STRUCTURAL AND MAGNETIC PROPERTIES OF Fe/Si$_x$Fe$_{1-x}$ MULTILAYERS

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Abstract: The Fe/Si$_x$Fe$_{1-x}$ ($x = 1$, 0.66, 0.5) and Fe/Co/Si/Co multilayers deposited by magnetron sputtering have been studied. The strongest antiferromagnetic coupling appeared in Fe/Si multilayers for $d_{Si} = 1.35$ nm with saturation field $H_S = 1.5$ T. Based on the temperature dependencies of the remanent magnetization we have estimated the semiconductor spacer layer energy gap $E_g \approx 200$ meV. It suggests that the Fe-Si silicide responsible for the antiferromagnetic coupling in Fe/Si multilayers is a semiconducting amorphous-like or nanocrystalline Si$_x$Fe$_{1-x}$ phase rich in Si. Annealing the multilayers destroys antiferromagnetic interlayer exchange coupling.

1. INTRODUCTION

Although the Fe/Si heterostructures have been extensively studied for several years [1-4], their interlayer coupling mechanism still remains unclear. It has not been explained how the iron-silicides formation affects the interlayer coupling and magnetic properties. The aim of this report is to clarify whether the existence of the strong antiferromagnetic (AF) coupling in the Fe/Si$_x$Fe$_{1-x}$ multilayers (Mls) is due to the presence of intermetallic Fe-Si phases such as β–FeSi$_2$ and/or ε-FeSi, which may arise from the interdiffusion of Fe into Si sublayers.

2. EXPERIMENT

The multilayered samples consisting of {Fe(3 nm)/Si$_x$Fe$_{1-x}$(d_S)} × 15 ($x = 1$, 0.66 and 0.5) and {Fe(3 nm)/Co(d_{Co})/Si(1.25 nm)/Co(d_{Co})} × 15 were deposited in UHV by magnetron sputtering at room temperature onto oxidized Si substrates. The Si$_x$Fe$_{1-x}$ spacer thickness d_S varied between 0.5 and 3 nm and the thickness of Co layer d_{Co} was 0.25 and 0.5 nm, thus the Fe/Co/Si/Co Mls were, in fact, the Fe/Si Mls with thin Co layers inserted between Fe and Si sublayers. The interface and structural properties were investigated by the conversion electron Mössbauer spectroscopy (CEMS) and X-ray diffraction, respectively. Their magnetic properties were examined by vibrating sample magnetometer (VSM) and extraction magnetometer measurements. Temperature dependencies of resistance $R(T)$ and magnetization were performed in the temperature range 4.2-300 K.

3. RESULTS AND DISCUSSION

The multilayer periodicity and the crystalline structure were investigated by small- and high- angle X-ray diffraction (SAXRD and HAXRD), respectively. The SAXRD spectra shown in Fig. 1a exhibit Bragg-like maxima, which confirm well defined periodic structure of the

examined Mls. Comparing the SAXRD spectra we can see, that the Fe/Si$_x$Fe$_{1-x}$ plots exhibit smaller number of peaks than the Fe/Si spectrum. This can be due to the better contrast between Fe and nominally pure Si sublayers, in the case of Fe/Si Ml. The HAXRD spectra shown in Fig. 1b exhibit only a single weak maximum of the *bcc*-Fe(110) phase, and several peaks originating from the Si substrate. There is also a tiny maximum at about 84°, which may be related to iron silicide phases. The HAXRD results showed, that the examined Mls were amorphous-like or nanocrystalline with crystallite dimension in the direction perpendicular to the sample plane not exceeding Fe sublayer thickness.
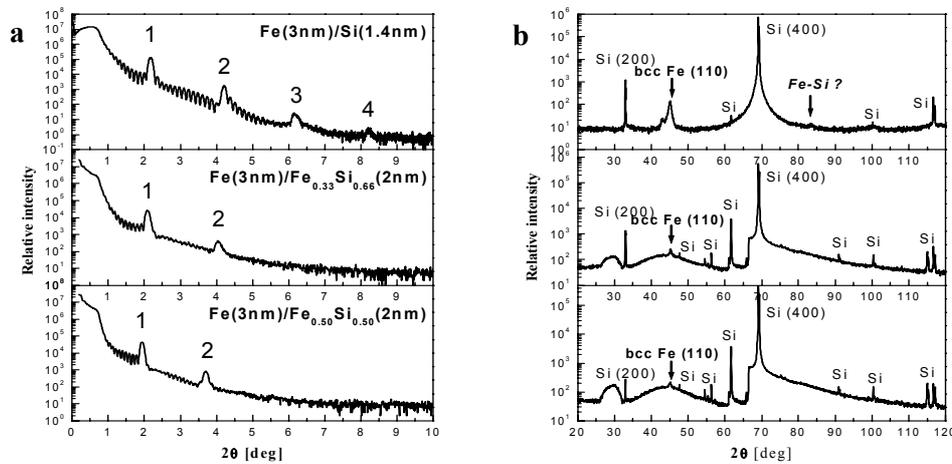

Fig. 1. SAXRD (a) and HAXRD (b) spectra of Fe/Si, Fe/Si$_{0.33}$Fe$_{0.66}$ and Fe/Si$_{0.50}$Si$_{0.50}$ Mls. The sublayers thicknesses as indicated above the plots

The interface structure of the examined Mls was investigated by CEMS. The CEMS spectra shown in Fig. 2a recorded at room temperature for Fe/Si Mls with d_{Fe} = 3 nm and d_{Si} = 1.2, 1.4 and 2.3 nm consist of 3 components: the Zeeman sextet with the hyperfine field $H_{hf} \approx 32.8$ T, and the isomer shift $\delta = 0.00$ mm/s, characteristic of the *bcc*-Fe phase of Fe layers, and two spectral components related to Fe-Si system at interfaces: (i) magnetic broadened sextet with $H_{hf} \approx 29$ T and $\delta \approx +0.05$ mm/s, originating most probably from Fe atoms at various interfacial step-sites and (ii) nonmagnetic component consisting of a quadrupole doublet with the splitting QS ≈ 0.60 mm/s and $\delta \approx +0.20$ mm/s. The last component corresponds to nonmagnetic iron silicide formed at interface and is related either to a small gap ε-FeSi semiconductor, the crystalline *c*-Fe$_{1-x}$Si$_x$ metallic phase or to the amorphous FeSi phase rich in Si [1, 2]. The CEMS spectra of Fe/Si$_{0.66}$Fe$_{0.33}$ Mls with two different thicknesses of SiFe layers (1.4 and 3 nm) consist of similar 3 components, but the spectral contribution of the QS doublet is significantly larger for the thicker SiFe layer (Fig. 2b). Based on the QS and δ values of the nonmagnetic spectral component it is difficult to determine the exact structure of the interfacial phase.

Magnetic properties were measured by vibrating sample magnetometer and extraction magnetometer. The saturation field H_S and F_{AF} factor ($F_{AF} = 1 - M_R/M_S$, where M_R and M_S denote the remanence and saturation magnetization, respectively) *versus* nonmagnetic spacer layer thickness d_S dependence is shown in Fig. 3.
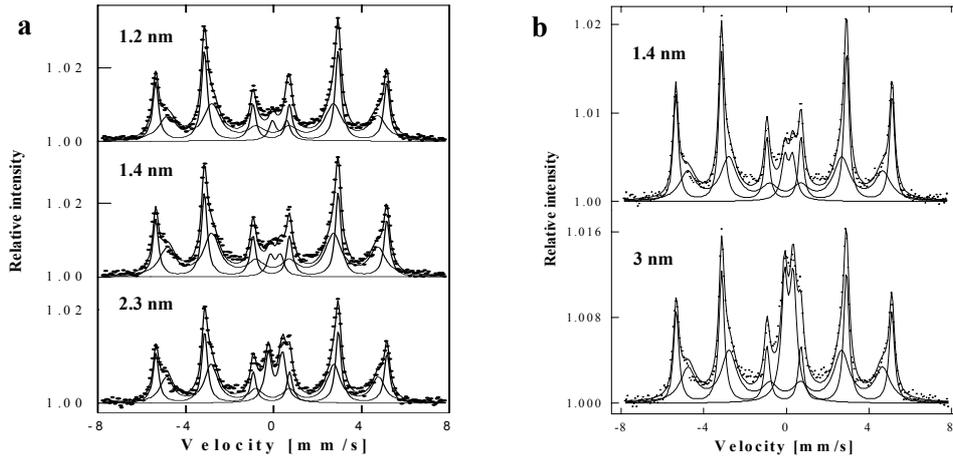

Fig. 2. CEMS spectra of Fe/Si (a) and Fe/Si$_{0.66}$Fe$_{0.33}$ (b) Mls with indicated spacer layer thicknesses

The existence of only single $H_S(d_S)$ maximum and exponential decay of H_S values above it suggests, that the exchange coupling in the examined Mls is not due to RKKY-like mechanism. It seems to correspond rather to the quantum interference model given by Bruno [5]. However, according to this model, in metal/insulator structures the *AF* coupling increases with increasing temperature, whereas our measurements (Fig. 4a) revealed, that the coupling decreases with increasing temperature. Therefore, the observed $H_S(d_S)$ behavior suggests, that the strong *AF* coupling in the Fe/Si Mls has another origin.
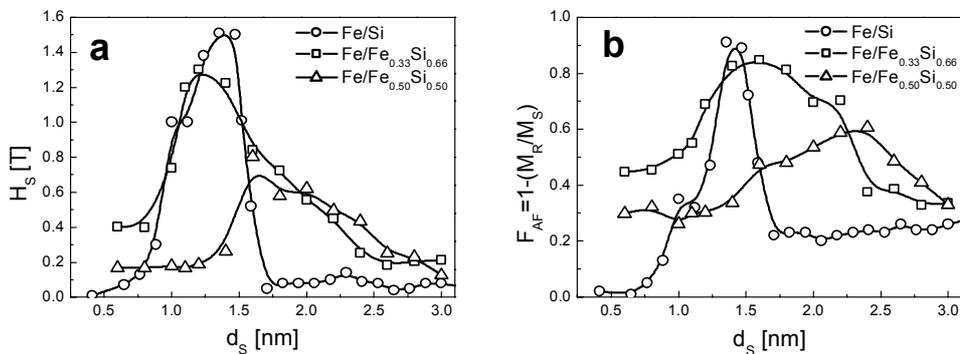

Fig. 3. Saturation field H_S (a) and F_{AF} factor (b) *vs* nonmagnetic spacer layer thickness d_S dependencies for Fe/Si$_x$Fe$_{1-x}$ Mls

In order to identify better the composition of an interfacial silicide phase the energy gap of this phase was estimated. According to Inomata *et al.* [3], the energy gap was determined from the analysis of the slope of $-\ln(M_R/M_S)$ *vs* $1/T$ which is equal to E_g/k_BT (E_g is the energy gap over which the carriers must be thermally excited). Our plots of $-\ln(M_R/M_S)$ *vs* $1/T$ for Fe/Si, Fe/Si$_{0.66}$Fe$_{0.33}$ and Fe/Si$_{0.50}$Fe$_{0.50}$ (Fig. 4b) are similar to the temperature dependence of the carrier concentration in an impurity semiconductor. It suggests that a spacer, which is semiconducting, may induce the coupling in Fe/Si$_x$Fe$_{1-x}$ Mls. The steep rise region of $-\ln(M_R/M_S)$ above 100 K corresponds to the intrinsic region in a semiconductor. Using this analysis we estimated that the energy gap is about 200 meV. This gap is larger than that found for ε-FeSi (50 meV [4]) and much smaller than that for the β-FeSi$_2$ (840 meV [4]).
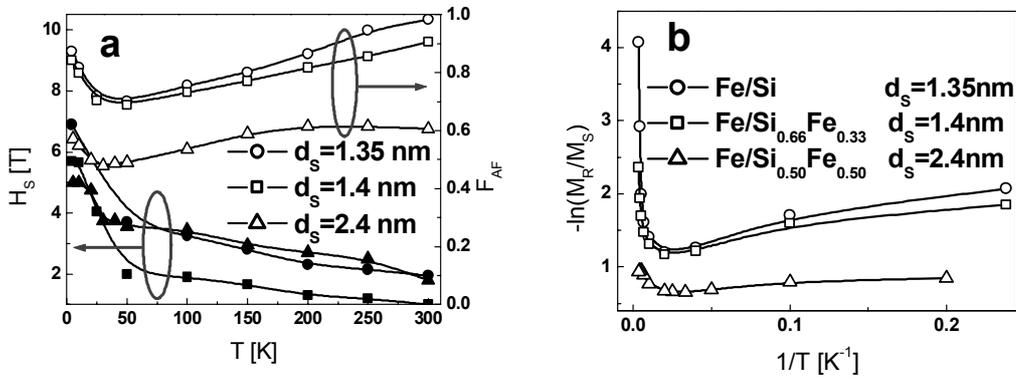

Fig. 4. Temperature dependencies of H_S and F_{AF} (a) and $-\ln(M_R/M_S)$ *vs* $1/T$ dependencies (b) for Fe/Si, Fe/Si$_{0.66}$Fe$_{0.33}$ and Fe/Si$_{0.50}$Fe$_{0.50}$ Mls, respectively

It may suggest therefore that the Fe-Si phase responsible for the *AF* coupling in the studied Mls is a semiconducting amorphous-like (or fine crystalline) Si$_x$Fe$_{1-x}$ phase rich in Si with $E_g \approx 200$ meV. The existence of the amorphous-like Si$_x$Fe$_{1-x}$ phase seems to be confirmed by thermal annealing of our Mls (1 h at 220°C) after which all examined multilayers showed the existence of the ferromagnetic coupling with strongly reduced F_{AF} value up to 0.2.

The $R(T)$ dependencies for all examined multilayered samples exhibit metallic behavior, whereas the single thin film of Si$_x$Fe$_{1-x}$ shows semiconducting behavior. Figure 5 displays the examples of $R(T)$ dependencies for Fe/Si$_{0.50}$Fe$_{0.50}$ Ml and Si$_{0.50}$Fe$_{0.50}$ thin film. It demonstrates, that the main contribution to the $R(T)$ dependencies in examined Mls arises mainly from Fe sublayers. At about 25 K a weak $R(T)$ minimum appears for Fe/Si$_x$Fe$_{1-x}$ Mls and persists even in the presence of magnetic field of 1 T. Therefore it testifies, that the resistance minimum is not due to the real Kondo effect, but may originate from the structural disorder.

According to [6], we could expect, that a thin layer of Co inserted between Fe and Si sublayers prevented the formation of Fe silicides. Actually, even 0.25 nm thick Co layer set at the interfaces disables the formation of the FeSi phases leading to the ferromagnetic coupling
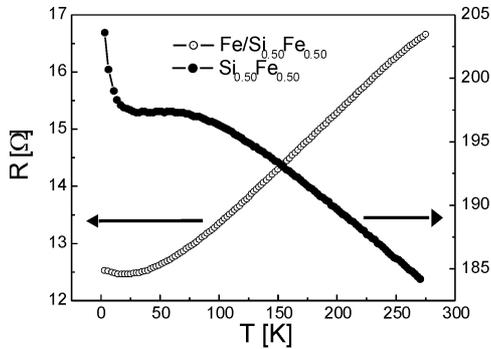
Fig. 5. $R(T)$ dependencies for Fe/Si$_{0.50}$Fe$_{0.50}$ (d_S = 2.4 nm) Ml and 30 nm Si$_{0.50}$Fe$_{0.50}$ thin film
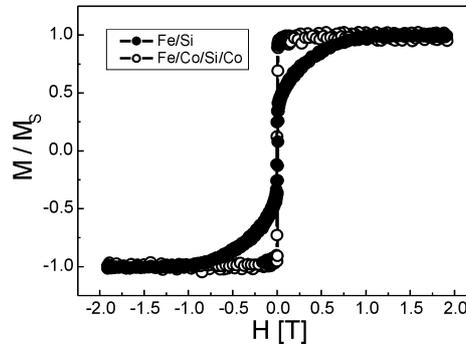
Fig. 6. Comparison of $M(H)$ dependencies for Fe/Si and Fe/Co/Si/Co Mls

only. The comparison of two histeresis loops of Fe/Si Mls with- and without Co sublayers is shown in Fig. 6.

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