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Scaling behavior and surface-plasmon modes in metal-insulator composites

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The ac dielectric response of metal-insulator composites is studied numerically, using the transfer-matrix algorithm of Derrida and Vannimenus. For two-dimensional random composites with site percolation, we verify numerically that the effective dielectric function can be written numerically in the form $\epsilon_e/\epsilon_1 = \xi^{-t/\nu} G_{\pm}((\epsilon_2/\epsilon_1)\xi^{(t+s)/\nu}, \xi/L)$, where ϵ_1 and ϵ_2 are the dielectric functions, ξ is the correlation length, L is the system size (or wavelength of the electric field), G_+ and G_- are universal functions above and below percolation, and t, s, and ν are standard percolation exponents. A similar form has been previously verified for bond percolation by Bug *et al.* We also study surface-plasmon resonances in a two-dimensional lattice model of a composite of Drude metal and insulator. The effective conductivity of the composite in this case is found to consist of a Drude peak which disappears below the metal percolation threshold, plus a band of surface-plasmon states separated from zero frequency by a gap which appears to vanish near the percolation threshold. The results in this case agree qualitatively with effective-medium predictions. The potential relation of these results to experiment, and the possibility of a Lifshitz tail in the surface-plasmon density of states, are briefly discussed.

I. INTRODUCTION

Composite materials have long been known to have electrical and optical properties very different from those of their constituents.[1] The differences are particularly marked near a percolation threshold, i.e., a point at which one of the two components of the composite first forms a closed connected path extending throughout the sample. For a composite comprised of two materials with conductivities σ_1 and σ_2, the effective conductivity σ_e exhibits the power law (in the limit in which σ_2 approaches zero)[2]

$$\sigma_e \approx \sigma_1 (p-p_c)^t \quad (p>p_c) \,, \tag{1.1}$$

where p is the volume fraction of material 1 and p_c is the percolation threshold for material 1. Conversely, if $\sigma_1 \gg \sigma_2$, then as p approaches p_c from below it is believed that σ_e diverges according to the law[3]

$$\sigma_e \approx \sigma_2 (p_c - p)^{-s} \quad (p<p_c) \,. \tag{1.2}$$

The exponents t and s are believed to depend on the dimensionality of the composite, but are otherwise "universal" in the sense that they are identical for all lattice models of the same dimensionality.[2] Exponents for a continuum composite are thought to depend on certain details of the microgeometry, and may differ from those expected for lattice models.[4] The percolation threshold p_c, on the other hand, is not universal, but varies from sample to sample.

The power-law behavior suggests a close analogy between the percolation transition and conventional phase transitions, with the percolation threshold as a kind of a critical point. Just as in the theory of phase transitions, there exists a correlation length in percolation phenomena which diverges as the percolation threshold is approached from either side. This correlation length, denoted ξ, describes the linear dimensions of a typical cluster for $p<p_c$; above p_c, it is the length scale below which the connected cluster has a fractal rather than a homogeneous, integer-dimensional geometry. ξ also obeys a power law near the percolation threshold, diverging on both sides of the threshold as[2]

$$\xi \approx |p-p_c|^{-\nu} \,, \tag{1.3}$$

where ν is another critical exponent with the same degree of universality as s and t.

This paper is concerned with several aspects of the connection between the percolation transition and conventional phase transitions. First, we consider a scaling form for the complex dielectric response of a metal-insulator composite. This form is expected to be valid at both finite sizes and finite frequencies. We provide numerical evidence to confirm that this form is valid in site percolation, using a transfer-matrix method recently developed to treat dc problems in composite media.[5] A similar demonstration has recently been carried out by Bug *et al.* for bond percolation in two dimensions.[6] Secondly, we examine a model for a composite of Drude metal and insulator near the percolation threshold. We present numerical evidence, using the transfer-matrix technique, that, in the limit of a long Drude relaxation time, there exists a gap in the absorption spectrum at low frequency, and that the frequency width of this gap goes to zero at p_c. Analytic arguments[7] suggest that the gap actually obeys a power law near the percolation transition. Our numerical results are not accurate enough to estimate this power.

All of our calculations are carried out using random impedance networks to model metal-insulator composites. These networks emerge naturally if one discretizes the appropriate electrostatic equations and solves them on a lattice.[8] Such a lattice representation of a continuum composite may lead to power laws near the percolation threshold that differ from the continuum in some cases. This difference appears to be less important in two dimensions, for which our lattice simulations are carried out.[3] Our results may thus have some relevance to real two-dimensional composites.

The use of a transfer-matrix technique to calculate the effective impedance of our model composite leads to great efficiency in ac problems, just as in static problems.[6,9] The method gives values for the percolation exponent t which are accurate, in two dimensions, to a few tenths of a percent,[10] in contrast to the earlier method, based on the solution of Kirchhoff's equations,[8] which are rarely accurate to better than 10%.

The scaling theory we study has its origin in the work of Straley.[3,11] For a two-component composite with constituent conductivities σ_1 and σ_2, Straley has proposed that the two variables characterizing the "phase diagram" are $p-p_c$ and σ_1/σ_2, which are analogous respectively to the temperature and magnetic field in a magnetic phase transition. Lobb and Frank[10] have applied finite-size-scaling arguments to obtain the exponents t and s precisely at $p=p_c$, using a homogeneous function representation for the function σ_e/σ_2, based on the two scaling variables. The transfer-matrix algorithm[5] permits the same finite-size-scaling idea to be used with much greater accuracy to calculate these exponents, and also eliminates the necessity of averaging over many realizations of a finite sample: this averaging is done automatically.

The scaling form we describe is appropriate for the effective complex dielectric function of a metal-insulator composite at finite sizes and finite frequencies. The extension of scaling ideas to finite frequency was first proposed by Bergman and Imry.[7] Further work has been carried out by Stroud and Bergman,[12] and by Wilkinson *et al.*;[13] the latter authors have used a random resistor-capacitor network as a representation of a metal-insulator composite near the percolation threshold, and have calculated the scaling function using a real-space renormalization-group approach. Several authors have discussed theoretically the ac properties of a metal-insulator composite near the percolation threshold,[9,14] and recently Bug *et al.*[6] have tested a finite-size, finite-frequency scaling form similar to that discussed below, for *bond* percolation in two dimensions. The fact that we are able to verify the same scaling form for site percolation is not surprising, but does provide further evidence in favor of the kind of universality expected in lattice percolation problems with short-range order.[2]

The latter part of the paper treats a lattice model of a composite of Drude metal and insulator. Such metal-insulator composites exhibit so-called surface-plasmon resonances and a corresponding absorption band at frequencies below the plasma frequency, ω_p.[15] We calculate this absorption band explicitly in two dimensions, using the transfer-matrix algorithm. The calculated absorption band compares well with a mean-field theory of the composite, the effective-medium approximation (EMA).[16] The bottom of the absorption band is found to go to zero frequency at the percolation threshold, in agreement with both the EMA and the scaling theory. Thus our results numerically confirm existing theories of absorption in random metal-insulator composites, within a lattice model.

We turn now to the body of the paper. Section II reviews the scaling form for the effective complex dielectric function, ϵ_e. Section III describes the model resistor-capacitor network on which to test the scaling form, and presents the results of numerical simulations based on the transfer-matrix method. Section IV gives analogous calculations for a network model of a Drude metal and insulator, and compares the results with the EMA. A discussion follows in Sec. V.

II. SCALING FORM FOR THE COMPLEX DIELECTRIC FUNCTION

We summarize the scaling formalism for a composite of two components having complex dielectric functions

$$\epsilon_1(\omega)=1+4\pi i\sigma_1(\omega)/\omega\ , \tag{2.1}$$

$$\epsilon_2(\omega)=1+4\pi i\sigma_2(\omega)/\omega\ , \tag{2.2}$$

where $\sigma_1(\omega)$ and $\sigma_2(\omega)$ are the frequency-dependent conductivities of the two constituents, which are present in volume fractions p and $1-p$. Since the formalism is largely equivalent to that described in Ref. 6, we simply quote the main results. When the wavelength of the applied field is large compared to any length scale within the composite (e.g., grain size, percolation correlation length), it is usually adequate to work in the quasistatic limit, in which the electric field $\mathbf{E}(x)$ and displacement $\mathbf{D}(x)$ satisfy the equations

$$\nabla\cdot\mathbf{D}=0\ , \tag{2.3}$$

$$\nabla\times\mathbf{E}=0\ , \tag{2.4}$$

$$\mathbf{D}(x)=\epsilon(x)\mathbf{E}(x)\ , \tag{2.5}$$

where $\epsilon(x)$ is the position- and frequency-dependent dielectric function. At finite frequencies, the displacement current is generally non-negligible. Thus a scaling function must be developed for the complex effective dielectric function

$$\epsilon_e(\omega)=\epsilon_{r,e}(\omega)+4\pi i\sigma_e(\omega)/\omega\ . \tag{2.6}$$

The scaling expression appropriate to a sample of finite linear dimensions L is[6]

$$\epsilon_e/\epsilon_1=\epsilon^{-t/\nu}G_+((\epsilon_2/\epsilon_1)\xi^{(t+s)/\nu},\xi/L),\quad \Delta p>0 \tag{2.7}$$

$$\epsilon_e/\epsilon_2=\epsilon^{s/\nu}G_-((\epsilon_2/\epsilon_1)\xi^{(t+s)/\nu},\xi/L),\quad \Delta p<0 \tag{2.8}$$

where $G_+(x,y)$ and $G_-(x,y)$ are functions of two variables which describe the frequency- and size-dependent dielectric function above and below the percolation threshold. In writing (2.7) and (2.8), we are considering both L and ξ to be expressed in units of some fundamental length, such as grain size, so that they can both be taken as dimensionless.

Precisely at $p=p_c$, the correlation length is infinite, and the complex dielectric function can be expressed in terms of a single variable. The result is

$$\epsilon_e/\epsilon_1=(\epsilon_2/\epsilon_1)^{t/(t+s)}J(L(\epsilon_2/\epsilon_1)^{\nu/(t+s)})\ , \tag{2.9}$$

where $J(u)$ is a complex function of the complex variable

u. Form (2.9) is readily tested numerically by calculating the complex conductivity of samples of different linear dimensions L. If the scaling form is valid, they can all be represented by the same universal function (2.9). This calculation provides a reasonably stringent test of the original scaling relation, Eqs. (2.7) and (2.8).

The scaling form is appropriate to a dielectric function at *infinite* wavelength in a *finite* medium. Presumably the same form holds at finite wavelength in an infinite medium. We then interpret Eqs. (2.7) and (2.8) as equations for the *wave-number- and frequency*-dependent effective dielectric function $\epsilon_e(q,\omega)$ near the percolation threshold, with L to be interpreted as the wavelength, $L=2\pi/q$.

This picture leads to some novel predictions for the ac response of composites near the percolation threshold. Assume $\Delta p>0$, $\xi/L\gg 1$, and $(\epsilon_2/\epsilon_1)\xi^{(t+s)/\nu}\ll 1$. The first inequality means that we are on the metallic side of the metal-insulator transition. The second implies that the role of the correlation length ξ is played by L, i.e., that ϵ_e is independent of ξ in this regime. The third inequality is the condition that ϵ_e should be independent of the insulating dielectric constant ϵ_2. Putting these results together, we find that $G_+(x,y)$ must vary as $u^{t/\nu}$ in this regime or

$$\epsilon_e(q,\omega)\sim\epsilon_1(\omega)L^{-t/\nu}\sim\epsilon_1(\omega)q^{t/\nu} . \tag{2.10}$$

To see how this result might influence electromagnetic wave transmission through a composite material, we note that the dispersion relation can be written self-consistently as

$$q^2=\omega^2\epsilon_e(q,\omega)/c^2 \tag{2.11}$$

where c is the speed of light. Substituting (2.22) into (2.23)

$$q\sim[\omega^2\epsilon_1(\omega)/c^2]^{1/(2-t/\nu)} . \tag{2.12}$$

If we assume $\epsilon_1(\omega)\sim 4\pi i\sigma/\omega$, characteristic of a metal at low frequencies, where σ is the conductivity, we get

$$\begin{aligned}\epsilon_e(q(\omega),\omega)&\sim\epsilon_1(\omega)q(\omega)^{t/\nu}\\&\sim\omega^{[1/(2-t/\nu)-1]} ,\end{aligned} \tag{2.13}$$

which describes very well the frequency dependence observed by Laibowitz and Gefen[17] in two-dimensional thin films of granular metals near the percolation threshold. However, the inequalities required for this form to be applicable are not satisfied in the frequency range studied by Laibowitz and Gefen; the agreement is therefore coincidental. Basically, (2.13) can apply only if (a) the wavelength of the radiation in the composite is small compared to a correlation length, and (b) the dielectric response of the insulator is so small in comparison to that of the metal grains that $(\epsilon_2/\epsilon_1)q^{-(t+s)/\nu}\ll 1$. Neither relation is true in the composites studied in reference in the frequency range of ~ 10 KHz investigated. Nevertheless, the present results show that novel frequency dependence is to be expected in metal-insulator composites near the percolation threshold when the wavelength is small compared to a correlation length, so that the fractal character of the percolation structure[18] can be probed.

III. CALCULATIONS OF THE SCALING FUNCTION FOR A METAL-INSULATOR MIXTURE AT FINITE FREQUENCIES

We will calculate the scaling function (2.16), and confirm the validity of the scaling form, by considering a random network of impedances in two dimensions. The impedances are chosen to model metallic and dielectric components at low frequencies. The metallic component is assigned a purely real impedance, $Z_m=R$, characteristic of a pure resistor. The dielectric, on the other hand, is given a purely capacitive impedance, $Z_d=1/(i\omega C)$, with C real. The impedances Z_m and Z_d are placed at random on the bonds of a two-dimensional rectangular network. We carry out the random placement according to the rules of site percolation. That is, we denote the sites on the network as "conducting" or "insulating" with probability p and $1-p$. The rules of bond assignment are that a bond between two conducting sites has impedance Z_m; a bond between a conducting and an insulating site, or between two insulating sites, has impedance Z_d.

To use the scaling form at finite frequencies, we must consider the effective dielectric function of the composite. Hence, rather than considering the impedances Z_m and Z_d, we must consider the effective "dielectric functions" of these bonds, defined by

$$\epsilon_m=1/(Z_m i\omega)=1/(i\omega R) , \tag{3.1}$$

$$\epsilon_d=1/(Z_d i\omega)=C . \tag{3.2}$$

In principle, a capacitance should also be added in parallel with the resistor across each metallic bond. But at sufficiently low frequencies such a capacitance will have little effect, and we do not include it here.

We calculate the effective dielectric function of the network, denoted $\epsilon_e(\omega)$, by applying an ac voltage difference across a particular realization of the random network and solving Kirchhoff's equations to obtain the electric displacement across each bond. The (complex) displacement across bond (ij), denoted $D_{ij}(\omega)$, is related to the (complex) potential difference $V_i(\omega)-V_j(\omega)$ by

$$D_{ij}(\omega)=\epsilon_{ij}(\omega)[V_i(\omega)-V_j(\omega)] , \tag{3.3}$$

where $\epsilon_{ij}=\epsilon_m$ or ϵ_d according as the bond (ij) is considered metallic or dielectric. The condition that no displacement current shall build up at the ith node is equivalent to the equation

$$\sum_j\epsilon_{ij}(V_i-V_j)=0 , \tag{3.4}$$

where the sum runs over all sites j with a nonzero bond connected to site i. The total displacement current across the network is that of an equivalent uniform network in which each bond has dielectric function $\epsilon_e(\omega)$.

Rather than solve Eqs. (3.4) directly to compute the voltage at each site—a procedure which consumes a great amount of computer time when done by a standard iterative procedure—we use the transfer-matrix algorithm suitably extended to finite frequencies.[6] To execute the method, we consider an $L\times M$ rectangular array of sites (L rows, M columns, with $M\gg L$). The potential is fixed at $V=0$ on the first row, $V=V_0\exp(i\omega t)$ on the

Lth row. The total displacement current flowing through the array is computed consecutively, column by column, with free boundary conditions on the first column of sites. The method requires no repeated iteration of linear equations, since only the total current is computed, not the voltages at each node, and thus is extremely fast. Also, because a very large number of columns can be accommodated by this procedure, the calculation in effect averages automatically over many $L \times L$ samples and thus makes unnecessary the averaging over many realizations required in earlier calculations of random impedance networks.

In order to verify the scaling form (2.7), we first assume, in agreement with established theory, that the correlation length ξ varies as $|\Delta p|^{-\nu}$ near $p=p_c$. We must graph $(\epsilon_e/\epsilon_m)|\Delta p|^{-t}$ against the variable $|\epsilon_d/\epsilon_m|\,|\Delta p|^{-(t+s)}$ for fixed values of the second variable $|\Delta p|^{-\nu}/L$, where L is the sample width. We have carried out such a calculation, but rather than displaying the ratio of dielectric functions, we plot instead the *ratio of admittances*

$$(g_e/g_m)|\Delta p|^{-t} \tag{3.5}$$

against the variable

$$|g_d/g_m|\,|\Delta p|^{-(t+s)}, \tag{3.6}$$

where the complex admittances are defined, for present purposes, by the relations

$$\begin{aligned} g_m &= 1/Z_m\ , \\ g_d &= 1/Z_d\ , \\ g_e &= 1/Z_e\ . \end{aligned} \tag{3.7}$$

The value of p_c for site percolation in two dimensions is known numerically to be 0.59297. . . . The ratio $t/\nu = 0.973$ from numerical studies based on the transfer-matrix algorithm and finite-size-scaling theory.[10] The exponents t and s are equal in two dimensions by an exact duality relation. Denoting the metallic admittance g_m and the insulating admittance as g_d, we have $g_d/g_m = i\omega RC$. The results of the calculations are shown in Figs. 1(a) and 1(b) with the values $R=1$, $C=1$. For $(\Delta p)^{-\nu}/L = 4.94$, we have carried out two calculations at different volume fractions of the metallic component but preserving the value of the ratio. Evidently the two curves lie atop one another as required by the scaling hypothesis. An additional calculation was carried out at $(\Delta p)^{-\nu}/L = 2.63$; this produces a scaling function which differs from the first at low frequencies. These results provide evidence that the form (2.16) is indeed valid, at least for $\Delta p > 0$ in two dimensions.

We have also carried out simulations precisely at $p=p_c$ in order to verify the scaling form (2.9) and to calculate the scaling function $J(u)$ explicitly. To verify (2.9) for the network under consideration, first note that it can be written in the equivalent form

$$\frac{g_e}{g_m} = \left[\frac{g_d}{g_m}\right]^{t/(t+s)} J(L(g_d/g_m)^{\nu/(t+s)})\ , \tag{3.8}$$

where the admittances and dielectric function are related as in Eq. (3.7). We must graph $(g_d/g_m)^{-t(t+s)}(g_e/g_m)$ against $L(g_d/g_m)^{\nu/(t+s)}$ for several sample widths L, adjusting the frequency ω so as to keep the variable $L(g_d/g_m)^{\nu/(t+s)}$ constant. The results are shown in Figs. 2(a) and 2(b). For each value of the scaled variable $L(g_d/g_m)^{\nu/(t+s)}$, the variable $(g_d/g_m)^{-t/(t+s)}(g_e/g_m)$ is indeed independent of the width L to within numerical accuracy. Thus we have perhaps even stronger evidence that the original scaling forms are valid, because we have eliminated the variable p by working precisely at $p=p_c$.

Finally, we show the scaling function itself at the percolation threshold. Defining $X = L(g_d/g_m)^{\nu/(t+s)}$, we write

$$J(X) = (g_d/g_m)^{-t/(t+s)}(g_e/g_m)\ , \tag{3.9}$$

which is plotted in Figs. 3(a) and 3(b). At small values of

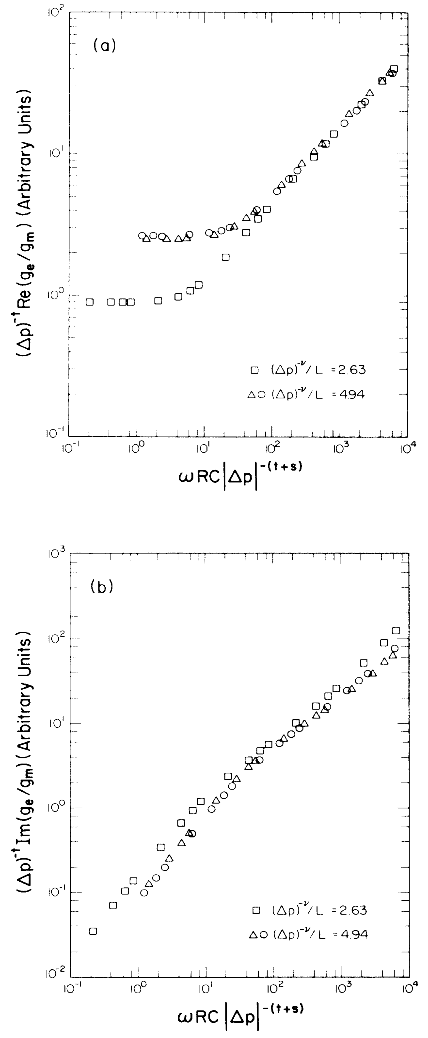

FIG. 1 (a) Scaled real part of the effective admittance, $(\Delta p)^{-t}\mathrm{Re}(g_e/g_m)$, plotted as a function of frequency for two values of the variable $(\Delta p)^{-\nu}/L$, where $(\Delta p)^{-\nu}$ is proportional to the correlation length and L is the width of the sample. For a fixed value of this variable, the conductivity is a universal function of $\omega RC\,|\Delta p|^{-(t+s)}$. (b) Scaled imaginary part of the effective conductivity, plotted as in (a).

X, Fig. 3(a) shows that Re$J(X)$ varies approximately as X^{-1}, while at large values it approaches a constant. The small-X behavior can be understood as follows. The two-dimensional strip, with $M \gg L$, is always above $p - p_c$, that is, the metallic portion percolates. Thus we expect that at small X the admittance g_e of the network should become independent of g_2, the admittance of the insulating component. This can occur only if $J(X)$ varies as $X^{-t/\nu}$ for small X. In two dimensions $t/\nu \sim 0.96$,[10] consistent with the behavior of Fig. 3(a), which shows the dominant part of $J(X)$ at small X. If we were below p_c, a complementary argument given in Ref. 6 shows that $g_e \sim g_d L^{s/\nu}$ at small X.

IV. SURFACE-PLASMON MODES IN A TWO-DIMENSIONAL METAL-INSULATOR COMPOSITE

As a second application of the transfer-matrix algorithm at finite frequencies, we have considered a model somewhat similar to that discussed in the preceding section, but generalized to take account of the so-called surface-plasmon modes known to be an important absorption mechanism in random metal-insulator composites.[15] In a bulk (three-dimensional) composite, such modes are easily understood by considering a small spherical metal particle, described by a Drude dielectric function,

$$\epsilon_m(\omega) = 1 - \omega_p^2/[\omega(\omega + i/\tau)] , \qquad (4.1)$$

embedded in an insulating matrix of dielectric constant unity. Here ω_p is the plasma frequency and τ is a characteristic relaxation time. If we imagine the medium subjected to an applied electric field $E_0 \exp(-i\omega t)$, then the solution of the electrostatic equations (2.3)–(2.5) leads to a uniform electric field $\mathbf{E}_{\text{in}}$ within the metal given by

$$\mathbf{E}_{\text{in}} = 3\mathbf{E}_0/(\epsilon_m + 2) . \qquad (4.2)$$

In the usual limit $\omega_p \tau \gg 1$, E_{in} becomes very large at frequencies in the vicinity of $\omega_p/(3)^{1/2}$, the surface-plasmon frequency for this geometry. At such frequencies, the applied field is in resonance with the surface-plasmon dipole mode of the small metal particle, and the result is a strong absorption at this frequency. If one calculates the effective dielectric function ϵ_e of a dilute suspension of small Drude metal spheres in dielectric, ϵ_e is found to have a large imaginary part near $\omega_p/(3)^{1/2}$. For larger concen-

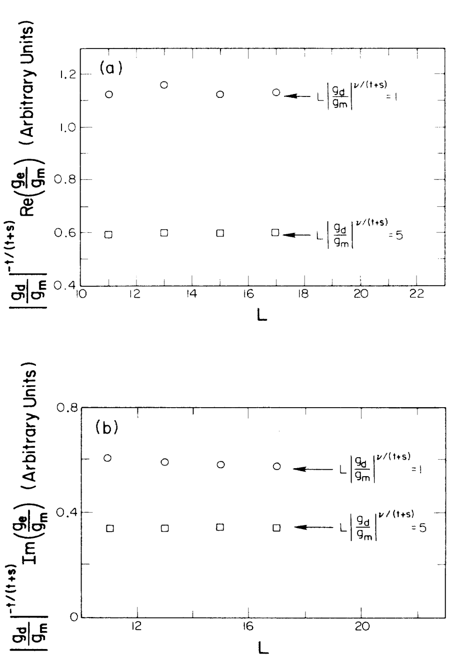

FIG. 2. (a) and (b) Real and imaginary parts of the variable $(g_d/g_m)^{-t/(t+s)}(g_e/g_m)$ plotted as a function of width L for two different values of the scaled frequency $L(g_d/g_m)^{\nu/(t+s)}$. The validity of scaling is demonstrated by the fact that the variable $(g_d/g_m)^{-t/(t+s)}(g_e/g_m)$ is independent of L for fixed values of the scaled frequency.

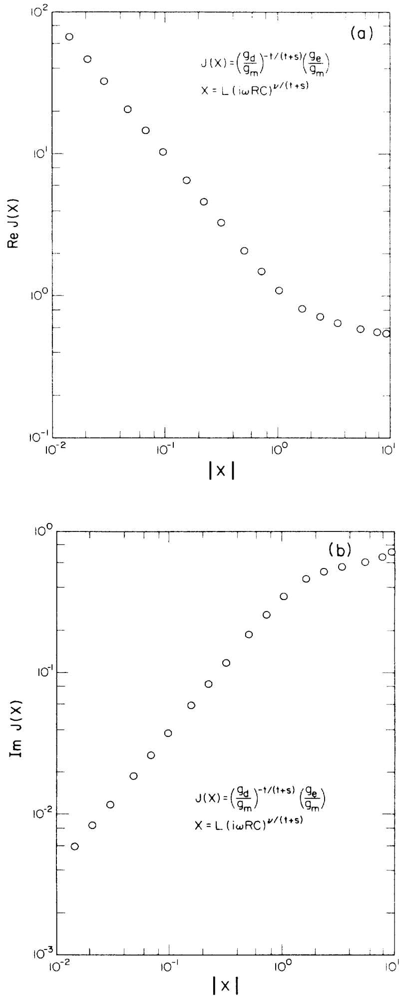

FIG. 3. (a) and (b) Real and imaginary parts of the scaling function $J(X)$ plotted against X at the percolation threshold, $p = p_c$. The scaling function and its argument are defined in the figure; both $J(X)$ and X are dimensionless.

trations of metal, this resonance broadens out and no fully satisfactory method exists for calculating the ϵ_e in such cases, although many approximations have been proposed.[19] It is therefore useful to have available some numerical results as standards for comparison with approximate theories.

We have carried out such numerical simulations in two dimensions, using the site percolation model described in Sec. III in conjunction with the transfer-matrix algorithm. However, the frequency-independent impedance used to represent the conductive bonds in Sec. III cannot lead to

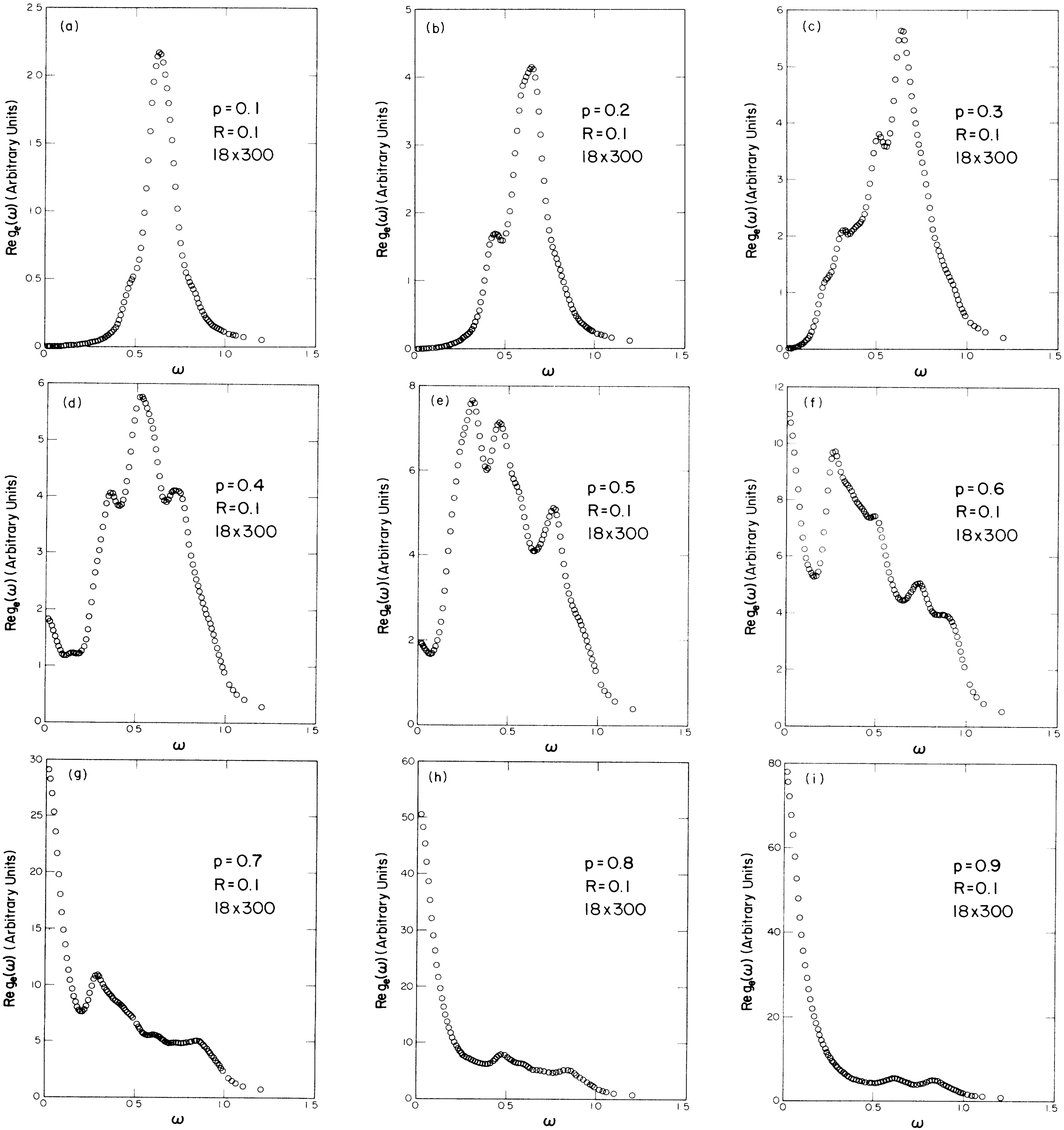

FIG. 4. Real part of the effective conductivity Reg_e (in arbitrary units), plotted against frequency for several concentrations of metal in the metal-insulator composite discussed in the text, as calculated numerically using the transfer-matrix algorithm. (The quantity actually plotted is the in-phase part of the current passing through the 18×300 network for unit voltage drop. The frequency is expressed in units of the plasma frequency, ω_p.)

surface-plasmon absorption; the model must be generalized to allow a *frequency-dependent* impedance. We therefore represent a metallic bond by a resistor and an inductor in *series*, and an insulating bond simply as a capacitance. Thus we have

$$Z_m = R + iL_{\text{ind}}/\omega \ , \tag{4.3}$$

$$Z_d = i\omega C \ . \tag{4.4}$$

The network corresponds to a random *LC* circuit with resistive damping in the inductive elements. To represent the Drude model more faithfully, we should, in principle, place a capacitance in parallel with the *LR* element in the metallic bonds. Such a capacitance does not affect the fundamental physics of the problem, however, and has not been included in our calculations. In the present model the surface-plasmon resonances will now manifest themselves as the *LC* resonances of the random impedance network.

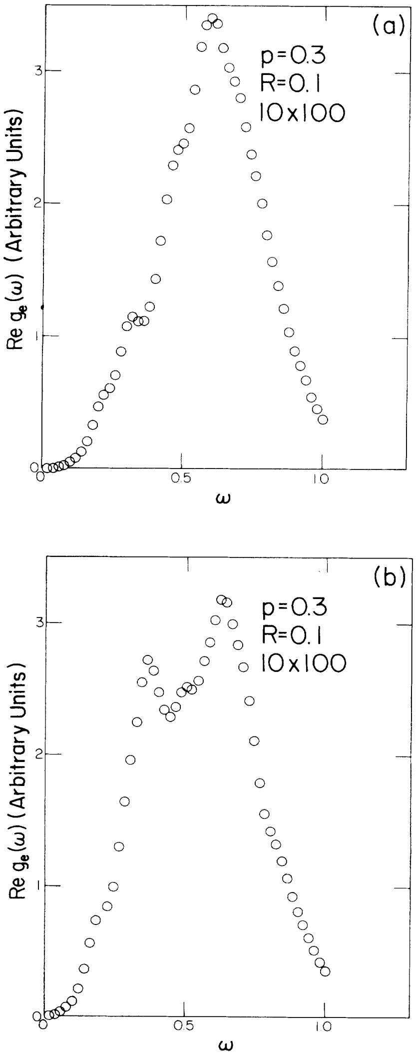

FIG. 5. (a) and (b) Same as Fig. 4, but for two realizations of a 10×100 network at $p=0.3$.

The results of our calculations are shown in Fig. 4. We assume without loss of generality that $L_{\text{ind}}=1$ and $C=1$. The figures show the real part of the effective admittances g_e of the random network. For the samples shown we have used the relatively small sizes $L=18$, $M=300$ because of the considerable expense in looking at larger samples over a broad range of frequencies. In particular, our samples are still too small to have fully converged to the expected infinite-sample behavior. Thus, we expect that many of the details of the structure shown for our rather small samples would vary from one realization of the disordered network to another. For example, the positions of many of the peaks shown in Fig. 4 (except for that at zero frequency) would be expected to differ from one realization to another. We have confirmed this hypotheses by carrying out two further runs at $p=0.3$, $L=10$, and $M=100$, each corresponding to a different realization of a composite with 30% of sites metallic. In each case, as may be seen in Fig. 5, the overall peak shapes are quite similar, and similar also to the single run at $L=18$, $M=300$. But the various details, such as the number and strengths of the peaks, do differ among the realizations.

Some features are independent of realization. These may be summarized as follows.

(i) The absorption band (i.e., the band of *LC* resonances, broadened by the resistive damping) can be divided into two parts. One is an "impurity band," consisting of the network analog of the surface-plasmon resonances described above, and spread over a typically broad range of frequencies. The other is the "Drude peak," centered at zero frequency, and appearing only above the metallic percolation threshold and corresponding to finite dc conductivity.

(ii) The bottom of the impurity band is separated from zero frequency by a gap which appears to go to zero at the percolation threshold, and to increase with $|p-p_c|$ on either side of the threshold. This feature is somewhat obscured by the resistive damping but certainly persists to the largest sample we have studied. We believe that in the limit of a very large sample and very low damping the feature would become even more prominent. Our results are not accurate enough to ascertain if the gap goes to zero according to some power law in $p-p_c$, as has been predicted by Bergman and Imry.[7]

(iii) There is a great deal of fine structure in the resonances within the impurity band. This structure is unmasked by reducing the damping in the calculation, but as noted above it is very sample dependent. For a very large sample, and very low damping, we believe, as stated above, that most of this structure would average out leaving a smooth impurity band with few prominent features. Our samples are not large enough to fully confirm this smoothness, however.

One other point should be made about the curves of Fig. 4. The nominal percolation threshold for an infinite sample is 0.592... (site percolation in two dimensions). For *finite* samples percolation of the metallic component will occur at a lower concentration. That is why the Drude peaks appear in the real part of the admittance for concentrations $p=0.4$ and greater: those at 0.4 and 0.5

would vanish in the limit of infinite sizes.

Our results can be put in perspective by comparing them with a simple analytic approximation, the effective-medium approximation. For a site percolation problem on a lattice, the effective-medium approximation has been worked out at dc by Watson and Leath;[20] their equations are readily generalized to our ac problem and the resulting effective conductivity is shown in Fig. 6. Our calculations agree qualitatively with the EMA; the extra structure, as noted above, may be sample size dependent. Our results do confirm the basic prediction of the effective-medium approximation for finite frequencies, namely, that the im-

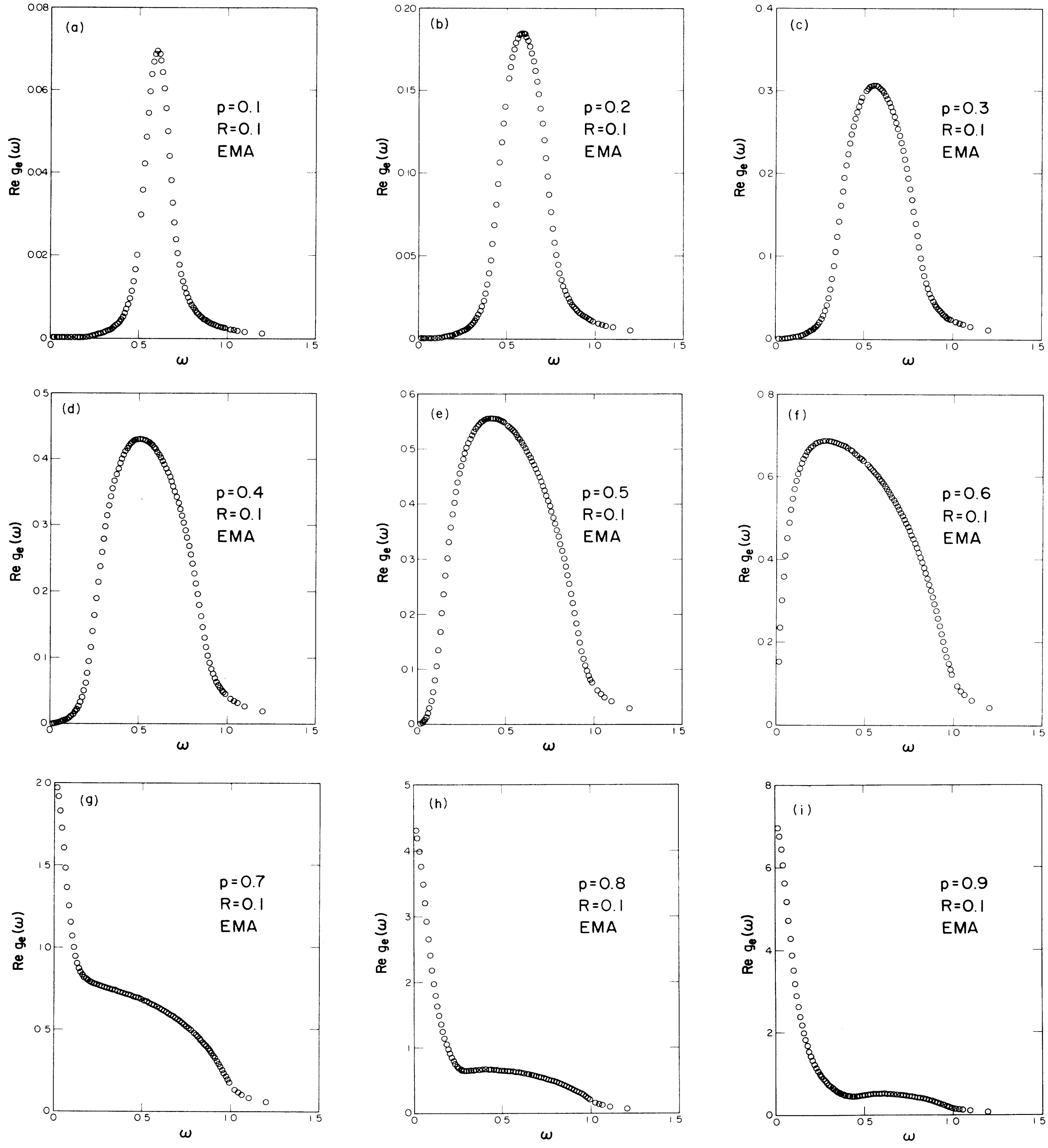

FIG. 6. Real part of the effective conductivity (in arbitrary units) as in Fig. 4, but as calculated in the effective-medium approximation.

purity band is separated from dc by a gap which goes to zero as the percolation threshold is approached.

Further work, using larger samples, would be useful to order to confirm that the resonance spectrum becomes smooth for large networks.

While the transfer-matrix algorithm is merely desirable in the calculations of Sec. III, it is essential here. The previously standard numerical technique for computing the effective conductivity of a random impedance network involves Gauss-Seidel relaxation (or overrelaxation) of the relevant linear equations (Kirchhoff's laws). When this technique is applied to a resonant random network, especially one with little damping, it does not converge, because of the presence of both positive and negative (imaginary) impedances. With the present technique, the impedance of a random network can be calculated at arbitrarily low damping. We include damping here only to smooth out the very complex structure found in the low-damping limit, not to achieve convergence.

V. DISCUSSION

We have considered two aspects of the finite-frequency response of metal-insulator composites. We have described a finite-size, finite-frequency scaling relation and verified it for two-dimensional composites with site percolation, using the transfer-matrix algorithm. Secondly, we have evaluated the surface-plasmon spectrum of a metal-insulator composite modeled as a random *LRC* network, again using the transfer-matrix algorithm.

In a real material, finite size will matter only if one of the sample dimensions is comparable to the percolation correlation length ξ. The same effect can be achieved in an infinite sample if the wavelength becomes comparable to ξ. However, it appears that the scaling relation discussed in Sec. II still cannot explain some of the anomalous experimental results,[17,21–23] in particular those of Ref. 17, relating the frequency dependence of the effective conductivity near the percolation threshold. Perhaps these experiments can be explained if one goes beyond the quasistatic approximation ($\nabla\times\mathbf{E}=0$) and includes eddy current effects, although this too seems unlikely in two dimensions, since the magnetic field of the incident radiation is typically parallel to the sample.

Our results for the surface-plasmon absorption spectrum are potentially relevant to real experiments. We have shown in two dimensions that the qualitative predictions of the effective-medium approximation are correct for this spectrum, provided that the geometrical assumption underlying the approximation (random distribution of dielectric and metallic cells) is satisfied.

Because of the small size of our surface-plasmon simulations, we cannot examine one intriguing possibility about this spectrum—the existence of a Lifshitz tail in the surface-plasmon density of states. Such tails are well known in electronic disordered systems.[24] A tail of this kind would probably fill up the gap in the surface-plasmon density of states, although the density of tail states would be exponentially small in the gap at low frequencies, just as in crystalline semiconductor alloys. The tail would be rather difficult to see in the surface-plasmon spectrum because, in a real composite, the surface-plasmon states always have a finite lifetime τ, i.e., a finite-frequency width. The tail would therefore be concealed under larger density of states arising from relaxation-time broadening.

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