the propagation direction z . To analyse pulse propagation in the structure, it is convenient to use Fourier expansion of the total electric field:

$$\vec{\mathcal{E}}(\vec{r}, t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} \mathbf{E}(\vec{r}, \omega) e^{-i\omega t} d\omega + c.c. \quad (1)$$

Each Fourier component \mathbf{E} solves Maxwell equations:

$$\vec{\nabla} \times \vec{\nabla} \times \mathbf{E} = \frac{\omega^2}{c^2 \epsilon_0} \mathbf{D}. \quad (2)$$

Optical response of all bulk materials is incorporated in the displacement vector \mathbf{D} . Atom-thick graphene layer is described by means of the surface current \mathbf{J} , the corresponding boundary condition is:

$$\vec{n} \times [\mathbf{H}_2 - \mathbf{H}_1] = \mathbf{J}, \quad (3)$$

where \vec{n} is the unit vector normal to the graphene layer and pointing from medium 1 to medium 2, which are on either side of the graphene layer.

In this work we will focus on instantaneous cubic (Kerr) nonlinearity of homogeneous isotropic media, so that the displacement vector and the induced current in graphene have the form, respectively:

$$\mathbf{D}(\omega) = \epsilon_0 \epsilon \mathbf{E}(\omega) + \epsilon_0 \hat{\chi}^{(3)} : \mathbf{E}(\omega_1) \mathbf{E}^*(\omega_2) \mathbf{E}(\omega_3), \quad (4)$$

$$\mathbf{J}(\omega) = \hat{\sigma}^{(1)} \mathbf{E}(\omega) + \hat{\sigma}^{(3)} : \mathbf{E}(\omega_1) \mathbf{E}^*(\omega_2) \mathbf{E}(\omega_3), \quad (5)$$

where $\omega_3 = \omega - \omega_1 + \omega_2$, nonlinear susceptibility tensor is given by: $\hat{\chi}_{ipjs}^{(3)} = (\chi_3/3) [\delta_{ip}\delta_{js} + \delta_{ij}\delta_{ps} + \delta_{is}\delta_{pj}]$, δ_{ij} is the Kronecker's delta. Considering graphene as the purely 2D material, and introducing local coordinates (ξ, τ, ζ) with ζ being orthogonal to the graphene layer and (ξ, τ) in-plane of graphene, we assume a similar structure of the nonlinear conductivity tensor $\hat{\sigma}_{ipjs}^{(3)}$, but with the indexes i, p, j, s each from the subset of in-plane coordinates (ξ, τ) . Also, the 2D symmetry of graphene and the requirement $J_\zeta = 0$ still permit six additional non-zero tensor components $\hat{\sigma}_{jj\zeta\zeta}^{(3)} = \hat{\sigma}_{j\zeta j\zeta}^{(3)} = \hat{\sigma}_{j\zeta\zeta j}^{(3)} = \tilde{\sigma}_3/3$, $j = \xi, \tau$. In absence of external magnetic fields, linear conductivity tensor has only two diagonal non-zero components: $\hat{\sigma}_{ii}^{(1)} = \sigma_1$, $i = \xi, \tau$.

It is convenient to decompose the displacement vector and the induced current as $\mathbf{D} = \mathbf{D}_0 + \mathbf{D}_p$, $\mathbf{J} = \mathbf{J}_0 + \mathbf{J}_p$, so that the solution of Maxwell equations with \mathbf{D}_0 and \mathbf{J}_0 gives the linear guided mode of the structure, while \mathbf{D}_p and \mathbf{J}_p are treated as perturbations. Developing perturbation expansion, we introduce a dummy small parameter s assuming $\mathbf{D}_p, \mathbf{J}_p \sim s^{3/2}$. Each Fourier component of the electric field is expanded in the perturbation series as:

$$\mathbf{E} = s^{1/2} I_\omega^{-1/2} A_\omega(z) \mathbf{e}_\omega(x, y) + s^{3/2} \mathbf{E}_1 + O(s^{5/2}), \quad (6)$$

and a similar expansion for the magnetic field is assumed. Here \mathbf{e}_ω is the mode of a given structure, obtained in the lowest $O(s^{1/2})$ order of perturbation analysis, I_ω is the normalization factor to be specified below. In the next order $O(s^{3/2})$ the propagation equation for modal amplitudes $A_\omega(z)$ is derived, which is the ultimate goal of this work. The overall procedure is explained in details in the following sections, starting with a simple planar geometry in Sec. 3, then proceeding to the fully 3D geometry of graphene nanoribbon waveguide in Sec. 4.

3. SURFACE GRAPHENE PLASMONS IN A PLANAR GEOMETRY

Consider a planar geometry with graphene layer sandwiched in-between two semi-infinite dielectrics, as illustrated in Fig. 1. We choose x axis to be perpendicular to the interfaces, z is the direction of propagation, and y is the unbound direction along which homogeneity of fields is assumed. The structure supports surface plasmon waves.^{12, 15, 16} In this case, linear graphene conductivity plays important role and should be included into the lowest order of perturbation procedure. Thus we take:

$$\mathbf{D}_0 = \epsilon_0 \epsilon \mathbf{E}, \quad \epsilon(x < 0) = \epsilon_s, \quad \epsilon(x > 0) = \epsilon_c, \quad (7)$$

$$\mathbf{J}_0 = \begin{bmatrix} 0, i\sigma_1^{(I)} E_y, i\sigma_1^{(I)} E_z \end{bmatrix}^T, \quad (8)$$

where we introduce real and imaginary parts of the linear graphene conductivity: $\sigma_1 = \sigma_1^{(R)} + i\sigma_1^{(I)}$, $\sigma_1^{(R)}$ is responsible for damping of plasmons and will be included in the next order of perturbation together with nonlinear terms (\mathbf{J}_p).

Below we discuss TM plasmons existing for $\sigma_1^{(I)} > 0$. In the lowest order $O(s^{1/2})$ of the perturbation expansion we obtain the boundary value problem for the linear mode $\mathbf{e}(x) = [e_x, 0, e_z]^T$ and the propagation

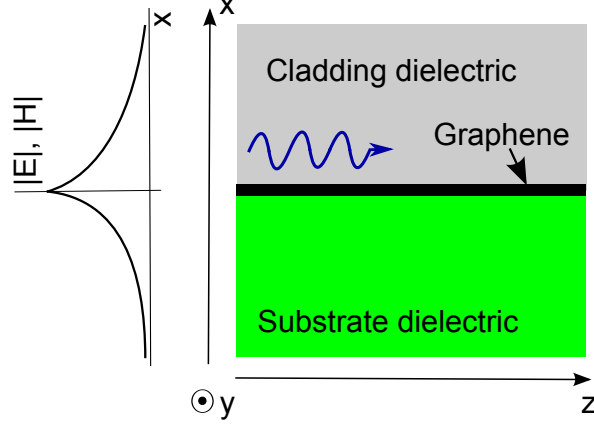


Figure 1. Schematic illustration of surface plasmon propagating along graphene sheet. Fields are exponentially localized in x (across the interface), as shown in the left panel.

constant $\beta(\omega)$:

$$\hat{L}_{TM} \cdot [e_x, e_z]^T = 0, \quad (9)$$

$$\hat{L}_{TM} = \begin{bmatrix} \beta^2 - \epsilon k^2 & i\beta \partial_x \\ i\beta \partial_x & -\partial_{xx}^2 - \epsilon k^2 \end{bmatrix}, \quad (10)$$

$$\Delta[e_z] = 0, \quad \Delta[i\beta e_x - \partial_x e_z] = \frac{\sigma_1^{(I)}}{\epsilon_0 \omega} \Theta[e_z]. \quad (11)$$

Here $k = \omega/c$, operators Δ and Θ are defined as:

$$\Delta[f(x)] = \lim_{\delta \rightarrow 0} (f(-\delta) - f(\delta)), \quad (12)$$

$$\Theta[f(x)] = \frac{1}{2} \lim_{\delta \rightarrow 0} (f(-\delta) + f(\delta)). \quad (13)$$

The corresponding mode can be found analytically in this case:

$$x < 0 : e_z = e^{q_s x}, \quad e_x = \frac{-i\beta}{q_s} e^{q_s x} \quad (14)$$

$$x > 0 : e_z = e^{-q_c x}, \quad e_x = \frac{i\beta}{q_c} e^{-q_c x} \quad (15)$$

$$q_{s,c} = \sqrt{\beta^2 - \epsilon_{s,c} k^2}, \quad (16)$$

where propagation constant β is defined through the dispersion relation:^{5,12}

$$\frac{\epsilon_s}{\sqrt{\beta^2 - \epsilon_s k^2}} + \frac{\epsilon_c}{\sqrt{\beta^2 - \epsilon_c k^2}} = \frac{\sigma_1^{(I)}}{\epsilon_0 \omega}. \quad (17)$$

We choose normalization factor as:

$$I_\omega = \frac{\epsilon_0 \omega}{2\beta} \int_{-\infty}^{+\infty} \epsilon |e_x|^2 dx, \quad (18)$$

so that the total power density (per unit length in the unbound y direction) P_ω carried in the propagation direction z by the mode at the given frequency is:¹²

$$P_\omega = \frac{1}{4} \int_{-\infty}^{+\infty} (\mathbf{E} \times \mathbf{H}^*) \cdot \hat{e}_z dx + c.c. = |A_\omega|^2 + O(s^2), \quad (19)$$

where \hat{e}_z is the unit vector along z -axis.

In the next order $O(s^{3/2})$ we obtain boundary value problem for the correction \mathbf{E}_1 :

$$\hat{L}_{TM} \cdot \begin{bmatrix} E_{1x} \\ E_{1z} \end{bmatrix} = I_\omega^{-1/2} \begin{bmatrix} \partial_z A_\omega (2i\beta e_x - \partial_x e_z) + \frac{\omega^2}{c^2 \epsilon_0} D_{px} \\ -\partial_z A_\omega \partial_x e_x + \frac{\omega^2}{c^2 \epsilon_0} D_{pz} \end{bmatrix} \quad (20)$$

$$\Delta[E_{1z}] = 0, \quad (21)$$

$$\Delta[i\beta E_{1x} + \partial_z A_\omega I_\omega^{-1/2} e_x - \partial_x E_{1z}] = -\frac{i\sigma_1^{(R)}}{\epsilon_0 \omega} I_\omega^{-1/2} A_\omega \Theta[e_z] + \frac{\sigma_1^{(I)}}{\epsilon_0 \omega} \Theta[E_{1z}] - \frac{i}{\epsilon_0 \omega} \Theta[J_{pz}], \quad (22)$$

where nonlinear polarization \mathbf{D}_p and current \mathbf{J}_p and given by:

$$\mathbf{D}_p = \frac{\epsilon_0 \chi_3}{16\pi} \int \int \frac{A_{\omega_1} A_{\omega_2}^* A_{\omega_3}}{\sqrt{I_{\omega_1} I_{\omega_2} I_{\omega_3}}} [(\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_2}^*) \mathbf{e}_{\omega_3} + (\mathbf{e}_{\omega_3} \mathbf{e}_{\omega_2}^*) \mathbf{e}_{\omega_1} + (\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_3}) \mathbf{e}_{\omega_2}^*] d\omega_1 d\omega_2, \quad (23)$$

$$J_{pz} = \frac{\sigma_3}{16\pi} \int \int \frac{A_{\omega_1} A_{\omega_2}^* A_{\omega_3}}{\sqrt{I_{\omega_1} I_{\omega_2} I_{\omega_3}}} [(\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_2}^*)_s e_{\omega_3,z} + (\mathbf{e}_{\omega_3} \mathbf{e}_{\omega_2}^*)_s e_{\omega_1,z} + (\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_3})_s e_{\omega_2,z}^*] d\omega_1 d\omega_2. \quad (24)$$

In the above expressions, (\mathbf{ab}) stands for the standard scalar product, while $(\mathbf{ab})_s = \eta a_x b_x + a_y b_y + a_z b_z$, deformation factor $\eta = \tilde{\sigma}_3/\sigma_3$ characterizes the relative impact of the orthogonal field component on the induced current in the graphene layer.

To proceed, we project Eq. (20) onto the mode \mathbf{e}_ω , split the full integral in the l.h.s. into two parts as $\int_{-\infty}^{\infty} dx = \int_{-\infty}^0 dx + \int_0^{+\infty} dx$ and take the resulting integrals by parts applying boundary conditions from Eqs. (11), (21), (22). As the result, we obtain the following equation describing evolution of the modal amplitude with propagation distance:

$$i\partial_z A_\omega = -(\beta + i\alpha_\omega) A_\omega - \frac{\omega}{2\pi} \int \int_{-\infty}^{\infty} \Gamma_{\omega\omega_1\omega_2\omega_3} A_{\omega_1} A_{\omega_2}^* A_{\omega_3} d\omega_1 d\omega_2, \quad (25)$$

where nonlinear coefficients $\Gamma_{\omega\omega_1\omega_2\omega_3}$ combine contributions from the dielectrics and graphene:

$$\Gamma_{\omega\omega_1\omega_2\omega_3} = \Gamma_{\omega\omega_1\omega_2\omega_3}^{(D)} + \Gamma_{\omega\omega_1\omega_2\omega_3}^{(G)}, \quad (26)$$

$$\Gamma_{\omega\omega_1\omega_2\omega_3}^{(D)} = \frac{\epsilon_0}{16\sqrt{I_\omega I_{\omega_1} I_{\omega_2} I_{\omega_3}}} \int_{-\infty}^{\infty} \chi_3 [(\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_2}^*) (\mathbf{e}_{\omega_3} \mathbf{e}_\omega^*) + (\mathbf{e}_{\omega_3} \mathbf{e}_{\omega_2}^*) (\mathbf{e}_{\omega_1} \mathbf{e}_\omega^*) + (\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_3}) (\mathbf{e}_{\omega_2}^* \mathbf{e}_\omega^*)] dx, \quad (27)$$

$$\Gamma_{\omega\omega_1\omega_2\omega_3}^{(G)} = \frac{1}{16\sqrt{I_\omega I_{\omega_1} I_{\omega_2} I_{\omega_3}}} \frac{i\sigma_3}{\omega} \Theta [(\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_2}^*)_s e_{\omega_3,z} e_{\omega,z}^* + (\mathbf{e}_{\omega_3} \mathbf{e}_{\omega_2}^*)_s e_{\omega_1,z} e_{\omega,z}^* + (\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_3})_s e_{\omega_2,z} e_{\omega,z}^*], \quad (28)$$

and coefficient α_ω gives attenuation of the graphene plasmon:

$$\alpha_\omega = \frac{\sigma_1^{(R)}}{4I_\omega} \Theta [|e_{\omega,z}|^2]. \quad (29)$$

4. GRAPHENE NANORIBBON WAVEGUIDE

The above described procedure can be generalized to fully 3D geometries. Consider a graphene nanoribbon waveguide, in which graphene layer has a finite width along y direction, see Fig. 2. The guided mode now has all three non-zero components of the electric field \mathbf{e}_ω , and needs to be computed numerically.¹⁷ Following similar steps as described in the above section, and applying nonlinear boundary conditions in the relevant region where the graphene nanoribbon is located ($x = 0, -L/2 < y < L/2$), we derive the modal propagation equation (25) with the following coefficients:

$$\Gamma_{\omega\omega_1\omega_2\omega_3}^{(D)} = \frac{\epsilon_0}{16\sqrt{I_\omega I_{\omega_1} I_{\omega_2} I_{\omega_3}}} \times$$

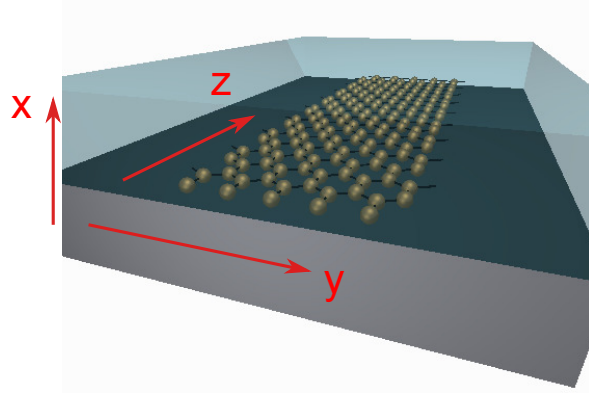


Figure 2. Schematic illustration of a graphene nanoribbon waveguide.

$$\int \int_{-\infty}^{\infty} \chi_3 [(\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_2}^*) (\mathbf{e}_{\omega_3} \mathbf{e}_{\omega}^*) + (\mathbf{e}_{\omega_3} \mathbf{e}_{\omega_2}^*) (\mathbf{e}_{\omega_1} \mathbf{e}_{\omega}^*) + (\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_3}) (\mathbf{e}_{\omega_2}^* \mathbf{e}_{\omega}^*)] dx dy , \quad (30)$$

$$\Gamma_{\omega\omega_1\omega_2\omega_3}^{(G)} = \frac{1}{16\sqrt{I_{\omega}I_{\omega_1}I_{\omega_2}I_{\omega_3}}} \frac{i\sigma_3}{\omega} \times \int_{-L/2}^{L/2} \Theta [(\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_2}^*)_s (\mathbf{e}_{\omega_3} \mathbf{e}_{\omega}^*)_G + (\mathbf{e}_{\omega_3} \mathbf{e}_{\omega_2}^*)_s (\mathbf{e}_{\omega_1} \mathbf{e}_{\omega}^*)_G + (\mathbf{e}_{\omega_1} \mathbf{e}_{\omega_3})_s (\mathbf{e}_{\omega_2}^* \mathbf{e}_{\omega}^*)_G] dy , \quad (31)$$

$$\alpha_{\omega} = \frac{\sigma_1^{(R)}}{4I_{\omega}} \int_{-L/2}^{L/2} \Theta [(\mathbf{e}_{\omega} \mathbf{e}_{\omega})_G] dy , \quad (32)$$

where $(\mathbf{ab})_G = a_y b_y + a_z b_z$ is the reduced scalar product involving only components in-plane of the graphene nanoribbon.

The normalization factor I_{ω} is computed as the total power P_{ω} carried in the propagation direction z by the mode:

$$I_{\omega} = \frac{1}{4} \int \int_{-\infty}^{\infty} (\mathbf{e} \times \mathbf{h}^*) \cdot \hat{\mathbf{e}}_z dx dy + c.c. \quad (33)$$

Note that in the fully 3D geometry I_{ω} is measured in watts, rather than in watts per meter as in the planar case in the previous section.

5. PULSE PROPAGATION EQUATION

Modelling of a nonlinear pulse propagation in a graphene plasmonic waveguide within the derived in previous sections modal propagation equation (25) is a computationally challenging task. A more convenient approach is to derive pulse propagation equation for the Fourier mirror $A(t, z)$ of the modal amplitudes $A_{\omega}(z)$. For spectrally narrow pulses, the common approximation is to neglect dispersion of nonlinear coefficients $\Gamma_{\omega\omega_1\omega_2\omega_3}$, replacing them by the constant value Γ_0 at the pulse reference frequency. A more advanced approach which still takes into account dispersion of nonlinearity, is based on the factorization approximation:¹⁸

$$\Gamma_{\omega\omega_1\omega_2\omega_3} \approx g_{\omega} g_{\omega_1} g_{\omega_2} g_{\omega_3}, \quad g_{\omega} = \Gamma_{\omega\omega\omega\omega}^{1/4} . \quad (34)$$

Accuracy of this factorization approximation must be checked separately for a given geometry and the range of frequencies of interest.

Introducing rescaled modal amplitudes:

$$A_{\omega} = (g_0/g_{\omega}) \Psi_{\omega} , \quad (35)$$

where subscript 0 relates to a reference frequency ω_0 , Eq. (25) becomes:

$$i\partial_z\Psi_\omega = -(\beta + i\alpha_\omega)\Psi_\omega - \frac{\gamma}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \Psi_{\omega_1}\Psi_{\omega_2}^*\Psi_{\omega_3}d\omega_1d\omega_2, \quad (36)$$

where the nonlinear coefficient γ is introduced as follows:

$$\gamma = \omega\sqrt{\Gamma_{0000}\Gamma_{\omega\omega\omega\omega}}. \quad (37)$$

Introducing polynomial fits of $\beta(\delta = \omega - \omega_0)$, $\alpha(\omega)$, $\gamma(\omega)$, and using inverse Fourier transform

$$\psi(t, z) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{+\infty} \Psi_\omega(z)e^{-i(\omega_0+\delta)t}d\delta, \quad (38)$$

the following pulse propagation equation is obtained:¹⁸

$$i\partial_z\psi = -\hat{D}(i\partial_t)\psi - \hat{G}(i\partial_t)(|\psi|^2\psi), \quad (39)$$

$$\hat{D} = \sum_{j=0}^{N_d} \frac{\beta_j + i\alpha_j}{j!} (i\partial_t)^j, \quad (40)$$

$$\hat{G} = \sum_{j=0}^{N_g} \frac{\gamma_j}{j!} (i\partial_t)^j, \quad (41)$$

where β_j , α_j and γ_j are coefficients of polynomial fits of the propagation constant β , attenuation α and nonlinear coefficient γ as functions of $\delta = \omega - \omega_0$.

6. SUMMARY

Treating optical response of graphene as the nonlinear boundary condition in Maxwell equations, we derive the pulse propagation equation for a layered graphene and graphene nanoribbon surface plasmon waveguides. The effective nonlinear coefficient and attenuation coefficient due to the graphene layer are obtained. Calculation of these coefficients is performed by contour integration along the graphene layer, as opposed to surface integration in case of corresponding coefficients for bulk materials. The overall procedure, and the derived coefficients, can be easily extended to other plasmonic and photonic geometries with graphene.

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