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Theoretical study of magnetoresistance for granular metals in the hopping regime

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Abstract

We present a theoretical study of magnetoresistance for granular metals deep in the hopping regime, in the limit of large metal/insulator volume ratio, within the framework of coherent electron backscattering. We adopt the one-electron approximation, and examine effects of a magnetic field and spin–orbit interaction on the backscattering, both in the semi-classical approximation. We derive an expression for magnetoresistance characteristic of zero-dimensional systems, i.e. dots. The result is compared with previous experiments.

1. Introduction

Coherent electron backscattering in disordered systems has many interesting consequences, among which is the magnetoresistance (MR) for systems in the weak-localization regime [1]. The origin of this MR is attributed to the destruction of backscattering, caused by magnetic-field-induced phase difference between quantum amplitudes of conjugate returning paths (clockwise and counterclockwise). When spin–orbit scattering is negligible, the backscattering enhances the resistance, and its destruction by a magnetic field results in negative MR, whereas in the presence of strong spin–orbit scattering, the backscattering reduces the resistance, and the corresponding MR is positive. On the other hand, for systems in the variable-range-hopping (VRH) regime, recent theoretical works [2] have shown that MR can arise from a different mechanism, namely, the quantum interference between various 'forwarding' paths. Since the effect depends on amount of magnetic flux encircled by paths, for two-dimensional (2D) systems, 'forwarding-MR' (FMR) theories predict anisotropic MR behaviour with respect to field direction. Experimental works in the past on strongly localized systems [3–6] were largely explained by these theories.

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Our motivation for the present study is to address the following questions. (1) For disordered systems such as doped semiconductors, a hopping path is primarily contained in the insulator, with the effect of a magnetic field on hopping path well described by FMR theories. In contrast, for some granular metals, the metal/insulator volume ratio can be very large. Does the magnetic influence on electron waves inside metal grains play any role? Is any phenomenon reminiscent of the 'backscattering MR' (BMR) observable? (2) There are experiments [7] showing isotropic MR for 2D granular metal films, which appear inconsistent with the anisotropy prediction of the FMR theory, and, hence, may belong to a category for which a different calculation is needed. In this work, we hope to examine carefully the role of backscattering and its relation with observed MR at low temperatures, for granular metals deep in the hopping regime, in the limit of large metal/insulator volume ratio.

Several important theoretical works are also relevant to the problem we are studying here. Raikh and Glazman considered the effect of a magnetic field on tunnelling between two smooth potential wells, and obtained a large MR [8]. Wang and Xie discussed the MR due to field-induced level crossing for a system of identical discs [9]. Eto developed an MR theory within a disordered interacting Anderson model [10]. In the calculation to be presented below, we assume (1) following [11], that metal grains are crystalline but with rough surfaces, (2) that neighbouring grains are not necessarily identical and (3) that the one-electron approximation is valid. The last assumption is made in view of the fact that it was employed for BMR calculations before [1].

In section 2, we present our theoretical model. In section 3, we compare the model with experiments. In section 4, we conclude the study.

2. Theoretical model

For a granular system in the strongly localized regime, the charge transport is determined by the so-called critical network of resistances, with each resistance corresponding mainly to a single coherent hopping path between two grains, labelled as 1 and N. Typically, N, the number of grains covered by the coherent hopping, is of the order of a few, for the temperature range 1–300 K. The smallness of N is due to the range of the parameters specific to the granular system, e.g., charging energy, intergrain spacing and wave function decay constant. This is in contrast to the case of doped semiconductors, where long-range hoppings occur at low temperatures. Although, in principle, there exists more than one hopping path between a given pair of grains, they are widely different in resistance, because the resistance, being proportional to

$$\exp\left(2\alpha \sum_{i=1}^{N-1} S_{i,i+1}^{\Gamma}\right),$$

varies drastically from path to path, where α is the wave function decay constant in the insulator, and $S_{i,i+1}^{\Gamma}$ the intergrain spacing between two neighbouring grains contained in the path Γ. To the approximation that we consider only the dominant path which gives the lowest resistance between grains 1 and N, the situation is as if we have a quasi-one-dimensional system, a string of grains with grains 1 and N at the ends. This is in contrast to the case of the impurity-doped semiconductors, where, between a given pair of sites (distant from each other), there are many other impurities between them, and zigzag forwarding paths via the intermediate sites with path length close to that of the shortest path (i.e. the direct path between the pair) can exist. Interference among such paths, including the shortest one, is important,

because they all make contributions of similar magnitude to the total hopping amplitude for the pair. This interference varies with magnetic field and gives rise to FMR. In our case, we have only one dominant string of grains. However, a possible source of MR can come from interference between conjugate returning paths constrained to this string. Note that, in this model, since the paths can only enclose the flux going through the grains of the string, it is obviously the same amount of magnetic flux that the string sees regardless of how the magnetic field is oriented, with the exception of disk-shape grains, of course. This explains why MR in this case can show isotropy. However, as we move away from the strong localization regime towards the metal–insulator transition, there can eventually appear more than one hopping path dominating the transport between two grains. The increase in the number of dominant paths will continue, and, when the metallic regime is finally reached, there are infinitely many equally important conducting paths between two points. So, closed loops composed of two different dominant paths can exist in the 'intermediate localization' regime, and rings of grains for such loops must be considered for an MR calculation. In this regime, the conjugate paths constrained to a ring can enclose the flux going through the area bounded by the ring, and the backscattering MR tends to show anisotropy. Continuing this trend finally brings us back to the ordinary backscattering MR in the weakly localized limit. Our calculation will be limited to the systems deep in the strongly localized regime.

Firstly, we discuss tunnelling between two neighbouring grains, say, 1 (in state $|1\rangle$) and 2 (in state $|2\rangle$). Let t be the tunnelling coupling. The hopping conductance between the grains is proportional to $|\langle 1|t|2\rangle|^2$, which, in coordinate representation, becomes $\Sigma_{X,Y}\langle 1|t|X\rangle\langle X|2\rangle\langle 2|Y\rangle\langle Y|t|1\rangle$. We make certain simplifications for this expression. Following Averin and Nazarov [12], we assume that the tunnelling matrix element is non-vanishing only if X and Y are close to the metal surface. Moreover, with short wavelength of atomic size and frequent random surface scattering, both $|1\rangle$ and $|2\rangle$ have phases which vary rapidly and chaotically with the position. So, the foregoing double sum reduces, within atomic resolution, to the single sum $|\langle 1|t|2\rangle|^2 \sim \Sigma_X|\langle 1|t|X\rangle|^2|\langle X|2\rangle|^2 = M|\langle 1|t|X_2\rangle\langle X_2|2\rangle|^2$, where X_2 is a representative surface atom on grain 2, with sizable tunnelling coupling to grain 1, and M is the effective number of such atoms. This summation of the squared terms means the neglect of possible interference among tunnelling paths ending at different X. Therefore, we shall regard the paths as if they are parallel, independent conducting channels and treat the interference effect, e.g., MR, for each channel separately. The overall MR can be obtained afterwards by a straightforward application of electric circuit theory. We stress that this independent channel approximation holds only with frequent surface scattering and short electron wavelength (compared with grain size).

Next, we regard a granular system as a conductor network and focus on one element corresponding to a coherent hopping covering N grains. The effective matrix element for a single conducting channel of the hopping is

$$T_{1N} = \sum_{2,\ldots N-1} t_{12}t_{23}\cdots t_{N-2,N-1}\langle N-1|t|X\rangle\langle X|N\rangle/(\Delta\varepsilon_{N,2}\Delta\varepsilon_{N,3}\cdots\Delta\varepsilon_{N,N-1}) \tag{1}$$

where the surface point X on grain N is written explicitly. $|1\rangle$ is a state on grain 1, coupled to the (initial) ground state $|1_g\rangle$ by electron–boson (plasmon or phonon) interaction. $\Delta\varepsilon_{N,i} = \varepsilon_N - \varepsilon_i + I/2\tau_{in}$ is the energy difference between the final state $|N\rangle$ (on grain N) and an intermediate state $|i\rangle$ (on grain i), with τ_{in} the dephasing time. We take ε_N to be higher than ε_{1g}, and assume that, prior to the hopping, the difference $\Omega = \varepsilon_N - \varepsilon_{1g}$ is compensated by boson absorption in grain 1. Other cases can be similarly treated. We denote the conductance associated with $T_{1\mathrm{N}}$ as g.

Backscattering for VRH systems is usually neglected as it is generally believed to be very small. However, let us examine it in detail for granular systems. We add a term to T_{1N}, with now $T_{1N}^{total} \sim T_{1N} + T_{1N}^{back}$, where T_{1N}^{back} corresponds to the hopping sequence $1 \to 2 \to \cdots \to N \to (N-1) \to \cdots \to 1 \to 2 \to \cdots \to N$. The leading-order correction, $\mathrm{d}g^{\mathrm{back}}$, to g derives from the cross term $T_{1N}\left(T_{1N}^{back}\right)^*$ +c.c. in $\left|T_{1N}^{total}\right|^2$. We now include electron–boson coupling H_{b} (in grain 1) explicitly and draw the following diagram for the cross term.

$$\begin{array}{llllllll} \text{---------o-------x-------x------x-------x-------o-------}\rangle & & & & & & & \\ 1_{\mathrm{g}},\Omega & 1 & N & 1' & N_1 & 1'' & 1_g,\Omega & (2) \end{array}$$

where 'o' is electron–boson interaction vertex and 'x' the tunnelling coupling between grains 1 and N similar to that defined in equation (1). The incoming state is an electron in $|1_{\mathrm{g}}\rangle$ and a boson of energy Ω. Let Σ be the self-energy of the diagram, then $[-2\int \mathrm{d}\Omega D(\Omega)\mathrm{Im}\Sigma]$ gives transition rate of the process corresponding to $T_{1N}\left(T_{1N}^{back}\right)^*$ +c.c., where $D(\Omega)$ is the boson density of states times the Bose–Einstein distribution. The diagram is summed with respect to N_1, $1'$ and $1''$, with states $|1\rangle$ and $|N\rangle$ fixed. N_1, $1'$ and $1''$ denote intermediates states on grains N and 1. With the chaotic property of electron states, the diagram is finite only for $|1\rangle = |1'\rangle = |1''\rangle$, because otherwise it would be random in phase and become zero upon averaging with respect to grain surface configuration. So,

$$\Sigma = \frac{|\langle 1_g,\Omega|H_b|1\rangle|^2}{(\varepsilon_0-\varepsilon_1)^2}\left\{\frac{1}{(\varepsilon_0-\varepsilon_1-\mathrm{i}/2\tau_{\mathrm{in}})}\sum_{N1}\frac{|T_{1,N}|^2}{(\varepsilon_0-\varepsilon_N+\mathrm{i}/2\tau_{in})}\frac{|T_{1,N1}|^2}{(\varepsilon_0-\varepsilon_{N1}-\mathrm{i}/2\tau_{in})}\right\} \quad (3)$$

where $\varepsilon_0 \equiv \varepsilon_{1g} + \Omega$, and the expression in { } describes tunnelling between grains 1 and N. With $\varepsilon_0 = \varepsilon_N - \mathrm{i}/2\tau_{in}$ (since the main contribution to the integral $\int \mathrm{d}\Omega D(\Omega)\mathrm{Im}\Sigma$ comes from this pole), we find the expression contains the factor

$$\left[\sum_{N1}\langle N|X\rangle\langle X|N_1\rangle\langle N_1|X\rangle\langle X|N\rangle/(\varepsilon_N-\varepsilon_{N1}-\mathrm{i}/\tau_{in})\right],$$

or alternatively,

$$\int \mathrm{d}w\frac{1}{(w-\mathrm{i}/\tau_{in})}\left\{\sum_{N1}\langle N|\delta(\vec{r}-X)|N_1\rangle\langle N_1|\delta(\vec{r}-X)|N\rangle\delta(\varepsilon_N-\varepsilon_{N1}-w)\right\}. \quad (4)$$

With the electron wavelength much smaller than the grain size, it is possible to adopt a certain semi-classical approximation treating electrons as classical particles. For metal grains, quasi-classical methods indeed were applied to reduce quantum expressions, such as that in { } of equation (4), to classical description of electron motion [11, 12]. Without going into details, we simply state that this expression is basically the Fourier transform, $p(w)$, of the classical time-dependent backscattering probability $P(t) \equiv \langle\delta[\vec{r}(0)-X]\,\delta[\vec{r}(t)-X]\rangle$, in grain N. We obtain

$$\mathrm{d}g^{back} \propto \int \mathrm{d}t\,P(\mathrm{t})\exp(-\mathrm{t}/\tau_{\mathrm{in}}). \quad (5)$$

For grains, $P(t) \sim$ constant for $t > \tau, \tau$ being the surface scattering time, and we have $\mathrm{d}g^{back} \propto \int \mathrm{d}t\exp(-t/\tau_{\mathrm{in}}) \propto \tau_{\mathrm{in}}$.

Computing $\int \mathrm{d}\Omega D(\Omega)\mathrm{Im}\Sigma$ gives

$$\frac{\mathrm{d}g^{back}}{g} \sim -\mathrm{Im}\left[\frac{|T_{1N}|^2}{\varepsilon_N-\varepsilon_1-I/\tau_{in}}\right]\tau_{in} \sim -(g/\tau_{in})\tau_{in} \quad (6)$$

with g in units of $2e^2/h$. We note again that the single-channel conductance g is different from the network conductance. For low-resistance systems which contain as many as, say, M, parallel channels, between grains 1 and N, with conductances $g_1 \sim g_2 \sim \cdots \sim g_M \sim g$, the circuit conductance between the grains is $G \sim Mg$, and simple circuit theory gives $\mathrm{d}G^{back}/G \sim \mathrm{d}g^{back}/g \sim -(G/\tau_{in})\tau_{in}/M$. On the other hand, for high-resistance systems to which percolation theory applies, a link G between critical subnetworks consists of M serial conductances among which one element, say, g_1, dominates [13]. With $G \sim g_1 \ll g_2, g_3, \ldots, g_M$, circuit theory gives $\mathrm{d}G^{\mathrm{back}}/G \sim -(G/\tau_{in})\tau_{in}M$. In summary, generally we have $\mathrm{d}G^{\mathrm{back}}/G \sim -(1/M)(G/\tau_{in})\tau_{in}$, with M a network-dependent circuit factor which can vary over a wide range of magnitudes. Later, when the theory is compared with experiments, M will be used as an adjustable parameter, and it will be seen that the parameter varies systematically from that characteristic of parallel circuits to that of serial circuits as the system resistance increases.

Now, we treat the effect of magnetic fields. Because we limit ourselves to the regime of large metal/insulator volume ratio, we neglect the magnetic influence on electron waves in the insulator [8]. We focus on the effect inside grains, and, with equation (5) established, we follow [1] and calculate quasi-classically the variation in $\mathrm{d}g^{\mathrm{back}}$ due to field-induced backscattering destruction. The destruction is induced by the phase difference between a pair of conjugated returning paths, $(2\mathrm{i}e/\hbar)\oint A\,\mathrm{d}r$, where A is the vector potential for the magnetic field and the line integration is carried out along a closed trajectory $X \to X$, traversed during a time length of t. Averaging the phase difference over all the trajectories, we obtain $\langle \exp[(2\mathrm{i}e/\hbar)\oint A\,\mathrm{d}r]\rangle \sim \exp(-t/t_H)$. For a spherical grain, we obtain the following magnetic dephasing rate:

$$1/t_H = \frac{e^2 v_f R_g^3}{8\hbar^2} H^2 \tag{7}$$

with v_{f} the Fermi velocity and R_{g} the grain radius. This result is independent of the field orientation except when it is extended to the case of disklike grains.

We now include the effect of spin–orbit (s.o.) scattering and calculate MR. We shall again adopt the semi-classical approximation. In contrast to the early works which were based on k-space formulation [14, 15], however, we consider electron motion in coordinate space, which is more compatible with the confining geometry of grains. We associate a quantum amplitude Q with the classical trajectory inside a grain, $(x_1 = X) \to (x_2) \to \cdots \to (x_{n-1}) \to (x_n = X)$, with Q written as a product of amplitudes, $Q = \langle x_1 \to x_2\rangle\langle x_2, k_1, s_1 \to x_2, k_2, s_2\rangle \cdots \langle x_{n-1} \to x_n\rangle\langle x_n, k_{n-1}, s_{n-1} \to x_n, k_n, s_n\rangle$, where x_{i}' are surface coordinates at which surface scattering occurs, k_{i}' momenta and s_{i}' spins. $\langle x_{\mathrm{i}} \to x_{\mathrm{i+1}}\rangle$ describes (free particle) orbital motion between scatterings, and $\langle x_{\mathrm{i+1}}, k_{\mathrm{i}}, s_{\mathrm{i}} \to x_{\mathrm{i+1}}, k_{\mathrm{i+1}}, s_{\mathrm{i+1}}\rangle$ corresponds to the scattering at $x_{\mathrm{i+1}}$, causing both momentum and spin to change. We take the s.o. coupling to be $V(k_{\mathrm{i}}, k_{\mathrm{i+1}})[\mathrm{i}c(k_{\mathrm{i}} \times k_{\mathrm{i+1}}) \cdot \sigma]$ [15], where $V(k_{\mathrm{i}}, k_{\mathrm{i+1}})$ is the surface scattering matrix element for spinless electrons, c a real number and σ the Pauli spin matrix. With s.o. scattering occurring only near the grain surface while the primary magnetic effect affects phases of orbital motion amplitudes $\langle x_{\mathrm{i}} \to x_{\mathrm{i+1}}\rangle$, we can separate magnetic and s.o. scattering effects. The prescription for treating the magnetic effect on orbital motion is the same as that we discussed earlier. To treat the s.o. scattering effect, we now focus on surface scattering $\langle k_{\mathrm{i}}, s_{\mathrm{i}} \to k_{\mathrm{i+1}}, s_{\mathrm{i+1}}\rangle$ (abbreviation of notations shall be adopted from here on unless possible confusion arises). We write, for the total surface (potential and s.o.) scattering amplitude,

$$\begin{aligned}\langle k_{\mathrm{i}} s_{\mathrm{i}} \to k_{\mathrm{i+1}} s_{\mathrm{i+1}}\rangle &= V(k_{\mathrm{i}}, k_{\mathrm{i+1}})[\delta(s_{\mathrm{i}}, s_{\mathrm{i+1}}) + \mathrm{i}c(k_{\mathrm{i}} \times k_{\mathrm{i+1}}) \cdot \sigma(s_{\mathrm{i}}, s_{\mathrm{i+1}})] \\ &\equiv V(k_{\mathrm{i}}, k_{\mathrm{i+1}}) V_s(k_{\mathrm{i}}, k_{\mathrm{i+1}}, s_{\mathrm{i}}, s_{\mathrm{i+1}}).\end{aligned}$$

Next, we consider the backscattering probability $P(t)$ for an electron having an initial spin, say, $s_1 = u$ (u = up, d = down) and returning to the starting point X with spin s after a time t. We write $P(t) = \sum_s |\langle X, s, t|X, u\rangle_1 + \langle X, s, t|X, u\rangle_2|^2$, where the subscripts '1' and '2' represent a pair of conjugated paths. Expanding $P(t)$, we obtain the sum of cross terms $A + A^*$, which is backscattering interference, with $A \equiv (\langle u, t|u\rangle_2)^*\langle u, t|u\rangle_1 + (\langle d, t|u\rangle_2)^*\langle d, t|u\rangle_1 \equiv A_1 + A_2$. For A_1, we can write (with orbital motion amplitudes dropped)

$$A_1 \propto \sum_{s_1=s_1'=u,s_2,s_2',\ldots,s_{n-1},s_{n-1}',s_n=s_n'=u} \Pi_j |V(k_{j-1}, k_j)|^2 V_s(k_{j-1}, k_j, s_{j-1}, s_j) V_s(k_{j-1}, k_j, s_{j-1}', s_j').$$

It is easy to verify that, assuming the vector product $(k_{j-1} \times k_j)$ to be random due to scattering and taking the average over this random distribution, only the diagonal term in the spin sum with intermediate spins $s_2 = s_2' = \cdots = s_{n-1} = s_{n-1}' = u$ survives in the last equation. The average of the quadratic V_s factor in A_1 gives

$$\langle V_s(k_{j-1}, k_j, s_{j-1} = u, s_j = u) V_s(k_{j-1}, k_j, s_{j-1}' = u, s_j' = u)\rangle \propto 1 - 4\tau/3\tau_{so}$$

where $1/\tau$, now, is the total surface scattering rate, and $1/\tau_{so}$ the s.o. scattering rate. Therefore, $\langle A_1\rangle \propto (1 - 4\tau/3\tau_{so})^{n-1} \sim \exp(-4t/3\tau_{so})$, where $(n-1)\tau$ has been set equal to t. Similarly, for A_2, one can derive that $\langle A_2\rangle \propto [\exp(-4t/3\tau_{so}) - 1]/2$. So, overall, $\langle A\rangle \propto [3\exp(-4t/3\tau_{so}) - 1]/2$. Adding up the s.o. and magnetic dephasing effects, we obtain the following MR:

$$\mathrm{MR} \sim (1/M)(G/\tau_{in}) \left\{\tfrac{3}{2}[\tau_\varphi^*(H) - \tau_\varphi^*(H=0)] - \tfrac{1}{2}[\tau_\varphi(H) - \tau_\varphi(H=0)]\right\} \tag{8}$$

where $1/\tau_\varphi(H) \equiv 1/\tau_{in} + 1/t_H$, $1/\tau_\varphi^*(H) \equiv 1/\tau_{in} + 4/3\tau_{so} + 1/t_H$ and the circuit factor M has been explicitly included.

We note that equation (8) contains three parameters, the circuit factor M, and the scattering times τ_{so} and τ_{in}. Because M is an multiplicative factor, it only affects the amplitude of an MR curve. On the other hand, the shape of the curve is determined by the scattering times.

According to equation (8), when $2/3 < \tau_{so}/\tau_{in} < 4/3(\sqrt{3} - 1)$, there is a positive MR at small H, reaching maximum at H_m with corresponding magnetic dephasing $1/t_{Hm} = 4/[3(\sqrt{3} - 1)\tau_{so}] - 1/\tau_{in}$, and zero at H_0 with $1/t_{H0} = [(\sqrt{3} - 1)/2][(\sqrt{3} + 1)\tau_{so}/\tau_{in} + 4/3]/[(\tau_{so}/\tau_{in} - 2/3)t_{Hm}]$. These relations, together with equation (7), permit us to determine τ_{so} and τ_{in} from experimental values H_m, H_0 and R_g, without any parameter adjustment.

3. Comparison with experiments

The MR predicted by the above theory has several important features. (A) In the presence of s.o. interaction, it shows sign variation with magnetic field, starting with positive values in small fields and turning to negative values for large fields, which is similar to the typical MR in weakly localized systems. (B) It is expected to be smaller than the typical MR of weakly localized systems. (C) It decreases with the resistance of the system. (B) and (C) follow from the fact that the backhopping probability of an electron decreases exponentially with the insulator layer thickness traversed in the hopping path. (D) It shows isotropic behaviour with respect to the orientation of the magnetic field. There are MR measurements performed on granular systems which show the aforementioned features and, hence, are regarded by us as cases where we may test the theory. Comparison between the theory and such experiments is presented below.

In figure 1, we compare our theory with experiments by Kobayashi's group for granular copper films of various resistances, with $R = 7.57$, 19.0, 42.6, 1130 and 1860 kΩ at $T = 4.22$ K,

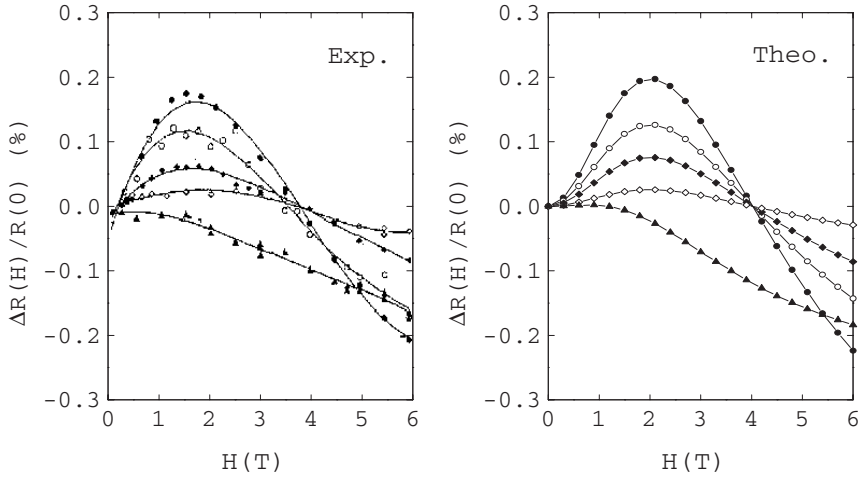

Figure 1. On the left are the experimental MR data for granular Cu films at 4.22 K, reproduced from [7]. On the right is the theoretical result of equation (8). Grain diameter = 40 A. For resistance of films, $R = 7.57$ (closed circles), 19.0 (open circles), 42.6 (closed squares), 1130 (open squares), and 1860 kΩ (closed triangles). The MR decreases in amplitude with increasing film resistance, and turns almost completely negative for the $R = 1860$ kΩ film.

all with the same grain diameter = 40 Å [7]. They indicated that the structure of Cu particles in their samples was crystalline and essentially spherical, surrounded by oxide, with resistances controlled by degree of oxidation. Fairly isotropic MR was observed. With material parameters for copper, equation (7) gives $1/t_{\rm H} = 3.6 \times 10^9 H^2\ {\rm s}^{-1}$, where H is in units of tesla. We take $H_{\rm m} \sim 2$ T and $H_0 \sim 4$ T from experiments, and obtain $\tau_{\rm so} \sim 5.3\times10^{-11}$ s and $\tau_{\rm in} \sim 5.1\times10^{-11}$ s, for the four low-R films. In contrast, earlier experiments of Gershenzon *et al* for Cu films in the weak-localization regime, with film thickness 42 A, gave $\tau_{\rm in} \sim 1.6 \times 10^{-11}$ and $\tau_{\rm so} \sim 0.84 \times 10^{-11}$ [16]. The difference between our $\tau_{\rm so}$ and theirs is consistent with the phenomenon of large suppression of s.o. scattering by quantum size effects (QSEs), which was theoretically explained by Kawabata [17] and experimentally verified [18]. For the 1860 kΩ film, we take $\tau_{\rm in}$ to remain unchanged, and obtain $\tau_{\rm so}$ which increases up to 8.3×10^{-11} s, reasonable if we assume that QSEs increase with increasing oxidation.

The amplitude of MR is fitted with adjustment of circuit factors, which are $M = 43.4$ (for 7.57 kΩ), 27 (for 19.0 kΩ), 20 (for 42.6 kΩ), 2.2 (for 1130 kΩ) and 0.77 (for 1860 kΩ). As we see, this factor decreases systematically, consistent with the picture that the conductor network structure varies continuously from that of a parallel circuit to that of a serial circuit with increasing degree of oxidation. Since the network structure obviously depends on the growth mechanism, magnitudes of M are not to be stressed. In fact, it has been observed that MR amplitude can change by an order of magnitude when growth conditions changes [7].

In figure 2, we compare our theory with experiments by Valles' group for granular Ag films of various resistances, 20, 40, and 140 kΩ, at $T = 3$ K [6]. We take $R_{\rm g} \sim 25$ Å [19], and obtain $\tau_{\rm so} \sim 3.9 \times 10^{-11}$ s, for the 20 and 40 kΩ films, a larger $\tau_{\rm so} \sim 4.08 \times 10^{-11}$ s for the 140 kΩ film and $\tau_{\rm in} \sim 4.9 \times 10^{-11}$ s for all three cases. In contrast, $\tau_{\rm in} = 6.7 \times 10^{-11}$ and $\tau^{\rm so} = 0.94 \times 10^{-11}$ s were obtained by Gershenzon *et al* for Ag films in the weak-localization regime [16], with film thickness 42 Å. The circuit factors are $M = 13.5$ (for 20 kΩ), 4.8 (for 40 kΩ), and 1.1 (for 140 kΩ), again showing the same systematic trend as that in the Cu case.

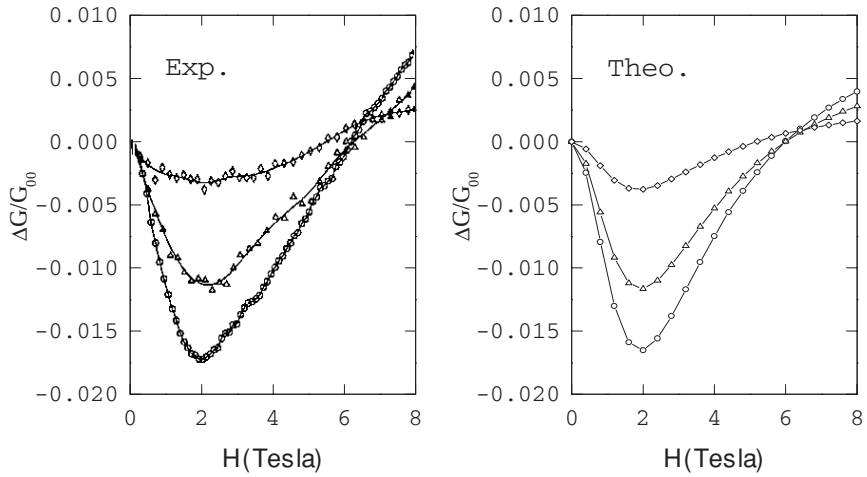

Figure 2. On the left are the experimental MR data for granular Ag films at 3 K, reproduced from [6]. On the right is the theoretical result of equation (8). Grain diameter = 50A. For resistance of films, $R = 20$ (open circles), 40 (open triangles), 140 kΩ (open squares). The MC amplitude decreases with increasing film resistance. $G_{00} \equiv e^2/\pi h$.

In both the above cases, although it appears that two of our parameters, $\tau_{\rm in}$ and $\tau_{\rm so}$, obtained from the fitting, fall in the range of measured values of Gershenzon *et al* and the third one, M, obeys a systematic variation, we do not naively take this to mean that our theory is 'the theory' for the experiments considered above. Instead, we should be cautious and take the agreement to mean that our theory only offers plausible explanation, in light of the following. (A) There are uncertainties in the magnitude of MR, both in the experiments and in the theory. Recall that the theory contains a multiplicative factor M, and the experimental MR value depends on the growth conditions of systems. The uncertainties make it difficult at the present stage to perform a rigorous comparison in magnitudes. (B) The fitting of theory to experiments contains three free parameters, $\tau_{\rm in}$ and $\tau_{\rm so}$ and M, which may be too many. (C) Other MR mechanisms, e.g., those considered in [2] and [8]–[10], could also be present but are not taken into account in our comparison. Because of (A)–(C), we must be conservative about the validity of the comparison. It will take further theoretical and experimental studies to close these loopholes.

4. Conclusion

In conclusion, we have presented a BMR calculation for granular systems in the hopping regime. We note that FMR theories and the present BMR calculation apply to different regimes. With a large metal/insulator volume ratio, MR phenomena, reminiscent of that in the weak-localization regime and studied in this work, can prevail. As the volume ratio approaches zero, we have $R_{\rm g} \to 0$, implying $t_{\rm H} \to \infty$ and $G \to 0$. Then, the BMR described by equation (8) becomes negligible. In this limit, the charging energy is large and favours long-range hopping. The energy level spacing in each grain also becomes large with the one nearest to the Fermi level dominant in a hopping process. The situation is then back to that of the impurity-doped semiconductors and FMR dominates. We hope that this work calls attention to the importance of backscattering in granular systems with large metal/insulator volume ratios, previously neglected by most groups.

Moreover, we remark that the study of metal particles, as a random system, has a long history. Important foundations were laid down by R Kubo [20] and by L P Gorkov and G M Elliashberg [21]. This is still an on-going field, with a view to the understanding of random zero-dimensional many-electron systems. Our study derives a formula which permits experimentalists to extract important quantities such as various scattering times from MR measurement, for these systems. We think that these quantities, if measured systematically, will aid future studies of metal particles in a significant way.

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