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Modal analysis of Mie resonators: Pole-expansion of scattering operators

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Abstract. Most of the resonant photonic structures behave like open cavities for light where light is trapped for some time before leaking or being absorbed. Their modes are called quasi-normal modes and are associated with complex eigenfrequencies $\omega_n = \omega'_n + i\omega''_n$, ω''_n characterizing the rate at which the energy leaks from the structure. As a consequence, one can show that the fields of these modes diverge far away from the scatterer. This is problematic when one attempts to develop a theoretical description of the resonant interaction between light and resonant photonic structures in terms of their quasi-normal modes. Moreover, the existence or not of a non-resonant term in addition to these resonant contributions is still an open problem. Here, we address these two problems by deriving pole-expansions of the scattering operators of resonant optical structures. We evince the existence of a non-resonant term and we solve the problem of the divergence by studying the scattered field in the time domain and by using the causality principle. The quasi-normal mode expansion that we obtain will be of a great use to study light-matter interactions since it allows to determine the optical response of a photonic resonator both in the time and frequency domain.

1. Introduction

The resonant interaction between light and nanoscale photonic structures is a fundamental process in photonics. Examples of such resonances are plasmonic resonances hosted by metallic nanostructures, whispering gallery modes also called morphological-dependent resonances occurring in small index dielectric microspheres/microdisks or low order Mie resonances featured by high-refractive index nanoparticles. One can explain the existence of such resonances because nanostructures behave like open cavities for light. These cavities consequently suffer from significant leakage of electromagnetic energy due to scattering into their environment. Besides scattering losses, some optical resonators such as plasmonic resonators can also suffer from additional absorption losses. Resonances in photonics consequently all share a common mathematical description. They can in fact all be considered to be modes of open optical cavities often referred as quasi-normal modes or resonant states that are associated with complex eigenfrequencies $\omega_n = \omega'_n + i\omega''_n$ where $\omega''_n < 0$ if the time-dependence of the fields is $e^{-i\omega t}$. These modes also fulfill the outgoing boundary conditions and thus asymptotically behave like $e^{ik_n r} = e^{i\frac{\omega_n}{c} r}$ when $r \rightarrow \infty$. This condition coupled to the fact that $\omega''_n < 0$ yields an exponential divergence of the eigen-vectors. This divergence has long been known as “exponential catastrophe” [1] but



is one of the main problems one faces when attempting to derive a theoretical description of the interaction between light and resonant photonic structures based on their quasi-normal modes. Such a quasi-normal mode expansion would however be of a great interest to strengthen the understanding of the enhancement of light-matter interactions near a resonant photonic structure [2]. Here, we will derive such a modal expansion and we are consequently going to address these two major problems:

- (i) Does the scattered field only contain resonant terms or is there also a non-resonant term?
- (ii) The second problem is related to the already mentioned exponential catastrophe: how is it possible to expand the scattered field on a set of exponentially diverging fields? We will start by establishing the pole expansion of the scattering operators characterizing the optical response of a resonant scatterer. This result will allow us to address problem (i). We will subsequently study the optical response of the scatterer in the time-domain and we will show that some considerations based on the causality principle will allow us to address the problem of the “exponential catastrophe”. The usefulness and the accuracy of the QNM analysis will be shown by studying the optical response of a dispersionless spherically symmetric scatterer.

2. Results:

2.1. Pole expansion of scattering operators:

Let us consider the case of light-scattering by a 3D scatterer of dielectric permittivity ϵ_s in the frequency domain. An excitation field $\mathbf{E}_{exc}(k\mathbf{r})$ that is usually a plane wave or a superposition of plane waves interacts with a scatterer. This process produces a scattered field $\mathbf{E}_{scat}(k\mathbf{r})$ that is radiated away from the scatterer. In the case of 3D scattering, it is more adapted to expand these fields on the set of regular $(\mathbf{M}_{n,m}^{(1)}(k\mathbf{r}), \mathbf{N}_{n,m}^{(1)}(k\mathbf{r}))$ and outgoing VPWs $(\mathbf{M}_{n,m}^{(+)}(k\mathbf{r}), \mathbf{N}_{n,m}^{(+)}(k\mathbf{r}))$:

$$\mathbf{E}_{exc}(k\mathbf{r}) = E_0 \sum_{n,m} e_{n,m}^{(h)}(\omega) \mathbf{M}_{n,m}^{(1)}(k\mathbf{r}) + e_{n,m}^{(e)}(\omega) \mathbf{N}_{n,m}^{(1)}(k\mathbf{r}) \quad (1)$$

$$\mathbf{E}_{scat}(k\mathbf{r}) = E_0 \sum_{n,m} f_{n,m}^{(h)}(\omega) \mathbf{M}_{n,m}^{(+)}(k\mathbf{r}) + f_{n,m}^{(e)}(\omega) \mathbf{N}_{n,m}^{(+)}(k\mathbf{r}) \quad (2)$$

The definition of $(\mathbf{M}_{n,m}^{(1)}(k\mathbf{r}), \mathbf{N}_{n,m}^{(1)}(k\mathbf{r}))$ and $(\mathbf{M}_{n,m}^{(+)}(k\mathbf{r}), \mathbf{N}_{n,m}^{(+)}(k\mathbf{r}))$ is provided in [3]. It is particularly useful to introduce the T-matrix that relates the scattered field to the incident field. For a spherically-symmetric scatterer, the T-matrix is diagonal and its coefficients are equal to $T_n^{(i)}(\omega) = \frac{f_{n,m}^{(i)}(\omega)}{e_{n,m}^{(i)}(\omega)}$ where $i = e$ or h . The total field outside the scatterer is equal to the superposition of the excitation field and the scattered field: $\mathbf{E}_{tot}(k\mathbf{r}) = \mathbf{E}_{exc}(k\mathbf{r}) + \mathbf{E}_{scat}(k\mathbf{r})$. One can also make the choice to express the total field as a superposition of incoming $\mathbf{E}_{in}(k\mathbf{r})$ and outgoing fields $\mathbf{E}_{out}(k\mathbf{r})$: $\mathbf{E}_{tot}(k\mathbf{r}) = \mathbf{E}_{in}(k\mathbf{r}) + \mathbf{E}_{out}(k\mathbf{r})$. $\mathbf{E}_{in}(k\mathbf{r})$ and $\mathbf{E}_{out}(k\mathbf{r})$ may be expanded on the set of incoming $(\mathbf{M}_{n,m}^{(-)}(k\mathbf{r}), \mathbf{N}_{n,m}^{(-)}(k\mathbf{r}))$ and outgoing $(\mathbf{M}_{n,m}^{(+)}(k\mathbf{r}), \mathbf{N}_{n,m}^{(+)}(k\mathbf{r}))$ VPWs. One can introduce the S-matrix that relates the outgoing field to the incoming field and its diagonal coefficients can be shown to be equal to $S_n^{(i)}(\omega) = 1 + 2T_n^{(i)}(\omega)$. In this study, we want to describe the light scattering by a nanoparticle in terms of its quasi-normal modes. It is thus necessary to establish a link between the quasi-normal modes of a scatterer and the scattering operators *i.e.* the S and T matrices. Since quasi-normal modes are the sourceless solutions of Maxwell equations, the QNM eigen-frequencies necessarily correspond to the poles of the S and T matrices *i.e.* the conditions for which a scattered field can exist without any excitation field. A quasi-normal mode description of light-scattering by a nanoparticles can consequently be obtained by deriving the pole expansion of its S and T matrix coefficients [4, 6, 7, 8]. We derived this pole expansion for a dispersionless spherically-symmetric scatterer and found the

following expressions [3]:

$$S_n^{(i)}(\omega) \approx e^{-2ikR} \left(S_{n.r.}^{(i)} + \sum_{\alpha=-M}^M \frac{r_{n,\alpha}^{(i)}}{\omega - \omega_{p,n,\alpha}^{(i)}} \right) \quad (3)$$

$$T_n^{(i)}(\omega) \approx \frac{S_{n.r.}^{(i)} e^{-2ikR} - 1}{2} + \frac{e^{-2ikR}}{2} \sum_{\alpha=-M}^M \frac{r_{n,\alpha}^{(i)}}{\omega - \omega_{p,n,\alpha}^{(i)}} \quad (4)$$

where $\omega_{p,n,\alpha}^{(i)}$ are the poles of the S and T matrices and correspond to the eigen-frequencies of the quasi-normal modes of the scatterer. $r_{n,\alpha}^{(i)}$ is the residue associated with the pole $\omega_{p,n,\alpha}^{(i)}$ whose expressions are provided in [3]. $S_{n.r.}^{(i)}$ is the non-resonant term and is equal to $S_{n.r.}^{(i)} \equiv 1 + \sum_{\alpha=-M}^M \frac{r_{n,\alpha}^{(i)}}{\omega_{p,n,\alpha}^{(i)*}}$ and is calculated by using the sum rules reported in [3, 5]. The poles $\omega_{p,n,-\alpha}^{(i)}$ are equal to $-\omega_{p,n,\alpha}^{(i)*}$. One clearly sees that these formulas predict the existence of a non-resonant term for both the T and S matrix coefficients. This expression consequently provides an answer to question (i) in the introduction. These formula are quite useful since they allow for an accurate prediction of the optical response of a scatterer in terms of a finite number of eigen-frequencies $\omega_{p,n,\alpha}^{(i)}$. Thanks to these formulas, resonances in the optical response of a photonic structure may be easily explained thanks to a small number of modes. This is illustrated in Fig. 1 where the dipole electric partial scattering cross-section of a $\varepsilon = 16$ spherical dielectric scatterer is shown. Overall, a very accurate prediction of the partial by taking $M = 50$ *i.e* by taking into account 100 poles.

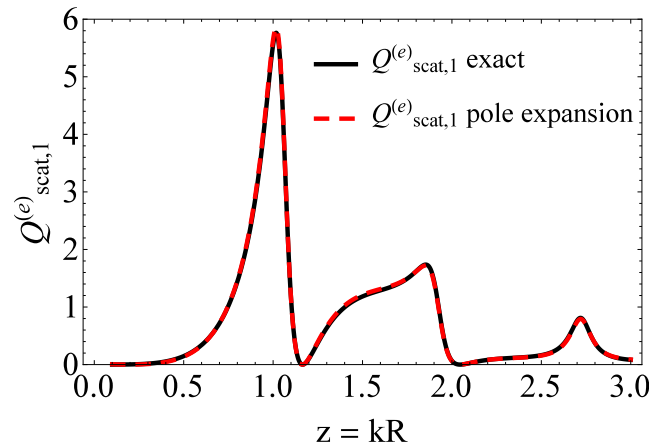


Figure 1. Comparison between exact calculations (solid black line) and the quasi-normal mode expansion (dashed red line) of the electric dipolar scattering efficiency for a $\varepsilon = 16$ dielectric spherical scatterer. The quasi-normal mode expansion is carried by setting $M = 50$ in Eq. (4).

2.2. Exponential catastrophe and scattered field in the time domain:

In the previous section, the existence of a non-resonant term was established. Now, let us consider the problem of the exponential divergence of the quasi-normal modes. Since these modes satisfy the outgoing boundary condition, their field is proportional to $e^{i\frac{\omega n}{c}r}$ when $r \rightarrow \infty$ as a consequence it exponentially diverges far away from the scatterer. One could think that this divergence forbids the derivation of the scattered field on the set of quasi-normal modes. In [3],

we showed that it was possible to derive such an expansion when one considers light-scattering in the time domain. In fact, if the excitation field is causal, meaning that it possesses a cutoff in time, one can establish that the scattered field admits a divergence-free modal expansion of the scattered field. In order to obtain such a divergence-free expansion of the time-dependent scattered field, we start by expressing the frequency dependent scattered field using the pole expansion of the S and T matrices previously obtained. We then evaluate its inverse-Fourier transform by means of the residue theorem. We thus found that there are two types of terms in the time-dependent optical response of a scatterer: a non-resonant term that possesses the same temporal envelope as the excitation field and resonant terms that correspond to the convolution between the excitation and exponentially decreasing terms.

3. Conclusion:

Here, we have reported our results concerning the quasi-normal mode analysis of the optical response of Mie resonators. By deriving pole-expansion of the scattering operators, we evinced the existence of a non-resonant term in the quasi-normal mode expansion of the scattered field. We also achieved to derive a divergence-free expansion of the time-dependent scattered field by using the causality principle.

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