Modal Analysis of photonic and plasmonic resonators

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Quasi-normal modes (QNMs) are ubiquitous throughout photonics and are utilized in a wide variety of applications, but determining these modes remains a formidable task in general. Here we show that by exploiting the structure of Maxwell’s equations it is possible to effectively compute QNMs of photonic and plasmonic nanoresonators. The symmetry of Maxwell’s equations allows for a reduction to a system of small order via a Lanczos reduction process through which dominant QNMs can be identified. A closed-form reduced-order model for the spontaneous decay (SD) rate of a quantum emitter is also obtained, which does not require an a priori QNM expansion of the fields. The model is parametric in wavelength and field expansions in dominant QNMs are determined a posteriori. We demonstrate and validate that QNMs of open resonators and the SD rate of a quantum emitter are accurately predicted.

Optical nanoresonators enable us to confine electromagnetic energy to subwavelength domains and give rise to locally enhanced fields that may stimulate various optical processes in a wide variety of applications and research areas such as biophotonics, optical antennas, and diffraction gratings [1–3]. Resonators consisting of metallic nanoparticles that are excited by femtosecond laser pulses are often of particular interest [4], since such resonators allow for the control of light-matter interactions with nanometer and subfemtosecond precision in space and time, respectively, thereby enabling new and exciting applications in cell biology and quantum optics, for example. Moreover, metallic nanoparticles are also often used in resonating structures designed to enhance the SD rate of a quantum emitter that is embedded in such a structure, since this rate depends on the surroundings of the emitter and can be enhanced by an electromagnetic resonance (Purcell effect). The spontaneous decay of a quantum emitter is a purely quantum mechanical process in a reference medium, $\gamma$ denoting the decay rate of the emitter in the resonator configuration of interest and $\gamma_0$ the decay rate of the same emitter in a reference medium, $\gamma/\gamma_0 = P/P_0$, where $P$ and $P_0$ are the time-averaged powers radiated by an electric dipole positioned at the location of the emitter in the resonator configuration and reference medium, respectively. Explicitly, for an emitter located at $x = x_S$ and an electric dipole of the form $J^{ext} = \delta t \delta(x - x_S)$ with dipole moment $p(t) = p(t)\hat{n}_S$, $p(t) = |p(t)|$, and $\hat{n}_S$ a unit vector, we have in steady-state $\hat{J}^{ext} = -i\omega \hat{p}(\omega)\delta(x - x_S)\hat{n}_S$ and the time-averaged radiated power is given by

$$P(\omega) = \frac{\omega}{2} \text{Im} \left[ \hat{p}^{*}(\omega) \hat{E}(x_S, \omega) \cdot \hat{n}_S \right].$$

(1)

To evaluate this power over a frequency or wavelength interval of interest, the electric field strength at the dipole location is required for all frequencies belonging to this interval.

To investigate what local field or decay rate enhancements can be realized, a modal analysis of a resonating structure is typically carried out. For open resonator structures these modes are called Quasi Normal Modes or QNMs and are characteristic of the structure at hand and independent of the excitation. An external source (or incident field) determines what resonant modes are actually excited, while the contribution of these excited modes to a measured field response is determined by the receiver. In open resonant structures, typically only a small number of QNMs are necessary to accurately model measured field responses [6–11] and in SD rate computations the source and receiver location actually coincide, since the electric field strength at the source (dipole) location is required to determine the radiated power (see Eq. (1)).

In this letter we show that by exploiting the symmetry of the first-order Maxwell system, it is possible to efficiently determine QNMs of open resonating structures consisting of dispersive metallic nanoparticles. In addition, we show that the SD rate can be computed without any a priori mode selection, that is, the decay rate can be computed without an explicit mode expansion of the fields as is more commonly done in decay rate computations (see, e.g., [7]).

To describe the reaction of a metallic nanoparticle to the presence of an electromagnetic field, we write the electric displacement vector in Maxwell’s equations as $\hat{D} = \varepsilon \hat{E} + \hat{P} = \varepsilon_\infty \hat{E}$ with $\varepsilon = \varepsilon_0 \varepsilon_\infty$, where $\varepsilon_\infty$ is the instantaneous (high-frequency) permittivity and a polarization vector $\hat{P}$ that is related to the electric field strength via the generic constitutive relation $-\omega^2 \hat{P} = \kappa \hat{E} + \beta_1 \hat{P} + \beta_0 \hat{E}$, where the coefficients $\beta_i$...
determine what type of relaxation is considered (Drude, Lorentz). For a Drude model, for example, we have \( \beta_0 = \varepsilon_0 \omega_p^2 \), \( \beta_1 = 0 \), and \( \beta_2 = \gamma_p \), where \( \omega_p \) is the volume plasma frequency and \( \gamma_p \) the collision frequency of the metal.

Introducing the auxiliary field variable \( \hat{U} = i\omega \hat{P} \), we can write the above constitutive relation and Maxwell’s equations in the consistent first-order form \[ \begin{bmatrix} -i\omega \varepsilon & -1 & -\nabla \times \\ -i\omega & 1 & 0 \\ \beta_0 & -\beta_1 & \beta_2 - i\omega \\ \nabla \times & \beta_1 & -i\omega \mu \end{bmatrix} \begin{bmatrix} \hat{E} \\ \hat{P} \\ \hat{U} \\ \hat{H} \end{bmatrix} = - \begin{bmatrix} 0 \\ 0 \\ 0 \end{bmatrix} + \hat{j}^{\text{ext}} \] \tag{2}

which can be written as \((\mathbf{D} + \mathbf{S} - i\omega \mathbf{M}) \hat{F} = -\hat{Q}\), where \( \mathbf{S} \) and \( \mathbf{M} \) are medium matrices, and the curl operators of Maxwell’s equations are contained in the spatial differentiation operator \( \mathbf{D} \). The electromagnetic field quantities and external sources are collected in the field vector \( \hat{F} \) and source vector \( \hat{Q} \), respectively. For most external sources used in practice (electric dipole, for example), the frequency dependence of the source can be factored out and we write \( \hat{Q} = \hat{p}(\omega) \hat{Q}' \), where \( \hat{p}(\omega) \) is the source wavelet and \( \hat{Q}' \) is frequency independent.

We note that the partial differential operator in Eq. (2) can be symmetrized by scaling the second row with \( \beta_1 \beta_0^{-1} \), the third row with \( -\beta_0^{-1} \), and the fourth row by \(-1\). The efficiency of our method is based upon this symmetry. Furthermore, measured (causal) material behavior can be modeled using this formulation by fitting a rational function representation (i.e. a multipole expansion consisting of a superposition of Lorentz and Drude models) for the complex permittivity to permittivity measurements. This leads to the introduction of multiple auxiliary field variables and the resulting system can be symmetrized in a similar manner as described above.

To carry out a modal analysis of arbitrarily-shaped open resonators, we discretize the first-order Maxwell system in space using a staggered finite-difference Yee mesh. We discretize on such a mesh, since it can be shown that the discretization procedure is mimetic, that is, it is structure preserving and conservation laws and important physical symmetry properties of Maxwell’s equations (symmetry related to energy conservation or symmetry related to reciprocity, for example) have a counterpart after discretization \[ \text{[12]} \text{[13]} \]. Other discretization schemes (finite elements, for example) can also be used, of course, so long as these schemes are mimetic as well.

In addition, radiation towards infinity has to be taken into account, since we are interested in open nanoresonators. Typically, this is realized by surrounding the domain of interest by a so-called Perfectly Matched Layer (PML) \[ \text{[14]} \] in which the spatial coordinates are stretched using frequency dependent stretching functions \[ \text{[15]} \]. However, a disadvantage of such an approach is that in two- and three-dimensional problems this leads to nonlinear eigenvalue problems that need to be solved to find dominant QNMs. Therefore, our approach is to apply the PML technique of \[ \text{[16]} \text{[17]} \] which uses complex spatial step sizes to realize a perfectly matched layer, which do not explicitly depend on frequency and leads to linear eigenproblems. Incorporating this PML technique into our spatial discretization scheme then leads to the discretized first-order Maxwell system

\[
(D + S - i\omega M) \hat{f}_m = -\hat{p}(\omega)q',
\] \tag{3}

where \( D \) contains the discretized curl operators, \( S \) and \( M \) are the discretized medium matrices, and \( \hat{f}_m \) and \( q' \) are the discretized field and source vector, respectively. The above system is not conjugate-symmetric with respect to frequency and its time-domain counterpart is unstable due to the application of a frequency-independent PML. However, conjugate-symmetric frequency-domain field approximations can be obtained from the above system as \[ \text{[16]} \]

\[
\hat{f}(\omega) = -\hat{p}(\omega)\hat{G}(\omega)q,
\] \tag{4}

where \( A = M^{-1}(D + S) \) is the first-order Maxwell system matrix, \( q = M^{-1}q' \) is the scaled source vector, and \( \hat{G}(\omega, \omega') = \hat{R}(\omega, \omega') + \hat{R}^*(\omega', -\omega) \) is the Green’s tensor of the configuration with \( \hat{R} \) the filtered resolvent of matrix \( A \) given by \( \hat{R}(\omega, \omega') = \chi(\omega)(\omega - i\omega)^{-1} \), in which \( \chi(z) \) is the complex Heaviside unit step function defined as \( \chi(z) = 1 \) for \( \text{Re}(z) > 0 \) and \( \chi(z) = 0 \) for \( \text{Re}(z) < 0 \). Note that \( \hat{f}(\omega) \) is conjugate-symmetric, that is, it satisfies \( \hat{f}^*(\omega) = \hat{f}(\omega) \), provided that \( \hat{p} \) is conjugate-symmetric.

For practical three-dimensional problems direct evaluation of Eq. (4) is usually not feasible, since the order \( n \) of the Maxwell system matrix \( A \) is simply too large (in 3D, typically \( n = O(10^{6-7}) \)). It can be shown, however, that matrix \( A \) satisfies a particular symmetry property that allows for efficient Lanczos model-order reduction. In particular, defining the bilinear form \( \langle x | y \rangle_{\text{WM}} = x^T W M y, \) \( x, y \in \mathbb{C}^n \), where \( W \) is a specific step size matrix \[ \text{[12]} \text{[13]} \], it can be shown that \( \langle A x | y \rangle_{\text{WM}} = \langle x | A y \rangle_{\text{WM}} \) for all vectors \( x, y \in \mathbb{C}^n \). Moreover, the bilinear form \( \langle f | f \rangle_{\text{WM}} \) is a discrete approximation of the integral

\[
\mathcal{L} = \int \varepsilon \hat{E}^2 + \beta_1 \beta_0^{-1} \hat{P}^2 - \beta_0^{-1} \hat{U}^2 - \mu \hat{H}^2 dV
= \int \hat{E} \cdot \frac{\partial \omega \varepsilon_c(\omega)}{\partial \omega} \cdot \hat{E} - \mu \hat{H} \cdot \hat{H} dV
\] \tag{5}

which in the literature is used to normalize QNMs \[ \text{[7]} \].

In 1931, Krylov \[ \text{[19]} \] used what are now called polynomial Krylov subspaces in his analysis of oscillations of mechanical systems (e.g. ships). Here, the symmetry of matrix \( A \) allows us to follow a similar approach. Specifically, the symmetry of \( A \) can be used to reduce this matrix to tridiagonal form using a three-term Lanczos-type recurrence relation \[ \text{[12]} \text{[18]} \]. Carrying out \( m \) steps of this reduction process, we obtain the decomposition

\[
A V_m = V_m T_m + \beta_{m+1} V_{m+1} T^T m,
\] \tag{6}
where \( T_m \) is a tridiagonal matrix of order \( m \ll n \) containing the Lanczos recurrence coefficients and \( V_m \) is a tall \( n \)-by-\( m \) matrix with a column partitioning \( V_m = (v_1, v_2, \ldots, v_m) \). The columns of matrix \( V_m \) are referred to as Lanczos vectors, which are taken to be quasi-orthonormal i.e., \( \langle v_i | v_j \rangle_{WM} = \delta_{ij} \), where \( \delta_{ij} \) is the Kronecker delta. Furthermore, \( \beta_{m+1} \) in Eq. (6) is a Lanczos recurrence coefficient and \( e_m \) is the \( m \)th canonical basis vector.

To find an approximate spectrum of the Maxwell system matrix \( A \), the Lanczos reduction process can be started with any (randomly generated) starting vector \( v_1 \) satisfying \( \langle v_1 | v_i \rangle_{WM} = 1 \). If, however, modes excited by a given external source are of interest (as in SD rate computations, for example) then we take \( v_1 = (q | q)^{-1/2} q \) as a starting vector in the reduction process.

The Lanczos decomposition of Eq. (6) serves as a starting point for our modal analysis and SD rate computations. First, as is well known \[20\], the decomposition can be used to find approximate QNMs of the open resonator system. Specifically, if \( (\theta_{jm}^{[m]}, z_{jm}^{[m]}) \) is an eigenpair of the reduced matrix \( T_m \), then postmultiplication of \( z_{jm}^{[m]} \) by \( z_{jm}^{[m]} \) shows that \( (\theta_{jm}^{[m]}, V_m z_{jm}^{[m]}) \) is an approximate eigenpair of \( A \) with a residual given by \( \beta_{m+1} (e_m | z_{jm}^{[m]}) | v_{m+1} \rangle \).

Converged QNMs \( y_j = V_m z_{jm}^{[m]} \) can be identified by computing the norm of this residual. Note that normalizing the eigenvectors \( z_{jm}^{[m]} \) of \( T_m \) such that \( (z_{jm}^{[m]} | z_{jm}^{[m]}) = \delta_{ij} \) ensures that the approximate QNMs \( y_j \) are normalized with respect to the bilinear form \[5\], i.e., \( \langle y_j | y_j \rangle_{WM} = \delta_{ij} \).

Second, for a given external source \( q \) the decomposition can be used to construct the reduced-order model (ROM) Eq. (6)

\[
\hat{m}(\omega) = i\omega \hat{p}(\omega) (q | q)^{1/2} \times \left[ V_m \hat{R}(T_m, \omega) | e_1 \rangle + V_m^* \hat{R}^*(T_m, -\omega) | e_1 \rangle \right],
\]

which gives an approximation of the three-dimensional field of order \( m \). In SD rate computations, however, only the projection of the electric field onto the direction of the dipole moment at the dipole location is of interest. For this projection, we have \( \hat{E}(x_S, \omega) \cdot n_S = \langle \hat{m}(\omega) | q \rangle_{WM} \) and substitution in Eq. (6) gives the ROM for the radiated power

\[
P_m(\omega) = P_R \text{Re} \left[ \langle e_1 | \hat{G}(T_m, \omega) | e_1 \rangle \right]
\]

with \( P_R = 0.5 \omega^2 | \hat{p}(\omega) |^2 (q | q)_{WM} \). Only filtered resolvents of the reduced tridiagonal matrix \( T_m \) need to be computed to evaluate this power over a complete frequency (wavelength) interval of interest and no \textit{a priori} expansion of the fields in QNMs is required. Explicitly, assuming that \( T_m \) can be diagonalized and arranging its eigenvectors as columns in matrix \( Z_m = (z_{1m}^{[m]}, z_{2m}^{[m]}, \ldots, z_{nm}^{[m]}) \), we have

\[
P_m(\omega) = P_R \text{Re} \left[ \sum_{k=1}^m w_k^2 \hat{R}(\theta_k^{[m]}, \omega) + (w_k^*)^2 \hat{R}^*(\theta_k^{[m]}, -\omega) \right],
\]

where \( w_k \) is the \( k \)th element of \( Z_m^T e_1 \). As mentioned above, converged QNMs can be identified by computing the residual of the approximate QNMs and their contribution to the radiated power \( P_m(\omega) \) can be determined using the spectral expansion of Eq. (9).

In Fig. 1 the computed Purcell factor over a complete wavelength interval of interest is shown. The solid line signifies the result obtained with the FEM-RCWA method \[17\], while the dashed line shows the converged reduced-order model response obtained via Lanczos reduction. The computed enhancement factors of both methods are in good agreement with each other. The un-reduced Maxwell system has an order of \( n = 8.6 \) mil-
metric resonances, where the wavelength of the anti-symmetric resonance is larger than the wavelength of the symmetric resonance in accordance with the theory of electronic oscillators. In particular, the wavelengths of the fundamental anti-symmetric and symmetric resonances are $\lambda = 1034 + 34i$ nm and $\lambda = 891 + 68i$ nm, respectively. Figure 3(a) shows an isosurface plot of $\Re(\hat{E}_x)$ of the anti-symmetric resonance. Isosurface plots of $\Re(\hat{E}_x)$ of the dominant QNM, whereas isosurface plots of $\Re(\hat{E}_x)$ of the anti-symmetric resonance are shown Figs. 3(b) - (e). Finally, a higher harmonic anti-symmetric resonance is depicted in Fig. 3(b).

In conclusion, we have shown that the symmetry of Maxwell’s equations can be used to effectively compute QNMs of three-dimensional arbitrarily-shaped dispersive nanoresonators. A mimetic discretization of the first-order Maxwell equations for dispersive media leads to a large-scale discretized Maxwell system that is symmetric with respect to a particular bilinear form. This symmetry property allows us to reduce the large-scale Maxwell system to a system of much smaller order via a Lanczos-type reduction process and to find QNMs that are quasi-orthonormal with respect to the bilinear form. Moreover, we have presented a new closed-form reduced-order model for the SD rate of a quantum emitter that is parametric in wavelength meaning that a single model approximates the SD rate over a complete wavelength interval of interest, i.e. the model allows for wavelength sweeps. This feature is important in many applications in quantum optics, where the SD rate is controlled and optimized by modifying the background configuration of the quantum emitter. Specifically, for each background realization a single ROM provides an SD rate response over a complete wavelength interval of interest, which can significantly speed up the design and optimization of the resonating environment. Furthermore, the ROM does not require an a priori expansion of the electric field in terms of QNMs. It is not necessary to determine beforehand which QNMs contribute the most to the electric field at the dipole location. In fact, which modes actually contribute on a given wavelength interval can be determined from the reduced Lanczos system and super-
imposing the most contributing modes until a specified error criterion is met. In this manner, the ROM for the SD rate gives us control over the error that is introduced when a subset of QNMs is used to approximate the SD rate of a quantum emitter.

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