Comment on "Finite-element modeling method for the study of dielectric relaxation at high frequencies of heterostructures made of multilayered particle" [J. Appl. Phys. 102, 124107 (2007)]

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(Received 10 April 2008; accepted 28 August 2008; published online 10 November 2008)

The article of Fourn *et al.* [J. Appl. Phys. **102**, 124107 (2007)] uses the numerical finite-element method to study the dielectric relaxation of a square array of coated cylinders. The comment here shows that similar results can be calculated immediately from the appropriate analytical expression. Our results are also compared to some figures in the article of Fourn *et al.* [J. Appl. Phys. **102**, 124107 (2007)]. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3009672]

I. ANALYTIC RESULTS

In Ref. 1, first-principles calculations of the effective complex permittivity of a regular array of two-layered cylinders are made, by the finite-element method. Here, the equivalent analytical formula is used to show that similar results can be reproduced immediately. The analytic method is explicit of the intrinsic physical principles and easier to manipulate than the numerical method. Moreover, analytical methods provide a reliable means of verifying the validity of a numerical result.

We denote the volume fraction of the cylindrical phase as Φ, for consistency with Ref. 1. The analytical expression for the effective permittivity of a square array of *homogeneous* cylinders in Ref. 2 includes multipole terms up to Φ^8. We employ this formula and generalize it to the case of a square array of two-layered cylindrical inclusions. Like the expression given for the case of a simple-cubic structure of multilayered spheres given in Ref. 3, we here obtain a similar expression for a square array of coated cylinders.

The unit cell of the coated cylindrical inclusion embedded in the matrix is illustrated in Fig. 1. Denote the ratio $t = e/R$, where e is the thickness of the shell and R is the outer radius of the cylinder.

The effective permittivity of the square array of two-layered cylinders is given by

$$\frac{\varepsilon}{\varepsilon_1} = 1 - 2\Phi\left[\frac{1}{T} + \Phi + C_1\Phi^4 + C_2\Phi^8 + O(\Phi^{12})\right]^{-1}, \tag{1}$$

$$T = -\frac{1 + (1-t)^2 L_{32}L_{21}}{(1-t)^2 L_{32} + L_{21}}, \tag{2}$$

$$L_{32} = \frac{\rho_{32} - 1}{\rho_{32} + 1}, \quad L_{21} = \frac{\rho_{21} - 1}{\rho_{21} + 1}, \quad \rho_{ij} = \frac{\varepsilon_i}{\varepsilon_j}, \tag{3}$$

$$C_1 = -\frac{0.305\,827T}{T^2 - 1.402\,958\Phi^8}, \quad C_2 = -\frac{0.013\,362}{T}. \tag{4}$$

In Eqs. (1)–(4), term T is obtained by applying mixture formula with the dipole approximation, terms L_{32} and L_{21} are obtained by applying continuity conditions of the electromagnetic field at the interfaces of the two-layered cylinders, and terms C_1 and C_2 are given in Ref. 2 by lattice sum techniques. Incidentally, Ref. 2 also includes analysis of a hexagonal array of cylinders which yields an expression for ε similar to that in Eq. (1), but with different coefficients and exponents of volume fraction. Further, the analytic method of solution can be employed to obtain a solution for an array of elliptic cylinders.[4]

We denote F as the frequency, and use the same horizontal axis, $\log(F/\mathrm{GHz})$, as in Ref. 1. To assess the accuracy of our analytical expression (1), we plot the real part of the effective permittivity of the mixture versus $\log(F/\mathrm{GHz})$ to various orders. Approximations up to order Φ^1, Φ^4, and Φ^8 in the square-bracketed term on the right-hand side of Eq. (1) are plotted in Fig. 2. As shown, when Φ is less than 0.5, there is nearly no difference between these curves for the dielectric parameters used. However, when Φ grows larger than 0.5, the multipole terms should be taken into account and the difference between these curves becomes significant. It can also be seen from Fig. 2 that there is no observable difference between curves calculated with approximation up

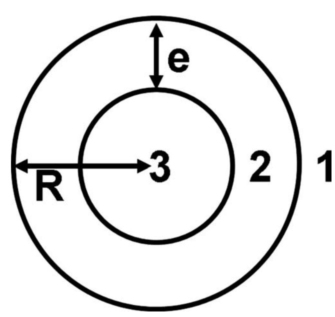

FIG. 1. Schematic illustration of one unit cell of a square array of two-layered cylinders embedded in a matrix. Indices 1, 2, and 3 denote the matrix, shell, and core, respectively.

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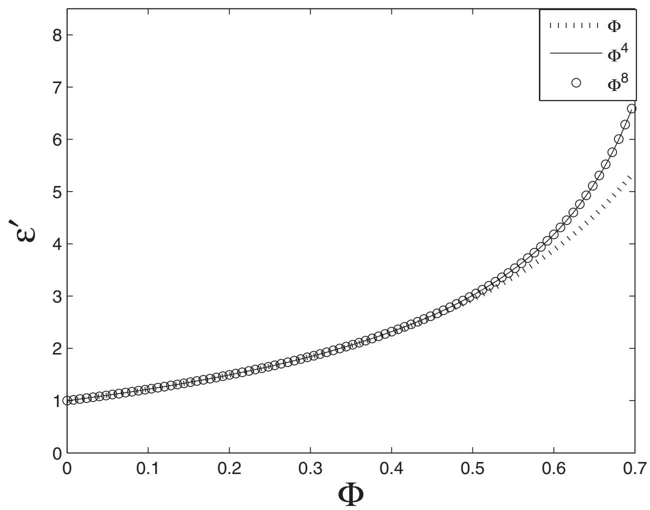

FIG. 2. The relative effective permittivity of the mixture vs volume fraction with approximation up to Φ, Φ^4, and Φ^8. The values of permittivity of core, background, and shell used in calculation are: $\varepsilon_3=10$, $\varepsilon_1=1$, and $\varepsilon_2=1+j\sigma/(\omega\varepsilon_0)$ with $\sigma=10^3$ S/m and $\varepsilon_0=8.85\times10^{-12}$ F/m. The ratio between shell thickness and radius of particle used in calculation is $t=0.3$. The value of $\log(F/\mathrm{GHz})$ used in calculation is $\log(F/\mathrm{GHz})=3$.

to Φ^4, and up to Φ^8, even when the volume fraction Φ is 0.7. This shows that the approximation including terms of order Φ^8 is accurate enough in this problem where the contrast between the core permittivity and the matrix permittivity is $\varepsilon_3/\varepsilon_1=10$. In the case of higher contrast, terms of higher order than Φ^8 may be needed to maintain accuracy. The maximum value of Φ considered in Ref. 1 is 0.7, which is close to the theoretical maximum $\pi/4\approx0.785$.

II. COMPARISON OF THE RESULTS

To compare the results, we here use the same parameters as in Fig. 2 (the case with $\Phi=0.4$), Figs. 7 and 8 of Ref. 1. In all three of these figures, $\Phi=0.4$, $\varepsilon_2=1+j\sigma/(\omega\varepsilon_0)$ with $\sigma=10^3$ S/m and $\varepsilon_0=8.85\times10^{-12}$ F/m.

In Figs. 3 and 4, the value of t is tuned whereas $\varepsilon_1=1$ and $\varepsilon_3=10$. In Figs. 5 and 6, the value of ε_3 is tuned with $\varepsilon_1=1$ and $t=0.1$. In Figs. 7 and 8, the value of ε_1 is tuned with $\varepsilon_3=10$ and $t=0.1$.

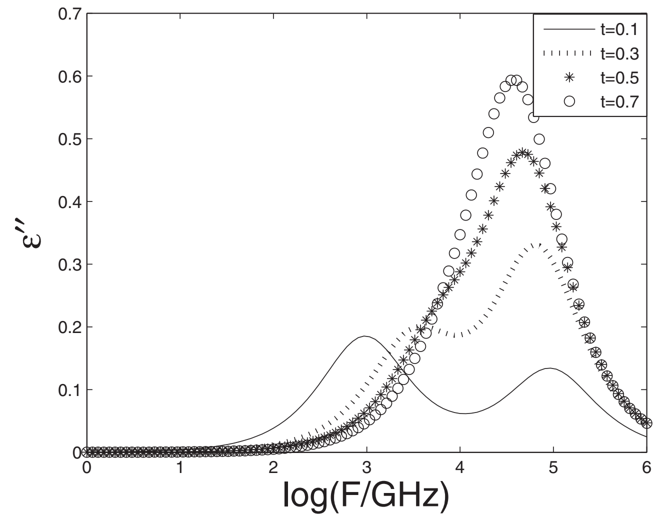

FIG. 4. As for Fig. 3 but for the imaginary part of the permittivity.

Comparing with the corresponding figures in Ref. 1, we obtain the same results, exhibiting the same characteristic double-Debye-like relaxation process. The analytic formula presented here is, however, much more readily computed than those obtained using a finite-element method.

There are two particular comments that need to be made regarding statements made in Ref. 1. First, it is not appropriate to compare the analytic results in Fig. 4 of Ref. 3 with the numerical results in Ref. 1, because the treatment of Ref. 3 is for two-layered spheres arranged on a simple-cubic lattice, which is different from the case of a square array of cylinders as treated in Ref. 1. Second, the results presented in Ref. 3 are calculated using analysis that takes into account multipolar interactions between the particles, not on the basis of a dipole approximation, as stated in Ref. 1. The multipole

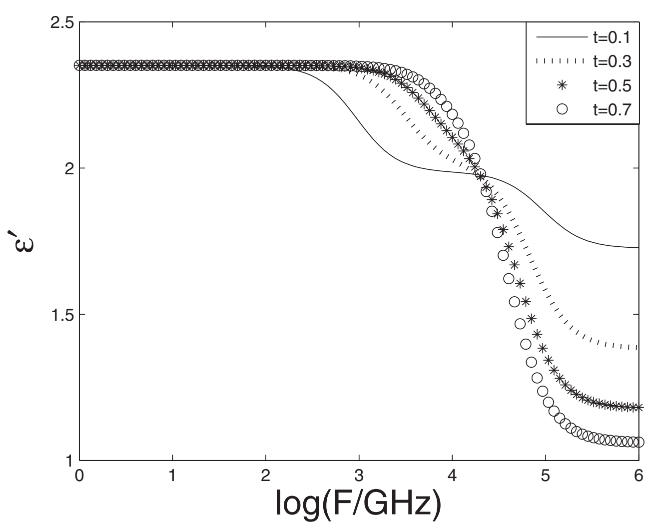

FIG. 3. Real part of permittivity calculated from analytic expression (1) for a square array of two-layered cylinders with different values of ratio $t=0.1$, 0.3, 0.5, and 0.7. Other values used in calculation are: $\varepsilon_3=10$, $\varepsilon_1=1$, and $\Phi=0.4$.

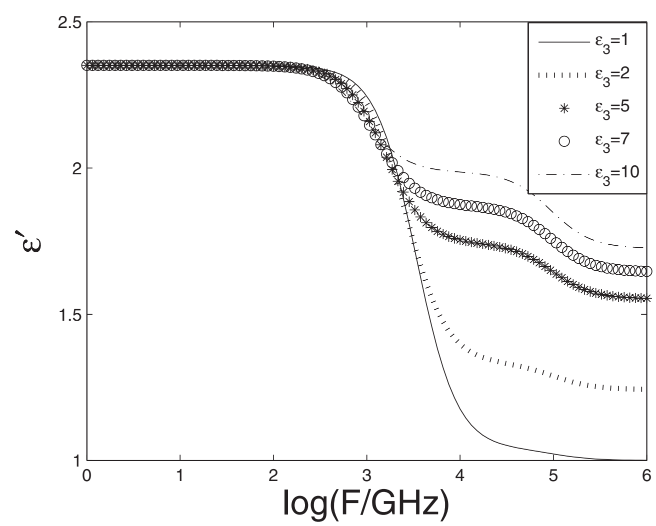

FIG. 5. Real part of permittivity calculated from analytic expression (1) for a square array of two-layered cylinders with different values of the permittivity $\varepsilon_3=1$, 2, 5, 7, and 10. Other values used in calculation are: $\varepsilon_1=1$, $t=0.1$, and $\Phi=0.4$.

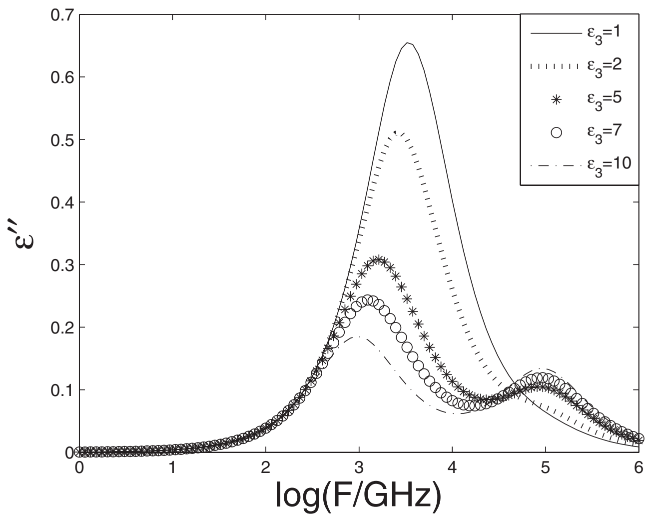

FIG. 6. As for Fig. 5 but for the imaginary part of the permittivity.

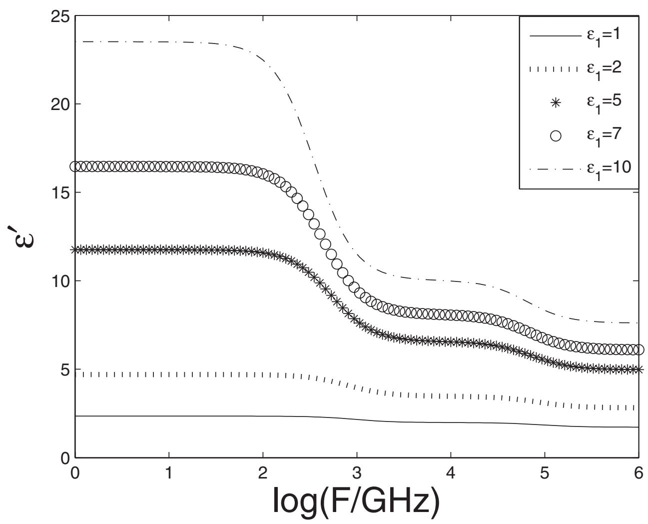

FIG. 7. Real part of permittivity calculated from analytic expression (1) for a square array of two-layered cylinders with different values of the permittivity $\varepsilon_1=1$, 2, 5, 7, and 10. Other values used in calculation are: $\varepsilon_3=10$, $t=0.1$, and $\Phi=0.4$.

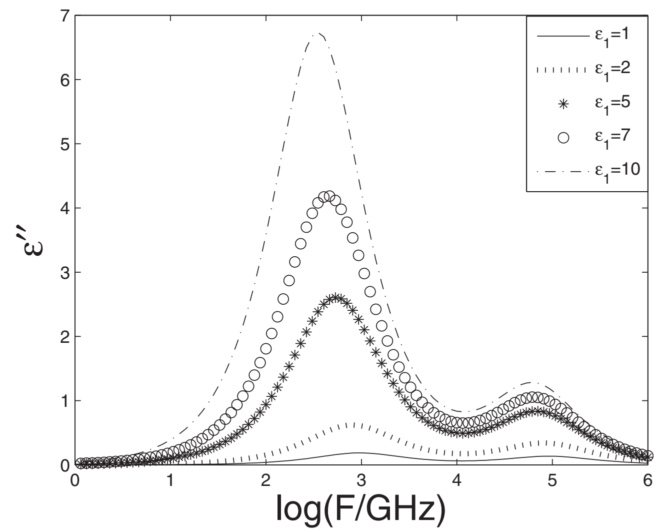

FIG. 8. As for Fig. 7 but for the imaginary part of the permittivity.

equation used in Ref. 3 is similar to that shown here in Eq. (1), but appropriate for coated spheres on a simple-cubic lattice.

Although the numerical methods do well in solving complex problems when it is hard to solve by hand, analytic methods provide insight into the underlying physics, and are more simple to implement. As for the study on dielectric behavior of a regular array of coated cylinders, it seems that analytical solutions are sufficient. If the problem becomes more complicated, however, such as in the case of surface roughness on the inclusion, or if the coating contains an aperture, then numerical methods provide a powerful tool to obtain solutions. Nonetheless, it should be kept in mind that the validity of numerical methods should be tested, and analytic methods provide one means of verification in limiting cases.

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