Giant magnetoresistance in magnetic granular alloys

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Numerical simulation on resistance and giant magnetoresistance in magnetic granular alloys is performed for finite-size systems by making use of the real-space Green's-function method based on the Kubo formalism. Spin-dependent scattering causes the giant magnetoresistance in the magnetic granular alloys as in the magnetic multilayers. It is shown by using the microscopic theory that the magnetoresistance increases with decreasing the size of magnetic grains, in agreement with experiment. The theoretical results indicate that scattering at the surfaces of magnetic grains governs the resistance and magnetoresistance in granular alloys. Difference between the giant magnetoresistances in the granular alloys and multilayers is discussed.

The giant magnetoresistance (GMR) in magnetic multilayers[1] has stimulated a great deal of investigation of the magnetotransport properties in the multilayered structures. The electrical resistance decreases with the reorientation of magnetic moments in magnetic layers from an antiferromagnetic to ferromagnetic alignment by an external magnetic field. Spin-dependent scattering at the interfaces between layers has been pointed out to be responsible for GMR in the multilayers.[1,2] Recent observation of GMR in magnetic/nonmagnetic granular alloy films of Co/Cu,[3,4] Co/Ag,[5,6] Fe/Ag, and Fe/Cu (Ref. 7) has further developed a new field of magnetotransport phenomena in magnetic films. This phenomenon has been observed even in melt spun ribbons.[8] GMR is a new phenomenon brought by an interplay of the charge and spin degrees of freedom of electrons in heterogeneous systems. In the granular alloys, the geometry or size of the scatterers is found to play an important role in GMR by analyzing the experimental data as described below. Fundamental study of GMR in the granular alloys is significant not only to clarify the origin and mechanism of GMR but to develop a new research field of spin-dependent quantum transport in metallic materials with nano-scale heterogeneity as well as to provide useful information for technical applications.

Magnetic atoms in the granular alloys form grains in nonmagnetic matrices. Each grain has a magnetic moment and its direction is random in the absence of the external magnetic field. When the magnetic field aligns the magnetic moments of the grains ferromagnetically, the resistance decreases as in the multilayers. Furthermore, GMR in the granular systems may be also due to the spin-dependent scattering. GMR in the granular alloys depends on the annealing temperature T_A. The magnitude of GMR increases with decreasing T_A. Below $T_A \sim 200\,^{\circ}\mathrm{C}$, however, GMR shows a tendency to decrease with decreasing T_A, which can be attributed to magnetic coupling between the magnetic grains. Slow saturation of the resistivity with increasing the magnetic field supports this interpretation. The experiments of transmission electron micrograph[5] and small-angle x-ray scattering[9] have revealed that the average radius r_C of grains decreases with decreasing T_A. From these experimental results, we may deduce that GMR increases with decreasing r_C.

So far, only a phenomenological model has been used for an explanation of GMR in the granular alloys.[10] The aim of the present study is, by using a microscopic model, to clarify what governs the essential ingredient of GMR in the granular alloys, that is, the increase of GMR with decreasing r_C. We put an emphasis on the effects of finite size of scatters (magnetic grains) and spin-dependent potential on the resistivity and GMR. We perform numerical calculations of the conductance of the granular alloys at zero temperature based on the Kubo formalism by using the recursion method.[11,12] In this method, the conductance is obtained exactly without any approximation although the system has finite size. We will show that the resistivity and the GMR in the granular alloys are governed by the scattering at surfaces of magnetic grains and increase with increasing the surface-to-volume ratio of magnetic grains.

Let us consider the tight-binding Hamiltonian on a simple cubic lattice,

$$H=-t\sum_{\langle i,j\rangle,\alpha} c_{i,\alpha}^{+}c_{j,\alpha}+\sum_{i\in\text{matrix},\alpha}\varepsilon_B c_{i,\alpha}^{+}c_{i,\alpha} + \sum_{i\in\text{cluster},\alpha,\beta}(\varepsilon_A c_{i,\alpha}^{+}c_{i,\alpha}\delta_{\alpha\beta}-\mathcal{M}c_{i,\alpha}^{+}\sigma_{\alpha,\beta}c_{i,\beta}) . \quad (1)$$

Here i and j label the sites, $c_{i,\alpha}^{+}(c_{i,\alpha})$ is the creation (annihilation) operator of an electron with spin α at site i, t denotes the transfer integral, σ's are the Pauli matrices, ε_A and ε_B are the on-site potentials in the magnetic atoms and the nonmagnetic atoms, respectively, and $\mathcal{M}$ is the exchange potential in the magnetic atoms. The summation $\langle i,j\rangle$ runs over the nearest-neighbor sites.

We introduce a relation

$$\varepsilon_A+|\mathcal{M}|\approx\varepsilon_B . \quad (2)$$

Due to this relation, electrons in the minority spin band are less scattered by the magnetic atoms which form grains than those in the majority spin band when the magnetic moments of the grains are aligned ferromagnet-

ically. It has been pointed out that in Fe/Cr multilayers,[2,13] a relation equivalent to Eq. (2) holds. The characteristic features of the parallel and perpendicular magnetoresistance in the multilayers have been successfully explained[14,15] by using the relation. The spin-dependent scattering may depend on the details of electron structure of the systems. However, the essence of the asymmetry of the spin-dependent scattering is included in Eq. (2).

In the calculation, we take a model system as shown in Fig. 1. The lengths of a sample in the directions parallel and normal to the current are chosen as $L=60$ and $M=12$, respectively, in units of the lattice constant. Two perfect lead wires with semi-infinite length are attached to both sides of the sample. In the lead wires, the on-site potential and the exchange potential are chosen to be zero. In the present formalism, the conductance is calculated exactly by using real-space Green's functions which are expressed $M\times M$ matrices for the present system and obtained recursively. We prepare several types of magnetic clusters which model the grains as shown in Table I. The conductance is first calculated in a sample where magnetic clusters of one type are randomly distributed, and then averaged over 20 different random distributions of the clusters and the directions of the magnetization of the clusters. We assume that each cluster has magnetization.

Let us first examine the resistance (inverse of the conductance) as a function of R_{SV} for several concentrations of magnetic atoms, where R_{SV} is a measure of the surface-to-volume ratio of clusters and is defined as follows. Each cluster is connected with the matrix by bonds. The number of such bonds N_S and the number of the atoms for each cluster are given in Table I. We take N_S and the number of atoms of the cluster as the area of the surface and the volume of the cluster, respectively, and define R_{SV} as the ratio of N_S to the number of the atoms in each cluster. R_{SV} smoothly links to the ratio $4\pi r_C^2/(4\pi r_C^3/3)=3/r_C$ of large clusters with radius r_C. Note that, in a large cluster with the simple cubic lattice, the unit area of the surface corresponds to one bond connecting the cluster with the matrix. In the 27-site cluster, $3/r_C$ is given to be 1.6 when we estimate its radius by the equation $4\pi r_C^3/3=27$.

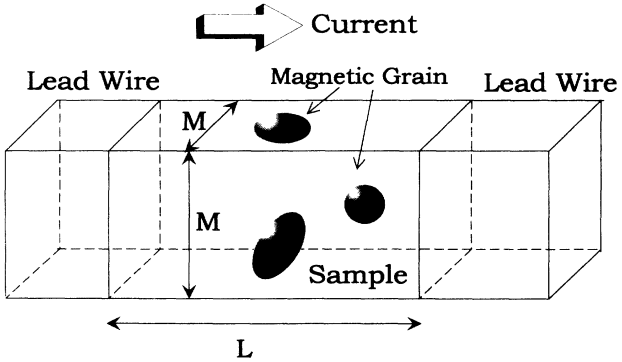

FIG. 1. Geometry of systems used in the calculation, L and M are the lengths of the sample in the directions parallel and normal to the current, respectively. Two infinitely long perfect lead wires are attached to the both sides of the sample.

TABLE I. Magnetic clusters used in the calculation. Volume is the number of atoms of a cluster and N_S denotes the number of bonds which connect each cluster with the matrix, R_{SV} is the ratio of N_S to volume.

Cluster	•				
Volume	1 site	7 site	8 site	19 site	27 site
N_S	6	30	24	54	54
R_{SV}	6.0	4.3	3.0	2.8	2.0

Calculated results of the resistance in the systems where the direction of the magnetic moments of clusters is distributed randomly (**R** configuration) and those in the ferromagnetic (**F**) configuration are shown in units of h/e^2 in Figs. 2(a) and 2(b), respectively. The systems in the **R** and **F** configurations correspond to the granular alloys in the absence of the magnetic field and in the saturation field H_S, respectively. The potentials of the cluster and the matrix are chosen to satisfy the relation Eq. (2) as $\varepsilon_A=-1.0$, $\varepsilon_B=0.0$, and $|\mathcal{M}|=1.0$, and the Fermi energy E_F is taken to be zero, where the energy is measured in units of the transfer integral t. Comparing with the band width (i.e., 12.0), the difference in the potentials, $|\varepsilon_A-\varepsilon_B|$, is comparable to that of the actual materials.[2]

The resistance in the **R** configuration increases with increasing R_{SV} for fixed concentration. The resistance in Fig. 2(a) is replotted in the inset of Fig. 2 as a function of the total number of the bonds which connect the clusters with the matrices in alloys ($N_{AB}\equiv N_S\times$number of clusters in an alloy). It can be seen that the resistance is well scaled by N_{AB}, irrespective of a variety of the cluster size and the concentration.[16] The results in the inset indicate that the surface-to-volume ratio of the cluster R_{SV} characterizes the resistance in the granular alloys. This is because the Fermi wavelength ($1/k_F$) is of the order of

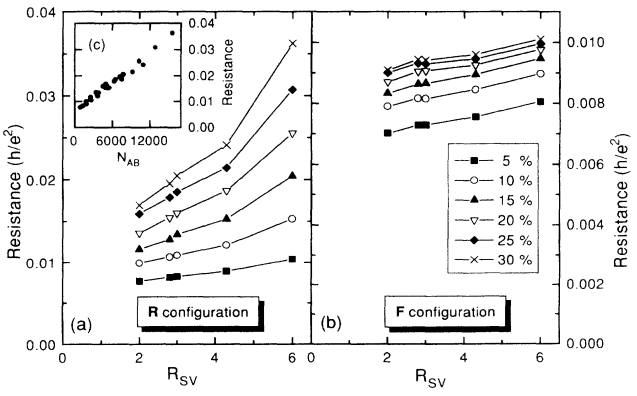

FIG. 2. Calculated results of the resistance versus the surface-to-volume ratio of cluster R_{SV} in the random magnetic (**R**) and ferromagnetic (**F**) configurations for several concentrations of the magnetic atoms In the inset, the resistance in the **R** configuration is replotted as a function of the total number of the bond N_{AB} (see text).

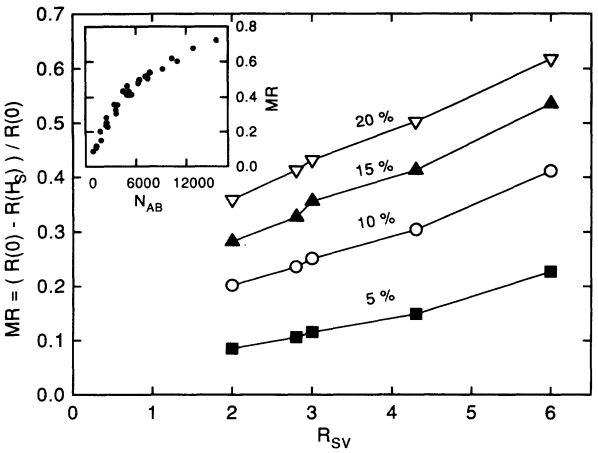

FIG. 3. Calculated results of the magnetoresistance vs the surface-to-volume ratio of clusters R_{SV} for 5, 10, 15, and 20% concentrations of the magnetic atoms. The inset shows N_{AB} dependence of magnetoresistance.

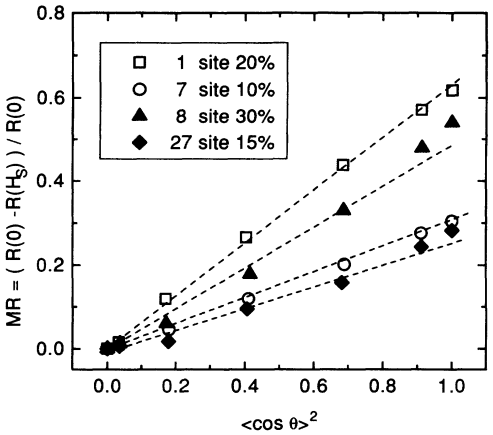

FIG. 4. Calculated results of the magnetoresistance as a function of $\langle\cos\theta\rangle^2$. Dashed lines are a guide for the eyes.

the lattice constant and the resistance is determined by the scattering within a range of $1/k_F$. The resistance is well correlated with N_{AB} when E_F is around the band center. When E_F is close to the band edge and $1/k_F$ is larger than the lattice constant, the correlation tends to be weak. Note that the contact between sample and lead wires causes the resistance even if there is no cluster in a sample. The contact resistance may weaken the magnitude of GMR discussed below. The dependence of GMR on cluster size, however, can well be studied since the contact resistance is constant for all samples and small enough as compared with the resistance due to magnetic clusters. The resistance in the **F** configuration also increases with increasing R_{SV} and is much smaller than that in the **R** configuration. This is due to the fact that electrons in the minority spin band are not scattered by the clusters in the **F** configuration. In both **R** and **F** configurations, the resistance increases with decreasing the grain size, in agreement with experiment.[5]

We define the magnetoresistance by the equation $\mathrm{MR}=\{R(0)-R(H_S)\}/R(0)$, where $R(0)$ and $R(H_S)$ are the resistance in the **R** and **F** configurations, respectively. The magnetoresistance for several concentrations is shown as a function of R_{SV} in Fig. 3. It increases with increasing R_{SV} for all concentrations. In the inset, MR in Fig. 3 is replotted as a function of N_{AB}. The MR is scaled by N_{AB}. This is because the resistance is scaled by N_{AB} as shown in the inset of Fig. 2. Thus, we find that the magnetoresistance in the granular alloys increases with the surface-to-volume ratio of magnetic clusters. The results in Fig. 3 agree with experiment.[7,8]

The surface-to-volume ratio of grains governs GMR in the granular alloys. In order to compare the GMR in the granular alloys with the parallel GMR in the multilayers, let us consider a rectangular lattice with dimensions $l\times l\times d$ as a magnetic layer in the multilayers, where d is the thickness of the layer. The surface-to-volume ratio of the layer is roughly estimated to be $2l^2/dl^2=2/d\leq 2.0$ in the limit of $l\gg 1$. On the other hand, the ratio of clusters in the granular alloys can be larger than 2.0 as seen in Table I. Therefore, we find that the granular alloys will exhibit larger magnetoresistance than the multilayers. Note that the perpendicular GMR in the multilayers cannot be compared with that in the granular alloys, since the miniband structure due to the periodicity also causes the perpendicular GMR.[15]

The relation between the magnetoresistance and the magnetization of the system, $\mathrm{MR}\propto(\text{magnetization})^2\propto\langle\cos\theta\rangle^2$, has been observed,[3] where θ is the relative angle between the magnetization axis of the grains and the external field, and $\langle\cdots\rangle$ denotes the average over the magnetic grains. In order to reveal the relation, we assume that θ is randomly distributed in the range $0\leq\theta\leq\theta_0$ and calculate the magnetoresistance and $\langle\cos\theta\rangle$ independently. θ_0 is chosen to be π and zero in the **R** and **F** configurations, respectively. Figure 4 shows the relation between the magnetoresistance and $\langle\cos\theta\rangle^2$ for several choices of the concentration of magnetic atoms and the types of clusters. The magnetoresistance is proportional to $\langle\cos\theta\rangle^2$, consistent with the experimental results. The physical reason is as follows. When the quantum axis of each cluster is taken along θ direction, the up- and down-spin states mix and the average transition probability changes like $\langle\cos^2(\theta/2)\rangle$ if the interference between clusters is neglected. Using the two-current model, we obtain that $\mathrm{MR}\propto\langle\cos\theta\rangle^2$. The result in Fig. 4 that MR is almost linearly proportional to $\langle\cos\theta\rangle^2$ may indicate that each magnetic cluster contributes to MR almost independently.

As mentioned, the scattering of electrons is governed by a range of $1/k_F$ which is of the order of the lattice constant in the present calculations. Because the diameter of the large clusters used is larger than $1/k_F$, the qualitative tendency would be unchanged even for larger clusters and the present results can well reproduce the essential tendency of GMR observed. Furthermore, we have used symmetrical clusters since the grains in real materials are more or less spherical in shape. The dependence of the resistivity on the shape of the clusters, if controlled experimentally, is another interesting issue, a study of which is in progress.

In conclusion, it has been shown that the spin-

dependent scattering causes the GMR in the granular alloys as well as in the multilayers and that the scattering at surface of magnetic grains governs the resistivity and GMR in the granular alloys. The results explain the observed tendency that the resistivity and magnetoresistance increase with decreasing the size of magnetic grains. Further study of the transport phenomena in heterogeneous materials is encouraged.

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[1]M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).

[2]J. Inoue, A. Oguri, and S. Maekawa, J. Phys. Soc. Jpn. **60**, 376 (1991).

[3]John Q. Xiao, J. Samuel Jiang, and C. L. Chien, Phys. Rev. Lett. **68**, 3749 (1992).

[4]A. E. Berkowitz, J. R. Mitchell, M. J. Carey, A. P. Young, S. Zhang, F. E. Spada, F. T. Parker, A. Hutten, and G. Thomas, Phys. Rev. Lett. **68**, 3745 (1992).

[5]P. Xiong, G. Xiao, J. Q. Qang, J. Q. Xiao, J. S. Jiang, and C. L. Chien, Phys. Rev. Lett. **69**, 3220 (1992).

[6]J. A. Barnard, A. Waknis, M. Tan, E. Haftek, M. R. Parker, and M. L. Watson, J. Magn. Magn. Mater. **114**, L230 (1992).

[7]J. Q. Wang, P. Xiong, and G. Xiao, Phys. Rev. B **47**, 8341 (1993).

[8]B. Dieny, A. Chamberod, J. B. Genin, B. Rodmacq, S. R. Teixeira, S. Auffret, P. Gerard, O. Redon, J. Pierre, R. Ferrer, and B. Barbara, J. Magn. Magn. Mater. **126**, 433 (1993).

[9]T. A. Rabedeau, M. F. Toney, R. F. Marks, S. S. P. Parkin, R. F. C. Farrow, and G. R. Harp, Phys. Rev. B **48**, 16 810 (1993).

[10]S. Zhang, Appl. Phys. Lett. **61**, 1855 (1992); S. Zhang and P. M. Levy, J. Appl. Phys. **73**, 5315 (1993).

[11]P. A. Lee and D. S. Fisher, Phys. Rev. Lett. **47**, 882 (1981).

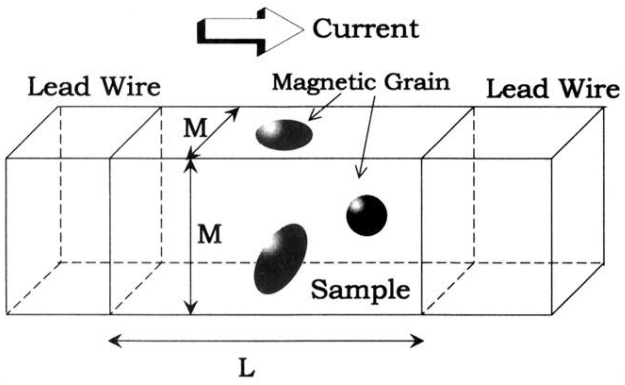
[12]D. S. Fisher and P. A. Lee, Phys. Rev. B **23**, 6851 (1981).

[13]H. Itoh, J. Inoue, and S. Maekawa, Phys. Rev. B **47**, 5809 (1993).

[14]A. Oguri, Y. Asano, and S. Maekawa, J. Phys. Soc. Jpn. **61**, 2652 (1992).

[15]Y. Asano, A. Oguri, and S. Maekawa, Phys. Rev. B **48**, 6192 (1993).

[16]Numerical errors occur due to cluster touching for large clusters in high concentrations, but they are estimated to be less than 10% even for the highest concentration studied.



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