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CPA theory for the giant Hall effect in disordered nanoscale systems

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Abstract. We present a theory based on the Green's-function formalism to study the so-called giant Hall effect in disordered nanoscale systems in which the metallic sites build up percolating networks. The disordered Green's functions are solved in the coherent potential approximation (CPA). This method is an extension of the Blackman-Esterling-Beck approach. The crucial point is that we are able to predict the quantum percolation threshold near which the Hall coefficient is enhanced by several orders of magnitude.

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In recent years many experimental works have been presented on the so-called giant Hall effect in granular metal-insulator films or binary alloys near the percolation threshold, in which the granular size is scaled about several nanometers [1–4]. The Hall coefficient in such systems is 3 or 4 orders of magnitude greater than that in a pure metal. Pakhomov et al. firstly reported the giant Hall effect in Ni-SiO$_2$ films in 1995 [1]. With the metal volume fraction x near the metal-insulator transition, the saturated value of the Hall resistivity is almost 4 orders of magnitude greater than that of the pure nickel. Afterwards, nearly 3 orders of magnitude enhancement in the Hall coefficient was observed in Cu-SiO$_2$ granular films, which are nonmagnetic [2]. It has been found that the Hall coefficient varies little in both the magnetic and nonmagnetic metal granular systems throughout a quite wide range of temperature. Phenomena similar to these cases were also found in disordered binary alloys near some special volume fraction by experimentalists [3].

Theoretically, Wan and Sheng studied the giant Hall effect in nonmagnetic films [4]. They introduced the competition between the local quantum interference and the percolation to explain the giant Hall effect. Since the weak-localization of electrons caused by their wave nature decreases the effective electron density, the effective Hall coefficient is enhanced. The Hall coefficient was calculated by the Kubo formula in which the eigenfunctions and eigenenergies of a disordered system were solved by the numerical diagonalization of the Hamiltonian. The numerical calculation process needs a large amount of sampling average.

The basic physics for the giant Hall effect is now becoming clear. In a metallic percolation network the wave nature of charge carriers can significantly modify the classical picture of the Hall effect, especially when the metallic concentration x is around the quantum-percolation threshold x_q. In general, the quantum percolation threshold x_q is greater than the classical percolation threshold x_c. In this paper, we would analytically study the Hall effect in nonmagnetic disordered metal-insulator composites. However, unlike many previous studies in disordered binary alloys or metal-insulator composite systems, not only the diagonal disorder should be taken into account, but also the nondiagonal disorder plays a quite important part in the present case. These characteristics increase difficulties for analytic treatment of the giant Hall effect.

It is reasonable to assume that the structure of a metal-insulator composition system for study is subjected to a crystallographic description. A strictly periodic lattice containing N equivalent sites is randomly occupied by two kinds of atoms, the metallic and the insulated. The respective concentrations are x and $(1-x)$. These considerations define a whole ensemble of possible arrangements of atoms. We are interested in the physical properties of the disordered system averaged over this ensemble.

Since the metallic atoms are randomly distributed in the insulator background, the model Hamiltonian can be written as

$$\mathcal{H} = \sum_{i,j} t_{ij} \left[\exp\left(iA_{ij}/\phi_0\right) a_i^{+} a_j + \exp\left(-iA_{ij}/\phi_0\right) a_j^{+} a_i\right] + \sum_i \epsilon_i a_i^{+} a_i, \tag{1}$$

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where ϵ_i and t_{ij} are the on-site energy and hopping integral, respectively, and are all random variables. The on-site energy ϵ_i is an finite value ϵ for metallic sites, but is infinite for insulated sites. In the nearest-neighbor approximation, if site i and site j are all metallic, $t_{ij} = 1$; otherwise, $t_{ij} = 0$. The other parameter in equation (1) is A_{ij} which represents the gauge field, induced by a uniform magnetic field B along the z-axis, $\phi_0 = hc/e$ is the unit quantum flux. We choose the gauge to give $A_{i,i+\{0,0,1\}} = A_{i,i+\{0,1,0\}} = 0$, and $A_{i,i+\{1,0,0\}} = -Bb_y b_x j$, where b_x, b_y and b_z are the lattice separations in the three orthogonal directions. For convenience, we take the cubic lattice, so $b_x = b_y = b_z = b$.

The conductivity tensor σ in such disordered system is given by the well-known Kubo formula [5]

$$\sigma_{\alpha\beta} = \frac{2\pi e^2\hbar}{\Omega}\mathrm{Tr}[v_\alpha \mathrm{Im}\langle G\rangle v_\beta \mathrm{Im}\langle G\rangle], \tag{2}$$

where e is the electron charge, Ω is the volume of the whole system, the Greek letters α, β refer to the Cartesian coordinates. Here v_α and v_β are the velocity operators determined by the Heisenberg equation of motion, whereas G is the Green's function, and the symbol $\langle\ \rangle$ is used to express the ensemble average.

The most important quantity is G_{ij} which represents the matrix element of the Green's function G or can be directly called the site Green's function. To get the conductivity tensor we have to calculate $\langle G_{ij}\rangle$ in the coherent potential approximation (CPA). In the theory of the Green's function, we can write the recursion equation for the site Green's function as

$$G_{ij} = g_i\left(\delta_{ij} + \sum_{im} W_{im}G_{mj}\right), \tag{3}$$

where

$$W_{im} = \begin{cases} \exp(iA_{im}/\phi_0), & \text{if } i \text{ is nearest to } m,\\ 0, & \text{otherwise,}\end{cases} \tag{4}$$

and

$$g_i = \frac{1}{z-\epsilon_i}. \tag{5}$$

The latter is the bare locator for site i. If the disordered distribution of metal atoms is taken into account, the site Green's function and bare locator can be rewritten, like in the Blackman-Esterling-Beck's approach [6,7], as the following equations

$$\tilde{G}_{ij} = x_i G_{ij} x_j, \tag{6}$$

and

$$\tilde{g}_i = x_i g_i. \tag{7}$$

Here the projection operator x_i is equal to one if site i is occupied by a metallic atom and is zero otherwise.

Obviously the disorder is only associated with a single-site quantity now. The locator $\tilde{g}_i$ varies from site to site. However, W_{ij} is assumed to be translationally invariant, depending only on the vector $\boldsymbol{R}_{ij}$ between sites i and j. The diagonal matrix element $\tilde{G}_{ii}$ can be written as

$$\tilde{G}_{ii} = \frac{\tilde{g}_i}{1-\tilde{g}_i\Delta_i}, \tag{8}$$

where Δ_i is the fully renormalized interaction defined by

$$\Delta_i = \sum_{i\neq j} W_{ij}\tilde{g}_j W_{ji} + \sum_{j\neq i}\sum_{m\neq i} W_{ij}\tilde{g}_j W_{jm}\tilde{g}_m W_{mi} + \cdots \tag{9}$$

From this expansion, we can understand that Δ_i arises from the hopping of electrons away from and then back to site i, avoiding site i in the midway. It provides a self-energy correction to the site energy ϵ_i.

We have to make an approximation to evaluate the expansion in equation (9). An effective-medium locator is introduced by

$$\bar{g} = \frac{1}{z-\epsilon_0}, \tag{10}$$

in terms of a renormalized energy ϵ_0. The renormalized interaction can also be obtained and is denoted by $\bar{\Delta}$. Replacing Δ_i by $\bar{\Delta}$ in equation (8), one obtains the result

$$\tilde{G}_{ii} = \frac{\tilde{g}_i}{1-\tilde{g}_i\bar{\Delta}}. \tag{11}$$

It is consistent with the spirit of the CPA to replace locator $\tilde{g}_i$ in equation (7) by the effective-medium locator $\bar{g}$, because the renormalized interaction $\bar{\Delta}$ depends only on the effective-medium locator $\bar{g}$. Then, an effective-medium Green's function is obtained as

$$\langle\tilde{G}_{ii}\rangle_{\mathrm{eff}} = \frac{\bar{g}}{1-\bar{g}\bar{\Delta}}. \tag{12}$$

From equation (11) one can define

$$\gamma \equiv \langle\tilde{G}_{ii}\rangle, \tag{13}$$

where $\langle\ \rangle$ means the average over the occupation of a single site. So γ is independent of site index i. The effective-medium Green's function $\langle\tilde{G}_{ii}\rangle_{\mathrm{eff}}$ must be equal to $\langle\tilde{G}_{ii}\rangle$, so

$$\gamma = \frac{1}{z-\epsilon_0-\bar{\Delta}} = \frac{x}{z-\epsilon-\bar{\Delta}}. \tag{14}$$

From this equation, ϵ_0 can be found.

In the momentum representation, we have

$$\gamma = \frac{1}{N}\sum_{\boldsymbol{k}} G(\boldsymbol{k}) = \frac{1}{N}\sum_{\boldsymbol{k}}\frac{1}{z-\epsilon-W_{\boldsymbol{k}}}, \tag{15}$$

where the sum for $\boldsymbol{k}$ is taken over the first Brillouin zone. The self-consistent condition for the renormalized interaction $\bar{\Delta}$ is derived as

$$\frac{1}{\bar{\Delta}} = \sum_{\boldsymbol{k}}\frac{1}{z-\epsilon-2\bar{\Delta}-xW_{\boldsymbol{k}}}, \tag{16}$$

where the Fourier transform of $W_{\boldsymbol{k}}$ is defined by the expression

$$W_{\boldsymbol{k}} = \frac{1}{N}\sum_{i,j} W_{ij} e^{i\boldsymbol{k}\cdot\boldsymbol{R}_{ij}}, \tag{17}$$

which can be evaluated by combining with equation (4).

By the numerical calculation for equations (16) and (17), we can get the renormalized interaction $\bar{\Delta}$, and then γ from equation (14). As usual, in the Green's function approach, the variable z must be replaced by $z + i\eta$ where η is an infinitesimal quantity, and the limit $z \longrightarrow 0$ must be taken. Finally, the average site Green's function can be expressed in the form

$$\langle G_{ij}\rangle = \gamma + \gamma W_{ij}\gamma + \gamma W_{im}\gamma W_{mj}\gamma + \cdots \tag{18}$$

Here still the nearest neighboring sites are taken into account. Since γ and W_{ij} are not dependent on the site indices after average, $\langle G_{ij}\rangle$ is only dependent on the vector $\boldsymbol{R}_{ij}$. A further restriction is needed. In each term of equation (18), any site index is not allowed to repeat, and all γs of a particular term refer to different sites. We use the first four terms for our calculations, because equation (18) is an infinite series, a cut-off must be made to get numerical results. The number of terms to be included is relevant to the dephasing length of the system.

In nanoscale metal-insulator composites, a conductivity transition is predicted, if charge carriers are assumed to be classical particles at the geometric percolation threshold x_c, below which the metallic component can no longer form a infinite network. But the real picture is quite different, when the wave nature of the charges is important taken into account, and the inelastic scattering is absent in a region larger than several granules, that is, the phase coherence is conserved in the elastic mean free path. Localization is caused by the multiple scatterings of the electronic waves in the random metallic network. Even if the metallic volume fraction x is above the geometric percolation threshold x_c and the infinite metallic network is formed, an electron is still in its localized area until $x > x_q$, which is the quantum percolation threshold. When $x_c < x < x_q$, the classical-percolation predicts metallic behavior while the quantum-percolation predicts nonmetallic behavior. Since the phase factor is associated with the external magnetic field, the Hall coefficient also depends on the external magnetic field.

Now we can calculate the average site Green's function self-consistently by the numerical iterative method. And then the site Green's function is substituted into the Kubo formula in order to get the conduction tensor, especially the transverse conductor σ_{xy}, and the Hall coefficient R_{H} is given by

$$R_{\mathrm{H}} = \frac{1}{\sigma_{xy}B}. \tag{19}$$

The $\log R_{\mathrm{H}}$ is plotted versus $(x - x_c)$ in Figure 1. In general, the classical percolation threshold x_c is about 0.31. Above the classical percolation threshold x_c, the Hall efficient R_{H} increases with increasing metallic volume fraction until x is larger than a certain value. This value here is about 0.41 which is the quantum percolation threshold.

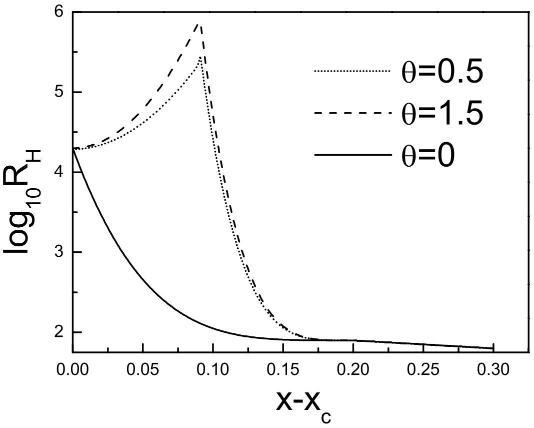

Fig. 1. Effective Hall coefficient versus metallic concentration calculated in the coherent potential approximation. The phase factor $\theta = -Bb^2/\phi_0$, where b is the bond length in the three directions. The effect of the phase factor is shown in the present figure.

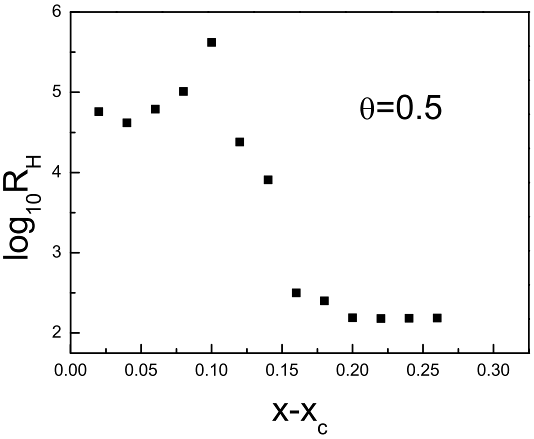

Fig. 2. Effective Hall coefficient versus metallic concentration calculated by the numerical diagonalization method.

We use θ to label the phase factor. It could be seen in Figure 1 that, near the quantum percolation threshold x_q, the Hall coefficient R_{H} peaks for several finite θ. Afterwards, R_{H} begins to decrease as in the classical cases. The value of the Hall coefficient at x_q is about 3 orders greater than the pure metal case. The dependence of the magnetic filed is also exhibited in Figure 1. These results are in excellent agreement with the formerly experimental and theoretical results.

For comparison, we also use the numerical diagonalization method to deal with the Hamiltonian in equation (1) [4]. The calculated results for R_{H} versus $(x - x_c)$ are shown in Figure 2. These totally numerical calculations by sampling and average also indicate that both the quantum interference and the classical percolation are essential in the giant Hall effect. The competition between the two kind of behaviors leads to the great enhancement of the effective Hall coefficient. These results agree well with the results obtained from the CPA and Green's function above.

In summary, we have presented an analytic method to study the local quantum interference and percolation in the electron transport in granular metal-insulator composites. The conductivity, Hall coefficient and other physical quantities can be numerically calculated by our method. We have shown that the quantum interference and percolation can change the electron transport behavior dramatically, which offers a quantitative explanation of the giant Hall effect in nonmagnetic granular metal-insulator systems.

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