Design of an efficient single photon source from a metallic nanorod dimer: a quasi-normal mode finite-difference time-domain approach

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We describe how the finite-difference time-domain (FDTD) technique can be used to compute the quasi-normal mode (QNM) for metallic nano-resonators, which is important for describing and understanding light–matter interactions in nanoplasmonics. We use the QNM to model the enhanced spontaneous emission rate for dipole emitters near a gold nanorod dimer structure using a newly developed QNM expansion technique. Enhanced single photon emission factors of around 1500 and output β-factors of around 60% are found near the localized plasmon resonance. © 2014 Optical Society of America

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Resonant cavity structures have the ability to trap light in very small spatial volumes, which has a wide range of applications in nanophotonics [1]. Various miniaturized cavity structures have been developed over the years to manipulate light at the subwavelength scale, and extreme nanoscale confinement is now possible with metallic nano-resonators (MNRs). In a frequency regime near a localized surface plasmon (LSP) [2], the local density of optical states (LDOS) can be increased dramatically. Consequently, the spontaneous emission (SE) rate of a dipole emitter can be enhanced via the Purcell effect. The resonant enhancement from MNRs has applications in chemical sensing [3], high-resolution imaging [4,5], optical antennas [6], and single photon emission [7].

Although the optical properties of MNRs are being actively pursued, the numeric modeling of the basic cavity physics is extremely demanding, and analytical solutions of the modes only exist for very simple structures, such as spheres. For resonant cavity structures, the natural modes of the system are called quasi-normal modes (QNMs) [8,9], defined as the frequency domain solutions to the wave equation with open boundary conditions (the Silver–Müller radiation condition). Kristensen et al. [10] first used the QNMs to introduce a rigorous definition of the “generalized effective mode volume” and Purcell factor [11], and applied these results to photonic cavity structures. For MNRs, the QNMs also form the natural starting point for developing analytical theories of light–matter interactions in nanoplasmonics [12–14].

One of the most common numerical techniques for obtaining the cavity mode for dielectric cavities is the finite-difference time-domain (FDTD) technique [15]. The FDTD technique allows one to simulate open boundary conditions with “perfectly matched layers” (PMLs), located at the leaky mode region outside the cavity. For dielectric cavities, this open-boundary FDTD approach has been shown to yield excellent agreement with direct integral equation methods [10]. Other time-domain techniques, such as the discontinuous Galerkin time-domain approach, can also use PMLs [16]. For metals, a major problem occurs when using the usual mode calculation approach with a dipole excitation source, since the extracted mode depends sensitively on the dipole position [17] and is, therefore, incorrect. Thus, it has been common practice to excite the MNR with a plane wave source and obtain the scattered field. However, this scattered field is not the same field as the QNM field and it cannot be properly normalized for use in quantum optics, e.g., for obtaining the Purcell factor and effective mode volume [12]—two well-known quantities that help describe the underlying physics of cavity light–matter interactions. Although some frequency-domain techniques exist for computing the QNMs of MNRs [18,19], it is highly desirable to be able to compute the QNMs using the commonly employed and general FDTD technique.

The FDTD method is already widely used by the plasmonics community, and its accuracy for obtaining the enhanced field has been verified against other numerical techniques, such as the multipole expansion technique [20]. In addition, the LDOS can be calculated by employing a dipole excitation source [21–23], which can also model local field effects, e.g., associated with finite-size photon emitters inside a MNR [21]. While direct dipole calculations are feasible, they are very time-consuming and require a new simulation (which may require many hours of computational time) for each spatial position of the dipole emitter; thus, a QNM picture would be much more efficient, since it allows one to simulate dipole responses both as a function of space and frequency as soon as the QNM is obtained and properly normalized.

In this Letter, we first describe how the FDTD technique [24] can be efficiently employed to obtain QNMs of a MNR by filtering the scattered field with a temporal window function. We compute the spatial dependence of the QNM and the effective mode volume, and show excellent agreement with full-dipole calculations for the enhanced SE factor of a dipole emitter. We then show how a gold nanorod dimer can act as an efficient single photon source with large Purcell factors (1500) and impressive output β-factors (around 60%). In contrast with spherical dimer structures [25], we find that the nanorod dimer acts to increase the β-factor for good photon
emitters (in comparison with a single resonator), and has the additional advantage of yielding resonant frequencies in the visible spectrum.

The QNM $\tilde{f}_a$ has a complex eigenfrequency, $\tilde{\omega}_a = \omega_a - i\gamma_a$, with a quality factor $Q = \omega_a/2\gamma_a$. The QNM is normalized through [8,9]

$$\langle \tilde{f}_a | \tilde{f}_a \rangle = \lim_{V \to \infty} \int_V \left( \frac{1}{2\omega} \frac{\partial (\varepsilon(\omega) \omega^2)}{\partial \omega} \right)_{\omega=\omega_a} \tilde{f}_a(r) \cdot \tilde{f}_a(r) \text{d}r$$

$$+ \frac{ic}{2\omega} \int_V \sqrt{\varepsilon(r)} \tilde{f}_a(r) \cdot \tilde{f}_a(r) \text{d}r = \delta_{\mu\nu}. \quad (1)$$

Since the eigenfrequencies are complex, the QNMs diverge in space and each part of Eq. (1) diverges, but the total sum converges quickly in space [12,14]. This convergence occurs approximately when the outgoing field becomes purely oscillatory, rather than evanescent. For spatial points near the resonator, the transverse part of the photon Green function can be expanded as [9,14]

$$G^F(r_1, r_2; \omega) = \sum_\mu (\omega^2/2\omega \tilde{\omega}_\mu) f_\mu(r_1) f_\mu(r_2).$$

One can then derive the effective mode volume $V_{\text{eff}}$ and enhanced SE factor $F_a(r_a, \omega)$, where $r_a$ is the spatial position of a dipole emitter. For dipole positions near the resonator, these quantities are defined through [10,14]

$$\frac{1}{V_{\text{eff}}} = \text{Re} \left( \frac{1}{v_Q} \right), \quad v_Q = \frac{\langle \tilde{f}_a | \tilde{f}_a \rangle}{c B^2 F_a(r_a)}, \quad (2)$$

and

$$F_a(r_a, \omega) = F_{\text{PQ}}(r_a, n_a; \omega) + 1, \quad (3)$$

where

$$F_{\text{PQ}}(r_a) = \frac{3Q/4\pi^2 (\lambda_c^2/n_B^2)(\omega_c^2 \gamma_c/\omega) \text{Im}([n_a - e_B \tilde{f}_a(r_a) \tilde{f}_a(r_a) - n_a]/(\tilde{\omega}_a - \omega))}{}\text{Purcell factor} \times F_\mu \times \text{factor}$$

for any deviations from the field maximum $r_\infty$ for $\omega_c$ is obtained from FDTD. For dipole positions near the resonator, the Purcell factor $F_\mu$ is found that a good choice is to simply choose $n_B = 2\sqrt{\ln 2\tau_{\text{win}}}$ gives the FWHM (full-width at half-maximum) of the time window, and $t_{\text{off}}$ is the time offset from the center of the source pulse to the center of the time window function. One criterion for the selection of time window parameters is to set $\omega_c^2 \tau_{\text{win}}^2/2\tau_{\text{win}} \to Q$ while keeping $\tau_{\text{win}}$ as large as possible. To understand why this works, consider the ideal case for which the integral in Eq. (5) is carried out from $-\infty$ to $+\infty$, and assume that the scattered field is given by $E^\text{S}(t) = \sum_i\Delta\omega_i e^{(-i\omega_i t)}$ ($\omega_i, \gamma_i$ are the frequencies and amplitudes of the resonances), with $\Delta\omega_i = \omega_i - \omega_c$ and $\Delta\gamma_i = \gamma_i - t_{\text{off}}/\tau_{\text{win}}$. If the resonances of the system are well-separated, so that $\Delta\omega_i/\tau_{\text{win}} \gg 1$, then the QNM resonance $\omega_c = \omega_n - i\gamma_n$ will be the only surviving term after applying the windowing function. This is achieved by having $t_{\text{off}}/\tau_{\text{win}} \approx \tau_{\text{win}}$, and $\Delta\omega_i \approx \Delta\gamma_i$ for $i \neq n$, with $\omega_n$ being the only surviving term after applying the windowing function. We have found that a good choice is to simply choose $t_{\text{off}} = 4\pi/\gamma_n$, i.e., two times the lifetime of the QNM.

Next, we apply the above technique to investigate a gold dimer structure made up of two identical nanorods.
We choose a rod radius \( r_a = 15 \text{ nm} \) with an axis length \( l = 100 \text{ nm} \) (along \( y \)). We use the Drude model 
\[ \varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)} \], with parameters similar to gold, with \( \omega_p = 1.26 \times 10^{16} \text{ rad/s} \) (plasmon frequency), and \( \gamma = 1.41 \times 10^{14} \text{ rad/s} \) (collision rate). To get a larger enhancement of the SE for a quantum dipole, we consider two nanorods that are parallel to each other. The separation gap is set to 20 nm, which helps minimize nonradiative decay and also allows sufficient space to embed a quantum emitter, such as a quantum dot (see dipole arrow in Fig. 2). From FDTD analysis, we find the dipole mode is around \( \tilde{\omega}_y / 2\pi = 291.06 - 120.28 \text{ THz} \) \( (\omega_0 \approx 1.2 \text{ eV}) \), which is redshifted (by about 34 THz) with respect to a single nanorod \([14]\) due to the bonding effect; the corresponding quality factor is \( Q \approx 7.2 \), which is smaller than that for a single nanorod \((Q_{\text{single}} \approx 10)\).

To obtain the scattered field and the QNM, a \( y \)-polarized plane wave with frequency \( \omega / 2\pi = 291.06 \text{ THz} \) is employed, incident in the \( x \) direction. The simulation domain size is \( 12 \mu\text{m} \times 8 \mu\text{m} \times 6 \mu\text{m} \) (Fig. 1) and \( 2.4 \mu\text{m} \times 2.4 \mu\text{m} \times 2.4 \mu\text{m} \) (Fig. 2), and we use a conformal meshing scheme with a maximum step size 40 nm in all directions; a smaller refined mesh of 1 nm is used around the metal dimer; one hundred perfectly matched layers have been used with symmetric (antisymmetric) boundary condition in \( x \) (\( y \)) direction for both simulation and the time step is 1.8875 as. We use a time window with parameters \( t_{\text{off}} = 100 \text{ fs} \) and \( t_{\text{win}} = 23.3/\sqrt{m} \) fs. The left and right panels of Fig. 1 show the scattered field and QNM, respectively. As anticipated, the QNM is seen to have an increasing field value for positions further away from the resonator, which is caused by the outgoing boundary conditions and the complex eigenfrequency. A close-up view of the QNM near the resonator is shown in the left panel of Fig. 2, and the correspondent mode volume is calculated to be \( V_{\text{mode}} \approx 1.9 \times 10^{-4} (\lambda_c / n_B)^3 \) (which is about double that of the single nanorod). The oscillating charge distribution is of opposite sign at both ends of each nanorod; in addition, the oscillating charge at the bottom of the upper nanorod has a different sign from the charge at the top of the lower nanorod, which makes the LSPs of the individual nanorods couple effectively. Consequently, the field inside the cavity (gap) is significantly enhanced and \( y \)-polarized. The middle and right panels in Fig. 2 show the \( y \) and \( x \) components of the QNM at \( z = 0 \). The node of the \( y \)-component sits around both ends of the nanorods,

![Fig. 1. Spatial profile of the dipole mode of a gold nanorod dimer, showing both the scattered field and QNM. Left: \(|E_x(x, y, 0; \omega_0)|\) and right: \(|E_y(x, y, 0; \omega_0)|\) at \( \omega_0 = 291.06 \text{ THz} \). The excitation source is a \( y \)-polarized plane wave incident in the \( x \) direction.](image)

![Fig. 2. Close-up view of the QNM profile of the gold dimer. Left: \(|E(x, y, 0; \omega_0)|\), middle: \(|E_y(x, y, 0; \omega_0)|\), and right: \(|E_y(x, y, 0; \omega_0)|\) at \( \omega_0 = 291.06 \text{ THz} \). A \( y \)-polarized dipole at the center of the gap is shown by the white dot and double arrow in the left panel.](image)

![Fig. 3. Enhanced SE factor for the gold nanorod dimer structure, with the blue (dashed) curve given by direct FDTD calculation [Eq. (4)] and the orange (solid) curve given by the mode expansion technique [Eq. (3)]. The emission dipole is \( y \)-polarized at the center of the gap of the dimer (see left panel of Fig. 2).](image)

slightly inside this metal. Similar nodal lines appear for an electric dipole composed of charge \( \mp q \).

We remark that, for these calculations, it is important to choose a \( y \)-polarized plane wave that efficiently excites the QNM of interest. If we use an \( x \)-polarized plane wave, with the parameters same as above, then a rather strange pseudo-mode is obtained. The failure of using \( x \)-polarized plane wave to obtain the QNM can be explained by the fact that the QNM cannot be efficiently excited since the dipole moment lies along the \( y \) direction. Therefore, some care is needed in choosing the correct excitation source, though, in practice, this is easy to do when exciting the QNM of interest. We have also checked that our mode calculation approach successfully reproduces the mode profile and QNMs for dielectric cavity structures, e.g., in Ref. [10], and also for other MNRs.

**Single photon enhancement factors.** As a possible application of using MNRs for single photon emitters, a large enhancement of the SE factor is desired. This can be achieved by having a quantum dipole at the gap center of the dimer. In Fig. 3, the enhanced SE factor...
$F_y$ of a $y$-polarized dipole emitter at the center of the gap is shown by the blue (dashed) curve, using a full-dipole numerical calculation with no approximations, i.e., $F_y^{\text{FDTD}}$ [22,23]. Next, we use the QNM to obtain the enhanced SE factor [14]. The result is shown by the orange (solid) curve in Fig. 3 and agrees extremely well with the full-dipole FDTD calculations. Indeed, the agreement also confirms that the entire response is dominated by a single QNM, which is highly desired for single photon source applications. From our results, we obtain impressive peak emission factors of around 1500. Note that, although the $Q/V$ factor of the dimer structure is about three times smaller than the single nanorod, the emission factor of the dimer is around three times larger than for a single nanorod at an equivalent dipole position. Thus, the $Q/V$ is clearly not the main metric to describe the emission factors for dipole away from the field antinode; the reason for not choosing the field antinode is that the emission would be severely quenched from Ohmic heating, and a single mode Purcell factor ceases to have any meaning [14]. In this regard, the other important figure-of-merit for emitting single photons is the output-coupling $\beta$-factor (probability of photon emission via radiative decay in the far-field) of the dipole emitter, which is computed here to be $\beta \approx 58\%$; again, this is better than the single gold nanorod by about 10%. With respect to the single nanorod, the electric field of the dimer is repelled outside the lossy nanorods (especially around the hot spots) due to opposite charge distribution between them. This leads an overall nonradiative decay reduction ($\propto \text{Im}[e(r, \omega)] |E^{\text{inc}}(r, \omega)|^2 dV$), even though the volume of the integral is doubled.

In summary, we have described how the commonly employed FDTD method can be used to efficiently obtain the QNMs for metallic resonators by applying a filtering function to the scattered field. These calculations are verified by comparing the resulting SE emission factor with full-dipole FDTD numerical calculations, where we find excellent agreement. Using this technique, we have proposed a metal nanorod dimer structure that acts as an efficient single photon source, yielding a large Purcell factor of 1500 and an output $\beta$-factor of around 60%.

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References