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Proposal for a self-excited electrically driven surface plasmon polariton generator

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We propose a generator of surface plasmon polaritons (SPPs) which, unlike spasers or plasmon lasers, does not require stimulated emission in the system. Its principle of operation is based on a positive feedback which an ensemble of classical oscillating dipoles experiences from a reflective surface located in its near field. The generator design includes a nanocavity between two metal surfaces which contains metal nanoparticles in its interior. The whole structure is placed on a prism surface that allows one to detect the generated SPPs in the Kretschmann configuration. The generation process is driven by a moderate DC voltage applied between the metal covers of the cavity. Both the generation criterion and the steady-state operation of the generator are investigated. Published by AIP Publishing. [http://dx.doi.org/10.1063/1.4974454]

The progress of nanophotonics develops towards smaller photonic elements and higher packing density of photonic circuitries. Metal nanostructures, which support strongly localized electromagnetic excitations known as surface plasmon polaritons (SPPs), provide unique opportunities in this direction.1 Utilization of SPPs allows one to overcome the diffraction limit in photonics2 and can be implemented for extreme light energy concentration,3 ultra-sensitive sensing,4 high-resolution microscopy,5 ultra-fast computations, and a plenty of other applications.

This strategy faces, however, challenges because of high dissipation which is inevitable for all metallic structures. To compensate optical losses, it was suggested to introduce a gain medium into a metal nanostructure6—the idea which stems from the conventional approach in laser physics.7 The same principle forms the basis of Surface Plasmon Amplification by Stimulated Emission of Radiation (SPASER)8,9 or plasmon laser.10 In such a case, the stimulated emission in the gain medium incorporated into the metal nanostructure leads to generation of SPP quanta.

This mechanism is, however, not the only possible way of generating SPPs. A classical dipole oscillating above a conductive surface excites SPPs at the surface - the result which dates back to Sommerfeld’s paper from 1909.11,12 The electroductive surface excites SPPs at the surface - the result which stems from the conventional approach in laser physics.7 A classical dipole oscillating above a metal nanostructure leads to generation of SPP quanta. A classical dipole oscillating above a metal nanostructure leads to generation of SPP quanta.

The design of such a generator is shown in Fig. 1. A rectangular cavity of cross-section $L_x \times L_y$ in the $xy$ plane is formed in the subwavelength gap of thickness $d$ between two metals with the dielectric function $\epsilon_m$. From the other sides, the cavity is enclosed by a dielectric material to ensure an electrical isolation between the metal plates. To provide the possibility of detection of generated SPPs in the Kretschmann configuration, the whole structure is placed onto a prism with the dielectric function $\epsilon_p$, so that the substrate metal thickness, $h$, is of the order of the wavelength. We assume that the cavity interior is filled with the material of the dielectric function $\epsilon_b$ and contains identical spherical metal nanoparticles (NPs) randomly distributed with the volume fraction $f$.

Suppose now that at the moment of time $t=0$, one applies a DC voltage $\mathcal{E}$ between the metal covers of the cavity. This leads to the charging of the nanocapacitor formed by the cavity between the metal plates, so that the electric field applied to the cavity varies as $E_0(t) = e_0[1 - \exp(-t/\tau)]$ with $\tau = RC$, $R$ being the effective resistance of the electric circuit, and $C$ being the capacitance of the nanocapacitor. This field polarizes nanoparticles in the cavity and their polarization, $P(t)$, can be described in the framework of the harmonic oscillator model as follows:13
The oscillating part of the polarization can be represented in the form $\mathbf{P}_1(t) = \mathbf{P}_1(t) \exp(-i\omega_0 t)$, where the amplitude $\mathbf{P}_1(t)$ varies on the time scale $\sim \Gamma^{-1}$ and satisfies the equation

$$
\frac{i}{2\omega_0} \frac{d^2 \mathbf{P}_1}{dt^2} + \frac{d\mathbf{P}_1}{dt} + \frac{\Gamma}{2} \mathbf{P}_1 \approx i\beta \tilde{\mathbf{E}}_R
$$

with $\beta = a/(2\omega_0)$ and the initial conditions $\mathbf{P}_1(0) = 0$ and $d\mathbf{P}_1/dt(0) = -d\mathbf{P}_0/dt(0)$, where we have neglected the terms of the order of $\Gamma/\omega_0$. The slowly varying amplitude of the reflected field, $\tilde{\mathbf{E}}_R$, is in turn expressed in terms of $\mathbf{P}_1(t)$ through the approximate equation

$$
\tilde{\mathbf{E}}_R(r, t) \approx \int \tilde{\mathbf{F}}^R(r, r'; \omega_0) \mathbf{P}_1(r', t) dr',
$$

where $\tilde{\mathbf{F}}^R(r, r'; \omega)$ is the reflected contribution to the field susceptibility tensor and the radius vectors $r$ and $r'$ specify points in the cavity. The quantity $\tilde{\mathbf{F}}^R(r, r'; \omega)$ relates the reflected electric field at the point $r$ generated by a classical dipole, oscillating at frequency $\omega$, with the dipole moment itself, located at $r'$. Its explicit form is known for a dipole between two parallel reflective surfaces.

We assume that the dimensions of the cavity along the $x$ and $y$ axes are much larger than its height, i.e., $L_x, L_y \gg d$. In such a case, to a good approximation, the dipole emission in the cavity can be regarded as the one in an infinitely extended cavity.

Equation (2) should be considered jointly with Eq. (3) in order to analyze the stability of the cavity field. To investigate the field in the cavity, we expand it in the Fourier series over the intervals $-L_x/2 < x < L_x/2$ and $-L_y/2 < y < L_y/2$ as follows:

$$
\tilde{\mathbf{E}}_R(r, t) = \sum_{m,n=\infty}^{\infty} \mathbf{e}_m^n(z, t) \exp(i \frac{2\pi m}{L_x} x) \exp(i \frac{2\pi n}{L_y} y).
$$

Similar expansions can be written for the quantities $\mathbf{e}_0^0(r)$ and $\mathbf{P}_1(r, t)$ with the coefficients $\mathbf{e}_m^n(z, t)$ and $\mathbf{p}_m^n(z, t)$, respectively. Substituting these expansions into Eqs. (2) and (3) and taking into account the inequalities $L_x, L_y \gg d$, one obtains the equations for the Fourier coefficients

$$
\frac{i}{2\omega_0} \frac{d^2 \mathbf{p}_m^n}{dt^2} + \frac{d\mathbf{p}_m^n}{dt} + \frac{\Gamma}{2} \mathbf{p}_m^n \approx i\beta \tilde{\mathbf{e}}_m^n
$$

and

$$
\mathbf{e}_m^n(z, t) \approx \int_{-d/2}^{d/2} \tilde{\mathbf{F}}^R(z, z'; \omega_0, \kappa_{mn}) \mathbf{p}_m^n(z', t) dz',
$$

where $\tilde{\mathbf{F}}^R(z, z'; \omega, \kappa)$ is the Fourier transform of the field susceptibility tensor (supplementary material) and

$$
\kappa_{mn} = 2\pi \sqrt{(m/L_x)^2 + (n/L_y)^2}
$$

is the absolute value of the wave vector of the field mode $(m, n)$. 

FIG. 1. (a) Design of the SPP generator (side view). The leakage radiation of the generated SPPs can be observed in the Kretschmann configuration. (b) Top view of the cavity. The red arrows show the propagation directions of the generated modes $(m, n)$. 

\[\text{Equation}(1)\] 

\[\text{Equation}(2)\] 

\[\text{Equation}(3)\] 

\[\text{Equation}(4)\] 

\[\text{Equation}(5)\] 

\[\text{Equation}(6)\] 

\[\text{Equation}(7)\]
Taking the Laplace transform of Eqs. (5) and (6) in time and performing necessary integrations and summations, one comes to the vector equation

\[ \mathbf{M} - \sigma(s) \mathbf{I} \mathbf{A}(s) = \mathbf{B}, \]  

(8)

where \( \sigma(s) = [(i/2 \omega_0)^2 + s + \Gamma/2]/i\beta \), \( \mathbf{A}(s) = (\mathbf{E}^{\perp}_n(s), \mathbf{E}^{\parallel}_n(s), \mathbf{E}^{\perp}_z(s), \mathbf{E}^{\parallel}_z(s))^T \), \( \mathbf{B} = (\mathbf{P}^{\perp}_n, \mathbf{P}^{\parallel}_n, \mathbf{P}^{\perp}_z, \mathbf{P}^{\parallel}_z)^T \) with

\[ \mathbf{E}^{\pm}_j(s) = \sum_k \int_{-d/2}^{d/2} \mathcal{H}^{\pm}_{jk}(z) e^{ikx/\varepsilon_0} e^{ikz} d z, \]  

(9)

\[ \mathcal{P}^{\pm}_j = \frac{1}{\varepsilon_0^2} \int_{-d/2}^{d/2} \int_{-d/2}^{d/2} \mathcal{H}^{\pm}_{jk}(z) \mathcal{F}^{R}_{kl}(z, z') e^{ikl(z')} d z' d z. \]  

(10)

Here \( \mathbf{M} \) is the unit 4 \times 4 matrix, the explicit forms of the matrix \( \mathbf{M} \) and the functions \( \mathcal{H}^{\pm}_{jk}(z) \) are given in Ref. 14, the latter ones take into account the different reflection coefficients for the upper and lower Ag films, the tilde denotes the Laplace transform, and we have omitted everywhere the subscripts \( mn \) for the sake of brevity (supplementary material).

As it follows from Eq. (8), the time evolution of the cavity field, which enters the vector \( \mathbf{A}(s) \) by means of its Laplace transform \( \mathbf{E}(s) \), is determined by the poles of \( \mathbf{A}(s) \) or, equivalently, by the zeros \( s_j \) of the determinant of the matrix \( \mathbf{M} - \sigma(s) \mathbf{I} \). On the other hand, at these zeros, the quantity \( \sigma(s) \) gives the eigenvalues \( \lambda_j \) \((j = 1, \ldots, 4) \) of the matrix \( \mathbf{M} \) by definition. Taking into account the inequalities \( \Gamma, \beta < \omega_0 \), the poles of \( \mathbf{A}(s) \) can be expressed as \( s_j = -\Gamma/2 + i\beta \lambda_j \). Consequently, if the matrix \( \mathbf{M} \) has at least one eigenvalue with a negative imaginary part, \( \text{Im}(\lambda_j) < 0 \), and

\[ -\text{Im}(\lambda_j) > \frac{\Gamma}{2\beta} = \frac{4\pi}{3} \frac{\Gamma}{\epsilon_0 \omega_0}, \]  

(11)

then the cavity field will increase with time as \( \exp(gt) \) with the generation rate \( g = -\Gamma/2 - \beta \text{Im}(\lambda_j) \). In such a case, the imaginary part of the corresponding pole will determine the frequency pulling effect for the frequency of generation, \( \omega_g = \omega_0 - \beta \text{Re}(\lambda_j) \), which is known for lasers as well.\(^7\)

Equation (2) describes a linear regime of the NPs excitation when the cavity field is not too strong. If the generated field is very intensive, it should be corrected to take into account the nonlinear terms. The nonlinear optical response of metal NPs is manifested, in particular, as the saturation of absorption of metal-nanoparticle composite.\(^20\)\(^21\) It becomes essential when the exciting field intensity, \( I \), is comparable with the saturation intensity, \( I_s \). Then the nanocomposite absorption coefficient can be well described as \( \alpha(l) = \alpha_0/(1 + I/I_s) \) with \( \alpha_0 \) being the absorption coefficient in the linear regime. This effect can be introduced in Eq. (2) by means of multiplying the coefficient \( \beta \) by the factor \( (1 + I/I_s)^{-1} \) with \( I_R \) being the intensity of the reflected field. Then, considering the saturation regime where the cavity field amplitude \( \mathbf{E}_R \) varies very slowly in time and performing the Laplace transform, one comes to the equation

\[ [(1 + I_R/I_s)^{-1} \mathbf{M} - \sigma(s) \mathbf{I}] \tilde{\mathbf{A}}(s) = \tilde{\mathbf{B}}, \]  

(12)

instead of Eq. (8). Here the vector \( \tilde{\mathbf{B}} \) is determined by the value of the NPs polarization at some moment of time \( t = t_1 \) which corresponds to the saturation regime, and we have neglected the time dependence of \( I_R \) in the equation coefficient for the sake of simplicity. The condition of the steady-state operation, \( \text{Re}(\lambda_j) = 0 \), gives the intensity of the generated field in the steady-state regime, \( I_{ss} \)

\[ \frac{I_{ss}}{I_s} = -\frac{2\beta}{\Gamma} \text{Im}(\lambda_j) - 1 \]  

(13)

with \( \lambda_j \) being the eigenvalue of the matrix \( \mathbf{M} \) which corresponds to generation. The generation frequency undergoes saturation as well

\[ \omega_g = \omega_0 - \beta \text{Re}(\lambda_j) (1 + I_{ss}/I_s)^{-1}. \]  

(14)

Let us note that in the steady-state regime, the current in the electric circuit of the generator can be described in terms of the effective cavity capacitance \( C_{eff}(t) \) which oscillates with the frequency \( \omega_{ss} \). The current oscillates as well and the average power consumed from the battery for a period is always positive (supplementary material).

We investigate the criterion of generation for a cavity between two silver films. One of them is deposited onto a prism surface (\( \epsilon_p = 1.45^2 \)) and has the thickness \( h = 100 \) nm, whereas the other is much thicker than the wavelength of operation and is assumed to be semi-infinite in the \( z \)-direction. The cavity interior is filled with glass (\( \epsilon_g = 1.45^2 \)) and contains Ag NPs with the volume fraction \( f \). The dielectric function of the Ag films is taken in the Drude model, \( \epsilon_m(\omega) = \epsilon_{\infty} - \omega_p/|\omega(\omega + i\Gamma)| \) with \( \epsilon_{\infty} = 5, \omega_p = 14.0 \times 10^{13} \) s\(^{-1} \) and \( \gamma = 0.032 \times 10^{15} \) s\(^{-1} \). For silver NPs the relaxation constant can be written as \( \Gamma = \gamma + b_{np} R \), where the Fermi velocity \( v_F = 1.4 \times 10^4 \) cm/s, \( R \) is the NP radius and \( b = 1.22 \). For the given parameters and \( f = 0.01 \), the LSPP wavelength is found as \( \lambda_{ls} = 2\pi c/\omega_0 = 2\pi c (\epsilon_{\infty} + 2\epsilon_g)^{1/2}/|\omega_p(1 - f)^{1/2}| \approx 410 \) nm.

Figure 2 shows the contour plots in the parameter plane \( \kappa - d \) which according to Eq. (11) determines the threshold for generation. The other eigenvalues of the matrix \( \mathbf{M} \), \( \lambda_{3,4} = 0 \), correspond to the polarization oscillations decaying with time as \( \exp(-\Gamma t/2) \). The threshold is increased with the increase in \( \Gamma \) (or, equivalently, with the decrease in the NP radius \( R \)) and with the decrease in \( f \). Accordingly, to ensure a single-mode operation, one can decrease either the size of NPs or their volume fraction. Additional room for parameter optimization can be provided by shrinking the cavity sizes \( L_x \) and \( L_y \), which leads to a more sparse distribution of the cavity modes.

The values of the modes wave numbers, Eq. (7), calculated for a cavity with \( L_x = 1300 \) nm and \( L_y = 650 \) nm are shown in Fig. 2(b) by arrows. One can see that for the cavity thickness \( d \approx 50 \) nm, the generation condition can be realized only for the mode \((2,2)\). However, due to the degeneracy with respect to the signs of \( m \) and \( n \), the same condition is also fulfilled for the modes \((-2,2), (2,-2), \) and \((-2,-2)\). These four modes propagate along the different
directions specified by the azimuthal angles $\phi = \pm 63.4^\circ$, $\pm 116.6^\circ$ in the $xy$ plane [see Fig. 1(b)].

The initial intensity of the generated wave depends on the constant voltage, $E_0$, applied to the cavity. The calculation for $d = 50 \text{ nm}$ and $k_{22} = 0.0216 \text{ nm}^{-1}$ gives for its mean amplitude at $t = 0$ $|e^p(0)| \approx 0.30 \times |e^0(0)|$, that corresponds to the initial wave intensity $I(0) \approx 1.1 \times 10^8 \times |e^0(0)|^2$ in Gaussian units. If, for example, $E_0 = 1 \text{ mV}$, then $I(0) \approx 5 \text{ W/cm}^2$.

For the parameters given above, one finds the generation rate $g \approx 8.3 \times 10^{13} \text{ s}^{-1}$. When the saturation comes into play, the intensity of the generated field in the steady-state regime can be obtained from Eq. (13) as $I_s \approx 0.33 I_\nu$. Taking into account the frequency pulling effect, Eq. (14), one finds the wavelength of generation $\lambda_g \approx 407 \text{ nm}$. From here one calculates the polar angle $\theta$ relative to the $z$-axis, at which the generated wave can be detected in the Kretschmann configuration, as $\theta = \arcsin{\{k_{22}/[2\pi/\lambda_g (\varepsilon_p)^{1/2}])\} = 74.8^\circ$.

For estimates we take the results of the self-consistent calculations of the saturable absorption in silica glass doped with Ag nanoparticles, which give $I_s \approx 100 \text{ MW/cm}^2$ at $\lambda = 430 \text{ nm}$. Despite a very high intensity of the generated wave ($I_{st} \approx 33 \text{ MW/cm}^2$), the corresponding consumed power is rather low: $P = I_{st} (L_\nu / \cos \phi) d \approx 26 \text{ mW}$.

In conclusion, we have proposed and analyzed a self-excited generator of gap surface plasmon polaritons which is driven by a DC applied voltage. Its scheme is based on a plasmonic nanocavity doped with metal nanoparticles whose polarization undergoes a positive feedback from the reflective cavity walls. In contrast to spasers or plasmon lasers, such a generator does not exploit stimulated emission and does not require therefore powerful pumping, which is necessary to create a population inversion in a system with fast relaxation. The generation frequency is dictated by the LSPP frequency of the NPs and can be tuned by changing their metal composition, size, and shape. The advantage of this approach, among other things, is the possibility to trigger the self-excitation (self-oscillation) process by applying a moderate electric field, that is a significant gain for practical applications.

See supplementary material for triggering the self-oscillation, power consumption from the battery, field susceptibility tensor, and the matrix $M$.

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