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Double-negative acoustic metamaterials*

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Abstract

The aim of this paper is to provide a mathematical theory for understanding the mechanism behind the double-negative refractive index phenomenon in bubbly fluids. The design of double-negative metamaterials generally requires the use of two different kinds of subwavelength resonators, which may limit the applicability of double-negative metamaterials. Herein we rely on media that consists of only a single type of resonant element, and show how to turn the acoustic metamaterial with a single negative effective property obtained in [H. Ammari and H. Zhang, Effective medium theory for acoustic waves in bubbly fluids near Minnaert resonant frequency. SIAM J. Math. Anal., 49 (2017), 3252–3276.] into a negative refractive index metamaterial, which refracts waves negatively, hence acting as a superlens. Using bubble dimers made of two identical bubbles, it is proved that both the effective mass density and the bulk modulus of the bubbly fluid can be negative near the anti-resonance of the two hybridized Minnaert resonances for a single constituent bubble dimer. A rigorous justification of the Minnaert resonance hybridization, in the case of a bubble dimer in a homogeneous medium, is established. The acoustic properties of a single bubble dimer are analyzed. Asymptotic formulas for the two hybridized Minnaert resonances are derived. Moreover, it is proved that the bubble dimer can be approximated by a point scatterer with monopole and dipole modes. For an appropriate volume fraction of bubble dimers with certain conditions on their configuration, a double-negative effective medium when the frequency is near the anti-resonance of the hybridized Minnaert resonances can be obtained.

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1 Introduction

Metamaterials are man-made composite media structured on a scale much smaller than a wavelength. They raise the possibility of an unprecedented level of control when it comes to engineering the propagation of waves. A particularly interesting capability is the prospect of focusing or imaging beyond the diffraction limit. Metamaterials can manipulate and control sound waves in ways that are not possible in conventional materials. Their study has also drawn increasing interest in recent times due to their potential application in cloaking [21, 19, 25].

Metamaterials can be assembled into structures (typically periodic but not necessarily so) that are similar to continuous materials, yet have unusual wave properties that differ substantially from those of conventional media. Subwavelength resonators are the building blocks of metamaterials. Because of the subwavelength scale of the resonators, it is possible to describe the marcoscopic behavior of a metamaterial using homogenization theory, and this results in an effective medium having negative or high contrast parameters. Metamaterials with negative or high contrast refractive indices offer new possibilities for imaging and for the control of waves at deep subwavelength scales [16].

In acoustics, it is known that air bubbles are subwavelength resonators [27]. Due to the high contrast between the air density inside and outside an air bubble in a fluid, a quasi-static acoustic resonance known as the Minnaert resonance occurs [2]. At or near this resonant frequency, the size of a bubble can be up to three orders of magnitude smaller than the wavelength of the incident wave, and the bubble behaves as a strong monopole scatterer of sound. The Minnaert resonance phenomenon makes air bubbles good candidates for acoustic subwavelength resonators. They have the potential to serve as the basic building blocks for acoustic metamaterials, which include bubbly fluids [23, 22, 24]. This motivated our series of bubble studies [2, 4, 5, 6, 14]. We refer to [2] for a rigorous mathematical treatment of Minnaert resonance and the derivation of the monopole approximation in the case of a single, arbitrary shaped bubble in a homogeneous medium.

As shown in [14], around the Minnaert resonant frequency, an effective medium theory can be derived in the dilute regime. Furthermore, above the Minnaert resonant frequency, the real part of the effective bulk modulus is negative, and consequently the bubbly fluid behaves as a diffusive medium for the acoustic waves. Meanwhile, below the Minnaert resonant frequency, with an appropriate bubble volume fraction, a high contrast effective medium can be obtained, making the sub-wavelength focusing or super-focusing of waves achievable [15]. These properties show that the bubbly fluid functions like an acoustic metamaterial, and indicate that a sub-wavelength bandgap opening occurs at the Minneaert resonant frequency [23]. We remark that such behavior is rather analogous to the coupling of electromagnetic waves with plasmonic nanoparticles, which results in effective negative or high contrast dielectric constants for frequencies near the plasmonic resonance frequencies [1, 12, 13].

In [4], the opening of a sub-wavelength phononic bandgap is demonstrated by considering a periodic arrangement of bubbles and exploiting their Minnaert resonance. It is shown that there exists a subwavelength band gap in such a bubbly crystal. This subwavelength band gap is mainly due to the cell resonance of the bubbles in the quasi-static regime and is quite different from the usual band gaps in photonic/phononic crystals, where the gap opens at a wavelength which is comparable to the period of the structure [18, 10, 9]. In [11], the homogenization theory of the bubbly crystal near the frequency where the band gap opens is further investigated. It is shown that the band gap opens at the corner (edge in two dimensions) of the Brillouin zone. Moreover, explicit formulas for the Bloch eigenfunctions are derived. This makes both the homogenization theory and the justification of the superfocusing phenomenon in the non-dilute case possible. We also refer to [5] for the related work on bubbly metasurfaces in which a homogenization theory is developed for a thin layer of periodically arranged bubbles mounted on a perfectly reflecting surface.

In this paper, we aim to understand the mechanism behind the double-negative refractive index phenomenon in bubbly fluids. The design of double-negative metamaterials generally requires the use of two different kinds of building blocks or specific subwavelength resonators presenting multiple overlapping resonances. Such a requirement limits the applicability of double-negative metamaterials. Herein we rely on media that consists of only a single type of resonant element, and show how to turn the acoustic metamaterial with a single negative effective property obtained in [14] into a negative refractive index metamaterial, which refracts waves negatively, hence acting as a superlens [31, 28, 26].

Our main result is to prove that, using bubble dimers, the effective mass density and bulk modulus of the bubbly fluid can both be negative over a non empty range of frequencies. A bubble dimer is a system consists of two identical separated bubbles. It features two slightly different subwavelength resonances, called the hybridized Minnaert resonances. We establish a rigorous mathematical justification of the Minnaert resonance hybridization in the case of a bubble dimer in a homogeneous medium. We analyze the acoustic properties of the bubble dimer, derive asymptotic formulas for the two resonances, and prove that the bubble dimer can be approximated as a point scatterer with monopole and dipole modes. The hybridized Minnaert resonances are fundamentally different modes. The first mode is, as in the case of a single bubble, a monopole mode, while the second mode is a dipole mode. The resonance associated with the dipole mode is usually referred to as the anti-resonance. For an appropriate volume fraction, when the excitation frequency is close to the anti-resonance, we obtain a double-negative effective mass density and bulk modulus for bubbly media consisting of a large number of bubble dimers with certain conditions on their distribution. The dipole modes in the background medium contribute to the effective mass density while the monopole modes contribute to the effective bulk modulus.

The paper is organized as follows. In Section 2 we introduce some preliminaries on layer potentials. In Section 3 we describe the hybridization phenomenon for a bubble dimer, prove that two subwavelength resonances occur, and compute their asymptotic expansions in terms of the mass density contrast. In Section 4 we prove that the bubble dimer can be approximated as the sum of a monopole source and a dipole source. In

Section 5 we derive a double-negative effective medium theory for bubbly media. Finally, in Section 6 we compute effective mass density and bulk modulus dispersion curves near the hybridized Minnaert resonances to illustrate the double-negative property of bubbly media.

2 Preliminaries

2.1 Layer potentials

For a given bounded domain D in $\mathbb{R}^3$, with Lipschitz boundary ∂D, the single layer potential of the density function $\varphi \in L^2(\partial D)$ is defined by

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

where $G(x,k)$ is the fundamental solution to $\triangle + k^2$, i.e.,

$$G(x,k) = -\frac{1}{4\pi|x|}\exp(ik|x|). \tag{2.1}$$

The following jump relation holds:

$$\frac{\partial}{\partial \nu}\mathcal{S}_D^k[\varphi]\bigg|_{\pm}(x) = \left(\pm\frac{1}{2}I + \mathcal{K}_D^{k,*}\right)[\varphi](x), \quad x \in \partial D, \tag{2.2}$$

where the Neumann-Poincaré operator $\mathcal{K}_D^{k,*}$ is defined by

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

Here $\frac{\partial}{\partial \nu}$ denotes the normal derivative on ∂D, and the subscripts $+$ and $-$ indicate the limits from outside and inside D, respectively.

We make use of low frequency asymptotic expansions of the layer potentials. For a small parameter ϵ, $G(x,\epsilon k)$ can be expanded as

$$G(x,\epsilon k) = -\frac{1}{4\pi}\sum_{n=0}^{\infty}\frac{(i\epsilon k)^n}{n!}|x|^{n-1}, \tag{2.3}$$

and it follows that

$$\mathcal{S}_D^{\epsilon k}[\phi] = \sum_{n=0}^{\infty}(\epsilon k)^n \mathcal{S}_D^n[\phi], \quad \mathcal{K}_D^{\epsilon k}[\phi] = \sum_{n=0}^{\infty}(\epsilon k)^n \mathcal{K}_D^n[\phi], \tag{2.4}$$

where

$$\mathcal{S}_D^n[\phi](x) := -\frac{i^n}{4\pi n!}\int_{\partial D}|x-y|^{n-1}\phi(y)d\sigma(y), \tag{2.5}$$

$$\mathcal{K}_D^n[\phi](x) := -\frac{i^n(n-1)}{4\pi n!}\int_{\partial D}\langle x-y,\nu_x\rangle|x-y|^{n-3}\phi(y)d\sigma(y). \tag{2.6}$$

It is known that $\mathcal{S}_D^0 : L^2(\partial D) \to H^1(\partial D)$ is invertible and that its inverse is bounded.

Throughout the paper we assume that D has two connected components D_1 and D_2, and we use the following notation:

$$\int_{\partial D_1 - \partial D_2} f(y) d\sigma(y) = \int_{\partial D_1} f(y) d\sigma(y) - \int_{\partial D_2} f(y) d\sigma(y), \tag{2.7}$$

where f is a function defined on $\partial D = \partial D_1 \cup \partial D_2$.

Finally, we present some useful formulas which are frequently used in the sequel.

Lemma 2.1. *The following identities hold for any $\phi \in L^2(\partial D)$: for j = 1, 2,*

(i) $\int_{\partial D_j} \left(\frac{1}{2}I - \mathcal{K}_D^{0,*}\right) \phi(y) d\sigma(y) = 0$;

(ii) $\int_{\partial D_j} \left(\frac{1}{2}I + \mathcal{K}_D^{0,*}\right) \phi(y) d\sigma(y) = \int_{\partial D_j} \phi(y) d\sigma(y)$;

(iii) $\int_{\partial D_j} \mathcal{K}_D^2[\phi] = -\int_{D_j} \mathcal{S}_D^0[\phi]$;

(iv) $\int_{\partial D_j} \mathcal{K}_D^3[\phi] = i|D_j|/(4\pi) \int_{\partial D} \phi$.

Proof. (i) follows from the jump relation $(-\frac{1}{2}I + \mathcal{K}_D^{0,*})[\phi] = \partial \mathcal{S}_D^0[\phi]/\partial\nu|_{\partial D}^-$ and the fact that $\mathcal{S}_D^0[\phi]$ is harmonic in D. (ii) immediately follows from (i). For (iii), we have

$$\int_{\partial D_j} \mathcal{K}_D^2 \phi = \frac{1}{8\pi} \int_{\partial D_j} \frac{\partial}{\partial \nu_x} \int_{\partial D} |x-y| \phi(y) = \frac{1}{8\pi} \int_{D_j} \int_{\partial D} \frac{2}{|x-y|} \phi(y) = -\int_{D_j} \mathcal{S}_D^0[\phi].$$

Finally, it holds that

$$\int_{\partial D_j} \mathcal{K}_D^3[\phi] = \frac{i}{12\pi} \int_{\partial D_j} \int_{\partial D} \langle\, x-y, \nu_x \rangle \phi(y) = \frac{i|D_j|}{4\pi} \int_{\partial D} \phi,$$

which proves (iv). $\square$

2.2 Capacitance coefficients

Let ψ_1, $\psi_2 \in L^2(\partial D)$ be given by

$$\mathcal{S}_D^0[\psi_1] = \begin{cases} 1 & \text{on } \partial D_1, \\ 0 & \text{on } \partial D_2, \end{cases} \qquad \mathcal{S}_D^0[\psi_2] = \begin{cases} 0 & \text{on } \partial D_1, \\ 1 & \text{on } \partial D_2. \end{cases} \tag{2.8}$$

Then, using (2.2), it is easy to check that

$$\ker \left(-\frac{1}{2}I + \mathcal{K}_D^{0,*} \right) = \text{span } \{\psi_1, \psi_2\}. \tag{2.9}$$

We define the capacitance coefficients matrix $C = (C_{ij})$ by

$$C_{ij} := -\int_{\partial D_j} \psi_i, \quad i,j = 1,2.$$

We remark that the matrix C is positive definite and symmetric.

Suppose D_1 and D_2 are identical balls of radius r_0 separated by a distance d_0 such that the distance between the centers of the balls is $d_0 + 2r_0$. Then $C_{11} = C_{22},\ C_{12} = C_{21},\ C_{11} > 0$, and $C_{12} < 0$. Explicit formulas for the capacitance coefficients for two balls can be obtained using bispherical coordinates [20]. We have the following result:

$$
\begin{aligned}
C_{11} &= 8\pi\alpha \sum_{n=0}^{\infty} \frac{e^{(2n+1)T}}{e^{2(2n+1)T} - 1}, \\
C_{12} &= -8\pi\alpha \sum_{n=0}^{\infty} \frac{1}{e^{2(2n+1)T} - 1}, \\
C_{21} &= C_{12}, \quad C_{22} = C_{11},
\end{aligned}
$$

where

$$
\alpha = \sqrt{d_0(r_0 + d_0/4)}, \quad T = \sinh^{-1}(r_0/a).
$$

Lemma 2.2. *Suppose that D_1 and D_2 are two identical balls. Then the following identities hold for any $\phi \in L^2(\partial D)$:*

(i) $\int_{\partial D_j} \mathcal{K}_D^3[\psi_1 - \psi_2] d\sigma = 0, \quad j = 1, 2;$

(ii) $\int_{\partial D_1 - \partial D_2} (\mathcal{S}_D^0)^{-1} \mathcal{S}_D^1[\phi] d\sigma = 0;$

(iii) $\int_{\partial D_1 - \partial D_2} (\mathcal{S}_D^0)^{-1}[y_i] d\sigma(y) = \int_{\partial D} y_i(\psi_1 - \psi_2) d\sigma(y).$

Proof. (i) and (ii) follow from Lemma 2.1 (iv), the definition of $\mathcal{S}_D^1$ and the symmetry of $D_1 \cup D_2$. For (iii), by letting $\phi_i(x) := (\mathcal{S}_D^0)^{-1}[y_i](x)$ and using the jump relations (2.2) and the fact that $\mathcal{S}_D^0[\phi]$ is harmonic in D, we can check that

$$
\begin{aligned}
\int_{\partial D_1 - \partial D_2} (\mathcal{S}_D^0)^{-1}[y_i] d\sigma(y) &= \int_{\partial D_1 - \partial D_2} 1 \cdot \left(\frac{\partial \mathcal{S}_D^0[\phi_i]}{\partial \nu}\Big|_+ - \frac{\partial \mathcal{S}_D^0[\phi_i]}{\partial \nu}\Big|_- \right) \\
&= \int_{\partial D_1 - \partial D_2} 1 \cdot \frac{\partial \mathcal{S}_D^0[\phi_i]}{\partial \nu}\Big|_+ = \int_{\partial D} \mathcal{S}_D[\psi_1 - \psi_2] \cdot \frac{\partial \mathcal{S}_D^0[\phi_i]}{\partial \nu}\Big|_+.
\end{aligned}
$$

Then, since and $\mathcal{S}_D^0[\phi_i]|_{\partial D} = y_i$, the Green's identity yields (iii). □

3 Resonance for a dimer consisting of two identical bubbles

In this section, we consider the quasi-static resonances of a bubble dimer. Throughout, we denote by D the normalized bubble dimer which consists of two identical balls D_1 and D_2, both with volume one. We assume that the two balls D_1 and D_2 are symmetric with respect to the origin. Moreover, D_1 and D_2 are aligned with respect to the x_1-axis. For a general dimer featuring two identical balls, we call the unit direction vector along which the two balls are aligned the orientation of the dimer. Let (κ_b, ρ_b) and (κ, ρ) be the bulk modulus and density of air and water, respectively. Let u^{in} be the incident wave which we assume to be a plane wave for simplicity:

$$u^{in}(x) = e^{i\omega\sqrt{\rho/\kappa}x\cdot\theta},$$

where θ is a unit vector in $\mathbb{R}^3$. Then the acoustic wave propagation can be modeled as

$$\begin{cases} \nabla\cdot\left(\frac{1}{\rho_b}\chi_D + \frac{1}{\rho}\chi(\mathbb{R}^3\setminus\overline{D})\right)\nabla u + \omega^2\left(\frac{1}{\kappa_b}\chi_D + \frac{1}{\kappa}\chi(\mathbb{R}^3\setminus\overline{D})\right)u = 0 \quad \text{in } \mathbb{R}^3, \\ u - u^{in} \text{ satisfies the Sommerfeld radiation condition at infinity.} \end{cases} \tag{3.1}$$

Recall that the Sommerfeld radiation condition at infinity can be expressed by

$$\left|\frac{\partial}{\partial|x|}(u-u^{in})(x) - \omega\sqrt{\rho/\kappa}(u-u^{in})(x)\right| = O(\frac{1}{|x|^2}),$$

uniformly in $x/|x|$ as $|x|\to+\infty$.

Let

$$v = \sqrt{\frac{\kappa}{\rho}}, \quad v_b = \sqrt{\frac{\kappa_b}{\rho_b}}, \quad k = \omega\sqrt{\frac{\rho}{\kappa}}, \quad k_b = \omega\sqrt{\frac{\rho_b}{\kappa_b}}. \tag{3.2}$$

We assume that

$$\delta := \frac{\rho_b}{\rho} \ll 1 \tag{3.3}$$

and

$$v, v_b = O(1). \tag{3.4}$$

From, for example, [8], we know that the solution to (3.1) can be represented using the single layer potentials $\mathcal{S}_D^{k_b}$ and $\mathcal{S}_D^{k}$ as follows:

$$u(x) = \begin{cases} u^{in}(x) + \mathcal{S}_D^k[\psi](x), & x\in\mathbb{R}^3\setminus\bar{D}, \\ \mathcal{S}_D^{k_b}[\phi](x), & x\in D, \end{cases} \tag{3.5}$$

where the pair $(\phi,\psi)\in L^2(\partial D)\times L^2(\partial D)$ is a solution to

$$\begin{cases} \mathcal{S}_D^{k_b}[\phi] - \mathcal{S}_D^k[\psi] = u^{in} \\ \dfrac{1}{\rho_b}\left(-\dfrac{1}{2}I + \mathcal{K}_D^{k_b,*}\right)[\phi] - \dfrac{1}{\rho}\left(\dfrac{1}{2}I + \mathcal{K}_D^{k,*}\right)[\psi] = \dfrac{1}{\rho}\dfrac{\partial u^{in}}{\partial\nu} \end{cases} \quad \text{on } \partial D. \tag{3.6}$$

We denote by

$$\mathcal{A}_\delta^\omega := \begin{bmatrix} \mathcal{S}_D^{k_b} & -\mathcal{S}_D^k \\ \left(-\frac{1}{2}I + \mathcal{K}_D^{k_b,*}\right) & -\delta\left(\frac{1}{2}I + \mathcal{K}_D^{k,*}\right) \end{bmatrix}. \tag{3.7}$$

Then (3.6) can be written as

$$\mathcal{A}_\delta^\omega \begin{pmatrix} \phi \\ \psi \end{pmatrix} = \begin{pmatrix} u^{in} \\ \delta\frac{\partial u^{in}}{\partial\nu} \end{pmatrix}.$$

It is well-known that the above integral equation has a unique solution for all real frequencies ω.

The resonance of the bubble dimer D in the scattering problem (3.1) can be defined as all the complex numbers ω with negative imaginary part such that there exists a nontrivial solution to the following equation:

$$\mathcal{A}_\delta^\omega \begin{pmatrix} \phi \\ \psi \end{pmatrix} = 0. \tag{3.8}$$

These can be viewed as the characteristic values of the operator-valued analytic function $\mathcal{A}_\delta^\omega$ (with respect to ω); see [9].

It can be shown that $\omega = 0$ is a characteristic value of $\mathcal{A}_0^\omega$ when $\delta = 0$. Then Gohberg-Sigal theory [9] tells us that there exists a characteristic value $\omega_0 = \omega_0(\delta)$ such that $\omega_0(0) = 0$ and ω_0 depends on δ continuously. We call this characteristic value the quasi-static resonance (or Minnaert resonance) [2]. We now present the main result on the quasi-static resonances of the bubble dimer D.

Theorem 3.1. *There are two quasi-static resonances with positive real part for the bubble dimer D. Moreover, they have the following asymptotic expansions as δ, defined by (3.3), goes to zero:*

$$\omega_1 = \sqrt{\delta v_b^2 (C_{11} + C_{12})} - i\tau_1 \delta + O(\delta^{3/2}), \tag{3.9}$$

$$\omega_2 = \sqrt{\delta v_b^2 (C_{11} - C_{12})} + \delta^{3/2}\hat{\eta}_1 + i\delta^2 \hat{\eta}_2 + O(\delta^{5/2}), \tag{3.10}$$

where

$$\tau_1 = \frac{v_b^2}{4\pi v}(C_{11} + C_{12})^2,$$

and $\hat{\eta}_1$ and $\hat{\eta}_2$ are real numbers which are determined by D, v, and v_b. The two resonances ω_1 and ω_2 are called the hybridized resonances of the bubble dimmer D. The resonance ω_2 is referred to as the anti-resonance.

Proof. Step 1. Suppose that $(\phi,\ \psi)$ is a nontrivial solution to (3.8) for some small $\omega = \omega(\delta)$.

Using the asymptotic expansions (2.4) of the single layer potential and the Neumann-Poincaré operator, together with the fact that $k = O(\omega)$, we have

$$\left\{ \begin{array}{l} \mathcal{S}_D^0[\phi - \psi] + k_b \mathcal{S}_D^1[\phi] - k\mathcal{S}_D^1[\psi] = O(\omega^2), \\ \left(-\frac{1}{2}I + \mathcal{K}_D^{0,*} + k_b^2 \mathcal{K}_D^2 + k_b^3 \mathcal{K}_D^3\right)[\phi] - \delta\left(\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)[\psi] = O(\omega^4 + \delta\omega^2). \end{array} \right. \tag{3.11}$$

From the first equation of (3.11) and the definition of $\mathcal{S}_D^1$, it holds that

$$\psi = \phi - \frac{1}{4\pi i}(\mathcal{S}_D^0)^{-1}\left(k_b \int_{\partial D} \phi - k \int_{\partial D} \psi\right) + O(\omega^2).$$

Then, from the fact that $\phi - \psi = O(\omega)$ and the definition of ψ_j, we obtain

$$\psi = \phi + \frac{(k_b - k)}{4\pi i}(\psi_1 + \psi_2) \int_{\partial D} \phi + O(\omega^2). \tag{3.12}$$

Plugging (3.12) into the second equation of (3.11), we get

$$\begin{aligned} \left(-\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)[\phi] + \left(k_b^2 \mathcal{K}_D^2 + k_b^3 \mathcal{K}_D^3 - \delta\left(\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)\right)[\phi] \\ - \frac{\delta(k_b - k)}{4\pi i}(\psi_1 + \psi_2) \int_{\partial D} \phi = O(\omega^4 + \delta\omega^2), \end{aligned} \tag{3.13}$$

where we have used the identity

$$\begin{aligned} \psi_j &= \frac{\partial \mathcal{S}_D}{\partial \nu}\bigg|_{\partial D}^{+}[\psi_j] - \frac{\partial \mathcal{S}_D}{\partial \nu}\bigg|_{\partial D}^{-}[\psi_j] \\ &= \frac{\partial \mathcal{S}_D}{\partial \nu}\bigg|_{\partial D}^{+}[\psi_j] - 0 = \left(\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)[\psi_j]. \end{aligned}$$

In view of (2.9), a nontrivial solution ϕ to (3.13) can be written as

$$\phi = a\psi_1 + b\psi_2 + O(\omega^2 + \delta), \tag{3.14}$$

for some nontrivial constants $a,\ b$ with $|a| + |b| = O(1)$.

Step 2. Recall that $|D_1| = |D_2| = 1$, $C_{11} = C_{22}$, and $C_{12} = C_{21} < 0$. By integrating (3.13) over $\partial D_j,\ j = 1, 2$, and then using Lemma 2.1, we have, up to an error of order $O(\omega^4 + \delta\omega^2)$,

$$\begin{cases} -\dfrac{\omega^2}{v_b^2}a - \dfrac{i\omega^3}{4\pi v_b^3}C(a,b) + \delta(aC_{11} + bC_{12}) + \dfrac{i(v_b^{-1} - v^{-1})\delta\omega}{4\pi}(C_{11} + C_{12})C(a,b) = 0, \\ -\dfrac{\omega^2}{v_b^2}b - \dfrac{i\omega^3}{4\pi v_b^3}C(a,b) + \delta(aC_{12} + bC_{11}) + \dfrac{i(v_b^{-1} - v^{-1})\delta\omega}{4\pi}(C_{12} + C_{11})C(a,b) = 0, \end{cases} \tag{3.15}$$

where

$$C(a,b) := a(C_{11} + C_{12}) + b(C_{21} + C_{22}).$$

Then it follows that for $\delta \ll 1$, the characteristic values of $\mathcal{A}_\delta^\omega$ are given by

$$\omega_* = \omega_0 + O(\delta^{3/2}), \tag{3.16}$$

for some $\omega_0 = O(\sqrt{\delta})$. Solving (3.15) for ω, we find two characteristic values with positive real parts:

$$\omega_1 = \sqrt{\delta v_b^2 (C_{11} + C_{12})} + O(\delta), \quad \omega_2 = \sqrt{\delta v_b^2 (C_{11} - C_{12})} + O(\delta),$$

where the corresponding (a,b)'s are given by

$$(a,b) = (1,1),\ (1,-1), \tag{3.17}$$

up to an error of order $O(\delta)$. Plugging these values for a, b and ω_j into (3.15) and solving for the $O(\delta)$ term in ω_j, we obtain

$$\omega_1 = \sqrt{\delta v_b^2(C_{11}+C_{12})} - i\tau_1\delta + O(\delta^{3/2}), \quad \omega_2 = \sqrt{\delta v_b^2(C_{11}-C_{12})} + 0\cdot\delta + O(\delta^{3/2}).$$

The proof of the estimate (3.9) for ω_1 is concluded. We refine the estimate for ω_2 in the next step.

Step 3. Note that $\Im\omega_2$ is of order $\delta^{3/2}$. We perform a further calculation to find an explicit formula for $\Im\omega_2$. Since $(a,b) = (1,-1) + O(\delta)$, we write

$$\phi = \psi_1 - \psi_2 + \delta\phi_1 + \delta^{3/2}\phi_2 + \delta^2\phi_3 + \delta^{5/2}\phi_4 + O(\delta^3),$$

and

$$\omega_2 = \delta^{1/2}\eta_0 + \delta^{3/2}\eta_1 + \delta^2\eta_2 + O(\delta^{5/2}),$$

where $\eta_0 := \sqrt{v_b^2(C_{11}-C_{12})}$. From the symmetry of D, we can normalize ϕ so that

$$\phi_j \perp \operatorname{span}\{\psi_1,\psi_2\} = \ker\left(-\frac{1}{2}I + \mathcal{K}_B^{0,*}\right), \quad \text{for } j = 1,2,3,4, \quad \text{in } L^2(\partial D).$$

Step 4. By using (2.4), we have

$$\begin{cases} \mathcal{S}_D^0[\phi-\psi] + k_b\mathcal{S}_D^1[\phi] - k\mathcal{S}_D^1[\psi] + k_b^2\mathcal{S}_D^2[\phi] - k^2\mathcal{S}_D^2[\psi] + k_b^3\mathcal{S}_D^3[\phi] - k^3\mathcal{S}_D^3[\psi] = O(\delta^2), \\ \left(-\frac{1}{2}I + \mathcal{K}_D^{0,*} + k_b^2\mathcal{K}_D^2 + k_b^3\mathcal{K}_D^3 + k_b^4\mathcal{K}_D^4 + k_b^5\mathcal{K}_D^5\right)[\phi] - \delta\left(\frac{1}{2}I + \mathcal{K}_D^{0,*} + k^2\mathcal{K}_D^2 + k^3\mathcal{K}_D^3\right)[\psi] = O(\delta^3). \end{cases} \tag{3.18}$$

Hence, we get

$$\begin{aligned} \psi &= (\mathcal{S}_D^0 + k\mathcal{S}_D^1 + k^2\mathcal{S}_D^2 + k^3\mathcal{S}_D^3)^{-1}(\mathcal{S}_D^0 + k_b\mathcal{S}_D^1 + k_b^2\mathcal{S}_D^2 + k_b^3\mathcal{S}_D^3)[\phi] + O(\delta^2) \\ &= \phi + (\mathcal{S}_D^0 + k\mathcal{S}_D^1 + k^2\mathcal{S}_D^2)^{-1}((k_b-k)\mathcal{S}_D^1 + (k_b^2-k^2)\mathcal{S}_D^2 + (k_b^3-k^3)\mathcal{S}_D^3)[\phi] + O(\delta^2) \\ &= \phi + (\mathcal{S}_D^0 + k\mathcal{S}_D^1)^{-1}((k_b^2-k^2)\mathcal{S}_D^2 + (k_b^3-k^3)\mathcal{S}_D^3)[\psi_1-\psi_2] + \delta(k_b-k)(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^1[\phi_1] + O(\delta^2) \\ &= \phi + (k_b^2-k^2)(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^2[\psi_1-\psi_2] + (k_b^3-k^3)(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^3[\psi_1-\psi_2] \\ &\quad - k(k_b^2-k^2)(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^1(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^2[\psi_1-\psi_2] + \delta(k_b-k)(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^1[\phi_1] + O(\delta^2). \end{aligned}$$

Plugging this into the second equation of (3.18) and equating terms of the same order of δ, $\delta^{3/2}$, δ^2, $\delta^{5/2}$, we arrive at

- $O(\delta)$-terms:

$$\left(-\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)[\phi_1] + \eta_0^2 v_b^{-2}\mathcal{K}_D^2[\psi_1-\psi_2] - (\psi_1-\psi_2) = 0, \tag{3.19}$$

  whence ϕ_1 is uniquely determined.

- $O(\delta^{3/2})$-terms:

$$\left(-\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)[\phi_2] + \eta_0^3 v_b^{-3}\mathcal{K}_D^3[\psi_1 - \psi_2] = 0, \tag{3.20}$$

  whence ϕ_2 is uniquely determined.

- $O(\delta^2)$-terms:

$$\begin{aligned}&\left(-\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)[\phi_3] + 2\eta_0\eta_1 v_b^{-2}\mathcal{K}_D^2[\psi_1 - \psi_2] + \eta_0^2 v_b^{-2}\mathcal{K}_D^2[\phi_1] + \eta_0^4 v_b^{-4}\mathcal{K}_D^4[\psi_1 - \psi_2]\\ &- \left(\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)[\eta_0^2(v_b^{-2} - v^{-2})(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^2[\psi_1 - \psi_2] + \phi_1] - v^{-2}\eta_0^2\mathcal{K}_D^2[\psi_1 - \psi_2] = 0\end{aligned}$$

- $O(\delta^{5/2})$-terms:

$$\begin{aligned}&\left(-\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)[\phi_4] + 2\eta_0\eta_2 v_b^{-2}\mathcal{K}_D^2[\psi_1 - \psi_2] + \eta_0^2 v_b^{-2}\mathcal{K}_D^2[\phi_2] + \eta_0^3 v_b^{-3}\mathcal{K}_D^3[\phi_1]\\ &+ 3\eta_0^2\eta_1 v_b^{-3}\mathcal{K}_D^3[\psi_1 - \psi_2] + \eta_0^5 v_b^{-5}\mathcal{K}_D^5[\psi_1 - \psi_2]\\ &- \eta_0^3\left(\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)\Big[(v_b^{-3} - v^{-3})(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^3[\psi_1 - \psi_2]\\ &- v^{-1}(v_b^{-2} - v^{-2})(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^1(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^2[\psi_1 - \psi_2]\Big]\\ &- \eta_0^3\left(\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)\left[(v_b^{-1} - v^{-1})(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^1[\phi_1]\right] - \left(\frac{1}{2}I + \mathcal{K}_D^{0,*}\right)[\phi_2] - \eta_0^3 v^{-3}\mathcal{K}_D^3[\psi_1 - \psi_2] = 0.\end{aligned}$$

Step 5. We consider the terms of order $O(\delta^2)$. Integrating over $\partial D_1 - \partial D_2$ and using Lemma 2.2, we get

$$\begin{aligned}-2\eta_0\eta_1 v_b^{-2} + \int_{\partial D_1 - \partial D_2}\left(\eta_0^2 v_b^{-2}\mathcal{K}_D^2[\phi_1] - \phi_1\right) + \eta_0^4 v_b^{-4}\int_{\partial D_1 - \partial D_2}\mathcal{K}_D^4[\psi_1 - \psi_2]\\ -\eta_0^2(v_b^{-2} - v^{-2})\int_{\partial D_1 - \partial D_2}(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^2[\psi_1 - \psi_2] + \eta_0^2 v^{-2} = 0.\end{aligned} \tag{3.21}$$

It follows that η_1 is uniquely determined from the above equation. Moreover, we can check that η_1 is a real number.

Step 6. Finally, we consider the $\delta^{5/2}$ terms. Integrating over $\partial D_1 - \partial D_2$ and using Lemma 2.2 again, we get

$$\begin{aligned}&- 4\eta_0\eta_2 v_b^{-2} + \int_{\partial D_1 - \partial D_2}\eta_0^2 v_b^{-2}\mathcal{K}_D^2[\phi_2] - \phi_2 + \eta_0^3 v_b^{-3}\mathcal{K}_D^3[\phi_1]\\ &+ \int_{\partial D_1 - \partial D_2}\eta_0^5 v_b^{-5}\mathcal{K}_D^5[\psi_1 - \psi_2] - \eta_0^3(v_b^{-3} - v^{-3})(\mathcal{S}_D^0)^{-1}\mathcal{S}_D^3[\psi_1 - \psi_2] = 0.\end{aligned} \tag{3.22}$$

Therefore, η_2 is uniquely determined. We also note that η_2 is a purely imaginary number. Indeed, from (3.19), (3.20), we see that $\Im\phi_1 = 0, \Re\phi_2 = 0$. This combined with the integral representation of $\mathcal{K}_D^2, \mathcal{K}_D^3, \mathcal{K}_D^5$ yields the desired result.

Thus we conclude that the resonance frequency ω_2 has the following asymptotic expansion:

$$\omega_2 = \sqrt{\delta v_b^2 (C_{11} - C_{12})} + \delta^{3/2}\hat{\eta}_1 + i\delta^2\hat{\eta}_2 + O(\delta^{5/2}), \tag{3.23}$$

where $\hat{\eta}_1 := \eta_1$ and $\hat{\eta}_2 := -i\eta_2$ are real numbers given by (3.21) and (3.22), respectively. $\square$

Remark 1. *The above approach is applicable to a general bubble dimer which may consist of two non-identical spherical bubbles.*

4 The point scatterer approximation of bubbles

In this section we prove an approximate formula for the solution u to the scattering problem for the bubble dimer $D = D_1 \cup D_2$.

We need the following lemma which can be proved using a simple symmetry argument.

Lemma 4.1. *We have*

$$\int_{\partial D} y_2(\psi_1 - \psi_2) d\sigma(y) = \int_{\partial D} y_3(\psi_1 - \psi_2) d\sigma(y) = 0,$$

while

$$\int_{\partial D} y_1(\psi_1 - \psi_2) d\sigma(y) = P,$$

for some nonzero real number P.

In the next theorem, we prove that the bubble dimer can be approximated as a point scatterer with monopole and dipole modes.

Theorem 4.2. *For $\omega = O(\delta^{1/2})$ and a given plane wave u^{in}, the solution u to (3.1) can be approximated as $\delta \to 0$ by*

$$u(x) - u^{in}(x) = g^0(\omega) u^{in}(0) G(x,k) + \nabla u^{in}(0) \cdot g^1(\omega) \nabla G(x,k) + O(\delta |x|^{-1}), \tag{4.1}$$

when $|x|$ is sufficiently large, where

$$g^0(\omega) := \frac{C(1,1)}{1 - \omega_1^2/\omega^2}(1 + O(\delta^{1/2})), \tag{4.2}$$

$$g^1(\omega) = (g_{ij}^1(\omega)),\ g_{ij}^1(\omega) := \int_{\partial D} (\mathcal{S}_D^0)^{-1}[x_i](y) y_j + \frac{\delta v_b^2}{|D|(\omega_2^2 - \omega^2)} P^2 \delta_{i,1}\delta_{j,1}. \tag{4.3}$$

Proof. Step 1. Let $(\phi,\ \psi)$ be the solution to (3.6). Using the asymptotic expansions (2.4), we have

$$\begin{cases} \mathcal{S}_D^0[\phi-\psi]+k_b\mathcal{S}_D^1[\phi]-k\mathcal{S}_D^1[\psi]=u^{in}+O(\delta), \\ \left(-\frac{1}{2}I+\mathcal{K}_D^{0,*}+k_b^2\mathcal{K}_D^2+k_b^3\mathcal{K}_D^3\right)[\phi]-\delta\left(\frac{1}{2}I+\mathcal{K}_D^{0,*}\right)[\psi]=\delta\frac{\partial u^{in}}{\partial \nu}+O(\delta^2), \end{cases} \tag{4.4}$$

where the remainders $O(\delta)$ and $O(\delta^2)$ are in the operator norm.

From the first equation of (4.4), and following the same approach used to derive (3.12), we obtain

$$\psi=\phi+\frac{(k_b-k)}{4\pi i}(\psi_1+\psi_2)\int_{\partial D}\phi-(\mathcal{S}_D^0)^{-1}[u^{in}]+O(\delta). \tag{4.5}$$

Plugging (4.5) into the second equation of (4.4), we get

$$\mathcal{C}_\delta^\omega[\phi]=-\delta\left(\frac{1}{2}I+\mathcal{K}_D^{0,*}\right)(\mathcal{S}_D^0)^{-1}[u^{in}]+\delta\frac{\partial u^{in}}{\partial\nu}+O(\delta^2), \tag{4.6}$$

where $\mathcal{C}_\delta^\omega$ is defined by

$$\begin{aligned}\mathcal{C}_\delta^\omega[\phi]:=&\left(-\frac{1}{2}I+\mathcal{K}_D^{0,*}[\phi]\right)+\left(k_b^2\mathcal{K}_D^2+k_b^3\mathcal{K}_D^3-\delta\left(\frac{1}{2}I+\mathcal{K}_D^{0,*}\right)\right)[\phi]\\ &-\frac{\delta(k_b-k)}{4\pi i}(\psi_1+\psi_2)\int_{\partial D}\phi.\end{aligned}$$

Note that $\mathcal{C}_\delta^\omega[\phi]$ is equal to the left-hand side of (3.13).

Step 2. Using the Taylor expansion of u^{in} at the origin, and the fact that $\nabla u^{in}=O(\omega)=O(\delta^{1/2})$ and $\nabla^2u^{in}=O(\omega^2)=O(\delta)$, the right-hand side of (4.6) can be approximated by

$$\begin{aligned}&-\delta\left(\frac{1}{2}I+\mathcal{K}_D^{0,*}\right)(\mathcal{S}_D^0)^{-1}[u^{in}](x)+\delta\frac{\partial u^{in}}{\partial\nu}(x)\\ &=-\delta\left(\frac{1}{2}I+\mathcal{K}_D^{0,*}\right)(\mathcal{S}_D^0)^{-1}[u^{in}(0)+\nabla u^{in}(0)\cdot y](x)+\delta\nabla(u^{in}(0))\cdot\nu(x)+O(\delta^{3/2})\\ &=-\delta u^{in}(0)(\psi_1+\psi_2)-\delta(\mathcal{S}_D^0)^{-1}[\nabla u^{in}(0)\cdot y]+0+O(\delta^{3/2}).\end{aligned}$$

Therefore, (4.6) becomes

$$\mathcal{C}_\delta^\omega[\phi]=-\delta u^{in}(0)(\psi_1+\psi_2)-\delta(\mathcal{S}_D^0)^{-1}[\nabla u^{in}(0)\cdot y]+O(\delta^{3/2}). \tag{4.7}$$

Step 3. In the statement and proof of Theorem 3.1, we have verified that the characteristic values of $\mathcal{C}_\delta^\omega$ are given by (3.9) and (3.10), and that their corresponding singular functions are ϕ_1, ϕ_2, i.e.,

$$\mathcal{C}_\delta^{\omega_1}[\phi_1]=\mathcal{C}_\delta^{\omega_2}[\phi_2]=0.$$

Recall from (3.14) and (3.17) that

$$\phi_1 = \psi_1 + \psi_2 + O(\delta), \;\; \phi_2 = \psi_1 - \psi_2 + O(\delta).$$

We decompose the solution $\phi \in L^2(\partial D)$ to (4.7) as

$$\phi = a\phi_1 + b\phi_2 + \phi_3 \tag{4.8}$$

with $\langle \phi_1, \phi_3 \rangle = 0$ and $\langle \phi_2, \phi_3 \rangle = 0$, where $\langle \;, \; \rangle$ denotes the L^2-inner product on ∂D.

We have

$$\begin{aligned} &a(\mathcal{C}_\delta^\omega - \mathcal{C}_\delta^{\omega_1})[\phi_1] + b(\mathcal{C}_\delta^\omega - \mathcal{C}_\delta^{\omega_2})[\phi_2] + \mathcal{C}_\delta^\omega[\phi_3] \\ &\qquad = -\delta u^{in}(0)(\psi_1 + \psi_2) - \delta(\mathcal{S}_D^0)^{-1}[\nabla u^{in}(0)\cdot y]. \end{aligned} \tag{4.9}$$

Since

$$(\mathcal{C}_\delta^\omega - \mathcal{C}_\delta^{\omega_j})[\phi_j] = \frac{\omega^2 - \omega_j^2}{v_b^2}\mathcal{K}_D^2[\phi_j] + O(\delta^{3/2}), \quad j = 1, 2,$$

we have $\|(\mathcal{C}_\delta^\omega - \mathcal{C}_\delta^{\omega_j})[\phi_j]\| = O(\delta)$, and hence we conclude from (4.9) that

$$\|\phi_3\| = O((|a| + |b| + 1)\delta). \tag{4.10}$$

Integrating (4.9) over ∂D and then using Lemma 2.1, we obtain

$$-a\left(\frac{\omega^2 - \omega_1^2}{v_b^2}|D| + O(\delta^{3/2})\right) + bO(\delta^2) + \|\phi_3\|O(\delta) = 2\delta u^{in}(0)(C_{11} + C_{12}). \tag{4.11}$$

Similarly, by integrating (4.9) over $\partial D_1 - \partial D_2$, we have

$$aO(\delta^2) - b\left(\frac{\omega^2 - \omega_2^2}{v_b^2}|D| + O(\delta^{3/2})\right) + \|\phi_3\|O(\delta) = -\delta\int_{\partial D_1 - \partial D_2}(\mathcal{S}_D^0)^{-1}[\nabla u^{in}(0)\cdot y]. \tag{4.12}$$

We observe that

$$a = O(1), \quad b = O(\delta^{1/2}), \quad \|\phi_3\| = O(\delta). \tag{4.13}$$

By solving (4.11) and (4.12) for a and b, and then using (3.9), we get

$$a = -\frac{2\delta v_b^2(C_{11} + C_{12})}{|D|(\omega^2 - \omega_1^2)}(u^{in}(0) + O(\delta)) = -\frac{\omega_1^2 + O(\delta^{3/2})}{\omega^2 - \omega_1^2}(u^{in}(0) + O(\delta)), \tag{4.14}$$

$$b = \frac{\delta v_b^2}{|D|(\omega^2 - \omega_2^2)}\left(\int_{\partial D_1 - \partial D_2}(\mathcal{S}_D^0)^{-1}[\nabla u^{in}(0)\cdot y] + O(\delta)\right). \tag{4.15}$$

Now we can calculate ψ. By using (4.5), (4.13), (4.14) and (4.15), we obtain

$$\begin{aligned} \psi &= a(1 + O(\delta^{1/2}))\phi_1 + b\phi_2 + \phi_3 - (\mathcal{S}_D^0)^{-1}[u^{in}(0) + \nabla u^{in}(0)\cdot y] + O(\delta) \\ &= -\frac{\omega_1^2 + O(\delta^{3/2})}{\omega^2 - \omega_1^2}(u^{in}(0) + O(\delta))(1 + O(\delta^{1/2}))\phi_1 - u^{in}(0)\phi_1 \\ &\quad + b\phi_2 + (\mathcal{S}_D^0)^{-1}[\nabla u^{in}(0)\cdot y] + O(\delta) \\ &= \frac{\omega^2 + O(\delta^{3/2})}{\omega_1^2 - \omega^2}(u^{in}(0) + O(\delta))(1 + O(\delta^{1/2}))\phi_1 + b\phi_2 - \nabla u^{in}(0)\cdot(\mathcal{S}_D^0)^{-1}[y] + O(\delta). \end{aligned}$$

Step 4. Finally, we consider the scattered field $u^s(x) := u(x) - u^{in}(x) = \mathcal{S}_D^k[\psi](x)$. It is enough to consider $\mathcal{S}_D^k[\phi_1]$ and $\mathcal{S}_D^k[\phi_2]$.

Note that

$$G(x-y,k) = G(x,k) - \nabla G(x,k) \cdot y + O(\delta|x|^{-1}).$$

Note also that, due to the symmetry of D_1 and D_2, we have

$$\int_{\partial D} y(\psi_1 + \psi_2)(y) d\sigma(y) = 0, \quad \int_{\partial D} \psi_1 - \psi_2 = 0.$$

Therefore, for sufficiently large $|x|$, we have

$$\begin{aligned}
\mathcal{S}_D^k[\phi_1](x) &= \mathcal{S}_D^k[\psi_1 + \psi_2](x) + O(\delta|x|^{-1}) \\
&= \int_{\partial D} G(x-y,k)(\psi_1+\psi_2)(y) d\sigma(y) + O(\delta|x|^{-1}) \\
&= \int_{\partial D} G(x,k)(\psi_1+\psi_2)(y) d\sigma(y) \\
&\quad - \nabla G(x,k) \cdot \int_{\partial D} y(\psi_1+\psi_2)(y) d\sigma(y) + O(\delta|x|^{-1}), \\
&= -2(C_{11} + C_{12}) G(x,k) + 0 + O(\delta|x|^{-1}).
\end{aligned}$$

Similarly, we have

$$\begin{aligned}
\mathcal{S}_D^k[\phi_2](x) &= \mathcal{S}_D^k[\psi_1 - \psi_2](x) + O(\delta|x|^{-1}) \\
&= \int_{\partial D} G(x-y,k)(\psi_1-\psi_2)(y) d\sigma(y) + O(\delta|x|^{-1}) \\
&= -\left(\int_{\partial D} (\psi_1 - \psi_2) y \right) \cdot \nabla G(x,k) + O(\delta|x|^{-1}).
\end{aligned}$$

The proof is then complete. □

Corollary 4.3. *For the rescaled bubble dimer sR_dD with size s and orientation d, where R_d represents a rotation transform which aligns the bubble dimer D in the direction d, we have*

$$\omega_1(\delta, sR_dD) = \frac{1}{s}\omega_1(\delta, D), \tag{4.16}$$

$$\omega_2(\delta, sR_dD) = \frac{1}{s}\omega_2(\delta, D), \tag{4.17}$$

$$g^0(\omega, \delta, sR_dD) = \frac{2(C_{11}+C_{12})s}{1 - \omega_1(\delta, sR_dD)^2/\omega^2}(1 + O(\delta^{1/2})) \tag{4.18}$$

$$g^1(\omega, \delta, sR_dD) = s^3 \int_{\partial D} (\mathcal{S}_D^0)^{-1}[(R_dx)_i](y)(R_dy)_j + \frac{\delta v_b^2 s}{|D|(\omega_2(\delta, sR_dD)^2 - \omega^2)} P^2 d_i d_j. \tag{4.19}$$

Proof. By a scaling argument, one can show that

$$C_{ij}(sD) = sC_{ij}(D), \quad P(sD) = s^2 P(D),$$

and

$$\int_{\partial(sR_d D)} (\mathcal{S}^0_{sR_d D})^{-1}[x_i](y) y_j = s^3 \int_{\partial D} (\mathcal{S}^0_D)^{-1}[(R_d x)_i](y)(R_d y)_j.$$

The proof is then complete. □

Remark 2. *The coefficients C_{11}, C_{12} and P can be explicitly computed using bispherical coordinates. Explicit formulas for C_{11} and C_{12} are given in subsection 2.2. Using the same approach as in the derivation of C_{ij} [20], we can also obtain an explicit formula for P. It holds that*

$$P = -4\pi r_0(r_0 + d_0/2) - 8\pi\alpha^2 \sum_{n=0}^{\infty} (2n+1) e^{-(2n+1)T} \coth((n+1/2)T).$$

5 Homogenization theory

We consider the scattering of an incident acoustic plane wave u^{in} by N identical bubble dimers with different orientations distributed in a homogeneous fluid in $\mathbb{R}^3$. The N identical bubble dimers are generated by scaling the normalized bubble dimer D by a factor s, and then rotating the orientation and translating the center. More precisely, the bubble dimers occupy the domain

$$D^N := \cup_{1 \le j \le N} D_j^N,$$

where $D_j^N = z_j^N + sR_{d_j^N} D$ for $1 \le j \le N$, with z_j^N being the center of the dimer D_j^N, s being the characteristic size, and $R_{d_j^N}$ being the rotation in $\mathbb{R}^3$ which aligns the dimer D_j^N in the direction d_j^N. Here, d_j^N is a vector of unit length in $\mathbb{R}^3$.

We assume that $0 < s \ll 1$, $N \gg 1$ and that $\{z_j^N : 1 \le j \le N\} \subset \Omega$ where Ω is a bounded domain. Let u^{in} be the incident wave which we assume to be a plane wave for simplicity. The scattering of acoustic waves by the bubble dimers can be modeled by the following system of equations:

$$\begin{cases} \nabla \cdot \frac{1}{\rho} \nabla u^N + \frac{\omega^2}{\kappa} u^N = 0 \quad \text{in } \mathbb{R}^3 \backslash D^N, \\ \nabla \cdot \frac{1}{\rho_b} \nabla u^N + \frac{\omega^2}{\kappa_b} u^N = 0 \quad \text{in } D^N, \\ u_+^N - u_-^N = 0 \quad \text{on } \partial D^N, \\ \frac{1}{\rho} \frac{\partial u^N}{\partial \nu}\Big|_+ - \frac{1}{\rho_b} \frac{\partial u^N}{\partial \nu}\Big|_- = 0 \quad \text{on } \partial D^N, \\ u^N - u^{in} \text{ satisfies the Sommerfeld radiation condition,} \end{cases} \tag{5.1}$$

where u^N is the total field and ω is the frequency. Then the solution u^N can be written as

$$u^N(x) = \begin{cases} u^{in} + \mathcal{S}^k_{D^N}[\psi^N], & x \in \mathbb{R}^3 \backslash \overline{D^N}, \\ \mathcal{S}^{k_b}_D[\psi_b^N], & x \in D^N, \end{cases} \tag{5.2}$$

for some surface potentials $\psi, \psi_b \in L^2(\partial D^N)$. Here, we have used the notations

$$L^2(\partial D^N) = L^2(\partial D_1^N) \times L^2(\partial D_2^N) \times \cdots \times L^2(\partial D_N^N),$$
$$\mathcal{S}_{D^N}^k[\psi^N] = \sum_{1 \le j \le N} \mathcal{S}_{D_j^N}^k[\psi_j^N],$$
$$\mathcal{S}_D^{k_b}[\psi_b^N] = \sum_{1 \le j \le N} \mathcal{S}_{D_j^N}^k[\psi_{bj}^N].$$

Using the jump relations for the single layer potentials, it is easy to derive that ψ and ψ_b satisfy the following system of boundary integral equations:

$$\mathcal{A}^N(\omega, \delta)[\Psi^N] = F^N, \tag{5.3}$$

where

$$\mathcal{A}^N(\omega, \delta) = \begin{pmatrix} \mathcal{S}_{D^N}^{k_b} & -\mathcal{S}_{D^N}^k \\ -\frac{1}{2}Id + \mathcal{K}_{D^N}^{k_b,*} & -\delta(\frac{1}{2}Id + \mathcal{K}_{D^N}^{k,*}) \end{pmatrix}, \ \Psi^N = \begin{pmatrix} \psi_b^N \\ \psi^N \end{pmatrix}, \ F^N = \begin{pmatrix} u^{in} \\ \delta \frac{\partial u^{in}}{\partial \nu} \end{pmatrix} |_{\partial D^N}.$$

One can show that the scattering problem (5.1) is equivalent to the system of boundary integral equations (5.3) [9, 7]. Furthermore, it is well-known that there exists a unique solution to the scattering problem (5.1), or equivalently to the system (5.3).

We are concerned with the case when there is a large number of small identical bubble dimers distributed in a bounded domain and the frequency of the incident wave is close to the hybridized Minnaert resonances for a single bubble dimer.

We recall that for a bubble dimer $z + sR_dD$, there exist two quasi-static resonances which are given by

$$\omega_1(\delta, z + sR_dD) = \frac{1}{s}\omega_1(\delta, D),$$
$$\omega_2(\delta, z + sR_dD) = \frac{1}{s}\omega_2(\delta, D).$$

We are interested in the limit when the size s tends to zero while the frequency is of order one. In order to fix the order of the resonant frequency, we make the following assumption.

Assumption 5.1. $\delta = \mu^2 s^2$ *for some positive number* $\mu > 0$.

As a result, the two hybridized resonances have the following asymptotic expansions:

$$\omega_1(\delta, D_j^N) = \omega_{M,1} - i\tau_1\mu^2 s + O(s^2),$$
$$\omega_2(\delta, D_j^N) = \omega_{M,2} + \mu^3\hat{\eta}_1 s^2 - i\mu^4\hat{\eta}_2 s^3 + O(s^4),$$

where

$$\omega_{M,1} = v_b\mu\sqrt{(C_{11} + C_{12})}, \quad \omega_{M,2} = v_b\mu\sqrt{(C_{11} - C_{12})}.$$

Moreover, the monopole and dipole coefficients are given by

$$g^0(\omega, \delta, D_k^N) := \frac{2s(C_{11} + C_{12})}{1 - \omega_1(\delta, D_k^N)^2/\omega^2}(1 + O(\delta^{1/2})), \tag{5.4}$$

$$g^1(\omega, \delta, D_k^N) = (g_{ij}^1(\omega, \delta, D_k^N)), \tag{5.5}$$

where

$$g_{ij}^1(\omega, \delta, D_k^N) := s^3 \int_{\partial D} (\mathcal{S}_D^0)^{-1}[(R_{d_k^N} x)_i](y)(R_{d_k^N} y)_j + \frac{\mu^2 v_b^2 s^3}{2(\omega_2(\delta, D_k^N)^2 - \omega^2)} P^2 (d_k^N)_i (d_k^N)_j.$$

Assumption 5.2. *$\omega = \omega_{M,2} + as^2$ for some real number $a \neq \mu^3 \hat{\eta}_1$.*

Then

$$g^0(\omega, \delta, D_k^N) := \frac{2s(C_{11} + C_{12})}{1 - \omega_{M,1}^2/\omega_{M,2}^2}(1 + O(s)), \tag{5.6}$$

$$g_{ij}^1(\omega, \delta, D_k^N) := \frac{\mu^2 v_b^2 s}{2|D|\omega_{M,2}\left((\mu^3\hat{\eta}_1 - a) - i\mu^4\hat{\eta}_2 s\right)} P^2 (d_k^N)_i (d_k^N)_j + O(s^3). \tag{5.7}$$

We introduce the two constants

$$\tilde{g}^0 = \frac{2(C_{11} + C_{12})}{1 - \omega_{M,1}^2/\omega_{M,2}^2}, \quad \tilde{g}^1 = \frac{\mu^2 v_b^2}{2|D|\omega_{M,2}(\mu^3\hat{\eta}_1 - a)} P^2.$$

We now impose conditions on the distribution of the bubble dimers.

Assumption 5.3. *$sN = \Lambda$ for some positive number $\Lambda > 0$.*

Note that the volume fraction of the bubble dimers is of the order of $s^3 N$. The above assumption implies that the bubble dimers are very dilute with the average distance between neighboring dimers being of the order of $\frac{1}{N^{1/3}}$.

Assumption 5.4. *The bubble dimers are regularly distributed in the sense that*

$$\min_{i \neq j} |z_i^N - z_j^N| \geq \eta N^{-\frac{1}{3}},$$

for some constant η independent of N. Here, $\eta N^{-\frac{1}{3}}$ can be viewed as the minimum separation distance between neighbouring bubble dimers.

In addition, we also make the following assumptions on the regularity of the distribution $\{z_j^N : 1 \leq j \leq N\}$ and the orientation $\{d_j^N : 1 \leq j \leq N\}$.

Assumption 5.5. *There exists a function $\tilde{V} \in C^1(\bar{\Omega})$ such that for any $f \in C^{0,\alpha}(\Omega)$ with $0 < \alpha \leq 1$,*

$$\max_{1 \leq j \leq N} \Big| \frac{1}{N} \sum_{i \neq j} G(z_j^N - z_i^N, k) f(z_i^N) - \int_\Omega G(z_j^N - y, k)\tilde{V}(y) f(y) dy \Big| \leq C \frac{1}{N^{\frac{\alpha}{3}}} \|f\|_{C^{0,\alpha}(\Omega)} \tag{5.8}$$

for some constant C independent of N.

Assumption 5.6. *There exists a matrix valued function $\tilde{B} \in C^1(\bar{\Omega})$ such that for $f \in (C^{0,\alpha}(\Omega))^3$ with $0 < \alpha \leq 1$,*

$$\max_{1\leq j\leq N} \left|\frac{1}{N}\sum_{i\neq j}(f(z_i^N)\cdot d_i^N)(d_i^N\cdot\nabla G(z_i^N-z_j^N,k))-\int_\Omega f(y)\tilde{B}\nabla_y G(y-z_j^N,k)dy\right| \leq C\frac{1}{N^{\frac{\alpha}{3}}}\|f\|_{C^{0,\alpha}(\Omega)} \tag{5.9}$$

for some constant C independent of N.

Remark 3. *If we let $\{z_j^N : 1 \leq j \leq N\}$ be uniformly distributed, then $\tilde{V}$ is a positive constant in Ω. We can also let the orientation be uniformly distributed in the unit sphere in the sense that the average of the matrix $d_j^N(d_j^N)^T$ in any neighborhood of any point in Ω tends to a multiple of the identity matrix as N tends to infinity. In that case, $\tilde{B}$ is a positive constant multiple of the identity matrix at each point.*

5.1 The homogenized equations

In the same spirit as the point interaction approximation [3, 14, 17, 29, 30], we now formally derive the homogenized equation.

For $1 \leq j \leq N$, denote by

$$u_j^{i,N} = u^{in} + \sum_{i\neq j}\mathcal{S}_{D_i^N}^k[\psi_i^N], \tag{5.10}$$

$$u_j^{s,N} = \mathcal{S}_{D_j^N}^k[\psi_j^N]. \tag{5.11}$$

It is clear that $u_j^{i,N}$ is the total incident field which impinges on the bubble D_j^N, and $u_j^{s,N}$ is the corresponding scattered field. Denote by

$$\Omega_N = \Omega\backslash \cup_{1\leq j\leq N} B(z_j^N, \sqrt{s}).$$

Note that the volume fraction of the set $\cup_{1\leq j\leq N}B(z_j^N, \sqrt{s})$ is of the order

$$O(N\cdot s^{\frac{3}{2}}) = O(N\cdot s)\cdot s^{\frac{1}{2}},$$

which tends to zero as $N \to \infty$ under Assumption 5.3. We have

$$u^N(x) = u^{in} + \sum_{1\leq k\leq N} u_k^{s,N}(x) = u_j^{i,N}(x) + u_j^{s,N}(x), \text{ for each } 1 \leq j \leq N \text{ and } x \in \Omega_N.$$

Proposition 5.1. *Under Assumptions 5.1, 5.2, 5.3, we have that, for $x \in \Omega_N$,*

$$u_j^{s,N}(x) \approx g^0(\omega,\delta,D_j^N)u_j^{i,N}(z_j^N)G(x-z_j^N,k) + \nabla u_j^{i,N}(z_j^N)\cdot g^1(\omega,\delta,D_j^N)\nabla G(x-z_j^N,k).$$

We further assume that

Assumption 5.7. *There exists some macroscopic field $u \in C^{1,\alpha}(\Omega)$ such that*

$$u^N(x) \to u(x) \ \text{for}\ x \in \Omega_N.$$

Here the convergence is understood in the sense that for any $\epsilon > 0$, there exists N_0 such that for all $N \geq N_0$, we have

$$\|u^N(x) - u(x)\|_{C^{1,\alpha}(\Omega_N)} \leq \epsilon.$$

The above assumption implies in particular that $u_j^{i,N}(z_j^N) \to u(z_j^N)$. Therefore

$$g^0(\omega,\delta,D_j^N)u_j^{i,N}(z_j^N)G(x-z_j^N,k) \to \frac{1}{N}\Lambda u(z_j^N)\tilde{g}^0 G(x-z_j^N,k),$$
$$\nabla u_j^{i,N}(z_j^N)\cdot g^1(\omega,\delta,D_j^N,z_j^N)\nabla G(x-z_j^N,k) \to \frac{1}{N}\Lambda \tilde{g}^1(\nabla u(z_j^N)\cdot d_j^N)(d_j^N\cdot \nabla G(x-z_j^N,k)).$$

On the other hand, we have

$$u^N(x) \approx u^{in} + \sum_{1\leq j\leq N} g^0(\omega,\delta,D_j^N)u_j^{i,N}(z_j^N)G(x-z_j^N,k) + \sum_{1\leq j\leq N} \nabla u_j^{i,N}(z_j^N)\cdot g^1(\omega,\delta,D_j^N)\nabla G(x-z_j^N,k).$$

By letting $N\to\infty$, we obtain

$$u(x) = u^{in} + \int_\Omega \Lambda\tilde{g}^0 u(y)\tilde{V}(y)G(x-y,k)dy + \int_\Omega \Lambda\tilde{g}^1\nabla u(y)\tilde{B}\nabla G(x-y,k)dy. \quad (5.12)$$

By applying the operator $\triangle + k^2$ to both side of the above equation, we get

$$(\triangle + k^2)u(x) = \Lambda\tilde{g}^0\tilde{V}u(x) + \nabla\cdot\left(\Lambda\tilde{g}^1\tilde{B}\nabla u\right)(x), \quad \text{in } \Omega.$$

Or equivalently,

$$\nabla\cdot\left(I - \Lambda\tilde{g}^1\tilde{B}\right)\nabla u(x) + (k^2 - \Lambda\tilde{g}^0\tilde{V})u(x) = 0, \quad \text{in } \Omega. \quad (5.13)$$

Therefore, we have shown that the microscopic field u^N tends to a macroscopic field u which satisfies the following effective equation

$$\nabla\cdot M_1(x)\nabla u(x) + M_2(x)u(x) = 0, \quad \text{in } \mathbb{R}^3, \quad (5.14)$$

where

$$M_1 = \begin{cases} I, & \text{in } \mathbb{R}^3\setminus\Omega,\\ I - \Lambda\tilde{g}^1\tilde{B}, & \text{in } \Omega,\end{cases}$$

and

$$M_2 = \begin{cases} k^2, & \text{in } \mathbb{R}^3\setminus\Omega,\\ k^2 - \Lambda\tilde{g}^0\tilde{V}, & \text{in } \Omega.\end{cases}$$

Remark 4. *We have derived the effective media theory in a formal way under the crucial Assumption 5.7. We leave a rigorous justification as an open problem for future investigation. We refer to [14] for a similar but easier problem where a rigorous justification is possible.*

5.2 Double-negative refractive index media

If the bubble dimers are distributed such that $\tilde{B}$ is a positive matrix with $\tilde{B}(x) \geq C > 0$ for some constant C for all $x \in \Omega$, then we see that for ω in the form $\omega = \omega_{M,2} + as^2$ with $a < \mu^3 \hat{\eta}_1$, and sufficiently large Λ, both the matrix $Id - \Lambda \tilde{g}^1 \tilde{B}$ and the scalar function $k^2 - \Lambda \tilde{g}^0 \tilde{V}$ are negative. Therefore, we obtain an effective double-negative media with both negative mass density and negative bulk modulus. On the other hand, for ω in between $\omega_{M,1}$ and $\omega_{M,2}$ but away from the anti-resonance $\omega_{M,2}$, $\tilde{g}^1$ may be small enough such that the matrix $Id - \Lambda \tilde{g}^1 \tilde{B}$ is positive, while the matrix $k^2 - \Lambda \tilde{g}^0 \tilde{V}$ remains negative. Then the obtained effective media has negative mass density and positive bulk modulus.

6 Numerical illustrations

In this section, we illustrate the double-negative refractive index phenomenon in bubbly media by numerical examples.

We consider a cubic array of identical spherical bubble dimers. Suppose Ω is a cube with a side length of $L = 20$, *i.e.*, $\Omega = [0, 20]^3$. Let $a = 0.2$ and define a small cube $\Omega_a = [0, a]^3$. Then Ω can be considered as a union of small cubes as follows:

$$\Omega = \bigcup_{n_1, n_2, n_3 = 0, 1, \ldots, 99} \Omega_a + a(n_1, n_2, n_3).$$

We assume that a bubble dimer is centered in each of the small cubes. Then the total number of dimers is $N = 10^6$ and the periodicity of the dimer array is $a = 0.2$.

Recall that a bubble dimer is described by $z + sR_d D$, where z is the center of the dimer, s is its characteristic size, and R_d is a rotation in $\mathbb{R}^3$ which aligns the dimer in the direction d, where d is a unit vector. We set the characteristic size of the dimers to be $s = 0.1$. Since D has unit volume, the radius r_0 of the bubbles comprising the dimers is $r_0 = s(3/4\pi)^{1/3} \approx 0.005$.

We set $\tilde{\rho} = \tilde{\kappa} = 1$ and $\rho = \kappa = 5 \times 10^3$. Then $v = \tilde{v} = 1$, $k = \tilde{k} = \omega$, and $\delta = 2 \times 10^{-4}$. We assume that the two bubbles comprising each dimer are separated by a distance of $l = 5r_0 \approx 0.0248$. Moreover, each dimer is randomly oriented so that the unit vector d is uniformly distributed on the unit sphere. Under these assumptions, we can easily check that $\Lambda = 8 \times 10^3$, $\tilde{V} \approx |\Omega|^{-1} = 1.25 \times 10^{-4}$, and $\tilde{B} \approx (2|\Omega|)^{-1} I = 6.25 \times 10^{-5} I$.

Now we consider the effective properties of the homogenized media. Recall that the effective coefficients of the homogenized equation (5.13) are $k^2 - \Lambda \tilde{g}^0 \tilde{V}$ and $I - \Lambda \tilde{g}^1 \tilde{B}$. Using the above parameters, we have

$$\begin{aligned} & k^2 - \Lambda \tilde{g}^0 \tilde{V} \approx \omega^2 - \tilde{g}^0 = \omega^2 (1 - \tilde{g}^0 / \omega^2), \\ & I - \Lambda \tilde{g}^1 \tilde{B} \approx (1 - \tilde{g}^1 / 2) I. \end{aligned} \tag{6.1}$$

Note that the coefficient $I - \Lambda \tilde{g}^1 \tilde{B}$ can be roughly considered as a scalar quantity. The scattering functions $\tilde{g}_0$ and $\tilde{g}_1$ can be computed as follows. Since $\tilde{g}_0 \approx s g_0$ and $\tilde{g}_1 \approx s g_1$,

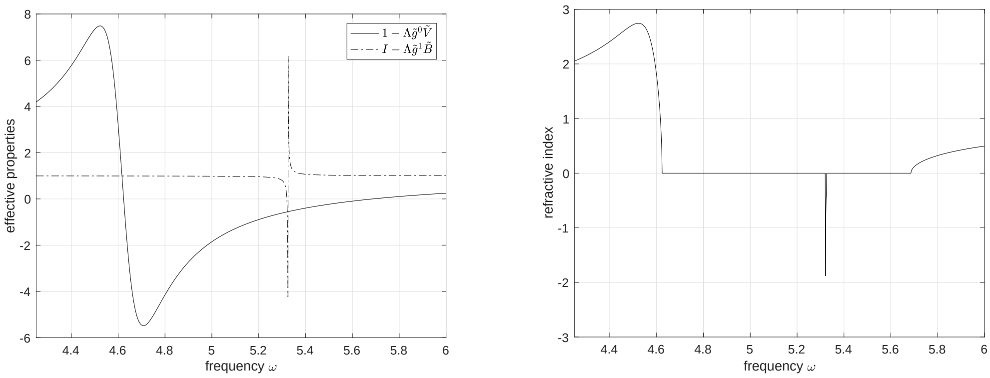

Figure 1: The effective properties of the homogenized media (left), and the refractive index (right).

we have from (4.18) and (4.19) that

$$\tilde{g}^0(\omega,\delta,sR_dD)\approx\frac{2(C_{11}+C_{12})}{1-\omega_1^2/\omega^2},$$
$$\tilde{g}^1(\omega,\delta,sR_dD)\approx\frac{\delta\tilde{v}^2}{2(\omega_2^2-\omega^2)}P^2d_id_j.$$

The resonance frequencies ω_1 and ω_2 can be easily calculated using a standard multipole expansion together with a root finding method such as Muller's method [9]. We find that $\omega_1\approx 4.6171-0.0926i$ and $\omega_2\approx 5.3253-0.0005i$. Then it is simple to compute $\tilde{g}^0$ and $\tilde{g}^1$.

In the left of Figure 1, we plot the two effective coefficients as functions of frequency. Clearly, there is a narrow region contained in the interval $[5.2, 5.4]$ in which both of the coefficients are negative. So we expect that the negative refraction occurs in this frequency region.

We next consider the refractive index. In view of (5.13) and (6.1), the effective mass density ρ_{eff} and the effective bulk modulus κ_{eff} can be computed approximately by

$$\rho_{eff}\approx 1-\tilde{g}^1/2,\quad \kappa_{eff}\approx(1-\tilde{g}^0/\omega^2)^{-1}.$$

As usual, we define the refractive index n_{eff} by

$$n_{eff}=\sqrt{\rho}_{eff}\sqrt{\kappa_{eff}^{-1}}.$$

In the right figure of Figure 1, we plot the refractive index as a function of frequency. It is clear that the refractive index becomes negative in a narrow region contained in the interval $[5.2, 5.4]$, as expected.

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