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Seminar for Applied Mathematics SAM

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Research Report No. 2017-45
September 2017

Seminar für Angewandte Mathematik
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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 implies the use of two different kinds of subwavelength resonators. 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 [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, it is proved that both the effective mass density and the bulk modulus of the bubbly fluid can be negative near the hybridized Minnaert resonances. 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 the bubble dimer are analyzed. Approximate formulas for the two hybridized Minnaert resonances are derived. Moreover, it is proved that the bubble dimer can be approximated as the sum of a monopole source and a dipole source. For an appropriate bubble volume fraction of randomly oriented bubble dimers, a double-negative effective medium theory near the hybridized Minnaert resonances is obtained.

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

Keywords. bubble, Minnaert resonance, hybridization, homogenization, double-negative metamaterial.

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*The work of Hyundae Lee was supported by National Research Fund of Korea (NRF-2015R1D1A1A01059357, NRF-2017R1A4A1014735). The work of Hai Zhang was partially supported by HK RGC GRF grant 16304517 and startup fund R9355 from HKUST.

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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 [19, 23, 24].

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 [25]. 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 [3]. 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 [21, 20, 22]. This motivated our series of bubble studies [3, 5, 6, 14]. We refer to [3] 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 [21]. 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 [2, 12, 13].

In [5], 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 [17, 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 [6] 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 implies 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 [28, 29, 30].

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 consists of two identical closely 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 approximate formulas for the two Minnaert resonances, and prove that the bubble dimer can be approximated as the sum of a monopole source and a dipole source. The hybridized Minnaert resonances correspond to 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. For an appropriate bubble volume fraction, when the excitation frequency is close to the hybridized Minnaert resonances we obtain a double-negative effective mass density and bulk modulus for bubbly media consisting of a large number of randomly distributed bubble dimers. The dipole sources in the background medium contribute to the effective mass density while the monopole sources 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:

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

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). □

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 [18]. 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

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

In addition, when D_1 and D_2 are identical balls, the next lemma provides some useful formulas.

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 realtions (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). $\square$

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 $(\tilde{\kappa}, \tilde{\rho})$ 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

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

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

$$v := \sqrt{\frac{\kappa}{\rho}}, \quad \tilde{v} := \sqrt{\frac{\tilde{\kappa}}{\tilde{\rho}}}, \quad k = \omega\sqrt{\frac{\rho}{\kappa}}, \quad \tilde{k} = \omega\sqrt{\frac{\tilde{\rho}}{\tilde{\kappa}}}. \tag{3.2}$$

We assume that

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

and

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

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

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

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

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

We denote by

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

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) [3]. 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:*

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

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

where

$$\tau_1 = \frac{\tilde{v}}{4\pi}(C_{11} + C_{12})^2 - \frac{1}{4\pi}\left(1 - \frac{\tilde{v}}{v}\right)(C_{11} + C_{12})^{3/2},$$

and $\hat{\eta}_1$ and $\hat{\eta}_2$ are real numbers which are determined by D, v, and $\tilde{v}$.

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

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

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

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

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

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

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

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

where we have used the identity

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

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 $O(\omega^4 + \delta\omega^2)$,

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

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:

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

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

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

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 $\eta_0 := \sqrt{\frac{\tilde{v}^2 \lambda_2}{\beta}}$. For the purpose of normalization, we require that

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

Step 4. By using (2.4), we have

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

Hence, we get

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

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:

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

  whence ϕ_1 is uniquely determined.

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

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

whence ϕ_2 is uniquely determined.

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

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

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

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

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

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

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

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

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:

$$\omega_2 = \sqrt{\delta\tilde{v}^2\lambda_2} + \delta^{3/2}\hat{\eta}_1 + i\delta^2\hat{\eta}_2 + O(\delta^{5/2}), \tag{3.23}$$

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

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

4 The point and dipole 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 the sum of a monopole source and a dipole source.

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

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

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

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

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

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

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

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

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

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 $\nabla^2 u^{in}=O(\omega^2)=O(\delta)$, 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

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

Since

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

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

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

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

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

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

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

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

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

$$\begin{aligned}
\mathcal{S}_D^k[\phi_2](x) &= \mathcal{S}_D^k[\psi_1-\psi_2](x) + O(\delta|x|^{-1}) \\
&= \int_{\partial D} G(x-y,k)(\psi_1-\psi_2)(y)d\sigma(y) + O(\delta|x|^{-1}) \\
&= -\left(\int_{\partial D} (\psi_1-\psi_2)y\right) \cdot \nabla G(x,0) + O(\delta|x|^{-1}).
\end{aligned}$$

The proof is then complete. □

Corollary 4.3. *For the rescaled bubble dimer sR_dD with size s and orientation d, we have*

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

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

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

$$g^1(\omega,\delta,sR_dD) = s^3\int_{\partial D}(\mathcal{S}_D^0)^{-1}[y_i]y_j + \frac{\delta\tilde{v}^2 s}{|D|(\omega_2(\delta,sR_dD)^2-\omega^2)}P^2 d_i d_j. \tag{4.19}$$

Proof. By a scaling argument, one can show that

$$C_{ij}(sD) = sC_{ij}(D), \quad P(sD) = s^2P(D), \quad \int_{\partial(sD)}(\mathcal{S}_{sD}^0)^{-1}[y_i]y_j = s^3\int_{\partial D}(\mathcal{S}_D^0)^{-1}[y_i]y_j.$$

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} [18], 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

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

where $D_j^N = z_j^N + sR_{d_j^N} D$ for $1 \leq j \leq N$, 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 $\{z_j^N : 1 \leq j \leq N\} \subset \Omega$ 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:

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

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

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

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

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

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

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

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 resonances have the following asymptotic expansions:

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

where

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

Moreover, the monopole and dipole coefficients are given by

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

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

where

$$g_{ij}^1(\omega,\delta,D_j^N) := s^3\int_{\partial D}(\mathcal{S}_D^0)^{-1}[y_i]y_j + \frac{\mu^2\tilde{v}^2 s^3}{2(\omega_2(\delta,D_j^N)^2-\omega^2)}P^2 d_i d_j.$$

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

Then

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

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

We introduce the two constants

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

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^3N. 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 assumption 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 $0 < \alpha \leq 1$,*

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

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 $0 < \alpha \leq 1$,*

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

for some constant C independent of N.

Assumption 5.7. *There exists a constant $C > 0$ such that*

$$\max_{1\le j\le N} \frac{1}{N}\sum_{i\neq j} \frac{1}{|z_j^N - z_i^N|} \le C,$$
$$\max_{1\le j\le N} \frac{1}{N}\sum_{i\neq j} \frac{1}{|z_j^N - z_i^N|^2} \le C,$$

for all $1 \le j \le N$.

Remark 3. *If we let $\{z_j^N : 1 \le j \le N\}$ be uniformly distributed, then $\tilde{V}$ is a positive constant in Ω. We can also let the orientation be uniformly distributed 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

We now formally derive the homogenized equation.

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

$$u_j^{i,N} = u^{in} + \sum_{i\neq j} \mathcal{S}^k_{D_i^N}[\psi_i^N], \tag{5.10}$$
$$u_j^{s,N} = \mathcal{S}^k_{D_j^N}[\psi_j^N]. \tag{5.11}$$

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

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

We have

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

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 assume that there exists some $u \in C^{1,\alpha}(\Omega)$ such that

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

In particular, $u_j^{i,N}(z_j^N) \to u(z_j^N)$. Hereafter, the convergence is in the sense that for any $\epsilon > 0$, there exists N_0 such that for all $N \ge N_0$, we have

$$|u^N(x) - u(x)| \le \epsilon$$

for all $x \in \Omega_N$.

Since $u_j^{s,N}(x) \to 0$ for $|x - z_j^N| \geq \sqrt{s}$, we can neglect the effect of the scattered field $u_j^{s,N}$ from each bubble D_j^N. Thus we have

$$u_j^{i,N}(z_j^N) \to u(z_j^N).$$

As a result,

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

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

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

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

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

Or equivalently,

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

In other words, u satisfies

$$\nabla \cdot M_1(x) \nabla u(x) + M_2(x) u(x) = 0, \quad \text{in } \mathbb{R}^2, \quad (5.14)$$

where

$$M_1 = \begin{cases} I, & \text{in } \mathbb{R}^2 \setminus \Omega, \\ I - \Lambda \tilde{g}^1 \tilde{B}, & \text{in } \Omega, \end{cases}$$

and

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

Remark 4. *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 medium with both negative mass and negative bulk modulus.*

5.2 Justification based on the point scatter approximation

In this section, we justify the homogenization theory. The rigorous theory for the original scattering system (5.10) is very technical. According to Proposition 5.1, each bubble dimer behaves as the sum of a monopole source and a dipole source when the frequency is close to the two quasi-static Minnaert resonances. We can simplify the presentation and analysis significantly by considering the following system:

$$v_j^{i,N}(x) = u^{in}(x) + \sum_{i\neq j} v_i^{s,N}(x), \tag{5.15}$$

$$v_j^{s,N}(x) = g^0(\omega,\delta,D_j^N)v_j^{i,N}(z_j^N)G^k(x,z_j^N) + \nabla v_j^{i,N}(z_j^N)\cdot g^1(\omega,\delta,D_j^N,z_j^N)\nabla G^k(x,z_j^N). \tag{5.16}$$

We denote by

$$v^N(x) = u^{in}(x) + \sum_i v_i^{s,N}(x).$$

We introduce the following four integral operators:

$$\mathcal{T}_1 : C^{1,\alpha}(\Omega) \to C^{1,\alpha}(\Omega), \quad \mathcal{T}_1 f(x) = \int_\Omega G(x-y,k)\tilde{V}(y)f(y)dy; \tag{5.17}$$

$$\mathcal{T}_2 : (C^{0,\alpha}(\Omega))^3 \to C^{1,\alpha}(\Omega), \quad \mathcal{T}_2 f(x) = \int_\Omega f(y)\tilde{B}(y)\nabla G(x-y,k)dy; \tag{5.18}$$

$$\mathcal{T}_3 : C^{1,\alpha}(\Omega) \to (C^{0,\alpha}(\Omega))^3, \quad \mathcal{T}_3 f(x) = \int_\Omega \nabla_x G(x-y,k)\tilde{V}(y)f(y)dy; \tag{5.19}$$

$$\mathcal{T}_4 : (C^{0,\alpha}(\Omega))^3 \to (C^{0,\alpha}(\Omega))^3, \quad \mathcal{T}_4 f(x) = \int_\Omega f(y)\tilde{B}(y)\nabla_x\nabla_y G(x-y,k)dy. \tag{5.20}$$

We also define $\mathcal{T} : C^{1,\alpha}(\Omega)\times(C^{0,\alpha}(\Omega))^3 \to C^{1,\alpha}(\Omega)\times(C^{0,\alpha}(\Omega))^3$ by

$$\mathcal{T}\begin{bmatrix} u \\ v \end{bmatrix} = \begin{bmatrix} \tilde{g}^0\Lambda\mathcal{T}_1 u + \tilde{g}^1\Lambda\mathcal{T}_2 v \\ \tilde{g}^0\Lambda\mathcal{T}_3 u + \tilde{g}^1\Lambda\mathcal{T}_4 v \end{bmatrix} = \Lambda\begin{bmatrix} \tilde{g}^0\mathcal{T}_1, \tilde{g}^1\mathcal{T}_2 \\ \tilde{g}^0\mathcal{T}_3, \tilde{g}^1\mathcal{T}_4 \end{bmatrix}\begin{bmatrix} u \\ v \end{bmatrix}. \tag{5.21}$$

Lemma 5.2. *The operators $\mathcal{T}_1$, $\mathcal{T}_2$, $\mathcal{T}_3$, $\mathcal{T}_4$ and $\mathcal{T}$ are compact.*

Lemma 5.3. *The following statements are equivalent:*

(i) There exists a unique solution to the integral equation (5.12);

(ii) There exists a unique solution to the differential equation (5.14) such that $u - u^{in}$ satisfies the Sommerfeld radiation condition at infinity;

(iii) There exists a unique solution to the system

$$\begin{bmatrix} u \\ v \end{bmatrix} = \mathcal{T}\begin{bmatrix} u \\ v \end{bmatrix} + \begin{bmatrix} u^{in} \\ \nabla u^{in} \end{bmatrix}.$$

We assume that the homogenized system is well-posed. More precisely, we make the following assumption.

Assumption 5.8. *For each incident field u^{in}, there exists a unique solution to (5.14) such that $u - u^{in}$ satisfies the Sommerfeld radiation condition at infinity.*

Lemma 5.4. *Under Assumption (5.8), the operator $Id - \mathcal{T}$ is invertible with a bounded inverse in $C^{1,\alpha}(\Omega) \times (C^{0,\alpha}(\Omega))^3$.*

We now introduce the discrete version of the operators $\mathcal{T}_1$, $\mathcal{T}_2$, $\mathcal{T}_3$, $\mathcal{T}_4$ and $\mathcal{T}$. We define

$$\mathcal{T}_1^N : \mathbb{C}^N \to \mathbb{C}^N; \mathcal{T}_2^N : \mathbb{C}^{3N} \to \mathbb{C}^N; \mathcal{T}_3^N : \mathbb{C}^N \to \mathbb{C}^{3N}; \mathcal{T}_4^N : \mathbb{C}^{3N} \to \mathbb{C}^{3N}$$

by the following formulas: for $x^N = (x_1^N, \cdots, x_N^N) \in \mathbb{C}^N$, $y^N = (y_1^N, \cdots, y_N^N) \in \mathbb{C}^{3N}$, we let

$$\left(\mathcal{T}_1^N x^N\right)_j = \frac{1}{N} \sum_{i \neq j} G(z_j^N - z_i^N, k) x_i^N; \tag{5.22}$$

$$\left(\mathcal{T}_2^N y^N\right)_j = \frac{1}{N} \sum_{i \neq j} (y_j^N \cdot d_j^N) \cdot (d_j^N \cdot \nabla G(z_j^N - z_i^N, k)); \tag{5.23}$$

$$\left(\mathcal{T}_3^N x^N\right)_j = \frac{1}{N} \sum_{i \neq j} \nabla G(z_j^N - z_i^N, k) x_i^N; \tag{5.24}$$

$$\left(\mathcal{T}_4^N y^N\right)_j = \frac{1}{N} \sum_{i \neq j} (y_j^N \cdot d_j^N) \cdot (d_j^N \nabla_x \nabla_y G(z_j^N - z_i^N, k)). \tag{5.25}$$

We also denote by $\mathcal{T}^N : \mathbb{C}^N \times \mathbb{C}^{3N} \to \mathbb{C}^N \times \mathbb{C}^{3N}$,

$$\mathcal{T}^N \begin{bmatrix} x^N \\ y^N \end{bmatrix} = \begin{bmatrix} \tilde{g}^0 \Lambda \mathcal{T}_1^N x^N + \tilde{g}^1 \Lambda \mathcal{T}_2^N y^N \\ \tilde{g}^0 \Lambda \mathcal{T}_3^N x^N + \tilde{g}^1 \Lambda \mathcal{T}_4^N y^N \end{bmatrix}. \tag{5.26}$$

We have

$$\begin{aligned} v_j^{i,N}(z_j^N) &= u^{in}(z_j^N) + \sum_{i \neq j} g^0(\omega, \delta, D_i^N) v_j^{i,N}(z_j^N) G(z_i^N - z_j^N, k) \\ &\quad + \sum_{i \neq j} \nabla v_i^{i,N}(z_i^N) \cdot g^1(\omega, \delta, D_i^N) \nabla G(z_i^N - z_j^N, k) \\ &= u^{in}(z_j^N) + \frac{\Lambda \tilde{g}^0}{N} \sum_{i \neq j} (1 + O(s)) v_i^{i,N}(z_i^N) G(z_i^N - z_j^N, k) \\ &\quad + \frac{\Lambda \tilde{g}^1}{N} \sum_{i \neq j} \nabla v_j^{i,N}(z_i^N) \cdot (1 + O(s)) \nabla G(z_i^N - z_j^N, k), \end{aligned}$$

and

$$
\begin{aligned}
\nabla v_j^{i,N}(z_j^N) &= \nabla u^{in}(z_j^N) + \sum_{i\neq j} g^0(\omega,\delta,D_i^N) v_i^{i,N}(z_i^N) \nabla\nabla_x G(z_i^N - z_j^N, k) \\
&\quad + \sum_{i\neq j} \nabla v_i^{i,N}(z_i^N)\cdot g^1(\omega,\delta,D_i^N)\nabla_x\nabla_y G(z_i^N - z_j^N, k) \\
&= \nabla u^{in}(z_j^N) + \frac{\Lambda\tilde{g}^0}{N}\sum_{i\neq j}(1+O(s)) v_j^{i,N}(z_i^N)\nabla\nabla_x G(z_i^N - z_j^N, k) \\
&\quad + \frac{\Lambda\tilde{g}^0}{N}\sum_{i\neq j}\nabla v_j^{i,N}(z_i^N)\cdot(1+O(s))\nabla_x\nabla_y G(z_i^N - z_j^N, k).
\end{aligned}
$$

If we denote by

$$
\begin{aligned}
x^N &= (x_1^N,\cdots,x_N^N) = (v_1^{i,N}(z_1),\cdots,v_N^{i,N}(z_N)),\\
y^N &= (y_1^N,\cdots,y_N^N) = (\nabla v_1^{i,N}(z_1),\cdots,\nabla v_N^{i,N}(z_N)),\\
b^N &= (u^{in}(z_1),\cdots,u^{in}(z_N)),\\
c^N &= (\nabla u^{in}(z_1),\cdots,\nabla u^{in}(z_N)),
\end{aligned}
$$

then $(x^N, y^N)^T$ satisfies the following equation:

$$
\begin{bmatrix} x^N \\ y^N \end{bmatrix} = \mathcal{T}_s^N \begin{bmatrix} x^N \\ y^N \end{bmatrix} + \begin{bmatrix} b^N \\ c^N \end{bmatrix}.
$$

Lemma 5.5. *Viewed as operators from $(\mathbb{C}^{4N}, \|\cdot\|_\infty)$ to $(\mathbb{C}^{4N}, \|\cdot\|_\infty)$, we have*

$$
\|\mathcal{T}_s^N - \mathcal{T}^N\|_\infty \leq Cs
$$

for some constant C independent of N.

On the other hand, we have the following lemma which follows by a standard interpolation argument.

Lemma 5.6. *Under Assumption (5.8), the operator $I - \mathcal{T}^N$ is invertible for N sufficiently large. Moreover,*

$$
\|I - \mathcal{T}^N\|_\infty \leq C
$$

for some constant C independent of N.

Lemma 5.7. *We have*

$$
(v_j^{i,N}(z_j^N), \nabla v_j^{i,N}(z_j^N)) \to (u(z_j^N), \nabla u(z_j^N))
$$

uniformly as N tends to infinity.

Finally, we prove our main result.

Theorem 5.8. *Under Assumptions 5.1, 5.2, 5.3, 5.4, 5.5, 5.6, and 5.8, we have* $v^N(x) \to u(x)$ *uniformly for* $x \in \Omega_N$.

Proof. For $x \in \Omega_N$, we have

$$\begin{aligned}
v^N(x) &= u^{in}(x) + \sum_i v_i^{s,N}(x) \\
&= u^{in}(x) + \frac{\Lambda \tilde{g}^0}{N} \sum_i (1 + O(s)) v_i^{i,N}(z_i^N) G(z - z_i^N, k) \\
&\quad + \frac{\Lambda \tilde{g}^1}{N} \sum_i \nabla v_j^{i,N}(z_i^N) \cdot (1 + O(s)) \nabla G(x - z_i^N, k) \\
&= u^{in}(x) + \frac{\Lambda \tilde{g}^0}{N} \sum_i v_i^{i,N}(z_i^N) G(x - z_i^N, k) \\
&\quad + \frac{\Lambda \tilde{g}^1}{N} \sum_i \nabla v_j^{i,N}(z_i^N) \cdot \nabla G(x - z_i^N, k) + O(s).
\end{aligned}$$

Define

$$\tilde{u}^N(x) = u^{in}(x) + \frac{\Lambda \tilde{g}^0}{N} \sum_i u(z_i^N) G(x - z_i^N, k) + \frac{\Lambda \tilde{g}^1}{N} \sum_i \nabla u(z_i^N) \cdot \nabla G(x - z_i^N, k).$$

By Lemma 5.7 and Assumption 5.7, we can show that $\tilde{u}^N(x) - v^N(x) \to 0$ uniformly.

On the other hand, we have

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

Using Assumptions 5.5–5.6, one can show that $u(x) - \tilde{u}^N(x) \to 0$ uniformly. Therefore, it follows that $v^N(x) \to u(x)$ uniformly. $\square$

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 $z + sR_dD$, 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 homgenized 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 $\tilde{g}_0 \approx sg_0$ and $\tilde{g}_1 \approx sg_1$, we have from (4.18) and (4.19) that

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

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.

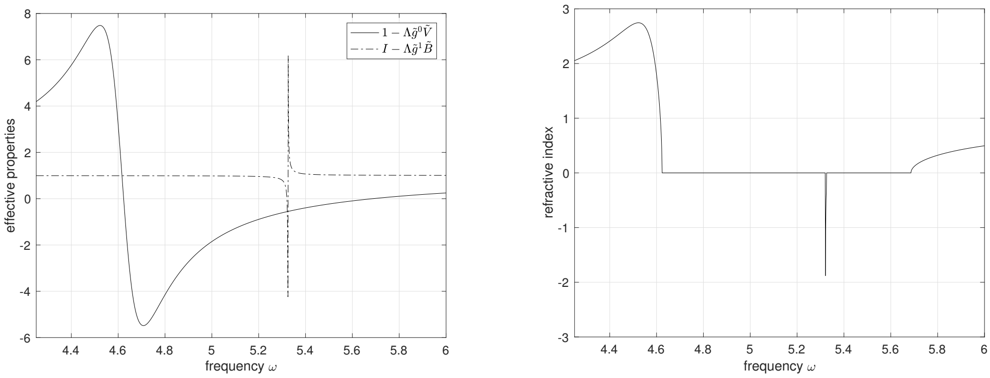

Figure 1: The effective properties of the homogenized media (left), and the refractive index (right).

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