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Exact calculation to second order of the effective dielectric constant of a strongly nonlinear inhomogeneous composite

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We study strongly nonlinear and inhomogeneous dielectrics that follow a power-law relation between the electric and the displacement fields, $\mathbf{D}=\epsilon|E|^{\beta}\mathbf{E}$. Considering a narrow distribution of local dielectric constants $\epsilon(\mathbf{r})$, we find the first correction to the potential field. Using this result we present an exact calculation of the effective dielectric constant of an isotropic system to second order in the fluctuations $\delta\epsilon(\mathbf{r})$. Our result is independent of details of the local geometry and represents the nonlinear analog of the exact calculation in linear dielectrics.

The linear properties of composite materials have received attention over the years from both scientists and engineers. In particular, properties of inhomogeneous conductors and dielectrics have been the target of intensive investigations.[1] Much effort centered around the value of the effective dielectric constant ϵ_{eff}. Its value was found to second order in the local fluctuations of $\epsilon(\mathbf{r})$, $\delta\epsilon(\mathbf{r})$, and was bounded by rigorous bounds,[2,3] independent of the microgeometry. On the other hand, nonlinear phenomena in composite dielectrics have received very little attention until very recently, even though they have been known to exist for a very long time. Excluding laser studies, nonlinearities of dielectrics are usually treated by assuming a small perturbation on the "pure" linear behavior.[4] Motivated by the scarcity of exact results in strongly nonlinear inhomogeneous dielectrics, we decided to investigate systems that have in general no linear regime, even in small fields. In this Rapid Communication we study inhomogeneous dielectric media that obey the following relation between the displacement and the electric fields:

$$\mathbf{D}=\epsilon(\mathbf{r})[(\mathbf{E})^2]^{\beta/2}\mathbf{E}. \tag{1}$$

This kind of relation may also describe a strongly nonlinear conductor where the current density $\mathbf{J}$ replaces $\mathbf{D}$ and the conductivity σ is the analog of ϵ. Such a $\mathbf{J}$-$\mathbf{E}$ relation has been found in some ceramic two-dimensional (2D) systems in low temperatures.[5] It is easy to show that if the exponent β characterizes the local dependence of D on E throughout the system, even while $\epsilon(\mathbf{r})$ varies in space, then (1) also characterizes the relation between the volume averages $\langle D\rangle$ and $\langle E\rangle$. Assuming a narrow spatial distribution of $\epsilon(\mathbf{r})$, we find the effective dielectric constant ϵ_{eff} of an isotropic system to second order in $\delta\epsilon$, independent of details of the microgeometry. We note that even in the linear case it is impossible to calculate higher-order terms in $\delta\epsilon$ without more detailed knowledge of the microstructure of the system.

The procedure will be to first establish the relation between the corrections to the potential field, Φ and ϵ_{eff}. We will show that for the second-order correction $\delta^2\epsilon_{\text{eff}}$ one needs to know only the first variation in Φ, $\delta\Phi$, as in linear systems.[3] Then we will solve for $\delta\Phi$ and use this solution to determine $\delta^2\epsilon_{\text{eff}}$.

The system under study is assumed to be composed of homogeneous grains made of different components. Altogether, the system comprises N nonlinear components having dielectric constants ϵ_i $(i=1,2,\dots,N)$. We further assume that the distribution of ϵ is narrow, i.e., $(\epsilon_i-\langle\epsilon\rangle)\ll\langle\epsilon\rangle$ for all i where, throughout the text, angular brackets denote volume averages. The system is confined in the z direction between two parallel plates located at $z=0$ and $z=L$, but is infinitely broad in the perpendicular directions. The boundary conditions are $\Phi=E_0L$ at $z=L$ and $\Phi=0$ at $z=0$. Under these conditions a homogeneous system ($\epsilon_i=\epsilon_0$ for all i) would display a constant field in the z direction, $\mathbf{E}_0$. We define ϵ_0 to be the dielectric constant of one of the components, and try to expand Φ and ϵ_{eff} in powers of $\delta\epsilon_i=\epsilon_i-\epsilon_0$. We shall see below that the exact value of ϵ_{eff} to second order in $\delta\epsilon_i$ is independent of this choice.

The bulk effective nonlinear dielectric constant ϵ_{eff} is defined by[6]

$$\epsilon_{\text{eff}}=\frac{1}{V}E_0^{-(\beta+2)}\int\epsilon(\mathbf{r})\,|\nabla\Phi|^{\beta+2}dV. \tag{2}$$

The effective dielectric constant may be naturally defined either via a volume average of the energy density, as in (2), or via the relation between the volume averages of D and E. In Ref. 6 it was shown that the two definitions coincide. This definition is useful as long as the boundary conditions vary appreciably in space only on length scales much larger than the grain sizes. To zero order in $\delta\epsilon$ we have $\epsilon_{\text{eff}}=\epsilon_0$. A first-order variation of both sides of (2) gives

$$\delta\epsilon_{\text{eff}}=\frac{1}{V}E_0^{-(\beta+2)}\int\{[\epsilon(\mathbf{r})-\epsilon_0]\,|\nabla\Phi_0|^{\beta+2}+\epsilon_0(\beta+2)\,|\nabla\phi_0|^{\beta}\nabla\Phi_0\cdot\nabla\delta\Phi\}dV\,.$$

The second term on the right-hand side (RHS) can be shown to vanish after integrating by parts and using $\nabla\cdot\delta\mathbf{D}=0$ and the condition that $\delta\Phi=0$ on the boundaries. Thus we are left with the first integral that can be calculated explicitly to yield

$$\delta\epsilon_{\text{eff}}=\langle\epsilon\rangle-\epsilon_0 \text{ or } \epsilon_{\text{eff}}=\langle\epsilon\rangle+O(\delta\epsilon^2)\,. \tag{3}$$

In a similar manner it can be shown that the second variation is

$$\delta^2\epsilon_{\text{eff}}=\frac{\beta+2}{VE_0^2}\int[\epsilon(\mathbf{r})-\epsilon_0]\nabla\Phi_0\cdot\nabla\delta\Phi\,dV\,. \tag{4}$$

Thus, to find $\delta^2\epsilon_{\text{eff}}$ we need only to know $\delta\Phi$. Note that relations (2)-(4) reduce to the linear results[3] when $\beta\to 0$.

We now proceed to find $\delta\Phi$ in terms of a Green's function. Expanding $\mathbf{D}$ to first order in $\delta\mathbf{E}$, assuming $\delta\mathbf{E}=-\nabla\delta\Phi$ and using $\nabla\cdot\mathbf{D}=0$, we find

$$\epsilon_0(\nabla^2+\beta\partial_{zz})\delta\Phi=\nabla\cdot(\delta\epsilon\mathbf{E}_0)\,. \tag{5}$$

Transforming to the new coordinate system:[6,7] $\xi=x$, $\eta=y$, and $\zeta=z/\sqrt{\beta+1}$, we can write (5) in the rescaled coordinates as

$$\nabla^2\delta\Phi=\frac{1}{\epsilon_0\sqrt{\beta+1}}\nabla\cdot(\delta\epsilon\mathbf{E}_0)\,. \tag{6}$$

This equation has the same form as the equation for $\delta\Phi$ in a linear dielectric composite where $\beta=0$ [see Eq. (II.43) in Ref. 3]. The same can be said about (4). Consequently, we can apply much of the formal machinery developed for linear media[3] to treat the nonlinear composites. Thus, we can use the Green's function for Laplace's equation, $G(\rho,\rho')$, to solve (6) as follows:

$$\begin{aligned}\delta\Phi&=-\frac{E_0}{\epsilon_0\sqrt{\beta+1}}\int d\Omega' G(\rho,\rho')\partial_{\zeta'}\delta\epsilon(\rho')\\&=\frac{E_0}{\epsilon_0\sqrt{\beta+1}}\int d\Omega'\delta\epsilon(\rho')\partial_{\zeta'}G(\rho,\rho')\\&=\frac{E_0}{\epsilon_0\sqrt{\beta+1}}\int d\Omega'[\delta\epsilon(\rho')-\langle\delta\epsilon\rangle]\partial_{\zeta'}G(\rho,\rho')\,.\end{aligned} \tag{7}$$

The second line is obtained after integrating parts and using $G=0$ on the boundary. Because of this boundary condition we also have $\int\partial_{\zeta'}G=0$, which explains the insertion of $\langle\delta\epsilon\rangle$ into the third line. A similar consideration, using $\delta\Phi=0$ on the boundary, allows ϵ_0 to be replaced by $\langle\epsilon\rangle$ in (4). Substituting for $\delta\Phi$ in (4), we get

$$\delta^2\epsilon_{\text{eff}}=-\frac{\beta+2}{V\epsilon_0\sqrt{\beta+1}}\int d\Omega\,d\Omega'\{[\delta\epsilon(\rho)-\langle\delta\epsilon\rangle]\times[\delta\epsilon(\rho')-\langle\delta\epsilon\rangle]\}\partial_{\zeta\zeta'}G(\rho,\rho')\,, \tag{8}$$

where the integration is performed in the new coordinates. For a macroscopically homogeneous composite, we can replace the term in the curly brackets by its volume average, i.e., by the correlation function

$$g(\rho-\rho')=\langle[\delta\epsilon(\rho)-\langle\delta\epsilon\rangle][\delta\epsilon(\rho')-\langle\delta\epsilon\rangle]\rangle\,,$$

which depends only on the relative vector $\mathbf{R}\equiv\rho-\rho'$. Assuming now that $g(\mathbf{R})$ decays to zero over a distance much smaller than the size of the system, we can replace G over most of the volume (except near the boundary) by

$$G_0(\mathbf{R})=1/4\pi|\mathbf{R}|\,.$$

Then the integration over one of the coordinates can be carried out immediately to yield the rescaled total volume $\Omega=V/\sqrt{\beta+1}$, and we are left with

$$\delta^2\epsilon_{\text{eff}}=-\frac{\beta+2}{\epsilon_0(\beta+1)}\int d^dR\,g(\mathbf{R})\partial_{\zeta\zeta'}G_0(\mathbf{R})\,. \tag{9}$$

This result depends only on the macroscopic homogeneity and the assumed short range of the geometrical correlations. To make further progress, we now assume that the composite is also isotropic, which implies that g is spherically symmetric in the original coordinates. Since (9) is performed in rescaled coordinates the correlation function g has an ellipsoidal symmetry. Namely, it is stretched $(-1<\beta<0)$ or contracted $(\beta>0)$ in the ζ direction. For $\beta=0$ the integral can be evaluated very easily, using the spherical symmetry,[3] leading to a result that depends only on $g(0)$.[3] Somewhat surprisingly, the integral can be evaluated exactly even when $\beta\neq0$. Consider the Fourier transforms of the Green's function $\tilde{G}_0=1/|\mathbf{k}|^2$ and of g:

$$\begin{aligned}\tilde{g}&=\int g_0(\mathbf{R})\exp[i(k_xx+k_yy+k_zz/\sqrt{\beta+1})]d^3R\\&=\frac{1}{\sqrt{1+\beta}}\tilde{g}_0\left[k_x,k_y,\frac{k_z}{\sqrt{\beta+1}}\right]\,,\end{aligned} \tag{10}$$

where g_0 is the spherically symmetric correlation function in the original coordinates. Since $\tilde{g}_0$ depends only on q^2, we conclude that

$$\tilde{g}(\mathbf{k})=(1+\beta)^{-1/2}\tilde{g}_0\left[k^2\left[1-\frac{\beta\cos^2\theta}{\beta+1}\right]\right]\,,$$

where θ is the polar angle between $\mathbf{k}$ and the ζ axis. Substituting this result and the Fourier transform of G_0, we can rewrite (9) as

$$\delta^2\epsilon_{\text{eff}}=-\frac{\beta+2}{(\beta+1)^{3/2}\epsilon_0}\int d^3R\frac{d^3q}{(2\pi)^3}\frac{d^3k}{(2\pi)^3}\exp(-i\mathbf{k}\cdot\mathbf{R})\tilde{g}_0(\mathbf{k})\exp(-i\mathbf{q}\cdot\mathbf{R})\frac{q_\zeta^2}{|q|^2}\,. \tag{11}$$

The integrations over **R** and **q** can now be carried out, and the remaining integral over **k** can be simplified in polar coordinates by a change of the radial variable, namely, $K \equiv [1-\beta\cos^2\theta/(\beta+1)]^{1/2}k$. This leads to

$$\delta^2\epsilon_{\text{eff}} = \frac{\beta+2}{2\epsilon_0(\beta+1)^{3/2}}\int_0^\infty \tilde{g}_0(K^2)\frac{4\pi K^2}{(2\pi)^3}\int_{-1}^{1} u^2[1-\beta u^2/(\beta+1)]^{-3/2}du \,. \tag{12}$$

The integral over K we identify as $g_0(0)$ while the second integral can be evaluated exactly. Thus we finally get

$$\delta^2\epsilon_{\text{eff}} = -\frac{\beta+2}{\epsilon_0\beta(\beta+1)^{1/2}}g_0(0)[\sqrt{\beta+1}-\sqrt{(\beta+1)/\beta}\arcsin\sqrt{\beta/(\beta+1)}] \,. \tag{13}$$

Note that this expression is valid for both $\beta>0$ and $-1<\beta<0$ by analytic continuation. Recalling our definition of the correlation function, we have

$$g_0(0) = \langle[\delta\epsilon(\rho)-\langle\delta\epsilon\rangle]^2\rangle = \langle\epsilon^2\rangle - \langle\epsilon\rangle^2 \,,$$

and summing the zero- and first-order contributions to ϵ_{eff}, we finally get

$$\epsilon_{\text{eff}} = \langle\epsilon\rangle - \frac{\beta+2}{2\epsilon_0\beta}\left[1-\frac{\arcsin\sqrt{\beta/(\beta+1)}}{\sqrt{\beta}}\right](\langle\epsilon^2\rangle-\langle\epsilon\rangle^2) \,, \tag{14}$$

which is exact to order $\delta\epsilon^2$. The generalization to other dimensions is straightforward and will be presented elsewhere.[7] As $\beta\rightarrow 0$, (14) reduces to the well-known expression for ϵ_{eff} of the linear problem: namely,[3]

$$\epsilon_{\text{eff}} = \langle\epsilon\rangle - \tfrac{1}{3}(\langle\epsilon^2\rangle-\langle\epsilon\rangle^2) \,.$$

To summarize, we have studied the behavior of an inhomogeneous nonlinear composite with small fluctuations in the local dielectric constant. We solved for the first correction to the potential field, in terms of the Green's function, and used it to evaluate the second-order correction to the bulk effective dielectric constant ϵ_{eff}. We find that to this order, the result is insensitive to the local microgeometry and depends only on the variance of the global distribution.

The method we employed to obtain our results is a generalization of a well-known procedure of treating linear composite dielectrics with no additional assumptions involved. As such it constitutes one of the very few cases where such a generalization is possible. An alternative approach to the problem studied here focuses on a calculation of $D(\mathbf{r}) = \nabla A(\mathbf{r})$ instead of $E(\mathbf{r}) = -\nabla\Phi(\mathbf{r})$. This approach leads to the same result as (14) (see Ref. 7) as is known from the study of the linear case.[3] An extension of Eqs. (6) and (9) to arbitrary order in $\delta\epsilon$ is possible and will be discussed elsewhere.

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[1]See, e.g., review by R. Landauer, in *Electrical, Transport, and Optical Properties of Inhomogeneous Media,* edited by J. C. Garland and D. B. Tanner, AIP Conf. Proc. No. 40 (American Institute of Physics, New York, 1978), and references therein.

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