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Mathematical Analysis of Plasmonic Nanoparticles: The Scalar Case

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Abstract

Localized surface plasmons are charge density oscillations confined to metallic nanoparticles. Excitation of localized surface plasmons by an electromagnetic field at an incident wavelength where resonance occurs results in a strong light scattering and an enhancement of the local electromagnetic fields. This paper is devoted to the mathematical modeling of plasmonic nanoparticles. Its aim is fourfold: (1) to mathematically define the notion of plasmonic resonance and to analyze the shift and broadening of the plasmon resonance with changes in size and shape of the nanoparticles; (2) to study the scattering and absorption enhancements by plasmon resonant nanoparticles and express them in terms of the polarization tensor of the nanoparticle; (3) to derive optimal bounds on the enhancement factors; (4) to show, by analyzing the imaginary part of the Green function, that one can achieve super-resolution and super-focusing using plasmonic nanoparticles. For simplicity, the Helmholtz equation is used to model electromagnetic wave propagation.

Contents

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1. Introduction

Plasmon resonant nanoparticles have unique capabilities of enhancing the brightness of light and confining strong electromagnetic fields [40]. A thriving interest in optical studies of plasmon resonant nanoparticles is due to their recently proposed use as labels in molecular biology [27]. New types of cancer diagnostic nanoparticles are constantly being developed. Nanoparticles are also being used in thermotherapy as nanometric heat-generators that can be activated remotely by external electromagnetic fields [17].

According to the quasi-static approximation for small particles, the surface plasmon resonance peak occurs when the particle's polarizability is maximized. Plasmon resonances in nanoparticles can be treated at the quasi-static limit as an eigenvalue problem for the Neumann–Poincaré integral operator, which leads to direct calculation of resonance values of permittivity and optimal design of nanoparticles that resonate at specified frequencies [2,6,25,34,35]. At this limit, they are size-independent. However, as the particle size increases, they are determined from scattering and absorption blow up and become size-dependent. This was experimentally observed, for instance, in [26,38,41].

In [6], we have provided a rigorous mathematical framework for localized surface plasmon resonances. We have considered the full Maxwell equations. Using layer potential techniques, we have derived the quasi-static limits of the electromagnetic fields in the presence of nanoparticles. We have proved that the quasi-static limits are uniformly valid with respect to the nanoparticle's bulk electron relaxation rate. We have introduced localized plasmonic resonances as the eigenvalues of the Neumann–Poincaré operator associated with the nanoparticle. We have described a general model for the permittivity and permeability of nanoparticles as functions of the frequency and rigorously justified the quasi-static approximation for surface plasmon resonances.

In this paper, we first prove that, as the particle size increases and crosses its critical value for dipolar approximation which is justified in [6], the plasmonic resonances become size-dependent. The resonance condition is determined from absorption and scattering blow up and depends on the shape, size and electromagnetic parameters of both the nanoparticle and the surrounding material. Then, we precisely quantify the scattering absorption enhancements in plasmonic nanoparticles. We derive new bounds on the enhancement factors given the volume and electromagnetic parameters of the nanoparticles. At the quasi-static limit, we prove that the averages over the orientation of scattering and extinction cross-sections of a randomly oriented nanoparticle are given in terms of the imaginary part of the polarization tensor. Moreover, we show that the polarization tensor blows up at plasmonic resonances and derive bounds for the absorption and scattering cross-sections. We also prove the blow-up of the first-order scattering coefficients at plasmonic resonances. The concept of scattering coefficients was introduced in [9] for scalar wave propagation problems and in [10] for the full Maxwell equations, rendering a powerful and efficient tool for the classification of the nanoparticle shapes. Using such a concept, we have explained in [3] the experimental results reported in [16]. Finally, we consider the super-resolution phenomenon in plasmonic nanoparticles. Super-resolution is meant to cross the barrier of diffraction limits by reducing the focal spot size. This resolution limit applies only to light that has propagated for a distance substantially larger than its wavelength [18,19]. Super-focusing is the counterpart of super-resolution. It is a concept for waves to be confined to a length scale significantly smaller than the diffraction limit of the focused waves. The super-focusing phenomenon is being intensively investigated in the field of nanophotonics as a possible technique to focus electromagnetic radiation in a region of order of a few nanometers beyond the diffraction limit of light and thereby causing an extraordinary enhancement of the electromagnetic fields. In [12,13], a rigorous mathematical theory is developed to explain the super-resolution phenomenon in microstructures with high contrast material around the source point. Such microstructures act like arrays of subwavelength sensors. A key ingredient is the calculation of the resonances and the Green function in the microstructure. By following the methodology developed in [12,13], we show in this paper that one can achieve super-resolution using plasmonic nanoparticles as well.

The paper is organized as follows. In Section 2 we introduce a layer potential formulation for plasmonic resonances and derive asymptotic formulas for the plasmonic resonances and the near- and far-fields in terms of the size. In Section 3 we consider the case of multiple plasmonic nanoparticles. Section 4 is devoted to the study of the scattering and absorption enhancements. We also clarify the connection between the blow up of the scattering frequencies and the plasmonic resonances. The scattering coefficients are simply the Fourier coefficients of the scattering amplitude [9,10]. In Section 5 we investigate the behavior of the scattering coefficients at the plasmonic resonances. In Section 6 we prove that using plasmonic nanoparticles one can achieve super resolution imaging. “Appendix A” is devoted to the derivation of asymptotic expansions with respect to the frequency of some boundary integral operators associated with the Helmholtz equation and a single particle. These results are generalized to the case of multiple particles in

"Appendix B". In "Appendix C" we provide the technical modifications needed in order to study the shift in the plasmon resonance in the two-dimensional case. In "Appendix D" we prove useful sum rules for the polarization tensor.

2. Layer Potential Formulation for Plasmonic Resonances

2.1. Problem formulation and some basic results

We consider the scattering problem of a time-harmonic wave incident on a plasmonic nanoparticle. For simplicity, we use the Helmholtz equation instead of the full Maxwell equations. The homogeneous medium is characterized by electric permittivity ε_m and magnetic permeability μ_m, while the particle occupying a bounded and simply connected domain $D \Subset \mathbb{R}^3$ (the two-dimensional case is treated in "Appendix C") of class $\mathcal{C}^{1,\alpha}$ for some $0 < \alpha < 1$ is characterized by electric permittivity ε_c and magnetic permeability μ_c, both of which may depend on the frequency. Assume that $\Re\mu_c < 0$, $\Im\mu_c > 0$, $\Im\varepsilon_c > 0$, and define

$$k_m = \omega\sqrt{\varepsilon_m\mu_m}, \quad k_c = \omega\sqrt{\varepsilon_c\mu_c},$$

and

$$\varepsilon_D = \varepsilon_m\chi(\mathbb{R}^3\backslash\bar{D}) + \varepsilon_c\chi(\bar{D}), \quad \mu_D = \varepsilon_m\chi(\mathbb{R}^3\backslash\bar{D}) + \varepsilon_c\chi(D),$$

where χ denotes the characteristic function. Let $u^i(x) = e^{ik_m d\cdot x}$ be the incident wave. Here, ω is the frequency and d is the unit incidence direction. Throughout this paper, we assume that ε_m and μ_m are real and strictly positive and that $\Im k_c > 0$.

Using dimensionless quantities, we assume that the particle D has size of order one and also that the following condition holds:

Condition 1. *We assume that the numbers $\varepsilon_m, \mu_m, \varepsilon_c, \mu_c$ are dimensionless and are of order one. In addition, $\Im\mu_c = o(1)$. We also assume that ω is dimensionless and is of order $o(1)$.*

It is worth emphasizing that in this section the variable ω refers to the ratio between the size of the particle and the incident wavelength. For real plasmonic nanoparticles made of noble metals such as silver and gold, their electric permittivity is only negative over a small range of frequencies in the optical regime. This is also the frequency range in which Condition 1 holds and plasmonic resonance occurs. For the frequencies that are beyond that range, especially those near the origin, we shall assume that ε_c and μ_c are constant there. This assumption avoids complicated discussion on the dispersive property of electromagnetic parameters in that regime, and enables us to focus on the interesting frequency range when plasmonic resonance occurs. We also note that $\omega = o(1)$ implies that the plamsmonic nanoparticles have a size much smaller than the incident wavelength. This is the case when plamsonic resonance occurs.

The scattering problem can be modeled by the following Helmholtz equation:

$$\begin{cases} \nabla \cdot \dfrac{1}{\mu_D} \nabla u + \omega^2 \varepsilon_D u = 0 \quad \text{in } \mathbb{R}^3 \backslash \partial D, \\ u_+ - u_- = 0 \quad \text{on } \partial D, \\ \dfrac{1}{\mu_m} \dfrac{\partial u}{\partial \nu}\Big|_+ - \dfrac{1}{\mu_c} \dfrac{\partial u}{\partial \nu}\Big|_- = 0 \quad \text{on } \partial D, \\ u^s := u - u^i \ \text{ satisfies the Sommerfeld radiation condition.} \end{cases} \tag{2.1}$$

Here, $\partial/\partial\nu$ denotes the normal derivative and the Sommerfeld radiation condition can be expressed in dimension $d = 2, 3$, as follows:

$$\left| \frac{\partial u}{\partial |x|} - i k_m u \right| \leqq C|x|^{-(d+1)/2}$$

as $|x| \to +\infty$ for some constant C independent of x.

The model problem (2.1) is referred to as the transverse magnetic case. Note that all the results of this paper hold true in the transverse electric case where ε_D and μ_D are interchanged.

Let

$$F_1(x) = -u^i(x) = -e^{ik_m d \cdot x},$$

$$F_2(x) = -\frac{1}{\mu_m} \frac{\partial u^i}{\partial \nu}(x) = -\frac{i}{\mu_m} k_m e^{ik_m d \cdot x} d \cdot \nu(x),$$

with $\nu(x)$ being the outward normal at $x \in \partial D$. Let G be the Green function for the Helmholtz operator $\Delta + k^2$ satisfying the Sommerfeld radiation condition. In dimension three, G is given by

$$G(x, y, k) = -\frac{e^{ik|x-y|}}{4\pi |x - y|}.$$

By using the following single-layer potential and the Neumann–Poincaré integral operator

$$\mathcal{S}_D^k[\psi](x) = \int_{\partial D} G(x, y, k)\psi(y) d\sigma(y), \qquad x \in \mathbb{R}^3,$$

$$(\mathcal{K}_D^k)^*[\psi](x) = \int_{\partial D} \frac{\partial G(x, y, k)}{\partial \nu(x)} \psi(y) d\sigma(y), \quad x \in \partial D,$$

we can represent solution u in the following form:

$$u(x) = \begin{cases} u^i + \mathcal{S}_D^{k_m}[\psi], & x \in \mathbb{R}^3 \backslash \bar{D}, \\ \mathcal{S}_D^{k_c}[\phi], & x \in D, \end{cases} \tag{2.2}$$

where $\psi, \phi \in H^{-\frac{1}{2}}(\partial D)$ satisfy the following system of integral equations on ∂D [7]:

$$\begin{cases} \mathcal{S}_D^{k_m}[\psi] - \mathcal{S}_D^{k_c}[\phi] = F_1, \\ \frac{1}{\mu_m}\big(\frac{1}{2}Id + \left(\mathcal{K}_D^{k_m}\right)^*\big)[\psi] + \frac{1}{\mu_c}\big(\frac{1}{2}Id - \left(\mathcal{K}_D^{k_c}\right)^*\big)[\phi] = F_2, \end{cases} \tag{2.3}$$

where Id denotes the identity operator. In the sequel, we set $\mathcal{S}_D^0 = \mathcal{S}_D$.

We are interested in the scattering in the quasi-static regime, that is, for $\omega \ll 1$. Note that for ω small enough, $\mathcal{S}_D^{k_c}$ is invertible [7]. We have $\phi = (\mathcal{S}_D^{k_c})^{-1}\big(\mathcal{S}_D^{k_m}[\psi] - F_1\big)$, whereas the following equation holds for ψ:

$$\mathcal{A}_D(\omega)[\psi] = f, \tag{2.4}$$

where

$$\mathcal{A}_D(\omega) = \frac{1}{\mu_m}\left(\frac{1}{2}Id + \left(\mathcal{K}_D^{k_m}\right)^*\right) + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \left(\mathcal{K}_D^{k_c}\right)^*\right)(\mathcal{S}_D^{k_c})^{-1}\mathcal{S}_D^{k_m}, \tag{2.5}$$

$$f = F_2 + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \left(\mathcal{K}_D^{k_c}\right)^*\right)\left(\mathcal{S}_D^{k_c}\right)^{-1}[F_1]. \tag{2.6}$$

It is clear that

$$\begin{aligned} \mathcal{A}_D(0) = \mathcal{A}_{D,0} &= \frac{1}{\mu_m}\left(\frac{1}{2}Id + \mathcal{K}_D^*\right) + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}_D^*\right) \\ &= \left(\frac{1}{2\mu_m} + \frac{1}{2\mu_c}\right)Id - \left(\frac{1}{\mu_c} - \frac{1}{\mu_m}\right)\mathcal{K}_D^*, \end{aligned} \tag{2.7}$$

where the notation $\mathcal{K}_D^* = (\mathcal{K}_D^0)^*$ is used for simplicity.

We are interested in finding $\mathcal{A}_D(\omega)^{-1}$. We first recall some basic facts about the Neumann–Poincaré operator $\mathcal{K}_D^*$ [7,14,29,31].

Lemma 2.1. *(i) The following Calderón identity holds: $\mathcal{K}_D\mathcal{S}_D = \mathcal{S}_D\mathcal{K}_D^*$;*

(ii) The operator $\mathcal{K}_D^*$ is self-adjoint in the Hilbert space $H^{-\frac{1}{2}}(\partial D)$ equipped with the following inner product:

$$(u, v)_{\mathcal{H}^*} = -(u, \mathcal{S}_D[v])_{-\frac{1}{2},\frac{1}{2}} \tag{2.8}$$

with $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$ being the duality pairing between $H^{-\frac{1}{2}}(\partial D)$ and $H^{\frac{1}{2}}(\partial D)$, which is equivalent to the original one;

(iii) Let $\mathcal{H}^*(\partial D)$ be the space $H^{-\frac{1}{2}}(\partial D)$ with the new inner product. Let (λ_j, φ_j), $j = 0, 1, 2, \ldots$ be the eigenvalue and normalized eigenfunction pair of $\mathcal{K}_D^*$ in $\mathcal{H}^*(\partial D)$, then $\lambda_j \in (-\frac{1}{2}, \frac{1}{2}]$ and $\lambda_j \to 0$ as $j \to \infty$;

(iv) The following trace formula holds: for any $\psi \in \mathcal{H}^*(\partial D)$,

$$\left(-\frac{1}{2}Id + \mathcal{K}_D^*\right)[\psi] = \frac{\partial \mathcal{S}_D[\psi]}{\partial \nu}\Big|_-;$$

(v) The following representation formula holds: for any $\psi \in H^{-1/2}(\partial D)$,

$$\mathcal{K}_D^*[\psi] = \sum_{j=0}^{\infty} \lambda_j (\psi, \varphi_j)_{\mathcal{H}^*} \otimes \varphi_j.$$

It is clear that the following result holds:

Lemma 2.2. *Let $\mathcal{H}(\partial D)$ be the space $H^{\frac{1}{2}}(\partial D)$ equipped with the following equivalent inner product*

$$(u, v)_{\mathcal{H}} = \Big((-\mathcal{S}_D)^{-1}[u], v \Big)_{-\frac{1}{2},\frac{1}{2}}. \tag{2.9}$$

Then, $\mathcal{S}_D$ is an isometry between $\mathcal{H}^*(\partial D)$ and $\mathcal{H}(\partial D)$.

We now present other useful observations and basic results. The following holds:

Lemma 2.3. *(i) We have $(-\frac{1}{2}Id + \mathcal{K}_D^*)\mathcal{S}_D^{-1}[\chi(\partial D)] = 0$ with $\chi(\partial D)$ being the characteristic function of ∂D;*
(ii) Let $\lambda_0 = \frac{1}{2}$. Then the corresponding eigenspace has dimension one and is spanned by the function $\varphi_0 = c\mathcal{S}_D^{-1}[\chi(\partial D)]$ for some constant c such that $||\varphi_0||_{\mathcal{H}^*} = 1$;
(iii) Moreover, $\mathcal{H}^*(\partial D) = \mathcal{H}_0^*(\partial D) \oplus \{\mu\varphi_0,\ \mu \in \mathbb{C}\}$, where $\mathcal{H}_0^*(\partial D)$ is the zero mean subspace of $\mathcal{H}^*(\partial D)$ and $\varphi_j \in \mathcal{H}_0^*(\partial D)$ for $j \geqq 1$, that is, $(\varphi_j, \chi(\partial D))_{-\frac{1}{2},\frac{1}{2}} = 0$ for $j \geqq 1$. Here, $\{\varphi_j\}_j$ is the set of normalized eigenfunctions of $\mathcal{K}_D^*$.

From (2.7), it is easy to see that

$$\mathcal{A}_{D,0}[\psi] = \sum_{j=0}^{\infty} \tau_j (\psi, \varphi_j)_{\mathcal{H}^*} \varphi_j, \tag{2.10}$$

where

$$\tau_j = \frac{1}{2\mu_m} + \frac{1}{2\mu_c} - (\frac{1}{\mu_c} - \frac{1}{\mu_m})\lambda_j. \tag{2.11}$$

We now derive the asymptotic expansion of the operator $\mathcal{A}(\omega)$ as $\omega \to 0$. Using the asymptotic expansions in terms of k of the operators $\mathcal{S}_D^k$, $(\mathcal{S}_D^k)^{-1}$ and $(\mathcal{K}_D^k)^*$ proved in "Appendix A", we can obtain the following result:

Lemma 2.4. *As $\omega \to 0$, the operator $\mathcal{A}_D(\omega) : \mathcal{H}^*(\partial D) \to \mathcal{H}^*(\partial D)$ admits the asymptotic expansion*

$$\mathcal{A}_D(\omega) = \mathcal{A}_{D,0} + \omega^2 \mathcal{A}_{D,2} + O(\omega^3),$$

where

$$\mathcal{A}_{D,2} = (\varepsilon_m - \varepsilon_c)\mathcal{K}_{D,2} + \frac{\varepsilon_m \mu_m - \varepsilon_c \mu_c}{\mu_c} \left(\frac{1}{2} Id - \mathcal{K}_D^* \right) \mathcal{S}_D^{-1} \mathcal{S}_{D,2}. \tag{2.12}$$

Proof. Recall that

$$\mathcal{A}_D(\omega) = \frac{1}{\mu_m}\left(\frac{1}{2}Id + \left(\mathcal{K}_D^{k_m}\right)^*\right) + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \left(\mathcal{K}_D^{k_c}\right)^*\right)\left(\mathcal{S}_D^{k_c}\right)^{-1}\mathcal{S}_D^{k_m}. \tag{2.13}$$

By a straightforward calculation, it follows that

$$\begin{aligned}\left(\mathcal{S}_D^{k_c}\right)^{-1}\mathcal{S}_D^{k_m} &= Id + \omega\big(\sqrt{\varepsilon_c\mu_c}\mathcal{B}_{D,1}\mathcal{S}_D + \sqrt{\varepsilon_m\mu_m}\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\big) + \omega^2\big(\varepsilon_c\mu_c\mathcal{B}_{D,2}\mathcal{S}_D \\ &\quad + \sqrt{\varepsilon_c\mu_c\varepsilon_m\mu_m}\mathcal{B}_{D,1}\mathcal{S}_{D,1} + \varepsilon_m\mu_m\mathcal{S}_D^{-1}\mathcal{S}_{D,2}\big) + O(\omega^3), \\ &= Id + \omega\big(\sqrt{\varepsilon_m\mu_m} - \sqrt{\varepsilon_c\mu_c}\big)\mathcal{S}_D^{-1}\mathcal{S}_{D,1} + \omega^2\big(\left(\varepsilon_m\mu_m - \varepsilon_c\mu_c\right) \\ &\quad \mathcal{S}_D^{-1}\mathcal{S}_{D,2} + \sqrt{\varepsilon_c\mu_c}\left(\sqrt{\varepsilon_c\mu_c} - \sqrt{\varepsilon_m\mu_m}\right)\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\big) \\ &\quad + O(\omega^3),\end{aligned}$$

where $\mathcal{B}_{D,1}$ and $\mathcal{B}_{D,2}$ are defined by (A.5). Using the facts that

$$\left(\frac{1}{2}Id - \mathcal{K}_D^*\right)\mathcal{S}_D^{-1}\mathcal{S}_{D,1} = 0$$

and

$$\frac{1}{2}Id - (\mathcal{K}_D^k)^* = \left(\frac{1}{2}Id - \mathcal{K}_D^*\right) - k^2\mathcal{K}_{D,2} + O(k^3),$$

the lemma immediately follows. □

We regard $\mathcal{A}_D(\omega)$ as a perturbation to the operator $\mathcal{A}_{D,0}$ for small ω. Using standard perturbation theory [39], we can derive the perturbed eigenvalues and their associated eigenfunctions. For simplicity, we consider the case when λ_j is a simple eigenvalue of the operator $\mathcal{K}_D^*$.

We let

$$R_{jl} = \big(\mathcal{A}_{D,2}[\varphi_j], \varphi_l\big)_{\mathcal{H}^*}, \tag{2.14}$$

where $\mathcal{A}_{D,2}$ is defined by (2.12).

As ω goes to zero, the perturbed eigenvalue and eigenfunction have the following form:

$$\tau_j(\omega) = \tau_j + \omega^2\tau_{j,2} + O(\omega^3), \tag{2.15}$$

$$\varphi_j(\omega) = \varphi_j + \omega^2\varphi_{j,2} + O(\omega^3), \tag{2.16}$$

where

$$\tau_{j,2} = R_{jj}, \tag{2.17}$$

$$\varphi_{j,2} = \sum_{l\neq j}\frac{R_{jl}}{\left(\frac{1}{\mu_m} - \frac{1}{\mu_c}\right)(\lambda_j - \lambda_l)}\varphi_l. \tag{2.18}$$

2.2. *First-order correction to plasmonic resonances and field behavior at the plasmonic resonances*

We first introduce different notions of plasmonic resonance as follows:

Definition 1. (i) We say that ω is a plasmonic resonance if

$$|\tau_j(\omega)| \ll 1 \quad \text{and is locally minimal for some } j;$$

(ii) We say that ω is a quasi-static plasmonic resonance if $|\tau_j| \ll 1$ and is locally minimized for some j. Here, τ_j is defined by (2.11);

(iii) We say that ω is a first-order corrected quasi-static plasmonic resonance if $|\tau_j + \omega^2 \tau_{j,2}| \ll 1$ and is locally minimized for some j. Here, the correction term $\tau_{j,2}$ is defined by (2.17).

Note that quasi-static resonances are size independent and is therefore a zero-order approximation of the plasmonic resonance in terms of the particle size while the first-order corrected quasi-static plasmonic resonance depends on the size of the nanoparticle (or equivalently on ω in view of the non-dimensionalization adopted herein).

We are interested in solving the equation $\mathcal{A}_D(\omega)[\phi] = f$ when ω is close to the resonance frequencies, that is, when $\tau_j(\omega)$ is very small for some j's. In this case, the major part of the solution would be the contributions of the excited resonance modes $\varphi_j(\omega)$. We introduce the following definition:

Definition 2. We call $J \subset \mathbb{N}$ index set of resonance if τ_j's are close to zero when $j \in J$ and are bounded from below when $j \in J^c$. More precisely, we choose a threshold number $\eta_0 > 0$ independent of ω such that

$$|\tau_j| \geqq \eta_0 > 0 \quad \text{for } j \in J^c.$$

Remark 2.1. Note that for $j = 0$, we have $\tau_0 = 1/\mu_m$, which is of size one by our assumption. As a result, throughout this paper, we always exclude 0 from the index set of resonance J.

From now on, we shall use J as our index set of resonances. We assume throughout that the following conditions hold:

Condition 2. *Each eigenvalue λ_j for $j \in J$ is a simple eigenvalue of the operator $\mathcal{K}_D^*$.*

Condition 3. *Let*

$$\lambda = \frac{\mu_m + \mu_c}{2(\mu_m - \mu_c)}. \tag{2.19}$$

We assume that $\lambda \neq 0$ or equivalently, $\mu_c \neq -\mu_m$.

Condition 3, which is crucial to our analysis, implies that the set J is finite. Otherwise, infinity resonance modes may be excited and the problem becomes unstable. We refer to [23,24,37] for detailed discussion on this case.

Remark 2.2. Note that in the ideal case when $\Im \mu_c = 0$, we know that $\tau_j = 0$ if λ defined in (2.19) is equal to λ_j. This the usual definition in the quasi-static limiting case when the wavelength is infinite. In the case $\Im \mu_c \neq 0$ but $\Im \mu_c = o(1)$, one may neglect the imaginary part and still use the definition to find the resonance frequency. The drawback of this definition is that the resonance frequency is independent of the size of the particle. Now, with the asymptotic expansion (2.15), we may find ω, the resonance frequency, according to the criterion in Definition 1 (i) in a small neighborhood of the resonant frequency of the quasi-static limiting case. The difference of the two frequency yields the shift of resonance frequency with respect to size of the particle.

We now define the projection $P_J(\omega)$ such that

$$P_J(\omega)[\varphi_j(\omega)] = \begin{cases} \varphi_j(\omega), & j \in J, \\ 0, & j \in J^c. \end{cases}$$

In fact, we have

$$P_J(\omega) = \sum_{j \in J} P_j(\omega) = \sum_{j \in J} \frac{1}{2\pi i} \int_{\gamma_j} (\xi - \mathcal{A}_D(\omega))^{-1} d\xi, \tag{2.20}$$

where γ_j is a Jordan curve in the complex plane enclosing only the eigenvalue $\tau_j(\omega)$ among all the eigenvalues.

To obtain an explicit representation of $P_J(\omega)$, we consider the adjoint operator $\mathcal{A}_D(\omega)^*$. By a similar perturbation argument, we can obtain its perturbed eigenvalue and its eigenfunction, which have the following form:

$$\widetilde{\tau}_j(\omega) = \overline{\tau_j(\omega)}, \tag{2.21}$$

$$\widetilde{\varphi}_j(\omega) = \varphi_j + \omega^2 \widetilde{\varphi}_{j,2} + o(\omega^2). \tag{2.22}$$

Using the eigenfunctions $\widetilde{\varphi}_j(\omega)$, we can show that

$$P_J(\omega)[x] = \sum_{j \in J} \big(x, \widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*} \varphi_j(\omega). \tag{2.23}$$

Throughout this paper, for two Banach spaces X and Y, by $\mathcal{L}(X, Y)$ we denote the set of bounded linear operators from X into Y.

We are now ready to solve the equation $\mathcal{A}_D(\omega)[\psi] = f$. First, it is clear that

$$\psi = \mathcal{A}_D(\omega)^{-1}[f] = \sum_{j \in J} \frac{\big(f, \widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*}}{\tau_j(\omega)} + \mathcal{A}_D(\omega)^{-1}[P_{J^c}(\omega)[f]]. \tag{2.24}$$

The following lemma holds:

Lemma 2.5. *The norm $\|\mathcal{A}_D(\omega)^{-1} P_{J^c}(\omega)\|_{\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}^*(\partial D))}$ is uniformly bounded in ω for ω sufficiently small.*

Proof. Consider the operator

$$\mathcal{A}_D(\omega)|_{J^c} : P_{J^c}(\omega)\mathcal{H}^*(\partial D) \to P_{J^c}(\omega)\mathcal{H}^*(\partial D).$$

For ω small enough, we can show that $\operatorname{dist}(\sigma(\mathcal{A}_D(\omega)|_{J^c}), 0) \geqq \frac{\eta_0}{2}$, where $\sigma(\mathcal{A}_D(\omega)|_{J^c})$ is the discrete spectrum of $\mathcal{A}_D(\omega)|_{J^c}$. Then, it follows that

$$\begin{aligned}\|\mathcal{A}_D(\omega)^{-1}(P_{J^c}(\omega)f)\| &= \|\big(\mathcal{A}_D(\omega)|_{P_{J^c}}\big)^{-1}(P_{J^c}(\omega)f)\| \\ &\lesssim \frac{1}{\eta_0}\exp\left(\frac{C_1}{\eta_0^2}\right)\|P_{J^c}(\omega)f\|,\end{aligned}$$

where the notation $A \lesssim B$ means that $A \leqq CB$ for some constant C.

On the other hand,

$$\begin{aligned}P_J(\omega)f &= \sum_{j\in J}\big(f, \widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*}\varphi_j(\omega) = \sum_{j\in J}\big(f, \varphi_j + O(\omega)\big)_{\mathcal{H}^*}\big(\varphi_j + O(\omega)\big) \\ &= \sum_{j\in J}\big(f, \varphi_j\big)_{\mathcal{H}^*}\varphi_j(\omega) + O(\omega).\end{aligned}$$

Thus,

$$\|P_{J^c}(\omega)\| = \|(Id - P_J(\omega))\| \lesssim (1 + O(\omega)),$$

from which the desired result follows immediately. □

Second, we have the following asymptotic expansion of f given by (2.6) with respect to ω:

Lemma 2.6. *Let*

$$f_1 = -i\sqrt{\varepsilon_m\mu_m}e^{ik_m d\cdot z}\left(\frac{1}{\mu_m}[d\cdot\nu(x)] + \frac{1}{\mu_c}\big(\frac{1}{2}Id - \mathcal{K}_D^*\big)\mathcal{S}_D^{-1}[d\cdot(x-z)]\right)$$

and let z be the center of the domain D. In the space $\mathcal{H}^*(\partial D)$, as ω goes to zero, we have

$$f = \omega f_1 + O(\omega^2),$$

in the sense that, for ω small enough,

$$\|f - \omega f_1\|_{\mathcal{H}^*} \leqq C\omega^2$$

for some constant C independent of ω.

Proof. A direct calculation yields

$$\begin{aligned}f &= F_2 + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \left(\mathcal{K}_D^{k_c}\right)^*\right)(\mathcal{S}_D^{k_c})^{-1}[F_1] \\ &= -\omega\frac{i}{\mu_m}\sqrt{\varepsilon_m\mu_m}e^{ik_m d\cdot z}[d\cdot\nu(x)] + O(\omega^2)\end{aligned}$$

$$
\begin{aligned}
&+\frac{1}{\mu_c}\Big(\big(\frac{1}{2}Id-\mathcal{K}_D^*\big)\big((\mathcal{S}_D)^{-1}+\omega\mathcal{B}_{D,1}\big)+O(\omega^2)\Big)\\
&\Big[-e^{ik_m d\cdot z}\big(\chi(\partial D)+i\omega\sqrt{\varepsilon_m\mu_m}\,[d\cdot(x-z)]\big)+O(\omega^2)\Big]\\
=&-\frac{e^{ik_m d\cdot z}}{\mu_c}\left(\frac{1}{2}Id-\mathcal{K}_D^*\right)\mathcal{S}_D^{-1}[\chi(\partial D)]-\frac{\omega e^{ik_m d\cdot z}}{\mu_c}\left(\frac{1}{2}Id-\mathcal{K}_D^*\right)\mathcal{B}_{D,1}[\chi(\partial D)]\\
&-\omega i\sqrt{\varepsilon_m\mu_m}e^{ik_m d\cdot z}\left(\frac{1}{\mu_m}[d\cdot\nu(x)]+\frac{1}{\mu_c}\left(\frac{1}{2}Id-\mathcal{K}_D^*\right)\mathcal{S}_D^{-1}[d\cdot(x-z)]\right)\\
&+O(\omega^2)\\
=&-\omega i\sqrt{\varepsilon_m\mu_m}e^{ik_m d\cdot z}\left(\frac{1}{\mu_m}[d\cdot\nu(x)]+\frac{1}{\mu_c}\left(\frac{1}{2}Id-\mathcal{K}_D^*\right)\mathcal{S}_D^{-1}[d\cdot(x-z)]\right)\\
&+O(\omega^2),
\end{aligned}
$$

where we have made use of the facts that

$$\left(\frac{1}{2}Id-\mathcal{K}_D^*\right)\mathcal{S}_D^{-1}[\chi(\partial D)]=0$$

and

$$\mathcal{B}_{D,1}[\chi(\partial D)]=c\mathcal{S}_D^{-1}[\chi(\partial D)]$$

for some constant c; again, see “Appendix A”. □

Finally, we are ready to state our main result in this section.

Theorem 2.1. *Let D has size of order one. Under Conditions 1, 2, and 3 the scattered field $u^s=u-u^i$ due to a single plasmonic particle D has the following representation in the quasi-static regime:*

$$u^s=\mathcal{S}_D^{k_m}[\psi],$$

where

$$
\begin{aligned}
\psi&=\sum_{j\in J}\frac{\omega\big(f_1,\widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*}\varphi_j(\omega)}{\tau_j(\omega)}+O(\omega),\\
&=\sum_{j\in J}\frac{ik_m e^{ik_m d\cdot z}\big(d\cdot\nu(x),\varphi_j\big)_{\mathcal{H}^*}\varphi_j+O(\omega^2)}{\lambda-\lambda_j+O(\omega^2)}+O(\omega)
\end{aligned}
$$

with λ being given by (2.19).

Proof. We have

$$
\begin{aligned}
\psi&=\sum_{j\in J}\frac{\big(f,\widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*}\varphi_j(\omega)}{\tau_j(\omega)}+\mathcal{A}_D(\omega)^{-1}(P_{J^c}(\omega)f),\\
&=\sum_{j\in J}\frac{\omega\big(f_1,\varphi_j\big)_{\mathcal{H}^*}\varphi_j+O(\omega^2)}{\frac{1}{2\mu_m}+\frac{1}{2\mu_c}-\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)\lambda_j+O(\omega^2)}+O(\omega).
\end{aligned}
$$

We now compute $\big(f_1, \varphi_j\big)_{\mathcal{H}^*}$ with f_1 given in Lemma 2.6. We only need to show that

$$\left((\frac{1}{2} Id - \mathcal{K}_D^*) \mathcal{S}_D^{-1}[d \cdot (x-z)]), \varphi_j \right)_{\mathcal{H}^*} = (d \cdot \nu(x), \varphi_j)_{\mathcal{H}^*}. \tag{2.25}$$

Indeed, we have

$$\begin{aligned}
&\left(\left(\frac{1}{2} Id - \mathcal{K}_D^* \right) \mathcal{S}_D^{-1}\left[d \cdot (x-z)\right], \varphi_j \right)_{\mathcal{H}^*} \\
&= -\Big(\mathcal{S}_D^{-1}\left[d \cdot (x-z)\right], \big(\frac{1}{2} Id - \mathcal{K}_D\big) \mathcal{S}_D[\varphi_j] \Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= -\Big(\mathcal{S}_D^{-1}[d \cdot (x-z)], \mathcal{S}_D\big(\frac{1}{2} Id - \mathcal{K}_D^*\big)[\varphi_j] \Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= -\Big(d \cdot (x-z), \big(\frac{1}{2} Id - \mathcal{K}_D^*\big)[\varphi_j] \Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= -\Big(d \cdot (x-z), -\frac{\partial \mathcal{S}_D[\varphi_j]}{\partial \nu}\Big|_- \Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= \int_{\partial D} \frac{\partial [d \cdot (x-z)]}{\partial \nu} \mathcal{S}_D[\varphi_j] d\sigma \\
&\quad - \int_D \Big(\Delta[d \cdot (x-z)] \mathcal{S}_D[\varphi_j] - \Delta \mathcal{S}_D[\varphi_j][d \cdot (x-z)] \Big) dx \\
&= -\Big(d \cdot \nu(x), \varphi_j \Big)_{\mathcal{H}^*},
\end{aligned}$$

where we have used the fact that $\mathcal{S}_D[\varphi_j]$ is harmonic in D. This proves the desired identity and the rest of the theorem follows immediately. □

Corollary 2.1. *Assume the same conditions as in Theorem 2.1. Under the additional condition that*

$$\min_{j \in J} |\tau_j(\omega)| \gg \omega^3, \tag{2.26}$$

we have

$$\psi = \sum_{j \in J} \frac{i k_m e^{i k_m d \cdot z} \big(d \cdot \nu(x), \varphi_j\big)_{\mathcal{H}^*} \varphi_j + O(\omega^2)}{\lambda - \lambda_j + \omega^2 \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1} \tau_{j,2}} + O(\omega).$$

More generally, under the additional condition that

$$\min_{j \in J} \tau_j(\omega) \gg \omega^{m+1},$$

for some integer $m > 2$, we have

$$\psi = \sum_{j \in J} \frac{i k_m e^{i k_m d \cdot z} \big(d \cdot \nu(x), \varphi_j\big)_{\mathcal{H}^*} \varphi_j + O(\omega^2)}{\lambda - \lambda_j + \omega^2 \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1} \tau_{j,2} + \cdots + \omega^m \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1} \tau_{j,m}} + O(\omega).$$

Rescaling back to original dimensional variables, we suppose that the magnetic permeability μ_c of the nanoparticle is changing with respect to the operating angular frequency ω while that of the surrounding medium, μ_m, is independent of ω. Then we can write

$$\mu_c(\omega) = \mu'(\omega) + i\mu''(\omega). \tag{2.27}$$

Because of causality, the real and imaginary parts of μ_c obey the following Kramer–Kronig relations:

$$\begin{aligned} \mu''(\omega) &= -\frac{1}{\pi}\text{p.v.} \int_{-\infty}^{+\infty} \frac{1}{\omega - s}\mu'(s)ds, \\ \mu'(\omega) &= \frac{1}{\pi}\text{p.v.} \int_{-\infty}^{+\infty} \frac{1}{\omega - s}\mu''(s)ds, \end{aligned} \tag{2.28}$$

where p.v. stands for the principle value.

The magnetic permeability $\mu_c(\omega)$ can be described by the Drude model; see, for instance, [40]. We have

$$\mu_c(\omega) = \mu_0 \left(1 - F\frac{\omega^2}{\omega^2 - \omega_0^2 + i\tau^{-1}\omega}\right), \tag{2.29}$$

where $\tau > 0$ is the nanoparticle's bulk electron relaxation rate (τ^{-1} is the damping coefficient), F is a filling factor, and ω_0 is a localized plasmon resonant frequency. When

$$(1 - F)(\omega^2 - \omega_0^2)^2 - F\omega_0^2(\omega^2 - \omega_0^2) + \tau^{-2}\omega^2 < 0,$$

the real part of $\mu_c(\omega)$ is negative.

We suppose that $D = z + \delta B$. The quasi-static plasmonic resonance is defined by ω such that

$$\Re \frac{\mu_m + \mu_c(\omega)}{2(\mu_m - \mu_c(\omega))} = \lambda_j$$

for some j, where λ_j is an eigenvalue of the Neumann–Poincaré operator $\mathcal{K}_D^*(= \mathcal{K}_B^*)$. It is clear that such definition is independent of the nanoparticle's size. In view of (2.15), the shifted plasmonic resonance is defined by

$$\operatorname{argmin}\left|\frac{1}{2\mu_m} + \frac{1}{2\mu_c(\omega)} - (\frac{1}{\mu_c(\omega)} - \frac{1}{\mu_m})\lambda_j + \omega^2\delta^2\tau_{j,2}\right|,$$

where $\tau_{j,2}$ is given by (2.17) with D replaced by B.

3. Multiple Plasmonic Nanoparticles

3.1. Layer potential formulation in the multi-particle case

We consider the scattering of an incident time harmonic wave u^i by multiple weakly coupled plasmonic nanoparticles in three dimensions. Our motivation is to demonstrate the principle of super-resolution in resonant media; see Section 6. The scattering from multiple weakly coupled, non-resonant small particles can be analyzed in the same way. However, no super-resolution can be achieved in this case.

For ease of exposition, we consider the case of L particles with an identical shape. We assume that Condition 1 holds. Moreover, in contrast to Section 2 where the size of the particle is assumed to be of order one, we assume the following condition in the section.

Condition 4. *All the identical particles have size of order δ which is a small parameter and the distances between neighboring ones are of order one.*

We write $D_l = z_l + \delta\widetilde{D}, l = 1, 2, \ldots, L$, where $\widetilde{D}$ has size one and is centered at the origin. Moreover, we denote $D_0 = \delta\widetilde{D}$ as our reference nanoparticle. Denote by

$$D = \bigcup_{l=1}^{L} D_l, \quad \varepsilon_D = \varepsilon_m \chi(\mathbb{R}^3 \backslash \bar{D}) + \varepsilon_c \chi(\bar{D}), \quad \mu_D = \mu_m \chi(\mathbb{R}^3 \backslash \bar{D}) + \mu_c \chi(D).$$

The scattering problem can be modeled by the following Helmholtz equation:

$$\begin{cases} \nabla \cdot \dfrac{1}{\mu_D} \nabla u + \omega^2 \varepsilon_D u = 0 \quad \text{in } \mathbb{R}^3 \backslash \partial D, \\ u_+ - u_- = 0 \quad \text{on } \partial D, \\ \dfrac{1}{\mu_m} \dfrac{\partial u}{\partial \nu}\Big|_+ - \dfrac{1}{\mu_c} \dfrac{\partial u}{\partial \nu}\Big|_- = 0 \quad \text{on } \partial D, \\ u^s := u - u^i \quad \text{satisfies the Sommerfeld radiation condition.} \end{cases} \tag{3.1}$$

Let

$$\begin{aligned} u^i(x) &= e^{ik_m d\cdot x}, \\ F_{l,1}(x) &= -u^i(x)\big|_{\partial D_l} = -e^{ik_m d\cdot x}\big|_{\partial D_l}, \\ F_{l,2}(x) &= -\frac{\partial u^i}{\partial \nu}(x)\Big|_{\partial D_l} = -ik_m e^{ik_m d\cdot x} d \cdot \nu(x)\big|_{\partial D_l}, \end{aligned}$$

and define the operator $\mathcal{K}^k_{D_p, D_l}$ by

$$\mathcal{K}^k_{D_p, D_l}[\psi](x) = \int_{\partial D_p} \frac{\partial G(x, y, k)}{\partial \nu(x)} \psi(y) \, \mathrm{d}\sigma(y), \quad x \in \partial D_l.$$

Analogously, we define

$$\mathcal{S}^k_{D_p,D_l}[\psi](x) = \int_{\partial D_p} G(x,y,k)\psi(y)\,\mathrm{d}\sigma(y), \quad x \in \partial D_l.$$

The solution u of (3.1) can be represented as follows:

$$u(x) = \begin{cases} u^i + \displaystyle\sum_{l=1}^{L} \mathcal{S}^{k_m}_{D_l}[\psi_l], & x \in \mathbb{R}^3 \backslash \bar{D}, \\ \displaystyle\sum_{l=1}^{L} \mathcal{S}^{k_c}_{D_l}[\phi_l], & x \in D, \end{cases}$$

where $\phi_l, \psi_l \in H^{-\frac{1}{2}}(\partial D_l)$ satisfy the following system of integral equations

$$\begin{cases} \mathcal{S}^{k_m}_{D_l}[\psi_l] - \mathcal{S}^{k_c}_{D_l}[\phi_l] + \displaystyle\sum_{p \neq l} \mathcal{S}^{k_m}_{D_p,D_l}[\psi_p] = F_{l,1}, \\ \dfrac{1}{\mu_m}\left(\dfrac{1}{2}Id + \left(\mathcal{K}^{k_m}_{D_l}\right)^*\right)[\psi_l] + \dfrac{1}{\mu_c}(\dfrac{1}{2}Id - \left(\mathcal{K}^{k_c}_{D_l}\right)^*)[\phi_l] \\ \qquad + \dfrac{1}{\mu_m}\displaystyle\sum_{p \neq l} \mathcal{K}^{k_m}_{D_p,D_l}[\psi_p] = F_{l,2}, \end{cases}$$

and

$$\begin{cases} F_{l,1} = -u^i \quad \text{on } \partial D_l, \\ F_{l,2} = -\dfrac{1}{\mu_m}\dfrac{\partial u^i}{\partial \nu} \quad \text{on } \partial D_l. \end{cases}$$

3.2. First-order correction to plasmonic resonances and field behavior at plasmonic resonances in the multi-particle case

We consider the scattering in the quasi-static regime, that is, when the incident wavelength is much greater than one. With proper dimensionless analysis, we can assume that $\omega \ll 1$. As a consequence, $\mathcal{S}^{k_c}_D$ is invertible. Note that

$$\phi_l = \left(\mathcal{S}^{k_c}_{D_l}\right)^{-1}\left(\mathcal{S}^{k_m}_{D_l}[\psi_l] + \sum_{p \neq l} \mathcal{S}^{k_m}_{D_p,D_l}[\psi_p] - F_{l,1}\right).$$

We obtain the following equation for ψ_l's:

$$\mathcal{A}_D(w)[\psi] = f,$$

where

$$\mathcal{A}_D(w) = \begin{pmatrix} \mathcal{A}_{D_1}(\omega) & & & \\ & \mathcal{A}_{D_2}(\omega) & & \\ & & \ddots & \\ & & & \mathcal{A}_{D_L}(\omega) \end{pmatrix}$$
$$+ \begin{pmatrix} 0 & \mathcal{A}_{1,2}(\omega) & \cdots & \mathcal{A}_{1,L}(\omega) \\ \mathcal{A}_{2,1}(\omega) & 0 & \cdots & \mathcal{A}_{2,L}(\omega) \\ \vdots & \cdots & 0 & \vdots \\ \mathcal{A}_{L,1}(\omega) & \cdots & \mathcal{A}_{L,L-1}(\omega) & 0 \end{pmatrix},$$
$$\psi = \begin{pmatrix} \psi_1 \\ \psi_2 \\ \vdots \\ \psi_L \end{pmatrix}, \quad f = \begin{pmatrix} f_1 \\ f_2 \\ \vdots \\ f_L \end{pmatrix},$$

and

$$\mathcal{A}_{l,p}(\omega) = \frac{1}{\mu_c}\left(\frac{1}{2}Id - \left(\mathcal{K}_{D_l}^{k_c}\right)^*\right)\left(\mathcal{S}_{D_l}^{k_c}\right)^{-1}\mathcal{S}_{D_p,D_l}^{k_m} + \frac{1}{\mu_m}\mathcal{K}_{D_p,D_l}^{k_m},$$
$$f_l = F_{l,2} + \frac{1}{\mu_c}\left(\frac{1}{2}Id - (\mathcal{K}_{D_l}^{k_c})^*\right)(\mathcal{S}_{D_l}^{k_c})^{-1}[F_{l,1}].$$

The following asymptotic expansions hold:

Lemma 3.1. *(i) Regarded as operators from $\mathcal{H}^*(\partial D_p)$ into $\mathcal{H}^*(\partial D_l)$, we have*

$$\mathcal{A}_{D_j}(\omega) = \mathcal{A}_{D_j,0} + O(\delta^2\omega^2);$$

(ii) Regarded as operators from $\mathcal{H}^*(\partial D_l)$ into $\mathcal{H}^*(\partial D_j)$, we have

$$\mathcal{A}_{l,p}(\omega) = \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}_{D_l}^*\right)\mathcal{S}_{D_l}^{-1}\left(\mathcal{S}_{p,l,0,1} + \mathcal{S}_{p,l,0,2}\right)$$
$$+ \frac{1}{\mu_m}\mathcal{K}_{p,l,0,0} + O(\delta^2\omega^2) + O(\delta^4).$$

Moreover,

$$\left(\frac{1}{2}Id - \mathcal{K}_{D_l}^*\right) \circ \mathcal{S}_{D_l}^{-1} \circ \mathcal{S}_{p,l,0,1} = O(\delta^2),$$
$$\left(\frac{1}{2}Id - \mathcal{K}_{D_l}^*\right) \circ \mathcal{S}_{D_l}^{-1} \circ \mathcal{S}_{p,l,0,2} = O(\delta^3),$$
$$\mathcal{K}_{p,l,0,0} = O(\delta^2).$$

Proof. The proof of (i) follows from Lemmas 2.4 and B.3. We now prove (ii). Recall that

$$
\begin{aligned}
\frac{1}{2}Id - \left(\mathcal{K}_{D_l}^{k_c}\right)^* &= \frac{1}{2}Id - \mathcal{K}_{D_l}^* + O(\delta^2\omega^2),\\
\left(\mathcal{S}_{D_l}^{k_c}\right)^{-1} &= \mathcal{S}_{D_l}^{-1} - k_c\mathcal{S}_{D_l}^{-1}\mathcal{S}_{D_l,1}\mathcal{S}_{D_l}^{-1} + O(\delta^2\omega^2),\\
\mathcal{S}_{D_p,D_l}^{k_m} &= \mathcal{S}_{p,l,0,0} + \mathcal{S}_{p,l,0,1} + \mathcal{S}_{p,l,0,2}\\
&\quad + k_m\mathcal{S}_{p,l,1} + k_m^2\mathcal{S}_{p,l,2,0} + O(\delta^4) + O(\omega^2\delta^2)\\
\mathcal{K}_{D_p,D_l}^{k_m} &= \mathcal{K}_{p,l,0,0} + O(\omega^2\delta^2).
\end{aligned}
$$

Using the identity

$$
\left(\frac{1}{2}Id - \mathcal{K}_{D_l}^*\right)\mathcal{S}_{D_l}^{-1}[\chi(D_l)] = 0,
$$

we can derive that

$$
\begin{aligned}
\mathcal{A}_{l,p}(\omega) &= \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}_{D_l}^*\right)(\mathcal{S}_{D_l}^{k_c})^{-1}\mathcal{S}_{D_p,D_l}^{k_m} + \frac{1}{\mu_m}\mathcal{K}_{p,l,0,0} + O(\delta^2\omega^2)\\
&= \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}_{D_l}^*\right)\mathcal{S}_{D_l}^{-1}\mathcal{S}_{D_p,D_l}^{k_m} + \frac{1}{\mu_m}\mathcal{K}_{p,l,0,0} + O(\delta^2\omega^2)\\
&= \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}_{D_l}^*\right)\mathcal{S}_{D_l}^{-1}\big(\mathcal{S}_{p,l,0,0} + \mathcal{S}_{p,l,0,1} + \mathcal{S}_{p,l,0,2}\\
&\quad + k_m\mathcal{S}_{p,l,1} + k_m^2\mathcal{S}_{p,l,2,0} + O(\delta^4)\big) + \frac{1}{\mu_m}\mathcal{K}_{p,l,0,0} + O(\delta^2\omega^2)\\
&= \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}_{D_l}^*\right)\mathcal{S}_{D_l}^{-1}\big(\mathcal{S}_{p,l,0,1} + \mathcal{S}_{p,l,0,2}\big)\\
&\quad + \frac{1}{\mu_m}\mathcal{K}_{p,l,0,0} + O(\delta^2\omega^2) + O(\delta^4).
\end{aligned}
$$

The rest of the lemma follows from Lemmas B.3 and B.6. □

Denote by $\mathcal{H}^*(\partial D) = \mathcal{H}^*(\partial D_1) \times \ldots \times \mathcal{H}^*(\partial D_L)$, which is equipped with the inner product

$$
(\psi, \phi)_{\mathcal{H}^*} = \sum_{l=1}^{L}(\psi_l, \phi_l)_{\mathcal{H}^*(\partial D_l)}.
$$

With the help of Lemma 3.1, the following result is obvious:

Lemma 3.2. *Regarded as an operator from $\mathcal{H}^*(\partial D)$ into $\mathcal{H}^*(\partial D)$, we have*

$$
\mathcal{A}(\omega) = \mathcal{A}_{D,0} + \mathcal{A}_{D,1} + O(\omega^2\delta^2) + O(\delta^4),
$$

where

$$
\mathcal{A}_{D,0} = \begin{pmatrix} \mathcal{A}_{D_1,0} & & & \\ & \mathcal{A}_{D_2,0} & & \\ & & \cdots & \\ & & & \mathcal{A}_{D_L,0} \end{pmatrix}, \quad \mathcal{A}_{D,1} = \begin{pmatrix} 0 & \mathcal{A}_{D,1,12} & \mathcal{A}_{D,1,13} & \cdots \\ \mathcal{A}_{D,1,21} & 0 & \mathcal{A}_{D,1,23} & \cdots \\ & & \cdots & \\ \mathcal{A}_{D,1,L1} & \cdots & \mathcal{A}_{D,1,LL-1} & 0 \end{pmatrix}
$$

with

$$\mathcal{A}_{D_l,0} = \left(\frac{1}{2\mu_m} + \frac{1}{2\mu_c}\right) Id - \left(\frac{1}{\mu_c} - \frac{1}{\mu_m}\right)\mathcal{K}^*_{D_l},$$
$$\mathcal{A}_{D,1,pq} = \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}^*_{D_p}\right)\mathcal{S}^{-1}_{D_p}\big(\mathcal{S}_{q,p,0,1} + \mathcal{S}_{q,p,0,2}\big) + \frac{1}{\mu_m}\mathcal{K}_{q,p,0,0}.$$

It is evident that

$$\mathcal{A}_{D,0}[\psi] = \sum_{j=0}^{\infty}\sum_{l=1}^{L} \tau_j (\psi, \varphi_{j,l})_{\mathcal{H}^*}\varphi_{j,l}, \tag{3.2}$$

where

$$\tau_j = \frac{1}{2\mu_m} + \frac{1}{2\mu_c} - \left(\frac{1}{\mu_c} - \frac{1}{\mu_m}\right)\lambda_j, \tag{3.3}$$
$$\varphi_{j,l} = \varphi_j e_l \tag{3.4}$$

with e_l being the standard basis of $\mathbb{R}^L$.

We take $\mathcal{A}(\omega)$ as a perturbation to the operator $\mathcal{A}_{D,0}$ for small ω and small δ. Using a standard perturbation argument, we can derive the perturbed eigenvalues and eigenfunctions. For simplicity, we assume that the following conditions hold:

Condition 5. *Each eigenvalue λ_j, $j \in J$, of the operator $\mathcal{K}^*_{D_1}$ is simple. Moreover, we have $\omega^2 \ll \delta$.*

In what follows, we only use the first order perturbation theory and derive the leading order term, that is, the perturbation due to the term $\mathcal{A}_{D,1}$. For each l, we define an $L \times L$ matrix R_l by letting

$$\begin{aligned} R_{l,pq} &= \big(\mathcal{A}_{D,1}[\varphi_{l,p}], \varphi_{l,q}\big)_{\mathcal{H}^*}, \\ &= \Big(\mathcal{A}_{D,1}[\varphi_l e_p], \varphi_l e_q\Big)_{\mathcal{H}^*}, \\ &= \big(\mathcal{A}_{D,1,pq}[\varphi_l], \varphi_l\big)_{\mathcal{H}^*}. \end{aligned}$$

Lemma 3.3. *The matrix $R_l = (R_{l,pq})_{p,q=1,\ldots,L}$ has the following explicit expression:*

$$\begin{aligned} R_{l,pp} &= 0, \\ R_{l,pq} &= \frac{3}{4\pi\mu_c}\left(\lambda_j - \frac{1}{2}\right)\sum_{|\alpha|=|\beta|=1}\int_{\partial D_0}\int_{\partial D_0}\frac{(z_p - z_q)^{\alpha+\beta}}{|z_p - z_q|^5}x^\alpha y^\beta \varphi_l(x)\varphi_l(y) \\ &\quad d\sigma(x)\, d\sigma(y) \\ &\quad + \left(\frac{1}{4\pi\mu_c} - \frac{1}{4\pi\mu_m}\right)\left(\lambda_j - \frac{1}{2}\right)\int_{\partial D_0}\int_{\partial D_0}\frac{x\cdot y}{|z_p - z_q|^3}\varphi_l(x)\varphi_l(y) \\ &\quad d\sigma(x)\, d\sigma(y) \\ &= O(\delta^3), \quad p \neq q. \end{aligned}$$

Proof. It is clear that $R_{l,pp} = 0$. For $p \neq q$, we have

$$R_{l,pq} = R^{I}_{l,pq} + R^{II}_{l,pq} + R^{III}_{l,pq},$$

where

$$\begin{aligned} R^{I}_{l,pq} &= \frac{1}{\mu_c}\Big(\big(\frac{1}{2}Id - \mathcal{K}^*_{D_p}\big)\mathcal{S}^{-1}_{D_p}\mathcal{S}_{q,p,0,1}[\varphi_l], \varphi_l\Big)_{\mathcal{H}^*(\partial D_l)}, \\ R^{II}_{l,pq} &= \frac{1}{\mu_c}\Big(\big(\frac{1}{2}Id - \mathcal{K}^*_{D_p}\big)\mathcal{S}^{-1}_{D_p}\mathcal{S}_{q,p,0,2}[\varphi_l], \varphi_l\Big)_{\mathcal{H}^*(\partial D_l)}, \\ R^{III}_{l,pq} &= \frac{1}{\mu_m}\big(\mathcal{K}_{q,p,0,0}[\varphi_l], \varphi_l\big)_{\mathcal{H}^*(\partial D_l)}. \end{aligned}$$

We first consider $R^{I}_{l,pq}$. By the identity

$$\left(\frac{1}{2}Id - \mathcal{K}^*_{D_p}\right)\mathcal{S}_{D_l}[\varphi_l] = \mathcal{S}_{D_l}\left(\frac{1}{2}Id - \mathcal{K}_{D_p}\right)[\varphi_l] = \left(\lambda_j - \frac{1}{2}\right)\varphi_l,$$

we obtain

$$\begin{aligned} R^{I}_{l,pq} &= -\frac{1}{\mu_c}\Big(\big(\frac{1}{2}Id - \mathcal{K}^*_{D_p}\big)\mathcal{S}^{-1}_{D_p}\mathcal{S}_{q,p,0,1}[\varphi_l], \mathcal{S}_{D_l}[\varphi_l]\Big)_{L^2(\partial D_l)}, \\ &= \frac{1}{\mu_c}\left(\lambda_j - \frac{1}{2}\right)\big(\mathcal{S}_{q,p,0,1}[\varphi_l], \mathcal{S}_{D_l}[\varphi_l]\big)_{L^2(\partial D_l)}. \end{aligned}$$

Using the explicit representation of $\mathcal{S}_{q,p,0,1}$ and the fact that $(\chi(\partial D_j), \phi_l)_{L^2(\partial D_j)} = 0$ for $j \neq 0$, we further conclude that

$$R^{I}_{l,pq} = 0.$$

Similarly, we have

$$\begin{aligned} R^{II}_{l,pq} &= \frac{1}{\mu_c}\left(\lambda_j - \frac{1}{2}\right)\big(\mathcal{S}_{q,p,0,2}[\varphi_l], \mathcal{S}_{D_l}[\varphi_l]\big)_{L^2(\partial D_l)}, \\ &= \frac{1}{\mu_c}\left(\lambda_j - \frac{1}{2}\right)\sum_{|\alpha|=|\beta|=1}\int_{\partial D_0}\int_{\partial D_0} \\ &\quad \Big(\frac{3(z_p - z_q)^{\alpha+\beta}}{4\pi|z_p - z_q|^5}x^\alpha y^\beta + \frac{\delta_{\alpha\beta}x^\alpha y^\beta}{4\pi|z_p - z_q|^3}\Big)\varphi_l(x)\varphi_l(y)\,\mathrm{d}\sigma(x)\,\mathrm{d}\sigma(y) \\ &= \frac{3}{4\pi\mu_c}\left(\lambda_j - \frac{1}{2}\right)\sum_{|\alpha|=|\beta|=1}\int_{\partial D_0}\int_{\partial D_0}\frac{(z_p - z_q)^{\alpha+\beta}}{|z_p - z_q|^5}x^\alpha y^\beta\varphi_l(x)\varphi_l(y) \\ &\quad \mathrm{d}\sigma(x)\,\mathrm{d}\sigma(y) \\ &\quad + \frac{1}{4\pi\mu_c}\left(\lambda_j - \frac{1}{2}\right)\sum_{|\alpha|=1}\int_{\partial D_0}\int_{\partial D_0}\frac{1}{|z_p - z_q|^3}x^\alpha y^\alpha\varphi_l(x)\varphi_l(y) \\ &\quad \mathrm{d}\sigma(x)\,\mathrm{d}\sigma(y). \end{aligned}$$

Finally, note that

$$\mathcal{K}_{q,p,0,0}[\varphi_l] = \frac{1}{4\pi|z_p - z_q|^3} a \cdot \nu(x) = \frac{1}{4\pi|z_p - z_q|^3} \sum_{m=1}^{3} a_m \nu_m(x),$$

where $a_m = \big((y - z_q)_m, \varphi_l\big)_{L^2(\partial D_q)}$, and $a = (a_1, a_2, a_3)^T$.

By identity (2.25), we have

$$\begin{aligned}
R^{III}_{l,pq} &= -\frac{1}{\mu_m}\big(\mathcal{K}_{q,p,0,0}[\varphi_l], \varphi_l\big)_{\mathcal{H}^*(\partial D_l)} \\
&= -\frac{1}{4\pi|z_p - z_q|^3\mu_m}\big(a \cdot \nu(x), \varphi_l\big)_{\mathcal{H}^*(\partial D_l)} \\
&= -\frac{1}{4\pi|z_p - z_q|^3\mu_m}\left((\frac{1}{2}Id - \mathcal{K}^*_{D_p})\mathcal{S}^{-1}_{D_p}(a \cdot (x - z_p)), \varphi_l\right)_{\mathcal{H}^*(\partial D_l)} \\
&= -\frac{1}{4\pi|z_p - z_q|^3\mu_m}\left(\lambda_j - \frac{1}{2}\right)\big(a \cdot (x - z_p), \varphi_l\big)_{L^2(\partial D_p)} \\
&= -\frac{1}{4\pi|z_p - z_q|^3\mu_m}\left(\lambda_j - \frac{1}{2}\right)\int_{\partial D_0}\int_{\partial D_0} x \cdot y\varphi_l(x)\varphi_l(y)\, d\sigma(x)\, d\sigma(y).
\end{aligned}$$

This completes the proof of the lemma. □

We now have an explicit formula for the matrix R_l. It is clear that R_l is symmetric, but not self-adjoint. For ease of presentation, we assume the following condition:

Condition 6. *R_l has L-distinct eigenvalues.*

We remark that Condition 6 is not essential for our analysis. Without this condition, the perturbation argument is still applicable, but the results may be quite complicated. We refer to [30] for a complete description of the perturbation theory.

Let $\tau_{j,l}$ and $X_{j,l} = (X_{j,l,1}, \cdots, X_{j,l,L})^T$, $l = 1, 2, \ldots, L$, be the eigenvalues and normalized eigenvectors of the matrix R_j. Here, T denotes the transpose. We remark that each $X_{j,l}$ may be complex valued and may not be orthogonal to other eigenvectors.

Under perturbation, each τ_j is split into the following L eigenvalues of $\mathcal{A}(\omega)$:

$$\tau_{j,l}(\omega) = \tau_j + \tau_{j,l} + O(\delta^4) + O(\omega^2\delta^2). \tag{3.5}$$

The associated perturbed eigenfunctions have the form

$$\varphi_{j,l}(\omega) = \sum_{p=1}^{L} X_{j,l,p}e_p\varphi_j + O(\delta^4) + O(\omega^2\delta^2). \tag{3.6}$$

We are interested in solving the equation $\mathcal{A}_D(\omega)[\psi] = f$ when ω is close to the resonance frequencies, that is, when $\tau_j(\omega)$ are very small for some j's. In this case, the major part of the solution would be based on the excited resonance modes

$\varphi_{j,l}(\omega)$. For this purpose, we introduce the index set of resonance J as we did in the previous section for a single particle case.

We define

$$P_J(\omega)\varphi_{j,m}(\omega) = \begin{cases} \varphi_{j,m}(\omega), & j \in J, \\ 0, & j \in J^c. \end{cases}$$

In fact,

$$P_J(\omega) = \sum_{j\in J} P_j(\omega) = \sum_{j\in J} \frac{1}{2\pi i} \int_{\gamma_j} (\xi - \mathcal{A}_D(\omega))^{-1}\, \mathrm{d}\xi, \tag{3.7}$$

where γ_j is a Jordan curve in the complex plane enclosing only the eigenvalues $\tau_{j,l}(\omega)$ for $l = 1, 2, \dots, L$ among all the eigenvalues.

To obtain an explicit representation of $P_J(\omega)$, we consider the adjoint operator $\mathcal{A}_D(\omega)^*$. By a similar perturbation argument, we can obtain its perturbed eigenvalue and eigenfunctions. Note that the adjoint matrix $\bar{R}_j^T = \bar{R}_j$ has eigenvalues $\overline{\tau_{j,l}}$ and corresponding eigenfunctions $\overline{X_{j,l}}$. Then the eigenvalues and eigenfunctions of $\mathcal{A}_D(\omega)^*$ have the form

$$\begin{aligned} \widetilde{\tau}_{j,l}(\omega) &= \tau_j + \overline{\tau_{j,l}} + O(\delta^4) + O(\omega^2\delta^2), \\ \widetilde{\varphi}_{j,l}(\omega) &= \widetilde{\varphi}_{j,l} + O(\delta^4) + O(\omega^2\delta^2), \end{aligned}$$

where

$$\widetilde{\varphi}_{j,l} = \sum_{p=1}^{L} \widetilde{X}_{j,l,p} e_p \varphi_j$$

with $\widetilde{X}_{j,l,p}$ being a multiple of $\overline{X_{j,l,p}}$.

We normalize $\widetilde{\varphi}_{j,l}$ in a way such that the following holds:

$$(\varphi_{j,p}, \widetilde{\varphi}_{j,q})_{\mathcal{H}^*(\partial D)} = \delta_{pq},$$

which is also equivalent to the condition

$$\overline{X_{j,p}}^T \widetilde{X}_{j,q} = \delta_{pq}.$$

Then, we can show that the following result holds:

Lemma 3.4. *In the space $\mathcal{H}^*(\partial D)$, as ω goes to zero, we have*

$$f = \omega f_0 + O(\omega^2 \delta^{\frac{3}{2}}),$$

where $f_0 = (f_{0,1}, \dots, f_{0,L})^T$ with

$$\begin{aligned} f_{0,l} &= -i\sqrt{\varepsilon_m \mu_m} e^{ik_m d\cdot z_l} \left(\frac{1}{\mu_m} d \cdot \nu(x) + \frac{1}{\mu_c} \left(\frac{1}{2} Id - \mathcal{K}_{D_l}^* \right) \mathcal{S}_{D_l}^{-1} \left[d \cdot (x - z) \right] \right) \\ &= O(\delta^{\frac{3}{2}}). \end{aligned}$$

Proof. We first show that

$$\|u\|_{\mathcal{H}^*(\partial D_0)} = \delta^{\frac{3}{2}+m}\|u\|_{\mathcal{H}^*(\partial \widetilde{D})}, \quad \|u\|_{\mathcal{H}(\partial D_0)} = \delta^{\frac{1}{2}+m}\|u\|_{\mathcal{H}(\partial \widetilde{D})}$$

for any homogeneous function u such that $u(\delta x) = \delta^m u(x)$. Indeed, we have $\eta(u)(x) = \delta^m u(x)$. Since $\|\eta(u)\|_{\mathcal{H}^*(\partial \widetilde{D})} = \delta^{-\frac{3}{2}}\|u\|_{\mathcal{H}^*(\partial D_0)}$ (see "Appendix B"), we obtain

$$\|u\|_{\mathcal{H}^*(\partial D_0)} = \delta^{\frac{3}{2}}\|\eta(u)\|_{\mathcal{H}^*(\partial \widetilde{D})} = \delta^{\frac{3}{2}+m}\|u\|_{\mathcal{H}^*(\partial \widetilde{D})},$$

which proves our first claim. The second claim follows in a similar way. Using this result, by an argument similar to the proof of Lemma 2.6, we arrive at the desired asymptotic result. □

Denote by $Z = (Z_1, \ldots, Z_L)$, where $Z_j = ik_m e^{ik_m d\cdot z_j}$. We are ready to present our main result in this section.

Theorem 3.1. *Under Conditions 1, 2, 3, 4 and 6, the scattered field by L plasmonic particles in the quasi-static regime has the following representation:*

$$u^s = \mathcal{S}_D^{k_m}[\psi],$$

where

$$\begin{aligned}\psi &= \sum_{j\in J}\sum_{l=1}^{L}\frac{\big(f, \widetilde{\varphi}_{j,l}(\omega)\big)_{\mathcal{H}^*}\varphi_{j,l}(\omega)}{\tau_{j,l}(\omega)} + \mathcal{A}_D(\omega)^{-1}(P_{J^c}(\omega)f)\\ &= \sum_{j\in J}\sum_{l=1}^{L}\frac{(d\cdot\nu(x), \varphi_j)_{\mathcal{H}^*(\partial D_0)} Z\overline{\widetilde{X}_{j,l}}\,\varphi_{j,l} + O(\omega^2\delta^{\frac{3}{2}})}{\lambda - \lambda_j + \big(\frac{1}{\mu_c} - \frac{1}{\mu_m}\big)^{-1}\tau_{j,l} + O(\delta^4) + O(\delta^2\omega^2)} + O(\omega\delta^{\frac{3}{2}}).\end{aligned}$$

Proof. The proof is similar to that of Theorem 2.1. □

As a consequence, we have

Corollary 3.1. *With the same notation as in Theorem 3.1 and under the additional condition that*

$$\min_{j\in J}|\tau_{j,l}(\omega)| \gg \omega^q\delta^p,$$

for some integer p and q, and

$$\tau_{j,l}(\omega) = \tau_{j,l,p,q} + o(\omega^q\delta^p),$$

we have

$$\psi = \sum_{j\in J}\sum_{l=1}^{L}\frac{(d\cdot\nu(x), \varphi_j)_{\mathcal{H}^*(\partial D_0)} Z\overline{\widetilde{X}_{j,l}}\,\varphi_{j,l} + O(\omega^2\delta^{\frac{3}{2}})}{\tau_{j,l,p,q}} + O(\omega\delta^{\frac{3}{2}}).$$

4. Scattering and Absorption Enhancements

In this section we analyze the scattering and absorption enhancements. We prove that, at the quasi-static limit, the averages over the orientation of scattering and extinction cross-sections of a randomly oriented nanoparticle are given by (4.10) and (4.11), where M given by (4.7) is the polarization tensor associated with the nanoparticle D and the magnetic contrast $\mu_c(\omega)/\mu_m$. In view of (4.15), the polarization tensor M blows up at the plasmonic resonances, which yields scattering and absorption enhancements. A bound on the extinction cross-section is derived in (4.17). As shown in (4.20) and (4.22), it can be sharpened for nanoparticles of elliptical or ellipsoidal shapes.

4.1. Far-field expansion

For simplicity, we assume throughout this section that D contains the origin. We first prove the following representation for the scattering amplitude:

Proposition 4.1. *Let $u^i = e^{ik_m d\cdot x}$ with d being a unit vector. Let $x \in \mathbb{R}^3$ be such that $|x| \gg 1/\omega$. Then, we have*

$$u^s(x) = \frac{e^{ik_m|x|}}{|x|} A_\infty\left(\frac{x}{|x|}, d\right) + O\left(\frac{1}{|x|^2}\right) \tag{4.1}$$

with

$$A_\infty\left(\frac{x}{|x|}, d\right) = -\frac{1}{4\pi}\int_{\partial D} e^{-ik_m\frac{x}{|x|}\cdot y}\psi(y)\,\mathrm{d}\sigma(y) \tag{4.2}$$

being the scattering amplitude and ψ being defined by (2.3).

Proof. We recall that the scattered field u^s can be represented as follows:

$$\begin{aligned} u^s(x) &= \mathcal{S}_D^{k_m}[\psi](x) \\ &= -\frac{1}{4\pi}\int_{\partial D}\frac{e^{ik_m|x-y|}}{|x-y|}\psi(y)\,\mathrm{d}\sigma(y). \end{aligned}$$

From

$$|x-y| = |x|\left(1 - \frac{x\cdot y}{|x|^2} + O\left(\frac{1}{|x|^2}\right)\right),$$

it follows that

$$u^s(x) = -\frac{e^{ik_m|x|}}{4\pi|x|}\int_{\partial D} e^{-ik_m\frac{x}{|x|}\cdot y}\psi(y)\left(1 + \frac{(x\cdot y)}{|x|^2}\right)\mathrm{d}\sigma(y) + o\left(\frac{1}{|x|^2}\right),$$

which yields the desired result. □

4.2. Energy flow

The following definitions are from [22]. We include them here for the sake of completeness. The analogous quantity of the Poynting vector in scalar wave theory is the *energy flux* vector; see [22]. We recall that for a real monochromatic field

$$U(x,t) = \Re\left[u(x)e^{-i\omega t}\right],$$

the averaged value of the energy flux vector, taken over an interval which is long compared to the period of the oscillations, is given by

$$F(x) = -iC\left[\overline{u}(x)\nabla u(x) - u(x)\nabla\overline{u}(x)\right],$$

where C is a positive constant depending on the polarization mode. In the transverse electric case, $C = \omega/\mu_m$ while in the transverse magnetic case $C = \omega/\varepsilon_m$. Assume that the particle is contained in the ball B_R of radius R and center the origin. We now consider the outward flow of energy through the sphere ∂B_R to be

$$\mathcal{W} = \int_{\partial B_R} F(x)\cdot\nu(x)\,\mathrm{d}\sigma(x),$$

where $\nu(x)$ is the outward normal at $x \in \partial B_R$.

As the total field can be written as $U = u^s + u^i$, the flow can be decomposed into three parts as follows

$$\mathcal{W} = \mathcal{W}^i + \mathcal{W}^s + \mathcal{W}',$$

where

$$\begin{aligned}
\mathcal{W}^i &= -iC\int_{\partial B_R}\left[\overline{u^i}(x)\nabla u^i(x) - u^i(x)\nabla\overline{u^i}(x)\right]\cdot\nu(x)\,\mathrm{d}\sigma(x),\\
\mathcal{W}^s &= -iC\int_{\partial B_R}\left[\overline{u^s}(x)\nabla u^s(x) - u^s(x)\nabla\overline{u^s}(x)\right]\cdot\nu(x)\,\mathrm{d}\sigma(x),\\
\mathcal{W}' &= -iC\int_{\partial B_R}\left[\overline{u^i}(x)\nabla u^s(x) - u^s(x)\nabla\overline{u^i}(x) - u^i(x)\nabla\overline{u^s}(x) + \overline{u^s}(x)\nabla u^i(x)\right]\\
&\qquad\cdot\nu(x)\,\mathrm{d}\sigma(x).
\end{aligned}$$

It is straightforward to check that $\mathcal{W}$, $\mathcal{W}^i$, $\mathcal{W}^a$ and $\mathcal{W}'$ in the above definitions are independent of the radius R as long as the particle is contained in B_R. In the case where u^i is a plane wave, we can see that $\mathcal{W}^i = 0$:

$$\begin{aligned}
\mathcal{W}^i &= -iC\int_{\partial B_R}\left[\overline{u^i}(x)\nabla u^i(x) - u^i(x)\nabla\overline{u^i}(x)\right]\;\mathrm{d}\sigma(x),\\
&= -iC\int_{\partial B_R}\left[e^{-ik_m d\cdot x}ik_m d e^{ik_m d\cdot x} + e^{ik_m d\cdot x}k_m d e^{-ik_m d\cdot x}\right]\cdot\nu(x)\,\mathrm{d}\sigma(x),\\
&= 2Ck_m d\cdot\int_{\partial B_R}\nu(x)\,\mathrm{d}\sigma(x),\\
&= 0.
\end{aligned}$$

In a non-absorbing medium with a non-absorbing scatterer, $\mathcal{W}$ is equal to zero because the electromagnetic energy would be conserved by the scattering process. However, if the scatterer is an absorbing body, the conservation of energy gives the rate of absorption as

$$\mathcal{W}^a = -\mathcal{W}.$$

Therefore, we have

$$\mathcal{W}^a + \mathcal{W}^s = -\mathcal{W}'.$$

Here, $\mathcal{W}'$ is called the extinction rate. It is the rate at which the energy is removed by the scatterer from the illuminating plane wave, and it is the sum of the rate of absorption and the rate at which energy is scattered.

4.3. Extinction, absorption, and scattering cross-sections and the optical theorem

Denote by U^i the quantity $U^i(x) = \left|\overline{u^i}(x)\nabla u^i(x) - u^i(x)\nabla\overline{u^i}(x)\right|$. In the case of a plane wave illumination, $U^i(x)$ is independent of x and is given by $U^i = 2k_m$.

Definition 3. The scattering cross-section Q^s, the absorption cross-section Q^a and the extinction cross-section are defined by

$$Q^s = \frac{\mathcal{W}^s}{U^i}, \quad Q^a = \frac{\mathcal{W}^a}{U^i}, \quad Q^{ext} = \frac{-\mathcal{W}'}{U^i}.$$

Note that these quantities are independent of x for a plane wave illumination.

Theorem 4.1 (Optical theorem). *If $u^i(x) = e^{ik_m d\cdot x}$, where d is a unit direction, then*

$$Q^{ext} = Q^s + Q^a = \frac{4\pi}{k_m}\Im\left[A_\infty(d,d)\right], \tag{4.3}$$

$$Q^s = \int_{\mathbb{S}^2} |A_\infty(\hat{x},d)|^2 \, \mathrm{d}\sigma(\hat{x}) \tag{4.4}$$

with A_∞ being the scattering amplitude defined by (4.2).

Proof. The Sommerfeld radiation condition gives, for any $x \in \partial B_R$,

$$\nabla u^s(x)\cdot\nu(x) \sim ik_m u^s(x). \tag{4.5}$$

Hence, from (4.1), we get

$$u^s(x)\nabla\overline{u^s}(x)\cdot\nu(x) - \overline{u^s}(x)\nabla u^s(x)\cdot\nu(x) \sim -\frac{2ik_m}{|x|^2}\left|A_\infty\left(\frac{x}{|x|},d\right)\right|^2,$$

which yields (4.4). We now compute the extinction rate. We have

$$\nabla u^i(x)\cdot\nu(x) = ik_m d\cdot\nu(x)e^{ik_m d\cdot x}. \tag{4.6}$$

Therefore, using 4.5 and 4.6, it follows that

$$
\begin{aligned}
&\overline{u^i}(x)\nabla u^s(x)\cdot\nu(x) - u^s(x)\nabla\overline{u^i}(x)\cdot\nu(x)\\
&\sim \big(ik_m\frac{e^{ik_m(|x|-d\cdot x)}}{|x|}d\cdot\nu + ik_m\frac{e^{ik_m(|x|-d\cdot x)}}{|x|}\big)A_\infty\left(\frac{x}{|x|},d\right)\\
&= \frac{ik_m e^{ik_m|x|-d\cdot\nu(x)}}{|x|}\left(d\cdot\nu(x)+1\right)A_\infty\left(\frac{x}{|x|},d\right).
\end{aligned}
$$

For $x\in\partial B_R$, we can write

$$
\begin{aligned}
&\overline{u^i}(x)\nabla u^s(x)\cdot\nu(x) - u^s(x)\nabla\overline{u^i}(x)\cdot\nu(x)\\
&\quad\sim \frac{ik_m e^{-ik_mR\nu(x)\cdot(d-\nu(x))}}{R}\left(d\cdot\nu(x)+1\right)A_\infty\left(\frac{x}{|x|},d\right).
\end{aligned}
$$

We now use Jones' lemma (see, for instance, [22, Chapter 13.3]) to write the following asymptotic expansion as $R\to\infty$:

$$
\frac{1}{R}\int_{\partial B_R}\mathcal{G}(\nu(x))e^{-ik_m d\cdot\nu(x)}\,\mathrm{d}\sigma(x)\sim\frac{2\pi i}{k_m}\left(\mathcal{G}(d)e^{-ik_mR}-\mathcal{G}(-d)e^{ik_mR}\right),
$$

to obtain

$$
\int_{\partial B_R}\left[\overline{u^i}(x)\nabla u^s(x)-u^s(x)\nabla\overline{u^i}(x)\right]\cdot\nu(x)\sim -4\pi A_\infty(d,d)\quad\text{as } R\to\infty.
$$

Therefore,

$$
\mathcal{W}' = -\,i4\pi C\left[A_\infty(d)-\overline{A_\infty}(d)\right] = 8\pi C\,\Im\left[A_\infty(d)\right].
$$

Since

$$
\left|\overline{u^i}(x)\nabla u^i(x)-u^i(x)\nabla\overline{u^i}(x)\right| = 2k_m,
$$

we get the result. □

4.4. The quasi-static limit

We start by recalling the small volume expansion for the far-field. Let λ be defined by (2.19) and let

$$
M(\lambda,D):=\int_{\partial D}(\lambda Id-\mathcal{K}_D^*)^{-1}[\nu]x\,\mathrm{d}\sigma(x) \tag{4.7}
$$

be the polarization tensor. The asymptotic expansion that follows holds. It can be proved by exactly the same arguments as those in [6].

Proposition 4.2. *Assume that $D = \delta B + z$. As δ goes to zero the scattered field u^s can be written as follows:*

$$u^s(x) = -k_m^2 \left(\frac{\varepsilon_c}{\varepsilon_m} - 1 \right) |D| G(x, z, k_m) u^i(z) - \nabla_z G(x, z, k_m) \cdot M(\lambda, D) \nabla u^i(z)$$
$$+ O\left(\frac{\delta^4}{\operatorname{dist}(\lambda, \sigma(\mathcal{K}_D^*))} \right) \tag{4.8}$$

for x away from D. Here, $\operatorname{dist}(\lambda, \sigma(\mathcal{K}_D^*))$ denotes $\min_j |\lambda - \lambda_j|$ with λ_j being the eigenvalues of $\mathcal{K}_D^*$.

We denote the first term in the right hand side of (4.8) by u_1^s and the second term by u_2^s. It is clear that u_1^s represent monopole radiation and u_2^s the dipole radiation. We explicitly compute the scattering amplitude A_∞ in (4.1). Take $u^i(x) = e^{ik_m d\cdot x}$ and assume again for simplicity that $z = 0$. Note that

$$u_2^s(x) = \frac{e^{ik_m|x|}}{4\pi|x|} ik_m \left(ik_m \frac{x}{|x|} - \frac{x}{|x|^2} \right) \cdot M(\lambda, D) d.$$

In the far-field region, that is for $|x| \gg \frac{1}{\omega}$,

$$u_2^s(x) = -k_m^2 \frac{e^{ik_m|x|}}{4\pi|x|} \left(\frac{x}{|x|} \cdot M(\lambda, D) d \right) + O\left(\frac{1}{|x|^2} \right).$$

On the other hand,

$$u_1^s(x) = k_m^2 \frac{e^{ik_m|x|}}{4\pi|x|} \left(\frac{\varepsilon_c}{\varepsilon_m} - 1 \right) \cdot |D|.$$

Throughout the paper, we are interested in the case when the frequency is near the plasmonic resonant frequency, then the polarization tensor $M(\lambda, D)$ blow up and hence the magnitude of the dipole part u_2^s is much greater than that of the monopole part u_1^s. Therefore, the leading term in the scattered field (4.8) is given by the dipole part, that is

$$u^s(x) \approx -k_m^2 \frac{e^{ik_m|x|}}{4\pi|x|} \left(\frac{x}{|x|} \cdot M(\lambda, D) d \right). \tag{4.9}$$

In the next proposition we write the extinction and scattering cross-sections in terms of the polarization tensor.

Proposition 4.3. *Near plasmonic resonant frequency, the leading-order term (as δ goes to zero) of the average over the orientation of the extinction cross-section of a randomly oriented nanoparticle is given by*

$$Q_m^{ext} = \frac{4\pi k_m}{3} \Im\left[\operatorname{Tr} M(\lambda, D) \right], \tag{4.10}$$

where Tr *denotes the trace of a matrix. The leading-order term of the average over the orientation scattering cross-section of a randomly oriented nanoparticle is given by*

$$Q_m^s = \frac{k_m^4}{9\pi} \left|\mathrm{Tr} M(\lambda, D)\right|^2 . \tag{4.11}$$

Proof. Remark from (4.9) that the scattering amplitude A_∞ in the case of a plane wave illumination is given by

$$A_\infty\left(\frac{x}{|x|}, d\right) = -\frac{k_m^2}{4\pi}\frac{x}{|x|} \cdot M(\lambda, D)d. \tag{4.12}$$

Using Theorem 4.1, we can see that for a given orientation

$$Q^{ext} = -4\pi k_m \Im\left[d \cdot M(\lambda, D)d\right].$$

Therefore, if we integrate Q^{ext} over all illuminations we find that

$$Q_m^{ext} = -\,k_m \Im\left[\int_{\mathbb{S}^2} d \cdot M(\lambda, D)d \, d\sigma(d)\right].$$

Since $\Im M(\lambda, D)$ is symmetric, it can be written as $\Im M(\lambda, D) = P^t N(\lambda) P$ where P is unitary and N is diagonal and real. Then, by the change of variables $d = P^t x$ and using spherical coordinates, it follows that

$$Q_m^{ext} = -k_m\left[\int_{\mathbb{S}^2} x \cdot N(\lambda)x \, \mathrm{d}\sigma(x)\right],$$

and therefore,

$$Q_m^{ext} = -\frac{4\pi k_m}{3}\left[\mathrm{Tr} N(\lambda)\right] = -\frac{4\pi k_m}{3}\Im\left[\mathrm{Tr} M(\lambda, D)\right]. \tag{4.13}$$

Now, we compute the averaged scattering cross-section. Let $\Re M(\lambda, D) = \widetilde{P}^t \widetilde{N}(\lambda)\widetilde{P}$ where $\widetilde{P}$ is unitary and $\widetilde{N}$ is diagonal and real. We have

$$\begin{aligned} Q_m^s &= \frac{k_m^4}{16\pi^2}\iint_{\mathbb{S}^2\times\mathbb{S}^2} |x \cdot M(\lambda, D)d|^2 \, \mathrm{d}\sigma(x)\, d\sigma(d), \\ &= \frac{k_m^4}{16\pi^2}\Bigg[\iint_{\mathbb{S}^2\times\mathbb{S}^2} \left|\widetilde{x} \cdot N(\lambda)\widetilde{d}\right|^2 \, \mathrm{d}\sigma(\widetilde{x})\, \mathrm{d}\sigma(\widetilde{d}) \\ &\quad + \iint_{\mathbb{S}^2\times\mathbb{S}^2} \left|\widetilde{x} \cdot \widetilde{N}(\lambda)\widetilde{d}\right|^2 \, \mathrm{d}\sigma(\widetilde{x})\, \mathrm{d}\sigma(\widetilde{d})\Bigg]. \end{aligned}$$

Then a straightforward computation in spherical coordinates gives

$$Q_m^s = \frac{k_m^4}{9\pi}\left|\mathrm{Tr} M(\lambda, D)\right|^2,$$

which completes the proof. □

From Theorem 4.1, we obtain that the averaged absorption cross-section is given by

$$Q_m^a = -\frac{4\pi k_m}{3}\Im\left[\mathrm{Tr} M(\lambda, D)\right] - \frac{k_m^4}{9\pi}\left|\mathrm{Tr} M(\lambda, D)\right|^2 .$$

Therefore, under the condition (2.26), Q_m^a blows up at plasmonic resonances.

4.5. An upper bound for the averaged extinction cross-section

The goal of this section is to derive an upper bound for the modulus of the averaged extinction cross-section Q_m^{ext} of a randomly oriented nanoparticle. Recall that the entries $M_{l,m}(\lambda, D)$ of the polarization tensor $M(\lambda, D)$ are given by

$$M_{l,m}(\lambda, D) := \int_{\partial D} x_l (\lambda I - \mathcal{K}_D^*)^{-1}[\nu_m](x)\, \mathrm{d}\sigma(x). \tag{4.14}$$

For a $\mathcal{C}^{1,\alpha}$ domain D in $\mathbb{R}^d$, $\mathcal{K}_D^*$ is compact and self-adjoint in $\mathcal{H}^*$ (defined in Lemma 2.1 for $d = 3$ and in Lemma C.1 for $d = 2$). Thus, we can write

$$(\lambda Id - \mathcal{K}_D^*)^{-1}[\psi] = \sum_{j=0}^{\infty} \frac{(\psi, \varphi_j)_{\mathcal{H}^*} \otimes \varphi_j}{\lambda - \lambda_j},$$

with (λ_j, φ_j) being the eigenvalues and eigenvectors of $\mathcal{K}_D^*$ in $\mathcal{H}^*$ (see Lemma 2.1). Hence, the entries of the polarization tensor M can be decomposed as

$$M_{l,m}(\lambda, D) = \sum_{j=1}^{\infty} \frac{\alpha_{l,m}^{(j)}}{\lambda - \lambda_j}, \tag{4.15}$$

where $\alpha_{l,m}^{(j)} := (\nu_m, \varphi_j)_{\mathcal{H}^*}(\varphi_j, x_l)_{-\frac{1}{2},\frac{1}{2}}$. Note that $(\nu_m, \chi(\partial D))_{-\frac{1}{2},\frac{1}{2}} = 0$. So, considering the fact that $\lambda_0 = 1/2$, we have $(\nu_m, \varphi_0)_{\mathcal{H}^*} = 0$ and so, $\alpha_{l,m}^{(0)} = 0$.

The following lemmas are useful for us:

Lemma 4.1. *We have*

$$\alpha_{l,l}^{(j)} \geqq 0, \quad j \geqq 1.$$

Proof. For $d = 3$, we have

$$\begin{aligned}
(\varphi_j, x_l)_{-\frac{1}{2},\frac{1}{2}} &= \Big(\big(\frac{1}{2} - \lambda_j\big)^{-1} \big(\frac{1}{2} Id - \mathcal{K}_D^*\big)[\varphi_j], x_l \Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= \frac{-1}{1/2 - \lambda_j} \Big(\frac{\partial \mathcal{S}_D[\varphi_j]}{\partial \nu}\Big|_-, x_l \Big)_{-\frac{1}{2},\frac{1}{2}} \\
&= \int_{\partial D} \frac{\partial x_l}{\partial \nu} \mathcal{S}_D[\varphi_j]\, \mathrm{d}\sigma - \int_D \Big(\Delta x_l \mathcal{S}_D[\varphi_j] - x_l \Delta \mathcal{S}_D[\varphi_j] \Big)\, \mathrm{d}x \\
&= \frac{(\nu_l, \varphi_j)_{\mathcal{H}^*}}{1/2 - \lambda_j},
\end{aligned}$$

where we used the fact that $\mathcal{S}_D[\varphi_j]$ is harmonic in D. The same result holds for $d = 2$ if we change $\mathcal{S}_D$ by $\widetilde{\mathcal{S}}_D$ (see "Appendix C"). Since $|\lambda_j| < 1/2$ for $j \geqq 1$, we obtain the result. □

Lemma 4.2. *Let*

$$M_{l,m}(\lambda, D) = \sum_{j=1}^{\infty} \frac{\alpha_{l,m}^{(j)}}{\lambda - \lambda_j}$$

be the (l, m)*-entry of the polarization tensor* M *associated with a* $\mathcal{C}^{1,\alpha}$ *domain* $D \Subset \mathbb{R}^d$*. Then, the following properties hold:*

(i)

$$\sum_{j=1}^{\infty} \alpha_{l,m}^{(j)} = \delta_{l,m}|D|;$$

(ii)

$$\sum_{j=1}^{\infty} \lambda_i \sum_{l=1}^{d} \alpha_{l,l}^{(j)} = \frac{(d-2)}{2}|D|;$$

(iii)

$$\sum_{j=1}^{\infty} \lambda_j^2 \sum_{l=1}^{d} \alpha_{l,l}^{(j)} = \frac{(d-4)}{4}|D| + \sum_{l=1}^{d} \int_D |\nabla \mathcal{S}_D[\nu_l]|^2 \, \mathrm{d}x.$$

Proof. The proof can be found in "Appendix D". □

Let $\lambda = \lambda' + i\lambda''$. We have

$$\left|\Im(\mathrm{Tr}(M(\lambda, D)))\right| = \sum_{j=1}^{\infty} \frac{|\lambda''| \sum_{l=1}^{d} \alpha_{l,l}^{(j)}}{(\lambda' - \lambda_j)^2 + \lambda''^2}. \tag{4.16}$$

For $d = 2$ the spectrum $\sigma(\mathcal{K}_D^*)\backslash\{1/2\}$ is symmetric. For $d = 3$ this is no longer true. Nevertheless, for our purposes, we can assume that $\sigma(\mathcal{K}_D^*)\backslash\{1/2\}$ is symmetric by defining $\alpha_{l,l}^{(j)} = 0$ if λ_j is not in the original spectrum.

Without loss of generality we assume for ease of notation that Conditions 2 and 3 hold. Then we define the bijection $\rho : \mathbb{N}^+ \to \mathbb{N}^+$ such that $\lambda_{\rho(j)} = -\lambda_j$ and we can write

$$\begin{aligned}
\left|\Im(\mathrm{Tr}(M(\lambda, D)))\right| &= \frac{1}{2}\left(\sum_{j=1}^{\infty} \frac{|\lambda''|\beta_j}{(\lambda' - \lambda_j)^2 + \lambda''^2} + \sum_{j=1}^{\infty} \frac{|\lambda''|\beta^{(\rho(j))}}{(\lambda' + \lambda_j)^2 + \lambda''^2}\right) \\
&= \frac{|\lambda''|}{2} \sum_{j=1}^{\infty} \frac{(\lambda'^2 + \lambda''^2 + \lambda_j^2)(\beta^{(j)} + \beta^{(\rho(j))}) + 2\lambda'\lambda_j(\beta^{(j)} - \beta^{(\rho(j))})}{\big((\lambda' - \lambda_j)^2 + \lambda''^2\big)\big((\lambda' + \lambda_j)^2 + \lambda''^2\big)},
\end{aligned}$$

where $\beta_j = \sum_{l=1}^{d} \alpha_{l,l}^{(j)}$.

From Lemma 4.1 it follows that

$$\frac{(\lambda'^2+\lambda''^2+\lambda_j^2)(\beta^{(j)}+\beta^{(\rho(j))})+2\lambda'\lambda_j(\beta^{(j)}-\beta^{(\rho(j))})}{\big((\lambda'-\lambda_j)^2+\lambda''^2\big)\big((\lambda'+\lambda_j)^2+\lambda''^2\big)} \geqq 0.$$

Moreover,

$$\begin{aligned}&\frac{(\lambda'^2+\lambda''^2+\lambda_j^2)(\beta^{(j)}+\beta^{(\rho(j))})+2\lambda'\lambda_j(\beta^{(j)}-\beta^{(\rho(j))})}{\big((\lambda'-\lambda_j)^2+\lambda''^2\big)\big((\lambda'+\lambda_j)^2+\lambda''^2\big)}\\ &\quad\leqq \frac{(\lambda'^2+\lambda''^2+\lambda_j^2)(\beta^{(j)}+\beta^{(\rho(j))})+2\lambda'\lambda_j(\beta^{(j)}-\beta^{(\rho(j))})}{\lambda''^2(4\lambda'^2+\lambda''^2)}\\ &\qquad+O(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}).\end{aligned}$$

Hence,

$$\begin{aligned}&\big|\Im(\mathrm{Tr}(M(\lambda,D)))\big|\\ &\leqq \frac{|\lambda''|}{2}\sum_{j=1}^{\infty}\frac{(\lambda'^2+\lambda''^2+\lambda_j^2)(\beta^{(j)}+\beta^{(\rho(j))})+2\lambda'(\lambda_j\beta^{(j)}+\lambda_{\rho(j)}\beta^{(\rho(j))})}{\lambda''^2(4\lambda'^2+\lambda''^2)}\\ &\quad+O\left(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}\right).\end{aligned}$$

Using Lemma 4.2 we obtain

Theorem 4.2. *Let $M(\lambda, D)$ be the polarization tensor associated with a $\mathcal{C}^{1,\alpha}$ domain $D \Subset \mathbb{R}^d$ with $\lambda = \lambda' + i\lambda''$ such that $|\lambda''| \ll 1$ and $|\lambda'| < 1/2$. Then,*

$$\begin{aligned}&\big|\Im(\mathrm{Tr}(M(\lambda,D)))\big|\\ &\quad\leqq \frac{d|\lambda''||D|}{\lambda''^2+4\lambda'^2}+\frac{1}{|\lambda''|(\lambda''^2+4\lambda'^2)}\\ &\quad\left(d\lambda'^2|D|+\frac{(d-4)}{4}|D|+\sum_{l=1}^{d}\int_D|\nabla\mathcal{S}_D[\nu_l]|^2\,\mathrm{d}x+2\lambda'\frac{(d-2)}{2}|D|\right)\\ &\quad+O\left(\frac{\lambda''^2}{4\lambda'^2+\lambda''^2}\right).\end{aligned}$$

The bound in the above theorem depends not only on the volume of the particle but also on its geometry. Nevertheless, we remark that, since $|\lambda_j| < \frac{1}{2}$,

$$\sum_{j=1}^{\infty}\lambda_j^2\sum_{l=1}^{d}\alpha_{l,l}^{(j)} < \frac{d|D|}{4}.$$

Hence, we can find a geometry independent, but not optimal, bound.

Corollary 4.1. *We have*

$$\left|\Im(\mathrm{Tr}(M(\lambda, D)))\right| \leqq \frac{1}{|\lambda''|(\lambda''^2 + 4\lambda'^2)} \left(d|D|\big(\lambda'^2 + \frac{1}{4}\big) + 2\lambda' \frac{(d-2)}{2}|D| \right)$$
$$+ \frac{d|\lambda''||D|}{\lambda''^2 + 4\lambda'^2} + O\left(\frac{\lambda''^2}{4\lambda'^2 + \lambda''^2} \right). \tag{4.17}$$

4.5.1. Bound for ellipses If D is an ellipse whose semi-axes are on the x_1- and x_2- axes and of length a and b, respectively, then its polarization tensor takes the form [7]

$$M(\lambda, D) = \begin{pmatrix} \dfrac{|D|}{\lambda - \frac{1}{2}\frac{a-b}{a+b}} & 0 \\ 0 & \dfrac{|D|}{\lambda + \frac{1}{2}\frac{a-b}{a+b}} \end{pmatrix}. \tag{4.18}$$

On the other hand, it is known that in $\mathcal{H}^*(\partial D)$ [31]

$$\sigma(\mathcal{K}_D^*)\backslash\{1/2\} = \left\{ \pm\frac{1}{2}\left(\frac{a-b}{a+b}\right)^j, \quad j = 1, 2, \dots \right\}.$$

Then, from (4.15), we also have

$$M(\lambda, D) = \begin{pmatrix} \displaystyle\sum_{j=1}^{\infty} \frac{\alpha_{1,1}^{(j)}}{\lambda - \frac{1}{2}\left(\frac{a-b}{a+b}\right)^j} & \displaystyle\sum_{j=1}^{\infty} \frac{\alpha_{1,2}^{(j)}}{\lambda - \frac{1}{2}\left(\frac{a-b}{a+b}\right)^j} \\ \displaystyle\sum_{j=1}^{\infty} \frac{\alpha_{1,2}^{(j)}}{\lambda - \frac{1}{2}\left(\frac{a-b}{a+b}\right)^j} & \displaystyle\sum_{j=1}^{\infty} \frac{\alpha_{2,2}^{(j)}}{\lambda - \frac{1}{2}\left(\frac{a-b}{a+b}\right)^j} \end{pmatrix}.$$

Let $\lambda_1 = \dfrac{1}{2}\dfrac{a-b}{a+b}$ and $\mathcal{V}(\lambda_j) = \{i \in \mathbb{N} \text{ such that } \mathcal{K}_D^*[\varphi_i] = \lambda_j \varphi_i\}$. It is clear now that

$$\sum_{i\in\mathcal{V}(\lambda_1)} \alpha_{1,1}^{(i)} = \sum_{i\in\mathcal{V}(-\lambda_1)} \alpha_{2,2}^{(i)} = |D|, \quad \sum_{i\in\mathcal{V}(\lambda_j)} \alpha_{1,1}^{(i)} = \sum_{i\in\mathcal{V}(-\lambda_j)} \alpha_{2,2}^{(i)} = 0 \tag{4.19}$$

for $j \geqq 2$ and

$$\sum_{i\in\mathcal{V}(\lambda_j)} \alpha_{1,2}^{(i)} = 0$$

for $j \geqq 1$.

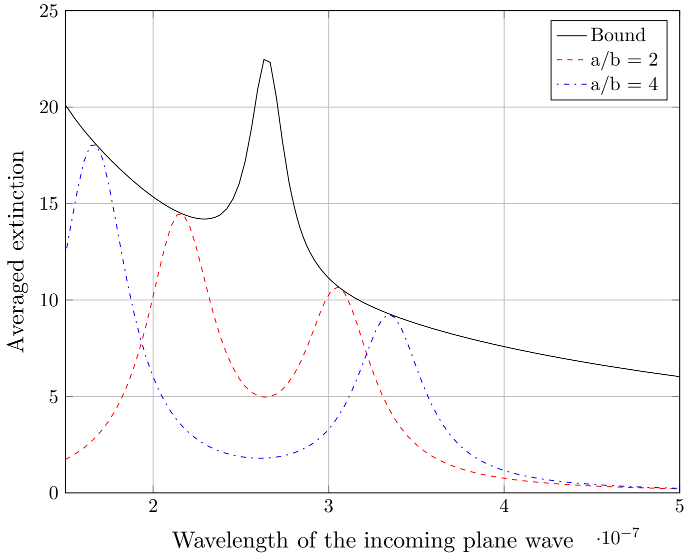

Fig. 1. Optimal bound for ellipses.

In view of (4.19), we have

$$\frac{\beta^{(j)}}{(\lambda' - \lambda_j)^2 + \lambda''^2} + \frac{\beta^{(\rho(j))}}{(\lambda' + \lambda_j)^2 + \lambda''^2} \leqq \frac{4\lambda'^2 \beta^{(j)} + \lambda''^2(\beta^{(j)} + \beta^{(j)})}{\lambda''^2(4\lambda'^2 + \lambda''^2)} + O\left(\frac{\lambda''^2}{4\lambda'^2 + \lambda''^2}\right).$$

Hence,

$$|\Im(\mathrm{Tr}(M(\lambda, D)))| \leqq \frac{|\lambda''|}{2} \sum_{j=1}^{\infty} \frac{4\lambda'^2 \beta^{(j)} + \lambda''^2(\beta^{(j)} + \beta^{(j)})}{\lambda''^2(4\lambda'^2 + \lambda''^2)} + O\left(\frac{\lambda''^2}{4\lambda'^2 + \lambda''^2}\right).$$

Note that for for any ellipse $\widetilde{D}$ of semi-axes of length a and b, $\Im(\mathrm{Tr}(M(\lambda, \widetilde{D}))) = \Im(\mathrm{Tr}(M(\lambda, D)))$. Then using Lemma 4.2 we obtain the following result:

Corollary 4.2. *For any ellipse $\widetilde{D}$ of semi-axes of length a and b, we have*

$$|\Im(Tr(M(\lambda, \widetilde{D})))| \leqq \frac{|\widetilde{D}| 4\lambda'^2}{|\lambda''|(\lambda''^2 + 4\lambda'^2)} + \frac{2|\lambda''||\widetilde{D}|}{\lambda''^2 + 4\lambda'^2} + O\left(\frac{\lambda''^2}{4\lambda'^2 + \lambda''^2}\right). \tag{4.20}$$

Figure 1 shows (4.20) and the average extinction of two ellipses of semi-axis a and b, where the ratio $a/b = 2$ and $a/b = 4$, respectively.

We can see from (4.16), Lemma 4.1 and the first sum rule in Lemma 4.2 that for an arbitrary shape B, $|\Im(\mathrm{Tr}(M(\lambda, B)))|$ is a convex combination of $\frac{|\lambda''|}{(\lambda' - \lambda_j)^2 + \lambda''^2}$ for

$\lambda_j \in \sigma(\mathcal{K}_B^*)\backslash\{1/2\}$. Since ellipses put all the weight of this convex combination in $\pm\lambda_1 = \pm\frac{1}{2}\frac{a-b}{a+b}$, we have for any ellipse $\widetilde{D}$ and any shape B such that $|B| = |\widetilde{D}|$,

$$|\Im(\mathrm{Tr}(M(\lambda^*, B)))| \leqq |\Im(\mathrm{Tr}(M(\lambda^*, \widetilde{D})))|$$

with $\lambda^* = \pm\frac{1}{2}\frac{a-b}{a+b} + i\lambda''$.

Thus, bound (4.20) applies for any arbitrary shape B in dimension two. This implies that, for a given material and a given desired resonance frequency ω^*, the optimal shape for the extinction resonance (in the quasi-static limit) is an ellipse of semi-axis a and b such that $\lambda'(\omega^*) = \pm\frac{1}{2}\frac{a-b}{a+b}$.

4.5.2. Bound for ellipsoids Let D be an ellipsoid given by

$$\frac{x_1^2}{p_1^2} + \frac{x_2^2}{p_2^2} + \frac{x_3^2}{p_3^2} = 1. \tag{4.21}$$

The following holds [7]:

Lemma 4.3. *Let D be the ellipsoid defined by (4.21). Then, for $x \in D$,*

$$\mathcal{S}_D[\nu_l](x) = s_l x_l, \quad l = 1, 2, 3,$$

where

$$s_l = -\frac{p_1 p_2 p_3}{2}\int_0^\infty \frac{1}{(p_l^2 + s)\sqrt{(p_1^2 + s)(p_2^2 + s)(p_3^2 + s)}}\,\mathrm{d}s.$$

Then we have

$$\sum_{l=1}^{3}\int_D |\nabla\mathcal{S}_D[\nu_l]|^2 dx = (s_1^2 + s_2^2 + s_3^2)|D|.$$

For a rotated ellipsoid $\widetilde{D} = \mathcal{R}D$ with $\mathcal{R}$ being a rotation matrix, we have $M(\lambda, \widetilde{D}) = \mathcal{R}M(\lambda, D)\mathcal{R}^T$ and so $\mathrm{Tr}(M(\lambda, \widetilde{D})) = \mathrm{Tr}(M(\lambda, D))$. Therefore, for any ellipsoid $\widetilde{D}$ of semi-axes of length p_1, p_2 and p_3, we have

Corollary 4.3. *For any ellipsoid $\widetilde{D}$ of semi-axes of length p_1, p_2 and p_3, we have*

$$\Im(\mathrm{Tr}(M(\lambda, \widetilde{D}))) \leqq \frac{|\widetilde{D}|\left(3\lambda'^2 + \lambda' - \frac{1}{4} + (s_1^2 + s_2^2 + s_3^2)\right)}{|\lambda''|(\lambda''^2 + 4\lambda'^2)} + \frac{3|\lambda''||\widetilde{D}|}{\lambda''^2 + 4\lambda'^2} + O\left(\frac{\lambda''^2}{4\lambda'^2 + \lambda''^2}\right), \tag{4.22}$$

where for $j = 1, 2, 3$,

$$s_j = -\frac{p_1 p_2 p_3}{2}\int_0^\infty \frac{1}{(p_j^2 + s)\sqrt{(p_1^2 + s)(p_2^2 + s)(p_3^2 + s)}}\,\mathrm{d}s.$$

5. Link with the Scattering Coefficients

Our aim in this section is to exhibit the mechanism underlying plasmonic resonances in terms of the scattering coefficients corresponding to the nanoparticle. The concept of scattering coefficients was first introduced in [9]. It plays a key role in constructing cloaking structures. It was extended in [10] to the full Maxwell equations. The scattering coefficients are simply the Fourier coefficients of the scattering amplitude A_∞. In Theorem 5.1 we provide an asymptotic expansion of the scattering amplitude in terms of the scattering coefficients of order ± 1. Our formula shows that, under physical conditions, the scattering coefficients of orders ± 1 are the only scattering coefficients inducing the scattering cross-section enhancement. For simplicity we only consider here the two-dimensional case.

5.1. The notion of scattering coefficients

From Graf's addition formula [7] and (2.2) the following asymptotic formula holds as $|x| \to \infty$

$$u^s(x) = (u - u^i)(x) = -\frac{i}{4} \sum_{n\in\mathbb{Z}} H_n^{(1)}(k_m|x|)e^{in\theta_x} \int_{\partial D} J_n(k_m|y|)e^{-in\theta_y}\psi(y)\, \mathrm{d}\sigma(y),$$

where $x = (|x|, \theta_x)$ in polar coordinates, $H_n^{(1)}$ is the Hankel function of the first kind and order n, J_n is the Bessel function of order n and ψ is the solution to (2.4).

For $u^i(x) = e^{ik_m d\cdot x}$ we have

$$u^i(x) = \sum_{m\in\mathbb{Z}} a_m(u^i) J_m(k_m|x|)e^{im\theta_x},$$

where $a_m(u^i) = e^{im(\frac{\pi}{2}-\theta_d)}$. By the superposition principle, we get

$$\psi = \sum_{m\in\mathbb{Z}} a_m(u^i)\psi_m,$$

where ψ_m is solution to (2.4) replacing f by

$$f^{(m)} := F_2^{(m)} + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \left(\mathcal{K}_D^{k_c}\right)^*\right)\left(\mathcal{S}_D^{k_c}\right)^{-1}\left[F_1^{(m)}\right]$$

with

$$F_1^{(m)}(x) = -J_m(k_m|x|)e^{im\theta_x},$$
$$F_2^{(m)}(x) = -\frac{1}{\mu_m}\frac{\partial J_m(k_m|x|)e^{im\theta_x}}{\partial\nu}.$$

We have

$$u^s(x) = (u - u^i)(x) = -\frac{i}{4}\sum_{n\in\mathbb{Z}} H_n^{(1)}(k_m|x|)e^{in\theta_x}\sum_{m\in\mathbb{Z}} W_{nm}e^{im(\frac{\pi}{2}-\theta_d)},$$

where

$$W_{nm} = \int_{\partial D} J_n(k_m|y|)e^{-in\theta_y}\psi_m(y)\,\mathrm{d}\sigma(y). \tag{5.1}$$

The coefficients W_{nm} are called the scattering coefficients.

Lemma 5.1. *In the space $\mathcal{H}^*(\partial D)$, as ω goes to zero, we have*

$$\begin{aligned} f^{(0)} &= O(\omega^2), \\ f^{(\pm 1)} &= \omega f_1^{(\pm 1)} + O(\omega^2), \\ f^{(m)} &= O(\omega^m), \quad |m| > 1, \end{aligned}$$

where

$$f_1^{(\pm 1)} = \mp \frac{\sqrt{\varepsilon_m \mu_m}}{2}\Big(\frac{1}{\mu_m}e^{i\pm\theta_\nu} + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}_D^*\right)\widetilde{\mathcal{S}}_D^{-1}\left[|x|e^{i\pm\theta_x}\right]\Big).$$

Proof. Recall that $J_0(x) = 1 + O(x^2)$. By virtue of the fact that

$$\left(\frac{1}{2}Id - \left(\mathcal{K}_D^{k_c}\right)^*\right)\left(\mathcal{S}_D^{k_c}\right)^{-1}[\chi(\partial D)] = O(\omega^2),$$

we arrive at the estimate for $f^{(0)}$ (see "Appendix C"). Moreover,

$$J_{\pm 1}(x) = \pm\frac{x}{2} + O(x^3),$$

together with the fact that

$$\left(\frac{1}{2}Id - \left(\mathcal{K}_D^{k_c}\right)^*\right)\left(\mathcal{S}_D^{k_c}\right)^{-1} = \left(\frac{1}{2}Id - \mathcal{K}_D^*\right)\widetilde{\mathcal{S}}_D^{-1} + O(\omega^2\log\omega),$$

gives the expansion of $f^{(\pm 1)}$ in terms of ω (see "Appendix C").

Finally, $J_m(x) = O(x^m)$ immediately yields the desired estimate for $f^{(m)}$. □

From Theorem C.1, it is easy to see that

$$\psi_m = \sum_{j\in J}\frac{\left(f^{(m)}, \widetilde{\varphi}_j(\omega)\right)_{\mathcal{H}^*}\varphi_j(\omega)}{\tau_j(\omega)} + \mathcal{A}_D(\omega)^{-1}(P_{J^c}(\omega)f). \tag{5.2}$$

Hence, from the definition of the scattering coefficients,

$$\begin{aligned} W_{nm} = \sum_{j\in J}&\frac{\left(f^{(m)}, \widetilde{\varphi}_j(\omega)\right)_{\mathcal{H}^*}\left(\varphi_j(\omega), J_n(k_m|x|)e^{-in\theta_x}\right)_{-\frac{1}{2},\frac{1}{2}}}{\tau_j(\omega)} \\ &+ \int_{\partial D} J_n(k_m|y|)e^{-in\theta_y}O(\omega)\,\mathrm{d}\sigma(y). \end{aligned} \tag{5.3}$$

Since

$$J_m(x) \sim \frac{1}{\sqrt{(2\pi|m|)}}\left(\frac{ex}{2|m|}\right)^{|m|}$$

as $m \to \infty$, we have

$$|f^{(m)}| \leqq \frac{C^{|m|}}{|m|^{|m|}}.$$

Using the Cauchy-Schwarz inequality and Lemma 5.1, we obtain the following result:

Proposition 5.1. *For $|n|, |m| > 0$, we have*

$$|W_{nm}| \leqq \frac{O\left(\omega^{|n|+|m|}\right)}{\min_{j\in J} |\tau_j(\omega)|} \frac{C^{|n|+|m|}}{|n|^{|n|}|m|^{|m|}}$$

for a positive constant C independent of ω.

5.2. The leading-order term in the expansion of the scattering amplitude

In the following, we analyze the first-order scattering coefficients.

Lemma 5.2. *Assume that Conditions 1 and 2 hold. Then,*

$$\psi_0 = \sum_{j\in J} \frac{O(\omega^2)}{\tau_j(\omega)} + O(\omega),$$

$$\psi_{\pm 1} = \sum_{j\in J} \frac{\pm\omega\frac{\sqrt{\varepsilon_m\mu_m}}{2}\left(\frac{1}{\mu_m} - \frac{1}{\mu_c}\right)(e^{\pm i\theta_v}, \varphi_j)_{\mathcal{H}^*}\varphi_j + O(\omega^3\log\omega)}{\tau_j(\omega)} + O(\omega).$$

Proof. The expression of ψ_0 follows from (5.2) and Lemma 5.1. Changing $\mathcal{S}_D$ by $\widetilde{\mathcal{S}}_D$ in Theorem 2.1 gives $\left((\frac{1}{2}Id - \mathcal{K}_D^*)\widetilde{\mathcal{S}}_D^{-1}[|x|e^{i\theta_x}], \varphi_j\right)_{\mathcal{H}^*} = -(e^{i\theta_v}, \varphi_j)_{\mathcal{H}^*}$. Using now Lemma 5.1 in (5.2) yields the expression of $\psi_{\pm 1}$. □

Recall that in two dimensions,

$$\tau_j(\omega) = \frac{1}{2\mu_m} + \frac{1}{2\mu_c} - \left(\frac{1}{\mu_c} - \frac{1}{\mu_m}\right)\lambda_j + O(\omega^2\log\omega),$$

where λ_j is an eigenvalue of $\mathcal{K}_D^*$ and $\lambda_0 = 1/2$. Recall also that for $0 \in J$ we need $\tau_j \to 0$ and so $\mu_m \to \infty$, which is a limiting case that we can ignore. In practice, $P_J(\omega)[\varphi_0(\omega)] = 0$. We also have $(\varphi_j, \chi(\partial D))_{-\frac{1}{2},\frac{1}{2}} = 0$ for $j \neq 0$.

It follows then from the above lemmas and the expression (5.3) of the scattering coefficients that

$$W_{00} = \sum_{j\in J} \frac{O(\omega^4\log\omega)}{\tau_j(\omega)} + O(\omega),$$

$$W_{0\pm 1} = \sum_{j\in J} \frac{O(\omega^3\log\omega)}{\tau_j(\omega)} + O(\omega),$$

$$W_{\pm 10} = \sum_{j \in J} \frac{O(\omega^3)}{\tau_j(\omega)} + O(\omega^2).$$

Note that $W_{\pm 1 \pm 1}$ has a special structure. Indeed, from Lemma 5.2 and equation (5.3), we have

$$\begin{aligned}
&W_{\pm 1 \pm 1} \\
&= \sum_{j \in J} \frac{\pm \pm \, \omega \frac{\sqrt{\varepsilon_m \mu_m}}{2} \left(\frac{1}{\mu_m} - \frac{1}{\mu_c} \right) \left(\varphi_j, J_1(k_m |x|) e^{\mp i\theta_x} \right)_{-\frac{1}{2}, \frac{1}{2}} \left(e^{\pm i\theta_\nu}, \varphi_j \right)_{\mathcal{H}^*} + O(\omega^4 \log \omega)}{\tau_j(\omega)} \\
&\quad + O(\omega^2), \\
&= \sum_{j \in J} \frac{\pm \pm \, \omega^2 \frac{\varepsilon_m \mu_m}{4} \left(\frac{1}{\mu_m} - \frac{1}{\mu_c} \right) \left(\varphi_j, |x| e^{\mp i\theta_x} \right)_{-\frac{1}{2}, \frac{1}{2}} \left(e^{\pm i\theta_\nu}, \varphi_j \right)_{\mathcal{H}^*} + O(\omega^4 \log \omega)}{\tau_j(\omega)} \\
&\quad + O(\omega^2), \\
&= \frac{k_m^2}{4} \left(\sum_{j \in J} \frac{\pm \pm \left(\varphi_j, |x| e^{\mp i\theta_x} \right)_{-\frac{1}{2}, \frac{1}{2}} \left(e^{\pm i\theta_\nu}, \varphi_j \right)_{\mathcal{H}^*} + O(\omega^2 \log \omega)}{\lambda - \lambda_j + O(\omega^2 \log \omega)} + O(1) \right),
\end{aligned}$$

where λ is defined by (2.19). Now, assume that $\min_{j \in J} |\tau_j(\omega)| \gg \omega^2 \log \omega$. Then,

$$W_{\pm 1 \pm 1} = \frac{k_m^2}{4} \left(\sum_{j \in J} \frac{\pm \pm \left(\varphi_j, |x| e^{\mp i\theta_x} \right)_{-\frac{1}{2}, \frac{1}{2}} \left(e^{\pm i\theta_\nu}, \varphi_j \right)_{\mathcal{H}^*}}{\lambda - \lambda_j} + O(1) \right). \tag{5.4}$$

Define the contracted polarization tensors by

$$N_{\pm,\pm}(\lambda, D) := \int_{\partial D} |x| e^{\pm i\theta_x} (\lambda I - \mathcal{K}_D^*)^{-1} [e^{\pm i\theta_\nu}](x) \, \mathrm{d}\sigma(x).$$

It is clear that

$$\begin{aligned}
N_{+,+}(\lambda, D) &= M_{1,1}(\lambda, D) - M_{2,2}(\lambda, D) + i2M_{1,2}(\lambda, D), \\
N_{+,-}(\lambda, D) &= M_{1,1}(\lambda, D) + M_{2,2}(\lambda, D), \\
N_{-,+}(\lambda, D) &= M_{1,1}(\lambda, D) + M_{2,2}(\lambda, D), \\
N_{-,-}(\lambda, D) &= M_{1,1}(\lambda, D) - M_{2,2}(\lambda, D) - i2M_{1,2}(\lambda, D),
\end{aligned}$$

where $M_{l,m}(\lambda, D)$ is the (l, m)—entry of the polarization tensor given by (4.7).

Finally, considering the above we can state the following result.

Theorem 5.1. *Let A_∞ be the scattering amplitude in the far-field defined in (4.2) for the incoming plane wave $u^i(x) = e^{ik_m d \cdot x}$. Assume Conditions* 1 *and* 2 *and*

$$\min_{j \in J} |\tau_j(\omega)| \gg \omega^2 \log \omega.$$

Then, A_∞ admits the following asymptotic expansion

$$A_\infty\left(\frac{x}{|x|} \right) = \frac{x}{|x|}^T W_1 d + O(\omega^2),$$

where

$$W_1 = \begin{pmatrix} W_{-11} + W_{1-1} - 2W_{11} & i\left(W_{1-1} - W_{-11}\right) \\ i\left(W_{1-1} - W_{-11}\right) & -W_{-11} - W_{1-1} - 2W_{11} \end{pmatrix}.$$

Here, W_{nm} are the scattering coefficients defined by (5.1).

Proof. From (4.12), we have

$$A_\infty\left(\frac{x}{|x|}\right) = -k_m^2 \frac{x}{|x|}^T M(\lambda, D)d.$$

Since $\mathcal{K}_D^*$ is compact and self-adjoint in $\mathcal{H}^*$, we have

$$\begin{aligned} N_{\pm,\pm}(\lambda, D) &= \sum_{j=1}^{\infty} \frac{\left(\varphi_j, |x|e^{\pm i\theta_x}\right)_{-\frac{1}{2},\frac{1}{2}} \left(e^{\pm i\theta_\nu}, \varphi_j\right)_{\mathcal{H}^*}}{\lambda - \lambda_j} \\ &= \sum_{j\in J} \frac{\left(\varphi_j, |x|e^{\pm i\theta_x}\right)_{-\frac{1}{2},\frac{1}{2}} \left(e^{\pm i\theta_\nu}, \varphi_j\right)_{\mathcal{H}^*}}{\lambda - \lambda_j} + O(1). \end{aligned}$$

We have then from (5.4) that

$$\begin{aligned} -\frac{k_m^2}{4} N_{+,+}(\lambda, D) &= W_{-11} + O(\omega^2), \\ -\frac{k_m^2}{4} N_{+,-}(\lambda, D) &= -W_{11} + O(\omega^2), \\ -\frac{k_m^2}{4} N_{-,+}(\lambda, D) &= -W_{11} + O(\omega^2), \\ -\frac{k_m^2}{4} N_{-,-}(\lambda, D) &= W_{1-1} + O(\omega^2). \end{aligned}$$

In view of

$$\begin{aligned} M_{11} &= \frac{1}{4}\left(N_{+,+} + N_{-,-} + 2N_{+,-}\right), \\ M_{22} &= \frac{1}{4}\left(-N_{+,+} - N_{-,-} + 2N_{+,-}\right), \\ M_{12} &= \frac{-i}{4}\left(N_{+,+} - N_{-,-}\right), \end{aligned}$$

we get the result. □

6. Super-Resolution (Super-Focusing) by Using Plasmonic Particles

It is known that the resolution limit (or the diffraction limit) in a general inhomogeneous space is determined by the imaginary part of the Green function in the associated space [1]. By modifying the homogeneous spaces with subwavelength resonators, we can introduce propagating subwavelength resonance modes to the space which encode subwavelength information in a neighborhood of the space embedded by the subwavelenghth resonators, thus yielding a Green's function whose imaginary part exhibits subwavelength peaks and therefore breaks the resolution limit (or diffraction limit) in the homogeneous space. The principle has been mathematically demonstrated in [12]. Here, using the fact that plasmonic particles are ideal subwavelength resonators, we consider the possibility of super-resolution (super-focusing) by using a system of identical plasmonic particles. The results in this section can be viewed as a consequence of the results in Section 3. The mechanism of super-resolution in the case considered in this section is due to propagating subwavelength resonant modes that are generated by weakly coupled plasmonic particles. For non-resonant small particles, no subwavelength resonance can be excited and hence no super-resolution can be achieved. The analysis here is for a point source and can be easily extended by a convolution argument to general sources.

6.1. Asymptotic expansion of the scattered field

In order to illustrate the superfocusing phenomenon, we set

$$u^i(x) = G(x, x_0, k_m) = -\frac{e^{ik_m|x-x_0|}}{4\pi|x-x_0|}.$$

Lemma 6.1. *In the space $\mathcal{H}^*(\partial D)$, as ω goes to zero, we have*

$$f = f_0 + O(\omega\delta^{\frac{3}{2}}) + O(\delta^{\frac{5}{2}}),$$

where $f_0 = (f_{0,1}, \dots, f_{0,L})^T$ with

$$\begin{aligned} f_{0,l} = &-\frac{1}{4\pi|z_l - x_0|^3} \\ &\left(\frac{1}{\mu_m}(z_l - x_0)\cdot\nu(x) + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}^*_{D_l}\right)\mathcal{S}^{-1}_{D_l}\left[(z_l - x_0)\cdot(x - z_l)\right]\right) \\ = &\, O\left(\delta^{\frac{3}{2}}\right). \end{aligned}$$

Proof. The proof is similar to that of Lemma 2.6. Recall that

$$f_l = F_{l,2} + \frac{1}{\mu_c}\left(\frac{1}{2}Id - (\mathcal{K}^{k_c}_{D_l})^*\right)(\mathcal{S}^{k_c}_{D_l})^{-1}[F_{l,1}].$$

We can show that

$$F_{l,2} = -\frac{1}{\mu_m}\frac{\partial u^i}{\partial \nu} = -\frac{1}{4\pi\mu_m|z_l - x_0|^3}(z_l - x_0)\cdot\nu(x) + O\left(\delta^{\frac{5}{2}}\right)$$

$$+O(\omega\delta^{\frac{3}{2}}) \quad \text{in } \mathcal{H}^*(\partial D_l).$$

Besides,

$$\begin{aligned} u^i(x)|_{\partial D_l} = &-\frac{e^{ik_m|z_l-x_0|}}{4\pi|z_l-x_0|}\chi(\partial D_l) + \frac{1}{4\pi|z_l-x_0|^3}(z_l-x_0)\cdot(x-z_l) + O\left(\delta^{\frac{5}{2}}\right) \\ &+O(\omega\delta^{\frac{3}{2}}) \quad \text{in } \mathcal{H}(\partial D_l). \end{aligned}$$

Using the identity $(\frac{1}{2}Id - \mathcal{K}^*_{D_l})\mathcal{S}^{-1}_{D_l}[\chi(\partial D_l)] = 0$, we obtain that

$$\begin{aligned} &\frac{1}{\mu_c}\left(\frac{1}{2}Id - \left(\mathcal{K}^{k_c}_{D_l}\right)^*\right)\left(\mathcal{S}^{k_c}_{D_l}\right)^{-1}[F_{l,1}] \\ &\quad = -\frac{1}{4\pi|z_l-x_0|^3\mu_c}\left(\frac{1}{2}Id - \mathcal{K}^*_{D_l}\right)\mathcal{S}^{-1}_{D_l}\left[(z_l-x_0)\cdot(x-z_l)\right]. \end{aligned}$$

This completes the proof of the lemma. □

We now derive an asymptotic expansion of the scattered field in an intermediate regime which is neither too close to the plasmonic particles nor too far away. More precisely, letting C be a fixed, sufficiently large positive number, we consider the domain

$$\mathrm{D}_{\delta,k,C} = \Big\{x \in \mathbb{R}^3;\ \min_{1\leqq l\leqq L}|x-z_l| \geqq C\delta,\ \ \max_{1\leqq l\leqq L}|x-z_l| \leqq \frac{1}{Ck}\Big\}.$$

Lemma 6.2. *Let* $\psi_l \in \mathcal{H}^*(\partial D_l)$ *and let* $v(x) = \mathcal{S}^k_{D_l}[\psi_l](x)$. *Then we have for* $x \in \mathrm{D}_{\delta,k,C}$,

$$\begin{aligned} v(x) = &\, G(x,z_l,k)\left(\frac{1}{|x-z_l|} - ik\right)\frac{x-z_l}{|x-z_l|}\cdot\int_{\partial D_0} y\psi_l(y)d\sigma(y) \\ &+O(\delta^{\frac{5}{2}})\|\psi_l\|_{\mathcal{H}^*(\partial D_l)} + G(x,z_l,k)\int_{\partial D_0}\psi_l(y)d\sigma(y). \end{aligned}$$

Moreover, the following estimates hold:

$$\begin{aligned} v(x) &= O(\delta^{\frac{3}{2}}) \quad \text{if } \int_{\partial D_0}\psi_l(y)\,\mathrm{d}\sigma(y) = 0, \\ v(x) &= O(\delta^{\frac{1}{2}}) \quad \text{if } \int_{\partial D_0}\psi_l(y)\,\mathrm{d}\sigma(y) \neq 0. \end{aligned}$$

Proof. We only consider the case when $l = 1$. The other case follows similarly or by coordinate translation. We have

$$\begin{aligned} v(x) = \mathcal{S}^k_D[\psi](x) &= \int_{\partial D_0} G(x,y,k)\psi(y)\,\mathrm{d}\sigma(y) \\ &= -\int_{\partial D_0}\frac{e^{ik|x-y|}}{4\pi|x-y|}\psi(y)\,\mathrm{d}\sigma(y). \end{aligned}$$

Since

$$G(x, y, k) = G(x, 0, k) + \sum_{|\alpha=1|} \frac{\partial G(x, 0, k)}{\partial y^\alpha} y^\alpha + \sum_{m \geqq 2} \sum_{|\alpha=m|} \frac{\partial^m G(x, 0, k)}{\partial y^\alpha} y^\alpha,$$

and

$$\frac{\partial G(x, 0, k)}{\partial y^\alpha} = -\frac{e^{ik|x|}}{4\pi |x|} \left(\frac{1}{|x|} - ik \right) \frac{x}{|x|} = G(x, 0, k) \left(\frac{1}{|x|} - ik \right) \frac{x^\alpha}{|x|},$$

we obtain the required identity for the case $l = 1$. The estimate follows from the fact that

$$\| y^\alpha \|_{\mathcal{H}(\partial D_0)} = O\left(\delta^{\frac{2|\alpha|+1}{2}} \right).$$

This completes the proof of the lemma. □

Denote by

$$\begin{aligned}
S_{j,l}(x, k) &= G(x, z_l, k) \frac{x - z_l}{|x - z_l|^2} \cdot \int_{\partial D_0} y \varphi_j(y) \, \mathrm{d}\sigma(y), \\
S_l(x, k) &= G(x, z_l, k) \int_{\partial D_0} \varphi_0(y) \, \mathrm{d}\sigma(y), \\
H_{j,l}(x_0) &= -\frac{1}{4\pi |z_l - x_0|^3} \big((z_l - x_0) \cdot \nu(x), \varphi_j \big)_{\mathcal{H}^*(\partial D_0)}.
\end{aligned}$$

It is clear that the following size estimates hold:

$$\begin{aligned}
S_{j,l}(x, k) &= O(\delta^{\frac{3}{2}}), \quad S_l(x, k) = O(\delta^{\frac{1}{2}}), \\
H_{j,l}(x_0) &= O(\delta^{\frac{3}{2}}) \quad \text{for } j \neq 0, \quad H_{O,l}(x_0) = 0.
\end{aligned}$$

Theorem 6.1. *Under Conditions 1, 2, 3, 4 and 6, the Green function $\Gamma(x, x_0, k_m)$ in the presence of L plasmonic particles has the following representation in the quasi-static regime: for $x \in \mathrm{D}_{\delta,k_m,C}$,*

$$\begin{aligned}
\Gamma(x, x_0, k_m) &= G(x, x_0, k_m) \\
&\quad + \sum_{j \in J} \sum_{l=1}^{L} \frac{H_{j,p}(x_0) \widetilde{X}_{j,l,p} X_{j,l,q} S_{j,q}(x, k_m) + O(\delta^4) + O(\omega \delta^3)}{\lambda - \lambda_j + \left(\frac{1}{\mu_c} - \frac{1}{\mu_m} \right)^{-1} \tau_{j,l} + O(\delta^4) + O(\delta^2 \omega^2)} \\
&\quad + O(\delta^3).
\end{aligned}$$

Proof. With $u^i(x) = G(x, x_0, k_m)$, we have

$$\psi = \sum_{j \in J} \sum_{1 \leqq l \leqq L} a_{j,l} \varphi_{j,l} + \sum_{1 \leqq l \leqq L} a_{0,l} \varphi_{0,l} + O\left(\delta^{\frac{3}{2}} \right),$$

where

$$a_{j,l} = (f, \widetilde{\varphi}_{j,l})_{\mathcal{H}^*(\partial D)} = (f_0, \widetilde{\varphi}_{j,l})_{\mathcal{H}^*(\partial D)} + O(\omega \delta^{\frac{3}{2}}) + O\left(\delta^{\frac{5}{2}} \right),$$

$$= \left(\frac{1}{\mu_c} - \frac{1}{\mu_m} \right) \widetilde{X}_{j,l,p} H_{j,p}(x_0) + O(\omega \delta^{\frac{3}{2}}) + O\left(\delta^{\frac{5}{2}}\right),$$
$$a_{0,l} = (f, \widetilde{\varphi}_{0,l})_{\mathcal{H}^*(\partial D)} = O\left(\delta^{\frac{5}{2}}\right).$$

By Lemma 6.2,

$$\mathcal{S}_D^{k_m}[\varphi_{j,l}](x) = \sum_{1 \leqq p \leqq L} \mathcal{S}_D^{k_m}\left[X_{j,l,p} \varphi_j e_p\right](x) = \sum_{1 \leqq p \leqq L} X_{j,l,p} \mathcal{S}_{D_p}^{k_m}[\varphi_j](x)$$
$$= \sum_{1 \leqq p \leqq L} X_{j,l,p} S_{j,p}(x, k_m) + O\left(\delta^{\frac{5}{2}}\right) + O\left(\omega \delta^{\frac{3}{2}}\right).$$

On the other hand, for $j = 0$, we have

$$\mathcal{S}_D^{k_m}[\varphi_{0,l}](x) = O\left(\delta^{\frac{1}{2}}\right),$$
$$\tau_{0,l}(\omega) = \tau_0 + O(\delta^4) + O\left(\delta^2 \omega^2\right) = O(1).$$

Therefore, we can deduce that

$$u^s = \mathcal{S}_D^{k_m}[\psi](x) = \sum_{j \in J} \sum_{1 \leqq l \leqq L} a_{j,l} \mathcal{S}_D^{k_m}[\varphi_{j,l}] + \sum_{1 \leqq l \leqq L} a_{0,l} \mathcal{S}_D^{k_m}[\varphi_{0,l}] + O(\delta^3),$$
$$= \sum_{j \in J} \sum_{l=1}^{L} \frac{1}{\tau_{j,l}(\omega)} \Big(\left(\frac{1}{\mu_c} - \frac{1}{\mu_m} \right) H_{j,p}(x_0) \widetilde{X}_{j,l,p} X_{j,l,q} S_{j,q}(x, k_m) + O(\omega \delta^3) + O(\delta^4) \Big)$$
$$+ O(\delta^3),$$
$$= \sum_{j \in J} \sum_{l=1}^{L} \frac{H_{j,p}(x_0) \widetilde{X}_{j,l,p} X_{j,l,q} S_{j,q}(x, k_m) + O(\omega \delta^3) + O(\delta^4)}{\lambda - \lambda_j + \left(\frac{1}{\mu_c} - \frac{1}{\mu_m}\right)^{-1} \tau_{j,l} + O(\delta^4) + O(\delta^2 \omega^2)} + O(\delta^3).$$

□

6.2. Asymptotic expansion of the imaginary part of the Green function

As a consequence of Theorem 6.1, we obtain the following result on the imaginary part of the Green function:

Theorem 6.2. *Assume the same conditions as in Theorem 6.1. Under the additional assumption that*

$$\lambda - \lambda_j + \left(\frac{1}{\mu_c} - \frac{1}{\mu_m} \right)^{-1} \tau_{j,l} \gg O(\delta^4) + O(\delta^2 \omega^2),$$
$$\Re \left(\lambda - \lambda_j + \left(\frac{1}{\mu_c} - \frac{1}{\mu_m} \right)^{-1} \tau_{j,l} \right) \lesssim \Im \left(\lambda - \lambda_j + \left(\frac{1}{\mu_c} - \frac{1}{\mu_m} \right)^{-1} \tau_{j,l} \right)$$

for each l and $j \in J$, we have

$$\Im \Gamma(x, x_0, k_m) = \Im G(x, x_0, k_m) + O(\delta^3) +$$

$$\sum_{j\in J}\sum_{l=1}^{L}\Re\Big(H_{j,p}(x_0)\widetilde{X}_{j,l,p}X_{j,l,q}S_{j,q}(x,0)+O(\omega\delta^3)+O(\delta^4)\Big)$$
$$\times\Im\left(\frac{1}{\lambda-\lambda_j+\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)^{-1}\tau_{j,l}}\right),$$

where $x, x_0 \in \mathrm{D}_{\delta,k_m,C}$.

Note that $\Re\big(H_{j,p}(x_0)\widetilde{X}_{j,l,p}X_{j,l,q}S_{j,q}(x,0)\big) = O(\delta^3)$. Under the conditions in Theorem 6.2, if we have additionally that

$$\Im\left(\frac{1}{\lambda-\lambda_j+\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)^{-1}\tau_{j,l}}\right)=O\left(\frac{1}{\delta^3}\right)$$

for some plasmonic frequency ω, then the term in the expansion of $\Im\Gamma(x, x_0, k_m)$ which is due to resonance has size one and exhibits subwavelength peak with width of order one. This breaks the diffraction limit $1/k_m$ in the free space. We also note that the term $\Im G(x, x_0, k_m)$ has size $O(\omega)$. Thus, we can conclude that super-resolution (super-focusing) can indeed be achieved by using a system of plasmonic particles.

7. Concluding Remarks

In this paper, based on perturbation arguments, we studied the scattering by plasmonic nanoparticles when the frequency is close to a resonant frequency. We have shown that plasmon resonant nanoparticles provide a possible way not only of super-resolved imaging but also of scattering and absorption enhancements.

We have derived the shift and broadening of the plasmon resonance with changes in size. We have also consider the case of multiple nanoparticles under the weak interaction assumption. The localization algorithms developed in [7,8,20] can be extended to the problem of imaging plasmonic nanoparticles. We have precisely quantified the scattering and absorption cross-section enhancements and gave optimal bounds on the enhancement factors. We have also linked the plasmonic resonances to the scattering coefficients and showed that the leading-order term of the scattering amplitude can be expressed in terms of the ±-one order of the scattering coefficients.

The generalization to the full Maxwell equations of the methods and results of the paper is under consideration and will be reported elsewhere. Another challenging problem will be to optimize the super-focusing phenomenon in terms of the organization of the nanoparticles. This will also be the subject of a forthcoming publication.

Appendix A: Asymptotic Expansion of the Integral Operators: Single Particle

In this section, we derive asymptotic expansions with respect to k of some boundary integral operators defined on the boundary of a bounded and simply connected smooth domain D in dimension three whose size is of order one.

We first consider the single layer potential

$$\mathcal{S}_D^k[\psi](x) = \int_{\partial D} G(x, y, k)\psi(y)\,\mathrm{d}\sigma(y), \quad x \in \partial D,$$

where

$$G(x, y, k) = -\frac{e^{ik|x-y|}}{4\pi|x-y|}$$

is the Green function of Helmholtz equation in $\mathbb{R}^3$, subject to the Sommerfeld radiation condition. Note that

$$G(x, y, k) = -\sum_{j=0}^{\infty} \frac{(ik|x-y|)^j}{j!4\pi|x-y|} = -\frac{1}{4\pi|x-y|} - \frac{ik}{4\pi}\sum_{j=1}^{\infty}\frac{(ik|x-y|)^{j-1}}{j!}.$$

We get

$$\mathcal{S}_D^k = \mathcal{S}_D + \sum_{j=1}^{\infty} k^j \mathcal{S}_{D,j}, \tag{A.1}$$

where

$$\mathcal{S}_{D,j}[\psi](x) = -\frac{i}{4\pi}\int_{\partial D}\frac{(i|x-y|)^{j-1}}{j!}\psi(y)\,\mathrm{d}\sigma(y).$$

In particular, we have

$$\mathcal{S}_{D,1}[\psi](x) = -\frac{i}{4\pi}\int_{\partial D}\psi(y)\,\mathrm{d}\sigma(y), \tag{A.2}$$

$$\mathcal{S}_{D,2}[\psi](x) = -\frac{1}{4\pi}\int_{\partial D}|x-y|\psi(y)\,\mathrm{d}\sigma(y). \tag{A.3}$$

Lemma A.1. $\|\mathcal{S}_{D,j}\|_{\mathcal{L}((\mathcal{H}^*(\partial D),\mathcal{H}(\partial D))}$ *is uniformly bounded with respect to* j. *Moreover, the series in (A.1) is convergent in* $\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}(\partial D))$.

Proof. It is clear that

$$\|\mathcal{S}_{D,j}\|_{\mathcal{L}(L^2(\partial D),H^1(\partial D))} \leqq C,$$

where C is independent of j. On the other hand, a similar estimate also holds for the operator $\mathcal{S}_{D,j}^*$. It follows that

$$\|\mathcal{S}_{D,j}\|_{\mathcal{L}(H^{-1}(\partial D),L^2(\partial D))} \leqq C.$$

Thus, we can conclude that $\|\mathcal{S}_{D,j}\|_{\mathcal{L}(H^{-\frac{1}{2}}(\partial D),H^{\frac{1}{2}}(\partial D))}$ is uniformly bounded by using interpolation theory. By the equivalence of norms in the $H^{-\frac{1}{2}}(\partial D)$ and $H^{\frac{1}{2}}(\partial D)$, the lemma follows immediately. □

Note that $\mathcal{S}_D$ is invertible in dimension three, so is $\mathcal{S}_D^k$ for small k. By formally writing

$$(\mathcal{S}_D^k)^{-1} = \mathcal{S}_D^{-1} + k\mathcal{B}_{D,1} + k^2\mathcal{B}_{D,2} + \dots, \tag{A.4}$$

and using the identity $(\mathcal{S}_D^k)^{-1}\mathcal{S}_D^k = Id$, we can derive that

$$\mathcal{B}_{D,1} = -\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\mathcal{S}_D^{-1}, \quad \mathcal{B}_{D,2} = -\mathcal{S}_D^{-1}\mathcal{S}_{D,2}\mathcal{S}_D^{-1} + \mathcal{S}_D^{-1}\mathcal{S}_{D,1}\mathcal{S}_D^{-1}\mathcal{S}_{D,1}\mathcal{S}_D^{-1}. \tag{A.5}$$

We can also derive the other lower-order terms $\mathcal{B}_{D,j}$.

Lemma A.2. *The series in* (A.4) *converges in* $\mathcal{L}(\mathcal{H}(\partial D), \mathcal{H}^*(\partial D))$ *for sufficiently small* k.

Proof. The proof can be deduced from the identity

$$\left(\mathcal{S}_D^k\right)^{-1} = \left(Id + \mathcal{S}_D^{-1}\sum_{j=1}^{\infty} k^j \mathcal{S}_{D,j}\right)^{-1} \mathcal{S}_D^{-1}.$$

□

We now consider the expansion for the boundary integral operator $(\mathcal{K}_D^k)^*$. We have

$$\left(\mathcal{K}_D^k\right)^* = \mathcal{K}_D^* + k\mathcal{K}_{D,1} + k^2\mathcal{K}_{D,2} + \dots, \tag{A.6}$$

where

$$\begin{aligned}\mathcal{K}_{D,j}[\psi](x) &= -\frac{i}{4\pi}\int_{\partial D} \frac{\partial (i|x-y|)^{j-1}}{j!\partial\nu(x)}\psi(y)\,\mathrm{d}\sigma(y)\\ &= -\frac{i^j(j-1)}{4\pi j!}\int_{\partial D} |x-y|^{j-3}(x-y)\cdot\nu(x)\psi(y)\,\mathrm{d}\sigma(y).\end{aligned}$$

In particular, we have

$$\mathcal{K}_{D,1} = 0, \quad \mathcal{K}_{D,2}[\psi](x) = \frac{1}{4\pi}\int_{\partial D} \frac{(x-y)\cdot\nu(x)}{|x-y|}\psi(y)\,\mathrm{d}\sigma(y). \tag{A.7}$$

Lemma A.3. *The norm* $\|\mathcal{K}_{D,j}\|_{\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}^*(\partial D))}$ *is uniformly bounded for* $j \geqq 1$. *Moreover, the series in (A.6) is convergent in* $\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}^*(\partial D))$.

Appendix B: Asymptotic Expansion of the Integral Operators: Multiple Particles

In this section, we consider the three-dimensional case. We assume that the particles have size of order δ which is a small number and the distance between them is of order one. We write $D_j = z_j + \delta\widetilde{D}$, $j = 1, 2, \dots, M$, where $\widetilde{D}$ has size one and is centered at the origin. Our goal is to derive estimates for various boundary integral operators considered in the paper that are defined on small particles in terms of their size. For this purpose, we denote by $D_0 = \delta\widetilde{D}$. For each function f defined on ∂D_0, we define a corresponding function on $\widetilde{D}$ by

$$\eta(f)(\widetilde{x}) = f(\delta\widetilde{x}).$$

We first state some useful results.

Lemma B.1. *The following scaling properties hold:*

(i) $\|\eta(f)\|_{L^2(\partial\widetilde{D})} = \delta^{-1}\|f\|_{L^2(\partial D_0)}$*;*

(ii) $\|\eta(f)\|_{\mathcal{H}(\partial\widetilde{D})} = \delta^{-\frac{1}{2}}\|f\|_{\mathcal{H}(\partial D_0)}$*;*

(iii) $\|\eta(f)\|_{\mathcal{H}^*(\partial\widetilde{D})} = \delta^{-\frac{3}{2}}\|f\|_{\mathcal{H}^*(\partial D_0)}$*.*

Proof. The proof of (i) is straightforward and we only need to prove (ii) and (iii). To prove (iii), we have

$$\begin{aligned}\|f\|^2_{\mathcal{H}^*(\partial D_0)} &= \int_{\partial D_0}\int_{\partial D_0} \frac{f(x)f(y)}{4\pi|x-y|}\,\mathrm{d}\sigma(x)\,\mathrm{d}\sigma(y)\\ &= \delta^3 \int_{\partial\widetilde{D}}\int_{\partial\widetilde{D}} \frac{\eta(f)(\widetilde{x})\eta(f)(\widetilde{y})}{4\pi|\widetilde{x}-\widetilde{y}|}\,\mathrm{d}\sigma(\widetilde{x})\,\mathrm{d}\sigma(\widetilde{x})\\ &= \delta^3\|\eta(f)\|^2_{\mathcal{H}^*(\partial\widetilde{D})},\end{aligned}$$

whence (iii) follows. To prove (ii), recall that

$$\|f\|_{\mathcal{H}(\partial D_0)} = \|\mathcal{S}_{D_0}^{-1} f\|_{\mathcal{H}^*(\partial D_0)}.$$

Let $u = \mathcal{S}_{D_0}^{-1}[f]$. Then $f = \mathcal{S}_{D_0}[u]$. We can show that

$$\eta(f) = \delta\mathcal{S}_{\widetilde{D}}(\eta(u)).$$

As a result, we have

$$\begin{aligned}\|\eta(f)\|_{\mathcal{H}(\partial\widetilde{D})} &= \delta\|\mathcal{S}_{\widetilde{D}}(\eta(u))\|_{\mathcal{H}(\partial\widetilde{D})} = \delta\|\eta(u)\|_{\mathcal{H}^*(\partial\widetilde{D})}\\ &= \delta^{-\frac{1}{2}}\|u\|_{\mathcal{H}^*(\partial D_0)} = \delta^{-\frac{1}{2}}\|f\|_{\mathcal{H}(\partial D_0)},\end{aligned}$$

which proves (ii). □

Lemma B.2. *Let X and Y be bounded and simply connected smooth domains in $\mathbb{R}^3$. Assume $0 \in X, Y$ and $X = \delta\widetilde{X}$, $Y = \delta\widetilde{Y}$. Let $\mathcal{R}$ and $\widetilde{\mathcal{R}}$ be two boundary integral operators from $\mathcal{D}'(\partial Y)$ to $\mathcal{D}'(\partial X)$ and $\mathcal{D}'(\partial\widetilde{Y})$ to $\mathcal{D}'(\partial\widetilde{X})$, respectively. Here, $\mathcal{D}'$ denotes the Schwartz space. Assume that both operators have the same Schwartz kernel R with the following homogeneous scaling property*

$$R(\delta x, \delta y) = \delta^m R(x, y).$$

Then,

$$\begin{aligned}\|\mathcal{R}\|_{\mathcal{L}(\mathcal{H}^*(\partial Y),\mathcal{H}^*(\partial X))} &= \delta^{2+m}\|\widetilde{\mathcal{R}}\|_{\mathcal{L}(\mathcal{H}^*(\partial\widetilde{Y}),\mathcal{H}^*(\partial\widetilde{X}))},\\ \|\mathcal{R}\|_{\mathcal{L}(\mathcal{H}^*(\partial Y),\mathcal{H}(\partial X))} &= \delta^{1+m}\|\widetilde{\mathcal{R}}\|_{\mathcal{L}(\mathcal{H}^*(\partial\widetilde{Y}),\mathcal{H}(\partial\widetilde{X}))}.\end{aligned}$$

Proof. The result follows from Lemma B.1 and the identity

$$\mathcal{R} = \delta^{2+m} \eta^{-1} \circ \widetilde{\mathcal{R}} \circ \eta.$$

$\square$

We first consider the operators $\mathcal{S}_{D_j}^k$ and $(\mathcal{K}_{D_j}^k)^*$. The following asymptotic expansions hold:

Lemma B.3. *(i) Regarded as operators from $\mathcal{H}^*(\partial D_j)$ into $\mathcal{H}(\partial D_j)$, we have*

$$\mathcal{S}_{D_j}^k = \mathcal{S}_{D_j} + k\mathcal{S}_{D_j,1} + k^2 \mathcal{S}_{D_j,2} + O(k^3\delta^3),$$

where $\mathcal{S}_{D_j} = O(1)$ and $\mathcal{S}_{D_j,m} = O(\delta^m)$;
(ii) Regarded as operators from $\mathcal{H}(\partial D_j)$ into $\mathcal{H}^*(\partial D_j)$, we have

$$\left(\mathcal{S}_{D_j}^k\right)^{-1} = \mathcal{S}_{D_j}^{-1} + k\mathcal{B}_{D_j,1} + k^2 \mathcal{B}_{D_j,2} + O(k^3\delta^3),$$

where $\mathcal{S}_{D_j}^{-1} = O(1)$ and $\mathcal{B}_{D_j,m} = O(\delta^m)$;
(iii) Regarded as operators from $\mathcal{H}^*(\partial D_j)$ into $\mathcal{H}^*(\partial D_j)$, we have

$$\left(\mathcal{K}_{D_j}^k\right)^* = \mathcal{K}_{D_j}^* + k^2 O(\delta^2),$$

where $\mathcal{K}_{D_j}^* = O(1)$.

Proof. The proof immediately follows from Lemmas B.2, A.1 and A.3. $\square$

We now consider the operator $\mathcal{S}_{D_j,D_l}^k$. By definition,

$$\mathcal{S}_{D_j,D_l}^k[\psi](x) = \int_{\partial D_j} G(x, y, k)\psi(y)\, \mathrm{d}\sigma(y), \quad x \in \partial D_l.$$

Using the expansion

$$G(x, y, k) = \sum_{m=0}^{\infty} k^m Q_m(x, y),$$

where

$$Q_m(x, y) = -\frac{i^m |x - y|^{m-1}}{4\pi},$$

we can derive that

$$\mathcal{S}_{D_j,D_l}^k = \sum_{m\geqq 0} k^m \mathcal{S}_{j,l,m},$$

where

$$\mathcal{S}_{j,l,m}[\psi](x) = \int_{\partial D_j} Q_m(x, y)\psi(y)\, \mathrm{d}\sigma(y).$$

We can further write

$$\mathcal{S}_{j,l,m} = \sum_{n\geqq 0} \mathcal{S}_{j,l,m,n},$$

where $\mathcal{S}_{j,l,m,n}$ is defined by

$$\begin{aligned}&\mathcal{S}_{j,l,m,n}[\psi](x)\\&\quad = \int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!} \frac{\partial^{|\alpha|+|\beta|}}{\partial x^\alpha \partial y^\beta} Q_m(z_l, z_j)(x-z_l)^\alpha (y-z_j)^\beta \psi(y)\, \mathrm{d}\sigma(y).\end{aligned}$$

In particular, we have

$$\begin{aligned}
\mathcal{S}_{j,l,0,0}[\psi](x) &= -\frac{1}{4\pi|z_j - z_l|}(\psi, \chi(\partial D_j))_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \chi(D_l),\\
\mathcal{S}_{j,l,0,1}[\psi](x) &= \sum_{|\alpha|=1} \frac{(z_l - z_j)^\alpha}{4\pi|z_l - z_j|^3}\Big((x-z_l)^\alpha (\psi, \chi(\partial D_l))_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)}\\
&\qquad + \big((y-z_j)^\alpha, \psi\big)\chi(D_l)\Big),\\
\mathcal{S}_{j,l,0,2}[\psi](x) &= \sum_{|\alpha|+|\beta|=2} \frac{1}{\alpha!\beta!} \frac{\partial^2 Q_0(z_l, z_j)}{\partial x^\alpha \partial y^\beta}(x-z_l)^\alpha (y-z_j)^\beta \psi(y)\, \mathrm{d}\sigma(y),\\
\mathcal{S}_{j,l,1}[\psi](x) &= -\frac{i}{4\pi}(\psi, \chi(\partial D_j))_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \chi(D_l),\\
\mathcal{S}_{j,l,2,0}[\psi](x) &= \frac{1}{4\pi}|z_l - z_j|(\psi, \chi(\partial D_j))_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \chi(D_l).
\end{aligned}$$

The following estimate holds:

Lemma B.4. *We have* $\|\mathcal{S}_{j,l,m,n}\|_{\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}(\partial D))} \lesssim O(\delta^{n+1})$.

Proof. After a translation of coordinates, the stated estimate immediately follows from Lemma B.2. □

Similarly, for the operator $\mathcal{K}^{k_m}_{D_j, D_l}$, defined as

$$\mathcal{K}^k_{D_j, D_l}[\psi](x) = \int_{\partial D_j} \frac{\partial G(x, y, k)}{\partial \nu(x)} \psi(y)\, \mathrm{d}\sigma(y), \quad x \in \partial D_l,$$

we have

$$\mathcal{K}^k_{D_j, D_l} = \sum_{m\geqq 0} k^m \sum_{n \geqq 0} \mathcal{K}_{j,l,m,n},$$

where

$$\mathcal{K}_{j,l,m,n}[\psi](x) = \int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!} \frac{\partial^n K_m(z_l, z_j)}{\partial x^\beta \partial y^\alpha}(x-z_l)^\beta (y-z_j)^\alpha (x-y)$$

$$\cdot \nu(x)\psi(y)\, d\sigma(y)$$

with

$$K_m(x,y) = -\frac{i^m(m-1)|x-y|^{m-3}}{4\pi m!}.$$

In particular, we have

$$\begin{aligned}\mathcal{K}_{j,l,0,0}[\psi](x) = \frac{1}{4\pi|z_l - z_j|^3}\Big[&(x-z_l)\cdot\nu(x)\big(\psi, \chi(\partial D_j)\big)_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \\ &-\big(\psi, (y-z_j)\cdot\nu(x)\big)_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)} \\ &+(z_l - z_j)\cdot\nu(x)\big(\psi, \chi(\partial D_j)\big)_{H^{-1/2}(\partial D_j), H^{1/2}(\partial D_j)}\Big], \qquad \text{(B.1)}\\ \mathcal{K}_{j,l,1,m}[\psi] = 0 \quad &\text{for all } m. \qquad \text{(B.2)}\end{aligned}$$

Lemma B.5. *We have* $\|\mathcal{K}_{j,l,m,n}\|_{\mathcal{L}(\mathcal{H}^*(\partial D_j), \mathcal{H}^*(\partial D_l))} \lesssim O(\delta^{n+2})$.

Proof. Note that

$$\begin{aligned}\mathcal{K}_{j,l,m,n}[\psi](x) = &\int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!}\frac{\partial^n K_m(z_l,z_j)}{\partial x^\beta \partial y^\alpha}(x-z_l)^\beta (y-z_j)^\alpha (x-z_l) \\ &\cdot\nu(x)\psi(y)\, d\sigma(y), \\ &-\int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!}\frac{\partial^n K_m(z_l,z_j)}{\partial x^\beta \partial y^\alpha}(x-z_l)^\beta (y-z_j)^\alpha (y-z_j) \\ &\cdot\nu(x)\psi(y)\, d\sigma(y), \\ &+\int_{\partial D_j} \sum_{|\alpha|+|\beta|=n} \frac{1}{\alpha!\beta!}\frac{\partial^n K_m(z_l,z_j)}{\partial x^\beta \partial y^\alpha}(x-z_l)^\beta (y-z_j)^\alpha (z_l-z_j) \\ &\cdot\nu(x)\psi(y)\, d\sigma(y).\end{aligned}$$

After a translation of coordinates, we can apply Lemma B.2 to each one of the three terms above to conclude that $\mathcal{K}_{j,l,m,n} = O(\delta^{n+3}) + O(\delta^{n+2})$. This completes the proof of the lemma. □

To summarize, we have proven the following results:

Lemma B.6. *(i) Regarded as an operator from $\mathcal{H}^*(\partial D_j)$ into $\mathcal{H}(\partial D_l)$ we have,*

$$\mathcal{S}^k_{D_j,D_l} = \mathcal{S}_{j,l,0,0} + \mathcal{S}_{j,l,0,1} + \mathcal{S}_{j,l,0,2} + k\mathcal{S}_{j,l,1} + k^2\mathcal{S}_{j,l,2,0} + O(\delta^4) + O(k^2\delta^2).$$

Moreover,

$$\mathcal{S}_{j,l,m,n} = O(\delta^{n+1}).$$

(ii) Regarded as an operator from $\mathcal{H}^*(\partial D_j)$ into $\mathcal{H}^*(\partial D_l)$, we have

$$\mathcal{K}^k_{D_j,D_l} = \mathcal{K}_{j,l,0,0} + O(k^2\delta^2).$$

Moreover,

$$\mathcal{K}_{j,l,0,0} = O(\delta^2).$$

Appendix C: Adaptation of Results to the Two-Dimensional Case

In this section we adapt the layer potential formulation to plasmonic resonances in two dimensions. We only consider the single particle case. For the multiple particle case, a similar analysis holds.

Recall that in $\mathbb{R}^2$ the single-layer potential $\mathcal{S}_D : H^{-1/2}(\partial D) \to H^{1/2}(\partial D)$ is not, in general, invertible nor injective. Hence, $-(u, \mathcal{S}_D[v])_{-\frac{1}{2},\frac{1}{2}}$ does not define an inner product and the symmetrization technique described in Lemma 2.1 is no longer valid.

To overcome this difficulty, a substitute of $\mathcal{S}_D$ can be introduced as in [14] by

$$\widetilde{\mathcal{S}}_D[\psi] = \begin{cases} \mathcal{S}_D[\psi] & \text{if } (\psi, \chi(\partial D))_{-\frac{1}{2},\frac{1}{2}} = 0, \\ \chi(\partial D) & \text{if } \psi = \varphi_0, \end{cases} \tag{C.1}$$

where φ_0 is the unique (in the case of a single particle) eigenfunction of $\mathcal{K}_D^*$ associated with eigenvalue $1/2$ such that $(\varphi_0, \chi(\partial D))_{-\frac{1}{2},\frac{1}{2}} = 1$. Note that, from the jump relations of the layer potentials, $\mathcal{S}_D[\varphi_0]$ is constant.

The operator $\widetilde{\mathcal{S}}_D : H^{-1/2}(\partial D) \to H^{1/2}(\partial D)$ is invertible. Moreover, the following Calderón identity holds: $\mathcal{K}_D\widetilde{\mathcal{S}}_D = \widetilde{\mathcal{S}}_D\mathcal{K}_D^*$. With this, define

$$(u, v)_{\mathcal{H}^*} = -(u, \widetilde{\mathcal{S}}_D[v])_{-\frac{1}{2},\frac{1}{2}}.$$

Thanks to the invertibility and positivity of $-\widetilde{\mathcal{S}}_D$, this defines an inner product for which $\mathcal{K}_D^*$ is self-adjoint and $\mathcal{H}^*$ is equivalent to $H^{-1/2}$. Then, if D is $\mathcal{C}^{1,\alpha}$, we have the following results:

Lemma C.1. *Let D be a $\mathcal{C}^{1,\alpha}$ bounded simply connected domain of $\mathbb{R}^2$ and let $\widetilde{\mathcal{S}}_D$ be the operator defined in C.1. Then,*

(i) The operator $\mathcal{K}_D^*$ is compact self-adjoint in the Hilbert space $\mathcal{H}^*(\partial D)$ equipped with the inner product defined by

$$(u, v)_{\mathcal{H}^*} = -(u, \widetilde{\mathcal{S}}_D[v])_{-\frac{1}{2},\frac{1}{2}} \tag{C.2}$$

with $(\cdot,\cdot)_{-\frac{1}{2},\frac{1}{2}}$ being the duality pairing between $H^{-1/2}(\partial D)$ and $H^{1/2}(\partial D)$, which is equivalent to the original one;

(ii) Let (λ_j, φ_j), $j = 0, 1, 2, \ldots,$ be the eigenvalue and normalized eigenfunction pair of $\mathcal{K}_D^*$ with $\lambda_0 = \frac{1}{2}$. Then, $\lambda_j \in (-\frac{1}{2}, \frac{1}{2}]$ and $\lambda_j \to 0$ as $j \to \infty$;

(iii) $\mathcal{H}^*(\partial D) = \mathcal{H}_0^*(\partial D) \oplus \{\mu\varphi_0,\ \mu \in \mathbb{C}\}$, where $\mathcal{H}_0^*(\partial D)$ is the zero mean subspace of $\mathcal{H}^*(\partial D)$;

(iv) The following representation formula holds: for any $\psi \in H^{-1/2}(\partial D)$,

$$\mathcal{K}_D^*[\psi] = \sum_{j=0}^{\infty} \lambda_j (\psi, \varphi_j)_{\mathcal{H}^*} \otimes \varphi_j.$$

Lemma C.2. *Let $\mathcal{H}(\partial D)$ be the space $H^{\frac{1}{2}}(\partial D)$ equipped with the following equivalent inner product*

$$(u, v)_{\mathcal{H}} = (-\widetilde{\mathcal{S}}_D^{-1}[u], v)_{-\frac{1}{2},\frac{1}{2}}. \tag{C.3}$$

Then, $\widetilde{\mathcal{S}}_D$ is an isometry between $\mathcal{H}^*(\partial D)$ and $\mathcal{H}(\partial D)$.

Remark C.1. Note that $\widetilde{\mathcal{S}}_D^{-1}[\chi(\partial D)] = \varphi_0$ and $(-\frac{1}{2}Id + \mathcal{K}_D^*) = (-\frac{1}{2}Id + \mathcal{K}_D^*)\mathcal{P}_{\mathcal{H}_0^*}$, where $\mathcal{P}_{\mathcal{H}_0^*}$ is the orthogonal projection onto $\mathcal{H}_0^*(\partial D)$. In particular, we have $(-\frac{1}{2}Id + \mathcal{K}_D^*)\widetilde{\mathcal{S}}_D^{-1}[\chi(\partial D)] = 0$.

Let us now consider the single-layer potential for the Helmholtz equation in $\mathbb{R}^2$

$$\mathcal{S}_D^k[\psi](x) = \int_{\partial D} G(x, y, k)\psi(y)\,\mathrm{d}\sigma(y), \quad x \in \partial D,$$

where $G(x, y, k) = -\frac{i}{4}H_0^{(1)}(k|x-y|)$ and $H_0^{(1)}$ is the Hankel function of first kind and order 0. We have

$$-\frac{i}{4}H_0^{(1)}(k|x-y|) = \frac{1}{2\pi}\log|x-y| + \tau_k + \sum_{j=1}^{\infty}(b_j \log k|x-y| + c_j)(k|x-y|)^{2j},$$

where

$$\begin{aligned}
\tau_k &= \frac{1}{2\pi}(\log k + \gamma - \log 2) - \frac{i}{4}, \quad b_j = \frac{(-1)^j}{2\pi}\frac{1}{2^{2j}(j!)^2}, \\
c_j &= -bj\left(\gamma - \log 2 - \frac{i\pi}{2} - \sum_{n=1}^{j}\frac{1}{n}\right),
\end{aligned}$$

and γ is the Euler constant. Thus, we get

$$\mathcal{S}_D^k = \hat{\mathcal{S}}_D^k + \sum_{j=1}^{\infty}\left(k^{2j}\log k\right)\mathcal{S}_{D,j}^{(1)} + \sum_{j=1}^{\infty}k^{2j}\mathcal{S}_{D,j}^{(2)}, \tag{C.4}$$

where

$$\begin{aligned}
\hat{\mathcal{S}}_D^k[\psi](x) &= \mathcal{S}_D[\psi](x) + \tau_k\int_{\partial D}[\psi]\,\mathrm{d}\sigma, \\
\mathcal{S}_{D,j}^{(1)}[\psi](x) &= \int_{\partial D} b_j|x-y|^{2j}\psi(y)\,\mathrm{d}\sigma(y), \\
\mathcal{S}_{D,j}^{(2)}[\psi](x) &= \int_{\partial D}|x-y|^{2j}(b_j\log|x-y| + c_j)\psi(y)\,\mathrm{d}\sigma(y).
\end{aligned}$$

Lemma C.3. *The norms $\|\mathcal{S}_{D,j}^{(1)}\|_{\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}(\partial D))}$ and $\|\mathcal{S}_{D,j}^{(2)}\|_{\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}(\partial D))}$ are uniformly bounded with respect to j. Moreover, the series in (C.4) is convergent in $\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}(\partial D))$.*

Proof. The proof is similar to that of Lemma A.1. □

Observe that

$$\begin{aligned}
\left(\mathcal{S}_D - \widetilde{\mathcal{S}}_D\right)[\psi] &= \left(\mathcal{S}_D - \widetilde{\mathcal{S}}_D\right)[\mathcal{P}_{\mathcal{H}_0^*}[\psi] + (\psi, \varphi_0)_{\mathcal{H}^*}\varphi_0] \\
&= (\psi, \varphi_0)_{\mathcal{H}^*}\left(\mathcal{S}_D[\varphi_0] - \chi(\partial D)\right).
\end{aligned}$$

Then it follows that

$$\begin{aligned}\hat{\mathcal{S}}_D^k[\psi] &= \widetilde{\mathcal{S}}_D[\psi] + (\psi, \varphi_0)_{\mathcal{H}^*}\left(\mathcal{S}_D[\varphi_0] - \chi(\partial D)\right) + \tau_k \int_{\partial D} \psi_0 + (\psi, \varphi_0)_{\mathcal{H}^*}\varphi_0 \, \mathrm{d}\sigma \\ &= \widetilde{\mathcal{S}}_D[\psi] + \Upsilon_k[\psi],\end{aligned}$$

where

$$\Upsilon_k[\psi] = (\psi, \varphi_0)_{\mathcal{H}^*}\left(\mathcal{S}_D[\varphi_0] - \chi(\partial D) + \tau_k\right). \tag{C.5}$$

Therefore, we arrive at

Lemma C.4. *For k small enough $\hat{\mathcal{S}}_D^k : \mathcal{H}^*(\partial D) \to \mathcal{H}(\partial D)$ is invertible.*

Proof. Υ_k is clearly a compact operator. Since $\widetilde{\mathcal{S}}_D$ is invertible, the invertibility of $\hat{\mathcal{S}}_D^k$ is equivalent to that of $\hat{\mathcal{S}}_D^k \widetilde{\mathcal{S}}_D^{-1} = Id + \Upsilon_k \widetilde{\mathcal{S}}_D^{-1}$. By the Fredholm alternative we only need to prove the injectivity of $Id + \Upsilon_k \widetilde{\mathcal{S}}_D^{-1}$.

Since $\forall\ v \in H^{1/2}$, $\Upsilon_k \widetilde{\mathcal{S}}_D^{-1}[v] \in \mathbb{C}$, for $\left(Id + \Upsilon_k \widetilde{\mathcal{S}}_D^{-1}\right)[v] = 0$, we need $v = \widetilde{\mathcal{S}}_D[\alpha\varphi_0] = \alpha \in \mathbb{C}$.

We have

$$\left(Id + \Upsilon_k \widetilde{\mathcal{S}}_D^{-1}\right)\left[\widetilde{\mathcal{S}}_D[\alpha\varphi_0]\right] = \alpha(\mathcal{S}_D[\varphi_0] + \tau_k) = 0 \quad \text{iff} \quad \mathcal{S}_D[\varphi_0] = -\tau_k \text{ or } \alpha = 0.$$

Since we can always find a small enough k such that $\mathcal{S}_D[\varphi_0] \neq -\tau_k$, we need $\alpha = 0$. This yields the stated result. □

Lemma C.5. *For k small enough, the operator $\mathcal{S}_D^k : \mathcal{H}^*(\partial D) \to \mathcal{H}(\partial D)$ is invertible.*

Proof. The operator $\mathcal{S}_D^k - \hat{\mathcal{S}}_D^k$ is a compact operator. Because $\hat{\mathcal{S}}_D^k$ is invertible for k small enough, by the Fredholm alternative only the injectivity of $\mathcal{S}_D^k$ is necessary. From the uniqueness of a solution to the Helmholtz equation we get the result. □

We can write (C.4) as

$$\mathcal{S}_D^k = \hat{\mathcal{S}}_D^k + \mathcal{G}_k,$$

where $\mathcal{G}_k = k^2 \log k \mathcal{S}_{D,1}^{(1)} + k^2 \mathcal{S}_{D,1}^{(2)} + O(k^4 \log k)$. From the two lemmas above we get the identity

$$(\mathcal{S}_D^k)^{-1} = \left(Id + (\hat{\mathcal{S}}_D^k)^{-1}\mathcal{G}_k\right)^{-1}(\hat{\mathcal{S}}_D^k)^{-1}.$$

It is clear that $\|(\hat{\mathcal{S}}_D^k)^{-1}\|_{\mathcal{L}(\mathcal{H}(\partial D), \mathcal{H}^*(\partial D))}$ is bounded in k. Thus, for k small enough, we can formally write

$$(\mathcal{S}_D^k)^{-1} = (\hat{\mathcal{S}}_D^k)^{-1} - (\hat{\mathcal{S}}_D^k)^{-1}\mathcal{G}_k(\hat{\mathcal{S}}_D^k)^{-1} + O(k^4 \log^2 k).$$

We have the identity

$$(\hat{\mathcal{S}}_D^k)^{-1} = \underbrace{\left(\widetilde{\mathcal{S}}_D^{-1}\hat{\mathcal{S}}_D^k\right)^{-1}}_{\Lambda_k^{-1}} \widetilde{\mathcal{S}}_D^{-1}.$$

Here,

$$\Lambda_k = Id - (\cdot, \varphi_0)_{\mathcal{H}^*}(\mathcal{S}_D[\varphi_0] + \chi(\partial D) + \tau_k)\varphi_0.$$

Then,

$$\Lambda_k^{-1} = Id - (\cdot, \varphi_0)_{\mathcal{H}^*}\frac{\mathcal{S}_D[\varphi_0] + \chi(\partial D) + \tau_k}{\mathcal{S}_D[\varphi_0] + \tau_k}\varphi_0,$$

and therefore,

$$(\hat{\mathcal{S}}_D^k)^{-1} = \widetilde{\mathcal{S}}_D^{-1} - (\widetilde{\mathcal{S}}_D^{-1}[\cdot], \varphi_0)_{\mathcal{H}^*}\varphi_0 + \frac{(\widetilde{\mathcal{S}}_D^{-1}[\cdot], \varphi_0)_{\mathcal{H}^*}}{\mathcal{S}_D[\varphi_0] + \tau_k}\varphi_0.$$

Finally, we get

$$\begin{aligned}(\mathcal{S}_D^k)^{-1} &= \mathcal{L}_D + \mathcal{U}_k - k^2 \log k \mathcal{L}_D \mathcal{S}_{D,1}^{(1)} \mathcal{L}_D \\ &\quad -k^2 \left(\mathcal{L}_D \mathcal{S}_{D,1}^{(2)} \mathcal{L}_D - \log k (\mathcal{U}_k \mathcal{S}_{D,1}^{(1)} \mathcal{L}_D + \mathcal{L}_D \mathcal{S}_{D,1}^{(1)} \mathcal{U}_k)\right) \\ &\quad + O(k^2 \log^{-1} k)\end{aligned}$$

with $\mathcal{L}_D = \mathcal{P}_{\mathcal{H}_0^*}\widetilde{\mathcal{S}}_D^{-1}$ and $\mathcal{U}_k = -\dfrac{(\widetilde{\mathcal{S}}_D^{-1}[\cdot], \varphi_0)_{\mathcal{H}^*}}{\mathcal{S}_D[\varphi_0] + \tau_k}\varphi_0$. We note that $\mathcal{U}_k = O(\log^{-1} k)$.

We now consider the expansion for the boundary integral operator $(\mathcal{K}_D^k)^*$. We have

$$(\mathcal{K}_D^k)^* = \mathcal{K}_D^* + \sum_{j=1}^{\infty}\left(k^{2j} \log k\right)\mathcal{K}_{D,j}^{(1)} + \sum_{j=1}^{\infty} k^{2j}\mathcal{K}_{D,j}^{(2)}, \tag{C.6}$$

where

$$\begin{aligned}\mathcal{K}_{D,j}^{(1)}[\psi](x) &= \int_{\partial D} b_j \frac{\partial |x-y|^{2j}}{\partial \nu(x)}\psi(y)\,\mathrm{d}\sigma(y), \\ \mathcal{K}_{D,j}^{(2)}[\psi](x) &= \int_{\partial D} \frac{\partial\left(|x-y|^{2j}(b_j \log|x-y| + c_j)\right)}{\nu(x)}\psi(y)\,\mathrm{d}\sigma(y).\end{aligned}$$

Lemma C.6. *The norms $\|\mathcal{K}_{D,j}^{(1)}\|_{\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}^*(\partial D))}$ and $\|\mathcal{K}_{D,j}^{(2)}\|_{\mathcal{L}(\mathcal{H}^*(\partial D),\mathcal{H}^*(\partial D))}$ are uniformly bounded for $j \geqq 1$. Moreover, the series in (C.6) is convergent in $\mathcal{L}(\mathcal{H}^*(\partial D), \mathcal{H}^*(\partial D))$.*

Proof. The proof is similar to that of Lemma A.1. □

Recalling (2.5) and (2.6), we can show that the following result holds:

Lemma C.7. *Regarding $\mathcal{A}_D(\omega)$ as an operator from $\mathcal{H}^*(\partial D)$ to $\mathcal{H}^*(\partial D)$, we have*

$$\mathcal{A}_D(\omega) = \mathcal{A}_{D,0} + \omega^2(\log \omega)\mathcal{A}_{D,1} + O(\omega^2),$$

where

$$\mathcal{A}_{D,0} = \left(\frac{1}{2\mu_m} + \frac{1}{2\mu_c}\right) Id + \left(\frac{1}{\mu_m} - \frac{1}{\mu_c}\right)\mathcal{K}_D^*,$$

$$\mathcal{A}_{D,1} = \mathcal{K}_{D,1}^{(1)}\left(\varepsilon_m Id - \varepsilon_c \mathcal{P}_{\mathcal{H}_0^*}\right) + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}_D^*\right)\widetilde{\mathcal{S}}_D^{-1}\mathcal{S}_{D,1}^{(1)}$$
$$\left(\mu_m\varepsilon_m Id - \mu_c\varepsilon_c\mathcal{P}_{\mathcal{H}_0^*}\right).$$

Proof. We have

$$(\mathcal{S}_D^{k_c})^{-1} = \mathcal{L}_D + \mathcal{U}_{k_c} - \omega^2(\log\omega)\varepsilon_c\mu_c\mathcal{L}_D\mathcal{S}_{D,1}^{(1)}\mathcal{L}_D + O(\omega^2)$$
$$\mathcal{S}_D^{k_m} = \widetilde{\mathcal{S}}_D + \Upsilon_{k_m} + \omega^2(\log\omega)\varepsilon_m\mu_m\mathcal{S}_{D,1}^{(1)} + O(\omega^2).$$

Also, $\mathcal{L}_D\Upsilon_{k_m} = \mathcal{P}_{\mathcal{H}_0^*}(\widetilde{\mathcal{S}}_D)^{-1}\Upsilon_{k_m} = 0$, where Υ_{k_m} is defined by (C.5). Hence,

$$\begin{aligned}
&(\mathcal{S}_D^{k_c})^{-1}\mathcal{S}_D^{k_m}\\
&\quad= \mathcal{P}_{\mathcal{H}_0^*} + \mathcal{U}_{k_c}\widetilde{\mathcal{S}}_D + \mathcal{U}_{k_c}\Upsilon_{k_m} + \omega^2(\log\omega)\big(\varepsilon_m\mu_m\mathcal{L}_D\mathcal{S}_{D,1}^{(1)} - \varepsilon_c\mu_c\mathcal{L}_D\mathcal{S}_{D,1}^{(1)}\mathcal{L}_D\widetilde{\mathcal{S}}_D\big)\\
&\qquad+O(\omega^2)\\
&\quad= \mathcal{P}_{\mathcal{H}_0^*} + \mathcal{U}_{k_c}\widetilde{\mathcal{S}}_D + \mathcal{U}_{k_c}\Upsilon_{k_m} + \omega^2\log\omega\mathcal{L}_D\mathcal{S}_{D,1}^{(1)}\big(\varepsilon_m\mu_m Id - \varepsilon_c\mu_c\mathcal{P}_{\mathcal{H}_0^*}\big)\\
&\qquad+O(\omega^2).
\end{aligned}$$

From Remark C.1, it follows that

$$\left(\frac{1}{2}Id - \mathcal{K}_D^*\right)\mathcal{U}_{k_c} = 0.$$

Since $\frac{1}{2}Id - (\mathcal{K}_D^k)^* = \left(\frac{1}{2}Id - \mathcal{K}_D^*\right) - k^2\log k\mathcal{K}_{D,1}^{(1)} + O(k^2)$, we get the desired result. $\square$

Under Conditions 2 and 3, the perturbed eigenvalues and eigenfunctions of $\mathcal{A}_D(\omega)$ have the form

$$\tau_j(\omega) = \tau_j + \omega^2(\log\omega)\tau_{j,1} + O(\omega^2), \tag{C.7}$$
$$\varphi_j(\omega) = \varphi_j + \omega^2(\log\omega)\varphi_{j,1} + O(\omega^2), \tag{C.8}$$

where

$$\tau_{j,1} = R_{jj}, \tag{C.9}$$
$$\varphi_{j,1} = \sum_{l\neq j}\frac{R_{jl}}{\left(\frac{1}{\mu_m} - \frac{1}{\mu_c}\right)(\lambda_j - \lambda_l)}\varphi_l, \tag{C.10}$$

and

$$R_{jl} = (\mathcal{A}_{D,1}[\varphi_j], \varphi_l)_{\mathcal{H}^*}.$$

It is clear that Lemma 2.5 holds in the two-dimensional case. We also have the following asymptotic expansion for f in terms of ω:

Lemma C.8. *In the space $\mathcal{H}^*(\partial D)$, as ω goes to zero, we have*

$$f = \omega f_1 + O(\omega^2),$$

where

$$f_1 = -i e^{ik_m d\cdot z}\sqrt{\varepsilon_m \mu_m}\left(\frac{1}{\mu_m}[d\cdot\nu(x)] + \frac{1}{\mu_c}\left(\frac{1}{2}Id - \mathcal{K}_D^*\right)\widetilde{\mathcal{S}}_D^{-1}[d\cdot(x-z)]\right)$$

and z is the center of the domain D.

Finally, the following result holds:

Theorem C.1. *Under Conditions 1, 2, and 3, the scattered field by a single plasmonic particle, $u^s = u - u^i$, has in the quasi-static limit the following representation:*

$$u^s = \mathcal{S}_D^{k_m}[\psi],$$

where

$$\psi = \sum_{j\in J}\frac{ik_m e^{ik_m d\cdot z}\big(d\cdot\nu(x),\varphi_j\big)_{\mathcal{H}^*}\varphi_j + O(\omega^3\log\omega)}{\lambda - \lambda_j + O(\omega^2\log\omega)} + O(\omega)$$

with λ being defined by (2.19).

Proof. We have

$$\begin{aligned}\psi &= \sum_{j\in J}\frac{\big(f,\widetilde{\varphi}_j(\omega)\big)_{\mathcal{H}^*}\varphi_j(\omega)}{\tau_j(\omega)} + \mathcal{A}_D(\omega)^{-1}(P_{J^c}(\omega)f)\\ &= \sum_{j\in J}\frac{\omega\big(f_1,\varphi_j\big)_{\mathcal{H}^*}\varphi_j + O(\omega^3(\log\omega))}{\frac{1}{2\mu_m}+\frac{1}{2\mu_c}-\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)\lambda_j + O(\omega^2\log\omega)} + O(\omega).\end{aligned}$$

Since $d\cdot(x-z)$ is a harmonic function, changing $\mathcal{S}_D$ by $\widetilde{\mathcal{S}}_D$ in Theorem 2.1 yields

$$\Big((\frac{1}{2}Id - \mathcal{K}_D^*)\mathcal{S}_D^{-1}[d\cdot(x-z)],\varphi_j\Big)_{\mathcal{H}^*} = -(d\cdot\nu(x),\varphi_j)_{\mathcal{H}^*}.$$

Then the proof is complete. □

Corollary C.1. *Assume the same conditions as in Theorem 2.1. Then, under the additional condition*

$$\min_{j\in J}|\tau_j(\omega)| \gg \omega^2,$$

we have

$$\psi = \sum_{j\in J}\frac{ik_m e^{ik_m d\cdot z}\big(d\cdot\nu(x),\varphi_j\big)_{\mathcal{H}^*}\varphi_j + O(\omega^3\log\omega)}{\lambda - \lambda_j + \omega^2\log\omega\big(\frac{1}{\mu_c}-\frac{1}{\mu_m}\big)^{-1}\tau_{j,1}} + O(\omega).$$

Appendix D: Sum Rules for the Polarization Tensor

Let f be a holomorphic function defined in an open set $U \subset \mathbb{C}$ containing the spectrum of $\mathcal{K}^*_{\partial D}$. Then, we can write $f(z) = \sum_{j=0}^{\infty} a_j z^j$ for every $z \in U$.

Definition 4. Let

$$f(\mathcal{K}^*_D) := \sum_{j=0}^{\infty} a_j (\mathcal{K}^*_D)^j,$$

where $(\mathcal{K}^*_D)^j := \underbrace{\mathcal{K}^*_D \circ \mathcal{K}^*_D \circ .. \circ \mathcal{K}^*_D}_{j\ times}$.

Lemma D.1. *We have*

$$f(\mathcal{K}^*_D) = \sum_{j=1}^{\infty} f(\lambda_j)(\cdot, \varphi_j)_{\mathcal{H}^*}\varphi_j.$$

Proof. We have

$$\begin{aligned} f(\mathcal{K}^*_D) = \sum_{i=0}^{\infty} a_i (\mathcal{K}^*_D)^i &= \sum_{i=0}^{\infty} a_i \sum_{j=1}^{\infty} \lambda_j^i (\cdot, \varphi_j)_{\mathcal{H}^*}\varphi_j \\ &= \sum_{j=1}^{\infty} \left(\sum_{i=0}^{\infty} a_i \lambda_j^i \right) (\cdot, \varphi_j)_{\mathcal{H}^*}\varphi_j \\ &= \sum_{j=1}^{\infty} f(\lambda_j)(\cdot, \varphi_j)_{\mathcal{H}^*}\varphi_j. \end{aligned}$$

□

From Lemma D.1, we can deduce that

$$\int_{\partial D} x_l f(\mathcal{K}^*_D)[\nu_m](x)\ \mathrm{d}\sigma(x) = \sum_{j=1}^{\infty} f(\lambda_j)\alpha_{l,m}^{(j)}. \tag{D.1}$$

Equation (D.1) yields the summation rules for the entries of the polarization tensor.

In order to prove that $\sum_{j=1}^{\infty} \alpha_{l,m}^{(j)} = \delta_{l,m}|D|$, we take $f(\lambda) = 1$ in (D.1) to get

$$\sum_{j=1}^{\infty} \alpha_{l,m}^{(j)} = \int_{\partial D} x_l \nu_m(x)\ \mathrm{d}\sigma(x) = \delta_{l,m}|D|.$$

Next, we prove that

$$\sum_{j=1}^{\infty} \lambda_j \sum_{l=1}^{d} \alpha_{l,l}^{(j)} = \frac{(d-2)}{2}|D|.$$

Taking $f(\lambda) = \lambda$ in (D.1), we obtain

$$\sum_{j=1}^{\infty} \lambda_j \sum_{l=1}^{d} \alpha_{l,l}^{(j)} = \sum_{l=1}^{d} \int_{\partial D} x_l \mathcal{K}_D^*[\nu_l](x)\ \mathrm{d}\sigma(x),$$

$$\begin{aligned}\int_{\partial D} x_l \mathcal{K}_D^*[\nu_l](x)\ \mathrm{d}\sigma(x) &= \int_{\partial D} x_l \left(\frac{1}{2}\nu_l(x) + \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_{-}(x)\right) \mathrm{d}\sigma(x), \\ &= \frac{|D|}{2} + \int_{\partial D} x_l \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_{-}(x)\, \mathrm{d}\sigma(x). \end{aligned} \tag{D.2}$$

Integrating by parts we arrive at

$$\int_{\partial D} x_l \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_{-}(x)\, \mathrm{d}\sigma(x) = \int_D e_l(x) \cdot \nabla \mathcal{S}_D[\nu_l](x)\, \mathrm{d}x + \int_D x_l \Delta \mathcal{S}_D[\nu_l](x)\, \mathrm{d}x.$$

Since the single-layer potential is harmonic on D,

$$\int_{\partial D} x_l \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_{-}(x)\, \mathrm{d}\sigma(x) = \int_D e_l(x) \cdot \left(\int_{\partial D} \nabla_x \Gamma(x, x')\nu_l(x')\, \mathrm{d}\sigma(x')\right) \mathrm{d}x.$$

Summing on i and using $\nabla_x \Gamma(x, x') = -\nabla_{x'} \Gamma(x, x')$, we get

$$\begin{aligned}\sum_{l=1}^{d} \int_{\partial D} x_l \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_{-}(x)\, \mathrm{d}\sigma(x) &= -\int_D \left(\int_{\partial D} \nu(x') \cdot \nabla_{x'} \Gamma(x, x')\, \mathrm{d}\sigma(x')\right) \mathrm{d}x, \\ &= -\int_D \mathcal{D}_D[1](x)\, \mathrm{d}x, \\ &= -|D|, \end{aligned} \tag{D.3}$$

where $\mathcal{D}_D$ is the double-layer potential. Hence, summing equation (D.2) for $i = 1, \ldots, d$, we get the result.

Finally, we show that

$$\sum_{j=1}^{\infty} \lambda_j^2 \sum_{l=1}^{d} \alpha_{l,l}^{(j)} = \frac{d-4}{4}|D| + \sum_{l=1}^{d} \int_D |\nabla \mathcal{S}_D[\nu_l]|^2\, \mathrm{d}x.$$

Taking $f(\lambda) = \lambda^2$ in (D.1) yields

$$\begin{aligned}\sum_{j=1}^{\infty} \lambda_j^2 \sum_{l=1}^{d} \alpha_{l,l}^{(j)} &= \sum_{l=1}^{d} \int_{\partial D} x_l (\mathcal{K}_D^*)^2[\nu_l](x)\ \mathrm{d}\sigma(x) \\ &= \sum_{l=1}^{d} \int_{\partial D} \mathcal{K}_D[y_l](x) \mathcal{K}_D^*[\nu_l](x)\ \mathrm{d}\sigma(x) \\ &= \sum_{l=1}^{d} \int_{\partial D} \mathcal{K}_D[y_l] \frac{\nu_l}{2}\, \mathrm{d}\sigma + \sum_{l=1}^{d} \int_{\partial D} \mathcal{K}_D[y_l] \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}|_{-}\, \mathrm{d}\sigma \end{aligned}$$

$$= \frac{(d-2)}{4}|D| - \underbrace{\sum_{l=1}^{d} \int_{\partial D} \frac{y_l}{2} \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_- \mathrm{d}\sigma}_{I_1}$$

$$+ \underbrace{\sum_{l=1}^{d} \int_{\partial D} \mathcal{D}_D[y_l]\Big|_- \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_- \mathrm{d}\sigma}_{I_2}.$$

From (D.3) it follows that

$$I_1 = -\frac{|D|}{2}.$$

Since x_l is harmonic, we have $x_l = \mathcal{D}_D[y_l](x)|_- - \mathcal{S}_D[\nu_l](x)$ on ∂D, and thus,

$$\begin{aligned} I_2 &= \sum_{l=1}^{d} \int_{\partial D} (x_l + \mathcal{S}_D[\nu_l](x)) \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_- (x)\, \mathrm{d}\sigma(x), \\ &= -|D| + \sum_{l=1}^{d} \int_{\partial D} \mathcal{S}_D[\nu_l] \frac{\partial \mathcal{S}_D[\nu_l]}{\partial \nu}\Big|_- \mathrm{d}\sigma, \\ &= -|D| + \sum_{l=1}^{d} \int_{D} |\nabla \mathcal{S}_D[\nu_l]|^2\, \mathrm{d}x. \end{aligned}$$

Replacing I_1 and I_2 by their expressions gives the desired result.

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