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# Classical wave propagation in periodic structures

E. N. Economou and A. Zdetsis
*Department of Physics, University of Crete and Research Center of Crete, P.O. Box 1527, 711 10 Heraklio, Crete, Greece*
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We considered a muffin-tin periodic potential ($-\delta$ inside the spheres and zero outside). Using the augmented-plane-wave method we systematically studied the location of gaps for positive energy and for various values of $\delta$ and the sphere radius $a$. Our results are applicable to the problem of classical-wave propagation in composite media and relevant to the problem of optical localization.

The question of classical-wave localization (CWL), in particular, light localization,[1,2] has received considerable attention recently both experimentally[3-8] and theoretically.[9-11] In spite of this, no proof exists that CWL does indeed take place in systems characterized by a positive definite random dielectric function. In a recent paper Soukoulis *et al.*[12] demonstrated numerically that CWL does take place in a lattice model. This numerical work, together with approximate calculations based on the coherent-potential approximation (CPA) and the potential-well analogy[13] (PWA), provided for the first time strong evidence that CWL is possible in a composite system consisting of spheres of radius $a$ and of dielectric constant $\epsilon_2$ embedded randomly into a medium of dielectric constant $\epsilon_1$ ($\epsilon_2 > \epsilon_1$). Drake and Genack[14] reported measurements recently of the optical diffusion coefficient in a system of close-packed titania spheres strongly suggesting that the critical regime very close to localization has been reached for the first time.

These recent developments indicate that a clear experimental demonstration of optical localization is imminent. As a result it is quite important to find out which are the optimum and/or the more easily realizable values of the parameters for achieving optical localization or CWL. The relevant parameters are the following: (a) The volume fraction $x$, i.e., the percentage of space occupied by the spheres; for periodic arrangement of the spheres (of equal size) $x$ can reach up to 74%, while for a random arrangement $x \leq 63.7\%$ provided that there is no overlap of the spheres; (b) the ratio $\mu \equiv \epsilon_2/\epsilon_1 \geq 1$; and (c) the frequency $\omega$ [in units of $c/a(\epsilon_1)^{1/2}$]. The dependence on the frequency is highly nonmonotonic because of resonances in the scattering from a single sphere (Mie resonances). For each spherical harmonic $l$ there is an infinite number of resonant frequencies $\omega_{ln}$ ($l=s,p,d,\ldots$ for $l=0,1,2,\ldots$; $n=1,2,3,\ldots$). The higher the ratio $\epsilon_2/\epsilon_1$, the more pronounced the resonances are. There is also a near degeneracy between $\omega_{l+2,n}$ and $\omega_{l,n+1}$. The numerical results and the CPA-PWA treatment show that the single-sphere resonances persist in strongly influencing transport quantities even for rather high values of $x$, which for the CPA-PWA approximate results can reach up to the close-packed limit.

It must be pointed out that the high-$x$ regime cannot be easily studied by the numerical technique of Ref. 12 because the range of this technique cannot exceed the second Mie resonance,[12] while for high $x$ the most important resonance seems to be the sixth. On the other hand, for the approximate CPA-PWA methods, which have given very interesting results in the high-$x$ regime,[12] we have no way of systematically estimating their accuracy.

Thus, there is a great need for an independent reliable way to check existing results and methods. We can reliably calculate wave-propagation quantities if the spheres are placed in a periodic arrangement such that their centers form a periodic lattice. If the wave is a scalar one, we can then employ one of the standard techniques for band structure in solids [such as the augmented-plane wave[15] (APW)] to calculate the position of the gaps and other relevant quantities. One may ask himself what is the connection between the gaps in a periodic system and the ranges of localized states in a random system. Although we cannot claim that there is complete coincidence between the two sets, nevertheless, we point out that both the gaps and the regions of localized states are due to the same physical mechanism, namely, destructive multiple scattering interference. The interconnection of gaps (in periodic systems) and regions of localized states (in random systems) becomes more obvious if we start from a periodic arrangement of the spheres and gradually introduce some disordering process regarding their positions. For weak disorder, it is well known that the bands will be practically unaffected by the disorder, while tails of localized states will be developed in the gaps. Thus, in this case of weak positional disorder, as described above, the regions of localized states will practically coincide with the positions of the gaps. Let us point out also that at very high $x$ (approaching the close-packed limit) there is very little room to move the spheres around out of their periodic position; thus, at high $x$ we are close to the above-mentioned weak positional disorder. We also point out that John and co-workers[16] have proposed the periodic arrangement of the spheres as a starting point followed by a weak disordering process in order to achieve optical localization in relatively low $x$. Their calculations, based on the Korringa-Kohn-Rostoker method,[17] gave an optimum $x$ around 0.12, in disagreement with earlier estimations.

In this paper we report results based on a systematic study of bands and gaps in periodic configurations of spheres (in the results reported here the periodic lattice was fcc; however, we made calculations for the bcc structure as well with similar results). We studied the elec-

 

tronic problem, where the potential inside each sphere is $-\delta$ ($\delta \geq 0$, and measured in units of $\hbar^2/2ma^2$, where $a$ is the radius of each sphere), the potential in the interstitial region between the spheres is taken as zero. The quantity $x$ is given by the ratio $a^3/r_{WS}^3$, where $r_{WS}$ is the so-called Wigner-Seitz radius. For fixed $\delta$ and $x$ (or equivalently $a$) we have scanned the positive energy $E$ (measured in units of $\hbar^2/2ma^2$) semiaxis to find the gap positions (parallel solid lines in Fig. 1). We have employed the APW technique. As is shown in Fig. 1, for a given $x$ (i.e., $a$) there is a strong dependence of our results on $\delta$, the depth of the potential well of each sphere. This behavior can be interpreted in terms of the resonant scatterings from a single sphere. These resonances are, in turn, associated with those values of $\delta$ for which a new bound state appears in the single potential well. These values of $\delta, \delta_{ln}$ are given as solutions of the equation $j_{l-1}(k_{ln}a)=0$ ($l \geq 1$) where $\delta_{ln}=\hbar^2 k_{ln}^2/2m$ and $j_l$ is the spherical Bessel function. For $l=0$, $k_{0n}a=(2n-1)\pi/2$. The explicit results are as follows: for $s$ waves ($l=0$), $\delta_{0n}=2.47$, 22.21, 61.69 for $n=1,2,3$, respectively; for $p$ waves ($l=1$), $\delta_{1n}=9.87$, 39.48, 88.83 for $n=1,2,3$, respectively; for $d$ waves ($l=2$), $\delta_{2n}=20.19$, 59.68 for $n=1,2$, respectively. These $\delta$ values are in close correspondence with the values of $\delta$ in Fig. 1, for which gaps appear for high positive values of the energy. An independent check of these resonance assignments can be made by omitting the corresponding partial wave in the APW calculation. We then find that the corresponding gap disappears, proving that the appearance of gaps for positive high energy is due to strong resonant scattering by each sphere. It is rather surprising that the single-sphere scattering is the dominant factor in determining the gap favoring values of $\delta$ even for values of $x$ so high as to approach the close-packed limit ($x \approx 0.74$). A possible explanation for this dominant role of the single-sphere scattering may be associated with its spherical geometry. Indeed, the spherical scatterers, as opposed, e.g., to the cubic scatterers, cannot form new well-connected shapes by clustering together. Thus, new cluster resonances cannot appear easily. This geometrical effect is also related with the fact that nonoverlapping spheres cannot form percolation channels even in the close-packed limit of $x \approx 0.74$, while for other geometries a percolation channel opens up for $x \approx 0.15$. It must be pointed out that the persistence of the dominant role of the single scatterer even for very high $x$ makes the CPA approach more reliable.

Our results for the muffin-tin electronic problem are directly applicable to the scalar-wave equation $\nabla^2 u + \omega^2 \epsilon u/c^2 = 0$ by observing that each $E, \delta$ point in Fig. 1 is mapped to an $\omega, \mu$ ($\equiv \epsilon_2/\epsilon_1$) point by the equations[12]

$$\mu = \frac{\delta}{E} + 1 , \tag{1a}$$

$$\frac{\omega^2 \epsilon_1 a^2}{c^2} = E/E_0 , \quad E_0 = \hbar^2/2ma^2 . \tag{1b}$$

Thus, the absolute threshold contrast $\mu$ for which a frequency gap (or CWL in the corresponding weak disorder case) just appears, is obtained from Fig. 1 by drawing the minimum slope straight line through the origin which intersects a gap segment [straight lines $s1$, $p1$, $p3$ in Figs.

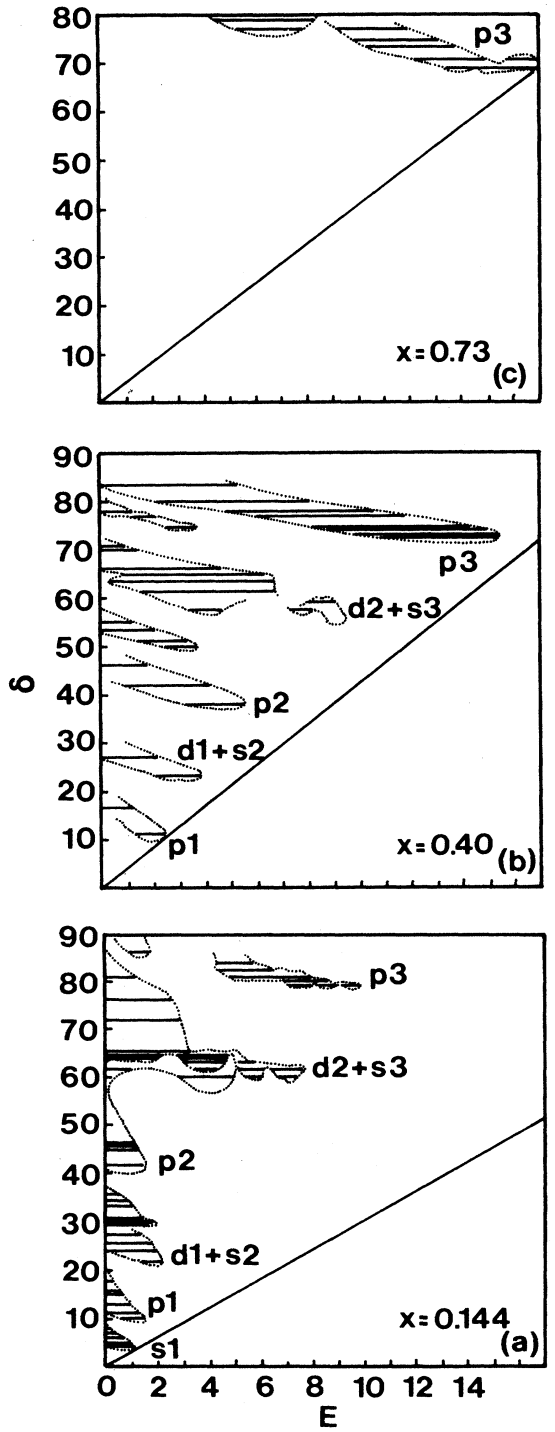


FIG. 1. Positions of the gaps (solid-horizontal lines) in an fcc muffin-tin periodic potential ($-\delta$ inside each sphere, zero in the interstitial region). The quantity $x$ is the volume fraction occupied by the spheres. The energy $E$ and the depth $\delta$ are measured in units of $\hbar^2/2ma^2$, where $a$ is the muffin-tin radius. The thin line passing through the ends of the horizontal lines is a guide to the eye, roughly indicating the trajectory(ies) of the band edges for positive $E$. The symbols indicate the dominant resonant scatterings responsible for the corresponding gap (see text).

1(a)–1(c) respectively]. We can also find from our data and Eq. (1a) the threshold value of $\mu$ for each particular resonance structure of Fig. 1 ($s1$, $p1$, $s2+d1$, $p2$, $s3+d2$, $p3$).[18] These various threshold values of $\mu,\mu_{ln}$, are plotted in Figs. 2(a) and 2(b) versus the volume-fraction $x$. For each $\mu_{ln}$ (with the possible exception of $\mu_{01}$) there is a threshold value of $x,x_{ln,t}$, above which the $ln$ gap appears for some finite value of $\mu$. As $x$ increases beyond $x_{ln,t}$, the threshold value of $\mu$ decreases and reaches a minimum, $\mu_{ln,m}$, for an optimum value of $x,x_{ln,0}$. As $x$ increases further beyond $x_{ln,0}$, $\mu_{ln}$ increases and approaches infinity for an upper cutoff value of $x,x_{ln,c}$. Thus the specific gap $ln$ appears only for $x_{ln,t} < x < x_{ln,c}$ and for $\mu > \mu_{ln}(x)$. For example, for the $p1$ gap $x_{11,t} \approx 0.1$ and $x_{11,c} \approx 0.5$; the optimum value $x_{11,0}$ is approximately 0.3 and the corresponding minimum value of $\mu_{11}$ is approximately 5. The $s1$ gap has an optimum value of $x \approx 0.14$ corresponding to $\mu_{01,m} \approx 4$, which is the absolute minimum value of $\mu$ for the appearance of any gap.

We see from Figs. 2(a) and 2(b) that the general tendency is for the lower gaps to appear and disappear for lower values of $x$. This is definitely true for the lowest gap, the $s1$, although for higher gaps the situation is more complicated. It should be pointed out that the $p3$ gap is the only one (up to $\delta \approx 100$) which persists beyond $x \approx 0.5$ all the way to the close-packed limit of $x \approx 0.74$. This gap also has a rather flat minimum of $\mu_{13}$ ($\mu_{13,m} \approx 5$) at $x_{13,0} \approx 0.5$ to 0.7.

If we compare our present reliable results for the gaps with our previous CPA-PWA results[12] for the localized segments we find a reassuring coincidence. (The CPA-PWA values for $x_{ln,0}$ are 0.2, 0.3, 0.35 vs 0.14, 0.3, 0.32 for the present case for the first three gaps; the corresponding values of $\mu_{ln,m}$ are 6.7, 6.2, 9 for the CPA-PWA vs 4, 4.8, 6.2 for the present case.) The main disagreement is with the $p3$ gap: The CPA-PWA $p3$ localized segment does not seem to survive beyond $x \approx 0.5$ in contrast to the present result where it extends to beyond $x \approx 0.74$. We have no explanation for this apparent discrepancy. We only note that the CPA-PWA for the electromagnetic case show that the localized segments due to the combined action of the $p2$ and $d2$ resonances persist all the way up $x \approx 0.75$.

In conclusion, we made a systematic study of the frequency spectrum (in particular, the position of the gaps) for a classical wave propagating in a periodic arrangement of spheres of radius $a$ and dielectric constant $\epsilon_2$ embedded in a medium of dielectric constant $\epsilon_1$. Besides the frequency $\omega$, the relevant parameters are the contrast $\mu = \epsilon_2/\epsilon_1$ and the volume fraction $x$ ($\equiv a^3/r_{WS}^3$). Our results for the gaps (the main features of which were presented here) are closely related to the localization regions in a disordered system resulting from the periodic one by randomizing the position of the spheres. This is true in particular for the very interesting case of the high-$x$ regime near the close-packed limit.

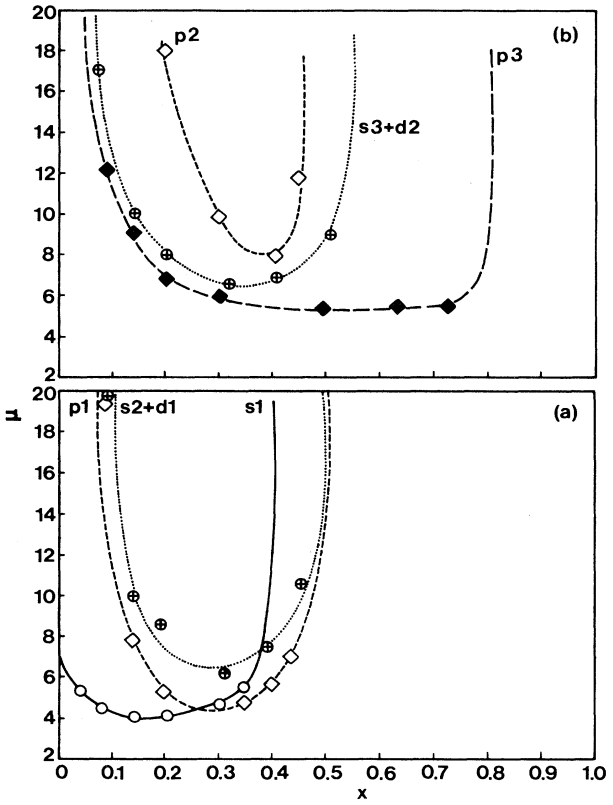


FIG. 2. Each curve represents the threshold value of the contrast $\mu$ for which a frequency gap of a specific type just opens up plotted against the volume fraction $x$. The type of the gap (and consequently the corresponding curve) is characterized by the dominant resonance (see text and Fig. 1) responsible for its existence.

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[1]S. John, Phys. Rev. Lett. **53**, 2169 (1984); Phys. Rev. B **31**, 304 (1985).

[2]P. W. Anderson, Philos. Mag. B **52**, 505 (1985).

[3]Y. Kuga and A. Ishimaru, J. Opt. Soc. Am. A **1**, 831 (1984).

[4]M. P. Van Albada and A. Lagendijk, Phys. Rev. Lett. **55**, 2692 (1985).

[5]P. E. Wolf and G. Maret, Phys. Rev. Lett. **55**, 2696 (1985).

[6]S. Etemad, R. Thompson, and M. J. Andrejco, Phys. Rev. Lett. **57**, 575 (1986).

[7]A. Z. Genack, Phys. Rev. Lett. **58**, 2059 (1987).

[8]M. Kaveh, M. Rosenbluh, I. Edvei, and I. Freund, Phys. Rev. Lett. **57**, 2049 (1986).

[9]Ping Sheng and Z. Q. Zhang, Phys. Rev. Lett. **57**, 1879 (1986).

[10]K. Arya, Z. B. Su, and J. L. Birman, Phys. Rev. Lett. **57**, 2725 (1986).

[11]C. A. Condat and T. R. Kirkpatrick, Phys. Rev. Lett. **58**, 226 (1987).

[12]C. M. Soukoulis, E. N. Economou, G. S. Grest, and M. H.

Cohen, Phys. Rev. Lett. **62**, 575 (1989); see also E. N. Economou and C. M. Soukoulis (unpublished).

[13]E. N. Economou, C. M. Soukoulis, and A. D. Zdetsis, Phys. Rev. B **30**, 1686 (1984); **31**, 6483 (1985); E. N. Economou, C. M. Soukoulis, M. H. Cohen, and A. D. Zdetsis, *ibid.* **31**, 6172 (1985); E. N. Economou, *ibid.* **31**, 7710 (1985).

[14]J. M. Drake and A. Z. Genack (unpublished).

[15]See, e.g., D. A. Papaconstantopoulos, *Handbook of the Band Structure of Elemental Solids* (Plenum, New York, 1986).

[16]S. John, Phys. Rev. Lett. **58**, 2486 (1987); S. John and R. Rangavajan, Phys. Rev. B **38**, 10101 (1988).

[17]J. Korringa, Physica **13**, 392 (1947); W. Kohn and N. Rostoker, Phys. Rev. **94**, 1111 (1954).

[18]We note also that our data seem to indicate [see, e.g. the $d2+s3$ gap structure in Fig. 1(b)] that there may be two threshold values of $\mu$ for the same type of gap, i.e., a lower threshold value of $\mu$ for which this specific gap opens up for the first time followed by a higher value of $\mu$ for which this gap disappears; and then a higher threshold value of $\mu$ for which this gap opens up again. Such a behavior corresponds to the existence of disconnected closed loops in the band edge trajectory(ies). Our data, although indicative of the existence of such behavior, are not so detailed as to establish it beyond any doubt.