## Air Bubbles in Water: A Strongly Multiple Scattering Medium for Acoustic Waves

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Using a newly developed multiple scattering scheme, we calculate band structure and transmission properties for acoustic waves propagating in bubbly water. We prove that the multiple scattering effects are responsible for the creation of wide gaps in the transmission even in the presence of strong positional and size disorder.

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In this paper we present results obtained by employing a novel method [1] which fully incorporates the multiple scattering effects, and is applicable to acoustic wave propagation in media consisting of nonoverlapping spherical scatterers embedded in a fluid. The method, based on the well known (to solid state physicists) Korringa-Kohn-Rostoker (KKR) approach [2,3], is capable of calculating (a) the sound transmission through a finite cluster of regularly or irregularly placed scatterers (up to a few hundred of them), and (b) the band structures (i.e., the  $\omega$  vs k relation) for an infinite periodic lattice of scatterers.

Acoustic and elastic waves in complex media have attracted recently a wider interest [4–16] because of (a) the so-called phononic band gap crystals (i.e., periodic structures exhibiting gaps in their acoustic spectra) and (b) the opportunity they offer to study under diverse and controlled conditions the major problem of wave localization arising as a result of multiple scattering. The latter is expected to be important in bubbly liquids, because the single bubble is a strong sound scatterer exhibiting a huge s-wave resonance, the so-called Minnaert resonance, at a frequency  $\omega_o$  such that  $\omega_o r_s/c_i = \sqrt{3\rho_i/\rho_o}$ ;  $r_s$  is the bubble radius,  $c_i$ ,  $\rho_i$  and  $c_o$ ,  $\rho_o$  are the sound velocity and the density of the gas inside and the liquid outside the bubble, respectively [17].

Besides the resonance, the isotropic scattering creates an almost uniform and large background in the scattering cross section,  $\sigma$  ( $\sigma/\pi r_s^2 \approx 4$  for  $0.05 \lesssim \omega r_s/c_o \lesssim 1$ ), interrupted by some very sharp higher frequency resonances (the first two at  $\omega r_s/c_o \simeq 0.48$  and  $\omega r_s/c_o \simeq$ 0.77). As has been pointed out [10,13] such a frequency dependence of the single scatterer cross section provides ideal conditions for the appearance of wide spectral (density of states or propagation) gaps in a multiple scattering environment, because in the frequency region between resonances neither the host material allows propagation, nor coherent hopping to neighboring scatterers utilizing the resonances can take place. Notice that the mean free path, l, at the resonance, as obtained from the approximate formula  $l = 1/n\sigma$  (where n is the average concentration of the bubbles), equals to the average nearest neighbor distance already at an air volume fraction, f, of the order of 0.0002. This means that multiple scattering effects are important even for f as low as 0.0002. Thus, acoustic waves in bubbly liquids (especially water), in addition to being very important per se [17], provide an almost ideal case for examining in detail the localization question. Up to now the extensive studies of bubbly water (or other bubbly liquids) have been analyzed in the framework of single scattering [17]. Only recently Ruffa [18], employing a finite element analysis, has calculated among other quantities the low frequency dependence (for  $\omega < \omega_0$ ) of sound velocity along the [100] direction of an infinite periodic simple cubic arrangement of bubbles in water; also Kushwaha et al. [19], employing the plane wave method [20], and assuming again an infinite medium with periodic placement of the bubbles (in fcc, bcc, and sc lattices), calculated the  $\omega$  vs **k** relation for the first thirty lowest bands.

We applied first our multiple scattering method to sound waves in infinite periodic bubbly water in order to check our results against those of Refs. [18] and [19]. In Fig. 1(a) we show the first nine bands for a simple cubic arrangement of bubbles with volume fraction f = 0.1. The lowest

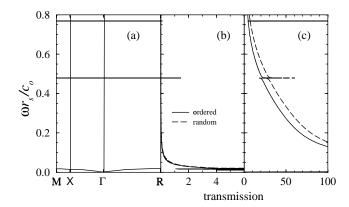


FIG. 1. (a): Dispersion relation along the MXΓR directions for a sc periodic composite consisting of air bubbles (of radius  $r_s$ ) in water.  $\omega$  is the frequency and  $c_o$  is the wave velocity in the water. Air volume fraction f = 10%. (b): Generalized transmission coefficient through a finite cluster consisting of 147 air bubbles in water in ordered (solid line) or random (dotted line) arrangement and volume fraction f = 10%. (c): The same as in panel (b) but with the multiple scattering switched off.

band is essentially confined between  $\omega = 0$  and  $\omega \approx \omega_0$ . The next three very flat and degenerate looking bands [19] at  $\omega r_s/c_o \approx 0.48$  arise from the p-type sharp scattering resonance (which can be viewed as approximate single bubble eigenmodes) through a tight-binding procedure [3]. The flatness of these bands is due to the fact that these approximate eigenmodes are confined mainly inside the bubble with a minute escape to the surrounding liquid. This is reflected in the very small width of the single bubble resonance indicating extremely weak radiation. The next five [19] bands at  $\omega r_s/c_o \approx 0.77$  arise from the d-type approximate eigenmodes and their flatness is due again to the minute escape of the field outside each bubble and the subsequent very weak coupling between neighboring bubbles. Our results in Fig. 1(a) are in agreement with those of Ref. [19] and they verify the impressive feature that the bubbly water with f = 0.1 is essentially impenetrable to sound waves of frequency  $\omega$ in the range  $\omega_o \leq \omega \leq 60\omega_o$  (with the exception of very narrow passing bands). From the lowest band (of  $\omega \leq \omega_0$ ) one can easily obtain both the phase velocity,  $c = \omega/k$ , and the group velocity,  $c_g = d\omega/dk$ . It should be pointed out that the long wavelength sound velocity can be obtained very easily and accurately by employing mean field theories, more specifically the so-called coherent potential approximation (CPA) [14,21]. The CPA replaces the actual random bubbly water by a uniform effective medium characterized by bulk modulus  $B_e$  and density  $\rho_e$  so that  $c = \sqrt{B_e/\rho_e}$ . These effective parameters are obtained by setting equal to zero the average scattering cross section resulting from the local replacement of the effective medium by actual configurations of the inhomogeneous system. The long wavelength limit of  $B_e$  coincides with the well-known Wood's law [14],  $B_e^{-1} = fB_i^{-1} + (1 - f)B_o^{-1}$  ( $B_i$ and  $B_o$  are the bulk moduli of the bubbles and the fluid, respectively). The long wavelength limit of  $\rho_e$  depends on which local actual configurations we use. We found that it is imperative to respect the topology and consider an air bubble surrounded by a shell of water, i.e., to employ the so-called "coated CPA" [14,21], which led to the following expression for the  $\rho_e$ :  $\rho_e = \rho_o [f(\rho_i - \rho_o) + 2\rho_i +$  $[\rho_o]/[2f(\rho_o-\rho_i)+2\rho_i+\rho_o]$ . Thus, the coated CPA gave c = 468, 43, and 35 m/s for the Silberman and Hall values of  $f = 5.84 \times 10^{-4}$ , f = 0.1, and f = 0.2, respectively [18]. This dramatic slowdown of the sound propagation in bubbly water is due to the fact that the restoring pressure is mainly determined by the bulk modulus of air, while the inertia is determined mainly by the density of water, as shown from the above formulas for  $B_e$  and  $\rho_e$ . In Fig. 2 we plot our results for c vs f which are in agreement with each other and with the experimental and computational data presented in Ref. [18], thus reinforcing confidence in our multiple scattering approach. The vanishing of the group velocity,  $d\omega/dk$ , for  $\omega \approx \omega_o$  can be attributed to the strong resonance at  $\omega = \omega_0$  and the related long trapping of the wave around each bubble.

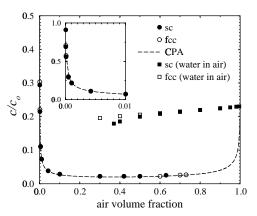


FIG. 2. Long wavelength sound velocity, c, for a system consisting of air bubbles in water as a function of the bubbles volume fraction. The opaque circles show the phase velocity for a sc lattice and the open circles for a fcc lattice. The dashed line shows a corresponding "coated CPA" result for a random system. The inset graph is an enlargement of the low volume fraction regime.  $c_o$  is the wave velocity in the water. For comparison, the sound speed in air containing suspended drops of water is shown (squares).

We turn now to novel results made possible by our method which takes fully into account the multiple scattering effects in a finite cluster of nonoverlapping spherical bubbles.

We have considered first a finite cluster of  $7 \times 7 \times 3 = 147$  periodically placed (in sc lattice) air bubbles embedded in an infinite mass of water. A point source emitting a spherical wave was placed very close to the  $7 \times 7$  face of the cluster. The energy flux vector,  $\mathbf{J}$ , was calculated behind the cluster in the far field region and along the line perpendicular to the  $7 \times 7$  faces and passing through its center (and through the point source). Assuming a time dependence of the form  $e^{-i\omega t}$ ,  $\mathbf{J}$  is given in terms of the pressure field,  $p(\mathbf{r})$ , by the formula

$$\mathbf{J} = -\frac{1}{2\rho\omega} \operatorname{Im}[p^*(\nabla p)]. \tag{1}$$

Following Ref. [22], we define a generalized transmission coefficient, T, as the ratio  $J/J_0$  of the energy flux with and without the cluster of air bubbles.

We repeated the above calculation by placing the 147 bubbles in a random but not overlapping way and within the boundaries of the same imaginary rectangular box as in the case above. We allowed also the radius of each bubble to be random ranging from  $0.75r_s$  to  $1.25r_s$ . In Fig. 1(b) we show results for T vs  $\omega$  for the ordered and the random (with common radius) cases. The ordered results are consistent with the band structure since the  $T \approx 0$  frequency regions practically coincide with the gaps. This shows that three finite size layers of bubbles approximate acceptably the infinite periodic system. Disordering the position of the bubbles does not change the results appreciably showing that a relatively thin wall of randomly placed bubbles with  $f \approx 0.1$  is enough to

inhibit sound propagation except at low frequencies ( $\omega \lesssim \omega_o$ ) and possibly at very narrow windows at  $\omega r_s/c_o \approx 0.48$ , 0.77, etc. In the panel of Fig. 1(c) we recalculated the transmission T neglecting altogether the multiple scattering effects. The difference from panel 1(b) is dramatic both in size and in the width around the main resonance. This proves how important the interference of multiple scattered waves is.

The most significant and novel results we obtained are shown in the panel of Fig. 3(b) corresponding to f = 0.01. We see again in this case a narrow acoustic band extending a little beyond the frequency of the strong resonance which gives rise to the maximum transmission. Then a wide gap follows [Fig. 3(a)] which corresponds to  $T \approx 0$  transmission for both the ordered and disordered cases [Fig. 3(b)]. It is impressive that this gap survives almost intact the positional disorder. When, in addition, we allowed the radius of each bubble to vary randomly between 0.75 to 1.25 of  $r_s$  the gap still survived although it was reduced by about 20% mostly from the upper side [see Fig. 3(b)]. Note that this gap is due entirely to multiple scattering since it disappears when multiple scattering is switched off [Fig. 3(c)]. Above the gap, a complex wide band appears dominated by branches with an  $\omega/k$  slope close to  $c_o$ ; this shows that the propagation for  $\omega r_s/c_o \gtrsim 0.2$  takes place mostly through the water with the air bubbles playing a less important role except that of superimposing (with very weak hybridization) very flat bands at the frequencies of the higher resonances.

In the case of 1% volume fraction there are noticeable differences in the transmission through the ordered and the disordered clusters: There is higher transmission in the disordered case than in the ordered one (i) at the upper part of the gap due probably to a tail of localized eigenstates, and (ii) around Bragg points, where the disorder tends to diminish destructive interference. In the rest of the spectrum the disorder decreases the transmission as expected.

In the present treatment we have omitted surface tension, thermal and viscous absorption, and the nonlinear effects.

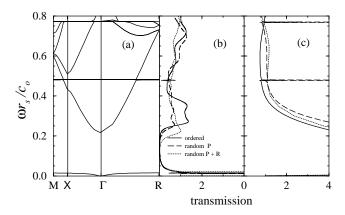


FIG. 3. The same as in Fig. 1, but with air volume fraction f=1%, and with both positional (P) and bubble radius (R) disorder (dotted line) in panels (b) and (c).

Surface tension becomes important only for very small size bubbles ( $r_s \leq 0.1$  mm); thermal and viscous absorption are important for small bubbles but tend to become insignificant for larger bubbles ( $r_s \geq 1$  cm) [17]. In any case these effects can be easily incorporated within our multiple scattering scheme. Nonlinear effects on the other hand pose severe difficulties, however, they are less important in a multiple bubble system than in a single bubble [18].

In conclusion, by applying our multiple scattering scheme we were able to obtain, among other results, the sound transmission through a cloud of randomly placed and of random size spherical bubbles in liquids. The spectral gaps are entirely due to multiple scattering and surprisingly survive in the presence of disorder. These results, besides their relevance to the water acoustic community, have wider interest due to their direct connection to the fundamental problem of Anderson localization.

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- [1] M. Kafesaki and E. N. Economou, Phys. Rev. B **60**, 11 993 (1999).
- [2] J. Korringa, Physica (Utrecht) 13, 392 (1947); W. Kohn and N. Rostoker, Phys. Rev. 94, 1111 (1951).
- [3] N.W. Ashcroft and N.D. Mermin, *Solid State Physics* (Holt, Rinehart and Winston, Philadelphia, 1976), pp. 202–206 and 176–190.
- [4] J. Liu, L. Ye, D. A. Weitz, and P. Sheng, Phys. Rev. Lett. 65, 2602 (1990).
- [5] M. M. Sigalas and E. N. Economou, J. Sound Vib. 158, 377 (1992); Solid State Commun. 86, 141 (1993).
- [6] E. N. Economou and M. M. Sigalas, in *Photonic Band Gaps and Localization*, edited by C. M. Soukoulis (Plenum, New York, 1993), pp. 317–338; J. Acoust. Soc. Am. 95, 1734 (1994).
- [7] J. O. Vasseur, B. Djafari-Rouhani, L. Dobrzynski, M. S. Kushwaha, and P. Halevi, J. Phys. Condens. Matter 6, 8759 (1994).
- [8] M. S. Kushwaha, P. Halevi, G. Martinez, L. Dobrzynski, and B. Djafari-Rouhani, Phys. Rev. B 49, 2313 (1994);
  M. S. Kushwaha, B. Djafari-Rouhani, L. Dobrzynski, and J. O. Vasseur, Eur. Phys. J. B 3, 155 (1998).
- [9] M. Kafesaki, M.M. Sigalas, and E.N. Economou, Solid State Commun. 96, 285 (1995).
- [10] M. Kafesaki, E. N. Economou, and M. M. Sigalas, in *Photonic Band Gap Materials*, edited by C. M. Soukoulis (Kluwer Academic Publishers, Dordrecht, 1996), pp. 143–164.
- [11] J. V. Sanchez-Perez et al., Phys. Rev. Lett. 80, 5325 (1998).
- [12] D. O. Riese and G. H. Wegdam, Phys. Rev. Lett. **82**, 1676 (1999).
- [13] M. Kafesaki and E. N. Economou, Phys. Rev. B 52, 13317 (1995).

- [14] P. Sheng, Introduction to Wave Scattering, Localization and Mesoscopic Phenomena (Academic Press, San Diego, 1995).
- [15] F. R. Montero de Espinosa, E. Jiménez, and M. Torres, Phys. Rev. Lett. 80, 1208 (1998); M. Torres, F. R. Montero de Espinosa, D. García-Pablos, and N. García, Phys. Rev. Lett. 82, 3054 (1999); D. García-Pablos, M. Sigalas, F. R. Montero de Espinosa, M. Torres, and N. García (to be published).
- [16] M. Kafesaki and E. N. Economou, Europhys. Lett. 37, 7 (1997); Ann. Phys. (Leipzig) 7, 383 (1998).
- [17] See, e.g., T. G. Leighton, *The Acoustic Bubble* (Academic Press, London, 1994).

- [18] A. A. Ruffa, J. Acoust. Soc. Am. 91, 1 (1992).
- [19] M. S. Kushwaha, B. Djafari-Rouhani, and L. Dobrzynski, Phys. Lett. A 248, 252 (1998).
- [20] The plane wave method presents some convergence difficulties for low f. This is the reason for a discrepancy between the density of states result shown in Fig. 1(c) of Ref. [5] and the position of the single scattering resonances.
- [21] C.M. Soukoulis, S. Datta, and E.N. Economou, Phys. Rev. B 49, 3800 (1994).
- [22] Lie-Ming Li and Zhao-Qing Zhang, Phys. Rev. B 58, 9587 (1998); Lie-Ming Li, Zhao-Qing Zhang, and Xiang-dong Zhang, Phys. Rev. B 58, 15589 (1998).