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Magnetic domain walls in thin molybdenum permalloy films observed under external magnetic fields by electron microscopy

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Dynamic observations of domain walls have been made using the Lorentz Microscopy technique. The aim of the investigations was to study the influence of magnetic fields on the domain wall stability and also to study the influence of structural features such as grain boundaries, twins, inclusions and second-phase particles on the initial permeability. Most of the thin foils used had thicknesses less than 100 nm hence the specimens could be referred to as thin films. Cross-tie walls which were noted to occur in materials with high permeability, i.e. materials of low crystal anisotropy, K_i, were observed at intermediate thicknesses of the thin foils. Both the cross-tie length and period are dependent upon film thickness, each becoming shorter with increasing thickness. The influence of the applied field on the cross-tie wall stability depends on both the magnitude and direction of the applied field. Secondly, it was observed experimentally that besides the transition from the Néel wall to the Bloch wall, transitions also occur as a function of the applied field. Structural features such as grain boundaries, twins, inclusions, which may take many forms as holes or cracks, etc., had a very strong pinning effect on the domain wall motion, leading to a reduction in permeability.

1. Introduction

Observations of domain walls in thin ferromagnetic films are usually made by various techniques which fall into two main groups: (i) those which disclose domain walls, the Bitter or powder method (using electron microscopy), and (ii) those which disclose domain (optical methods). This involves Kerr or Faraday effects. A new method for the observation of domain walls by the transmission electron microscope was developed by Fuller and co-workers [1–3]. The technique utilizes a standard electron microscope in a deformed mode of operation and is known as Lorentz microscopy. This is because when an electron interacts with a magnetic material it experiences a force known as Lorentz force. When an electron beam passes through a thin magnetized film the beam undergoes a deflection by interaction with the magnetization of the specimen, in addition to random deflections by normal scattering processes. The Lorentz force F on, and the maximum angle of deflection θ of, the electron beam as given by Fuller and Rubeinstein [4] are

$$F = -e\mu_0 V M_s \quad (1)$$

$$\theta = \frac{e\mu_0}{m}\frac{M_s t}{V} \quad (2)$$

where e is the electron charge, V is the electron velocity in the direction of the optical axis (Z-axis), M_s is the saturation magnetization in the plane of the specimen (xy plane) and t is the foil thickness.

The deflection force was assumed to act only during the transit of the electron beam through the thickness of the magnetic material and to be uninfluenced by demagnetizing fields. The later assumption is only valid in simple but important cases but not in general. The expressions hold for the time interval during which the electron is traversing the thickness of the specimen. In deriving Equation 2 the normal scattering mechanisms are neglected.

Equations 1 and 2 are known to give only semi-quantitative agreement with experimental observations because the beam velocity is a function of the beam accelerating voltage. The amount of deflection depends on film thickness and the local magnitude of the component of magnetization perpendicular to the beam direction and the direction of the deflection depends on the direction of the local component of magnetization in the plane of the film. The domain wall is observed because of the direct interaction of the electron beam with the specimen magnetization. The walls are delineated as black or white lines (see Fig. 1). Three types of domain walls can exist in thin films: the Néel, Cross-tie and Bloch walls. The type of domain walls that are more stable at a given thickness depends on the total wall energy, γ, which contains magnetostatic, exchange and anisotropy terms, and also on the direction and magnitude of the applied field. Whereas the width of the observed domain walls in the bulk magnetic material is a compromise between the exchange and the anisotropy energy terms, in thin

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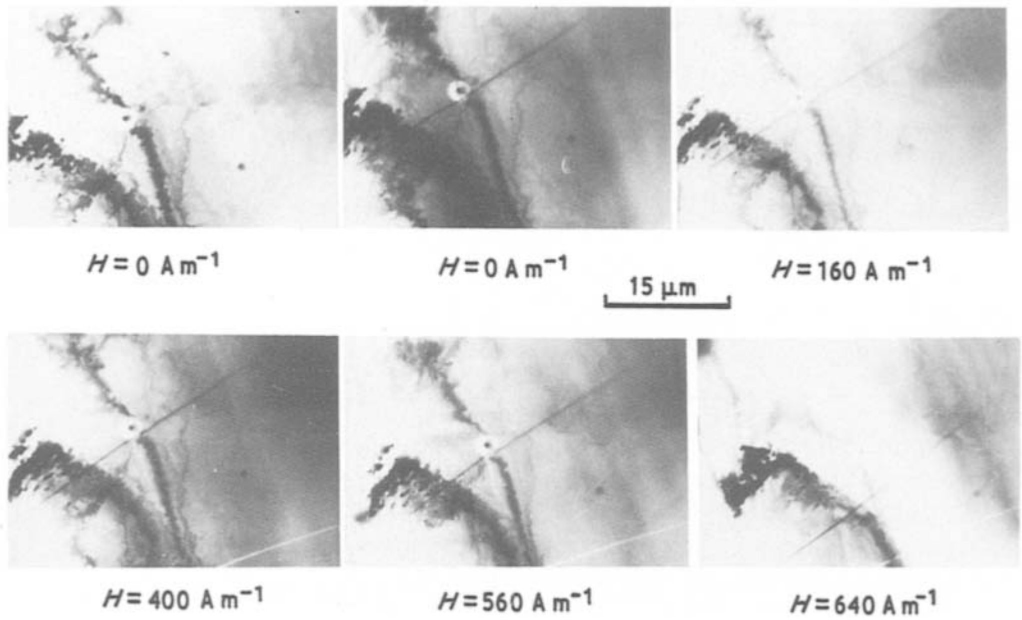

Figure 1 Interaction of black and white walls in a thin ferromagnetic film.

films, the wall width also depends on the film thickness and the angle through which the magnetization turns in the wall by an applied field [5]. Calculations by Soohoo [6], Prutton [7] and Middelhoek [5] give the kind of domain wall which is more stable at a given thickness.

The ease of domain wall movement determines the initial permeability. In a bulk material, the motion of the domain walls under the application of a field is influenced by the magnetic anisotropy and magnetostriction, and microstructural features such as dislocations, grain boundaries, twins, inclusions, etc. [8–10]. These microstructural features can pin domain walls and decrease the permeability.

It is the aim of this work to: (i) investigate the influence of magnetic field on wall mobility and stability, and (ii) investigate the influence of structural features such as grain boundaries, twins, inclusions, etc., on wall motion and permeability.

2. Experimental techniques

Thin foil specimens were used. The thin foils were made from 3 mm discs which were electrochemically polished by a jetting rig. The thin foils were thinned and perforated at the centre.

Studies of domain wall interactions were carried out on the specimens by Lorentz microscopy, or Fresnel phase contrast. This technique was employed because in the normal operating condition of the microscope, the magnetic field of the objective lens is usually so large that the specimen becomes completely extinguished. To prevent this from happening, one or more of the following steps were employed:

(a) the objective lens is switched off and the diffraction lens used as an objective;

(b) the strength of the objective lens is weakened sufficiently so that the specimen is not saturated;

(c) the specimen is raised from the normal position into a region where the objective lens field is less.

The three techniques above were used with the help of two electron microscopes. Each of these microscopes had different electron optical modes of operation.

In the first instance, a Philips EM 200 microscope was used. Its principle of operation was (a), i.e. the objective lens was removed and the diffraction lens used for focusing. A special Lorentz attachment, designed and supplied by the maker, was used in conjunction with the microscope. The attachment consisted of a pair of vacuum-cored coils with which a uniform magnetic field could be applied parallel to the plane of the specimen.

The second electron microscope used was a 1×10^6 V high voltage electron microscope (HVEM), AEI type EM7. Its mode of operation was a combination of (b) and (c). With the help of a specially constructed top entry cartridge specimen holder, it was possible to raise the specimen up to about 10 mm above the objective lens, the objective lens field being reduced to a negligibly small value at the specimen. In order to obtain sharp domain wall images the incident beam was made as parallel as possible by defocusing the condenser system to give effectively a point source a long way from the specimen. In the above operational mode, focusing was carried out by the objective lens and magnification by the intermediate lens. The advantages of the HVEM over the Philips EM 200 microscope are that since the electrons are accelerated by a much higher voltage, they have greater penetrating power and a thicker specimen could be used. Secondly, a higher magnification could be used. Thirdly, the applied external field on the specimen did not affect the electron beam as much as in the lower voltage microscope.

Domain wall images are formed in both cases by deflection of the beam as it passes through the specimen. The amount of deflection depends on both the film thickness and the component of the magnetization perpendicular to the electron beam. The direction of this deflection is a function of the magnetization

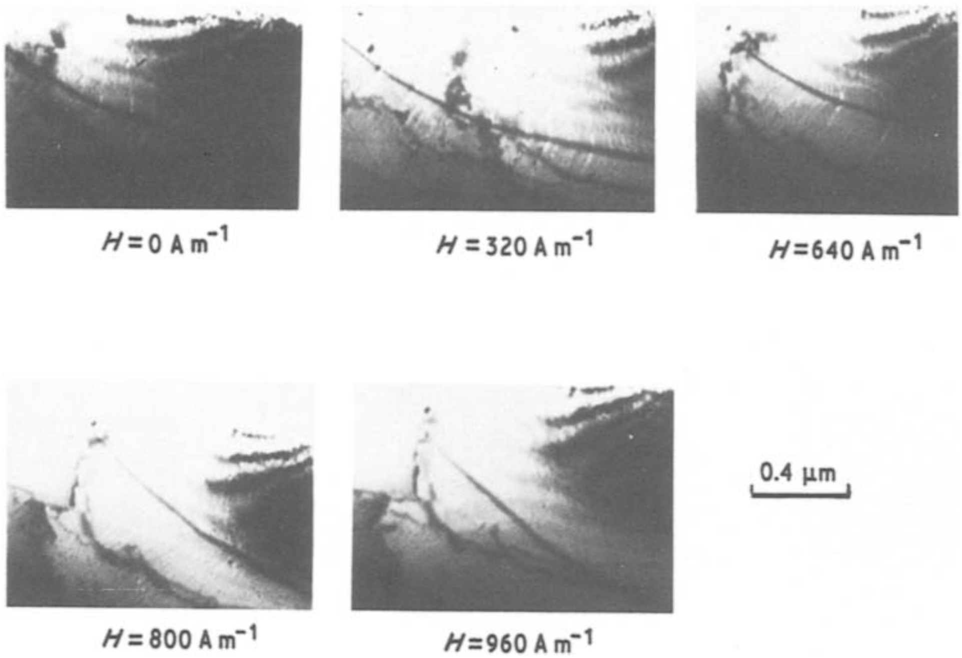

Figure 2 The influence of applied field on domain walls stability.

in the plane of the foil, within each domain. The domain walls are delineated as black or white lines. The deflection of the beam gives rise to overlapping or diverging beams near regions of the specimen where there are changes of direction of magnetization in the plane of the film. This overlap or divergence gives rise to white or black lines when the microscope is slightly out of focus (see Fig. 1). Confirmation that the white or black lines are not due to bend contours is obtained if the contrast of the wall changes when going from underfocus to overfocus (or vice versa) of the objective (or diffraction) lens. As the defocusing is slight and the projector has a large depth of focus the image observed by this method still retains most of the film features.

3. Results and discussion

As stated previously, the technique of Lorentz electron microscopy developed by Fuller *et al.* [1] was used to observe domain walls in thin ferromagnetic alloys. The influence of applied magnetic field and microstructural factors such as dislocations, grain boundaries, twins, inclusion, etc., on the wall mobility and stability were also investigated. Thin foils were used. But since most of the thin foils used had thickness less than 100 nm (i.e. the thickness range normally referred to as thin films) the specimens could be called thin films. In the present work, a window electropolishing technique was used. In order to study the type of domain walls which are stable at any given film (or foil) thickness, the resultant thin foil could be likened to a wedge-shaped specimen, with the thin edge at the hole in the centre of the specimen and the edge of the specimen represents the thicker end of the wedge.

Fig. 2 shows a selection of Lorentz electron micrographs obtained during the investigations and which shows cross-tie walls. From these micrographs one may conclude:

(a) that there are three types of domain walls which can exist in thin films. Near the hole (i.e. in the thinner area) cross-tie walls were not observed, but Néel walls were observed. As can be seen from Fig. 2, the dark domain wall consists of a chain of crosses, the leg of which are Néel walls known as cross-ties in its centre region but these are absent at the thinner area to the left and in the thicker area toward the right. The absence of cross-ties near the edge is also apparent in the dark domain wall;

(b) the separation of the cross-ties decreases, and their length decreases as can be seen in the figure. This confirms the experimental work of Prutton, [7], Fuller *et al.* [1], Middlehoek [5], Huber *et al.* [11] and Rubeinstein and Spain [12];

(c) the cross-ties were more frequently observed in specimens with high initial permeabilities ($\mu_i > 4000$) and so probably appear in areas where the anistropy energy is relatively small. This observation confirms the results of Huber *et al.* [11], Rubeinstein and Spain [12] and Middlehoek [5]. Middelhoek has shown that both the cross-tie length and period are influenced by the anisotropy constant, K_1. He indicated that the cross-tie length, p, is inversely proportional to the anisotropy constant, K_1;

(d) near the edge no cross-ties were observed (see Fig. 2). The wall at this part of the specimen is the stable Bloch wall.

It should be pointed out at this stage that the thickness at which transition from one type of wall to another occurred could not be accurately measured and so this cannot be discussed here.

The effects of a field applied in the film plane on the cross-ties were investigated. The results show that the influence of the applied field on the cross-tie wall stability depends on both the magnitude and direction of the applied field. In Fig. 2, the density of the cross-tie increases initially before they finally vanished under the influence of increasing field. It was

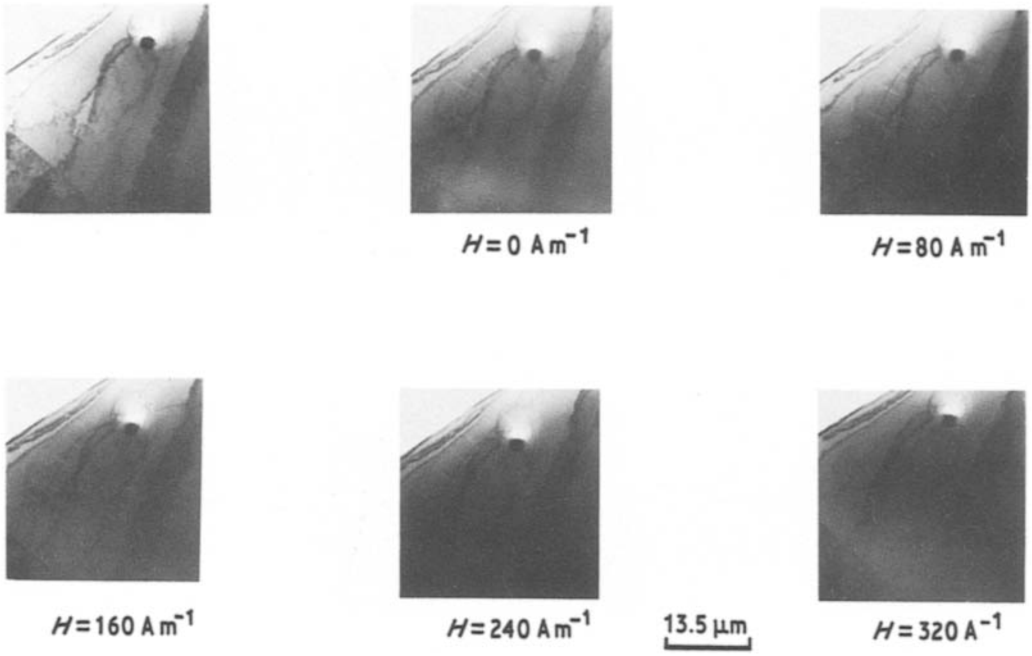

Figure 3 **Pinning effect of domain walls by a particle and a grain boundary.**

also noted in all the specimens that a large fraction of cross-ties failed to reappear on the removal of the field. On the average, application of a reverse field resulted in the return of cross-ties that had been removed. Some interesting cases also emerged in which some simple walls become cross-tie bearing walls in their motion toward an apparently highly stressed region of the thin films. The probable cause of this could be attributed to magnetostrictive effect. It is a function of both the applied field's direction and magnitude. The material could expand when the field is applied in one direction and contract when the field is reversed. The expansion could cause the film thickness to be below the critical value for the stability of cross-ties to disappear. When the material contracts, this can cause both the main wall and magnetization to buckle. If magnetization buckling occurs, then the cross-ties could be formed. Rubeinstein and Spain [12] and Fuller and Rubeinstein [4] have also observed similar reappearance of cross-ties when the reversed field was increased and a magnetization buckling model was also suggested.

As stated previously, removal of the field resulted in a small fraction of the cross-ties reappearing. The new stable wall configuration could probably correspond to the eliminating of some of the magnetization rotations along the wall, since the Néel segments are not observed with maximum applied field.

In order to investigate the influence of microstructural features such as dislocation, inclusions, grain boundaries, twins, etc., on the initial permeability it was decided to investigate the influence of these features on the domain wall motion. Their influence on the wall mobility is an indirect way of relating them to the initial permeability because the ease of domain wall movement determines the initial permeability, because any factor which impedes domain wall motion will lead to a drop in the permeability of the material.

Figs 3 to 7 show a few micrographs for the study of domain wall motion. It can be seen from all micrographs that in the absence of an applied external field the domain walls are curved. This is because the magnetization direction in a ferromagnetic material is influenced by the anisotropy energy, E_k. The anisotropy is such that the magnetization tends to be direct along a certain crystallographic direction and the domain wall tends to be straight. In the permalloy type of alloys the anisotropy energy is very small and the domain boundaries are often curved and lie only approximately parallel to the easy axis. The anisotropy is a function of composition and the degree of short-range order in the material. At a critical degree of order the anisotropy is vanishingly small. Secondly, the domain walls curve because they are under the influence of imperfections and discontinuities in the material.

Fig. 1 shows the pinning effect of a thin circular region on a black wall. The white wall was able to move when the applied field was increased, but the black wall was only able to move at a field of about $640\ \mathrm{A\,m^{-1}}$. When the two walls met on increasing the field, there was no cross over and no more wall movement. This could be due to a repulsion between the black and white walls. Fig. 3 shows the pinning effect of a black wall by a particle, a hole and a grain boundary. One leg of the wall is pinned by the grain boundary, and the other leg is pinned by a hole and particle, and the wall bows between them and its motion is restricted. The cross-ties disappear with increasing field but the black wall is unable to pull away from the pinning sites and it straightens as the field increases.

Fig. 4 shows the pinning effect of two holes which is a form of an inclusion on a white wall. It is an inclusion because in the domain the hole acts as a region with a different magnetization from the surrounding material, or none at all, i.e. the hole is simply a nonmagnetic region in the domain. Closure and parabolic domains are formed around the holes in order to reduce the magnetostatic energy. Negative

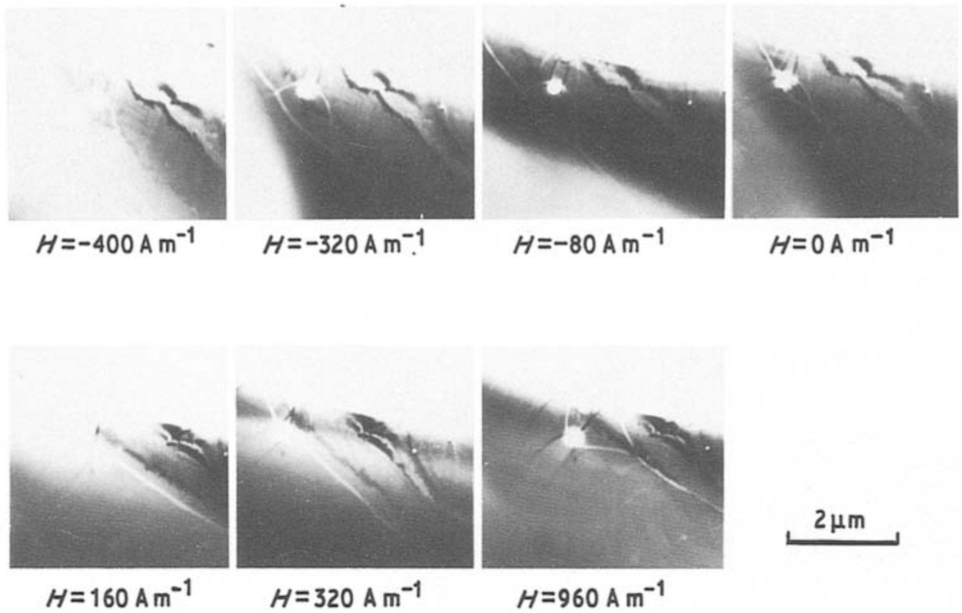

Figure 4 Pinning of domain walls by two holes.

fields caused the pair of black walls joining the holes to expand and the white wall to move to the left drawing out a white closure domain into a parabolic domain whose pinning effect can be seen by the kinks in the main white wall. A field in the opposite direction caused the black walls to contract and the white wall to move to the right. The parabolic wall elongates with increasing field in both direction. Within the limit of field used the motion is reversible.

In Fig. 5 a domain wall is driven across a hole. The white parabolic wall stretches from the hole to the domain wall and there is a black parabolic wall on the opposite side. As the field is increased the wall passes across the hole and the white parabolic domain changes over to the other side of the hole. We can see here that the wall motion is impeded by the inclusion because of the interaction of the moving wall with the parabolic walls normally attached to the inclusion rather than by interaction with the inclusion themselves. The closure and parabolic domains are formed around the inclusion in order to decrease its area, and hence the magnetostatic energy of the wall. The interaction between the main wall and the parabolic wall can be seen from the kinks in the main wall where they join. At $240\,\mathrm{A\,m^{-1}}$ the motion of the parabolic domains and that of the main wall is still reversible at this field, but if the field is continually increased, the parabolic domains do not continue to lengthen indefinitely, because their increasing surface area adds too much wall energy to the system. This assumption is validated because when the field was increased to $320\,\mathrm{A\,m^{-1}}$ the main wall suddenly snapped off the

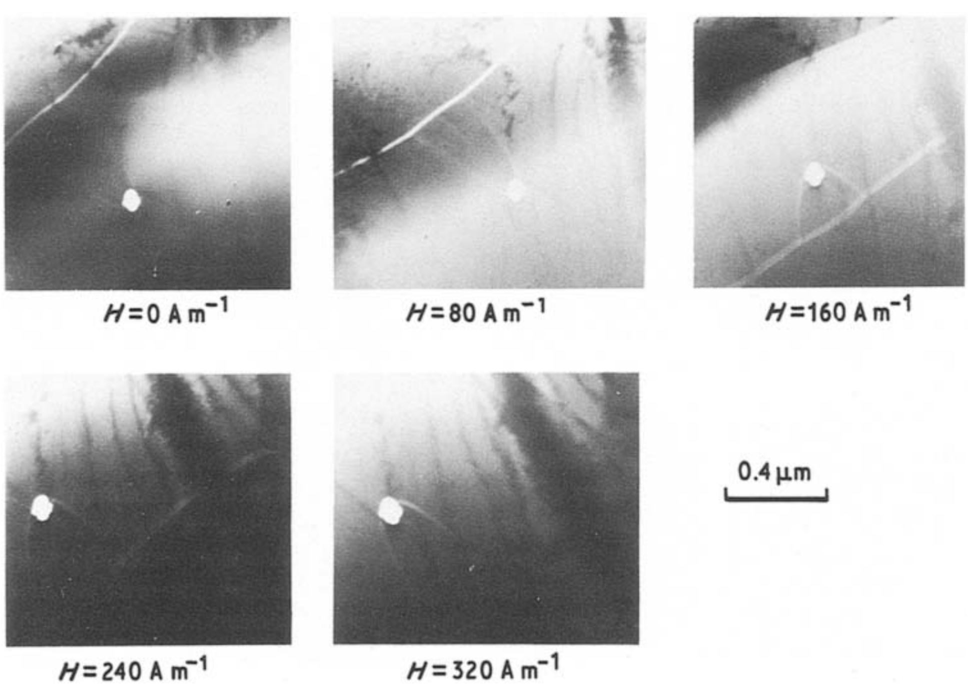

Figure 5 Pinning of domain wall by an inclusion (note the parabolic wall formed around the inclusion in order to reduce magnetostatic energy of the wall).

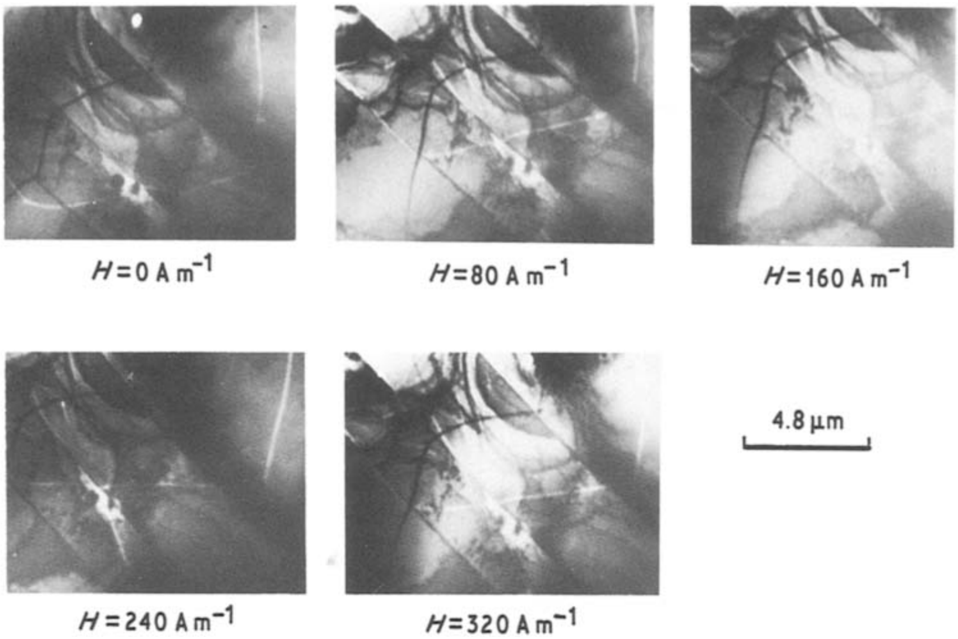

Figure 6 **Influence of twin boundaries on domain motion.**

parabolic domains irreversibly, and jumped away to the right, leaving the parabolic domains strongly attached around the hole. This is Barkhausen jump, i.e. the wall motions are rather jerky and discontinuous.

Figs 6 and 7 show the pinning of a domain wall at twin boundaries. In Fig. 6, the dark wall bends as it crosses the lower twin boundary. On increasing the field the dark wall is strongly pinned where it crosses the three twin boundaries and is only partially free to move in the lower region while the white wall is pulled into coincidence with the lower twin boundary and is strongly pinned there. In Fig. 7 the walls run through a twin and become coincident with the lower twin boundary. The 180° wall forms two 90° walls within the twin but as the field was increased to 400 $\mathrm{A\,m^{-1}}$ the junction of the three walls becomes coincident with the twin boundary leaving a domain wall crossing the twin and a wall coincident with the lower boundary, so that further domain motion could not occur. Since twins are seen to impede wall motion, it is expected that the greater the number of twins, the greater the probability of wall pinning and the smaller the permeability of the material. Conversely, a low twin frequency in a material would be expected to lead to a higher permeability. Consequently, permeability measurements were carried out on the cold-rolled (as-received), recovered and annealed materials. Results of permeability measurements for materials with high dislocation densities (i.e. the cold-rolled materials) show that permeability measurements for such materials were very poor (of the order of 10^2), thus supporting the micrograph results. According to domain theory developed by Chikazumi and Charap [13], the initial permeability is given by

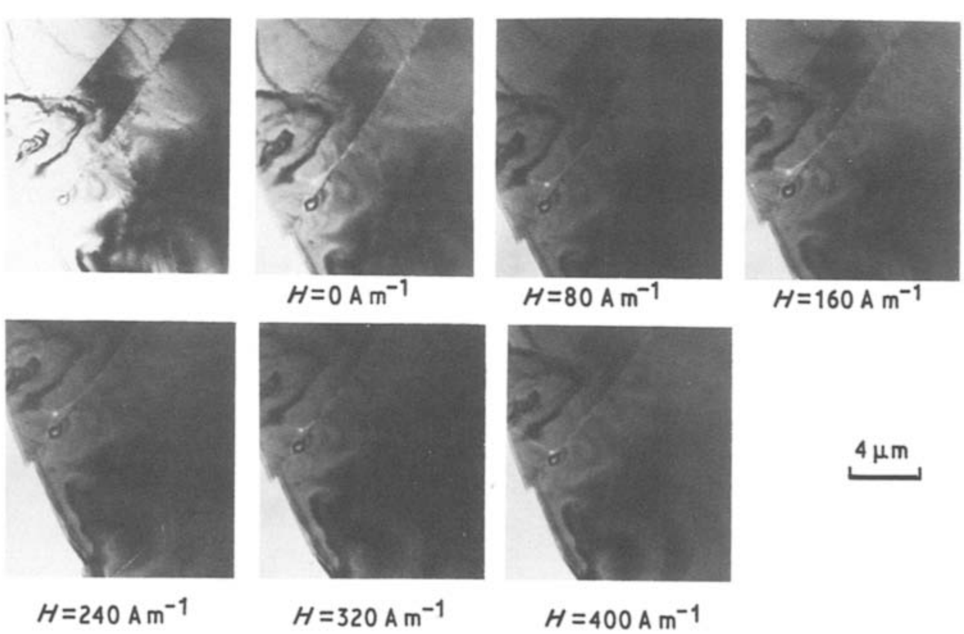

Figure 7 **Pinning of domain walls by twins.**

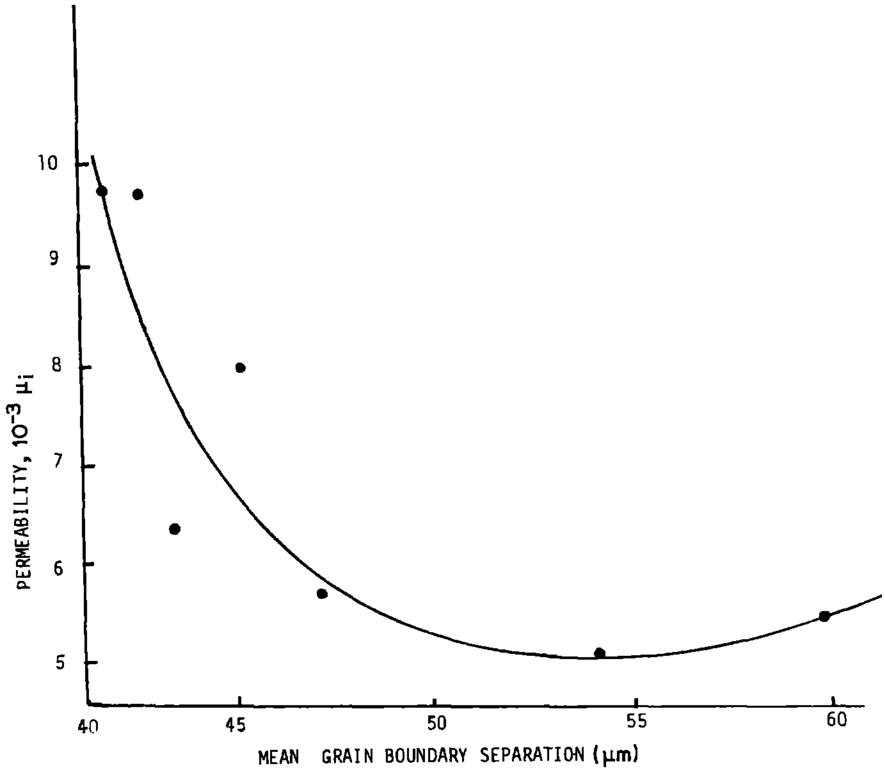

Figure 8 Influence of mean boundary separation on the initial permeability.

$$\mu_i = \frac{2M_s^2}{\pi^2 \lambda_s \sigma \mu_0} \tag{3}$$

if the magnetization occurs by the movement of 180° domain walls and by

$$\mu_i = \frac{CM_s^2}{3\pi \sigma \mu_0} \tag{4}$$

for movement of 90° walls, where $C = 4/\lambda_{100}$ for a material with [1 0 0] as easy direction, and $C = 2/\lambda_{111}$ for a material with [1 1 1] as easy direction, σ is the residual stress in the material, and λ_s is the saturation magnetostriction constants in the 1 0 0 and [1 1 1] directions.

The strong pinning effect of dislocations on the movement of the domain wall which leads to very low permeability in the materials with high dislocation density as a result of high internal stress in the material, as supported by Equations 3 and 4, can now be discussed. The equations indicate that the higher the dislocation density the higher the internal stress and the lower the permeability.

In order to investigate the influence of twins on the permeability, the mean boundary separation as a function of permeability was investigated. It was found that the permeability decreases as the frequency of the twins increases. The relationship between the frequency of twin and the initial permeability was not linear. (see Fig. 8).

The influence of grain boundaries on the initial permeability was investigated, and the results are shown in Fig. 9. It is seen that as the grain size

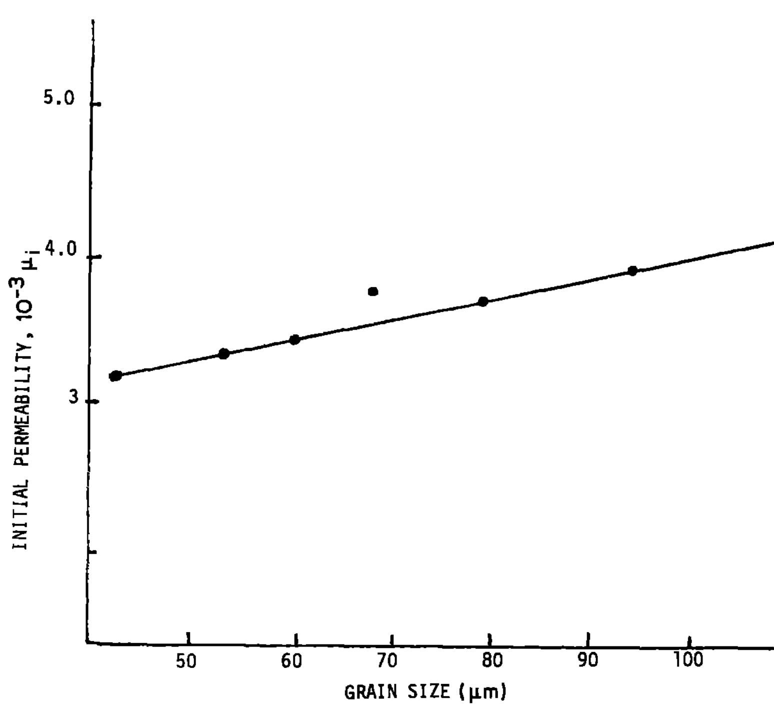

Figure 9 Influence of grain size on the initial permeability.

increases, permeability also increases. The bigger the grains, the fewer grain boundaries in the material and the higher the initial permeability. The permeability measurement results also support the results of the effects of the grain boundaries on domain wall motion, which causes strong pinning of the walls thereby reducing their easy movement and consequently leading to a material of low permeability.

Hence it could be concluded at this stage that any factor such as second-phase particles, inclusions (which introduce stress and strain the lattice of the material) dislocations, grain boundaries, twins, etc., which could impede domain wall motion would lead to smaller permeability.

4. Conclusion

In thin permalloy films the cross-tie walls were noted to occur at intermediate thicknesses and only in materials with high permeability, i.e. materials with low crystal anisotropy, K_i. The stability of the cross-tie walls under the action of an applied field depends on both the magnitude and direction of the field. Transitions of Néel to cross-tie and cross-tie to Bloch walls occurred in addition, as a result of the application of external field. Inclusions, precipitate, and second-phase particles have a strong pinning effect on the domain walls which consequently leads to low permeability in the materials. One reason why an inclusion causes an impediment to wall motion is that the walls tend to cling to the inclusion in order to decrease the area, and hence the wall energy. Dislocations, grain boundaries, and twins are imperfections in the material, that is, they form discontinuities in the material and they also impede domain wall motion by introducing residual microstress in the material.

References

1. H. W. FULLER, M. E. HALE and H. R. RUBEINSTEIN, *J. Appl. Phys.* **30** (1959) 789.
2. H. W. FULLER and M. E. HALE, *ibid.* **31** (1960) 308 S.
3. *Idem, ibid.* **31** (1960) 238.
4. H. W. FULLER and H. RUBEINSTEIN, *ibid.* **30** (1960) 84 S.
5. S. MIDDELHOEK, *ibid.* **34** (1963) 1054.
6. R. F. SOOHOO, "Magnetic Thin Films" (Butterworth, London, 1965).
7. M. PRUTTON, "Thin Ferromagnetic Films" (Butterworth, London, 1964).
8. T. AKOMOLAFE, PhD thesis, Leeds University (1983).
9. B. D. CULLITY, "Introduction to Magnetic Materials" (Addison-Wesley, London, 1972).
10. E. G. TAYLOR, PhD thesis, Leeds University (1977).
11. E. E. HUBER Jr, D. O. SMITH and J. B. GOODENOUGH, *J. Appl. Phys.* **29** (1958) 294.
12. H. RUBEINSTEIN and R. B. SPAIN, *ibid.* **31** (1960) 306 S.
13. S. CHIKAZUMI and S. H. CHARAP, "Physics of Magnetism" (Wiley, London, 1964).

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