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The bulk effective response of non-linear random resistor networks: numerical study and analytic approximations

Ohad Levy and David J Bergman

School of Physics and Astronomy, Raymond and Beverly Sackler Faculty of Exact Sciences, Tel-Aviv University, Tel-Aviv 69978, Israel

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Abstract. The effective response of non-linear random resistor networks consisting of two different types of resistor is studied numerically and compared with some simple approximations. One type of resistor is assumed to be ohmic, while the other is assumed to have a non-linear I–V response of the form $i = \chi v^{\beta+1}$. The effective response is calculated by solving Kirchhoff's equations for the voltages at each node of the network. Numerical results, for $\beta = 2$ and $\beta = 4$, are compared to theoretical predictions of a recently derived Clausius–Mossotti approximation for such networks. The Clausius–Mossotti results are found to provide a good description of the results of the simulations in cases of low contrast between the two components or a small fraction of the non-linear component in the network. It is also found that the range of validity of this non-linear Clausius–Mossotti approximation is larger than that of the classical Clausius–Mossotti approximation for linear two-component random resistor networks. An extension of Bruggeman's effective-medium approximation to this case is found to give somewhat better agreement with the numerical results.

1. Introduction

The non-linear properties of small particle composites have been extensively studied in recent years [1]. The composites of interest are usually made of non-linear (metal or semiconductor) particles embedded in a linear host. They are of particular interest because their non-linearities may be strongly enhanced relative to bulk samples of the same materials [1] and phenomena of intrinsic bistability may arise in them under certain conditions [2, 3]. These effects are the results of a possibly great enhancement of the electric field within the particles, which can be produced by an appropriate ratio of the host-to-particle conductivity and by a modification of the field inside a given particle by other neighbouring particles. The effective properties of such composites with particles of weak power-law non-linearity were theoreticaly treated by an effective medium type approximation [4, 5]. A recently published numerical study [6] concluded that this approximation agrees with numerical results for random resistor networks (RRNs) with low contrast between the two components and can also describe the qualitative behaviour of high-contrast networks. The properties of composites with particles of arbitrary non-linearity can be calculated by a recently derived extension of the Clausius–Mossotti (CM) approximation [7].

In this paper we report on numerical studies of mixed linear–non-linear RRNs with strong power-law non-linearity. The results are compared to those predicted by the CM approximation. It is well known that, for linear dielectrics, the CM approach gives a good description of the effective properties of composites in cases of small contrast between the two components or of small volume fraction of one of the components. The higher the contrast between the components, the smaller is the range, in terms of volume fractions, in which the approximation is good. We found that the non-linear CM approximation also provides a good description of the numerical results in these cases and that its range of usefulness, in terms of the volume fraction of the non-linear component, is even *larger* than that of the CM approximation for a linear composite with the *same* contrast between the components. We also propose a new type of approximation for systems of this kind, which is an extension of Bruggeman's symmetric effective-medium theory (EMT) [8]. It too is compared with the simulations and found to be better than the CM approximation for all the cases we studied.

The remainder of this paper is organized as follows. In section 2 we briefly describe the CM approximation for a linear host with non-linear inclusions and compare its predictions with results of simulations of an RRN model. In section 3 we present our extension of EMT for such systems, and compare it with the same numerical results. Our conclusions are briefly summarized in section 4.

2. Clausius–Mossotti approximation and numerical simulations

Consider a two-dimensional square lattice with fraction p of non-linear conductors and fraction $1-p$ of linear conductors. The linear conductors have an I–V response of the form $i = g_0 v$, where g_0 is the constant linear conductance. The I–V response of the non-linear conductors is $i = g(v)\,v$, where $g(v)$ is the voltage-dependent non-linear conductance and v is the voltage across the conductor. The effective response $g_e(v_e)$ of the random network is defined such that the network is equivalent to a full uniform network made of conductors that have this response. It can be calculated using a generalization of the CM approximation [7]. The CM (also known as the Maxwell-Garnett) approximation is one of the most widely used methods for calculating the bulk properties of linear composites [9]. It involves an exact calculation of the field induced in the uniform host by a *single* spherical or ellipsoidal inclusion and an approximate treatment of the interaction between the effects of different inclusions, which results in a *uniform* field *inside* all the inclusions. The generalization of this approximation to materials composed of a linear host and inclusions of arbitrary non-linearity is based on the well known observation [10] that the field inside an isolated inclusion of the above-mentioned shapes is uniform even when the inclusion is a non-linear material, irrespective of the form of the non-linearity. For a composite with many non-linear inclusions we can thus solve the electrostatic problem for the Lorentz local field and use that in order to find the uniform field inside the inclusions. In the discrete case of an RRN the problem can be similarly solved and the voltage v in each of the non-linear conductors is found to be given by [7]

$$(1-p)\left[g(v)-g_0\right]v + (z/2)g_0 v = (z/2)g_0 v_e \tag{1}$$

where z is the coordination number of the network and v_e is the average voltage. The effective conductance function of the network is found by a simultaneous solution of this relation and the equation

$$g_e(v_e) = g_0 + (z/2)pg_0\frac{g(v)-g_0}{(1-p)\left[g(v)-g_0\right]+(z/2)g_0}. \tag{2}$$

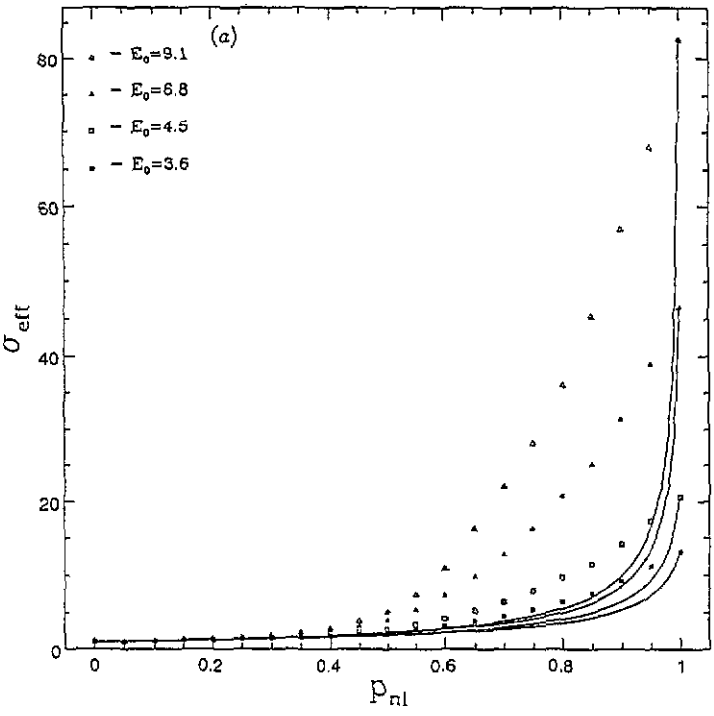
This conductance is voltage dependent. Its functional form depends on the magnitude of the applied or average voltage v_e and is in general different from that of the non-linear elements of the network [7]. In the linear case $g(v)$ is constant and equation (2) gives the well known linear CM result.

This result can be applied to any type of functional dependence $g(v)$. In this study the non-linear conductors are assumed to be strongly non-linear with $g(v) = g_1 v^{\beta}$, where g_1 and $\beta > -1$ are constants. (If $\beta \leq -1$ the uniqueness of the solution of Kirchhoff's equations cannot be guaranteed [11].) Two-dimensional square networks ($z = 4$) possess a special symmetry called duality [12]. The duality transformation consists of replacing each conductor g by a perpendicular conductor $g_d = 1/g$ and adjusting the boundary conditions so that the voltage on every new conductor is equal to the current flowing through the original one. It is clear that the network (named the dual network) satisfies Kirchhoff's equations. Its effective conductivity is given by $g_{ed}(v_{ed}) = 1/g_e(v_e)$. The CM approximation presented above preserves this invariance. $g_{ed}(v_{ed})$ can be calculated by equation (2) where g_0 is replaced by $1/g_0$ and $g(v)$ by $1/g(v)$. The voltage $v_{ed} = g_e(v_e)\,v_e$ is given by equation (1) where v_e is replaced by v_{ed}, g_0 by $1/g_0$ and v by $v_d = g(v)\,v$. This will give the same result as $g_{ed}(v_{ed}) = 1/g_e(v_e)$. In our case of power low non-linearity the local constitutive relation in the dual network is $g_d(v_d) = (1/g_1^{\beta+1})v^{-\beta/(\beta+1)}$. Thus, it is possible to relate networks of two different non-linearity exponents β and $-\beta/(\beta+1)$.

Simulations were carried out for $\beta = 0, 2$ and 4 on 10×10 square lattices with boundary conditions of zero voltage on one edge and a constant finite voltage on the opposite edge. At the two perpendicular edges a zero-current boundary condition was applied at all nodes. Both g_0 and g_1 were kept finite and calculations were performed for various values of p and applied external voltage. Kirchhoff's equations for the voltages at each node were solved self-consistently using an iterative relaxation algorithm and the value of the effective conductance was extracted from the calculated total dissipation in the network. The final result for each calculation is an average over 100 different configurations. The error bars in all following graphs are of the order of the size of the data points or less. The values two and four for β were chosen because they represent the lowest-order non-linear corrections to the linear response of materials with spatial inversion symmetry. In these materials the non-linear effects begin with cubic terms in the expansion of the electric current. In general, this expansion contains all odd powers of the electric field.

The algorithm used converges very slowly because of the non-linearity of the equations. It becomes slower as the contrast between the components increases. In this study the contrast between the components was kept finite so that we are always away from the percolation critical point. The systems were also kept small due to practical limitations of computing power. We did not try to investigate the percolation behaviour of such systems nor to extract critical exponents. Thus, finite size effects should have minor significance and our choice of 10×10 square lattices is sufficient to show the effective response of such systems.

We computed the effective conductance of two-dimensional RRNs as described above. Typical results are shown in figure 1 for networks with $g_0 = g_1 = 1$, $\beta = 2$ and various external electric fields. The external applied electric field E_0 is the average voltage v_e divided by the size of the unit cell. In these cases the contrast between the components is completly determined by the value of E_0. In electric fields larger (smaller) than unity the non-linear component is a better (worse) conductor than the linear component. The results in cases where $g_0 \neq g_1$ are similar and their important characteristics are the same. Only the value of the threshold field above (below) which the non-linear component is a better (worse) conductor than the linear component changes. Results for several fields larger than unity (which we shall call high fields) are shown in figure 1(a). The numerical results appear as points and the CM predictions as full curves. As in networks with linear



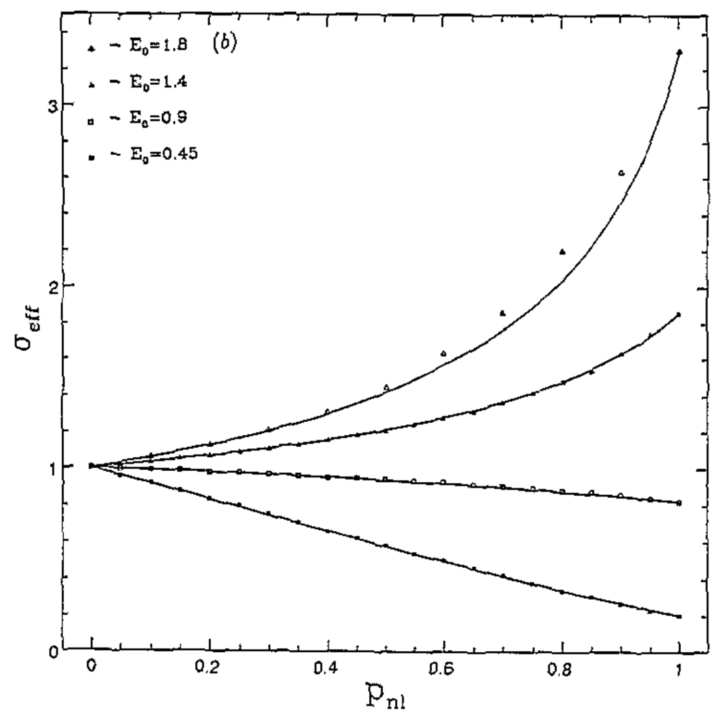


Figure 1. The effective conductance of non-linear networks versus the fraction of the non-linear component for $g_0 = g_1 = 1$, $\beta = 2$ and various applied fields E_0. The full curves are the CM predictions for each case in 1(*a*) and 1(*b*) and for the highest-contrast ($E_0 = 0.045$) case in 1(*c*).

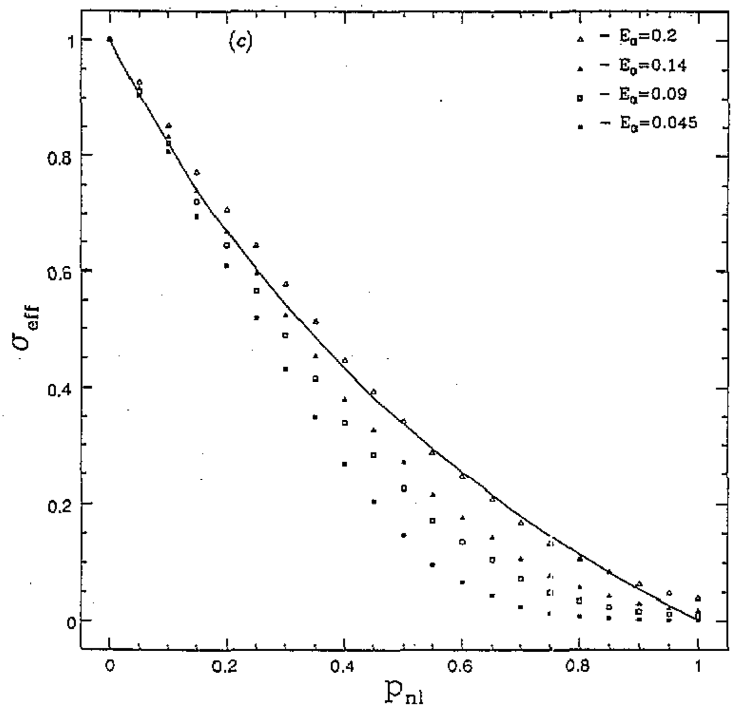


Figure 1. *Continued.*

components, the CM approximation gives the correct results for the pure networks of either component. It is apparent that at high fields there is good agreement between the numerical results and the CM predictions only when the fraction of the non-linear component in smaller than ~ 0.4. In this region the linear component, which is the bad conductor, dominates the conductance. The contribution of the non-linear component (the good conductor) to the conductance becomes appreciable only at higher concentrations, where there are large deviations from the CM results. As expected these deviations become larger as the applied field and the contrast between the components increase. At high fields the CM predictions *underestimate* the effective conductance of the network. Results for fields close to unity are shown in figure 1(*b*). These networks have low contrast and their effective conductance is very well described by the CM approximation at all concentrations. Results for fields much smaller than unity (low fields) are shown in figure 1(*c*). Here the linear component is the good conductor, the non-linear component the bad one, and the contrast between them is high. The CM predictions are close to the numerical results only for small concentrations of the non-linear component. The discrepancies between the two increase as the electric field decreases and the contrast between the components increases. At high concentrations the non-linear component is dominant. The range of good agreement is smaller than in high-field cases with similar contrast. In low-field cases the CM predictions *overestimate* the effective conductance of the network.

Similar results are obtained for networks in which the non-linear component has a non-linearity exponent β that differs from two. The qualitative behaviour described above does not depend on the precise value of β. However, there are some interesting β-dependent differences in the details. Two examples of high-contrast networks with $\beta = 0$, 2 and 4 are shown in figures 2 and 3. The applied electric fields are chosen such that the conductance ratio of the homogeneous networks of the components is the same for all values of β. In figure 2 the contrast in the non-linear cases is caused by high fields and the non-linear component is a better conductor. In all cases the bad conductor is dominant in a wide region of low good-conductor concentration and the CM predictions are good only in that region. However it is clear that the deviation from these predictions decreases with increasing β. This is even more apparent in figure 2(*b*), which is a magnification of the low good-conductor concentration regime. The discrepancy between the numerical results and the CM predictions is much larger, and becomes appreciable at lower concentrations in the linear case ($\beta = 0$) as compared to the non-linear cases ($\beta = 2, 4$). As β increases this discrepancy decreases.

This behaviour can be explained by the following intuitive argument. The cases compared in figure 2 all have the same conductance ratio of the full uniform networks made of one of their components. In the linear case this ratio does not depend on electric field and thus exists also between single elements of the two components in the mixed networks. In the non-linear cases this contrast is field dependent. In the mixed networks the non-linear component is a better conductor and the electric field in it is thus smaller than the applied field. As a result the conductance ratio between a single non-linear element in the mixed network and a linear element is smaller than the ratio between the same non-linear element in the homogeneous non-linear network and the linear element. Because of this the effective contrast between the components in the mixed non-linear networks is smaller than in the mixed linear network and their effective conductances are better described by the CM approximation. This effect is stronger when β is larger since the local conductance ratio is given by the local field to the power β.

A similar behaviour is observed in high-contrast, low-field cases. An example is shown in figure 3. Here the CM predictions are almost identical for all values of β but the simulation results are appreciably different. The largest differences between predictions and numerical results are obtained in the linear case. These differences decrease as β increases. This is a result of the same effect as discussed above for high fields. At low fields the non-linear elements are poorer conductors than the linear elements and the electric field in them is thus larger. This again causes a smaller effective contrast in the mixed networks and a better agreement with CM predictions.

The field dependence of the effective conductivity $\sigma_e(E_0)$ is more complicated than the simple power-law form of the non-linear elements. The CM approximation predicts that at high fields $\sigma_e(E_0)$ is a concave function of the field with an asymptotic behaviour of the form

$$c_1 + c_2 E_0^{-\beta/(\beta+1)}$$

at very high fields. At low fields it is predicted to be a convex function with an asymptotic behaviour of the form $d_1 + d_2 E_0^{\beta}$ for very low fields. The values c_1, c_2, d_1 and d_2 depend only on g_0, g_1 and p. In figure 4 we compare these predictions with numerical results for $p = 0.3$ and $\beta = 2, 4$. At high fields (figure 4*a*) these results are described by a concave function, but as expected they deviate from the predictions more and more as E_0 increases and the above asymptotic behaviour is not observed. For larger p these deviations are larger and the concave form disappears. At low fields (figure 4(*b*)) the numerical results are not described by a convex function and they disagree rather strongly with the CM predictions.

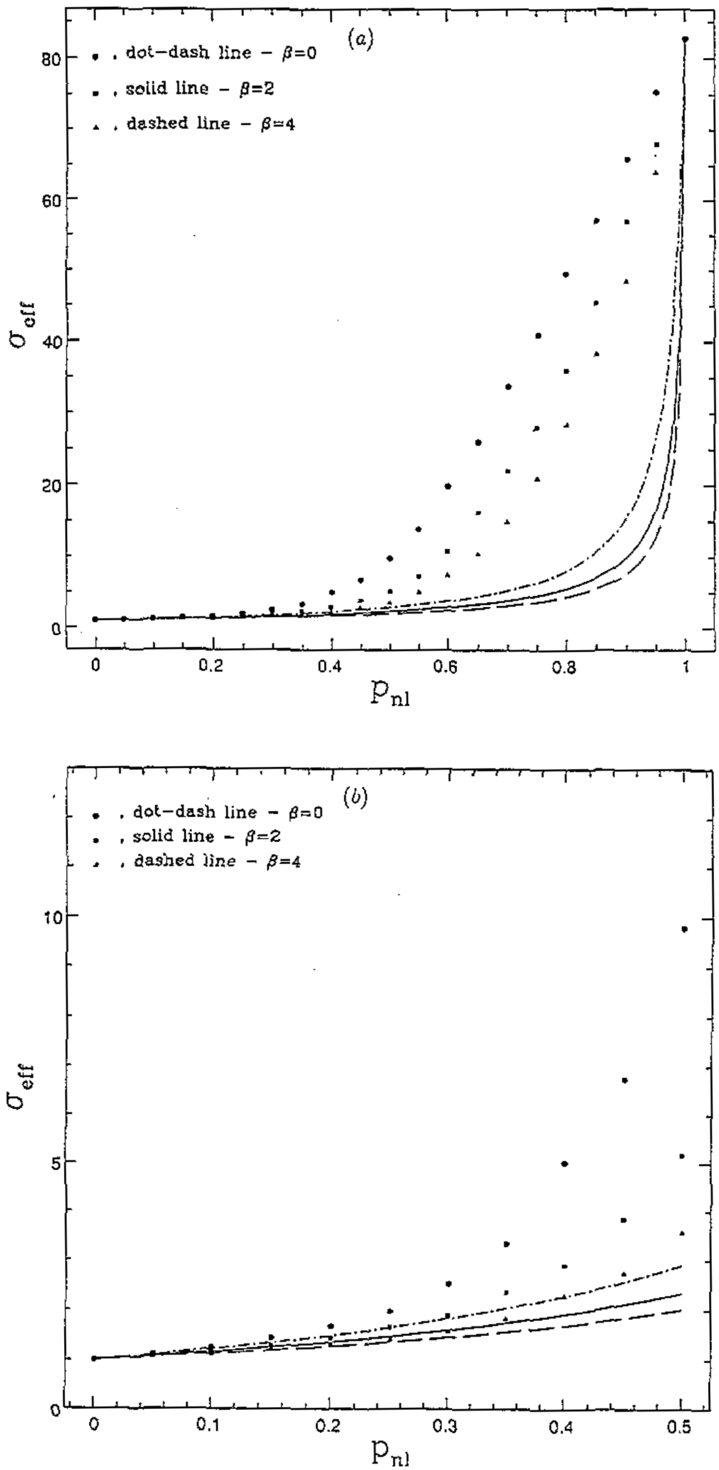


Figure 2. The effective conductance versus the fraction of the non-linear component for $\beta = 0$, 2 and 4 in high applied fields. For $\beta = 2$ $E_0 \simeq 9.1$ and for $\beta = 4$ $E_0 \simeq 3.0$, $g_0 = g_1 = 1$ and the contrast in all cases is approximately 82 to 1. Points mark numerical results and lines mark corresponding CM predictions. 2(*b*) is a magnification of the low-concentration region of 2(*a*).

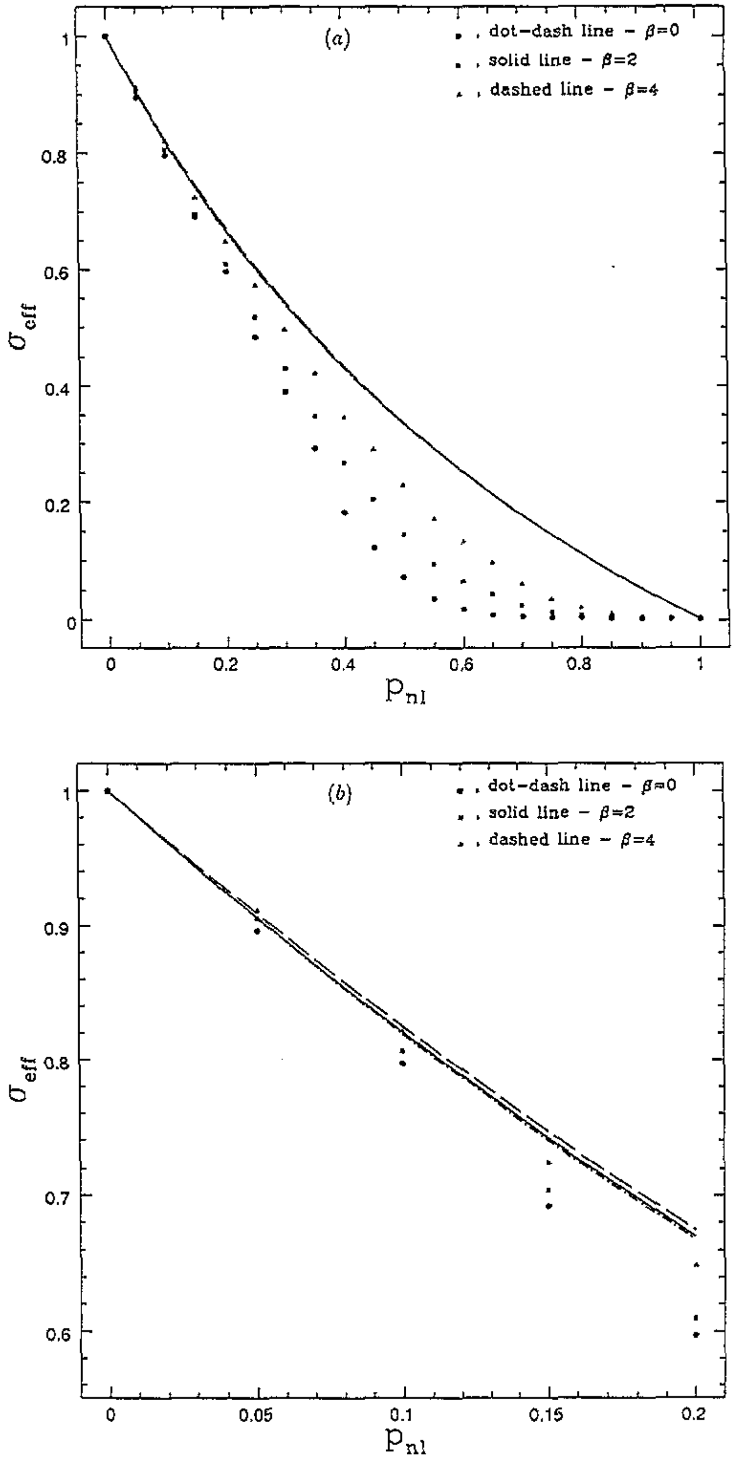


Figure 3. The effective conductance versus the fraction of the non-linear component for $\beta = 0$, 2 and 4 in low applied fields. For $\beta = 2$ $E_0 \simeq 0.05$ and for $\beta = 4$ $E_0 \simeq 0.2$; $g_0 = g_1 = 1$ and the contrast in all cases is approximately 0.002 to 1. Points mark numerical results and curves mark corresponding CM predictions. 3(*b*) is a magnification of the low-concentration region of 3(*a*).

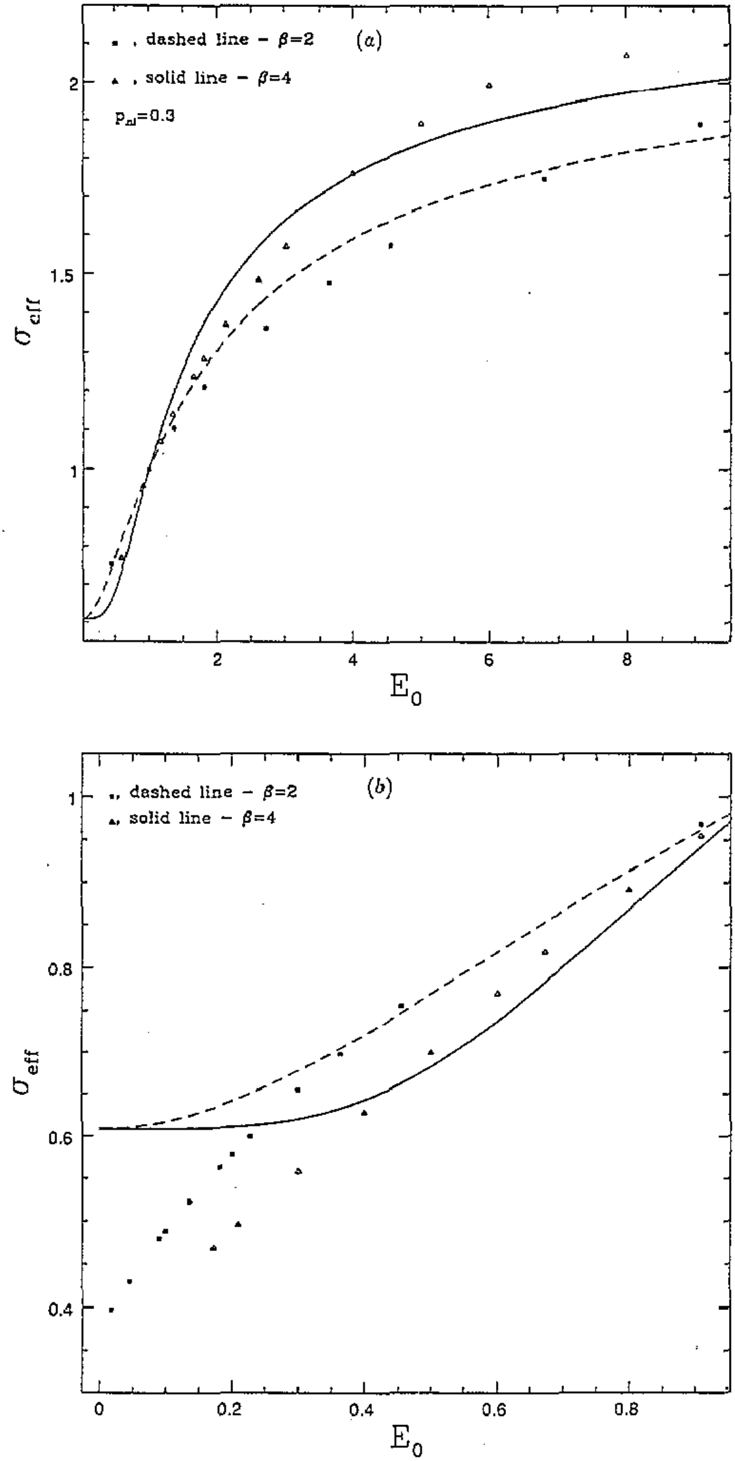


Figure 4. The effective conductance as a function of applied electric field in high fields (*a*) and low fields (*b*).

3. The effective medium approximation

Another simple analytic approximation, which often gives very good results for the bulk effective conductivity of linear composite materials, is Bruggeman's symmetric EMT [8]. As in the case of the CM approximation, EMT is also based on an exact analytical calculation of the field distortion caused by a single spherical or ellipsoidal inclusion, but the host material is now the fictitious effective medium rather than one of the actual components. For this reason it is in general impossible to extend EMT to non-linear composites even when only one component is non-linear: the effective medium itself is now non-linear. Therefore the problem of a single inclusion becomes analytically intractable.

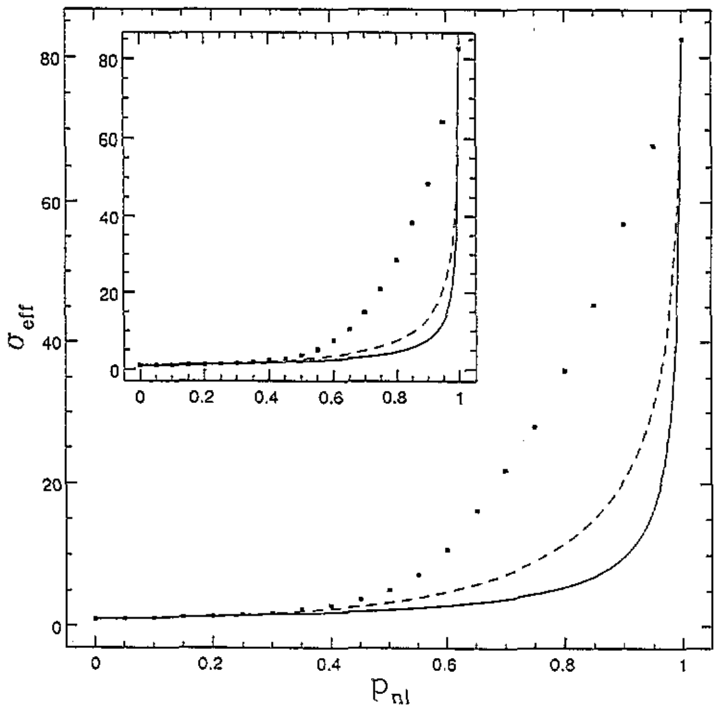


Figure 5. Comparison of the CM approximation (full line) and EMT (dashed line) for $\beta = 2$ in a high-field case. In the inset $\beta = 4$.

Here we nevertheless attempt to extend EMT to a mixture of a linear and a non-linear component by noting that, from the discussion of the CM approximation for such composites [7] it follows that in many cases, even though the non-linear component is *strongly non-linear*, the effective medium turns out to be only *weakly non-linear*. In particular, this is always the case if the non-linear component is present with a small volume fraction or if the contrast between the physical properties of the composites is low. Whenever the effective medium is only weakly non-linear, we can solve the problem of a single inclusion by ignoring the non-linearity of the fictitious uniform host medium. In this way we obtain the well known equation of Bruggeman [8] but with one of the component conductances

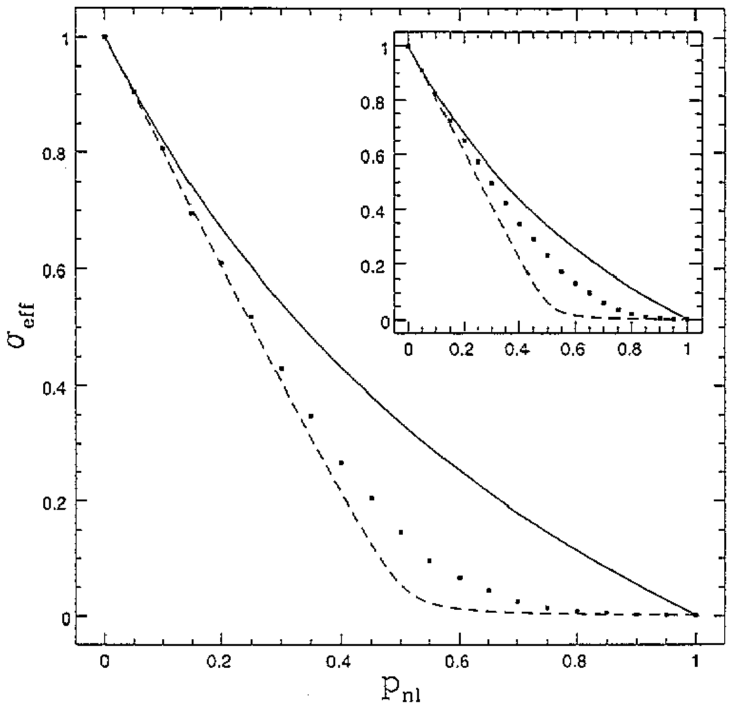


Figure 6. Comparison of the CM approximation (full line) and EMT (dashed line) for $\beta = 2$ in a low-field case. In the inset $\beta = 4$.

$g(v)$ depending on the local voltage drop v across such an isolated inclusion

$$g_e(v_e) = \frac{1}{2\left(\frac{z}{2}-1\right)}\left(g_0\left(\frac{z}{2}(1-p)-1\right)+g(v)\left(\frac{z}{2}p-1\right)\right) + \frac{1}{2\left(\frac{z}{2}-1\right)}\left[\left(g_0\left(\frac{z}{2}(1-p)-1\right)+g(v)\left(\frac{z}{2}p-1\right)\right)^2 + 4\left(\frac{z}{2}-1\right)g_0 g(v)\right]^{1/2}. \quad (3)$$

As in the case of the non-linear CM approximation, we have to supplement this equation by another equation, i.e. equation (1), which gives the connection between the average voltage v_e and the local voltage v across the non-linear conductor. To obtain this equation no assumption had to be made regarding the behaviour of the effective medium. The only assumption is, as in the linear case, that the field incident upon each of the non-linear conductors is uniform. These equations are solved simultaneously by standard numerical methods to give the explicit functional form of $g_e(v_e)$.

Typical results are shown in figures 5 and 6 in which we compare numerical results of two high-contrast cases to predictions of the CM approximation (equations 2 and 1) and EMT (equations 3 and 1). In figure 5 the field is high and the non-linear component is the good conductor. For both $\beta = 2$ and 4 the EMT predictions are slightly better than those of the CM approximation. They too are close to the numerical results only in the low-concentration range of the non-linear component and become better as β increases. In high-contrast,

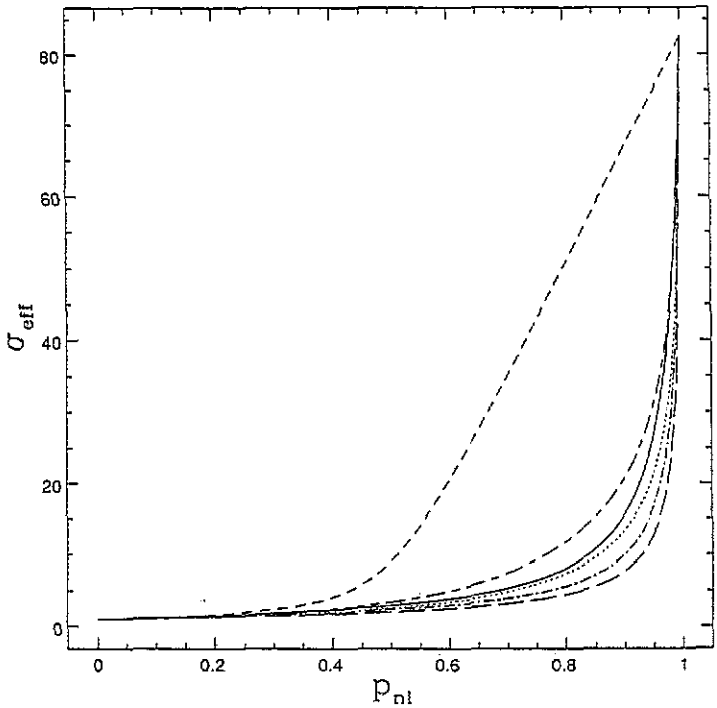


Figure 7. The CM approximation and EMT for a high-field case. CM: linear case (full line), $\beta = 2$ (chain curve) and $\beta = 4$ (long dashed curve). EMT: linear case (dashed line), $\beta = 2$ (long dash–short dash) and $\beta = 4$ (dotted curve).

low-field cases (figure 6) the EMT approach is much better than the CM. It is closer to the numerical results at both ends of the mixture range. In contrast to the CM predictions, those of the EMT become worse as β increases and underestimate the effective conductance at low fields too. At low-field cases the numerical results lie between the EMT and the CM predictions. The larger is β, the closer they are to those of the CM approximation.

It is interesting to see how the results of both approximations are affected by the non-linearity exponent β. This is shown in figure 7 for a high-field case. It is seen that the EMT is most strongly affected by the replacement of a linear component by a non-linear one. For non-linear components with different β both predictions are similar and decrease with increasing β.

4. Conclusions

We have determined the effective conductance properties of mixed linear–non-linear RRNs with a strong power-law non-linear component by numerical simulations, and compared them with predictions of a previously proposed CM approximation and a new EMT approximation. It is shown that both approximations work well for cases of either low contrast between the components or low concentration of the non-linear component. For high-contrast cases it is shown that the CM approximation is better for such mixed networks than for linear networks with the same contrast, and that it improves as the power β of non-linearity of the non-linear component increases. The extension of Bruggeman's symmetric

EMT to these high contrast cases gives better results than the CM approach, even though it requires one further assumption concerning the non-linear behaviour of the effective medium. Thus it is shown that the EMT approximation, and to a somewhat lesser extent the CM approximation, can be used to calculate the electrical properties of non-linear small-particle composites, and to study the interesting and potentially useful non-linear phenomena that they produce.

The analytic and numerical predictions presented above should be compared to results of systematic experiments on non-linear composites. At present, no such experimental studies have been reported and we hope a change in this state of affairs will be encouraged by this paper.

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