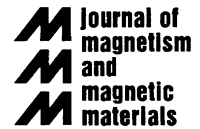




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# Perpendicular magnetization in Fe/Ni bilayers on GaAs(001)

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## Abstract

Following the concept of spin-injection into a semiconductor-based device, a ferromagnetic element (like a GMR multilayer structure) can be used as a spin filter. A high spin-polarization of the electrons can be realized by the preparation of a monocrystalline multilayer structure consisting of ultrathin films of a high magnetic polarization. In the case of ultrathin films, the manipulation of the easy-axis of magnetization is possible, by changing the anisotropy terms contributing to the effective anisotropy of the structure. We report on the structural and magnetic properties of Ni/Fe and Fe/Ni bilayers epitaxially grown on GaAs(001). By a proper choice of Fe and Ni sequences (Fe/Ni/GaAs) and their thickness (up to 3 ML of Fe on the top of Ni), the rotation of magnetization from the in-plane to the out-of-plane direction was achieved. © 2001 Elsevier Science B.V. All rights reserved.

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*Keywords:* Ferromagnetic thin films; Semiconducting substrates; Anisotropy; Spin-polarized transport

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## 1. Introduction

Spin electronic devices operate by coding magnetic information on the itinerant electrons that are used to transfer spin information from one part of the device to write to another part where it is read. These write and read operations are performed by small ferromagnetic elements whose relative magnetic orientation can be varied by application of an external magnetic field. By

changing the magnetization of the two ferromagnetic elements from parallel to antiparallel, the current flow undergoes a bipolar modulation [1]. A ferromagnetic element on a nonmagnetic substrate can be used as a spin filter. The concept of spin-injection into a semiconductor-based structure can lead to the idea of a spin-polarized field-effect transistor or any other circuits integrating ferromagnetic thin film elements into semiconductors.

The operation of spin electronic devices is critically dependent on the efficient spin transfer through the interface between the ferromagnetic and nonmagnetic elements. Thus, to achieve spin-polarized electron transport across the interface, a high magnetic polarization, i.e. ferromagnetically

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ordered moments even in the first atomic layer of ferromagnetic film on semiconducting substrate is required.

Only a few studies have been carried out on spin-dependent electron transport through ferromagnet/semiconductor thin film junctions. This is due to expected difficulties in growing metals on nonconducting substrates. A lot of work is reported concerning Fe growth on GaAs [2–5]. However, as Fe forms a rectifying contact on GaAs, the Schottky barrier of about 0.8 V prevents efficient current injection from the Fe ferromagnetic film into the semiconducting GaAs substrate. Recent studies provide evidence that a spin-dependent current is transmitted over the Schottky barrier in ferromagnet/semiconductor direct interfaces based on, e.g. NiFe/GaAs [6,7] and through magnetic semiconductors used as spin aligning materials [8]. In particular, for 3 nm Au/5 nm Ni<sub>80</sub>Fe<sub>20</sub>/GaAs(110), a significant transport current was detected, whose magnitude depends on the relative orientation of the spin polarization and the magnetization vector [6].

Another practical requirement for spin electronic devices is to have perpendicularly magnetized ferromagnetic films, at least in view of realizing high-density information storage media. Also, in the case of electrical spin injection, their polarization can be proven by helicity of the light emitted out of the plane of a semiconductor. Injection of the spins oriented out-of-plane improves the detection efficiency of spin-polarization and can be easily realized with an out-of-plane easy-axis of the ferromagnetic “spin aligner”. In the case of an in-plane easy-axis, a high external magnetic field is required for achieving perpendicular saturation. The changes in the signal for opposite photon helicity could indicate that the transport between the ferromagnetic layer and the semiconducting substrate depends on the magnetization direction. Thus, the preparation of ferromagnetic layers on semiconducting substrates, with no magnetically dead layers and with the easy-axis of magnetization perpendicular to the film plane is of great practical importance.

The competition between different anisotropy energies determines the orientation of the easy-axis

of magnetization. In ultrathin films, manipulation of the easy-axis is possible by changing the anisotropy constants. This can be done by the proper choice of the film thickness and by altering the surface anisotropy with a capping layer. Perpendicular magnetization of monolayer films can be expected if the spin-orbit-induced anisotropy at the surface could overcome the magnetic dipole interaction, which tends to favor in-plane magnetization. This phenomenon has been observed for a number of ultrathin magnetic films of typically 2–5 monolayers (ML) thickness, in particular for Fe on Ag(001) [9], Cu(001) [10] and Pd(001) [11]. For thicker films, the easy-axis lies in the film plane due to the volume effect of the magnetostatic energy (shape anisotropy arising from the dipolar fields, which, in the case of homogeneously magnetized films, forces the magnetization to lie in the plane). The changes in anisotropy caused by capping are due to the surface anisotropy, which is determined by the capping layer and the demagnetizing energy [12]. The demagnetizing energy due to the capping layer, per unit volume of the film, depends on its magnetization and varies with thickness. The interface anisotropy of the film/capping-layer can also contribute to the total anisotropy energy. Perpendicular magnetization arising from interface anisotropy (which is very close to the surface anisotropy) is found in magnetic multilayers like, e.g. Co/Pd [13].

The aim of this work was to orient the magnetization of ferromagnetic Fe-based films grown on GaAs(001) out-of-plane, by changing the magnitude of one of the anisotropy energy terms contributing to the effective anisotropy of the system. To achieve this, we have used different sequences of ultrathin Ni and Fe films to make the resulting anisotropy energy favorable for out-of-plane magnetization.

## 2. Experimental details

An atomically clean and flat GaAs(001) surface cannot be achieved by solely annealing as assessed in the case of some metal substrates and silicon. The removal of carbon requires sputtering, low

energy ions being recommended to minimize the damage. Due to its zinc blend crystallographic structure with tetrahedral coordination in the bulk, the polar GaAs(001) surface could be terminated with either As or Ga atoms. Depending on the experimental conditions, the surface exhibits a number of reconstructions, starting with the most As-rich phase which has a  $c(4 \times 4)$  symmetry, through the  $2 \times 4$ ,  $1 \times 6$ ,  $4 \times 6$ , and the  $4 \times 2$  Ga-stabilized phase, which can influence the growth of the material deposited on in the early stages [14].

All of our experiments, including the GaAs(001) substrate preparation, were carried out in an ultrahigh vacuum (UHV) multichamber system equipped with molecular beam epitaxy (MBE) for sample preparation, Auger electron spectroscopy (AES), low energy electron diffraction (LEED), scanning tunneling microscopy (STM) for sample characterization and in situ polar magneto-optical Kerr effect (MOKE) for the magnetic analysis. The commercially available GaAs(001) wafers<sup>1</sup> were introduced into UHV without any chemical treatment. The substrates were mounted on a transferable sample holder allowing its movement between different UHV chambers and sample stages. The GaAs substrates were prepared in three different ways: (a) by atomic hydrogen (hydrogen irradiation at 155 W source power, at  $p = 7 \times 10^{-7}$  mbar in the so-called two-step atomic hydrogen irradiation technique [15]), (b) by preannealing at 520°C in order to degas the sample holder and then sputtering at room temperature (RT) with low energy (850 eV) Ar ions (at  $p = 5 \times 10^{-7}$  mbar) for about 3 h and (c) by preannealing at 520°C, subsequent annealing up to approximately 590°C and sputtering at the same temperature for 45 min only, again with low energy (850 eV) Ar ions.

Epitaxial growth of Fe on GaAs(001) is possible due to the fact that the size of the BCC Fe unit cell is 2.866 Å, which is approximately half of the lattice spacing of GaAs (5.654 Å). Pseudomorphic Fe films on GaAs(001) substrates are

therefore compressed by 1.34%. Along the  $\langle 001 \rangle$  growth direction, 1 ML of BCC bulk Fe corresponds to a thickness of 1.433 Å. As reported earlier, the major problem is the Ga and As diffusion into the Fe overlayers and, finally, As segregation. This influences the growth mode of the next layers, as well as their magnetic properties. Epitaxial growth of Ni on GaAs(001) is more questionable due to its FCC structure. The lattice spacing of Ni does not fit to that of GaAs(001), neither in the  $\langle 100 \rangle$  nor in the  $\langle 110 \rangle$  direction. The only possibility is to force a pseudomorphic growth of BCC-Ni in the ultrathin film limit. Intermixing between GaAs and Ni is again possible in such a case. The epitaxial growth of Ni on GaAs, to the best of our knowledge, had not been reported previously.

Fe and Ni films were deposited on carefully prepared atomically clean GaAs(001) substrates. The atomic cleanliness of the substrates, as well as of the overlayer films was confirmed with AES after preparation of each GaAs substrate and after completion of each film growth. No carbon and oxygen (within the detection limit) were detected on the GaAs substrate after sputtering with low energy Ar ions at elevated temperature. The base pressure in the preparation chamber was  $4 \times 10^{-11}$  mbar and during deposition increased to  $9 \times 10^{-10}$  mbar. Both Fe and Ni were deposited at a rate of about 1–1.3 Å (below the thickness of 1 ML) per minute, by electron beam evaporation of a thoroughly outgassed high-purity iron and nickel rod, respectively. Fe was always grown at room temperature, whereas Ni was deposited at gradually increasing temperature from RT for the deposition of the first 3–4 monolayers, to 400 K at the end of the preparation. The film thickness was controlled using a quartz crystal monitor placed at the sample position before and after each preparation. LEED images were taken with a charge coupled device (CCD) camera. All STM images were acquired in the constant current mode at currents of 0.1–1 nA and bias voltages between –2.5 and –3.0 V. MOKE loops were collected in situ in the polar geometry using an electromagnet with a maximum field of 750 Oe, and an intensity stabilized laser diode (wavelength 670 nm). The

<sup>1</sup> Distributed in Germany by Freiberger Compound Materials GmbH.

angle of incidence was about  $20^\circ$  with respect to the surface normal.

### 3. Results

The STM images taken for GaAs(001) prepared as described above showed remarkable differences in the surface quality: for different preparation methods the atomically flat surface was never observed after RT sputtering, whereas the STM images of the GaAs(001) substrates sputtered at elevated temperature showed atomically flat terraces of hundreds of nanometers width, separated by double-layer steps (Fig. 1a). The orientation of the stripes is uniform over the whole sample. The preparation of GaAs(001) by sputtering at elevated temperature (as described above) resulted in a  $p(4 \times 6)$  surface reconstruction, clearly seen from the image via the 4-fold periodicity along the  $\langle \bar{1}10 \rangle$  direction (Fig. 1b). This characteristic  $p(4 \times 6)$  reconstruction is also seen in the LEED patterns. The LEED pattern of our cleaned surface is shown in Fig. 2a. We stress the fact that the quality of our LEED pattern equals that of MBE grown GaAs(001) [16].

#### 3.1. Fe/GaAs(001) and Ni/GaAs(001) systems

For Fe grown on GaAs(001) at RT, LEED patterns of clear 4-fold symmetry, although a little bit diffuse, were observed even for the 3 ML thick films (Fig. 2b). They became sharper for the 4–5 ML thick films (Fig. 2c) and were observed up to the thickest Fe film deposited (about 10 ML). A lot of work has been reported concerning the magnetic properties of Fe grown on GaAs(001). A “magnetically dead” layer is often reported to exist at the interface [17,18]. Only recently, it was reported that RT growth of Fe on GaAs(100) prepared by bombardment with low energy ions prevents the formation of magnetically dead layers [16,19]. Our experience is that even an  $n = 2$  ML thick Fe film grown on a carefully prepared GaAs(001) is magnetically “alive”. By polar MOKE, a clear magneto-optical response of s-shape with no hysteresis loop was observed at 150 K, which is typical for a superparamagnetic

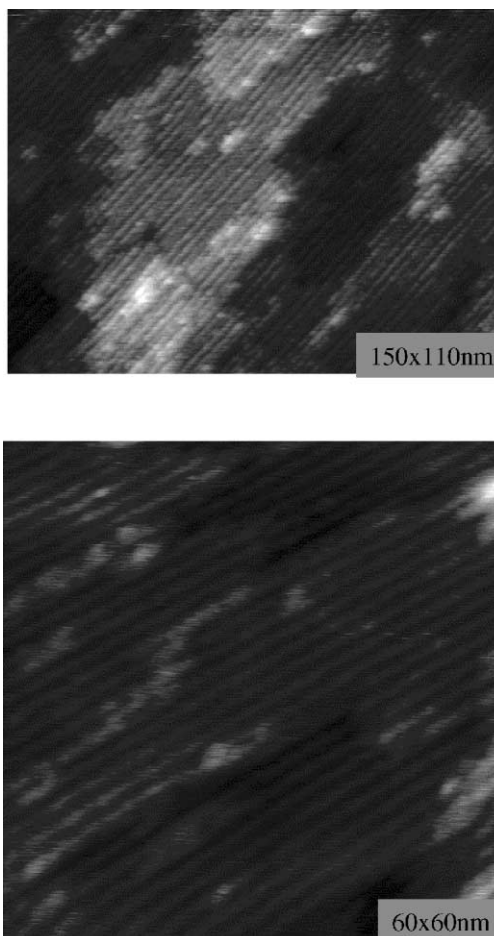


Fig. 1. STM image of GaAs(001) surface of a commercially available wafer, prepared by sputtering at  $590^\circ\text{C}$ , for 45 min, with low energy (850 eV) Ar ions.

system (Fig. 3). This is consistent with the earlier studies that have indicated the formation of small three-dimensional clusters in the initial stages of Fe growth on GaAs(001) [20]. The measurable polar magneto-optical response at room temperature was first detected for  $n = 3$  ML of Fe (3 ML Fe/GaAs(001), Fig. 4). When the thickness of Fe was increased up to about  $n = 9–10$  ML, the magneto-optical response became less pronounced and gradually reduced. Above  $n = 10$  ML, the typical hard-axis magnetization linearly proportional to the magnetic field was obtained, and the slope was found to depend on the film thickness (Fig. 4).

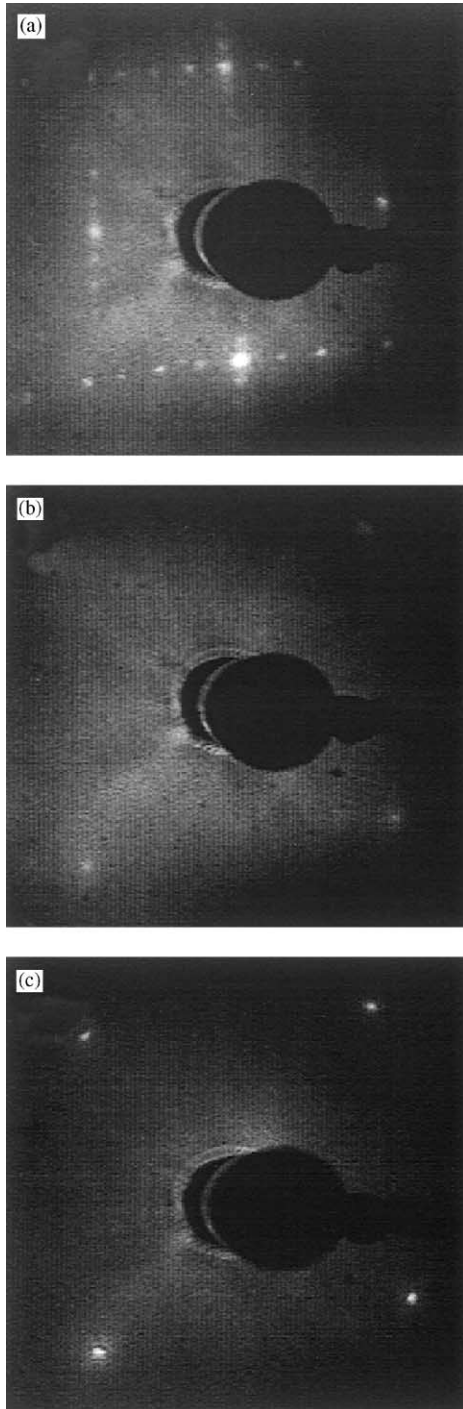


Fig. 2. (a) LEED patterns of the GaAs(001) surface shown in Fig. 1. (b, c) LEED patterns after deposition of  $n = 3$  and 5 ML of Fe on GaAs(001) at RT.

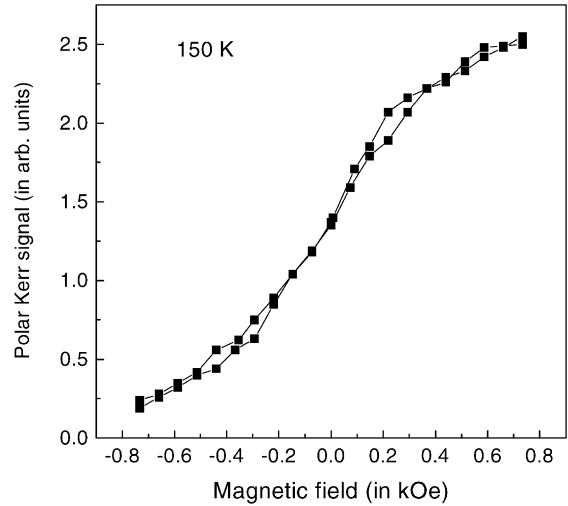


Fig. 3. Polar MOKE signal measured at 150 K for 2 ML of Fe deposited on GaAs(001) at RT.

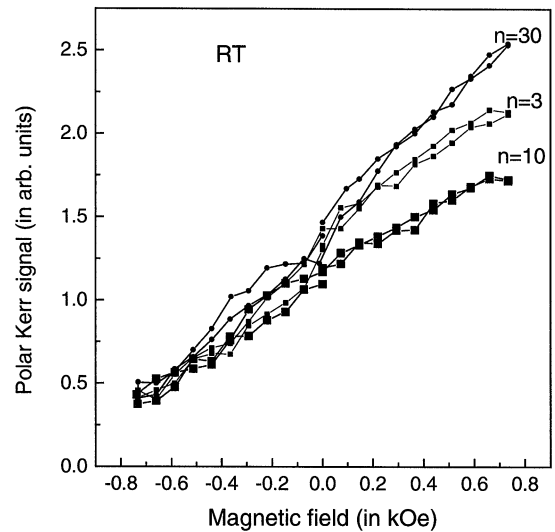


Fig. 4. Polar MOKE signal measured at RT for various thicknesses ( $n = 3, 10$  and 30) of the Fe film deposited on GaAs(001) at RT.

Then we tried epitaxy of Ni on the GaAs(001) which is also a ferromagnetic material (however, spin polarization at the Fermi level is less than that of Fe), in order to check its magnetic properties in the ultrathin film regime, i.e. to check its ability for spin-polarized injection. For Ni on GaAs(001) no

LEED spots were observed below a Ni thickness of about  $m = 8$  ML at which a diffuse pattern appeared. Up to  $m = 8$  ML of Ni, no detectable magnetic response was measured by MOKE applied in polar geometry, neither at room temperature nor at 150 K. Thus, the Ni/GaAs(001) films are suggested to be magnetically dead or to have their easy-axis in plane. This question cannot be answered with our in situ MOKE system that was set up in the polar configuration, due to the weak signal caused by the weak magneto-optic interaction of Ni.

Following the idea of anisotropy engineering, we tried to rotate the magnetization from in-plane (expected for BCC ferromagnetic films on GaAs(001) [16,19]) to out-of-plane by a capping layer of a ferromagnetic material. We deposited Ni and Fe layers of various thicknesses in a number of different sequences, on the carefully prepared atomically flat GaAs(001) surface. The idea was to play with two ferromagnetic layers but in different sequences allowing to vary the total anisotropy energy, first of all by the different surface/interface anisotropy terms introduced in these cases.

### 3.2. Ni/Fe/GaAs(001) system

In order to save the magnetically “alive” Fe layer existing at the interface which warrants the spin-polarization of the injected electrons, we tested first the structure and magnetic properties of Ni/Fe bilayers, i.e. of the  $m$ Ni/ $n$ Fe/GaAs(001) system. No LEED patterns were observed after the deposition of Ni on the top of Fe films. Only coating with maximum  $m = 3$  ML Ni of less than  $n = 3$  ML Fe resulted in a restored LEED pattern similar to that observed for  $n = 3$  ML Fe originally grown on GaAs(001). This occurs even in the case of  $n = 2$  ML of Fe, i.e., at that thickness of Fe grown on GaAs(001) for which LEED patterns were not observed (Fig. 5). Further covering with 2 ML of Fe (2 ML Fe/3 ML Ni/2 ML Fe/GaAs(001) sample) improves the quality of the LEED pattern (Fig. 5). The LEED patterns disappeared again after the deposition of a Ni film thicker than 3 ML on the top of Fe. The preparation of sequences of  $(m$ Ni/ $n$ Fe) $_x$  bilayers

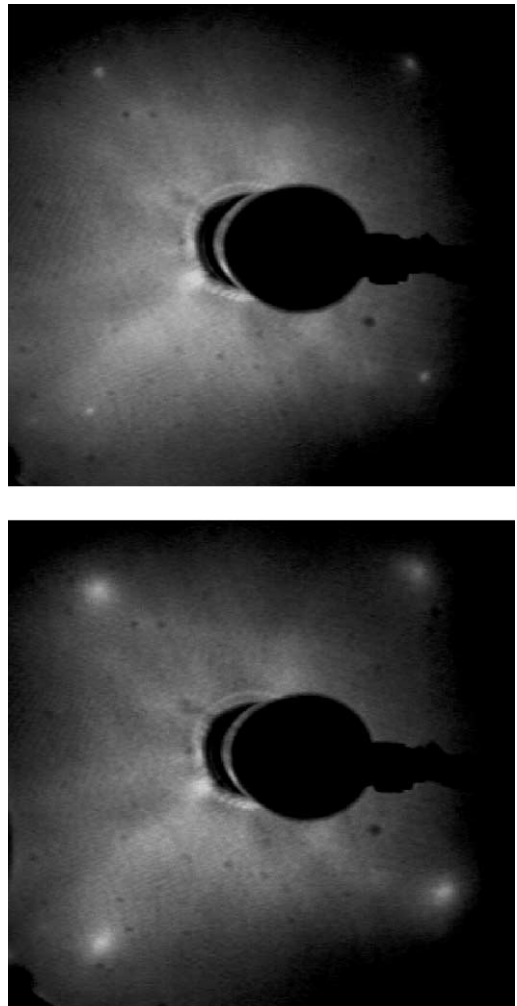


Fig. 5. LEED patterns of 3 ML Ni/2 ML Fe/GaAs(001) and 2 ML Fe/3 ML Ni/2 ML Fe/GaAs(001), both Fe and Ni prepared at RT.

on GaAs(001) always leads to in-plane orientation of the easy-axis, independent of the Fe and Ni thickness and the number ( $x$ ) of sequences.

In the case of Fe deposited on GaAs(001), a small hysteresis loop superimposed onto a clear hard-axis signal was observed occasionally by polar MOKE. This could be due to an occasionally perpendicular orientation of the spins (e.g. those close to the island edges) corresponding to the local GaAs wafer quality. They contribute to the perpendicular magnetization also after

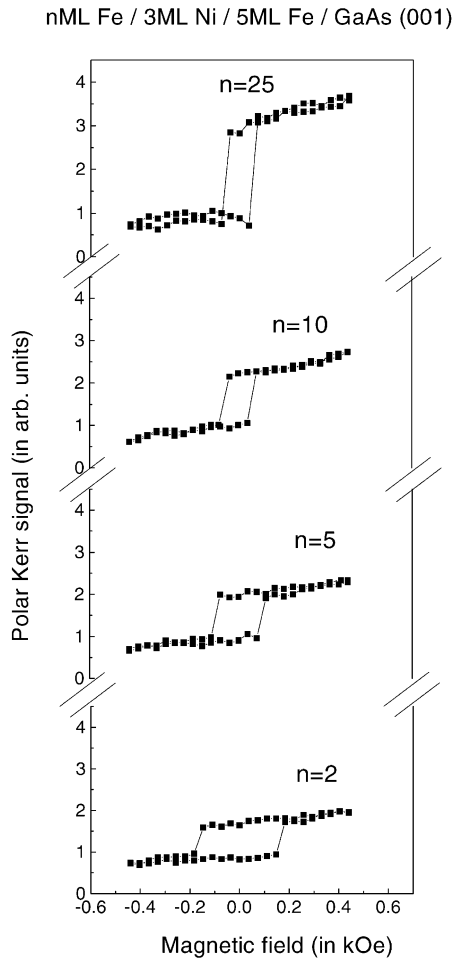


Fig. 6. Polar MOKE signal measured at RT for  $n$ ML Fe/3ML Ni/5ML Fe/GaAs(001) films deposited at RT,  $n = 2, 5, 10$  and 25.

covering Fe with Ni and afterwards with a second Fe layer (e.g. the sample  $n$ ML Fe/3ML Ni/5ML Fe/GaAs(001)), in proportion to their increasing thickness  $n$  (Fig. 6). The coercivity decreases with increasing thickness and saturates around  $n = 10$  ML of the top Fe layer. If this is actually caused by the local perpendicular spin orientation, this means that the forthcoming layers preserve the orientation, which is fairly unlikely. The occurrence of a tilt angle of the easy-axis orientation with respect to the surface plane seems to be more probable. This cannot result from the competition between the second-order surface and volume out-

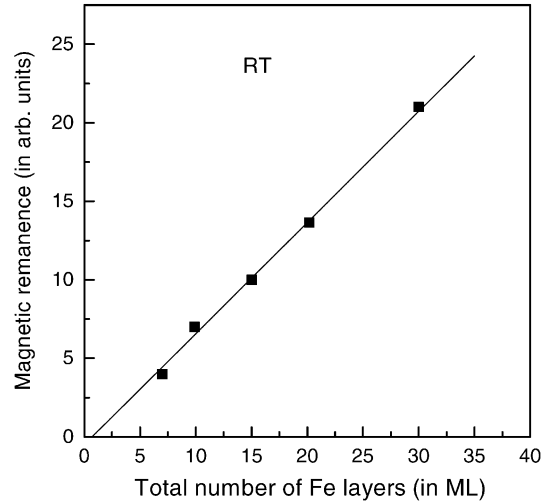


Fig. 7. Kerr signal corresponding to “remanence” effectively in-plane (taken from Fig. 6) for  $n$ ML Fe/3ML Ni/5ML Fe/GaAs(001) plotted vs. the total number ( $= n + 5$ ) of Fe layers.

of-plane anisotropies of different signs, but requires additional fourth-order terms, either of out-of-plane volume or surface anisotropy [21]. On the other hand, an inhomogeneity of the external magnetic field (that is created by a unipolar piece magnet) results in a field component parallel to the film plane, even in the polar geometry. Due to an irregular sample shape and an uncertainty of the sample positioning with respect to the magnet axis, a field gradient could result in the area where the laser beam is focused. Thus, we suggest that the loops correspond to the in-plane easy-axis of magnetization probed by the in-plane contribution of the magnetic field. The uniaxial in-plane anisotropy ( $k_s > 0$ ) typical for Fe/GaAs(001) causes the loop to appear if the external field is, by chance, parallel to the  $\langle \bar{1}10 \rangle$  direction. For other sample orientations, off the in-plane easy-axis of magnetization, we observed only very minor loops. Hence, the present observation confirms the uniaxial in-plane anisotropy of Fe films grown on GaAs(001) [19]. A plot of the Kerr signal in remanence (from the in-plane magnetization loops) vs. the total number of Fe layers confirms that no magnetically dead layer exists at the Fe/GaAs(001) interface (Fig. 7). A linear

extrapolation intersects the coverage axis at 0.7 ML indicating that almost all of the Fe atomic layers are magnetically “alive”. This confirms the chemical inertness of the properly prepared GaAs(001) surface thus preserving magnetic properties of the deposited films. Ferromagnetic order in the interface is required for an efficient spin-dependent electron injection into the GaAs.

### 3.3. Fe/Ni/GaAs(001) system

We studied also  $n\text{Fe}/m\text{Ni}/\text{GaAs}(001)$  films, which is the reverse sequence of the Ni/Fe/GaAs(001) reported above. A reduced effective in-plane anisotropy was expected in this case relative to that observed for Fe films grown directly on top of GaAs(001). This could be simply the result of the lack of the Fe/GaAs(001) interface in the Fe/Ni/GaAs(001) system, because the strong uniaxial in-plane anisotropy of Fe films on GaAs(001) is suggested to originate exclusively from the Fe/GaAs interface [19]. In addition, a possible contribution of the Fe surface anisotropy to the effective anisotropy of the system has to be examined.

Specifically,  $m = 5$  ML of Ni were deposited directly on GaAs(001): (a) at room temperature and (b) at temperature gradually increasing from RT to 400 K. No LEED patterns were detected, indicating that the Ni film does not grow epitaxially. However, it fully covers the GaAs surface (the GaAs spots disappear after depositing  $m = 3$  ML of Ni). In both the cases, (a) and (b), no magnetic response was detected by polar MOKE. In addition to the weak sensitivity, which is a characteristic feature of Ni, it could also happen that the films were magnetically dead or magnetized in plane. The Ni films were further covered with  $n = 3$  ML of Fe at room temperature (3 ML Fe/5 ML Ni/GaAs(001)). A clear LEED pattern typical for Fe(001) was then observed at an electron beam energy of 130 eV (the same LEED pattern was observed for  $n = 2$  ML of Fe as described below and shown in Fig. 8). For higher energies, the spots were diffuse showing a film of worse quality, although grown epitaxially. It indicates that a thin Ni layer still permits the

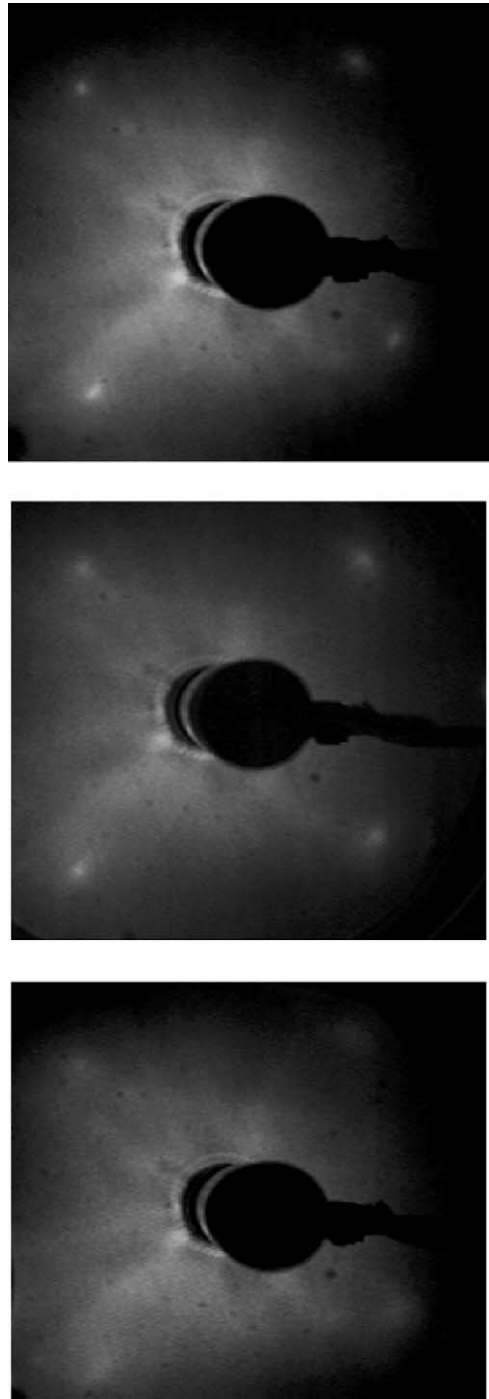


Fig. 8. LEED patterns after 2 ML of Fe deposited on the top of Ni film of various thicknesses  $m$  ( $m = 3, 5$  and  $8$ ), both Ni and Fe deposited at RT.



epitaxial growth of Fe similar to that of the direct deposition of Fe on GaAs(001). The polar MOKE measurements showed that the hard-axis of magnetization is oriented out-of-plane. Further coating with 3 ML of Ni (3 ML Ni/3 ML Fe/5 ML Ni/GaAs(001)) again kills the LEED patterns. The weak MOKE signal compared to that measured for 3 ML Fe/5 ML Ni/GaAs(001) might indicate either no magnetic response or a considerably reduced slope of the hard-axis hysteresis loop. This may be due to the increased saturation field, which is a measure of anisotropy. Finally, the sample was coated with 5 ML of Fe (5 ML Fe/3 ML Ni/3 ML Fe/5 ML Ni/GaAs(001)). The typical LEED patterns of Fe (of good quality) were recovered again. However, no magnetic response was detected at RT in the polar geometry in this case.

Finally, the thickness of the capping Fe layer was reduced to  $n = 2$  ML only. We found out that already 2 ML of Fe, deposited at RT on top of Ni film (with less than  $m = 8$  ML), leads to the occurrence of the clear LEED pattern of 4-fold symmetry which is characteristic for epitaxial Fe(001) grown on GaAs(001) (Fig. 8, in comparison with Fig. 2b).

The hysteresis loops measured by polar MOKE after covering of  $m$  layers of Ni with  $n = 2$  ML of Fe are shown in Fig. 9a. The curves at room temperature are of s-like shape exhibiting a paramagnetic behavior. However, as it is seen in Fig. 9a (right side), the polar MOKE measurements at 150 K resulted in rectangular hysteresis loops of nearly the same remanence for all Ni thicknesses (between  $m = 3$  and 8 ML). This clearly confirms that the 2 ML Fe films on  $m$ Ni/GaAs(001) exhibit an out-of-plane easy-axis of magnetization at low temperature. Any coating of Ni/GaAs(001) with more than 3 ML of Fe rotates the magnetization from out-of-plane back to in-plane. This is clearly shown in Fig. 9b for 4 ML Fe/3 ML Ni/GaAs(001) sample: no magneto-optical response is detected by polar MOKE in this case even at low temperature. Moreover, another coating of the 2 ML Fe/ $m$ Ni/GaAs(001) system with Ni recovers the in-plane orientation of magnetization, which remains in-plane also after several  $n$ Fe/ $m$ Ni sequences.

## 4. Discussion

The interpretation of the LEED data of the films of Fe, Ni  $m$ Ni/ $n$ Fe and  $n$ Fe/ $m$ Ni on GaAs(001) is crucial for understanding the magnetic properties of this system.

### 4.1. Fe/GaAs(001)

The first appearance of a LEED pattern at an Fe thickness of 3 ML (i.e. before the complete coalescence) (Fig. 2b) is possible if the islands are large enough. The magnetic properties of the Fe layers grown on GaAs are sensitive to substrate preparation and growth conditions [3,18,22]. Earlier, Xu et al. [16] have shown that, for Fe films grown on a GaAs(001) surface of high quality, the in situ longitudinal MOKE measurements show an evolution of the magnetic phase as a function of thickness. Thin films of less than 3.5 ML of Fe are reported [16] to be nonmagnetic at room temperature. A significant longitudinal MOKE signal was first detected at a thickness of 3.5 ML, with the intensity linearly proportional to the magnetic field in the range investigated ( $\pm 2000$  Oe). The lack of magnetic signal for the first 3.5 ML was suggested to be due to the small initial cluster size, which prevents the development of magnetic ordering, or the ordering below room temperature [16]. This is in agreement with our results. For thinner films (i.e. less than 3.5 ML), the present polar MOKE curves become s-shaped when they are measured at low temperatures (e.g. at 150 K, Fig. 3). The exchange interaction within the clusters becomes strong enough at 150 K to lead to an internal ferromagnetic ordering in the superparamagnetic phase. The lack of hysteresis indicates that the ferromagnetic phase did not yet develop at this temperature or that the easy-axis lies in the film plane. The loops taken in longitudinal geometry clearly show hysteresis indicating the onset of ferromagnetism at room temperature (with in-plane easy-axis) only after 4.8 ML of Fe [16]. This is in agreement with a recent report on the first anisotropic magnetoresistance signal detected at a thickness of 4.9 ML that clearly marks the onset of the ferromagnetic phase [23]. This study indicates that a superparamagnetic to ferromagnetic phase

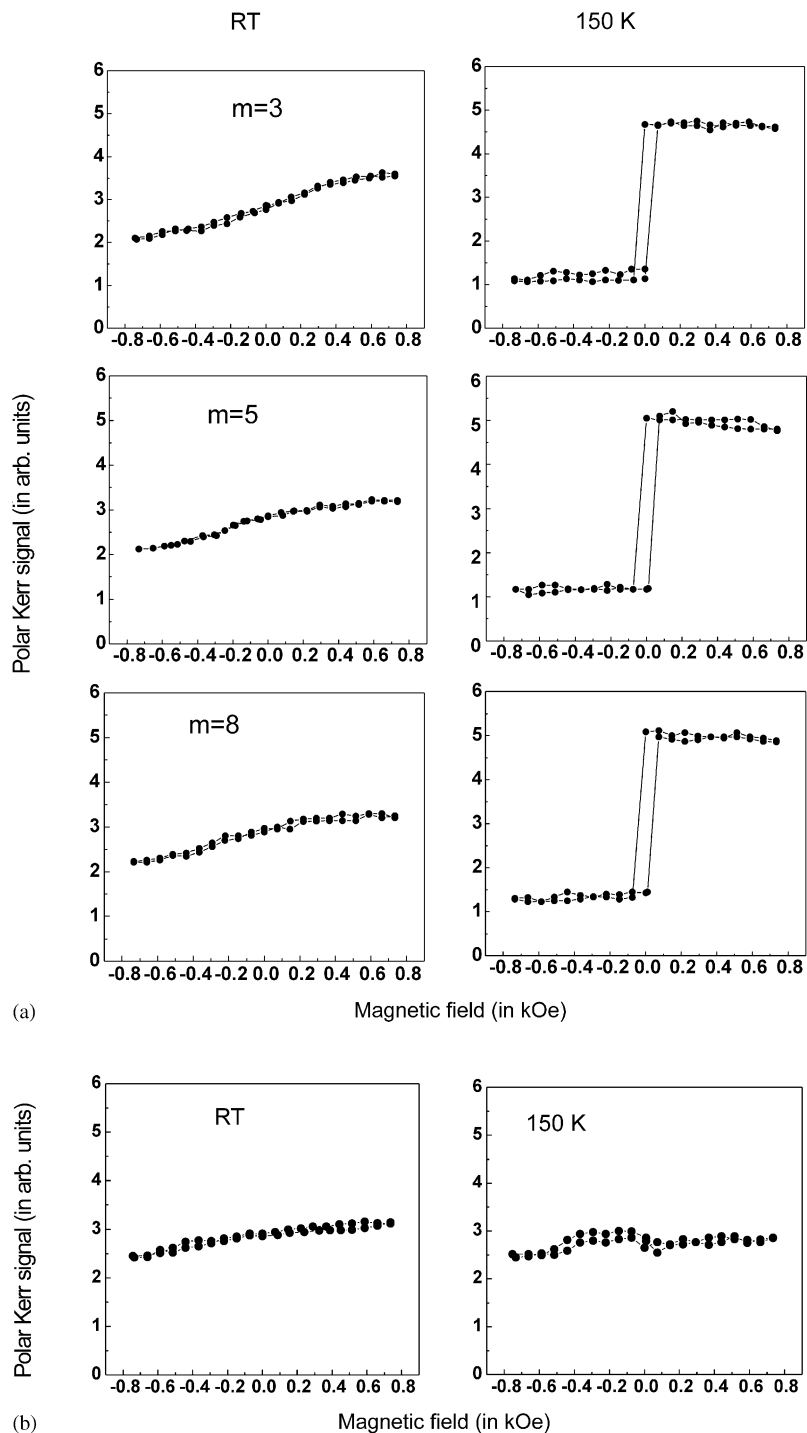


Fig. 9. (a) Polar MOKE signal measured at RT and 150 K for 2 ML Fe/*m* ML Ni/GaAs(001), *m* = 3, 5 and 8, both Ni and Fe prepared at RT. (b) Polar MOKE signal measured at RT and 150 K for 4 ML Fe/3 ML Ni/GaAs(001), included for comparison.

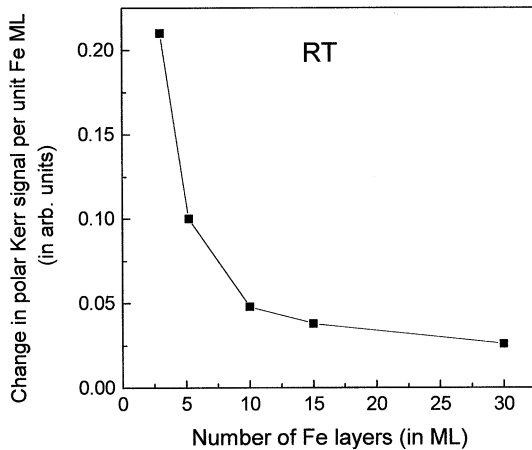


Fig. 10. Total change of polar Kerr signal per unit Fe monolayer measured at RT for  $n$ ML Fe/GaAs(001) in the range of applied field ( $\pm 750$  Oe) vs. the number ( $n$ ) of Fe layers.

transition is proven by the direct observation of a GMR effect in the thickness range between 4.9 and 6.3 ML. Finally, it is reported there that the transition from the superparamagnetic to the ferromagnetic phase is completed after the deposition of more than 6.3 ML [23].

This is consistent with the present results obtained by polar MOKE, which shows that the  $M$ – $H$  curve for  $n = 3$  is “easiest”. The total change of the Kerr signal over the range of the applied field ( $\pm$  maximum field available in the experimental setup; data taken from Fig. 4) is plotted per unit Fe monolayer as a function of the Fe film thickness (Fig. 10). For  $n = 3$  the difference signal is maximum, then decreases fast with increasing thickness. We think that this is due to the islands’ coalescence forcing a long-range ferromagnetic order in the film plane, rather than due to a superparamagnetic behavior of almost isolated clusters. Remarkably, change of slope of this characteristic corresponds to the thickness where the clusters are ferromagnetically coupled and magnetized in-plane. The further decrease of the signal, which is actually detected along the hard-axis, simply reflects the increasing thickness of the ferromagnetic film ( $M$ – $H$  curves become “harder”). The hysteresis loop observed by Xu et al. [16] at low temperature (in longitudinal geometry) reflects long-range ferromagnetic order

observed before the islands coalesce, due to the interparticle interaction.

In order to obtain a good ferromagnetic order at room temperature, which is necessary for an effective spin polarization, the Fe films on GaAs(001) should be at least 7 ML thick, i.e. above the percolation limit at room temperature. On the other hand, in order to promote its perpendicular magnetization, the film should be as thin as possible (only then an out-of-plane surface anisotropy can overcome the shape anisotropy of the BCC-Fe film). Unfortunately, Fe has its coalescence limit at a thickness beyond the limit of perpendicular magnetization (which is determined by the roughness dependence of effective  $k_s$  also) [24].

#### 4.2. Ni/Fe/GaAs(001)

Further deposition of Ni on the top of Fe films of up to 3 ML thickness (even for  $n = 2$  ML of Fe, i.e., for that thickness where the LEED pattern is not yet visible if Fe is grown on GaAs(001)) restores the LEED patterns (Fig. 5). This could mean that Fe and Ni form a kind of BCC-clustered system that adapts its atomic spacing to that of GaAs(001) and permits epitaxial growth, or that the patterns simply reflect the Fe three-dimensional cluster existing in this thickness range. By increasing the thickness of Fe above the coalescence limit the epitaxial growth of Ni is suppressed. Hence, Ni growth has to be considered now as the Ni growing on the Fe(001) buffer layer, although with a distorted structure. Gordon et al. [25] have observed a significant strain relaxation occurring between 3.5 and 6 ML of Fe deposited on GaAs(001) which coincides with the switching of growth mode from Volmer–Weber type to a quasi Frank–van der Merwe type. This is accompanied by a significant improvement of the local ordering. The thicker films (6–15 ML) are reported there [25] to have stabilized BCT symmetry, with a  $c/a$  ratio of 1.03. This kind of structure modification might be responsible for the non-epitaxial growth of Ni, contrary to its pseudomorphic growth on monocrystalline Fe(001) in this thickness range reported by Mijiritskii et al. [26].

Coating of Fe films with Ni reduces the polar magneto-optical response of the system. This is due to an additional volume anisotropy of Ni, which favors the in-plane magnetization and thus increases the saturation field along the hard axis. In particular, the growth of a Ni capping layer on 2 ML Fe/GaAs(001) makes the film continuous. This results in a long-range ferromagnetic order with the magnetization oriented in the film plane. At room temperature, the magneto-optical response suggests paramagnetic behavior. At 150 K, i.e., below the Curie temperature of the system, the magneto-optical response we detected in the polar geometry corresponds to hard-axis behavior of the ferromagnetic film. For comparison, the Ni/Fe(001) bilayer films grown on Ag(001) are reported to be magnetized in plane with the enhanced in-plane anisotropy exceeding that of bulk Fe [27]. The surface anisotropy constant of Ni is reported to be positive [24]. Thus a perpendicular magnetization induced by the Ni overlayer is hardly expected.

#### 4.3. Fe/Ni/GaAs(001)

The most interesting system appears to be 2 ML Fe/*m*Ni/GaAs(001) due to the surface anisotropy of Fe(001) which creates perpendicular magnetization in the ultrathin film limit ( $k_s$  of the Fe/UHV interface is well known to be strongly negative [24]), whereas the relatively strong in-plane anisotropy of the Fe/GaAs(001) interface is simply ruled out from the system (by the Ni spacer placed between the Fe layer and GaAs substrate). The question is about the actual structure of Fe grown on the top of the Ni film. The arguments of imperfect Ni structure (a kind of mixture of BCC and FCC phases?) could explain the occurrence of the LEED patterns of BCC Fe(001) after an Fe overlayer is deposited onto the Ni film (even  $m = 8$  ML thick) (e.g. 2 ML Fe/Ni/GaAs(001), see Fig. 8). The lack of a LEED pattern before covering with Fe and its appearance afterwards leads to the conclusion that this is caused by Ni growth persisting in a slightly “disordered” manner. Nevertheless, while the structure of the Ni film remains unclear, the Fe/*m*Ni/GaAs(001) system exhibits a clear BCC(001) crystallographic

order. The strong capability of Fe to maintain its crystallographic BCC order and to make Ni adapt its structure to the one of GaAs(001) may be responsible for this effect. It is known from literature that covering material dictates crystallographic structure throughout the whole film while this costs only small energy compared to that of the original stacking of the film (e.g. FCC stacking of Co in Cu/Co/Cu(111) sandwich structures [28]). We stress the fact that the lack of a LEED pattern is an insufficient argument to rule out that BCC-Ni(001) has grown, in particular in the case of an extremely rough surface. It is suggested, based on the careful RHEED studies of Fe films grown on GaAs(001) at RT, that actually BCC-Ni(001) growth proceeds in this way [29].

O’Brien et al. reported that, for the Fe/Ni/Cu(001) system, the Fe films of less than 5 ML thickness couple ferromagnetically to Ni/Cu(001) [12]. Ultrathin films (up to 2.5 ML) are homogeneously magnetized and show an FCC structure which is induced by the FCC-Ni underlayer. Considering all anisotropy energy terms contributing to the effective anisotropy of the system, they found that, for 2 ML thick capping layer of Fe, the Ni film below about 50 ML thick is expected to have out-of-plane easy-axis [12]. Unfortunately, the agreement with experiment was poor since the Fe capping layer has no effect on the Ni magnetization direction and, below 7 ML, the Ni film remains magnetized in plane [12]. On the contrary, Sander et al. [30] showed that, with less than 1 ML of Fe capping layer, the easy-axis of magnetization of FCC Ni(111) film on W(110) reorients from in-plane to out-of-plane. This is exactly what we observed except for the structure of the Ni film which is clearly FCC and, what is more important, of (111) orientation if grown on W(110) [30]. Thus the comparison is less significant than that with, e.g. Fe/Ag(001) system of clearly negative  $k_s$  for both the Fe/UHV and the Fe/Ag interfaces [24]. In the case of a Ni film grown on GaAs(001), its structure remains questionable, although the subsequently deposited Fe film exhibits a clear BCC structure [31] and (001) orientation (Fig. 8). This can mean that actually a BCC-Ni(001) film is generated and the system

follows the property expected for BCC-Fe(001) ultrathin films on (001)-oriented metallic substrates. The magnetization curves measured for 2 ML Fe/*m*Ni/GaAs(001) films by polar MOKE at room temperature indicate the paramagnetic state, which is expected due to the reduced Curie temperature (Fig. 9a, left side). In the thickness range of 2 ML for continuous Fe layer, the Curie temperature is expected to be reduced due to the finite size effect, in particular in the case where only the Fe capping layer is ferromagnetically coupled. However, at low temperatures the easy-axis of magnetization is perpendicular to the film plane, independent of the thickness of the Ni layer underneath (Fig. 9a, right side). Thus, the out-of-plane magnetization in the present 2 ML Fe/*m*Ni/GaAs(001) system is interpreted rather as the result of a strong perpendicular surface anisotropy of 2 ML film of Fe on top of the Ni layer, than the temperature driven spin reorientation transition. This is proved by a set of experiments provided in longitudinal MOKE geometry and described elsewhere [31]. Furthermore, a negative volume anisotropy, which favors a perpendicular magnetization, was earlier observed for BCC-Fe films on Ni (above the thickness of Fe corresponding to FCC–BCC transition), arising because of the strain in the Fe overlayer [32]. The role of the Ni underlayer can be qualitatively considered as a seed-layer, which cancels the in-plane anisotropy existing at the Fe/GaAs interface. However, the Ni layer introduces new terms in the effective anisotropy energy balance of the Fe/Ni/GaAs(001) system. These new terms, that likely support the out-of-plane magnetization, can originate in a volume anisotropy of Ni, as well as in an Fe/Ni interface anisotropy. No quantitative analysis of the Fe/Ni interface anisotropy was possible with the experimental setup being used. However, analysis for two sample series: (DML Fe/DML Ni)<sub>*x*</sub>/GaAs(001), for *x* = 1 and 2, is in progress now and *k<sub>s</sub>*/(Fe/Ni) will be obtained. It is worth mentioning that substitution of the Ni layer with a nonmagnetic noble metal (NM) introduces a strong perpendicular Fe/NM interface anisotropy term. This leads to the easy-axis of magnetization again oriented out-of-plane within the same thickness range of the Fe overlayer [31]. However,

this results in an FM/NM/SC structure that is unsuitable for a spin-dependent carrier transport; at least in the case where the NM layer thickness is above the length spins lose their coherency.

The almost constant value of remanence measured for systems of various Ni thicknesses (Fig. 9a, right side) indicates that no more than 3 ML of Ni contribute to the polar magnetization. This can be caused by exchange coupling between the Ni film and Fe overlayer that extends over a couple of the topmost atomic layers of Ni, even in the case where the BCC-Ni is nonmagnetic. The only available data allow comparing it with the magnetic properties of the 1 ML Ni/Fe and 2 ML Ni/Fe slabs calculated with the FLAPW method by Lee et al. [33]. BCC-Ni is found to be ferromagnetic in this case. Its magnetic moment is even increased above the bulk value due to the Ni–Fe hybridization. Another possibility is that Ni contribution to the magneto-optical response of the system comprises the Ni film as a whole; however, it is overridden by the strong signal from the Fe overlayer. Its variation with the increasing Ni thickness could not be seen in this case. This is again in agreement with the experiment of Sander et al. [30] who found an increase of the remanent polar Kerr signal increasing Ni thickness up to *m* = 3 ML, but a further increase of the Ni coverage up to 5 ML did not increase the Kerr intensity. In both cases (Fe/Ni/GaAs(001) and Fe/Ni/W(110)), the in-plane magnetization is recovered at a critical thickness of the BCC-Fe overlayer close to 4 ML (Fig. 9b). However, the origin of this easy-axis rotation can be different for both systems. This seems to be in agreement with the simple argument of the shape anisotropy of a homogeneously magnetized BCC-film that favors the in-plane magnetization already in this thickness range. The in-plane magnetization detected at the same thickness of Fe (above 4 ML) grown on Ni(111)/W(110) can be induced also by an FCC to BCC transition of the Fe film [30].

In the case of FCC-Ni, which is ferromagnetic, the exchange coupling between the Ni and the Fe overlayer should result in ferromagnetic ordering at RT (the Curie temperature of a bulk FCC-Ni equals 627 K), but only if the Ni film is thick enough [34]. On the other hand, any further

increase of the Ni film thickness can lead to a different anisotropy energy balance resulting in an in-plane magnetization. In the case of BCC-Ni, that is expected if grown on GaAs(001), nothing is known about its magnetic properties. The exceptional out-of-plane magnetization of BCC-Fe films occurs only in the limit of a few monolayers [9–11]. Usually, the thickness dependence of Curie temperature is a strong one in the ultrathin film regime [35]. Even in the case of the 2 ML Fe/8 ML Ni bilayer, that includes 8 ML of Ni but likely no more than three of them are magnetically alive, the actually ferromagnetic film consists of five atomic layers only. Then, the “2 ML Fe + 3 ML Ni” has to be considered as a separate ferromagnetic film and its reduced Curie temperature is easy to be understood. By careful deposition one might find the thickness of Fe capping layer that preserves ferromagnetic order at RT with the easy-axis still perpendicular to the film plane. Also, the observed independence of the total anisotropy of the 2 ML Fe/Ni system on its thickness can be explained, assuming that BCC-Ni film is nonmagnetic except for a constant number of the atomic layers just interfacing with Fe.

In addition, any coating with different materials reduces the out-of-plane surface anisotropy of Fe and favors an in-plane magnetization. Thus the out-of-plane magnetized Fe-based film exhibiting ferromagnetic order at and above room temperature is rather unexpected. For practical applications, systems exhibiting the out-of-plane magnetization in an extended thickness range are highly required.

## 5. Summary

The structural and magnetic properties of Fe and Ni, as well as of Ni/Fe and Fe/Ni bilayers, carefully prepared on GaAs(001), were examined. The lack of a LEED pattern before covering of the Ni film with the Fe overlayer, and its appearance afterwards, leads to the conclusion that this is caused by Ni growth persisting in a BCC-structure, although in a lightly disordered manner. The Fe/*m*Ni/GaAs(001) system exhibits a clear BCC(001) crystallographic order. The strong capability of Fe to maintain its BCC-structure

and to make Ni adapt its structure to the one of GaAs(001) is suggested to be responsible for this effect. However, any coating of Fe/GaAs(001) with Ni does not result in its pseudomorphic growth and a BCC-Ni cannot be stabilized in this way.

We showed that the Fe/Ni bilayer on GaAs(001) could be magnetized out-of-plane. The observed rotation of the easy-axis of magnetization from in-plane to out-of-plane is due to the strong perpendicular surface anisotropy of Fe. It is found that 2 ML of Fe deposited on top of a Ni film are sufficient to magnetize the system out-of-plane. Any further coating with Fe or Ni recovers the in-plane magnetization. The almost constant value of remanence indicates that no more than 3 ML of Ni contribute to the polar magnetization. This could mean that the BCC-Ni film is nonmagnetic or only its three topmost atomic layers are magnetically alive due to the Fe/Ni hybridization.

There is sufficient novelty and future scope of work in the Fe/Ni/GaAs(001) system to clarify the mode of Ni growth, its magnetic properties and its effect on the magnetic properties of the Fe overlayer.

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## References

- [1] G.A. Prinz, *Phys. Today* 48 (1995) 58.
- [2] E.M. Kneeder, T. Jonker, P.M. Thibado, R.J. Wagner, B.V. Shanabrook, L.J. Whitman, *Phys. Rev. B* 56 (1997) 8163.
- [3] B. Lepine, C. Lallaizon, S. Ababou, A. Guivarch, S. Deputier, A. Filipe, F. Nguyen Van Dau, A. Schuhl, F. Abel, C. Cohen, *J. Crystal Growth* 201/202 (1999) 702.
- [4] M. Gester, C. Daboo, R.J. Hicken, S.J. Gray, A. Ercole, J.A. Bland, *J. Appl. Phys.* 80 (1996) 347.
- [5] A. Filipe, A. Schuhl, P. Galtier, *Appl. Phys. Lett.* 70 (1997) 129.
- [6] A. Hirohata, Y.B. Xu, C.M. Guertler, J.A.C. Bland, *J. Appl. Phys.* 85 (1999) 5804.

- [7] P.R. Hammar, B.R. Bennett, M.Y. Yang, M. Johnson, *Phys. Rev. Lett.* 83 (1999) 203.
- [8] R. Fiederling, M. Keim, G. Reuscher, W. Ossau, G. Schmidt, A. Waag, L.W. Molenkamp, *Nature* 402 (1999) 787.
- [9] N.C. Koon, B.T. Jonker, F.A. Volkening, J.J. Krebs, G.A. Prinz, *Phys. Rev. Lett.* 59 (1987) 2463.
- [10] D. Li, M. Freitag, J. Pearson, Z.Q. Qiu, S.D. Bader, *Phys. Rev. Lett.* 72 (1994) 3112.
- [11] C. Liu, S.D. Bader, *J. Magn. Magn. Mater.* 93 (1991) 307.
- [12] W. O'Brien, T. Droubay, B.P. Tonner, *Phys. Rev. B* 54 (1996) 9297.
- [13] H.J.G. Draaisma, W.J.M. de Jonge, F.J.A. den Broeder, *J. Magn. Magn. Mater.* 66 (1987) 351.
- [14] Qi-Kun Xue, T. Hashizume, T. Sakurai, *Appl. Surf. Sci.* 141 (1999) 244.
- [15] K. Yamaguchi, Z. Qin, H. Nagano, M. Kobayashi, A. Yoshikawa, K. Takahashi, *Jpn. J. Appl. Phys.* 36 (1997) L1367.
- [16] Y.B. Xu, E.T.M. Kernohan, D.J. Freeland, A. Ercole, M. Tselepi, J.A.C. Bland, *Phys. Rev. B* 58 (1998) 890.
- [17] E.M. Kneedler, T. Jonker, *J. Appl. Phys.* 81 (1997) 4463.
- [18] J.J. Krebs, B.T. Jonker, G.A. Prinz, *J. Appl. Phys.* 61 (1987) 2596.
- [19] M. Zolfl, M. Brockmann, M. Kohler, S. Kreuzer, T. Schweinbock, S. Miethaner, F. Bensch, G. Bayreuther, *J. Magn. Magn. Mater.* 175 (1997) 16.
- [20] H. Takeshita, H. Akinaga, M. Ehinger, Y. Suzuki, K. Ando, K. Tanaka, *Jpn. J. Appl. Phys.* 34 (1995) 1119.
- [21] H. Fritzsche, J. Kohlhepp, H.J. Elmers, U. Gradmann, *Phys. Rev. B* 49 (1994) 15665.
- [22] G. Bayreuther, unpublished results.
- [23] Ch.Gurtler et al., 44th MMM, San Jose, USA. BG-05 paper, 1999, p. 89.
- [24] U. Gradmann, in: K.H.J. Buschow (Ed.), *Handbook of Magnetic Materials*, Vol. 7, Elsevier, Amsterdam, 1993, pp. 1–96.
- [25] R.A. Gordon, E.D. Crozier, D.T. Jiang, T.L. Monchesky, B. Heinrich, *Phys. Rev. B* 62 (2000) 2151.
- [26] A.V. Mijiritskii, P.J.M. Smulders, V.Ya. Chumanov, O.C. Rogojanu, M.A. James, D.O. Boerma, *Phys. Rev. B* 58 (1998) 8960.
- [27] B. Heinrich, S.T. Purcell, J.R. Dutcher, K.B. Urquhart, J.F. Cochran, A.S. Arrott, *Phys. Rev. B* 38 (1988) 12879.
- [28] K. Heinz, S. Muller, L. Hammer, *J. Phys.: Condens. Matter* 11 (1999) 9437.
- [29] X.F. Jin, private communication.
- [30] D. Sander, A. Enders, C. Schmidhals, J. Kirschner, *J. Appl. Phys.* 81 (1997) 4702.
- [31] M. Przybylski, S. Chakraborty, F. Zavaliche, J. Kirschner, to be published.
- [32] Yi Li, C. Polaczyk, D. Riegel, *J. Magn. Magn. Mater.* 165 (1997) 227.
- [33] J.I. Lee, S.C. Hong, A.J. Freeman, C.L. Fu, *Phys. Rev. B* 47 (1993) 810.
- [34] R. Zhang, R.F. Willis, *Phys. Rev. Lett.* 86 (2001) 2665.
- [35] M. Przybylski, *Hyper Interact.* 113 (1998) 135.