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Optical and magneto-optical study of Fe/Si multilayers

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Experimental and theoretical investigations of the optical and magneto-optical (MO) properties of sputter deposited Fe/Si_xFe_{1-x} multilayers (MLS) are presented. The diagonal and off-diagonal components of the optical conductivity tensor of the MLS have been determined in the photon energy range 0.8–5.8 eV from the measurements of the magneto-optical complex Kerr angles and the optical data measured by the spectroscopic ellipsometry and compared with the theoretical ones calculated from first principles in density functional theory by the LMTO method within the supercell approach. The calculations have been performed for different models of iron silicide structures. In particular, various spacer layer structures: metallic FeSi and semiconducting $FeSi_2$ iron-silicide phases, as well as pure Si and Fe were investigated. The comparison of the recorded and calculated spectra confirm the conclusion inferred from other studies that the spacer layer structures represent semiconducting ε-FeSi and β-$FeSi_2$ phases rather than the metallic FeSi phase.

Key words: *magnetic multilayer; interface; magneto-optical spectrum; ab-intio calculations*

1. Introduction

Multilayered films consisting of transition metals and semiconductors have attracted a lot of interest because of their unique properties and possible application in the semiconductor industry. The basic phenomenon of the Fe/Si multilayer systems that will decide about the future application of the structure in the area of spintronics is the strong antiferromagnetic (AF) coupling between Fe sublayers within a wide

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range of nonmagnetic spacer layer thicknesses [1]. Many studies have been done to understand the mechanism of interlayer coupling in the system and its relation to the transformation of Si spacer layer into iron silicides [1–3]. Fe and Si form a rich variety of stable bulk binary alloys and several iron silicides have been identified. The nonmagnetic $FeSi_2$ compound exists in a stable orthorhombic semiconducting phase known as β-$FeSi_2$ and an unstable tetragonal metallic α-$FeSi_2$ phase. FeSi silicides exist in two phases: a stable semimetallic nonmagnetic ε-FeSi with cubic *B*20 type of structure and a metastable metallic with *B*2 (CsCl) structure. Moreover, there are few ferromagnetic phases that include Fe_5Si_3 alloy with the Mn_5Si_3 structure, Fe_3Si with the DO_3 structure, and metallic hexagonal Fe_2Si phase.

The nature of iron-silicides spontaneously formed during MLS growth responsible for the observed Fe/Si MLS magnetic and electrical properties was the subject of many studies [1–7]. A strong AF coupling in the Fe/Si MLS is mediated by Fe–Si silicides which may arise from the interdiffusion during the growth. There is an experimental evidence that Fe-Si silicides responsible for the AF coupling between Fe layers can exhibit semiconducting highly resistive character [2–4] or (depending on sublayer thicknesses and preparation technique) metallic properties [1, 5–7].

It is clear that the composition of the spacer layer formed during deposition of films is nonuniform [1, 3]. There is still some controversy about the structure and stoichiometry of the Fe/Si MLS interface region and the goal of the present work was to provide additional information about the chemical composition of the interface by applying experimental optical and magneto-optical spectroscopy methods, phenomenological multireflection calculations and *ab-initio* calculations of some model structures [8]. Effective optical and magneto-optical response of the layered structures is directly related to the thickness and properties of constituent sublayers that, in turn, depend on the nominal thickness of Fe and Si, interfacial mixing and roughness. The structure of Fe/Si MLSs can be verified by comparing the results of model calculations with the experiment.

2. Experimental

The set of Fe/Si_xFe_{1-x} MLS films was prepared in UHV chamber by magnetron-sputtering method on oxidized, (001)-oriented Si substrate. Various spacer layer compositions (x = 100, 66, 50) were intentionally deposited and the spacer layer thickness d was varied within the range from 0.5 nm to 3.0 nm. Each film was composed of 15 (Fe/Si) bilayer periods, the nominal Fe sublayer thickness being fixed at 3 nm. Crystal structures of the samples and their periodicity were examined by the high- and low angle X-ray diffraction. The analysis of the X-ray data indicates that the resulting film structure is close to amorphous. The films after deposition were characterized by many techniques [2, 3]. In this work, the optical and magneto-optical spectroscopy and magneto-optical magnetometry studies were carried out to study the composition of Fe/Si_xFe_{1-x} MLS spacer layers.

The films were studied experimentally by means of the Kerr effect in polar and longitudinal geometry as a function of magnetic field up to 2.4 T and photon energy in the range of 0.74–5.8 eV. The diagonal and off-diagonal components of the optical conductivity tensor of the MLS were determined from the measured Kerr rotation and ellipticity and the optical data were recorded by spectroscopic ellipsometry. The theoretical spectra were calculated from first principles in the density functional theory by the LMTO method with the use of the supercell approach. Details of the experimental and theoretical procedures have been published elsewhere [9].

3. Results and discussion

Magnetic, optical and magneto-optical properties were studied at room temperature for Fe/Si_xFe_{1-x} MLS with nominal thicknesses d_{Fe} = 3 nm and d_{Si} = 0.9, 1.35, 2.55 nm. Figure 1 shows the dependence of the Kerr rotation on the magnetic field applied parallel and perpendicular to the film surface for three various spacer layer compositions. The measured polar and longitudinal Kerr hysteresis loops for the Fe/Si_xFe_{1-x} MLS films exhibit behaviour typical of the AF coupled sublayers. The strength of the interlayer coupling depends strongly on thickness and composition of the spacer layer and agrees with the results obtained from magnetometry measurements [2]. The in-plane saturation field determines the strength of the AF interlayer exchange coupling. The strongest AF coupling has been found for Fe/Si MLS with nominal Si thickness equal to 1.35 nm. The Kerr rotation dependence on the magnetic field applied perpendicular to the films shows that the Fe/Si sample with the strongest AF has no saturation in the field of up to 2.4 T, suggesting the presence of an important paramagnetic fraction with high susceptibility or strong in-plane anisotropy. As is seen from Fig. 1, the magnetization is in the sample plane. It has been verified experimentally that no easy axis exists in the MLS film planes.

To answer the question which Fe–Si phase promotes the MLS interlayer exchange coupling, the optical properties of the spacer layer prepared by the same technique were investigated at first step. In Figure 2, the measured optical conductivity components are shown for Fe, $Fe_{50}Si_{50}$ and $Fe_{33}Si_{66}$ alloy films, and for Si(100) substrate. The results of the energy dependence of optical conductivity show for both the $Fe_{50}Si_{50}$ and $Fe_{33}Si_{66}$ alloys large and positive dispersive parts, and strongly decreasing absorptive parts with the decrease of photon energy in the IR region of spectra, characteristic of a semiconducting-like behavior. These alloys represent semiconducting ε-FeSi and β-$FeSi_2$ phases rather than metallic α-FeSi and c-FeSi with CsCl structure.

To support the observation, optical conductivity spectra were calculated from first principles for ε-FeSi, β-$FeSi_2$, c-FeSi, and *bcc* Fe. The results are presented in Fig. 3 together with the calculated spectra of the crystalline and amorphous silicon. From the comparison of the recorded and calculated spectra it can be seen that the results confirm the above stated conclusion, despite the existing differences in the amplitudes, widths and energy positions of the spectral peaks. In particular, the optical conductivity spectra of the metallic c-FeSi system exhibit different behaviour in the UV-spectral range as compared to semiconducting ε-FeSi alloy.

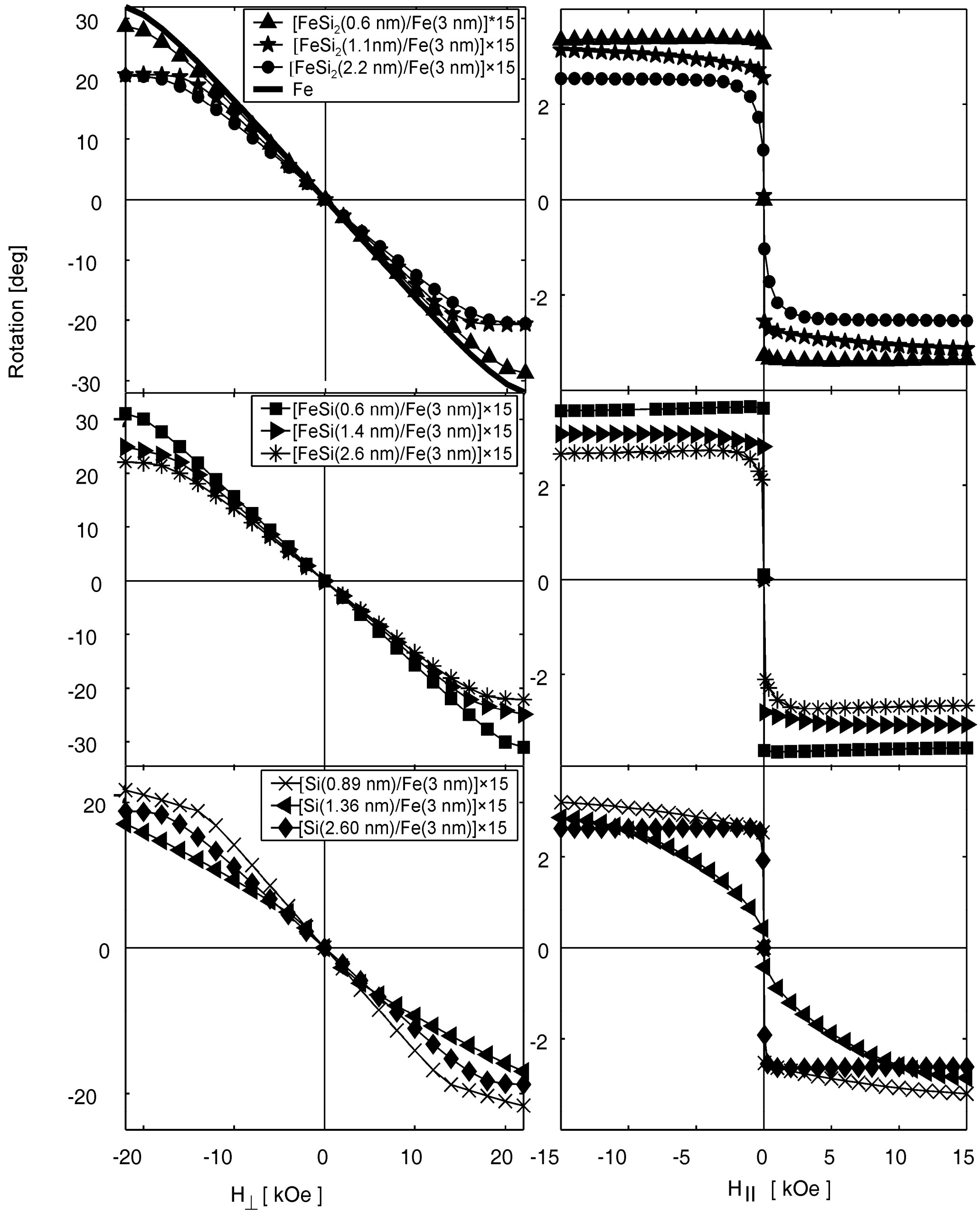

Fig. 1. The Kerr rotation dependence on the magnetic field applied perpendicular (left panel) and parallel (right panel) to the sample surface for the series of Fe/Si_xFe_{1-x} MLS

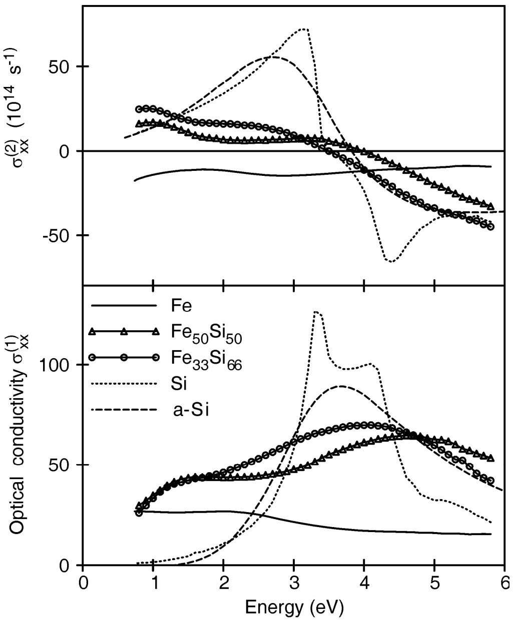

Fig. 2. Recorded spectra of the optical conductivity tensor components for sputter deposited Fe, $Fe_{50}Si_{50}$ and $Fe_{33}Si_{66}$ alloy films, and Si(100) substrate. Data for a-Si (amorphous) were taken from literature

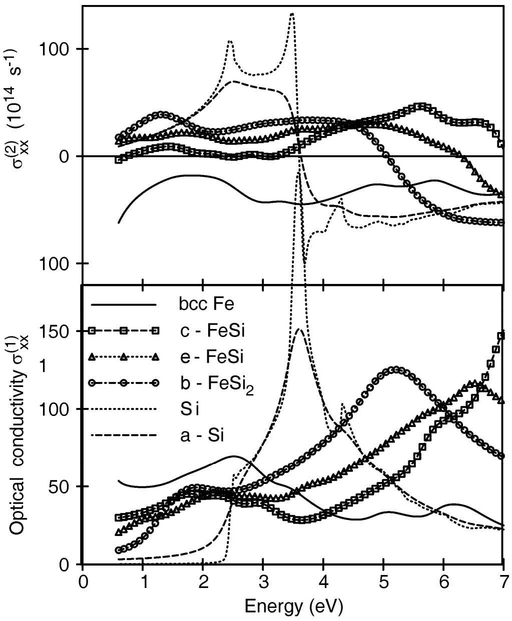

Fig. 3. Calculated spectra of the optical conductivity for Fe, metallic c-FeSi, semiconducting β-$FeSi_2$, ε-FeSi, Si crystal and amorphous a-Si

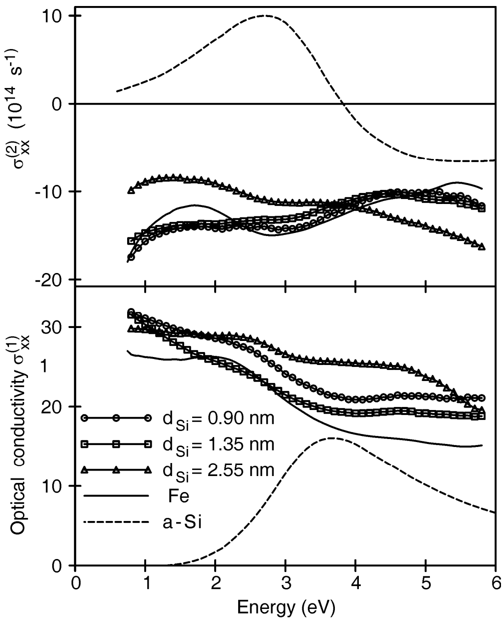

Fig. 4. Measured optical conductivity tensor components for as-deposited Fe/Si multilayers with fixed d_{Fe}= 3 nm and varied d_{Si}. The spectra of the Fe films and bulk amorphous a-Si (multiplied by the factor of 0.18) are included for comparison

The above conclusion obtained from the analysis of the optical conductivity spectra is in line with the results of CEMS study. The CEMS spectra [3] indicate that AF coupling can be mediated by formation of nonmagnetic Fe–Si compounds at the interfaces and/or in spacer layers, e.g., semiconducting ε-FeSi, non-stoichiometric metallic c-Si_xFe_{1-x} and intermixed crystalline or amorphous Fe–Si.

These alloy systems constitute spacer sublayers in the MLS structures. The situation is more difficult to analyze and to determine the exact structure of the interfacial phase because the formed interface region is inhomogeneous. In Figure 4, the exemplary optical conductivity spectra are presented for three Fe/Si MLS with increasing spacer layer thickness. The spectra carry information on the effective optical conductivity of the MLS. As can be seen in Figure 4, this effective optical response, even for the thickest Si spacer, is closer to the Fe spectral dependence. Overall evolution of the conductivity is in agreement with the prediction of effective medium approximation theory [10]. It is clear that, on average, the MLS structures exhibit metallic-like behaviour. This conclusion was verified by multireflection calculation results, not presented in the paper. The optical conductivity spectra of the MLS films prepared with intentionally chosen alloy spacer layers of the $Fe_{50}Si_{50}$ and $Fe_{33}Si_{66}$ compositions have similar to that energy dependences seen in the case of the Si spacer.

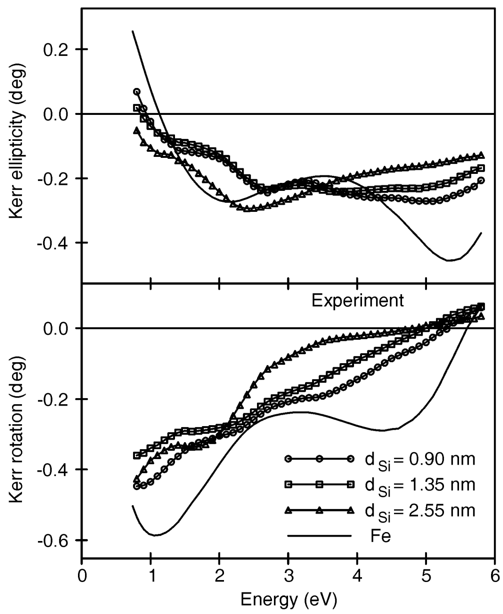

Fig. 5. Recorded complex Kerr rotation spectra for pure iron thick film and Fe/Si MLS with fixed d_{Fe} = 3 nm and varied d_{Si}

The measured magneto-optical response of the Fe/Si MLS is presented in Fig. 5, together with the spectrum of Fe that is the source of magnetism in the system. It is clearly seen from the Kerr rotation that the peaks at ~1.1 eV and ~3.5 eV characteristic of *bcc* Fe disappear and the amplitude of the spectra diminishes, although generally it is not scaled with the Fe content in the whole spectral range. It can be concluded that the main contribution to the magneto-optical response comes from the intrinsic part of Fe sublayers, and iron silicides formed at Fe-Si interface contribute significantly to the spectra. To elucidate the magneto-optical nature of the interface, the first principles calculations are necessary. Such calculations are difficult to perform owing to the large Fe sublayer thickness.

In conclusion, the comparison of the measured and *ab initio* calculated optical conductivity spectra of the alloys shows that the alloys represent semiconducting ε-FeSi and β-$FeSi_2$ phases rather than a metallic FeSi phase, which confirms the conclusions inferred from other methods. The measured magneto-optical response of the MLS studied comes mainly from the intrinsic part of Fe sublayers that mask the contribution of iron silicides formed at the Fe-Si interface. The detailed *ab initio* modeling of the magneto-optical response of complex multilayer structures is in progress.

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