I. INTRODUCTION

Epitaxial growth of Fe on GaAs(001) was demonstrated long ago. A renewed interest in this material arose since it has been established as a prototype system for successful spin injection from a ferromagnet into a semiconductor, which is a key requirement for the development of spintronic devices.

An unexpected in-plane uniaxial magnetic anisotropy (UMA) superimposed on the expected fourfold (cubic) anisotropy was recognized in Fe/GaAs(001) from the beginning, which dominates in ultrathin films of 20 monolayers (ML) (atomic ML) and less. It was subsequently confirmed in numerous studies. After early confusion about the crystalline axes, a general agreement emerged that the cubic axes of the Fe and GaAs lattice were parallel to the respective GaAs axes. Magnetization loops, \( M(\mathbf{H}) \), measured in UHV (\( p = 10^{-10} \) mbar) at 550°C with simultaneous Ar ion sputtering was applied until the reflection high energy electron diffraction (RHEED) pattern of the substrate showed sharp spots on Laue circles, as shown in Fig. 1(a), indicative of a clean and flat surface. Our measurements were performed on GaAs surfaces with a predominant (4 \( \times \) 6) surface reconstruction. The presence of atomically flat terraces several 100 nm wide separated by ML steps was verified by \textit{in situ} STM, as shown in Fig. 1(b). After the growth of Fe and Fe\textsubscript{32}Co\textsubscript{68} films of 30 ML and 100 ML by MBE at 300 K a 20-ML-thick protective layer of Au(001) was deposited. The cubic axes of the Fe and Fe\textsubscript{32}Co\textsubscript{68} lattice were parallel to the respective GaAs axes. Magnetization loops, \( M(\mathbf{H}) \), of Fe/GaAs(001) substrates by molecular beam epitaxy (MBE) using previously developed procedures.

II. EXPERIMENT

Fe(001) and Fe\textsubscript{32}Co\textsubscript{68}(001) films were epitaxially grown on GaAs(001) substrates by molecular beam epitaxy (MBE) using previously developed procedures. To facilitate the measurements of magnetoelastic stress from the substrate curvature, a commercial GaAs wafer was thinned to \( t_s = 180 \) \( \mu \)m and cleaved into stripes of 2 mm \( \times \) 12 mm. Annealing in UHV (\( p = 10^{-10} \) mbar) at 550°C with simultaneous Ar ion sputtering was applied until the reflection high energy electron diffraction (RHEED) pattern of the substrate showed sharp spots on Laue circles, as shown in Fig. 1(a), indicative of a clean and flat surface. Our measurements were performed on GaAs surfaces with a predominant (4 \( \times \) 6) surface reconstruction. The presence of atomically flat terraces several 100 nm wide separated by ML steps was verified by \textit{in situ} STM, as shown in Fig. 1(b). After the growth of Fe and Fe\textsubscript{32}Co\textsubscript{68} films of 30 ML and 100 ML by MBE at 300 K a 20-ML-thick protective layer of Au(001) was deposited. The cubic axes of the Fe and Fe\textsubscript{32}Co\textsubscript{68} lattice were parallel to the respective GaAs axes. Magnetization loops, \( M(\mathbf{H}) \), of Fe/GaAs(001) substrates by molecular beam epitaxy (MBE) using previously developed procedures.

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An early speculation about the presence of oriented defects was soon discarded. Two different ideas have emerged about the origin of the UMA: (i) magnetoelastic coupling due to the lattice mismatch between GaAs and Fe and resulting anisotropic strain, or (ii) electronic hybridization between Fe and GaAs. The conclusion is that the magnetoelastic interaction—while possibly present—is not the dominating contribution to the uniaxial magnetic anisotropy in these systems.
of the films were measured by longitudinal magneto-optic Kerr effect (MOKE), superconducting quantum interference device (SQUID), alternating gradient magnetometry (AGM), and vibrating sample magnetometry (VSM). Anisotropy constants were determined from the $M(H)$ loops, as described in Sec. III. The magnetoelastic coupling constant $B_2$ was measured by using a cantilever method.\textsuperscript{15,16} The film-GaAs substrate composite stripe was clamped at one end to a sample manipulator in air and alternately magnetized along its short and long side. The external magnetic fields up to 85 mT ensured magnetic saturation along the width (i.e., [110]) and length (i.e., [110]) of the film, as controlled by simultaneous longitudinal MOKE measurements. The in-plane magnetization reorientation induced a corresponding change of the magnetoelastic stress, which induced a change of curvature $\Delta \frac{1}{R}$ on the order of 1/5000 m$^{-1}$. From the change in curvature measured by the deflection of two laser beams the magnetoelastic coupling coefficient, $B_2$ was determined\textsuperscript{3,4} according to $B_2 = \frac{B}{t} \Delta \frac{1}{R}$, where the Young modulus of the GaAs substrate along [110], $Y$, and its Poisson ratio along [110], $\nu$, were given by $Y = 121.3$ GPa and $\nu = 0.021$;\textsuperscript{17} $t_S$ and $t$ are the thickness of the substrate and the ferromagnetic film, respectively.

Our technique measured a curvature change of a thin substrate, which was induced by the magnetoelastic stress. This curvature led to a negligible change of film strain. The data of Fig. 4 indicate a radius of curvature on the order of 5000 m, the substrate thickness on the order of 0.0001 m, thus the stress-induced curvature change corresponded to a change of surface strain on the order of $2 \times 10^{-8}$. Thus, although the substrate curved, the resulting change of strain is 6 orders of magnitude smaller than typical misfit strains. Therefore, the technique measured the magnetoelastic coupling coefficient $B_2$ at a constant strain, which was given by the growth conditions. The curvature measurement technique has been used successfully to measure $B_i$ in single ultrathin films, as demonstrated in Refs. 15 and 16.

### III. EVIDENCE FOR UMA FROM MAGNETIZATION MEASUREMENTS

To check the presence of magnetic anisotropies and to determine the respective anisotropy constants magnetization loops, $M(H)$, were measured by longitudinal MOKE, SQUID, and VSM for different in-plane angles $\phi$ between the measurement and field axis and the [110] axis. The magnetizing energy was determined by integrating the anhysteretic $M(H)$ loops, as demonstrated in Fig. 2 for a 14 ML Fe film on GaAs(001). The (red/dark gray) hatched area represents $W_{\text{mag}}$ along [110], which is indeed free of hysteresis. In other cases the hysteresis was removed by averaging between the right and the left branch of the loop. The magnetic anisotropy directly showed up in the angular dependence of the magnetizing energy, $W_{\text{mag}}(\phi)$, with the difference between the maximum and minimum values being the anisotropy energy. The polar diagrams for 75 ML Fe and 100 ML Fe$_{32}$Co$_{68}$ in Fig. 3 clearly show the superposition of a fourfold contribution expected from the cubic anisotropy and a uniaxial component with the easy axis always along [110]. From a numerical fit with the corresponding expression

$$W_{\text{mag}}(\phi) = -\frac{1}{4}K_1^\text{eff} \sin^2(2\phi) + K_4^\text{eff} \sin^2 \phi + \text{const},$$

shown as a (red/dark gray) solid line in Fig. 3(b), the effective anisotropy constants of the fourfold ($K_1^\text{eff}$) and the uniaxial ($K_4^\text{eff}$) terms are obtained. $\phi$ denotes the angle between the magnetization and the [110] direction. The fourfold anisotropy constant, $K_1^\text{eff}$, has the opposite sign for Fe$_{32}$Co$_{68}$ compared to

![Image](image_url)
Fe ($K_{1}^{\text{eff}} > 0$), which corresponds to a rotation of the easy axes by 45°. Furthermore, it has been shown previously\(^1\) that $K_{1}^{\text{eff}}$ scales linearly with the inverse film thickness and changes sign at a thickness of 6 ML, where the anisotropy is purely uniaxial and the hard axis loop along [110] is linear.

Once the symmetry of the anisotropies is known the respective anisotropy constants $K_{1}^{\text{eff}}$ and $K_{2}^{\text{eff}}$ can be determined more conveniently by fitting the hard-axis magnetization loop with an expression of the inverted curve, $H(m)$,

$$H(m) = 2K_{1}^{\text{eff}}(2m^3 - m)/M_S + 2K_{2}^{\text{eff}} m/M_S,$$

as described in detail in Ref.\(^1\). Here, $m = M/M_S$ denotes the magnetization component along the axis of the applied field normalized to the saturation magnetization, $M_S$. The results in Fig. 3 clearly prove that a substantial uniaxial anisotropy, which dominates at thicknesses below 20 ML, is still present in thicker films.

**IV. LATTICE STRAIN AND MAGNETOElastic COUPLING**

Both Fe and Fe\(_{32}\)Co\(_{68}\) grow epitaxially on GaAs(001) with a stable bcc lattice in the range of pseudomorphic growth, which extends to at least 100 ML, as verified by RHEED and x-ray diffraction data. The epitaxial misfit is compressive: $-1.4\%$ for Fe/GaAs and $-0.67\%$ for Fe\(_{32}\)Co\(_{68}\).\(^1\)\(^2\) For a cubic lattice the magnetoelastic energy density is given by\(^1\)\(^5\)

$$f_{\text{ME}}(\varepsilon, \alpha) = B_1(\alpha_1^2\varepsilon_1 + \alpha_2^2\varepsilon_2 + \alpha_3^2\varepsilon_3) + B_2(\alpha_1\alpha_2\varepsilon_5 + \alpha_2\alpha_3\varepsilon_6 + \cdots),$$

where $\alpha_i$ are the direction cosines of the magnetization relative to the cubic axes, $\varepsilon_j$ are the strain components, and $B_{1,2}$ are the magnetoelastic coupling coefficients in first order in strain. The dots symbolize that higher order terms may contribute.\(^1\)\(^6\) The magnetization of the present films always

\begin{align*}
\text{FIG. 2.} & \quad \text{(Color online) Normalized in-plane magnetization loops at } T = 300 \text{ K measured by longitudinal MOKE for a 14 ML epitaxial Fe(001) film on GaAs(001) along the [110] and [1\bar{1}0] axes (inset shows the [110] loop with enlarged field scale). The (red/dark gray) hatched area represents the magnetizing energy } W_{\text{mag}} \text{ along [110]. For the [110] orientation the anhysteretic } M(H) \text{ loop is obtained by averaging between the right and the left branch of the loop leading to } W_{\text{mag}} \approx 0. \\
\end{align*}

\begin{align*}
\text{FIG. 3.} & \quad \text{(Color online) Polar diagrams of the magnetizing energy, } W_{\text{mag}}(\phi), \text{ at } 295 \text{ K for 75 ML Fe (a) and 100 ML Fe}_{32}\text{Co}_{68} \text{ (b) on GaAs(001) in the (001) plane. The constant in Eq. (1) is set to } \frac{1}{2} K_{1}^{\text{eff}} \text{ for Fe and to zero for Fe}_{32}\text{Co}_{68} \text{ in order to always keep } W_{\text{mag}} > 0. \text{ The (red/dark gray) solid line in Fig. 3(b) represents a numerical fit assuming a superposition of a uniaxial and a fourfold in-plane magnetic anisotropy. The fourfold easy axes are along [110] for the Fe film and along [110] for Fe}_{32}\text{Co}_{68}, \text{ respectively; the uniaxial easy axis is along [110] in both cases.} \\
\end{align*}

\begin{align*}
\text{lies in the film plane, i.e., } \alpha_3 = 0, \text{ which results in} \\
\end{align*}

$$f_{\text{ME}} = B_1(\varepsilon_1 \cos^2 \phi + \varepsilon_2 \sin^2 \phi) + \frac{1}{2} B_2 \varepsilon_6 \sin(2\phi),$$

where $\phi$ denotes the angle between the cubic [100] axis and the magnetization.

From detailed RHEED investigations for Fe films up to 100 ML thickness the in-plane strain was found to be isotropic within experimental error of $<1\%$, which means that

$$\varepsilon_1 = \varepsilon_2$$

and the azimuthal dependence of the magnetoelastic energy density becomes

$$f_{\text{ME}} = C + c B_2 \varepsilon_6 \sin^2 \phi,$$

where $\varepsilon_6$ denotes a shear strain component.\(^1\)\(^5\) $C$ and $c$ are numerical factors resulting from the conversion of trigonometric functions.

The comparison of this expression with the energy density of a UMA

$$f_U = K_U \sin^2 \phi$$
reveals that the magnetoelastic energy is equivalent to a UMA with the anisotropy constant
\[ K_U \propto B_2 \varepsilon_6. \]

In view of the simple epitaxial relation with an isotropic in-plane strain, at first sight it is not obvious that a shear strain \( \varepsilon_6 \) needs to be expected. However, given the atomic and electronic structure at the GaAs surface, which deviates sharply from that of a simple cubic metal, shear strains near the interface cannot be ruled out \textit{a priori}, and they should have the same sign for both materials and would contribute equally to \( K_U^{\text{eff}} \) via Eq. (6).

The same sign of the strain components \( \varepsilon_6 \) is expected for Fe and Fe\(_{32}\)Co\(_{68}\), as the lattice misfit is of the same sign for both materials. Thus, we may suggest that the sign of the UMA depends on the sign of \( B_2 \), provided that the magnetoelastic coupling is the dominant anisotropy contribution.

Figure 4 shows the result of the magnetoelastic stress measurements for (a) 100 ML Fe and (b) Fe\(_{32}\)Co\(_{68}\). Here the magnetization was sequentially switched in-plane from along the length to along the width of the stripe, as indicated by the sketch. The data reflect the change of curvature upon the magnetization reorientation. Obviously, the change in curvature has the opposite sign for the two materials. This indicates an opposite sign of the magnetoelastic coupling constant \( B_2 \) for Fe(001) and Fe\(_{32}\)Co\(_{68}\)(001) for this film thickness. The values of \( B_2 \) for films of 30 ML and 100 ML are listed in Table I. Within the experimental uncertainty the values are the same for both thicknesses. The results for Fe deviate slightly from the bulk value (\( B_2 = +7.83 \text{ MJ/m}^2 \)), we are not aware of any available bulk data for FeCo).

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|}
\hline
\textbf{t} material & \textbf{Fe} & \textbf{Fe\(_{32}\)Co\(_{68}\)} \\
\hline
100 ML & +10.6 ± 1.3 & -28.4 ± 2.8 \\
30 ML & +8.4 ± 1.4 & -23.2 ± 3.7 \\
\hline
\end{tabular}
\caption{Magnetoelastic coupling constant \( B_2 \) [MJ/m\(^2\)] for Fe(001) and Fe\(_{32}\)Co\(_{68}\)(001) films on GaAs(001) with two thickness values, t.}
\end{table}

The important point, however, is the fact that both Fe(001) and Fe\(_{32}\)Co\(_{68}\)(001) films show a UMA with the same sign of \( K_U^{\text{eff}} \) for the entire thickness range, i.e., the easy axis of the UMA is always along [110]. This is in direct contradiction to the expectation of a mainly magnetoelastic origin of the UMA based on the \( B_2 \) data listed in Table I. We conclude that the magnetoelastic interaction may contribute to the effective UMA, but it cannot be the dominant contribution.

V. MAGNETIC ANISOTROPY: RESULTS AND DISCUSSION

The UMA constant, \( K_U^{\text{eff}} \), extracted from room temperature measurements, is plotted as a function of the inverse film thickness for a series of Fe(001) and Fe\(_{32}\)Co\(_{68}\)(001) films in Fig. 5. The data follow straight lines through the origin for both materials. The observed proportionality between the anisotropy constant and the inverse thickness indicates that the UMA is strictly a surface or interface effect according to
\[ K_U^{\text{eff}} = \frac{K_S}{t}, \]
with \( K_S \) being the interface anisotropy constant. \( K_U^{\text{eff}} \) vanishes for infinitely thick films as expected for a cubic material. Deviations from the 1/t scaling for films of 6 ML and thinner are due to reduced Curie temperature and the gradual decrease of the interface anisotropy constant \( K_S \) below 6 ML.\(^{6,19} \)

A seeming volume UMA reported recently\(^{20} \) is presumably related to structural defects which show up in increased coercivities along the easy and hard axes (\( H_C = 80–90 \) Oe in Ref. 20 as compared to \( H_C < 5 \) Oe in Fig. 2).

The important point, however, is the fact that both Fe(001) and Fe\(_{32}\)Co\(_{68}\)(001) films show a UMA with the same sign of \( K_U^{\text{eff}} \) for the entire thickness range, i.e., the easy axis of the UMA is always along [110]. This is in direct contradiction to the expectation of a mainly magnetoelastic origin of the UMA based on the \( B_2 \) data listed in Table I. We conclude that the magnetoelastic interaction may contribute to the effective UMA, but it cannot be the dominant contribution.
In order to support the leading contribution of the magnetoelastic interaction to the UMA it was argued in Ref. 12 that the presence of shear strain in ultrathin Fe films on GaAs had indeed been experimentally verified by Gordon and Crozier\textsuperscript{19} who have determined the shear strain by extended x-ray absorption fine structure (EXAFS) via an in-plane anisotropy of the nearest neighbor distance in 2 ML and 5 ML Fe on GaAs. While such a shear strain was clearly observed for a 2 ML film, it was below the detection limit in the 5 ML film. This result implies that the resulting magnetoelastic contribution to the magnetic anisotropy should be much stronger in a 2 ML film as compared with a 5 ML film, as it is expected to scale with lattice strain, as exemplified in Eq. (8). This prediction can be experimentally checked by a comparison with earlier results reported in Ref. 21. There it has been shown that the interface anisotropy energy constant, $K_s$, defined as the uniaxial anisotropy energy per unit area, decreases with decreasing Fe thickness below 8 ML and vanishes around 2.5 ML, i.e., at the same thickness where the ferromagnetic long-range order vanishes and the Curie temperature $T_C$ becomes zero. (It should be noted that the uniaxial anisotropy persists in the paramagnetic state far above $T_C$, as shown in Fig. 3 in Ref. 21.) The fact that the observed shear strain at 2 ML is accompanied by a vanishing anisotropy, whereas at 5 ML a nondetectable shear coincides with a strong UMA, clearly contradicts the assumption that the UMA originates from an in-plane lattice shear and is in contrast to a dominant magnetoelastic contribution to the UMA.

Therefore, the most likely origin of the observed anisotropy is the presence of oriented bonds at the interface between GaAs and the film material. This mechanism was shown in calculations to be responsible for the in-plane UMA in the similar system Fe/ZnSe.\textsuperscript{14} Recently, it was experimentally demonstrated that already 1–2 ML of MgO epitaxially grown between the GaAs substrate and the Fe film are sufficient to essentially quench the UMA of the Fe layer.\textsuperscript{23} This is indeed to be expected if oriented Fe–As bonds are the main symmetry breaking mechanism being the source of the UMA. A different effect of an MgO interlayer deposited between GaAs and Fe on the UMA has been reported recently;\textsuperscript{25} however, this study lacks a structural characterization, which substantially limits its relevance. Finally, the minor role of epitaxial strain for the amount of UMA has also been evidenced by studying the effect of postgrowth annealing on the UMA in Fe/GaAs:\textsuperscript{24} while a 1 h anneal at 200 °C resulted in a drastic strain relaxation the UMA remained practically unchanged.\textsuperscript{21}

VI. CONCLUSION

The experiments reported here demonstrate that although magnetoelastic effects may possibly contribute to the in-plane UMA in Fe/GaAs(001), the magnetoelastic interaction is not the dominating mechanism to the UMA in ultrathin Fe(001) films on GaAs(001). Rather, the UMA is mainly attributed to oriented covalent bonds at the metal-GaAs interface. This view, which is also held in a recent review article on ferromagnetic metal–compound semiconductor hybrid structures,\textsuperscript{25} has now been firmly substantiated by the direct measurement of magnetoelastic coupling constants reported here. Understanding the underlying mechanisms allows us to control magnetic anisotropies in ferromagnet-semiconductor hybrid systems via film thickness and composition, which in turn has proved to be extremely useful in spin injection experiments.\textsuperscript{26}

To quantitatively evaluate the magnitude of the magnetoelastic contribution to the UMA in Fe/GaAs, \textit{in situ} measurements of strain, magnetoelastic coupling, and magnetic anisotropy are called for. Also, an extension of the present work to thicker Fe films could elucidate the origin of the observed deviation of the magnetoelastic coupling constant $B_2$ from its bulk value. \textit{Ab initio} calculations of the electronic structure at Fe/GaAs and FeCo/GaAs interfaces for different Fe-Co compositions would be highly desirable to further advance the understanding of the origin and strength of the UMA on the electronic level.

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