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# MATHEMATICAL MODELING OF THE PHOTOACOUSTIC EFFECT GENERATED BY THE HEATING OF METALLIC NANOPARTICLES

By

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**Abstract.** This paper is devoted to the modeling of the photoacoustic effect generated by the electromagnetic heating of metallic nanoparticles embedded in a biological tissue. We first derive an asymptotic model for the plasmonic resonances and the electromagnetic fields. We then describe the acoustic generation created by the electromagnetic heating of the nanoparticle. Precisely, we derive the model equations that describe the coupling between the temperature rise in the medium and the acoustic wave generation. We obtain a direct relation between the acoustic waves and the electromagnetic external sources. Finally, we solve the multiwave inverse problem that consists in the recovery of the electric permittivity of the biological tissue from the measurements of the generated acoustic waves on the boundary of the sample.

## Contents



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**1. The photoacoustic model and main results.** Photoacoustic imaging [4, 5, 18, 22, 25, 26, 35, 37, 55, 58] is a recent hybrid imaging modality that couples electromagnetic waves with acoustic waves to achieve high-resolution imaging of optical properties of heterogeneous media such as biological tissues. Our objective in this paper is to derive a realistic complete mathematical model for the photoacoustic generation by a single nanoparticle embedded in a biological tissue. We introduce the mathematical framework and give the main result in the first section. In the second section we describe the mechanism of enhancement of light through the optical scattering properties of metallic nanoparticles. The third section is devoted to the thermal modeling of the part of the electromagnetic energy converted into heat. We precisely derive a theoretical model for the generation of acoustic waves by the thermal expansion of the tissue around the metallic nanoparticles. The inverse photoacoustic problem is solved asymptotically in section 4. We finally give useful technical results in the appendix.

We now give a mathematical framework for the whole photoacoustic effect. Let $\Omega$ be a bounded $C^2$ domain in $\mathbb{R}^2$. The outward unit normal at $x$ to $\partial\Omega$ is denoted by $\nu_\Omega(x)$. The domain $\Omega$ is referred to as the biological sample that we aim to image by the non-invasive photoacoustic modality. Assume that $\Omega$ contains a single nanoparticule, of the form $B_\alpha := z^\star + \alpha B$, where $B$ is a bounded, $C^2$ smooth domain containing the origin, $\alpha > 0$ is a small constant that represents the size of the nanoparticle, and $z^\star$ is the position of the nanoparticle. The first step in the photoacoustic imaging system is to illuminate the sample by an electromagnetic wave produced by a laser source. The time dependent, linear Maxwell's equations take the form

$$\begin{aligned} \nabla \times \mathbf{E} &= -\mu_0 \frac{\partial}{\partial t}\mathbf{H}, \\ \nabla \times \mathbf{H} &= \varepsilon \frac{\partial}{\partial t}\mathbf{E}, \end{aligned}$$

where $\mathbf{E}$ and $\mathbf{H}$ are the total electric field and the total magnetic field, respectively. The coefficients $\varepsilon$ and $\mu$ are the electric permittivity and magnetic permeability of the sample. The magnetic permeability is assumed to be a constant equal to $\mu_0$, the permeability of the free space, while the electric permittivity is given by

$$\varepsilon(x) = \begin{cases} \varepsilon_s(x) & \text{for } x \in \mathbb{R}^2 \setminus \overline{B_\alpha}, \\ \varepsilon_m & \text{for } x \in B_\alpha, \end{cases}$$

where $\varepsilon_m$ is the permittivity of the metal that will be specified later, and $\varepsilon_s(x)$ is the permittivity of the sample that is assumed of class $C^2$ and is a constant equal to $\varepsilon_0 > 0$, the permittivity of the free space, outside $\Omega$. We assume throughout that $0 < c_0 < \Re(\varepsilon_s(z^\star)) < |\Re(\varepsilon_m)|$ for all $x \in \Omega$, $\Im(\varepsilon_s(x))$ belongs to $C_0^2(\Omega)$ and satisfies $\Im(\varepsilon_s(z^\star)) > c_0$. The imaginary part of the electric permittivity $\Im(\varepsilon_s(x))$, is related to the absorption of the electromagnetic energy, and provides a good description of the state of the biological tissue. Our objective in this paper is to recover this parameter around the nanoparticles.

We assume that during the illumination of the sample a part of the electromagnetic energy is dissipated by absorption inside the biological tissue and inside the nanoparticle. The absorption of the electromagnetic energy by the biological tissue is transformed into heat and leads through the thermo-elastic expansion of the tissue to the generation of an acoustic pressure $p(x,t)$ that propagates to the detectors on the boundary $\partial\Omega$. The measurements of $p(x,t)$ on the boundary allow the reconstruction of the absorption and diffusion coefficients in the conventional pulsed photoacoustic imaging system. In practice, it has been observed in various experiments that the imaging depth, i.e. the maximal depth of the sample at which features can be resolved at expected resolution, is still fairly limited, usually on the order of millimeters. This is mainly due to the limitation on the penetration ability of the electromagnetic waves in the tissue: optical signals are attenuated significantly by absorption and scattering. In [24], the authors showed that the resolution is proportional to the magnitude of the laser fluence in the sample, and recently in [54] the mechanism of depth resolution was mathematically investigated. Metallic nanoparticles are very attractive as photoacoustic contrast agents because of their large capacity to absorb light and convert it to heat and their spectral selectivity. When they are illuminated at their plasmonic resonances their absorption of light is amplified and their temperature increases significantly leading to various phenomena including heating the surrounding media. For example, in Hyperthermia therapy for cancer treatments one seeks to destroy tumors through heating metallic nanoparticles [52]. In the context of photoacoustic imaging the heat of the surrounding biological tissue will generate a strong acoustic pressure wave $p(x,t)$ that can also be detected on the boundary $\partial\Omega$. The principal idea for the use of metallic nanoparticles in photoacoustic imaging is that one can insert them at any position inside the sample and obtain strong acoustic sources inside the sample. This will overcome the problem of the limitation in the penetration resolution depth of the conventional photoacoustic imaging modality based on the illumination of only the biological tissue. There are already several related results in the physicists community [23, 53].

Our objective in this paper is to study the inverse problem to recover $\Im(\varepsilon_s(x))$ at $z^\star$ from measurements of the pressure $p(x,t)$ on the boundary $\partial\Omega$.

Assuming that $|\nabla \mathbf{H}(z^\star)| \neq 0$, $B$ is ball, and that $z^\star$ is known we derive the following global stability estimate. It shows how the errors in measurements can effect the reconstruction of the electric permittivity at $z^\star$.

THEOREM 1.1. Let $\tau_p > \tau_\Omega$ where $\tau_p = \sup_{x,y\in\Omega} |x-y|$. Let $p_a(x,t)$ (resp., $p_b(x,t)$) be the acoustic pressure generated by an external electromagnetic source in a medium with electric permittivity $\varepsilon_{s,a}(x)$ (resp., $\varepsilon_{s,b}(x)$).

Then, there exists a constant $C > 0$ that does not depend on $\alpha$ and the boundary measurements such that

$$|\Im(\varepsilon_{s,a}(z^\star)) - \Im(\varepsilon_{s,b}(z^\star))| \\ \leq C \left( \left\| \frac{\partial p_a}{\partial t} - \frac{\partial p_b}{\partial t} \right\|_{L^2(\partial\Omega\times(0,\tau_p))} + \|\nabla p_a - \nabla p_b\|_{L^2(\partial\Omega\times(0,\tau_p))} \right)^{\frac{1}{4}} + O(\alpha).$$

The proof of the theorem is given in section 4. It is based on asymptotic expansion of the electromagnetic fields when $\alpha$ tends to zero. The coupling between the acoustic and electromagnetic waves allows us to retrieve the inner asymptotic expansion of the electromagnetic fields in a small neighborhood of $z^\star$ (Theorem 4.1, and subsection 4.2.2). Since $\alpha$, the size of the nanoparticle is small, the stability estimate of Hölder type shows that the reconstruction of $\Im(\varepsilon_s(z^\star))$ from measurements of the pressure $p(x,t)$ on the boundary $\partial\Omega$ is in fact a well-posed inverse problem. In subsection 2.1 we derived the asymptotic expansion of the plasmonic resonances of the system nanoparticle and biological tissue. Later on in subsection 4.2.2, we showed that choosing the incident wave frequency close to the real part of a plasmonic resonance enhances the photoacoustic signal measured on the boundary. Finally, the stability result can be easily extended to cover the case where many well separated nanoparticles are embedded in the sample.

**2. Electromagnetic excitation.** The first syntheses of metallic small particles date back to the fourth or fifth century B.C., where gold specimens were reported in China and Egypt. Their optical properties were used for coloration of glass, ceramics, china, and pottery (see [44] and the references therein).

It is now well known that the interesting diffractive properties of these particles are linked to resonances phenomena. In fact, plasmon resonances may occur in metallic particles if the dielectric permittivity inside the particle is negative and the wavelength of the incident excitation is much larger than the dimension of the particle. For nanoscale metallic particles, these resonances occur in the optical frequency range and they result in an extremely large enhancement of the electromagnetic field near the boundary of the particles. This phenomena has applications in many areas such as nanophotonics, nanolithography, near field microscopy, and biosensors. The desired resonance frequencies as well as the local fields enhancement can be achieved by controlling the geometry of the metallic nanostructure.

From a mathematical point of view these resonance values are the complex eigenvalues of Maxwell's equations that only occur when the dielectric permittivity of the nanoparticles is negative and the size of the nanoparticles is less than the incident wavelength. A formal asymptotic expansion in [42, 43] showed that if the ratio between the incident wavelength and the size of the nanoparticle tends to zero the plasmonic resonances approch the eigenvalues of the Neumann-Poincaré operator or the variational Poincaré operator [42, 43]. In [21], the authors have derived a rigorous justification of the quasi-static approximation in harmonic frequency regime [42, 43]. It is well known that the resonance phenomena occur only in transverse magnetic polarization ((TM)

polarization). Here we consider the time harmonic regime in (TM) polarization, that is, $\mathbf{E} = \Re(\mathbb{E}e^{i\omega t})$ and $\mathbf{H} = \Im(\mathbb{H}e^{i\omega t})$, where $\mathbb{E} = (E(x_1, x_2), 0)$ and $\mathbb{H} = (0, 0, H(x_1, x_2))$.

The total magnetic field can be decomposed into two parts $H = H_i + H_s$ where $H_i$ and $H_s$ are, respectively, the incident and scattered waves.

The homogeneous frequency-domain, linear Maxwell's equations, in the transverse magnetic polarization (TM) and in absence of internal sources, take the form

$$\nabla \cdot \left(\frac{1}{\varepsilon}\nabla H\right) + \omega^2 \mu_0 H \quad = 0 \qquad \text{in} \quad \mathbb{R}^2 \tag{2.1}$$

with the Sommerfeld radiation condition as $|x| \to +\infty$ [46]:

$$\frac{\partial H_s}{\partial |x|} - i\omega\sqrt{\varepsilon_0 \mu_0} H_s \quad = \quad O(\frac{1}{\sqrt{|x|}}). \tag{2.2}$$

Recall that the electric permittivity is given by

$$\varepsilon(x) \; = \; \begin{cases} \varepsilon_0 & \text{for } x \in \mathbb{R}^2 \setminus \overline{\Omega}, \\ \varepsilon_s(x) & \text{for } x \in \Omega \setminus \overline{B_\alpha}, \\ \varepsilon_m(\omega) & \text{for } x \in B_\alpha, \end{cases}$$

where $\varepsilon_0$ is the permittivity of the free space. The incident field $H_i$ satisfies

$$\Delta H_i + \omega^2 \mu_0 \varepsilon_0 H_i \quad = 0 \qquad \text{in} \quad \mathbb{R}^2.$$

The electric field $E$ can be deduced directly from the magnetic field through the relation

$$E(x) \quad = \quad \begin{pmatrix} \partial_{x_2} H(x) \\ -\partial_{x_1} H(x) \end{pmatrix}. \tag{2.3}$$

The metal that fills the nanoparticle is assumed to be real and its dielectric constant is described by the Drude model:

$$\varepsilon_m(\omega) \quad = \quad \varepsilon_0 \left(\varepsilon_\infty - \frac{\omega_P^2}{\omega^2 + i\omega\Gamma}\right), \tag{2.4}$$

where $\varepsilon_\infty > 0$, $\omega_P > 0$, and $\Gamma > 0$ are the metal parameters that are usually fitted utilizing experiment data [44]. The dielectric constant $\varepsilon_m$ depends on the frequency $\omega$, and so incident waves can cause a change in the metal behavior. Media having such a property are termed dispersive media.

The Drude model considered here describes well the optical properties of many metals within relatively wide frequency range. For example, the function $\varepsilon_m(\omega)$ with effective parameters: $\varepsilon_\infty = 9.84\ eV$, $\omega_P = 9.096\ eV$, $\Gamma = 0.072\ eV$ for gold, and $\varepsilon_\infty = 3.7\ eV$, $\omega_P = 8.9\ eV$, $\Gamma = 0.021\ eV$ for silver reproduce quite well the experimental values of the dielectric constant in the frequency range $0.8\ eV$ to $4\ eV$ (see, for instance, [32]).

2.1. *Plasmonic resonances.* When the frequency lies in the upper half complex space, that is, $\Im(\omega) \geq 0$, the system (2.1) has a unique solution. The resolvent of the differential operator (2.1) with condition (2.2) has a meromorphic continuation in the lower complex plane.

The complex number $\omega$ is said to be a plasmonic resonant frequency of the nanoparticle $B_\alpha$ if there exists a non-trivial solution $H$ to the system (2.1)-(2.2) with zero incident wave.

It is known that the set of scattering resonances $\{\omega_j\}$ of the above Helmholtz equation in the absence of dispersion ($\varepsilon_m$ does not depend on $\omega$) is discrete and symmetric in the complex plane about the imaginary axis. Further, it can be easily seen that all the resonant frequencies $\{\omega_j\}$ are in the lower half-space $\Im\omega < 0$. They can be found explicitly for a circular or ellipsoid shape and are connected in this case with the zeros of certain Bessel functions. More elaborate results assert that for strictly convex shapes in dimension three the resonant frequencies accumulate rapidly on the real axis as $|\Re\omega| \to \infty$ [51].

It has been shown in dimension one that the scattering resonances of a non-dispersive medium satisfy [29, 48]

$$\Im(\omega) \geq C_1 e^{-C_2|\Re(\omega)|^2},$$

where the constants $C_i,\ i = 1, 2$ only depend on $\varepsilon$ and the size of the domain.

The imaginary part of a resonance gives the decay rate of the associated resonant states. Thus, resonances close to the real axis give information about long term behavior of waves. In particular, since the work of Lax-Phillips [38] and Vainberg [57], resonance free regions near the real axis have been used to understand decay of waves. Several works in nano-optics have related the amplification and enhancement of light to the behavior of the imaginary part of the scattering resonances close to the real axis [14, 19, 20].

Like the non-dispersive case, the plasmonic resonances form a set of discrete and isolated complex values $(\omega_j(\alpha))_j$. In [21] the authors have derived the asymptotic expansion of the plasmonic resonant frequencies as $\alpha$ tends to zero and when the nanoparticle is surrounded by a homogeneous medium with a constant electric permittivity. In the following paragraph we adapt their techniques to our problem and derive the first term in the asymptotic expansion of the plasmonic resonances. We refer the reader to [3, 8, 11, 16] for recent and interesting mathematical results on plasmonic resonances for nanoparticles.

Making the change of variables $x = z^\star + \alpha\xi$ in the spectral problem (2.1), we get

$$\nabla \cdot \left(\frac{1}{\tilde{\varepsilon}_\alpha}\nabla\widetilde{H}\right) + \alpha^2\omega^2\mu_0\widetilde{H} \quad = 0 \qquad \text{in} \quad \mathbb{R}^2, \tag{2.5}$$

with the radiation condition

$$\frac{\partial\widetilde{H}}{\partial|\xi|} - i\alpha\omega\sqrt{\varepsilon_0\mu_0}\widetilde{H} \quad = \quad O(\frac{1}{\sqrt{|\xi|}}) \quad \text{as} \quad |\xi| \to +\infty, \tag{2.6}$$

where $\widetilde{H}(\xi) = H(z^\star + \alpha\xi)$, and $\tilde{\varepsilon}_\alpha(\xi) = \varepsilon(z^\star + \alpha\xi)$ is given by

$$\tilde{\varepsilon}_\alpha(\xi) \ = \ \begin{cases} \varepsilon_0 & \text{for} \quad \xi \in \mathbb{R}^2 \setminus \overline{\Omega_\alpha}, \\ \varepsilon_s(z^\star + \alpha\xi) & \text{for} \quad \xi \in \Omega_\alpha \setminus \overline{B}, \\ \varepsilon_m(\omega) & \text{for} \quad \xi \in B. \end{cases}$$

Here $\Omega_\alpha$ denotes $\left\{\frac{x-z^\star}{\alpha}; x \in \Omega\right\}$. It contains zero and tends to the whole space when $\alpha$ approaches zero. Similarly, the piecewise smooth function $\tilde{\varepsilon}_\alpha(\xi)$ converges in $L^\infty_{loc}(\mathbb{R}^2)$

to the piecewise constant function

$$\tilde{\varepsilon}(\xi) \;=\; \begin{cases} \varepsilon_s(z^\star) & \text{for} \quad \xi \in \mathbb{R}^2 \setminus \overline{B}, \\ \varepsilon_m(\omega) & \text{for} \qquad \xi \in B. \end{cases}$$

In the quasi-static regime $\alpha\omega \ll \omega \ll 1$, the above spectral problem formally converges to the quasi-static spectral problem

$$\nabla \cdot \left( \frac{1}{\tilde{\varepsilon}} \nabla \widetilde{H}_0 \right) \;= 0 \qquad \text{in} \quad \mathbb{R}^2, \tag{2.7}$$

where the field $\widetilde{H}_0(x)$ belongs to $W_0^{1,-1}(\mathbb{R}^2)$, where

$$W_0^{1,-1}(\mathbb{R}^2) := \left\{ u \in H^1_{loc}(\mathbb{R}^2) : \;\; u/(1+|\xi|^2)^{\frac{1}{2}} \;\; \ln(1+|\xi|^2) \in L^2(\mathbb{R}^2); \nabla u \in L^2(\mathbb{R}^2); \;\; \lim_{|\xi| \to +\infty} u = 0 \right\}.$$

Next, we define the integral operator $\mathcal{T}_0 : W_0^{1,-1}(\mathbb{R}^2) \to W_0^{1,-1}(\mathbb{R}^2)$ by

$$\int_{\mathbb{R}^2} \nabla \mathcal{T}_0 w \nabla v d\xi = \int_B \nabla w \nabla v d\xi \qquad \text{for all } v \in W_0^{1,-1}(\mathbb{R}^2).$$

We introduce the single layer vector space

$$\mathfrak{H} := \{ u \in W_0^{1,-1}(\mathbb{R}^2) : \Delta u = 0 \;\text{ in } B \cup \mathbb{R}^2 \setminus \overline{B}; \;\; u|_+ = u|_- \;\text{ on } \partial B \}.$$

We deduce from [21] that the restriction of $\mathcal{T}_0$ to $\mathfrak{H}$ is a self-adjoint operator of Fredholm type with index zero. In fact, $\frac{1}{2}I - \mathcal{T}_0$ is a compact operator.

Let us denote as $\left\{\beta_j^{\pm}\right\}_{j\geq 1}$ the eigenvalues of $\mathcal{T}_0 : \mathfrak{H} \to \mathfrak{H}$, ordered in the following way:

$$0 = \beta_1^- \leq \beta_2^- \leq ... \leq \beta_\infty^+ = \frac{1}{2},$$

and

$$\beta_\infty^+ = \frac{1}{2} \leq ... \leq \beta_2^+ \leq \beta_1^+ < 1,$$

and satisfies $\lim_{j\to+\infty} \beta_j^{\pm} = \beta_\infty^{\pm} = \frac{1}{2}$. We deduce immediately from the min-max principle for the compact, self-adjoint operator $\frac{1}{2}I - \mathcal{T}_0$ the following characterization of the spectrum of $\mathcal{T}_0$ [21].

Proposition 2.1. Let $\left\{w_j^{\pm}\right\}_{j\geq 1}$ be the set of corresponding eigenfunctions of the operator $\mathcal{T}_0$, associated to the eigenvalues $\left\{\beta_j^{\pm}\right\}_{j\geq 1}$. The following equalities hold:

$$\beta_j^- = \min_{\substack{u\in\mathfrak{H} \\ u\perp w_1^-,\ldots,w_{j-1}^-}} \frac{\displaystyle\int_D |\nabla u|^2 \, dx}{\displaystyle\int_\Omega |\nabla u|^2 \, dx} = \max_{\substack{F_j \subset \mathfrak{H} \\ \dim(F_j)=j-1}} \min_{u\in F_j^\perp} \frac{\displaystyle\int_D |\nabla u|^2 \, dx}{\displaystyle\int_\Omega |\nabla u|^2 \, dx}$$

and

$$\beta_j^+ = \max_{\substack{u\in\mathfrak{H} \\ u\perp w_1^+,\ldots,w_{j-1}^+}} \frac{\displaystyle\int_D |\nabla u|^2 \, dx}{\displaystyle\int_\Omega |\nabla u|^2 \, dx} = \min_{\substack{F_j \subset \mathfrak{H} \\ \dim(F_j)=j-1}} \max_{u\in F_j^\perp} \frac{\displaystyle\int_D |\nabla u|^2 \, dx}{\displaystyle\int_\Omega |\nabla u|^2 \, dx}$$

for all $j \geq 1$.

We define the quasi-static resonances $\left(\omega_j^{\pm}(0)\right)_{j\geq 1}$ of the spectral problem (2.7) the complex roots of the following dispersion equations:

$$\frac{\varepsilon_m(\omega)}{\varepsilon_s(z^\star)} \quad = \quad k_j^{\pm} := \frac{\beta_j^{\pm}}{\beta_j^{\pm}-1}, \qquad 1\leq j\leq \infty. \tag{2.8}$$

We first remark that since $\beta_j^{\pm}$ belong to $[0,1)$ the values on the right side of the equality $k_j^{\pm}$ are negative reals. Thus $\Re(\varepsilon_m(\omega))$, the real part of the electric permittivity, at the quasi-static plasmonic resonances $\left(\omega_j^{\pm}(0)\right)_{j\geq 1}$ takes negative reals. This is exactly what one would expect in such a situation, and the existence of the plasmonic resonances cannot occur if the material inside the nanoparticle is a modest electric permittivity that always has a strictly positive real part.

LEMMA 2.1. The complex roots to the dispersion relation (2.8) are explicitly given by

$$-i\frac{\Gamma}{2} \pm \sqrt{\frac{\omega_p^2}{\varepsilon_\infty - k_j^{\pm}\varepsilon_s(z^\star)} - \frac{\Gamma^2}{4}}, \tag{2.9}$$

where $\sqrt{z}$ is the complex square root function defined on $\mathbb{C}\setminus i(0,\infty)$.

We note that the quantities $k_j$ and $\varepsilon_\infty - 4\frac{\omega_p^2}{\Gamma^2}$ only depend, respectively, on the shape of the particle and the nature of the metal that fills the particle. Based on this calculation we remark that the circular shape has only four quasi-static resonances given by $-i\frac{\Gamma}{2}\pm\sqrt{\frac{\omega_p^2}{\varepsilon_\infty}-\frac{\Gamma^2}{4}}$, and $-i\frac{\Gamma}{2}\pm\sqrt{\frac{\omega_p^2}{\varepsilon_\infty+\varepsilon_s(z^\star)}-\frac{\Gamma^2}{4}}$. They satisfy, respectively, the dispersion equation with $k_1^- = 0$, and $k_\infty^{\pm} = -1$. We remark that only the resonances related to $k_\infty^{\pm} = -1$ depend on the surrounding media electric permittivity $\varepsilon_s(z^\star)$ and may provide later information on it. Finally, the eigenfunctions associated to $k_1^- = 0$, are constant on the boundary $\partial B$.

We follow the same steps as in the proof of Theorem 2.1 in [13, 14, 21] and prove the following asymptotic result.

PROPOSITION 2.2. Let $\omega(0)$ be a quasi-static resonance with multiplicity $m$. Then there exists a constant $\alpha_0 > 0$ such that for $0 < \alpha < \alpha_0$ there exist $m$ plasmonic resonances $(\omega_j(\alpha))_{1\leq j\leq m}$ satisfying the following asymptotic expansion as $\alpha \to 0$:

$$\frac{1}{m}\sum_{j=1}^{m}\omega_j(\alpha) = \omega(0) + o(1). \tag{2.10}$$

Next, we derive the asymptotic expansion of the electromagnetic fields when the size of the nanoparticle tends to zero.

2.2. *Small volume expansion of the EM fields.* Our strategy here is to use the tools developed in [6,9] and the references therein to derive the leading terms in the asymptotic expansion of electromagnetic fields when the volume of the nanoparticle tends to zero. Since the frequency of the incident wave is real and thus far away from the complex plasmonic resonances we expect that the remaining terms of the asymptotic expansion stay uniformly bounded.

Let $H_0 = H_i + H_{0s}$, be the total electric field in the absence of the nanoparticle. It satisfies the system

$$\nabla \cdot \left(\frac{1}{\varepsilon_s}\nabla H_0\right) + \omega^2 \mu_0 H_0 \quad = 0 \qquad \text{in} \quad \mathbb{R}^2 \tag{2.11}$$

with the Sommerfeld radiation condition as $|x| \to +\infty$:

$$\frac{\partial H_{0s}}{\partial |x|} - i\omega\sqrt{\varepsilon_0 \mu_0} H_{0s} \quad = \quad O(\frac{1}{\sqrt{|x|}}). \tag{2.12}$$

Recall that in the quasi-static regime the scattering resonances are far away from the real axis. Consequently, the system (2.11)-(2.12) above has a unique solution $H$ for any given real frequency $\omega$. Hence the following Green function $G(x, y)$ is well defined:

$$\nabla \cdot \left(\frac{1}{\varepsilon_s}\nabla G\right) + \omega^2 \mu_0 G \quad = \delta_y(x) \qquad \text{in} \quad \mathbb{R}^2 \tag{2.13}$$

with the Sommerfeld radiation condition as $|x| \to +\infty$:

$$\frac{\partial G}{\partial |x|} - i\omega\sqrt{\varepsilon_0 \mu_0} G \quad = \quad O(\frac{1}{\sqrt{|x|}}). \tag{2.14}$$

A simple integration by parts in the system (2.11)-(2.12) yields

$$H(x) = H_0(x) + \int_{B_\alpha} \left(\frac{1}{\varepsilon_m} - \frac{1}{\varepsilon_s(x)}\right) \nabla H(y) \nabla_y G(x, y) dy, \tag{2.15}$$

which leads to the following result.

Proposition 2.3. There exists a constant $C > 0$, independent of $\alpha$ and $H_i$ such that

$$\|H(x) - H_0(x)\|_{H^1(\Omega)} \leq C\alpha \|H_i\|_{H^1(\Omega)}.$$

This proposition shows that if $\omega$ is real, the field $H_0(x)$ is the first term in the asymptotic expansion of $H(x)$ when $\alpha$ tends to zero. However, the constant $C$ in the proposition depends on $\varepsilon(x)$ and $\omega$ can be large. In fact, considering the results in Proposition 2.2 and Lemma 2.1 if the attenuation $\Gamma$ tends to zero the plasmonic resonances will approach the real axis and then the constant $C$ may blow up. In such a situation one needs to take into account further terms in the asymptotic expansion of $H(x)$ when $\alpha$ tends to zero in order to improve the approximation. Here we will derive formally the first and second terms in the asymptotic expansion. In [9] a uniform asymptotic expansion of the magnetic field is derived using the method of matched asymptotic expansions for $\alpha$ small enough. Here we apply the same approach to obtain a formal asymptotic expansion of the electromagnetic fields. We shall represent the field $H(x)$ by two different expansions, an inner expansion for $x$ near $z^\star$, and an outer expansion for $x$ far away from $z^\star$.

The outer expansion takes the form

$$H(x) = H_0(x) + \alpha H_1(x) + \alpha^2 H_2(x) + \cdots \qquad \text{for} \quad |x - z^\star| \gg O(\alpha), \tag{2.16}$$

where $H_1, H_2$ satisfy the following Helmholtz equation:

$$\nabla \cdot \left(\frac{1}{\varepsilon_s}\nabla H_i\right) + \omega^2 \mu_0 H_i \quad = 0 \qquad \text{in} \quad |x - z^\star| \gg O(\alpha),$$

with the Sommerfeld radiation condition as $|x| \to +\infty$:

$$\frac{\partial H_i}{\partial |x|} - i\omega\sqrt{\varepsilon_0\mu_0}H_i \quad = \quad O(\frac{1}{\sqrt{|x|}}).$$

Introducing the microscale variable $\xi = (x - z^\star)/\alpha$, then the inner expansion can be written as

$$H(z^\star + \alpha\xi) = h_0(\xi) + \alpha h_1(\xi) + \alpha^2 \ln(\alpha) h_2(\xi) + \cdots \qquad \text{for} \quad |\xi| = O(1), \tag{2.17}$$

where the functions $h_0, h_1, h_2$ satisfy the following divergence form equations:

$$\nabla \cdot \left(\frac{1}{\tilde{\varepsilon}}\nabla h_0\right) \quad = 0 \qquad \text{in} \quad \mathbb{R}^2, \tag{2.18}$$

$$\nabla \cdot \left(\frac{1}{\tilde{\varepsilon}}\nabla h_1\right) + \nabla \cdot (\eta_1(\xi)\nabla h_0) \quad = 0 \qquad \text{in} \quad \mathbb{R}^2, \tag{2.19}$$

$$\nabla \cdot \left(\frac{1}{\tilde{\varepsilon}}\nabla h_2\right) \quad = 0 \qquad \text{in} \quad \mathbb{R}^2, \tag{2.20}$$

where $\eta_1(\xi)$ and $\eta_2(\xi)$ are the coefficients of the inner expansion of $\frac{1}{\varepsilon(z^\star + \alpha\xi)}$ given by

$$\frac{1}{\varepsilon(z^\star + \alpha\xi)} = \frac{1}{\tilde{\varepsilon}(\xi)} + \eta_1(\xi)\alpha + \eta_2(\xi)\alpha^2 + \cdots \tag{2.21}$$

with

$$\eta_1(\xi) = \begin{cases} \nabla(\frac{1}{\varepsilon_s})(z^\star)\xi & \text{in} \quad \mathbb{R}^2 \setminus \overline{B}, \\ 0 & \text{in} \quad B, \end{cases}$$

and

$$\eta_2(\xi) = \begin{cases} \nabla^2(\frac{1}{\varepsilon_s})(z^\star)\frac{\xi^2}{2} & \text{in} \quad \mathbb{R}^2 \setminus \overline{B}, \\ 0 & \text{in} \quad B. \end{cases}$$

Obviously the inner and outer expansions are not valid everywhere and the systems of equations satisfied by the functions $H_i$ and $h_i$ are not complete. In order to determine these functions uniquely, we need to equate the inner and the outer expansions in some overlap domain within which the microscale variable $\xi$ is large and $x - z^\star$ is small. In this domain the matching conditions are:

$$H_0(y) + \alpha H_1(y) + \alpha^2 H_2(y) + \cdots \sim h_0(\xi) + \alpha h_1(\xi) + \alpha^2 \ln(\alpha) h_2(\xi) + \cdots .$$

A change of variables in the Lippman-Schwinger integral representation formula (2.15) yields

$$\begin{aligned} &H(z^\star + \alpha\xi) \\ &= H_0(z^\star + \alpha\xi) + \alpha \int_B \left(\frac{1}{\varepsilon_m} - \frac{1}{\varepsilon_s(z^\star + \alpha\xi')}\right) \partial_{\xi_k}\left(H(z^\star + \alpha\xi')\right) \partial_{x_k} G(z^\star + \alpha\xi, z^\star + \alpha\xi') d\xi'. \end{aligned} \tag{2.22}$$

An asymptotic expansion of the quantities above gives

$$H_0(z^\star + \alpha\xi) = H_0(z^\star) + \partial_{x_i} H_0(z^\star)\xi_i\alpha + \partial^2_{x_i x_j} H_0(z^\star)\xi_i\xi_j\frac{\alpha^2}{2} + o(\alpha^2),$$

and

$$\alpha\partial_{\xi_k}G(z^\star+\alpha\xi,z^\star+\alpha\xi)=\varepsilon_s(z^\star)\partial_{\xi_k}\Phi_0(\xi,\xi')+\frac{1}{4\pi}\partial_{x_k}\varepsilon_s(z^\star)\alpha\ln(\alpha)+\alpha\Phi_1(\xi,\xi')+o(\alpha),$$

where $\Phi_0(\xi,\xi')=\frac{1}{2\pi}\ln(|\xi-\xi'|)$ is the Green function of the Laplacian in the whole space, and $\Phi_1(\xi,\xi')$ is a weakly singular function (see Theorem 5.1 in Appendix).

Inserting now the inner expansion of $H$, and the above asymptotic expansion into (2.22) we obtain

$$h_0(\xi)=H_0(z^\star),$$
$$h_1(\xi)=\partial_{x_i}H_0(z^\star)\xi_i+\left(\frac{\varepsilon_s(z^\star)}{\varepsilon_m}-1\right)\int_B\partial_{\xi_k}\Phi_0(\xi,\xi')\partial_{\xi_k}h_1(\xi')d\xi',$$

and

$$h_2(\xi)=\left(\frac{\varepsilon_s(z^\star)}{\varepsilon_m}-1\right)\int_B\partial_{\xi_k}\Phi_0(\xi,\xi')\partial_{\xi_k}h_2(\xi')d\xi'+\frac{1}{4\pi}\left(\frac{1}{\varepsilon_m}-\frac{1}{\varepsilon_s(z^\star)}\right)\partial_{x_k}\varepsilon_s(z^\star)\int_B\partial_{\xi_k}h_1(\xi')d\xi'.$$

Now we suppose that the functions $h_0, h_1$, and $h_2$ are defined not just in the domain $B$, but everywhere in $\mathbb{R}^2$. Considering the asymptotic expansions obtained from the Lipmann-Schwinger equation and matching conditions, we obtain

$$h_0(\xi)=H_0(z^\star), \tag{2.23}$$

$$\nabla\cdot\left(\frac{1}{\tilde{\varepsilon}}\nabla h_1(\xi)\right)=0\quad\text{in }\mathbb{R}^2, \tag{2.24}$$

$$\lim_{\xi\to+\infty}(h_1(\xi)-\partial_{x_i}H_0(z^\star)\xi_i)=0, \tag{2.25}$$

and

$$\nabla\cdot\left(\frac{1}{\tilde{\varepsilon}}\nabla h_2(\xi)\right)=0\text{ in }\mathbb{R}^2, \tag{2.26}$$

$$\lim_{\xi\to+\infty}\left(h_2(\xi)-\frac{1}{4\pi}\left(\frac{1}{\varepsilon_m}-\frac{1}{\varepsilon_s(z^\star)}\right)\partial_{x_k}\varepsilon_s(z^\star)\int_B\partial_{\xi_k}h_1(\xi')d\xi'\right)=0. \tag{2.27}$$

Using a variational approach in the Hilbert space $W_0^{1,-1}(\mathbb{R}^2)$ one can prove that the systems (2.24)- (2.25) and (2.26)- (2.27) have unique solutions. Precisely, it can be shown that $h_1(\xi)$ satisfies the following volume integral equation:

$$\left(\frac{\varepsilon_m(\omega)}{\varepsilon_s(z^\star)-\varepsilon_m(\omega)}I+\mathcal{T}_0\right)(h_1(\xi)-\partial_{x_i}H_0(z^\star)\xi_i)=\partial_{x_i}H_0(z^\star)\hat{\xi}_i(\xi), \tag{2.28}$$

where $\hat{\xi}_i(\xi)\in W_0^{1,-1}(\mathbb{R}^2)$ is the orthogonal projection of $\xi_i\chi_B(\xi)$ onto $W_0^{1,-1}(\mathbb{R}^2)$, which can be defined as the unique solution to the system

$$\int_{\mathbb{R}^2}\nabla\hat{\xi}_i\nabla v d\xi=\int_B\nabla\xi_i\nabla v d\xi\qquad\text{for all } v\in W_0^{1,-1}(\mathbb{R}^2). \tag{2.29}$$

Since $\mathcal{T}_0$ is self-adjoint and $\frac{\varepsilon_m(\omega)}{\varepsilon_s(z^\star)-\varepsilon_m(\omega)}$ has a non-zero imaginary component, the equation (2.28) has a unique solution.

Similarly, a forward calculation yields

$$\left(\frac{\varepsilon_m(\omega)}{\varepsilon_s(z^\star)-\varepsilon_m(\omega)}I+\mathcal{T}_0\right)\left(h_2(\xi)-\frac{1}{4\pi}\left(\frac{1}{\varepsilon_m}-\frac{1}{\varepsilon_s(z^\star)}\right)\partial_{x_k}\varepsilon_s(z^\star)\int_B\partial_{\xi_k}h_1(\xi')d\xi'\right)=0,$$

and, consequently,

$$h_2(\xi)=\frac{1}{4\pi}\left(\frac{1}{\varepsilon_m}-\frac{1}{\varepsilon_s(z^\star)}\right)\partial_{x_k}\varepsilon_s(z^\star)\int_B\partial_{\xi_k}h_1(\xi')d\xi', \tag{2.30}$$

is indeed a constant function.

Now, we shall determine the outer expansion functions $H_1$ and $H_2$. To do so we again consider the Lipmann-Schwinger equation

$$H(x)=H_0(x)+\alpha\int_B\left(\frac{1}{\varepsilon_m}-\frac{1}{\varepsilon_s(z^\star+\alpha\xi')}\right)\partial_{\xi_k}\left(H(z^\star+\alpha\xi')\right)\partial_{x_k}\left(G(x,z^\star+\alpha\xi')\right)d\xi'. \tag{2.31}$$

Using the inner expansion of $H$ and the regularity of the Green function $G$ we obtain

$$H_1(x)=0, \tag{2.32}$$

$$H_2(x)=\left(\frac{1}{\varepsilon_m}-\frac{1}{\varepsilon_s(z^\star)}\right)\int_B\partial_{\xi_k}h_1(\xi')d\xi'\partial_{x_k}G(x,z^\star). \tag{2.33}$$

It is well known that the inner and outer expansions are not valid uniformly in $x$ [9]. In order to obtain an asymptotic expansion of the fields as $\alpha$ tends to zero that is valid uniformly in space variable, we merge the two expansions together. Thus, adding the outer and inner expansions and subtracting out the common part, we formally find the following uniform expansions: for all $x\in\Omega$:

$$H(x)=H_0(x)+\alpha\mathcal{H}_1(\frac{x-z^\star}{\alpha})+\alpha^2\ln(\alpha)\mathcal{H}_2(\frac{x-z^\star}{\alpha})+\alpha^2H_2(x)+O(\alpha^2\ln(\alpha)), \tag{2.34}$$

where

$$\mathcal{H}_1(\xi)=h_1(\xi)-\xi_i\partial_{x_i}H_0(z^\star)+\left(\frac{\varepsilon_s(z^\star)}{\varepsilon_m}-1\right)\frac{1}{\pi}\int_B\partial_{\xi_i}h_1(\xi')d\xi'\frac{\xi_i}{|\xi|^2},$$

$$\mathcal{H}_2(\xi)=\frac{1}{4\pi}\left(\frac{1}{\varepsilon_m}-\frac{1}{\varepsilon_s(z^\star)}\right)\partial_{x_k}\varepsilon_s(z^\star)\int_B\partial_{\xi_k}h_1(\xi')d\xi'.$$

Following the steps of the proof of Theorem 2.1 in [9] one can obtain the following uniform asymptotic expansion.

THEOREM 2.1. For $\delta\in(0,1)$, there exists a constant $C>0$, independent of $\alpha$ and $H_i$ such that

$$\|H(x)-H_0(x)-\alpha\mathcal{H}_1(\frac{x-z^\star}{\alpha})-\alpha^2\ln(\alpha)\mathcal{H}_2(\frac{x-z^\star}{\alpha})-\alpha^2H_2(x)\|_{H^1(\Omega)}\leq C\alpha^2\|H_i\|_{H^1(\Omega)}.$$

The approximation can be improved by considering the inner expansion term of order $\alpha^2$ and computing the limit of $\Phi_1(\xi,\xi')$ as $\xi$ tends to $+\infty$. Opposite of the first impression, the term $\alpha^2H_2(x)$ on the right hand side is necessary to cancel out the singularity of $\mathcal{H}_1(\xi)$ when $\xi$ tends to zero. Finally, if $\partial_{x_k}\varepsilon_s(z^\star)=0$ one can recover the results of [9] by adding the order $\alpha^2$ inner term.

2.3. *The radial case.* Here we assume that $\Omega$ and $B$ are the unit disc, and $z^\star = 0$. We also assume that the electric permittivity $\varepsilon$ is piecewise constant.

Let $(r, \theta)$ be the polar coordinates in $\mathbb{R}^2$, let $m$ be a fixed integer larger than 1, and consider

$$H_i(r, \theta) = J_m(\frac{\omega}{c_0} r) e^{im\theta}$$

to be the magnetic incident field, where $J_m(\xi)$ is the Bessel function of the first kind of order $m$, and $c_0 = \frac{1}{\sqrt{\varepsilon_0 \mu_0}}$ is the speed of light in the free space.

Then, the total magnetic field takes the form $H(r, \theta) = h_\alpha(r) e^{im\theta}$, with

$$h(r) = \begin{cases} \kappa_1 H_m(\frac{\omega}{c_0} r) + J_m(\frac{\omega}{c_0} r) & \text{for } r \geq 1, \\ \kappa_2 H_m(\frac{\omega}{c_s} r) + \kappa_3 J_m(\frac{\omega}{c_s} r) & \text{for } \alpha \leq r \leq 1, \\ \kappa_4 J_m(\frac{\omega}{c_m} r) & \text{for } r \leq \alpha, \end{cases}$$

where $c_s = \frac{1}{\sqrt{\varepsilon_s \mu_0}}$, and $c_m = \frac{1}{\sqrt{\varepsilon_m \mu_0}}$ are the speed of light in the dielectric coating and in the metallic nanoparticle, respectively. $H_m(\xi)$ is the Hankel function of the first kind of order $m$.

The transmission conditions for $r = 1$ and $r = \alpha$ give the following system:

$$\begin{pmatrix} H_m(\frac{\omega}{c_0}) & -H_m(\frac{\omega}{c_s}) & -J_m(\frac{\omega}{c_s}) & 0 \\ \frac{c_0}{c_s} H'_m(\frac{\omega}{c_0}) & -H'_m(\frac{\omega}{c_s}) & -J'_m(\frac{\omega}{c_s}) & 0 \\ 0 & H_m(\frac{\omega}{c_s}\alpha) & J_m(\frac{\omega}{c_s}\alpha) & -J_m(\frac{\omega}{c_m}\alpha) \\ 0 & \frac{c_s}{c_m} H'_m(\frac{\omega}{c_s}\alpha) & \frac{c_s}{c_m} J'_m(\frac{\omega}{c_s}\alpha) & -J'_m(\frac{\omega}{c_m}\alpha) \end{pmatrix} \overrightarrow{\kappa} = \begin{pmatrix} -J_m(\frac{\omega}{c_0}) \\ -J'_m(\frac{\omega}{c_0}) \\ 0 \\ 0 \end{pmatrix}.$$

The plasmonic resonances, in this case, are exactly the zeros of the determinant $d_\alpha(\omega)$, of the scattering matrix. An asymptotic expansion of the latter when $\alpha$ tends to zero gives

$$d_\alpha(\omega) = \frac{d_0(\omega)}{\alpha} + o(\frac{1}{\alpha}),$$

where

$$d_0(\omega) := \left( -H_m(\frac{\omega}{c_0}) J'_m(\frac{\omega}{c_s}) + \frac{c_s}{c_0} H'_m(\frac{\omega}{c_0}) J_m(\frac{\omega}{c_s}) \right) \frac{c_s^m}{\pi\omega} (c_m^2 + c_s^2) \frac{1}{c_m^{m+1}}.$$

Hence a limiting value $\omega(0)$ of a sequence of plasmonic resonances has to be finite and satisfy the dispersion equation $d_0(\omega(0)) = 0$. We remark that the complex roots of the function

$$-H_m(\frac{\omega}{c_0}) J'_m(\frac{\omega}{c_s}) + \frac{c_0}{c_s} H'_m(\frac{\omega}{c_0}) J_m(\frac{\omega}{c_s})$$

are exactly the scattering resonances of the domain $\Omega$ in the absence of the nanoparticle. If we drop the assumption that $\omega$ is small, and if the material that fills the nanoparticle is non-dispersive, we obtain the well-known convergence of the scattering resonances to the non-perturbed ones (see, for instance, [6, 9]).

A careful analysis of the zeros of $d_0(\omega)$ in the quasi-static regime leads to $\varepsilon_m(\omega(0)) = -\varepsilon_s$ or $\varepsilon_m(\omega(0)) = 0$, which correspond exactly to the plasmonic values of the circular shape nanoparticle $\beta_\infty^\pm = \frac{1}{2}$ and $\beta_1^- = 0$ (see, for instance, (2.8)).

In the case where $m$ is equal to one the determinant $d_\alpha(\omega)$ has the following asymptotic expansion $d_\alpha(\omega) = d_0(\omega)\ln(\alpha) + o(\ln(\alpha))$ as $\alpha$ tends to zero. Using the Rouché Theorem one can determine the complete asymptotic expansion of the plasmonic resonances in the case of a circular shape.

**3. Photoacoustic effect.** In this section we consider a metallic nanoparticle in a liquid medium and we want to describe the photoacoustic generation created by the electromagnetic heating of the nanoparticle. We derive the model equations that describe the coupling between the temperature rise in the medium and the acoustic wave generation.

3.1. *Acoustic sources.* We write the fundamental equations of acoustics as explained in [53], i.e., the equation of continuity, the Euler equation, and the continuity equation for heat flow

$$\frac{\partial \rho}{\partial t} = -\rho_0 \mathrm{div}(v), \tag{3.1}$$

$$\rho_0 \frac{\partial v}{\partial t} = -\nabla p, \tag{3.2}$$

$$\rho_0 T \frac{\partial s}{\partial t} = \mathrm{div}(\kappa \nabla T) + P_v, \tag{3.3}$$

where $\rho$ is the mass density, $p(r,t)$ is the acoustic pressure, $v(r,t)$ is the acoustic displacement velocity, $s(r,t)$ is the specific entropy, $T(r,t)$ is the temperature, and $P_v$ is the heat source. The change of density is assumed small ($\frac{\rho-\rho_0}{\rho_0} \ll 1$). The thermal conduction $\kappa$ is given by

$$\kappa(x) \;=\; \begin{cases} \kappa_s(x) & \text{for } x \in \Omega \setminus \overline{B_\alpha}, \\ \kappa_0 & \text{for } x \in B_\alpha, \end{cases}$$

where $\kappa_s(x) > 0$ is the thermal conduction of the liquid and $\kappa_0 > 0$ is the thermal conduction of the metal that fills the nanoparticle, and verifies $\kappa_0 \gg \kappa_s$.

We can write the two equations of state giving the change of density $\delta\rho$ and the change of entropy $\delta s$ in terms of $\delta p$ and $\delta T$ [45]

$$\delta\rho = \frac{\gamma}{c_s^2}\delta p - \rho_0 \beta \delta T, \tag{3.4}$$

$$\delta s = \frac{c_p}{T}(\delta T - \frac{\gamma - 1}{\rho_0 \beta c_s^2}\delta p), \tag{3.5}$$

where $c_p = T\left(\frac{\partial s}{\partial T}\right)_p$ is the specific heat capacity at constant pressure, $c_v = T\left(\frac{\partial s}{\partial T}\right)_\rho$ is the specific heat capacity at constant volume, $\gamma = \frac{c_p}{c_v}$, $\beta = -\frac{1}{\rho}\left(\frac{\partial \rho}{\partial T}\right)_p$ is the thermal expansion coefficient, and $c_s$ is the isentropic sound velocity.

We deduce from equations (3.4) and (3.5) the two following equations:

$$\frac{\partial \rho}{\partial t} = \frac{\gamma}{c_s^2}\frac{\partial p}{\partial t} - \rho_0 \beta \frac{\partial T}{\partial t}, \tag{3.6}$$

$$\frac{\partial s}{\partial t} = \frac{c_p}{T}(\frac{\partial T}{\partial t} - \frac{\gamma - 1}{\rho_0 \beta c_s^2}\frac{\partial p}{\partial t}). \tag{3.7}$$

We can make the assumption for liquids that $\gamma = 1$. With this assumption and combining equations (3.3) and (3.7), we obtain the following equation for the temperature field $T$:

$$\rho_0 c_p \frac{\partial T}{\partial t} = \operatorname{div}(\kappa \nabla T) + P_v. \tag{3.8}$$

We now use equations (3.1) and (3.2) to get $\frac{\partial^2 \rho}{\partial t^2} - \Delta p = 0$. We can transform this equation thanks to equation (3.4) and we obtain:

$$\frac{\gamma}{c_s^2}\frac{\partial^2 p}{\partial t^2} - \Delta p = \rho_0 \frac{\partial}{\partial t}\left(\beta \frac{\partial T}{\partial t}\right). \tag{3.9}$$

With the assumption that $\gamma = 1$ and that $\beta = \beta_0$, we finally have the following system of coupled equations for the generation of photoacoustic waves in a liquid medium:

$$\rho_0 c_p \frac{\partial T}{\partial t} = \operatorname{div}(\kappa \nabla T) + P_v, \tag{3.10}$$

$$\frac{1}{c_s^2}\frac{\partial^2 p}{\partial t^2} - \Delta p = \rho_0 \beta_0 \frac{\partial^2 T}{\partial t}. \tag{3.11}$$

3.2. *Electromagnetic sources.* The source term $P_v$ in equation (3.3) is the energy produced by electromagnetic heating. It can be written as follows [52]:

$$P_v = Q_{gen} + Q_{met}, \tag{3.12}$$

where $Q_{gen}$ is the volumetric power density of the electromagnetic source, and $Q_{met}$ is the metabolic heat generated by biological tissues. We consider here that $Q_{met} = 0$.

The electromagnetic coefficients of the medium are the complex electric permittivity $\varepsilon_s$, the magnetic permeability $\mu_0$. Since the electromagnetic wave is time pulsed and because of the difference of time scales between the acoustic and electromagnetic waves, the volumetric power density is described by the time averaging of the real part of the divergence of the Poynting vector $\mathbf{S} = \mathbf{E} \times \overline{\mathbf{H}}$ times the Dirac function at zero. On the other hand the divergence of $\mathbf{S}$ is given by

$$-\nabla \cdot \mathbf{S} = i\omega\overline{\varepsilon}|\mathbf{E}|^2 + i\omega\overline{\mu}_0|\mathbf{H}|^2. \tag{3.13}$$

By taking the real part and time averaging of the divergence of the Poynting vector we finally have

$$Q_{gen} = \omega \Im(\varepsilon)\langle|\mathbf{E}|^2\rangle\delta_0(t) = \omega\Im(\varepsilon)|E|^2\delta_0(t), \tag{3.14}$$

where the time averaging is defined by $\langle f\rangle := \lim_{\tau\to+\infty}\int_0^\tau f(t)dt$, and $\delta_0$ is the Dirac function at 0.

We can finally write the following system of coupled equations that describes the photoacoustic generation by the electromagnetic heating of a metallic nanoparticle

$$\rho_0 c_p \frac{\partial T}{\partial t} = \operatorname{div}(\kappa \nabla T) + \omega\Im(\varepsilon)|E|^2\delta_0(t), \tag{3.15}$$

$$\frac{1}{c_s^2}\frac{\partial^2 p}{\partial t^2} - \Delta p = \rho_0\beta_0\frac{\partial^2 T}{\partial t}, \tag{3.16}$$

with the initial conditions at $t = 0$:

$$T = p = \frac{\partial p}{\partial t} = 0. \tag{3.17}$$

Following the same analysis as in [10] one can show that the temperature $T$ approaches $T_0$ as $\alpha$ tends to zero, where $T_0$ is the solution to

$$\rho_0 c_p \frac{\partial T_0}{\partial t} = \operatorname{div}(\kappa_s \nabla T_0) + \omega \Im(\varepsilon)|E|^2 \delta_0(t),$$

with initial boundary condition $T_0 = 0$ at $t = 0$, and $\lim_{|x|\to+\infty} T_0(x) = 0$. Here we did not consider the first and second terms in the small volume asymptotic expansion because the thermal conduction $\kappa$ is frequency independent, and hence the limiting problems are well-posed compared to the ones in the asymptotic expansion of the EM fields.

Since the conductivity $\kappa_s$ of the biological is very small compared to the other quantities we neglect it and find the following equation for the temperature:

$$\rho_0 c_p \frac{\partial T_0}{\partial t} = \omega \Im(\varepsilon)|E|^2 \delta_0(t),$$

which combined with the acoustic waves (3.16), provides at the end the following model for the photoacoustic effect by a metallic nanoparticle:

$$\begin{cases} \frac{1}{c_s^2}\frac{\partial^2 p}{\partial t^2}(x,t) - \Delta p(x,t) = 0 & \text{in} \quad \mathbb{R}^2 \times \mathbb{R}_+, \\ p(x,0) = \frac{\omega\beta_0}{c_p}\Im(\varepsilon)(x)|E(x)|^2 & \text{in} \quad \mathbb{R}^2, \\ \frac{\partial p}{\partial t}(x,0) = 0 & \text{in} \quad \mathbb{R}^2. \end{cases} \tag{3.18}$$

The system above (3.18) coupled with the Helmholtz equation (2.1)-(2.2) represents the forward problem. Next, we study the photoacoustic inverse problem.

**4. The photoacoustic inverse problem.** In this section we study the inverse problem of the reconstruction of the electric permittivity $\varepsilon$ from the measurements of the acoustic waves $p(x,t)$, $(x,t) \in \partial\Omega \times (0,\tau_p)$, generated by the photoacoustic effect from the heating of the small metallic nanoparticle $B_\alpha$ in the presence of electromagnetic fields at a frequency close to a plasmonic resonance. Here $\tau_p > 0$ is the period of time where the measurements are taken, that will be specified later. We have two inversions: the acoustic inversion where we assume that the speed of the wave is a known constant $c_s$ and reconstruct the initial pressure $\Im(\varepsilon(x))|E(x)|^2$, $x \in \Omega$ from the knowledge of $p(x,t)$, $(x,t) \in \partial\Omega \times (0,\tau_p)$; the second step is to recover the electric permittivity $\varepsilon(x)$ from the internal data $\Im(\varepsilon(x))|E(x)|^2$, $x \in \Omega$.

4.1. *Acoustic inversion.* Recall that $\Im(\varepsilon)(x)$ is a compactly supported function in $\Omega$, and that we have assumed that the acoustic wave speed in the tissue takes a constant value $c_p$ that corresponds to the isentropic acoustic speed in the water, that is, 1400 m/s. These two assumptions allow us to use well-known results from control theory to derive a stability estimate for the acoustic inversion. The following result is based on the multiplier method and can be found in [30, 39].

THEOREM 4.1. Let $\tau_p > \tau_\Omega$ where $\tau_p = \sup_{x,y\in\Omega} |x-y|$. Then, there exists a constant $C = C(\Omega) > 0$ such that

$$\frac{\omega\beta_0}{c_p}\|\Im(\varepsilon(x))|E(x)|^2\|_{L^2(\Omega)} \leq C\|\frac{\partial p}{\partial t}\|_{L^2(\partial\Omega\times(0,\tau_p))} + \|\nabla p\|_{L^2(\partial\Omega\times(0,\tau_p))}.$$

We refer the readers to the survey [36] on related reconstruction methods and different approaches based on integral equations for constant acoustic speed. The stability result shows that the reconstruction of the electromagnetic energy responsible for the generation of the acoustic signal by heating the nanoparticle from boundary measurements of the acoustic waves is stable if the observation time $\tau_p$ is large enough. This result can be extended to a non-constant acoustic speed as well as measurements of the acoustic waves on a small part of the boundary [4, 33, 49]. In this paper for the sake of simplicity we do not handle such general cases.

We further assume that the constants $\beta_0$ and $c_p$ are given. Let $\mathcal{O}_M$ denote the ball centered at 0 and of radius $M > 0$ in $H^2(B_R(z^\star))$, where $R > 0$ is large enough such that $\overline{\Omega} \subset B_R(z^\star)$.

COROLLARY 4.1. Assume that $\varepsilon \in B_M(0)$, and let $\tau_p > \tau_\Omega$. Then, there exists a constant $C = C(\omega, M, \beta_0, c_p) > 0$ such that the following estimate:

$$\|\Im(\varepsilon)|\nabla H|^2\|_{C^0(\overline{\Omega})} \leq C\left(\left\|\frac{\partial p}{\partial t}\right\|_{L^2(\partial\Omega\times(0,\tau_p))} + \|\nabla p\|_{L^2(\partial\Omega\times(0,\tau_p))}\right)^{\frac{1}{4}}, \tag{4.1}$$

holds.

*Proof.* A simple calculation yields $|E(x)|^2 = |\nabla H(x)|^2$ over $\Omega$. Using the interpolation between Sobolev spaces [40], we estimate $\Im(\varepsilon)|\nabla H|^2$ in $H^{\frac{3}{2}}(\Omega)$ in terms of its norms in $L^2(\Omega)$ and $H^2(\Omega)$, respectively. Thus we deduce (4.1) from elliptic regularity of the system (2.1) and the estimate in Theorem 4.1. □

4.2. *Optical inversion.* In this part of the paper we assume that the internal electromagnetic energy

$$\Im(\varepsilon(x))|\nabla H(x)|^2$$

for $x \in \Omega$ is recovered, and we study the inverse problem of determining $\varepsilon(x)$ over $\Omega$ using the small volume asymptotic expansion of the EM fields in the previous section. In fact in applications we only need to recover the imaginary part of the electric permittivity which is related to the absorption of the EM fields and the generation of the photoacoustic wave.

Recall that the absorption of EM energy by only the biological tissue is negligible inside $\Omega$. In practice the photoacoustic signal generated by such absorption is weak inside $\Omega$ and cannot be used to image the tissue itself.

From section 2 we deduce the inner and outer asymptotic expansions of the magnetic field $|\nabla H(x)|^2$. Our strategy here is to first analyze the information about the medium and the nanoparticle contained in the outer asymptotic expansion. This problem is a classical boundary/internal inverse problem, and has some known limitations. Then we

complete the recovery of the optical properties of the medium using information retrieved from the inner expansion of the magnetic field and the a priori information about the shape of the nanoparticle.

4.2.1. *Inversion using the outer expansion.* Recall the outer asymptotic expansion (2.16)- (2.32) of the magnetic field:

$$H(x) = H_0(x) + \alpha^2 H_2(x) + o(\alpha^2) \qquad \text{for } x \in \partial\Omega,$$

where $H_0(x)$ is the solution to the system (2.11)- (2.12), and $H_2(x)$ is given by

$$H_2(x) = \left(\frac{1}{\varepsilon_m(\omega)} - \frac{1}{\varepsilon_s(z^\star)}\right) \int_B \partial_{\xi_k} h_1(\xi') d\xi' \partial_{x_k} G(x, z^\star)$$

with $h_1(\xi)$ the unique solution to the system (2.24)-(2.25).

In fact the asymptotic expansion above is valid in a neighboring region of the boundary $\partial\Omega$, but since the internal data is of the form $\Im(\varepsilon(x))|\nabla H(x)|^2$, where $\Im(\varepsilon)$ is compactly supported in $\Omega$, we can only retrieve information about the magnetic field on the boundary $\partial\Omega$. Note that since $\varepsilon_0$ is given one can retrieve the Cauchy data of the magnetic field on $\partial\Omega$ from the knowledge of its trace on the same set.

The function $H_2(x)$ can be rewritten in terms of the first order polarization tensor $M(\frac{\varepsilon_m(\omega)}{\varepsilon_s(z^\star)}) = (M_{kl})_{1\le k,l\le 2}$, as follows (see, for instance, [7] and the references therein):

$$H_2(x) = \left(\frac{1}{\varepsilon_m(\omega)} - \frac{1}{\varepsilon_s(z^\star)}\right) \nabla G(x, z^\star) \cdot M \nabla H_0(z^\star),$$

where

$$M_{kl} = \int_B \partial_{\xi_k} \phi_l(\xi') d\xi', \tag{4.2}$$

and $\phi_l(\xi), l = 1, 2$ are the unique solutions to the system

$$\nabla \cdot \left(\frac{1}{\tilde{\varepsilon}} \nabla \phi_l(\xi)\right) = 0 \quad \text{in } \mathbb{R}^2, \tag{4.3}$$

$$\lim_{\xi \to +\infty} (\phi_l(\xi) - \xi_l) = 0. \tag{4.4}$$

On the other hand, $\phi_l(\xi), l = 1, 2$ can be rewritten as follows:

$$\phi_l(\xi) = \xi_l - \left(\frac{\varepsilon_m(\omega)}{\varepsilon_s(z^\star) - \varepsilon_m(\omega)} I + \mathcal{T}_0\right)^{-1} \hat{\xi}_l(\xi), \tag{4.5}$$

where $\hat{\xi}_l(\xi) \in W_0^{1,-1}(\mathbb{R}^2)$ is the orthogonal projection of $\xi_l \chi_B(\xi)$ onto $W_0^{1,-1}(\mathbb{R}^2)$ defined in (2.29).

Regarding the integral equation (4.5), we observe that when $\omega$ tends to a plasmonic resonance $\omega_j(\alpha)$ the functions $\phi_l(\xi)$, and, consequently, the polarization tensor $M$ will most likely blow up. Since in applications $\omega$ is real, and the plasmonic resonances of the nanoparticle embedded in the medium approaches the quasi-static resonances $\omega_j(0)$ when $\alpha$ tends to zero (Proposition 2.2, we expect that the coefficient $M$ becomes large in the case where $\omega$ coincides with $\Re(\omega_j(0))$, and $\Gamma \ll 1$.

Many works have considered the localization of small inhomogeneities in a known background medium, and most of the proposed methods are based on an appropriate averaging of the asymptotic expansion by using particular background solutions as weights [7, 12].

In other words, the position $z^\star$ of the nanoparticle can be uniquely determined from the outer expansion of $H(x)$, that is, $H_0(x) + \alpha^2 H_2(x),\ x \in \partial\Omega$, if the electric permittivity of the background medium $\varepsilon_s(x)$ is known everywhere. But this is not the case in our problem, since our objective is to determine $\varepsilon_s(x)$, while $\varepsilon_m(\omega)$ is known (which is the complete opposite of the setting where small inhomogeneities are imaged). Here to overcome these difficulties we may propose the use of multifrequency measurements $H_2(x), \omega \in (\underline{\omega}, \overline{\omega})$ to localize $z^\star$ [15, 28], where $\underline{\omega}, \overline{\omega}$ are two strictly positive constants satisfying $\underline{\omega} \ll \overline{\omega}$. We will study this specific inverse problem in future works. From now on we assume that the position $z^\star$ of the nanoparticle is known.

Note that in general if $\varepsilon_s(x)$ is known, it is still not possible to recover simultaneously the shape of the nanoparticle $\partial B$ and the contrast $\frac{\varepsilon_s(z^\star)}{\varepsilon_m(\omega)}$ from only the measurement of the outer expansion $H_0(x) + \alpha^2 H_2(x),\ x \in \partial\Omega$. Meanwhile in our setting the shape of the nanoparticle is assumed to be known. For example, if we consider the circular shape, that is, $B$ is the unit disc, $\hat{\xi}_l(\xi)$, $l = 1, 2$, and hence $\phi_l(\xi)$, $l = 1, 2$, can be determined explicitly

$$\hat{\xi}_l(\xi) = \begin{cases} \frac{\xi_l}{2} & \text{for } \xi \in B, \\ \frac{\xi_l}{2|\xi|^2} & \text{for } \xi \in \mathbb{R}^2 \setminus \overline{B}, \end{cases} \tag{4.6}$$

$$\phi_l(\xi) = \begin{cases} \frac{2\varepsilon_m(\omega)}{\varepsilon_s(z^\star)+\varepsilon_m(\omega)}\xi_l & \text{for } \xi \in B, \\ \xi_l - \frac{\varepsilon_s(z^\star)-\varepsilon_m(\omega)}{\varepsilon_s(z^\star)+\varepsilon_m(\omega)}\frac{\xi_l}{|\xi|^2} & \text{for } \xi \in \mathbb{R}^2 \setminus \overline{B}, \end{cases} \tag{4.7}$$

which implies that the polarization tensor can be simplified into

$$M_{kl} = \frac{2\varepsilon_m(\omega)}{\varepsilon_s(z^\star) + \varepsilon_m(\omega)} |B| \delta_{kl},$$

where $\delta_{kl}$ is the Kronecker delta. Assuming that $H_0(x),\ x \in \partial\Omega$ is given, we deduce from the outer expansion the following approximation [7, 12]:

$$\frac{1}{\varepsilon_0} \int_{\partial\Omega} \left( H \frac{\partial H_0}{\partial \nu_\Omega} - \frac{\partial H}{\partial \nu_\Omega} H_0 \right) ds(x) \tag{4.8}$$

$$= \alpha^2 \left( \frac{1}{\varepsilon_m(\omega)} - \frac{1}{\varepsilon_s(z^\star)} \right) \nabla H_0(z^\star) \cdot M \nabla H_0(z^\star) + o(\alpha^2)$$

$$= 2|B| \frac{\varepsilon_s(z^\star) - \varepsilon_m(\omega)}{\varepsilon_m(\omega) + \varepsilon_s(z^\star)} \frac{1}{\varepsilon_s(z^\star)} \left| \nabla H_0(z^\star) \right|^2 \alpha^2 + o(\alpha^2). \tag{4.9}$$

To ensure that the first term of the asymptotic expansion does not vanish, and to guarantee the success of the identification procedure, it becomes necessary to assume the following non-degeneracy condition:

$$\left| \nabla H_0(z^\star) \right|^2 \neq 0.$$

For a circular shape nanoparticle we can immediately see from the explicit expression of the first term in the asymptotic expansion that when $\omega$ is close to a plasmonic resonance, that is, $\varepsilon_m(\omega) = -\varepsilon_s(z^\star)$, the polarization tensor constant blows up. In the next paragraph we investigate the inner expansion of the magnetic field which represents our photoacoustic data, in order to derive the contrast $\frac{\varepsilon_s(z^\star)}{\varepsilon_m(\omega)}$.

4.2.2. *Inversion using the inner expansion.* We further assume that the position $z^\star$, the size $\alpha$, and the shape $\partial B$ of the nanoparticle are known. Recall the inner expansion (2.17):

$$H(z^\star+\alpha\xi) = H_0(z^\star) + \alpha h_1(\xi) + \alpha^2 \ln(\alpha) h_2(\xi) + O(\alpha^2) \qquad \text{for} \quad |\xi| = O(1),$$

where $h_1(\xi)$ is the unique solution to the system (2.24)-(2.25), that is,

$$\begin{aligned} \nabla \cdot \left( \frac{1}{\tilde{\varepsilon}} \nabla h_1(\xi) \right) &= 0 \quad \text{in } \mathbb{R}^2, \\ \lim_{\xi \to +\infty} \left( h_1(\xi) - \partial_{x_i} H_0(z^\star) \xi_i \right) &= 0, \end{aligned}$$

and $h_2(\xi)$ is a constant function given by

$$h_2(\xi) = \frac{1}{4\pi} \left( \frac{1}{\varepsilon_m} - \frac{1}{\varepsilon_s(z^\star)} \right) \partial_{x_k} \varepsilon_s(z^\star) \int_B \partial_{\xi_k} h_1(\xi') d\xi'.$$

Using the functions $\phi_l, l = 1, 2$ solutions to the system (4.3)-(4.4), we can rewrite $h_1(\xi)$ as

$$h_1(\xi) = \phi_k(\xi) \partial_{x_k} H_0(z^\star). \tag{4.10}$$

Recall that the acoustic inversion provides the internal function

$$\Psi(x) = \Im(\varepsilon(x)) |\nabla H(x)|^2, \qquad x \in \Omega.$$

Combining (2.17) and (2.21), we obtain the following inner expansion:

$$\begin{aligned} \Psi(z^\star + \alpha\xi) &= \Im(\varepsilon(z^\star + \alpha\xi)) |\nabla H(z^\star + \alpha\xi)|^2 \\ &= \Im(\tilde{\varepsilon}(\xi)) |\nabla_\xi h_1(\xi)|^2 + O(\alpha^2) \qquad \text{for} \quad |\xi| = O(1). \end{aligned} \tag{4.11}$$

We further assume that $B$ is the unit disc. Our objective is to recover $\varepsilon_s(z^\star)$ from the knowledge of $\Im(\tilde{\varepsilon}(\xi)) |\nabla_\xi h_1(\xi)|^2$ for $\xi \in 2B$, where $2B$ is the disc of center zero and radius 2.

Combining (4.10) and (4.7), we find

$$h_1(\xi) = \begin{cases} (1-\kappa)\, \xi \cdot \nabla H_0(z^\star) & \text{for } \xi \in B, \\ \left(1 - \frac{\kappa}{|\xi|^2}\right) \xi \cdot \nabla H_0(z^\star) & \text{for } \xi \in 2B \setminus \overline{B}, \end{cases}$$

where

$$\kappa := \frac{\varepsilon_s(z^\star) - \varepsilon_m(\omega)}{\varepsilon_s(z^\star) + \varepsilon_m(\omega)}.$$

Hence

$$\Psi(z^\star + \alpha\xi) + o(\alpha) = \begin{cases} \Im(\varepsilon_m(\omega)) |1-\kappa|^2 |\nabla H_0(z^\star)|^2 & \text{for } \xi \in B, \\ \Im(\varepsilon_s(z^\star)) \left| \nabla_\xi \left( \left(1 - \frac{\kappa}{|\xi|^2}\right) \frac{\xi}{|\xi|^2} \cdot \nabla H_0(z^\star) \right) \right|^2 & \text{for } \xi \in 2B \setminus \overline{B}. \end{cases}$$

A forward calculation yields

$$\Psi(z^\star + \alpha\xi) = \Im(\varepsilon_s(z^\star)) \left| \left(1 - \frac{\kappa}{|\xi|^2}\right) \nabla H_0(z^\star) + 2\kappa \frac{\xi}{|\xi|^2} \cdot \nabla H_0(z^\star) \frac{\xi}{|\xi|^2} \right|^2 + O(\alpha)$$

for $\xi \in 2B \setminus \overline{B}$.

Now taking the ratio between $\Psi|^{+}_{\partial B_\alpha}$ and $\Psi(z^\star) = \fint_{B_\alpha} \Psi(x)dx$, we obtain

$$\begin{aligned}\frac{\Psi(z^\star+\alpha\xi)|_+}{\Psi(z^\star)} &= \frac{\Im(\varepsilon_s(z^\star))}{\Im(\varepsilon_m(\omega))}\left(\left|\frac{1+\kappa}{1-\kappa}\right|^2\left|\frac{\nabla H_0(z^\star)}{|\nabla H_0(z^\star)|}\cdot\xi\right|^2+\left|\frac{\nabla H_0(z^\star)}{|\nabla H_0(z^\star)|}\cdot\xi^\perp\right|^2\right)+O(\alpha),\\ &= \Psi_0(\xi)+O(\alpha) \qquad (4.12)\end{aligned}$$

for $\xi\in\partial B=\left\{\xi'\in\mathbb{R}^2;\ \ |\xi'|=1\right\}$, where $\xi^\perp$ is a $\frac{\pi}{2}$ counterclockwise rotation of $\xi$.

Now, assuming that $|\Re(\varepsilon_m(\omega))|>|\Re(\varepsilon_s(z^\star))|$, we have

$$\left|\frac{1+\kappa}{1-\kappa}\right|>1,$$

and thus the function $\Psi_0(\xi)$ takes its maximum and minimum on $\partial B$ at $\xi=\pm\frac{\nabla H_0(z^\star)}{|\nabla H_0(z^\star)|}$ and $\xi=\pm\frac{\nabla H_0(z^\star)^\perp}{|\nabla H_0(z^\star)|}$, respectively.

Consequently,

$$\begin{aligned}\frac{\Im(\varepsilon_s(z^\star))}{\Im(\varepsilon_m(\omega))} &= \frac{\Psi(z^\star+\alpha\frac{\nabla H_0(z^\star)^\perp}{|\nabla H_0(z^\star)|})|_+}{\Psi(z^\star)}+O(\alpha) \qquad (4.13)\\ &= \min_{\xi\in\partial B}\frac{\Psi(z^\star+\alpha\xi)|_+}{\Psi(z^\star)}+O(\alpha) \qquad (4.14)\end{aligned}$$

and

$$\begin{aligned}\frac{\Im(\varepsilon_s(z^\star))}{\Im(\varepsilon_m(\omega))}\left|\frac{1+\kappa}{1-\kappa}\right|^2 &= \frac{\Psi(z^\star+\alpha\frac{\nabla H_0(z^\star)}{|\nabla H_0(z^\star)|})|_+}{\Psi(z^\star)}+O(\alpha) \qquad (4.15)\\ &= \max_{\xi\in\partial B}\frac{\Psi(z^\star+\alpha\xi)|_+}{\Psi(z^\star)}+O(\alpha). \qquad (4.16)\end{aligned}$$

Since $\varepsilon_m(\omega)$ is given, we can retrieve $\Im(\varepsilon_s(z^\star))$ from equality (4.13), and then $\Re(\varepsilon_s(z^\star))$ from equality (4.14). Now, we are able to prove the main stability estimate.

4.3. *Proof of the main Theorem* 1.1. We deduce from equalities (4.13)-(4.14) the following estimates.

THEOREM 4.2. Under the same assumptions as in Theorem 1.1, there exists a constant $C>0$ that does not depend on $\alpha$, such that

$$|\Im(\varepsilon_{s,a}(z^\star))-\Im(\varepsilon_{s,a}(z^\star))|\le C\|\Psi_a-\Psi_b\|_{L^\infty(2B_\alpha)}+O(\alpha).$$

*Proof.* Equalities (4.13)-(4.14) imply

$$\begin{aligned}\Psi_{0,a}\left(\frac{\nabla H_{0,a}(z^\star)^\perp}{|\nabla H_{0,a}(z^\star)|}\right) &= \min_{\xi\in\partial B}\Psi_{0,a}(\xi)\\ &= \min_{\xi\in\partial B}\left(\Psi_{0,b}(\xi)+\Psi_{0,a}(\xi)-\Psi_{0,b}(\xi)\right).\end{aligned}$$

Therefore,

$$\begin{aligned}\min_{\xi\in\partial B}\left(\Psi_{0,b}(\xi)-|\Psi_{0,a}(\xi)-\Psi_{0,b}(\xi)|\right) &\le \Psi_{0,a}\left(\frac{\nabla H_{0,a}(z^\star)^\perp}{|\nabla H_{0,a}(z^\star)|}\right)\\ &\le \min_{\xi\in\partial B}\left(\Psi_{0,b}(\xi)+|\Psi_{0,a}(\xi)-\Psi_{0,b}(\xi)|\right),\end{aligned}$$

which implies

$$|\Psi_{0,a}(\frac{\nabla H_{0,a}(z^\star)^\perp}{|\nabla H_{0,a}(z^\star)|}) - \Psi_{0,b}(\frac{\nabla H_{0,b}(z^\star)^\perp}{|\nabla H_{0,a}(z^\star)|})| \leq \max_{\xi\in\partial B} |\Psi_{0,a} - \Psi_{0,b}|, \tag{4.17}$$

and, consequently,

$$|\Psi_{0,a}(\frac{\nabla H_{0,a}(z^\star)^\perp}{|\nabla H_{0,a}(z^\star)|}) - \Psi_{0,a}(\frac{\nabla H_{0,b}(z^\star)^\perp}{|\nabla H_{0,a}(z^\star)|})| \leq 2\max_{\xi\in\partial B} |\Psi_{0,a} - \Psi_{0,b}|.$$

Using the explicit expression of $\Psi_{0,a}(\xi)$ given in (4.12), we find

$$\left|\frac{\nabla H_{0,a}(z^\star)^\perp}{|\nabla H_{0,a}(z^\star)|} - \frac{\nabla H_{0,b}(z^\star)^\perp}{|\nabla H_{0,b}(z^\star)|}\right| \leq C\max_{\xi\in\partial B} |\Psi_{0,a} - \Psi_{0,b}|.$$

Since $\varepsilon_{s,a}(z^\star)$ is lower bounded, combining the estimate above and (4.17), we obtain the desired result. □

Now, by combining the results of Theorem 4.1, Corollary 4.1, and Theorem 4.2, we have the main stability estimate in Theorem 1.1.

**5. Appendix.** In this section we derive the asymptotic expansion of the gradient of the Green function $\nabla_x G(z^\star + \alpha\xi, z^\star + \alpha\xi')$ when $\alpha$ tends to zero.

THEOREM 5.1. Let $G(x,y)$ be the Green function solution to the system (2.13)-(2.14). Then, the following asymptotic expansion holds:

$$\alpha\partial_{x_k} G(z^\star + \alpha\xi, z^\star + \alpha\xi') = \varepsilon_s(z^\star)\partial_{\xi_k}\Phi_0(\xi,\xi') + \frac{1}{4\pi}\partial_{x_k}\varepsilon_s(z^\star)\alpha\ln(\alpha) + \alpha\Phi_1(\xi,\xi') + o(\alpha)$$

for all $\xi, \xi' \in B$ satisfying $\xi \neq \xi'$, and $o(\alpha)$ is uniform in $\xi, \xi' \in B$.

$\Phi_0(\xi,\xi') = \frac{1}{2\pi}\ln(|\xi - \xi'|)$ is the Green function of the Laplacian in the whole space, and $\Phi_1(\xi,\xi')$ has a logarithmic singularity on the diagonal $\xi = \xi'$, that is, $|\Phi_1(\xi,\xi')| \leq C|\Phi_0(\xi,\xi')|$ for all $\xi, \xi' \in B$, with $C > 0$ a constant that only depends on $\varepsilon_s(x)$.

*Proof.* We first use the Liouville transformation and substitute the Green function $G(x,y)$ by

$$\underline{G}(x,y) = \frac{1}{\varepsilon_s^{\frac{1}{2}}(x)\varepsilon_s^{\frac{1}{2}}(y)} G(x,y)$$

in the system (2.13)-(2.14), to obtain

$$\Delta\underline{G}(x,y) + V(x)\underline{G}(x,y) \quad = \delta_y(x) \qquad \text{in} \quad \mathbb{R}^2 \tag{5.1}$$

with the Sommerfeld radiation condition as $|x| \to +\infty$:

$$\frac{\partial\underline{G}}{\partial|x|} - i\omega\sqrt{\varepsilon_0\mu_0}\underline{G} \quad = \quad O(\frac{1}{\sqrt{|x|}}) \tag{5.2}$$

and where

$$V(x) \quad := \quad \omega^2\mu_0\varepsilon_s(x) - \frac{\Delta\varepsilon_s^{\frac{1}{2}}(x)}{\varepsilon_s^{\frac{1}{2}}(x)}. \tag{5.3}$$

For simplicity, we assume that $V(z^\star) \neq 0$. If it is not the case the proof can be slightly modified.

Let $\underline{G}_0(x,y)$ be the Green function of the Helmholtz equation in the free space, solution to the system

$$\Delta \underline{G}_0(x,y) + V(y)\underline{G}_0(x,y) \quad = \delta_y(x) \qquad \text{in} \quad \mathbb{R}^2 \tag{5.4}$$

with the Sommerfeld radiation condition as $|x| \to +\infty$:

$$\frac{\partial \underline{G}_0}{\partial |x|} - i\sqrt{V(y)}\underline{G}_0 \quad = \quad O(\frac{1}{\sqrt{|x|}}). \tag{5.5}$$

The function $\underline{G}_0(x,y)$ is given by

$$\underline{G}_0(x,y) = -\frac{i}{4}H_0^{(1)}(\sqrt{V(y)}|x-y|) \qquad \text{for } x \neq y,$$

where $H_0^{(1)}(t)$ is the Hankel function of the first kind of order zero.

Now, we shall derive the asymtotic expansion of $\partial_{x_k}\underline{G}(x,y)$ as $x$ tends to $y$.

Let

$$\mathcal{G}(\xi,\xi') := \underline{G}(x,y) - \underline{G}_0(x,y).$$

It satisfies the Helmholtz equation

$$\Delta \mathcal{G}(x,y) + V(x)\mathcal{G}(x,y) \quad = -(V(x)-V(y))\underline{G}_0(x,y) \qquad \text{in} \quad B_R(z^\star) \tag{5.6}$$

with the boundary condition

$$\mathcal{G}(x,y) = \underline{G}(x,y) - \underline{G}_0(x,y) \quad \text{on} \quad \partial B_R(z^\star). \tag{5.7}$$

Further, we fix $R > 1$ such that the system (5.6)-(5.7) has a unique solution. Since the $H_0^{(1)}(t)$ has a logarithmic singularity as $t$ tends to zero, the right hand side belongs to $C^{0,\iota}(\overline{B_R(z^\star)})$ for any $\iota \in [0,1)$, uniformly in $y \in B_1(z^\star)$ (see, for instance, Proposition 4.1 in [20]).

Considering the fact that $\underline{G}(x,y) - \underline{G}_0(x,y) \in C^\infty(\partial B_R(z^\star) \times B_1(z^\star))$, we deduce from elliptic regularity that $\mathcal{G}(x,y) \in C^{2,\iota}(\overline{B_R(z^\star)})$ uniformly in $y \in B_1(z^\star)$ [41]. In addition, due to the explicit expression of the right hand side in equation (5.6), one can prove easily that $\partial_{x_k}\mathcal{G}(z^\star + \alpha\xi, z^\star + \alpha\xi')$ has a finite continuous limit when $\alpha$ tends to zero, denoted by $\Phi_{11}(\xi,\xi')$.

From known asymptotic expansions of Hankel functions, we have [1]

$$\partial_{x_k}\underline{G}_0(z^\star + \alpha\xi, z^\star + \alpha\xi') = \frac{1}{\alpha}\partial_{x_k}\Phi_0(\xi,\xi') + \alpha\ln(\alpha)|\xi - \xi'| + O(\alpha),$$

where $O(\alpha)$ is uniform in $\xi, \xi' \in B$.

Consequently,

$$\alpha\partial_{x_k}\underline{G}(z^\star + \alpha\xi, z^\star + \alpha\xi') = \partial_{x_k}\Phi_0(\xi,\xi') + \alpha\Phi_{11}(\xi,\xi') + o(\alpha),$$

which combined with the regularity of $\varepsilon_s(x)$ achieves the proof of the theorem. □

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