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Research Report No. 2020-19  
March 2020

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# Modal expansion for plasmonic resonators in the time domain\*

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## Abstract

We study the electromagnetic field scattered by a metallic nanoparticle with dispersive material parameters placed in a homogeneous medium in a low frequency regime. We use asymptotic analysis and spectral theory to diagonalise a singular integral operator, which allows us to write the field inside and outside the particle in the form of a complete and orthogonal modal expansion. We find the eigenvalues of the volume operator to be associated, via a non-linear relation, to the resonant frequencies of the problem. We prove that all resonances lie in a bounded region near the origin. Finally we use complex analysis to compute the Fourier transform of the scattered field and obtain its modal expansion in the time domain.

**Mathematics Subject Classification (MSC2000).** 35R30, 35C20.

**Keywords.** plasmonic resonance, normal modes, dispersive scatterer, time domain expansion

## 1 Introduction

### 1.1 Position of the problem

Modal analysis has been a useful tool in wave physics to understand the behaviour of complex systems and to numerically compute the response to an excitation. For a bounded, lossless system, the operator  $\Delta^{-1}$  associated with the wave equation with Dirichlet or Neumann boundary conditions is compact and self-adjoint when studied in the right functional spaces. Hence it can be diagonalised and a complete basis of eigenmodes with real eigenfrequencies can be exhibited. The response of the system to an excitation can then be computed by summing the response of each mode to the excitation.

However, when the system exhibits loss (by absorption or radiation), the operator cannot be diagonalised with the classical spectral theorem and the eigenfrequencies have a negative imaginary part. Using sophisticated micro-local analysis and building on the Lax-Phillips scattering theory [21], several authors have obtained *resonance expansions* in various cases.

We refer to [28, 33] for a general presentation of resonance expansions and to the recent book [17] for the state of the art. These expansions rely on *high frequency estimates* of the resolvent and, to the best of our knowledge, rigorous resonance expansions for the transmission problem in unbounded domains have so far only been obtained for the local scalar wave equation [26, 25] with non-negative coefficients until the recent progress of [8, 9], which deals with *non-locality* in time. One should also note that the spectral analysis used in these papers is far from elementary and hardly accessible to the non-specialist of semi-group theory and micro-local analysis, thus making these results difficult to use.

Nevertheless, in the physics community modal analysis is heavily used in numerical nano-photonics (see the review paper [20] and references therein) to study the interaction of light with resonant structures such as *nanoparticles* and *metamaterials*, which are described by the *non-local* Maxwell

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\*This work was supported in part by the Swiss National Science Foundation grant number 200021-172483.

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equations. In practice, modes (or generalised eigenvectors) are computed by solving, in the frequency domain, the source-free Maxwell's equations satisfying the outgoing radiation condition. Several practical and theoretical issues naturally arise with this approach. Fields oscillating at a complex frequency with negative imaginary part solving the radiation condition diverge exponentially in space (Lamb's so called *exponential catastrophe* [29]), making physical interpretations difficult and numerical computations in large domain very problematic without renormalisation techniques [30]. Generalised modes are not an orthogonal family, making energy considerations difficult. The density of the linear span of the family of modes has not been shown, raising questions about the possibility of representation of any electromagnetic field as a sum of modes.

In this article, we study the electromagnetic field scattered by a metallic nanoparticle in a *low frequency regime*, which is the one relevant for applications as it roughly corresponds to the visible/infrared frequency range and will be defined more precisely in section 3.4.

## 1.2 Main contributions

For a metallic nanoparticle  $D$  under the illumination  $\mathbf{E}^{\text{in}}$  at frequency  $\omega$ , we give a Laurent series expansion in  $\omega$  for the electromagnetic field inside the particle:

$$\mathbf{E}(\omega) = \sum_n \frac{\alpha_n(\omega)}{\omega - \omega_n} \langle \mathbf{E}^{\text{in}}(\omega), \mathbf{e}_n \rangle \mathbf{e}_n \text{ in } D,$$

where  $\mathbf{e}_n$  are eigenvectors of some singular integral operator,  $\alpha_n$  are holomorphic functions and the poles  $\omega_n$  belong to the lower complex half plane and depend on the size and shape of  $D$  (see Lemma 4.1). We also give a similar expansion for the scattered field outside the particle (see Proposition 4.1)). Note that  $(\omega_n)$  are not the *static resonances* predicted by the static theory of electromagnetic fields, but *dynamic resonances* that take into account retardation effects due to the non-zero ratio size of the particle over the wavelength of the incoming field in the frequency range considered (see Section 3). This is important since the static theory does not give a good approximation of resonant frequencies for moderate size nanoparticles [22, Section IV-B]. We also show that all the resonances lie in a bounded region near the origin of the lower complex half plane. From this pole expansion, using only elementary complex analysis tools (Paley-Wiener and residue theorems), we give a resonance expansion for the *low-frequency part* of the electromagnetic field in the time domain in Theorem 5.1. Since the poles lie in a bounded region near the origin, we can capture all the resonances with a low frequency approximation, without the need for the usual high-frequency resolvent estimates required for resonance expansions in the Lax-Phillips setting. To the best of our knowledge, it is the first time that an expansion of this type is obtained using only integral operator theory and elementary spectral analysis. We show that in the time domain, causality ensures that the electromagnetic field does not diverge exponentially in space. Similar results were obtained in [12] for a non-dispersive dielectric spherical scatterer in any frequency range. Our result is valid for an arbitrarily shaped scatterer (with some regularity conditions) and for dispersive media, but only for *low frequencies*. We would like to point out that despite the apparent similarities, our approach is very different from diagonalising the volume integral operator from the Lippmann-Schwinger equation at a fixed real frequency  $\omega \in \mathbb{R}$ , as it is done in [10], since, in these expansions, the poles obtained depend on the working frequency  $\omega$  and cannot be used to compute the electric field in the time domain with a residue theorem.

## 1.3 Sketch of the article

The paper is structured in the following way: We start from the classical volume integral equation at frequency  $\omega$  for the electric field  $\mathbf{E}$ , which reads, for a scatterer  $D$  constituted of a dispersive material under the excitation  $\mathbf{E}^{\text{in}}$ ,

$$(I - \gamma^{-1}(\omega)\mathcal{T}^\omega) \mathbf{E} = \mathbf{E}^{\text{in}} \quad \text{in } D,$$

where  $\mathcal{T}^\omega$  is a singular integral operator and  $\gamma$  is a non-linear function of  $\omega$  that depends on the model for the permittivity of the scatterer (Section 2.3). We begin by studying the static-limit operator  $\mathcal{T}^0$ . Using links with the Neumann-Poincaré operator and the Plemelj symmetrisation principle in  $H^{-1/2}(\partial D)$ , we build an eigenbasis for  $\mathcal{T}^0$  (Section 2.4). Then, writing  $\mathcal{T}^\omega$  as a perturbation of  $\mathcal{T}^0$  and using a perturbative spectral analysis technique, we compute the eigenvalues of  $\mathcal{T}^\omega$  (Section 2.5). We then compute the roots of the equation  $\gamma(\omega) \in \sigma(\mathcal{T}^\omega)$  and define these as the resonant frequencies of the system (Section 3). In Section 4 we give the Laurent series for the electric field. Finally, in Section 5 we give the resonance expansion for the low-frequency part of the electromagnetic field in the time domain.

## 2 Maxwell's equations for a metallic resonator

### 2.1 Problem geometry

We are interested in the scattering problem of an incident spherical wave on a plasmonic nanoparticle. The homogeneous medium is characterised by the electric permittivity  $\varepsilon_m$  and the magnetic permeability  $\mu_m$ . Let  $D$  be a smooth bounded domain in  $\mathbb{R}^3$ , of class  $C^{1,\alpha}$  for some  $\alpha > 0$ , characterised by electric permittivity  $\varepsilon_c$  and magnetic permeability  $\mu_c$ . We assume the particle to be non-magnetic, i.e.,  $\mu = \mu_c = \mu_m$  in  $\mathbb{R}^3$ . We define the wavenumbers  $k_c = \omega\sqrt{\varepsilon_c\mu_c}$  and  $k_m = \omega\sqrt{\varepsilon_m\mu_m}$ . Let  $\varepsilon = \varepsilon_c\chi(D) + \varepsilon_m\chi(\mathbb{R}^3 \setminus \bar{D})$ , where  $\chi$  denotes the characteristic function. We denote by  $c_0$  the speed of light in vacuum,  $c_0 = 1/\sqrt{\varepsilon_0\mu_0}$ , and by  $c$  the speed of light in the medium,  $c = 1/\sqrt{\varepsilon_m\mu_m}$ . Let  $D = z + \delta B$ , where  $B$  is the reference domain and contains the origin and  $D$  is located at  $z \in \mathbb{R}^3$  and has a characteristic size  $\delta$  small compared to the operating wavelength  $\delta k_m \ll 1$ . Let  $\nu$  be the normal vector. Throughout this paper, we assume that  $\varepsilon_m$  and  $\mu_m$  are real and positive. We also assume that  $\Im\varepsilon_c \leq 0$ .

Hereafter we use the Drude model [24] to express the electric permittivity of the particle:

$$\varepsilon_c(\omega) = \varepsilon_0 \left( 1 - \frac{\omega_p^2}{\omega^2 + i\omega T^{-1}} \right), \quad (1)$$

where the positive constants  $\omega_p$  and  $T$  are the plasma frequency and the collision frequency or damping factor, respectively. We write  $\varepsilon_m = \sqrt{n}\varepsilon_0$  where  $n$  is the refractive index of the medium.

### 2.2 Formulation

For a given incident wave ( $\mathbf{E}^{\text{in}}, \mathbf{H}^{\text{in}}$ ) solution to Maxwell's equations

$$\begin{cases} \nabla \times \mathbf{E}^{\text{in}} = i\omega\mu_m\mathbf{H}^{\text{in}} & \text{in } \mathbb{R}^3, \\ \nabla \times \mathbf{H}^{\text{in}} = -i\omega\varepsilon_m\mathbf{E}^{\text{in}} - i\frac{1}{\omega\mu_m}\mathbf{p}\delta_s & \text{in } \mathbb{R}^3, \end{cases}$$

where the source at  $s$  has a dipole moment  $\mathbf{p} \in \mathbb{R}^3$ , let  $(\mathbf{E}, \mathbf{H})$  be the solution to the following Maxwell equations:

$$\begin{cases} \nabla \times \mathbf{E} = i\omega\mu\mathbf{H} & \text{in } \mathbb{R}^3 \setminus \partial D, \\ \nabla \times \mathbf{H} = -i\omega\varepsilon\mathbf{E} & \text{in } \mathbb{R}^3 \setminus \partial D, \\ [\boldsymbol{\nu} \times \mathbf{E}] = [\boldsymbol{\nu} \times \mathbf{H}] = 0 & \text{on } \partial D, \end{cases} \quad (2)$$

subject to the Silver-Müller radiation condition:

$$\lim_{|x| \rightarrow \infty} |x|(\sqrt{\mu_m}(\mathbf{H} - \mathbf{H}^{\text{in}}) \times \hat{x} - \sqrt{\varepsilon_m}(\mathbf{E} - \mathbf{E}^{\text{in}})) = 0,$$

where  $\hat{x} = x/|x|$ . Here,  $[\boldsymbol{\nu} \times \mathbf{E}]$  and  $[\boldsymbol{\nu} \times \mathbf{H}]$  denote the jump of  $\boldsymbol{\nu} \times \mathbf{E}$  and  $\boldsymbol{\nu} \times \mathbf{H}$  along  $\partial D$ , namely,

$$[\boldsymbol{\nu} \times \mathbf{E}] = (\boldsymbol{\nu} \times \mathbf{E})|_+ - (\boldsymbol{\nu} \times \mathbf{E})|_-, \quad [\boldsymbol{\nu} \times \mathbf{H}] = (\boldsymbol{\nu} \times \mathbf{H})|_+ - (\boldsymbol{\nu} \times \mathbf{H})|_-.$$

**Proposition 2.1.** *If  $\mathcal{I}[\varepsilon_c] \neq 0$ , then problem (2) is well-posed. Moreover, if we denote by  $(\mathbf{E}, \mathbf{H})$  its unique solution, then  $(\mathbf{E}, \mathbf{H})|_D \in H(\text{curl}, D)$  and  $(\mathbf{E}, \mathbf{H})|_{\mathbb{R}^3 \setminus D} \in H_{\text{loc}}(\text{curl}, \mathbb{R}^3 \setminus \overline{D})$ .*

*Proof.* The well-posedness is addressed in [31, 15, 6]. □

### 2.3 Volume integral equation for the electric field

We now recall the well-known Lippmann-Schwinger equation [4] satisfied by the electric field for a non-magnetic particle:

$$\mathbf{E}(x) = \mathbf{E}^{\text{in}}(x) + \frac{\varepsilon_m - \varepsilon_c}{\varepsilon_m} \left(\frac{\omega}{c}\right)^2 \int_D \mathbf{\Gamma}^{\frac{\omega}{c}}(x, y) \mathbf{E}(y) dy, \quad x \in \mathbb{R}^3, \quad (3)$$

where  $\mathbf{\Gamma}^{\frac{\omega}{c}}$ , the dyadic Green's function, is defined in Appendix A.2. Consequently, it suffices to derive an approximation for the electric field  $\mathbf{E}$  inside  $D$  and insert it in the right-hand side of (3) to obtain an expression for  $\mathbf{E}$  for all points outside.

**Lemma 2.1.** *Using the dyadic Green's function, one can express the incident field as*

$$\mathbf{E}^{\text{in}}(x) = \mathbf{\Gamma}^{\frac{\omega}{c}}(x, s) \mathbf{p}, \quad x \in \mathbb{R}^3. \quad (4)$$

*Proof.* For  $x \in \mathbb{R}^3$ , the incident fields solves

$$\nabla \times \nabla \times \mathbf{E}^{\text{in}}(x) - \left(\frac{\omega}{c}\right)^2 \mathbf{E}^{\text{in}}(x) = \mathbf{p} \delta_s(x)$$

so

$$\mathbf{E}^{\text{in}}(x) = (\mathbf{\Gamma}^{\frac{\omega}{c}} * \mathbf{p} \delta_s)(x) = \int_{\mathbb{R}^3} \mathbf{\Gamma}^{\frac{\omega}{c}}(x - y) \mathbf{p} \delta_s(y) dy = \mathbf{\Gamma}^{\frac{\omega}{c}}(x, s) \mathbf{p}.$$

□

**Definition 2.1.** *We denote the contrast  $\gamma$  by*

$$\gamma(\omega) = \frac{\varepsilon_m}{\varepsilon_m - \varepsilon_c(\omega)}.$$

Restricting equation (3) to  $D$  yields the following integral representation.

**Proposition 2.2.** *The electric field inside the particle satisfies the volume integral equation:*

$$\left(\gamma(\omega)I - \mathcal{T}_D^{\frac{\omega}{c}}\right) \mathbf{E} = \gamma(\omega) \mathbf{E}^{\text{in}} \quad \text{in } D, \quad (5)$$

where  $\mathcal{T}_D^{\frac{\omega}{c}} : L^2(D, \mathbb{R}^3) \rightarrow L^2(D, \mathbb{R}^3)$  is a singular integral operator of the Calderón-Zygmund type, defined in Appendix B.1.

*Proof.* See [13, Chapter 9] or [14]. □

$\mathcal{T}_D^{\frac{\omega}{c}}$  is neither compact nor self-adjoint on  $L^2(D)$  so diagonalising  $\mathcal{T}_D^{\frac{\omega}{c}}$  directly to find a modal expansion for  $\mathbf{E}$  is not possible. However, when  $\omega = 0$  (in the *static regime*), the operator  $\mathcal{T}_D^0$  has nice properties.

## 2.4 The static regime

### 2.4.1 Main results

**Lemma 2.2.** *In the static limit, when  $\omega\delta c^{-1} \rightarrow 0$ , (5) becomes:*

$$(\gamma(\omega)I - \mathcal{T}_D^0) \mathbf{E} = \gamma(\omega)\nabla U^{in} + \mathcal{O}(\omega\delta c^{-1}) \quad \text{in } D, \quad (6)$$

where  $\nabla U^{in}$  is the orthogonal projection of  $\mathbf{E}^{in}$  on the space  $\mathbf{W} \subset L^2(D, \mathbb{R}^3)$  ( $\mathbf{W}$  is the space of gradient of harmonic functions, see Lemma 2.3 for more details).

The goal of this section is to show that the following theorems hold:

**Theorem 2.1** (Spectral decomposition of  $\mathcal{T}_D^0$ ). *The set of eigenvalues  $(\gamma_n)_{n \in \mathbb{N}}$  of  $\mathcal{T}_D^0|_{\mathbf{W}}$  is discrete, and the associated eigenfunctions  $(\mathbf{e}_n)_{n \in \mathbb{N}}$  form an orthonormal basis of  $\mathbf{W}$ . Hence we have:*

$$\mathcal{T}_D^0|_{\mathbf{W}} = \sum_n \gamma_n \langle \mathbf{e}_n, \cdot \rangle_{L^2} \mathbf{e}_n,$$

and  $\gamma_n \in ]0, 1]$ .

**Theorem 2.2** (Modal decomposition in the static regime). *When  $\omega\delta c^{-1} \rightarrow 0$ ,*

$$\mathbf{E} = \sum_n \frac{\gamma(\omega)}{\gamma(\omega) - \gamma_n} \langle \mathbf{E}^{in}, \mathbf{e}_n \rangle_{L^2} \mathbf{e}_n \quad \text{in } D.$$

*Proof.* Theorem 2.2 is a direct consequence of Theorem 2.1 and equation (6). □

### 2.4.2 Proof of Theorem 2.1

Recall the following orthogonal decomposition.

**Lemma 2.3.** *We have*

$$L^2(D, \mathbb{R}^3) = \nabla H_0^1(D) \oplus \mathbf{H}(\text{div } 0, D) \oplus \mathbf{W},$$

where  $\mathbf{H}(\text{div } 0, D)$  is the space of divergence free  $L^2$  vector fields and  $\mathbf{W}$  is the space of gradients of harmonic  $H^1$ -functions.

We start with the following result from [15]:

**Proposition 2.3.**  $\mathcal{T}_D^0$  is a bounded self-adjoint map on  $L^2(D, \mathbb{R}^3)$  with  $\nabla H_0^1(D)$ ,  $\mathbf{H}(\text{div } 0, D)$  and  $\mathbf{W}$  as invariant subspaces. On  $\nabla H_0^1(D)$ ,  $\mathcal{T}_D^0[\mathbf{e}] = \mathbf{e}$ , on  $\mathbf{H}(\text{div } 0, D)$ ,  $\mathcal{T}_D^0[\mathbf{e}] = 0$  and on  $\mathbf{W}$ :

$$\boldsymbol{\nu} \cdot \mathcal{T}_D^0[\mathbf{e}] = \left( \frac{1}{2}I + \mathcal{K}_D^* \right) [\mathbf{e} \cdot \boldsymbol{\nu}] \text{ on } \partial D,$$

where  $\mathcal{K}_D^*$  is the Neumann-Poincaré operator defined in Section B.1.

*Proof.* The proof can be found in [18, 15]. □

From this, it immediately follows that the following corollary holds.

**Corollary 2.1.** *Let  $\gamma \neq 1$ . Let  $\mathbf{e} \neq 0$  be such that*

$$\gamma \mathbf{e} - \mathcal{T}_D^0[\mathbf{e}] = 0 \quad \text{in } D.$$

Then,

$$\begin{aligned}
\mathbf{e} &\in \mathbf{W}, \\
\nabla \cdot \mathbf{e} &= 0 && \text{in } D, \\
\gamma \mathbf{e} &= \nabla \mathcal{S}_D[\mathbf{e} \cdot \boldsymbol{\nu}] && \text{in } D, \\
\gamma \mathbf{e} \cdot \boldsymbol{\nu} &= \left( \frac{1}{2} I + \mathcal{K}_D^* \right) [\mathbf{e} \cdot \boldsymbol{\nu}] && \text{on } \partial D,
\end{aligned}$$

where  $\mathcal{S}_D$  is the single-layer potential defined in Section B.1.

**Remark 2.1.** It has been shown in [1, 6] that the plasmonic resonances are linked to the eigenvalues of the Neumann-Poincaré operator. Corollary 2.1 shows that the volume integral approach and the surface integral approach are consistent with one another.

We now have all the tools to prove Theorem 2.1. With Proposition B.2 one can build a basis of eigenvectors  $(\phi_n)_{n \in \mathbb{N}}$  for  $\mathcal{K}_D^*$  in  $\mathcal{H}^*(\partial D)$ , associated to the eigenvalues  $(\lambda_n)_{n \in \mathbb{N}}$ . Using Corollary 2.1 one can see that the gradient of the single layer potentials of this eigenbasis is a basis of  $\mathbf{W}$  constituted of eigenvectors of  $\mathcal{T}_D^0$  with eigenvalues  $\gamma_n = \frac{1}{2} + \lambda_n$ ,  $n \in \mathbb{N}$ .

## 2.5 Dynamic regime

Our strategy is to work in a perturbative regime where  $\omega \delta c^{-1}$  is small but not zero, where we compute the eigenvalues of  $\mathcal{T}_D^{\frac{\omega}{c}}$  as perturbations of eigenvalues of  $\mathcal{T}_D^0$  and use the eigenvectors of  $\mathcal{T}_D^0$  to build a modal basis.

Recall that when the ratio of the size of the particle over the wavelength in the surrounding medium  $\omega \delta c^{-1}$  is small but not zero, the governing equation for the electric field is

$$\left( \gamma(\omega) I - \mathcal{T}_D^{\frac{\omega}{c}} \right) \mathbf{E} = \gamma(\omega) \mathbf{E}^{\text{in}} \quad \text{in } D.$$

In order to get a modal decomposition similar to the static case one, we need to study the eigenvalues of  $\mathcal{T}_D^{\frac{\omega}{c}}$  as perturbations of eigenvalues of  $\mathcal{T}_D^0$ . This was done in [5, Section 4] and we recall here the main results.

**Lemma 2.4.** Let  $\gamma_{n_0}$  be a simple eigenvalue for  $\mathcal{T}_D^0$ . Then, if  $|\omega \delta c^{-1}|$  is small enough, there exists a neighbourhood  $V \subset \mathbb{C}$  of  $\gamma_{n_0}$  such that  $\mathcal{T}_D^{\frac{\omega}{c}}$  has exactly one eigenvalue in  $V$  denoted  $\lambda_{n_0}(\omega)$ .

**Proposition 2.4.** The following asymptotic formula for the perturbed eigenvalues holds:

$$\gamma_{n_0}(\omega) = \gamma_{n_0} + \left\langle \left( \mathcal{T}_D^{\frac{\omega}{c}} - \mathcal{T}_D^0 \right) \mathbf{e}_{n_0}, \mathbf{e}_{n_0} \right\rangle_{L^2(B, \mathbb{R}^3)} + o(\omega^2 \delta^2 c^{-2}), \quad (7)$$

where  $\mathbf{e}_{n_0} \in L^2(D, \mathbb{R}^3)$  is a unitary eigenvector for  $\mathcal{T}_D^0$  associated with  $\gamma_{n_0}$ .

**Remark 2.2.** In [5] the remainder is of order  $\omega \delta c^{-1}$  only. Nevertheless, it has been shown in [7], in the formalism of layer potentials and boundary integrals, that the perturbation of the eigenvalues is actually of order  $\omega^2 \delta^2 c^{-2}$ .

**Proposition 2.5.** Let  $\mathbf{g} \in L^2(D, \mathbb{R}^3)$ . If  $\mathbf{f} \in L^2(D, \mathbb{R}^3)$  is a solution of

$$\left( \gamma(\omega) I - \mathcal{T}_D^{\frac{\omega}{c}} \right) \mathbf{f} = \mathbf{g},$$

then

$$\begin{aligned}
\langle \mathbf{f}, \mathbf{e}_{n_0} \rangle_{L^2} &\sim \frac{\langle \mathbf{g}, \mathbf{e}_{n_0} \rangle_{L^2}}{\gamma(\omega) - \gamma_{n_0}(\omega)} \\
&\sim \frac{\langle \mathbf{g}, \mathbf{e}_{n_0} \rangle_{L^2}}{\gamma(\omega) - \gamma_{n_0} - \left\langle \left( \mathcal{T}_D^{\frac{\omega}{c}} - \mathcal{T}_D^0 \right) \mathbf{e}_{n_0}, \mathbf{e}_{n_0} \right\rangle_{L^2} + o(\omega^2 \delta^2 c^{-2})}.
\end{aligned}$$

Using the governing equation for the field inside the particle we immediately get a modal expansion inside the particle.

**Corollary 2.2** (Modal expansion in the dynamic case). *When  $\omega\delta c^{-1} \ll 1$ ,*

$$\mathbf{E} = \sum_n \frac{\gamma(\omega)}{\gamma(\omega) - \gamma_n(\omega)} \langle \mathbf{E}^{in}, \mathbf{e}_n \rangle_{L^2(D, \mathbb{R}^3)} \mathbf{e}_n + \mathcal{O}\left(\frac{\omega\delta}{c}\right) \quad \text{in } D$$

with

$$\gamma_n(\omega) = \gamma_n + \left\langle \left( \mathcal{T}_D^{\frac{\omega}{c}} - \mathcal{T}_D^0 \right) \mathbf{e}_n, \mathbf{e}_n \right\rangle_{L^2(D, \mathbb{R}^3)} + o(\omega^2 \delta^2 c^{-2}).$$

The Lippmann-Schwinger equation allows us to extend the expansion outside the particle:

**Corollary 2.3** (Modal expansion outside the particle).

$$\mathbf{E}(x) = \mathbf{E}^{in}(x) + \left(\frac{\omega}{c}\right)^2 \sum_n \frac{1}{\gamma(\omega) - \gamma_n(\omega)} \langle \mathbf{E}^{in}, \mathbf{e}_n \rangle_{L^2} \int_D \mathbf{\Gamma}^{\frac{\omega}{c}}(x, y) \mathbf{e}_n(y) dy + \mathcal{O}\left(\frac{\omega\delta}{c}\right), \quad x \in \mathbb{R}^3 \setminus \bar{D}.$$

Before we can study the electric field in the time domain, we need to determine the localisation of the poles. In the next section we study the roots of the equations:

$$\begin{aligned} \gamma(\omega) &= \gamma_n && \text{(static regime),} \\ \gamma(\omega) &= \gamma_n(\omega) && \text{(dynamic regime).} \end{aligned}$$

**Remark 2.3.** (Equivalent method with boundary integral operators). *Corollary 2.1 shows that there is a strong link between the surface integral operator and the volume integral operator. A similar type of modal expansion can be obtained using layer potential operators. The layer potential operators describing the scattering problem act on  $L_T^2(\partial D)$  the space of vector fields in  $L^2$  tangential to the particle. The vectorial equivalent of the Neumann-Poincaré operator that appears cannot be symmetrised as easily as  $\mathcal{K}_D^*$  in the scalar case. One has to perform a Helmholtz type decomposition on the  $L_T^2(\partial D)$  vector fields, and the symmetrisation is only valid on one of the subspaces, see [1, section 4] for more details. The computation of the perturbed spectrum can be carried through, as in [7]. Nevertheless, the computations are quite technical, making the result more difficult to read and interpret.*

### 3 Static and dynamic plasmonic resonances

**Definition 3.1.** *A static plasmonic resonance can occur when  $\gamma(\omega) \in \sigma(\mathcal{T}_D^0)$ . A dynamic plasmonic resonance can occur when the contrast parameter  $\gamma(\omega) \in \sigma\left(\mathcal{T}_D^{\frac{\omega}{c}}\right)$ .*

In what follows we use the lower-case character  $\omega$  for real frequencies and the upper-case character  $\Omega$  for complex frequencies.

#### 3.1 Static plasmonic resonances

**Proposition 3.1.** *Assuming  $\varepsilon_m \in \mathbb{R}^+$  and using the Drude model (1) the static plasmonic resonances have an explicit formula:  $\Omega_n = \Omega'_n + i\Omega''_n$  such that for  $n \geq 1$ ,*

$$\begin{aligned} \Omega'_n &= \sqrt{\frac{\omega_p^2}{1 - \frac{\gamma_n - 1}{\gamma_n} \frac{\varepsilon_m}{\varepsilon_0}} - \frac{1}{4\mathbb{T}^2}}, \\ \Omega''_n &= -\frac{1}{2\mathbb{T}}. \end{aligned}$$

*The static plasmonic resonances all lie in the lower part of the complex plane and their real parts are bounded.*

*Proof.* Let  $\gamma_n \in \sigma(\mathcal{T}_D^0)$ :

$$\begin{aligned} \frac{\varepsilon_m}{\varepsilon_m - \varepsilon_c(\omega)} = \gamma_n &\Leftrightarrow \varepsilon_c(\omega) = \varepsilon_m \left(1 - \frac{1}{\gamma_n}\right) \\ &\Leftrightarrow \frac{\omega_p^2}{\omega^2 + i\omega T^{-1}} = 1 - \frac{\gamma_n - 1}{\gamma_n} \frac{\varepsilon_m}{\varepsilon_0} \\ &\Leftrightarrow \Omega'^2 - \Omega''^2 + 2i\Omega'\Omega'' + i\Omega'T^{-1} - \Omega''T^{-1} = \frac{\omega_p^2}{1 - \frac{\gamma_n - 1}{\gamma_n} \frac{\varepsilon_m}{\varepsilon_0}}, \end{aligned}$$

which gives the result.  $\square$

In the non-restrictive case where the particle is embedded in vacuum,  $\varepsilon_m = \varepsilon_0$ , the expression for the static resonant frequency simplifies to

$$\begin{aligned} \Omega'_n &= \sqrt{\gamma_n \omega_p^2 - \frac{1}{4T^2}}, \\ \Omega''_n &= -\frac{1}{2T}. \end{aligned}$$

### 3.2 First approximation of dynamic plasmonic resonances

Finding the frequencies  $\omega$  at which a dynamic plasmonic resonance can occur is the non-linear eigenvalue problem of

$$\text{finding } \omega \text{ s.t. } \gamma(\omega) \in \sigma\left(\mathcal{T}_D^{\frac{\omega}{c}}\right). \quad (8)$$

In the case where the particle is small compared to the wavelength of the incoming electric field (i.e. in a regime where  $\delta k_m \ll 1$ ), it is possible to get an approximation of the dynamic plasmonic resonant frequencies  $(\Omega_n(\delta))_{n \in \mathbb{N}}$  solutions of (8) by the following perturbative approach:

1. Compute the eigenvalues  $\gamma_n$  of  $\mathcal{T}_D^0$  on  $\mathbf{W}$ :

$$\sigma(\mathcal{T}_D^0) = (\gamma_n)_{n \in \mathbb{N}},$$

as well as the associated eigenfunctions  $\mathbf{e}_n \in \mathbf{W}$ .

2. Compute  $\Omega_n$  solutions of

$$\gamma(\Omega_n) = \gamma_n.$$

3. Compute the eigenvalues of the perturbed operator

$$\gamma_n(\delta\Omega_n) = \gamma_n + \left\langle \left( \mathcal{T}_D^{\frac{\Omega_n}{c}} - \mathcal{T}_D^0 \right) \mathbf{e}_n, \mathbf{e}_n \right\rangle_{L^2}.$$

4. Compute  $\Omega_n(\delta)$  solutions of

$$\gamma(\Omega_n(\delta)) = \gamma_n(\delta\Omega_n).$$

5. Iterate the process if higher accuracy is needed.

### 3.3 Convergence analysis

This method can be seen as the first algorithmic step of a fixed-point method to determine the correct plasmonic resonant frequency. In the following proposition we show that the algorithm is convergent. Assume, without loss of generality, that  $\varepsilon_m = \varepsilon_0$ , and that  $\varepsilon_c$  is given by the Drude model (1).

**Lemma 3.1.** *The function  $\gamma : \Omega \mapsto \left(1 - \frac{\varepsilon_c(\Omega)}{\varepsilon_0}\right)^{-1}$  has an inverse*

$$\gamma^{-1} : \left\{ \Omega \in \mathbb{C}, \Re(\Omega) > \frac{1}{4(\omega_p \mathbb{T})^2}, \Im(\Omega) \leq 0 \right\} \longrightarrow \{ \Omega \in \mathbb{C}, \Re(\Omega) > 0, \Im(\Omega) \leq 0 \}.$$

Moreover, for any  $\gamma_{max} > \frac{1}{4(\omega_p \mathbb{T})^2}$  and for  $\Re(\Omega) \in \left[ \frac{1}{4(\omega_p \mathbb{T})^2}, \gamma_{max} \right]$ ,

$$0 < \Re(\gamma^{-1}(\Omega)) \leq \sqrt{\gamma_{max}} \omega_p.$$

*Proof.* We first write:

$$\frac{\varepsilon_0}{\varepsilon_0 - \varepsilon_c(\Omega)} = Z \iff \Omega^2 + i\Omega \mathbb{T}^{-1} = \omega_p^2 Z \iff \Omega = \frac{-i\mathbb{T}^{-1} \pm \sqrt{-\mathbb{T}^{-2} + 4\omega_p^2 Z}}{2}.$$

Define  $\gamma^{-1} := \Omega \mapsto \frac{1}{2} \left( -i\mathbb{T}^{-1} + \sqrt{-\mathbb{T}^{-2} + 4\omega_p^2 \Omega} \right)$ . □

**Proposition 3.2.** *Let  $(\Omega_n^{(k)})_{k \in \mathbb{N}}$  be defined by*

$$\begin{aligned} \Omega_n^{(0)} &= \gamma^{-1}(\gamma_n) \\ \Omega_n^{(k+1)} &= \gamma^{-1} \left( \gamma_n + \left\langle \left( \mathcal{T}_D^{\frac{\Omega_n^{(k)}}{c}} - \mathcal{T}_D^0 \right) \mathbf{e}_n, \mathbf{e}_n \right\rangle_{L^2} \right). \end{aligned}$$

If  $\delta \frac{\omega_p}{c} \leq \frac{1}{2}$  and  $\gamma_n - (\omega_p \mathbb{T})^{-2} > 2 \left( \frac{\delta \gamma^{-1}(\gamma_n)}{c} \right)^2 = 2 \left( \frac{\delta \Omega_n^{(0)}}{c} \right)^2$  then the sequence is convergent and we denote  $\Omega_n(\delta)$  its limit.

**Remark 3.1.** *Note that  $\gamma_n$  is a sequence in  $]0, 1]^{\mathbb{N}}$  converging to  $1/2$ . In two dimensions, the convergence is algebraic for circles, geometric for ellipses. For an ellipse of semi-axes  $a, b$  with  $a < b$ , the eigenvalues are simply:*

$$\gamma_n = \frac{1}{2} + \frac{1}{2} \left( \frac{a-b}{a+b} \right)^n.$$

*In three dimensions, the formula for the eigenvalues is more complicated and uses elliptic integrals but the behaviour is the same. See [2, Chapter 2] for more details.*

**Remark 3.2.** *These convergence bounds are not sharp, they are just sufficient conditions. They show that the range of validity of the method is much better than the static approach. The first condition gives a condition on the size of the particle for convergence. The second condition is a condition on both the size and the shape. The smallest eigenvalue of the operator  $\mathcal{T}_D^0$  needs to be bounded away from zero.*

*Indeed, to study plasmonic resonances of a metallic nanoparticle in the static regime at optical frequencies, one has to take  $\omega \in [2, 5] \cdot 10^{15}$  Hz, and has to consider a particle whose size is negligible compared to the wavelength of the surrounding medium  $\delta\omega/c \ll 1$ . This means that the particle has*

a radius of a couple of nanometers at most (see for instance [11, 23]). Here, since  $\omega_p = 2 \cdot 10^{15}$  Hz, the first condition is satisfied for particles up to several hundreds of nanometers. The second condition depends on the shape of the particle. Assuming that  $\gamma_n$  is somewhere between 0.2 and 0.8 (so not too close to 0, true for any ellipse with eccentricity  $e \leq 0.8$  for instance) and  $\Omega_n^{(0)}$  is in the optical frequencies  $[\omega_p, 2\omega_p]$  then the second criterion is valid for  $(\delta\omega_p/c)^2 \leq 1/20$  which is  $\delta \lesssim 100$  nm.

*Proof.* Define  $F(\omega) := \gamma^{-1} \left( \gamma_n + \left\langle \left( \mathcal{T}_D^{\frac{\omega}{c}} - \mathcal{T}_D^0 \right) \mathbf{e}_n, \mathbf{e}_n \right\rangle_{L^2} \right)$  and  $G(\omega) := \gamma_n + \left\langle \left( \mathcal{T}_D^{\frac{\omega}{c}} - \mathcal{T}_D^0 \right) \mathbf{e}_n, \mathbf{e}_n \right\rangle_{L^2}$ .  $\gamma^{-1}$  is meromorphic and  $(\gamma^{-1})'(z) = \omega_p^2 (-T^{-2} + 4\omega_p^2 z)^{-1/2}$ . Noting that  $G(\omega) - \gamma_n \sim \delta^2 \omega^2 / c^2$ , we have that  $G'(\omega) \sim 2\delta^2 \omega / c^2$ . Therefore we have:

$$\begin{aligned} F'(\omega) &= G'(\omega) (\gamma^{-1})'(G(\omega)) \\ &= \frac{\omega_p G'(\omega)}{2\sqrt{G(\omega) - (2T\omega_p)^{-2}}} \\ &\leq \frac{\delta\omega_p}{c}. \end{aligned}$$

Now, taking  $\Re(\omega) \leq N\omega_p$  for any  $N \in \mathbb{N}^*$  and  $\delta\omega_p/c < 1 - 1/N^2$  we get that  $|G(\omega)| \leq 1 + N^2 \delta^2 \omega_p^2 / c^2 \leq N^2$ . Using Lemma 3.1 with  $\gamma_{max} = N^2$  we have  $|F(\omega)| \leq N\omega_p$ . So we have the stability of the region  $\{\omega \in \mathbb{C}, \Re(\omega) \leq N\omega_p, \Im(\omega) < 0\}$  and the fact that  $F$  is a contraction. Hence the convergence of the sequence holds.  $\square$

**Remark 3.3.** *The assumption that  $\varepsilon_c(\omega)$  follows a simple Drude model is not restrictive. One could take a more sophisticated model for the permittivity of the metallic nanoparticle, such as the one given in [27]. The computations to find sufficient conditions for the convergence of the method would be a lot heavier, but essentially  $\varepsilon_c(\omega)$  would have a similar behaviour and one can check numerically that the iterative process converges for particles that satisfy similar size conditions.*

### 3.4 Range of validity

For a particle  $D = z + \delta B$ , the perturbed quasi-static method to describe the field at a frequency  $\omega$  in the optical frequencies is pertinent if the following two conditions are met:

1. The sufficient conditions of Proposition 3.2 on  $\delta$  are met, for accurate computations of the retarded quasi-static resonances. This could be written as  $\delta \leq \delta_{max}(B)$ .
2. The frequency  $\omega$  corresponds to a perturbed quasi-static regime  $\omega \lesssim c/\delta$ .

We can then define, for each particle shape  $B$ , a maximum radius  $\delta_{max}(B)$  and for each radius a maximum frequency

$$\begin{aligned} ]0, \delta_{max}(B)[ &\longrightarrow \mathbb{R}^+ \\ \delta &\longmapsto R(\delta) := \frac{c}{\delta}. \end{aligned} \tag{9}$$

The fact that the sequence is converging can be interpreted this way. If the operator  $\mathcal{T}^0$ 's eigenvalues are not too small, the corresponding static resonant frequencies are not too big. And if the sequence above is convergent, then the perturbed resonant frequencies are all in a range where the behaviour of the electromagnetic field is described by the perturbative approach.

**Corollary 3.1** (Localisation of the poles in the complex plane). *Under the convergence conditions given in proposition 3.2, the plasmonic resonances are all located in the following bounded region of the lower complex half plane. More over, all resonances are in a bounded region near the origin: if  $\delta\omega_p c^{-1} < 1/2$  then  $|\Omega_n(\delta)| < R(\delta)$ .*

*Proof.* Assume that the sequence

$$\begin{aligned}\Omega_n^{(0)} &= \gamma^{-1}(\gamma_n) \\ \Omega_n^{(k+1)} &= \gamma^{-1} \left( \gamma_n + \left\langle \left( \mathcal{T}_D^{\frac{\Omega_n^{(k)}}{c}} - \mathcal{T}_D^0 \right) \mathbf{e}_n, \mathbf{e}_n \right\rangle_{L^2} \right).\end{aligned}$$

is convergent (i.e. under the conditions of proposition 3.2). Then the convergence is geometric and

$$\begin{aligned}|\Omega_n(\delta)| &\leq |\Omega_n^0| + |\Omega_n(\delta) - \Omega_n^0| \\ &\leq |\Omega_n^0| + \sum_k |\Omega_n^{(k+1)} - \Omega_n^{(k)}| \\ &\leq |\Omega_n^0| + \sum_k \left( \frac{\delta \omega_p}{c} \right)^{2k}\end{aligned}$$

□

## 4 Modal expansion for the electric field

We can now give a pole expansion of the scattered electric field.

**Lemma 4.1.** *In the quasi-static limit, when  $\omega\delta/c \ll 1$ , the following modal decomposition for the electric field inside the particle holds:*

$$\mathbf{E} = \sum_n \frac{\gamma(\omega)}{\gamma(\omega) - \gamma_n(\Omega_n(\delta)) + o(\omega^2\delta^2c^{-2})} \langle \mathbf{E}^{in}, \mathbf{e}_n \rangle_{L^2} \mathbf{e}_n + \mathcal{O}\left(\frac{\omega\delta}{c}\right) \text{ in } D,$$

where  $(\mathbf{e}_n)_{n \in \mathbb{N}}$  is an orthonormal basis of  $\mathbf{W}$  for the usual  $L^2(D, \mathbb{R}^3)$  scalar product, and  $\gamma_n(\Omega_n(\delta))$  are the second order approximation eigenvalues of  $\mathcal{T}_D^{\frac{\omega}{c}}$  at the dynamic plasmonic resonant frequency  $\omega = \Omega_n(\delta)$  on  $\mathbf{W}$  associated with the eigenvectors  $\mathbf{e}_n$ .

**Proposition 4.1.** *In the quasi-static regime, when  $\omega\delta/c \ll 1$ , the following modal decomposition for the scattered field holds for  $x \in \mathbb{R}^3$ :*

$$\begin{aligned}\mathbf{E}^{sca}(x) &= \mathbf{E}(x) - \mathbf{E}^{in}(x) \\ &= \sum_n \frac{1}{\gamma(\omega) - \gamma_n(\Omega_n(\delta)) + o(\omega^2\delta^2c^{-2})} \langle \mathbf{E}^{in}, \mathbf{e}_n \rangle_{L^2} \left( \frac{\omega}{c} \right)^2 \int_D \Gamma^{\frac{\omega}{c}}(x, y) \mathbf{e}_n(y) dy + \mathcal{O}\left(\frac{\omega\delta}{c}\right).\end{aligned}\tag{10}$$

*Proof.* Substituting the field inside  $D$  from Lemma 4.1 into (3) concludes the proof. □

**Remark 4.1.** *Note that this method of constructing the modes by defining a modal basis inside the resonator for  $L^2(D)$  and expanding the modes outside of the resonators leads to non-diverging eigenmodes for  $\omega \in \mathbb{R}$ .*

One can see that the function  $\omega \mapsto \mathbf{E}^{sca}(\omega)$  is meromorphic and has simple poles at  $\omega = \Omega_n(\delta)$ . One can define the so-called *quasi-normal modes* as the excitation independent part of the residue of  $\mathbf{E}^{sca}$  at each pole.

**Definition 4.1.** *The quasi-normal modes are the functions defined by*

$$\mathbf{E}_n(x) = \begin{cases} \mathbf{e}_n(x), & x \in D, \\ \left( \frac{\Omega_n(\delta)}{c} \right)^2 \int_D \Gamma^{\frac{\Omega_n(\delta)}{c}}(x, y) \mathbf{e}_n(y) dy, & x \in \mathbb{R}^3 \setminus \bar{D}. \end{cases}\tag{11}$$

**Remark 4.2.** *About the completeness of quasi-normal modes. Lemma 4.1 states that  $\{\mathbf{E}_n\}$  is a basis of the subspace  $\mathbf{W} \subset L^2(D)$ . It is a complete basis for the space of solutions of the Maxwell equations in  $D$ .*

**Remark 4.3** (About the infinite sum). *In this article we do not need to consider a far-field approximation. The closer the source to the scatterer, the larger the number of modes the sum needs to take into account. Nevertheless, the sum is not really infinite as in most papers only the first couple of modes are considered.*

Since  $\Omega_n(\delta)$  has a negative imaginary part the quasi-normal modes do not belong to  $L^2$  and diverge exponentially as  $|x| \rightarrow \infty$ .

In the next section we will show that in the time domain, the scattered field can be expressed using a resonance expansion without any divergence problems by using a residue theorem.

## 5 Time domain approximation

Given a wideband signal  $\hat{f} : t \mapsto \hat{f}(t) \in C_0^\infty([0, C_1])$ , for  $C_1 > 0$ , we want to express the time domain response of the electric field to an oscillating dipole placed at a source point  $s$ . We assume that most of the energy of the excitation is concentrated in the low frequencies (i.e., in frequencies corresponding to wavelengths that are much larger than the particle, such that the response of the particle can be studied via the perturbed quasi-static theory). This means that for a fixed  $\delta$  we can pick  $\eta \ll 1$  and  $\rho$  such that

$$\int_{\mathbb{R} \setminus [-\rho, \rho]} |f(\omega)|^2 d\omega \leq \eta,$$

$$\frac{\rho\delta}{c} \leq 1,$$

where  $f : \omega \mapsto f(\omega)$  is the Fourier transform of  $\hat{f}$ . The goal of this section is to establish a resonance expansion for the low-frequency part of the scattered electric field in the time domain. Introduce, for  $\rho > 0$ , the truncated inverse Fourier transform of the scattered field  $\mathbf{E}^{\text{sca}}$  given by

$$P_\rho \left[ \widehat{\mathbf{E}}^{\text{sca}} \right] (x, t) = \int_{-\rho}^{\rho} \mathbf{E}^{\text{sca}}(x, \omega) e^{-i\omega t} d\omega.$$

Recall that  $z$  is the centre of the resonator and  $\delta$  its radius. Let us define

$$t_0^\pm(s, x) := \frac{1}{c} (|s - z| + |x - z| \pm 2\delta),$$

the time it takes to the signal to reach first the scatterer and then observation point  $x$ . The term  $\pm 2\delta/c$  accounts for the maximal timespan spent inside the particle.

**Proposition 5.1.** *The incident field has the following form in the time domain:*

$$\begin{aligned} \widehat{\mathbf{E}}^{\text{in}}(x, t) &= \int_{\mathbb{R}} \mathbf{\Gamma}^{\frac{\omega}{c}}(x, s) \mathbf{p} f(\omega) e^{-i\omega t} d\omega \\ &= \frac{\hat{f}(t - |x - s|/c)}{4\pi|x - s|} \mathbf{p} + c^2 \mathbf{D}_x^2 \frac{\hat{f}''(t - |x - s|/c)}{4\pi|x - s|} \mathbf{p}. \end{aligned}$$

*Proof.* See Appendix A.3. □

**Theorem 5.1.** For a particle of size  $\delta \leq \delta_{max}$ , the scattered field has the following form in the time domain for  $x \in \mathbb{R}^3 \setminus \overline{D}$ :

$$P_\rho \left[ \widehat{\mathbf{E}}^{sca} \right] (x, t) = \begin{cases} \mathcal{O} (R(\delta)^{-N}), & t \leq t_0^-, \\ 2\pi i \sum_{n=1}^{\infty} \text{Res}(\mathbf{E}^{sca}(x, \Omega), \Omega_n(\delta)) e^{-i\Omega_n(\delta)t} + \mathcal{O} \left( \frac{1}{t} R(\delta)^{-N} \right), & t \geq t_0^+, \end{cases} \quad (12)$$

for  $N$  a large positive integer with  $\Omega_l(\delta)$  the plasmonic resonant frequencies of the particle, given by Proposition 3.2 and  $R(\delta)$  is given by equation (9).

**Remark 5.1.** The resonant frequencies  $\Omega_l(\delta)$  have negative imaginary parts, so Theorem 5.1 expresses the scattered field as the sum of decaying oscillating fields. The imaginary part of  $\Omega_l(\delta)$  accounts for absorption losses in the particle as well as radiative losses.

**Remark 5.2** (About the remainder  $R(\delta)$ ). Since for a particle of finite size  $\delta$  our expansion only holds for a range of frequencies  $|\omega| \leq R(\delta)$ , we cannot compute the full inverse Fourier transform and we have a remainder that depends on the maximum frequency that we can use. Since that maximum frequency  $R(\delta)$  behaves as  $c/\delta$  we can see that the remainder gets arbitrarily small for small particles. For a completely point-like particle one would get a zero remainder.

**Remark 5.3.** If we had access to the full inverse Fourier transform of the field, of course, since the inverse Fourier transform of a function which is analytic in the upper-half plane is causal we would find that in the case  $t \leq (|s - z| + |x - z| - 2\delta)/c$ ,  $\mathbf{E}^{sca}(x, t) = 0$ . Nevertheless, our method only works for a truncated low frequency estimate of the scattered field, hence the arbitrarily small remainder.

*Proof.* We start by studying the time domain response of a single mode to a causal excitation at the source point  $s$ . Therefore, according to Proposition 4.1 we need to compute the contribution  $\Xi_n$  of each mode  $\mathbf{e}_n$ , that is,

$$\begin{aligned} & \int_{-\rho}^{\rho} \Xi_n(x, \omega) e^{-i\omega t} d\omega \\ & := \int_{-\rho}^{\rho} \left[ \frac{1}{\gamma(\omega) - \gamma_n(\delta\Omega_n) + o(\omega^2\delta^2c^{-2})} \langle \Gamma^{\frac{\omega}{c}}(\cdot, s) \mathbf{p}f(\omega), \mathbf{e}_n \rangle_{L^2} \left( \frac{\omega}{c} \right)^2 \int_D \Gamma^{\frac{\omega}{c}}(x, y) \mathbf{e}_n(y) dy \right] e^{-i\omega t} d\omega. \end{aligned}$$

We need the following lemma:

**Lemma 5.1.**

$$\Gamma^{\frac{\omega}{c}}(x, z) = -e^{i\frac{\omega}{c}|x-z|} \frac{\mathbf{A}(x, z, \omega/c)}{4\pi|x-z|},$$

where  $\mathbf{A}$  is given in Appendix A.2, and behaves like a polynomial in  $\omega$ .

One can then write:

$$\begin{aligned} & \langle \Gamma^{\frac{\omega}{c}}(\cdot, s) \mathbf{p}f(\omega), \mathbf{e}_n \rangle_{L^2} \left( \frac{\omega}{c} \right)^2 \int_D \Gamma^{\frac{\omega}{c}}(x, y) \mathbf{e}_n(y) dy = \\ & \left( \frac{\omega}{c} \right)^2 f(\omega) \int_{D \times D} e^{i\frac{\omega}{c}(|x-y|+|s-v|)} \frac{\mathbf{A}(x, y, \omega/c) \mathbf{e}_n(y)}{4\pi|x-y|} \mathbf{e}_n(v) \cdot \frac{\mathbf{A}(s, v, \omega/c) \mathbf{p}}{4\pi|s-v|} dv dy. \end{aligned} \quad (13)$$

Now we want to apply the residue theorem to get an asymptotic expansion in the time domain. Note that:

$$\int_{-\rho}^{\rho} \Xi_n(x, \omega) e^{-i\omega t} d\omega = \oint_{\mathcal{C}^\pm} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega - \int_{\mathcal{C}_\rho^\pm} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega,$$

where the integration contour  $\mathcal{C}_\rho^\pm$  is a semicircular arc of radius  $\rho$  in the upper (+) or lower (-) half-plane, and  $\mathcal{C}^\pm$  is the closed contour  $\mathcal{C}^\pm = \mathcal{C}_\rho^\pm \cup [-\rho, \rho]$ . The integral on the closed contour is the main contribution to the scattered field by the mode  $\mathbf{e}_n$  and can be computed using the residue theorem to get, for  $\rho \geq \Re[\Omega_n(\delta)]$ ,

$$\begin{aligned} \oint_{\mathcal{C}^+} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega &= 0, \\ \oint_{\mathcal{C}^-} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega &= 2\pi i \text{Res}(\Xi_n(x, \Omega) e^{-i\Omega t}, \Omega_n(\delta)). \end{aligned}$$

Since  $\Omega_n(\delta)$  is a simple pole of  $\omega \mapsto \frac{\gamma(\omega)}{\gamma(\omega) - \gamma_n(\omega\delta)}$  we can write:

$$\oint_{\mathcal{C}^-} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega = 2\pi i \text{Res}(\Xi_n(x, \Omega), \Omega_n(\delta)) e^{-i\Omega_n(\delta)t}.$$

To compute the integrals on the semi-circle, we introduce:

$$\mathbf{B}_n(y, v, \Omega) = \frac{\Omega^2}{\gamma(\Omega) - \gamma_n(\delta)} \frac{\mathbf{A}(x, y, \Omega/c) \mathbf{e}_n(y) \mathbf{e}_n(v) \cdot \mathbf{A}(s, v, \Omega/c) \mathbf{p}}{16c^2\pi^2|x-y||s-v|} \quad (y, v) \in D^2.$$

Note that  $\mathbf{B}_n(\cdot, \cdot, \Omega)$  behaves like a polynomial in  $\Omega$  when  $|\Omega| \rightarrow \infty$ . Given the regularity of the input signal  $\hat{f} \in C_0^\infty([0, C_1])$ , the Paley-Wiener theorem [32, p.161] ensures decays properties of its Fourier transform at infinity. For all  $N \in \mathbb{N}^*$  there exists a positive constant  $C_N$  such that for all  $\Omega \in \mathbb{C}$

$$|f(\Omega)| \leq C_N(1 + |\Omega|)^{-N} e^{C_1|\Im(\Omega)|}.$$

We now re-write the integrals on the semi-circle

$$\int_{\mathcal{C}_\rho^\pm} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega = \int_{\mathcal{C}_\rho^\pm} f(\Omega) \int_{D \times D} \mathbf{B}_n(y, v, \Omega) e^{i\Omega(\frac{|x-y|+|s-v|}{c}-t)} dv dy d\Omega.$$

Two cases arise.

**Case 1:** For  $0 < t < t_0^-$ , i.e., when the signal emitted at  $s$  has not reached the observation point  $x$ , we choose the upper-half integration contour  $\mathcal{C}^+$ . Transforming into polar coordinates,  $\Omega = \rho e^{i\theta}$  for  $\theta \in [0, \pi]$ , we get:

$$\left| e^{i\Omega(\frac{|x-y|+|s-v|}{c}-t)} \right| \leq e^{(t_0^- - t)\Im(\Omega)} \quad \forall (y, v) \in D^2,$$

and

$$\begin{aligned} \left| \int_{\mathcal{C}_\rho^+} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega \right| &\leq \int_0^\pi |f(\rho e^{i\theta})| e^{-\rho(t_0^- - t)\sin\theta} \int_{D \times D} |\mathbf{B}_n(y, v, \Omega)| dv dy d\theta, \\ &\leq \rho C_N (1 + \rho)^{-N} \max_{\theta \in [0, \pi]} \|\mathbf{B}_n(\cdot, \cdot, \rho e^{i\theta})\|_{L^\infty(D \times D)} \pi \frac{1 - e^{\rho[C_1 - (t_0^- - t)]}}{\rho(t_0^- - t - C_1)}, \end{aligned}$$

where we used that for  $\theta \in [0, \pi/2]$ , we have  $\sin\theta \geq 2\theta/\pi \geq 0$  and  $-\cos\theta \leq -1 + 2\theta/\pi$ . The usual way to go forward from here is to take the limit  $\rho \rightarrow \infty$ , and get that the limit of the integral on the semi-circle is zero. However, we work in the quasi-static approximation here, and our modal expansion is not uniformly valid for all frequencies. So we have to work with a fixed maximum frequency  $\rho$ . However, the maximum frequency  $\rho$  depends on the size of the particle via the hypothesis  $\rho \leq R(\delta)$ .

Since  $N$  can be taken arbitrarily large and that  $\mathbf{B}_n$  behaves like a polynomial in  $\rho$  whose degree does not depend on  $n$ , we get that, uniformly in  $n \in \mathbb{N}$ :

$$\left| \int_{\mathcal{C}_\rho^\pm} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega \right| = \mathcal{O} \left( \frac{1}{t_0 - t - C_1} R(\delta)^{-N} \right).$$

Of course if one has to consider the full inverse Fourier transform of the scattered electromagnetic field, by causality, one should expect the limit to be zero. However, one would need high-frequency estimates of the electromagnetic field, as well as a modal decomposition that is uniformly valid for all frequencies. Since our modal expansion is only valid for a limited range of frequencies we get an error bound that is arbitrarily small if the particle is arbitrarily small, but not rigorously zero.

**Case 2:** For  $t > t_0^+$ , we choose the lower-half integration contour  $\mathcal{C}^-$ . Transforming into polar coordinates,  $\Omega = \rho e^{i\theta}$  for  $\theta \in [\pi, 2\pi]$ , we get

$$\left| e^{i\Omega \left( \frac{|x-y|+|s-v|}{c} - t \right)} \right| \leq e^{(t-t_0^+) \Im(\Omega)} \quad \forall (y, v) \in D^2,$$

and

$$\begin{aligned} \left| \int_{\mathcal{C}_\rho^\pm} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega \right| &\leq \int_\pi^{2\pi} |f(\rho e^{i\theta})| e^{\rho(t-t_0^+) \sin \theta} \int_{D \times D} |\mathbf{B}_n(y, v, \Omega)| ds dv d\theta, \\ &\leq \rho C_N (1 + \rho)^{-N} \max_{\theta \in [0, \pi]} \|\mathbf{B}_n(\cdot, \cdot, \rho e^{i\theta})\|_{L^\infty(D \times D)} \pi \frac{1 - e^{\rho(C_1 - (t-t_0^+))}}{\rho(C_1 - (t-t_0^+))}, \end{aligned}$$

Exactly as in Case 1, we cannot take the limit  $\rho \rightarrow \infty$ . However, the maximum frequency  $\rho$  depends on the size of the particle via the hypothesis  $\rho \leq R(\delta)$ . Since  $R(\delta) \rightarrow \infty$  ( $\delta \rightarrow 0$ ), using the fact that  $N$  can be taken arbitrarily large and that  $\mathbf{B}_n$  behaves like a polynomial in  $\rho$  whose degree does not depend on  $n$ , we get that, uniformly in  $n \in \mathbb{N}$ .

$$\left| \int_{\mathcal{C}_\rho^\pm} \Xi_n(x, \Omega) e^{-i\Omega t} d\Omega \right| = \mathcal{O} \left( \frac{1}{t} R(\delta)^{-N} \right).$$

The result of Theorem 5.1 is obtained by summing the contribution of all the modes.  $\square$

## 6 Concluding Remarks

In this paper we have shown, through the spectral analysis of singular integral operators of the Calderón-Zygmund type, that the electromagnetic field scattered by a small particle constituted of a dispersive media could be expanded in an orthogonal basis inside the particle, and that the modal expansion could be extended outside the particle by the Lippmann-Schwinger equation (Corollary 2.3). We have shown that the scattered electromagnetic field is a meromorphic function of the frequency, and that the poles are the solutions to a non-linear eigenvalue problem, and that they are all located in a bounded region of the complex lower-half plane. We have shown that the so-called *quasi-normal modes* that appear in the physics literature [20] can be defined from this expansion but it is not the only way [16]. We have then given a resonance expansion in the time domain for the low frequency part of the scattered electromagnetic field, as a sum of complex exponential (decaying in time) fields, using only results from surface integral operator theory. We have also shown with elementary complex analysis that there is no divergence problem at infinity once we are in the time domain.

## A Fundamental solutions

### A.1 Green's function

**Definition A.1.** Denote by  $\Gamma^k$  the outgoing Green function for the homogeneous medium, i.e., the unique solution of the Helmholtz operator:

$$(\Delta + k^2)\Gamma^k(\cdot, y) = \delta_y(\cdot) \quad \text{in } \mathbb{R}^3$$

satisfying the Sommerfeld radiation condition. In dimension three,  $\Gamma^k$  is given by

$$\Gamma^k(x, y) = -\frac{e^{ik|x-y|}}{4\pi|x-y|}, \quad x, y \in \mathbb{R}^3.$$

### A.2 Dyadic Green's function

**Definition A.2.** Using the scalar function  $\Gamma^k$  defined in Appendix A.1 as the fundamental solution to the Helmholtz equation, we now define the matrix-valued function, referred to as the Dyadic Green's function, as

$$\mathbf{\Gamma}^k(x, y) = -\Gamma^k(x, y)\mathbf{I} - \frac{1}{k^2}\mathbf{D}_x^2\Gamma^k(x, y), \quad x, y \in \mathbb{R}^3, \quad (14)$$

where  $\mathbf{I}$  is the  $3 \times 3$  identity matrix and  $\mathbf{D}_x^2$  denotes the Hessian.

**Proposition A.1.**  $\mathbf{\Gamma}^{k_m}$  is a Green's function for the background electric problem, i.e., it satisfies:

$$\nabla \times \nabla \times \mathbf{\Gamma}^{k_m} - k_m^2 \mathbf{\Gamma}^{k_m} = \delta_y \mathbf{I} \quad \text{in } \mathbb{R}^3.$$

**Lemma A.1.** The matrix  $\mathbf{A}(x, z, \omega) = (A)_{p,q=1}^3$  introduced in Lemma 5.1 is with entries

$$\begin{aligned} A_{pp} &= \frac{1}{\omega^2|x-z|^4} \left[ -3(x_p - z_p)^2 + |x-z|^2 + 3i\omega(x_p - z_p)^2|x-z| + \omega^2(x_p - z_p)^2|x-z|^2 \right. \\ &\quad \left. - i\omega|x-z|^3 - \omega^2|x-z|^4 \right], \\ A_{pq} &= \frac{1}{|x-z|^4} (x_p - z_p)(x_q - z_q) \left[ -3 + 3i\omega|x-z| + \omega^2|x-z|^2 \right], \quad \text{for } p \neq q. \end{aligned}$$

### A.3 In the time domain

In this subsection we compute the inverse Fourier transform of the Green function. For a source located in  $s$ :

$$\begin{aligned} \widehat{\mathbf{E}}^{\text{in}}(x, t) &= \int_{\mathbb{R}} \mathbf{\Gamma}^{\frac{\omega}{c}}(x, s) \mathbf{p} f(\omega) e^{-i\omega t} d\omega \\ &= - \int_{\mathbb{R}} \mathbf{\Gamma}^{\frac{\omega}{c}}(x, s) \mathbf{I} \mathbf{p} f(\omega) e^{-i\omega t} d\omega - \int_{\mathbb{R}} \frac{c^2}{\omega^2} \mathbf{D}_x^2 \mathbf{\Gamma}^{\frac{\omega}{c}}(x, s) \mathbf{p} f(\omega) e^{-i\omega t} d\omega \\ &= \int_{\mathbb{R}} \frac{e^{i\omega|x-s|/c}}{4\pi|x-s|} f(\omega) e^{-i\omega t} d\omega \mathbf{p} + c^2 \mathbf{D}_x^2 \int_{\mathbb{R}} \frac{e^{i\omega|x-s|/c}}{4\pi|x-s|} \frac{f(\omega)}{\omega^2} e^{-i\omega t} d\omega \mathbf{p} \\ &= \int_{\mathbb{R}} \frac{e^{-i\omega[t-|x-s|/c]}}{4\pi|x-s|} f(\omega) d\omega \mathbf{p} + c^2 \mathbf{D}_x^2 \int_{\mathbb{R}} \frac{e^{-i\omega[t-|x-s|/c]}}{4\pi|x-s|} g(\omega) d\omega \mathbf{p} \\ &= \frac{\widehat{f}(t - |x-s|/c)}{4\pi|x-s|} \mathbf{p} + c^2 \mathbf{D}_x^2 \frac{1}{4\pi|x-s|} \widehat{g}(t - |x-s|/c) \mathbf{p} \\ &= \frac{\widehat{f}(t - |x-s|/c)}{4\pi|x-s|} \mathbf{p} + c^2 \mathbf{D}_x^2 \frac{\widehat{f}''(t - |x-s|/c)}{4\pi|x-s|} \mathbf{p}, \end{aligned}$$

where  $g(\omega) := f(\omega)/\omega^2$  and  $\widehat{f}''$  is the second derivative of  $\widehat{f}$ . Note that  $\widehat{f}$  and  $\widehat{f}''$  vanish for negative arguments, which is physically meaningful since for  $t < |x - s|/c$  the direct signal has not reached the observation point yet.

## B Properties of integral operators

### B.1 Definitions

We start by defining a singular integral operator, sometimes known as the *magnetization integral operator* [18].

**Definition B.1.** *Introduce*

$$\mathcal{T}_D^k : L^2(D, \mathbb{R}^3) \longrightarrow L^2(D, \mathbb{R}^3) \\ \mathbf{f} \longmapsto -k^2 \int_D \Gamma^k(\cdot, y) \mathbf{f}(y) dy - \nabla \nabla \cdot \int_D \Gamma^k(\cdot, y) \mathbf{f}(s) dy.$$

We also need to define the classical *single-layer potential operator*

**Definition B.2.**

$$\mathcal{S}_D^k : L^2(\partial D) \longrightarrow L^2(\partial D) \\ \varphi \longmapsto \int_{\partial D} \Gamma^k(\cdot, y) \varphi(y) d\sigma(y),$$

where  $\sigma$  is the Lebesgue measure on  $\partial D$ .

We also recall the definition of the *Neumann-Poincaré operator*

**Definition B.3.**

$$\mathcal{K}_D^k : L^2(\partial D) \longrightarrow L^2(\partial D) \\ \phi \longmapsto \int_{\partial D} \frac{\partial \Gamma^k(x, y)}{\partial \nu(x)} \phi(y) d\sigma(y), \quad x \in \partial D.$$

When  $k = 0$ , we just write  $\mathcal{S}_D$  and  $\mathcal{K}_D^*$  for simplicity. The following lemmas can be found in [3, Chapter 2]:

**Lemma B.1.** *The single-layer potential  $\mathcal{S}_D$  is a unitary operator in  $H^{-1/2}(\partial D)$  in three dimensions.*

**Lemma B.2.** *Calderón identity*

$$\mathcal{S}_D \mathcal{K}_D^* = \mathcal{K}_D \mathcal{S}_D \quad \text{in } H^{-1/2}(\partial D).$$

For more on the symmetrisation property see also [19].

### B.2 Spectral properties of $\mathcal{K}_D^*$

**Proposition B.1.** *If  $\partial D$  has  $\mathcal{C}^{1,\alpha}$  regularity for  $\alpha > 0$  then  $\mathcal{K}_D^*$  is a compact operator.*

*Proof.* See [3, Chapter 2] □

In this whole paper we stand under the  $\mathcal{C}^{1,\alpha}$  regularity hypothesis for  $\partial D$ . We now recall the *Plemelj symmetrisation principle*:

**Proposition B.2.** Let  $\mathcal{H}^*(\partial D)$  be the Hilbert space  $H^{-1/2}(\partial D)$  equipped with the following inner product:

$$\langle u, v \rangle_{\mathcal{H}^*} = -\langle u, \mathcal{S}_D[v] \rangle_{-1/2, 1/2}.$$

Then  $K_D^*$  is a self-adjoint operator on  $\mathcal{H}^*$ .

*Proof.* This is a direct consequence of Lemmas B.2 and B.1.  $\square$

**Theorem B.1** (Diagonalisation of  $\mathcal{K}_D^*$ ).  $\mathcal{K}_D^*$  has a discrete set of real eigenvalues  $\lambda_n$  with associated eigenvector  $\phi_n$  and

$$\mathcal{K}_D^*[\phi] = \sum_n \lambda_n \langle \phi, \phi_n \rangle_{\mathcal{H}^*} \phi_n, \quad \phi \in H^{-1/2}(\partial D)$$

with  $\lambda_n \in ]-1/2, 1/2]$ ,  $\lambda_0 = 1/2$  and  $|\lambda_n| \rightarrow 0$  as  $n \rightarrow +\infty$ .

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