Frequency dependence of the polarization catastrophe at a metal-insulator transition and related problems

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A scaling assumption is made in order to analyze the critical behavior of the complex dielectric constant $\epsilon(\omega)$ at low frequencies near a metal-insulator or a superconductor–normal-conductor transition in a disordered system. Percolative transitions as well as microscopic or quantum transitions are discussed. On either side of the metal-insulator transition, $\mathrm{Re}\epsilon(\omega)$ is found to have a peak at $\omega=0$ whose width tends to zero and whose height diverges at the transition. Similar behavior is found for the normal component of conductivity on both sides of the superconducting transition, and the effective penetration depth is found to diverge as $(p-p_c)^{-t/2}$ near the percolation threshold in a composite superconductor.

Following theoretical predictions, based on percolation theory, that the dc dielectric constant would diverge at a metal-insulator transition,[1,2] such behavior has recently been observed to occur in a real system of small Ag particles randomly dispersed throughout a KCl host.[3] Somewhat earlier, a similar behavior was observed in P-doped Si crystals,[4] although in that system, the transition is apparently not percolative.

In this Communication, we discuss theoretically the frequency dependence of the dielectric constant $\epsilon(\omega)$ at low frequencies near the metal-insulator transition both for the case of a metal-insulator composite near the percolation threshold of the metal, and for a microscopic or quantum type of homogeneous transition due to electron localization. From general considerations, we show that a characteristic peaked structure in $\mathrm{Re}\epsilon(\omega)$ can be expected centered at $\omega=0$, both for a percolation and for a microscopic type of metal-insulator transition. Moreover, this structure is expected both in the insulating and in the metallic phases (i.e., both above and below the transition), although in the metallic phase $\mathrm{Re}\epsilon(\omega)$ will be masked by a huge value of $\mathrm{Im}\epsilon(\omega)$ at low frequencies arising from the finite dc conductivity.

We also discuss, using precisely analogous mathematical considerations, the case of a composite of normal metal and superconductor near the percolation threshold of the superconducting component. Here we find, in addition to the well-known characteristic divergence of the dc conductivity below the threshold, a peak in $\mathrm{Re}\sigma(\omega)$ centered at $\omega=0$ with a half-width that vanishes as the percolation threshold is approached from either side. This is the analog of ac fluctuation paraconductivity well known in the theory of homogeneous superconductors above T_c. Above the percolation threshold, the same argument leads to a prediction of the dependence of the effective superfluid density and London penetration depth on volume fraction of superconductor, which may have been seen experimentally.

In the case of a percolative transition in a two-component composite medium, we use a scaling form for the bulk effective dielectric constant ϵ_e, valid for $|\epsilon_1/\epsilon_2| \ll 1$ and $|p_2-p_c| \ll 1$ (ϵ_e, ϵ_1, ϵ_2 are in general complex, p_1, p_2 are the volume fractions, and p_c is the percolation threshold for the ϵ_2 component[2,5,6])

$$\frac{\epsilon_e}{\epsilon_2} = |p_2-p_c|^t f\left(\frac{\epsilon_1/\epsilon_2}{|p_2-p_c|^{t+s}}\right) . \tag{1}$$

The scaling function $f(z)$, where z is a complex variable, has the following asymptotic forms:

$$f(z) = \begin{cases} A + Bz + \cdots & \text{for } |z| \ll 1, \quad p_2 > p_c \\ B'z + \cdots & \text{for } |z| \ll 1, \quad p_2 < p_c \\ A'' z^{t/(t+s)} & \text{for } |z| \gg 1, \text{ any } p_2 \end{cases} . \tag{2}$$

We now focus on the case of metallic inclusions ϵ_2 in an insulating host ϵ_1, writing

$$\epsilon_2 = \frac{4\pi i \sigma_m}{\omega}, \quad \epsilon_1 = \epsilon_i, \quad \frac{\epsilon_1}{\epsilon_2} = \frac{\epsilon_i \omega}{4\pi i \sigma_m} \ll 1 , \tag{3}$$

where ϵ_i and σ_m are real. From the asymptotic forms of $f(z)$, we deduce

$$\mathrm{Re}\epsilon_e(\omega) = \begin{cases} A''\epsilon_1^{t/(t+s)} \left(\dfrac{4\pi\sigma_m}{\omega}\right)^{s/(t+s)} \cos\left(\dfrac{\pi}{2}\dfrac{s}{t+s}\right), & \left|\dfrac{\epsilon_1}{\epsilon_2}\right| \gg |p_2-p_c|^{t+s} \\ B'\epsilon_1 |p_2-p_c|^{-s}, & \left|\dfrac{\epsilon_1}{\epsilon_2}\right| \ll |p_2-p_c|^{t+s}, \quad p_2 < p_c \\ B\epsilon_1 |p_2-p_c|^{-s}, & \left|\dfrac{\epsilon_1}{\epsilon_2}\right| \ll |p_2-p_c|^{t+s}, \quad p_2 > p_c \end{cases} . \tag{4}$$

Viewed as a function of ω, $\mathrm{Re}\epsilon_e$ thus has a peak at $\omega=0$ whose half-width

$$\Delta\omega \cong \frac{4\pi\sigma_m}{\epsilon_i}|p_2-p_c|^{t+s} \tag{5}$$

decreases to zero at p_c, and whose height is proportional to $\epsilon_1|p_2-p_c|^{-s}$, diverging at p_c. These results hold for p_2 either below or above p_c. Above p_c the peak in $\mathrm{Re}\epsilon_e$ will of course be masked by the huge value of $\mathrm{Im}\epsilon_e$ at small ω. The peak should nevertheless be observable even then in an appropriate experiment.

To estimate $\Delta\omega$, assume that $\epsilon_i=1$, $\sigma_m=10^{17}$ sec^{-1} and $|p_2-p_c|=0.01$. Calculations on three-dimensional (3D) random-resistor-networks lead to[7] $t+s\cong 2.4$. Using these data, $\Delta\omega/2\pi c\cong 100\ \mathrm{cm}^{-1}$; i.e., the peak falls off considerably when far-infrared frequencies are reached. This estimate assumes, of course, that σ_m itself is not frequency dependent on the scale of 100 cm^{-1}. A typical Drude peak width, however, is of the order of 1000 cm^{-1} in a pure metal, and probably more in a composite (due to a shortening of the relaxation time when the metal is in the form of small grains) so that sufficiently close to p_c, $\Delta\omega$ will be principally determined by the critical phenomenon just described.

We turn next to another application of the ideas just presented, namely, composites of normal (N) and superconducting (S) metals. In this case, it is more convenient to consider the critical behavior of the complex conductivity $\sigma_e(\omega)$ rather than of the complex dielectric constant $\epsilon_e(\omega)$. For the normal metal and superconductor we take

$$\sigma_1=\sigma_n,\quad \sigma_2=\frac{iG_0}{\omega}\ . \tag{6}$$

The S component thus has an inductive contribution arising from the first London equation of superconductivity. The scaling relation is now given by (1) and (2), with σ_1, σ_2 substituted for ϵ_1, ϵ_2. From this we deduce the following relations:

$$\mathrm{Re}\sigma_e(\omega)=\begin{cases} A''\sigma_1^{t/(s+t)}\left(\dfrac{G_0}{\omega}\right)^{s/(t+s)}\cos\left(\dfrac{\pi}{2}\dfrac{s}{s+t}\right), & \left|\dfrac{\sigma_1}{\sigma_2}\right|\gg|p_2-p_c|^{t+s} \\ B'\sigma_1|p_2-p_c|^{-s}, & p_2<p_c \text{ and } \left|\dfrac{\sigma_1}{\sigma_2}\right|\ll|p_2-p_c|^{t+s} \\ B\sigma_1|p_2-p_c|^{-s}, & p_2>p_c \text{ and } \left|\dfrac{\sigma_1}{\sigma_2}\right|\ll|p_2-p_c|^{t+s}\ . \end{cases} \tag{7}$$

In addition, we have

$$\mathrm{Im}\sigma_e(\omega)=\frac{G(p_2)}{\omega}\ ,$$
$$G(p_2)\propto G_0(p_2-p_c)^t,\quad p_2>p_c\ , \tag{8}$$

which states that the strength of the inductive part of the conductivity vanishes as $(p_2-p_c)^t$ on the superconducting side of the percolation threshold.

Several immediate experimental consequences can be deduced from Eqs. (7) and (8). First, from Eq. (7), $\mathrm{Re}\sigma_e(\omega)$ has a peak at $\omega=0$ whose half-width on *both* sides of the percolation threshold is

$$\Delta\omega\cong\frac{G_0}{\sigma_n}|p_2-p_c|^{s+t}$$
$$=\frac{\sigma_n'}{\sigma_n}\frac{\pi\Delta}{\hbar}\tanh\left(\frac{\Delta}{2k_BT}\right)|p_2-p_c|^{s+t}\ . \tag{9}$$

The last estimate expresses G_0 in terms of the superconducting energy gap Δ, temperature T, and normal conductivity σ_n' of the S constituent, using the low-frequency limit of the Mattis-Bardeen expression for the ac conductivity of a superconductor.[8] Particularly interesting is the peak on the *normal* side of the transition, which amounts to a *percolative analog of the usual Aslamasov-Larkin fluctuation paraconductivity*[9] in an ordinary homogeneous superconductor above T_c. For $p-p_c$ of order 0.05, typical values of σ_n'/σ_n and Δ yield peak widths in the GHz region.

A possible probe of Eq. (8) for the *inductive* part of the conductivity would be a measurement of the penetration depth in a composite superconductor. The effective coefficient $G(p_2)$ plays the role of n_se^2/m in the London equations, where n_s is the superfluid number density. From this connection, the London penetration depth λ_L should vary with p_2 according to the rule

$$\lambda_L(p_2)\propto n_s^{-1/2}\propto(p_2-p_c)^{-t/2}\ . \tag{10}$$

This exponent is about 0.86 in three dimensions and 0.55 in two.[7]

To observe such phenomena would require a percolative transition in a real N-S composite. In some

composites, it is possible that the effective volume fraction of superconductor is actually a linear function of temperature. In such materials, the peak in the paraconductivity should satisfy an equation similar to (9) but with the variable $p_2 - p_c$ replaced by $T - T_c$. Similarly, below the transition temperature, the London penetration depth should diverge as $(T_c - T)^{-t/2}$. It is possible that the unusual temperature dependence of λ_L recently observed by Raboutou *et al.*[10] may be due to such a percolative transition.

We now turn to a discussion of $\epsilon(\omega)$ in a system that has a quantum or microscopic metal-insulator transition. Viewed as a function of the complex frequency z, $\epsilon(z)$ must always be analytic for z in the upper half plane, with a pole at $z=0$ in the metallic state. The experimental results of Capizzi *et al.*,[4] who observed the onset of a polarization catastrophe as the metal-insulator transition in a doped semiconductor is approached from the insulating side, indicate that $\epsilon(z)$ has a singularity at a point in the lower half plane that is approaching $z=0$ at the transition.

One possibility to consider is that $\epsilon(z)$ has a pole whose position approaches the origin and whose residue tends to zero as the transition is approached from the insulating side; i.e.,

$$\epsilon(z) \cong \frac{A_0}{\omega_0 - iz}, \quad A_0 > 0, \quad \omega_0 > 0 \ , \qquad \omega, A_0 \to 0 \text{ as } n \to n_{MI} \ , \tag{11}$$

where n is the concentration of microscopic impurities. In a dc measurement, one would then observe

$$\mathrm{Re}\epsilon(\omega=0) \cong \frac{A_0}{\omega_0} \propto (n_{MI} - n)^{-\zeta} \ . \tag{12}$$

The experimental value found for the exponent is $\zeta = 1.09$ (see Ref. 4).

In order to determine whether such a description is consistent, we argue as follows (following a similar argument due to Harris[11]): We subdivide our system into volumes of size ξ^d where ξ is the localization length of individual electrons in the insulating state and d is the dimensionality. The length ξ is obviously the smallest scale on which one can still discuss bulk properties. Because the impurities are randomly distributed, each subvolume will have a slightly different value of n, and hence of ω_0. Since ω_0 is an intensive bulk property, its rms fluctuation is given by

$$(\Delta\omega_0)_{\mathrm{rms}} \propto \xi^{-d/2} \propto (n_{MI} - n)^{d\nu/2} \ . \tag{13}$$

For consistency of the single-pole description, $(\Delta\omega_0)_{\mathrm{rms}}$ should be much smaller than the average value of ω_0, and therefore we must have

$$A_0 > (n_{MI} - n)^{d\nu/2 - \zeta} \ . \tag{14}$$

Since we also require that $A_0 \to 0$ as $n \to n_{MI}$, we get an inconsistency unless

$$d\nu > 2\zeta \ . \tag{15}$$

Experimentally, $\nu = 0.55$ (see Ref. 12) and $\zeta = 1.09$ (see Ref. 4) for a metal-insulator transition in a doped bulk semiconductor. In that case, the above inequality is clearly violated, and it is thus impossible to have a single value of ω_0 that characterizes the behavior of the entire system. In 2D systems the situation is as yet unclear because there are no results available for the exponent ζ.

Having shown that it is inconsistent to assume Eq. (11) for $\epsilon(z)$, we turn to the other possibility $-\epsilon(z)$ has a branch point singularity in the lower half plane that approaches $z=0$ at the transition. In that case, we are able to assume a scaling form for $\epsilon(z)$, valid when both $|n - n_{MI}|$ and $|z|$ are small, namely,[13]

$$\epsilon(z) = |n - n_{MI}|^{-\zeta} f\left(\frac{-iz}{|n - n_{MI}|^{\mu+\zeta}}\right) \ . \tag{16}$$

Here $f(v)$ is a function of the complex variable v with the following forms of asymptotic behavior:

$$f(v) = \begin{cases} B - Cv + \cdots & \text{for } |v| \ll 1, \quad n < n_{MI} \\ \dfrac{A'}{v} + B' - C'v + \cdots & \text{for } |v| \ll 1, \quad n > n_{MI} \\ \dfrac{A''}{v^{\zeta/(\zeta+\mu)}} & \text{for } |v| \gg 1, \text{ any } n \ . \end{cases} \tag{17}$$

Clearly, the exponent μ characterizes the behavior of the dc conductivity above n_{MI}, while ζ characterizes the divergent behavior of $\mathrm{Re}\epsilon$ at $\omega = 0$ (we expect it to diverge as $n \to n_{MI}$ from either side).

It is useful to contrast the present approach with other scaling approaches to the metal-insulator transitions that have been put forward in recent years.[14–18] Rather than being based upon correlation lengths associated with electronic wave functions, our scaling assumption arises from a more phenomenological approach that focuses on the experimentally controllable parameters ω and n. For this reason, Eq. (16) is similar in form to Eq. (1), which embodies the scaling assumption for the metal-insulator transition in a percolative system.

We can now use Eq. (16) to discuss the behavior of $\mathrm{Re}\epsilon(\omega)$ as a function of frequency. As in the case of the percolative transition, discussed before, $\mathrm{Re}\epsilon(\omega)$ must have a peak at $\omega = 0$ whose width tends to zero and whose height diverges at $n = n_{MI}$:

$$\mathrm{Re}\epsilon(\omega) \cong \begin{cases} \dfrac{A''}{\omega^{\zeta/(\zeta+\mu)}} \cos\left(\dfrac{\pi}{2}\dfrac{\zeta}{\zeta+\mu}\right) \text{ for } \dfrac{\hbar\omega}{E_c} >> \left|(n-n_{MI})\dfrac{1}{E_c}\dfrac{dE_c}{dn}\right|^{\mu+\zeta} \\ \dfrac{B}{|n-n_{MI}|^{\zeta}} \text{ for } \dfrac{\hbar\omega}{E_c} << \left|(n-n_{MI})\dfrac{1}{E_c}\dfrac{dE_c}{dn}\right|^{\mu+\zeta}, \quad n < n_{MI}\ , \\ \dfrac{B'}{|n-n_{MI}|^{\zeta}} \text{ for } \dfrac{\hbar\omega}{E_c} << \left|(n-n_{MI})\dfrac{1}{E_c}\dfrac{dE_c}{dn}\right|^{\mu+\zeta}, \quad n > n_{MI} \end{cases} \tag{18}$$

$$\hbar\Delta\omega \cong E_c\left|(n-n_{MI})\frac{1}{E_c}\frac{dE_c}{dn}\right|^{\mu+\zeta} . \tag{19}$$

Using the results of Ref. 12, namely, $\mu=\frac{1}{2}\zeta=0.55$, we could get an estimate of $\Delta\omega$ if we knew the rate of change of the mobility edge E_c with concentration n. Conversely, by measuring $\Delta\omega$, we would in effect be measuring the quantity $E_c^{-1}dE_c/dn$.

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