

# PLASMON ZEBRA RESONANCES AND NANOPAINTING

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## Abstract

We investigate metal-dielectric metasurfaces composed from periodic metal nanostrips deposited on a dielectric substrate. The metasurface can be termed as plasmon zebra (PZ). The metasurface operates as a set of open plasmon resonators. The theory of plasmon, excited in the open, interconnect resonators, is developed. The large local electromagnetic field is predicted for optical frequencies when plasmon is excited. The reflectance of PZ is much enhanced at the frequency of plasmon resonance and PZ ascribe the color corresponding to the resonance frequency. We propose PZ as simplest but easy tuning plasmon painting.

**Keywords:** plasmon resonance, nanopaint, SERS

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## 1. Introduction

Optical surface waves, known as surface plasmons (SP), can get excited in metal films. The metal permittivity  $\varepsilon_m = \varepsilon'_m + \varepsilon''_m$  is mainly negative for good optical metals like silver or gold where  $\varepsilon'_m < 0$  and  $\varepsilon''_m \ll |\varepsilon_m|$ . SP, which is electromagnetic field bounded with electric charges, can propagate in the metal nanofilms that thickness  $2h$  is much less than the wavelength  $\lambda$ . For example, the symmetric SP, where surface charges have the same signs on both film sides, propagates in a metal nanofilm. The wavevector  $q$  of the symmetric SP is proportional to  $q \sim -\varepsilon_d/(\varepsilon_m h)$ , recall that  $\varepsilon'_m < 0$ . The esteem is obtained for the thin film ( $d \ll \lambda$ ) by considering the film as an inductive plane. Mean free-path estimates as  $l_p = 1/\Im q \sim h |\varepsilon_m|^2 / (\varepsilon_d \varepsilon''_m) \gg h$ , so that it is much larger than the film thickness. The incident light cannot excite SP in the unbound, infinite metal plane since SP velocity is always less than the speed of light. Yet, SP can be easily excited in any finite piece of the of the metal film since SP transfers momentum to the environment by reflecting from the edges. The propagating SP reflects from the edges of the metal film and forms a standing wave. That is a finite patch of the metal film operates as an open plasmon resonator in optical and infrared frequency bands. For example, the simple system of the parallel metal strips operates as a set of the interconnecting plasmon resonators. This plasmon zebra (PZ) system is shown in Fig. 1. SP being excited in  $x$  direction, which is perpendicular to the PZ strips, reflects from edges of a strip. It can also jump from one strip to the neighboring strips. That is SP propagates in the transversal direction ( $x$  direction in Fig. 1). The phase speed of the transversal SP depends on the PZ parameters. Therefore its wavevector  $q$  can be fitted to an arbitrary value by varying the width, thickness of the metal strips as well as by change the gap between strips. The reflectance has maximum(s) at the resonance(s). Being illuminate by natural light PZ ascribe the color in reflection corresponding to the frequency of the plasmon resonance.

To simplify the consideration the quasistatic approximation is used that conveys the main features of the plasmon resonance. We assume that the PZ period is less than  $\lambda/2$ . Then the incident light does not diffract at the PZ but just produces reflected and transmitted EM waves. The evanescent waves, localized around the subwavelength PZ, are mainly due to excitation SP in the PZ. EM properties can be discussed in terms of the effective permittivity  $\varepsilon_e$ , i.e., the film conductance  $\Sigma_e$ , and effective permeability  $\mu_e$ . Note

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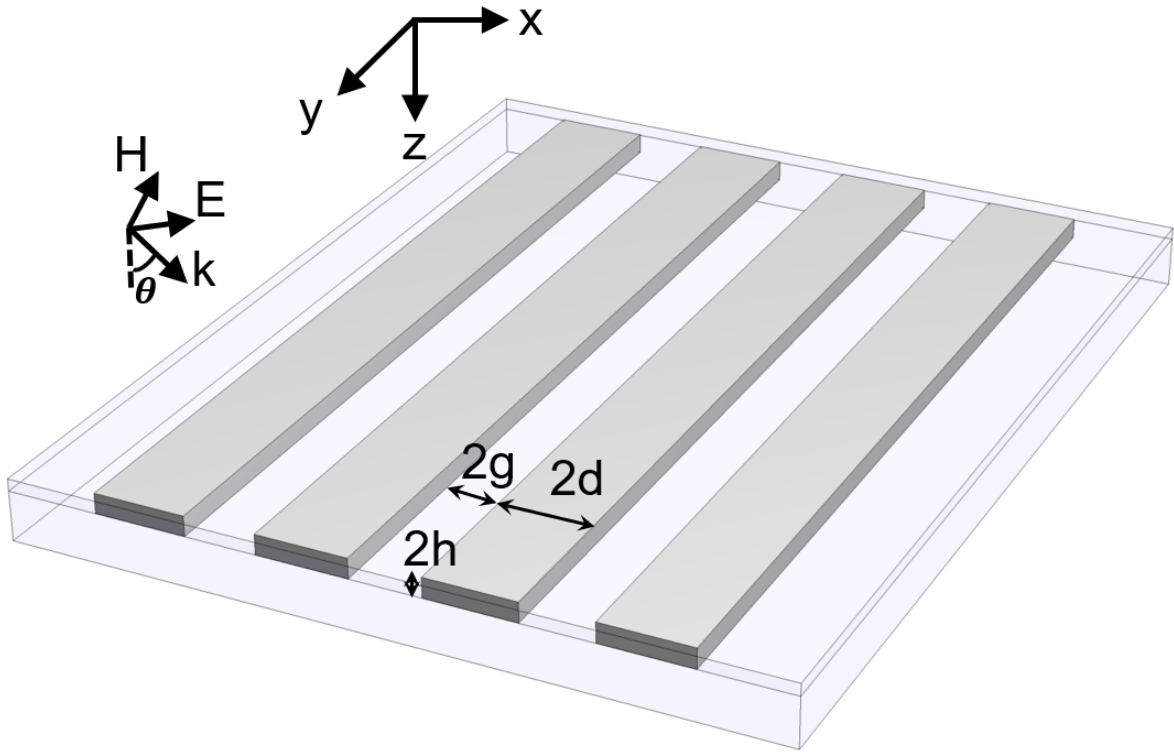


Рис. 1: Metal plasmon zebra (PZ) composed from parallel metal strips (gray color) with thickness  $2h$  and width  $2d$  that are separated by interstrip gates with width  $2g$ ; period of PZ equals to  $2(d+g)$ . Light is incident from above at angle  $\theta$  to the normal. The metal strips are staffed between substrate with permittivity  $\varepsilon_d$  and upper dielectric protective layer with permittivity  $\varepsilon_e$ .

the permeability  $\mu_e$  almost equal to one for the thin PZ. Optical reflectance from PZ achieves maximum when SP is excited in PZ strips. The natural light is composed from EM waves of various wavelengths. The light reflected from PZ is composed mainly from the wavelengths corresponding to the SP resonances. That is metal subwavelength PZ has a color that depends, in general, on the angle of the incidence and light polarization. By fitting the parameters of PZ can be used as a plasmon painting with any wanted color and almost zero thickness. The glass coloring by plasmon nanoparticles was known from the time of the ancient Egypt (see, e.g., Lycurgus Cup in the British museum [1]). One can see the plasmon paintings does not fade for many centuries. Plasmon structures are considered as main ingredient responsible for the beautiful European cathedral-stained glass windows [2]. Recent nanotechnology pave the way for mass production of the light, flexible, nanothin and almost eternal plasmon paintings. Moreover, plasmon paintings are environmentally friendly since they do not contains dies, which are very often rather toxic [3]. Various technologies were studied [4–6] including electron beam lithography [7–15] ion milling, [8, 16–18] and nanoimprint lithography [7]. In recent work of Shalaev’s group [19] a sustainable, lithography-free process is demonstrated for generating non fading plasmon colors with a prototype device that produces a wide range of vivid colors. The extended color palette is obtained through photo-modification by the heating of the localized SP under femtosecond laser illumination [20]. The proposed printing approach can be extended to other applications including laser marking, anti-counterfeiting, and chrome-encryption.

In this paper we present analytical theory for propagating or localized SP in PZ, which is the simplest possible periodic plasmon metasurface consisting of the parallel metal nanostrips. The explicit equations are derived for the reflectance as well as for the local EM field. We use the GOL approximation [21,22] considering EM field around PZ in self consistent way. SP propagating in the lateral directions along the metasurface are incorporated in GOL approach. The developed theory gives the value of the resonance local electric field that can be enhanced by orders on magnitude compared to the impinged light. The reflectance, local field and SERS were found in the system of silicone bars covered by the silver film shown in Fig. 2 [23–26]. Since the resonance fields are much enhanced in the silver bars of this PZ, it is used as SERS substrate. The smooth spatial structure of the film is convenient for the analyte deposition and can be tuned for effective adsorbing and sensing microscopic objects like protein molecules or viruses [27–29].

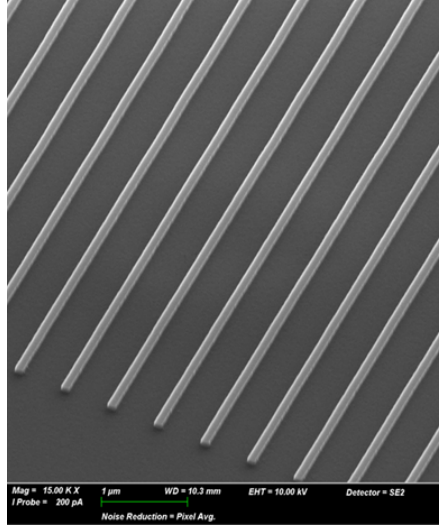


Рис. 2: Scanning electron microscopy image of the silver zebra on silicone substrate (reprint from [24]).

## 2. Quasistatic Theory of Plasmon Resonance

We consider the interaction of a nanothin-thin metal film with thickness  $2h$  with an incident light. As it was mentioned above the metal permittivity  $\varepsilon_m = \varepsilon'_m + \varepsilon''_m$  is mainly negative, namely,  $\varepsilon'_m < 0$  and  $\varepsilon''_m \ll |\varepsilon_m|$ . The film is deposited on the dielectric substrate with the permittivity  $\varepsilon_d$  and it is covered with a protective dielectric layer  $\varepsilon_e$ . The thickness  $2h$  of the metal film is chosen in such a way that the incident light infiltrate in the substrate. The propagation of SP in the metal film that thickness  $2h$  is less than the skin layer can be considered in the quasistatic approximation. In this case the electric field  $E$  of SP can be find in terms of the electric potential  $\varphi$  so that  $E = -\nabla\varphi$ . The electric potential is a solution of the Laplace equation  $\Delta\varphi = 0$ .

We consider the metal film placed at the plane  $z = 0$ . SP propagates over the film in  $x$  direction with wavevector  $q$ . The electric field is invariant under translation in  $y$  direction (see Fig. 1). SP field exponentially decays away from PZ. Electric potential  $\varphi_e$  above the film ( $z < -h$ ) equals to  $\varphi_e = A \exp(iqx) \exp(qz)$ , the potential below film ( $z > h$ ) equals to  $\varphi_d = B \exp(iqx) \exp(-qz)$ . Inside the metal film ( $-h < z < h$ ) the solution of the Laplace equation has the form  $\varphi_m = \exp(iqx)[C_1 \exp(qz) + C_2 \exp(-qz)]$ . To find the electric field  $E$  in SP that is the coefficients  $A, B, C_1$ , and  $C_2$  we use the boundary conditions

$$E_{e,x} = E_{m,x}, \quad \varepsilon_e E_{e,z} = \varepsilon_m E_{m,z}; \quad z = -h, \quad (1)$$

$$E_{d,x} = E_{m,x}, \quad \varepsilon_d E_{d,z} = \varepsilon_m E_{m,z}; \quad z = h, \quad (2)$$

where  $E_e, E_m$ , and  $E_d$  are the electric fields in the protective layer, metal film, and dielectric substrate correspondingly. Equations (1) and (2) have nontrivial solution if and only if the corresponding determinant  $Det$  equals to zero. Thus the SP wavevector  $q$  is obtained from the equation  $Det = 0$

$$q = \frac{1}{4h} \log \left[ \frac{(\varepsilon_e - \varepsilon_m)(\varepsilon_d - \varepsilon_m)}{(\varepsilon_e + \varepsilon_m)(\varepsilon_d + \varepsilon_m)} \right]. \quad (3)$$

Recall the optical metal permittivity is mainly negative  $\Re\varepsilon_m < 0$ . In the limit  $|\varepsilon_m| \gg \varepsilon_e, \varepsilon_d$ , which is typical for the visible and infrared range, the SP wavevector approximates as

$$q \simeq -\frac{\varepsilon_e + \varepsilon_d}{2h\varepsilon_m} \quad (4)$$

, In any case we suppose that the film thickness  $2h$  is much smaller than the SP wavelength that is  $hq \ll 1$ . Then the electric field inside the metal film  $E_m$  does not depend on the normal coordinate "z" and the electric current  $J(x)$  is function of "x", which is a solution of the wave equation for SP, namely,

$$\frac{d^2 J(x)}{dx^2} + q^2 J(x) = q^2 \Sigma E_e(x), \quad (5)$$

where the external field  $E_e$  is added to the r.h.s. The term  $\Sigma$  in r.h.s. of Eq. (5) is in general a linear operator which gives spatial harmonics of the electric nearfield when the external field  $E_e$  is an arbitrary function

of the coordinate "x". To simplify the consideration we take into account the field modulation due to the SP excitation only and approximate as  $\Sigma \simeq 2h\sigma_m$ , where  $\sigma_m - i\frac{\omega\varepsilon_m}{4\pi}$  is the metal conductivity. When the spatial scale of the external field  $E_e(x)$  is much larger than the plasmon wavelength  $\lambda_p = 2\pi/q$  the current  $J(x) = 2h\sigma_mE_e(x)$ . That is we assume that the film current follows the external field and this approach can be called one mode approximation.

The plane EM wave excites SP in the metal nanostrip that have finite width  $2d$  and thickness  $2h \ll 2d$ . Suppose that the strip is illuminated by the light which is incident under the angle  $\theta$  in respect to the normal to the  $x, y$  plane of the film (see Fig. 1). First we consider the plane of incidence, which is perpendicular to the metal strips, that is the wave vector  $k = n_e\omega/c$ , ( $n_e = \sqrt{\varepsilon_e}$ ) has  $x$  and  $z$  components  $\mathbf{k} = k \{ \sin\theta, 0, -\cos\theta \}$ . In  $P$  polarized light the incident electric field has component  $E_{0,x} = E_0 \cos\theta$  and the external electric field in Eq. (5) takes the following form  $E_e = E_{0,x} \exp(ik_x x)$ , where  $E_{0,x}$ , and  $k_x$  are the projections of the field  $E_0$  and the wave vector  $k = n_e\omega/c$  of the impingement light. Assuming that  $|\varepsilon_m| \gg \varepsilon_e, \varepsilon_d$  we apply zero boundary condition  $J(\pm d) = 0$  at edges of a strip (see discussion in the next section) and Eq. (5) gives

$$J(x) = J_1(x) + J_2(x), \quad (6)$$

where the current

$$J_1(x) = \Sigma_e E_e, \quad \Sigma_e = \frac{2hq^2\sigma_m}{q^2 - k_x^2}, \quad (7)$$

does not depend on the edge boundary conditions, and the current

$$J_2(x) = -E_e \Sigma_e \frac{e^{-ixk_x} [e^{iqx} \sin d(k_x + q) - e^{-iqx} \sin d(k_x - q)]}{\sin 2dq} \quad (8)$$

is the current due to the reflection of the plasmon from edges of the strip.

The PS wavevector  $q = q_1 + iq_2$  is in general a complex value, where the imaginary part  $q_2$  estimates from Eq. (3) as

$$q_2 = \frac{\varepsilon_m'' (\varepsilon_d + \varepsilon_e) (\varepsilon_m'^2 - \varepsilon_d \varepsilon_e)}{2h (\varepsilon_m'^2 - \varepsilon_d^2) (\varepsilon_m'^2 - \varepsilon_e^2)}. \quad (9)$$

It is easy to check that  $q_2/q_1 \sim \varepsilon_m''/|\varepsilon_m| \ll 1$ . We obtain from Eq. (8) "odd" plasmon resonances where  $q_1^{(od)} d = (\pi/2)m$ ,  $m = 1, 3, 5 \dots$  and "even" resonances where  $q_1^{(ev)} d = (\pi/2)m$ ,  $m = 2, 4, 6 \dots$ . The electric field  $E_{\max}$  in even and odd SP resonance estimates as

$$E_{\max}^{od} = -E_{0,x} \frac{2i^m q_1^3}{\pi m q_2 (k_x^2 - q_1^2)} \cos(q_1 x) \cos\left(\frac{\pi m k_x}{2q_1}\right) \quad (10)$$

and

$$E_{\max}^{ev} = E_{0,x} \frac{2i^m q_1^3}{\pi m q_2 (k_x^2 - q_1^2)} \sin(q_1 x) \sin\left(\frac{\pi m k_x}{2q_1}\right) \quad (11)$$

correspondingly. Since we consider narrow strips where  $d \ll \lambda$ , i.e.,  $q_1 \gg k_x$  the odd resonances estimate as  $|E_{\max}^{ev}/E_{0,x}| \sim |k_x/q_2|$  and it is much less than the even resonances  $|E_{\max}^{ev}/E_{0,x}| \sim q_1/q_2 \sim |\varepsilon_m'/\varepsilon_m''| \gg 1$ . When the light is impinged normal to the thin metal strip, i.e.,  $k_x = 0$  the even resonances get excited only. In  $S$  polarization when the electric field is perpendicular to the plane of incidence the external electric field  $\{0, E_0, 0\}$  is aligned with the strip direction. Then the current  $J_y = \Sigma_e E_0$  flows along continuous nanothin metal strip.

For example, we consider the strip embedded in the dielectric host with permittivity  $\varepsilon_e = \varepsilon_d$ . The condition for the first resonance  $qd = \pi/2$  can be rewritten by substituting the wavevector  $q$  from Eq. (4), obtaining  $\frac{\varepsilon_m h}{\varepsilon_d d} = \frac{2}{\pi}$ . This result close to the well known quasistatic result for the plasmon resonance in the prolate metal  $2D$  ellipsoid (semi-axes  $h \ll d$ ) which resonates when  $\frac{\varepsilon_m h}{\varepsilon_d d} = 1$  (see, e.g., [30]).

The similar consideration holds when  $\{y, z\}$  is the plane of incidence. That is EM wave is impinged along the strip. In this case  $S$  polarized EM wave excites SP since the  $S$  electric field is directed across the strip whereas the resonance conditions remains the same. Thus we obtain that the natural light, which contains EM waves with various polarization, excites SP in a metal strip for any direction of the incidence. Above equations are derived for a single metal strip.

Consider now PZ composed from the periodic system of the parallel metal nanostrips. It is still assumed that the strip thickness  $2h$  is much less than the skin depth so the electric field does not depend on the coordinate  $z$  in the metal strip. Recall the width of a strip equals  $2d$ , the gap between neighboring

strips equals to  $2g$  (see schematic Fig. 3), so the PZ period equals to  $2d + 2g < \lambda/2$ . Conductance of PZ is anisotropic: The conductance in  $y$  direction equals to  $\Sigma_{yy} = \Sigma_e p$ , where  $p = d/(d + g)$  is the part of the  $z = 0$  interface, which is covered by the metal strips, the surface conductance  $\Sigma_e$  is given by Eq. (6).

To find the conductance  $\Sigma_{xx}$  across the strips it is necessary to find the electric current when the transverse external field  $E_e$  is applied. The current inside the strip is still given by Eq. (6). However, the current  $J(x)$  does not now vanish at the edges of the strip since two neighboring strips have the capacity connection. The interstrip capacitance connects neighboring metal strips that results in collective response of

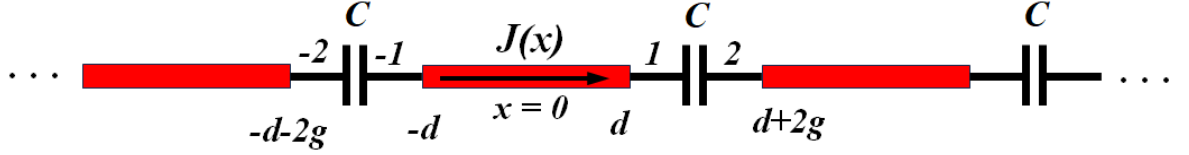


Рис. 3: Lumped circuit of plasmon zebra with period  $2d + 2g$ ; electric current  $J(x)$  flows in metal nanostraps that are connected via interstrip capacitance  $C$ .

PZ to the external field. The interstrip capacitance  $C$  shown in Fig. 3 we approximate as capacitance between two thin strips made of perfectly conducting metal. Two perfectly conducting strips are placed at interface  $z = 0$  between upper half space  $z > 0$  with permittivity  $\varepsilon_e$  and lower half -space  $z < 0$  with permittivity  $\varepsilon_d$ . First perfect strip has width  $2d$  and it is centered at the origin of the coordinate. Second perfectly conducting also has width  $2d$  and its center is at the coordinate  $x = 2(d + g)$ . The gap between right and left edges of the strips equal  $2g$ . To find the capacitance  $C$  we suppose left strip has the electric charge  $Q$  and right strip has charge  $-Q$ . Then the electric field is elementary found from complex variable theory. We introduce the complex variable  $u = x - d - g + iz$ , then the complex electric field  $E_g(u) = E_{gx}(u) + iE_{gz}(u)$  equals to

$$E_g(u) = E_{gx}(u) - iE_{gz}(u) = \frac{E_{0g} d(2d + g)}{\sqrt{(d^2 - u^2)((2d + g)^2 - u^2)}}, \quad (12)$$

where  $E_{gx}$  and  $E_{gz}$  take real values that are proportional to the charge  $Q$ . The complex electric field  $E_g$  is an analytical function. Therefore their components  $E_{gx}(u)$  and  $E_{gz}(u)$  are solutions of the Laplace equation. The branch of the analytical function  $E_g(u)$  is chosen so that  $E_g(0) = E_{0g}$ . The electric field  $E_g$  has only "z" component on the surface of the metal plates. Therefore, the electric charge equals to

$$Q_1 = i\alpha \int_{-2d-g}^{-g} E_g(u) du = E_{0g} \alpha g K \left( \frac{4d(d+g)}{(2d+g)^2} \right) \simeq E_{0,g} \alpha g \log \left( \frac{8d}{g} \right) \quad (13)$$

for the electric field  $E_g$  given by Eq. (12), where  $\alpha = \frac{\varepsilon_e + \varepsilon_d}{4\pi}$ . The last esteem holds for the narrow slit between the strips when  $g \ll d$ . The electric charge  $Q_2$  on the right strip has the opposite sign  $Q_2 = -Q_1$ .

The electric field  $E_g$  has  $x$  component only in the gap  $-g < u < g$  between the metal plates. The electric field  $E_s$  at the edge of the strip is estimated assuming that the strip thickness  $2h$  is much smaller than the gap width, i.e.,  $g \gg h$ . Strictly speaking the field given by Eq. (12) goes to infinity exactly at the edges where  $u = \pm g$ . Since a metal strip has the finite thickness  $2h$  we substitute the coordinate  $u_{1,2} = \pm(g - 2h)$  in Eq. (12) obtaining

$$E_{1,2} = E_g(u_{1,2}) = \mp E_{0,g} \frac{(2d + g)}{4\sqrt{d(d+g)}} \sqrt{\frac{g}{h}}, \quad (14)$$

where the condition  $g \gg h$  is taken into account. The current flows out of the edge  $u_2$  equals to  $J_2 = (\sigma_e + \sigma_d)hE_2$ , where  $\sigma_{e,d} = -i\frac{\omega\varepsilon_{e,d}}{4\pi}$ . The ratio of the derivative of the charge with respect to time  $-dQ/dt = i\omega Q_1$  to the current  $J$  equals to

$$N = -4 \frac{\sqrt{d(d+g)}}{(2d+g)} \sqrt{\frac{g}{h}} K \left( \frac{4d(d+g)}{(2d+g)^2} \right). \quad (15)$$

It is a dimensionless quantity, which depends on the geometry of the system, namely, the strip width  $2d$ , strip thickness  $2h$  and the gap  $2g$  between the neighboring trips.

On the other hand the electric charge  $Q$  at the edge of the strip (see Fig. 3) estimates from the charge conservation law as  $Q \sim \frac{l}{i\omega} \frac{dJ(x)}{dx} \Big|_{x=d}$ , where the current  $J(x)$  is given by Eq. (5),  $l$  is the characteristic length for the charge distribution. The capacity connection between the metal strips in PZ is important at the plasmon resonances when the number of maxima of the current  $|J(x)|$  equals to the order  $m$  of the

resonance. Then the length  $l$  could be estimated as  $l \sim d/m$ , where  $m = 1, 2, 3, \dots$  is the order of the resonance. We obtain the following boundary condition for the electric current at the strip edges.

$$\left. \frac{d}{J(x)} \frac{dJ(x)}{dx} \right|_{x=d} = - \left. \frac{d}{J(x)} \frac{dJ(x)}{dx} \right|_{x=-d} = \gamma m N = -\beta, \quad (16)$$

where  $\gamma$  is a numerical coefficient. The value of  $\gamma$  can be obtained by comparison with computer simulation. Below, to simplify consideration, we put  $\gamma \approx 1$ . The important parameter  $\beta$  approximates as  $\beta \simeq 2\pi\sqrt{d}/h$  for the isolated strips when the gap  $g \gg d$ . It is independent on the gap value  $g$  and much larger than one  $\beta \gg 1$ , which corresponds to the boundary conditions  $J(\pm d) = 0$  used in the previous section. In the opposite case of a narrow slit between the strips  $g \ll d$  the boundary parameter  $\beta \simeq 2\sqrt{\frac{g}{h}} \log\left(\frac{8d}{g}\right)$  so it is again much larger than one since we consider narrow strips which thickness  $h \ll g, d$ .

The plasmon current is obtained from the solution of Eq. (5) with Eq. (16) boundary conditions

$$J(x) = J_1(x) + J_2(x), \quad J_2(x) = -E_e \Sigma_e \frac{A(q)e^{iqx} - A(-q)e^{-iqx}}{2D_1(q)D_2(q)}, \quad (17)$$

$$A(q) = (\beta^2 - d^2 q k_x) \sin(d(k_x + q)) + \beta d(k_x + q) \cos(d(k_x + q)), \quad (18)$$

$$D_1(q) = \beta \cos(dq) - dq \sin(dq), \quad D_2(q) = \beta \sin(dq) + dq \cos(dq), \quad (19)$$

where the current  $J_1(x)$  and the conductance  $\Sigma_e$  are given by Eq. (7). The plasmon current  $J(x)$  has evident resonance when the projection of the wavevector  $k_x$  of the incident light equals to the plasmon wavevector  $\Re q = k_x$ . It could happen in so-called Kretschmann geometry where the upper half space ( $z < 0$ ) permittivity  $\varepsilon_e$  is larger than the lower half-space permittivity  $\varepsilon_e > \varepsilon_d$  and also the angle of incidence  $\theta$  (see Fig. 1) is large enough so that  $k_x = \sin \theta \frac{\omega}{c} \sqrt{\varepsilon_e} = q$ . Then EM wave, which is incident from above, excites plasmon in a metal film.

Yet, we consider in this paper the EM that propagating in all the space. The resonance can happen for any  $k_x$ , i.e., for any incident wave when the discriminant  $D_1(q)D_2(q)$  in Eq. (17) vanishes. Consider the plasmon electric field of the first order resonance where  $m = 1$ . To simplify the consideration we assume that  $k_x = 0$  that is the light is incident normal to the PZ plane, i.e.  $z = 0$  plane. Then the dimensionless plasmon electric field  $|E(x)|^2 = |J(x)/(E_e 2h\sigma_m)|^2$  reaches its maximum at the centers of PZ strips. Substituting the plasmon current from Eq. (17) we obtain

$$|E(0)|^2 = \left| 1 - \frac{\beta}{D_1(q)} \right|^2 \quad (20)$$

The real part of the discriminant  $D_1$  vanishes exactly at the resonance when  $D_1(q = q_r) = 0$ ,  $q_r = q_{r1} + iq_{r2}$ . Expanding  $D_1(q_r)$  in series of  $q_{r2}$  we obtain the maximum resonance field

$$|E_{max}|^2 \simeq \frac{\beta^4}{d^2 q_{r2}^2 (\beta^2 + \beta + d^2 q_{r1}^2)^2 \sin(dq_{r1})^2} \quad (21)$$

The wavelength  $\lambda_p$  of the plasmon is on the order of the width  $2d$  of a strip, that is  $dq_{r1} \sim d/\lambda \sim 1$  in the resonance. On the other hand the parameter  $\beta$  given by Eq. (16) is proportional to  $\beta \sim 1/\sqrt{g/h}$  and it could be rather large for a thin metal film, where the thickness  $h$  is much smaller than the inter strip gap  $g \gg h$ . The dispersion equation  $D_1(q_{r1}) = 0$  is expanded in reciprocal powers of  $\beta$ , which gives

$$q_{r1} = \frac{\pi}{2d} (1 - \beta^{-1} + \beta^{-2}), \quad (22)$$

and the maximum field estimates as  $E_{max} \sim 1/(dq_{r2})^2$ . Substituting here imaginary part  $q_{r2}$  from Eq. (9) we obtain the following

$$|E_{max}|^2 \simeq \frac{4h^2 (\varepsilon_m'^2 - \varepsilon_d^2)^2 (\varepsilon_m''^2 - \varepsilon_e^2)^2}{\varepsilon_m''^2 d^2 (\varepsilon_d + \varepsilon_e)^2 (\varepsilon_m'^2 - \varepsilon_d \varepsilon_e)^2} \sim \frac{\varepsilon_m'^4}{\varepsilon_m''^2} \left( \frac{2h}{d(\varepsilon_d + \varepsilon_e)} \right)^2, \quad (23)$$

where the last esteem holds for red and infrared spectral range where the metal permittivity is typically large  $|\varepsilon_m| \gg 1$ . For example, the silver permittivity estimates as  $\varepsilon_{Ag} \simeq -30 + i0.38$  [31], therefore, the factor  $\varepsilon_m'^4/\varepsilon_m''^2 > 10^6$  is huge in Eq. (23). Note the electric field enhancement in metal nanoparticles is typically restricted by the radiation loss that rapidly increases with the particle size. Yet, a radiation loss is almost zero for the fully periodic PZ, the radiation loss happens due to the manufacturing imperfections only. We speculate that the result for the huge resonance field enhancement obtained above in quasistatic approximation holds up to diffraction limit. The electric field  $E_{max}$  in PZ is shown in Fig. 4. It can be observed that local electric field can be enhanced in PZ for any part of visible spectrum.



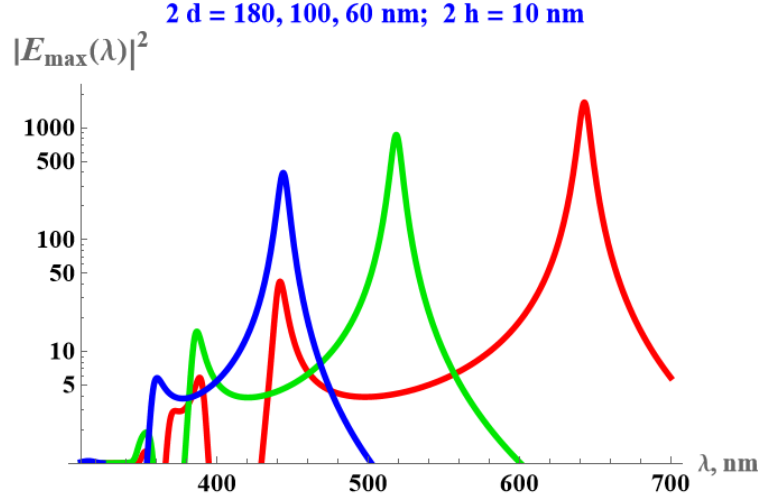


Рис. 4: Electric field enhancement in PZ illuminate by light impinged normal to PZ plane; metal strip thickness  $2h = 10 \text{ nm}$ , interstrip gap  $2g = 40 \text{ nm}$ , strip width  $2d = 180, 100, 60 \text{ nm}$  (red, green, blue); amplitude of incident light  $E_0 = 1$ .

### 3. Reflectance and Color of Plasmon Zebra

It is save to suppose that even thin PZ effectively reflects the light, which wavelength correspond to the plasmon resonance. That is the color of PZ corresponds to the resonance wavelength. The reflectance of PZ is defined by its effective surface conductance  $\Sigma^{(ef)}$ , which is anisotropic in this case  $\Sigma^{(ef)} = \{\Sigma_x, \Sigma_y\}$ . The effective surface conductance is obtained by average of the electric current over the PZ plane  $\Sigma_x = p \int_{-d}^d J(x)/E(x) dx/2d$ ,  $\Sigma_y = p\Sigma_e$ , where  $p = d/(d + g)$  is the fraction of  $z = 0$  plane, which is covered by the metal strips. Integration of the electric current given by Eq. (17) gives the effective  $x$  conductance

$$\Sigma_x = p\Sigma_e \left[ \frac{\sin dk_x}{dk_x} + \frac{\sin dq (dk_x \sin dk_x - \beta \cos dk_x)}{dq D_1(q)} \right], \quad (24)$$

which resonates as it is shown in Fig. 5. The PZ permittivity resonate when the discriminant  $D_1(q) \simeq 0$ , i.e., for the odd plasmon resonances. In this sense the even resonances ( $D_2(q) = 0$ ) are so-called dark resonances [32].

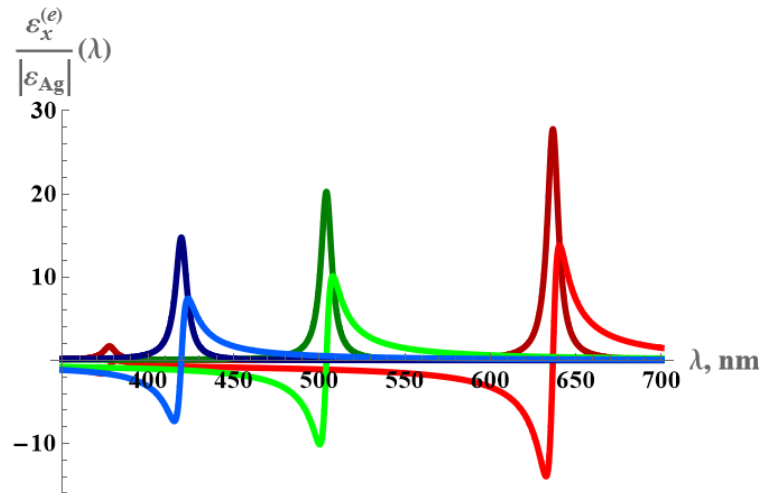


Рис. 5: Ratio of real and imaginary parts of the effective zebra permittivity  $\varepsilon_x^{(e)} = \varepsilon_x'^{(e)} + i\varepsilon_x''^{(e)}$  to silver permittivity  $\varepsilon_{Ag}$ ; red and dark red strip width  $2d = 180 \text{ nm}$ , green and dark green  $2d = 100 \text{ nm}$ , blue and dark blue  $2d = 60 \text{ nm}$ ; metal strip thickness  $2h = 10 \text{ nm}$ , interstrip gap  $2g = 40 \text{ nm}$ .

We consider here, for simplicity, the case when the natural light is incident normal to the plane of PZ. The reflectance  $r$  of a thin metal film, which thickness is less than the skin depth, is defined by the surface conductance (see, e.g., [33, 34]) or effective surface permittivity  $\varepsilon^{(ef)} = -i2\pi\Sigma^e/h\omega$ . The effective surface permittivity is anisotropic  $\varepsilon^e = \{\varepsilon_x^e, \varepsilon_y^e\}$  for a PZ. Recall we consider PZ composed of parallel metal strips, which are aligned with  $y$  axis as it is shown in Fig. 1. The reflectance  $r$  is obtained by matching the incident EM wave in the upper half space, where the electric field  $E_e \sim E_0 [\exp(ik_e z) + r \exp(-ik_e z)]$  and the transmitted wave in the lower half space  $E_d \sim E_0 t \exp(ik_d z)$ , where  $t$  is the transmittance of PZ; wavevectors of the incident and transmittance waves equal to  $k_e = n_e k$  and  $k_d = n_d k$ , where  $n_e = \sqrt{\varepsilon_e}$  and  $n_d = \sqrt{\varepsilon_d}$ . That is we extrapolate EM field to PZ surface, i.e., to the plane  $z = -h$  (see Fig. 1). By the same token we extrapolate farfield in the lower half space  $E_d \sim E_0 t \exp(ik_d z)$  to the lower PZ boundary  $z = h$ . Among all EM spatial harmonics excited in PZ we take into account those that are due to the plasmon resonance. The field of SP is added to the fields  $E_e$  and  $E_d$ . This approach is similar to GOL method [21, 22] and can be called one mode GOL approximation.

The vector of the electric field  $E_0 = \{E_{0x}, E_{0y}, E_{0z}\}$  is determined by the polarization of the incident light. To find the reflectance  $r$  and transmittance  $t$  of a thin film we equate the electric field of the incident light, electric field in the film, and the field in the transmitted light in the middle of the film at the plane  $z = 0$ . The reflectance  $r$  is defined by the polarization of the incident light when the direction of EM wave is normal to the film. Then solution of the Maxwell equations and matching the magnetic fields at the interfaces of the film gives

$$r_{x,y} = \frac{n_e - n_d + W_{x,y} (\varepsilon_{x,y}^e - n_d n_e)}{n_d + n_e - W_{x,y} (\varepsilon_{x,y}^e + n_d n_e)}, \quad (25)$$

where

$$W_{x,y} = i \tan(2hk \sqrt{\varepsilon_{x,y}^e}) / \sqrt{\varepsilon_{x,y}^e}, \quad (26)$$

where the effective surface permittivity  $\varepsilon_{x,y}^e = i \frac{2\pi}{\omega h} \{\Sigma_x, \Sigma_y\}$  is obtained from Eq. (24), wavevector  $k = \omega/c$ . The reflectance of PZ is shown in Fig. 6. The maxima of the reflectance correspond to the maxima of the effective PZ permittivity (Fig. 5). Note the reflectance of PZ with the strip width of 200 nm has two maxima

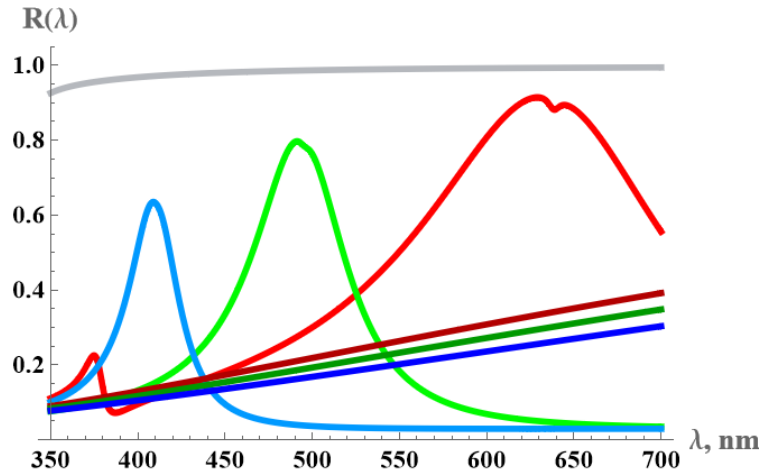


Рис. 6: Reflectance of silver zebra with thickness  $2h = 10 \text{ nm}$ , interstrip gap  $2g = 40 \text{ nm}$ , that is deposited on glass substrate  $\varepsilon_e = 1$ ,  $\varepsilon_d = 2$ ; red and dark-red are  $x$  and  $y$  reflectances of PZ with strip width  $2d = 180 \text{ nm}$ , green and dark-green are  $x$  and  $y$  reflectances with width  $2d = 100 \text{ nm}$ , blue and dark-blue are  $x$  and  $y$  reflectances with width  $2d = 60 \text{ nm}$ ; gray line is reflectance of bulk silver plate.

that correspond to the first  $m = 1$ ,  $\lambda \simeq 650 \text{ nm}$  and third  $m = 3$ ,  $\lambda \simeq 370 \text{ nm}$  plasmon resonances.

The reflectance of PZ is anisotropic as well as surface conductivity as it is shown in Figs. 5 and 6. We consider the color of PZ when it is illuminated by the natural light, which is composed from EM of various polarization. We assume that photons of different polarization are incoherent. Therefore, the total reflection coefficient being averaged over the polarization equals to  $R(\lambda) = (|r_x(\lambda)|^2 + |r_y(\lambda)|^2) / 2$ . We are interested in the PZ color, which is determined by behavior the reflection  $R(\lambda)$  as function of the wavelength. The coloring is well established problem discussed, for example, in the recent papers [19, 35–38].



Yet, everybody sees her/his own a color shadow. For qualitative consideration we adopt the simplest possible approach. All visible spectrum is divided on three parts: red  $\lambda_{G1} < \lambda < \lambda_R$ , green  $\lambda_{G2} < \lambda < \lambda_{G1}$ , and blue  $\lambda_B < \lambda < \lambda_{G2}$ , where  $\lambda_R = 680 \text{ nm}$ ,  $\lambda_{G1} = 590 \text{ nm}$ ,  $\lambda_{G2} = 480 \text{ nm}$ , and  $\lambda_B = 390 \text{ nm}$ . We calculate the  $\{\mathbf{R}, \mathbf{G}, \mathbf{B}\}$  color as  $\mathbf{R} = W_1 \int_{\lambda_{G1}}^{\lambda_R} R(\lambda) d\lambda$ ,  $\mathbf{G} = W_2 \int_{\lambda_{G2}}^{\lambda_{G1}} R(\lambda) d\lambda$ , and  $\mathbf{B} = W_3 \int_{\lambda_B}^{\lambda_{G1}} R(\lambda) d\lambda$ , where the color weights  $W_1, W_2$ , and  $W_3$ , are chosen to mimic real color. We consider here the silver PZ, then the color weights  $\{W_1, W_2, W_3\} = \{0.80, 0.83, 0.85\}$  are chosen in such a way that  $\{\mathbf{R}, \mathbf{G}, \mathbf{B}\}$  color of the bulk silver, calculated from the reflectance in Fig. 6 has the silver color indeed as it is shown in Fig. 7. The silver PZ



Рис. 7: Color of silver zebra with thickness  $2h = 10 \text{ nm}$ , interstrip gap  $2g = 40 \text{ nm}$ , that is deposited on glass substrate  $\varepsilon_e = 1$ ,  $\varepsilon_d = 2$ ; 1, 2, 3 – color of PZ with strip width  $2d = 200 \text{ nm}$ ,  $2d = 120 \text{ nm}$ , and  $2d = 80 \text{ nm}$ , 4–bulk silver plate.

has various colors in Fig. 7 that depends of the parameters that can be easily changed in the process of the manufacturing. Yet, thus obtained plasmon colors are not pure enough since the PZ reflectance has rather wide maxima at resonance wavelength.

#### 4. Conclusions

The simple analytical theory is presented for the plasmon resonances in the system of thin periodical metal strips and patches. In particular plasmon resonance are analytically calculated for the system of parallel metal strips we called plasmon zebra (PZ). The frequencies of the plasmon resonances correspond to the maxima of PZ reflectance. Simple PZ with all the strips of the same width gives RGB colors. Combination the metal strips of different parameters could produce the plasmon painting of arbitrary color.

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## ЗЕБРА-ПЛАЗМОННЫЕ РЕЗОНАНСЫ И НАНОКРАСКА

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### Аннотация

Мы исследуем металл-диэлектрические метаповерхности, состоящие из периодических металлических нанополосок, нанесенных на диэлектрическую подложку. Метаповерхность можно назвать плазмонной зеброй (ПЗ). Метаповерхность работает как набор открытых плазмонных резонаторов. Разработана теория плазмона, возбуждаемого в открытых резонаторах, соединенных между собой. Предсказано большое локальное электромагнитное поле для оптических частот, соответствующих возбуждению плазмона. Отражательная способность ПЗ значительно усиливается на частоте плазмонного резонанса, и ПЗ приписывают цвет, соответствующий резонансной частоте. Мы предлагаем ПЗ как простейшую, но легко настраиваемую плазмонную нанокраску.

**Ключевые слова:** зебра-плазмонный резонанс, усиление электромагнитного поля, нанокраска

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