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Magnetization Distribution of 180° Domain Walls at Fe(100) Single-Crystal Surfaces

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We present the first experimental analysis of domain-wall *fine* structure at the surface of single-crystal iron bulk material by means of a UHV microscope with spin-polarization analysis of the secondary electrons. It is found on Fe(100) that the domain wall of a 180° Bloch wall in the bulk is terminated in a Néel-type structure at the surface. In spite of this dramatic change of the wall structure, the wall width at the surface is about the same as inside the sample. For the 180° Bloch walls we find a width of 210 ± 40 nm.

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What happens to a magnetic domain wall in a material when it encounters a free surface? This question is as old as modern magnetism, dating back to the beginning of this century when Weiss[1] introduced the concept of magnetic domains. Pioneering experimental work on imaging of domain walls at surfaces was done by Schmidt, Rave, and Hubert[2] using Kerr microscopy and by Koike *et al.*[3] and Hembree *et al.*[4] using a scanning electron microscope with spin-polarization analysis of the secondaries. Two of these studies[2,3] could verify the magnetic "thick-film" model of Hubert.[5] The investigation of bulk material,[4] on the other hand, was confined to the imaging of a small stripe in between two oppositely magnetized domains at the surface of a not-well-specified technical material. A proof of the stripe not to be a small domain was not given nor was the internal structure analyzed. The result of this investigation was the existence of an in-plane magnetization *component* perpendicular to the magnetization of neighboring domains, thus not demonstrating the transition to be Néel type. From these considerations, the study[4] does not allow us to extract *general* results for the surface-wall behavior of a semi-infinite material. From the theoretical point of view, however, there is a strong demand for generality that could provide a major input to the micromagnetic description of domain walls at bulk surfaces. To yield such basic information, we studied the domain walls at the surface of a classical, well-specified, semi-infinite ferromagnet, an iron single crystal, going beyond the previous investigation by studying the internal structure of 180° walls.

The structure of the boundary between adjacent domains in bulk material was first analyzed theoretically by Bloch,[6] then Landau and Lifshitz,[7] Herring and Kittel,[8] Lilley,[9] and others. In particular, it has been shown[6,7] that between two large bulk domains of opposite magnetization direction the magnetization rotates in a so-called Bloch-type manner. The width of this 180° Bloch wall is of order 100 nm, usually determined by the interplay between exchange and anisotropy energies. As theoretically proven,[6,7] this configuration requires the lowest free energy if the domain wall is infinitely extended in two dimensions. Now, if we introduce a free surface by cutting the solid along a plane parallel to the magnetization orientation within the two domains and remove one half of the solid to infinity, the original Bloch wall will no more represent a minimum-free-energy configuration. At the surface the magnetization vector within the domain wall points out of the crystal. This generates magnetic stray fields, and in order to minimize the magnetic-stray-field energy the Bloch wall is expected to relax to some new configuration at and near the surface. While this problem was recognized quite early, up to the present time, there does not exist any realistic micromagnetic theory for the behavior of domain walls in the surface region of semi-infinite ferromagnets.

The very existence of perpendicular stray magnetic fields related to the rotation of the magnetic vector was never seriously put into question because of the great success of the decoration technique using fine magnetic particles clustering at domain walls, which was developed simultaneously by Hámos and Thiessen[10] and Bitter[11] in 1931. Particularly strong contrast is usually obtained from 180° Bloch walls on bulk samples. The requirement of free-energy minimization was partly taken into account by a model of Döring,[12] suggesting a *narrowing* of the Bloch wall at and near the surface by the order of 50%, thus reducing the spatial extension of the stray field and its energy content. Contemporary work on Fe single-crystal whiskers,[13] using improved decoration techniques, was interpreted as evidence for the existence of narrowed Bloch walls at the (100) surface of iron crystals.

In thin films, on the other hand, 180° Bloch walls are known to yield notoriously low Bitter contrast. This was explained by La Bonte[14] and Hubert,[15] who came to the same conclusion on different theoretical grounds: On both surfaces of the film the wall is of Néel-type character (i.e., with the magnetization vector rotating *in* the surface),[16] while in the center of the film the wall is of partly Bloch-type character. This model has been confirmed by numerous works using Lorentz microscopy with[17] or without differential phase contrast[18,19] in transmission electron microscopes. The existence of asymmetric walls up to film thicknesses of order 200 nm may thus be considered well established. Their characteristic features are the existence of a magnetic flux vortex in the center of the film, the virtual absence of stray fields, and an *enlarged* domain-wall width at the surface.[2,3] On the basis of his model for thick films, Hubert[5] conjectured that under certain circumstances semi-infinite Fe and Ni should also show asymmetric field-free surface domain walls, though this model is based on assumptions untenable for semi-infinite samples, and in spite of some model deficiencies (e.g., infinite surface wall thickness for infinite film thickness). This conjecture apparently is at variance with the interpretation of Bitter patterns quoted above.

This Letter reports on the first experimental test of these conflicting views with a clean, bulk single crystal. We built up a UHV scanning electron microscope with polarization analysis of the secondary electrons, similar to previous developments,[20,21] but with sufficient resolution to resolve the domain-wall structure of iron.

A major result is that on Fe(100) the bulk 180° Bloch walls end in a Néel-type surface domain wall. The implications of this finding for the interpretation of Bitter patterns are discussed at the end.

The concept of our microscope is similar to that of Koike and Haykawa.[20] It basically consists of a low-energy UHV field emission scanning-electron-microscope column (similar to the pioneering instrument of Ichinokawa and Ishikawa[22]) and the well-proven LEED detector[23] for spin-polarization analysis. Two polarization components are simultaneously detectable by measuring the intensity of the four {2,0} beams diffracted from the W(001) crystal. The sample is a Fe(100) single crystal with dimensions 5×9×2 mm^3. It was mechanically polished and heated beyond 760 °C in vacuum for 20 min. Before polarization analysis the surface was sputter cleaned by ion bombardment (Ar$^+$, 2 keV). This does not seriously affect the surface magnetization as was recently demonstrated by studying the ion-excited secondary-electron spin polarization.[24] The polarization contrast between oppositely magnetized domains was typically ±24% or above depending on contamination.[25] In one direction the sample was tilted by 45° against the detection axis (marked by crosses in the following figures). This allows us to measure any out-of-plane spin-polarization component, if existing.

A high-resolution image of the magnetic microstructure is shown in Fig. 1(a). The orientation of the measured spin polarization determines the color in the picture. The arrows show the orientation of the spin polarization for that color. As the spin polarization is opposite to the magnetization[26] the picture is exactly identical with the domain structure. Some roughening of the transitions between adjacent domains are caused by statistical uncertainty. In Fig. 1(a) one 180° wall and some 90° walls are visible. The most interesting part is the 180° wall, which shows a narrow yellow stripe between the neighboring oppositely magnetized domains. This means that in between the oppositely magnetized domains, a magnetization along the second easy axis in the surface (at 90° to the former ones) appears. A similar contrast, using this technique, was previously observed by Koike *et al.*[3] and by Hembree *et al.*,[4] as mentioned above.

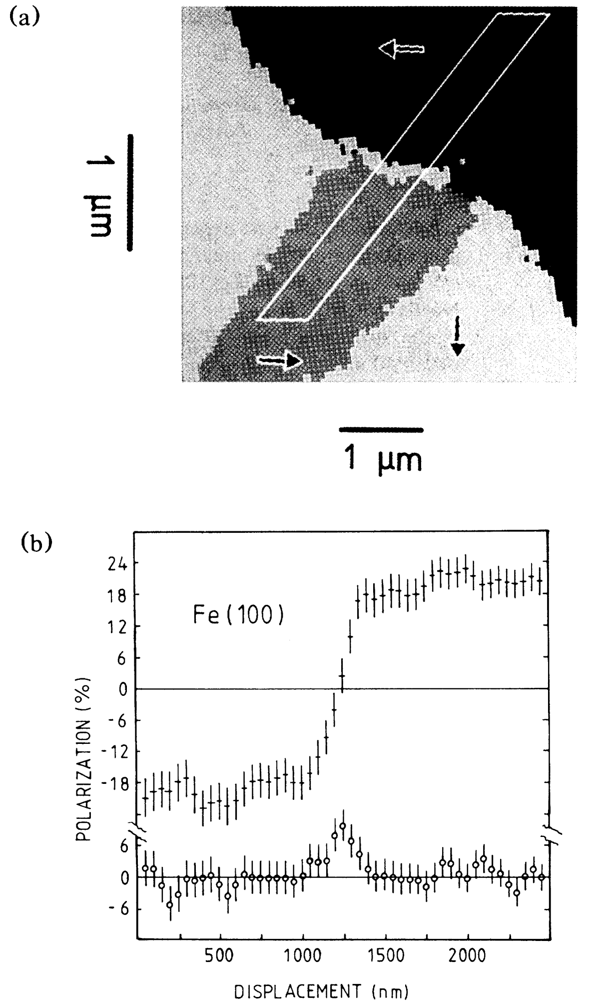

FIG. 1. (a) Magnetic surface domain structure on Fe(100). The arrows indicate the measured polarization orientation in the domains. The frame shows the area over which the polarization distribution of (b) is averaged. (b) Polarization distribution across a 180° domain wall, taken from (a). The vertical polarization component is indicated by the crosses. The circles show the horizontal polarization distribution. The step width is 50 nm. The error bars give the 1σ-statistical error.

To obtain more quantitative information about this region, we averaged over ten lines of the image running parallel to the long edge of the frame indicated in the picture. This distribution is shown in Fig. 1(b). The crosses indicate the polarization along the vertical direction, the circles show the perpendicular component lying in the surface.

Looking at the vertical component (crosses), we observe the change from negative [downward in Fig. 1(a)] polarization in one domain to the positive one on the other domain to be continuous. This proves that the transition region is not separated into two 90° walls with a small domain in between, but that there is a continuous transition between domains with opposite magnetization (180° wall). Near its zero crossing, the in-plane component (0) raises to its maximum value. This directly proves that the 180° wall is not Bloch type at the surface, because in case of Bloch-type behavior the in-plane component should be zero across the whole domain wall. These observations also rule out the above-mentioned models[12,13] based on an approximate solution of the micromagnetic equations. Starting with the one-dimensional solution (Bloch wall), these models consider the influence of minimizing the stray-field energy. This yields a very small center part of the wall with Bloch-type configuration. At both sides of this part the magnetic flux is shunted by the ferromagnetic material itself, thus leading to a component in the surface parallel to the wall normal. For symmetry reasons, the components in the surface point to opposite directions on each side. Such wall structure consequently would yield a change in sign in the in-plane component, which is not observable.

Comparing both distributions, the magnetization vector seems to rotate counterclockwise across the wall. A question arising from this result is if the rotation takes part in the surface or if the plane of rotation is tilted against the surface. From the maximum value of the in-plane component being smaller than on the domains, one could infer such a tilting (supposing $|M|$ =const everywhere in the sample). However, from the experimental point of view, this could also be a consequence of insufficient lateral resolution. Therefore, high-resolution line scans across this wall (and several others) were taken. The result is shown in Fig. 2. The meaning of the symbols are the same as in Fig. 1(b) (see inset in Fig. 2). The step width, e.g., the spacing between adjacent points, is 20 nm. From the sharp edge on the P_0 component, we estimate a lateral magnetic resolution of less than 40 nm. The in-plane component (P_0) has drastically increased, due to the superior resolution, and its maximum equals (within the statistical fluctuations) the polarization found at saturation of the component P_+, i.e., inside the domains. In view of the statistics, a small tilt angle ($\leq 20°$) cannot be ruled out. A second check for estimating the maximum tilt angle was made by comparing the shape of the P_+ and P_0 distribution curves, in particular, the relative positions of zero crossing (+ component) and maximum (0 component). As the P_+ component measures a projection of one surface magnetization component as well as the component perpendicular to the surface (see inset in Fig. 2), its shape would be influenced if a tilting, e.g., a perpendicular component, existed. Corresponding model calculations confirm the in-plane magnetization within an error margin of ±20°. Identical results with equal or slightly less resolution were found for a number of other 180° walls, including those showing the 180° change along the second easy [100] axis in the surface.

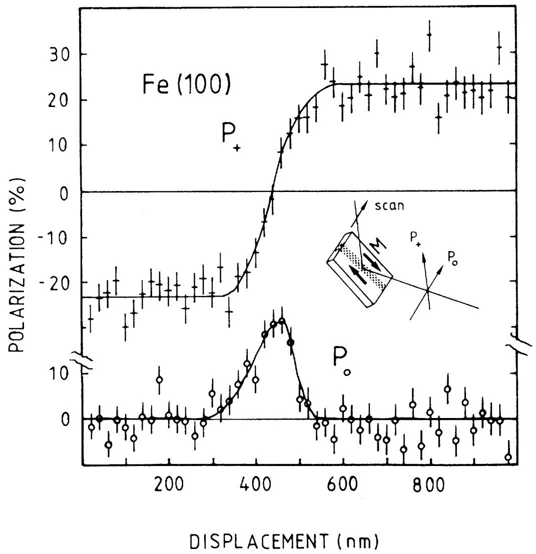

FIG. 2. High-resolution line scan across a 180° domain wall. Inset: The orientation of polarization detection axes relative to the sample. The P_+ component is corrected for the sample tilt. The lines are meant to guide the eye. The step width is 20 nm. The error bars gives the 1σ-statistical error.

In conclusion, the present investigations clearly demonstrate that the 180° domain walls at iron single-crystal surfaces are by no means Bloch type. The rotation of the magnetization within the wall occurs in a plane perpendicular to the wall plane, e.g., within the surface.

This result answers our question raised at the beginning: When a bulk 180° Bloch wall encounters a free surface, it changes its character into a Néel-type 180° wall at the surface. It should be emphasized that the magnetic probing depth of our method does not exceed the first ten atomic layers, a depth which is small compared to the distances over which rotations of the magnetization vector typically occur. We do not know where the transition from Bloch-type–to–Néel-type character takes place and over what depth it extends. Since our finding qualitatively agrees with Hubert's conjecture,[5] it is likely to find a vortex structure of the magnetic flux somewhere beneath the surface, but a new puzzle emerges with the width of the surface wall. As mentioned above, the thin-film model predicts the Néel-type surface wall to be substantially wider than the Bloch wall in the center of the film. Our experimental results do not confirm this prediction for iron: From the P_+ component in Fig. 2, one may infer a width $W = 135$ nm if conventionally one takes the intersection points of a tangent at the zero crossing with the extrapolated saturation values. We estimate the uncertainty in W to be ± 25 nm from drawing tangents with extremal slopes through the data points. As Hubert[27] pointed out, the relation $W = (2/\pi) W_L$ is valid, where W_L is the width with respect to the rotation angle of the magnetization, as defined by Lilley.[9] With this in mind, we obtain a width $W_L = 210 \pm 40$ nm. This value agrees quite well with the base width of the hump observed in the P_0 component. According to Lilley's[9] calculations for one-dimensional Bloch walls in cubic crystals, the width of a 180° Bloch wall in iron is given by $W_L = 10.87(A/K)^{1/2}$. With $K = 4.68 \times 10^4$ J/m^3 (Ref. 28) and $A = 2 \times 10^{-11}$ J/m (Ref. 29) for Fe, this amounts to $W_L = 225$ nm. The width of a 180° Bloch wall in the bulk has, to our knowledge, never been measured directly, but some indirect information is available. Hartmann and Mende[30] deduced recently an effective bulk domain-wall width of $\delta = 228$ nm from the susceptibility of 180° walls in iron whiskers. In Lorentz microscopy of single-crystal iron films, Suzuki and Suzuki[19] found a constant wall width of ~ 260 nm for film thicknesses above 200 nm. Comparing these data with our value for the surface domain-wall width, we find that apparently the wall width is the same in the bulk and at the surface—in disagreement with predictions from the thick-film model.

Given our result of the Néel-type surface termination of a 180° Bloch wall, which is expected from the thick-film model[5] to minimize the stray field, one is led to ask, Why does the Bitter technique, requiring a field gradient, work at all, and why does it work particularly well for 180° Bloch walls? We see two possible solutions to this dilemma: There remain sufficiently strong fields near the domain wall to attract the tracer particles. These fields would have to originate below the surface, most likely from the transition region between Bloch- and Néel-type walls. The other alternative would be that the ferromagnetic tracer particles themselves modify the domain-wall structure. For example, during the initial homogeneous deposition of the tracer, the Néel-type surface termination might be transformed into a more Bloch-type configuration with a stronger field, being shunted by the tracer material itself. Which of these alternatives applies will have to be decided in the future.

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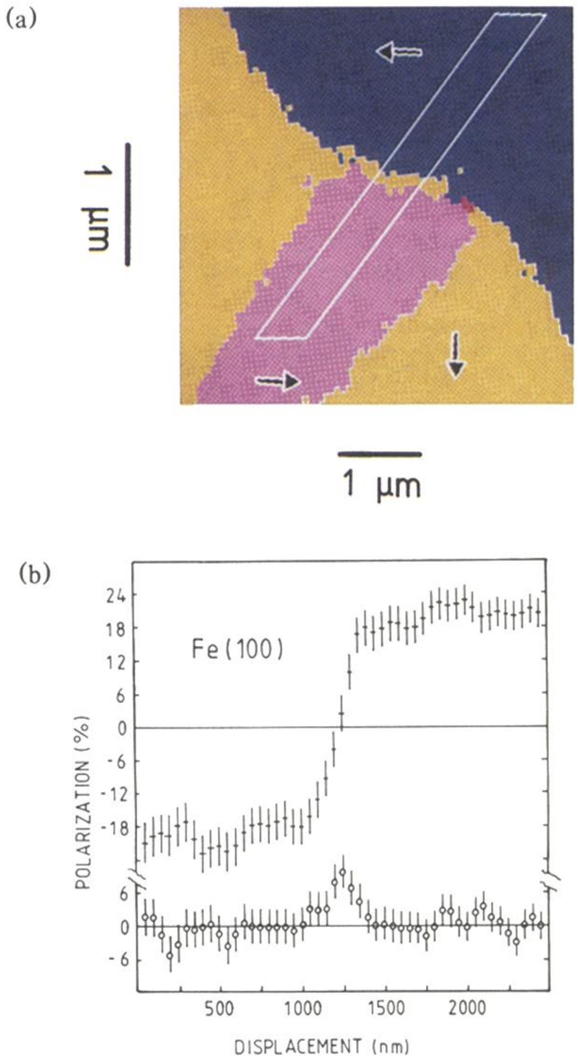

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