Magnetic Properties of Thin Iron Films

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The incorporation of magnetic layers in semiconductor heterostructures is an increasingly active area of study. There is a great interest in the catalytic, electronic, and magnetic properties of transition metal overlayers on semiconductor substrates in thin film form. Since the metal-semiconductor interface plays an important role in thin film heterostructures, the initial stages of overlayer growth determine the morphology and crystalline structure of subsequent growth. Thin film properties often differ significantly from bulk properties due to surface and interface effects dominating the overall behavior of these films. Iron films show a broad range of magnetic properties depending on film thickness and deposition conditions. We investigate the influence of the initial substrate surface reconstruction on the magnetic behavior of iron films both on GaAs substrates as well as on ZnSe epilayers. Surface reconstruction leads to magnetic anisotropies dominated by an in-plane uniaxial component. We study the growth of ferromagnetic iron to determine the mode of film growth, the interface formation, and the magnetic film properties. Films are characterized by various methods: surface reconstructions are determined by Reflection High Energy Electron Diffraction (RHEED) and magnetic properties by Superconducting Quantum Interference Device (SQUID) measurements with the magnetic field applied along different in-plane directions.

1. Introduction

The growth of single-crystal ferromagnetic films on semiconductor substrates and the incorporation of metal layers in semiconductor heterostructures has attracted considerable attention due to its compatibility with planar electronics (e.g. for inducing magnetic fields in dilute magnetic semiconductors) and due to its potential for spin-sensitive heterostructure devices.

Body-centered cubic (bcc) α -iron is preferentially deposited on GaAs and ZnSe due to the slight lattice mismatch (1.5 % and 1.1 %, respectively). By variation of film thickness and deposition conditions iron films show a broad range of magnetic properties. In this paper we present the influence of the surface reconstruction of the substrate and the influence of the film thickness on the magnetization curves obtained by superconducting quantum interference device measurements.

2. Experimental

A new ultrahigh vacuum chamber, designed for the growth of bcc α -iron and described earlier [1], has been attached to an existing MBE-system allowing us to grow iron directly on (001) GaAs substrates or on ZnSe epilayers.

Prior to growth the (001) GaAs substrates are heated up to 720 °C and kept at that temperature till a streaky RHEED pattern indicates deoxidation of the substrates, usually for a few seconds.

The ZnSe epilayer is grown by ALE (Atomic Layer Epitaxy) [2] in the following way: The shutter of the Zn effusion cell is kept open for 3 sec. After a delay time of 0.5 sec, in which all shutters are closed, the shutter of the Se effusion cell is open for 3 sec. Another delay time of 0.5 sec finishes one growth cycle during which one monolayer of ZnSe is grown. Therefore the substrate with a temperature of 300 °C is exposed to only one kind of source material at one time. The ALE-growth of ZnSe starts with Zn and finishes with Se leading to a Se-terminated surface. The thickness of the ZnSe epilayer is chosen to be 500 Å, which is below the critical thickness at which dislocations start to form.

Figure 1 shows a typical RHEED pattern observed after growth of ZnSe. The distance between the streaks corresponds to a (2x4) reconstruction.



Fig. 1: RHEED pattern along the two [110] directions observed after growth of 500 Å ZnSe in ALE-mode on (001) GaAs.

After ZnSe growth the sample is cooled down to 150 °C and then transferred to the iron chamber via an UHV tunnel without breaking the vacuum. The iron chamber is equipped with an electron beam evaporation source due to the low vapor pressure of iron and the high source temperature required. For details see [1].

The iron layer is grown at a substrate temperature of 165 °C and with a growth rate of approximately 10 Å/min. The thicknesses of the iron films are in the range of 20 - 120 nm. After growth of iron RHEED patterns are taken still in UHV without the effects of oxidation. Then the iron samples, which are not covered with a capping layer, are exposed to air and oxidize. The films are characterized by SQUID measurements with the magnetic field applied along the [100] and [110] in-plane directions.

3. Results and Discussion

Figure 2 shows the results of SQUID measurements taken on a 60 nm thick Fe film on a ZnSe epilayer. There is a shift in horizontal direction due to an offset of the SQUID magnetometer. (Without offset the curves are symmetric with respect to the vertical axis.) For all measurements the magnetic field is applied in the plane of the Fe film.



Fig. 2: Magnetic moment, m, versus applied magnetic field, H, obtained from SQUID measurements. (a) Iron film (60 nm thick) on a ZnSe epilayer; (b) magnification of (a) for low magnetic fields.

The curves of magnetization versus applied magnetic field indicate a saturation of magnetization at low magnetic fields (at 50 G and 750 G for the [100] and [110] direction, respectively). It is clearly visible that the [100] axis is the easy axis of magnetization while the [110] axis is the intermediate axis of magnetization as it is expected for a 60 nm thick film [3].

3.1 Magnetic Field Applied Along In-Plane [100] Directions

By taking a closer look at the magnetic behavior for low applied fields (see Fig. 2) we observe no rectangular hysteresis with one jump in magnetization, but a hysteresis containing two irreversible jumps of the magnetization when increasing/decreasing the magnetic field.

This behavior strongly depends on the thickness of the iron layer: For Fe films with a thickness between 20 and 30 nm we observe a hysteresis with only one jump, whereas the Fe films with a thickness between 60 and 120 nm show the two-jump-behavior.

This can qualitatively be explained by evaluating the energy density E of a magnetic thin film with the in-plane magnetization vector \vec{M} :

$$E = -\vec{M} \cdot \vec{H} + K_{1} \cdot (\alpha_{1}^{2} \alpha_{2}^{2} + \alpha_{2}^{2} \alpha_{3}^{2} + \alpha_{3}^{2} \alpha_{1}^{2}) + K_{u} \cdot \sin^{2}\theta$$

where \vec{H} is the applied magnetic field, K_1 is the fourth-order cubic anisotropy, α_i are the direction cosines of the magnetization with respect to the cubic axes, K_u is a uniaxial in-plane second-order anisotropy, and θ is the angle between the magnetization and the in-plane [110] axis.

The ratio K_u/K_1 is crucial for determining the hysteresis. For the magnetic field applied along the [100] direction the hysteresis shows one jump for $|K_u/K_1| > 1$ (range (1)), two

jumps for $1 > |K_u/K_1| > 0,3$ (range (2)) and again one jump for $0,3 > |K_u/K_1|$ (range (3)) [4]. In range (1) the magnetic field, at which the irreversible jump in the magnetization occurs, decreases with decreasing ratio K_u/K_1 . In range (2) the value of the magnetic field at the first jump increases and for the second jump decreases with decreasing ratio K_u/K_1 .

The evolution of the hysteresis with film thickness for our samples exactly agrees with the behavior predicted by the model: Films with thicknesses of 20 and 30 nm show a hysteresis with one irreversible jump of the magnetization with decreasing jump-field for increasing film thickness, whereas films with thicknesses of 60 and 120 nm show two jumps in the hysteresis. By assuming a decreasing ratio K_u / K_1 with increasing film thickness the fields for the jumps behave according to the model described above. Apparently the ratio K_u/K_1 is larger than 1 for Fe films between 20 nm and 30 nm thick while for the thicker films (60 – 120 nm) K_u/K_1 is between 0,3 and 1.

From this thickness behavior we conclude that the uniaxial anisotropy resulting in the inequivalence of the two in-plane [110]-directions is induced by the interface. Another important fact must be mentioned: We observe hysteresis curves with two irreversible jumps only for the Fe films on a ZnSe epilayer, whereas the Fe films grown directly on GaAs show only one jump in the magnetization for the complete thickness range !

This behavior can be explained in the following manner: By deoxidizing the GaAssubstrate prior to growth of iron directly on GaAs, there is no specific background pressure. Therefore after deoxidation we find Ga- and As-dimers at the surface, which are oriented along the [110] and $\overline{110}$ directions [5].

On the contrary by the insertion of a ZnSe epilayer, deposited by atomic layer epitaxy as described in the previous section, we have a Se-terminated surface due to the finishing epitaxy step with the Se shutter open, where the Se-dimers are oriented along the $[\bar{1}10]$ -direction [6].

For a (2x4)-reconstruction the dimer bond is parallel to the $[\bar{1}10]$ -axis as a result of the orientation of the dangling bonds, and the (2x4)-surface has a pronounced rowlike structure along $[\bar{1}10]$ due to the missing Se-dimer rows. Therefore, the axis perpendicular to the missing dimer rows, which characterize the surface reconstruction of the ZnSe, is an easy axis, whereas the axis parallel to the missing dimer rows is a hard one [5]. Due to the (2x4) surface reconstruction the two in-plane [110]-directions become inequivalent, expressed by the last term in Eq. 1 where θ is the angle between the magnetization and the [110] direction.

The uniaxial anisotropy originates in the interface between ZnSe and Fe and decreases with increasing film thickness, whereas the cubic anisotropy due to the crystal stays is independent of the film thickness. This leads to a decreasing ration K_u/K_1 with increasing film thickness, which has also qualitatively been determined from the evolution of the hysteresis curves.

3.2 Magnetic Field Applied Along In-Plane [110]-Directions

By comparing the hysteresis curves for the magnetic field applied along the two [110] in-plane directions (see Fig. 2), we notice that the magnetic fields of the irreversible jump of the magnetization are different for the two [110] directions by a factor of ap-

proximately 2.4. This also clearly indicates that one of the two in-plane [110] directions is an easy axis for the magnetization, while the other one is a hard axis. This confirms the discussion given above.

In summary there are two contributions determining the hardness of the different inplane axes: The cubic anisotropy, which distinguishes the [100] (easy) from the [110] (hard) axis, and the uniaxial axis, which leads to the inequivalence of the [110] and the 110 are two contributions combine in a way of the contribution [110]

[110] axis. These two contributions combine in a way as theoretically predicted in [7].

4. Conclusion

In summary we have presented (001) iron films with different thicknesses deposited both directly on GaAs as well as on ZnSe epilayers. While the iron films on GaAs show a rectangular hysteresis in the M versus H curves (magnetic field parallel [100]) with one irreversible jump of the magnetization for the whole thickness range, the iron films on the ZnSe epilayer exhibit a different magnetic behavior: The hystereses of thin films (20 - 30 nm) contain only one irreversible jump, but thicker films (60 - 120 nm) lead to hysteresis curves with two irreversible jumps.

This behavior originates in the Fe – ZnSe interface. The ZnSe epilayers are grown by atomic layer epitaxy, where the Se-terminated surface contains a rowlike composition of dimers resulting in inequivalence of the two [110] in-plane directions.

Measurements of M versus H curves with the magnetic field applied along the in-plane [110]-directions are in agreement with the results of the [100]-measurements.

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