As-grown thin epitaxial Fe$_3$Si films fabricated by molecular beam epitaxy on GaAs(001) are studied by grazing incidence x-ray diffraction. The long-range order parameters of the films of different stoichiometry are determined by measuring fundamental and superlattice crystal truncation rods and comparing them to simulations in dynamical approximation. Two order parameters, associated with the degree of migration of Si atoms into different Fe sublattices, are obtained. A residual intermixing of the sublattices is found, even near complete stoichiometry. The relative positions of the Fe$_3$Si and GaAs lattices are determined. Fe atoms in Fe$_3$Si are located at the positions of the Ga atoms in GaAs, with an additional shift of 0.1±0.04 Å of the tetragonally distorted Fe$_3$Si lattice normal to the interface.

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I. INTRODUCTION

The combination of magnetic and semiconducting materials opens up new pathways in the development of semicon-ductor devices utilizing the spin of the carriers.\textsuperscript{1} From the viewpoint of such spintronic device structures, it is highly desirable to explore epitaxial ferromagnet/semiconductor heterostructures that possess a rather perfect interface\textsuperscript{2} structure as well as high thermal stability. Fe$_3$Si on GaAs is a promising candidate for spintronic applications.\textsuperscript{3–5} It can be grown by molecular beam epitaxy at GaAs substrate temperatures near 200 °C.\textsuperscript{6} The Curie temperature of Fe$_3$Si is as high as 840 °C.\textsuperscript{7} Spin injection at room temperature has been observed.\textsuperscript{8} The interface between Fe$_3$Si and GaAs is found to be smooth and of good structural perfection: Fe$_3$Si on GaAs can be grown without misfit dislocations at the heterointerface.

Fe$_3$Si has the face-centered cubic D0$_3$ structure.\textsuperscript{9,10} This structure (see Fig. 1) can be considered as four interpenetrating fcc sublattices A, B, C, and D with origins at A(0, 0, 0), B(0.25, 0.25, 0.25), C(0.5, 0.5, 0.5), and D(0.75, 0.75, 0.75). In the ordered Fe$_3$Si crystal, Fe atoms occupy the three sublattices A, B, and C, while Si atoms fill the sublattice D. Bulk Fe$_3$Si crystals acquire a long-range order of the sublattices after an annealing in vacuum at 600 °C for about two hours.\textsuperscript{11} The ordered alloy can be regarded as a magnetic Heusler alloy Fe$_2$B(Fe$_2$A,C)$_2$Si(D) with the two distinct Fe sites (A,C) and (B).\textsuperscript{10,12} The order in Heusler alloys\textsuperscript{13} has a significant influence on their electric and magnetic properties: the magnetic moments of the Fe atoms at the positions Fe(A,C) linearly depend on the number of nearest Fe neighbors.\textsuperscript{14,15} Thus, the magnetic properties can be tuned: some of the local Fe magnetic moments are almost as large as in pure bcc iron, while others are reduced due to the presence of Si. Moreover, the introduction of small amounts of transition metal impurities into the Fe$_3$Si matrix opens up new possibilities\textsuperscript{11,16} to tune the magnetic properties. Impurities to the left of Fe in the periodic table prefer the Fe(B) site with eight first-neighbor Fe atoms. Those beneath and to the right of Fe in the periodic table enter the Fe(A, C) site with four Fe and four Si atoms as nearest neighbors. A highly ordered and stoichiometric Fe$_3$Si matrix is the starting point for most experiments with bulk Fe$_3$Si.

In as-grown thin epitaxial films of Fe$_3$Si on GaAs, the lattice mismatch and the long-range order depend on the stoichiometry. Significant changes of the saturation magnetization and the resistivity with film stoichiometry have been observed.\textsuperscript{17} While the saturation magnetization changes almost linearly with composition and the coercive field remains unchanged over a wide range of compositions, a clear minimum of the resistivity $\rho$ near stoichiometric conditions is found. It has been suspected that the minimum is due to alloy scattering, which is smallest for an ordered crystal structure, but detection of the degree of the order has been lacking. Moreover, an additional contribution to the planar Hall effect was recently found, which reflects magnetic in-
TABLE I. Structural parameters and the low temperature resistivities of the samples under investigation.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Si cell temperature (°C)</th>
<th>Si content in Fe$_3$Si (%)</th>
<th>Layer thickness (nm)</th>
<th>Misfit (%)</th>
<th>Order parameters $\alpha$ to Fe (B)</th>
<th>$\beta$ to Fe (A, C)</th>
<th>Relaxation $z$ Å</th>
<th>Resistivity (µΩ cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1395</td>
<td>25.5</td>
<td>32.6</td>
<td>−0.06</td>
<td>0</td>
<td>0.3</td>
<td>0.14</td>
<td>23</td>
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<tr>
<td>2</td>
<td>1390</td>
<td>22.5</td>
<td>28.4</td>
<td>0.15</td>
<td>0.25</td>
<td>0.28</td>
<td>0.06</td>
<td>50</td>
</tr>
<tr>
<td>3</td>
<td>1375</td>
<td>16.5</td>
<td>28.2</td>
<td>0.62</td>
<td>0.25</td>
<td>0.5</td>
<td>0.08</td>
<td>103</td>
</tr>
</tbody>
</table>

interactions with reduced crystal symmetry for certain Fe sublattices in ordered Fe$_3$Si on GaAs.$^{18}$

Our aim in the present work is the determination of long range order in thin Fe$_3$Si films by grazing incidence x-ray diffraction (GID) measurements. We use the same measurements for a precise determination of position of the Fe$_3$Si unit cell with respect to the GaAs substrate. We utilize the method of grazing incidence diffraction that is ideally suited for thin film investigation, since it gives access even to those reflections that cannot be measured by conventional diffraction methods.

II. EXPERIMENTAL

The Fe$_3$Si films were grown by solid source molecular beam epitaxy (MBE). The GaAs(001) templates were prepared in a separate III-V growth chamber using standard GaAs growth techniques. The sample was then transferred into an As-free chamber through UHV for the Fe$_3$Si deposition. The Fe$_3$Si layers were grown$^6$ on the As-rich GaAs surface at 200 °C with a growth rate of 24 nm h$^{-1}$. The Si cell temperature was varied in order to obtain different degrees of stoichiometry of the films. The Si content of the film was obtained$^6$ from the position of the Fe$_3$Si layer peak on the x-ray diffraction curve. For the present study, we have chosen three samples with different Si contents; see Table I. Samples 1 and 2 are nearly stoichiometric, sample 1 with a small Si surplus, sample 2 with a slight Fe excess. Sample 3 is clearly nonstoichiometric.

The x-ray measurements were performed at the wiggler beamline U125/2 KMC (PHARAO) at the storage ring BESSY in Berlin, Germany. A double crystal Si(111) monochromator was used. The energy of the radiation was 10 keV. A six-circle x-ray diffractometer$^{19}$ was used for grazing incidence diffraction measurements. The incidence angle was 1.2°. The acceptance angle of the detector was 0.1° perpendicular to the surface and 1° (open slits) parallel to the surface. Additional conventional coplanar x-ray diffraction measurements were made using a Panalytical X-Pert system with Ge (220) hybrid monochromator and CuKα$_1$ radiation.

III. THEORY

The stoichiometric compound Fe$_3$Si possesses three types of reflections. Fundamental reflections, which are not influenced by disorder, are given by $H+K+L=4n$, where $n$ is an integer and (HKL) are the Miller indices of the diffracting net planes. The structure amplitude of these reflections is

$$F_0 = 4(f_{\text{Si}} + 3f_{\text{Fe}}),$$

where $f_{\text{Si}}$ and $f_{\text{Fe}}$ are the atomic scattering factors of the respective elements.

The disorder is described by two order parameters $\alpha$ and $\beta$, which are fractions of Si atoms occupying the Fe(B) and Fe(A,C) sites, respectively. There are two distinct types of superlattice reflections.$^9$ Reflections with odd $H,K,L$ are sensitive to both types of disorder, the structure amplitude being

$$F_1 = 4i(1 - 2\alpha - \beta)(f_{\text{Si}} - f_{\text{Fe}}).$$

Reflections given by $H+K+L=2n$ (where $n$ is an integer) are sensitive to disorder in the Fe(A,C) sublattice,

$$F_2 = -4(1 - 2\beta)(f_{\text{Si}} - f_{\text{Fe}}).$$

In order to obtain the order parameters $\alpha$ and $\beta$, we measure diffraction curves of all three types of reflections and fit them by dynamical diffraction calculations. They are obtained for each sample by a simultaneous fit of the diffracted intensities of all the measured Fe$_3$Si peaks. The epitaxial layers themselves are thin enough and their diffraction peaks can be well fitted in the kinematical approximation. However, we want to fit simultaneously the layer and the substrate peaks, the latter serving as an internal reference to obtain the absolute values of the structure amplitudes (1)–(3). The dynamical calculation automatically solves this problem.

The dynamical diffraction theory for layer structures is well established, and we use the well-known algorithm described in Ref. 20 and references therein. One point of the dynamical calculations needs a special discussion. The scattering properties of each layer are described by the Fourier components of the polarizability $\chi_{\text{H}}$. In the computer programs for the analysis of x-ray diffraction experiments available so far, the coefficients $\chi_{\text{H}}$ are considered as eigenproperties of each layer. That means that the origin of the unit cell to calculate $\chi_{\text{H}}$ is chosen independently for each layer, usually according to the symmetry of the respective layer. The phase factor $\text{exp}(i\mathbf{H} \cdot \mathbf{R})$ that arises if the origin is shifted by a real-space distance $\mathbf{R}$ is not taken into consideration. Such a phase factor does not appear if all epitaxial layers have a similar structure (an AlAs/GaAs superlattice would be a simple example). However, if the structures are different, the relative positions of the unit cells are not known in advance and an additional phase factor in the polarizability of one layer with respect to another may appear.
The problem is resolved by choosing a common origin for all structures. This is a commonplace in surface diffraction, but not taken into consideration in practical dynamical diffraction calculations. Figure 2 shows the influence of the relative shift of the crystal lattices on the diffraction curves. We compare a crystal truncation rod (CTR) scan at the 111 reflection calculated for relative positions of the Fe$_3$Si and GaAs lattices shown in Fig. 1 with the same scan for the case when the Fe$_3$Si lattice is shifted by 1/2 of the unit cell in the interfacial plane. The corresponding shift of the layer thickness oscillations is obvious. Thus, we have included the relative positions of the layer and the substrate crystal lattices in the fits to obtain the registry of the layer on the substrate. The $z$ component (normal to the interface) of the shift is a relaxation at the interface. A similar analysis is commonly performed in x-ray surface structure studies to obtain the registry of a reconstructed layer with respect to the bulk structure, but has not been applied to studies of epitaxial layers so far.

**IV. RESULTS**

Figure 3 combines CTR scans performed on sample 1 at different bulk reflections. The width of the GaAs substrate peak is limited by the resolution of the experimental setup. The Fe$_3$Si layer peak is broadened due to the small thickness of the layer, and evidencing periodic side maxima thanks to the high quality of its top and bottom interfaces (see Table I). The layer peak is fairly close to the substrate peak, indicating that the sample is nearly stoichiometric. The 004 and 022 reflections are not sensitive to disorder. The 002 and 222 reflections are influenced by disorder in the Fe(A,C) sublattice, see Eq. (3), and the 111, 311, and 113 reflections are sensitive to disorder in both sublattices Fe(B) and Fe(A,C), as given by Eq. (2). A well-pronounced Fe$_3$Si layer reflection in all these scans indicates a high degree of long-range structural order in the layer.

All measured curves were fitted simultaneously by the dynamical diffraction calculations. We obtain the order parameters $\alpha$ and $\beta$ from the relative intensities of the layer peaks (with the substrate peak serving as an internal reference), the layer thickness from the oscillation period and the registry of the lattices from the phase of the oscillations. The order parameters are found to be $\alpha=0$ and $\beta=0.3$ (see Table I) with an accuracy of $\pm 0.05$. Thus, the as-grown Fe$_3$Si layer of sample 1 is not fully ordered, although it is nearly stoichiometric. Approximately 30% of Si has left its sublattice $D$ and exchanged with the Fe(A,C) atoms. The Si atoms did not exchange with Fe(B) atoms. This is an unexpected result, since in Fe$_3$Si powder samples an excess of Si was found to replace the Fe(B) sites.

From the positions of the intensity oscillations, we find that the Fe$_3$Si and GaAs lattices are positioned with respect to each other, as shown schematically in Fig. 1, i.e., the Fe(A) atoms at the positions of Ga atoms. The Fe$_3$Si unit cell is found to be shifted up vertically along $z$ by 0.14Å with respect to the GaAs unit cell. No signatures of an interface reconstruction are found.

Figure 4 shows the CTR scans of sample 2. The Fe$_3$Si film of sample 2 contains 22.55% Si, i.e., it is nearly stoichiometric with a small deficit of Si. The fundamental 022 and 004 reflections are similar to those of sample 1, as expected. A large amplitude of the thickness oscillations in the 002 reflection and weak contrast in the 111 reflection indicates that the disorder mainly involves the order parameter $\alpha$. The simultaneous fit of all experimental curves gives $\alpha = 0.25$ and $\beta = 0.28$ with an accuracy of $\pm 0.05$. Thus, sample 2 still shows some long-range order with considerable disorder. In this case about 25% of the Si atoms migrate into the
Fe(B) sublattice, whereas 28% of the Si atoms migrate into the Fe(A,C) sublattices. Figure 5 shows analogous scans of sample 3. The film contains only 16.5% Si, i.e., it is highly nonstoichiometric. The fundamental reflection 022 shows the layer peak with the adjacent thickness oscillations. All the measured superlattice reflections (002, 111, 131, and 311) of the layer vanish and only the GaAs substrate peaks remain. The fits to the experimental curves give $\alpha=0.25$ and $\beta=0.5$, i.e., a complete intermixing of Fe and Si in the sublattices. The film does not show any long-range order. Thus, the long-range order in Fe$_3$Si epitaxial layers strongly depends on their stoichiometry. Near stoichiometry the layers are ordered. It is important to note that the almost complete long-range order is observed without additional thermal treatment, which is usually required for the preparation of bulk samples. The slight remaining disorder may be due to the relatively low growth temperature, which has been chosen to avoid the possible intermixing of Fe and/or Si with Ga and/or As at the ferromagnet/semiconductor interface.

V. SUMMARY

In conclusion, we analyzed MBE-grown Fe$_3$Si films on GaAs using grazing incidence x-ray diffraction. We characterized the relative positions of the film and the substrate lattices as well as the long-range order in films with different stoichiometry. The Fe(A) atoms are positioned above the Ga atoms, as shown in Fig. 1. The Fe$_3$Si lattice is shifted with
respect to the GaAs lattice by 0.1±0.04 Å normal to and away from the interface. Long-range order is observed in the nearly stoichiometric as-grown films without additional thermal treatment, which seems to be required for the preparation of bulk samples. A slight deficit of Fe in the epitaxial Fe₃Si layer leads to the intermixing of Si and Fe from the Fe(A,C) sites. A small excess of Fe gives rise to the intermixing of Si and Fe on all the Fe sublattices. Better long-range order in the Fe₃Si films leads to lower resistivity $R_S$.

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