Two Oscillatory Behaviors as Functions of Ferromagnetic Layer Thickness in Fe/Cr(100) Multilayers

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Oscillatory magnetoresistance as a function of the Fe layer thickness has been found in Fe/Cr(100) multilayers deposited on MgO(100). It is mainly attributed to an oscillatory interlayer exchange coupling between adjacent Fe layers as a function of the Fe layer thickness. Further, oscillatory variation of the saturation resistivity with Fe layer thickness was also observed. Both the oscillation periods are approximately 8 Å. These two oscillations can be understood in terms of the partial confinement of the perpendicular motion to the layer plane of Fe majority spin electrons.

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In recent years metallic layered materials have been gaining interest for designing new materials with desirable properties. One of the essential features in magnetic multilayers is an exchange coupling between ferromagnetic layers across nonmagnetic spacer layers. Since the discovery of the long-period oscillations in exchange coupling as a function of the spacer thickness in Co/Ru, Co/Cr, and Fe/Cr multilayers [1], the oscillatory exchange coupling between ferromagnetic (F) and antiferromagnetic (AF) coupling has created widespread interest because of its universality in a wide variety of magnetic multilayers and the unusual oscillation periods (9–23 Å) [2,3]. Furthermore, the AF-coupled films exhibit "giant" negative magnetoresistance (MR) [4] which has potential applications in magnetic storage technology. The oscillatory exchange coupling is accompanied by oscillations in magnetoresistance.

Explanations of the oscillatory exchange coupling have generally been based on the RKKY theory. Although naive RKKY theory predicts a single period $\Lambda = \lambda_F/2 \approx 1$ ML which is much shorter than the experimental ones, the long periodicity of the oscillations can be obtained by considering the discreteness of the spacer thickness [5]. Further refinement by Bruno and Chappert [6] showed that the coupling can be understood in terms of the topological properties of the Fermi surface of the spacer material; oscillation is determined by one or more extremal wave vectors parallel to the growth direction which connect two points of the Fermi surface with antiparallel Fermi velocities. Multiperiodic oscillations predicted in [6] have been actually found in Co/Cu(100) [7] and Fe/Au(100) [8] systems, and predicted orientational dependence of the coupling strength [6] has been reported for the Co/Cu system [9]. Moreover, strong dependence on the Fermi surface dimensions of the oscillation period has been found in Cu-Ni alloy spacer materials [10].

On the other hand, the oscillatory exchange coupling based on the idea of quantum confinement of the electrons within the individual spacer layers has been proposed by Edwards *et al.* [11]. In general, the component of the wave vector of electrons in the direction perpendicular to the film plane becomes discretized by thinning the film thickness to confine the electrons in this direction. This leads to so-called quantum size effects. Since the first observation in a low-energy electron transmission experiment for thin Au films [12], quantum size effects in metal films have been experimentally observed in many phenomena: oscillatory variations of the electrical resistivity with increasing film thickness, low-energy electron tunneling experiments, and direct observations of quantum well states by means of photoemission and inverse photoemission spectroscopies (Refs. [13–15] and references therein). The treatment [11] provides another quantum size effect characteristic of the magnetic multilayers in which the spin polarized carriers are confined in the individual layers. Further treatment of the quantum confinement by Barnas [16] has predicted that the coupling strength oscillates also as a function of the ferromagnetic layer thickness. So far, the coupling strength has been observed to be constant, irrespective of the thickness of the ferromagnetic layers (e.g., [17]) and little attention has been paid to the ferromagnetic layer thickness.

Most recently, a unified theory of interlayer exchange coupling which includes the RKKY theory and the quantum confinement theory in terms of the spin asymmetry of the reflection at the interfaces has been presented by Bruno [18], in which it is indicated that the interlayer exchange coupling also oscillates as a function of the ferromagnetic layer thickness.

In this Letter, we present the first observation of the oscillatory magnetoresistance as a function of the ferromagnetic layer thickness in Fe/Cr(100) multilayers. It is mainly attributed to the oscillatory exchange coupling as a function of the Fe layer thickness. Furthermore, the saturation resistivity also oscillates significantly with the same period as the exchange coupling. These two oscillations are characteristic of the quantum size effect in thin films, and from the comparison between band structures of Fe and Cr, it is found that the Fe majority spin electrons are partially confined to Fe layers.

Fe/Cr(100) multilayers were grown epitaxially on

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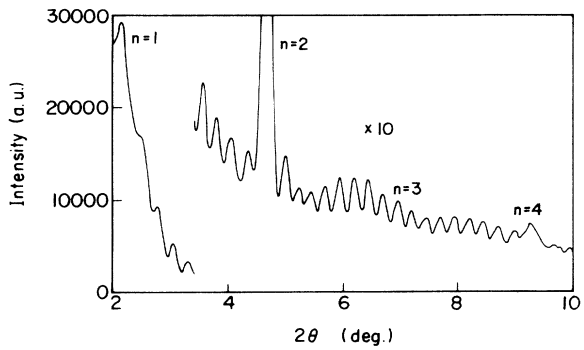

FIG. 1. Low-angle x-ray diffraction pattern for 8×[Fe(27 Å)/Cr(12 Å)] multilayer.

single crystal MgO(100) substrates using ion beam sputtering (IBS) at a base pressure of 5×10^{-7} Torr. Film orientation relative to the substrate confirmed by the transmission electron diffraction was Fe/Cr(100) ∥MgO(100) and Fe/Cr⟨100⟩∥MgO⟨110⟩. The ferromagnetic layer thickness was designed from 5 to 29 Å and the individual Cr layer thickness was held constant at 12 Å which is just thicker than the thickness at which the first maximum in AF coupling with Cr thickness occurs. On the top of the multilayers, 38 Å Cr layers were deposited to prevent oxidation. The thicknesses for each multilayer were confirmed by a chemical analysis using inductively coupled plasma-optical emission spectroscopy which revealed that the total Cr layer thickness for each multilayer was constant within ±0.4 Å throughout a series with varying Fe thicknesses. In addition to Fe/Cr multilayers, $Fe_{0.8}Cr_{0.2}$/Cr multilayers were also prepared for comparison. Figure 1 shows a typical low-angle x-ray diffraction pattern of an Fe/Cr multilayer. Superlattice Bragg peaks up to fourth order and finite-size peaks between them are clearly visible. High-angle x-ray diffraction patterns showed 200 peaks with first to third order satellites. These x-ray diffraction results suggest that the individual layers are *very* flat and have *relatively small* interfacial atomic mixing, although it is difficult to derive a quantitative interfacial structure from the x-ray diffraction results. Here, we should notice further the interfacial structure of the multilayers prepared by IBS deposition. In the case of the IBS deposition, the atoms arriving at the already deposited layers have higher kinetic energies on the average in comparison to those in molecular-beam epitaxy (MBE), permitting a little interfacial mixing depending on the energy of the arriving atoms. If interfaces are perfectly sharp, the coupling strength is very sensitive to the small change in the Cr layer thickness within 1 ML, because of the appearance of the 2 ML oscillation in interlayer exchange coupling with the Cr layer thickness which has been observed in the MBE-grown wedge-shaped sandwiches [19]. However, the interfacial mixing will smear the 2 ML oscillation and make the coupling strength less sensitive to the small error in the Cr thickness, which makes it possible to investigate the exchange

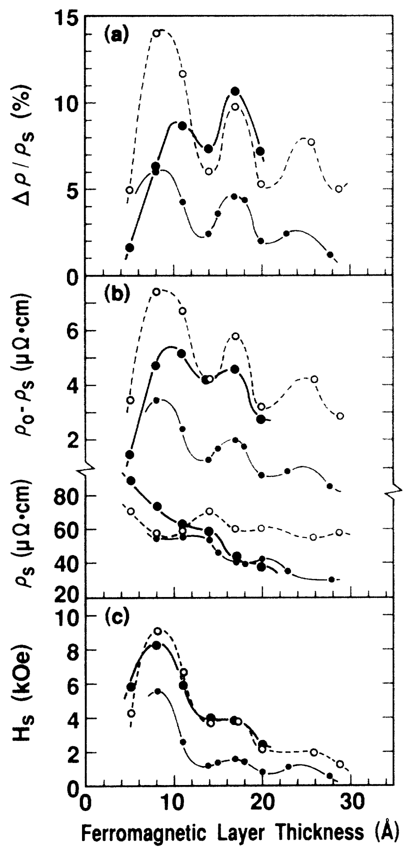

FIG. 2. (a) Magnetoresistance ratio (77 K), (b) resistivity change and saturation resistivity (77 K), and (c) saturation field (293 K) vs ferromagnetic layer thickness for 15×[Fe(t Å)/Cr(12 Å)] (100) multilayers deposited by 500 eV sputter Ar ions (large solid circles), 16×[Fe(t Å)/Cr(12 Å)] (100) multilayers deposited by 400 eV sputter Ar ions (small solid circles), and 15×[$Fe_{0.8}Cr_{0.2}$(t Å)/Cr(12 Å)] (100) multilayers deposited by 500 eV sputter Ar ions (open circles). The external magnetic field is applied parallel to the Fe(Cr) [100] in the film plane. Oscillations are also observed with the applied field parallel to the Fe(Cr) [110].

coupling and the MR ratio as a function of the ferromagnetic layer thickness by using the individually prepared multilayered samples. For the multilayers with an Fe layer thickness of more than 8 Å, the saturation magnetization M_s per volume unit of Fe at room temperature was constant at 1700 emu/cm^3, and a cubic in-plane magnetic anisotropy of the order of 10^5 erg/cm^3 was introduced with the easy axis of the magnetization parallel to the Fe⟨100⟩ direction. For $Fe_{0.8}Cr_{0.2}$/Cr multilayers with an $Fe_{0.8}Cr_{0.2}$ layer thickness of more than 8 Å, M_s was 1200 emu/cm^3. M_s in both cases dropped rapidly with decreasing ferromagnetic layer thickness from 8 to 5 Å.

Figure 2(a) presents the dependence of the MR ratio on the ferromagnetic layer thickness. MR was measured using a direct current four-terminal method with an applied magnetic field parallel to the Fe/Cr⟨100⟩ in the film plane. The MR ratio exhibits similar behavior for three sets of the multilayers, oscillatory behavior with ferromagnetic layer thickness: Peaks start at 9 Å with a period of 8 Å. Figure 2(b) shows a numerator of the MR ratio, resistivity change, $\Delta\rho = \rho_0 - \rho_s$ and a denominator, saturation resistivity, ρ_s, separately. Oscillations in $\Delta\rho$ corresponding to the MR ratio indicate that the oscillatory behavior in the MR ratio is mainly attributed to the change in $\Delta\rho$. However, ρ_s also varies weakly with peaks at the thicknesses corresponding to the valleys in $\Delta\rho$, also contributing to the oscillation in the MR ratio. The saturation field as a function of the ferromagnetic layer thickness is shown in Fig. 2(c). It exhibits peaks around 8 Å followed by weak oscillations superimposed on the largely decreasing curves with the ferromagnetic layer thickness. The decrease in the saturation field above 8 Å is due to the increasing ferromagnetic layer thickness, because $H_s = 4J/M_s t - 2K_1/M_s$ [20], where t and K_1 are the ferromagnetic layer thickness and cubic anisotropy constant of the ferromagnetic layers, respectively. Therefore the actual oscillation in the coupling strength is more pronounced than that in the saturation field; the coupling strength derived from Fig. 2(c) was roughly proportional to the MR ratio. Figure 2(c) indicates that the interlayer exchange interaction between ferromagnetic layers across Cr layers itself oscillates while maintaining AF coupling. Average AF-coupling strength ignoring the oscillation is 0.3 mJ/m^2 for Fe/Cr multilayers deposited by 500 eV Ar ions, which is equal to that of 12 Å Cr thickness for a Fe/Cr/Fe(100) MBE-grown wedge-shaped sandwich [21]. The AF-coupling strength determines the antiferromagnetically aligned component of the magnetization vectors of the adjacent ferromagnetic layers at zero field by which the spin-dependent scattering of the conduction electrons occurs. Therefore the oscillation in the coupling strength yields the oscillation in $\Delta\rho$. Above the saturation field, the external magnetic field which overcomes the AF coupling aligns ferromagnetic layers parallel to the applied field, to give ρ_s independent of the coupling strength. If the oscillation of ρ_s originates from fluctuations of the sample conditions such as layer thickness, interface sharpness, and crystal perfection, a temperature coefficient of ρ_s should also fluctuate with ferromagnetic layer thickness. However, the temperature coefficient of ρ_s between room temperature and 77 K increased linearly without any deviating points from the line with increasing ferromagnetic layer thickness for three sets of multilayers. The oscillation in ρ_s obviously indicates the occurrence of the size effect.

We interpret the observed oscillatory behaviors as functions of the ferromagnetic film thickness to be caused by the partial confinement of the majority spin band in the Fe layers, because the oscillation of the resistivity has been known as one of the typical quantum size effects since the first prediction by Sandomirskii [22], and the oscillation in the interlayer exchange coupling is expected as a result of the quantum confinement of the spin polarized electrons in ferromagnetic layers as shown by Barnas [16]. Making a comparison between Fe and Cr band structures, the minority spin-band structure of the Fe is quite similar to the Cr band structure, while the majority spin bands of Fe lie lower than the Cr bands [23]. Therefore minority spin electrons can move freely passing through interfaces, whereas, for majority spin electrons, interfaces become potential barriers. Although this barrier cannot truly confine the majority spin electrons to the Fe layers because there are electronic states in the neighboring layers to connect with, the partial reflection of the wave function at the interface can yield quantum well resonances giving rise to discrete states. Such quantum well resonance states have been observed by photoemission and inverse photoemission experiments for Ag films on Ni, Cu, Si, Ge, and Au substrates [13] and Cu films on Co and Fe substrates [15].

As a result of the partial confinement of the majority spin electrons in the Fe layers, the electron density of the majority spin electrons at the Fermi level fluctuates with respect to the Fe layer thickness. It should affect not only the saturation resistivity and the coupling strength but also the spin-dependent scattering directly. Most recently, oscillation in magnetoresistance with the layer thickness, directly affected by the quantization of the perpendicular electron motion to the film plane, was shown by Vedyayev *et al.* [24]. Their calculations are performed for perfectly antiferromagnetically aligned samples at zero field. However, actual multilayers are not often perfectly antiferromagnetically coupled; in that case, the antiferromagnetically aligned component of the magnetization is determined by the coupling strength; hence, spin-dependent scattering is also strongly affected by the coupling strength as mentioned earlier. In our result in which the MR ratio is roughly proportional to the derived coupling strength, the influence of the quantum effect on the spin-dependent scattering is thought to hide behind that of the coupling strength.

In the simple free electron model, an expected oscillation period in coupling strength with ferromagnetic layers is $\lambda_F/2$ [6] and an expected oscillation period in resistivity is also $\lambda_F/2$ [22,25]. However, $\lambda_F/2$ for Fe is 1.8 Å, which is too small to explain the observed oscillation period. Although the detailed theories which deal with the periodicity of the oscillatory behavior caused by quantum size effect have not been presented, to our knowledge, we suppose that the observed oscillation period corresponds to the extremal wave vector of the Fe majority spin Fermi surface, from an analogy with the de Haas–van Alphen effect presented by Edwards *et al.* [11]. In de Haas–van Alphen oscillations, the carrier energy is quantized by a magnetic field in a two-dimensional plane perpendicular to the field, whereas in the present problem, one-dimensional quantization in the direction perpendicular to the film is caused by thinning the film thickness. By analogy with de Haas–van Alphen oscillations in which the oscillation period is determined by the extremal area in the quantized plane, the oscillation period of the quantum well state would be expected to be determined by the extremal wave vector along the direction perpendicular to the film plane. In the majority spin Fermi surface of Fe, we tentatively assign such an extremal wave vector to a wave vector **q** connecting the large Γ-centered majority spin electron surface and the intermediate *H*-centered majority spin hole pocket along the ΓH line, because its length $0.38\Gamma H$ [26] leads an oscillation period $\Lambda = 2\pi/|\mathbf{q}| = 7.5$ Å and it gives rise to the strongest oscillatory behavior among the extremal wave vectors along the ΓH direction. In the case of a Fe-Cr alloy, the Fermi level for the majority spin bands decreases with increasing Cr content. Assuming that the change of the Fermi level is proportional to the Cr content, the length of this extremal vector does not change drastically up to 20% Cr because the $\Delta_1\uparrow$ band and the $\Delta_5\uparrow$ band in the E-k curve [23] are almost parallel just below the Fermi level. Thus the oscillation period would not be sensitive to small additions of the Cr.

In conclusion, we have observed the magnetoresistance in Fe/Cr(100) multilayers and found oscillatory interlayer exchange coupling and oscillatory saturation resistivity as functions of the ferromagnetic layer thickness. These oscillations can be understood in terms of the partial confinement of the majority spin electrons to the Fe layers.

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