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Magnetization reversal in ultrathin Co/Cu(0 0 1)-microstructures

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Abstract

The magnetic domain structure of ultrathin Co-microstructures on Cu(0 0 1) has been investigated by means of a scanning electron microscope with polarization analysis. In particular, the influence of a magnetic field on the domain structure has been studied. Here, we concentrate on squares of different size (3–30 μm) and variable thickness, although triangles and bars with aspect ratios up to 1:30 have been studied, too. In the as-grown state the microstructures with an edge length larger than 3 μm exhibit a multidomain structure, while the smaller ones are always single domain. After saturating the structures by a strong field, the magnetization can be switched homogeneously by either $\pm 90^\circ$ or 180° independent of size and shape. The microstructures remain in a single-domain state. Roughly 20% of the ultrathin particles show a two-step 180°-switching via a 90°-state. An analytical model is developed yielding coercivities of the right order of magnitude. The two-step reversal process can be explained in the framework of this model.

Keywords: Magnetic thin films; Microstructures; Magnetization reversal; Domain structure

1. Introduction

The understanding of magnetization reversal processes in ultrathin films is of basic interest both for fundamental research and for application as magnetic memory devices. One possibility to get information on the basic processes is to reduce the lateral dimensions of the films to values that are within experimental field of view, since in this case the magnetic state of the entire system can be fully determined. A vectorial analysis of the magnetization direction with high lateral resolution can be achieved by scanning electron microscopy with polarization analysis (SEMPA). From the domain image all contributions to the total energy of the system can be extracted. For highly symmetric shapes, e.g. parallelipipeds, even the magnetostatic self-energy can be calculated exactly [1] besides domain wall and Zeeman energies. Minimization of the total energy provides theoretical insight into both the energetically favourable states and possible transitions between them.

2. Experimental aspects

Experiments including sample preparation as well as domain structure investigation were carried

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out under ultrahigh vacuum conditions. The Cu(0 0 1) single crystal was cleaned by cycles of Ar^+ sputtering (600 eV) and annealing ($T \approx 650^\circ$C). Cobalt was deposited by e^--beam evaporation at room temperature (RT) with a growth rate of ≈ 1 monolayer (ML) per minute. Microstructures with thickness up to 10 ML were prepared. During evaporation, the pressure did not exceed 5×10^{-10} mbar. The microstructures were fabricated by evaporation through a mask. The mask is a 240 μm thick Si wafer that was structured by etching and lithography techniques [2]. It contains about 40 arrays of regularly arranged microstructures of different size and shape (squares, stripes, and triangles). The distance between neighbouring squares is of the same order of magnitude as their edge length, that is 3, 6, 9, 12, 15, and 30 μm. The edges of the Co-squares were oriented along the $\langle 1\,1\,0 \rangle$-directions of the Cu substrate which are the magnetically easy axes of FCC-Co films. The mask was put as close as possible to the Cu crystal. Its position was controlled with a telescope. The distance between mask and surface of the Cu single crystal was typically in the range of 100 μm. Due to this separation of mask and template shadowing effects occur yielding a continuous thickness variation at the edges. By means of scanning electron microscopy (SEM) images we have estimated the width of the thickness decrease at the edges.[2] We obtain ≈ 400 nm in good agreement with calculations based on the geometry of the preparation set-up (sharp edges for a parallel evaporation beam and ≈ 600 nm width for maximum divergence). These numbers point out how crucial the distance between mask and template is for the quality of the structures one can make by this technique. A contact was avoided to prevent any damage of the surface.

The magnetic domain pattern in the Co/Cu(0 0 1)-microstructures was determined in situ by SEM with spin polarization analysis of the secondary electrons (SEMPA) [3]. This technique does not work while a strong external field is applied. To overcome this obstacle we applied a field for 10 s and reduced it to zero. Then the magnetic microstructure was investigated, i.e. in the state of remanence. The field was increased stepwise ($\Delta H \approx 4$ Oe) from one to the next cycle and the influence of the field on the magnetic domain pattern was monitored. The external field was oriented either 90° or 180° to the magnetization of the microstructures.

3. Results

After growth, domains were found in all microstructures with an edge length larger than 3 μm independent of film thickness. The domains could be erased by applying a magnetic field in the range of 100 Oe along one of the easy axes transfering every structure into a single-domain state. More details about the features of the as-grown state and saturating the microstructures will be given in a forthcoming paper [4].

Starting from a single-domain configuration the reversal/switching behaviour was investigated. A sequence of images is shown in Fig. 1. The images show the same array after different fields have been applied. In small fields the microstructure remains in the state with magnetization pointing downward along the vertical direction, visualized by the black contrast with respect to the surrounding Cu-substrate (Fig. 1a). It turns out that for every structure in the array the magnetization is stable up to a certain field. For larger fields the structures start to switch as a whole. We never found that single-domain structures decompose into a multidomain pattern for fields applied along the easy axes. Applying a field of 8 Oe causes five squares to reverse (white contrast in Fig. 1b). Four additional microstructures switched after the field was increased to 12 Oe. Finally, all microstructures exhibit reversed magnetization direction after a field of 24 Oe was applied.

In the following we will present results of such SEMPA investigations in the form of the residual magnetization along the initial magnetization direction versus applied field, i.e. as magnetization

[2] The secondary electron contrast in Co/Cu(0 0 1) reveals a pronounced thickness dependence. While below 2 ML thickness Cobalt appears light with respect to the Cu-substrate it becomes dark for $D > 2$ ML. Measuring the width of the region with light contrast for 2 ML thick structures allows to determine the edge profile roughly.

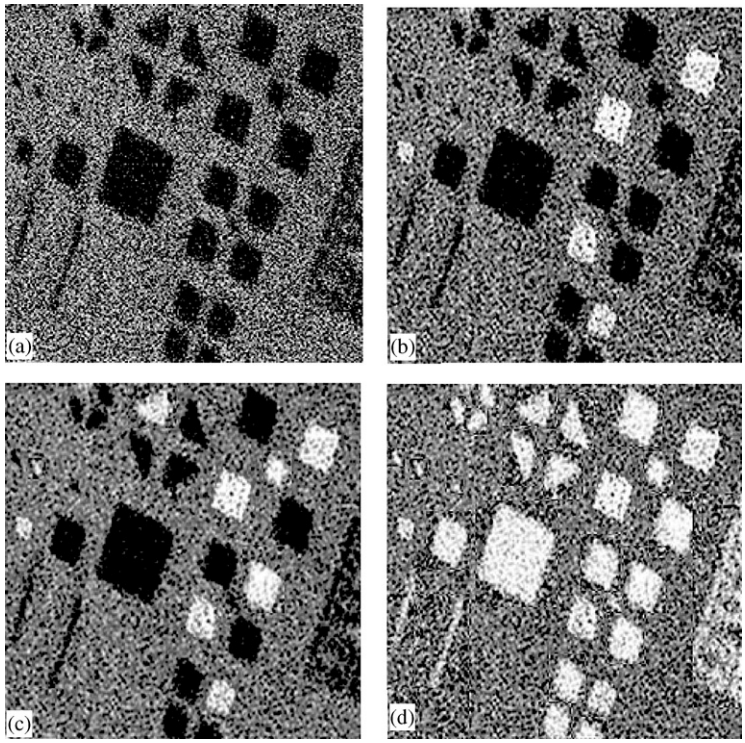

Fig. 1. Images showing the switching behaviour in a Co/Cu(0 0 1)-microstructure array. The thickness of the array is 4 ML. For every microstructure the initial state is stable up to a certain field and switches completely if this field is exceeded. The field strengths applied before investigation are 0, 8, 12, 24 Oe for the images (a)–(d), respectively.

curves. Fig. 2 displays the switching behaviour in reversed fields of three equivalent squares. The size of the squares is 15 μm and the thickness 10 ML. Only one half of each loop has been measured. The well-known hysteresis loop is generated by inversion.

Although the squares are nominally identical and were produced in a single evaporation step, the switching fields scatter by more than a factor of two from 6 to 14 Oe. We have carefully investigated all the arrays for spatial correlations of the switching behaviour as well as for crosstalk between adjacent structures. No dependence on position nor an effect of neighbouring structures could be identified.

The magnetization loops for structures with other size and thickness appear to be similar to those of Fig. 2. To find out if there is a dependence of the switching on size or thickness, we have

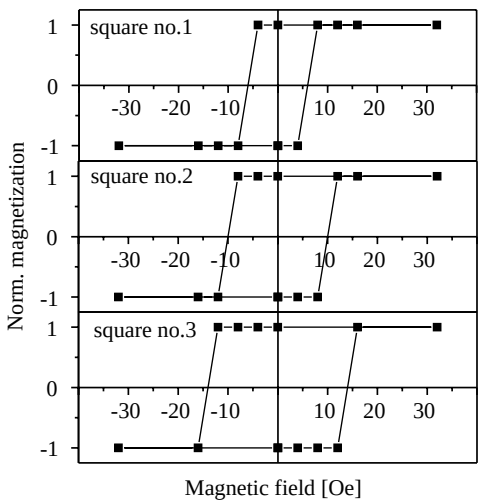

Fig. 2. Switching curves of different squares with edge length of 15 μm and thickness of 10 ML. The squares switch within a field window of 4 Oe. The lines are a guide to the eye.

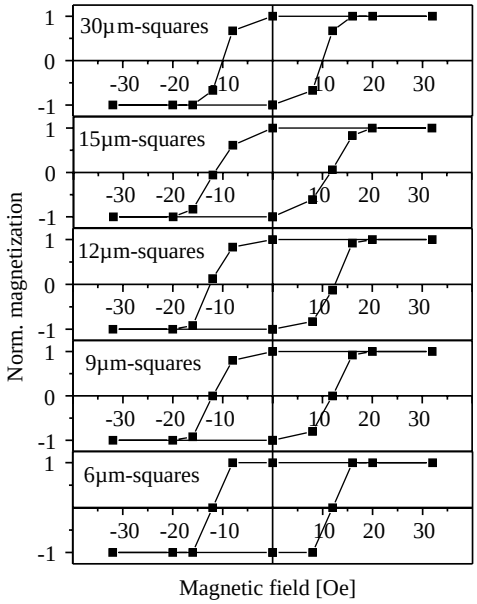

Fig. 3. Averaged switching loops for 4 ML thick squares with different edge length. The lines are a guide to the eye. Its crossings with the abscissa show that within the experimental accuracy there is no dependence of the average switching field on edge length.

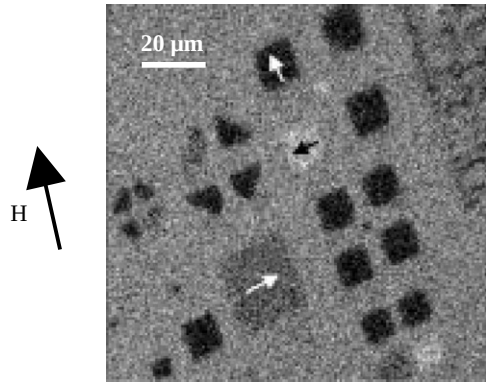

Fig. 4. Existence of a 90°-transition state during 180° reversal of an array of Co/Cu(0 0 1)-microstructures. While most of the structures are magnetized parallel to the applied field, in four squares the main magnetization component points either to the left (light grey) or to the right (dark grey).

created the averaged hysteresis loops of 12 identical squares. Fig. 3 shows for a thickness of 4 ML from bottom to top the loops for squares with an edge length of 6, 9, 12, 15, and 30 µm, respectively.

As can be seen from Fig. 3 there is no dependence of the averaged switching field on the edge length within the experimental accuracy. The Co/Cu(0 0 1)-microstructures switch at fields of approximately 12 Oe, independently of their size. The same behaviour is found for all the other thicknesses studied.

About 80% of the squares are found to switch directly by 180°. In the remaining 20% switching happens via a two step process, i.e. by two 90°-steps. Fig. 4 shows a 10 ML thick microstructure array after a field of 16 Oe had been applied. For this measurement the polarization sensitive axis of the spin detector was tilted slightly away from the easy axes of the Cobalt to make all four orientations of the magnetization visible.

In almost all structures the magnetization has been switched into the field direction (black contrast in Fig. 4). In four squares and one triangle, however, the magnetization points either to the left (light grey) or to the right (dark grey), i.e. the structure is flipped by 90°. The existence of both components perpendicular to the applied field proves that the 90°-state cannot be due to a misalignment of the applied field. The structures that show this behaviour are distributed randomly over the sample surface and no correlation with their size exists.

Obviously, during 180°-reversal 90°- and 180°-switching processes occur within the same range of field values. To verify this, we have applied fields that were oriented perpendicularly to the initial state. We find that within experimental uncertainty the direct 90°-switching occurs at the same field values as the 180°-reversal.

A small number of microstructures with other shapes have been investigated, too. Neither for equal-sided rectangular triangles with the same lateral dimensions as the squares nor for bars with aspect ratios up to 1:30 a change of the switching fields within our experimental accuracy was found. Thus, the switching is not depending on the shape of the structure.

4. Discussion

The most striking finding is that in remanence the microstructures stay in a single-domain configuration as soon as they have been saturated for the first time. Obviously, the single-domain state which corresponds surely to the absolute energy minimum in field still reflects a local minimum as the field is reduced to zero. In remanence, however, this state is energetically degenerate because of the fourfold anisotropy in Co/Cu(0 0 1). A transition between these states is possible if a field is applied. This transition can proceed either by rotation of magnetization which is homogenous within the whole microstructure (*coherent rotation*) or by a state with inhomogenous magnetization distribution. In the latter situation a domain is nucleated and switching proceeds via domain expansion. Another incoherent reversal mode, the classical *curling mode*, is not considered here, since in an ultrathin particle with in-plane magnetization curling around the field direction is not possible.

From literature it is well known that in small particles magnetization reversal via coherent rotation can be described by the Stoner–Wohlfarth fomalism. Applying that description of magnetization switching to the system Co/Cu(0 0 1) with fourfold anisotropy [5] we can calculate the switching field by means of the anisotropy values published in literature [6,7].[3] We obtain a field value of 2 kOe, which is several orders of magnitude larger than the experimentally observed values in the SEMPA investigation. Hence, in Co/Cu(0 0 1)-microstructures magnetization reversal by coherent rotation must be excluded.

Alternatively, the switching can proceed via nucleation of a domain with different magnetization direction and consecutive propagation of its domain wall. In this situation the switching field can be determined either by the nucleation process or by the pinning of domain walls while moving. The process with the highest activation energy is responsible for the coercive field. From the fact that we do not find any domains we can deduce that the domain walls can easily propagate and that domain wall pinning is of minor importance. The nucleation of domains with different magnetization orientation seems to cost the main energy. Hence, if we want to understand the switching in the microstructures we have to consider the details of the nucleation process. Due to the fact that the total energy of the microstructures can be calculated, a good approximation of all energies involved in the switching can be achieved. As the structures have highly symmetrical shapes the magnetostatic energy can also be calculated exactly [1]. The energy balance of the microstructure with nucleus is determined on the one hand by the gain of Zeeman and magnetostatic energy due to a domain with switched magnetization orientation and on the other hand by the cost of mainly exchange and anisotropy energy within the newly created domain wall. In ultrathin films the domain walls have Néel structure which creates a small amount of magnetostatic energy in the wall as well [8]. The interaction of the corresponding charge with those at the particle boundaries is neglected.

Counting all these contributions we can write down the total energy of such a microstructure in an external field H as a function of a geometrical factor a that describes the size of the nucleus with switched magnetization orientation

$$E_{\mathrm{tot}}(a,H) = -\alpha_1 f(a) H M_{\mathrm{s}} + \gamma g(a) + \sum_{i=0}^{n} \delta_i a^i + E_0 \quad (1)$$

The first expression is the gain in Zeeman energy due to the nucleus and depends on its geometry. The geometry factor $f(a)$ is given in Appendix A and α_1 is 1 or 2 depending on wether the nucleus is magnetized 90° or 180° to the initial magnetization of the structure. The second term is the total energy due to the additional domain wall with γ the domain wall energy per domain wall area (Appendix B) and $g(a)$ the domain wall area (Appendix A). The third term is a polynomial approximation of the expression for the magnetostatic energy obtained from the Rhodes and

[3] In Ref. [7] the same in-plane anisotropy for Cu covered Co/Cu(0 0 1) was found as in Ref. [6]. With the proof (see Ref. [6]) that the bulk fourfold anisotropy is independent of coverage we have taken this value as the best guess for our films. The surface contribution was taken from Ref. [6] although it is of minor importance in the thickness range under consideration.

Rowlands (R&R) model [1]. A second-order polynomial has been taken to enable simple analytical treatment (see below). In this form the dependence of the magnetostatic energy on a can be described within an error of 5% compared to the exact R&R expression. The pre-factors δ_i are fitting parameters and have different values for different microstructure dimensions.[4] Finally, E_0 includes all terms that are independent of a.

One consequence of the expression in Eq. (1) is that the situation of 90°-nucleation in a reversed field and field conforming nucleation in a field oriented perpendicular to the initial state are energetically equivalent. In both cases the absolute values of magnetostatic energy and domain wall energy are the same, while the Zeeman energy differs only by a constant HM_sL^2D (with L, D the edge length and thickness, respectively), which has no importance for the nucleation. Hence, the critical field for both processes can a priori be expected to be equal. This already confirms the experimental observation that on average 90°-switching and 180°-switching occur at the same field value.

Taking Eq. (1), the critical domain size a^* for a given field H can be obtained by determining the local maximum in the total energy with respect to the variable a, i.e. the energy barrier that prevents full expansion of the nucleus

$$\frac{\partial E_{\mathrm{tot}}(a,H)}{\partial a} = 0 \quad \Rightarrow \quad a = a^*(H) \tag{2}$$

and

$$\left(\frac{\partial^2 E_{\mathrm{tot}}(a,H)}{\partial a^2}\right)_{a=a^*(H)} \leqslant 0. \tag{3}$$

Hence, for $a > a^*$ the total energy decreases when a increases, i.e. a larger domain size is energetically favourable and the domain can expand. An increase in field strength of H reduces both the maximum of the total energy and the critical nucleus diameter. Due to the reduced lateral dimensions of the microstructure an upper limit for the critical diameter of the nucleus and thus a lower limit for the field exists above which domain formation becomes energetically favourable. At the critical field H_c the critical nucleus diameter has at least to be smaller than the edge length L of the microstructure, i.e.

$$a^*(H_c) < L. \tag{4}$$

For the calculations we have assumed that the domain wall is infinitely thin and that the domain is nucleated at the edges of the structure where magnetic poles exist (see Fig. 5(1)). A nucleation in the center of the structures has not been considered. Such a process is less favourable since the gain in magnetostatic energy is lower while the cost in domain wall energy is higher than in case of domain nucleation at the edges.

Due to the fourfold anisotropy of Co/Cu(0 0 1) we consider the magnetization in the nucleus not solely to be parallel to the applied field, but also the case of 90° orientation of magnetization. There are two reasons for taking the latter scenario into account. First, an alignment parallel to the microstructure boundary avoids magnetic poles and gains the largest amount of magnetostatic energy. Secondly, the cost in domain wall energy is smaller. On the other hand, the gain in Zeeman energy is reduced by a factor of two. The following geometries have been calculated:

1. Nucleation of a half-cylindrically shaped domain at a charged edge with its magnetization oriented either parallel or perpendicular to the applied field.

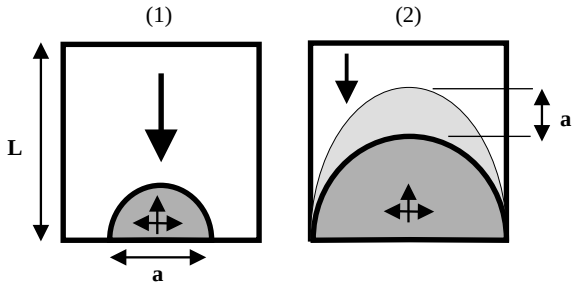

Fig. 5. Sketch of the processes that have been considered: Nucleation of a half-cylindrically shaped domain at a charged edge (1) and expansion of this domain through the center of the structure (2).

[4] For each set of variables L and D the magnetostatic energy has been calculated exactly with the R&R-formalism and was then approximated by a second-order polynomial.

Table 1
Critical field values in Oersted for the processes domain nucleation and expansion shown in Fig. 5 in case of a Co/Cu(0 0 1)-microstructure with atomically sharp edges (upper half) and a stairlike thickness decrease within 400 nm as observed experimentally (lower half). A thickness of 4 and 10 ML, an edge length of 3 and 15 μm, and an angle between field and magnetization in the nucleus of 90° and 180° have been considered. In addition, in the most right column the average switching field obtained from experiment is shown, i.e. ≈12 Oe independently of edge length, thickness

	Edge length	Thickness	Process				Exp.
			Nucleat.		Expans.		
			90°[a]	180°[a]	90°[a]	180°[a]	
Sharp edges		4 ML	3.4	9.1	3.1	3.6	
	3 μm	10 ML	1.8	14.2	3.9	4.7	
		4 ML	0.5	1.8	0.6	0.7	
	15 μm	10 ML	0	2.8	0.8	0.9	
		4 ML	5.5	7.3	3.1	3.6	12
Unsharp edges	3 μm	10 ML	7.0	11.2	3.9	4.7	12
		4 ML	1.0	1.4	0.6	0.7	12
	15 μm	10 ML	1.1	2.2	0.8	0.9	12

[a] (M,H)-angle.

2. Expansion of the fully extended half-circularly shaped domain wall through the centre of the structure.

To estimate the influence of a continuous thickness decrease at the edges we have considered both the ideal case, i.e. atomically sharp edges and a decrease in form of a steplike profile with equispaced monolayer steps. Since domain wall energy and Zeeman energy are only slightly affected by this correction, it has only been applied to the magnetostatic energy. The resulting critical fields (in Oe) for the two processes (see Fig. 5) are given in Table 1 in dependence on thickness and edge length of the Co/Cu(0 0 1)-microstructure.

The calculated values for the critical field for both processes are of the same order of magnitude as the average experimental value (see Table 1). A closer inspection of the calculated fields, however, shows that the critical field depends on both edge length and thickness. In case of 15 μm edge length almost all critical fields are rather small and for one parameter set (sharp edges - 15 μm—10 ML—90°) even a critical field of 0 Oe is calculated. This would mean spontaneous nucleation, i.e. domain formation even at remanence, which is in absolute disagreement with experiment, where a single-domain state is found. The calculations, however, demonstrate that in an ultrathin particle with absolutely sharp edges the magnetostatic energy might lead to domain formation and, hence, seems to be more important than assumed in recent reports [9,10].

If at the edges a rather gradual decrease of the thickness is assumed, the critical fields for process one will change remarkably. A direct comparison of the values for sharp and unsharp edges reveals, that the change of nucleation field depends on the relative orientation of the field to magnetization in the nucleus. While for the 90° configuration the critical fields are shifted to larger values, they become smaller for an antiparallel alignment. This somewhat surprising result is due to the different dependence of the magnetostatic energy on nucleus diameter of 90° and 180° nucleus magnetization:

For a 90°-nucleus the magnetostatic contribution to the total energy decreases monotonically with increasing diameter of the nucleus, since more and more magnetic poles are avoided. If unsharp edges are assumed both the absolute value of the magnetostatic energy and its gain by domain formation will be reduced. Hence, to create a nucleus in the particle the Zeeman energy has to

become larger, i.e. the field strength has to be increased. In case of a reversed magnetization of the nucleus charges of opposite sign are generated at the edge where the nucleus is created. This reduces stray field energy. However, above a certain diameter of the nucleus the stray field energy increases again because the ratio of charges with opposite sign becomes unfavourable. Finally, the edge is completely homogeneously reversed and the total energy is higher than in the starting configuration as the charges at opposite edges have the same sign. This causes an energy barrier for the nucleus to expand along the edge yielding the critical field for nucleation. Unsharp edges reduce the height of the barrier and allow the expansion of the nucleus at smaller fields.

In the following we will restrict ourselves to the case of unsharp edges since we believe that this model comes close to the experimental situation. A direct comparison of the critical fields for processes 1 and 2 shows that the value for domain nucleation is always larger than the one for its expansion. Accordingly, a domain will expand rather spontaneously once it has been nucleated. Hence, domain nucleation is the limiting process and in remanence always a single-domain state should occur as is found in the experiment.

Furthermore, the model predicts that for both nucleation and expansion the critical field is smaller for a domain being magnetized perpendicular to the direction of the field. Hence, during 180°-reversal nucleation of 90°-domains can be expected to occur preferentially. As has been discussed above, nucleation of these domains will occur only at a charged edge. After nucleation and consecutive expansion of a 90°-domain the initially charged edges become uncharged while former uncharged edges become charged. The nucleation of a second 90°-domain will take place at one of the latter edges. Hence, two edges of the square will be involved in the switching process. If for some reason, e.g. inhomogeneities within the unsharp edges, the critical field for the two nucleation processes are different a state with magnetization perpendicular to the applied field will be observed in remanence when a field has been applied which fits between the two fields. The fact that only 20% of the microstructures show this state, although the probability should be near 50%, might be due to the rather big field steps of 4 Oe used our in experiment.

The weak dependence of the critical field on thickness is due to an increase of the anisotropy and, hence, the domain wall energy with thickness [6,7]. This variation of critical field is of the same order of magnitude as the field steps and is most likely not resolved.

The dependence of the critical field on the edge length is much more pronounced in the calculations. The main reason for that behaviour is the upper limit of the critical domain diameter that was set equal to the edge length of the microstructure (see Eq. (4)). If an edge length independent value is assumed, the critical fields become independent of the edge length. Since a direct observation of the nucleation process was not possible no experimental evidence for such a limitation is at hand.

The reason for the rather large scattering of the switching fields of the microstructures is not yet clear. Apparently, each microstructure has its own and well-defined coercive field as has already been reported in case of ultrathin Au/Co/Au(1 1 1)-elements [11]. Our analytical model predicts and the experiments confirm that domain nucleation is the limiting process, but the nucleation process itself is hindered or accelerated rather randomly.

5. Conclusion

We have presented investigations of domain structures in ultrathin Co/Cu(0 0 1)-microstructures after magnetic fields have been applied. Experiments reveal that on average reversal occurs at a field of approximately 12 Oe independent of size, thickness, and shape. An analytical model for domain nucleation in ultrathin structures with in-plane magnetization is put forward. It is theoretically found that in a structured system with fourfold anisotropy a nucleus with magnetization at 90° to the applied field is energetically more favourable than the nucleus with reversed magnetization. The model can explain also the existence of a 90°-state and the equal switching fields for 90°- and 180°-reversal.

Appendix A. Geometrical factors

In Eq. (1) the functions $f(a)$ and $g(a)$ describe the increase in volume of the domain with different direction of magnetization and the change in area of the domain wall, respectively. For processes 1 and 2 they are

$$f_1(a) = \frac{\pi}{8} D\ a^2, \tag{A.1}$$

$$f_2(a) = \frac{\pi}{2} D\ a\, l \tag{A.2}$$

and

$$g_1(a) = \frac{\pi}{2} D\ a, \tag{A.3}$$

$$g_2(a) = \frac{\pi}{4} D \left(l + 3\,a - l\sqrt{1 + \frac{2\,a}{l}} \right), \tag{A.4}$$

where D and l are the thickness of the microstructure and the length of the domain wall, respectively.

Appendix B. Domain wall energy

According to Middelhoek [8] the energy density of a Néel-like domain wall γ_N can be expressed as

$$\gamma_N = \alpha_1 \sqrt{A\,K} + \alpha_2\, \pi\, D\, M_s^2. \tag{B.1}$$

The first term is the well-known exchange and anisotropy contribution with A and K the exchange and anisotropy constant, respectively. The second term is the magnetostatic energy. The parameter α_2 depends on the angle of rotation in the wall and is 0.09 and 1.00 for a 90°-wall and a 180°-wall, respectively.

It should be noted, that the domain wall involved in the 180° nucleation process is no ordinary 180°-wall, since the magnetization in the adjacent domains points head to head. As a consequence the divergence of the magnetization and the energy of the wall will be larger compared to the classical 180°-wall. Hence, the expression in Eq. (B.1) is a lower limit for energy of this type of wall.

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