Magnetic properties of ultrathin Fe$_3$Si films on GaAs(001)

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Abstract. For a detailed understanding of the interface between Fe$_3$Si and GaAs, we investigate Fe$_3$Si films in the ultrathin limit down to a few monolayers and compare the results to Fe$_3$Si/MgO(001) which serves as a reference in the present study. From X-ray magnetic circular dichroism measurements we determine averaged spin and orbital magnetic Fe moments. Further insight follows from SPR-KKR calculations. Conversion electron Mössbauer spectroscopy (CEMS) yields information on the chemical ordering and is able to distinguish inequivalent Fe lattice sites. The CEMS results indicate structural disorder which we attribute to an interdiffusion at the Fe$_3$Si/GaAs interface.

1. Introduction
The Heusler-like binary compound Fe$_3$Si has entered into the focus of current research as a promising candidate for spintronic applications. Due to the low lattice mismatch (theoretically 0.1%), GaAs(001) is a suitable substrate for which electrical spin injection from Fe$_3$Si contacts was observed [1]. Most important for improved spin injection efficiency are high quality and control of structural and magnetic properties at the interface. Fe$_3$Si crystallizes in the D0$_3$ structure and can be regarded as a quasi Heusler compound (Fe$_A$)$_2$(Fe$_B$)Si with two inequivalent Fe sites [2, 3]. Fe atoms (Fe$_B$) on sites B have 8 nearest neighbors (nn) Fe. Fe atoms (Fe$_A$) on sites A have only 4 nn Fe while the other 4 nn are Si. Fe$_3$Si can be grown epitaxially on GaAs(001) and MgO(001) [4, 5].

In the present work, we investigated the magnetic moments in epitaxial Fe$_3$Si films on GaAs(001) and MgO(001) by X-ray magnetic circular dichroism (XMCD) spectroscopy and correlate them with the chemical ordering as obtained from conversion electron Mössbauer spectroscopy (CEMS). To complete our study, XAS and XMCD spectra have been calculated within a multiple scattering theory. In the calculations, Ga impurities in Fe$_3$Si have been considered, as CEMS measurements yield evidence for perturbed growth on GaAs.

2. Methods
We deposited Fe$_3$Si films on MgO(001) and GaAs(001)-(4 × 6) by co-evaporation of $^{57}$Fe and Si at a substrate temperature of $T_S = 520$ K in ultrahigh vacuum conditions as described in detail elsewhere [6]. The samples were covered by a 40 Å gold layer to prevent oxidation during transportation to the measuring chamber. We monitored the growth by reflection high energy
Figure 1. XAS (top) and XMCD (bottom) Left: of 57 ML Fe₃Si on MgO and GaAs and a bulk-like Fe reference measured at RT, Right: from calculations considering various concentrations of Ga impurities in bulk Fe₃Si.

electron diffraction (RHEED), thereby confirming the layer-by-layer growth of Fe₃Si(001) on the MgO(001) substrate from the beginning of deposition. On GaAs the first Fe₃Si(001) reflections appear at a film thickness of about 6 ML when the initial islands start coalescing [7].

We measured thickness-dependent XAS and XMCD at the helical undulator beamline UE56/2-PGM 1 at BESSY by detecting the total electron yield at the Fe L_{2,3} edges. The easy axis of magnetization is in the film plane of Fe₃Si [8], we recorded the XMCD spectra at an angle of 20° between the photon wave vector and the surface of the sample. The spectra were divided by the incoming photon flux, i.e. the photocurrent of a Au mesh in the incident X-ray beam, normalized to unity and corrected for saturation effects. We determined spin and orbital magnetic moments per Fe atom averaging over the two different Fe sites by a standard sum rule procedure [9, 10].

Theoretical spectra are obtained from ab initio calculations using the Munich SPR-KKR code within the local density approximation in the parametrization of Vosko, Wilk, and Nussair [11, 12, 13]. To study the influence of Ga impurities on the magnetic structure of Fe₃Si KKR calculations within the coherent potential approximation (CPA) were carried out. The XAS and XMCD have been obtained from an expression based on Fermi’s Golden rule [12]. Further details will be published [6].

CEMS measurements in zero external field at room temperature revealed the chemical ordering of the Fe₃Si films. Due to the different surroundings of Fe atoms of type A and B, they have different hyperfine fields that can be distinguished by CEMS. Therefore it is possible to discriminate contributions from the two inequivalent Fe lattice sites [14]. For the final analysis we fitted the Mössbauer spectra in a two step procedure accounting (i) for highly ordered Fe₃Si (Fe_A and Fe_B sextets) and (ii) for an additional spectral contribution (hyperfine field distribution) that we attribute to an additional, non-Fe₃Si-like Fe phase due to disorder in the film [6].

3. Results
Fig. 1 shows the XAS and the XMCD spectra at the Fe L_{2,3} edges as obtained from measurements (left) and calculations (right). The experimental XAS lines of Fe₃Si are broadened compared to
bulk Fe with a somewhat lower maximum at the Fe \( L_3 \) edge (decrease by \( \sim 8 \% \) on MgO(001) and \( \sim 17 \% \) on GaAs-(4 \( \times \) 6)). We note that on GaAs the Fe-XMCD signal of Fe\(_3\)Si is lower than on MgO. In addition, a shoulder occurs as observed also in other Heusler systems [15]. A standard sum rule analysis yields an averaged total magnetic moment per Fe atom of 1.6\( \mu_B \) on MgO (error bar 10 \%) in agreement with literature values [2]. On GaAs the sum rules yield 1.5\( \mu_B \) per Fe atom – a value that, although still within the error bar of the sum rule analysis, follows the trend of reduced magnetization that the reduced XMCD spectrum indicates. The orbital to spin moment ratio is 9 \% and 6 \% on MgO and GaAs, respectively.

To investigate the reduction of the XMCD signal on GaAs in further detail, we calculated XAS and XMCD considering various various concentrations of Ga impurities in bulk Fe\(_3\)Si. The resulting spectra are shown on the right-hand side of fig. 1. The larger the number of impurities in the samples, the smaller the XAS intensity becomes. In the same manner, the XMCD decreases with increasing Ga concentration. These results indicate, that our experimental finding of reduced XMCD in Fe\(_3\)Si on GaAs may be related to an increased interdiffusion of substrate Ga atoms into the ferromagnetic film. According to the calculations, these impurities preferably tend to occupy Fe\(_A\) sites.

Fig. 2 (left) shows the CEM spectra of 57 ML Fe\(_3\)Si films on MgO and GaAs. The two samples were grown simultaneously so that the growth conditions for both samples were identical. Already from the bare experimental data (solid circles in the figure) it becomes obvious that the chemical ordering of Fe\(_3\)Si is higher on MgO than on GaAs. The Mössbauer lines are sharper and more pronounced. On MgO a satisfactory fit can be obtained from a calculated spectrum for nearly perfectly ordered Fe\(_3\)Si. No additional spectral contribution is required. In contrast, on GaAs a second sub-spectrum is needed to account for the non-perfect order in the film. From our fitting routine, that is based on the work by Arita and coworkers [16], we obtain long range order parameters \( S(D0_3) \) and \( S(B2) \) of 0.83 and 0.52, respectively, on MgO and 0.68 and 0.32, respectively, on GaAs. In the ideal case they should be \( S(D0_3) = 1 \) and \( S(B2) = 2/3 \) for Fe\(_3\)Si with perfect D0\(_3\) structure. This larger disorder in Fe\(_3\)Si on GaAs may explain the experimentally observed reduction of XMCD, possibly as a result of diffusion of substrate atoms into the Fe\(_3\)Si film. As discussed above, our calculations with Ga impurities support this explanation. In contrast, on MgO the Fe\(_3\)Si films appear quite well ordered and
may thus serve as a reference standard.

On the right-hand side of fig. 2 we plot the thickness dependence of the XMCD spectra of $\text{Fe}_3\text{Si}$ on GaAs measured at $T = 40$ K. The corresponding XAS (not shown) are normalized to unity. Within the experimental error they are identical to the one of 57 ML $\text{Fe}_3\text{Si}$. These thickness-dependent measurements show a clear decrease of the XMCD intensity and thus of the magnetization with decreasing thickness. Two possible reasons may lead to this reduction: (i) the well known finite size effect of thin film, and (ii) interdiffusion of substrate atoms into the film. Up to now, our investigations do not allow for discrimination between these two origins. In the future, CEMS with $^{57}\text{Fe}_3\text{Si}$ tracer layers at the interface will contribute to the clarification.

4. Conclusion

In conclusion, XMCD and CEMS together with DFT calculations yield a deeper insight into the relation between structural and magnetic properties of $\text{Fe}_3\text{Si}$ on GaAs and MgO. The case of $\text{Fe}_3\text{Si}$ on GaAs is especially interesting with respect to future spintronic devices, where ferromagnet/semiconductor interfaces play an important role. Our investigations yield evidence of disorder and/or interdiffusion at the interface of $\text{Fe}_3\text{Si}$/GaAs, while the magnetic moments of Fe are not much influenced. Thickness-dependent XMCD measurements show a decrease of the magnetization at low temperature with decreasing thickness. Further investigations will follow, e.g. by CEMS with tracer layers at the interface to clarify the origin of this reduction.

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