Conduction in anisotropic disordered systems: Effective-medium theory

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Some aspects of conduction in disordered systems are studied by investigating the properties of disordered classical resistance networks. The effective-medium theory of Kirkpatrick is extended to include networks with an anisotropic distribution of conductance values, and then applied to some model systems. In particular, it is shown that the "observable" anisotropy of the conductivity is limited by an expression which only depends on the concentration of interruptions along the highly conducting direction.

I. INTRODUCTION

Disordered classical resistance networks have recently become an important model for the investigation of transport phenomena in disordered physical systems.[1–17] In particular, the network model for variable-range hopping conduction in amorphous materials has been studied extensively.[1,3,4,7–12,16,17] Miller and Abrahams[1] have shown that the thermally activated hopping between two localized states i and j can be described by a conductance σ_{ij}, so that the resistance of a macroscopic sample is equivalent to that of the corresponding random network of conductances σ_{ij}. Ziman,[2] Eggarter and Cohen,[18] and others have suggested a connection between the conductivity of disordered systems and percolation problems. Mott's[19] famous $\ln\sigma \sim T^{-1/4}$ law for the variable-range hopping conductivity σ at low temperatures T has subsequently been derived on the basis of percolation arguments.[3,4,8,9] The study of nearest-neighbor hopping models[7,8,9,13,15] recently lead to an alternative explanation of the $T^{-1/4}$ behavior often observed in disordered materials.

Disordered resistance networks have also been studied from a more general point of view.[6–9,14,15] Analog simulations and numerical calculations on large networks have been performed[6–9,15] in order to study conduction in bond- and site-percolation models. Kirkpatrick[7] also investigated some general binary-disorder networks, and Stinchcombe[14] gave an exact solution to the conductivity of the bond-percolation model for tree-lattices.

A simple, but very powerful, theoretical description of conduction in disordered resistance networks is provided by a so-called *effective-medium theory*.[7–9,13,20–22] This theory, originally formulated to describe the conductivity of binary mixtures[20,21] and recently generalized to include the Hall effect,[22] has been extended and adapted to treat general disordered resistance networks.[7–9] Under a wide range of conditions, the effective-medium theory gives very accurate results for the conductivity in such network systems[7–9,13,14] It has been successfully applied to percolation models (outside the threshold region) and to general binary-disorder networks,[7–9,14] and also to nearest-neighbor hopping models at not too low temperatures.[7–9,13]

In the following, the effective-medium theory of Kirkpatrick[7–9] will be extended to include the description of conduction in *anisotropic* disordered networks (i.e., networks with an anisotropic distribution of conductance values).

Such an extension is not trivial and leads to interesting predictions concerning the conductivity and its anisotropy in anisotropic disordered systems. With the recent extensive studies on various quasi-one-dimensional conductors,[23] the interest in anisotropic disordered materials has become very great. There exist a few measurements of the anisotropy of the conductivity in such materials[23–27]; the theoretical explanations of the results are, however, far from clear. The effective-medium theory for anisotropic random networks can at least help to clarify some general aspects of conduction in anisotropic systems, even if their simulation by a resistance network is a rather crude approximation.

II. EFFECTIVE-MEDIUM THEORY

We shall be concerned with disordered network models of the following general type. To each bond (ij) of an infinite regular lattice we assign a conductance σ_{ij}. These σ_{ij} values are randomly and independently distributed according to a probability density $\rho(\sigma)$. We shall restrict ourselves to two-dimensional square and three-dimensional simple cubic lattices, and define an *anisotropic distribution* of conductance values by introducing *different probability densities* $\rho_\nu(\sigma)$ for the conductances aligned along the different lattice directions ν ($\nu = 1, \ldots, d$; where d is the dimensionality of the lattice).

In the effective-medium theory,[7–9,13] the average effects of the random conductances in such a disordered network will be represented by an *anisotropic "effective network"* in which all the conductances in ν direction have the same value $\bar{\sigma}_\nu$ (ν

$= 1, \ldots, d$). These effective conductivities $\bar{\sigma}_\nu$ are self-consistently determined by the requirement that the fluctuating "local fields" in the random network should average to zero. As in the isotropic case we can use two procedures to express this qualitative requirement in mathematical terms. The first method is an adaption of the classical theory of mixtures to random resistance networks,[7-9,13] and the second[8,9] is analogous to the coherent potential approximation[28,29] in the theory of electrons in alloys. Both procedures (the anisotropic version of the first method is sketched in Appendix A) lead to the following set of integral equations for the determination of the effective conductivities $\bar{\sigma}_1, \ldots, \bar{\sigma}_d$:

$$\int d\sigma \rho_\nu(\sigma) \frac{\bar{\sigma}_\nu - \sigma}{\sigma + S_\nu(\bar{\sigma}_1, \ldots, \bar{\sigma}_d)} = 0, \quad \nu = 1, \ldots, d, \tag{1}$$

where

$$S_\nu = 1/R_\nu - \bar{\sigma}_\nu \tag{2}$$

and $R_\nu(\bar{\sigma}_1, \ldots, \bar{\sigma}_d)$ is the total resistance between two neighboring nodes in ν direction in the infinite effective lattice.

In the isotropic case, where all the $\bar{\sigma}_\nu$ are equal, the problem of calculating R (or S) is easily solved by using symmetry arguments.[7-9,13] In the anisotropic case, however, the calculation of the R_ν's is much more complicated. Exact analytical expressions for R_ν can be obtained, for example, by Fourier transforming the appropriate Kirchhoff equations or by using a generalization of the method of Jelitto and Borm.[30-32] In Appendix B we describe the Fourier-transformation method from which we obtain

$$R_\nu = \frac{1}{\pi^d} \int_0^\pi dk_1 \ldots \int_0^\pi dk_d \, (1 - \cos k_\nu) \Big/ \sum_{l=1}^{d} \bar{\sigma}_l (1 - \cos k_l), \quad \nu = 1, \ldots, d. \tag{3}$$

For $d = 2$ (*two-dimensional square network*) the integrations in Eq. (3) can be carried out analytically, and we obtain

$$R_1 = \frac{2}{\pi \bar{\sigma}_1} \arctan(\bar{\sigma}_1/\bar{\sigma}_2)^{1/2} \quad \text{(and cyclic)} \tag{4}$$

or

$$S_1 = \bar{\sigma}_1 \frac{\arctan(\bar{\sigma}_2/\bar{\sigma}_1)^{1/2}}{\arctan(\bar{\sigma}_1/\bar{\sigma}_2)^{1/2}} \quad \text{(and cyclic)} . \tag{5}$$

For $d = 3$ (*three-dimensional simple cubic network*) the expression (3) for R_ν can be reduced to[33]

$$R_\nu = \frac{1}{\pi^2} \left(\frac{\bar{\sigma}_\nu}{\bar{\sigma}_1 \bar{\sigma}_2 \bar{\sigma}_3} \right)^{1/2} \int_0^\pi dx \, k_\nu K(k_\nu) , \tag{6}$$

where

$$k_1 = \left(\frac{4\bar{\sigma}_2 \bar{\sigma}_3}{[\bar{\sigma}_1(1 - \cos x) + \bar{\sigma}_2 + \bar{\sigma}_3]^2 - (\bar{\sigma}_2 - \bar{\sigma}_3)^2} \right)^{1/2} \tag{7}$$

and k_2, k_3 are obtained from Eq. (7) by cyclic permutation of the indices 1, 2, 3;

$$K(k) = \int_0^{\pi/2} d\theta \, (1 - k^2 \sin^2\theta)^{-1/2} \tag{8}$$

is the complete elliptic integral of the first kind. In general, formula (6) can be evaluated only numerically.

By using certain results obtained from calculations on a continuum with an anisotropic conductivity tensor we can, however, construct a simple approximate formula for the S_ν's corresponding to a three-dimensional simple cubic network:

$$S_1 \approx \bar{\sigma}_1 \cdot \frac{\arctan[\bar{\sigma}_1^{-1}(\bar{\sigma}_1\bar{\sigma}_2 + \bar{\sigma}_2\bar{\sigma}_3 + \bar{\sigma}_3\bar{\sigma}_1)]^{1/2}}{\arctan[\bar{\sigma}_1(\bar{\sigma}_1\bar{\sigma}_2 + \bar{\sigma}_2\bar{\sigma}_3 + \bar{\sigma}_3\bar{\sigma}_1)]^{-1/2}} \quad \text{(and cyclic)}. \tag{9}$$

Equation (9) reduces to the exact result (5) for two-dimensional square networks ($\bar{\sigma}_2 = 0$ or $\bar{\sigma}_3 = 0$) and gives the correct answer ($S = 2\bar{\sigma}$) for the isotropic case ($\bar{\sigma}_1 = \bar{\sigma}_2 = \bar{\sigma}_3 = \bar{\sigma}$).

To conclude this section, we shall discuss the important special case of *uniaxial anisotropy* in some detail. We set

$$\bar{\sigma}_1 = \bar{\sigma}_\parallel \text{ and } \bar{\sigma}_2 = \bar{\sigma}_3 = \bar{\sigma}_\perp \tag{10}$$

and obtain, from Eqs. (6) and (7),

$$R_\parallel = \frac{2}{\pi^2} \int_0^\pi dx \, \frac{1 - \cos x}{2\bar{\sigma}_\perp + \bar{\sigma}_\parallel(1 - \cos x)} \times K\left(\frac{2\bar{\sigma}_\perp}{2\bar{\sigma}_\perp + \bar{\sigma}_\parallel(1 - \cos x)} \right) . \tag{11}$$

$R_\perp$ is related to $R_\parallel$ by

$$R_\perp = (1/2\bar{\sigma}_\perp)(1 - \bar{\sigma}_\parallel R_\parallel) , \tag{12}$$

as is easily seen from the original expressions (3). From Eq. (12) we immediately obtain the isotropic result

$$R_\parallel = R_\perp = 1/3\bar{\sigma}_\parallel \quad \text{for } \bar{\sigma}_\perp = \bar{\sigma}_\parallel . \tag{13}$$

For small and large "effective anisotropies" U,

$$U \equiv \bar{\sigma}_\perp/\bar{\sigma}_\parallel , \tag{14}$$

we can derive the following exact asymptotic expansions:

$$R_\parallel \approx (1/\bar{\sigma}_\parallel)(1 - 0.9581\sqrt{U}) \quad \text{for } U \to 0 , \tag{15}$$

$$R_\parallel \approx \frac{1}{2\pi\bar{\sigma}_\parallel} \left(\frac{\ln U}{U} + 2.3557 \frac{1}{U} \right) \quad \text{for } U \to \infty . \tag{16}$$

Finally, we observe that, for uniaxial anisotropy, the "continuum-approximation" (9) leads to

$$R_\parallel = (2/\pi\bar{\sigma}_\parallel) \arctan[(2U + U^2)^{-1/2}] . \tag{17}$$

Its small U behavior

$$R_{\parallel} \approx (1/\bar{\sigma}_{\parallel})[1-(2\sqrt{2}/\pi)\sqrt{U}] \tag{18}$$

is qualitatively correct, and the coefficient of $\sqrt{U}$ differs from the exact value by only about 5%. The values of $R_{\parallel}$, obtained from the exact formula (11) and from the "continuum approximation" (17), respectively, coincide at $U=1$ and differ at most by 0.02 over the whole range of $U(0 \leq U < \infty)$. The large-U behavior of Eq. (17), $R_{\parallel} \sim 1/U$, is qualitatively different from the exact result (16), so that the relative error becomes very large in this limit. The effects of this discrepancy, however, are negligible in most applications.

III. APPLICATIONS

In the following we shall apply our effective-medium theory of Sec. II to some specific examples of anisotropic disordered network models. First, we consider the two-dimensional anisotropic bond-percolation model defined by

$$\rho_{\nu}(\sigma) = p_{\nu}\,\delta(\sigma - G_{\nu}) + (1-p_{\nu})\,\delta(\sigma), \quad \nu = 1, 2, \tag{19}$$

i.e., the conductances in ν direction assume the values G_{ν} and zero with probabilities p_{ν} and $1-p_{\nu}$, respectively. The results for the effective conductivities $\bar{\sigma}_1$, $\bar{\sigma}_2$ [as determined by Eqs. (1) and (5)] and the effective anisotropy $Q=\bar{\sigma}_1/\bar{\sigma}_2$ can be represented as follows:

$$\begin{aligned} &\bar{\sigma}_1 = G_1(p_1 - x)/(1-x), \\ &\bar{\sigma}_2 = G_2[p_2 - (1-x)]/x, \quad Q = \tan^2(\tfrac{1}{2}\pi x), \end{aligned} \tag{20}$$

so that x is the solution of

$$\tan^2(\tfrac{1}{2}\pi x) = \frac{G_1}{G_2}\,\frac{x(p_1-x)}{(1-x)[p_2-(1-x)]}\,. \tag{21}$$

The conductivities $\bar{\sigma}_1$ and $\bar{\sigma}_2$ both vanish for $p_1+p_2 \leq 1$ and are nonzero in the complementary region. Figure 1 shows the results for $p_1=p_2=p$, $G_1=10$, and $G_2=1$. The agreement with numerical calculations on networks with 20×20 nodes is remarkably good, except in a critical region near $p=\frac{1}{2}$.

For fixed values of p_1 and p_2 (with $p_1+p_2>1$), the anisotropy Q increases monotonically with increasing G_1/G_2, but it is bounded by $\tan^2(\frac{1}{2}\pi p_1)$ as $G_1/G_2 \to \infty$. This boundedness of the anisotropy Q has important consequences for the interrupted strand model which has been introduced to explain certain features of quasi-one-dimensional (i.e., highly anisotropic) conductors.[23] The simplest network-model of an interrupted strand system is represented by the following special case of a three-dimensional bond-percolation model:

$$\begin{aligned} &\rho_1(\sigma) = (1-p)\,\delta(\sigma-\sigma_{\parallel}) + p\,\delta(\sigma), \\ &\rho_2(\sigma) = \rho_3(\sigma) = \delta(\sigma-\sigma_{\perp}). \end{aligned} \tag{22}$$

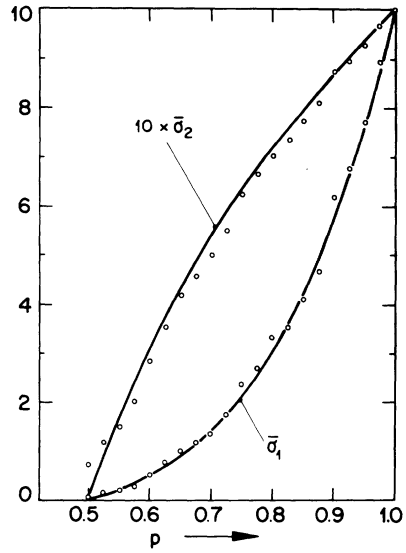

FIG. 1. Effective conductivities $\bar{\sigma}_1$, $\bar{\sigma}_2$ for the anisotropic bond-percolation model defined by Eq. (19), with $p_1=p_2=p$, $G_1=10$, $G_2=1$. The circles refer to numerical computations on networks with 20×20 nodes.

We assume a high uniaxial intrinsic anisotropy ($\sigma_{\parallel}/\sigma_{\perp} \gg 1$) and a small concentration of interruptions (missing conductances) in the highly conducting chains ($p \ll 1$). In these limits we obtain simple, but very instructive, relations between the intrinsic anisotropy $\sigma_{\parallel}/\sigma_{\perp}$ and the "observable" anisotropy $Q=\bar{\sigma}_{\parallel}/\bar{\sigma}_{\perp}$: If $\sigma_{\parallel}/\sigma_{\perp} \gg 1/p^2$, Q is approximately equal to

$$Q_{\infty} = 0.918/p^2\,, \tag{23}$$

i.e., Q is given by the defect concentration p alone, and is proportional to the square of the mean length $1/p$ of the strand segments. In the opposite limit $\sigma_{\parallel}/\sigma_{\perp} \ll 1/p^2$, Q is practically equal to the intrinsic anisotropy $\sigma_{\parallel}/\sigma_{\perp}$. As an illustration, Fig. 2 shows a plot of Q vs $\sigma_{\parallel}/\sigma_{\perp}$ for $p=0.002$.

These results offer a simple explanation to the anisotropy measurements of Zeller[23,25] on the quasi-one-dimensional conductor $K_2Pt(CN)_4Br_{0.3}\cdot 3(H_2O)$. In this compound, the anisotropy is strongly sample dependent at high temperatures where it has its maximum value. It decreases with decreasing temperature and becomes practically sample independent (intrinsic) below about 100 K. Within our simple model, these results can thus be explained as a consequence of the defect-limited observability of large intrinsic conductivity anisotropies.

Finally, we consider a two-dimensional network model for anisotropic thermally activated nearest-neighbor hopping. We assume the individual conductances $\sigma_{ij}^{(\nu)}$ ($\nu=1, 2$) to have the form

$$\sigma_{ij}^{(\nu)} = \sigma_0^{(\nu)}\, e^{-\beta E_{ij}^{(\nu)}}\,, \quad \beta = 1/k_B T\,, \tag{24}$$

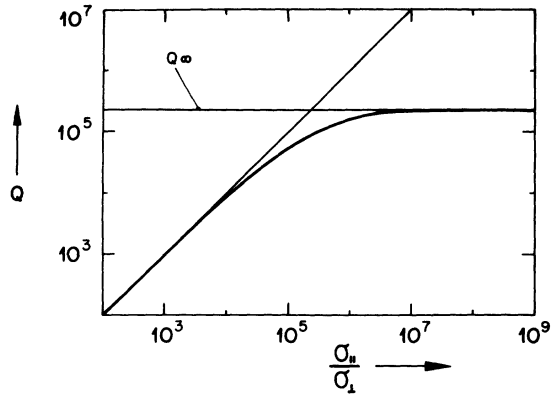

FIG. 2. "Observable" anisotropy $Q=\bar{\sigma}_\parallel/\bar{\sigma}_\perp$ vs intrinsic anisotropy $\sigma_\parallel/\sigma_\perp$ for the model defined by Eqs. (22), with $p=0.002$.

the activation energies $E_{ij}^{(\nu)}$ being distributed according to the probability density $\rho_\nu(E)$. For the simple case where the activation energies in both directions have the same uniform distribution

$$\rho_1(E)=\rho_2(E)=\begin{cases}1/E_m, & 0\le E\le E_m\\ 0, & \text{otherwise,}\end{cases} \tag{25}$$

and only the prefactor $\sigma_0^{(\nu)}$ is anisotropic, the results for the effective conductivities can be represented as follows:

$$\bar{\sigma}_1\cdot\bar{\sigma}_2=\sigma_0^{(1)}\sigma_0^{(2)}e^{-\beta E_m},\quad Q=\bar{\sigma}_1/\bar{\sigma}_2=\tan^2(\tfrac{1}{2}\pi x), \tag{26}$$

with x being the solution of

$$\tan^2(\tfrac{1}{2}\pi x)=\frac{\sigma_0^{(1)}}{\sigma_0^{(2)}}\left(\frac{x\sinh[\frac{1}{2}\beta E_m(1-x)]}{(1-x)\sinh(\frac{1}{2}\beta E_m x)}\right)^2. \tag{27}$$

In Fig. 3 we have plotted $\ln\bar{\sigma}_1$ and $\ln\bar{\sigma}_2$ as a function of the inverse temperature β for $\sigma_0^{(1)}=10^6$ and $\sigma_0^{(2)}=1$. The anisotropy Q is equal to $\sigma_0^{(1)}/\sigma_0^{(2)}$ at $\beta=0$ and tends to 1 as $\beta\to\infty$. In the case of two different uniform distributions $\rho_1(E)$ and $\rho_2(E)$ of the type (25), Q is still equal to $\sigma_0^{(1)}/\sigma_0^{(2)}$ at $\beta=0$, but tends to $\tan^2[\frac{1}{2}\pi(1+E_m^{(1)}/E_m^{(2)})^{-1}]$ as $\beta\to\infty$. As in the isotropic case,[13] the predictions of the effective-medium theory for such hopping models are quantitatively only reliable at not too large β values.

In the isotropic analog of our two-dimensional hopping model, the effective-medium result for the $\ln\bar{\sigma}$-vs-β curve is a straight line with slope $(-\frac{1}{2})$. Figure 3 therefore gives us some information about the influence of anisotropy on the shape of the conductivity curves. Conductivity measurements on triethylammonium-tetracyanoquimodimethan[26,27] [TEA(TCNQ)$_2$] seem to indicate such anisotropy effects.

The above simple examples demonstrate the usefulness of the effective-medium theory in dealing with quite general aspects of the conductivity and its anisotropy in disordered systems.

The theory gives an accurate description of a wide class of anisotropic mixture problems (except in critical regions). For a quantitative analysis of hopping models, however, it ought to be supplemented by an extension of the critical-path analysis[3,7–9,13] to anisotropic systems.

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APPENDIX A: DETERMINATION OF THE EFFECTIVE CONDUCTIVITIES $\bar{\sigma}_\nu$

In the effective network we change a single conductance, oriented along the external electric field (direction ν), back to its true value $\sigma_{ij}^{(\nu)}$, leaving all the other conductances equal to their respective mean values $\bar{\sigma}_1,\ldots,\bar{\sigma}_d$. Then we calculate the difference

$$\Delta V_\nu(\bar{\sigma}_{ij}^{(\nu)};\bar{\sigma}_1,\ldots,\bar{\sigma}_d)=V_{ij}^{(\nu)}-\bar{V}_\nu, \tag{A1}$$

where $V_{ij}^{(\nu)}$ is the voltage across $\sigma_{ij}^{(\nu)}$ and $\bar{V}_\nu$ is the voltage across a conductance $\bar{\sigma}_\nu$ that is far away from $\sigma_{ij}^{(\nu)}$. The effective conductivities $\bar{\sigma}_1,\ldots,\bar{\sigma}_d$ are determined by the requirement that the ΔV_ν's should average to zero.

$$\int d\sigma\,\rho_\nu(\sigma)\,\Delta V_\nu(\sigma;\bar{\sigma}_1,\ldots,\bar{\sigma}_d)=0,\quad \nu=1,\ldots,d. \tag{A2}$$

The quantity ΔV_ν is evaluated by using the same equivalent circuit diagram as in the isotropic case [Fig. 2(a) of Ref. 13]; this leads to

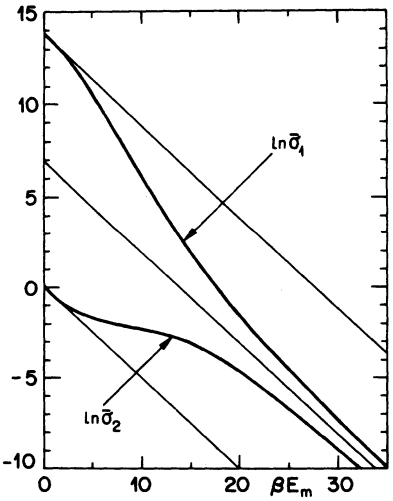

FIG. 3. Solutions $\bar{\sigma}_1$ and $\bar{\sigma}_2$ of Eqs. (26) and (27) as a function of βE_m for $\sigma_0^{(1)}=10^6$, $\sigma_0^{(2)}=1$.

$$\Delta V_\nu(\sigma;\bar{\sigma}_1,\ldots,\bar{\sigma}_d)=\bar{V}_\sigma\frac{\bar{\sigma}_\nu-\sigma}{\sigma+S_\nu(\bar{\sigma}_1,\ldots,\bar{\sigma}_d)}, \tag{A3}$$

where $S_\nu=1/R_\nu-\sigma_\nu$ and R_ν is the total resistance between two neighboring nodes in ν direction in the (infinite) effective lattice. R_ν depends on all effective conductances $\bar{\sigma}_1,\ldots,\bar{\sigma}_d$ and is calculated in Appendix B. Insertion of Eq. (A3) into (A2) finally leads to the system of integral equations that determines the $\bar{\sigma}_\nu$ values [Eq. (1)].

APPENDIX B: DERIVATION OF AN EXACT EXPRESSION FOR R_ν

We characterize the nodes of our effective network (Sec. II) by lattice vectors $\vec{n}=(n_1,\ldots,n_d)$, with n_ν integers. From outside we impose a current J which flows into the network at site $\vec{n}^+$ and out at site $\vec{n}^-$. The node voltages are denoted by $V_{\vec{n}}$, and the total resistance $R_{\vec{n}^+\vec{n}^-}$ between the sites $\vec{n}^+$ and $\vec{n}^-$ is then given by

$$R_{\vec{n}^+\vec{n}^-}=(V_{\vec{n}^+}-V_{\vec{n}^-})/J. \tag{B1}$$

To calculate the voltages $V_{\vec{n}}$ we consider the Kirchhoff equations corresponding to our problem:

$$\sum_{\vec{\delta}}\bar{\sigma}_{\nu(\vec{\delta})}(V_{\vec{n}}-V_{\vec{n}+\vec{\delta}})=J(\delta_{\vec{n},\vec{n}^+}-\delta_{\vec{n},\vec{n}^-}) \tag{B2}$$

for all lattice vectors $\vec{n}$ of our infinite effective lattice. The summation is over all nearest neighbors $\vec{n}+\vec{\delta}$ of site $\vec{n}$, and $\nu(\vec{\delta})$ denotes the lattice direction to which $\vec{\delta}$ is parallel.

If we introduce the Fourier transform g of V,

$$g(\vec{k})=\sum_{\vec{n}}e^{-i\vec{n}\vec{k}}V_{\vec{n}}, \tag{B3}$$

it follows from Eq. (B2) that

$$g(\vec{k})=J(e^{-i\vec{n}^+\vec{k}}-e^{-i\vec{n}^-\vec{k}})\Big/\sum_{\vec{\delta}}\bar{\sigma}_{\nu(\vec{\delta})}(1-e^{i\vec{k}\vec{\delta}}). \tag{B4}$$

The voltages $V_{\vec{n}}$ are then given by

$$V_{\vec{n}}=\frac{1}{(2\pi)^d}\int d\vec{k}\,e^{i\vec{n}\vec{k}}g(\vec{k}). \tag{B5}$$

From Eqs. (B1), (B5), and (B4) we can now calculate the resistance $R_{\vec{n}^+\vec{n}^-}$. If we specialize to the case where $\vec{n}^+$ and $\vec{n}^-$ are nearest neighbors in the ν direction, we obtain the desired exact expression for R_ν,

$$R_\nu=\frac{1}{\pi^d}\int_0^\pi dk_1\cdots\int_0^\pi dk_d\,(1-\cos k_\nu)\Big/\sum_l^d\bar{\sigma}_l(1-\cos k_l). \tag{B6}$$

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