Magnetic anisotropy of Co on Cu(1 1 17)

W. Wulfhekel,[a] S. Knappmann, and H. P. Oepen
Institut für Grenzflächenforschung und Vakuumphysik, Forschungszentrum Jülich, D-52425 Jülich, Germany

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The in-plane magnetic anisotropy of ultra-thin Co films, epitaxially grown on Cu(1 1 17), was determined *in situ* by means of the magneto-optic Kerr effect down to thicknesses as low as 2 monolayers. Uniaxial and biaxial anisotropy contributions were observed. At room temperature, the uniaxial component is dominant and the easy axis of magnetization is parallel to the step edges. Above 4 monolayers the magnetic anisotropy exhibits a thickness dependence which can be described by volume and interface contributions. For thinner films a pronounced deviation from that behavior is found. The anisotropy drops abruptly by one order of magnitude below 3 monolayers. Thickness dependent relaxations are proposed as driving forces for that behavior. © *1996 American Institute of Physics.* [S0021-8979(96)06902-X]

INTRODUCTION

Magnetic anisotropies play an important role in ultra-thin ferromagnetic films. Anisotropy is necessary in two dimensional ferromagnets to obtain long-range order. This has been rigorously proven by Mermin and Wagner.[1] Many properties of ferromagnets are directly or indirectly determined by the anisotropy, e.g., the magnetization orientation of domains, domain structure, coercivity, and the magnetization reversal process. Hence, the understanding of magnetic anisotropy and its correlation with film properties, such as structure and morphology, are very important for the interpretation of the magnetic behavior of ultra-thin films. Various mechanisms are known to influence film anisotropy. Besides magneto-crystalline and magneto-elastic anisotropy contributions, shape anisotropies play a crucial role. In thin films shape effects generally become preponderant as the dimensions differ strongly perpendicular and parallel to the film. Apart from this semi-classical argumentation, the continuum approach can fail for films of monolayer (ML) thickness. A discrete description of dipolar interaction has shown that the shape anisotropy depends strongly on the arrangement of atoms and film roughness in case of vanishing thickness.[2,3] Magneto-elastic and magneto-crystalline anisotropy contributions may exhibit pronounced thickness dependence as well. In the limit of vanishing thickness these anisotropies are solely determined by contributions from the surface and/or interface. Hence, the thickness dependence of film anisotropy is in general given by the phenomenological ansatz, distinguishing between bulk and surface/interface contributions:

$$K = K_v + \frac{(K_s + K_i)}{d} \tag{1}$$

with K_v as volume, K_s as surface, K_i as interface anisotropy contributions and d the film thickness. Discontinuities are not included in that ansatz. The surface contribution has been introduced by Néel when considering the reduced symmetry at the surface of a ferromagnet.[4]

Recently it was pointed out by Albrecht *et al.*[5,6] that micro-structures at the surface, like steps, should give rise to additional magnetic surface anisotropies. The symmetry breaking at steps should cause, ensuing from Néel's idea, local contributions to magnetic anisotropy with twofold symmetry. In case of preferentially aligned steps and a high step density, step anisotropies can influence and even determine the macroscopic properties of the film. This has been experimentally demonstrated for ferromagnetic films grown on vicinal Cu and W surfaces.[7,8]

For Co/Cu(1 1 13) the micro-magnetic structure as well as the magnetization behavior has been studied by means of Scanning Electron Microscopy with Polarization Analysis and Magneto-Optic Kerr Effect (MOKE).[7,9] The anisotropy constants for Co/Cu(1 1 13) at room temperature have been determined for film thicknesses above 4 ML with Brillouin Light Scattering (BLS).[10]

The purpose of our study was to investigate in more detail the thickness dependence of anisotropy. Particularly, the investigations were focused on films in the thickness range of a few monolayers where the transition from surface-to bulk-determined anisotropy should appear. This paper deals with the magnetic anisotropy of Co films grown epitaxially on vicinal Cu(1 1 17) surfaces. The magnetic anisotropies were determined *in situ* by means of the magneto-optic Kerr effect.

FILM PREPARATION AND CHARACTERIZATION

The experiments, including film preparation and investigation of the magnetic properties, were performed under Ultra High Vacuum (UHV) conditions (base pressure $p \approx 1 \times 10^{-10}$ Torr) in the same vacuum chamber. The UHV chamber was equipped with a Medium Energy Electron Diffraction (MEED) experiment and an Auger electron spectroscope for growth and film characterization.

Vicinal Cu(11n) surfaces were used as templates. These surfaces have been well studied by means of helium scattering[11] and Scanning Tunneling Microscopy (STM).[12,13] Microscopically, the surfaces consist of (001) orientated terraces, separated by monoatomic steps. The average terrace

[a]Electronic mail: w.wulfhekel@kfa-juelich.de

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width is n/2 atomic distances. Step bunching has not been observed.[12] The steps are aligned along the $[1\bar{1}0]$ in-plane direction.

The substrate was cleaned and prepared by cycles of Ar sputtering (600 eV) and subsequent annealing ($T>670$ °C). After ion bombardment, an ordered microstructure was obtained by careful annealing.[12] The quality of the surface crystal structure was checked with MEED. The MEED diffraction pattern shows pronounced splitting of the regular lattice spots indicating a periodic step arrangement on the copper surface.[14]

The films were grown at 45 °C with a rate of 1 ML/min. During electron beam evaporation the pressure did not exceed 5×10^{-10} Torr. The growth process was monitored by measuring MEED intensities. The MEED reflectivity remained high during all stages of growth, which means that the MEED pattern of high quality persisted. A similar behavior has been previously observed for the growth of Co on Cu(1 1 13).[7]

The magnetic properties of the films were investigated *in situ* by means of the longitudinal magneto-optic Kerr effect. The Kerr ellipticity was measured utilizing an optical set-up similar to the Kerr experiment used by Bader and coworkers.[15] Magnetic fields up to 140 Oe could be applied parallel to the film plane. The fields were created by current driven core-less coils mounted inside the vacuum chamber.

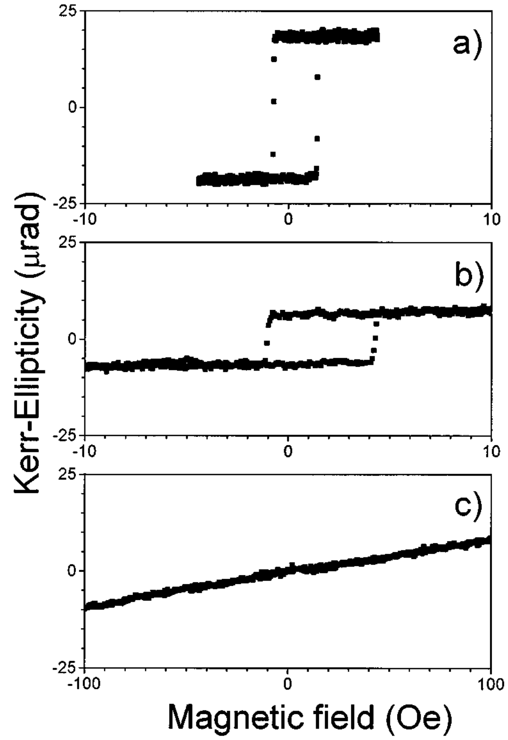

FIG. 1. In-plane magnetization loops for Co/Cu(1 1 17) ($d\approx$ 2.2 ML) taken at different azimuthal orientations. The angle φ measured with respect to the step edges is (a) 0°; (b) 65°; (c) 90°. The hysteresis loops have been obtained by means of the longitudinal magneto-optic Kerr effect at 40 °C.

MAGNETIZATION BEHAVIOR

As a first result we demonstrate qualitatively the strength of the in-plane uniaxial anisotropy in Co/Cu(1 1 17). Magnetization loops have been taken along different directions in the film plane. Fig. 1 shows the different hysteresis curves obtained parallel (a), at an angle of 65° (b) and perpendicular (c) to the step edges, respectively. The data were taken at 40 °C and a film thickness of $\approx$ 2.2 ML. Along the step edges (i.e., parallel to the $[1\bar{1}0]$ direction) the hysteresis loop exhibits a high squareness with an extremely sharp switching behavior [see Fig. 1(a)]. The film magnetization is completely reversed within a switching field of $\approx$ 2 Oe. With increasing in-plane angle φ to the step edges, the remanence decreases while the coercivity increases [see Fig. 1(b)]. Perpendicular to the step edges [Fig. 1(c)] no hysteresis loop can be observed. In this case, a small slope in the magnetization curve is resolved but no remanence found. Thus, it was not possible to magnetize the film perpendicular to the step edges with the magnetic fields we could apply. This demonstrates that an easy axis of magnetization exists parallel to the step edges.

Assuming that the film behaves purely uniaxial, one has to expect a cosφ dependence of the remanence signal due to the projection of the easy axis magnetization onto the axis of observation. A polar plot of the remanence is shown in Fig. 2 for a 3.6 ML film. The plot proves unambiguously the uniaxial behavior of the films, as the measured remanence (squares) fits the cosφ behavior (line) nearly perfectly. The increase of the switching field with φ [see Fig. 1(b)] is also a consequence of geometry. The projection of the applied field onto the easy axis has to overcome the switching field of the easy axis. Hence the as-grown films clearly have a uniaxial magnetic behavior. At room temperature the easy axis of magnetization is parallel to the step edges and magnetization reversal takes place along the easy axis only. The in-plane direction perpendicular to the step edges is a hard axis of

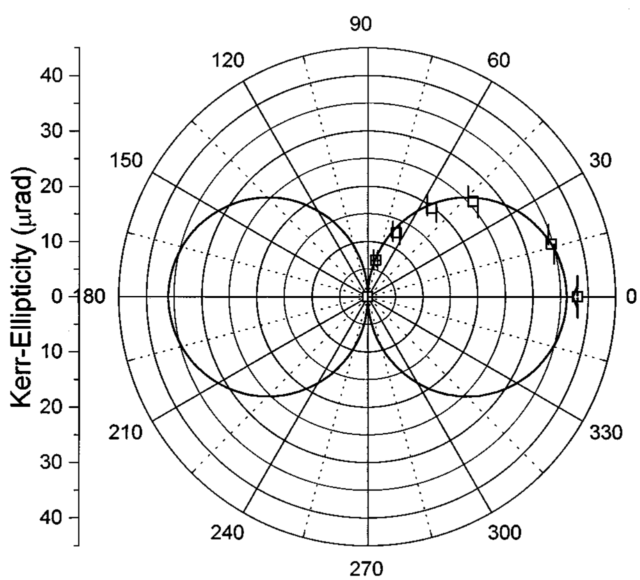

FIG. 2. Polar plot of the Kerr ellipticity for a Co/Cu(1 1 17) film of 3.6 ML thickness. The vertical bars give the 1σ-statistical uncertainty. The solid line gives the cosφ behavior fitted to the data points.

magnetization. The same behavior continues up to $\approx$ 15 ML, i.e., for all thicknesses studied. This is in agreement with previous findings for Co/Cu(1 1 13).[16]

MAGNETIC ANISOTROPY OF AS-GROWN FILMS

Perpendicular to the step edges the response of the magnetization to variations of the magnetic field is strictly reversible. The linear dependence in weak fields, which can be seen as a straight line of small slope shown in Fig. 1(c), is due to the rotation of the magnetization of the whole film. The same is true for the small increase of the signal above (below) the switching field in Fig. 1(b). Hence, any magnetization along the hard axis is induced via rotation of the magnetization away from the easy axis when applying an external field. No domains magnetized along the hard axis can be created and no hysteresis effects are found along the hard axis.

Using the magnetic free energy as a function of the orientation of the in-plane magnetization, that characteristic behavior can be used to determine quantitatively the anisotropy constants from the shape of the magnetization curve along the hard axis. In addition to a twofold in-plane anisotropy contribution, a fourfold contribution is considered. Such biaxial anisotropies have to be taken into account in order to properly describe the magnetic behavior of the (001) terraces, which should display a fourfold symmetry like Co/Cu(001).[16–18] Hence, the orientational free energy $V(\varphi)$ for Co films on vicinal Cu(11n) can be written in the simplest form as:

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

φ is the angle of magnetization with respect to the step edges. K_u is the uniaxial and K_4 the biaxial anisotropy constant. A positive K_u means that the easy axis is parallel to the step edges. If K_4 is negative the biaxial contribution favors magnetization along the $\langle 110\rangle$ directions, i.e., parallel and perpendicular to the step edges. A positive K_4 means that the biaxial contribution favors $\langle 100\rangle$ directions as easy axes (45° to the step edges). The ansatz is equivalent to the one used in Ref. 19.

At equilibrium the torque caused by the external field applied along the hard axis is counterbalanced by the torque due to magnetic anisotropy of the film. That means that the first derivative (with respect to the angle) of the free energy density

$$F(\varphi)=V(\varphi)-I_S H\cos\left(\frac{\pi}{2}-\varphi\right) \tag{3}$$

is zero. Here, φ is the in-plane angle of magnetization with respect to the easy axis, H the magnetic field strength applied along the hard axis and I_S the saturation magnetization of the film. With neutron diffraction experiments it has been shown that the magnetic moment of Co in Co/Cu(001) films is the same as in bulk cobalt.[20] As we are not able to determine I_S, we assume this to be the case also for Co/Cu(1 1 17).

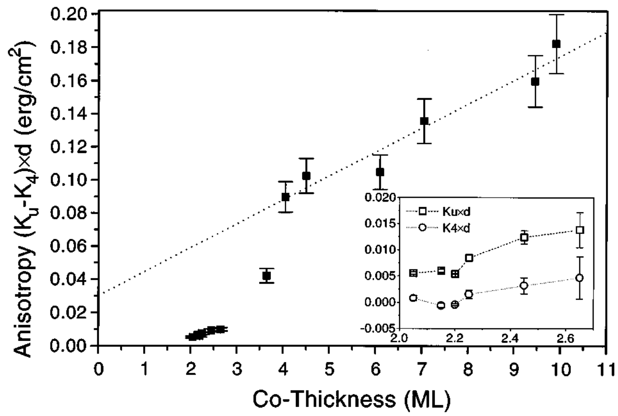

FIG. 3. In-plane magnetic anisotropy $(K_u-K_4)\times d$ versus thickness d for Co/Cu(1 1 17). The insert shows $K_u\times d$ (squares) and $K_4\times d$ (circles) for the very thin films. The error bars give the 1σ statistical errors.

The field dependent angle of rotation $\varphi(H)$ can be obtained from the Kerr signal along the hard axis. For small rotation angles a linear dependence is found which allows to determine easily the quantity (K_u-K_4) from Eqs. (2) and (3) in the small angle approximation.[21] In the case of larger rotation angles, a slight deviation from the linear behavior can be identified which makes a separation of K_u and K_4 possible. Using Eqs. (2) and (3) and the field dependent angle $\varphi(H)$ the anisotropy constants can be determined by a fit, analogous to the method used in Ref. 22–24. Due to the limitation of the magnetic field strength, the two- and fourfold anisotropy constants could be separated only for the thinnest films, i.e., below $\approx$3 ML.

In Fig. 3 $(K_u-K_4)\times d$ is plotted versus film thickness d. For film thicknesses above 4 ML $(K_u-K_4)\times d$ rises linearly with film thickness. Below 4 ML the quantity deviates strongly from the linear thickness dependence. The linear behavior proves that a constant magnetic anisotropy contribution adds up with increasing thickness which is usually attributed to a volume property. Hence, a separation of the anisotropy into surface, interface and volume contributions, in accord with Equation (1), can be made above 4 ML. A least-squares fit yields the dotted line in Fig. 3. The slope of the graph is the volume anisotropy $(K_u-K_4)_v$ which is 7.9×10^5 erg/cm^3. The sum of surface and interface anisotropies can be extrapolated from the intersection of the linear graph with the ordinate. The fit yields for the sum a value of about 3×10^2 erg/cm^2 for $(K_u-K_4)_{si}$. Below 4 ML the magnetic anisotropy (K_u-K_4) drops by almost one order of magnitude, from (0.09 ± 0.01)erg/cm^2 at 4 ML to less than 0.010 erg/cm^2 at $\approx$2.6 ML (Fig. 3). One might conjecture that this drop is due to the absence of the volume contribution, as the films below 3 ML consist of surface and interface layer only. The (K_u-K_4) value for the thinnest films ($\approx$0.007 erg/cm^2) is, however, much smaller than the extrapolated surface/interface quantity. The thinnest films obviously exhibit a completely different behavior and the steep decrease cannot be explained by a vanishing volume contribution.

The insert of Fig. 3 shows $K_u\times d$ and $K_4\times d$ for films thinner than 3 ML. It is obvious that the uniaxial contribution $K_u\times d$ is dominant. With increasing film thickness $K_u\times d$ gets larger. The biaxial contribution, however, is considerably smaller and varies only slightly with thickness. Within this thickness range K_4 has a positive value, though very small. Hence, the biaxial anisotropy, which has to be attributed to the influence of the terraces, favors an easy axes parallel to the $\langle 100\rangle$ directions. This behavior is in contrast to observations in Co/Cu(001) films where for all thicknesses the $\langle 110\rangle$ directions are easy axes of magnetization.[17,18,25]

DISCUSSION

First, we compare our data with published results for Co/Cu(1 1 13). The BLS investigations have been performed on thicker films (above 4 ML).[10] The dependence of anisotropy on film thickness has been found to be in good agreement with a linear dependence law. This is not surprising for that thickness range, in the light of the findings presented above. In the BLS experiment the volume contributions have been determined as $K_{uv}=(6.0\pm 0.7)\times 10^5$ erg/cm^3 and $K_{4v}=-(6.5\pm 0.2)\times 10^5$ erg/cm^3, which yields $(K_u-K_4)_v=(12.5\pm 0.9)\times 10^5$ erg/cm^3. This is close to the value we obtain by a least-squares fit to our data above 4 ML, i.e., $(8.0\pm 0.8)\times 10^5$ erg/cm^3. Scaling the uniaxial anisotropy value for Co/Cu(1 1 13) by the ratio of the step densities one would expect $(K_u-K_4)_v=11.1\times 10^5$ erg/cm^3 for Co/Cu(1 1 17). This value does not agree with our result within the given error margins. The deviation might be attributed to the different composition of the films (see discussion below).

Strong deviations between our results and the BLS data show up for the surface/interface anisotropy, i.e., the intersection with the ordinate. The BLS data yield $(K_u-K_4)_i=(-0.042\pm 0.008)$ erg/cm^2,[26] whereas we obtain a *positive* value of (0.03 ± 0.01) erg/cm^2. The most striking difference is the opposite sign. This discrepancy is most likely caused by the different interface anisotropies which are probed in the two experiments. In our experiments an uncovered film was investigated, which means that two different interfaces, Co-Cu and Co-vacuum, contribute to the anisotropy. The BLS experiments were performed on a Cu-covered film. Hence, a symmetric layer structure, consisting of two Co-Cu interfaces was investigated. One is led to conclude that the Cu-layer covering the Co film changes the surface/interface contribution. This assumption is confirmed by comparing previous measurements. While Cu covered films are expected to have an easy axis perpendicular to the step edges below a critical thickness of about 2.9 ML,[10] measurements with uncovered films on Cu(1 1 13) revealed an easy axis along the steps in that thickness regime.[9]

The different results can be reconciled, if we assume that the anisotropies attributed to the Co-Cu and Co-vacuum interfaces are of opposite signs. The Co/Cu interface apparently favors magnetization perpendicular to the step edges, but in the uncovered film the contribution of the Co/vacuum interface dominates. Upon covering the films, the Co/vacuum interface is eliminated and the interface anisotropy changes sign.

In the following, we turn to the discussion of two major characteristics of the thickness dependence of anisotropy, namely the strong deviation from the ''bulk''-like behavior in the thinner films and the positive value of the biaxial anisotropy in the very thin films.

The drop in (K_u-K_4) observed at 3 ML is far too large to be explained by the absence of the bulk anisotropy contribution only. The strong decrease has to be attributed to changes in the surface and/or interface anisotropy contributions. In the thickness range below 3 ML the discrete nature of the films seems to manifest in the magnetic properties, such that a separation of anisotropy into interface, bulk and surface contributions is very questionable. Additionally, strong structural changes and lattice relaxations occur in the first few layers and the structure of films below 3 ML are known to depend strongly on the amount of deposited material. It is to be expected that the structural variations will have a strong effect on the magnetic properties in the very thin films.

The biaxial anisotropy reflects such influences in the thickness range up to 3 ML. In this range $K_4\geqslant 0$ which means that the easy axes of magnetization lies along the $\langle 100\rangle$ directions. If the biaxial anisotropy is mainly determined by the film structure on the terraces, the positive value indicates that the structure on the (001) terraces is different from the arrangement of atoms on Cu(001).[18,25]

One is led to ask which anisotropy contribution is responsible for the change of the anisotropy (K_u-K_4) in the thickness range around 3 ML. The hysteresis loops indicate that the uniaxial anisotropy contribution gets considerably larger with thickness. From the temperature behavior of the differential susceptibility in the 4 ML film it can be deduced that the biaxial contribution also increases with thickness while the temperature dependence of the anisotropy indicates that K_4 is negative in that thickness range.[27] Hence, K_4 changes sign and becomes stronger around 3 ML in qualitative accordance with the negative fourfold magnetic anisotropies found in the BLS experiments for copper covered Co/Cu(1 1 13) above 4 ML.[10] Hence, we conclude that the abrupt increase of the anisotropy (K_u-K_4) is due to changes of both uniaxial and biaxial anisotropy contributions.

The increase of the uniaxial contribution could be easily explained by smoother steps in the thicker films, e.g., above 4 ML. Recent STM investigations have indeed shown that the thinner films are quite rough.[28] The values of the biaxial contribution for thin films, however, cannot be understood with this assumption. If the steps are rougher the uniaxial contribution will certainly become smaller and biaxial contributions will dominate. The fourfold lattice symmetry on the terraces, however, should not be altered by that effect. Hence, the behavior would be close to Co/Cu(001), i.e., negative K_4 which vanishes below 2 ML.[18,25] The measured biaxial anisotropy is, however, positive which indicates that the arrangement of atoms on the terraces is different from that in Co/Cu(001). Thus the decrease of anisotropy in the thinnest films cannot be completely explained by growth induced step disorder. As the biaxial anisotropy becomes negative above 4 ML, the arrangement of terrace atoms in the thicker films must be similar to that on Cu(001) surfaces. Hence, in the same thickness range where the strong increase in $(K_u - K_4)$ is observed, a structure on the terraces is established which is similar to that in Co/Cu(001) films. It seems likely that the origin of the changes of uniaxial and biaxial anisotropy are intimately linked.

What remains as a possible structural influence is the strain in the films which was also suggested as driving force for the restructuring of the vicinal Cu surface in the submonolayer coverage regime.[28] The strain is expected to change with thickness as thickness dependent relaxations are likely to occur, similar to those found for Co/Cu(001).[29] Kowalewski *et al.* have found a strong decrease of the fourfold anisotropy in Co/Cu(001) in the low thickness limit.[25] They have correlated the strong deviation from ''bulk''-like thickness dependence with such relaxations. Their results for Co/Cu(001), however, do not show the sign alteration in the biaxial anisotropy. Hence, if the same relaxations occur, the change of sign is particular to vicinal surfaces. The following scenario can be imagined: In case of vicinal surfaces the steps will influence the relaxation. Besides the mismatch within the surface plane, the film has to match the template lattice in the vertical direction at the step edges. If the Co film grows in perfect registry with the lattice on the terraces, the vertical mismatch at the steps would add up to 4.6%. On vicinal surfaces the Co films therefore have to fit both the strong mismatch in the vertical direction at the steps and the 2% mismatch within the film plane on the terraces.[30] This situation represents the important difference to films grown on low index surfaces. The high density of steps thus gives rise to additional strain contributions which cause lattice relaxations that differ from those taking place on the (001) surface. The strain originating from the steps will effect the strain and the local symmetry of atoms on the terraces, thus influencing the biaxial magnetic anisotropy. Additionally, if the strain is responsible for the film structure found in the STM study,[28] it should also cause a reduction of the uniaxial magnetic anisotropy. In any case, one has to expect that effects of relaxations on the magnetic properties should manifest in a completely different way on low index and vicinal surfaces. With increasing thickness the magnetic behavior changes and the biaxial contribution is more like that expected for Co on (001) terraces. That indicates that the Co-Co bond will dominate the Co-Cu bond and cause the lattice to become less influenced by the template above 4 ML. This was also concluded from the STM results.[28]

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