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Magnetic anisotropy in Co/Cu(1 1 17): Temperature dependence

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Abstract

The temperature dependence of the in-plane magnetic anisotropy of Co/Cu(1 1 17) has been studied by means of the magneto-optic Kerr effect. Strong variations of uniaxial and biaxial anisotropy contributions are found during the first heating. At two different temperatures the uniaxial anisotropy crosses zero causing a change of magnetic behavior. A correlation between the anisotropy behavior and the temperature dependence of the differential susceptibility is apparent. The susceptibility maxima appear at the temperatures where the magnetic anisotropies become negligible small.

Keywords: Anisotropy – temperature dependent; Anisotropy – uniaxial; Anisotropy – biaxial; Kerr effect; Thin films; Susceptibility

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1. Introduction

Current research on magnetism in thin films is largely focused on the magnetic anisotropy and its relation to structure, morphology and strain [1–12]. In films with perpendicular magnetization interface anisotropies and dipolar energies are the contributions which influence the magnetic behavior most. Their relative strengths determine weather the magnetization flips into the film plane or stays perpendicular [13,14]. As magneto-static energies play a major part in this scenario the creation of magnetic domains has to be considered and the micro-magnetic analysis is absolutely necessary for the understanding of the anisotropy transition [15,16]. The domain formation, however, plays a minor role in magnetic films with in-plane magnetization. In-plane magnetic anisotropies and anisotropy transitions can be easily investigated taking magnetization curves by means of techniques without spatial resolution, e.g. the magneto-optic Kerr effect.

Cobalt films grown on vicinal Cu($11n$) surfaces at room temperature exhibit in-plane uniaxial anisotropy with the easy axis of magnetization parallel to the step edges [17]. For Co on Cu($11n$) the orientational free energy can be written as:

$$V(\varphi) = -K_u\cos^2(\varphi) + K_4\left[\cos^2\left(\varphi-\frac{\pi}{4}\right) - \cos^4\left(\varphi-\frac{\pi}{4}\right)\right], \quad (1)$$

where φ is the in plane angle of magnetization with respect to the step edges and K_u is the uniaxial and K_4 the biaxial anisotropy constant. A positive K_u indicates an easy axis along the step edges while a negative K_u describes an easy axis perpendicular to the step edges. If K_4 is negative the biaxial contribution favors magnetization along the ⟨110⟩ directions, i.e. parallel and perpendicular to the step edges. If

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K_4 is positive the biaxial contribution favors $\langle 100 \rangle$ as easy axes (45° to the step edges). The ansatz is equivalent to the one used in Ref. [18].

Recent investigations of the temperature dependent susceptibility in Co/Cu(1 1 17) revealed unexpected peaks in the ferromagnetic phase. The peaks were attributed to changes of the magnetic anisotropy which was qualitatively proven by Kerr hysteresis loops [19].

The studies were resumed to investigate the temperature effects in more detail. Particularly, the magnetic anisotropy was measured quantitatively as a function of temperature to prove the suggestions of the previous study [19]. The effect of film thickness on the magnetic temperature behavior was addressed as well. The reversible and irreversible behavior of the individual anisotropy transitions were studied in more detail. In the following paragraph a brief summary of essential experimental details are given. In the succeeding chapter the results are presented and discussed in the last paragraph.

2. Film preparation and characterization

The experiments are performed under UHV conditions, base pressure $p \approx 1 \times 10^{-10}$ Torr. Surface preparation and film growth are monitored via medium energy electron diffraction (MEED) and Auger electron spectroscopy. The magnetic properties are investigated by means of the magneto-optic Kerr effect.

Vicinal Cu(11n) surfaces are well studied by means of helium scattering [20] and scanning tunneling microscopy [21,22]. Microscopically the surfaces consist of terraces with (001) orientation, separated by monatomic steps. The terrace width is $n/2$ atomic distances on the average. Step bunching has not been observed [21]. The steps are aligned with the $[1\bar{1}0]$ in-plane direction.

The substrate is cleaned and prepared by cycles of Ar^+ sputtering (600 eV) and subsequent annealing ($T > 670$°C). The procedure is repeated until no traces of any contamination (typically carbon and sulphur) can be detected with Auger electron spectroscopy. After ion bombardment the micro-structure is obtained by carefully annealing [21]. The quality of the surface crystal structure is checked via MEED. The MEED diffraction pattern show pronounced splitting of regular lattice spots indicating the periodic step arrangement on the copper surface [23].

The films are grown at ≈ 45°C with a rate of ≈ 1 ML/min. During electron beam evaporation the pressure does not exceed $p = 5 \times 10^{-10}$ Torr. While growing the films, the intensity of the specular MEED beam is monitored. Electrons with energy of 3 keV are used which hit the surface under an angle of a few degrees. No oscillations of the MEED intensity could be found. The MEED reflectivity remains on a high level in every stage of growth, which means that the MEED pattern are conserved with high quality. A similar behavior was previously found for the growth of Co on Cu(1 1 13) [24].

The magnetic properties of the films are investigated in situ by means of the longitudinal magneto-optic Kerr effect. The optical set-up is similar to the Kerr experiment used by Bader and co-workers [25]. The Kerr ellipticity is measured. Magnetic fields up to 140 Oe can be applied parallel to the film plane. The fields are created by current driven core-less coils mounted inside the vacuum chamber. The response of the magnetic film on small magnetic fields, i.e. the magnetic susceptibility, is measured utilizing the same optical set-up. An ac magnetic field with small amplitude is applied parallel to the film plane. The change of Kerr ellipticity is measured via a phase sensitive amplifier [26,27]. The signal is proportional to the slope of the hysteresis and/or magnetization curve at zero magnetic field. We call that quantity differential susceptibility [28,35]. No additional biasing fields are applied. A high sensitivity has been achieved using this technique. Changes of Kerr ellipticity in the range a 10 nrad can be resolved.

3. Temperature dependence of the anisotropy

Recent investigations of Co/Cu(1 1 17) films of thickness around 2.5 ML showed pronounced maxima in the temperature dependent susceptibility [19]. A typical result is shown in Fig. 1. Besides the susceptibility maximum at the critical temperature T_c, two additional maxima (named T_1 and T_2 in Fig. 1) were found in the ferromagnetic regime. The additional maxima below T_c could be observed only

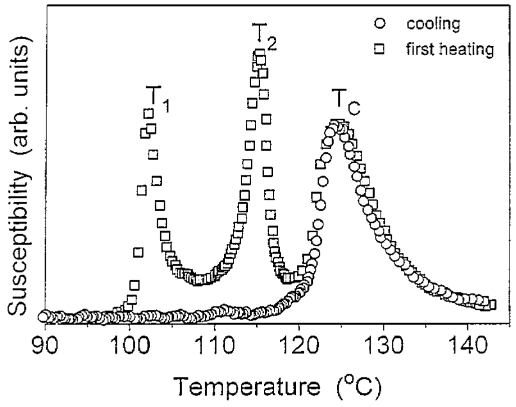

Fig. 1. Differential susceptibility versus temperature for Co/Cu(1 1 17) ($d = 2.3 \pm 0.1$ ML) measured parallel to the step edges. The squares show the data obtained during first heating. The circles represent the differential susceptibility taken during cooling.

in the first heat treatment (squares in Fig. 1). On cooling and in subsequent heating procedures these peaks did not appear any longer while the peak at the phase transition was always reproduced (circles in Fig. 1). The maxima in the ferromagnetic regime were interpreted as secondary maxima which appear when magnetic anisotropies become zero [29].

That suggestion was qualitatively proven by magnetization curves which showed anisotropy changes at the peak temperatures. Fig. 2 shows the magnetization curves in a 2.5 ML film obtained parallel (left-hand side) and perpendicular to the step edges within the film plane. At room temperature the as-grown films show a square hysteresis along the step edges with a small coercivity (Fig. 2a). Perpendicular to the step edges, in-plane, no hysteresis is found (Fig. 2b). Thus, the film exhibits the well known uniaxial behavior with remanence parallel to the step edges [17,24,30,31]. On heating the films above T_1 ($\approx$ 110°C) the magnetic behavior changes (Fig. 2c,d). Hysteresis loops are found in both directions, parallel and perpendicular to the step edges. Hence the magnetic anisotropy of the film has changed to a nearly biaxial behavior. In detail, one recognizes that the hysteresis perpendicular to the step edges (Fig. 2d) is already saturated in small magnetic fields, while the hysteresis observed along the step edges (Fig. 2c) exhibits a non-zero slope beyond the switching fields [32]. The slope of the curve indicates that the film is not saturated in that direction. That means that a small uniaxial anisotropy contribution is still acting which tends to favor the direction perpendicular to the step edges. Above T_1, K_u has to be negative and smaller than K_4. Hence, K_u changes sign which seems to be a necessary prerequisite for the maximum in the differential susceptibility at T_1.

Increasing temperature beyond T_2 gives a further change of magnetic anisotropy (see Fig. 2e,f). The film becomes uniaxial again with an easy axis along the step edges. No further changes of the easy axis could be observed neither on heating up to 160°C nor on cooling down from above T_2 to room temperature. Heating the films beyond 160°C has been avoided as interdiffusion has been identified to start in that temperature range in Co/Cu(001) [33].

Beyond that qualitative proof we investigated the temperature dependence of the anisotropy constants. Applying the procedure sketched in a recent paper [17], utilizing hard axis magnetization curves, we have determined the temperature dependency of K_u

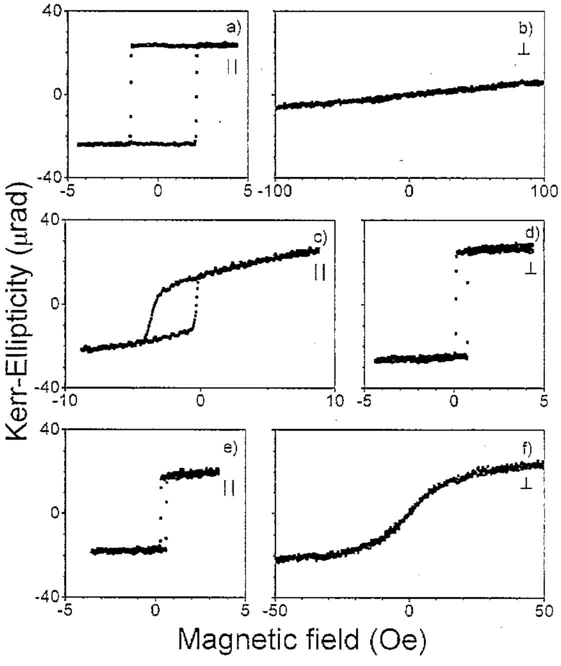

Fig. 2. In-plane hysteresis curves for Co/Cu(1 1 17) ($d \approx 2.5$ ML) measured parallel (left-hand side) and perpendicular (right-hand side) to the step edges. The magnetization curves have been taken at different temperatures. The temperatures are: (a, b) $T \approx 40$°C; (c, d) $T \approx 110$°C; (e, f) $T \approx 130$°C.

and K_4. The results are shown for 2.5 ML Co/Cu(1 1 17) in Fig. 3. The data have been obtained during the first heating procedure. The squares (circles) represent the uniaxial (biaxial) anisotropy constant as a function of temperature. The different temperature regimes found in the susceptibility can be seen in the plot of the anisotropy data as well. Below $T \approx 100°\text{C}$ and above $T \approx 128°\text{C}$ the uniaxial behavior is found which allows to determine the anisotropy constants. In between those temperatures the films exhibit the predominant biaxial behavior. Hysteresis loops are obtained in both directions which means that our procedure, to determine anisotropy constants, cannot be applied.

In the temperature range below $T \approx 70°\text{C}$ the twofold anisotropy is constant within the uncertainty of the measurement. Above $T \approx 70°\text{C}$ a strong decrease of the uniaxial anisotropy is observed. K_u drops by several orders of magnitude and becomes zero at around 100°C. At higher temperatures, above $T \approx 128°\text{C}$, the uniaxial anisotropy is again positive and increases with rising temperature. Compared to the room temperature value of the as-grown films the uniaxial anisotropy constant is reduced by about one order of magnitude.

The fourfold anisotropy is considerably smaller than the uniaxial contribution and positive at room temperature. At about 70°C the biaxial anisotropy constant begins to decrease like the uniaxial contribution and becomes zero around 90°C. Within the uncertainty of the measurements K_4 seems to stay zero up to the transition temperature T_1. In the high temperature range, above T_2, the value of K_4 is also zero within the error margins. In the range between $T \approx 100°\text{C}$ and $T \approx 128°\text{C}$ we may deduce qualitatively from the magnetization curves that the twofold anisotropy constant has changed sign, the fourfold contribution has to be non-zero and the absolute value of K_4 has to be larger than the twofold anisotropy constant. Hence in conclusion, the results verify that the uniaxial anisotropy changes sign at two temperatures. In that temperature range the biaxial contribution is found to be zero, within the experimental uncertainty. The experiments prove that the maxima in the differential susceptibility appear where both anisotropy constants vanish.

The insert of Fig. 3 shows K_u and K_4 versus temperature, measured while cooling. K_u increases slightly with falling temperature as one would expect. It does not cross zero anymore. K_4 shows only minor variations with decreasing temperature. The fourfold contribution is almost zero in the whole temperature range. As the total anisotropy never becomes zero again, the differential susceptibility

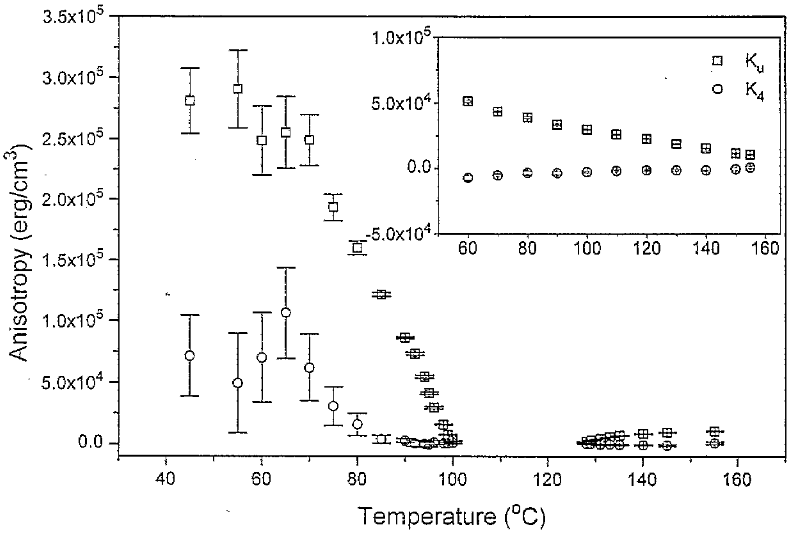

Fig. 3. Magnetic anisotropy constants versus temperature for Co/Cu(1 1 17) ($d \approx 2.5$ ML) during first heating. The insert shows the magnetic anisotropy while cooling. Squares represent the uniaxial anisotropy constant K_u. Circles show the biaxial anisotropy constant K_4. The error bars give the 1σ statistical error.

should not show any maxima in the ferromagnetic regime on cooling, which was actually found in the susceptibility experiments (see Fig. 1). If one compares the values of K_u before and after heat treatment (Fig. 3) one recognizes that K_u is strongly reduced, i.e. by more than a factor of five. The biaxial anisotropy is altered as well. Most remarkably is that K_4 has changed sign. After heating K_4 is negative, i.e. like in Co/Cu(001) films [34]. Hence film properties change drastically under heat treatment. Both magnetic anisotropy contributions exhibit an irreversible behavior with temperature.

4. Temperature effects in films above 3 ML

Next temperature effects in thicker Co/Cu(1 1 17) films are addressed. In the very thin films a strong deviation from 'bulk'-like behavior was published recently [17]. The onset of 'bulk'-like behavior was accompanied by a steep increase of $(K_u - K_4)$ around 3 ML. As the effects discussed above have been obtained with films below that transition thickness one is led to ask how the films behave on temperature changes in the thickness regime of 'bulk'-like behavior. For that purpose an ≈ 4 ML film has been investigated. The magnetization loops for different temperatures are shown in Fig. 4. Temperatures are very close to those of Fig. 2. It is evident that the 4 ML and the 2.5 ML film exhibit the same behavior. Fig. 4a–d exhibit qualitatively the same features as the thin film plots of Fig. 2, i.e. the uniaxial behavior changes to a nearly biaxial one in the same temperature range. At higher temperatures (Fig. 4e/f) the easy axis of magnetization switches back to the

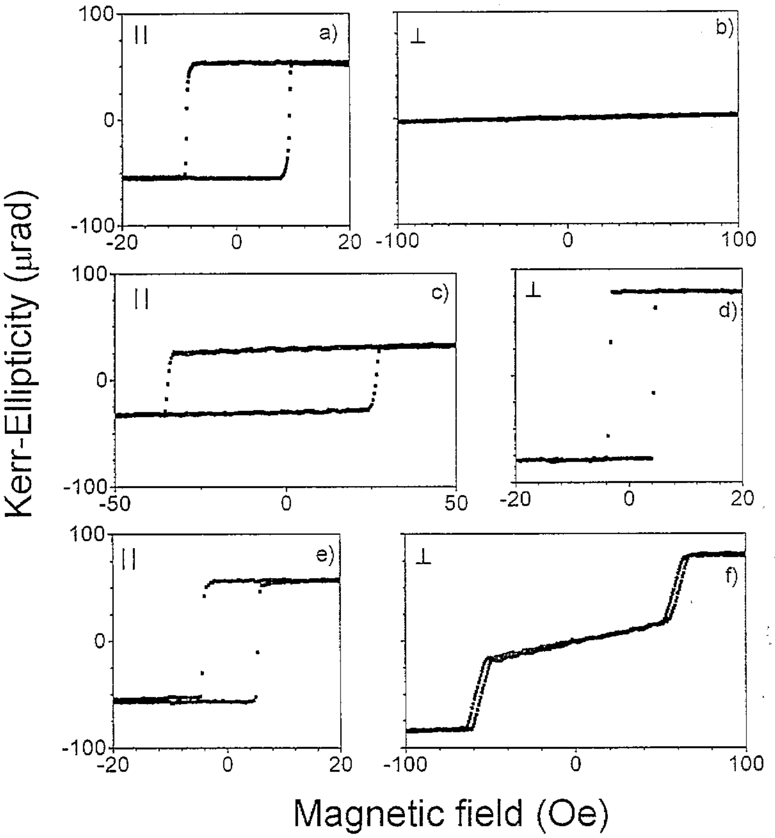

Fig. 4. In-plane hysteresis curves for Co/Cu(1 1 17) ($d \approx 4$ ML) measured parallel (left-hand side) and perpendicular (right-hand side) to the step edges. The magnetization curves have been taken at different temperatures. The temperatures are: (a, b) $T \approx 40$°C; (c, d) $T \approx 110$°C; (e, f) $T \approx 150$°C.

direction parallel to the step edges in complete agreement with the thin film behavior.

In contrast to the magnetization behavior the susceptibility exhibits a completely different temperature dependence. In films above 4 ML the susceptibility does not show any maximum in the ferromagnetic regime (up to 160°C). An almost structure-less temperature dependence is observed, which indicates differences of the magnetic properties compared to films below 3 ML. In the light of the above discussion one must conclude that the total magnetic anisotropy does not vanish. The hysteresis curves, however, demonstrate that the twofold contribution changes sign and thus crosses zero. Hence, the biaxial anisotropy has to be finite at the temperatures of vanishing uniaxial anisotropy, which means that in the 4 ML film a stronger fourfold anisotropy is established. The susceptibility indicates that in 2.3 ML the transitions happen via (almost) zero anisotropy states while in the 4 ML film the transitions are characterized by the persisting fourfold anisotropy contribution.

The increasing influence of the fourfold anisotropy contribution is obviously responsible for the shape of the hysteresis curve in Fig. 4f. The hard axis magnetization curve shows two loops at larger external fields. At the fields where the loops appear the magnetization switches into the hard direction. Around zero field a linear behavior is found indicating magnetization rotation in small fields. The loops at higher fields are caused by the biaxial part of the magnetic anisotropy which is no longer negligible small compared to the uniaxial anisotropy.

After heat treatment the magnetic anisotropy is weakened like in the thin films and the different anisotropy contributions can be separated again. Fig. 5 shows the temperature dependence of K_u and K_4 during cooling. It is obvious that the fourfold contribution has gained importance compared to films below 3 ML. While in thin films K_4 has been nearly zero in the whole temperature range, the biaxial contribution is even higher than K_u in the thicker films after heating. With decreasing temperature the biaxial value increases clearly while the uniaxial contribution rises only slightly.

5. Influence of temperature cycling on the anisotropy transitions

The last topic deals with the reversible or irreversible behavior of the anisotropy transitions. The susceptibility of thin films (thickness ≤ 2.5 ML) has been measured for different temperature cycles. Fig.

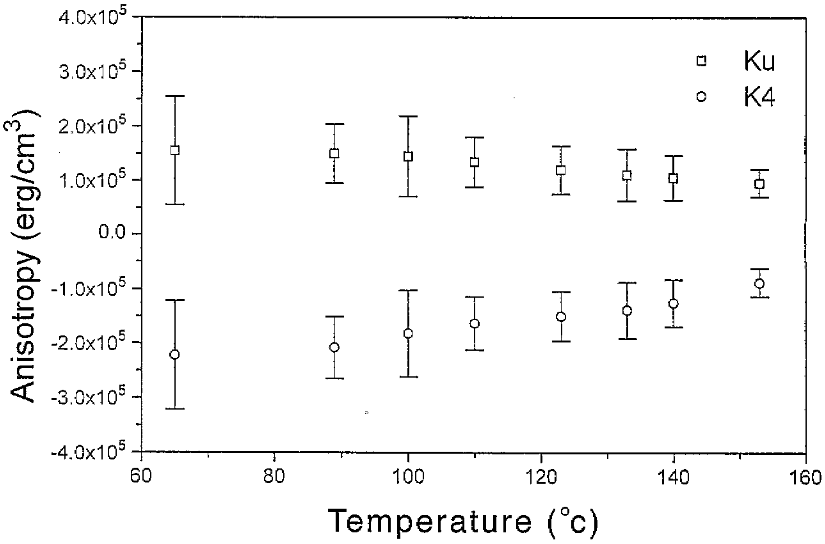

Fig. 5. Magnetic anisotropy constants versus temperature for Co/Cu(1 1 17) ($d \approx 4$ ML) during cooling. Squares indicate the uniaxial anisotropy constant K_u. Circles represent the biaxial anisotropy constant K_4.

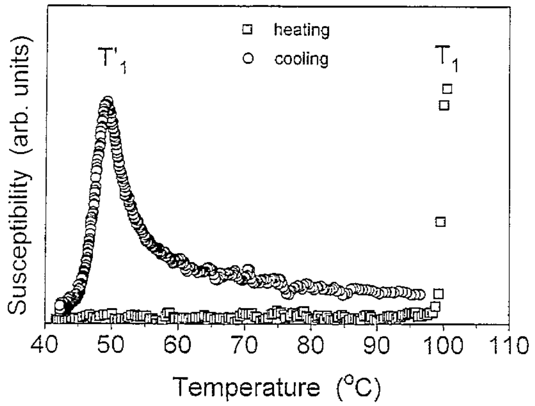

Fig. 6. Differential susceptibility versus temperature for Co/Cu(1 1 17) ($d = 2.3 \pm 0.1$ ML) measured parallel to the step edges. Squares represent the susceptibility of an as-grown film during first heating to T_1. Circles indicate the susceptibility during successive cooling.

6 shows the differential susceptibility for a film of 2.3 ML. The heating procedure is stopped when the first maximum in the susceptibility appears, i.e. at T_1. As temperature decreases a new maximum occurs at T_1', considerably below T_1. That new maximum is reproducible in following heating cycles (see Fig. 7) while the peak at T_1 is no longer found. The maximum at T_1' seems to shift slightly to higher temperature with each heating cycle (Fig. 7).

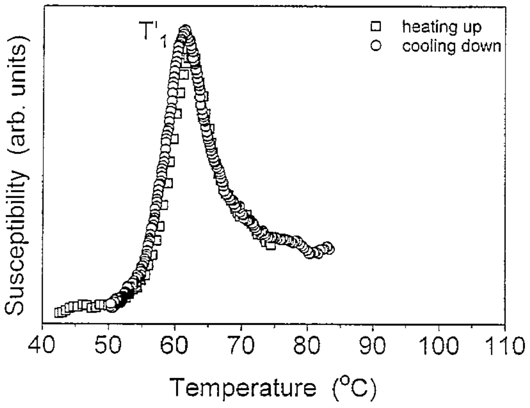

Fig. 7. Differential susceptibility versus temperature for Co/Cu(1 1 17) ($d = 2.3 \pm 0.1$ ML) measured parallel to the step edges. The temperature cycling has been performed immediately after the measurement shown in Fig. 6. Squares represent the susceptibility during heating. Circles show the susceptibility during cooling.

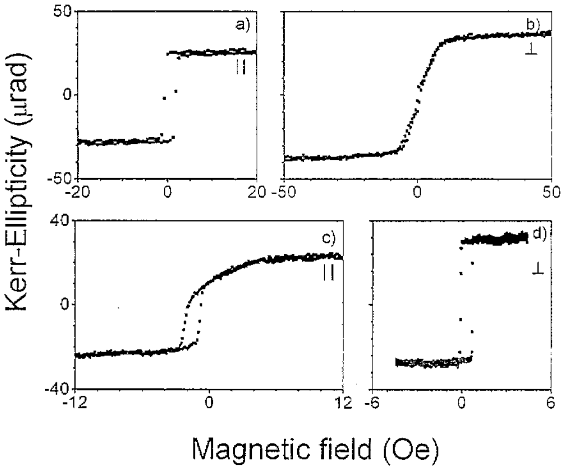

Fig. 8. Hysteresis loops for Co/Cu(1 1 17) ($d = 2.3 \pm 0.1$ ML) obtained below and above T_1' (see Figs. 6,7). The loops on the left-hand side have been measured parallel to the step edges. The magnetization curves on the right-hand side have been obtained perpendicular to the step edges. The loops have been measured at: (a, b) $T \approx 45$°C; (c, d) $T \approx 75$°C.

To find out what happens at the new susceptibility maximum, hysteresis loops have been taken at temperatures below and above T_1'. Fig. 8 shows the magnetization curves obtain parallel and perpendicular to the step edges. Comparing the plots of Fig. 8 with the magnetization curves obtained around T_1 during first heating (Fig. 2), it is obvious that a similar magnetic behavior produces both susceptibility maxima. In both cases a transition from uniaxial to nearly biaxial anisotropy happens. The two maxima represent the same anisotropy transition. During the first heating the transition is shifted to lower temperatures. The shifted transition exhibits a reversible behavior. The anisotropy can be changed back and forth by small temperature variations. Additionally to this obvious behavior the uniaxial anisotropy becomes extremely small. From the loops shown in Fig. 8 we obtain $K_u = 6400 \pm 100$ erg/cm^3 and $K_4 = 1600 \pm 300$ erg/cm^3.

Heating the film to considerably higher temperatures gives the maximum at T_2 besides that at T_1' (Fig. 9). The maximum at T_1 does not appear as the film is already switched to the nearly biaxial behavior at T_1'. The susceptibility peak at T_2, however, is neither affected by the previous heating cycles nor is it influenced by the position of the first anisotropy

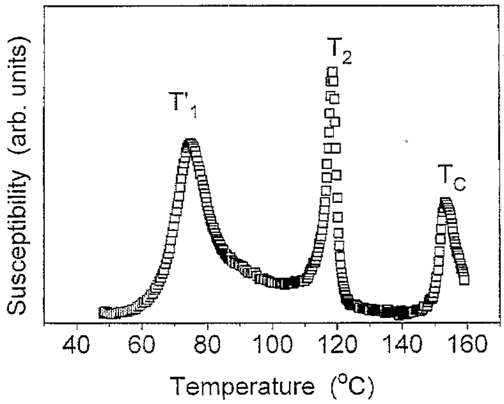

Fig. 9. Differential susceptibility versus temperature for Co/Cu(1 1 17) ($d = 2.3 \pm 0.1$ ML) measured parallel to the step edges. The film had been heated several times before the plotted data were taken. In the preceding heating procedures temperature had never exceeded T_1.

change. It is found at the same temperature. It turns out that at T_2 the films undergo an irreversible change into a stable configuration which does not show any further anisotropy alterations.

6. Discussion

As we find the same temperature induced changes of the anisotropy below and above 3 ML it follows that the origin of the transition is neither affected by the strength of the anisotropy nor depending on the change to 'bulk'-like behavior [17]. Moreover, as the changes of magnetic properties happen at about the same temperatures, it seems reasonable to assume that the transition is not driven by effects of magnetic origin. It is more likely to search for structural changes which should influence mainly the properties of the interfaces (including Co/vacuum and Co/Cu interface).

A reasonable process which can change the interface structure is the interdiffusion of cobalt and copper. It was found that in Co/Cu(001) interdiffusion appears at elevated temperatures [33]. Driven by the lower surface free energy of Cu, surface diffusion of Cu starts after holes are created in the Co film. The diffusion stops when the Co film is covered by Cu [33]. Although that process happens far above the temperatures where the anisotropy changes are found in Co/Cu(1 1 17), the interdiffusion of Cu and Co was studied by means of Auger electron spectroscopy. To obtain a high sensitivity for compositional changes the intensity of the low energy Auger lines was measured while temperature cycling and thin films (2 and 3 ML) were investigated. Diffusion processes should immediately appear in a 2 ML film due to a higher number of preexisting film imperfections. The 3 ML film guarantees an almost closed Co film [33] and the Auger peak ratios should be very sensitive to changes of film morphology, like hole creation. Fig. 10 shows the intensity ratios of the Cu(60 eV) to Co(53 eV) Auger transitions during heating. The temperature was raised remarkably slower than in typical susceptibility measurements. No indications for an increase of the Cu(60 eV)/Co(53 eV) ratio on heating to 160° could be found. For the 2 ML film a small decrease of the Cu/Co ratio is detected which might indicate a slight annealing effect. The on-set of hole creation and/or Cu diffusion in the 3 ML film can be seen in Fig. 10 after 2 h of keeping the film at elevated temperatures (varying between 100 and 180°C). As the anisotropy changes appear at lower temperatures and on considerably shorter time scales, the Auger experiment rules out that Cu diffusion is the driving mechanism for the anisotropy changes. Also the reversible character of the first transition at T_1' cannot be explained by interdiffusion.

The preponderant irreversible change of anisotropies due to heat treatment is the reduction of

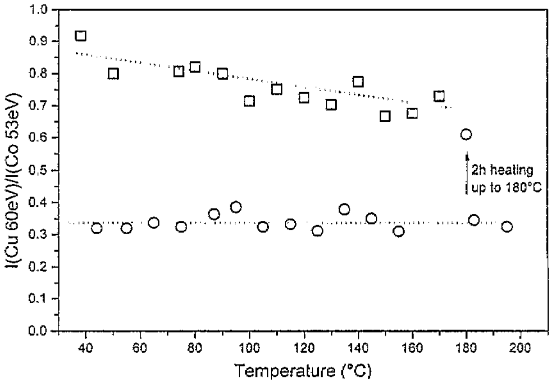

Fig. 10. Auger intensity ratio $I(\mathrm{Cu}_{60})/I(\mathrm{Co}_{53})$ versus temperature when heating a 2 ML (squares) and a 3 ML (circles) film. The value plotted at 180°C has been obtained after keeping the film for about 2 h at elevated temperatures (100°C $< T <$ 180°C).

the uniaxial anisotropy strength. Surprisingly, heating the films to T_1 gives a stronger reduction than heating to the higher transition temperature, T_2. The uniaxial anisotropy at room temperature is reduced by more than a factor of 30, after heating to T_1, whereas heating to T_2 gives a decrease by a factor of 4 (see foregoing paragraph). This result indicates that at T_2 the uniaxial anisotropy is reestablished.

The peculiarity of the transition temperature T_2 becomes also evident with the temperature behavior of the fourfold anisotropy in the thinnest films. Below T_2, K_4 is positive. This indicates a structural arrangement of terrace atoms which is different from that on Cu(001) surfaces. Heating the films to T_1 yields still a positive, however, strongly reduced biaxial anisotropy at room temperature. The biaxial anisotropy changes sign after the films had been heated to or above the second transition temperature (Fig. 3). The negative sign of K_4 means that the Co atoms on the terraces have rearranged to a structure similar to that for Co on Cu(001) surfaces. Hence, in the thinnest films a continuous, irreversible reduction of both anisotropy contributions are caused when heating to T_1. At T_1 the uniaxial anisotropy crosses zero while K_4 is negligible small, which is responsible for the maximum in the differential susceptibility. At T_2, however, the films undergo a irreversible, most likely structural, change which reestablishes the uniaxial anisotropy and reverses the sign of the biaxial anisotropy. Structural modifications with twofold as well as fourfold symmetry have to be incorporated in that transition at T_2.

In thicker films the temperature dependence of the uniaxial anisotropy is similar to that in the thin films. Important differences, however, manifest in the biaxial anisotropy. From the differential susceptibility and the temperature dependence of anisotropy it must be deduced that K_4 does not change sign on heating. After heating, the absolute value of the biaxial anisotropy is several orders of magnitude larger than in films below 3 ML. Thus, in the as-grown films (above 4 ML) the symmetry on the terraces is that of Co/Cu(001). The structure is most likely stabilized during film growth when the thickness is increased beyond 3 ML [17,36]. Consequently, we may conclude that the structural changes, which reestablish the uniaxial anisotropy, are not necessarily driven by the structural rearrangement of the atoms on terraces. Vice versa, it looks as if the twofold structural change initiates the modification on the terraces in the thinnest films. Hence, one might conjecture that the changes of the uniaxial behavior is mainly due to effects occurring at the steps. In the thinnest films the structural changes at the steps will affect the arrangement of atoms on the terraces. At the moment it cannot be clarified whether solely lattice relaxations at the steps or relaxations accompanied by temperature dependent roughness of steps at the surface and/or interface are responsible for the effects [19].

7. Conclusion

We have determined the magnetic anisotropy of Co/Cu(1 1 17) as a function of temperature for various film thicknesses. The uniaxial anisotropy shows extreme variations with temperature. At two temperatures the uniaxial anisotropy vanishes and an almost biaxial behavior is observed. In films below 3 ML thickness the zero crossing of K_u causes strong maxima in the differential susceptibility. In thicker films no susceptibility maxima could be found while a similar temperature dependent magnetization behavior is observed. The difference of susceptibility with thickness is attributed to strong changes of K_4. In the thinnest films the biaxial anisotropy is extremely small while with increasing thickness K_4 is established.

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