Chapter 7

Summary, conclusion and outlook

Motivated by the lack of microscopic data and by the possible relevance for spintronic applications the centrepiece of this thesis presents a multitechnique experimental study of ultrathin layers of Fe$_3$Si epitaxially grown on GaAs(001) and GaAs(110). Emphasis was laid on the extraction of structural and magnetic properties between which, where possible, correlations were elaborated.

Apart from the analysis of Fe$_3$Si/GaAs, a substantial amount of work done within the framework of this thesis was dedicated to modifications of the experimental setup as described in section 4.2. For the fabrication of Fe$_3$Si layers a stable silicon MBE source was build up on the basis of an electron beam evaporator. Both hardware and software were developed for attaining reproducible compositions of the iron silicide. In this context, the use of secondary substrates consisting of MgO for the determination of the exact composition by means of RBS was established. Although the resulting MBE setup allows the fabrication of iron silicide layers with arbitrary compositions, the analyses presented in this work are restricted to films close to stoichiometric Fe$_3$Si. Further modifications were done by enhancing the UHV system with a flow cryostat that can be used both for MOKE measurements as well as for film growth at low temperatures. Due to the fact that one part of the UHV chamber needed to be reconstructed in order to accommodate the flow cryostat, a revision of the MOKE setup associated with the UHV chamber was done at the same time. As a result, the revised MOKE setup provides a stronger electromagnet, easier sample transfer and sample alignment and, due to an increase of the angle of incidence of the MOKE laser beam onto the sample, an increased sensitivity to the magneto-optical Kerr effect. First experiments involving the use of the cryostat were shown in section 5.4.1 where the shape parameter of the temperature dependent magnetization according to the semi-empirical description by Kuz’min \cite{Kuz05, Kuz06} was determined for a 40 ML Fe$_3$Si/GaAs(001) sample. The current setup allows MBE growth and MOKE measurement at sample temperatures down to 96 K when operating the cryostat with liquid nitrogen. According to the considerations in section 4.2.2 it can be estimated that even without the use of cryogenic shields temperatures of approximately 30 K can be reached by using liquid helium as the
The main part of this thesis described the structural and magnetic properties of Fe$_3$Si/GaAs(0 0 1) with varying adsorbate layer thicknesses. The starting point for the structural analysis was the optimization of the layer morphology as a function of the growth parameters and post annealing processes under the observation of STM and LEED. This was motivated by the large range of growth parameters that can be found in literature (see section 1.1) and by the lack of microscopic data. In the literature it was already indicated by Monte Carlo simulations [Kag09] that a surface morphology corresponding to a pseudo Frank-van der Merwe growth mode can be achieved for layer thicknesses of above approximately 10 ML. However, at least in terms of the atomic ordering at the surface, it is found that this cannot be performed in a one-step process. On the other hand, the thermal energy at lower growth temperatures is insufficient to induce atomic ordering. On the other hand, the deposition at elevated growth temperatures leads to a layer morphology in which trenches down to the substrate level persist up to higher film thicknesses. The STM analysis points out that the layer morphology can be optimized by post-annealing the samples at 300 °C following the film growth where a deposition rate of 0.1 nm min$^{-1}$ and a growth temperature of 200 °C were found to be most suitable. The study of the influence of post-annealing also revealed that although the D0$_3$ structure of Fe$_3$Si/GaAs(0 0 1) was shown to be stable up to 425 °C [Her05] an incipient break-up of the Fe$_3$Si layer can already be observed at 400 °C or even below. After post annealing at an even increased temperature of 500 °C further breaking up of the layer can be observed alongside the formation of two different types of alloying between the substrate and the adsorbate. 

By means of STM, the surface atomic structure of Fe$_3$Si/GaAs(0 0 1) could be imaged in real space for the first time. The microscopic data is in agreement with the D0$_3$ structure of Fe$_3$Si but it also points out that regardless of the applied STM bias voltage always one and the same sublattice is imaged. This sublattice is tentatively assigned to the Si species as the analyses of different samples with slight compositional variations lead to this assumption. Up to now, there remains a discrepancy with STM simulations in terms of a contrast inversion which was discussed in section 5.3.1. The atomic scale images were furthermore used to assign surface defects which were previously reported in a study based on XRD measurements [Kag08]. Besides the obvious appearance of antisite defects the STM data points out that the majority of disorder must be ascribed to faulted stacking sequences of
larger areas which leads to a splitting of the iron silicide into bcc iron and B2 Fe-Si unit cells instead of the D0₃ structure.

While the STM study of the system Fe₃Si/GaAs(001) was clearly motivated by the lack of real space data there were also different incentives for its magnetic characterization. For one, it was essential to obtain a magnetic characterization of the same samples of which the structural properties were considered in order to make reliable statements about the correlations of these properties. Moreover, despite the fact that a lot of information about the magnetic behaviour of Fe₃Si/GaAs(001) can be gathered from literature as summarized in section 1.1, many open-ended questioned such as the origin of the uniaxial magnetic anisotropy or the thickness dependent magnetic moment remain up to date. The conclusions that are drawn in this thesis do not claim to ultimately answer these questions but to provide information based on the connections that can be established between structural and magnetic properties.

In situ MOKE measurements were used to obtain hysteresis loops and anisotropy plots on the basis of the angular dependent normalized magnetic remanences. Beyond that, the in situ MOKE measurements were able to point out that the method used for capping iron layers [Elm88, Spo04, Urb05], i.e. the deposition of Ag and Au layers with thicknesses of 2.5 nm, respectively, can also be applied to Fe₃Si layers as no notable influence on the hysteresis loops and on the anisotropy plots could be observed. More quantitative data could be obtained from ex situ SQUID magnetometry and FMR measurements. The average magnetic moment per atom of the Fe₃Si layers is found to be enhanced above the bulk value below thicknesses of 40 ML. A peak value of $(2.14 \pm 0.2) \mu_B$ at room temperature is found at a thickness of 5 ML. While it is possible that part of this increase can be ascribed to surface and interface anisotropies the STM data support an increase caused by unquenched orbital moments at the surface due the significantly higher densities of step edges at low coverage. It was furthermore possible to determine the thickness dependences of the magnetic anisotropy constants. The experimental data confirmed that the overall magnetic anisotropy is a superposition of the magnetocrystalline and a uniaxial magnetic anisotropy. Similar to [Her08] a reorientation of the UMA is observed. This points out that there is a finite volume contribution to the UMA the origin of which, however, still remains unknown. For thicker films above 40 ML the magnetocrystalline anisotropy constant $K_1$ reaches a bulk-like value.

One point of interest in this work consisted of the structural and magnetic properties at a coverage both below the onset of ferromagnetism and below the thicknesses at which the growth proceeds quasi layerwise. Here, the experimental data point out that the 3D island-wise growth is strongly linked to the magnetic behaviour which is comparable to a system of superparamagnetic nanoparticles. In fact, the superspin derived from a Langevin fit to the magnetization loop can clearly be brought into agreement with the average magnetic moment per atom and the size of the clusters as measured by STM (see section 5.5.1). In comparison to Fe₃Si/GaAs(001) of which many aspects have been studied to a large
extent the structural and magnetic data on Fe\textsubscript{3}Si/GaAs(110) represent the first experimental work that has been done so far. The growth parameters and the post annealing procedure which resulted in an optimized layer morphology in the case of Fe\textsubscript{3}Si/GaAs(001) were also applied to Fe\textsubscript{3}Si/GaAs(110). At a film thickness of 56 ML, where according to STM data one monolayer can be appointed as approximately 0.2 nm, the closest possible spacing between the (110) oriented atomic layers, the Fe\textsubscript{3}Si film covers the substrate entirely. However, compared to Fe\textsubscript{3}Si/GaAs(001) the surface is found to be rougher. STM and LEED data could furthermore determine that the crystallographic orientation of the substrate is sustained by the adsorbate, hence (110)[001]GaAs∥(110)[001]Fe\textsubscript{3}Si. In situ MOKE measurements point out that at a thickness of 28 ML the overall magnetic anisotropy of Fe\textsubscript{3}Si/GaAs(110) is strongly dominated by a uniaxial magnetic anisotropy while at a thickness of 112 ML it corresponds to the magnetocrystalline anisotropy. For low coverage, theoretical considerations determined an augmented average magnetic moment per atom and predicted that the bulk magnetic moment would be approached for higher thicknesses \cite{Her08a}. The latter could be verified by SQUID magnetometry measurements which at a thickness of 56 ML yielded an average magnetic moment per atom of \((1.06 \pm 0.20)\) µ\textsubscript{B} at room temperature.

In summary, structural and magnetic properties of Fe\textsubscript{3}Si/GaAs have been investigated upon establishing the technical requirements by modifying the experimental setup. Due to the fact that the series of experiments such as the optimization of the layer morphology or the thickness dependent analyses of structural and magnetic properties were time-consuming there remain projects that could not be accomplished within the scope of this thesis and which are therefore listed here as an outlook.

Throughout this work the influence of the substrate surface reconstructions in the case of GaAs(001) was neglected. Although it can be assumed that this is appropriate for thicker films it cannot be ruled out that influences on the structural and magnetic properties might be observed. This could be studied by varying the substrate surface reconstructions as a result of different preparation methods.

The magnetic analyses showed that a uniaxial anisotropy can be observed both for Fe\textsubscript{3}Si/GaAs(001) and Fe\textsubscript{3}Si/GaAs(110). Under the consideration of literature \cite{Len05} it can be summarized that the origin of the UMA might be ascribed to different mechanisms, namely the magnetoelastic anisotropy and anisotropic bonding at the interface the magnitude of which could be influenced by the exact silicon content of the Fe\textsubscript{3}Si adsorbate layers. This could be systematically investigated by using substrates with variable lattice constants like Ga\textsubscript{As\textsubscript{x}}P\textsubscript{1-x} as mentioned in section \ref{sec:6.3} and by varying the iron silicide composition.

Regarding Fe\textsubscript{3}Si/GaAs(110), clearly a more detailed and especially a quantitative magnetic analysis needs to be done in order to obtain a comprehensive description. Furthermore, the growth parameters were directly transferred from the Fe\textsubscript{3}Si/GaAs(001) samples. Considering that the diffusion barrier of the (110) oriented substrates is most likely to differ
from the (100) oriented substrates an independent optimization of the growth parameters is required for Fe$_3$Si/GaAs(110).

In the introduction it was already mentioned that the system Fe$_3$Si/GaAs is a promising candidate for spintronic applications, e.g. due to the quasi half-metallic behaviour of the Heusler alloy Fe$_3$Si. In total, the present work confirms this. The thickness dependent magnetic moments indicate a sharp interface with the substrate or, in other words, that magnetically dead layers which would counteract spin injection are not likely to be formed. Fe$_3$Si layers were furthermore proven to be thermally stable up to approximately 400$^\circ$C.

It was also shown in this thesis that under the appropriate choice of growth parameters atomically flat layers of Fe$_3$Si can be grown on GaAs(001) which is a good premise to create multilayer systems for technological applications. Although the surface was found to exhibit a slightly higher roughness it could be shown that closed layers of Fe$_3$Si can also be grown on GaAs(110) where the epitaxial relation is sustained across the interface. The magnetic anisotropy, which as in the case of Fe$_3$Si/GaAs(001) is a superposition of the magnetocrystalline anisotropy and a uniaxial magnetic anisotropy, can be tailored by changing the film thickness of the ferromagnet.