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ANTIFERROMAGNETIC EXCHANGE COUPLING IN Fe/Si_xFe_{1-x} MULTILAYERS

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Abstract: The Fe/Si_xFe_{1-x} multilayers with $x = 1$, 0.66, 0.5 and 0.4 have been examined. The strongest room temperature antiferromagnetic interlayer coupling, $J = -1.94$ mJ/m^2, was found in multilayers with nominally pure Si spacer layer. The analysis of the temperature dependences of remanence to saturation magnetization ratios allowed to find the value of semiconductor spacer layer energy gap ($E_g \approx 200$ meV) of the examined structures. It may suggest that the Fe-Si phase, responsible for antiferromagnetic coupling is an amorphous-like Fe-Si phase.

1. INTRODUCTION

Metal-semiconductor multilayers (Mls) are extensively studied because of their potential application in electronics. Recently the investigations have been focused on Fe/Si/Fe coupled heterostructures since they show a very strong antiferromagnetic (AF) interlayer coupling [1]. In spite of many efforts, the origin of the interlayer coupling in Fe/Si system has not been clarified [2-5]. Moreover, it is not well understood how the iron-silicides formation affects the interlayer coupling. Therefore the information about the spacer layer composition and its correlation with magnetic properties of this system is of particular interest. The main goal of our study was to test whether the existence of the strong AF coupling in Fe/Si Mls is related to (or mediated by) the appearance of the interfacial Fe-Si phases like FeSi and/or $FeSi_2$.

2. EXPERIMENT

The [Fe(3 *nm*)/$Si_xFe_{1-x}(d_S)$]*15 multilayers, (where Si_xFe_{1-x} alloys for $x = 1$, 0.66, 0.5 and 0.4 simulate Si and $FeSi_2$, FeSi, Fe_5Si_3 iron-silicide phases, respectively and d_S denotes the spacer layer thickness) were deposited in UHV by d.c. magnetron sputtering at room temperature onto oxidized Si wafers. The crystalline structure of our samples and their multilayer periodicity were examined using the high- and small-angle X-ray diffraction, respectively. Magnetization measurements were performed in the as-deposited state and after sequential annealing at 100 °C and 220 °C as a function of temperature ranging from 4.2 to 300 K.

3. RESULTS AND DISCUSSION

Figure 1 displays the saturation fields H_S and F_{AF} parameter values ($F_{AF} = 1 - M_R/M_S$, where M_R and M_S denote the remanence and saturation magnetizations, respectively) obtained at room temperature for the examined Fe(3 *nm*)/$Si_xFe_{1-x}(d_S)$ Mls with $x = 1$, 0.66, 0.5 as a function of the spacer layer thickness d_S. In metallic type Mls the F_{AF} parameter is usually treated as an indicator (or may be proportional to) of the antiferromagnetically coupled fraction of the sample and $F_{AF} = 1$ when the sample is completely AF coupled and $F_{AF} = 0$ for ferromagneticaly coupled

Mls. Saturation field, according to the relation $J = -(1/4)H_S M_S d_{Fe}$ is proportional to the AF interlayer exchange coupling J. We have found that the spacer layer with $Si_{40}Fe_{60}$ composition is ferromagnetic at room temperature leading to direct ferromagnetic coupling between Fe layers in Fe/$Si_{0.40}Fe_{0.60}$ multilayers. The strongest antiferromagnetic coupling $J = -1.94$ mJ/m^2 (at room temperature) accompanied by saturation field of H_S = 1200 kA/m (1.51 T) has been found for Fe/Si multilayers with d_S = 1.35 nm. Figure 1 shows also that for all the examined Fe/Si$_x$Fe$_{1-x}$ Mls (except Fe/ $Si_{0.40}Fe_{0.60}$) only a single H_S and F_{AF} maximum *versus* spacer layer thickness has been observed. Position of the $F_{AF}(d_S)$ maximum moves towards the thicker spacer layer with decreasing x i.e., with increasing Fe concentration in the Si$_x$Fe$_{1-x}$ spacer layer. Above the $H_S(d_S)$ maximum i.e., for a thicker spacer layer its value was found to decay exponentially. The reduction of F_{AF} values below their $F_{AF}(d_S)$ maximum points out that the neighboring Fe layers become gradually connected through pinholes. The above results may suggest that the appearance of the single H_S maximum *versus* spacer layer thickness is not due to RKKY-like coupling mechanism. The observed non-oscillatory but exponentially decaying saturation field values seems to correspond rather to the quantum interference model of exchange coupling in metal/insulator structures given by Bruno [6]. His model predicts however, that the AF interlayer exchange coupling in metal/insulator structures increases with increasing temperature in contrast to the AF coupling behavior predicted for entirely metallic types of AF coupled Mls.

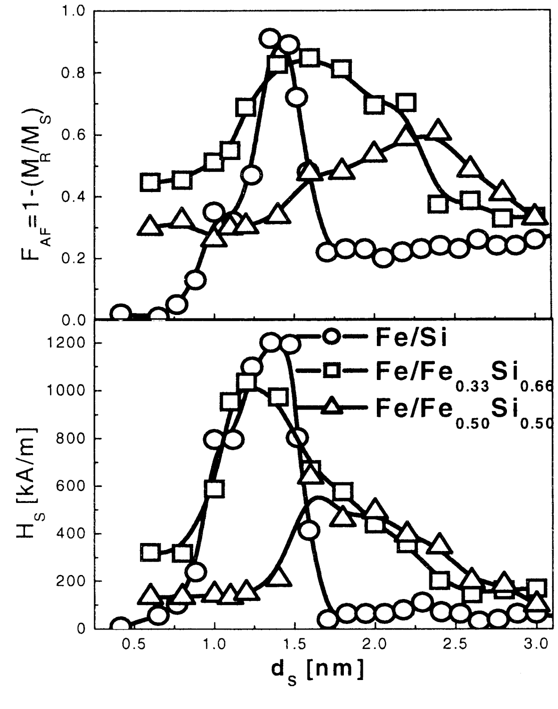
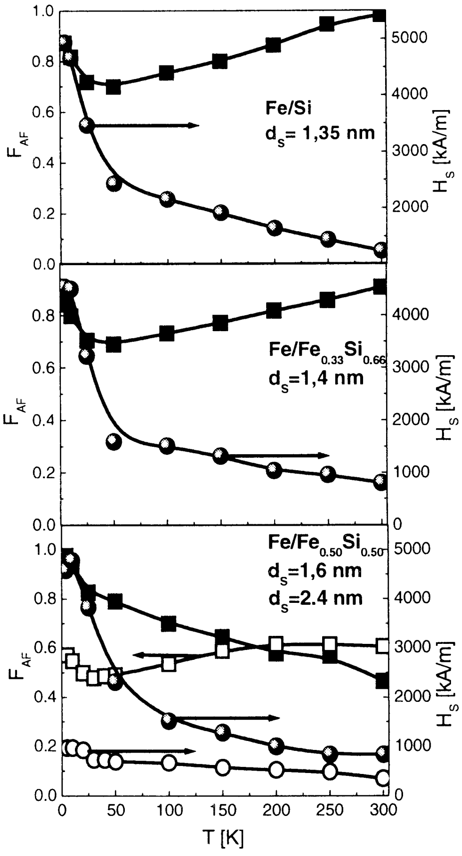


Fig. 1. Room temperature values of saturation field H_S and F_{AF} parameter of the examined [Fe(d_{Fe} = 3 nm)/Si$_x$Fe$_{1-x}$(d_S)]*15 Mls with x = 1, 0.66, 0.5 as a function of the spacer layer thickness d_S

Figure 2 shows the temperature dependences of the F_{AF} parameter and H_S for examined Mls with spacer layer thickness d_S representing the maximum values of $H_S(d_S)$ observed in our Fe(d_{Fe} = 3 nm)/Si$_x$Fe$_{1-x}$(d_S) Mls with x = 1, 0.66, 0.5. $F_{AF}(T)$ dependences show a very unusual



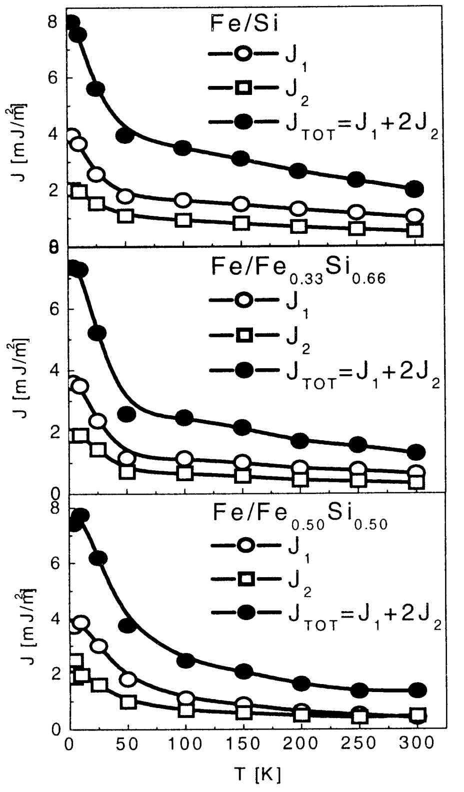


Fig. 2. Temperature dependences of the F_{AF} parameter and H_S of the examined MIs with spacer layer thickness d_S representing the maximum $H_S(d_S)$ values (from Fig. 1) of observed in our $[\mathrm{Fe}(d_{\mathrm{Fe}} = 3\mathrm{nm})/\mathrm{Si}_x\mathrm{Fe}_{1-x}(d_S)]*15$ MIs with $x = 1$, 0.66, 0.5. In case of $\mathrm{Fe/Si_{0.50}Fe_{0.50}}$ the $F_{\mathrm{AF}}(T)$ and $H_S(T)$ dependences for $d_S = 2.4$ nm (i.e., representing the $F_{\mathrm{AF}}(d_S)$ maximum) are also shown (open symbols)

Fig. 3. Temperature dependences of the bilinear and biquadratic coupling (J_1 and J_2, respectively) shown together with $J_{\mathrm{TOT}} = J_1 + 2J_2$

behavior. For Fe/Si and $\mathrm{Fe/Si_{0.66}Fe_{0.33}}$ MIs, $F_{\mathrm{AF}}(T)$ decreases with decreasing temperature up to about 50 K and then increases whereas in case of the $\mathrm{Fe/Si_{0.50}Fe_{0.50}}$ MI, $F_{\mathrm{AF}}(T)$ continuously increases with decreasing temperature. Dissimilar $F_{\mathrm{AF}}(T)$ behavior, observed for $\mathrm{Fe/Si_{0.50}Fe_{0.50}}$ MI with $d_S = 1.6$ nm (i.e., representing the $H_S(d_S)$ maximum) can be due the fact that the positions of the $H_S(d_S)$ and $F_{\mathrm{AF}}(d_S)$ maximum are distinctly different. However, for $\mathrm{Fe/Si_{0.50}Fe_{0.50}}$ MI with $d_S = 2.4$ nm (i.e., representing the $F_{\mathrm{AF}}(d_S)$ maximum) its $F_{\mathrm{AF}}(T)$ and $H_S(T)$ dependences are similar to those found for Fe/Si and $\mathrm{Fe/Si_{0.66}Fe_{0.33}}$ MIs. In metallic type of MIs with magnetic bridges (nonferromagnetic at given temperature), the reduction of the $F_{AF}(T)$ values with decreasing temperature usually suggests that the magnetic bridges become progressively ferromagnetic and that the neighboring ferromagnetic layers become gradually connected by pinholes [7]. In case of examined Mls however, this mechanism seems to be questionable since, below 50 K $F_{AF}(T)$ increases suggesting that the amount of the AF coupled phases increases. Therefore the magnetic pinholes mechanism may not be responsible for the observed unusual $F_{AF}(T)$ behavior because already active (ferromagnetic) magnetic pinholes can not be deactivated at lower temperatures.

Basing on the coupling energy expression given by Fullerton in [4] and on our magnetization measurements we have calculated bilinear (J_1) and biquadratic (J_2) coupling for [Fe(d_{Fe} = 3 nm)-/Si_xFe_{1-x}(d_S)]*15 Mls with x = 1, 0.66, 0.5 using the relations:

$$H_s = \frac{4(J_1 + 2J_2)}{M_s d_{Fe}}, \quad \text{and}$$
$$\frac{M_R}{M_S} = \begin{cases} 0 & \text{for } J_1 > 2J_2 \\ \sqrt{(2J_2 - J_1)/4J_2} & \text{for } J_1 < 2J_2 \end{cases} . \tag{1}$$

Shown in Figure 3 temperature dependences of the $J_1(T)$ and $J_2(T)$ (i.e., their decline with increasing temperature) are very similar to those observed in AF coupled metallic Mls.

The results presented above concerning $F_{AF}(T)$, $J(T)$ and $H_S(d_S)$ dependences show that the appearance of the AF interlayer coupling in the examined Fe/Si_xFe_{1-x} Mls is neither due to RKKY-like coupling mechanism nor due to the quantum interference mechanism of exchange coupling. It seems that the appearance of the AF coupling can be mediated by the formation of the nonmagnetic Fe-Si phases at Fe/Si_xFe_{1-x} interfaces. In our previous paper [8] it was shown that 0.25 nm thick, magnetically inactive layer is formed at each of the Fe/Si interfaces. It can be composed of nonmagnetic Fe-Si phases like ε-FeSi and/or β-$FeSi_2$ as confirmed by the Mössbauer spectroscopy. Inomata et al. [5] assumed that M_R/M_S is inversely proportional to the density of carriers in the spacer layer, and that the slope of the $-\ln(M_R/M_S)$ *versus* $1/T$ is equal to $-E_g/k_BT$, where E_g is the energy gap over which the carriers must be thermally excited. We have used this analysis for our Fe/Si_xFe_{1-x} Mls. Figure 4 displays the plot $-\ln(M_R/M_S)$ *versus* $1/T$ for Fe/Si and Fe/$Si_{0.66}Fe_{0.33}$ and Fe/$Si_{0.55}Fe_{0.50}$ Mls. For Fe/Si, Fe/$Si_{0.66}Fe_{0.33}$ and Fe/$Si_{0.50}Fe_{0.50}$ (with d_S = 2.4 nm i.e., representing the $F_{AF}(d_S)$ maximum) Mls the curves are similar to the temperature dependence of the carrier concentration in an impurity semiconductor (except the $-\ln(M_R/M_S)$ *versus* $1/T$ dependence for Fe/$Si_{0.55}Fe_{0.50}$ Ml with d_S = 1.6 nm i.e., representing the $H_S(d_S)$ maximum). It suggests that the semiconducting spacer may induce coupling in examined Fe/Si_xFe_{1-x} Mls. The steep rise region of $-\ln(M_R/M_S)$ above 100 K corresponds to the intrinsic region in a semiconductor. Using the described above correspondence we estimated the activation energy (energy gap) $E_g \approx 200$ meV. The estimated energy gap value is larger than that of ε-FeSi (50 meV [9]) and smaller than found for β-$FeSi_2$ (840 meV [9]) semiconducting Fe-Si phases. It may suggest that the Fe-Si phase responsible for the AF coupling in examined Mls is an amorphous-like (or fine-crystalline) Si_xFe_{1-x} phase with $E_g \approx 200$ meV. The existence of the amorphous-like Si_xFe_{1-x} phase seems to be confirm by the thermal treatment of our Mls. All the examined Fe/Si and Fe/$Si_{0.66}Fe_{0.33}$ Fe/$Si_{0.50}Fe_{0.50}$ Mls annealed by 1 hour in 220 °C showed only the existence of the ferromagnetic coupling and a strong reduction of the F_{AF} value up to 0.2-0.3.

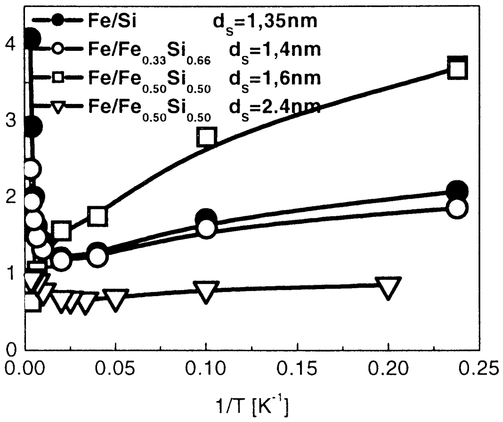


Fig. 4. $-\ln(M_R/M_S)$ *versus* $1/T$ dependences obtained for Fe/Si and Fe/$Si_{0.66}Fe_{0.33}$ and Fe/$Si_{0.50}Fe_{0.50}$ Mls

In summary, we have shown that the strongest room temperature AF interlayer coupling in examined $[Fe(d_{Fe} = 3\text{ nm})/Si_xFe_{1-x}(d_S)]*15$ Mls (with $x = 1, 0.66, 0.5$), $J = -1.94$ mJ/m^2 occurs for $x = 1$. Magnetic properties measurements showed the existence of only single saturation field $H_S(d_S)$ maximum with exponential decay above it. The analysis of the temperature dependences of M_R/M_S ratio allowed to find the value of semiconductor spacer layer energy gap in Fe/Si and Fe/$Fe_{0.33}Si_{0.66}$ multilayers ($E_g \approx 200$ meV). It may suggest that the Fe-Si phase, responsible for antiferromagnetic coupling in examined multilayers, is an amorphous-like Fe-Si phase.

Acknowledgments

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