Magnetoresistance Associated with Antiferromagnetic Interlayer Coupling Spaced by a Semiconductor in Fe/Si Multilayers

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Multilayer Fe/Si films with constant Fe thickness (2.6 nm) and variable Si thickness are investigated. Negative magnetoresistance is observed and two different temperature dependences are found as a function of Si thickness. For $t_{Si} = 1.2$ nm, the magnetoresistance decreases with temperature decrease. For $t_{Si} > 1.5$ nm, the magnetoresistance increases (weakly) with temperature decrease. The magnetoresistance is attributed to spin-dependent scattering caused by antiferromagnetic layer coupling across a semiconducting spacer: narrow gap iron silicide for thin Si spacer layers and amorphous Si for thicker spacer layers.

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The giant magnetoresistance (GMR) associated with antiferromagnetic (AF) interlayer exchange coupling in metallic multilayers formed by ferromagnetic and nonmagnetic metals has led to new important aspects in solid state physics. After the first report of the GMR in Fe/Cr multilayers [1], a number of metallic multilayers and spin-valve systems with noncoupling have been disclosed for providing GMR, including Co/Cu [2,3], NiFe/Cu/Co/Cu [4], NiFe/Cu/NiFe/FeMn [5], Co-Fe/Cu [6], NiFe/Cu [7], NiFe/Ag [8], and Ni-Fe-Co/Cu [9]. GMR has also been observed in heterogeneous thin films or rapidly quenched alloys such as Cu-Co alloys consisting of ultrafine Co-rich precipitate particles in a Cu matrix [10]. The mechanism of the GMR in the metallic multilayers and the heterogeneous alloy systems has been ascribed to spin-dependent scattering of conduction electrons [11].

As for magnetoresistance (MR) originating from spin-dependent scattering spaced by an insulator in a film, the ferromagnetic tunneling junction (ferromagnet/insulator/ferromagnet) has been reported, where GMR exceeding 16% at room temperature (RT) was reported recently in $Fe/Al_2O_3/Fe$ [12]. Furthermore, GMR in heterogeneous materials using insulating matrix was also reported recently in $Co\text{-}Al_2O_3$ [13]. In multilayers or heterogeneous systems using semiconducting spacers or matrices, however, MR induced by spin-dependent scattering has not been reported.

Recently, it was discovered that evaporated Fe/Si/Fe trilayers [14,15] and sputtered Fe/Si multilayers [16] exhibit AF interlayer coupling. The spacer inducing AF coupling is different in each case. AF coupling in the multilayers was observed only with crystalline spacer layers [16], attributed to iron silicide formed in the interface, while in the trilayers the spacer was claimed to be amorphous semiconducting Si (*a*-Si) [14,15]. The different spacer may originate from the difference in the preparation methods and the substrate temperatures used, because Fe and Si easily form alloys. It was also found in sputtered Fe/(Fe-Si) multilayers that the degree of AF coupling decreases dramatically as temperature decreases and that AF coupling at low temperature can be photoinduced [17]. Negative MR associated with AF coupling as in metallic multilayers, however, has not been reported in the Fe/Si/Fe trilayers or the Fe/Si and Fe/(Fe-Si) multilayers.

In this Letter we present the first observation of negative MR associated with AF coupling between ferromagnetic layers across semiconducting spacers in a series of $[(2.6\text{ nm Fe})/(t_{Si}\text{ nm Si})]_{22}$ multilayers as a function of the nominal Si layer thickness. We observed two different types of MR in the multilayers. One was observed at a nominal Si layer thickness of 1.2 nm at RT and significantly decreased at a low temperature. The other was observed for t_{Si} above 1.5 nm at RT, possessing a minimum around 2.5 nm, and was increased at a low temperature with weak temperature dependence. The spacer inducing the MR was attributed to a narrow gap semiconducting iron silicide, formed in the interface, for the former and to an *a*-Si for the latter.

The Fe/Si multilayers were grown at ambient temperature on water cooled thermally oxidized Si substrates by ion beam sputtering at a base pressure of 5×10^{-7} Torr. The sputtering was conducted using 1.8×10^{-4} Torr Ar ions and an acceleration voltage of 400 V. Individual Fe layer thicknesses were held constant at 2.6 nm, and the number of bilayers was 22 for all the films studied. The Si layer thickness was designed to be 0–3.5 nm. The thicknesses of Fe and Si layers were corrected on the basis of chemical analysis using inductively coupled plasma-optical emission spectroscopy and the bilayer thickness estimated from low-angle x-ray diffraction peaks. Magnetization and temperature-dependent magnetic hysteresis curves were measured using a vibrating sample magnetometer with a magnetic field in the film plane. Magnetoresistance measurements were carried out by the four point method with a field in the film plane.

In x-ray diffraction measurements, each sample exhibited second to sixth superlattice peaks at low-angle order

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depending on the nominal Si layer thickness, which indicates that the films are well layered. No satellite peaks, except for the Fe(110) main peak, were observed in the high-angle region for all the films. Cross-sectional transmission electron microscopy (TEM) also revealed the layered structure. Figure 1 shows the saturation magnetization M_s, the ratio of the remanent magnetization M_r to the M_s, and the saturation field H_s at RT for a [(2.6 nm Fe)/(t_{Si} nm Si)]$_{22}$ series. M_s takes the bulk value at t_{Si} = 0, decreases with increasing t_{Si}, and is saturated to the constant value of about 1200 emu/cm^3 above t_{Si} = 1.5 nm. This suggests that Fe-Si interdiffusion layers are formed in the interfaces up to 1.5 nm Si, above which the Si layer was identified to be amorphous by TEM observation. The maximum interdiffusion layer of 1.5 nm is consistent with the result of Dufour *et al.* [18]. There is a peak in H_s of 300 Oe and a minimum in M_r/M_s of 0.3 for t_{Si} = 1.2 nm, suggesting AF coupling, and M_r/M_s is nearly constant above t_{Si} = 1.5 nm. These results are in agreement with those of Fullerton *et al.* [16], except for the behavior of H_s. Our H_s in Fig. 1 is one order lower than that of Fullerton *et al.*, and exhibits a tendency to increase again with t_{Si} above 2.5 nm, while their H_s was constant above 1.5 nm.

We have estimated interlayer coupling J between Fe layers across the spacer from the saturation field using the relation of $H_s = 4J/dM_s$ (d is the Fe layer thickness) for the multilayers shown in Fig. 1, assuming AF coupling above 1.2 nm Si. The AF coupling above 1.2 nm Si was confirmed by ferromagnetic resonance (FMR) measurements using X-band (9.4 GHz) spectroscopy with a field applied in the film plane. The FMR for t_{Si} above 1.2 nm exhibited the optical and the acoustic resonance modes with the appearance of the optical mode at a higher field than that of the acoustic mode, demonstrating AF coupling. The interlayer coupling estimated from the resonance field separation between the optical and the acoustic modes was in agreement with that obtained from the saturation field. J is shown as a function of nominal Si layer thickness in Fig. 2. In this figure ferromagnetic (FM) coupling was assumed below 1.0 nm Si, of which value cannot be determined by H_s, because the M_r/M_s is large. The AF coupling seems to show an oscillation, possessing a minimum around t_{Si} = 2.5 nm. However, it is noted in Fig. 2 that the spacer is different depending on whether the Si thickness is less than or greater than 1.5 nm. The spacer is an iron silicide for the thickness up to 1.5 nm, as suggested in Fig. 1, and an amorphous Si for $t_{Si} > 1.5$ nm, as revealed by TEM observation. Thus, the spacer inducing the AF coupling for $t_{Si} > 1.5$ nm consists of an iron silicide and an a-Si layer.

We measured MR for the multilayers shown in Fig. 1, where negative MR was observed in the AF-coupled multilayers. Figure 3 shows MR curves at RT and 4 K for [(2.6 nm Fe)/(1.2 nm Si)]$_{22}$ and [(2.6 nm Fe)/(3.5 nm Si)]$_{22}$ multilayers as examples. The saturation fields at RT for both multilayers are in agreement with those in Fig. 1, obtained from the magnetization curve. The temperature dependences of the MR are significantly different from each other. The MR for t_{Si} = 1.2 nm is temperature sensitive with a decrease at a lower temperature, while the MR for t_{Si} = 3.5 nm is slightly temperature dependent with an increase at a lower temperature. Figure 4 exhibits resistivity at zero field and MR ratio at RT and 4 K as a function of nominal Si layer thickness for [(2.6 nm Fe)/(t_{Si} nm Si)]$_{22}$ multilayers. The resistivity increases with increasing Si thickness and

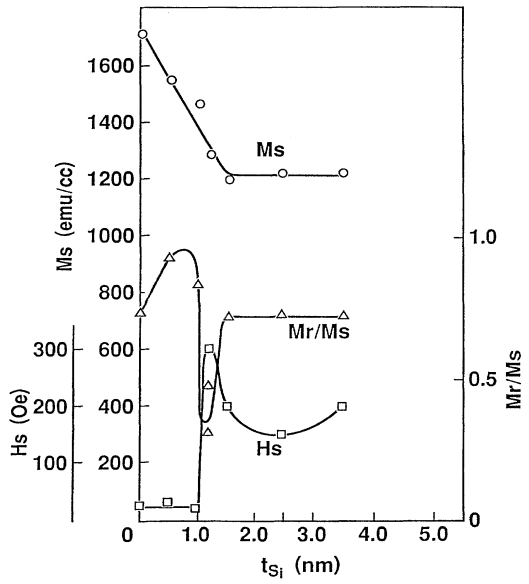

FIG. 1. Saturation magnetization M_s, M_r/M_s (M_r is the residual magnetization), and saturation field H_s at RT as a function of Si layer thickness in [(2.6 nm Fe)/(t_{Si} nm Si)]$_{22}$ multilayers.

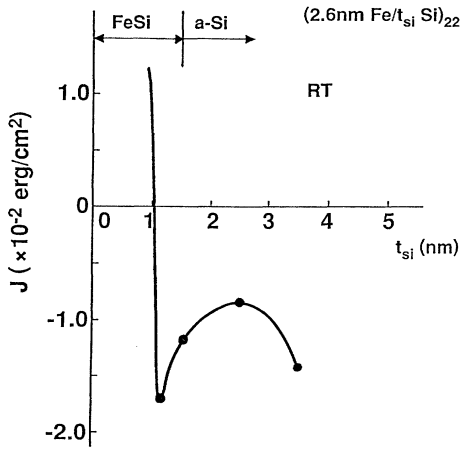

FIG. 2. Interlayer exchange coupling at RT as a function of Si layer thickness in [(2.6 nm Fe)/(t_{Si} nm Si)]$_{22}$, which was obtained from the saturation field in M-H curve and FMR measurements.

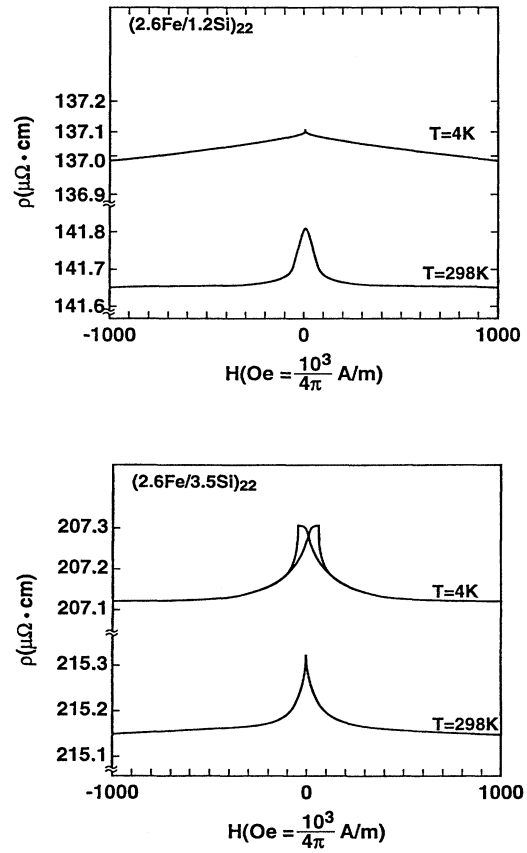

FIG. 3. MR curves at RT and 4 K for [(2.6 nm Fe)/(1.2 nm Si)]$_{22}$ and [(2.6 nm Fe)/(3.5 nm Si)]$_{22}$ multilayers.

provides a small change between RT and 4.2 K. The MR ratio demonstrates different temperature behaviors depending on whether the nominal Si thickness is less than or greater than 1.2 nm. The MR is temperature sensitive with the positive temperature coefficient for $t_{Si} = 1.2$ nm and weakly temperature dependent with the negative temperature coefficient for $t_{Si} \geq 1.5$ nm, both of which are significantly different from the behavior in metallic multilayers, where MR greatly increases with decreasing temperature. The small MR at RT and 4 K for $t_{Si} = 1.0$ nm is ascribed to the FM coupling. Thus, the origin of the negative MR may be attributed to spin-dependent scattering as in metallic multilayers. The difference in temperature dependence of the MR between $t_{Si} = 1.2$ nm and $t_{Si} \geq 1.5$ nm may originate from the difference in the spacer material as shown in Fig. 2.

In order to investigate the properties of the spacer and the cause of the temperature dependence of the MR for $t_{Si} = 1.2$ nm, we measured the magnetic hysteresis curve at various temperatures. We found that M_r/M_s dramatically increases from 0.3 at RT to 1.0 at 5 K with decreasing temperature, suggesting coupling conversion from AF to FM. When $-\ln(M_r/M_s)$ vs inverse temperature was

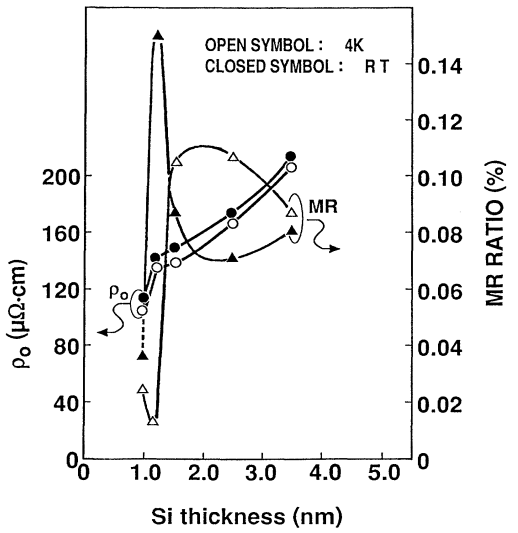

FIG. 4. MR ratio and resistivity at RT and 4 K as a function of Si layer thickness in [(2.6 nm Fe)/(t_{si} nm Si)]$_{22}$ multilayers.

plotted, the temperature dependence was similar to that of the carrier concentration in an impurity semiconductor. The carrier concentration n in an n-type semiconductor, for example, can be given by $n(n + N_A)/(N_D - N_A - n) = (N_c/2)\exp(\Delta E_D/k_B T)$ [19], where N_A, N_D, and N_c are the numbers of acceptors, donors, and conduction levels, respectively, and k_B and $\Delta E_D = E_c - E_D$ are the Boltzmann constant and ionization energy of the donors, respectively. This leads to a temperature-dependent carrier concentration curve similar to the temperature dependence of M_r/M_s, suggesting that the spacer is an impurity semiconductor in the multilayer. Using the correspondence, we estimated the activation energy of the spacer such that E_g, the energy gap in the intrinsic region, is 0.033 eV and ΔE is 0.015 eV. It is noted that E_g of 0.033 eV is similar to $E_g = 0.05$ eV of the bulk ϵ-FeSi silicide. Thus, we infer for the [(2.6 nm Fe)/(1.2 nm Si)]$_{22}$ multilayer that the spacer is a narrow gap semiconductor ϵ-FeSi, as suggested by Mattson *et al.* [17], and that the FeSi has a localized state in the energy gap, which is supported by the recent resistivity measurements by Chainai *et al.* [20]. We believe that the exchange coupling is dependent on the carrier concentration of the semiconducting spacer, and becomes FM coupling at a low temperature for which the excitation to the conduction band from the valence band is largely diminished, and thus the carrier concentration significantly decreases. If the number of carriers changes significantly, as expected in the present semiconductor, the Fermi wave vector also should change, which may alter the period of the coupling, leading to transition from AF to FM coupling when the temperature changes.

The weak temperature dependence of the MR with the negative temperature dependence of $t_{Si} > 1.5$ nm suggests that the semiconducting properties of the spacer are

different from those of $t_{Si} = 1.2$ nm. We revealed that the spacer for $t_{Si} > 1.5$ nm consists of the iron silicide and the a-Si layers. According to Chainai *et al.* [20] an a-Si in evaporated Fe/a-Si/Fe trilayers provides a positive temperature coefficient of the interlayer coupling, which is different from the present results, because their result should give a MR with a positive temperature coefficient. The conflicting behaviors might originate from the double spacers consisting of the iron silicide and the a-Si in our multilayers, or the difference in the impurity- or defect-induced states of the a-Si, caused by the different preparation methods. The mechanism of AF coupling through an a-Si and AF-biased–a-Si spacer is an important issue to be solved.

Very recently, Bruno proposed a theory for the interlayer magnetic coupling between ferromagnetic layers by an insulating spacer by extending his theory [21] for the metallic spacer, in which AF coupling was demonstrated to increase significantly with temperature [22]. This, however, cannot explain our results for the Fe/Si multilayers with $t_{Si} > 1.5$ nm. The disagreement may be attributed to the crystalline insulating spacer assumed in the theory, while we assumed, for $t_{Si} > 1.5$ nm, an a-Si spacer.

To summarize, we demonstrated two negative MR associated with AF couplings with different origins as functions of Si layer thickness in a series of [(2.6 nm Fe)/(t_{Si} nm Si)]$_{22}$ multilayers. One was observed at around $t_{Si} = 1.2$ nm at RT and decreased at a low temperature. The other was observed for t_{Si} thicker than 1.5 nm with a minimum around $t_{Si} = 2.5$ nm at RT, and slightly increased at a low temperature. The former originates from a narrow gap semiconducting spacer FeSi, with a localized state in an energy gap formed in the interface, and the latter is attributed to an amorphous Si spacer.

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