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1989 J. Phys. A: Math. Gen. 22 3681

(http://iopscience.iop.org/0305-4470/22/17/033)

J. Phys. A: Math. Gen. 22 (1989) 3681-3692. Printed in the UK

Hopping conduction in insulator–conductor–superconductor mixtures

A Puri† and T Odagaki‡§

† Department of Physics, University of New Orleans, New Orleans, LA 70148, USA
‡ Department of Physics, Brandeis University, Waltham, MA 02254-9110, USA

Received 11 July 1988

Abstract. The coherent medium approximation is applied to study the frequency-dependent diffusion constant in two- and three-dimensional ant-termite like mixtures, where three types of jump rates, 'normal' (metallic), 'superconducting' and 'insulating' are distributed randomly. The DC diffusion constant vanishes when $(p+r)\leq p_c$ and is proportional to $(p+r-p_c)/(r_c-r)$ when $p+r>p_c$, where p and r are the probabilities that a given bond is metallic and superconducting, respectively, and z is the coordination number of the lattice, $p_c=r_c\equiv 2/z$. In the low-frequency limit, the real and imaginary parts of the AC diffusion constant go as $A(z, d, p, r)f(\omega)$ and $B(z, d, p, r)g(\omega)$, respectively, a behaviour similar to the pure ant limit. The behaviour of $f(\omega)$ and $g(\omega)$ is examined below, at and above the percolation thresholds, in both two and three dimensions. The coefficients A and B of these leading terms diverge as $r\to r_c$ (superconducting region percolates), indicating enhancement in the dielectric constant.

1. Introduction

Stochastic transport in disordered media has been studied extensively in recent years (Havlin *et al* 1986, Haus and Kehr 1986, Hong *et al* 1986, Odagaki 1987, Havlin and Ben-Avraham 1987). Various physical and biological applications have been discussed by Shlesinger and West (1984) and Weiss and Rubin (1983). The stochastic motion of carriers is usually described by a random walk equation with random jump rates, i.e. carriers are assumed to perform a random walk in a random environment. The distribution of jump rates is selected to describe the actual stochastic nature of the system under consideration. For example, hopping conduction in doped semiconductors has been investigated via a random walk model in which the distribution of jump rates is determined by the exponential distance dependence of jump rates and by the Hertz distribution of nearest-neighbour distances (Scher and Lax 1973, Odagaki and Lax 1981). The percolation model where a jump rate has non-zero probability of vanishing has been extensively studied by Odagaki *et al* (1983). This represents a simple model of binary mixtures (Kirkpatrick 1973, Hong *et al* 1986). The bond percolation model is interesting in its own right, since the hopping model provides a physical description of the percolation process as a random walk (de Gennes 1976). De Gennes termed the transport process in normal (metallic) and insulating mixtures as an 'ant in a labyrinth'. This term arises from the fact that one can replace the

§ Present address: Department of Physics, Kyoto Institute of Technology, Kyoto 606, Japan.

conductivity problem with the diffusion problem using the Nernst–Einstein relation (Scher and Lax 1973).

Recently, in a similar spirit, de Gennes (1980) proposed a termite model for conductance in binary mixtures where one element is superconducting and the other element is normal. His results did not show percolation phenomena. De Gennes' work has been investigated further by Coniglio and Stanley (1984), Adler *et al* (1985), Bunde *et al* (1985), Sahimi and Saddiqui (1985), Hong *et al* (1986) and Havlin *et al* (1986). A close comparison between some of these models in one dimension has been carried out by Leyvraz *et al* (1986). Odagaki (1986) analysed the dynamic diffusion of the termite problem using the hopping model. His results show percolation phenomena in agreement with studies by previous authors except for de Gennes' treatment. He also showed that the termite limit of the trapping model leads to de Gennes result (Odagaki 1987).

There have been several experimental studies of random and inhomogeneous systems in recent years which in turn have aroused deep interest in the subject. A review of the experimental studies in the subject is given by Deutscher *et al* (1983). A wide variety of experiments has been discussed in this review, ranging from conductivities of thin films of lead deposited on an insulating substrate (Kapitulnik and Deutscher 1982) to thin films of superconductors deposited on a normal substrate (Orr *et al* 1985). There have been conductivity measurements in ionic conductors mixed with an insulating phase, where both ant and termite limits seem to play an important role as pointed out by Bunde *et al* (1985).

In this paper we address ourselves to the problem of ant-termite mixtures, i.e. a three-component system of normal (metallic), superconducting and insulating regions. This system is a special case of polychromatic percolation (Halley 1983). Following Odagaki and Lax (1981), in § 2 we obtain the coherent medium approximation (CMA) equation for hopping conduction in the ternary system. In § 3 the static diffusion constant is investigated. In § 4 the AC diffusion constant is studied for ant-termite mixtures in the square and cubic lattices. The pure ant and termite limits are recovered in appropriate limits. A brief summary is given in § 5.

2. CMA equation for a ternary system

The stochastic motion of a carrier is assumed to be governed by a random walk master equation (Scher and Lax 1973, Odagaki and Lax 1981):

$$\frac{\partial}{\partial t} P(s, t \mid s_0, 0) = -\Gamma_s P(s, t \mid s_0, 0) + \sum_{s' \neq s} w_{ss'} P(s', t \mid s_0, 0) \tag{1}$$

where the decay rate Γ_s is given by the sum of nearest-neighbour jump rates, $w_{s's}$

$$\Gamma_s = \sum_{s' \neq s} w_{s's}. \tag{2}$$

The quantity $P(s, t \mid s_0, 0)$ is the probability that a random walker is at site s after time t, when it started from site s_0 at $t = 0$. We consider hopping conduction in a ternary system where the distribution of jump rates obeys the probability distribution

$$P(w_{ss'}) = p\delta(w_{ss'} - w_p) + q\delta(w_{ss'} - w_q) + r\delta(w_{ss'} - w_r) \tag{3}$$

with $0 \le p, q, r \le 1$ and $p+q+r=1$. We set $w_p = 1$ and take ω_p as the unit of frequency. We consider the AC diffusion constant and its critical behaviour in the p, q, r domain using the CMA method of Odagaki and Lax (1981).

The CMA gives the self-consistent equation

$$\frac{1}{zw_c}(1-ug_{11}) \equiv \frac{1}{\Xi} = \left\langle \frac{1}{\Xi + 2(w_{12} - w_c)} \right\rangle \tag{4}$$

where $g_{11} \equiv P(1, u|1)$ is the Laplace transform of $P(1, t|1, 0)$, z is the coordination number, site 2 is the nearest neighbour of site 1, and the average $\langle \cdots \rangle$ is taken over w_{12}. The coherent jump rate $w_c(u)$ is to be self-consistently determined by the above equation. For distributions given by equation (3), the self-consistent condition for $w_c(u)$ is reduced to

$$\frac{1-ug_{11}}{zw_c} = \frac{2(a+b+c)}{2a(2w_c - w_q - w_r) + 2b(2w_c - w_r - w_p) + 2c(2w_c - w_p - w_q) \pm \sqrt{D}} \tag{5}$$

where

$$a = 2p(w_p - w_c) \qquad b = 2q(w_q - w_c) \qquad c = 2r(w_r - w_c) \tag{6a}$$

and

$$D = 4[(w_q + w_r)a + (w_r + w_p)b + (w_p + w_q)c]^2 - 16(a+b+c)(w_q w_r a + w_r w_p b + w_p w_q c). \tag{6b}$$

When $c = 0$, equation (5) is reduced to the self-consistent equation for the ant problem (Odagaki *et al* 1983).

The choice of + or − sign in equation (5) depends on whether $\mathrm{Re}\{2(w_r - w_q)a + 2(w_p - w_r)b\} > 0$ or < 0. The transition probability g_{11} in equation (5) is written in terms of the Hilbert transform $F(\xi)$ of the density of states $n(x)$ as (Odagaki *et al* 1983)

$$g_{11} = F(1 + u/zw_c(u))/zw_c(u) \tag{7}$$

where $F(\xi)$ is defined by

$$F(\xi) = \int_{-\infty}^{\infty} \frac{n(x)}{\xi - x} \, \mathrm{d}x. \tag{8}$$

The self-consistent condition for $w_c(u)$ given by equation (5) is a general expression for the ternary system. We consider here the ant-termite limit of the ternary system. Taking the limit of $w_q = 0$ and $w_r = \infty$ in the CMA equation (5), we find

$$\frac{1-ug_{11}}{zw_c} = \frac{w_c(1+r) - w_p(1-q) \mp \sqrt{D'}}{4(w_c - w_p)w_c} \tag{9}$$

where

$$D' = [w_c(1+r) - (1-q)w_p]^2 - 4rw_c(w_c - w_p). \tag{10}$$

3. Static diffusion constant

As can easily be seen, $ug_{11} \to 0$ as $u \to 0$. Therefore, equation (9) in the static limit becomes

$$\frac{1}{zw_c} = \frac{w_c(1+r) - w_p(1-q) \mp \sqrt{D'}}{4(w_c - w_p)w_c}. \tag{11}$$

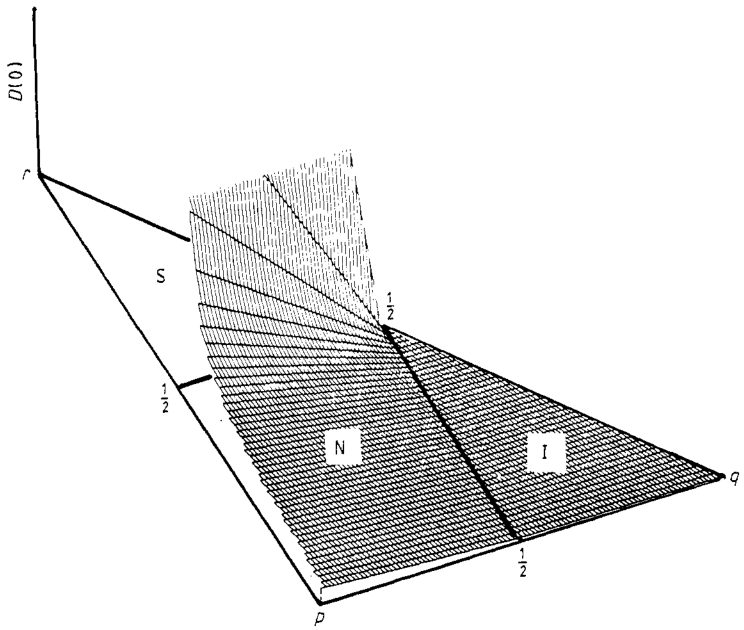

Figure 1. A perspective view of the DC diffusion constant in the (p, q, r) plane for $z=4$ obtained by the CMA method.

Supposing that $0<w_c<\infty$, we can simplify the above equation and obtain a solution for w_c in the static limit

$$w_c=\frac{2}{z-2}\frac{2/z-(p+r)}{(r-2/z)}. \tag{12}$$

When w_c given by equation (12) does not satisfy $0<w_c<\infty$, then $w_c=0$ or $w_c=\infty$ is the solution. Figure 1 shows the perspective view of w_c on the triangular region represented by (p, q, r) for $z=4$. Note that w_c diverges as r approaches $r_c\equiv 2/z$. The phase diagrams for the $z=4$ and $z=6$ cases are shown in figures 2(a) and (b), respectively. In the shaded regions of the triangles, none of the conducting phases percolate, though the system is normally conducting.

It is worth pointing out that when $r=0$, the pure ant limit (Odagaki *et al* 1983) is recovered

$$w_c=\frac{zp-2}{z-2}$$

and when $q=0$ $(p+r=1)$, the termite limit (Odagaki 1986) is recovered

$$w_c=\frac{2}{2-zr}.$$

4. Dynamic diffusion constant

In this section we investigate the dynamic or frequency-dependent diffusion constant in the ant-termite mixtures. The CMA equation (9) in the ant-termite limit can be

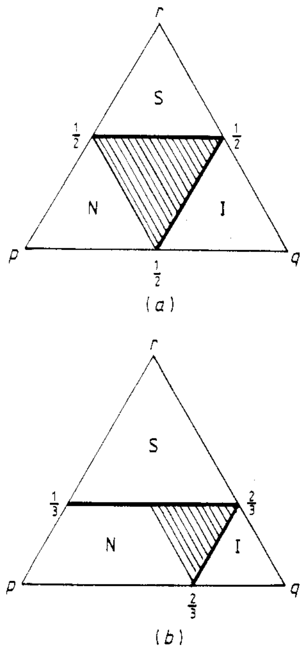

Figure 2. The phase diagram for the ternary system determined by the CMA method; (*a*) two dimensions, (*b*) three dimensions. Bold full lines indicate phase boundaries where the coefficients of the AC diffusion constant diverge. In the shaded area neither superconductor nor normal conductor percolates, though the DC diffusion constant does not vanish.

written as

$$\{w_c[-z^2r+2(1+r)z(1-ug_{11})-4(1-ug_{11})^2]+w_p[4(1-ug_{11})^2-2z(1-q)(1-ug_{11})]\}=0. \tag{13}$$

This equation is solved self-consistently for w_c for the square and simple cubic lattices.

4.1. Square lattice

The density of states of the square lattice is given by a complete elliptic integral of the first kind $K(x)$:

$$n(x)=\frac{2}{\pi^2}K(\sqrt{1-x^2}). \tag{14}$$

Therefore, the function $F(\xi)$ given by equation (8) is also expressed in terms of $K(x)$ as

$$F(\xi)=\frac{2}{\pi\xi}K\left(\frac{1}{\xi}\right). \tag{15}$$

The self-consistency equations (13), (7) and (15) were solved numerically for various values of p and r, and the dynamic diffusion constant $D(\omega)$ is obtained from (Odagaki and Lax 1981)

$$D(\omega)=a^2w_c(\mathrm{i}\omega) \tag{16}$$

where a is the nearest-neighbour distance. Figures 3, 4 and 5 show a frequency

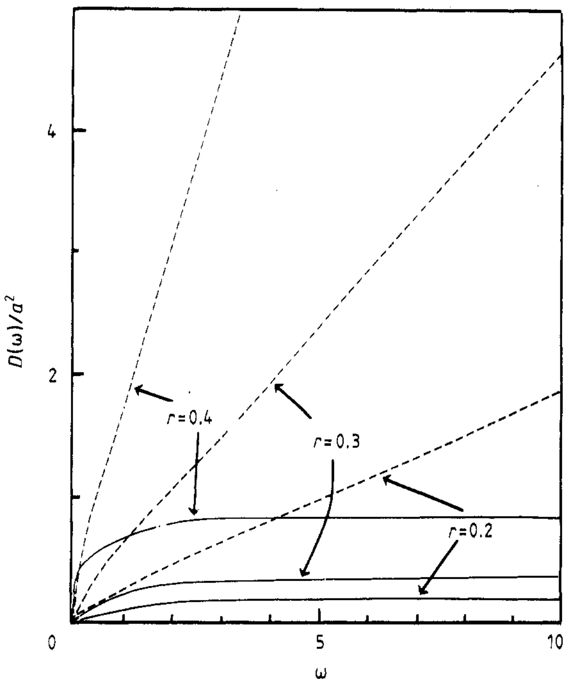

Figure 3. The AC diffusion constant for the square lattice determined by the CMA method. The normal jump rate w_p is set to unity and is used as the unit of frequency. Full curves are the real part and broken curves are the imaginary part for various values of r when $p=0.1$ satisfying $p+r \leqslant p_c$.

dependence of the real (full curve) and imaginary (broken curve) parts of the diffusion constant for the square lattice. These figures show results for several values of r for $p=0.1$ $(p+r \leqslant p_c)$, $p=0.3$ $(p+r \geqslant p_c)$, and $p=0.5$ $(p+r > p_c)$.

For $r=0.2$, the low-frequency part of these figures is expanded in figures 6(a) and (b) in the logarithmic scale, where $D(\omega)-D(0)$ is plotted, with $D(0)$ determined by equation (12) for $z=4$.

Examining the behaviour of $F(\xi)$ around $\xi=1$, we can easily determine the low-frequency behaviour of the AC diffusion constant for general two-dimensional lattices.

(i) For $p+r > p_c (\equiv \frac{1}{2})$

$$D(\omega)=a^2\left[\frac{(p+r)-p_c}{(r_c-r)}-\frac{1}{4\pi}\left(\frac{1-(p+r)}{(p+r)-p_c}+\frac{r}{(r_c-r)}\right)\mathrm{i}(\omega \ln \omega)\right.$$
$$\left.+\frac{1}{8}\left(\frac{1-(p+r)}{(p+r)-p_c}+\frac{r}{r_c-r}\right)\omega+\cdots\right]. \tag{17}$$

(ii) For $p+r=p_c$

$$D(\omega)\simeq\frac{a^2}{4\sqrt{\pi}}(1+\mathrm{i})\frac{(-\omega \ln \omega)^{1/2}}{(r_c-r)^{1/2}}. \tag{18}$$

(iii) For $p+r<p_c$

$$D(\omega)\simeq a^2\left(\alpha\omega\mathrm{i}+\frac{32\{\alpha^4 p[1-(p+r)]/(p+r)\}}{F'(1+(1/4\alpha))+32(p_c-(p+r))\alpha^2}\,\omega^2\right) \tag{19}$$

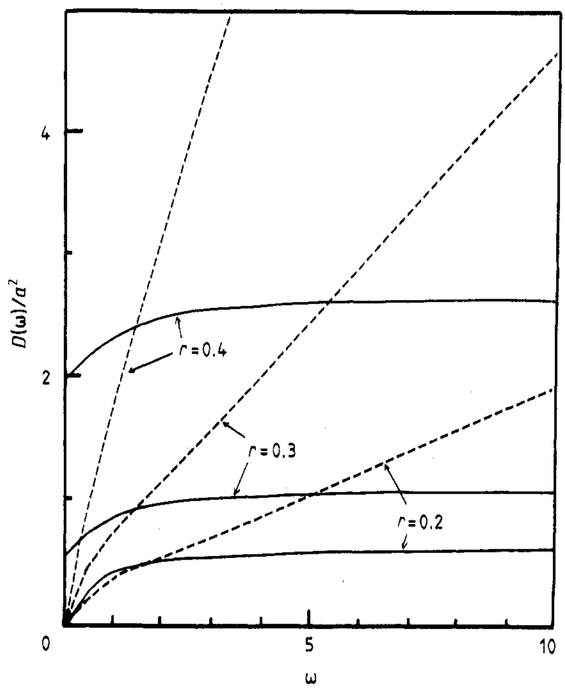

Figure 4. The same figure as figure 3 for $p = 0.3$ and various r satisfying $p + r \geq p_c$.

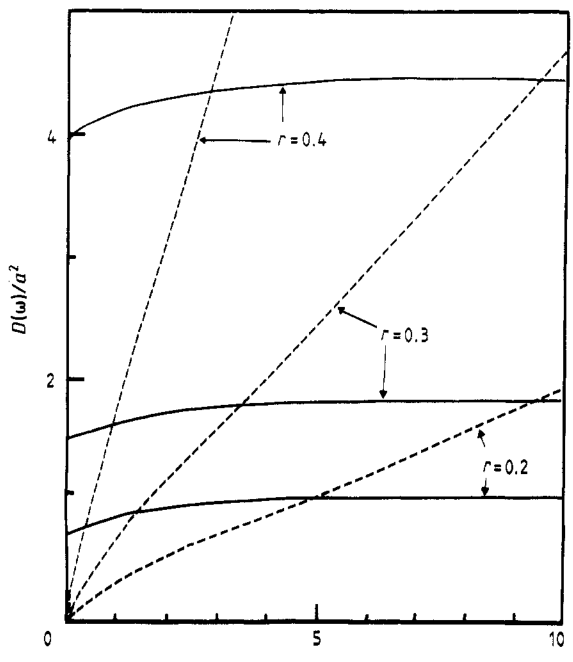

Figure 5. The same figure as figure 3 for $p = 0.5$ and various r satisfying $p + r > p_c$.

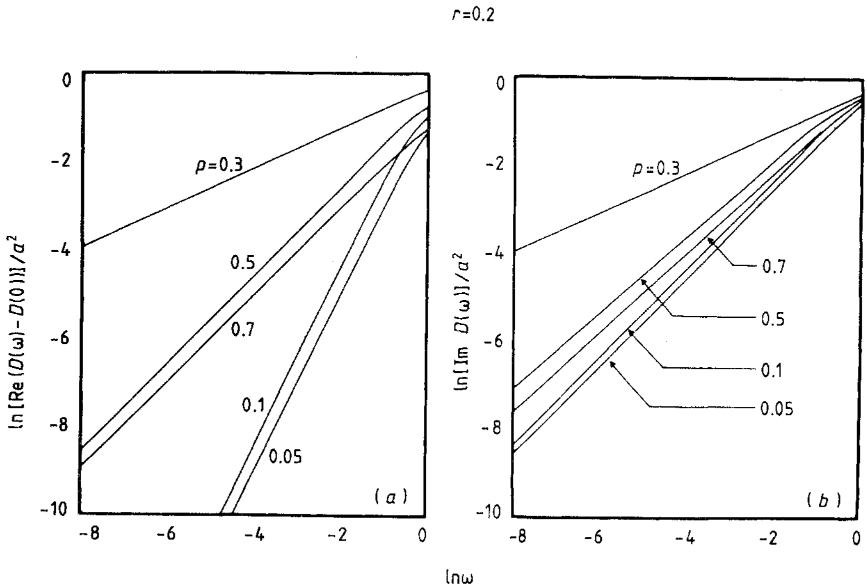

Figure 6. The log–log plot of the AC parts of the frequency-dependent diffusion constant near the static limit for the square lattice determined by the CMA method. The slope of the real part (a) is unity when $p+r>p_c$ and two when $p+r<p_c$; the imaginary part (b) is linear in ω when $p+r<p_c$ and behaves as $\omega \ln \omega$ when $p+r>p_c$. Both parts show logarithmic dependence on the frequency when $p+r=p_c$.

where α is a solution to

$$F\left(1+\frac{1}{4\alpha}\right)=8(p_c-(p+r))\alpha \tag{20}$$

and

$$F'(\xi)=\mathrm{d}F(\xi)/\mathrm{d}\xi. \tag{21}$$

Thus, the DC diffusion constant is zero when $p+r\leqslant p_c$ and is proportional to $[(p+r)-p_c]/(r_c-r)$ when $p+r>p_c$ (as shown in equations (12) and (17)). Here $r_c(\equiv\frac{1}{2})$ is the critical probability that superconducting bonds percolate. The coefficients of the AC part of the diffusion constant diverge as $p+r$ approaches p_c in equations (17) and (19) or as r approaches r_c in equations (17) and (18). In the pure ant limit, i.e. $r=0$, results obtained by Odagaki *et al* (1983) are recovered. The low-frequency behaviour is similar in both cases; however, as discussed above, the coefficients of the AC diffusion constant in ant-termite mixtures depend on both p and r. As r approaches r_c the coefficients of the leading terms in the AC part of the diffusion constant diverge, indicating enhancement in the dielectric constant, $\varepsilon(\omega)\propto \mathrm{Im}\, D(\omega)/\omega$. The static dielectric constant, $\varepsilon(0)$, is always infinity when $1>p+r\geqslant p_c$.

4.2. Simple cubic lattice

Next, we solve the CMA equation (13) for the simple cubic lattice. In the numerical calculation, we approximate g_{11} for the simple cubic lattice by

$$g_{11}=2[u+zw_c+(u(u+2zw_c))^{1/2}]^{-1}. \tag{22}$$

This approximation is equivalent to assuming a semi-elliptic density of states

$$n(x)=\frac{2}{\pi}(1-x^2)^{1/2}. \tag{23}$$

Substituting the value of g_{11} from equation (22) in equation (13), and solving for w_c self-consistently for various values of p and r, we obtain the dynamic diffusion constant. Figures 7 and 8 show frequency dependence of the real (full curve) and imaginary (broken curve) parts of the diffusion constant in three dimensions ($z=6$) for several values of r, for $p=0.1$ ($p+r\lesssim p_c$) and for $p=0.3$ ($p+r>p_c$), where $p_c=\frac{1}{3}$.

As in the two-dimensional case, examining the behaviour of $F(\xi)$ around $\xi=1$, we can easily determine the low-frequency behaviour of the AC diffusion constant for general lattices in three dimensions.

(i) For $p+r>p_c$ ($\equiv\frac{1}{3}$)

$$D(\omega)=a^2\left[\frac{1}{2}\frac{(p+r)-p_c}{(r_c-r)}+\frac{1}{6}\left(2\frac{\frac{2}{3}-(p+r)}{(p+r)-p_c}+\frac{r_c+r}{r_c-r}\right)\mathrm{i}\omega\right.$$
$$\left.+\frac{\sqrt{3}}{18}\left(2\frac{\frac{2}{3}-(p+r)}{(p+r)-p_c}+\frac{r_c+r}{r_c-r}\right)\left(\frac{r_c-r}{p+r-p_c}\right)^{1/2}(1-\mathrm{i})\omega^{3/2}\right]. \tag{24}$$

(ii) For $p+r=p_c$

$$D(\omega)=\frac{a^2}{6}(1+\mathrm{i})\frac{\omega^{1/2}}{(r_c-r)^{1/2}}. \tag{25}$$

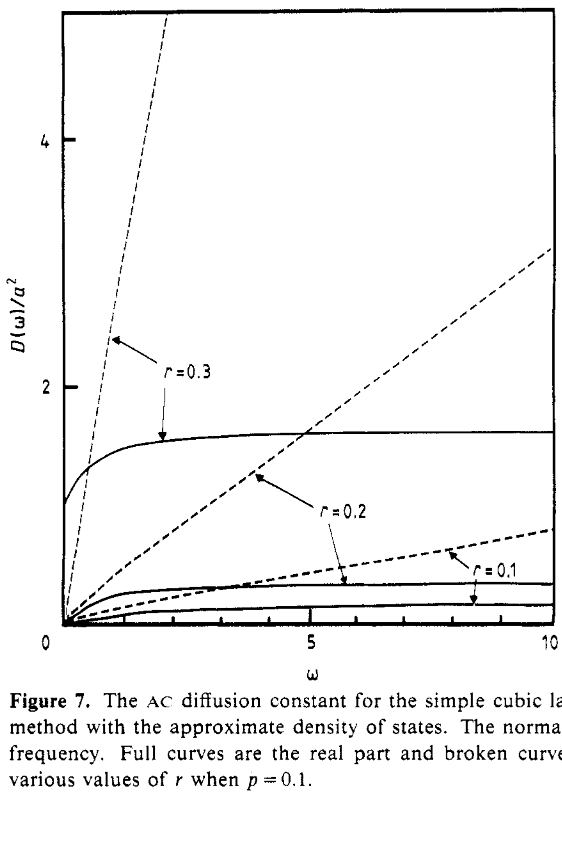

Figure 7. The AC diffusion constant for the simple cubic lattice determined by the CMA method with the approximate density of states. The normal jump rate w_p is the scale of frequency. Full curves are the real part and broken curves are the imaginary part for various values of r when $p=0.1$.

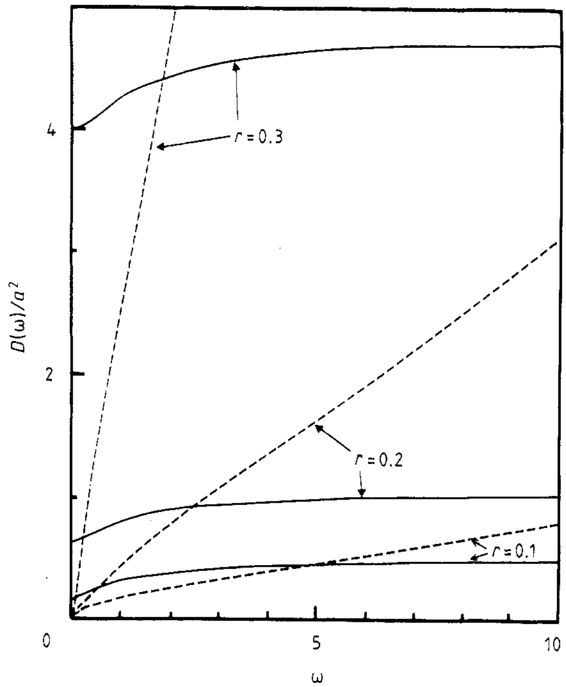

Figure 8. The same figure as figure 7 for $p = 0.3$ and various values of r satisfying $p + r > p_c$.

(iii) For $p+r<p_c$

$$D(\omega)=a^2\left(\alpha \mathrm{i}\omega+\frac{108\alpha^4 p[1-(p+r)]/(p+r)}{108[p_c-(p+r)]\alpha^2+F'(1+(1/6\alpha))}\,\omega^2\right) \tag{26}$$

where α is the solution of

$$F\left(1+\frac{1}{6\alpha}\right)=18[p_c-(p+r)]\alpha. \tag{27}$$

Here, $F(\xi)$ is defined by equation (8) and $F'(\xi)=\mathrm{d}F(\xi)/\mathrm{d}\xi$. Consequently, the DC diffusion constant is zero when $p+r\leqslant p_c$ and is proportional to $[(p+r)-p_c]/(r_c-r)$ when $p+r>p_c$. Here $r_c=(2/z)(\equiv\frac{1}{3})$. As in two dimensions, the coefficients of the AC part of the diffusion constant also diverge as $p+r$ approaches p_c or r approaches r_c. In the pure ant limit, i.e. $r=0$, the results obtained by Odagaki and Lax (1981) are recovered. In the pure termite limit, i.e. $p+r=1$, the results obtained by Odagaki (1986) are recovered. As r approaches r_c, the coefficients of the leading terms in the AC part of the diffusion constant diverge, indicating the enhancement in the dielectric constant. The static dielectric constant is found to be divergent at $p+r=p_c$.

5. Summary

In this paper we have applied the coherent medium approximation to a hopping conduction with ternary jump rates. For the sake of mathematical simplicity we specialised the distribution to ant-termite mixtures ($w_r\to\infty$, $w_q\to 0$), a model physical system. This system is also attractive in view of ceramic superconductors which may be a mixture of normal, superconducting and semiconducting phases.

The static diffusion constant diverges as the probability of a given bond being superconducting reaches the percolation limit, i.e. $r \to r_c - 0^+$ (figure 1). The phase diagram on a triangular region represented by (p, q, r) is shown in figures 2(a) and (b) for two and three dimensions, respectively. In the shaded region none of the conducting phases percolates, i.e. it is the region of mixed phase normal conductor and superconductor. Pure superconducting, normal and insulating regions are shown in different sections of the triangle.

The low-frequency behaviour in the ant-termite mixture is found to be similar to that in the ant model (Odagaki *et al* 1983). The coefficients of the leading terms diverge as $p+r \to p_c$ $(\equiv(2/z))$ or $r \to r_c$ $(\equiv(2/z))$ indicating the divergence in the dielectric constant. The static dielectric constant always diverges for $p_c < p+r < 1$ in two dimensions and for $p+r=p_c$ in three dimensions.

The AC diffusion constant is plotted in figures 3-6 for the square lattice, and in figures 7 and 8 for the simple cubic lattice. Note that as r approaches r_c, the imaginary part of the AC diffusion constant diverges, implying divergence in the dielectric constant.

To summarise, the transport properties become critical at $p+r=p_c$ and at $r=r_c$. Critical exponents are readily obtained from (17)-(19) and (25)-(27). A direct comparison of the present results is possible only for the DC diffusion constant in the pure ant and termite limits (Odagaki *et al* 1983, Odagaki 1986). Although some exponents (for example, u, an exponent characterising the divergence of the diffusion constant in the termite model) agree with computer simulation (Bunde *et al* 1985); other exponents determined by the CMA generally disagree with estimations by computer simulation or renormalisation group treatments (Bergman and Imry 1977, Kirkpatrick 1973). However, we expect very rich critical behaviours to be seen in the 'ant-termite' mixtures.

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