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Mössbauer and magnetic study of interface structure of Fe/Si_xFe_{1-x} multilayers with antiferromagnetic interlayer coupling

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Abstract

The structural and magnetic properties of Fe/Si_xFe_{1-x} multilayers (Mls) have been studied by the conversion electron Mössbauer spectroscopy (CEMS) and magnetic measurements. We believe that the Fe-Si silicides responsible for the AF coupling in Fe/Si multilayers are semiconducting Si_xFe_{1-x} phases rich in Si which appear in inhomogeneous spacer layer. Antiferromagnetically coupled Fe/Si multilayers have been successfully applied as artificial antiferromagnets in the magnetoresistive $(Fe/Si)_{15}$/Fe/Co1/Cu/Co2 pseudo-spin-valve system.

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Keywords: Multilayers; Interlayer coupling; Mössbauer spectroscopy

1. Introduction

The Fe/Si system is extensively studied [1–3] because the origin of antiferromagnetic (AF) interlayer coupling has not been clarified and due to its potential applications in electronics. The investigations have been focused on Fe/Si/Fe coupled heterostructures that show a very strong AF interlayer coupling [1]. The aim of our study concerns the question whether the exchange coupling in Fe/Si multilayers (Mls) is related to the appearance of the interfacial Fe-containing silicide phases. Therefore, information on the interface structure and its correlation with magnetic properties of this system is of particular interest.

The structural and magnetic properties of Fe/Si_xFe_{1-x} Mls have been studied by the conversion electron Mössbauer spectroscopy (CEMS) and magnetic measurements.

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2. Experiment

The samples of $[Fe(3\,nm)/Si_xFe_{1-x}(d_S)]_{15}$ ($x = 1$, 0.66, 0.5 and $0.5 \leqslant d_S \leqslant 5$ nm) were deposited in UHV chamber by DC magnetron sputtering at room temperature onto oxidized Si wafers. The crystalline structure of our samples and their multilayer periodicity were examined by the high- and low-angle X-ray diffraction, respectively. The interface structure and the formation of iron silicides were studied by CEMS at room temperature. Magnetization measurements were performed for Mls in the as-deposited state and after sequential annealing at 100 and 220 °C and as a function of temperature ranging from 4.2 to 300 K.

3. Results and discussion

The saturation fields H_s and interlayer exchange coupling 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 are shown in Fig. 1a. Saturation field, according to the relation $J = -(\frac{1}{4})H_sM_sd_{Fe}$, is proportional to the AF interlayer exchange coupling J (Fig. 1a). The strongest antiferromagnetic coupling $J = -1.94\,mJ/m^2$ (at room temperature) accompanied by saturation field of 1.51 T has been found for Fe/Si multilayers with $d_s = 1.35$ nm. Fig. 1a shows that for all studied Fe/Si_xFe_{1-x} Mls only a single maximum of H_s versus spacer layer thickness has been observed up to $d_s \leqslant 5$ nm. Above the $H_s(d_s)$ maximum the $H_s(d_s)$ values decay exponentially (Fig. 1a). The observed nonoscillatory but exponentially decaying saturation field values seem to correspond to the quantum interference model of exchange coupling in metal/insulator structures given by Bruno [4]. However, the model of exchange coupling in metal/insulator predicts that the coupling energy should increase with increasing temperature in contrast to the AF coupling behavior predicted for entirely metallic Mls (RKKY-like coupling mechanism) [4]. We have found (Fig. 2a) however, that H_s values decrease with increasing temperature, similarly to the AF-coupled metallic Mls. The results concerning

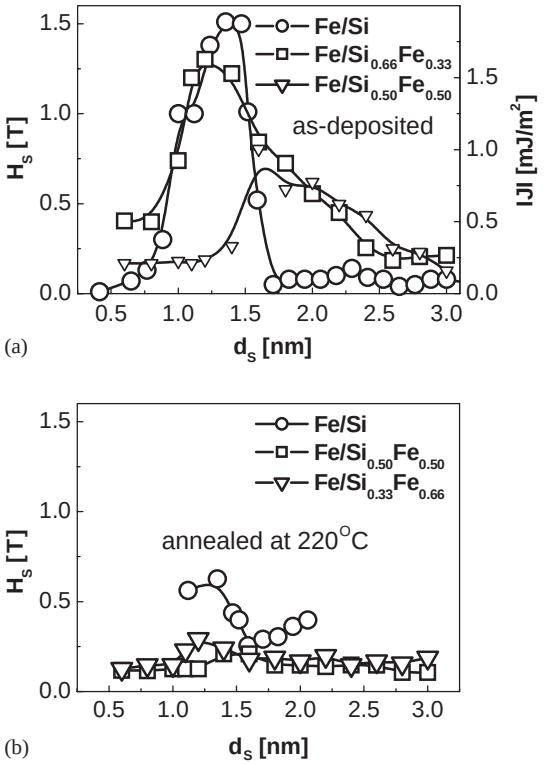

Fig. 1. Room temperature values of saturation field (and exchange coupling J_{AF}) of the examined $[Fe(d_{Fe} = 3\,nm)/Si_xFe_{1-x}(d_S)]*15$ Mls with $x = 1$, 0.66, 0.5 as a function of the spacer layer thickness d_S (a), and the influence of annealing on saturation field H_S (b).

$H_s(d_s)$ and $H_s(T)$ dependencies discussed above 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. The AF coupling can be mediated by the formation of nonmagnetic Fe–Si system at the interfaces and/or at spacer layers, e.g., ε-FeSi, nonstoichiometric c-$Fe_{1-x}Si_x$ crystalline silicides and/or intermixed crystalline or amorphous Fe–Si.

The spacer structure was investigated by CEMS. The CEMS spectra recorded at room temperature for Fe/Si Mls with $d_{Fe} = 3$ nm and $d_{Si} = 1.2$, 1.4 and 2.3 nm (Fig. 3) consist of three components: the Zeeman sextet with the hyperfine field $H_{hf} \approx 32.8$ T, and the isomer shift $\delta = 0.00$ mm/s,

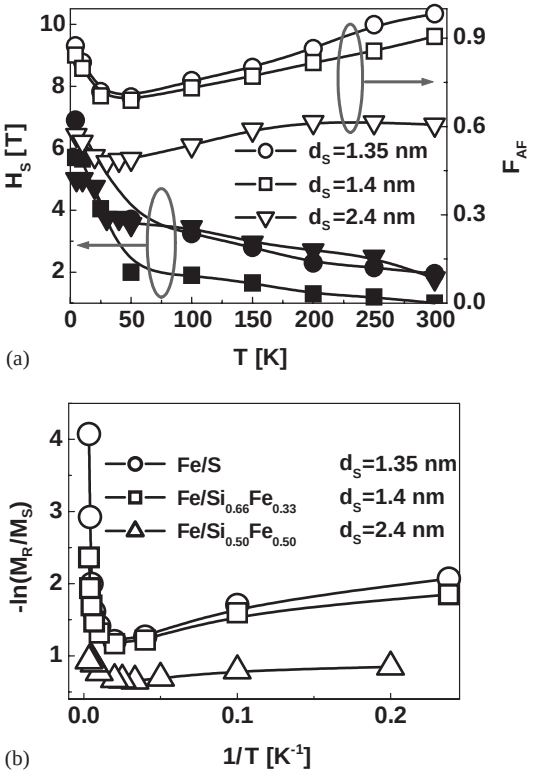

Fig. 2. Temperature dependencies of the H_S and F_{AF} parameter of examined [Fe($d_{Fe}=3$ nm)/Si$_x$Fe$_{1-x}$(d_S)]*15 Mls with $x=1$, 0.66, 0.5 Mls (a), and the $-\ln(M_R/M_S)$ versus $1/T$ dependencies for Fe/Si$_x$Fe$_{1-x}$ Mls with different x (spacer layer thicknesses d_S are shown) (b).

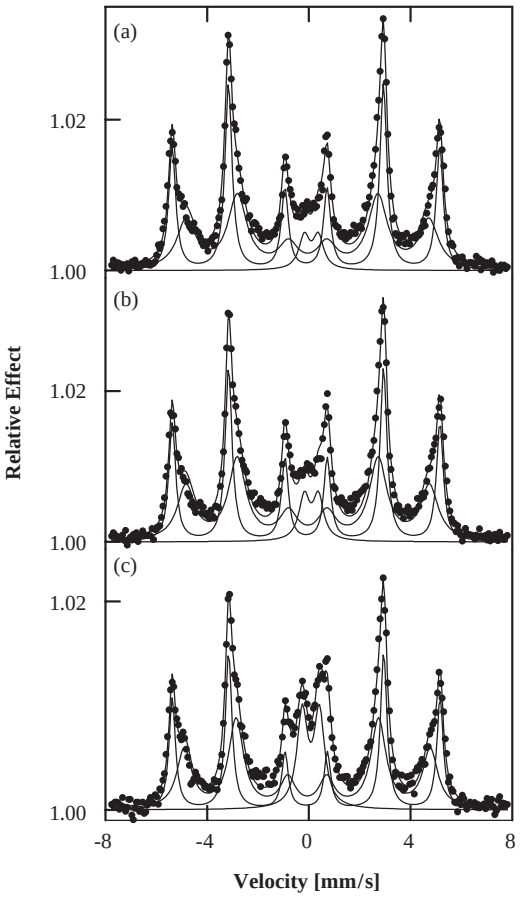

Fig. 3. CEMS spectra of Fe/Si Mls with Si layer thickness of 1.2 nm (a), 1.4 nm (b), and 2.3 nm (c), respectively.

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. The quadrupole splitting and the isomer shift for Fe/Si samples with $d_{Si}=1.2$ and 1.4 nm are almost identical ($QS\approx 0.55$ mm/s and $\delta\approx +0.20$ mm/s, Figs. 3a and b). The splitting of the quadrupole doublet increases to about 0.66 mm/s for the thickest Si spacer ($d_{Si}=2.3$ nm, Fig. 3c) suggesting that the iron silicide formed becomes richer in Si.

The last component corresponding to nonmagnetic iron silicide formed at interface could be related either to a small gap ε-FeSi semiconductor, the crystalline c-Fe$_{1-x}$Si$_x$ metallic phase or to the amorphous-like (or fine crystalline) FeSi phase rich in Si [5,6].

However, the quadrupole splitting for our Fe/Si Mls is somewhat larger than those typical for ε-FeSi [5,6] and c-Fe$_{1-x}$Si$_x$ [5] phases. It seems that the parameters of the nonmagnetic spectral component (QS and δ) correspond rather to inhomogeneous spacer containing nonmagnetic iron silicide phases.

The CEMS spectra of Fe/Si$_{0.66}$Fe$_{0.33}$ Mls with two different thickness of SiFe layers (1.4 and 3 nm) show the spectra consisting of similar three components, but the spectral contribution of the QS doublet is significantly larger for thicker

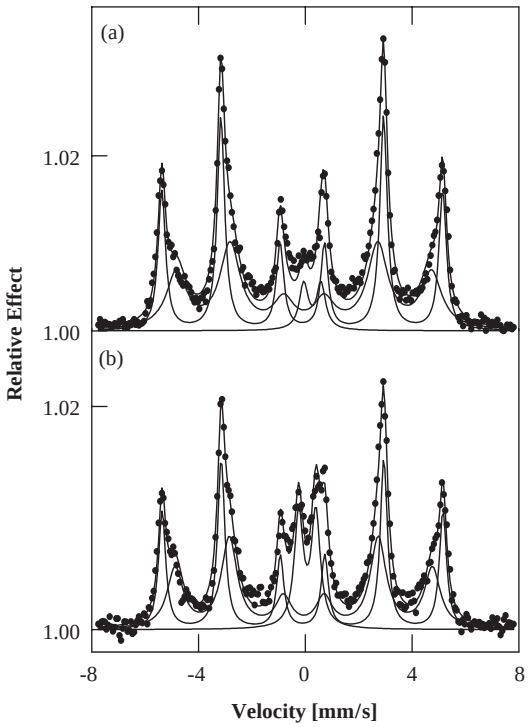

Fig. 4. CEMS spectra of $Fe/Si_{0.66}Fe_{0.33}$ Mls with thickness of SiFe layers of 1.4 nm (a) and 3 nm (b), respectively.

SiFe layer (Fig. 4). The smaller QS splitting ($QS \approx 0.42$ mm/s and $\delta \approx +0.24$ mm/s) in the $Fe/Si_{0.66}Fe_{0.33}$ Mls as compared with the Fe/Si samples may suggest that the composition of nonmagnetic silicide is somewhat different in the samples with $Si_{0.66}Fe_{0.33}$ spacer layers due to the interdiffusion during the deposition process (it contains more iron).

Basing on the QS and δ values of the nonmagnetic spectral component it is difficult to determine the exact structure of the interfacial phase. However, in our previous paper [7] it was shown that a 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 c-$Fe_{1-x}Si_x$ and/or intermixed Fe–Si.

In order to identify better the composition of an interfacial silicide phase the energy gap of this phase was estimated. The energy gap was determined, according to the Ref. [3], from the analysis of the slope of $-\ln(M_R/M_S)$ versus $1/T$ which is equal to E_g/k_B (where M_R and M_S denote the remanence and saturation magnetization, respectively, and E_g is the energy gap over which the carriers must be thermally excited). Our plots $-\ln(M_R/M_S)$ versus $1/T$ for Fe/Si, $Fe/Si_{0.66}Fe_{0.33}$ and $Fe/Si_{0.50}Fe_{0.50}$ (Fig. 2b) 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_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 that analysis we estimated that the energy gap is about (200 ± 50) meV. This gap is larger than that found for ε-FeSi (50 meV [8]) and much smaller than that for the β-$FeSi_2$ (840 meV [8]). Therefore we may conclude that the spacer layer is not homogeneous and contains semiconducting iron silicides rich in Si with an energy gap $E_g \approx 200$ meV which are responsible for the AF coupling in the studied Mls. After thermal annealing of our Mls by 1 h at 220 °C all examined multilayers show only the ferromagnetic coupling and a strong reduction of the saturation fields (Fig. 1b). It may be related to the existence of the fine-crystalline (or maybe amorphous-like) $Fe_{1-x}Si_x$ phases in the inhomogeneous spacer in the as-deposited samples. The magnetic pinhole mechanism is rather of secondary importance. The reduction of H_S and F_{AF} after annealing occurs similarly for thin and thick spacer layers (i.e., the differences ΔF_{AF} and ΔH_S of these parameters before and after annealing were almost the same for thin and thick spacer layers).

We demonstrate that the AF-coupled Fe/Si multilayers can be successfully applied as artificial antiferromagnets in the magnetoresistive $(Fe/Si)_{15}$/Fe/Co1/Cu/Co2 pseudo-spin- valve (PSV) system. We used a Co1/Cu/Co2 trilayer as a magnetoresistive structure with Co1(2) and Cu thickness of about 1.5 and 2.5 nm, respectively. Since the last Fe and Co1 layers are in the close contact, the Fe/Co1 bilayer behaves as a first magnetoresistive layer in the structure. The field dependence of the sample resistance, shown in Fig. 5, reflects a typical PSV giant magnetoresistance behavior. As can be seen, there are two switching fields: H_1

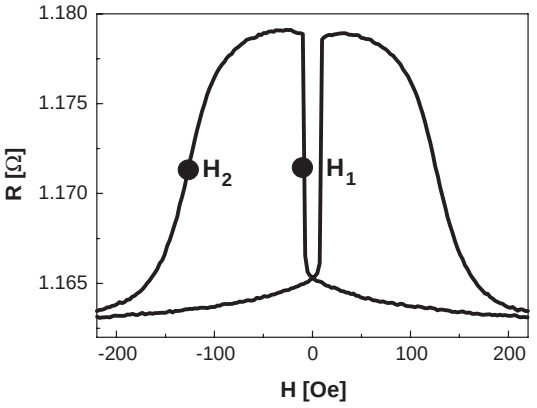

Fig. 5. Magnetoresistance effect of $(Fe/Si)_{15}Fe/Co/Cu/Co$ pseudo spin-valve structure with Fe/Si Ml used as a artificial antiferromagnet.

related to the magnetization reversal of the top Co2 layer, and H_2 which causes the switching of the Fe/Co1 pinned bilayer magnetization to the direction of the external magnetic field at higher fields. Since Fe/Co1 bilayer is AF coupled to the rest of Fe/Si Ml structure thus it reverses at higher fields than the top Co2 layer and an antiferromagnetic arrangement between magnetization of the Fe/Co1 bilayer and the Co2 layer occurs between fields H_1 and H_2.

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