Theory of optical bistability in a weakly nonlinear composite medium

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A variational approach is used to discuss bistable behavior in composite media where one component is a nonlinear dielectric while the other has a field independent complex dielectric constant with a negative real part. Under certain conditions, the physical parameters may be adjusted so that bistability appears even though the nonlinear behavior is everywhere weak, thus allowing it to be treated as a small perturbation to the leading linear behavior.

I. INTRODUCTION

There is a growing interest in optical materials with bistable behavior. This is due, in part, to their potential uses as materials for optical devices.[1] It has been recognized that one way to obtain such behavior is to combine in a composite medium a dielectric component together with a metallic or semiconducting component the dielectric constant of which has a negative real part and a small imaginary part.[2–4] Since the nonlinear behavior usually requires high field intensities in order to be non-negligible, bistable behavior is usually believed to appear only above a certain intensity threshold which can be very high. Proposals have been put forward for decreasing this threshold by exploiting the field enhancement produced by the surface plasmon resonance of the metallic component. This component was assumed to appear in the form of small particles (i.e., small compared to the wavelength) embedded in the dielectric host,[3,4] or as a thin metallic coating which covers similarly small dielectric inclusions.[5] It is difficult to study such materials theoretically because the nonlinear behavior of at least one of the components makes the usual methods developed for linear materials inapplicable. The only good quantitative analysis is for a very dilute composite of spherical or spheroidal inclusions in which the nonlinear behavior is restricted to the innermost core of the (possibly multilayered) inclusion.[2,4,5] The surface plasmon resonance is an extreme manifestation of the so-called local field effect, whereby the local electric field can be increased above its ambient or average value in the vicinity of a conducting inclusion which is embedded in a dielectric host medium. Such local field effects have been invoked in the past in connection with optical bistability also in other systems.[6–8]

In this paper we demonstrate the following results in the theory of metal-dielectric composites: (a) By appropriately tuning the material parameters and the frequency in the vicinity of a *sharp* resonance, bistable optical behavior can be achieved at field intensities so low that the local nonlinear behavior is *everywhere weak.* (b) In this weakly nonlinear regime, an accurate quantitative calculation of the optical properties can be performed by treating the nonlinear effects as a small perturbation to the leading linear behavior. (c) These conclusions are reached following a theoretical study of bistability in composite mixtures of a linear metal and a weakly nonlinear dielectric with three different microstructures: parallel slabs, a dilute suspension of metallic spheres in a nonlinear dielectric host, and a coated spheres assemblage with all spheres having the same core-to-shell volume ratio. (d) A type of variational principle is proposed and employed. This principle should be capable of generating useful approximations for the local electric field in many types of nonlinear dielectric composites besides those considered here.

II. EXAMPLES OF BISTABLE BEHAVIOR IN COMPOSITE MICROSTRUCTURES

In our analysis we always assume that the metal component has a frequency dependent but field independent complex dielectric constant

$$\epsilon_m = \epsilon'_m + i\epsilon''_m \text{ where } \epsilon'_m < 0,\ \epsilon''_m > 0 \text{ and } \epsilon''_m \ll |\epsilon'_m|. \tag{1}$$

The dielectric component is assumed to have a purely real, frequency independent but field dependent dielectric constant of the form

$$\epsilon_d(\mathbf{E}) = \epsilon_0 + b_0|E|^2 \text{ where } \epsilon_0, b_0 > 0 \text{ and } |E|^2 \equiv \mathbf{E}^* \cdot \mathbf{E}. \tag{2}$$

We also assume that the macroscopic properties of the medium can be obtained from the properties of the components in the quasistatic limit. This means that the grain sizes are all small compared to both the wavelength and the skin depth of the electromagnetic fields, and that scattering can be ignored.

Some of these assumptions are made purely for convenience. In particular, we could allow the dielectric constant of the metal to be weakly field dependent too, we could allow other values for the nonlinearity exponent, we could allow a small imaginary part in ϵ_d, and both ϵ_d and ϵ_m could depend on the frequency, all that without significantly changing our conclusions.

A. Parallel slabs microstructure

In this case, both fields **E** and **D** are uniform in each component and perpendicular to the slabs. (The case where these fields are parallel to the slabs is less interesting because the local electric field is then uniform, so that no enhancement is possible.) We denote the average or externally imposed values of the fields by $\mathbf{E}_0$, $\mathbf{D}_0$, the local fields in the two components by $\mathbf{E}_d$, $\mathbf{D}_d = \epsilon_d \mathbf{E}_d$ and $\mathbf{E}_m$, $\mathbf{D}_m = \epsilon_m \mathbf{E}_m$, and the volume fractions by p_d, $p_m = 1 - p_d$. The local field values are determined by the continuity conditions on the normal component of **D**

$$D_0 = \epsilon_m E_m = \epsilon_d E_d, \tag{3}$$

from which we get

$$E_0 = p_d E_d + p_m E_m = \left(p_d + p_m \frac{\epsilon_0 + b_0 |E_d|^2}{\epsilon_m} \right) E_d. \tag{4}$$

In order to solve this nonlinear equation for the complex field $\mathbf{E}_d$, we first derive from it an equation for $|E_d^2|$, namely,

$$E_0^2 = \left| p_d + p_m \frac{\epsilon_0 + b_0 |E_d|^2}{\epsilon_m} \right|^2 |E_d|^2, \tag{5}$$

which is rewritten in terms of a renormalized field intensity t as follows:

$$t \equiv \frac{b_0 |E_d|^2}{\left|\epsilon_0 + \frac{p_d}{p_m}\epsilon_m\right|} > 0 \ , \tag{6}$$

$$\mu \equiv -\frac{\mathrm{Re}\left(\epsilon_0 + \frac{p_d}{p_m}\epsilon_m\right)}{\left|\epsilon_0 + \frac{p_d}{p_m}\epsilon_m\right|}, \quad |\mu| < 1, \tag{7}$$

$$\alpha \equiv b_0 E_0^2 \frac{|\epsilon_m^2|/p_m^2}{\left|\epsilon_0 + \frac{p_d}{p_m}\epsilon_m\right|^3} > 0 \ , \tag{8}$$

$$f(t) \equiv t^3 - 2\mu t^2 + t = \alpha. \tag{9}$$

Any real solution of this equation for t, and hence for $|E_d|^2$, is then substituted into (4), which thus becomes a linear equation for E_d. Equation (9) always has at least one real, positive solution. In order for it to have (two) more such solutions, μ and α must satisfy the following additional inequalities:

$$\frac{\sqrt{3}}{2} \le \mu \ , \tag{10}$$

$$\alpha_+ \le \alpha \le \alpha_-, \tag{11}$$

where $\alpha_\pm = f(t_\pm)$ and t_- (t_+) is the position of the local minimum (maximum) of $f(t)$, given by

$$t_\pm = \tfrac{1}{3}[2\mu \pm (4\mu^2 - 3)^{1/2}] > 0. \tag{12}$$

Both of these values $t_\pm$ are positive and $O(1)$, and also $\alpha_- = O(1)$. Therefore all three solutions of (9) are $O(1)$, and the three corresponding fields E_d all satisfy

$$b_0 |E_d|^2 = O\left(\left|\epsilon_0 + \frac{p_d}{p_m}\epsilon_m\right|\right). \tag{13}$$

The quantity $|\epsilon_0 + p_d\epsilon_m/p_m|$ can be called the "detuning parameter," since it measures how far the system is from a perfect resonance condition, where it vanishes. If this detuning parameter is small, namely,

$$\left|\epsilon_0 + \frac{p_d}{p_m}\epsilon_m\right| \ll \epsilon_0, \tag{14}$$

then the dielectric component is in the weakly nonlinear regime. This property is not very important in the present case, where the local field can be calculated irrespective of whether the nonlinearity is weak or strong. However, in other microstructures, this property is crucial, since it enables us to perform an accurate evaluation of the local fields by treating the nonlinear behavior as a small perturbation. This is demonstrated below.

Since the three solutions of (9) and of (4) all provide possible values of the local field when the external or average field has the same value E_0, some other criterion must be used to decide which state the system will actually choose to be in. It is not easy to find such a criterion, since there is no simple stability principle in this nonequilibrium problem. By analogy with the somewhat similar problem of a first order phase transition in a thermodynamic system at equilibrium, we would guess that the two extreme solutions are the more stable ones, and that the system makes transitions between them, with hysteresis, as E_0 is varied. This problem clearly requires further study.

B. Low density of spherical inclusions

In this case we can neglect the interactions between different spheres and consider the problem of an isolated sphere of volume V_a embedded in a host of much larger volume $V \gg V_a$. When the host is a material with linear electrical properties and the externally applied field is uniform, this is a solvable problem even if the inclusion is nonlinear. This fact has been exploited to discuss the case of nonlinear metallic inclusions in a linear dielectric host.[2,4]

A more interesting case is the one where the embedded sphere is metallic or semiconducting with linear electrical properties but a negative value for $\mathrm{Re}(\epsilon)$, as in (1), while the host is a nonlinear dielectric, as in (2). This case cannot be solved exactly. We will now use an exact variational principle along with a trial potential function to generate a sensible approximation for this case.

The basic variational principle of electrostatic fields in dielectrics is that the energy functional

$$U[\phi] \equiv \frac{1}{8\pi}\int \epsilon(\mathbf{r})[\nabla\phi(\mathbf{r})]^2 dV \qquad (15)$$

achieves its absolute minimum value, among all continuous real scalar functions $\phi(\mathbf{r})$ that obey given real boundary conditions, when ϕ is the solution of the usual differential equation of electrostatics

$$\nabla\cdot[\epsilon(\mathbf{r})\nabla\phi] = 0. \qquad (16)$$

This applies when $\epsilon(\mathbf{r})$ is field independent and real. A generalization of this principle exists for the case where $\epsilon(\mathbf{r})$ is real but field dependent, as long as the scalar product $\mathbf{E}\cdot\mathbf{D} = \epsilon(E^2)E^2$ is everywhere an increasing function of E^2.[9] A generalization is also easily found for the case where $\epsilon(\mathbf{r})$ is complex but field independent. But when $\epsilon(\mathbf{r})$ is both complex and field dependent, having the form

$$\epsilon = \epsilon' + i\epsilon'' + b|E|^2, \qquad (17)$$

then an energylike functional $U[\phi]$ cannot be defined in general. We can nevertheless enunciate a variational principle by noting that the equation

$$\int \mathbf{D}\cdot\delta\mathbf{E}^* dV = 0 \qquad (18)$$

holds for arbitrary variations $\delta\phi(\mathbf{r})$ away from the solution of the equation

$$\nabla\cdot[\epsilon(\mathbf{r},\nabla\phi)\nabla\phi] = 0, \qquad (19)$$

as long as $\delta\phi = 0$ at the boundary.

We use this principle for the present problem by choosing as trial function a form that is suggested by the exact solution to the analogous linear problem, namely,

$$\phi(\mathbf{r}) = \begin{cases} E_0(1-B)r\cos\theta, & r < a \\ E_0 r\cos\theta - E_0 B a^3\frac{\cos\theta}{r^2}, & r > a, \end{cases} \qquad (20)$$

where E_0 is the (real) amplitude of the externally applied uniform field which is taken to point in the z direction and B is a complex variational parameter yet to be determined. Substituting this trial function into (18), we obtain the following equation for B:

$$0 = -(\epsilon_m - \epsilon_0) + (\epsilon_m + 2\epsilon_0)B + b_0E_0^2(1 + 2B + \tfrac{8}{5}\mathrm{Re}B + \tfrac{2}{5}|B^2| + \tfrac{4}{5}B\,\mathrm{Re}B + \tfrac{8}{5}|B^2|B). \qquad (21)$$

If we neglect the nonlinear effects, which appear in this equation as the terms multiplied by $b_0E_0^2$, we obtain the correct value of B for the linear problem. In the vicinity of the surface plasmon resonance, when $\epsilon_m + 2\epsilon_0 = 0$, this value becomes very large so that even if $b_0E_0^2$ is very small, the nonlinear effects may be important. In that case it is a good approximation to ignore all but the highest power of B among the terms that multiply $b_0E_0^2$. We thus get the following equation for B, valid near the surface plasmon resonance:

$$0 = -(\epsilon_m - \epsilon_0) + (\epsilon_m + 2\epsilon_0)B + \tfrac{8}{5}b_0E_0^2|B^2|B. \qquad (22)$$

This equation is very similar to (4), and can be analyzed in a similar way: We first get an equation for $|B^2|$

$$|\epsilon_m - \epsilon_0|^2 = \left|\epsilon_m + 2\epsilon_0 + \tfrac{8}{5}b_0E_0^2|B^2|\right|^2|B^2|, \qquad (23)$$

then use the definitions

$$t \equiv |B^2|\frac{\frac{8}{5}b_0E_0^2}{|\epsilon_m + 2\epsilon_0|} > 0\ , \qquad (24)$$

$$\mu \equiv -\frac{\mathrm{Re}(\epsilon_m + 2\epsilon_0)}{|\epsilon_m + 2\epsilon_0|},\ \ |\mu| < 1, \qquad (25)$$

$$\alpha \equiv \frac{\frac{8}{5}b_0E_0^2|\epsilon_m - \epsilon_0|^2}{|\epsilon_m + 2\epsilon_0|^3}\ , \qquad (26)$$

to get

$$f(t) \equiv t^3 - 2\mu t^2 + t = \alpha. \qquad (27)$$

Finally, we substitute any solution for t, and hence for $|B|^2$, into (22), which then provides a unique solution for B. Equation (27) has exactly the same form as (9), so the same analysis is applicable. In the dielectric host, the strongest field $E_{\max}$ is found at the poles of the spherical inclusion, and its value there is determined mostly by the induced dipole moment E_0Ba^3, so that

$$b_0|E_{\max}|^2 \approx b_0E_0^2 4|B^2| = O(|\epsilon_m + 2\epsilon_0|). \qquad (28)$$

Again, if α and μ are tuned to lie in the appropriate ranges and if the detuning parameter $|\epsilon_m + 2\epsilon_0|$ is small, namely, $|\epsilon_m + 2\epsilon_0| \ll \epsilon_0$, then we get bistable behavior under conditions where the local nonlinear behavior is weak everywhere.

We note that although the solution we obtained is approximate, its accuracy can be improved by including more spherical harmonics and more variational parameters in the trial function. We also note that the accuracy of our approximation is expected to be excellent in the case under consideration, when the nonlinearity is weak everywhere.

In order to detect bistable or multistable behavior in the bulk optical properties we must of course have a finite density of metallic inclusions. The multistable behavior will then manifest itself in that the volume average displacement field $\langle D\rangle$ will be able to attain different values for the same value of the applied or volume average electric field $\langle E\rangle = E_0$. In order for the discussion presented in this subsection to be valid, the density of inclusions must be small and their shapes as well as orientations must be similar, e.g., spheres.

C. Coated spheres assemblage

In this microstructure the material consists entirely of composite spheres with a spherical core made of one component and a concentric spherical shell made of the other component. The spheres must come in many different sizes but all have the same core-to-shell volume ratio. In the linear case the local fields can be found exactly.[10] In the nonlinear case we apply the variational principle of (18) to the following trial function for the potential in and around a coated sphere situated at the origin:

$$\phi = \begin{cases} E_0 A_1 r\cos\theta, & r < a \\ E_0 A_2 r\cos\theta - E_0 B a^3 \frac{\cos\theta}{r^2}, & a < r < b \\ E_0 r\cos\theta - E_0 B_3 b^3 \frac{\cos\theta}{r^2}, & b < r, \end{cases} \tag{29}$$

$$A_1 = A_2 - B \ , \tag{30}$$

$$1 - B_3 = A_2 - B\frac{a^3}{b^3}. \tag{31}$$

The latter two equalities are necessary to ensure that $\phi(\mathbf{r})$ is continuous at the two interfaces $r = a$, $r = b$, and they reduce to 2 the number of independent complex variational parameters. In the region $r > b$, all the other coated spheres in the assemblage are replaced by a fictitious uniform medium with a (field dependent) dielectric constant $\epsilon_e(E_0)$. The value of ϵ_e is determined so as to make the total dipole moment of the sphere under consideration vanish, i.e., $B_3 = 0$. But we can set $B_3 = 0$ only *after* implementing the variational principle, when B and B_3 are treated as the independent variational parameters. We have taken the core to be metallic and the coating to be dielectric, with dielectric constants ϵ_d, ϵ_m as in (1), (2), and with volume fractions $p_m \equiv a^3/b^3$, $p_d \equiv 1 - p_m$. The resultant equations, after setting $B_3 = 0$, are, respectively,

$$\begin{aligned} 0 = &-(\epsilon_m - \epsilon_0) + [\epsilon_m + 2\epsilon_0 - p_m(\epsilon_m - \epsilon_0)]B \\ &+ b_0 E_0^2\{[1 + (2 + p_m)B]|1 + p_m B|^2 + (\mathrm{Re}B + p_m|B^2|)[\tfrac{8}{5}(1 + 2p_m B) + \tfrac{4}{5}(1 + p_m)B] \\ &+ |B^2|[2p_m(1 + p_m B) + \tfrac{2}{5}(1 + p_m)(1 + 2p_m B) + \tfrac{8}{5}(1 + p_m + p_m^2)B]\} \ , \end{aligned} \tag{32}$$

$$\begin{aligned} 0 = &\ \epsilon_e(E_0) - \epsilon_0 - p_m(\epsilon_m - \epsilon_0) + p_m p_d(\epsilon_m - \epsilon_0)B \\ &+ b_0 E_0^2\{-p_d(1 + p_m B)|1 + p_m B|^2 - p_m p_d|B^2|[2(1 + p_m B) + \tfrac{2}{5}B(1 + p_m)] - \tfrac{8}{5}p_m p_d B(\mathrm{Re}B + p_m|B^2|)\} \ . \end{aligned} \tag{33}$$

The first of these must be solved for the unknown parameter B, and the second one then determines the other unknown ϵ_e. Both B and ϵ_e will depend on E_0^2. As before, when the nonlinear terms that appear multiplied by $b_0 E_0^2$ are neglected, one regains the correct solution for the linear case. The value thus found for B becomes very large in the vicinity of the resonance for this microstructure, i.e., when the detuning parameter $\epsilon_m + 2\epsilon_0 - p_m(\epsilon_m - \epsilon_0)$ satisfies

$$|\epsilon_m + 2\epsilon_0 - p_m(\epsilon_m - \epsilon_0)| \ll |\epsilon_m - \epsilon_0|. \tag{34}$$

In that case we cannot neglect the nonlinear terms, but we can get good results by including only the highest power of B. We thus get, instead of (32),

$$\begin{aligned} 0 = &-(\epsilon_m - \epsilon_0) + [\epsilon_m + 2\epsilon_0 - p_m(\epsilon_m - \epsilon_0)]B \\ &+ \tfrac{1}{5}(5p_m^3 + 52p_m^2 + 16p_m + 8)b_0 E_0^2|B^2|B, \end{aligned} \tag{35}$$

which is similar to (22) and (4), and can be reduced to (9) by making the following definitions:

$$t \equiv \frac{\frac{1}{5}b_0 E_0^2(5p_m^3 + 52p_m^2 + 16p_m + 8)}{|\epsilon_m + 2\epsilon_0 - p_m(\epsilon_m - \epsilon_0)|}|B^2| \ , \tag{36}$$

$$\mu \equiv -\frac{\mathrm{Re}[\epsilon_m + 2\epsilon_0 - p_m(\epsilon_m - \epsilon_0)]}{|\epsilon_m + 2\epsilon_0 - p_m(\epsilon_m - \epsilon_0)|}, \ |\mu| < 1, \tag{37}$$

$$\alpha \equiv \frac{|\epsilon_m - \epsilon_0|^2 \frac{1}{5}b_0 E_0^2(5p_m^3 + 52p_m^2 + 16p_m + 8)}{|\epsilon_m + 2\epsilon_0 - p_m(\epsilon_m - \epsilon_0)|^3}. \tag{38}$$

Again we find, as before, that in the vicinity of the resonance we can get bistable behavior while the nonlinearity is weak everywhere, namely,

$$b_0|E|^2 \le O(|\epsilon_m + 2\epsilon_0 - p_m(\epsilon_m - \epsilon_0)|) \ll 1. \tag{39}$$

III. QUANTITATIVE CONSIDERATIONS

The examples discussed in Sec. II show that an interesting type of bistable optical medium is a composite in which one component is a nonlinear dielectric while the other has a field independent dielectric constant with a negative real part and a very small imaginary part. A possible candidate to fill this prescription would be a dilute suspension of small silver spheres, each with a radius small compared to the wavelength, embedded in a matrix of optical glass. The glass should not be pure silica but a glass appropriately doped so as to enhance its cubic nonlinearity coefficient b_0.[11]

We assume the following values for the optical parameters:

$$\epsilon_m' = -8.4, \ \ \epsilon_m'' = 0.2, \ \ \epsilon_0 = 4, \ \ b_0 = 10^{-8} \text{ esu},$$

where the value of ϵ_m'' is close to the lowest value achievable in silver at visible frequencies,[12] while the values of ϵ_m' and ϵ_0 are actually nominal: ϵ_m' must be tuned by adjusting the frequency so as to make $\epsilon_m' + 2\epsilon_0$ have a value which is neither too large nor too small. On the one hand, it should be as small as possible so as to minimize the right-hand side (rhs) of (28), but on the other hand, it should be large enough so as to ensure that μ of (25) satisfies the inequality (10), which is necessary for bistability to occur. A good value to aim for is thus found to be $\epsilon'_m + 2\epsilon_0 = -0.4$. The value of b_0 is taken from a representative commercial doped glass.[11] Using these values for the case of the isolated sphere of Sec. II B we find

$$\epsilon'_m + 2\epsilon_0 = -0.4, \quad |\epsilon_m + 2\epsilon_0| = 0.447, \quad |\epsilon_m - \epsilon_0| = 12.40,$$
$$\mu = 0.8944, \quad t_+ = 0.7454, \quad t_- = 0.4472,$$
$$\alpha_+ \equiv f(t_+) = 0.1656, \quad \alpha_- \equiv f(t_-) = 0.1789.$$

Choosing $\alpha = 0.17$, which satisfies the inequality (11), we then find that there are three solutions for t, all of order 1. From (26) and (24) we then get

$$b_0 E_0^2 \approx 6 \times 10^{-5} ,$$
$$|B| \approx 68 ,$$

for all of these solutions. The incident energy flux required to produce bistable behavior is therefore given by

$$I = \frac{c}{4\pi} E_0^2 \approx 1.4 \times 10^6 \text{ W/cm}^2. \qquad (40)$$

The rather high value of flux that is required results from the fact that $|\epsilon_m + 2\epsilon_0|$ cannot be made small enough in this system, because ϵ''_m is never less than 0.186.[12] The obvious way to get a lower threshold for bistable behavior is to use, instead of silver inclusions, a nonmetallic material that has lower losses and is operated at a frequency slightly below a plasma edge (where $\epsilon'_m = 0$), so that $\epsilon''_m \ll 1$ and $\epsilon'_m < 0$.

IV. DISCUSSION AND CONCLUSIONS

As pointed out in the Introduction, the appearance of optical bistability in metal-dielectric composites has been predicted and discussed in a number of earlier references. Our contribution has been to introduce a variational principle that can be applied to some interesting microstructures that could not be discussed quantitatively in the previous approaches. In our examples, we found that by tuning of the material parameters and the frequency so as to be near the electric resonance of the system, we could achieve bistability even though the nonlinear properties were everywhere only a small perturbation to the leading linear behavior. Technically this happened because, even though the field dependent term $b_0|E|^2$ made only a small contribution to the local dielectric constant, the problem also had another small parameter, namely, the amount of detuning away from the perfect resonance condition. Because of this fact, the nonlinear terms involving $b_0|E|^2$ could not be treated perturbatively, as in Ref. 13, but had to be allowed to compete against the detuning parameter. Since the nonlinearity is everywhere weak, however, the field is everywhere close to what it would be in the linear approximation. Hence the choice of that form for the trial function should yield a good approximation to the exact field.

As shown by the example considered in Sec. III, it is difficult for strictly metallic inclusions to be sufficiently close to resonance, because ϵ''_m is not small enough. This problem may be overcome by using a different material which has a much smaller value of ϵ''_m along with a negative value of ϵ'_m.

We now speculate on how the results of this study might be generalized. In a nondilute composite there are an infinite number of resonances, each of which extends over the entire system.[9] When the microstructure is disordered, these resonances form a dense quasicontinuum, and the field enhancement near any one of them is insignificant. An exception to this statement may occur in the case of an exponentially localized state. However, such states do not contribute to the bulk effective dielectric constant in the quasistatic limit, and therefore any bistability associated with them will be difficult to detect macroscopically. A considerable enhancement, as found in the examples discussed in Sec. II, always occurs in the vicinity of a sharp, isolated resonance. Such sharp resonances which are extended over the entire system generally occur in two types of microstructure: (a) a dilute system of similarly shaped and identically oriented inclusions; (b) a nondilute composite mixture with a *periodic microstructure*. The example of Sec. II B clearly belongs to case (a). Examples belonging to case (b) remain to be studied. In doing so, it will presumably be helpful to use our solutions for the linear case as starting points for constructing good trial functions to use with the variational principle of (18).

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