Hall and Seebeck coefficients in a randomly disordered system

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Abstract. Formulae are obtained for calculating the effective Hall coefficient in a weak magnetic field and for the effective Seebeck coefficient neglecting the secondary thermo-current. A model of an infinite cluster on both sides of the percolation threshold with 'hot' and 'active' points is constructed. It is shown that the main contribution to the kinetic coefficient in the critical interval is given by a few separate small regions called 'active' or 'hot' points. For a given change of concentration in the critical interval, the number of these points correspondingly changes; near the percolation threshold there remains one active point in the volume ρ^3, where ρ is the correlation radius.

1. Introduction

This paper is a sequel to and a further development of Skal (1981) and Skal and Andreev (1982a, b). In this paper we try to answer the question of what information on the topology of a disordered system can be inferred from an experiment involving measurements of conductivity and Hall and Seebeck coefficients in a two-component random mixture. It seems reasonable to apply the percolation method to investigate this problem. The percolation problem is mainly of interest as a simple example of a second-order phase transition with associated critical phenomena.

The Hall effect and thermopower have been used intensively to investigate the metal–insulator and metal–superconductor transitions in a variety of disordered systems (see the pioneering theoretical work of Juretschke *et al* (1956) and Odolevskii (1951)). An effective-medium theory (EMT) and a simulation approach have been used to discuss the properties of the Hall effect and thermopower in conductors with macroscopic disorder (Straley 1980a, b, Orton and Powell 1980, Skal 1981, Skal and Andreev 1982a, b, Ottavi *et al* 1978). Reported in the literature are attempts at 'frontal attacks' on the solution of the complete conductivity tensor problem (Webman *et al* 1977). However, the results obtained cannot be considered as satisfactory because of poor convergence of the computation procedure near the percolation threshold.

In the present paper we solved a more particular problem, where the Hall coefficient is calculated, provided that variations in the current distributions occurring under the

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action of the magnetic field are completely neglected. Analogously, one can treat thermoelectric phenomena by neglecting variations in the temperature distribution which occur under the actions of thermoelectromotive force. Also a new model infinite cluster on both sides of the percolation threshold is introduced.

2. Calculation of the Hall effect in a disordered system

Consider a disordered system whose discontinuity scale is larger than the free path of charge carriers and whose free path is larger than the cyclotron radius. These conditions allow the introduction of local conductivity $\sigma(r)$ and Hall coefficient $R(r)$. The equation for the electric current in such a system can be written as

$$j(r) = \sigma(r)\nabla\varphi(r) + \sigma(r)R(r)[H \times j(r)] \tag{1}$$

where H is magnetic field and $-\varphi(r)$ is the electric potential. This equation is not valid for a very weak magnetic field, because cyclotron radius $R_c \sim 1/H$ and at low H it becomes larger than the free path and discontinuity length.

Consider a cubic sample with side lengths $L_{x_1}, L_{x_2}, L_{x_3}$ ($x = x_1, y = x_2, z = x_3$) with an applied electric field intensity E along the x_m axis. Denote by $-\varphi_m(r)$ the electrical potential caused by the electric field applied along x_m. In a weak magnetic field H one can express the current as

$$j_m^n(r) = \sigma(r)\nabla_n\varphi_m(r) + \sigma^2(r)R(r)[H \times \nabla\varphi_m(r)]_n. \tag{2}$$

The boundary conditions for the potential $\varphi_m(r)$ are $\varphi_m(r)|_{r\in\Gamma} = Ex_m$ where Γ is the boundary surface of the sample, $j_m^n(r)$ is the projection of $j_m(r)$ on the x_n axis.

With those conditions, if an electric field is applied along 'x' and Hall voltage is measured along 'y' the effective Hall coefficient can be expressed as

$$R^{eff}(p) = \int_V j_x^y(r)\, dV / [\sigma_x^{eff}(p)\sigma_y^{eff}(p)EHV] \tag{3}$$

where V is the volume of the sample and $\sigma_x^{eff}(p)$, $\sigma_y^{eff}(p)$ is the effective conductivity along axis 'x', 'y'.

Expanding the solution as a series up to the quadratic field term yields

$$j(r) = j_0(r) + j_1(r) + O(H^2) \tag{4a}$$

$$\varphi_m(r) = \varphi_{0,m}(r) + \varphi_{1,m}(r) + O(H^2). \tag{4b}$$

Equation (2) can now be written as

$$j_{0,m}^n(r) = \sigma(r)\nabla_n\varphi_{0,m}(r) \tag{5a}$$

$$j_{1,m}^n(r) = \sigma(r)\nabla_n\varphi_{1,m}(r) + \sigma^2(r)R(r)[H \times \nabla\varphi_{0,m}]_n \tag{5b}$$

with the boundary conditions

$$\varphi_{0,m}(r)|_{r\in\Gamma} = Ex_m \qquad \varphi_{1,m}(r)|_{r\in\Gamma} = 0. \tag{6}$$

To find the solution of equations (5) we introduce the Green function $G(r, r')$ such that

$$\nabla_n[\sigma(r)\nabla_n G(r, r')] = \delta^3(r - r')$$

$$G(r, r') = G(r', r) \qquad G(r, r')|_{r\in\Gamma} = 0. \tag{7}$$

Since $\nabla_n j_{0,m}^n(r) = 0$, then

$$\nabla_n[\sigma(r)\nabla_n\varphi_{0,m}(r)] = 0 \tag{8}$$

or it can be rewritten as

$$\nabla_n[\sigma(r)\nabla_n\varphi_{0,m}'(r)] = -\nabla_n[\sigma(r)\nabla_n(Ex_m)] \tag{9}$$

where

$$\varphi'_{0,m}(r) = \varphi_{0,m}(r) - Ex_m \qquad \varphi'_{0,m}(r)|_{r \in \Gamma} = 0.$$

Hence $\nabla_n(Ex_m) = E\delta^n_m$,

$$\varphi_{0,m}(r) = Ex_m + \varphi'_{0,m}(r) = Ex_m - \int_V G(r,r')\nabla'_n[\sigma(r')\nabla'_n(Ex'_m)] \, dV'$$

$$= Ex_m + E\int_V \sigma(r')\nabla'_m G(r,r') \, dV' \tag{10}$$

and

$$j^n_{0,m}(r) = \sigma(r)\nabla_n\varphi_{0,m}(r) = E\sigma(r)\delta^n_m + E\int_V \sigma(r')\sigma(r)\nabla_n\nabla'_m G(r,r') \, dV'. \tag{11}$$

Using the Green function one may calculate this integral. This is of primary importance both for the Hall effect and for the thermopower. For further applications the integral may be expressed in another form:

$$\int_V \sigma(r)\sigma(r')\nabla_n\nabla'_m G(r,r') \, dV = E^{-1}j^n_{0,m}(r) - \sigma(r)\delta^n_m. \tag{12}$$

Using the Green function one can now find a solution for $\varphi_{1,m}(r)$. Because $\nabla j_{1,m}(r) = 0$,

$$\nabla_n[\sigma(r)\nabla_n\varphi_{1,m}(r)] = -\nabla_n[\sigma^2(r)R(r)][H \times \nabla\varphi_{0,m}(r)]_n \tag{13}$$

$$\varphi_{1,m}(r) = \int_V \nabla'_n G(r,r')\sigma^2(r')R(r')[H \times \nabla\varphi_{0,m}(r')]_n \, dV'. \tag{14}$$

Now we are prepared to calculate the Hall effect to first order in the magnetic field. According to (3) we must determine

$$\int_V j^y_x(r) \, dV = \int_V j^y_{1,x}(r) \, dV + \int_V \sigma(r)\nabla_y\varphi_{0,x}(r) \, dV. \tag{15}$$

The second component on the right-hand side of equation (15) equals zero, owing to the boundary condition $\varphi_{0,x}(r)|_{r \in \Gamma} = Ex$ (or simply because it is independent of the magnetic field). Thus we have to calculate

$$\int_V j^y_{1,x}(r) \, dV = \int_V \sigma(r)\nabla_y\varphi_{1,x}(r) \, dV + \int_V \sigma^2(r)R(r)[H \times \nabla\varphi_{0,x}(r)]_y \, dV. \tag{16}$$

Using the result for $\varphi_{1,x}(r)$ from (14) one can obtain

$$\int_V \sigma(r)\nabla_y\varphi_{1,x}(r) \, dV = \int_V \int_{V'} \sigma(r)\sigma(r')\nabla_y\nabla'_n G(r,r')$$

$$\times \sigma(r')R(r')[H \times \nabla\varphi_{0,x}(r')]_n \, dV \, dV'$$

$$= \int_V [E^{-1}j^n_{0,y}(r') - \sigma(r')\delta^n_y]\sigma(r')R(r')[H \times \nabla\varphi_{0,x}(r')]_n \, dV'$$

$$= E^{-1} \int_V j^n_{0,y}(r)\sigma(r)R(r)[H \times \nabla\varphi_{0,x}(r)]_n \, dV$$

$$- \int_V \sigma^2(r)R(r)[H \times \nabla\varphi_{0,x}(r)]_y \, dV. \tag{17}$$

The last integral is the same as the second component on the right-hand side of (16) and is cancelled. Thus for the average Hall current, there is a very simple expression:

$$\int_V j^y_{1,x}(r) \, dV = E^{-1} \int_V R(r)j^n_{0,y}(r)[H \times j_{0,x}(r)]_n \, dV. \tag{18}$$

Now the effective Hall coefficient from equation (3) becomes

$$R^{\text{eff}}(p) = \frac{\int_V \{j_{0,y}(r) \cdot [j_{0,x}(r) \times H]\}R(r) \, dV}{V\sigma_x^{\text{eff}}(p)\sigma_y^{\text{eff}}(p)E^2 H}. \tag{19}$$

3. Calculation of the Seebeck coefficient in a disordered system

With the Green function technique used in the previous section one can easily obtain an analogous formula for the Seebeck coefficient. Assuming that the medium can be described by local thermoconductivity $\kappa(r)$, temperature $T(r)$ and Seebeck coefficient $\alpha(r)$ it is possible to write equations for the electric $j(r)$ and thermal flux $u(r)$ as follows:

$$j_k(r) = \sigma(r)\nabla_k\varphi(r) - \sigma(r)\alpha(r)\nabla_k T(r) \tag{20a}$$

$$u_k(r) = -\kappa(r)\nabla_k T(r) + \alpha(r)\sigma(r)T(r)\nabla_k\varphi(r). \tag{20b}$$

Suppose that opposite edges of the sample are electrically connected and the thermoelectric current is measured. The temperature of the edges is kept at T_1 and T_2. The boundary conditions for $\varphi(r)$ and $T(r)$ are

$$\varphi(r)|_{r\in\Gamma} = 0 \qquad T(r)|_{r\in\Gamma} = (T_2 - T_1)x_m/L_{x_m}. \tag{21}$$

Now assuming $\alpha(r)$ is small, i.e. second term in equation (20b) is much less than the first and can be neglected, system (20) can be written as

$$\nabla_k[\sigma(r)\nabla_k\varphi(r)] = \nabla_k[\sigma(r)\alpha(r)\nabla_k T(r)] \tag{22a}$$

$$\nabla_k[\kappa(r)\nabla_k T(r)] = 0. \tag{22b}$$

Equation (22b) for the temperature distribution is similar to that for the electric current with conductivity $\kappa(r)$. On a lattice it can be solved by iterations of the Kirchhoff equation.

From (22a) and using the Green function, it is easy to obtain

$$\varphi(r) = \int_V G(r, r')\nabla'_k[\sigma(r')\alpha(r')\nabla'_k T(r')] \, dV'$$

$$= -\int_V [\nabla'_k G(r, r')]\sigma(r')\alpha(r')\nabla_k T(r') \, dV'. \tag{23}$$

The effective Seebeck coefficient is proportional to the mean thermocurrent:

$$\langle j_m(r) \rangle = \int_V \sigma(r)\nabla_m \varphi(r) \, dV - \int_V \sigma(r)\alpha(r)\nabla_m T(r) \, dV. \tag{24}$$

Substituting $\varphi(r)$ from (23) and using (12) the first term in (24) becomes

$$\int_V \sigma(r)\nabla_m \varphi(r) \, dV = -\int_{V'} \alpha(r')\nabla'_k T(r') \int_V \sigma(r')\sigma(r)\nabla_m \nabla'_k G(r, r') \, dV$$

$$= E^{-1} \int_V j^k_{0,m}(r)\alpha(r)\nabla_k T(r) \, dV + \int_V \sigma(r)\alpha(r)\nabla_m T(r) \, dV. \tag{25}$$

The second term in (25) is cancelled by the second term in (24) so for the effective Seebeck coefficient a simple expression is obtained:

$$\alpha^{\text{eff}}(p) = \frac{\int_V \alpha(r)[j_{0,m}(r) \cdot \nabla T(r)] \, dV}{\sigma^{\text{eff}}_m(p)(T_2 - T_1)VE}. \tag{26}$$

In the case of a two-component medium the volume integral can be transformed into:

$$\alpha^{\text{eff}}(p) = \frac{\int_S \alpha(r)[n \cdot j_{0,m}(r)][T(r) - T_1] \, dS}{\sigma^{\text{eff}}_m(p)(T_2 - T_1)VE} \tag{27}$$

where S is the surface between the two phases, and n is normal to S.

4. Model of an infinite cluster

We are considering a two-component system with 'high' conducting component '1' and 'low' conducting component '2'. At $p < p_c$ one considers a metal–superconductor transition, in which case component '1' is superconductor and component '2' is metal. At $p > p_c$ component '1' is metal and component '2' is insulator. To define the 'special' points for the Hall and Seebeck coefficients and for the Joule heat we introduce

$$w^{\text{eff}}(p) = \int_V j^2(r)/\sigma(r) \, dV. \tag{28}$$

One can assign to each bond the value for the integrand expressions (19), (26) and (28). This can be done after solving the system of Kirchhoff equations and calculating the current density on each bond. In the case of Joule heat the physical meaning of the integrand expression is the Joule heat density; for the Hall and Seebeck coefficients there is no direct physical meaning for the integrand expression.

The distribution function of those integrand expressions on volume for all bonds can be found. It turns out that one can divide all volumes into three classes. For the purpose of this classification one has to introduce a cutting parameter ε, $0 \leqslant \varepsilon \leqslant 1$. Taking $\varepsilon = 0.985$ we investigate the question: what part of the volume gives off $\varepsilon w^{\text{eff}}(p)$ of the Joule heat.

It is necessary to make the distribution of all bonds in order of their decreasing Joule heat and begin to summarize this row from the 'hottest' point. We put into the first (small) class '1' all 'hottest' bonds that together give a value of Joule heat equal to exactly $\varepsilon w^{\text{eff}}(p)$. The remaining bonds are divided into two further classes.

The second class '2' is the largest part of the volume, but gives off very little of the Joule heat: $(1 - \varepsilon)w^{\text{eff}}(p)$.

The relatively small third class '3' does not give any Joule heat—these bonds are called 'dead ends'. This class exists only for $p > p_c$.

This classification allows us to build a single model of the phase transition on both sides of the percolation threshold: $p = p_c - \Delta p$ (metal–superconductor), $p = p_c + \Delta p$ (metal–insulator).

It is known that the order parameter for $p > p_c$, $P(p)$, is the relative volume of the infinite cluster and many papers have been devoted to its topological structure (Stauffer 1979, Stanley and Coniglio 1983, Kapitulnik and Deutcher 1984). Our calculations show that, for each kinetic coefficient, different geometrical properties are important and it is impossible to develop a single universal model for all the kinetic coefficients.

Even the 'dead ends' for the Hall effect (the volume for which the value of the integrand expression (19) is equal to zero) do not coincide with the 'dead ends' for conductivity, because the dead ends for the longitudinal conductivity $\sigma_x^{\text{eff}}(p)$ may carry current for the transverse conductivity $\sigma_y^{\text{eff}}(p)$.

Each kinetic coefficient has its own 'special points' that make up its specific model of an infinite cluster. Let us begin with constructing the infinite cluster model for conductivity.

For an increasing concentration in the critical interval, we investigate the dynamics of the phase transition for conductivity. What is changing in the critical interval when we add Δp of the component '1' bonds? Below the percolation threshold at $p = p_c - \Delta p$ there are large finite clusters from component '1' that are placed in class '2' and there are thin leads of component '2' between them. It turns out that those leads are 'hot points' (class '1'). This classification allows us to introduce a new determination of a conducting infinite cluster which consists of two components and carry the current. To change the concentration we add component '1' bonds; if this bond is placed on a 'hot point' it becomes 'cold' and the quantity of 'hot points' decreases as the percolation threshold approaches. Very surprisingly, it turns out that a two-component infinite cluster decreases to zero owing to changing current flow. The number of 'hot points' decreases; near threshold there remains one, consisting of component '2' in the volume $\rho^3(p)$, where $\rho(p)$ is the correlation radius, which diverges towards the percolation threshold.

With further increasing of concentration at $p > p_c$ the 'hot points' consist of component '1' and their quantity increases. But each of them becomes colder and colder; the distribution of heating changes as well as current flow. So only investigation of the Joule heat distribution allows one to obtain an exact solution of the problem of infinite cluster topology. In our model one can imagine an 'infinite cluster' as a superlattice consisting of big finite clusters of 'ideal metal' that are almost unchanged in the critical interval. All changes occur at the 'hot points' which connect large clusters like bridges.

It is possible to build similar distribution functions for the Hall effect and for thermopower, using instead Joule heat integrand expressions (19) and (26). After that the same classification for the volume is made and points with a high value for the integrand expression are called 'active points'. The 'active points' for the Hall effect turn out not to be in the same place as 'hot points' because the places where the current density $j_{0,x}(r)$ is high are not the same as where $j_{0,y}(r)$ is high and we are interested in their multiplication. But calculation shows that the dynamics of Hall 'active points' is the same as that of 'hot points'. On both sides of the transition their quantity decreases as the percolation threshold approaches, and at p_c there remains only one 'active point'.

For thermopower, the existence of 'active points' depends on the relation of the thermoconductivities. If $\kappa_1(r)/\kappa_2(r) \to \infty$ then the existence of 'active points' for thermopower can be found and geometrically they coincide with the 'hot points'; if $\kappa_1(r)/\kappa_2(r) \to 1$ there are no 'active points' for thermopower and we cannot speak about a superlattice for thermopower.

5. Conclusions

In this paper we have shown a method for calculating the effective values of two effective coefficients using their local values. A similar method can be used for all these kinetic coefficients, which can be introduced through a local value. The special features of those physical quantities near the percolation threshold are the existence of the 'special' points, whose existence is due to irregular current flow through the sample.

An infinite cluster consists of large finite clusters (referred to as class '2'), which almost do not change in the critical interval. All changes in phase transition happen at the 'hot points', which are situated at the same places below and above the threshold, because they are narrow bridges between large clusters. At $p < p_c$ the 'hot points' consist of component '2', and at $p > p_c$ of component '1'. Given a change of concentration in the critical interval, the quantity of 'hot points' and heat released by each one is changed.

Acknowledgments

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