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Neutron Diffraction Studies on a Non-Coupled Giant Magnetoresistance System, Co/Cu/NiFe/Cu Multilayer

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By polarized neutron diffraction, a long range antiparallel alignment of Co and NiFe(permalloy) layer magnetizations is confirmed in a non-coupled Co(10 Å)/Cu(36 Å)/NiFe(10 Å)/Cu(36 Å) giant magnetoresistance system. A general relationship between the magnetoresistance and the magnetization vectors of the Co and NiFe layers under various magnetic fields and temperatures is experimentally derived. The observed large temperature change in the magnetoresistance ratio is understood with this relationship.

[neutron diffraction, giant magnetoresistance, non-coupled giant magnetoresistance system, multilayer, antiparallel alignment of magnetizations, magnetization process, coercive field]

§1. Introduction

Multilayer films consisting of Co/Cu/NiFe (permalloy)/Cu layers indicate the giant magnetoresistance (GMR) effect.[1)] One of the necessary conditions for the GMR effect is an antiparallel alignment of adjacent magnetizations which are separated by a non-magnetic spacer layer. Up to now, two mechanisms are known to realize the antiparallel alignment. One is an oscillatory indirect exchange coupling between two magnetic layers. Typical examples are Fe/Cr[2)] and Co/Cu[3,4)] multilayers. The other is a difference in the coercive fields of two magnetic layers. The GMR effect of Co/Cu/NiFe/Cu multilayers is considered to occur mainly with this mechanism,[5)] though a contribution of the indirect exchange coupling was reported in some Co/Cu/NiFe/Cu multilayers with thin Cu layers.[6)] Hereafter we will concentrate on the Co(10 Å)/Cu(36 Å)/NiFe(10 Å)/Cu(36 Å) multilayer in which the exchange coupling is negligible at first approximation.

The existence of the antiparallel alignment state of the Co and NiFe magnetizations is judged from the two-step features of the magnetization curves. In some of the exchange coupled GMR systems, the antiparallel long range order of the magnetization vectors is directly observed by neutron diffraction.[7-11)] In non-coupled GMR systems, it is not obvious whether the antiparallel alignment state has a long range order or not. Even if there is no long range order between the Co and NiFe magnetizations, the GMR effect may apparently be induced by magnetic multi-domain structures. Furthermore the Co/Cu/NiFe/Cu multilayer indicates a very large temperature variation of the magnetoresistance ratio $[\Delta\rho/\rho(77\ \mathrm{K})]/[\Delta\rho/\rho(300\ \mathrm{K})]\sim 3$. So far the reason was not clear. Neutron diffraction studies are desired to reveal the origin of the enhancement of the magnetoresistance in this system.

In this paper we will deal with three subjects. The first concerns with the existence of the long range order of the antiparallel alignment state of the Co and NiFe magnetizations. The second is the reason for the large temperature dependence of the magnetoresistance ratio. The third is a general relationship between the magnetoresistance and magnetic structure as a function of applied field and temperature. A brief report about the neutron diffraction

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study of this system was already published.[12)]

§2. Experimental

The sample used for the present study was made by ultrahigh vacuum deposition. The source metals were successively deposited on glass substrates at room temperature. Neutron diffraction measurements were done using a film of 15×15 mm^2 in size. The glass substrate was cut into 5×5 mm^2 square for magnetization measurements. A film strip with the size of 10×0.3 mm^2 with electrical terminals was prepared for magnetoresistance measurements. All films were prepared in the same batch. The samples have a film structure of Cr(50 Å)/[Co(10 Å)/Cu(36 Å)/NiFe(10 Å)/Cu(36 Å)] × 20. The Cr buffer layer is necessary to increase the coercive field of the Co layer and induces a well defined antiparallel alignment state of the Co and NiFe magnetizations.[13)]

Magnetization curves were measured by a SQUID magnetometer with applying magnetic field along the in-plane direction. Magnetoresistance was measured by standard four-terminal method with applying the magnetic field up to 3 kOe parallel to the film plane and perpendicular to the current direction. Neutron diffraction measurements were carried out with the TOP spectrometer installed at the BSF of National Laboratory for High Energy Physics (KEK). The time of flight method was used to record diffraction profiles. Magnetic field was applied in the film plane. The scattering vector is perpendicular to the film plane.

§3. Results and Discussion

3.1 Magnetization and magnetoresistance curves

Magnetization curves at 300 K and 77 K are shown in Fig. 1. Prior to the measurements, the magnetic field of -1.5 kOe is applied and the magnetizations of the Co and NiFe layers are saturated in the negative direction. With increasing the external field, the *M-H* curves indicate two-step features. For example at 77 K, the *M-H* curve (solid line) shows a jump between the applied field of 0 and 150 Oe, and then the curve gradually increases to the saturation. As was discussed,[1,5)] the jump is expected to be a magnetization reversal of the NiFe layer and the gradual increase corresponds to the gradual magnetic saturation process of the Co layer. Therefore the Co and NiFe magnetizations are expected to be aligned antiparalell around the applied field of 150 Oe by the difference in the coercive fields of two magnetic layers. The qualitative features of the *M-H* curve at 300 K (broken line) are similar to that at 77 K. However the two-step features are less clear at 300 K.

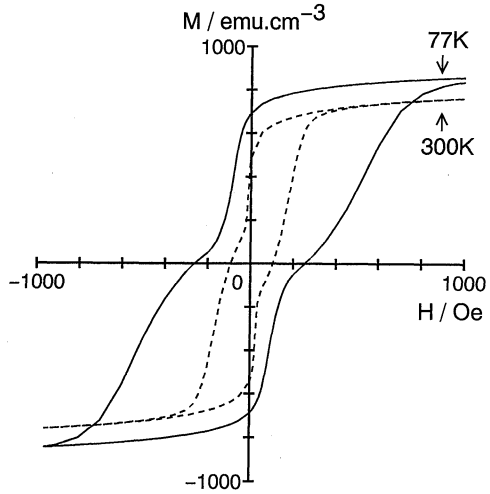

Fig. 1. Magnetization curves of the Co(10 Å)/Cu(36 Å)/NiFe(10 Å)/Cu(36 Å) multilayer film measured at 77 K (solid line) and 300 K (broken line). The magnetization is normalized to the total volume of the magnetic layers.

Magnetoresistance curves at 300 K and 77 K are indicated in Fig. 2. The curves are recorded after applying the magnetic field of -3.0 kOe. The enhancement of the electric resistivity is observed, as is expected from the magnetization measurements. The peak positions correspond to the steps observed in the *M-H* curves. Therefore the enhancement of the resistivity is thought to be the giant magnetoresistance effect, i.e. the enhancement is originated from the difference in the spin-dependent electron scatterings between the parallel and antiparallel alignment states of the magnetizations of two magnetic layers. The resistivity change $(\rho_{max}-\rho_{min})/\rho_{min}$ is 13.2% at 300 K and 41.3% at 77 K.

So far, we discussed qualitative characteristics of *M-H* and *ρ-H* curves. However if we quantitatively examine the temperature evolu-

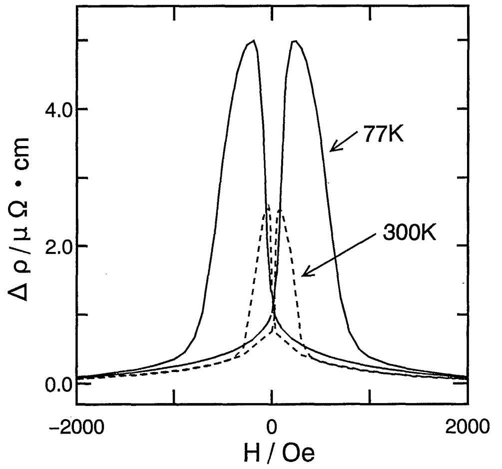

Fig. 2. Magnetoresistance curves measured at 77 K (solid line) and 300 K (broken line). The base resistivities are 12.1 $\mu\Omega\cdot$cm at 77 K and 19.3 $\mu\Omega\cdot$cm at 300 K.

tion of two curves, some disagreement is found. The temperature change in the saturation magnetization $M_s(77\text{ K})/M_s(300\text{ K})$ is 1.12, while that of the resistivity change $\Delta\rho(77\text{ K})/\Delta\rho(300\text{ K})$ is 1.91. The temperature change in the resistivity is much larger than that in the magnetization. To explain this discrepancy is important for the understanding the nature of the spin-dependent scattering. If the spin-dependent scattering occurs mainly at interface sites, the temperature dependence of interface magnetization should be considered. The temperature dependence of the interface magnetization may be larger than that of the bulk magnetization. However the difference estimated from Mössbauer spectroscopy is not so large to explain the temperature dependence of resistivity.[14]

3.2 *Neutron diffraction*

To make clear the reason for the large temperature dependence in the resistivity change $\Delta\rho$, we have used polarized neutron diffraction technique. Using this technique, the magnetization process of the Co and NiFe layer was investigated at 77, 150, 200 and 300 K. Apart from the reason for the large temperature change in $\Delta\rho$, it is significant to confirm that the antiparallel alignment state of the Co and NiFe magnetizations is a long range order, because the existence of the long range ordered antiparallel alignment state is not self-evident in non-coupled multilayers.

Neutron diffraction profiles of the first order Bragg peaks of the sample were measured with changing the applied field and temperature. First the magnetic field of −960 Oe was applied to saturate the Co and NiFe magnetizations in the negative direction, then measurements were done along the hysteresis loop. Using the polarized neutron, two intensities I_+ and I_- are obtained in every fixed temperature and magnetic field, where I_+ (I_-) is a Bragg peak intensity when the neutron polarization is parallel (antiparallel) to the applied field. For convenience, the intensity I_{sum} is defined as $I_{sum} \equiv I_+ + I_-$ and $I_{sub} \equiv (I_+ - I_-)/P_o$, where P_o is a magnitude of an incident neutron polarization. In Fig. 3, two intensities I_{sum} and I_{sub} at 77 K are plotted against the applied field. The solid lines are calculated intensities based on the followings.

To reproduce the intensities, we assume that the magnetic state of j layer is expressed with a single (averaged) magnetization vector M_j. With this assumption, the intensities can be calculated by a general method.[15] In the present case, we obtain

$$I_{sum} = G\,[I_{nuclear} + A\,(M_{Co} - M_{NiFe})^2] \tag{1}$$

$$I_{sub} = G\cdot B\,(M^z_{Co} - M^z_{NiFe}), \tag{2}$$

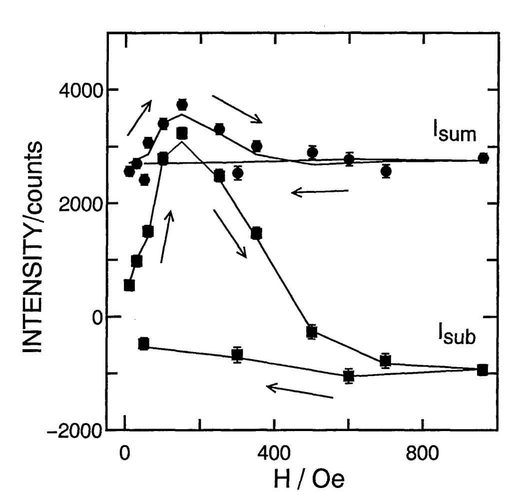

Fig. 3. Magnetic field dependence of neutron diffraction intensities I_{sum} (circle) and I_{sub} (square) measured at 77 K. The definition of I_{sum} and I_{sub} is shown in the text. The solid lines are the calculated intensities.

where $I_{nuclear}$ is an intensity coming from the nuclear scattering. A and B are constants. These three quantities are expressed with the nuclear scattering amplitudes, number densities and thicknesses of the constituent layers of the multilayer. These quantities are known. G is a geometrical constant, which is independent of the magnetic field. M_j^z is a parallel component of the magnetization vector of j layer to the applied field. Besides the intensities I_{sum} and I_{sub}, we can measure the total magnetization M_{tot} at every fixed field and temperature. In the present sample, M_{tot} is given as

$$M_{tot}=(M_{Co}^z+M_{NiFe}^z)/2. \qquad (3)$$

Equations (1)-(3) exactly hold good within the assumption. To reproduce the observed neutron intensities and total magnetization, we make a second assumption of $M_j=M_j^z z$, where z is a unit vector parallel to the applied field. This means that the transverse component of the magnetization vector to the applied field has no long range order and does not contribute to the diffraction intensity. In other words, only the parallel component (z-component) of the magnetization vector is coherent. If we adopt the second assumption, three unknown quantities M_{Co}^z, M_{NiFe}^z and G are determined at every applied field and temperature. The magnetization curves of the Co and NiFe layers are thus obtained. The results at 77 K and 300 K are shown in Fig. 4. In the figure, the magnetization is expressed as a magnetic moment $\mu=M^z/(\rho\mu_B)$, where ρ is a number density and μ_B Bohr magneton. The value of G should be field independent. However obtained G's are not exactly the same. To determine the value of G, measurements were done in the field of 9.4 kOe at each temperature, where the assumption of $M_j=M_j^z z$ is surely satisfied. Using the value of G determined from the measurement at 9.4 kOe, the calculated intensities are plotted in Fig. 3 with solid lines. The good fitting supports the validity of the overall procedure of the analysis.

In addition, the validity of the method and the analysis used above was confirmed with using magnetic/non-magnetic multilayers such as Fe/Mg.[15] In this case, the magnetization curve of the magnetic layer can be determined only from the neutron diffraction measurements. On the other hand, the magnetization curve is easily determined by conventional magnetization measurements. A satisfactory agreement was obtained between the curves determined by neutron diffraction and magnetization measurements. Therefore it is expected that the magnetization curves of the Co and NiFe layers are obtained by the analysis stated above without any further correction.

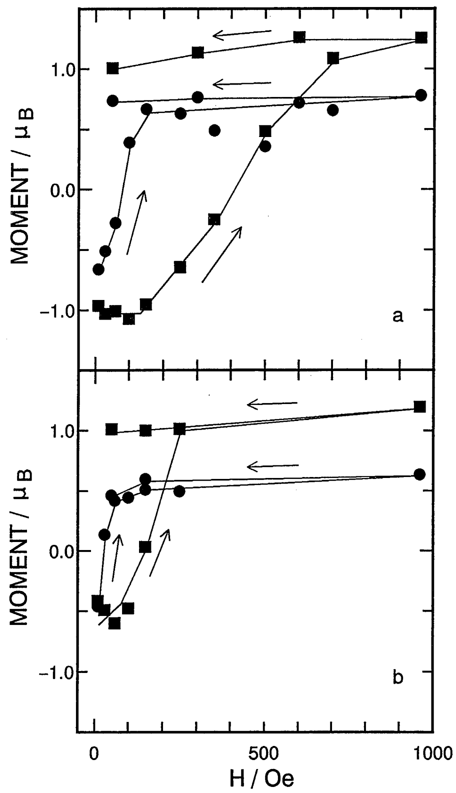

Fig. 4. Magnetization process of the Co (square) and NiFe (circle) layers at 77 K (a) and 300 K (b) determined from the magnetization and neutron diffraction measurements.

Figure 4(a) indicates the magnetization process of the individual magnetic layers at 77 K. First both the Co and NiFe layer magnetizations are saturated to the negative direction. With applying magnetic field to the positive direction, the magnetization reversal of the NiFe layer is completed around the applied field of 100 Oe. In contrast, the magnetization of the Co layer is antiparallel to the applied field and does not show a magnetization reversal up to 150 Oe. The long range order of the antiparallel alignment state of the Co and

NiFe magnetizations is thus confirmed by polarized neutron diffraction. The magnetic moments of the Co and NiFe layers in the antiparallel alignment state are almost the same as those at 940 Oe, respectively. This means that all magnetic moments in each layer are oriented to the same direction. In other words, the antiparallel alignment state is almost perfect for the applied field from 100 to 150 Oe. Above the applied field of 150 Oe, the Co magnetization gradually rotates to the applied field direction. These facts were inferred from the *M-H* curve features and are now experimentally confirmed.

The NiFe magnetization decreases around the applied field of 500 Oe. At present stage, we point out two possibilities for this. One is that the NiFe magnetization vector is really canted from the applied field direction. This may occur if the antiferromagnetic exchange interaction acts between the Co and NiFe magnetizations, though we have no experimental evidence for the interaction. The other is that this is only an artifact of the analysis. We assume that the magnetic state is expressed with a single magnetization vector $M_j = M_j^z z$. This approximation is not perfectly appropriate if the magnetization process is an inhomogeneous nucleation of magnetization reversal centers and a growth of proper magnetic domains. Such situations are expected in the magnetization reversal of the Co layer.

The magnetization process at 300 K is plotted in Fig. 4(b). The overall features of the curves are similar to that at 77 K. The antiparallel alignment state is realized around the applied field of 50 Oe. However the rotation of the Co magnetization already starts around the applied field of 50 Oe. The magnetic moment of the Co layer below 100 Oe is about 0.5 μ_B antiparallel to the external field, which is much smaller than that at 940 Oe. This indicates that not all the moments in the Co layer take the same direction. In other words, the long range order of the antiparallel alignment state at 300 K is less perfect than that at 77 K. Furthermore the saturation moment of the NiFe layer at 300 K is slightly smaller than that at 77 K.

3.3 *Magnetoresistance and magnetic structure*

From the magnetization processes shown in Figs. 4(a) and 4(b), it is suggested that the large temperature dependence of the magnetoresistance ratio is related to the degree of the antiparallel order. A possible definition of the antiparallel order parameter is $(M_{Co} - M_{NiFe})^2$ in the present case. As is known from eq. (1), this is related to I_{sum}. To make a quantitative comparison, we define the temperature dependence of the magnetoresistance change as $\Delta\rho(T) \equiv \rho(H_m, T) - \rho(960\text{ Oe}, T)$ and that of the diffraction intensity as $\Delta I_{sum}(T) \equiv I_{sum}(H_m, T) - I_{sum}(960\text{ Oe}, T)$, where H_m is the applied field corresponding to the resistivity maximum. A comparison of $\Delta I_{sum}(T)$ with $\Delta\rho(T)$ is made at 77, 150, 200 and 300 K. Results are plotted in Fig. 5. As $\Delta I_{sum}(T)/I_{nuclear}$ is a few tens percent, experimental errors in $\Delta I_{sum}(T)$ are relatively large. However a linear relationship between $\Delta I_{sum}(T)$ and $\Delta\rho(T)$ is obvious. The large temperature dependence of the magnetoresistance change is quantitatively related to the antiparallel order parameter $(M_{Co} - M_{NiFe})^2$.

So far we have concentrated on the parallel and antiparallel alignment states, i.e. the

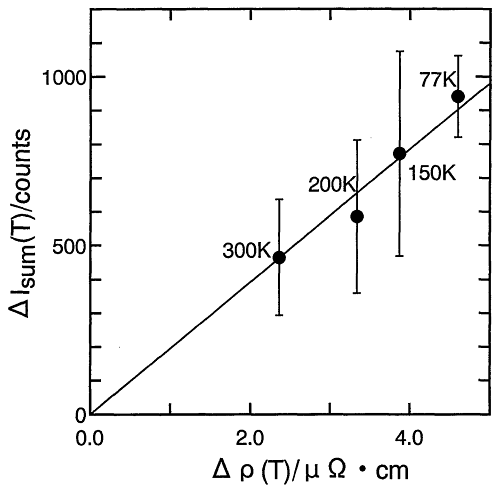

Fig. 5. Comparison of the magnetoresistance change and the change in the neutron diffraction intensity at various temperatures. The measurement temperatures are shown in the figure. The solid line is the least square fitted curve.

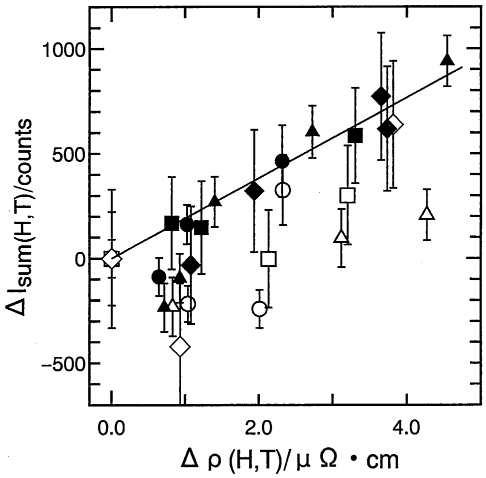

Fig. 6. Relationship between the magnetoresistance and neutron diffraction intensity measured at various temperatures and applied fields. The circle is measured at 300 K, square at 200 K, lozenge at 150 K and triangle at 77 K. The closed marks correspond to the field region where the NiFe moments are reversed. The open marks correspond to the region where the Co moments gradually rotate. The solid line is an eye guide.

limited field region. Now we consider a relation between the magnetoresistance and magnetic structure under arbitrary magnetic fields and temperatures. As $\Delta I_{\text{sum}}(T) \propto [M_{\text{Co}}(H_{\text{m}}, T) - M_{\text{NiFe}}(H_{\text{m}}, T)]^2 - [M_{\text{Co}}(960\ \text{Oe}, T) - M_{\text{NiFe}}(960\ \text{Oe}, T)]^2$ (see eq. (1)), the linear relationship implies that the resistivity $\rho(H, T)$ and the magnetization vectors $M_j(H, T)$ in the field H and at temperature T are connected with an equation of

$$\rho(H, T) = \rho_0(T) + C[M_{\text{Co}}(H, T) - M_{\text{NiFe}}(H, T)]^2, \quad (4)$$

where $\rho_0(T)$ is a field independent term and C is a constant. To examine this relation, the neutron intensity difference $\Delta I_{\text{sum}}(H, T) \equiv I_{\text{sum}}(H, T) - I_{\text{sum}}(960\ \text{Oe}, T)$ is compared with the resistivity difference $\Delta\rho(H, T) \equiv \rho(H, T) - \rho(960\ \text{Oe}, T)$ with using all measured data. Results are shown in Fig. 6. The deviation from the linear relationship is not small at some data points. Let us classify the magnetization process into two region. One is a region where the NiFe magnetization is reversed. The other corresponds to a gradual rotation of the Co layer moments. The former is indicated with closed marks and the latter open marks. The closed marks lie nearly on the linear relation. The NiFe layer is magnetically soft and the magnetization reversal range is relatively narrow. This implies that the magnetization reversal is rather uniform, i.e. the distribution of the direction of the NiFe layer moments is small. In such conditions, the antiparallel order parameter $(M_{\text{Co}} - M_{\text{NiFe}})^2$ is meaningful. On the other hand, the order parameter $(M_{\text{Co}} - M_{\text{NiFe}})^2$ is supposed to be improper in the Co magnetization reversal region. Since the distribution of the direction of the Co moments seems to be large in this field region, a single magnetization vector M_{Co} is not enough to represent the magnetic state of the Co layer.

In conclusion, the relationship between the magnetoresistance and magnetic structure as a function of magnetic field and temperature is expressed with eq. (4) when the single magnetization vector M_j properly represents the magnetic structure of j layer. A similar relation was already found at 300 K in Fe/Cr[12] and Fe/Au multilayers.

3.4 *Comparison with theoretical predictions*

There are several theoretical predictions between the magnetoresistance change and the magnetic structure. In most cases, theory deals with two identical magnetic layers. Nevertheless it is worth comparing our empirical relationship, eq. (4), with the theoretical predictions, because the applicability of eq. (4) is not confined with Co/Cu/NiFe/Cu multilayers. As for a magnetic field dependence, the following relation is proposed;[16]

$$\sigma(\theta) = \sigma^{\uparrow\downarrow} + (\sigma^{\uparrow\uparrow} - \sigma^{\uparrow\downarrow}) \cos^2(\theta/2), \quad (5)$$

where σ is an electric conductivity, θ an angle between two magnetization vectors. The symbols $\uparrow\downarrow$ and $\uparrow\uparrow$ represent the antiparallel and parallel alignment states, respectively. When two magnetic layers are identical, then $\cos^2(\theta/2)$ is expressed as $1 - (\Delta M)^2/(4M_s^2)$, where ΔM is a difference in the magnetization vectors of adjacent magnetic layers and M_s is a saturation magnetization. In this condition, eq. (5) leads to

$$\rho(\Delta M)=\left(\frac{1}{\sigma^{\uparrow\uparrow}}\right)\left\{\frac{1}{1-\frac{1}{4}\left(\frac{\sigma^{\uparrow\uparrow}-\sigma^{\uparrow\downarrow}}{\sigma^{\uparrow\uparrow}}\right)\left(\frac{\Delta M}{M_s}\right)^2}\right\}$$
$$=\left(\frac{1}{\sigma^{\uparrow\uparrow}}\right)\left\{1+\frac{1}{4}\left(\frac{\sigma^{\uparrow\uparrow}-\sigma^{\uparrow\downarrow}}{\sigma^{\uparrow\uparrow}}\right)\left(\frac{\Delta M}{M_s}\right)^2 +\frac{1}{16}\left(\frac{\sigma^{\uparrow\uparrow}-\sigma^{\uparrow\downarrow}}{\sigma^{\uparrow\uparrow}}\right)^2\left(\frac{\Delta M}{M_s}\right)^4+\cdots\right\}, \quad (6)$$

where $\rho(\Delta M)$ is a resistivity in the magnetic structure of ΔM. Equation (6) is equivalent to eq. (4) if the higher order terms of $(\Delta M)^2$ can be neglected, i.e. the magnetoresistance ratio is small.

The temperature dependence of magnetoresistance change between the parallel and antiparallel alignment states was also theoretically considered.[17] From the empirical eq. (4), the magnetoresistance change at temperature T is proportional to the square of the saturation magnetization $[M_s(T)]^2$. This is theoretically derived under the very limited conditions.

The empirical eq. (4) seems to have some theoretical supports concerning the field dependence. However as for the temperature dependence, theoretical basis of eq. (4) is not clear.

§4. Summary

The existence of the long range ordered antiparallel alignment state of the Co and NiFe magnetizations is clearly detected in the Co/Cu/NiFe/Cu GMR system by polarized neutron diffraction. The observed large temperature evolution of the magnetoresistance change $\Delta\rho(T)$ is attributed to the temperature change in the antiparallel order parameter $(M_{Co}-M_{NiFe})^2$. A general relationship between the magnetoresistance and magnetic structure, $\rho(H, T)=\rho_o(T)+C[M_{Co}(H, T)-M_{NiFe}(H, T)]^2$, is experimentally derived under the condition that the magnetization vector M_j properly represents the magnetic structure of j layer.

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