

**Spectral representation of the effective dielectric constant of graded composites**

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We generalize the Bergman-Milton spectral representation, originally derived for a two-component composite, to extract the spectral density function for the effective dielectric constant of a graded composite. This work has been motivated by a recent study of the optical absorption spectrum of a graded metallic film [Huang and Yu, *Appl. Phys. Lett.* **85**, 94 (2004)] in which a broad surface-plasmon absorption band was shown to be responsible for enhanced nonlinear optical response and an attractive figure of merit. It turns out that, unlike in the case of homogeneous constituent components, the characteristic function of a graded composite is a continuous function because of the continuous variation of the dielectric function within the constituent components. Analytical generalization to three-dimensional graded composites is discussed, and numerical calculations for multilayer composites are given as a simple application.

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**I. INTRODUCTION**

In graded materials [1] the physical properties may vary continuously in space making them distinctly different from homogeneous materials. Hence composite media consisting of graded inclusions have attracted much interest in various engineering applications, such as reduced residual and thermal barrier coatings of high temperature components in gas turbines, surface hardening for tribological protection, and graded interlayers used in multilayered microelectronic and optoelectronic components [2–4].

Like graded materials, thin films are of great interest in many practical applications and often possess different optical properties in comparison to bulk materials [5]. In particular, and of interest from the point of view of this study, it was found in a recent experiment that graded thin films may have high relative dielectric permittivity as well as a flatter temperature characteristic of permittivity than single-layer films [6,7]. The traditional theories used to deal with homogeneous materials [8,9] are not adequate for the treatment of composites with graded inclusions. To treat them properly, we have recently developed a first-principles approach [10,11] and a differential effective dipole approximation [12].

This work has been motivated by a recent study of the optical absorption spectrum of a graded metallic film [13]. In that work, a broad surface plasmon absorption band was observed in addition to a strong Drude absorption peak at zero frequency. Such a broad absorption band has been shown to be responsible for the enhanced nonlinear optical response as well as an attractive figure of merit (the degree of optical absorption). Yuen *et al.* [14] pointed out that such an absorption spectrum, being related to the imaginary part of the ef-

fective dielectric constant, should be also reflected in the Bergman-Milton spectral representation of the effective dielectric constant [15,16].

The Bergman-Milton spectral representation was originally developed for calculating the effective dielectric constant and other response functions of two-component composites. However, in that representation both of the two components concerned are assumed to be homogeneous. It is therefore worth extending the spectral representation to graded composite materials. This work on graded films is an example of a more general graded composite in three dimensions. One of the main purposes of this work is to help to identify the physical origin of the broad absorption band. It turns out that, unlike in the case of homogeneous materials, the characteristic function of a graded composite is a continuous function due to the continuous variation of the dielectric function within the constituent component.

Moreover, we apply our theory to a special case of graded composites, namely multilayer materials. From the practical point of view, they are more convenient to fabricate than graded materials [17] and, in addition, there are many algorithms available for designing multilayer coatings [18,19]. The present work is necessary in the sense that we shall discuss the multilayer effects as the number of layers inside the material increases. In this regard, this work should be expected to have practical relevance. We will show that as the number of layers  $N$  increases, a gradual transition from sharp peaks to a broad continuous band occurs. The graded composite results are recovered at the limit of  $N \rightarrow \infty$ .

The rest of this paper is organized as follows. In Sec. II the general derivation of the spectral representation for graded composites is presented. In Sec. III we describe the

model and present analytical results for the spectral representation of the effective dielectric constant of a graded film with an interface, as well as a graded sphere. In those cases, the Bergman-Milton formalism has been modified for graded composites. We further obtain an analytic form for the spectral density function of a multilayer film and a multilayer sphere in Sec. IV. Numerical results are presented in Sec. V, and discussion and conclusions are given in Sec. VI.

## II. FORMALISM

We consider a two-component composite in which graded inclusions of dielectric constant  $\epsilon_1(\mathbf{r})$  are embedded in a homogeneous host medium of dielectric constant  $\epsilon_2$ . It is important to notice that the dielectric constant  $\epsilon_1(\mathbf{r})$  is a gradation profile as a function of position  $\mathbf{r}$ . We will restrict our discussion and calculations to the quasistatic approximation, i.e.,  $dc/\omega \leq 1$ , where  $d$  is the characteristic size of the inclusion,  $c$  is the speed of light in vacuum, and  $\omega$  is the frequency of the applied field. In the quasistatic approximation, the whole graded structure can be regarded with an effective (overall) linear dielectric constant defined as [20]

$$\epsilon_e = \frac{1}{V} \int \frac{\mathbf{E} \cdot \mathbf{D}}{E_0^2} dV, \quad (1)$$

where  $E_0$  is the applied electric field along the  $z$  direction,  $\mathbf{E}$  and  $\mathbf{D}$  are the local electric field and the local displacement, respectively.

The objective of the present section is to solve the Laplace equation

$$\nabla \cdot [\epsilon(\mathbf{r}) \nabla \phi(\mathbf{r})] = 0 \quad (2)$$

subject to the boundary condition  $\phi_0 = -E_0 z$ . The dielectric function  $\epsilon(\mathbf{r})$  in Eq. (2) varies from component to component but has a fixed mathematical form for a given component. It can be given as [15]

$$\epsilon(\mathbf{r}) = \epsilon_2 \left[ 1 - \frac{1}{s} \eta(\mathbf{r}) \right], \quad (3)$$

where  $s = [1 - \epsilon_{\text{ref}}/\epsilon_2]^{-1}$  is the material parameter and  $\epsilon_{\text{ref}}$  is some reference dielectric constant in the graded component. The characteristic function  $\eta(\mathbf{r})$  may be written in terms of a real function  $f(\mathbf{r})$  as

$$\eta(\mathbf{r}) = \begin{cases} 1 + f(\mathbf{r}) & \text{in inclusion} \\ 0 & \text{in host,} \end{cases}$$

which accords for the microstructure of graded composites. The function  $f(\mathbf{r})$  depends on the variation of the dielectric constant in the inclusion component. For a homogeneous constituent component [i.e.,  $f(\mathbf{r})=0$ ],  $\eta(\mathbf{r})=1$  in the inclusion component. For graded systems,  $\eta(\mathbf{r})$  can be a continuous function in the inclusion component due to the continuous variation of the dielectric function within it. Thus Eq. (2) can be solved

$$\phi(\mathbf{r}) = -E_0 z + \frac{1}{s} \int dV' \eta(\mathbf{r}') \nabla' G(\mathbf{r} - \mathbf{r}') \cdot \nabla' \phi(\mathbf{r}'), \quad (4)$$

where  $G(\mathbf{r} - \mathbf{r}')$  is a Green's function satisfying

$$\nabla^2 G(\mathbf{r} - \mathbf{r}') = -\delta^3(\mathbf{r} - \mathbf{r}') \quad \text{for } \mathbf{r} \text{ in } V,$$

$$G = 0 \quad \text{for } \mathbf{r} \text{ on the boundary.}$$

In order to obtain a solution for Eq. (2), we introduce an integral-differential Hermitian operator  $\hat{\Gamma}$ ,

$$\hat{\Gamma} \equiv \int dV' \eta(\mathbf{r}') \nabla' G(\mathbf{r} - \mathbf{r}') \cdot \nabla',$$

and define an inner product as

$$\langle \phi | \psi \rangle = \int dV \eta(\mathbf{r}) \nabla \phi^* \cdot \nabla \psi. \quad (5)$$

With the above definitions, Eq. (4) can be simplified to

$$\phi(\mathbf{r}) = -E_0 z + \frac{1}{s} \hat{\Gamma} \phi(\mathbf{r}).$$

Let  $s_n$  and  $|\phi_n\rangle$  be the  $n$ th eigenvalue and eigenfunction of the operator  $\hat{\Gamma}$ . Then, the generalized eigenvalue problem becomes

$$\nabla \cdot [\eta(\mathbf{r}) \nabla \phi_n] = s_n \nabla^2 \phi_n.$$

The potential  $|\phi\rangle$  can be expanded in series of eigenfunctions,

$$|\phi\rangle \equiv \sum_n \left( \frac{s}{s_n - s} \right) \frac{|\phi_n\rangle \langle \phi_n | z \rangle}{\langle \phi_n | \phi_n \rangle}, \quad (6)$$

where we choose  $E_0=1$  for convenience. Since  $\eta(\mathbf{r})$  is a real function, the eigenvalues  $s_n$  will be real. Also, for the graded component,  $\eta(\mathbf{r})$  is a continuous function, which will cover the full region, i.e.,  $-\infty \leq \eta(\mathbf{r}) \leq \infty$ . Therefore the eigenvalues  $s_n$ , which depend on the continuously graded microstructure  $\eta(\mathbf{r})$ , do not lie within the interval  $[0, 1]$  but extend to  $-\infty \leq s_n \leq \infty$  as first pointed by Gu and Gong [21] for the case of three-component composites. However, eigenvalues  $s_n$  still lie in  $[0, 1]$  for  $0 \leq \eta(\mathbf{r}) \leq 1$ .

We are now in the position to find an analytical representation for the effective dielectric constant  $\epsilon_e$  in Eq. (1). We take advantage of Green's theorem, the boundary condition  $\phi_0 = -z$ , and the Maxwell equation  $\nabla \cdot \mathbf{D} = 0$  to obtain the effective dielectric constant,

$$\begin{aligned} \frac{\epsilon_e}{\epsilon_2} &= \frac{1}{\epsilon_2 V} \int (-\nabla \phi) \cdot \mathbf{D} dV \\ &= \frac{-1}{V} \int \hat{\mathbf{z}} \cdot \left[ \left( 1 - \frac{1}{s} \eta(\mathbf{r}) \right) \nabla \phi \right] dV = 1 + \frac{1}{sV} \langle z | \phi \rangle. \end{aligned} \quad (7)$$

If we now introduce the reduced response [15]

$$F(s) = 1 - \frac{\epsilon_e}{\epsilon_2}, \quad (8)$$

and substitute Eq. (6) into Eq. (7) we find

$$F(s) = \frac{1}{V} \sum_n \frac{|\langle z | \phi_n \rangle|^2}{\langle \phi_n | \phi_n \rangle} \left( \frac{1}{s - s_n} \right).$$

We can now express the effective dielectric constant as

$$\epsilon_e = \epsilon_2 \left( 1 - \sum_n \frac{f_n}{s - s_n} \right), \quad (9)$$

where  $f_n$  is given by

$$f_n = \frac{1}{V} \frac{|\langle z | \phi_n \rangle|^2}{\langle \phi_n | \phi_n \rangle}.$$

Using the above equations, we obtain the following sum rule:

$$\sum_n f_n = \frac{1}{V} \langle z | z \rangle = \frac{1}{V} \int dV \eta(\mathbf{r}) \nabla z \cdot \nabla z = \frac{1}{V} \int dV \eta(\mathbf{r}). \quad (10)$$

It is worth noting that the sum rule will not equal the volume fraction of the inclusion. This is different from the Bergman-Milton spectral representation for two homogeneous systems, in which the sum rule equals the volume fraction of the inclusion.

When the operator  $\hat{\Gamma}$  has a continuous spectrum, Eq. (9) should be replaced with the integral form

$$\epsilon_e = \epsilon_2 \left( 1 - \int ds' \frac{m(s')}{s - s'} \right), \quad (11)$$

where  $m(s')$  is the spectral density function. Then, the reduced response [Eq. (8)] becomes

$$F(s) = \int ds' \frac{m(s')}{s - s'}. \quad (12)$$

If we write  $s$  as  $s + i0^+$ , the right side of Eq. (12) becomes

$$\mathcal{P} \int ds' \frac{m(s')}{s - s'} - i\pi m(s),$$

and thus  $m(s')$  is given through the limiting process

$$m(s') = -\frac{1}{\pi} \text{Im}[F(s' + i0^+)]. \quad (13)$$

This final result is identical in form to Bergman's expression for the analogous function in scalar composite materials. However, there are differences in the derivation, namely, the definition of the inner product Eq. (5), the continuous graded microstructure  $\eta(\mathbf{r})$ , the sum rule, as well as the range of eigenvalues  $s_n$ .

From Eq. (11) it is evident that if the spectral density function  $m(s')$  is known, the effective dielectric constant can be obtained accurately, and vice versa. The spectral representation has been used to analyze the effective dielectric properties of composites. Recently, Levy and Bergman [22] also used it in their study of nonlinear optical susceptibility. In this regard, Sheng and co-workers [23] developed a practical algorithm for calculating the effective dielectric constants based on the spectral representation. In what follows, we

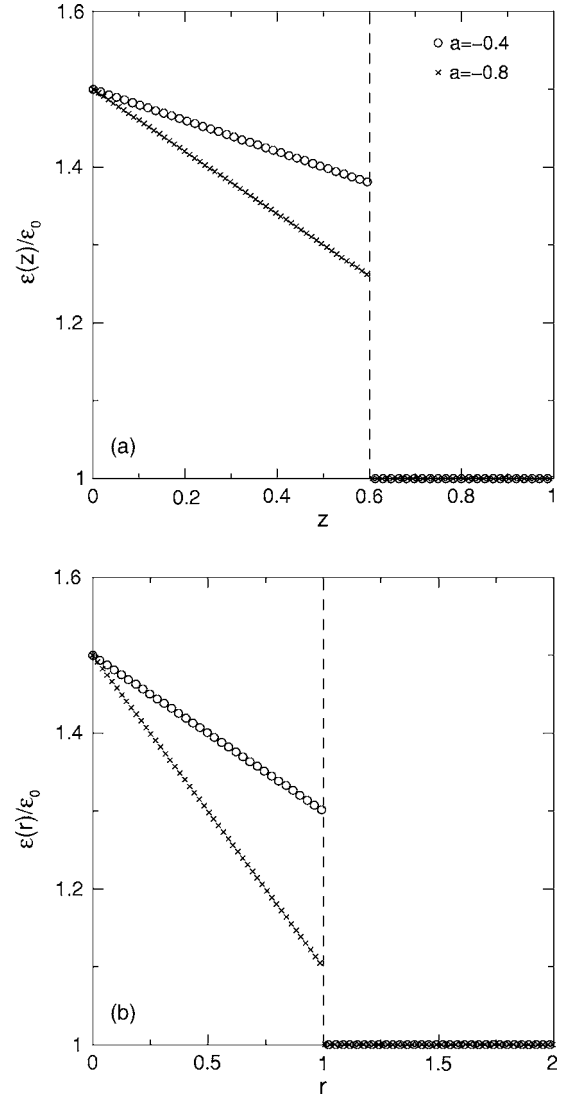


FIG. 1. (a) Dielectric profiles of two different graded films with an interface at  $h=0.6$ . (b) Dielectric profiles of two graded spheres with unit radius. Parameters used: The dielectric constant of the host medium is set to  $\epsilon_2=1$  and the material parameter to  $s=-2$ .

restrict ourselves to graded composites in one and three dimensions, and the corresponding multilayer composites.

### III. SPECTRAL DENSITY FUNCTION OF GRADED COMPOSITES

#### A. Spectral density function of a graded film

We consider a graded dielectric film of width  $L$ , in which two media meet at a planar interface as shown in Fig. 1(a). The first medium  $\epsilon_1(z)$  varies along the  $z$  axis, while the second medium  $\epsilon_2$  is homogeneous. We define the graded microstructure as

$$\eta(z) = \begin{cases} 1 + az, & 0 < z \leq h, \\ 0, & h < z < L, \end{cases} \quad (14)$$

where  $a$  and  $h$  are real constants. They can be varied to describe different graded films. Thus, according to Eq. (3),

the dielectric function of graded film can be expressed as

$$\epsilon(z) = \epsilon_2 \left( 1 - \frac{\eta(z)}{s} \right). \quad (15)$$

Owing to the simple geometry of a graded film, we can use the equivalent capacitance of a series combination to obtain the effective dielectric constant as

$$\frac{1}{\epsilon_e} = \frac{1}{L} \int_0^L \frac{1}{\epsilon(z)} dz. \quad (16)$$

Substituting Eqs. (14) and (15) into Eq. (16), we obtain

$$\frac{1}{\epsilon_e} = \frac{1-h}{\epsilon_2} + \frac{s \left[ \ln \left( 1 - \frac{\eta(0)}{s} \right) - \ln \left( 1 - \frac{\eta(h)}{s} \right) \right]}{a \epsilon_2},$$

with the assumption  $L=1$ .

We are now in a position to extend the Bergman-Milton spectral representation of the effective dielectric constant [15,16] to a graded film. For a graded system,  $\eta(z)$  can be a continuous function in the inclusion medium. Using Eqs. (11)–(13), we obtain the spectral density function for a graded film as

$$m(s') = - \frac{as' \arg \left( \frac{s-1}{s-ah-1} \right)}{\pi \left\{ \left[ s' \arg \left( \frac{s-1}{s-ah-1} \right) \right]^2 + \left[ a(h-1) - s' \ln \left( \frac{s'-1}{s'-ah-1} \right) \right]^2 \right\}},$$

where  $s' = \text{Re}[s]$  and  $\arg[\dots]$  denotes the arguments of complex functions.

### B. Spectral density function of a graded sphere

The above theory can be generalized to graded composites in three dimensions. We consider a graded sphere with dielectric constant  $\epsilon_1(r)$  embedded into a homogeneous host medium with dielectric constant  $\epsilon_2$ . The dielectric constant of the graded sphere  $\epsilon_1(r)$  varies along the radius  $r$ . We can obtain the effective dielectric constant of a graded sphere using the spectral representation. We consider the graded microstructure by using the characteristic function as

$$\eta(r) = \begin{cases} 1 + ar, & 0 < r \leq R, \\ 0, & r > R, \end{cases}$$

where  $R$  is the radius of the graded sphere. Thus from Eq. (3) the dielectric constant in the graded sphere is given by

$$\epsilon(r) = \epsilon_2 \left( 1 - \frac{\eta(r)}{s} \right). \quad (17)$$

In the dilute limit the effective dielectric constant of a small volume fraction  $p$  of graded spheres embedded in a host medium is given by [24,25]

$$\epsilon_e = \epsilon_2 + 3\epsilon_2 pb, \quad (18)$$

where  $b$  is the dipole factor of graded spheres embedded in a host as given in Ref. [10]. Using Eqs. (8) and (18), the reduced response can be obtained as

$$F(s) = -3\epsilon_2 pb. \quad (19)$$

Thus the spectral density function of a graded sphere can be given through a numerical evaluation of Eq. (13).

### IV. SPECTRAL DENSITY FUNCTION OF MULTILAYER COMPOSITES

A multilayer composite is a special case of graded composites. The gradation becomes continuous as the number of layers approaches infinity. To investigate the multilayer effect, we use a finite difference approximation for the graded profile [Eqs. (15) and (17)] for a finite number of layers. To mimic a multilayer system, we divide the interval  $[0, 1]$  into  $N$  equally spaced subintervals,  $[0, z_1], (z_1, z_2], \dots, (z_{N-1}, 1]$ . Then we adopt the midpoint value of  $\epsilon(z)$  for each subinterval as the dielectric constant of that sublayer. In this way, we calculate the effective dielectric constant, eigenvalues, as well as the spectral density function for each  $N$ . It is worth noting that as  $N \rightarrow \infty$  the results for graded composites are recovered.

In addition to multilayer films, we can use the above approach to study the much simpler problem of a two-layer film. In this system, we have two layers of dielectric constants  $\epsilon_1$  and  $\epsilon_2$ , and host  $\epsilon_0$ . The thicknesses are  $hy$ ,  $h(1-y)$ , and  $1-h$ , respectively, where  $y$  is the length ratio between component  $\epsilon_1$  and component  $\epsilon_2$ . We also define two microstructure parameters,  $\eta_1$  and  $\eta_2$ . If we let  $s = 1/(1 - \epsilon_1/\epsilon_0)$ , then  $\eta_1 = 1$ , and  $\eta_2 = (\epsilon_0 - \epsilon_2)/(\epsilon_0 - \epsilon_1)$ . According to Eq. (15), the effective dielectric constant of the two-layer film is now given by

$$\frac{1}{\epsilon_e} = \frac{hy}{\epsilon_1} + \frac{h(1-y)}{\epsilon_2} + \frac{1-h}{\epsilon_0}.$$

According to Eq. (8), the reduced response can be given by

$$F(s) = \frac{F_1}{s-s_1} + \frac{F_2}{s-s_2}, \quad (20)$$

where

$$F_1 = \frac{h[s_1(y-y\eta+\eta)-\eta]}{s_1-s_2},$$

$$F_2 = -\frac{h[s_2(y-y\eta+\eta)-\eta]}{s_1-s_2},$$

$$s_1 = \frac{1}{2}\{1 - h(y-y\eta+\eta) + \eta - \sqrt{4\eta(-1+h) + [1 - h(y-y\eta+\eta) + \eta]^2}\},$$

$$s_2 = \frac{1}{2}\{1 - h(y-y\eta+\eta) + \eta + \sqrt{4\eta(-1+h) + [1 - h(y-y\eta+\eta) + \eta]^2}\}.$$

From the sums of  $F_1$  and  $F_2$ , and the integral of the graded microstructure  $\eta(z)$  given by Eq. (14), we can check that the sum rule expressed by Eq. (10) is obeyed. It should also be noted that there are two poles in the expression for the reduced response corresponding to the two peaks in the spectral density function. If  $h=1$ , then  $s_1=0$ , i.e., one peak is located at zero, which is explicitly shown in Fig. 2(a).

Similarly, we can also apply our graded spectral representation to a single-shell sphere of core dielectric constant  $\epsilon_1$ , covered by a shell of  $\epsilon_2$ , and suspended in a host of  $\epsilon_0$ . In this example, we can also define two microstructure parameters  $\eta_1$  and  $\eta_2$ . If we let  $s=1/(1-\epsilon_1/\epsilon_0)$ , then  $\eta_1=1$ , and  $\eta_2=(\epsilon_0-\epsilon_2)/(\epsilon_0-\epsilon_1)$ . The dipole factor of a single-shell sphere is given by [24,25]

$$b = \frac{\epsilon_2 - \epsilon_0 + (\epsilon_0 + 2\epsilon_2)xf^3}{\epsilon_2 + 2\epsilon_0 + 2(\epsilon_2 - \epsilon_0)xf^3},$$

where  $f$  is the ratio between radius core and radius shell, and  $x$  is given by

$$x = \frac{\epsilon_1 - \epsilon_2}{\epsilon_1 + 2\epsilon_2}.$$

Then, we can also write Eq. (19) similarly to Eq. (20), where the residues and eigenvalues are given by

$$F_1 = \frac{-3ps_1[(-1+\eta)y^3 - \eta] - \eta p[1 - 2(-1+\eta)y^3 + 2\eta]}{3(s_1 - s_2)},$$

$$F_2 = \frac{3ps_2[(-1+\eta)y^3 - \eta] + \eta p[1 - 2(-1+\eta)y^3 + 2\eta]}{3(s_1 - s_2)},$$

$$s_1 = \frac{1}{6}[1 + 3\eta - \sqrt{1 + (2-8y^3)\eta + (1+8y^3)\eta^2}],$$

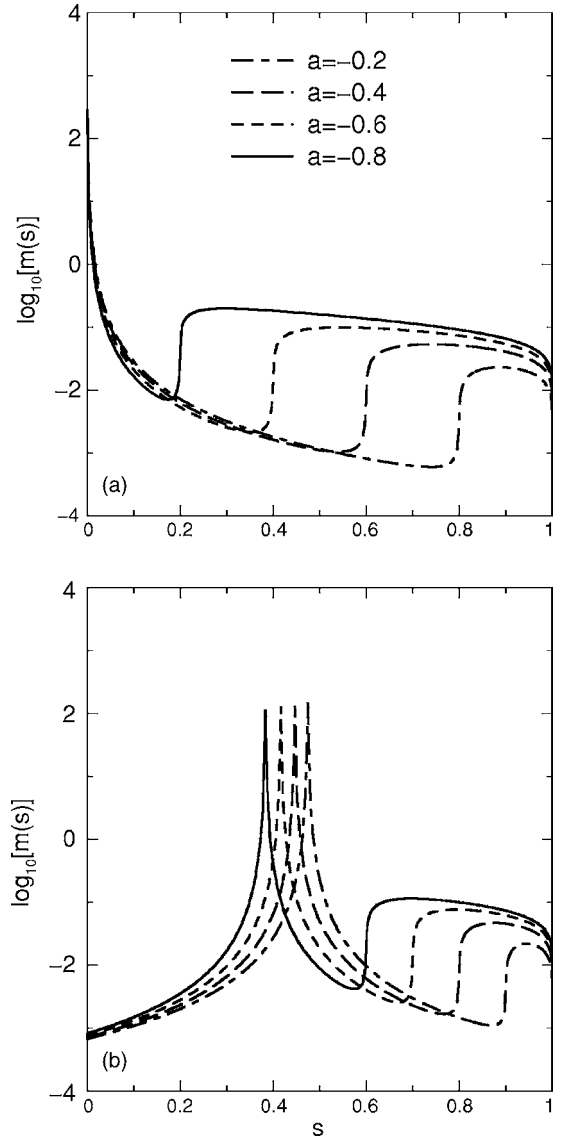


FIG. 2. (a) Spectral density functions of graded films without an interface, i.e.,  $h=1.0$  (the width of the film was chosen to be  $L=1$ ). (b) Spectral density function of a graded film meeting a homogeneous medium at an interface at  $h=0.5$ . The dielectric constant of the host medium is  $\epsilon_2=1$ .

$$s_2 = \frac{1}{6}[1 + 3\eta + \sqrt{1 + (2-8y^3)\eta + (1+8y^3)\eta^2}].$$

Analysis shows that the spectral representation for  $N=2$  contains two simple poles corresponding to two peaks in the spectral density function. Therefore we draw the conclusion that  $N$  peaks are a result of  $N$  layers. Moreover,  $N-1$  peaks will accumulate into a continuous broad absorption spectrum when  $N$  tends toward infinity, which can be seen from Figs. 4(f) and 5(f).

## V. NUMERICAL RESULTS

We are now in a position to do some numerical calculations of the spectral density function from Eqs. (11) and (13).

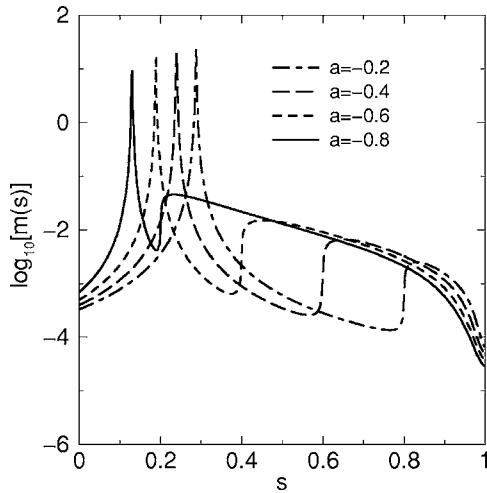


FIG. 3. The spectral density function of a graded sphere with volume fraction  $p=0.1$ .

A small but finite imaginary part in the complex parameter has been used in the calculations. Without any loss of generality, we choose  $L=1$  for the width of the dielectric film and  $R=1$  for the radius of the graded sphere. We show the effect of different graded profiles, as well as the effect of the thickness of the inclusion. It should be noted that in all figures the range of  $s$  is limited to  $[0, 1]$ , since we chose  $-1 < a < 0$  which limits the values of  $\eta$  into  $[0, 1]$ .

Figure 1 displays the dielectric profile of a graded film [Fig. 1(a)] and a graded sphere [Fig. 1(b)]. The figure shows that the dielectric constant varies with the position in the inclusion while it remains constant in the host medium. Also, different values of  $a$  accord with different graded materials.

In Fig. 2(a) we plot the spectral density function  $m(s)$  of a graded film without an interface against the spectral parameter for various graded microstructures  $\eta(z)$ . It is evident that there is always a broad continuous band in the spectral density function. Both the strength as well as the width of the

continuous part of  $m(s)$  increase with the gradient of the dielectric profile. Thus the previous results of the broad surface-plasmon band can be expected. Note that there is a sharp peak at  $s=0$ , which is also present in a homogeneous film. In Fig. 2(b) we plot the spectral density function of a graded film meeting a homogeneous medium at an interface for various graded microstructures  $\eta(z)$ . Again, there is always a broad continuous band in the spectral density function. However, the sharp peak has now shifted to a finite value of  $s$ , which is also present in a homogeneous film.

In Fig. 3 the spectral density function of a graded sphere is displayed for a volume fraction  $p=0.1$ . In this case, the interface always exists. It is clear that a broad continuous function in the spectral density function is always observed, as well as the shift of the sharp peak. However, the decrease of the broad continuous function is more abrupt for a graded sphere than for graded film with increasing  $s$ .

Figures 4 and 5 display the spectral density function for a multilayer film and a sphere, respectively. It is clear that there are always  $N$  sharp peaks for  $N$  layers. Moreover, it is worth noting that there occurs a transition from sharp peaks to a broad continuous band with increasing  $N$  [see Figs. 4(f) and 5(f)], that is, the graded results are recovered as  $N \rightarrow \infty$ . In particular, we had obtained the analytical expression of spectral density function for  $N=2$ . There are two resonances corresponding to the two peaks in Figs. 4(a) and 5(a).

### VI. DISCUSSION AND CONCLUSIONS

We have investigated a graded composite film and a sphere by means of the Bergman-Milton spectral representation. It was shown that the spectral density function can be obtained analytically for a graded system. However, unlike in the case of homogeneous constituent components, the characteristic function is a continuous function due to the presence of gradation. Moreover, the derivation as well as some salient properties, namely, the sum rule, the definition

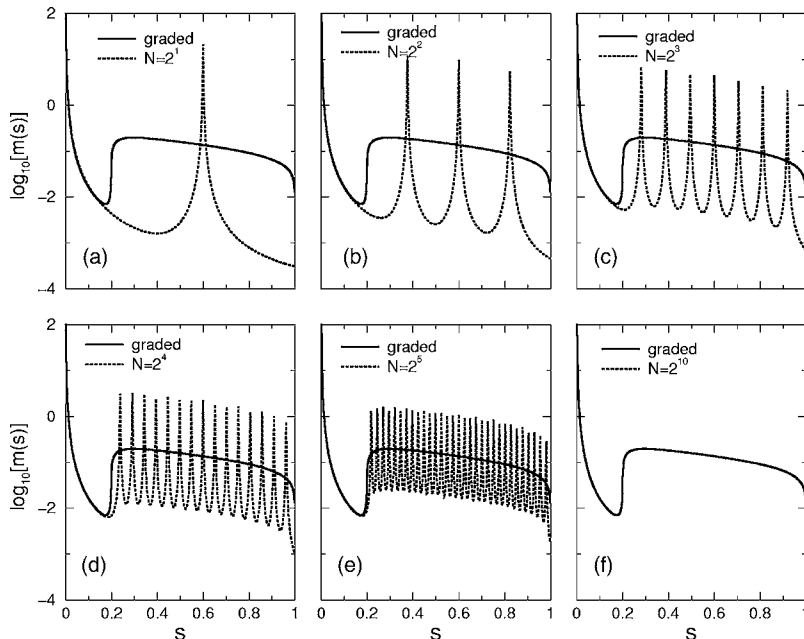


FIG. 4. Spectral density functions of multilayer films with a varying number of layers ( $N$ ). The dielectric constant of the host medium is set to  $\epsilon_0=1$  and the parameter defining the slope of the gradation to  $a=-0.8$ . The solid line shows the  $N \rightarrow \infty$  limit.

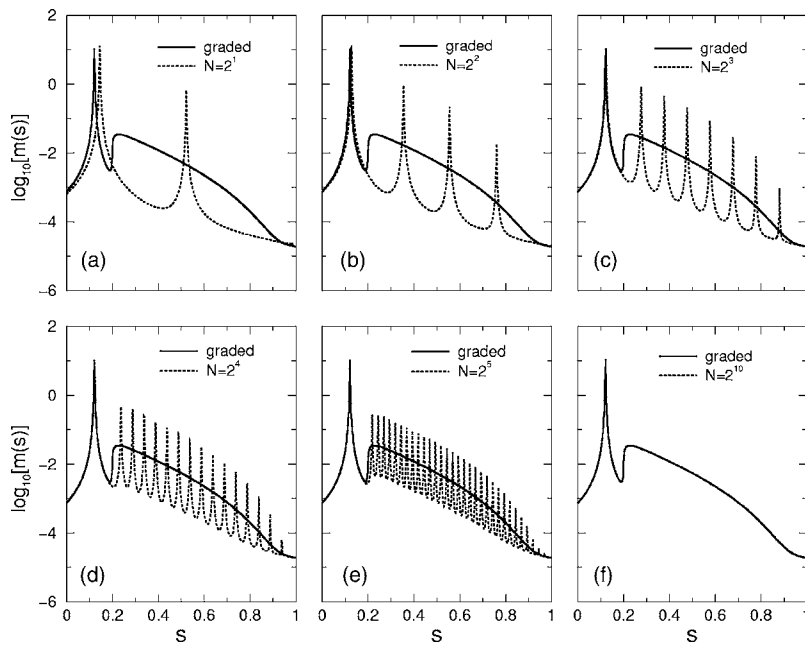


FIG. 5. Spectral density functions of various multishell spheres with a varying number of shells ( $N$ ). The dielectric constant of the host medium is set to  $\epsilon_0=1$  and the parameter defining the slope of the gradation to  $a=-0.8$ . The volume fraction of embedded graded spheres is  $p=0.1$ . The solid line shows the  $N \rightarrow \infty$  limit.

of the inner product, the definition of the integral-differential operators, and the range of spectral parameters, do change because of the continuous variation of the dielectric profile within the constituent components. It should be noted that in a graded composite the eigenvalues are not limited to  $[0, 1]$  and they can be extended to  $-\infty \leq s_n \leq \infty$  for the full region of  $\eta$ , i.e.,  $-\infty \leq \eta \leq \infty$ . In this work, for simplicity, we investigated the spectral density function in  $0 \leq s \leq 1$  by choosing  $-1 < a < 0$  to limit the values of  $\eta$  into  $[0, 1]$ .

We also studied multilayer composites and calculated the spectral density function versus the number of layers in order to explicitly demonstrate that the broad continuous spectrum arises from the accumulation of poles when the number of layers tends to infinity. This finding coincides with the broad surface-plasmon absorption band associated with the optical properties of graded composites.

To sum up, we have investigated the spectral density function of graded films and graded spheres as well as multilayers. There is always a broad continuous function in the spectral density function in graded composite, but simple poles in multilayer composite, and the number of poles, depend on the number of layers. Moreover, there is a gradual transition from sharp peaks to a broad continuous band until the graded composite results are recovered in the limit of  $N \rightarrow \infty$ .

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- [1] G. W. Milton, *The Theory of Composites* (Cambridge University Press, Cambridge, 2002), Chap. 7.
- [2] M. Yamanouchi, M. Koizumi, T. Hirai, and I. Shioda, in *Proceedings of the First International Symposium on Functionally Graded Materials* (Sendi, Japan, 1990).
- [3] J. B. Holt, M. Koizumi, T. Hirai, and Z. A. Munir, *Ceramic Transaction: Functionally Gradient Materials* (The American Ceramic Society, Westerville, OH, 1993), Vol. 34.
- [4] B. Ilschner and N. Cherradi, *Proceedings of the Third International Symposium on Structure and Functional Gradient Materials, Lausanne, Switzerland* (Presses Polytechniques et Universitaires Romands, 1994).
- [5] D. R. Kammler, T. O. Mason, D. L. Young, T. J. Coutts, D. Ko, K. R. Poeppelmeier, and D. L. Williamson, *J. Appl. Phys.* **90**, 5979 (2001).
- [6] A. Kawasaki and R. G. Ford, in *Functionally Graded Materials: Design, Processing, and Application*, edited by Y. Miyamoto, W. A. Kaysser, B. H. Rabin, A. Kawasaki, and R. G. Ford (Kluwer, Boston, 1999), p. 97.
- [7] S. G. Lu, X. H. Zhu, C. L. Mak, K. H. Wong, H. L. W. Chan, and C. L. Choy, *Appl. Phys. Lett.* **82**, 2877 (2003).
- [8] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975).
- [9] See, for example, the articles in *Electric Transport and Optical Properties of Inhomogeneous Media*, edited by J. C. Garland and D. B. Tanner, AIP Conference Proceedings No. 40 (AIP, New York, 1978).
- [10] L. Dong, G. Q. Gu, and K. W. Yu, *Phys. Rev. B* **67**, 224205 (2003).
- [11] G. Q. Gu and K. W. Yu, *J. Appl. Phys.* **94**, 3376 (2003).
- [12] J. P. Huang, K. W. Yu, G. Q. Gu, and M. Karttunen, *Phys. Rev. E* **67**, 051405 (2003).
- [13] J. P. Huang and K. W. Yu, *Appl. Phys. Lett.* **85**, 94 (2004).
- [14] K. P. Yuen, M. F. Law, K. W. Yu, and P. Sheng, *Phys. Rev. E*

- 56**, R1322 (1997).
- [15] D. J. Bergman and D. Stroud, in *Solid State Physics*, edited by H. Ehrenreich and D. Turnbull (Academic Press, New York, 1992), Vol. 46, p. 147.
- [16] G. W. Milton, *J. Appl. Phys.* **52**, 5286 (1980); R. C. McPhe-dran and G. W. Milton, *Appl. Phys. A: Solids Surf.* **26**, 207 (1981).
- [17] M. P. Hobson and J. E. Baldwin, *Appl. Opt.* **43**, 2651 (2004).
- [18] S. Martin, J. Rivory, and M. Schoenauer, *Appl. Opt.* **34**, 2247 (1995).
- [19] P. G. Verly, *Appl. Opt.* **37**, 7327 (1998).
- [20] D. Stroud and P. M. Hui, *Phys. Rev. B* **37**, 8719 (1988).
- [21] Y. Gu and Q. H. Gong, *Phys. Rev. B* **67**, 014209 (2003).
- [22] O. Levy and D. J. Bergman, *Physica A* **207**, 157 (1994).
- [23] H. Ma, B. Zhang, W. Y. Tam, and P. Sheng, *Phys. Rev. B* **61**, 962 (2000).
- [24] K. W. Yu, P. M. Hui, and D. Stroud, *Phys. Rev. B* **47**, 14150 (1993).
- [25] X. Zhang and D. Stroud, *Phys. Rev. B* **49**, 944 (1994).