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Abstract

Molecular emission enhancement is generally obtained by proper coupling with external resonances. Here we propose the idea of a plasmonic nanolauncher, i.e., a metamaterial-inspired ultranarrow channel at cutoff. Its peculiar operation provides uniform phase and drastic amplitude increase all over the channel, allowing high emission enhancement independent of the position of an individual or group of molecules along the channel, and of its length and geometry. This may provide a fascinating mechanism for efficient molecular detection and enhanced optical fluorescence.

Disciplines

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Boosting Molecular Fluorescence with a Plasmonic Nanolauncher

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Molecular emission enhancement is generally obtained by proper coupling with external resonances. Here we propose the idea of a plasmonic nanolauncher, i.e., a metamaterial-inspired ultranarrow channel at cutoff. Its peculiar operation provides uniform phase and drastic amplitude increase all over the channel, allowing high emission enhancement independent of the position of an individual or group of molecules along the channel, and of its length and geometry. This may provide a fascinating mechanism for efficient molecular detection and enhanced optical fluorescence.

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Detecting the presence of molecules at optical frequencies is usually made possible by collecting their fluorescent spontaneous emission. Drastic emission enhancement may be achieved when an atom or a molecule is coupled with an external resonance, as first verified by Purcell [1,2], allowing its efficient detection and emission, which is at the basis of various optical applications of current interest, e.g., fluorescence microscopy [3], DNA sequencing [4], single molecule detection [5], and low-threshold lasing [6]. In this sense, the use of resonant cavities [6] or the coupling with plasmonic resonant nanolayers, nanoparticles, and nanoshells [7–10] has been proposed by various groups. As one obvious limitation of these scenarios, the specific location of the molecule of interest, which should be placed near the resonant system at the point where the electric field is maximized, is fundamental in achieving maximized emission. Too close of a distance between the molecule and the resonant system may also produce quenching [9].

In a different field of metamaterials, a novel counter-intuitive resonant phenomenon, named supercoupling, based on resonant transmission, tunneling, and energy squeezing through subwavelength narrow channels has been recently introduced and developed by employing metamaterials with near-zero permittivity [11–17]. In [12–16], moreover, it was shown that similar energy squeezing may be achieved in *hollow* waveguide channels near their cutoff frequencies, demonstrating how the tunneling mechanism described in [11] may conceptually be obtained in a simple microwave waveguide at its cutoff, which may behave effectively as an ϵ -near-zero metamaterial with “infinite” phase velocity. The energy squeezing, in particular, has been shown to lead to a large increase in the electric field inside the narrow channel, and due to its large phase velocity at cutoff, almost zero phase and nearly uniform field amplitude were obtained along the channel [13]. By applying reciprocity, one may heuristically expect to achieve a corresponding increase in the radiation or

emission from a source embedded in the channel, with the interesting property of almost no dependence on its specific position.

Applying these concepts to molecular optics may provide us with a fundamentally novel way to achieve drastic enhancement in optical emission from an individual or a group of molecules coupled to the plasmonic cutoff mode in the channel, envisioning the concept of a plasmonic *nanolauncher*. The important and relevant advantage of such structure over other resonant systems coupled to molecules would be represented by the zero phase variation and nearly uniform distribution of enhanced electric field amplitude inside the channel at cutoff, independent of the channel length and geometry. This may allow a huge emission enhancement that would remain *independent* of the position of the molecule(s) along the channel, with the possibility of achieving constructive interference and maximized emission from collections of molecules radiating in phase placed anywhere inside the channel.

Consider a subwavelength plasmonic rectangular channel of height a_{ch} and width b , carved inside a metal block of length l_{ch} that is placed inside a parallel-plate metal-insulator-metal (MIM) waveguide of height $a \gg a_{\text{ch}}$, as depicted in Fig. 1. The rectangular narrow channel is closed on the sides by metallic implants of width $t/2$, which is larger than the metal skin depth δ , ensuring wave confinement inside the narrow channel. In the following we assume the metal blocks (gray in the figure) are made of silver, with a Drude model for its permittivity $\epsilon_{\text{Ag}} = \epsilon_0(\epsilon_\infty - f_p^2/[f(f + i\gamma)])$, with $f_p = 2175$ THz, $\gamma = 4.35$ THz and $\epsilon_\infty = 5$, which is close to experimentally measured bulk permittivity of silver in the frequency range of interest [18]. The gap filling both the waveguide and the channel is assumed to be free space with permittivity ϵ_0 (although this gap may be filled with any insulating dielectric). An emitting fluorescent molecule is assumed to be positioned inside the subwavelength chan-

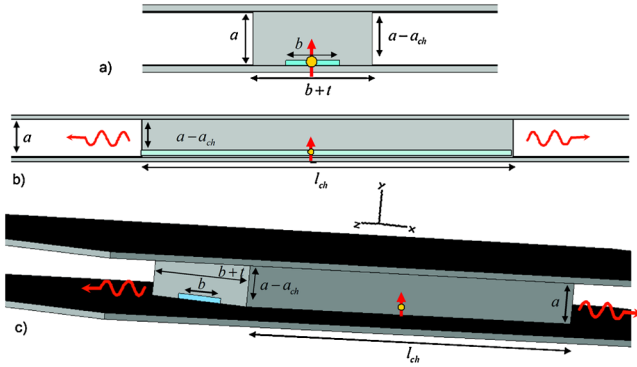


FIG. 1 (color online). Geometry of the structure. Enhanced molecular emission in a plasmonic narrow rectangular channel operating near its cutoff frequency and embedded in a metal-insulator-metal waveguide: (a) front view, (b) side view, (c) perspective.

nel, as sketched in the figure, and it may be adequately modeled as an electric dipole of amplitude p .

In order to first heuristically and intuitively predict the molecular emission properties of this setup, we may first consider the reciprocal problem of wave transmission through such a narrow plasmonic channel. Since silver is a plasmonic material with negative real part of permittivity and moderate losses, the channel is mostly opaque below UV and it is in general hugely mismatched to the outside MIM waveguide sections, since $a \gg a_{\text{ch}}$. Therefore, most of the impinging optical energy guided by the plasmonic MIM waveguide is expected to be reflected at the channel entrance. In [12–15], however, we have shown, for an analogous geometry at microwave frequencies, how a proper design of a subwavelength ultranarrow rectangular channel carved inside a perfectly conducting waveguide and connecting two much thicker waveguide sections may lead to supercoupling, i.e., dramatic transmission enhancement and energy squeezing, despite the small height of the channel and the huge mismatch between the different sections. Such supercoupling at microwave frequencies may be achieved if the lateral width b of the rectangular channel is at the cutoff of its dominant TE_{10} mode, which happens when its lateral dimension $b = \lambda_0/2$, with λ_0 being the wavelength in the material filling the waveguide. In [16] we have extended these concepts to optical frequencies, where we need to take into account the plasmonic nature of the metallic walls, i.e., the finite conductivity and losses of the channel walls, which strongly modify the dispersion properties of the waveguide and of the channel. Using our accurate analytical model [16], here we have designed a narrow subwavelength channel as in Fig. 1 to have its quasi- TE_{10} cutoff around $f = 400$ THz, yielding $a_{\text{ch}} = 20$ nm and $b = 200$ nm. The side implant width $t/2 = 150$ nm is sufficiently larger than $\delta = 24$ nm for silver at this frequency. The structure, embedded in a 2D metal-insulator-metal waveguide with height $a = 200$ nm, was numerically simulated with finite-integration technique CST Microwave StudioTM [19]. The transmis-

sion through the channel for a length $l_{\text{ch}} = 2 \mu\text{m}$ shows a first peak of transmission around $f_0 = 360$ THz, fairly close to our design. A uniform phase distribution along the channel confirms the presence of cutoff tunneling. Higher-frequency transmission peaks are also found in these numerical simulations, consistent with the presence of Fabry-Perot resonances due to the finite length of the channel, analogous to what we found in our microwave experiment [13] and our extended numerical results related to this transmission problem [16] (although in [16] the geometry was composed of an infinite array of such channels). It should be underlined that these higher-order resonances are drastically different in nature from the supercoupling phenomenon based on cutoff resonant tunneling, due to their nonuniform phase and amplitude distributions along the channel [15]. Applying reciprocity, it is expected that a drastic fluorescence enhancement may be achieved at the cutoff tunneling frequency f_0 for a molecule placed in such a narrow channel, operating as a nanolauncher.

In this sense, we report in Fig. 2(a) our numerical results for the transmission enhancement for a molecule placed inside and in the middle of this same narrow channel, by comparing the level of induced (i.e., radiated) electric field evaluated outside the channel (but in the MIM waveguide) at a distance of 10 nm from its end, for three different scenarios: (a) the channel as described above in Fig. 1 (black solid line), (b) the same geometry, but removing the side implants that produce the channel cutoff (blue dotted line), (c) no channel, i.e., removing also the upper block or wall of the channel, i.e., measuring the molecular fluorescence in a uniform plasmonic MIM waveguide with height a (red dashed line). In these simulations, the molecule is modeled as a vertically oriented dipole p with fixed amplitude, emitting at frequency ω . It can be seen that maximum emission is obtained around the cutoff frequency f_0 of the narrow channel. Higher-frequency peaks are also obtained at some of the Fabry-Perot resonances of the channel, but these are strongly dependent on the channel length and on the position of the molecule, due to the nonuniform field distribution of electric field along the channel length. However, the first peak, located at the cutoff frequency of the channel and associated with the supercoupling properties of the ultranarrow channel, possesses no phase variation and nearly uniform field amplitude variation along the channel. The absence of side walls in the channel (blue dotted line) avoids the cutoff, and consequently we get Fabry-Perot resonant peaks all over the spectrum of interest. Also, in this case, the peak amplitudes and positions are strongly dependent on l_{ch} and on the position of the molecule in the channel. Compared to the case in the absence of the channel (red line), transmitted field enhancement of more than 100 times is achieved outside the channel. Transmission enhancement in this setup compared to the same source in free space would be over 4 orders of magnitude. Even larger enhancement values may be achieved by increasing the ratio a/a_{ch} , even though

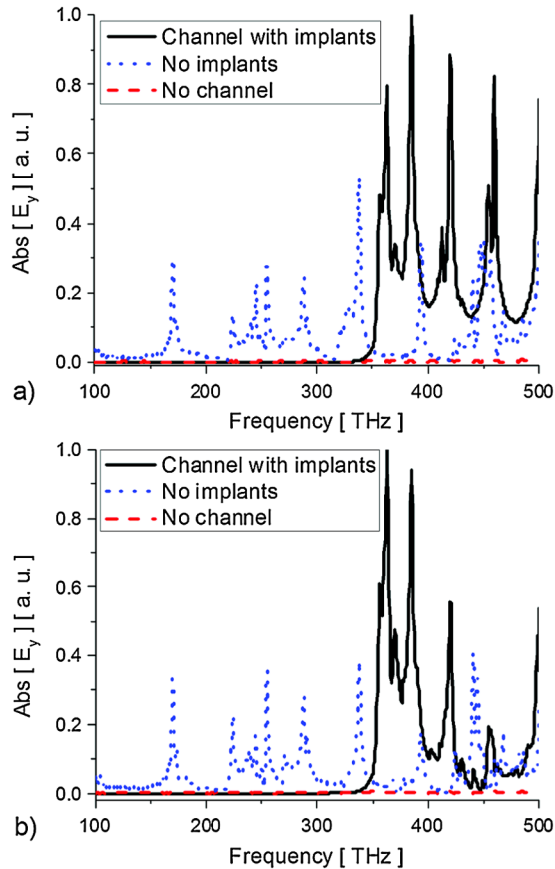


FIG. 2 (color online). Simulation results of electric field amplitude sampled at a distance of (a) 10 nm outside the channel exit; (b) inside the channel at a distance of 900 nm from a molecule placed in the middle of the channel of Fig. 1. The black line refers to the scenario of Fig. 1, the blue dotted line to the same geometry but with no side implants, and the red dashed line to the absence of the narrow channel. For this example: $a_{\text{ch}} = 20$ nm, $b = 200$ nm, $t = 300$ nm, $l_{\text{ch}} = 2$ μm .

a too narrow channel will at some point be limited by the increased absorption, causing quenching, and too large an outer MIM waveguide may support higher-order spurious modes that affect the achievable enhancement. However, large emission enhancement is expected in a plasmonic design at optical frequencies, as numerically shown in Fig. 2(a). It must be noted that this field enhancement here is the result of two factors: (a) the high directionality of the nanolauncher, that may guide the power at will to the desired location (and it can even “bend” the direction of radiation); (2) the overall emission enhancement of the molecule. To separate the two mechanisms, we have also evaluated the total emitted power by the molecule in the three cases of Fig. 2. We have found that at the design frequency the nanolauncher with implants may indeed extract over 33 times more power than the case without implants and 253 times more than the case in the standard parallel-plate waveguide.

Figure 2(b) reports the field amplitude [normalized to the same peak value as in Fig. 2(a)] sampled at a distance

of 900 nm from the molecule, which falls inside the narrow channel for the black and blue lines. It is evident that the field is very much enhanced also inside the channel at the resonance frequencies. The presence of lateral walls in general allows a higher increase of the field values inside the nanolauncher, due to the supercoupling associated with the cutoff modes in the channel, as proposed here, and this is reflected in larger emission enhancement shown in Fig. 2(a). Added to the large enhancement obtainable outside the channel, the key advantage of the nanolauncher resides in the zero phase variation and nearly uniform amplitude of the electric field along the channel. Figure 3 shows, as an example, the electric field E_y (snapshot in time, top view) at the cutoff frequency f_0 for the three geometries of Fig. 2. The launching mechanism and zero phase variation along the channel associated with its cutoff properties are clearly evident in the case of Fig. 3(a). In [20] we have reported the phase of the electric field (side view) for three different channel lengths $l_{\text{ch}} = 2$ μm , $l_{\text{ch}} = 1$ μm , and $l_{\text{ch}} = 500$ nm. There it was shown that independent of the channel length, the phase velocity in the channel is very large, providing an improved emission enhancement mechanism with emission in phase with a molecule placed *anywhere* inside the channel.

One fundamental advantage offered by these in-phase emission properties consists in the possibility of placing the emitting molecule anywhere inside the channel, since its emission properties are interestingly independent of its location. This is fundamentally different from any other emission enhancement methods that rely on resonant structures, for which the location and orientation of the molecule plays a fundamental role in the achievable enhancement. To highlight this interesting feature, Fig. 4

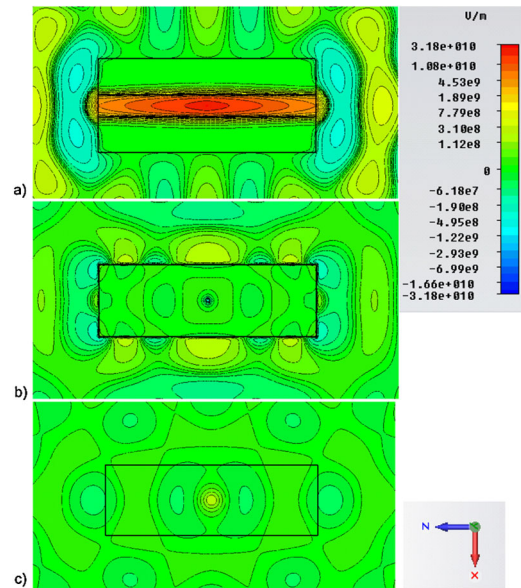


FIG. 3 (color online). Field distribution (snapshot in time) of E_y for a molecule emitting in the center of the narrow channel of Fig. 1 at the cutoff frequency f_0 in the three geometries of Fig. 2.

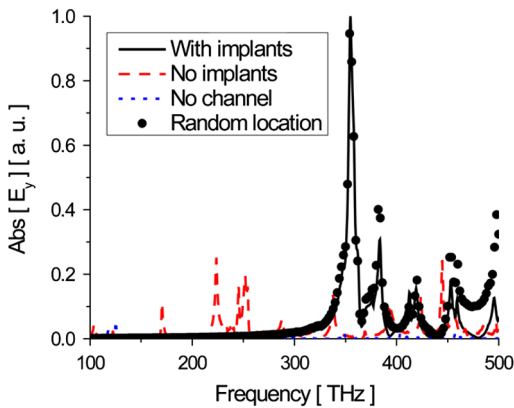


FIG. 4 (color online). For the same geometry as in Fig. 2(a), electric field amplitude sampled at a distance of 10 nm outside of the channel for a collection of nine molecules equispaced inside the narrow channel of Fig. 2. The black dotted line refers to the case in which the molecules are randomly displaced inside the channel.

shows the numerical simulation of emission from a collection of nine equispaced emitters placed inside the nanolauncher of Fig. 2(a) (distant 200 nm from each other). These molecules, which are considered as classical oscillating dipoles, are assumed to emit coherently and in phase. (Here we treat this problem classically.) It may be clearly seen that in this scenario the cutoff tunneling mode described here provides a drastically higher level of emission when compared with any other Fabry-Perot resonance, due to constructive interference among all the different emissions. This is made possible by the unique property of the anomalous cutoff mode excited inside the plasmonic channel, which ensures uniform-phase distribution all over its length.

In Fig. 4 we have also added a black thick dotted line that corresponds to the case for which the nine molecules are randomly distributed in the narrow channel with implants, instead of being equispaced. It is evident that the enhancement peak achieved at the cutoff frequency f_0 is not modified much by the random distribution of the molecules, due to the “staticlike” uniform-phase properties of the resonant electric field inside the channel. This is a unique property of the nanolauncher, operating near its cutoff frequency. It may be added that these features are independent of the possible arbitrary presence of bends and curvatures in the narrow channel, due to its uniform-phase emission features, as widely discussed in [12,14–16]. This implies that the nanolauncher may also be employed to direct the enhanced emission from an individual or a group of molecules in any direction of interest, by bending or curving the narrow channel in any desired way, without affecting the emission enhancement and its frequency of operation.

We predict that the proposed method for enhancing the emission of arbitrarily located molecules and for achieving in-phase constructive interference from collections of in-

phase emitting molecules may have important potential applications in several fields of research. For instance, one may open a tiny subwavelength hole in the side of the channel of Fig. 1, and then insert molecules or DNA samples in the channel. This may greatly enhance the emission of the fluorescent samples. The relatively high Q of the tunneling, as noticed in Figs. 2 and 4, may also be of importance for optically filtering and tuning the molecular emission. One other advantage of this configuration for enhancement of optical fluorescence is that here the molecular fluorescence is already tunneled and guided inside a plasmonic waveguide, and it may be rerouted at will for further optical processing with high efficiency, when compared with other available techniques in which the molecules radiate in an open background.

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- [20] See EPAPS Document No. E-PRLTAO-103-004931 for a phase distribution of the y component of the electric field for a molecule emitting in the center of the narrow channel of Fig. 1 at the cutoff frequency f_0 for three different channel lengths: (a) 2 μm , (b) 1 μm , and (c) 0.5 μm . For more information on EPAPS, see <http://www.aip.org/pubservs/epaps.html>.