Local field probability distribution in random media

Marc Barthélémy
Commissariat à l'Energie Atomique, CELV–Service de la Matière Condensée, 94195 Villeneuve-Saint-Georges Cedex, France

Henri Orland
Commissariat à l'Energie Atomique, Service de Physique Théorique, 91191 Gif-sur-Yvette Cedex, France
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We address the problem of the analytical determination of the local field distribution in random dielectrics. This distribution was shown numerically to exhibit a double peak behavior. We have determined analytically this distribution which is essentially the convolution of the distribution due to the first neighbors (i.e., the effect of the microstructure) with a Gaussian distribution describing the background. Our results are in good agreement with known numerical results. [S1063-651X(97)14209-X]

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I. INTRODUCTION

In the study of transport properties of disordered systems (and for nonlinear disordered systems), the random resistor network (RRN) is of the utmost importance. First, it provides a simple model for composite media such as conductor-dielectric mixtures and exhibits a percolation transition (in the dc case). Second, it can be generalized to a large number of situations such as ac properties, optical properties, etc. (see for instance [1]).

The main approach to this problem was to determine the effective conductivity of the network (see the review [2] and references therein). The effective medium concept is already an old one (one can find a historical review of effective medium in [3]) and consists in replacing the heterogeneous medium by a homogeneous one with the same macroscopic conductivity (this is in general possible when the observation length is greater than the disorder correlation length). The knowledge of the effective conductivity is equivalent to the knowledge of the averaged electric field and is therefore remotely related to the microstructure of the medium. More precisely, the usual effective medium theories neglect the microstructure by assuming a homogeneous local environment. During the past few years, the effect of the random microstructure has been investigated (see, for example, [4]). One way to study this problem is to look at the moments of the electric field. This information is of interest in many problems such as the weakly nonlinear composites (see [5–7] and references therein). Indeed, since the nonlinear susceptibility is related to higher moments of the electric field, it is much more sensitive to the fluctuations due to the microstructure. Moreover, in the random fuse problem (a model for dielectric breakdown and for rupture in heterogeneous media), the knowledge of the probability distribution allows for the determination of very important quantities such as the macroscopic breakdown threshold (for a review on fracture in disordered media, see, for example, [8]). Most of the approaches to these problem are based on numerics, scaling, extreme statistics (Lifchitz defects).

In spite of the importance of the subject there are very few analyses of this problem; even the weak-disorder regime is not well understood. We propose here a perturbative approach to the calculation of the full probability distribution. This approach could be used as a starting point for a strong disorder regime analysis. We determine analytically the probability distribution of the local field (which is the sum of the fields seen by the particle). In general, when an electric field is applied on a dielectric, the local field differs from the macroscopic field. This difference comes from the local environment of the particle. However it is not difficult to compute the average of this local field. This average has been first calculated by Lorentz and leads to the usual solution for the local field and to the Clausius-Mossotti relation [3]. Since the local environment is random, one expects the local field to be random too. In 1991, Chen and Sheng [9] proposed a numerical study of this problem (using a generalized Onsager approach) and found a double peak distribution for the local field. This double peak structure was also found numerically in the context of the RRN [10]. It can be related to the existence of two different environments relative to the applied field. We will return to the physical interpretation of this phenomenon. Up to now analytical efforts were made on the probability distribution tail ([11] and references therein) but not on the full distribution. We propose here a perturbative approach to the calculation of the full probability distribution.

The layout of the paper is the following. In Sec. II, we present the perturbative approach to the calculation of the probability distribution of the local electric field and compare it to previous numerical results. Section III summarizes the main conclusions of this work.

II. PERTURBATIVE STUDY OF THE PROBABILITY DISTRIBUTION

A. Calculation

We present a perturbative approach for the calculation of the local field probability distribution. Note that the electric field (as will be shown below) is simply related to the local field.

We will show that this problem reduces to the calculation of the probability distribution of a weighted sum of random variables. We then evaluate perturbatively the probability distribution of such a sum.

We model here the random dielectric as a three-dimensional cubic lattice (with spacing $a=1$) where the nodes are occupied randomly by polarizable particles (with polarizability α). We suppose that a particle is present with probability p and absent with probability $1-p$. The polarizabilities $\alpha(\mathbf{r})$ at $\mathbf{r}$ are independent (from site to site) random variables distributed according to a binary law

$$P(\alpha(\mathbf{r}))=p\,\delta(\alpha(\mathbf{r})-\alpha)+q\,\delta(\alpha(\mathbf{r})). \tag{1}$$

We apply a uniform and constant electric field $\mathbf{E}_0$ to this dielectric medium. The local field $\mathbf{E}_l(\mathbf{r}_i)$ at a point $\mathbf{r}_i$ is given by the sum of all the fields created by the other dipoles (the dipole $\mathbf{r}_i$ is excluded from the sum)

$$\mathbf{E}_l(\mathbf{r}_i)=\mathbf{E}_0+\sum_{\mathbf{r}_j\neq\mathbf{r}_i}\hat{G}(\mathbf{r}_i-\mathbf{r}_j)\mathbf{p}(\mathbf{r}_j), \tag{2}$$

where the dipolar moment $\mathbf{p}(\mathbf{r}_i)$ at point $\mathbf{r}_i$ is given by

$$\mathbf{p}(\mathbf{r}_i)=\alpha(\mathbf{r}_i)\mathbf{E}_l(\mathbf{r}_i) \tag{3}$$

and where $\hat{G}$ is the dipolar tensor (in real space and for $\mathbf{r}\neq 0$)

$$G_{\mu\nu}(\mathbf{r})=\frac{3r_\mu r_\nu-r^2\delta_{\mu\nu}}{r^5} \tag{4}$$

and $G_{\mu\nu}(r=0)=-\delta_{\mu\nu}/3$. The local field thus satisfies the following integral equation:

$$\mathbf{E}_l(\mathbf{r}_i)=\mathbf{E}_0+\sum_{\mathbf{r}_j\neq\mathbf{r}_i}\hat{G}(\mathbf{r}_i-\mathbf{r}_j)\alpha(\mathbf{r}_j)\mathbf{E}_l(\mathbf{r}_j). \tag{5}$$

The electric field is simply related to the local field by $E(\mathbf{r}_i)=E_l(\mathbf{r}_i)+G(0)\alpha(\mathbf{r}_i)E_l(\mathbf{r}_i)$, where $\mathbf{E}_l(\mathbf{r})$ and $\alpha(\mathbf{r})$ are independent random variables.

Equation (5) gives formally the local field as a function of the disorder. It seems out of reach to solve exactly this equation in order to extract the probability distribution of the local field $\mathbf{E}_l$. Due to translational invariance, the disorder averaged probability distribution is position independent and we choose $\mathbf{r}_i=0$ in Eq. (5). We study Eq. (5) perturbatively. To first order in α, we have

$$\mathbf{E}_l(0)\simeq\mathbf{E}_0+\sum_{\mathbf{r}_j\neq 0}\hat{G}(\mathbf{r}_j)\alpha(\mathbf{r}_j)\mathbf{E}_0. \tag{6}$$

At first sight, this approximation seems too simple but we will show that it is surprisingly accurate, at least for the average field. To discuss this point, let us assume that the dielectric material has permittivity ϵ_0 and the particles have a permittivity ϵ. In this case, the polarizability at $\mathbf{r}$ reads (see, for example, [3])

$$\alpha(\mathbf{r})=\frac{3[\epsilon(\mathbf{r})-\epsilon_0]}{2\epsilon_0+\epsilon(\mathbf{r})}. \tag{7}$$

It is then easy to compute the average fluctuation of the total field $\overline{\delta\mathbf{E}}=\overline{\mathbf{E}-\mathbf{E}_0}\simeq\overline{\alpha}\sum_{\mathbf{r}}G(\mathbf{r})\mathbf{E}_0$ to first order in α [where the bar denotes the average over the distribution (1)]. This fluctuation should be equal to zero if we replace the surrounding medium of permittivity ϵ_0 by an effective medium of permittivity ϵ^*. We find

$$\overline{\frac{\epsilon^*-\epsilon}{1+(\epsilon-\epsilon^*/3\epsilon^*)}}=0 \tag{8}$$

which is the well known Bruggeman formula [3]. This shows that, at least for the average field, the approximation (6) is not trivial and contains a lot of information. We also note that this approximation consists of considering all dipoles polarized along the applied field $\mathbf{E}_0$. The same approximation, restricted to the first neighbors, was proposed in [9] in order to justify the double peak character of the local field distribution. In the following, we will study Eq. (5) more thoroughly without restricting ourselves to the first neighbors.

We assume that the applied field is directed along the z axis and we first look at the z component of the local field $\mathbf{E}_l$ denoted by $\mathbf{E}_\parallel$. This component satisfies the equation

$$\mathbf{E}_\parallel=\mathbf{E}_0+\sum_{\mathbf{r}_i\neq 0}f(\mathbf{r}_i)\alpha(\mathbf{r}_i)\mathbf{E}_0, \tag{9}$$

where

$$f(\mathbf{r})=G_{zz}(\mathbf{r})=\frac{3z^2-r^2}{r^5}. \tag{10}$$

Equation (9) is the basic equation we study here. Let us introduce the quantity Y given by

$$Y=\sum_{\mathbf{r}_i\neq 0}f(\mathbf{r}_i)\alpha(\mathbf{r}_i). \tag{11}$$

The problem is thus reduced to the determination of the probability distribution of the quantity Y. This quantity is the sum of random binary variables $\alpha(\mathbf{r})$ weighted by $f(\mathbf{r})$ which is a decreasing function of r (roughly like $1/r^3$). The quantity Y remains distributed since f decreases sufficiently rapidly so that the central limit theorem (CLT) cannot apply. This is easily seen by the following two extreme cases. (i) The quantity f is local, for example f is nonzero only for one value of $\mathbf{r}$. In this case, Y is equal (up to a constant) to $\alpha(\mathbf{r})$ and will therefore be a random binary variable. (ii) The quantity f is approximately constant. The CLT can therefore apply and Y will be distributed according to a Gaussian law with a relative dispersion varying like $1/\sqrt{N}$, where N is the number of dipoles.

Our present case is intermediate, since at fixed r the number of dipoles is of order r^2 and the total contribution of this shell is of order $r^2\times 1/r^3\sim 1/r$, which is not integrable. Thus sufficiently far away from the origin, the variable associated to a shell is given by the CLT. We thus expect a contribution due to the first shell (the first neighbors) and a Gaussian contribution coming from all other shells. This is the central argument which allows us to study the probability distribution of Y, namely, $P(Y)$.

From Eq. (11), the expression for $P(Y)$ is

$$P(Y)=\int \frac{dk}{2\pi}\, e^{ikY+\Sigma_{\mathbf{r}_i\neq 0}\ln[pe^{-ik\alpha f(\mathbf{r}_i)}+q]}. \tag{12}$$

Let us note here that if f is constant the sum reduces to a factor N which allows us to use a saddle-point method and leads to the usual Gaussian distribution (CLT).

Following the preceding arguments, we divide the sum in two parts,

$$\sum_{\mathbf{r}_i\neq 0} \ln[p^{-ik\alpha f(\mathbf{r}_i)}+q]=\sum_{|\mathbf{r}_i|=1} \ln[p^{-ik\alpha f(\mathbf{r}_i)}+q] + \sum_{|\mathbf{r}_i|>1} \ln[p^{-ik\alpha f(\mathbf{r}_i)}+q], \tag{13}$$

where the first sum of the right-hand side is over the first six neighbors. We keep this first sum in the integral and apply a saddle-point method to the second sum. Let us briefly justify this point. The second term can be written as

$$\sum_{|\mathbf{r}_i|>1} \ln[p^{-ik\alpha f(\mathbf{r}_i)}+q]=\sum_{\text{shells}\ n} N(n)\ln[p^{-ik\alpha f_n}+q], \tag{14}$$

where $N(n)$ is the number of sites belonging to the shell number n (we mean here by shell the positions in space for which f has a constant value) and where f_n is the value of f on this shell. We assume that all these $N(n)$ are sufficiently large in order to enable the use of the saddle-point method [$N(n)$ is roughly varying like n^2]. The saddle point is at $k=0$. This leads to

$$P(Y)\simeq \int \frac{dk}{2\pi}\,(pe^{-ik\alpha f_1}+q)^4(pe^{-ik\alpha f_2}+q)^2 \times e^{ikY-(k^2/2)m_2' pq}, \tag{15}$$

where

$$m_2'=\sum_{|\mathbf{r}_i|>1}\left[f(\mathbf{r}_i)\right]^2 \tag{16}$$

and $f_1=-1$ is the value of G_{zz} for the four dipoles in the plane perpendicular to the z axis and $f_2=2$ is the value of G_{zz} for the two first neighbors on the z axis. The fact that f_1 and f_2 have different signs explains the double peak character of $P(Y)$. Indeed, the local environment comprising mainly dipoles in the (x,y) plane will produce a negative field and therefore a value of the local field lower than its average. On the other hand, the local microstructure with the dipoles on the z axis will produce a local field above its average value. The competition between these two effects leads to the double peaked distribution. It is now easy to compute $P(Y)$ and we obtain

$$P(Y)\simeq \frac{1}{\sqrt{2\pi pqm_2'}}\sum_{l=-4}^{+4} C_l e^{-(1/2\mu_2)(Y+l\alpha)^2}, \tag{17}$$

where

$$\begin{aligned} C_{-4}&=p^2q^4,\\ C_{-3}&=4p^3q^3,\\ C_{-2}&=2pq^5+6p^4q^2,\\ C_{-1}&=4p^5q+8p^2q^4,\\ C_0&=q^6+12p^3q^3+p^6,\\ C_1&=4pq^5+8p^4q^2,\\ C_2&=2p^5q+6p^2q^4,\\ C_3&=4p^3q^3,\\ C_4&=p^4q^2, \end{aligned} \tag{18}$$

and where $\mu_2=m_2' pq$ (numerically for a lattice spacing $a=1$, one finds $m_2'\simeq 1.36$).

Equation (17) [with Eq. (18)] is our final result. Equation (17) is essentially the convolution of the local probability distribution (which is the signature of the microstructure) with a Gaussian distribution representing the background. The width μ_2 of the Gaussian distributions is finite in the infinite volume limit, in contrast with the usual case (CLT), where this quantity is extensive. Moreover, this quantity is zero when $p=0$ or $p=1$, as expected since there is no disorder in these cases. Note that $P(Y)$ is invariant under the transform $p\to q$, $q\to p$, and $Y\to -Y$. This indicates that the function $P(Y)$ is even for $p=q=1/2$.

We can apply the same argument to the transverse field. The total field produced by the first neighbors is equal to zero. Thus the effect of the local microstructure is zero. The only contribution is thus the background, which is a Gaussian distribution with zero mean and a variance given by

$$\Delta=\alpha^2 pq\sum_{r>1}[G_{zz}(r)]^2=\alpha^2 pq\sum_{r>1}\left(\frac{3xz}{r^5}\right)^2 \tag{19}$$

(where $\Sigma_{r>1}[G_{zz}]^2\simeq 1.72$). This variance is therefore of order α^2 in agreement with [9].

The averaged local field is $\mathbf{E}_0$ and the normalized field $E_\parallel/E_0$ (which is still denoted by $E_\parallel$ in the following) is simply given by $E_\parallel=1+Y$.

B. Discussion and comparison with numerical results

Let us recall that in our calculation, two different approximations are made: a low polarizability expansion and a saddle-point method.

First of all, we checked that our analysis of Eq. (11) is correct (validity of the saddle-point method). For this, we studied numerically the probability distribution of Y and compared it with the distribution we obtained analytically [Eq. (17)]. The results for differents values of the concentration p are shown in Figs. 1(a)–1(c). The agreement is excellent. This shows that only the first neighbors contribute to the nonuniversal (non-Gaussian) behavior. Let us comment briefly these three curves. We call type-I sites the two sites at $z=-1,1$ (and $x=y=0$) and type-II sites the four nearest neighbors lying in the (x,y) plane (this is the same denomi-

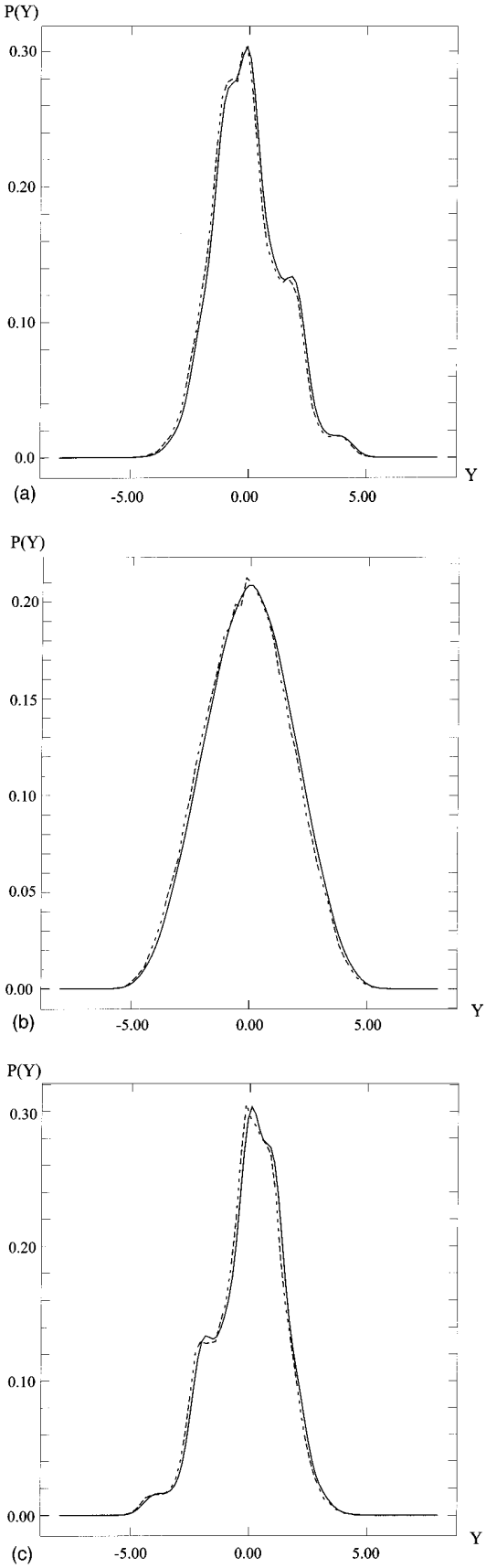

FIG. 1. Probability distribution $P(Y)$ for differents values of p with $\alpha=1$: (a) $p=0.2$, (b) $p=0.5$, (c) $p=0.8$. The solid line represents the analytical result [Eq. (17)] and the dotted line represents the numerical simulation of $P(Y)$ from Eq. (17).

nation as in [9]). A dipole on a type-II site creates a field at $r=0$ equal to -1 (for a dipole moment equal to 1).

(i) For $p=0.2$ [Fig. 1(a)], we have on the average one dipole (on the six neighbors). Since there are four type-II sites (over six) the probability is high to find this dipole on a type-II site and will produce a negative contribution to the local field. This corresponds to the high peak below $Y=0$. There is, however, a small probability (equal to 1/3) that the dipole will be on a type-I site and produce a positive field. This explains the double peak character of $P(Y)$.

(ii) For $p=0.5$ [Fig. 1(b)], there are approximately three dipoles. The most probable configuration is two dipoles lying on type-II sites and one lying on a type-I site. This will contribute to the local field with a value given by $1\times(2)+2\times(-1)=0$. This explains the one peak distribution centered at $Y=0$.

(iii) For $p=0.8$ [Fig. 1(c)], there are about five dipoles. The most probable configuration is three dipoles on type-II sites and two dipoles lying on type-I sites. This will create a field equal to $3\times(-1)+2\times(2)=+1$, which explains the highest peak in the positive region. The lower peak corresponds to a less probable configuration for which the contribution to the local field is equal to $4\times(-1)+1\times(2)=-2$.

We now compare our results with known numerical results [9]. We first plotted $P(E_\parallel)$ versus $E_\parallel$ for different values of p [see Figs. 2(a)–2(c)]. For any concentrations, the agreement is good, at least qualitatively. The positions of the peaks are well reproduced by our approximation. However, we observe slight differences. They may be attributed to the error bars in the numerical results of [9] or more probably to the approximation (6) which assumes essentially a low polarizability. We then plotted $P(E_\parallel)$ versus $E_\parallel$ for different values of α [see Figs. 3(a)–3(c)]. We observe that for a low polarizability, the approximation (6) is good, which is not surprising since (6) is a low polarizability approximation. In order to improve our approximation, one would need to expand to second order in α. It has been shown [12] that a calculation to second order in α (which is technically much more involved) does not improve significantly the results (less than a few percent). The agreement between our analytical result Eq. (17) and the numerical results [9] might seem surprising, since Chen and Sheng did not use a low polarizability approximation. Let us make here a few comments. First, one can view the generalized Onsager method (used in [9]) as separating the first neighbors contribution (which we treated exactly at the first order in α) from the background contribution (which we treat by a saddle-point method and which was treated by an effective medium calculation in [9]). Second, one has to study the first neighbors contribution. It can be seen that the work [9] (which treats this contribution exactly) is restricted to low polarizabilities (all polarizabilities in [9] are less than 0.15). This may explain the good agreement between our result and [9]. One expects a worse agreement for higher polarizabilities. Moreover, it is difficult to quantify the range of validity of our approximation, or equivalently the convergence radius of the expansion in α, since the spectrum of the random operator $\hat{G}(\mathbf{r}_i-\mathbf{r}_j)\alpha(\mathbf{r}_j)$ is unknown. In any case, our approximation is expected to be valid for small α.

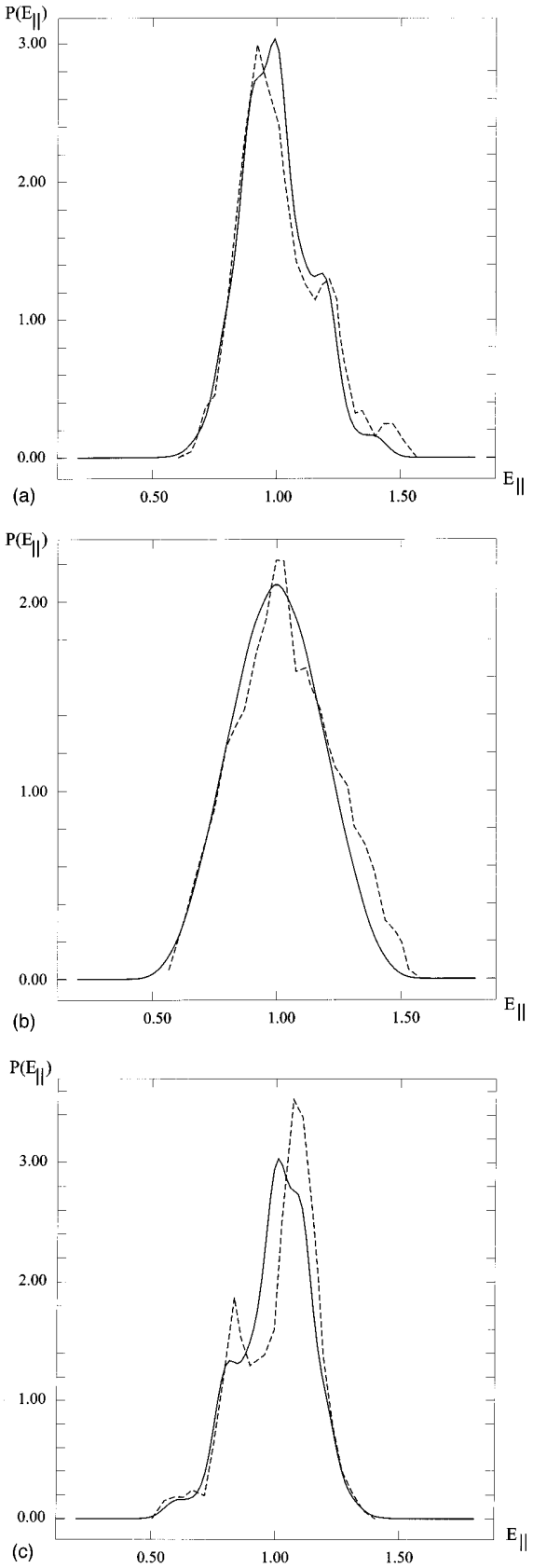

FIG. 2. $P(E_{\|})$ versus $E_{\|}$ for $\alpha=0.1$ and for different values of p: (a) $p=0.2$, (b) $p=0.5$, (c) $p=0.8$. The quantity $E_{\|}$ is here normalized to its average value. The solid line represents the analytical result [Eq. (17)] and the dotted line represents the numerical simulation of [9].

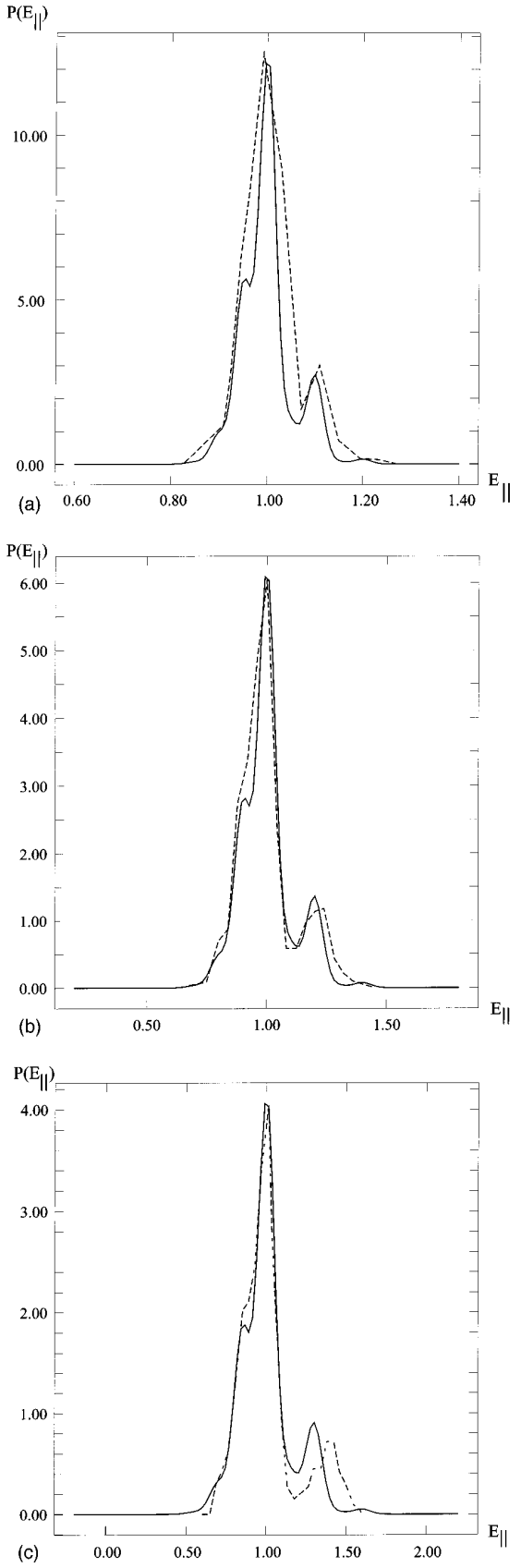

FIG. 3. $P(E_{\|})$ versus $E_{\|}$ for different values of α for the same value of $p=0.1$: (a) $\alpha=0.01$, (b) $\alpha=0.1$, (c) $\alpha=0.15$. The quantity $E_{\|}$ is here normalized to its average value. The solid line represents the analytical result [Eq. (17)] and the dotted line represents the numerical simulation of [9].

Finally, we see that for $E \gg 1$, the probability distribution behaves as e^{-cE^2}. From extreme statistics [11], one expect a decay like e^{-bE^d} (d is the space dimension and c and b are positive constants). This discrepancy may be attributed to the saddle-point approximation.

III. CONCLUSION

We have adressed the problem of the analytical determination of the local field distribution in random dielectrics. We find that for small polarizabilities it is a sum of Gaussian distributions and is essentially the convolution of the local distribution (effect of the local microstructure) with a Gaussian distribution which represents the background. We checked that our study is correct and compared our results with known numerical results. The agreement is good. In all cases, we reproduce the double peak character which arises from the existence of two main different local environments. This method can be easily applied to the current distribution in the RRN. This study is in progress.

It seems difficult to improve on our method, but it has the advantage of being easy to use in other cases, such as the quasistatic case. In that case, we expect important differences with the static case, since the dipolar field would be quite different. Another interesting point is that usual effective medium theories neglect the microstructure of the medium by assuming a homogeneous local environment. It would be interesting to incorporate the information embedded in $P(E_{\|})$ in an effective medium calculation.

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