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Out-of-plane magnetized Fe-based multilayers on GaAs(0 0 1)

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Abstract

We report on the out-of-plane magnetization in Fe/Ni and Fe/Au bilayers grown on GaAs(0 0 1). For Ni thicknesses of several monolayers, the magnetization of the Fe/Ni bilayer increases linearly with the Fe film thickness, which is typically observed when Fe grows in the BCC-structure. For thicknesses up to 8 ML, Ni is nonmagnetic such as in the Fe/Ni/GaAs(0 0 1) structure. This could be due diffusion of Ga. Up to thickness of 2 ML the Fe overlayer is magnetized out-of-plane, however at low temperatures only. The nature of the out-of-plane spin orientation is discussed, also in view of the crystallographic structure of the Fe/Ni bilayer and by comparison with the Fe/Au bilayer grown on GaAs(0 0 1).

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1. Introduction

The potential integration of magnetic elements with semiconductor-based circuits leads to an increased interest in the growth of epitaxial ferromagnetic metal films (FM) onto semiconductor substrates (SC) and in the experiments on spin-dependent electron transport at the FM/SC interface. However, the spin polarization of the electrons injected into SC, detected via the polarization of light emitted due to the recombination of electrons and holes, have only shown effects of less than 1–2% [1,2]. This is according to the theoretical obstacles originating in the less than 100% spin-polarization of the electrons in metal ferromagnets, and in the conductivity mismatch between metal and semiconductor, both preventing diffusive spin-polarized electrons being effectively injected [3]. Thus several modifications of the concept are proposed, like the integration of a tunnel junction to produce hot electrons as a part of the spintronic device, and the search for 100%

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spin-polarized materials that are necessary to obtain an on/off resistance ratio comparable with that of currently available semiconductor devices.

The perpendicularly magnetized films appear to be of greater interest both for recording and future spin-electronics applications. Even for efficient optical detection of spin polarization of the electrons injected from the FM film into the SC element only, a magnetic easy-axis normal to the film plane is highly desirable. The orientation of the easy-axis of magnetization is determined by the competition between different anisotropy energies. Perpendicular magnetization for monolayer films can be expected if the spin–orbit-induced anisotropy at the surface could overcome the magnetic dipole interaction. To achieve this in the case of metal ferromagnets grown on a GaAs(0 0 1) substrate, we used a sequence of ultrathin Ni and Fe films to make the resultant anisotropy energy favorable for the out-of-plane magnetization. We showed that the Fe/Ni bilayer on GaAs(0 0 1) could be magnetized out-of-plane [4]. In this paper, we discuss the nature of this out-of-plane spin orientation, also in the view of a crystallographic structure of the Fe/Ni bilayer and by comparison with the Fe/Au bilayer grown on GaAs(0 0 1).

2. Experimental

All of our experiments including GaAs(0 0 1) 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 magneto optical Kerr effect (MOKE) for the magnetic analysis. MOKE loops were collected by using an electromagnet (with a maximum field of 30 mT), whose axis makes a non-zero angle with respect to the specimen surface. Therefore, also the perpendicular magnetization can be detected, since it gives an ellipticity about one order of magnitude higher than the in-plane magnetization. The GaAs substrates were prepared by preannealing at 520°C, subsequent sputtering for 1.5 h at the same temperature and for 1 h at 590°C, with low energy (500 eV) Ar ions. Ni and Fe were deposited at a rate of about 1–1.3 Å/min, by electron beam evaporation from a thoroughly outgassed high-purity nickel and iron rod, respectively. Au was evaporated from a molybdenum crucible at nearly the same rate. The growth was performed at room temperature (RT), under UHV conditions ($p < 10^{-10}$ mbar).

3. Results and discussion

The STM images of the GaAs(0 0 1) substrates sputtered at elevated temperature show atomically flat terraces of tens of nanometers width separated by bi-atomic steps [4]. A Ga-rich $p(4 \times 6)$ surface reconstruction, resulting from a considerable As depletion compared to the As terminated surface, was clearly deduced from the STM images (via the 4-fold periodicity along [−1 1 0] direction) and from the LEED patterns [4].

For Ni films grown on GaAs(0 0 1) no LEED spots were observed below a Ni thickness of about 8 ML. Up to 8 ML of Ni, no detectable magnetic response was measured by MOKE, neither at RT nor at 150 K (our lowest achievable temperature). Nevertheless, we found that the RT deposition of about 2 ML of Fe, on the top of Ni film (less than 8 ML), leads to the occurrence of a clear LEED pattern of 4-fold symmetry which is characteristic for epitaxial BCC-Fe(0 0 1) grown on GaAs(0 0 1) surface [4]. While the structure of the Ni film remains unclear, the Fe/Ni/GaAs(0 0 1) system exhibits a clear crystallographic order up to about 5 ML of Fe if Fe is grown at RT. The BCC-like structure of the Fe overlayer is suggested by the resemblance between the LEED patterns of the epitaxial BCC-Fe(0 0 1) films grown on GaAs(0 0 1) with and without Ni underneath. If this is actually the case, this indicates at least two possibilities: (1) ultrathin Ni layer, independently of its crystallographic structure and "degree" of disorder, still permits an epitaxial growth of BCC-Fe by some preferable energy balance for the BCC-stacking of the Fe/Ni system on GaAs(0 0 1) or (2) Ni grows on GaAs(0 0 1) as a BCC-Ni(0 0 1).

The high capability of Fe to maintain its crystallographic order and to make Ni adopt its structure to the one of GaAs(0 0 1) can be responsible for this forced BCC-stacking effect after covering the Ni film with Fe. The most probable explanation of the LEED pattern recovering as the result of Fe deposition might be provided by an interdiffusion driven recrystallization, maybe a spontaneous formation of an ordered BCC-FeNi compound which works as a template for the epitaxial growth of Fe. In the case of BCC-Ni growth on GaAs(0 0 1), which is actually expected [5], a further growth of BCC-Fe for low coverage is easy to be understood.

The crystallographic structure of the Fe overlayer cannot be decided from visual LEED inspection because only the two-dimensional translational symmetry in the film plane can be deduced, which is the same for BCC and FCC structures (which are both special cases of the body centered tetragonal lattice). Thus we decided to examine the crystallographic structure of the Fe overlayer indirectly, by probing its magnetic properties with MOKE. In the hypothesis of FCC-Ni growth on GaAs(0 0 1) followed by FCC-Fe growth on the top of it, the magnetic properties of the Fe overlayer should reflect its structure through a typical disappearance of ferromagnetic order above a certain thickness and appearance of a low-spin antiferromagnetic phase [6]. If Ni grows BCC on GaAs(0 0 1) and the Fe capping layer grows also BCC (also in the case of FCC–BCC transition of Ni as the result of Fe growth), the magnetic properties of the BCC-Fe should be reflected in a linear dependence of the remanent magnetization on the Fe thickness over the whole thickness range. The corresponding experiment was performed for Fe capping layers grown on a 5 ML thick Ni film previously deposited on GaAs(0 0 1). Both Ni and Fe were deposited at RT. The longitudinal MOKE measurements performed at RT along [1 1 0] direction (i.e., along the easy-axis of magnetization of Fe if deposited directly on GaAs(0 0 1)), showed rectangular hysteresis loops above the thickness of about 3 ML of Fe. The results are summarized in Fig. 1 by plotting Kerr ellipticity at remanence vs. the number of Fe atomic layers deposited on the top

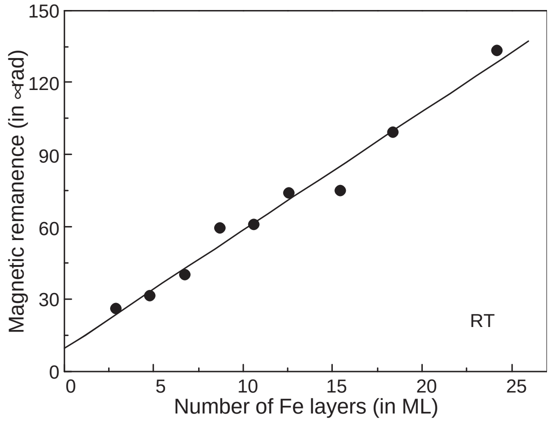

Fig. 1. Kerr ellipticity corresponding to in-plane "remanence" for Fe deposited on the top of 5 ML thick Ni film previously grown at RT on GaAs(0 0 1) plotted vs. the number of Fe monolayers.

of Ni film. It is seen that the ellipticity at remanence increases linearly with the film thickness which is typically observed when Fe films keep the BCC-structure independently of their thickness. In particular, the disappearance of ferromagnetic order above the thickness of about 4 ML as reported for Fe grown on the single-crystalline FCC-Ni(0 0 1) substrate [6], is not observed . Thus, a linear fit performed through the ellipticity data points supports the growth of BCC-Fe on Ni/GaAs(0 0 1).

Our previous experiments have clearly shown that the 2 ML thick Fe film on the top of Ni/GaAs(0 0 1) exhibits an out-of-plane easy-axis of magnetization at low temperatures [4]. To identify the origin of the out-of-plane spin orientation, the magnetization behavior of the Fe overlayer on a 4 ML thick Ni film was probed with respect to both thickness and temperature in a MOKE setup which enables us to distinguish between the in-plane and out-of-plane magnetization. In the case of a 1.5 ML thick Fe film grown under the same conditions as above, neither in-plane, nor out-of-plane loops were detected at RT. With gradually decreasing the temperature, paramagnetic loops of increasing ellipticity were measured from about 190 K (the open squares in Fig. 2), which open up in out-of-plane hysteresis loops at about 165 K whose ellipticity eventually reaches 150 μrad at about 150 K (the solid line in Fig. 2). This behavior

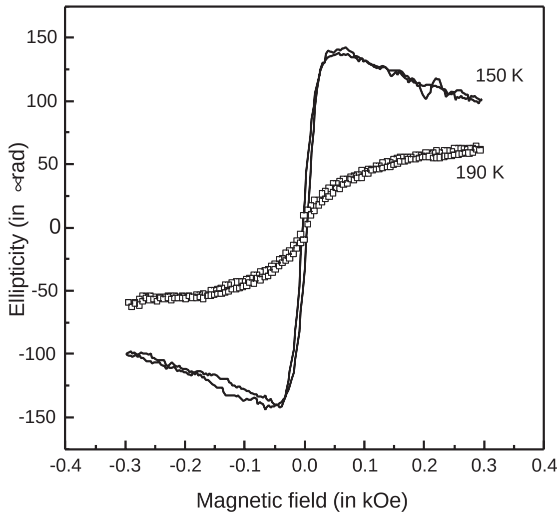

Fig. 2. MOKE ellipticity (measured in a configuration enabling to distinguish between the in-plane and out-of-plane magnetization) measured at 150 K (solid line) and 190 K (open squares) for 1.5 ML of Fe deposited on the top of a 4 ML thick Ni film on GaAs(0 0 1).

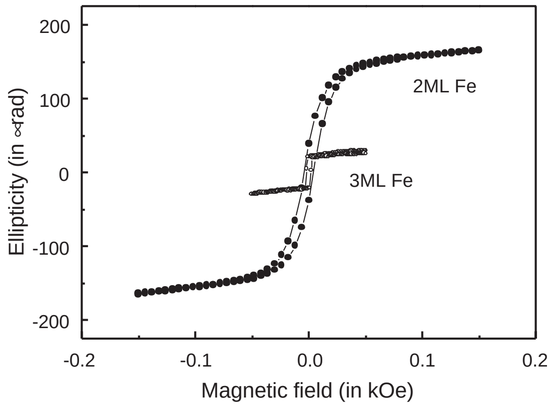

Fig. 3. MOKE ellipticity (measured in a configuration enabling to distinguish between the in-plane and out-of-plane magnetization) measured at RT for 2 ML of Fe (dots) and 3 ML of Fe (open circles), deposited on the top of a 4 ML thick Au film on GaAs(0 0 1).

excludes the possibility of a temperature driven spin reorientation transition in the Fe film and clearly confirms that the reduced Curie temperature effect is responsible for the absence of magnetization at RT (previously seen with a polar MOKE only [4]) with decreasing the thickness of the Fe overlayer. Due to the MOKE geometry we used (a component of the field perpendicular to the film surface is present), the magnetization rotates towards the film plane with increasing magnetic field, giving rise to the observed decrease in the measured ellipticity at higher fields.

Any further coating of 2 ML Fe/Ni/GaAs(0 0 1) with additional Fe rotates the magnetization from out-of-plane back to in-plane. Thus, the out-of-plane magnetization is interpreted as a result of a strong perpendicular anisotropy (surface anisotropy constant $k_s < 0$) induced by the surface of the Fe film deposited on the top of the Ni layer, that establishes the total anisotropy energy balance of the system in the thinnest Fe films regime. A comparison to the known k_s values for Fe(0 0 1)/X interface with X = Ag, Au and UHV, which is always negative and increases for the uncoated films [7], seems to be useful for a qualitative understanding of the observed behavior. Following this idea, any substitution of Ni in the Fe/Ni/GaAs(0 0 1) system with Ag or Au should result in an out-of-plane easy-axis of magnetization as well, which is actually the case. The corresponding experiment was performed for 4 ML thick Au film grown on GaAs(0 0 1), covered afterwards with 2 ML of Fe, both deposited at RT. Contrary to the case of Fe/Ni/GaAs, any coating of Au with Fe does not result in a visible LEED pattern. This can be caused by a very complex structure of Au buffer layer if grown on GaAs(0 0 1) and Au segregation. Moreover, any recrystallization caused by the Au and Fe intermixing cannot be expected due to the broad miscibility gap in this case. The MOKE loop measured at RT is shown in Fig. 3. Again, the Fe overlayer is magnetized out-of-plane even at RT in this case, as deduced from the ellipticity values higher with about one order of magnitude in comparison with the corresponding signal for an in-plane magnetized film of the same thickness. By the deposition of 3 ML of Fe on the Au film of the same thickness, the magnetization rotates to the film plane (as it is seen in Fig. 3). This is in agreement with the simple argument of the shape anisotropy that overrides the out-of-plane surface anisotropy of Fe and favors the in-plane magnetization in this thickness range.

From the comparison between the results obtained for 2 ML of Fe deposited on the top of

Ni and on Au, we can conclude about the Curie temperature of 2 ML thick Fe film with respect to the substrate the film is grown on. In the case of 2 ML of Fe grown on a Au-buffer layer at RT, the ferromagnetic order at RT we found is in agreement to the early observation of Bader et al. [8] and Duerr et al. [9]. However, only the latter one claimed the out-of-plane easy-axis, and only for Fe grown at 100 K. Nevertheless, one has to note that our Fe capping layer was grown on an Au-buffer layer deposited on GaAs(0 0 1) that cannot be compared with the monocrystalline Au-substrate. Despite the lower surface free energy of Au in comparison with that of Fe, the layer-by-layer growth of Fe occurs in this case and is explained by presence of the Au surfactant layer during the Fe growth [10]. This growth induced atomic exchange may play a significant role in the formation of a continuous layer and in the resulting onset of the ferromagnetism at RT at a Fe thickness below 2 ML. In the case of 2 ML of Fe grown on Ni/GaAs(0 0 1), the Curie temperature is definitely below 300 K. Nevertheless, the Fe thickness dependence of the Curie temperature for the Fe/5MLNi/GaAs(0 0 1) system is very strong (T_c varies from about 200 K for 1.5 ML to about 400 K for 3 ML of Fe). This is a typical variation in the Fe monolayer regime. In combination with the observed independence of the total anisotropy of the 2 ML Fe/Ni system on its total thickness [4], one can conclude that the BCC-Ni film (of the thickness up to about 8 ML) on GaAs(0 0 1) is nonmagnetic. Our preliminary X-ray magnetic circular dichroism (XMCD) measurements, which enable the separate determination of magnetic properties of different elements in the same sample, confirm that Ni is nonmagnetic in the Fe/Ni/GaAs(0 0 1) system [11]. However, this should be clarified whether our BCC-Ni film is nonmagnetic itself (suggested by theoretical calculations [12]), or nonmagnetic phases form due to the inter diffusion of Ga and/or As. We made some experiments showing that Ga diffusion into the Ni film plays some role when the films are grown at RT, and could be remarkably reduced by decreasing the deposition temperature. This results in the extended thickness range at which the Fe overlayers keep their perpendicular magnetization. Further details concerning the interdiffusion and its influence on magnetic properties of such systems will be published elsewhere [11].

4. Conclusions

We found that Fe films, in monolayer regime, can be magnetized out-of-plane while grown on a several monolayer thick buffer layer previously deposited on GaAs(0 0 1). In this way the Fe/GaAs(0 0 1) interface anisotropy (that forces the in-plane magnetization of Fe), is avoided and the surface anisotropy of Fe establishes the out-of-plane magnetization in the Fe overlayer. Moreover, the Ni film grows BCC on GaAs(0 0 1) (with the crystallographic order significantly improved after covering with Fe), and seems to be nonmagnetic except for the topmost atomic layers just interfacing with Fe. The replacement of Ni with Au does not change (qualitatively) the anisotropy energy balance, but the 2 ML thick Fe layer exhibits the out-of-plane ferromagnetic order at RT in this case.

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