Controlling Strong Electromagnetic Fields at Subwavelength Scales

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We investigate the optical response of two subwavelength grooves on a metallic screen, separated by a subwavelength distance. We show that the cavity, Fabry-Perot-like mode, already observed in one-dimensional periodic gratings and known for a single slit, splits into two resonances in our system: a symmetric mode with a small Q factor, and an antisymmetric one which leads to a much stronger field enhancement. This behavior results from the near-field coupling of the grooves. Moreover, the use of a second incident wave allows control of the localization of the photons in the groove of our choice, depending on the phase difference between the two incident waves. The system acts exactly as a *subwavelength optical switch* operated from far field.

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Surface enhancement Raman scattering (SERS) still remains in a large part a mystery, even though it is now accepted that the basic mechanism involves excitation of localized electromagnetic modes of irregular metallic surfaces [1,2]. Optical excitation of such modes can indeed lead to an important concentration of electromagnetic energy in volumes (cavities) much smaller than λ^3 where λ is the excitation wavelength, as is the case for SERS active surfaces. These specific regions of very strong electromagnetic field localization are called "active sites" or "hot spots." However, the debate on the origin of these hot spots remains open, as well as the hope of one day controlling this phenomenon. In the particular case of deep metallic gratings, the electromagnetic resonances are of two kinds: surface plasmons and cavity modes. In the latter the field is localized within the cavities, which act as waveguides [3]. The large interest raised by this fundamental physics is also increased by its wide potential application in biochips, sensors, nanoantennae, optoelectronics, or energy transport on nanostructured surfaces.

In this Letter, we consider a simple system, consisting only of two deep grooves on a plane gold surface, which allows one to *produce and control* such hot spot phenomena (Fig. 1). The excited modes appear, for this geometry, in the infrared region where the metal is a good reflector. Under this condition, we can use the modal method using surface impedance boundary conditions [4]. This method has already demonstrated its ability to give good quantitative agreement with the measured reflectivity of metallic gratings [5–7]. The case of one groove only was considered a long time ago [8], while the treatment of transmission for one [9] and two slits [10] is very recent. In contrast with [10], the distance between our two grooves is small with respect to the incident wavelength. Very recently, it was also shown [11] that sharp and deep resonances appear in the transmission response of gratings with more than one slit per period, or in gold dipole antennae [12]. Herein we analyze the physical origin of these new resonances for a two slit system. As we will see, this allows us to point out some very fundamental aspects of electromagnetic resonances on metallic surfaces. We consider a p-polarized incident plane wave (electric field in the plane of incidence) with a wave vector $k = 2\pi/\lambda$ impinging on the surface at an angle θ (Fig. 1). The knowledge of the magnetic field in the z direction completely solves the problem since $H_x = H_y = E_z = 0$, $E_x = (i/ck\varepsilon_0)\partial H_z/\partial y$, and $E_y = (-i/ck\varepsilon_0)\partial H_z/\partial x$. In region ($I$), the field is expressed as the sum of the incident wave and the reflected ones as:

$$H_z^{(I)}(x, y) = e^{ik(\sin\theta x - \cos\theta y)} + \int_{-\infty}^{+\infty} R(Q) e^{i(Qx+qy)} dQ,$$

where the distribution $R(Q)$ represents the amplitude of the reflected field at the wave vector (Q, q) with $q = \sqrt{k^2 - Q^2}$. In region (II) one has:

$$H_z^{(II)}(x, y) = A_1[e^{iky} + \alpha e^{-iky}]I_1(x) + A_2[e^{iky} + \alpha e^{-iky}]I_2(x),$$

where $I_1(x)$ and $I_2(x)$ equal 1 in the intervals $[(-w - d)/2, (w - d)/2]$ and $[(d - w)/2, (d + w)/2]$, respectively, and zero elsewhere. The term $\alpha = [(1 + Z)/(1 - Z)]\Phi^2$, where $\Phi = e^{-ikh}$, $Z = 1/\sqrt{\epsilon}$ is the surface imped-

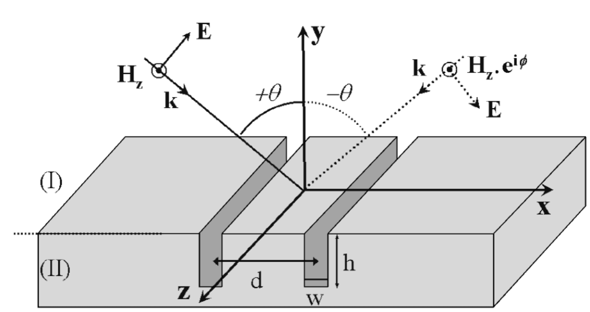

FIG. 1. Geometrical configuration and parameters. Region (I) and (II) correspond, respectively, to the region above and below the metallic surface. At the end of the Letter, we consider the case where a second incident plane wave is sent. ϕ is the phase difference between the two incident waves.

ance of the metal and ϵ its dielectric constant. The expression for $H_z^{(II)}$ assumes that the field is constant along x within each groove, which is a good approximation in the limit where $w \ll \lambda$ [5,6]. To illustrate our results numerically, we have fixed $w = 0.2\ \mu$m, $h = 1.5\ \mu$m, and $d = 0.5\ \mu$m throughout the Letter. The values of the complex dielectric constant $\epsilon(\lambda)$ are taken from [13].

The unknown variables are the distribution $R(Q)$ and the field amplitudes A_1 and A_2 in the first and second groove, respectively. A set of equations is obtained by applying the boundary conditions at the interface $y = 0$: $H_z^{(I)} = H_z^{(II)}$ at the mouth of each groove, and $\partial H_z^{(I)}/\partial y + ikZH_z^{(I)} = \partial H_z^{(II)}/\partial y + ikZH_z^{(II)}$ along the whole interface. After some elementary algebra [see [6] for the detailed procedure], the vector $\mathbf{A} = (A_1, A_2)$ is related to the excitation vector $\mathbf{V} = (V_1, V_2)$, by the matrix relation $\mathbf{A} = \mathbf{M}^{-1}\mathbf{V}$, where $\mathbf{M}$ is the 2×2 symmetric matrix which satisfies $m_{11} = m_{22}$ with:

$$m_{11} = (1 + \alpha) - \Gamma(1 + Z)(1 - \Phi^2) \int_{-\infty}^{+\infty} \frac{\sec^2(Qw/2)}{q + kZ} dQ$$

$$m_{12} = -\Gamma(1 + Z)(1 - \Phi^2) \int_{-\infty}^{+\infty} \frac{\sec^2(Qw/2)}{q + kZ} e^{iQd} dQ,$$

where $\Gamma = w/\lambda$. The coordinates of $\mathbf{V}$ are $V_1 = e^{-i\varphi}V_0$, $V_2 = e^{i\varphi}V_0$, with

$$V_0 = \frac{2\cos\theta}{\cos\theta + Z} \sec[k\sin(\theta)w/2],$$

where the angle $\varphi = kd\sin(\theta)/2$.

The matrix $\mathbf{M}$ has two eigenvalues $m_\pm = -i(1 - \Phi^2)e_\pm$ with eigenvectors $\mathbf{U}_\pm = (1, \pm 1)$, respectively, and:

$$e_\pm = \frac{1}{1 - Z}[\cot(kh) - iZ] - 2i\Gamma(1 + Z) \times \int_0^{+\infty} [1 \pm \cos(kdu)] \frac{\sec^2(kwu/2)}{\sqrt{1 - u^2} + Z} du.$$

We have made the variable change $Q = ku$ in the integrals. The solution of the problem is then:

$$A_{n=1,2} = \left[\frac{1}{e_+} + (-1)^n i \tan\varphi \frac{1}{e_-}\right] \frac{i\cos(\varphi)}{1 - \Phi^2} V_0$$

$$R(Q) = \frac{\cos\theta - Z}{\cos\theta + Z} \delta(Q - k\sin\theta) + \Gamma(1 + Z)(1 - \Phi^2) \times (e^{iQd/2}A_1 + e^{-iQd/2}A_2) \frac{\sec(Qw/2)}{q + kZ}. \quad (1)$$

From Eq. (1), one can see that the system has two electromagnetic resonances at $k = k_\pm$, which appear when $\mathrm{Re}\,(e_+) = 0$ and $\mathrm{Re}\,(e_-) = 0$, with line shapes governed, respectively, by $\mathrm{Im}\,(e_+)$ and $\mathrm{Im}\,(e_-)$. The fields in the cavities are always a linear combination of the two eigenvectors $\mathbf{A} \sim a_-\mathbf{U}_- + a_+\mathbf{U}_+$. However, when $k = k_\pm$, the vector $\mathbf{A}$ is almost collinear with $\mathbf{U}_\pm$, respectively, since the amplitudes in the two cavities are dominated by either the first or the second term in the square brackets in (1). We will thus call the resonance occurring at $k = k_-$ the $(-)$ antisymmetric mode and that occurring at $k = k_+$ the $(+)$ symmetric one. In contrast to the $(+)$ mode, which always exists, the $(-)$ one only develops for $\theta \neq 0$ (see Fig. 2) since it vanishes at normal incidence with $\tan\varphi = 0$. Its bandwidth is much thinner than that of the symmetric mode and its enhancement factor is much larger. The enhancement factor (EF), defined as $EF = |E_x/E_0|^2$, where E_0 is the incident electric field, reflects the amount of electromagnetic energy in the resonances. For convenience, we call EF_1 and EF_2 the enhancement factors calculated at the mouth of each cavity, i.e., at $x = \pm d/2$ and $y = 0$, where they are expressed as $EF_{n=1,2} = |A_{n=1,2}(1 - \alpha)|^2$. At the frequency of the $(-)$ mode, EF_2 is shown in Fig. 2(a). It increases with θ and its value can reach more than 10^3, whereas that of the symmetric mode stays at around 100. Another important point is that around the $(-)$ resonance, the fields in the two cavities are not identical. Figure 2(b) displays EF_1 and EF_2 close to $k = k_-$. At 1484 cm^{-1}, EF_2 reaches a maximum, whereas EF_1 is still low; at 1490 cm^{-1}, $EF_1 = EF_2$. Around this mode, the system develops a high sensitivity: with very little variation of wave number (here less than 1%), the field "jumps" from one cavity to the other.

In the following, we consider the metal to be a perfect reflector, i.e., $Z = 0$. This approximation induces only small quantitative modifications and allows an analytic study which helps to clarify the physics of the problem. However, our numerical results are obtained without this approximation, i.e., using the finite value of $\epsilon(\lambda)$. We first compare the two-groove system to the one where there is only one groove centered at $x = 0$. In this case, the amplitude of the field A_0 in the simple cavity is given by $A_0 = i[1 - \Phi^2]^{-1}V_0/e$, with:

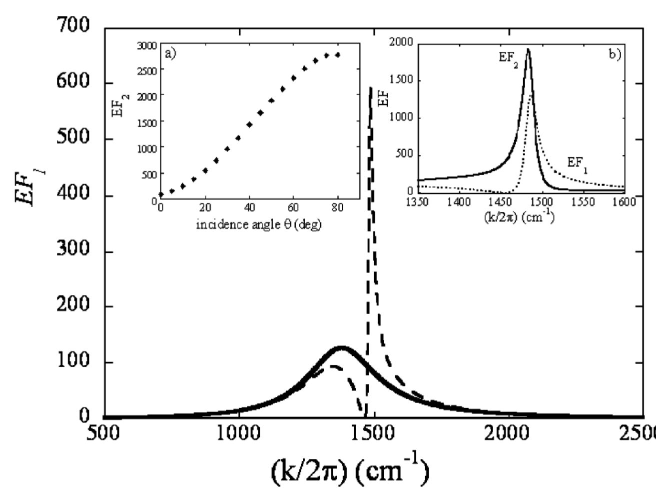

FIG. 2. Enhancement factor EF_1 of the cavity centered at $x = -d/2$, calculated as a function of the wave number, for $\theta = 0°$ (full line) and $\theta = 30°$ (dotted line). Inset (a) represents EF_2 (cavity centered at $x = +d/2$) as a function of the incidence angle θ, for $k = k_-$. Inset (b) gives the enhancements of both cavities, EF_1 and EF_2, respectively, calculated for 50°.

$$e = \cot(kh) - 2i\Gamma \int_0^{+\infty} \frac{\sec^2(kwu/2)}{\sqrt{1-u^2}} du.$$

The resonance of this cavity occurs at $k = k_0 = \omega_0/c$, for which $\mathrm{Re}\,(e) = 0$. Close to k_0, the field A_0 can be expanded around ω_0 as

$$A_0 \approx \frac{C_0}{\omega_0 - \omega - i\gamma_0/2},$$

with $C_0 \approx icV_0/2h$, and where we have taken advantage of the fact that at the resonance $k_0 h \approx \pi/2$ [6]. This equation is typical of a forced oscillator and, since the electric field inside the cavity is proportional to A_0, it indicates that the cavity behaves as a forced oscillating dipole with a radiation damping $\gamma_0 = 2w\omega_0^2/\pi c$, and an effective electromagnetic radius $r_0 = 2w/\pi$. The effective dipolar momentum, parallel to the interface, has a maximum at the mouth of the groove and decreases along the vertical walls. The maximum intensity is, at $\omega = \omega_0$, $|A_0|^2 \approx 4/(k_0 w)^2$, typically of order 100 for our geometrical parameters. We now expand, in the same manner, the values of $e_\pm$ around the same k_0 for the two-groove system. We easily get:

$$\begin{aligned} e_+ &\approx (\omega_+ - \omega - i\gamma_+/2)h/c, \\ e_- &\approx (\omega_- - \omega - i\gamma_-/2)h/c, \end{aligned} \tag{2}$$

with $\omega_\pm = \omega_0 \mp \Delta$, $\gamma_+ = 2\gamma_0$, and $\gamma_- = \gamma_0(k_0 d/2)^2$. The shift Δ, of the order of $\gamma_0 \ll \omega_0$, is

$$\Delta = \frac{\gamma_0}{\pi} \int_1^{+\infty} \frac{\cos(k_0 du)\sec^2(k_0 wu/2)}{\sqrt{u^2-1}} du. \tag{3}$$

Equations (2) and (3) confirm our numerical observation. They show that the width of the $(-)$ mode, driven by γ_-, is much lower than that of the $(+)$ mode, driven by γ_+, owing to the small factor $(k_0 d)^2$ (recall our subwavelength coupling hypothesis: $\lambda_0 \gg d$). A physical picture of these resonances can be given noticing that our results are analogous to those obtained by Lyuboshitz [14] for *two near-field coupled oscillating dipoles*. Our resonances thus arise from the near-field coupling of two identical grooves, individually resonating at ω_0. The symmetric $(+)$ mode corresponds to the in-phase oscillation of each cavity whereas the second one corresponds to an antiphase oscillation. The electric field distribution in the cavities is sketched in Fig. 3. As a consequence of this coupling, the $(+)$ mode has a dipolar character, with an effective dipolar moment close to twice that of a unique cavity and a large electromagnetic radius $r_+ = 2r_0$. In contrast, the $(-)$ mode has almost zero effective dipolar moment, with an electromagnetic radius reduced to $r_- = r_0(k_0 d/2)^2$. Its radiation pattern is essentially that of a quadrupole. This explains why this mode is weakly radiative and has an extremely narrow line shape, very different from the width of the in-phase mode. Searching for the location of the maximum of the field in each cavity around the $(-)$ mode, ones gets for $\theta \neq 0$:

$$\omega_{\max}^{n=1,2} \approx \omega_- - \frac{(-1)^n (\frac{k_0 d}{2})^3 \gamma_0}{4\sin\theta[1 + (\frac{2\Delta}{\gamma_0})^2]} + O[(k_0 d)^4]$$

$$|A_{\max}|^2 \approx \frac{16\sin^2\theta}{(k_0 w)^2 (k_0 d)^2},$$

where $|A_{\max}|^2$ is proportional to the intensity of the field in both cavities at $\omega_{\max}$. The two maxima $\omega_{\max}^1$ and $\omega_{\max}^2$ are separated by a very small frequency difference of order $(k_0 d)^3 \gamma_0$, which, together with the narrow line shape of the resonance, allows one to understand why the profile of the field strongly varies in this region. The magnitude of $|A_{\max}|^2$ requires some comment. Indeed, for a typical oscillator with damping γ, the intensity maximum of the oscillation scales as γ^{-2}, so that $|A_{\max}|^2$ should scale as $\gamma_-^{-2} \sim (k_0 d)^{-4}$ instead of $(k_0 d)^{-2}$. The field intensity of the $(+)$ mode scales, as expected, as γ_+^{-2} [Eq. (1)]. The reason for this is that the $(+)$ and $(-)$ modes *are not sensitive to the same parts of the incident electric field*. Since $d/\lambda \ll 1$, the latter can be express at the interface as $E_0(1 + ikx)$ at the scale of our two-groove system. The even term, corresponding to the mean value of the field, excites the $(+)$ mode, and the odd one, corresponding to the local variations of the field, excites the $(-)$ mode. This mode is thus sensitive to an "effective" field of intensity $\sim E_0^2 (k_0 d)^2$ at the mouth of the grooves, whereas the $(+)$ mode is excited by an effective field of intensity E_0^2. This is the origin of the loss of a factor $(k_0 d)^{-2}$ in the intensities of the $(-)$ mode.

We now take advantage of our understanding to control—from far field—the light localization in one or both cavities. We introduce a new free parameter by sending a

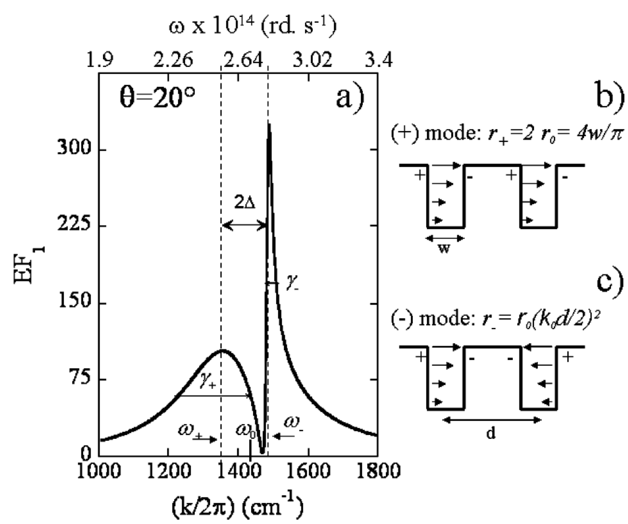

FIG. 3. Enhancement factor EF_1, calculated for $\theta = 20°$, showing the resonances $(+)$ and $(-)$ characterized by their eigenfrequencies $\omega_\pm$, located on both sides of the frequency resonance ω_0 of a unique cavity, and their bandwidth $\gamma_\pm$ (a). The right part schematically represents the in-phase coupling of the $(+)$ mode (b) and the antiphase coupling of the $(-)$ mode (c) and their corresponding equivalent dipole.

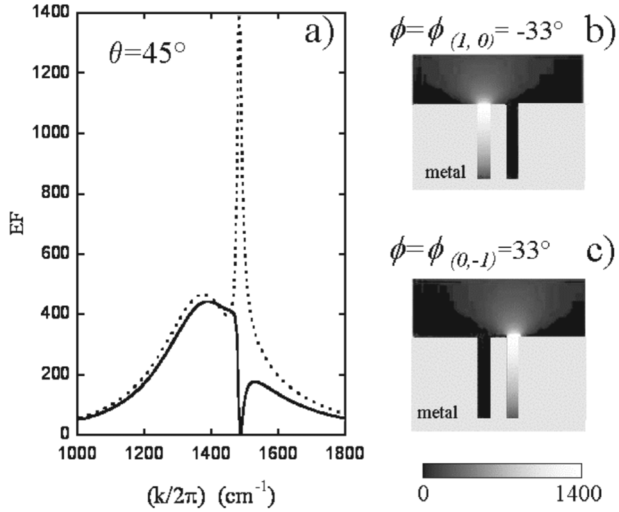

FIG. 4. Enhancements EF_1 and EF_2 represented as a function of the wave number, at $\theta = 45°$, for two incoming waves. For $\phi = \phi_{(1,0)}$ the full line is EF_2 and the dotted line is EF_1. For $\phi = \phi_{(0,-1)}$ it is the opposite (a). Related maps of the electric field amplitude E_x at 1490 cm^{-1} when the first cavity is lit (b) and when the second one is lit (c).

second incident plane wave, at the same frequency, with an incidence angle $-\theta$, and with a phase difference ϕ with respect to the first incident wave (Fig. 1). Changing ϕ, we can control the incident effective fields exciting, respectively, one mode or the other. Different states, which we will code as: (1, 1), (1, −1), (1, 0), and (0, −1) can thus be achieved. The first two, (1, 1) and (1, −1), correspond, respectively, to the case where only the pure (+) or the pure (−) resonances are excited. The cavities are then completely in phase or in antiphase. The other two correspond to cases where only *one of the cavities* is lit [cavity 1 for (1, 0), and cavity 2 for (0, −1)]. As ϕ is a parameter simple to modify, for instance, by changing the optical path, we can easily control the field localization.

With two incoming waves, the field becomes:

$$H_{\mathrm{inc}} = [e^{ik\sin\theta x} + e^{i(\phi - k\sin\theta x)}]e^{-ik\cos\theta y},$$

and the solution for each cavity can be written as:

$$A_{n=1,2} \sim \frac{\cos(\phi/2)}{e_+} + (-1)^n \tan\varphi \frac{\sin(\phi/2)}{e_-}, \qquad (4)$$

where we omit some unimportant prefactors common to both cavities. From these equations, it is easy to see that for $\phi = \phi_{(1,1)} = 0$, one gets $A_1 = A_2 \sim 1/e_+$, so that at $k = k_+$ we have the pure (+) resonance. In the same manner, the pure (−) resonance can be excited at $k = k_-$ when $\phi = \phi_{(1,-1)} = \pi$, where $A_1 = -A_2 \sim -\tan\varphi/e_-$. This last state presents an extremely high enhancement, $EF \sim 10^4$ at $\theta = 80°$.

A more subtle possibility is the control of the extinction of the field in only one of the cavities while the other one is resonating. Equation (4) shows that this can be achieved provided that $\cot\phi/2 = \pm(e_+/e_-)\tan\varphi$, the sign "+" corresponding to the (0, −1) state and the "−" sign to the (1, 0) state. This condition can be satisfied provided that e_+/e_- is real. This is obtained for $\omega \approx \omega_- + 2\Delta(\gamma_-/\gamma_+)$, which is very close to ω_-. Figure 4 shows EF in each cavity, choosing either $\phi = \phi_{(1,0)}$ or $\phi = \phi_{(0,-1)}$, together with the related maps of the electric field amplitude E_x. These maps show how the field can be strongly localized in only one of the cavities, while the second one is completely extinguished, even though the cavities are identical and separated by a subwavelength distance.

In conclusion, we have demonstrated that the near-field coupling of two metallic resonating cavities leads to a resonance with an extremely thin spectral width, with very intense localized fields. This could be a key point in the understanding of the SERS, as this physics should remain valid in the visible region, except for a scaling factor. Moreover, we propose a way to control the near field of each cavity, enabling this system to act as a subwavelength optical switch operated from the far field. Finally, we point out that this physics implies a medium supporting surface polaritons, coming either from the light coupling with free carriers, or from surface phonons. Similar effects are thus expected, in doped semiconductors or ionic crystals in the spectral range where their dielectric constant is negative.

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