Stability of the perpendicular magnetic anisotropy of ultrathin Ni films on Cu(100) upon multiple magnetization reversals

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Ultrathin Ni films with perpendicular magnetic anisotropy were deposited on Cu(100) at room temperature. The magnetic properties of the films were measured upon multiple magnetization reversals in an alternating magnetic field using the magneto-optic Kerr effect. All magnetic properties remain virtually constant for up to 10^6 magnetization reversals for all films studied. This finding is supported by a simple theoretical model.

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In recent years, substantial effort has been focused on the preparation and characterization of ultrathin magnetic films with perpendicular magnetic anisotropy because of their potential application as high density storage media.[1] A vast number of studies has discussed the origin of the perpendicular anisotropy, its thickness and temperature dependence, and related properties.[2,3] On the other hand, the stability of magnetic properties upon multiple magnetization reversals has to our knowledge not been studied for thin magnetic films. Nevertheless, it is a fundamental requirement for data storage materials that their magnetic properties should have high stability under their working conditions. Magnetic recording media undergo a large number of rewrite and erase processes. Therefore, if their magnetic properties, including the perpendicular anisotropy, the remanent magnetization, and the coercive field, change with the number of magnetization cycles, recording failure or data loss can occur. It is hence of key importance to check the magnetic stability of thin films that are prospective recording media. With this goal in mind, we have investigated the magnetic stability of thin Ni films on Cu(100). Ni films on Cu(100) show a perpendicular magnetic anisotropy over a wide thickness range,[4–7] which makes them interesting candidates for a study to determine the stability of the magnetic anisotropy versus multiple magnetization reversals.

Several other film systems have been found to possess a perpendicular magnetic easy axis as well. A popular example is CoCr films grown in the *c*-axis orientation, where the anisotropy has a magnetocrystalline origin.[8] Epitaxial superlattices of Co/Pt, Co/Pd, and Co/Au have also been studied extensively since they offer a large Kerr rotation. Their perpendicular anisotropy is believed to originate from the interface anisotropy due to the reduced coordination number of the interface atoms.[9] The reduced coordination number also makes ultrathin Fe and Co films exhibit a magnetic anisotropy with an easy-axis perpendicular to the film plane. For these films, there exists a critical thickness below which the surface anisotropy is sufficient to overcome the demagnetization field and the films are preferentially magnetized in perpendicular direction.[10,11] However, the critical thickness is usually only a few monolayers (ML's). This thickness limitation is disadvantageous for practical applications. A different behavior is found in Ni films on Cu(100), which show a transition from in-plane magnetization to perpendicular magnetization with increasing film thickness.[4–7] Within the thickness range from 7–10 ML to 37–70 ML, the Ni films show a perpendicular easy axis.[12] This large thickness range and the particular origin of the magnetic anisotropy outlined below make Ni films on Cu(100) an interesting candidate for studies of stability with respect to magnetization reversal. Hence we have checked if the magnetic properties show any change after the films were subject to 2×10^6 magnetization reversals in an alternating sweeping field.

The magnetic stability upon multiple magnetization reversals has recently been studied for several systems.[13,14] For spin-dependent tunneling devices, where a hard magnetic Co alloy layer is coupled over an aluminum oxide film to a soft magnetic Co layer, a decay of remanent magnetization is observed upon multiple magnetization reversals.[13] It has been suggested that the decay of the moment of the hard layer is caused by the demagnetizing field at the hard layer associated with domain walls in the free layer.[13] Aging effects caused by structural alterations have also been observed in metastable amorphous phases.[15] Concerning the stability of the magnetic properties of ultrathin Ni films on Cu(100), to our knowledge, no work has been reported as yet. These films are prone to considerable elastic strain that could influence the magnetic properties and might be a source of metastable structures.

Ultrathin Ni films grown on Cu(100) show a peculiar behavior regarding the magnetic anisotropy. For a film thickness between 7 ML and 9 ML, a sharp spin reorientation transition from in-plane to out-of-plane is observed. At much larger film thicknesses between 37 ML and 70 ML, the pref-

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erential magnetization direction gradually rotates back to the film plane. Hence in the range from 9 ML to 37 ML, the films show a perpendicular easy axis. Using ferromagnetic resonance, Schulz and Baberschke[4] found that this unusual behavior results from the competition between the perpendicular magnetoelastic volume anisotropy due to mismatch-induced strain and the sum of shape anisotropy plus surface and interface anisotropy. The latter favors in-plane magnetization.[4] In the pseudomorphic region, Ni films have an in-plane lattice constant expanded by 2.5 % to match that of the Cu substrate, causing a corresponding contraction of 3.2% in the perpendicular direction.[16] The tetragonal stress-induced perpendicular uniaxial anisotropy is determined by $K_u = \frac{3}{2}\lambda_{100}(c_{11}-c_{12})(\varepsilon_2-\varepsilon_1)$. Here λ_{100} is the magnetostriction constant, c_{11} and c_{12} are the cubic elastic constants, and ε_2 and ε_1 are the in-plane compressive strain and tensile strain in perpendicular direction, respectively.[4] Therefore the strain state directly determines the value of the perpendicular anisotropy and is closely correlated to the preferential magnetization direction. When the thickness increases beyond the pseudomorphic region, strain relief by dislocation formation begins and the perpendicular anisotropy is gradually reduced. At last, the easy axis returns back to the plane. This immediately implies that strain relief will affect the magnetic anisotropy. While the previous studies have mainly focused on strain relief by dislocation formation with increasing thickness, it should also be possible to cause strain relief via dislocation formation by magnetic field sweeps. During the sweep of an hysteresis loop an energy of $4\mu_0\mu_s H_c$ is released. This energy could facilitate dislocation formation and hence would change the film strain and magnetic anisotropy. To investigate the relevance of this mechanism, Ni films with a number of different thicknesses, which all showed a perpendicular magnetic anisotropy, were exposed to a large number of magnetization reversals.

Ni films were prepared and analyzed in a ultrahigh vacuum chamber, which has already been described elsewhere.[17] The chamber has a base pressure of 6×10^{-9} Pa and is equipped with a low-energy electron diffraction (LEED) and medium-energy electron diffraction (MEED) system, a cyclindrical mirror analyzer (CMA) for Auger analysis, and a pair of Helmholtz coils to apply magnetic fields up to 1000 Oe. The substrate was a polished Cu(100) single crystal, approximately 7 mm in diameter and 2.5 mm thick. The sample was cleaned by Ar-ion sputtering with an ion energy of 2 keV at room temperature. Subsequently the Cu crystal was annealed at 900 K for 5 min to reduce the sputter damage. High-purity Ni (99.98%) was evaporated from a thin Ni platelet of 10 mm diameter and 0.2 mm thickness, heated by the radiation of a tungsten filament. The Ni films were deposited at room temperature using a deposition rate of 0.3 ML/min. During evaporation, the pressure did not exceed 3×10^{-8} Pa. The growth of the films was monitored by MEED, employing an electron energy of 3 keV at an angle of incidence of 5° against the surface. Magnetic properties of the films were characterized using the magneto-optic Kerr effect (MOKE). Hysteresis loops were recorded in polar geometry. The light source was a He-Ne laser with a wavelength of 632.8 nm.

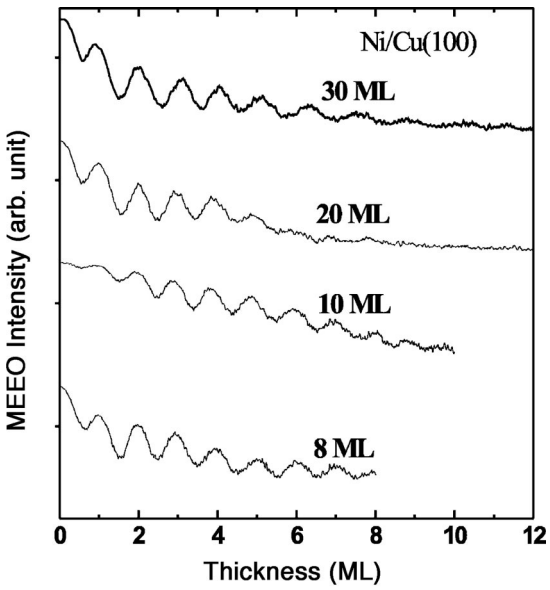

FIG. 1. MEED intensity of the $(2,\overline{3})$ beam for four different deposition runs. The film thickness is estimated from the intensity oscillations. See text for details.

Ni films with various thicknesses have been studied. Figure 1 shows the MEED measurement of one particular beam $(2,\overline{3})$ for four different deposition runs. Comparison of the different curves facilitates the identification of generic features. The MEED intensity is characterized by regular oscillations superimposed on a decrease of average intensity with growing film thickness. The oscillation in MEED intensity corresponds to an oscillation in average terrace width. The maximum intensity of each oscillation is indicative for the completion of each successive monolayer. The decreasing amplitude of the oscillations with thickness suggests an increasing surface roughness. Hence oscillations can only be observed up to approximately 10 ML. This finding is in agreement with a previous study on the growth of Ni on Cu(100).[18] In Fig. 1 MEED data are recorded for films with a thickness of 8, 10, 20, and 30 ML, but MEED data are only shown up to 12 ML. For the former two curves the film thickness can be determined precisely from the MEED oscillations. For the film thicknesses of 20 and 30 ML the thickness is extrapolated from the last five discernible oscillations. This leads to an error bar of less than 6% for the total thickness. Once the film was fabricated, a hysteresis loop was recorded. Subsequently the film was subjected to continuous magnetization cycles in an alternating applied field, which is perpendicular to the film plane. For a typical field sweeping rate of 0.1 s per cycle, it usually takes about 30 h to perform 10^6 magnetization cycles. Hence we have first checked if the magnetic properties of Ni films change with time due to the absorption of residual gas. As soon as the Ni film was fabricated, a hysteresis loop was recorded. After 30 h a second hysteresis loop was recorded. Figure 2 shows both hysteresis loops. The comparison reveals that the magnetic properties (M_r, M_s, H_c) are not affected by the 30 h exposure to residual gases.

Having excluded the influence of the absorption of gases

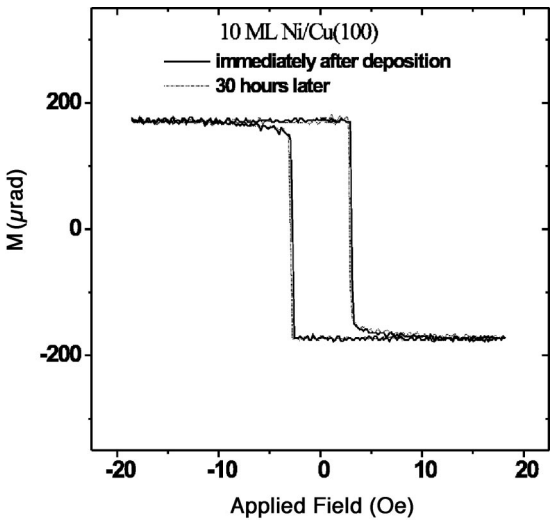

FIG. 2. A comparison of the magnetic hysteresis measured immediately after film fabrication and 30 h later. The film is 10 ML thick.

at the film surface, the magnetic stability with respect to magnetization reversal was examined. The amplitude of the alternating field is twice as large as the coercive field of the film studied. The changing rate of the applied field is 500 Oe/s. All experiments were carried out at room temperature, which is 31 °C. A small increase of sample temperature by $1-2$ °C was observed after the film stayed in the alternating field for more than 2 h. Hysteresis loops were measured after certain numbers of magnetizing cycles (Fig. 3). One can see that the shape of the hysteresis remains unchanged even after 2×10^6 magnetization reversals. Basic parameters such as

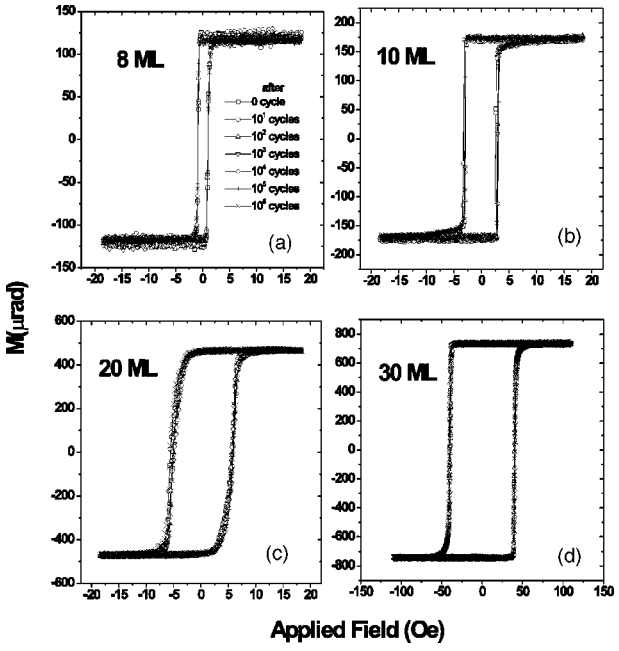

FIG. 3. Hysteresis loops recorded after various numbers of magnetization cycles for the films with a thickness of (a) 8 ML, (b) 10 ML, (c) 20 ML, and (d) 30 ML. The same symbols are used in (b), (c), and (d) as in (a).

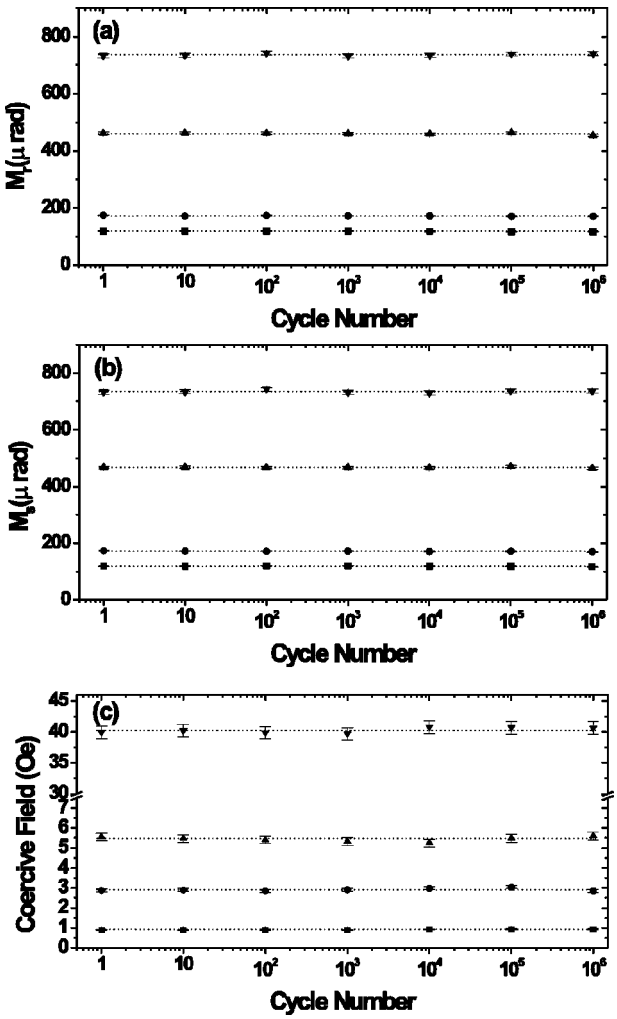

FIG. 4. Plots of (a) remanent magnetization, (b) saturation magnetization, and (c) coercive field against the number of magnetization cycles. The solid squares describe the 8 ML film, while solid circles, solid down triangles, and solid up triangles characterize the 10 ML film, 20 ML film, and 30 ML film, respectively. The dotted lines are generated assuming constant magnetic properties.

saturation magnetization, remanent magnetization, and coercive field are determined from the loops. They are presented in Fig. 4. The error bar for the remanent and saturation magnetization is 1%, while the error bar for the coercive field is 3%.[19] As can be seen from Fig. 4, the data can be fitted by a constant value. The scatter in the data for the three quantities studied here lies well within the error bars. This implies that the remanence, the saturation magnetization, and the coercivity remain constant. We have also tried to fit the data with fit functions other than a constant behavior but could not get a significant improvement in the fit. Hence it is safe to say that the perpendicular magnetic anisotropy of Ni films on Cu(100) as well as the magnetic properties do not change even after a large number of magnetization reversals. This finding is crucial both for an application of magnetic thin films in storage media and for basic studies of ultrathin films. In such studies the magnetic signal is often averaged over a large number of loops to improve the signal-to-noise ratio. Our study shows that at least for Ni films on Cu(100), this

approach is fully justified. In the last section of this report we will discuss how the result presented above can be explained by a simple analysis.

For Ni films on Cu(100), the magnetic anisotropy is governed by competition between the surface and interface magnetic anisotropy and two volume contributions, namely, the shape anisotropy and the magnetoelastic anisotropy. It is the interplay between the latter two quantities that determines the magnetic anisotropy. The shape anisotropy linearly increases with thickness, while the magneto-elastic contribution shows a somewhat more complicated behavior. It depends on the film thickness and the strain of the film, which itself is thickness dependent. The film strain is caused by the mismatch between the lattice parameters of the film and the substrate. For ultrathin films below the critical thickness pseudomorphic growth is expected. Above the critical thickness the strain energy will be reduced by dislocation formation. Both will contribute to the total energy of the film system. By minimizing the sum of the strain energy and the energy due to dislocations in the film the thickness dependence of both strain and interface dislocation density can be determined. For a small mismatch, a critical thickness exists, below which the film grows pseudomorphically. Above the critical thickness dislocations appear and their density increases with film thickness. Therefore the strain is uniquely decided by the film thickness and so is the perpendicular anisotropy. However, the energy of $\Delta E = 2\mu_0 M_s H_c$ released in the magnetization reversal process can perturb the system. Hence we have to compare ΔE to the elastic energy and the dislocation energy. The energy is given as an average per Ni atom. The values are listed in Table I. The energy release upon magnetization reversal has been derived from an estimated magnetic moment of $0.6\mu_B$ per Ni atom to obtain an upper limit of the energy release. Theoretical studies[20,21] have predicted the magnetic moment of Ni on Cu(100) to be only reduced directly at the Ni/Cu interface but find otherwise bulklike moments of around $0.6\mu_B$ in the Ni film. Experimental studies show a considerable scatter with some reports of reduced average moments in the Ni film[22,23]. Recent reports indicate that there might be a strain effect on the magnetic moment of the Ni film, where relaxed films have larger moments.[24] Since we are mainly interested in an upper estimate we calculate the energy release using a high moment of $0.6\mu_B$ for the entire film. The data reported for the critical thickness of pseudomorphic growth show considerable scatter. The critical thickness was either determined by experiments or predicted theoretically. Here we use O'Brien's data for which h_c is 13 ML.[12] For film thicknesses h larger than h_c, ε is approximated by $\varepsilon = \eta h_c / h$,[25,26] where η is the mismatch between the substrate and the deposited film. The equation for ε determines the remaining film strain and is used to calculate the dislocation density. The elastic constants are assumed to be the same as that for the bulk. In addition, a Poisson's ratio of 1/3 is assumed. The Burgers vector is considered to be of $\frac{1}{2}\langle 110\rangle$ type, which is typical of fcc-structured films, yielding $b = (1/\sqrt{2})a_0$, where b is the length of the Burgers vector and a_0 is the length of the cubic unit cell of Cu. A close look at Table I reveals that the energy per atom released in one hysteresis loop is significantly smaller than the energy of the dislocations E_d or the elastic energy of the strained film. Hence it is unplausible that magnetization reversal will have a profound effect on the film strain and the perpendicular magnetic anisotropy. This conclusion should even remain valid if a large number of magnetization reversals have been performed. In this case there will be a considerably higher energy release, but only for very fast magnetization experiments will significant power be absorbed.

TABLE I. Comparison between the energy released during magnetization reversal (ΔE) and the elastic energy (E_e) or the dislocation energy (E_d) of the films. The energy is given as an average per atom.

Thickness of film (ML)	8	10	20	30
ΔE (meV)	6.6×10^{-6}	2×10^{-5}	3.8×10^{-5}	2.9×10^{-4}
E_e (meV)	9.0	9.0	3.8	1.7
E_d (meV)	0	0	11.5	13.2

It is interesting to compare our experimental finding with a previous study that shows a decay of remanent magnetization upon multiple magnetization reversals.[13] In this case, however, the coupling between two magnetic layers leads to a new decay channel for the magnetization. Such a channel is not available in the present system. Aging effects have also been reported for metastable phases.[15] In such systems, the energy released during magnetization reversal accelerates the microstructural change. Metastable structures possibly also exist in Ni films on Cu(100), if, for example, the dislocation density would be lower than equilibrium density of dislocations. Under those circumstances, the energy released during magnetization reversal could be used to form dislocations and hence change the strain and the magnetic properties of the films. That we do not observe any change in magnetic properties indicates that the dislocation density is close to the equilibrium density. This is supported by the thickness dependence of film strain,[27] which can be fairly well described by an equilibrium theory for the residual in-plane film strain.

In conclusion, Ni ultrathin films have been prepared with a perpendicular easy axis on a Cu(100) substrate using molecular beam epitaxy. The magnetic properties of the films were monitored during continuous magnetization reversals in an alternating magnetic field. The results show that the films keep the magnetic properties unchanged even after 10^6 sweeps independent of the detailed strain state of the films. This finding can be understood based on the small energy released during each individual magnetization loop.

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[19] The coercivity of Ni films presented in this work is considerably smaller than the value reported by O'Brien *et al.* (see Ref. 12) but is in reasonable agreement with measurements by Huang *et al.* (Ref. 7) if we take into account that (H_c) increases by a factor of 4 or 5 when the samples are cooled from room temperature to 160 K, as was observed in our temperature-dependent measurement. The discrepancy with the result of O'Brien *et al.* could either come from the fact that they used a wedge sample or is due to different densities of defects such as impurities.

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