PHYSICAL REVIEW B VOLUME 38, NUMBER 14 15 NOVEMBER 1988-I

Optimal structures for classical wave localization: An alternative to the Ioffe-Regel criterion

Sajeev John and Raghavan Rangarajan*
Joseph Henry Laboratories of Physics, Jadwin Hall, Princeton University, Princeton, New Jersey 08544
(Received 30 June 1988)

Using the Korringa-Kohn-Rostoker method we compute the band structure for a classical scalar wave scattering from a periodic array of dielectric spheres in a uniform background. The optimal volume filling fraction f of spheres for the creation of a total gap in the density of states, and hence localization, is found to be approximately 11%. This gap persists for refractive index ratios as small as 2.8.

The static structure factor of a disordered medium plays a crucial role in the determination of transport properties of waves in strongly scattering media.[1,2] For weakly scattering media, defined by the criterion that the classical elastic mean free path l is long compared to the wavelength λ, various approximation schemes adequately describe wave propagation in terms of classical diffusion on long length scales. These approximations, in general, associate no statistical correlation between scatterers at different positions. For optical propagation in relatively dilute or low dielectric contrast microstructures, this "white-noise" approximation to the disorder captures the essential physics of coherent multiple scattering on length scales long compared to l.

This approximation has been successfully applied to describe various correlation and fluctuation phenomena in the weak-scattering regime.[3-10] Among these are the optical analog of universal conductance fluctuations[3-5,10] observed in electronic systems and the intensity time autocorrelation function for light scattering from dilute mobile dielectric spheres.[6-10] Even in the absence of localization phenomena, it has been suggested that time autocorrelations may be used as a spectroscopic tool to probe the static and dynamic structure of complex fluids exhibiting multiple light scattering.[9]

The possibility of photon localization, however, requires that light propagate through a relatively dense collection of high dielectric constant scatterers of size comparable to the optical wavelength.[11] The first correction to the simple "white-noise" picture is to associate a form factor with individual scatterers but to let their positions remain essentially uncorrelated. This allows the possibility of single-scattering Mie resonances which can significantly reduce the elastic mean free path.[12,13] It is, nevertheless, problematic to achieve the criterion $2\pi l/\lambda \simeq 1$ for localization proposed by Ioffe and Regel[14] at densities sufficiently low that the scatterers do not become optically connected.

It has been recently suggested[1] that the resolution to this dilemma may be found by considering carefully prepared dielectric superlattice structures. Here localization may occur even for very weak disorder relative to the otherwise periodic structure. This is based on the fundamental assertion that the criterion for a mobility edge proposed by Ioffe and Regel[14] is in fact inapplicable in strongly correlated scattering media. Any significant perturbation of the photon density of states by the scattering medium will significantly alter the criterion for localization. This is most easily seen in the extreme case of a strong periodic modulation which creates a gap in the photon spectrum. At a band edge, light of wave vector $\mathbf{k}$ is Bragg scattered into $\mathbf{k}-\mathbf{G}$ where $\mathbf{G}$ is a reciprocal lattice vector of the medium. The resulting superposition is a standing wave. At frequencies ω slightly above the gap in the photon density of states $\rho(\omega)$, the wave reacquires a propagating character which is expressed by a long-wavelength envelope which modulates this standing wave. As ω approaches the band edge, the wavelength λ' of the envelope diverges. For sufficiently weak disorder that the gap persists, the criterion for localization is no longer $2\pi l/\lambda \simeq 1$ but rather $2\pi l/\lambda' \simeq 1$. It is highly plausible that there is continuous crossover of the localization criterion from one form to the other as the static structure factor $S(\mathbf{q})$ of the medium varies from being independent of wave vector $\mathbf{q}$ (white-noise model) to having sharp Bragg peaks (periodic superlattice).

From an experimental point of view it is, therefore, of particular interest to determine the optimal scattering structure for the creation of either a gap or a pseudogap in $\rho(\omega)$. The above argument suggests that any significant departure of the photon group velocity from its phase velocity can significantly enhance the prospect for the observation of a mobility edge.

We consider the scattering of a scalar classical wave from an fcc lattice of identical dielectric spheres of dielectric constant ϵ_a and radius R_s embedded in a uniform background dielectric with $\epsilon_b = 1$. The method discussed below employs a variation-iteration method developed independently by Korringa[15] and Kohn and Rostoker[16] (KKR) for computing the band structure of electrons in a periodic lattice. The wave equation for a classical scalar wave of amplitude $\phi(\mathbf{r})$ and frequency ω propagating in such a dielectric medium can be expressed in the form of a Schrödinger equation:

$$-\nabla^2\phi(\mathbf{r})+U(\mathbf{r})\phi(\mathbf{r})=\frac{\omega^2}{c^2}\epsilon_b\phi(\mathbf{r}) \tag{1a}$$

where

$$U(\mathbf{r}) = -\frac{\omega^2}{c^2}\sum_{\mathbf{R}}\epsilon_a(|\mathbf{r}-\mathbf{R}|) \tag{1b}$$

and

$$\epsilon_a(r) = \begin{cases} \epsilon_a - \epsilon_b, & r < R_s, \\ 0, & r \geq R_s. \end{cases}$$

Here $\mathbf{R}$ runs over all translation vectors of the lattice and c is the vacuum speed of "light."

Equation (1a) can be formally reexpressed in terms of the associated free-photon Green's function

$$\Gamma(\mathbf{r},\mathbf{r}') \equiv -\frac{1}{4\pi}\exp\left(\frac{ik_b|\mathbf{r}-\mathbf{r}'|}{|\mathbf{r}-\mathbf{r}'|}\right), \tag{2}$$

as a linear integral equation:

$$\phi(\mathbf{r}) = \int d^3\mathbf{r}'\Gamma(\mathbf{r},\mathbf{r}')U(\mathbf{r}')\phi(\mathbf{r}'). \tag{3}$$

Here $k_b \equiv [(\omega^2/c^2)\epsilon_b]^{1/2}$ and integration in (3) is over all space. Since Bloch's theorem requires that $\phi(\mathbf{r}+\mathbf{R}) = \exp(i\mathbf{k}\cdot\mathbf{R})\phi(\mathbf{r})$ for some photon "crystal momentum" $\mathbf{k}$, the integral equation can be reduced to one involving integration only over a single unit cell of the lattice

$$\phi(\mathbf{r}) = \int_{|\mathbf{r}|<R_s} d^3\mathbf{r}' G(\mathbf{r},\mathbf{r}')U(\mathbf{r}')\phi(\mathbf{r}') \tag{4}$$

and

$$G(\mathbf{r},\mathbf{r}') \equiv \sum_{\mathbf{R}}\Gamma(\mathbf{r},\mathbf{r}'+\mathbf{R})e^{i\mathbf{k}\cdot\mathbf{R}}. \tag{5}$$

Since the new Green's function G satisfies the Bloch condition, it is straightforward to verify that $\phi(\mathbf{r})$ does as well.

As discussed by KKR, the integral Eq. (4) may be derived from a variational principle. The solution of (4) corresponds to a stationary point of the functional

$$\Lambda[\phi] = \int_{|\mathbf{r}|<R_s}\phi^* U\phi - \int_{|\mathbf{r}'|<R_s}\int_{|\mathbf{r}|<R_s}\phi^*(\mathbf{r})U(\mathbf{r})G(\mathbf{r},\mathbf{r}')U(\mathbf{r}')\phi(\mathbf{r}'), \tag{6}$$

with respect to variations in $\phi(\mathbf{r}')$. As approximate solution may be obtained by introducing a trial wave function

$$\phi(\mathbf{r}) = \sum_{l=0}^{l_{\max}}\sum_{m=-l}^{l} i^l C_{lm} j_l(k_a r) Y_l^m(\theta,\phi), \tag{7}$$

where C_{lm} are undetermined expansion coefficients, $k_a \equiv [(\omega^2/c^2)\epsilon_a]^{1/2}$, j_l is a spherical Bessel function and Y_l^m is a spherical harmonic. Truncating this expansion at $l_{\max}=2$ yields a 9×9 determinantal condition on the "eigenvalues" $(\omega^2/c^2)\epsilon_b$ of Eq. (1a) for a fixed "crystal momentum" $\mathbf{k}$. This may be expressed as

$$\det\Lambda_{l,m;l',m'} = 0, \tag{8a}$$

where

$$\begin{aligned}\Lambda_{l,m;l',m'} = {} & [j_l(k_a r), j_l(k_b r)] \\ & \times \{B_{lm;l'm'}[j_{l'}(k_a r), j_{l'}(k_b r)] \\ & + k_b \delta_{ll'}\delta_{mm'}[j_l(k_a r), n_l(k_b r)]\}.\end{aligned} \tag{8b}$$

Here primes denote derivatives, n_l is a spherical Neumann function, and the square brackets denote the Wronskian evaluated on the surface of a scattering sphere:

$$[X,Y] \equiv \left(X\frac{dY}{dr} - Y\frac{dX}{dr}\right)_{r=R_s}. \tag{8c}$$

The coefficients $B_{l,m;l',m'}$ are determined for a given $\mathbf{k}$ and ω entirely by the static structure factor of the medium and are independent of the nature of the individual scatterers. They can be expressed in terms of a smaller number of independent structure constants D_{LM}:

$$B_{l,m;l'm'} = 4\pi\sum_{L,M} D_{L,M} C_{lm;l'm';LM}, \tag{9a}$$

where

$$C_{l,m;l'm';LM} = \int d\Omega\, Y_l^m Y_{l'}^{m'} Y_L^M. \tag{9b}$$

Convergence of the infinite lattice sums is facilitated by expressing D_{LM} as the sum of reciprocal lattice sum $D_{LM}^{(1)}$ and a real-space sum $D_{LM}^{(2)}$. Following Ham and Segall[17] we write $D_{LM} = D_{LM}^{(1)} + D_{LM}^{(2)} + \delta_{L,0}D_{00}^{(3)}$:

$$D_{LM}^{(1)} = -\frac{4\pi}{V}k_b^{-L}e^{-k_b^2/\eta}\sum_{\mathbf{G}}\frac{|\mathbf{G}+\mathbf{k}|^2\exp[-(\mathbf{G}+\mathbf{k})^2/\eta]}{(\mathbf{G}+\mathbf{k})^2-k_b^2}Y_L^M(\mathbf{G}+\mathbf{k}), \tag{10a}$$

where V is the volume of the unit cell, η is an arbitrary real positive convergence parameter (D_{LM} is in fact independent of η), and $\mathbf{G}$ runs over the entire reciprocal lattice:

$$D_{LM}^{(2)} = \frac{i^L(-2)^{L+1}}{\sqrt{\pi}k_b^L}\sum_{\mathbf{R}\neq 0}e^{i\mathbf{k}\cdot\mathbf{R}}R^L Y_L^M(\hat{R})\int_{\sqrt{\eta}/2}^{\infty}d\xi\,\xi^{2L}\exp\left(-R^2\xi^2+\frac{k_b^2}{4\xi^2}\right), \tag{10b}$$

and

$$D_{00}^{(3)} = \frac{-\sqrt{\eta}}{2\pi}\sum_{n=0}^{\infty}\frac{(k_b^2/\eta)^n}{n!(2n-1)}. \tag{10c}$$

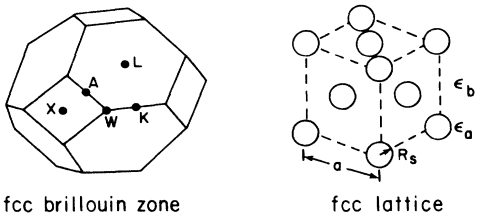

FIG. 1. Brillouin zone for an fcc lattice showing the extremal **k** points of the irreducible part: $L(0.5,0.5,0.5)$, $X=(1.0,0.0,0.0)$, $K=(0.75,0.75,0)$, $A=(1.0,0.25,0.25)$, and $W=(1.0,0.5,0.0)$. The triangles LKW and XAW subtend the region of the irreducible Brillouin zone. The above wave vectors are given in untis of $2\pi/a$ where a is the length of the side of the cube of the fcc lattice.

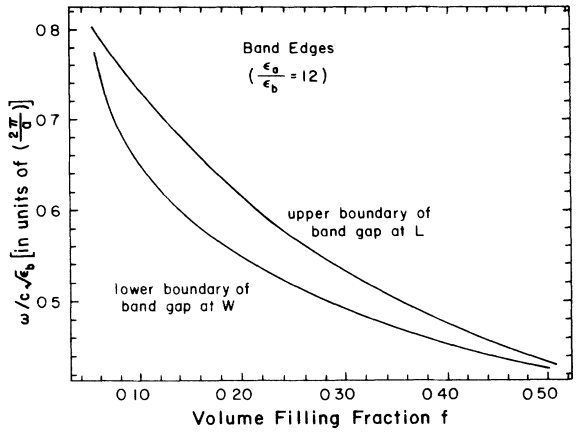

FIG. 3. Plot of the upper boundary of the band gap in $(\omega/c)\sqrt{\epsilon_b}$ at L and the lower boundary at W as a function of volume filling fraction f (in units of $2\pi/a$).

For the choice $\eta=0.6$, we found that convergence could be achieved by summing over 125 points in reciprocal space and 26 points in real space.[18]

Depicted in Fig. 1 is the first Brillouin zone for an fcc lattice. For the dielectric contrasts considered $6<\epsilon_a/\epsilon_b<12$ we find that the size of the photonic band gap is determined by the eigenvalue spectrum at points L and W, these corresponding to the extremal points of the zone surface. For fixed $\epsilon_a=12$ and $\epsilon_b=1$, the band structure for classical wave propagation was calculated for various values of the volume filling fraction f of spheres. At this large dielectric contrast a complete gap in the spectrum occurs over a wide range of f with a maximum occurring at $f\simeq 11\%$. This is depicted in Fig. 2. The band gap is expressed in terms of the "energy" eigenvalue $E\equiv(\omega^2/c^2)\epsilon_b$ of Eq. (1a) and is measured in units of $(2\pi/a)^2$ where a is the side length of the elementary cube of the fcc lattice (Fig. 1). In Fig. 3 we show the frequencies [in units of $(2\pi/a)$] for the upper and lower band edges as a function of f for $\epsilon_a/\epsilon_b=12$. Finally in Fig. 4 we show the computed ratio of the band gap in frequency to the center frequency of the gap. A maximum ratio of 13% is found at $f=0.15$. In all cases the lattice constant a was kept fixed and the radius R_s of the spheres was varied to change the volume filling fraction.

At the optimum volume-filling fraction $f=0.11$ the refractive index contrast was reduced until the gap disappeared. The band gap persists down to a ratio $(\epsilon_a/\epsilon_b)^{1/2}$ of 2.8. Since high refractive index materials of this nature are available[19] both in the microwave and optical regimes it is highly plausible that strong localization of photons may be observed in carefully prepared, weakly disordered arrays of scattering spheres. In general, large refractive index spheres may be fabricated from semiconducting materials with electron band gaps slightly larger than the energies of the relevant electromagnetic waves to be scattered. A trade-off is nevertheless required between large real part of the dielectric constant and an increasing imaginary part of the dielectric constant as the photon frequency enters the regime of the absorption edge of the semiconductor.

Application of these ideas to physical electromagnetic waves requires the generalization of the KKR technique to the case of a propagating vector field. It is likely that

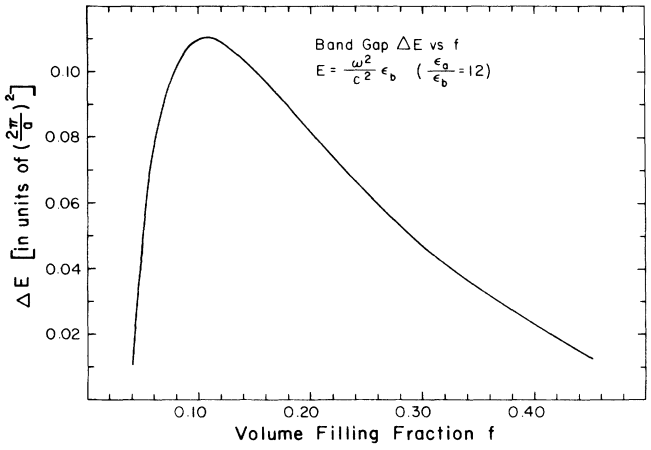

FIG. 2. Plot of the band gap in $(\omega^2/c^2)\epsilon_b$ in units of $(2\pi/a)^2$ as a function of volume filling fraction f. The largest gap occurs at $f\simeq 0.11$ $(\epsilon_a/\epsilon_b=12)$.

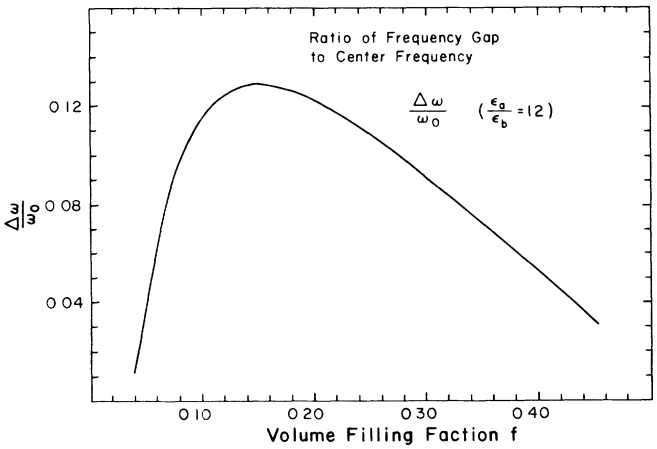

FIG. 4. Plot of the ratio of band gap $\Delta\omega$ in frequency to central frequency of band gap ω_0.

the existence of two polarization states will require a slightly higher refractive index ratio than that required to produce a band gap for a scalar wave.[1] However, the scalar wave calculations may provide a valuable guide in determining the optimal volume filling fraction. This conjecture is consistent with recent studies of photonic band structure by Yablonovitch using microwaves.[20]

Recently, Economou[21] has suggested that the photon band gap obtained for the periodic structure is in fact the remnant of a Mie resonance obtained for a single sphere. That is to say, although the single-scattering resonance is removed by the optical connectivity of the spheres it reappears as a perturbation on the total photon density of states. This argument suggests the existence of a number of higher frequency band gaps associated with each of the higher angular momentum Mie resonances of the single sphere. The precise volume-filling fraction required to optimize the size of these higher gaps and the robustness of these gaps with the introduction of disorder remain as important open questions. One immediate distinction between scalar waves and electromagnetic waves is the absence of an s-wave resonance due to the transverse nature of the electromagnetic wave. This may lead to an increase in the optimum volume-filling fraction from $(0.1 < f < 0.15)$ for the observation of the lowest-order band gap for true electromagnetic waves.

In summary, the existence of a nontrivial static structure factor of a disordered medium can have profound consequences on the nature of wave transport in it. Since the traditional Ioffe-Regel criterion is almost attainable for classical waves in an uncorrelated random medium, it is plausible that nearly any significant depression of the photon density of states from its effective medium value (obtained by single-scattering theory) will considerably enhance the prospects for the experimental observation of localization. The numerical results presented here strongly support the feasibility of producing the materials required for such an experiment as well as the need for further band-structure calculations in the case of physical electromagnetic waves.

We are grateful to Dr. B. Segall for providing some unpublished tables of the coefficients $C_{lm;l'm';LM}$ and to Dr. A. R. Williams for advice concerning computational methods. We are also grateful to Dr. E. Yablonovitch for communicating some of his experimental results on photonic band gaps prior to publication. This work was supported in part by the National Science Foundation under Grant No. DMR 8518163.

---

*Present address: Department of Physics, University of California, Santa Barbara, CA 91306.

[1] S. John, Phys. Rev. Lett. **58**, 2486 (1987).
[2] S. John, Comments Condens. Matter Phys. (to be published).
[3] S. Etemad, R. Thompson, and H. J. Andrejco, Phys. Rev. Lett. **57**, 575 (1986).
[4] M. Stephen and G. Cwilich, Phys. Rev. Lett. **59**, 285 (1987).
[5] S. Feng, C. Kane, P. A. Lee and A. D. Stone, Phys. Rev. Lett. **61**, 834 (1988).
[6] G. Maret and P. Wolf, Z. Phys. B **65**, 409 (1987).
[7] M. J. Stephen, Phys. Rev. B **37**, 1 (1988).
[8] I. Freund, M. Kaveh, and M. Rasenblush, Phys. Rev. Lett. **60**, 1130 (1988).
[9] D. J. Pine, D. A. Weitz, P. M. Chaikin, and E. Herbolzheimer, Phys. Rev. Lett. **60**, 1134 (1988).
[10] A. Z. Genack and M. Drake (unpublished).
[11] S. John, Phys. Rev. Lett. **53**, 2169 (1984).
[12] P. Sheng and Z. Zhang, Phys. Rev. Lett. **57**, 1879 (1986).
[13] K. Arya, Z. B. Su, and J. L. Birman, Phys. Rev. Lett. **57**, 2725 (1986).
[14] A. F. Ioffe and A. R. Regel, Prog. Semicon. **4**, 237 (1960).
[15] J. Korringa, Physica **13**, 392 (1947).
[16] W. Kohn and N. Rostoker, Phys. Rev. **94**, 1111 (1954).
[17] F. Ham and B. Segall, Phys. Rev. **124**, 1786 (1961).
[18] R. Rangarajan, undergraduate thesis, Princeton University, 1988 (unpublished).
[19] A. Z. Genack, Phys. Rev. Lett. **58**, 2043 (1987).
[20] E. Yablonovitch, Bull. Am. Phys. Soc. **33**, 421 (1988); Phys. Rev. Lett. **58**, 2059 (1987).
[21] E. N. Economou and C. M. Soukoulis (private communication).