Negative Magnetoresistance in the Nearest-neighbor Hopping Conduction

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Abstract

We propose a size effect which leads to the negative magnetoresistance in granular metal-insulator materials in which the hopping between two nearest neighbor clusters is the main transport mechanism. We show that the hopping probability increases with magnetic field. This is originated from the level crossing in a few-electron cluster. Thus, the overlap of electronic states of two neighboring clusters increases, and the negative magnetoresistance is resulted.
The magnetoresistance in various transport mechanisms has been the subject of many studies. For band conduction where electrons move from one place to another through diffusion, it is textbook knowledge that the resistance will increase with the strength of a strong magnetic field because electrons can be deflected by a Lorentz force. However, the negative magnetoresistance has been observed and explained in many systems in the past two decades. The negative magnetoresistance in dirty metals is related to weak localization phenomena. The negative magnetoresistance in the Mott’s variable-range hopping (VRH) region is also possible though the backscattering is unimportant there because electron states are highly localized and probability of backscattering is exponentially small. In the VRH case, it was pointed out that the average should be on the logarithm of the conductance so that the interference of forward tunneling paths is important. Replica treatment shows that the pairing of the tunneling paths is important to the average. A magnetic field can introduce phases to the tunneling paths. Thus, the pairing is weaken, the localization length increases, and the negative magnetoresistance is resulted. Recently, there is a report that the negative magnetoresistance was observed in Al/Al$_2$O$_3$ and gold granular materials in which the nearest neighbor hopping is the main transport mechanism. There are several interesting features in the observations. The negative magnetoresistance occurs only in a system with large metal grains and near the percolation threshold. It is unlikely that these experimental results can be explained either by the mechanism for the band conduction or by that for the VRH conduction. In this letter, we present a theory for the negative magnetoresistance in the nearest neighbor hopping conduction in granular materials.

For electron tunneling in a homogeneous material under a nonrandom potential, introduction of a magnetic field leads usually to a positive magnetoresistance. This is due to both the destructive quantum interference between various tunneling paths and that a strong magnetic field can shrink a wavefunction through the magnetic confinement. The calculation on a hydrogen-like state shows that a magnetic field can dramatically modify the asymptotic behavior of a wavefunction. A gigantic positive magnetoresistance in a doped semiconductor is explained by this shrinkage. However, a granular metal-insulator material with the nearest neighbor hopping conduction may behave differently in a magnetic field when the size of metallic grains are large enough. A magnetic field can interact with electron orbital motion. When the interaction is comparable with the typical level spacing (which is inverse proportional to the square of the grain size), a magnetic field may induce level crossing. The level crossing may profoundly affect the transport properties of granular materials. Consider two identical metallic grains 1 and 2, separated by a distance such that electrons move from one grain to another mainly by hopping. Let $\psi_1$ and $\psi_2$ be two states of each grain. Suppose that $\psi_1$ is right at the Fermi level while $\psi_2$ is just above the Fermi level in the absence of a magnetic field. Thus, the size of $\psi_2$ is, in general, larger than that of $\psi_1$. The electron hopping probability depends on the overlap of $\psi_1$ of the two grains. Increasing the strength of the magnetic field might cause $\psi_2$ to be at the Fermi energy, and $\psi_1$ to be above the Fermi level. In this case, the electron hopping probability will be dominated by the overlap of $\psi_2$ of the two grains. Therefore, the tunneling probability increases in the magnetic field, and the negative magnetoresistance in the nearest hopping conduction should be observed. In order to test the validity of the picture mentioned above, we study a simple exact solvable model for two-dimensional granular metal-insulator systems. We will demonstrate that the size of the wavefunction at the Fermi level of a metallic cluster goes
The eigenfunction and eigenenergies are several micrometers, \( \omega \) readily solved in a uniform magnetic field, \( B \), with symmetric gauge \( \vec{A} \) with the field. It is easy to see that the first crossing occurs between state of \( n \) or \( N \) electrons reside in the potential. The larger \( \omega_0 \) is, the smaller the metallic grain size will be. It should be pointed out that, when one compares with a real experiment, the order of \( \omega_0 \) can be chosen in such way that \( < r^2 > = N \hbar/(m \omega_0) \) is roughly equal to grain sizes, where \( N \) is a quantity determined by the number of electrons on one grain. For a grain of several micrometers, \( \omega_0 \) is order of \( 10^{12} \text{Hz} \). The Schrodinger equation corresponding to (1) is readily solved in a uniform magnetic field, \( B \), with symmetric gauge \( \vec{A} = (-B y/2, B x/2, 0) \). The eigenfunction and eigenenergies are

\[
\psi_{n,l}(r, \theta) = \frac{1}{\sqrt{2\pi}} e^{-i \theta} (\alpha r)^{|l|} e^{-\frac{1}{2}\alpha^2 r^2} F(-n, |l| + 1, \alpha^2 r^2) \tag{2}
\]

and

\[
E_{n,l} = (2n + |l| + 1)\hbar \omega - l\hbar \omega_L, \tag{3}
\]

where \( F(a, b, x) \) is the confluent hypergeometric function, \( \alpha = \sqrt{m \omega/\hbar} \), \( \omega^2 = \omega_0^2 + \omega_L^2 \), \( n = 0, 1, 2, \ldots \), and \( l = 0, \pm 1, \pm 2, \ldots \) with \( \omega_L = Be/(2mc) \). In the absence of a magnetic field, \( \omega_L = 0 \), \( E_{n,l} \) is \( 2n + |l| + 1 \) fold degenerated. The degeneracies are broken due to the second term in (3) which is from the magnetic-field-orbital interaction. This is the term which will be responsible to the level crossing. The size of wavefunction with quantum numbers \( n \) and \( l \) can be found to be

\[
< r^2 > = \frac{\hbar}{m \omega}(2n + |l| + 1). \tag{4}
\]

For a fixed \( 2n + |l| \), the sizes \( < r^2 > \) of these wavefunctions are the same though their energies may be different, and \( < r^2 > \) will shrink in a magnetic field because \( \omega \) increases with the field.

With a proper strength of a magnetic field, the energy of a state with a large \( 2n + |l| \), say \( N + 1 \), can be smaller than that with \( 2n + |l| = N \) because of the second term in equation (3). It is easy to see that the first crossing occurs between state of \( (n = N + 1, l = N + 1) \) and state of \( (n = N, l = -N) \) when \( (N + 1)\hbar \omega - (N + 1)\hbar \omega_L = N \hbar \omega + N \hbar \omega_L \), i.e., \( (2N + 1)\hbar \omega_L = \hbar \omega \), or \( B = (m \omega_0)/(e\sqrt{N(N + 1)}) \). Similarly, the energy level of at least one of states with \( 2n + |l| = N + 2 \) is below some of \( 2n + |l| = N \) states when \( B > (2m \omega_0)/(e\sqrt{N(N + 2)}) \). In general, at least one of \( 2n + |l| = N + k \) states is below some of \( 2n + |l| = N \) states when \( B > (km \omega_0)/(e\sqrt{N(N + k)}) \). To see how the size of the wavefunction depends on the magnetic field. Let us assume that all \( 2n + |l| = N \) states are occupied while higher states are empty in the absence of a magnetic field. Then it can be shown that the highest occupied state corresponds to \( 2n + |l| = N + 1 \) when \( B > m \omega_0/(e\sqrt{N(N + 1)}) \). According
to (4), \( < r^2 > \) will jump from \((N + 1)\hbar/(m\omega)\) to \((N + 2)\hbar/(m\omega)\). Increasing the magnetic field further to \( B > 3mc\omega_0/(e\sqrt{N(N + 3)}), \) \( < r^2 > \) will jump to \((N + 3)\hbar/(m\omega)\). In general, \( < r^2 > \) will jump to \((N + k)\hbar/(m\omega)\) when \( B \) is approximately larger than value \((k^2 + 2k + 2Nk - N)mc\omega_0/(e\sqrt{(N^2 + 3N)(N^2 + 2kN + 2N + k^2 + 2k)})\). The size \( < r^2 > \) between two jumps decreases with the magnetic field strength because of the magnetic confinement, but the overall trend of \( < r^2 > \) is increasing with the magnetic field. However, \( < r^2 > \) can also decrease when the field is very strong \((\omega_L >> \omega_0)\). The increase in \( < r^2 > \) implies the larger overlap in the wavefunction of two hopping states if the functional form of the wavefunction remains the same. Thus, an overall negative magnetoresistance is expected.

In order to be more precise, let us calculate the tunneling matrix element \( t_{12} \) between two states, \( \psi_1 \) and \( \psi_2 \), of two metallic grains separated by a distance \( d \). When an electron tunnels from an initially occupied state, say \( \psi_1 \), to the empty state, \( \psi_2 \), it will contribute to the hopping probability \( P \) (per unit time). The contribution will be proportional to \( |t_{12}|^2 \exp(-\Delta\epsilon_{12}/(KT)) \), where \( \Delta\epsilon_{12} \) describes the relative energy level with respect to the Fermi energy.\(^3\) The hopping conduction can be regarded as an electron diffusion process in which an electron undergoes a Brownian motion from one cluster to another, and the diffusion constant \( D \) relates to \( P \) as \( D = Pd^2 \), where \( d \) should be regarded as the average distance between two neighboring clusters. According to the Einstein relation, the electron mobility \( \mu \) is given by \( \mu = eD/(KT) \) which is related to the conductivity in the conventional way.\(^4\) Therefore, we can concentrate on how the tunneling matrix element near the Fermi energy depends on the magnetic field in order to study the magnetoresistance of the system. Let the centers of two clusters be at \((-d/2, 0)\) and \((d/2, 0)\), respectively. Assume that an electron tunnels from state \( \psi_1 \) of the left cluster to state \( \psi_2 \) of the right cluster, in the tight-binding approximation, the tunneling matrix element \( t_{12} \) is given by

\[
t_{12} = \frac{\hbar^2}{m} \int_{-\infty}^{\infty} \left[ (\psi_1^* \frac{\partial \psi_2}{\partial x} - \psi_2 \frac{\partial \psi_1^*}{\partial x}) - \frac{2i}{\phi_0} (\vec{A} \cdot \hat{x}) \psi_1^* \psi_2 \right]_x=0 dy,
\]

where \( \phi_0 = ch/e \) is the flux quanta, and \( \vec{A} \) is the vector potential. For small \( \vec{A} \), the second term will be small in comparison with the first term. Then equation (5) can be simplified to

\[
t_{12} = \frac{\hbar^2}{m} \int_{-\infty}^{\infty} \left[ \psi_1^* \frac{\partial \psi_2}{\partial x} - \psi_2 \frac{\partial \psi_1^*}{\partial x} \right]_x=0 dy.
\]

It is easy to show that \( \psi_1 \) and \( \psi_2 \) with quantum number \((n, l)\) can be expressed by solution (5), in the symmetric gauge \( \vec{A} = (-By/2, Bx/2, 0) \), as

\[
\psi_1 = \exp(i \frac{e}{\hbar c} \vec{A}_0 \cdot \hat{r}) \psi_{n,l}(r_1, \theta_1),
\]

\[
\psi_2 = \exp(-i \frac{e}{\hbar c} \vec{A}_0 \cdot \hat{r}) \psi_{n,l}(r_2, \theta_2),
\]

where \( \vec{A}_0 = Bd\hat{y}/4 \), \( r_1 \) and \( \theta_1 \) are the polar coordinates of \((r \cos(\theta) - d/2, r \sin(\theta))\), and \( r_2 \) and \( \theta_2 \) are the polar coordinates of \((r \cos(\theta) + d/2, r \sin(\theta))\). The phase factors in equations
and (8) are due to the magnetic field. They will give the usual interference on the tunneling matrix element.

At a low temperature, tunnelings between states close to the Fermi energy dominate the electron transport. For the simplicity, we will consider magnetic field dependence of the tunneling matrix element between two highest occupied states of two identical clusters. Therefore, the wavefunction should be replaced by a new one in calculating $t_{12}$ whenever the level crossing occurs at the Fermi level. There are two possible cases, two clusters with small and large separations. In the limit of large separation between grains, that is $d \gg \sqrt{<r^2>}$, the shrinkage of wavefunction due to the magnetic confinement dominates over jumps in quantum number $N$. Figure 1a is the semilog plot of magnetic field dependence of the hopping coefficient for $d = 600l_0$, and $N = 100$ when $B = 0$. $t_{lr}$ is in the unit of $\hbar^2/(2ml_0^2)$, and magnetic field is in the unit of $B_0$. Overall, $t_{lr}$ decreases with the magnetic field as expected. However, in the region of $d$ to be several $2\sqrt{<r^2>}$, the jumps in quantum number $N$ dominate, and $t_{lr}$ behaves similarly as that of $<r^2>$. Figure 1b is the semilog plot of the magnetic field dependence of the hopping coefficient for $d = 60l_0$, and $N = 100$ when $B = 0$. This may explain why the negative magnetoresistance in the nearest-neighbor-hopping region has been observed only near the percolation threshold point 7. It may be interesting to point out that the transport mechanism is mainly through band conduction when $d \approx 2\sqrt{<r^2>}$ because the overlap integrals between two clusters are large, and eigenstates in each cluster are broaded into energy bands. A rapid oscillation of $t_{lr}$ on $B$ is due to the oscillations of wavefunctions in the overlap region. Of course, the validity of our formula of tunneling matrix element is in question in this case.

Before summary, we address some important issues. The physics of magneto-transport of granular metal-insulator materials is rich in the nearest neighbor hopping conduction. Depending on the size of metallic grains and the distance between two adjacent grains, both positive magneto-resistance and negative magneto-resistances are possible. When the inter-distance of grains is very large, the magnetic confinement dominates and an overall positive magnetoresistance is expected. On the other hand, if level crossing dominates, the magnetoresistance is negative which happens when the potential barrier of two adjacent grains is small. Therefore, the negative magnetoresistance of a granular metal-insulator material should be expected only near the percolation threshold point. Unlike the weak localization effect for a dirty metal where the resistance can change slightly in a weak magnetic field, this proposed mechanism could greatly change the resistance if conditions are right. Another possible mechanism for negative magnetoresistance which is not addressed in this paper is as follows: A magnetic field may also increase the Fermi energy of a grain such that electrons need to tunnel through a lower potential barrier. In this work, we point out that the level crossings can come from the magnetic-field-orbital interaction. The level crossings occur when this interaction is comparable with the level spacing of the system. Since level spacing is proportional to the $\hbar^2/(2mr^2)$, where $r$ is the grain size, the field require to induce a level crossing depends on the grain size. In this model, for grain size of nanometers, the typical field for level crossing is of order of $10T$. Therefore, it is better to use material with grain size of tens nanometers to observe such jumps. Although we only studied a specific 2D model, but the physics discussed in this paper is expected to carry over to other 2D models, as well as 3D models. In particular, we use a simple harmonic oscillator metallic grain model to show that a magnetic field can induce the level crossings such that the highest
occupied state in a metallic grain has a larger size in a magnetic field than that without a magnetic field. This may lead to a series of jumps in the hopping coefficient as one increases the magnetic field. Such jumps might have already been observed in a real experiment. Of course, our analysis is zero temperature. In a real experiment with a finite temperature, these jumps are expected to be smoothed out. However, at a high temperature, the level crossing may become unimportant because a lot of states can participate in the hopping conduction. In such case, magnetic confinement effects may dominates. We have neglected Coulomb interaction in this work. The Coulomb energy is order of $\epsilon_c \sim \frac{e^2}{\kappa r}$ for a grain of size $r$, where $\kappa$ is the permittivity. The Coulomb interaction should be important when it is comparable with thermal energy $kT$, or level spacing. It is known that Coulomb interaction can have many important effects on the transport properties of quantum dots and granular metallic systems, such as Coulomb blockade and the interesting I-V characteristic in granular metallic systems. Therefore, it is important to consider both the Coulomb interaction and temperature effects in order to make a detail analysis of real experiment results. Further studies are needed on the interplay of the level crossing, Coulomb interaction, and the temperature effects.

In summary, we propose a level crossing effect which leads to negative magnetoresistance in granular materials where nearest neighbor hopping is the main transport mechanism.

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FIGURES

FIG. 1. The magnetic field dependence of the hopping coefficient between highest occupied states in two adjacent grains. The magnetic field is in the unit of $B_0 = mc_0/e$, and $< r^2 >$ in the figure is in the unit of $l_0^2 = \hbar/(m_0 \omega_0)$. (a) The semilog plot of $t_{tr}$ vs. $B$ for the case of $N = 2n + |l| = 100$ at $B = 0$ and $d = 600l_0$. $t_{tr}$ is dominated by the wavefunction shrinkage due to magnetic confinement. An overall positive magnetoresistance is observed. (b) The semilog plot of $t_{tr}$ vs. $B$ for the case of $N = 2n + |l| = 100$ at $B = 0$ and $d = 60l_0$. The magnetic field dependence of the hopping coefficient is dominated by the jumps in $< r^2 >$ in this case, and an overall negative magnetoresistance is resulted.