Uniaxial magnetic anisotropy in Fe/GaAs(001): Role of magnetoelastic interactions

Günther Bayreuther,[1,2,*] Jörg Premper,[1] Matthias Sperl,[2] and Dirk Sander[1]
[1]*Max-Planck-Institut für Mikrostrukturphysik, 06120 Halle, Germany*
[2]*Institut für Experimentelle und Angewandte Physik, Universität Regensburg 93040 Regensburg, Germany*
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The origin of the in-plane uniaxial magnetic anisotropy of epitaxial Fe(001) films on GaAs(001) has been controversially attributed either to a magnetoelastic interaction or to oriented interface bonds. In order to check the relevance of magnetoelastic contributions we performed magneto-optic Kerr effect (MOKE) and magnetoelastic stress measurements on Fe(001) and $Fe_{32}Co_{68}$(001) *bcc* epitaxial films on GaAs(001) in a thickness range from several monolayers (ML) up to 100 ML. The magnetoelastic coupling coefficient B_2 of 30 ML films is $+(8.4 \pm 1.4)\,MJ/m^3$ for Fe(001) and $-(23.2 \pm 3.7)\,MJ/m^3$ for $Fe_{32}Co_{68}$(001), respectively. In spite of the opposite sign of B_2 for Fe and $Fe_{32}Co_{68}$, the same sign of the uniaxial anisotropy constant $K_U^{\rm eff}$ is experimentally observed. It is concluded that the magnetoelastic interaction is not the dominating contribution to the uniaxial magnetic anisotropy in these systems.

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I. INTRODUCTION

Epitaxial growth of Fe on GaAs(001) was demonstrated long ago.[1,2] A renewed wide interest in this material arose since it has been established as a prototype system for successful spin injection from a ferromagnet into a semiconductor,[3] 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,[2] which dominates in ultrathin films of 20 monolayers (ML) (atomic ML) and less. It was subsequently confirmed in numerous studies.[4–7] After early confusion about the crystalline axes, a general agreement emerged that the UMA easy axis is always oriented along the [110] axis of the GaAs substrate, which is parallel to the Fe [110] axis with [1$\bar{1}$0] being the uniaxial hard axis. Furthermore, the role of different surface reconstructions of the GaAs substrate was investigated. It was concluded that the surface reconstruction of GaAs does not affect the orientation of the magnetic easy axis.[8,9] It could even be shown that different surface reconstructions (e.g., 4×2 and 6×4) on a Ga-rich GaAs(001) surface produce nearly identical values of the uniaxial anisotropy constant $K_U^{\rm eff}$.[9] It was also realized that the anisotropy constant $K_U^{\rm eff}$, defined as the anisotropy energy per volume, is proportional to the inverse Fe thickness $t_{\rm Fe}$ (Refs. 4,6,7,10–12) in a wide range, indicating that the UMA originates from the Fe/GaAs interface.

We note that the sheer existence of a uniaxial magnetic interface anisotropy is not surprising from symmetry arguments: although Fe and GaAs both have a cubic lattice the GaAs zincblende lattice lacks the fourfold rotational symmetry present in the Fe bcc lattice. Consequently, it is intuitively clear that the specific orientations of As and Ga dangling bonds at an ideal GaAs(001) surface define preferred axes for the electronic orbitals at the interface that locally break the fourfold symmetry of the natural Fe bulk lattice. The remaining twofold symmetry can be expected to give rise to a magnetic anisotropy with the same symmetry, which corresponds to the observed UMA.

Despite the general agreement on the phenomenology, the microscopic origin of the UMA remains a controversial topic. 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,[12,13] or (ii) electronic hybridization between Fe and GaAs.[2,7,14]

This article experimentally assesses the contribution of the magnetoelastic interaction to the uniaxial anisotropy. To this end we measure the magnetoelastic coupling of two different film systems, Fe/GaAs(001) and $Fe_{32}Co_{68}$/GaAs(001). Both grow epitaxially on GaAs(001) with a compressive lattice strain of -1.4% and -0.67%, respectively, in the case of pseudomorphic growth. They will be shown to have the opposite sign of the magnetoelastic coupling constant B_2 but the same sign of the UMA constant $K_U^{\rm eff}$. The conclusion is that the magnetoelastic interaction—while possibly present—is not the dominating contribution to the UMA. Rather, the electronic hybridization between the ferromagnetic films and the GaAs substrate appears to be decisive.

II. EXPERIMENT

Fe(001) and $Fe_{32}Co_{68}$(001) films were epitaxially grown on GaAs(001) substrates by molecular beam epitaxy (MBE) using previously developed procedures.[5,6,9] To facilitate the measurements of magnetoelastic stress from the substrate curvature,[15,16] a commercial GaAs wafer was thinned to $t_s = 180\ \mu$m and cleaved into stripes of 2 mm × 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 (4×6) surface reconstruction. The presence of atomically flat terraces several 100 nm wide separated by ML steps was verified by *in situ* STM,[6,9] as shown in Fig. 1(b). After the growth of Fe and $Fe_{32}Co_{68}$ 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 $Fe_{32}Co_{68}$ lattice were parallel to the respective GaAs axes. Magnetization loops, $M(H)$,

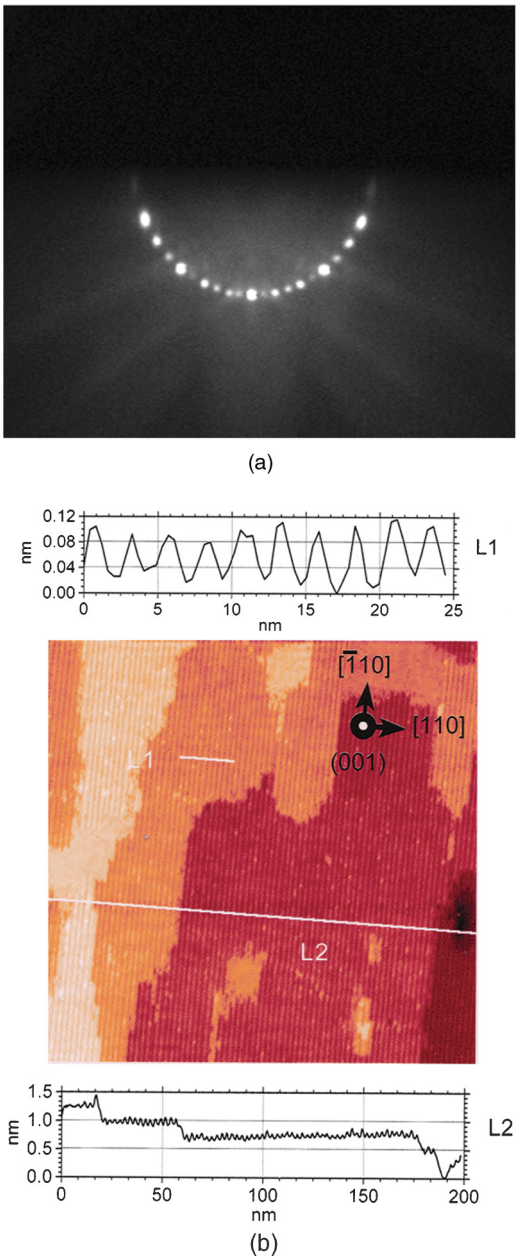

FIG. 1. (Color online) GaAs(001) surface after Ar ion sputtering ($E = 1$ keV, $j = 25\ \mu$A/cm^2, $t = 1$ h) at 550 °C under sample rotation and subsequent annealing in UHV at 550 °C for 15 min. (a) RHEED diffraction pattern along the [110] azimuth (first Laue circle) indicating a very flat surface; (b) 200 nm × 200 nm STM image and line scans showing atomically flat terraces separated by monoatomic steps. The stripe pattern is due to As dimer rows. The line scans identify the increased periodicity of the (4 × 6) surface reconstruction along [110].

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.[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., $[1\bar{1}0]$) 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 $1/5000\ \mathrm{m}^{-1}$. From the change in curvature measured by the deflection of two laser beams the magnetoelastic coupling coefficient, B_2 was determined[15,16] according to $B_2 = \frac{Yt_s^2}{6(1+\nu)t}\Delta\frac{1}{R}$, where the Young modulus of the GaAs substrate along [110], Y, and its Poisson ratio along $[1\bar{1}0]$, ν, were given by $Y = 121.3$ GPa and $\nu = 0.021$;[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 2×10^{-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 ϕ between the measurement and field axis and the [110] axis. The magnetizing energy was determined by integrating the anhysteretic $M(H)$ loops according to $W_{\mathrm{mag}} = \int_0^{M_S} H \cdot dM$, as demonstrated in Fig. 2 for a 14 ML Fe film on GaAs(001). The (red/dark gray) hatched area represents W_{mag} along $[1\bar{1}0]$, 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, $W_{\mathrm{mag}}(\phi)$, 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

$$W_{\mathrm{mag}}(\phi) = -\tfrac{1}{4}K_1^{\mathrm{eff}}\sin^2(2\phi) + K_U^{\mathrm{eff}}\sin^2\phi + \mathrm{const}, \quad (1)$$

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

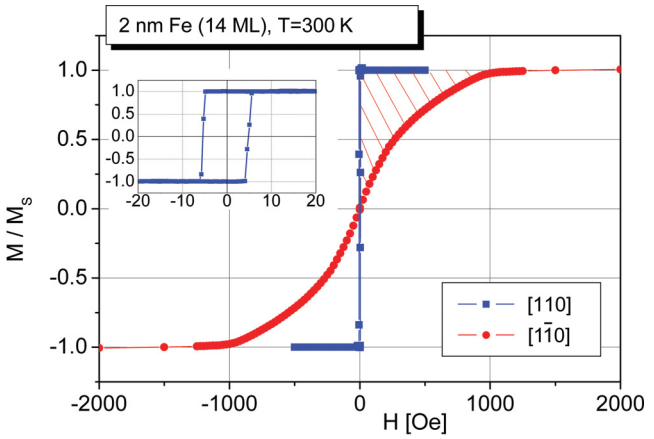

FIG. 2. (Color online) Normalized in-plane magnetization loops at $T = 300$ K measured by longitudinal MOKE for a 14 ML epitaxial Fe(001) film on GaAs(001) along the [110] and [1$\bar{1}$0] axes (inset shows the [110] loop with enlarged field scale). The (red/dark gray) hatched area represents the magnetizing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

Fe ($K_1^{\mathrm{eff}} > 0$), which corresponds to a rotation of the easy axes by 45°. Furthermore, it has been shown previously[10] that K_1^{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 [1$\bar{1}$0] is linear.

Once the symmetry of the anisotropies is known the respective anisotropy constants K_1^{eff} and K_U^{eff} can be determined more conveniently by fitting the hard-axis magnetization loop with an expression of the inverted curve, $H(m)$,

$$H(m) = 2K_1^{\mathrm{eff}}(2m^3 - m)/M_S + 2K_U^{\mathrm{eff}} m/M_S, \qquad (2)$$

as described in detail in Ref. 10. 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 $Fe_{32}Co_{68}$.[18] For a cubic lattice the magnetoelastic energy density is given by[15]

$$f_{\mathrm{ME}}(\varepsilon,\alpha) = B_1\left(\alpha_1^2\varepsilon_1 + \alpha_2^2\varepsilon_2 + \alpha_3^2\varepsilon_3\right) + B_2(\alpha_1\alpha_2\varepsilon_6 + \alpha_1\alpha_3\varepsilon_5 + \alpha_2\alpha_3\varepsilon_4) + \cdots, \qquad (3)$$

where α_i are the direction cosines of the magnetization relative to the cubic axes, ε_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.[16] The magnetization of the present films always

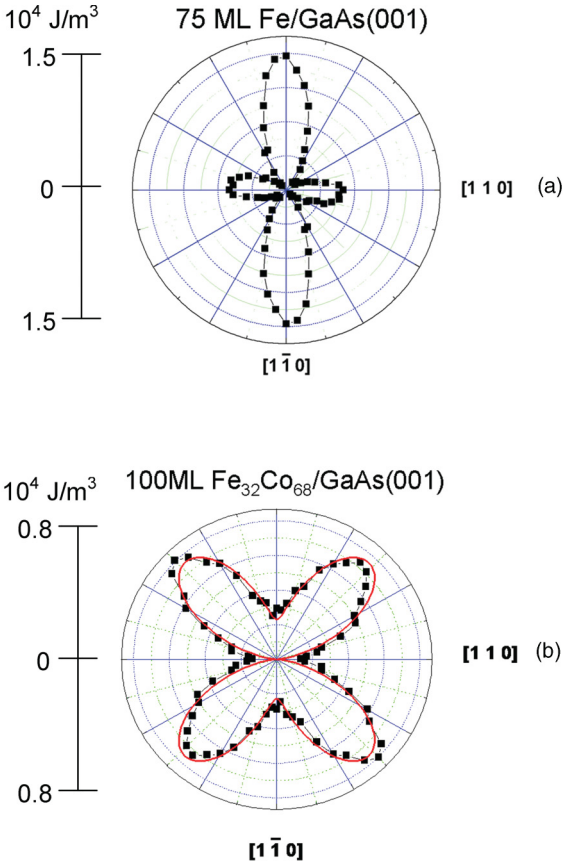

FIG. 3. (Color online) Polar diagrams of the magnetizing energy, $W_{\mathrm{mag}}(\phi)$, at 295 K for 75 ML Fe (a) and 100 ML $Fe_{32}Co_{68}$ (b) on GaAs(001) in the (001) plane. The constant in Eq. (1) is set to $\frac{1}{4}K_1^{\mathrm{eff}}$ for Fe and to zero for $Fe_{32}Co_{68}$ in order to always keep $W_{\mathrm{mag}} > 0$. 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 ⟨100⟩ for the Fe film and along ⟨110⟩ for $Fe_{32}Co_{68}$, respectively; the uniaxial easy axis is along [110] in both cases.

lies in the film plane, i.e., $\alpha_3 = 0$, which results in

$$f_{\mathrm{ME}} = B_1(\varepsilon_1 \cos^2\varphi + \varepsilon_2 \sin^2\varphi) + \tfrac{1}{2}B_2\varepsilon_6 \sin(2\varphi), \qquad (4)$$

where φ 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

$$\varepsilon_1 = \varepsilon_2 \qquad (5)$$

and the azimuthal dependence of the magnetoelastic energy density becomes

$$f_{\mathrm{ME}} = C + cB_2\varepsilon_6 \sin^2\varphi, \qquad (6)$$

where ε_6 denotes a shear strain component;[15] 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

$$f_U = K_U \sin^2\varphi \qquad (7)$$

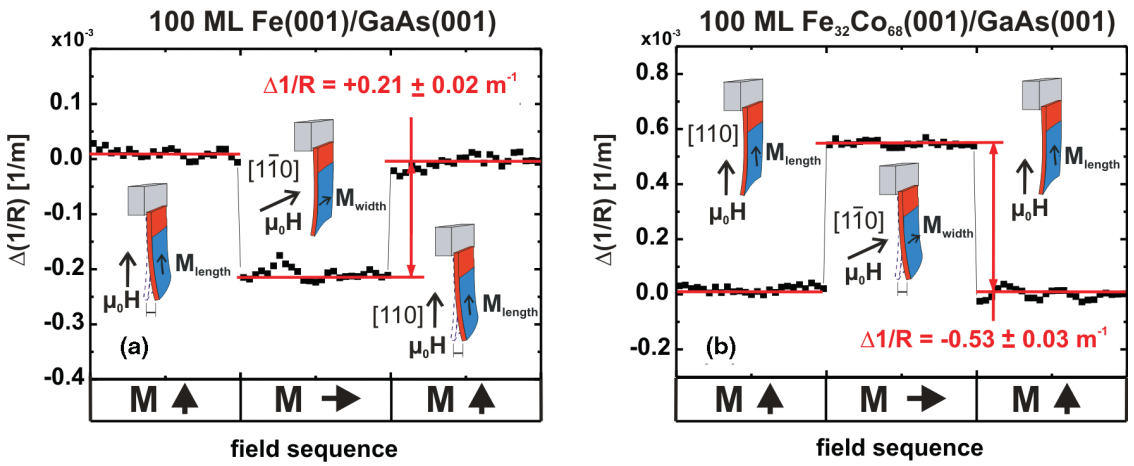

FIG. 4. (Color online) Change in curvature of a 12 mm × 2 mm GaAs stripe of 180 μm thickness with 100 ML Fe(001) (left) or $Fe_{32}Co_{68}$(001) (right) epitaxially grown on top when the magnetization is switched from [110] to [1$\bar{1}$0] and back.

reveals that the magnetoelastic energy is equivalent to a UMA with the anisotropy constant

$$K_U \propto B_2 \varepsilon_6. \tag{8}$$

In view of the simple epitaxial relation with an isotropic in-plane strain, at first sight it is not obvious that a shear strain ε_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 *a priori*, and they should have the same sign for both materials and would contribute equally to K_U^{eff} via Eq. (6).

The same sign of the strain components ε_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 ($B_2 = +7.83$ MJ/m^3;[15] we are not aware of any available bulk data for FeCo). We

TABLE I. Magnetoelastic coupling constant B_2 [MJ/m^3] for Fe(001) and $Fe_{32}Co_{68}$(001) films on GaAs(001) with two thickness values, t.

t material	Fe	$Fe_{32}Co_{68}$
100 ML	$+10.6 \pm 1.3$	-28.4 ± 2.8
30 ML	$+8.4 \pm 1.4$	-23.2 ± 3.7

may tentatively ascribe this deviation to the lattice strain of the films, which is roughly constant in this thickness range.[16] This assessment should be checked by future comparative measurements on thick Fe films with and without strain relief under thermal treatment. We conclude that if a magnetoelastic origin of the UMA is assumed, then these results suggest that its contribution to K_U^{eff} should be of the opposite sign for both materials. However, as will be shown below, the same sign and a comparable magnitude are found for the UMA, disqualifying magnetoelastic coupling as the main source of the UMA.

V. MAGNETIC ANISOTROPY: RESULTS AND DISCUSSION

The UMA constant, K_U^{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

$$K_U^{\text{eff}} = \frac{K_S}{t}, \tag{9}$$

with K_S being the interface anisotropy constant. K_U^{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^{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.

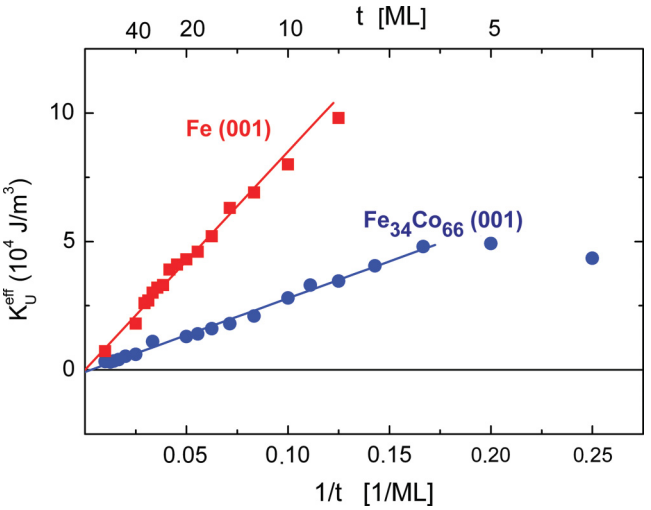

FIG. 5. (Color online) Uniaxial anisotropy constant, K_U^{eff}, versus inverse film thickness of epitaxial Fe(001) and $Fe_{32}Co_{68}$(001) films on GaAs(001) measured at 300 K. The slope is proportional to the interface anisotropy constant, K_S. Deviations from the straight line at small film thicknesses are due to reduced Curie temperature and the gradual decrease of the interface anisotropy constant K_S below 6 ML (Refs. 6 and 19). Error bars are represented by the size of the symbols.

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[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.[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.[22] 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;[23] 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:[24] while a 1 h anneal at 200 °C resulted in a drastic strain relaxation the UMA remained practically unchanged.[24]

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,[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.[26]

To quantitatively evaluate the magnitude of the magnetoelastic contribution to the UMA in Fe/GaAs, *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. *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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*Guenther.Bayreuther@physik.uni-regensburg.de

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