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LETTER TO THE EDITOR

Dynamics of random media: anomalous wave propagation and damping near the percolation threshold

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Abstract. It is shown that a description of the dynamics of random media, consisting of a mixture of conductors and dielectrics or of polymers close to the sol–gel transition, involves both the conductivity and the superconductivity exponents t and s. The mixing of the two exponents is derived explicitly for wave propagation and dissipation close to the percolation threshold. At high frequencies propagation and dispersion are equally important and the disperison relation, $k^2 \sim \omega^\alpha$, is anomalous and α depends on *both* t and s. At low frequencies there is both propagation and dissipation above and below the threshold.

The dynamic properties of percolation clusters, and fractal structures in general, has recently received considerable theoretical [1–4] and experimental [5–9] attention. Much of this activity was prompted by the introduction of the concept of *fractons* by Alexander and Orbach [3], who made the observation that diffusion on fractals is anomalous. The importance of this anomalous behaviour lies in the relation between diffusion and conduction that was first recognised by de Gennes [1, 2] in the percolation problem. As he pointed out [2, 10, 11], depending on whether the system is a random-resistor network or a random-superconducting network, two different cases must be considered. For the random-resistor network [10, 12, 13] the DC conductivity Σ goes to zero as we approach the percolation threshold from above with a characteristic exponent t:

$$\Sigma \sim (p - p_c)^t \qquad (p > p_c). \tag{1}$$

Here $(p - p_c)$ is the distance to the percolation threshold p_c. In the case of a mixture of resistors and superconductors [11–13] Σ diverges as we approach p_c from below with an exponent s:

$$\Sigma \sim (p_c - p)^{-s} \qquad (p < p_c). \tag{2}$$

Because of the Einstein relation between conductivity and diffusion [1], it was then possible to define not one, but two, *independent* diffusion problems, namely the ant [1, 3, 4] and termite [2, 14, 15] models of conduction. Because these two types of diffusion are decoupled from each other, we get two possible dynamics for the systems. This decoupling is clearly not a general result. For example, the viscoelastic behaviour of randomly branched polymers and gels in their reaction bath may be related to a

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combination of the random-resistor and the random-superconducting network problems [16]. This implies that the two problems must be treated simultaneously. Similarly, mixtures of dielectric and metallic materials exhibit non-trivial frequency-dependent properties [17]. It seems that for such systems there is a strong coupling between both problems and that they should not be considered separately. Indeed, we know that very generally they are related to the real and imaginary parts of the same response function and are related to each other by the Kramers–Kronig (KK) relations [18]. Recently Laibowitz and Gefen [5] used such a condition for a random-resistor network. Consistency then imposes that $s = 2\nu - \beta$, in disagreement with experimental results and numerical simulations. The difficulty resides in the moment when to introduce the KK relations. A correct approach has to take into account these relations from the start. Thus, even though relations (1) and (2) are valid for DC properties, as soon as the frequency is non-zero, the simple diffusion models are not longer valid, because both energy dissipation and storage have to be taken into account.

As a first step in treating both propagation and dissipation in random systems, in this letter we report the results of a study of wave propagation close the percolation threshold. This might be an elastic wave for a sol or a gel in its reaction bath, or an electromagnetic wave in the case of a mixture of dielectric and metallic components. We find that propagation is critically slowed down near the threshold, and because of the Kramers–Kronig relations between the real and the imaginary parts of the dielectric constant, an extremely rich situation occurs in which the dynamics depends on both s and t, i.e. the two anomalous transport laws are coupled. At low frequencies below the threshold, we find that the group velocity is reduced and vanishes at the percolation threshold.

Let k and ω be the wavevector and the frequency of the wave, respectively. One may write down the usual dispersion relation [18]

$$k^2 = \bar{\varepsilon}(\omega)c^{-2}\omega^2 \tag{3}$$

where c is the velocity in the vacuum and $\bar{\varepsilon}$ is the complex response function, which is the dielectric constant in the case of a dielectric, and is related to the elastic constant for a sol (or gel), for scalar elasticity. In the case of a dielectric,

$$\bar{\varepsilon}(\omega) = \varepsilon + i\sigma/\omega \tag{4}$$

and for the case of a sol or gel,

$$\bar{\varepsilon}(\omega) = \eta - iG/\omega \tag{4'}$$

where $i^2 = -1$, ε and σ are the real part of the dielectric constant and the conductivity of the dielectric, respectively, and η and G are the viscosity and the elastic modulus of the sol (or the gel), respectively. The complex dielectric constant in the percolation case was considered first by Efros and Shklovskii [17] some years ago and studied more recently by the Marseille [19] and Schlumberger [20] groups. The basic result is that $\bar{\varepsilon}(\omega)$ close to p_c has the following scaling form [17]:

$$\bar{\varepsilon}(\omega) = (p - p_c)^{-s}f_{\pm}(i(\omega/\omega_0)(p - p_c)^{-s-t}) \tag{5}$$

where ω_0 is a characteristic (molecular) frequency, $\omega \ll \omega_0$, and s and t are defined in (1) and (2). For small x, the scaling function $f_{\pm}(x)$ reduces to appropriate forms which are consistent with (1) and (2). In particular, below p_c, $f_{-}(x)$ has the following form:

$$f_{-}(x \ll 1) = 1 + ax + \ldots \tag{6}$$

and above p_c, it reduces to

$$f_+(x \ll 1) = \frac{1}{x} + \frac{a'}{x^2} + \dots \qquad (6')$$

where a and a' are real constants. In the opposite limit of high frequencies, the complex dielectric constant behaves as a power law of ω, without any dependence on $(p - p_c)$:

$$f_\pm(x \gg 1) \sim x^{-s/(s+t)}. \qquad (7)$$

The high-frequency behaviour of the scaling relation (7) was recently discussed by Laugier *et al* [19] and checked experimentally by Durand *et al* [8] on polymeric systems close to the gelation threshold.

Relation (5) may be written in a form exhibiting a characteristic time τ:

$$\bar{\varepsilon}(\omega) = (p - p_c)^{-s} f_\pm(i\omega\tau) \qquad (8a)$$

with

$$\omega_0 \tau \sim (p - p_c)^{-s-t}. \qquad (8b)$$

Several points should be noted at this time. First, the real and the imaginary parts of the response function are not independent, but are related by the Kramers–Kronig relations. Therefore, the characteristic time of the mixture depends on both the real and the imaginary parts, as may be seen from (5) or (8). The interpretation of this time is readily obtained by first combining (3) and (4) to find

$$k^2 = c^{-2}\omega^2(\varepsilon(\omega) - i\sigma(\omega)/\omega). \qquad (9)$$

Relations (6), (7) and (9) show that for high and low frequencies two distinct behaviours are generally expected. For small ω, the solution is propagative and the group velocity $V(\omega)$ is

$$V(\omega) = c\varepsilon(\omega)^{-1/2} \qquad (\omega\tau \gg 1) \qquad (10)$$

where τ is the characteristic time, to be discussed below. For larger frequencies, the solution is dispersive. This corresponds to a broadening of the evanescent wave and is directly related to the existence and frequency dependence of the conductivity. The crossover frequency between the two regimes corresponds to the characteristic time τ, relation (8), which may also be obtained by combining (2), (6) and (9):

$$\omega_0 \tau \sim (p - p_c)^{-s-t}. \qquad (8b)$$

Note that the crossover time depends on both s and t, in agreement with the experimental results [8]. Keeping only the constant term in relation (6) would lead to a propagating wave with constant velocity V without any attenuation, and crossing over to a regime with strong damping at frequencies higher than τ^{-1}. In fact, even for small frequencies, relation (6) shows that there is a finite contribution to $\sigma(\omega)$, which implies that the penetration depth is finite.

Let us first consider the high-frequency regime where the response function has a power-law behaviour. Using (5), (7) and (9), we find

$$(kc)^2 \sim \omega^2(i\omega/\omega_0)^{-s/(s+t)} \qquad (\tau^{-1} \ll \omega \ll \omega_0). \qquad (11)$$

Setting $k = k_r + i k_{im}$ where k_r and k_{im} correspond, respectively, to propagation and attentuation, we find $k_r \approx k_{im} \approx \omega^\alpha$ with $\alpha = (s + 2t)/2(s + t)$. Therefore propagation and damping behave in the same way. The real part implies that the group velocity is

$$V(\omega) \sim c(\omega/\omega_0)^{s/2(s+t)} \qquad (\tau^{-1} \ll \omega \ll \omega_0) \tag{12}$$

and from the imaginary part, from $k^2 \sim D(\omega)\omega$, we define a dispersion constant:

$$D(\omega) \sim (\omega/\omega_0)^{-t/(s+t)} \omega_0^{-1} c^2 \qquad (\tau^{-1} \ll \omega \ll \omega_0). \tag{13}$$

These relations show that wave propagation is dramatically slowed down in these mixtures. At the same time, if a wavepacket is sent in, its width broadens with a dispersion constant $D(\omega)$. Both relations (12) and (13) may be checked experimentally. It is important to note that, because of (11), propagation and attentuation are of the same order of magnitude. Note also that the dispersion constant is inversely proportional to the conductivity of the medium [18]. Finally, we can calculate the characteristic penetration length,

$$\Lambda \sim V(\omega)/\omega \sim (D(\omega)/\omega)^{1/2} \sim (\omega/\omega_0)^{-(s+2t)/2(s+t)} c \omega_0^{-1}. \tag{14}$$

As ω is decreased, starting from this high-frequency regime, the anaomalous propagation–dispersion is present as long as $\omega\tau$ is larger than unity. For lower frequencies, $\omega\tau \ll 1$, there is a crossover to other types of behaviour that depend on whether the system is below or above the threshold. For $p < p_c$, the velocity of the wave is a constant. From (10) and (2) we get

$$V \sim c(p - p_c)^{s/2}. \tag{15}$$

Combining (12) and (15), $V(\omega)$ can be written in the following scaled form:

$$V(\omega) \sim c(p - p_c)^{s/2} g((\omega/\omega_0)(p - p_c)^{-s-t}). \tag{16}$$

Similarly, from (4)–(6) one can determine the conductivity [20]

$$\sigma \sim (\omega^2/\omega_0)(p_c - p)^{-2s-t} \qquad (p < p_c, \omega\tau \ll 1). \tag{17}$$

The penetration depth Λ is given by

$$\Lambda \sim (\sigma\omega)^{-1/2}$$
$$\Lambda \sim \omega^{-3/2} \omega_0^{1/2} (p - p_c)^{(2s+t)/2} \qquad (p < p_c, \omega\tau \ll 1) \tag{18}$$

which diverges as ω vanishes, as it should, because the medium is a pure dielectric at zero frequency. Equation (18) may also be generalised in a scaled form involving $\omega\tau$ as the variable.

For $p > p_c$, on the other hand, the medium is a conductor with a conductivity given by (1). Thus for low frequencies, Λ is

$$\Lambda \sim (\omega\sigma)^{-1/2} \sim \omega^{-1/2}(p - p_c)^{-t/2} \qquad (p > p_c, \omega\tau \ll 1). \tag{19}$$

From the second term in the $1/x$ expansion of $\bar{\varepsilon}$, equation (6'), the real part of the dielectric constant can be determined:

$$\varepsilon \sim (\omega/\omega_0)^{-2}(p - p_c)^{s+2t} \qquad (p > p_c, \omega\tau \ll 1). \tag{20}$$

The above result implies that there is propagation no longer: inserting (20) in (3) and (4) leads to

$$k^2 \sim (p - p_c)^{s+2t} \qquad (p > p_c, \omega\tau \ll 1) \tag{21}$$

corresponding to the crossover expression for the penetration depth for $\omega\tau \sim 1$.

The above considerations are also applicable to the gelation transition. But, in this case it was conjectured by de Gennes [2] that, for scalar elasticity, the real part of the modulus G behaves as the conductance above p_c and the viscosity behaves as the dielectric constant below p_c, so that all the laws discussed above are inverted above and below p_c.

In summary, we considered the problem of dynamics at the percolation transition by studying propagation and dispersion of waves in a random mixture of conductors and dielectric materials†. Our main point was that, although the ant and termite models of conduction provide a good description of the DC properties of random materials, they fail to describe the frequency dependencies for most materials. The difficulty lies in the fact that they are usually related to the storage and dissipation of energy and are not independent of each other, as is implicitly assumed in the random walk models. we found that, at high frequencies, propagation and dispersion are equally important and the dispersion relation is anomalous with an exponent that depends on *both* the conductivity exponent t and the superconductivity exponent s, i.e. that the dynamics couples dispersion and propagation. The longest relaxation time also depends on both s and t. At low frequencies below the threshold, the group velocity is reduced and vanishes at the threshold. There is still attenuation, with a penetration length that diverges as $\omega \to 0$. The above results are amenable to experimental verification in a wide variety of random systems, including mixtures of good and poor conductors and branched polymers in the reaction bath near the sol–gel transition.

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† Sound wave propagation in the decoupled limit was considered in [21].

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