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LETTER TO THE EDITOR

The Bergman spectrum of the effective dielectric constant in two-dimensional composite media

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Abstract. The Bergman spectrum of a two-dimensional composite with parallel cylinders immersed in the matrix was calculated and analysed. We found that the spectrum is discrete away from the percolation threshold and tends towards being continuous as the cylinders become closer and finally touch. We also calculated the effective dielectric constant and found that close to the percolation threshold the effective dielectric constant is scaled with an exponent that is half of the distance between nearest cylinder surfaces, and this was explained by a simple physical argument.

The Bergman spectrum is very important in studies of effective properties of composites; it reflects all of the microstructure properties of the composite considered. As proved by Bergman, the effective dielectric constant of a two-component composite can be written as a pole expansion of the following form:

$$\overline{\epsilon}/\epsilon_2 = 1 - F(s) \tag{1}$$

where $s = \epsilon_2/(\epsilon_2 - \epsilon_1)$ is a material parameter. The spectrum function $F(s)$ can be expressed as

$$F(s) = \sum_\alpha \frac{F_\alpha}{s - s_\alpha} \tag{2}$$

where the pair (s_n, F_n) are the poles and the weights of the spectrum function $F(s)$ respectively; it is called Bergman's spectrum and is solely determined by the microstructure of the composite [1]. Bergman's spectrum is not necessarily discrete; it can also be continuous. In the latter case the summation in formula (2) is replaced by an integral and the spectrum is given by the density of states. There are some general properties of the spectrum; for example, the sum of the weights F_α gives the volume fraction of medium 1, and for cubic-symmetry systems in 3D and square or hexagonal symmetry in 2D, the following sum rule holds:

$$\sum_\alpha s_\alpha F_\alpha = \frac{p(1-p)}{d} \tag{3}$$

where d is the dimensionality of the system. Other forms of spectrum functions can be defined when we change the rules for medium 1 and medium 2 as well as ϵ_1 and ϵ_2 [1]. Using the spectrum function, one can derive in a very general manner many useful results for the composite, including the bounds of the effective dielectric constant [2], the microstructure enhancement of the nonlinear optical coefficients [3] and the yield stress of electrorheological fluids [7]. If the effective dielectric constant as a function of ϵ_1 and ϵ_2 for a composite is known, then one can extract from it the corresponding Bergman spectrum, and use the spectrum in the calculation of other properties of interest. Recently, Ma *et al* used the spectrum extracted from different approximation formulae for the effective dielectric constant in a study of the enhancement of the third-order nonlinear optical coefficient, and fairly good results were obtained [3].

The calculation of Bergman's spectrum is difficult in general cases; in some special cases, it can be calculated from Bergman's formulation by a matrix diagonalization.

In an early publication [4], Bergman calculated the pole spectrum of a simple cubic array of spheres and discussed the behaviour of the spectrum of contacting spheres on the basis of his calculations. McPhedran and McKenzie, in an invited paper published in *Applied Physics* [5], calculated the pole spectra of square and hexagonal arrays of cylinders by means of a different formulation; they also derived from their calculations some general features of the spectrum.

In the following, we will use Bergman's formulation [6] to calculate the Bergman spectrum of a two-dimensional composite, which consists of spherical cylinders arranged in square lattices. We found that the spectrum gets denser as the cylinders are condensed, and that the spectrum may become continuous in the limit where nearest cylinders contact. We also found that the effective dielectric constant with $s = 0$ is scaled according to the separation between nearest cylinder surfaces with an exponent close to half of that separation, which can be explained by a simple physical argument.

Now we explain our calculation and analysis in detail. Consider a two-component composite with cylinders of dielectric constants ϵ_1 immersed, parallel, in a medium of dielectric constant ϵ_2, with the whole system placed in an infinite parallel-plate capacitor. The y-axis is chosen to be perpendicular to the electrodes. The electrostatic potential ϕ is given by the solution of the following boundary value problem:

$$\begin{aligned} &\nabla \cdot (\epsilon\, \nabla\phi) = 0 \\ &\phi\left(x, y = -\frac{L}{2}\right) = \frac{L}{2} E_0 \\ &\phi\left(x, y = \frac{L}{2}\right) = -\frac{L}{2} E_0 \end{aligned} \tag{4}$$

where

$$\epsilon = \epsilon_2 \left(1 - \frac{1}{s}\eta(\boldsymbol{r})\right) \tag{5}$$

and

$$s = \frac{\epsilon_2}{\epsilon_2 - \epsilon_1} \tag{6}$$

is the only material parameter of the system and $\eta(\boldsymbol{r})$ is the indicator function defined to be 1 in medium 1 and 0 otherwise. With the help of the Green Function $G(\boldsymbol{r}, \boldsymbol{r}')$ of Laplace's operator, equation (4) can be transformed into an integral equation:

$$\phi = z + \frac{1}{s}\hat{\Gamma}\phi. \tag{7}$$

where the applied field was taken to be -1 and the operator $\hat{\Gamma}$ defined as

$$\hat{\Gamma}\phi = \int \mathrm{d}\boldsymbol{r}'\ \eta(\boldsymbol{r}')\, \nabla' G(\boldsymbol{r}, \boldsymbol{r}') \cdot \nabla'\phi(\boldsymbol{r}') \tag{8}$$

which is a Hermitian operator with the following definition of the inner product:

$$\langle \phi, \psi \rangle = \int \mathrm{d}\boldsymbol{r}\ \eta(\boldsymbol{r}) \nabla\phi^* \cdot \nabla\psi. \tag{9}$$

The Green function in the limit where the system tends to infinity is

$$G(\boldsymbol{r},\boldsymbol{r}') = \frac{1}{2\pi}\frac{1}{|\boldsymbol{r}-\boldsymbol{r}'|}. \tag{10}$$

The effective dielectric constant can be expressed as

$$\bar{\epsilon} = \frac{1}{S}\int \mathrm{d}S\,\epsilon_2\left(1-\frac{1}{s}\theta(\boldsymbol{r})\right)\frac{\partial\phi}{\partial z} = \epsilon_2\left(1-\frac{F_n}{s-s_n}\right) \tag{11}$$

where S is the area of the cross section of the sample, s_n is the nth eigenvalue of the operator $\hat{\Gamma}$ and F_n is related to the nth eigenfunction ϕ_n of the operator $\hat{\Gamma}$ by $F_n = |\langle\phi_n|y\rangle|^2$. This is the Bergman representation of the effective dielectric constant.

Following Bergman [1], we use a scheme for expanding the potential in medium 1 in eigenfunctions of $\hat{\Gamma}$ for single domains. The single-domain eigenfunction is given by

$$\phi_{\boldsymbol{R}n} = \begin{cases} \dfrac{1}{\sqrt{n\pi}}\dfrac{|\boldsymbol{r}-R|^n}{a^n}\cos n\theta_{\boldsymbol{r}-R} & |\boldsymbol{r}-R| < a \\ \dfrac{1}{\sqrt{n\pi}}\dfrac{a^n}{|\boldsymbol{r}-R|^n}\cos n\theta_{\boldsymbol{r}-R} & |\boldsymbol{r}-R| > a \end{cases} \tag{12}$$

with the eigenvalue $s_n = 1/2$. The final result for a square lattice can be obtained by diagonalization of the following symmetric matrix:

$$\begin{aligned}\Gamma_{n,n'}(\boldsymbol{k}=0) &= \sum_{\boldsymbol{R}}\Gamma_{0n,\boldsymbol{R}n'} = s_{n'}\sum_{\boldsymbol{R}}\int \mathrm{d}\boldsymbol{r}\,\eta^{+}_{\boldsymbol{R}}\,\boldsymbol{\nabla}\phi_{\boldsymbol{R}n}(\boldsymbol{r}-R)\cdot\boldsymbol{\nabla}\phi_{\boldsymbol{R}'n'}(\boldsymbol{r}-R) \\ &= (-1)^{n'}s_{n'}\sqrt{nn'}\frac{(n+n'-1)!}{n!n'!}\sum_{\boldsymbol{R}}\cos(n+n')\theta_{\boldsymbol{R}}\frac{a^{n+n'}}{|\boldsymbol{R}|^{n+n'}}+\frac{1}{2}\delta_{n,n'}. \end{aligned} \tag{13}$$

The eigenvalue of the above matrix is the s_n that we needed and F_n is given by $F_n = a^2\pi|U_{n,1}|^2$, where $U_{n,m}$, $m = 1, 2, 3, \ldots$, is the nth eigenvector of the matrix $\boldsymbol{\Gamma}$.

When $n + n'$ is equal to its smallest possible value 2, the sum over $\boldsymbol{R}$ in equation (13) is only conditionally convergent; this sum can be evaluated by the Lorentz effective-medium method [8]. For a square lattice the result is

$$\sum_{\boldsymbol{R}\neq 0}\frac{\cos 2\theta_{\boldsymbol{R}}}{R^2} = \frac{\pi}{s} \tag{14}$$

where s is the area of the unit cell. When $n + n'$ is greater than 2, the sum is absolutely convergent and can be evaluated by the brute force method; however, we have used the Ewald summation method [9] to accelerate the convergence when $n + n'$ is small.

For an array of spherical cylinders arranged in a square lattice, we calculated and analysed the Bergman spectrum. The centre-to-centre distance between nearest cylinders in our study is $2a(1 + \delta)$, where a is the radius of the cylinders and δ is a positive number. The spectrum consists of discrete eigenvalues s_n and corresponding weights F_n, which for a typical case were as plotted in figure 1. In order to get converged results, we increased the order of the $\boldsymbol{\Gamma}$ matrix for every fixed value of δ until the calculated s_n with nonzero weight F_n converged. Further increase in the order of the $\boldsymbol{\Gamma}$ matrix will result in extra eigenvalues at position 1/2 with zero weight, i.e. the accumulation of poles at 1/2.

The accumulation of poles at 1/2 was also observed and discussed by Bergman in the calculation of the pole spectrum of arrays of spheres [4, 6]; the phenomenon is even more prominent here and can be understand by examining formula (12). We see that when $n \to \infty$, $\phi_{\boldsymbol{R}n}(\boldsymbol{r}) \to 0$ unless $\boldsymbol{r}$ is near the cylinder boundary, since the nearest cylinder surfaces are at least $2a\delta$ apart. The sum in (13) tends to zero when n is large, so $\Gamma_{nn'} = (1/2)\delta_{n,n'}$ when n is large enough.

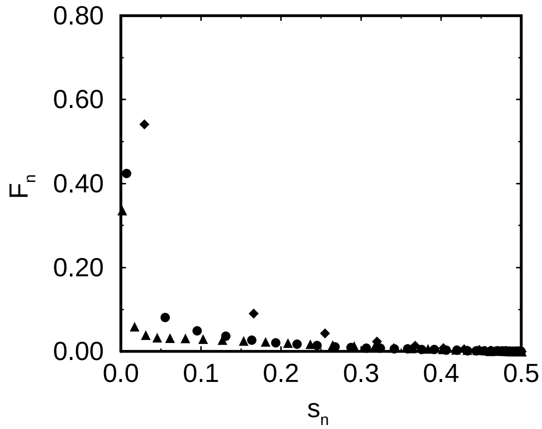

Figure 1. The Bergman spectrum s_n and F_n for different cylinder separations. $\delta = 10^{-2}$ (diamonds), 10^{-3} (circles), 10^{-4} (triangles).

The maximum order of the $\mathbf{\Gamma}$ matrix required to get converged results is sensitively dependent on the value of δ. For $\delta \sim 10^{-2}$, the maximum order of $\mathbf{\Gamma}$ is about 700, while for $\delta \sim 10^{-4}$, the maximum order of the $\mathbf{\Gamma}$ matrix can be as large as 2200. Further decrease of δ will cause the $\mathbf{\Gamma}$ matrix to become ill conditioned, and accurate results will be hard to obtain.

As shown in figure 1, the total number of poles with nonzero weight is increased when δ is decreased. We suspect that the spectrum will become continuous when the percolation threshold is reached; i.e., $\delta \to 0$ when the cylinders contact. There is a sharp corner of medium 2 at the contact point. The electric field is divergent at the corner, and the result is a continuous spectrum. The situation for the contacting cylinders can be compared to the two-dimensional checkerboard microgeometry, for which the effective dielectric constant is known exactly [10] and the corresponding spectrum is continuous. The lowest pole, s_1, has the largest weight for the square lattice that we studied here. The position of this pole tends to 0 when δ tends to 0; also, the weight of this pole decreases with δ. From (11) we see that

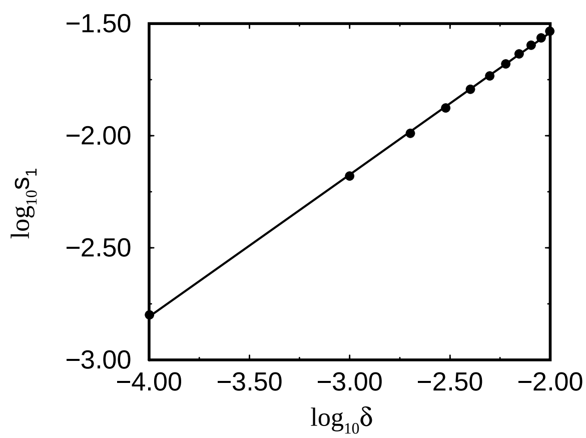

Figure 2. A log–log plot of the lowest eigenvalue s_1 as a function of δ. Points are from the calculation and the line is a straight-line fit.

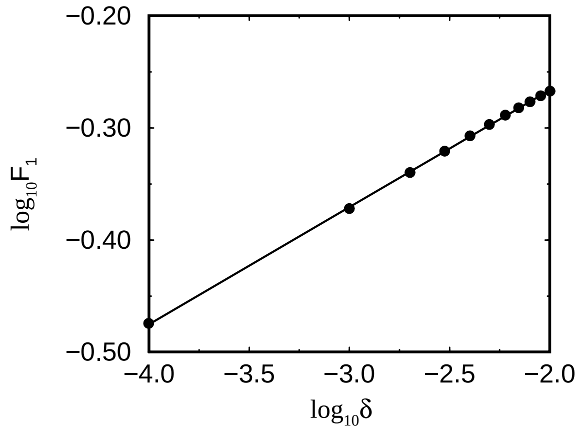

Figure 3. A log–log plot of the weight F_1 corresponding to the lowest eigenvalue s_1 as a function of δ. Points are from the calculation and the line is a straight-line fit.

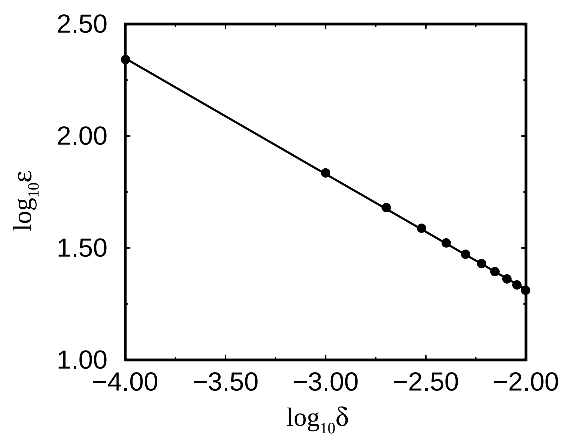

Figure 4. A log–log plot of the effective dielectric constant as a function of δ. Points are from the calculation and the line is a straight-line fit.

when $s = 0$, corresponding to $\epsilon_1 \to \infty$, the effective dielectric constant is dominated by the lowest pole s_1 and its weight F_1, i.e. $\overline{\epsilon}/\epsilon_2 \approx -F_1/s_1$. We systematically investigated s_1 and F_1 as functions of δ. For small enough δ, s_1 and F_1 scale as power laws with δ, as figure 2 and figure 3 indicate. The asymptotic behaviour of the pole position and its weight were fitted from calculated data, as $s_1 \approx 0.54\delta^{0.63}$ and $F_1 \approx 0.88\delta^{0.10}$. From these results, we estimate that the effective dielectric constant at $s = 0$ will be $\epsilon_1/\epsilon_2 \approx 1.6\delta^{-0.53}$. On the other hand, calculation of the effective dielectric constant at $s = 0$ using all of the poles gives a similar result. Figure 4 is a log–log plot of the effective dielectric constant at $s = 0$ as a function of δ. From the plot we get $\overline{\epsilon} = 1.9\delta^{-0.52}$. The exponents in the two cases are very close to $1/2$, which can be understood on the basis of the following argument. The capacitance of two infinitely long metal cylinders with radius a is given by [11]

$$\frac{1}{C} = 2\cosh^{-1}\left[\frac{c^2 - 2a^2}{2a^2}\right] \tag{15}$$

where c is the distance between the two cylinder centres. The smallest distance between the two cylinder surfaces $h = c - 2a$, while δ as defined above is given by $\delta = h/2a$. When h is small, we get

$$\frac{1}{C} = 2\cosh^{-1}\left[\frac{(2a+h)^2 - 2a^2}{2a^2}\right] = 4\sqrt{\frac{2h}{a}} + \mathrm{O}(h/a) = 8\delta^{1/2} + \mathrm{O}(\delta) \qquad (16)$$

which explains why the exponent of the effective dielectric constant at $s = 0$ is close to $1/2$.

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