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Domain wall energy densities of ferromagnetic films

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DOMAIN WALL ENERGY DENSITIES OF FERROMAGNETIC FILMS

by

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I. INTRODUCTION

A. Thin Magnetic Films Research

In ferromagnetic materials, it has been found that hysteresis phenomenon is exhibited when magnetization is coordinated within a magnetic field relationship. Below a certain temperature, known as the ferromagnetic Curic temperature, these materials show a spontaneous magnetization; above this temperature, however, the spontaneous magnetization vanishes and the material becomes paramagnetic.

Seeking to explain this phenomenon, P. Weiss (21) proposed in 1907, a phenomenological theory of ferromagnetism called the "domain theory." He assumed that a ferromagnetic sample of macroscopic dimensions contains a certain number of small regions (which he called domains) that are spontaneously magnetized. According to the theoretical construction of his theory, this magnetization is constant in both magnitude and direction within each domain. The net magnetization of the total sample can be calculated as the vector sum of the magnetizations of the individual domains.

Experimental support for the domain theory was first given by F. Bitter (2) in 1931. Using a magnetic powder technique which he developed, Bitter was able to observe a visible picture of the domain structure as it existed on the surface of his samples. This new procedure was named the "Bitter magnetic powder technique." It consists of depositing a drop

of a colloidal suspension of very fine magnetite (Fe_3O_4) particles onto the surface of the sample. A thin microscope cover glass is then placed over the droplet in order to spread the colloidal suspension evenly over the surface of the sample. Because the colloidal particles collect in proximity to the magnetic free poles along the magnetic domain walls, the position and outline of the domain walls become visible and the postulation of their existence becomes operationally valid. The suspended particles do not have an intrinsic magnetization high enough to seriously affect the pattern of wall formation. It is necessary, nevertheless, to make the observation quickly following the application of the solution to the sample in order that the surface will remain chemically unaffected by the solution. These domain patterns can be observed by using a microscope with a magnification in the range of 50 to 150 times.

In spite of this experimental confirmation, a really accurate technique for observing domain structure remained problematical for some time. A major difficulty was finding a way to eliminate surface roughness problems which would cause the magnitite particles to collect on the peaks and ridges of the surface, thereby forming spurious Bitter patterns. Mechanical polishing was found to completely disrupt the original domain structure by introducing mechanical stresses at the surface. By 1937, however, W. C. Elmore (8)

had introduced several improvements in the preparation of ferromagnetic colloidal suspensions and in the use of electrolytic polishing technique. These refinements led to relatively true surface domain observations.

Also in question was whether the observed surface domain structure truly represented the domain structure within a bulk sample. It was found that the domain pattern of the entire sample was accurately revealed by observation of the surface domain pattern only when the thickness of the observed sample was very thin. In 1955, M. S. Blois (4) studied the preparation and magnetic properties of thin Ni-Fe films which had been vacuum-deposited to a thickness of about 1000 Å. Because their behavior is of basic interest in computer technology, these thin magnetic films have been extensively investigated since that time. Magnetic thin films, have also been used as variable reactance elements in parametrons to generate the first subharmonic frequency as reported by Read et al. (16).

Perhaps the most important reason for the intense interest aroused by the development of thin magnetic films, however, is its promising application as a memory element in large-scale, high-speed, digital computers. An understanding of the equations and techniques involved in determining the direction of film magnetization has obvious practical consequences for binary computation. Making use of an analogy with the

Stoner-Wohlfarth model (20), it has been found possible to derive an expression for the direction of magnetization θ for a single-domain thin-film of uniaxial anisotropy. The direction is a function of an applied field. Changing the field applied, then, would make binary computation possible.

According to this modification of the Stoner-Wohlfarth model, the total energy of a single domain uniaxial film consists of the sum of the magnetostatic and the anisotropy energies. In order to find the conditions for an energy minimum, the first derivative of the total energy with respect to θ must be zero, where θ is the angle between the magnetization and easy axis of the film. Several values of θ may satisfy this condition. Only those values of θ for which the second partial derivative with respect to θ is positive correspond to energy minimum. Such values of θ are said to be stable angles. Those values of θ for which the second partial derivative with respect to θ is negative correspond to energy maximum and are said to be unstable angles.

Within the framework of this model, the dividing line between stable and unstable occurs when conditions are such that both the first and second partial derivatives with respect to θ are simultaneously equal to zero. A plot of these conditions in terms of externally applied hard axis versus applied easy axis fields gives rise to what is commonly spoken of as the "switching asteriod." The procedure outlined above gives what

is sometimes called the "rotational model of thin magnetic film behavior." Details of this procedure have been given in a number of references (13, 17, 19).

Figure 1 shows the hysteresis loops predicted by this rotational model for the special cases of externally applied fields in the easy direction and in the hard direction. It is seen that the hard axis loop has no hysteresis and that it saturates at a field

$$H_k = \frac{2K}{M}.$$

The easy axis loop is seen to be perfectly rectangular with the field instantly reversing its state of magnetization whenever the easy axis applied field exceeds H_k. In practice, however, it is observed that the film begins to reverse its state of magnetization at a field H_c. In all films for which the rotational model applies, H_c has always been found to be less than H_k. This is because films under the influence of only easy direction fields typically reverse by domain wall motion rather than by coherent rotation. More involved hysteresis loops can be constructed by considering external fields to be simultaneously applied along both the hard and easy axes. Such loops, however, will not be considered here.

H_c is the domain wall coercivity of the film while H_k is defined to be the effective anisotropy field and is sometimes spoken of as the rotational coercivity. Typical values of H_c and H_k for various films may range anywhere from a fraction

of an oersted to several hundred oersteds. Numerous and interrelated factors such as the film thickness, the film composition, the deposition rate, the substrate temperature, and the angle of incidence of the deposition vapor beam all influence the final values of H_c and H_k. Under controlled deposition conditions, however, films of 81% Ni - 19% Fe composition can regularly be deposited to have H_c's and H_k's of only a few oersteds.

A thin film of 81% Ni - 19% Fe permalloy exhibits essentially zero magnetostriction. When such a film is evaporated in the presence of a dc magnetic field uniaxial anisotropy is induced. The magnetization can easily be reversed by an externally applied switching field from one stable state, arbitrarily called a "zero" state, to the other stable state, called a "one" state. Switching fields may be applied in any of a number of directions. The time required for the process of reversing a "zero" state to a "one" state or vice versa takes only a few nanoseconds. This compares favorably with the switching time for electronic circuits and magnetic core elements.

The feature of rapid switching together with its small physical size and binary magnetization characteristics has made the utilization of thin magnetic films a major goal of many computer scientists.

B. Domain Walls

The domain wall is the transition layer between adjacent ferromagnetic domains. It is more favorable energetically for magnetic dipoles in the transition region to gradually change their direction from being parallel to one domain to being parallel to the other domain. The details of the domain wall widths and dipole orientations are determined by the magnetization orientation of the adjacent domains and by the anisotropy and quantum mechanical exchange constants of the film material.

Because of the technological potential of certain types of thin magnetic films, it was and continues to be necessary to investigate these domain structures in detail. In the past, at least two extreme theories have been entertained. Kittel's 1946 work (9) lead some initial investigators to believe that thin films could not contain walls and that, therefore, a thin film must be considered as a single domain. They assumed that crystal and magnetostrictive anisotropy could be neglected in comparison to the magnetostatic and quantum mechanical exchange interactions between the magnetizations of neighboring crystallites. This assumption proved invalid since experimental investigation showed that for "good films" H_c was less than H_k. This implied that the magnetization reversal in the easy direction was due, not to coherent rotation of the magnetization, but simply to domain wall motion.

The other extreme, the assumption that the film consists of many crystallites with no magnetostatic or exchange interaction, was then proposed.

Middelhoek (14, p. 9-1-1) pointed out in 1964 that neither of these extreme assumptions could be correct:

> A film could not be considered as a single domain nor as consisting of a multitude of independent crystallites. A film consists of many crystallites which are in very strong magnetostatic and exchange interaction. These interactions, however, are not yet large enough to completely suppress the anisotropy due to the crystallites.

Bloch (3) was able to show that, as a result of the exchange forces between neighboring dipoles, the walls of the domains are not abrupt discontinuities, but are spread out over a region many atoms thick. The portion of wall energy resulting from quantum mechanical exchange interaction decreases as the width of the region over which the dipole rotation takes place is increased. Inasmuch as the dipoles in the wall are rotated away from a local easy direction, domain walls also involve a certain amount of anisotropy energy. This anisotropy energy increases with domain wall width. The final wall width and energy occur at a width for which the total wall energy is a minimum.

The angle through which the magnetization turns in the wall may be represented by $2\Psi_o$. For a thin film with uniaxial anisotropy, the magnetizations outside the wall always lie in one of the easy directions so that $2\Psi_o = 180^\circ$ so far as the

external field is absent. However, $2\Psi_o$ may be smaller than 180° if external fields are applied or if magnetostatic stray fields are present. Variations in the relationship between Ψ_o and the thickness of the film have been found to produce three basic types of domain walls (Figure 15).

(1) Bloch wall - the magnetization in the wall turns around an axis which is perpendicular to the plane of the wall.

(2) Néel wall - the magnetization in the wall turns around an axis which is perpendicular to the plane of the film.

(3) Cross-tie wall - the main wall is cut at regular intervals by short, right-angle "cross-ties" which terminate in free, single ends (Figure 10).

When this variable relationship involves Ψ_o greater than approximately 75°, a transitional width of about 850 Å appears to demarcate the type of domain wall which is formed: below this transitional film thickness, the formation of a cross-tie wall is most favored; Bloch walls are typically formed above this thickness. For angles of Ψ_o smaller than approximately 75°, the formation of the Néel wall holds regularly more favorable (Figure 3).

C. Domain Wall Energy Densities

Films which possess uniaxial anisotropy form closure and elongated domains (Figure 2) which tend to minimize the total energy of the film. These domains are bounded by 180° walls in which the magnetic dipoles rotate by 180° from one domain to another, and by 90° walls in which the magnetic dipoles

rotate by 90° or so. For films of negligible magnetostriction, simple calculation shows that when K is the uniaxial anisotropy constant of the film and "a" and ℓ are the width of the domain and the width of the strip respectively, as illustrated in Figure 2, the 180° domain wall energy density is:

$$\gamma_{180°} = \frac{Ka^2}{2\ell}.$$

Since $\gamma_{180°}$ for any particular film is constant, "a" should be a simple function of ℓ. However, experiments by Middelhoek (13) and by Kuwahara et al. (10), have shown that "a" increases at a rate much less than the square root of ℓ. The corresponding $\gamma_{180°}$ value based on these experiments was only about one-fifth of that predicted from the theoretical model discussed later in this thesis.

Middelhoek's experiment involved saturating the film in the hard direction by an external dc field. When the field was subsequently turned off, the film split into fine, elongated, oppositely-magnetized domains. When a small ac field was applied, also in the hard direction, 90° closure domains formed. Middelhoek, working on films of anisotropy constant K equal to 2000 erg/cm^2, observed that a = 0.1 mm for ℓ = 1 mm. $\gamma_{180°}$ calculated from these data and the preceding equation have a value of 1.0 erg/cm^2 compared to an expected value of five times this amount.

Kuwahara et al. produced narrow thin films of 80-20 Ni-Fe by evaporation onto a glass substrate through a mask having several slits. The resultant films were between 15 and 300 μ wide and 400 and 1500 Å thick. Closure domains were formed after which $\gamma_{180°}$ was calculated to be 0.8 erg/cm^2. This figure was also too small to compare respectably with the theoretical prediction. Furthermore, Kuwahara's $\gamma_{180°}$ was independent of the film thickness in the 400 Å to 1500 Å range which was not in agreement with Middelhoek's predictions (12).

In the experiments performed by the writer, chemical etching techniques were used to etch the film to various widths from 0.02 mm to 2.0 mm (Figure 6). A 60 cps ac demagnetization field was applied in the easy direction of the film to demagnetize the sample. This experiment produced results in much closer agreement with the theoretical values than those of Middelhoek and Kuwahara et al. (Figure 5). Some deviation between the results obtained from the writer's experiments and the theories of Middelhoek and of Brown and LaBonte (6) can be explained by the factors of wall coercive force, anisotropy dispersion, and the possibility that the theoretical derivations may have been over-simplified.

II. THEORY AND EXPERIMENT

A. Theoretical Derivations

The total energy of a thin magnetic film consists of the sum of several different types of energy including magnetostatic energy, quantum mechanical exchange energy, anisotropy energy, domain wall energy, and magnetostriction energy. The magnetostriction energy can be neglected when the film is of an approximate 81% Ni - 19% Fe composition. When and if the film possesses uniaxial anisotropy and has a sufficiently small dimension along the easy axis (e.g., ℓ in Figure 2), closure domains will be formed which will, in turn, help reduce the magnetostatic energy.

The closure domains are illustrated in Figure 2 along with the longitudinal domains in the easy direction. Because the normal components of the magnetic flux are continuous across the closure domain boundaries, the magnetostatic energy there is zero. On the other hand, the closure domains, being magnetized in the hard direction, introduce anisotropy energy. Thus, for the closure domain configuration, the total energy of the film is equal to the sum of the anisotropy and wall energies. This relationship may be defined as follows:

(1) E_{anis}

E_{anis} is the anisotropy energy of the triangular domains. For these domains the magnetization is in the hard directions.

$$E_{anis} = K \sin^2\theta \times \text{Volume}$$
$$= K \times \frac{1}{2}a \times \frac{a}{2} \times 2T$$
$$= \frac{1}{2} Ka^2T$$

where K = anisotropy constant

θ = the angle between the easy direction and the magnetization, in this instance 90°

T = thickness of the film.

(2) $E_{180°}$

$E_{180°}$ is the 180° wall energy

$E_{180°} = \gamma_{180°} [\ell - a]T$

where $\gamma_{180°}$ = energy density of 180° wall.

(3) $E_{90°}$

$E_{90°}$ is the 90° wall energy

$$E_{90°} = \gamma_{90°} \quad 4\left(\sqrt{\left(\frac{a}{2}\right)^2 + \left(\frac{a}{2}\right)^2}\right)T$$

$$= 2\sqrt{2} \quad \gamma_{90°} \quad aT$$

Hence the total energy is

$$E = \frac{1}{2} Ka^2T + \gamma_{180°} [\ell - a]T + 2\sqrt{2}\, \gamma_{90°}\, aT$$

and the energy density is therefore

$$E' = \frac{1}{2} \frac{Ka}{\ell} + \frac{\gamma_{180°} [\ell - a]}{\ell a} + \gamma_{90°} \frac{2\sqrt{2}}{\ell}.$$

The condition for minimum energy is such that when $\frac{\partial E'}{\partial a} = 0$ then:

$$\gamma_{180°} = \frac{Ka^2}{2\ell}. \qquad (1)$$

As will be shown later, the values of K, a, and ℓ can be experimentally obtained. Therefore, the domain wall energy $\gamma_{180°}$ can be calculated by using the above equation.

For a film in a multidomain state, it is disadvantageous energetically for a magnetic dipole to change its direction abruptly from a particular domain to that of an adjacent domain. A gradual change is favored. For this reason, there always exists a finitely extended transition region (domain wall) between any two domains.

The domain wall energy consists of three parts: the anisotropy energy, the exchange energy, and the magnetostatic stray field energy. The exact calculation of these energies is rather difficult, but approximate methods can be used. Middelhoek (13) has shown that when M_s is the saturated magnetization and D and "a" are, respectively, the thickness and domain wall width of the film, then the domain wall energy density expression for a Bloch wall is

$$\gamma_B = A\left(\frac{\pi}{a}\right)^2 a + \frac{1}{2}aK + \frac{\pi a^2}{a+D} M_s^2$$

and for a Néel wall

$$\gamma_N = A\left(\frac{\pi}{a}\right)^2 a + \frac{1}{2}aK + \frac{\pi aD}{a+D} M_s^2.$$

In both equations above, the first term is the exchange energy density, the second term the anisotropy energy density, and the third term the magnetostatic stray field energy.

The stray field energy decreases as the thickness of the film increases for the Bloch wall. Since the reverse is true for the Néel wall, the expectation arises that domain wall energies are functions of thickness. Curves of the calculated

180° wall energy densities as functions of thickness for the Bloch and Néel walls are shown together with that of the cross-tie wall in Figure 4. The calculation for the energy density of the cross-tie wall is similar to that for the Bloch and Néel walls although considerably more complicated. It may be seen from the figure that cross-tie walls should occur for thicknesses below about 900 Å and that Bloch walls should occur for thicknesses of 900 Å and above. This thickness is called the transition thickness because it denotes a change in the type of wall formation. The theoretically expected domain wall density as a function of film thickness is shown in Figure 5.

With the aid of a digital computer, W. F. Brown and A. E. LaBonte (6) computed domain wall energy densities which were lower than those calculated by Middelhoek. For comparison, the wall energy densities for a Bloch wall as computed both by Middelhoek and Brown and LaBonte are shown in Figure 5.

In Brown and LaBonte's work, the continuous magnetization of a 180° wall was replaced by a discrete distribution for which the magnetostatic energy could be calculated. The general functional form of the direction of magnetization within the wall was not restricted by this replacement and the final minimization of the total energy yielded both the equilibrium wall structure and its corresponding energy.

By referring to Equation 1, it will be seen that since $\gamma_{180°}$ is constant for any particular film, "a" therefore should be a simple function of ℓ. Previous experiments (10, 13), however, had shown that "a" varied with ℓ only slightly, and that the γ value based on these experiments was only about one-fifth of that theoretically predicted.

In the present investigation, a chemical method was used to etch the film into strips of various widths from 0.02 mm to 2.0 mm as illustrated in Figure 6. A decreasing ac field was then used to demagnetize the film. Domain wall configurations were observed with a microscope and photomicrographs taken to aid in the analysis. The dimensions of "a" and ℓ were measured and the energy density γ was calculated. The values of γ thus obtained are shown in Table 1 and are plotted in Figure 5. In contrast to previous experiments, the results show a close similarity to the theoretical predictions of Middelhoek and Brown-LaBonte. The details of the experimental procedure will be defined in the next section.

B. Experimental Procedure

1. Film evaporation

To avoid any accidental contamination of the film, the glass substrates which were used (GRAF APSCO micro cover glasses No. 2; 7/8" square) were carefully precleaned. After about ten minutes of ultrasonic agitation in a detergent

(Alconix) solution followed by several minutes rinse in running water, the substrates were placed in distilled water and given an additional ten minutes of ultrasonic agitation. They were then placed in an alcohol bath for ten minutes. Although a more elaborate cleaning procedure may be employed, the above process usually results in thin films having suitable magnetic characteristics.

These substrates were then placed in a vacuum chamber heated to a temperature of approximately 300° C. Films of 81% Ni - 19% Fe composition were then deposited on these substrates under a vacuum pressure of about 10^{-5} Torr. and a deposition magnetic field of about 80 oersted in the plane of the substrates. The source of the film material was a Ni-Fe wire twisted around a tungsten filament. When the temperature of the tungsten filament was slowly raised, the Ni-Fe wire at first melted and then vaporized as the alloy temperature was further increased.

The vacuum required was such that the mean free path of the evaporated atom was longer than the distance between the source and the substrate upon which the film was to be deposited. Accordingly, the vast majority of the evaporated atoms traveled in straight lines from the source to the substrate without collision with residual gas atoms.

The chemical composition of the film is important for two reasons: first, the principal contribution to uniaxial

anisotropy comes from directed Fe pairs in the Ni matrix and the number of such pairs depends on a factor $c^2(1 - c)^2$, where c is the atom fraction of Fe in the alloy; second, magnetostriction is extremely sensitive to the composition, going through zero and changing sign near the composition 81% Ni - 19% Fe. Eighty-three percent Ni - 17% Fe wire was used to obtain the required 81% Ni - 19% Fe nonmagnetostrictive permalloy films. Since the boiling points of Ni and Fe are different, the vapor pressures are also different. This in turn leads to the variance in the composition between the melted source and the deposit.

The thickness of the film was initially estimated prior to the evaporation by weighing the amount of Ni-Fe wire and measuring the distance between the substrates and the tungsten filament. The measured distance was about 13 cm. The thickness of the film after deposition was measured by magnetic comparison with a permalloy film of a known thickness. This measuring method will be described later.

2. Film etching

KPR (Kodak Photo Resist) was evenly applied to a Ni-Fe film and after drying exposed to an ultraviolet light for one and one-half minutes through a "Photo negative mask." (This mask, as required by the experiment, included several strips of various widths from 0.02 mm to 2.0 mm.) The exposed film was then developed in KPR developing solution for two minutes.

It was rinsed in cold water for about one minute and then dried. The exposure to light polymerized the photoresist thereby providing a protective coating over that portion of the film which was not to be etched away. The developing process removed the unpolymerized photoresist exposing the portion of the film to be etched. The film was then etched by dipping it into a small beaker of ferric chloride solution. The etching process itself takes only a few seconds. Excess ferric chloride was removed by rinsing the film in cold water for about two minutes. The exposed KPR was then peeled off, leaving the film with the etched pattern desired. Occasionally, it was necessary to spray acetone on the film to aid in peeling off the exposed KPR. For the purpose of this investigation, a well-etched film would have the geometry shown in Figure 6.

3. Domain patterns observation

A drop of Bitter solution was applied to a well-etched film and a microscope slide cover placed over the surface. After the earth's magnetic field had been compensated for by a horseshoe magnet, the film was placed under the microscope. A 60 cps ac demagnetizing field (discussed in the following section) was then applied. Closure domain formations could be observed. With a special adaptor which permitted a 35 mm camera to be fitted to the microscope, photographs were taken before the Bitter solution dried. Although occasionally a magnification of up to 200 times was used for the observation

of domain configurations, a magnification of between 40 and 100 times was generally used for photographing. Since a large number of domains in a single picture are required to obtain a satisfactory average of the physical dimensions of a domain, a low magnification was preferable. Photographs of stage micrometers were also taken for use in establishing a scale for measuring the dimensions of domains and strip widths. Domain pictures were taken for film thickness ranging from 200 Å to 2700 Å, and for strip widths of 0.02 mm to 2.0 mm for each film thickness.

Domain pattern observation for the 100 Å range was not successful in the current investigation. No regular 180° or 90° domain walls seem to have formed although some features resembling wall structures appeared randomly in the strips. The first few atomic layers of film deposited on the substrate would be expected to conform to the contours and imperfections of the substrate. Thus, the properties of a film only 100 Å in thickness may well be determined by the film's sensitivity to the presence of scratches and other domain nucleation centers on the substrate.

Since free poles are established at the intersection of the domain wall and the sample surface, the magnetostatic fields associated with these free poles attract the magnetic particles in the colloidal suspension thereby allowing the domain wall positions to be observed. It was found that for

strip widths of about 0.05 mm and above, Lifshitz domains occurred. The Lifshitz domains are believed to represent for wide strips a lower energy state than a closure domain (11). These will be discussed further in the section of this paper dealing with summary and conclusions (Chapter III).

For strip widths below 0.05 mm, closure domains were regularly observed. Typical examples of closure domains are shown in Figure 11. There was little difficulty in recognizing closure domains since they are easily identified. The peculiar shape of the cross-tie wall made it readily recognizable. There was little difficulty in distinguishing between Bloch and Néel wall types due to the great difference in their respective visibilities. The visibility of the Néel wall in the photographs was much higher than that of the Bloch wall. The cause for this difference apparently rests with the difference in field direction above the wall even though the magnetic moments and the magnitude of the stray fields above the film plane do not greatly vary for the two types.

In making these identifications, a notable distinction between regular 90° closure domains and edge domains in a remanent state was utilized. In contrast with the 90° closure domains of Figure 2, the edge domains of the remanent state shown in Figure 12(a) are, by and large, 180° reversed domains. These edge domains grow inward toward the interior of the strip when a magnetic field H which is slightly larger

than the coercive force H_c is applied in the direction opposite to that of the magnetization of the main body of the film (see Figures 7 and 13). This mode of growth when the magnetization is reversed indicates rather clearly that these edge domains are, in effect, 180° domains reversed and orientated along the easy axis, not 90° closure domains as might have been thought at first glance.

With these distinctions relative to different domain wall types in mind, it was observed in this investigation that the films having a thickness of less than 900 Å tended to have cross-tie walls as their 180° walls and Néel walls as their 90° walls. Films having a thickness greater than 900 Å tended to have Bloch walls as their 180° walls and Néel walls as their 90° walls. These results are in agreement with the theoretical results presented in Figures 3 and 5.

Although the maximum value of the earth's field in the plane of the film was only about 0.5 oersted, the domain patterns were greatly influenced by its presence. The domains in the earth's field direction increased, while those in the opposite direction decreased. Figure 12 illustrates this phenomenon.

The anisotropy constant of each film was measured before chemical etching. Once the dimensions of "a" and ℓ were known from the measurements of the microscope photographs, the domain wall energy density γ was calculated by using Equation

1. All the measured data are shown in Table 1. In Figure 5, experimental γ is plotted as a function of thickness. In this figure, the similarity between the theoretical expectation and the experimental results of the present procedure is apparent.

4. Demagnetization methods

A pair of rectangular coils was designed (Appendix B) for an ac field demagnetization procedure. To assure uniformity in the magnetic field applied to the film, this pair of coils was designed so that the inhomogeneity of the field would not exceed 0.7% on any part of the film.

Three methods of demagnetization were investigated:

(1) The first method of demagnetization consisted in the application of a dc field of 15 oersted in the hard direction for three minutes. Immediately after the dc field was turned off, an ac field of one oersted was applied in the same direction and left on during the observation.

The dc field created the single domain state aligned parallel to the hard direction. When the dc field was removed, the film split into a multidomain magnetic state with adjacent domains being antiparallel to each other, i.e., they were 180° domains. In all strips with widths in the range between 0.02 mm and 2.0 mm (Figure 9) closure domains were observed.

(2) In the second demagnetization procedure, an ac field of approximately 25 oersted was applied in the hard direction

of the film for a full minute; during the following minute, this field was gradually reduced to zero. Closure domains were again formed in the narrow strips (Figure 10), while regular and irregular Lifshitz domains were observed in the wider strips.

(3) The third demagnetization procedure was the same as 2 except that the ac field was applied in the easy direction instead of the hard direction of the film. The domain configurations observed (Figure 11) were the same as 2, but the domain wall width "a" was considerably larger than that obtained from 2.

In all three cases, regular closure domains were obtained in the strip width range between 0.02 mm and 0.5 mm. The domain width in Case 3, however, was considerably larger than that of 1 or 2. Thus, the domain wall energy densities γ calculated from Equation 1 are quite different in each of the three cases. In Cases 1 and 2, γ was less than 1 erg/cm^2, while in Case 3 γ's range from 1 to 9 erg/cm^2 depending upon the thickness of the film. In addition, domain width a varied with strip width ℓ only slightly in Cases 1 and 2, while considerable variation occurred in Case 3.

The results obtained from the method of demagnetization procedurally outlined under Case 3 seemed close enough to the theory to confirm it. The data in Table 1 and the experimental

wall density curve in Figure 5 are based upon this latter method of demagnetization.

5. Film thickness measurement

Although in this experiment an estimated thickness of the film was typically projected prior to its deposition, a more reliable value was obtained by measurement after the film was deposited. This involved a magnetic comparison of the film thickness with a permalloy film of a known thickness.

The thickness of a circular permalloy film to be used as a standard was calculated first. If Φ represents the total magnetic flux, B the flux density, M the magnetization, and A the cross section area at the diameter D of the film, then $\Phi = BA = 4\pi MDT$, all values appearing as cgs units. Since Φ could be measured by a flux meter and M = 800 gauss for an 81% Ni - 19% Fe permalloy, the thickness T could thus be determined as soon as the diameter D was measured.

The magnetic flux Φ is proportional to DT. Therefore, for any two films having the same diameter D, the thickness T will be directly proportional to Φ. This, then, provides a convenient method of measuring the thickness of magnetic films. The magnetic fluxes of two films can be easily compared on the screen of an oscilloscope by measuring the saturated magnetization of their easy axis hysteresis loops.

Errors are easily introduced by using the above equation when the two film diameters are different. Due to the

magnetic flux distribution which is different for different diameters of a film, it is preferable to compare films of the same diameter even though films of different diameters can be used. In the present experiment, films whose thickness was to be measured were mechanically cut to the same size as the film used as a standard.

6. Measurement of the anisotropy constant of a film

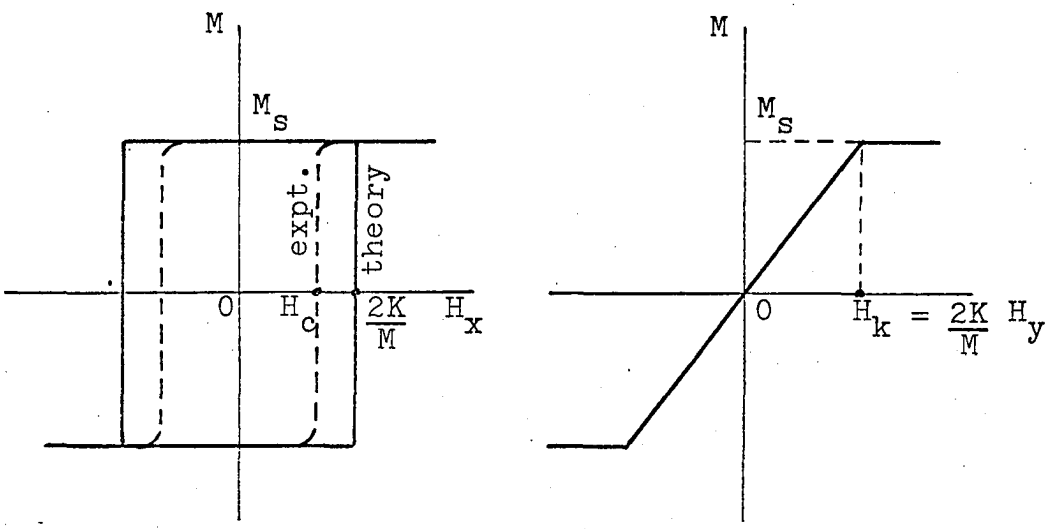
The hysteresis loop of a thin film in the hard direction (Figure 1) is, according to the single domain model, a straight line without hysteresis. It saturates at

$$H_k = \frac{2K}{M}.$$

H_k is the anisotropy field and can be conveniently measured by a hysteresis loop tracer; M is the saturation magnetization and is equal to 800 gauss for 81% Ni - 19% Fe permalloy film. Thus, from this relationship, the anisotropy constant K of the film can easily be determined.

Figure 1. Hysteresis loops of thin magnetic films

Figure 2. Closure domain configuration



(a) Easy-axis loop (b) Hard-axis loop

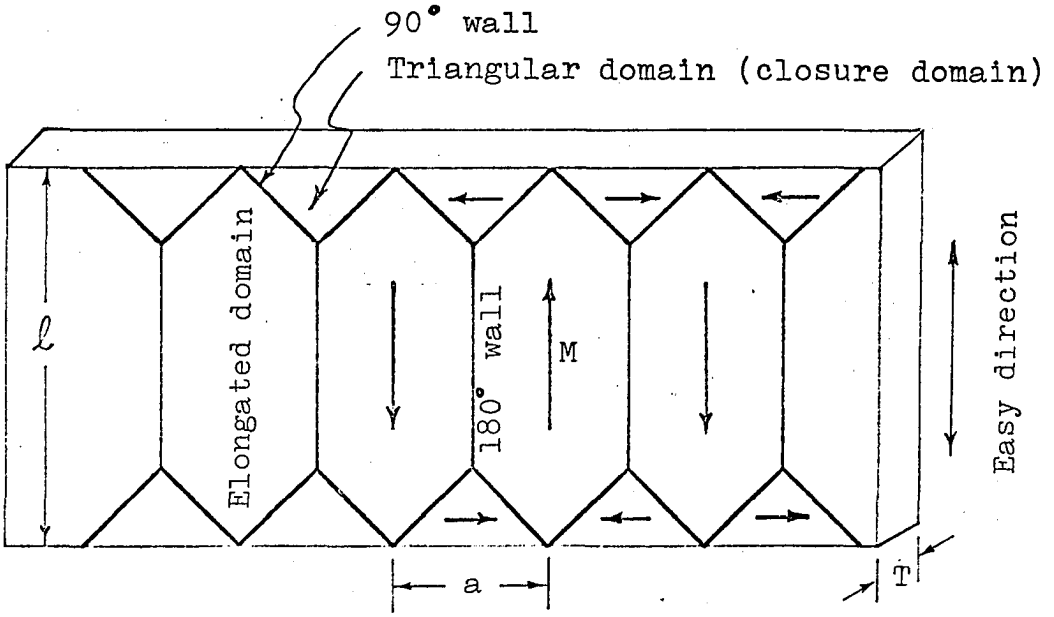
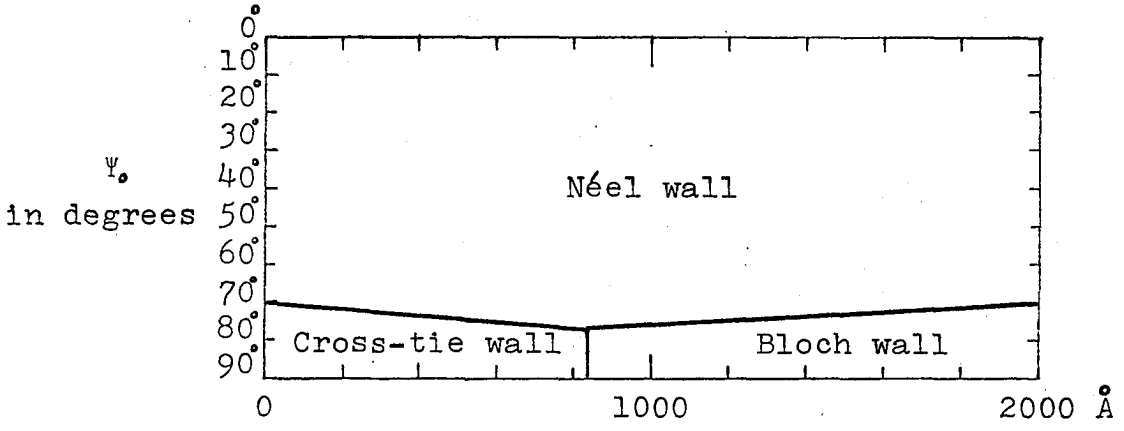


Figure 3. Diagram which shows which type of wall is stable as a function of the thickness T and the angle Ψ_o. $2\Psi_o$ is defined as the angle through which the magnetization turns in the wall

Figure 4. Calculated Bloch, Néel and cross-tie wall energy densities [exchange constant $A = 10^{-6}$ erg/cm saturation magnetization $M_s = 800$ gauss]. After Middelhoek


Ψ_o
in degrees
0°
10°
20°
30°
40°
50°
60°
70°
80°
90°
Néel wall
Cross-tie wall
Bloch wall
0
1000
2000 Å
film thickness T

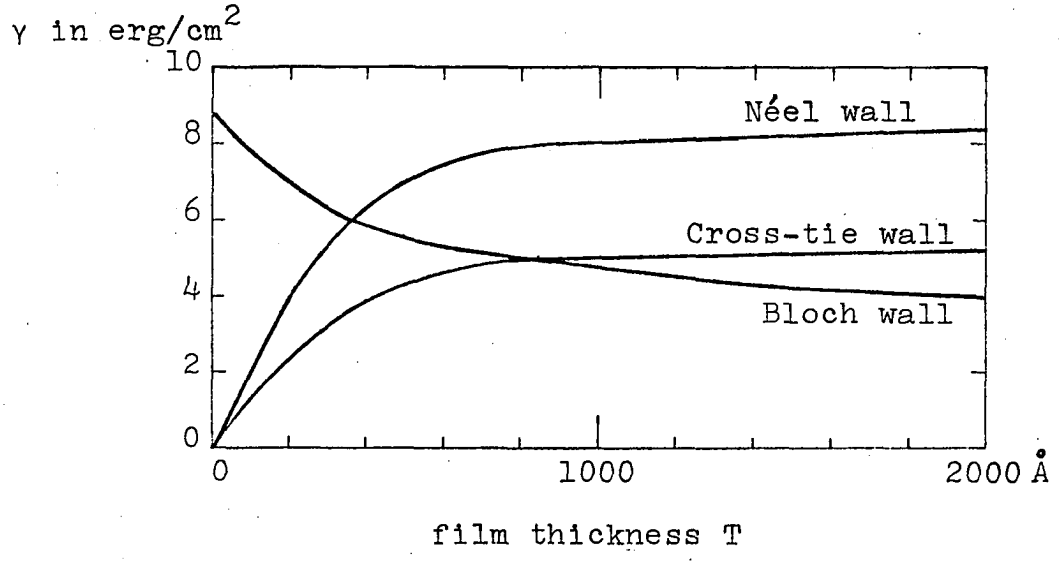
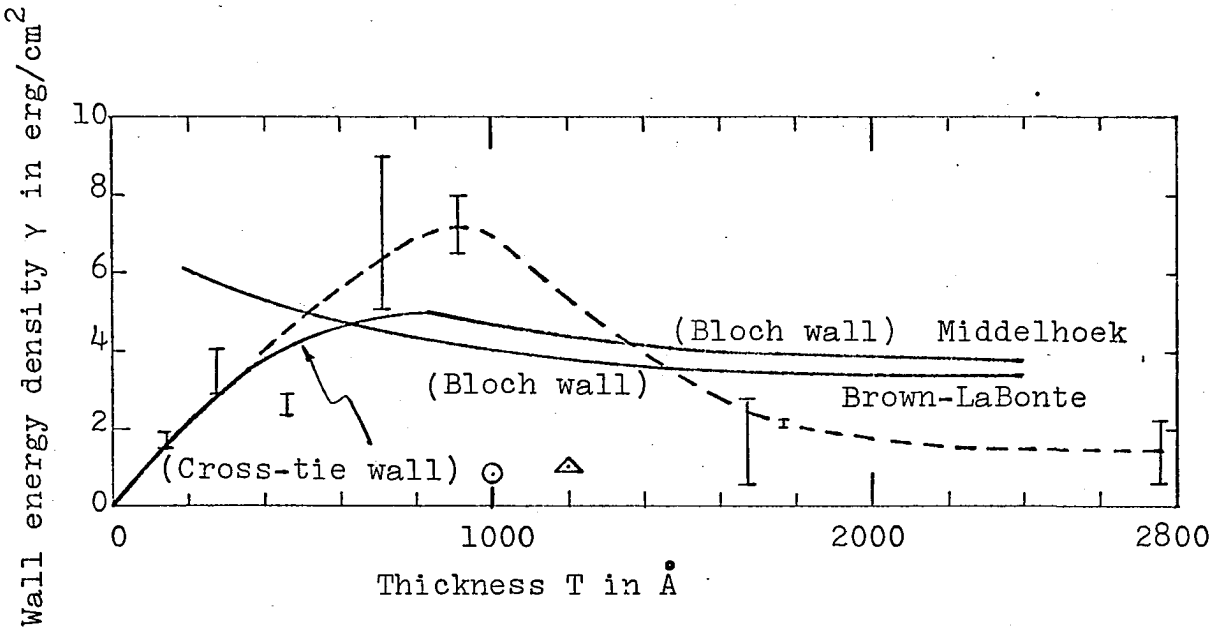

γ in erg/cm^2
10
8
6
4
2
0
Néel wall
Cross-tie wall
Bloch wall
0
1000
2000 Å
film thickness T

Figure 5. Comparison of experiment with theoretical derivations



△ Experiment result after Middelhoek

⊙ Experiment result after Kuwahara et al.

——— Theory

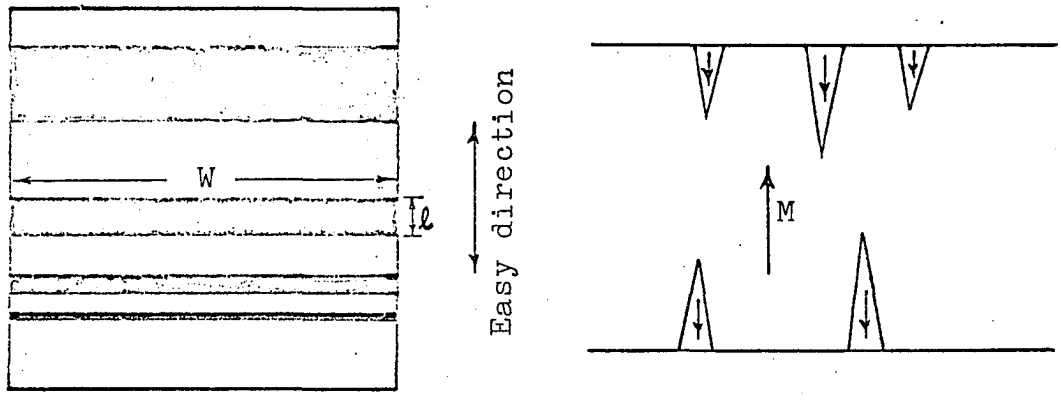
------ Experiment: Bloch walls when T > 922 Å

Cross-tie walls when T ≤ 922 Å

Figure 6. Well-etched Ni-Fe film

Figure 7. Schematic diagram of edge domains

Figure 8. Schematic diagram of Lifshitz domain structure


W
l
Easy direction
M

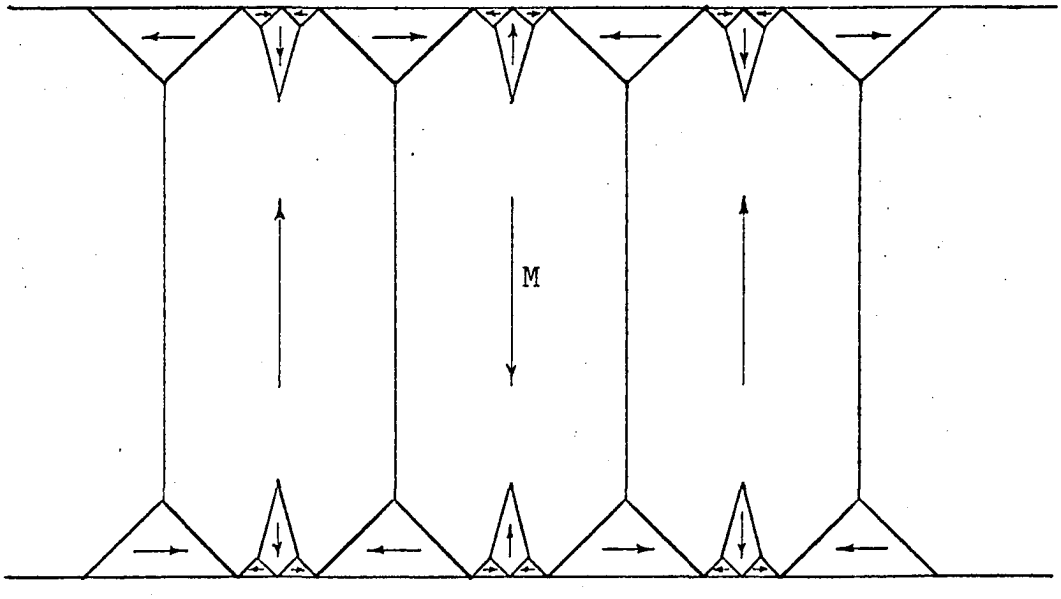
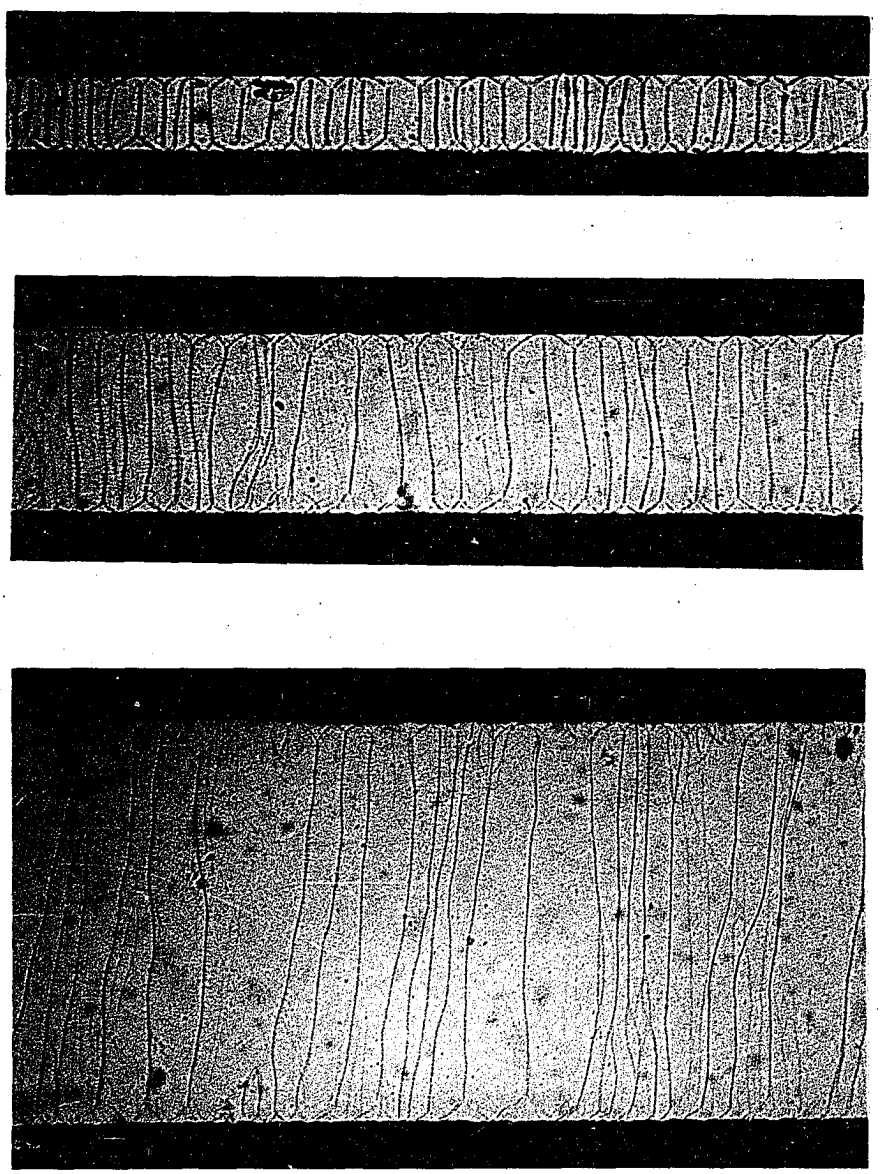
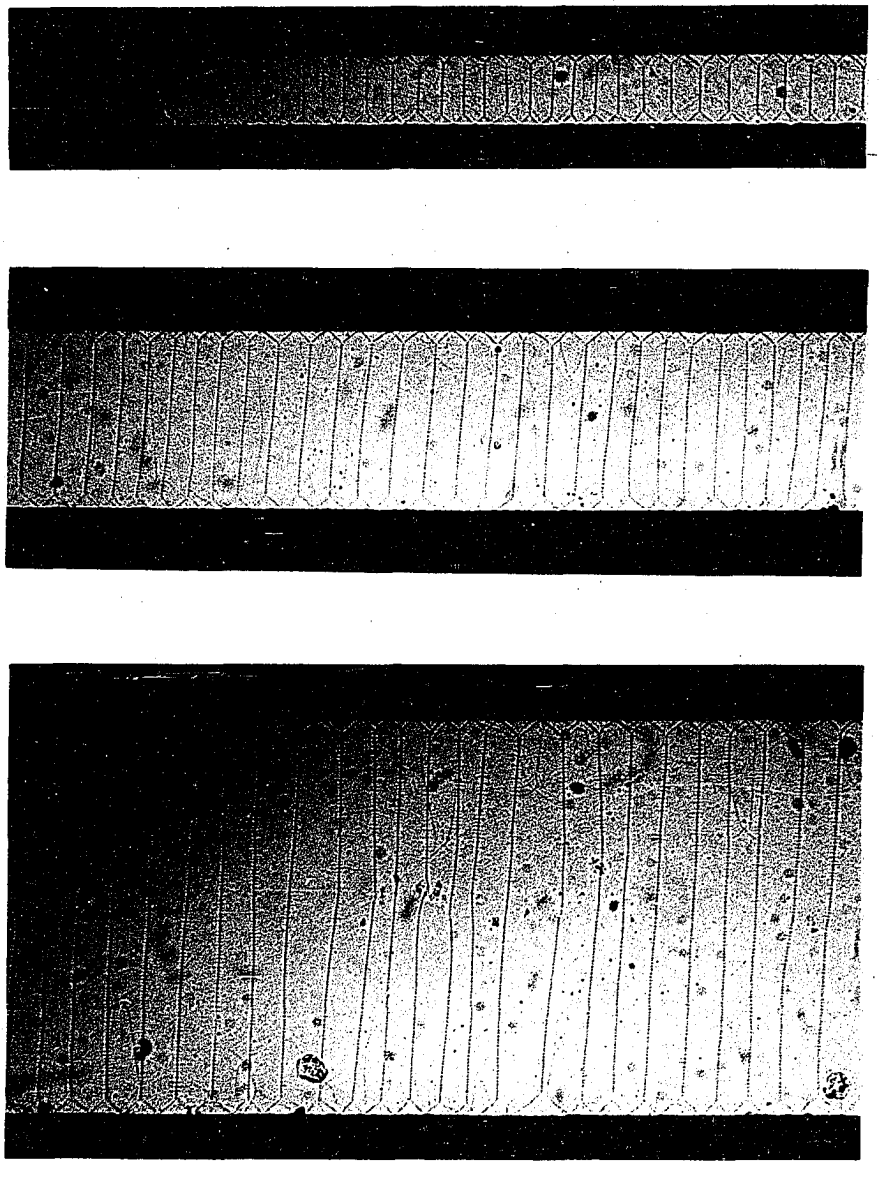

M

Figure 9. Closure domain pattern of 81% Ni - 19% Fe film dc field of 15 oersted is applied to the hard direction for 3 minutes. Then an ac field is applied in the hard direction immediately after the dc field is turned off. The ac field here remains on. (Sample #5-4-10 thickness 720 Å strip widths about 0.1, 0.2 and 0.5 mm respectively.)



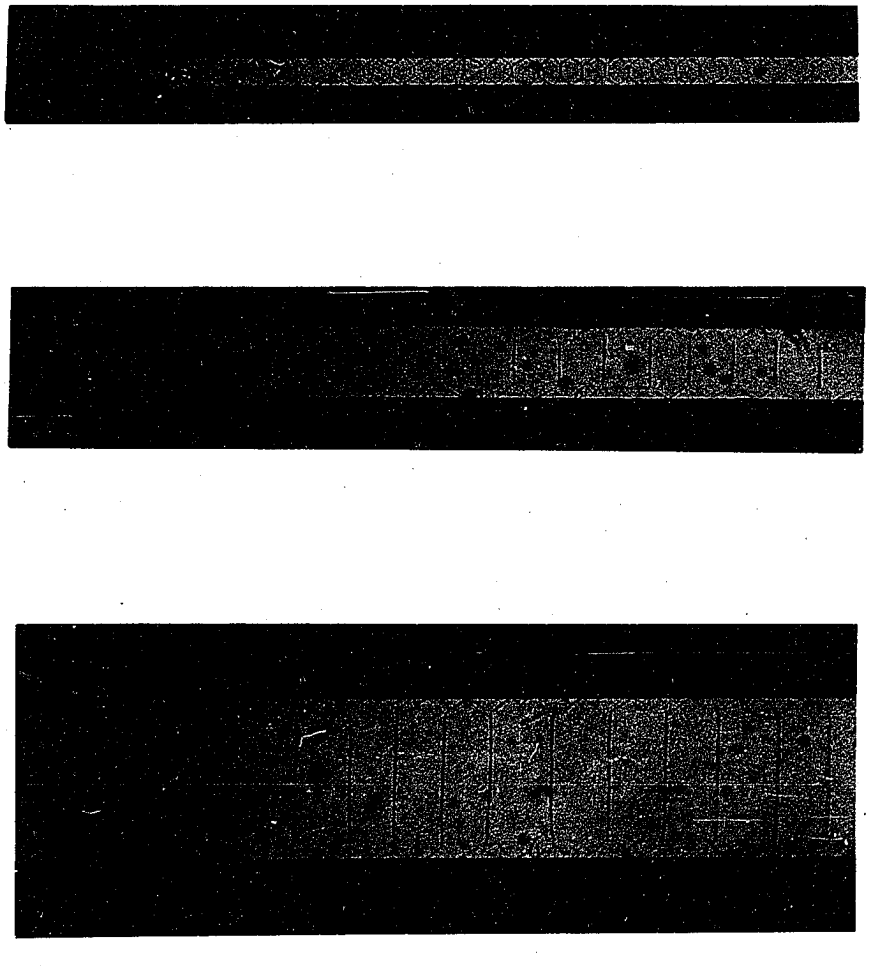
0 0.1 mm

Figure 10. Closure domain pattern of 81% Ni - 19% Fe film. ac field of 25 oersted is applied in the hard direction for 1 minute. Then the field is gradually reduced to zero within 1 minute. (Sample #5-4-9 Thickness 720 Å Strip widths about 0.1, 0.2 and 0.5 mm respectively)



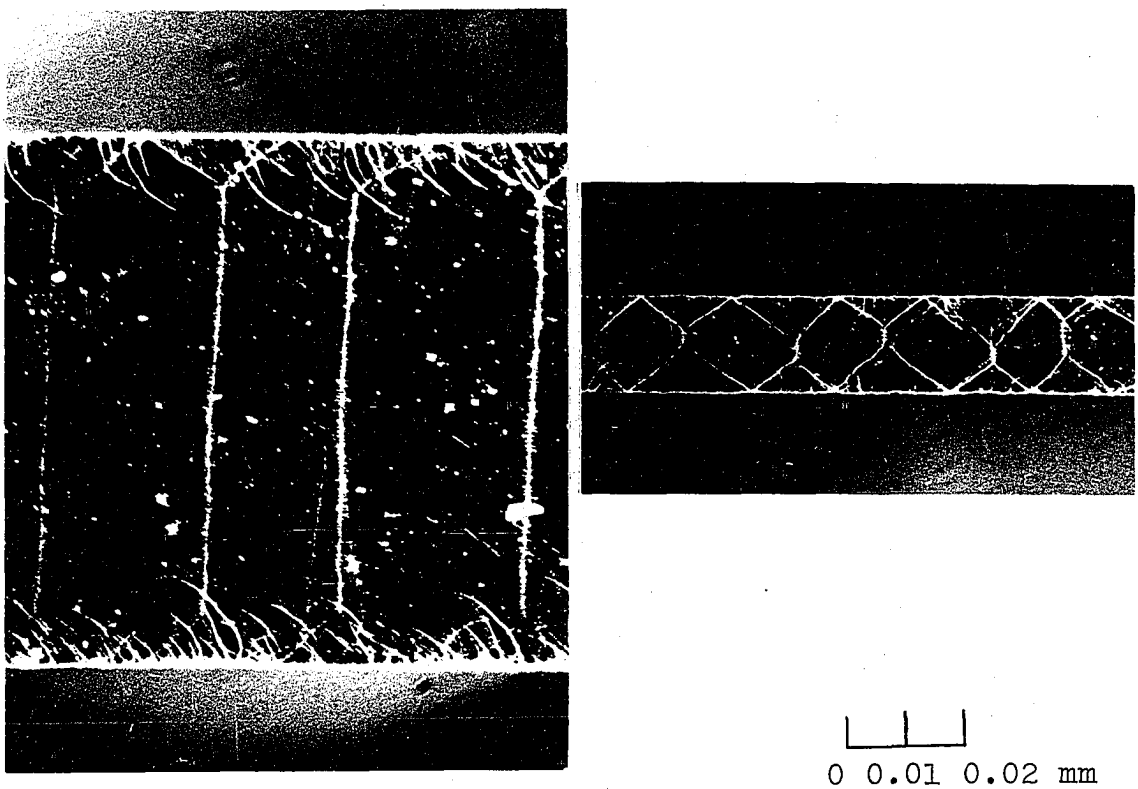
0 0.1 mm

Figure 11. Closure domain pattern of 81% Ni - 19% Fe film ac field of 25 oersted is applied in the easy direction for 1 minute. Then the field is gradually reduced to zero within 1 minute. (Sample #5-4 Thickness 720 Å Strip widths about 0.1, 0.2 and 0.5 mm respectively)



0 0.1 0.2 mm

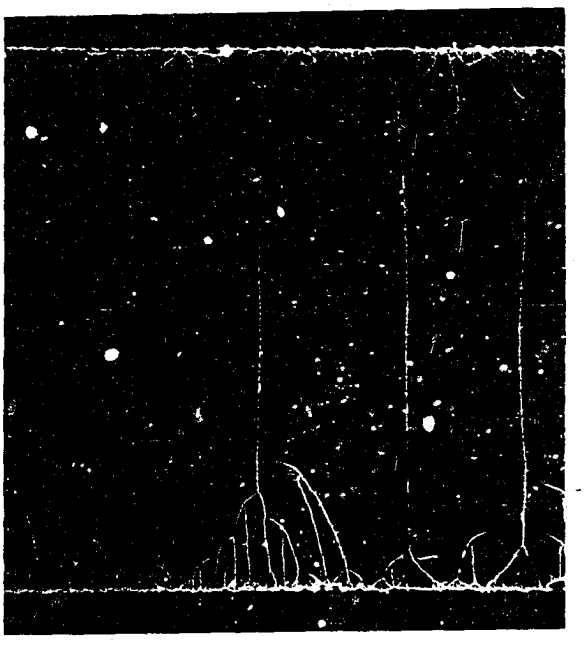
Figure 12. Domain pattern rearranged due to the earth magnetic field. Domains whose magnetization is the same as the earth field increase their size while those opposite to the earth field decrease. (a) Strip width about 0.1 mm (b) Strip width about .02 mm. In (a), edge domains can be seen



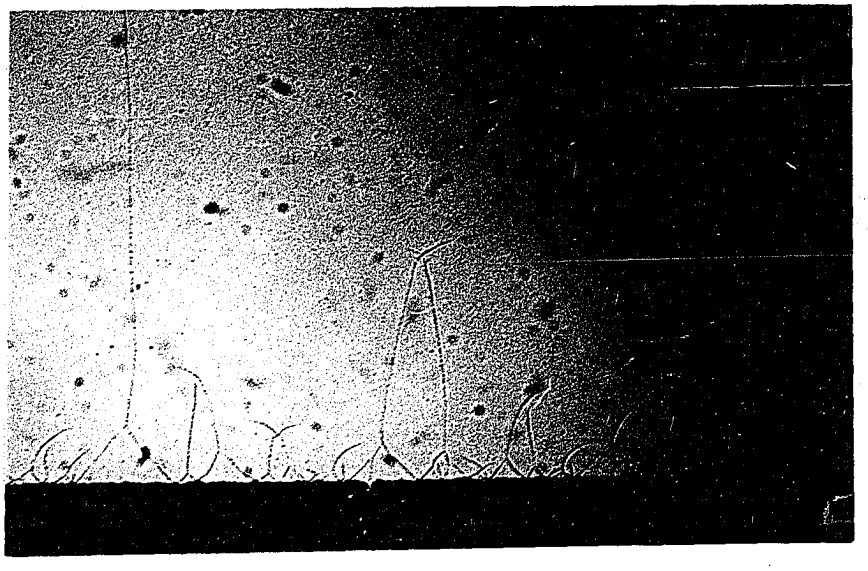
(a) (b)

Figure 13. 90° domains, Lifshitz domains and edge domains. Film thickness about 1000 Å

Figure 14. Several Lifshitz domains between 90° domains for the wide strips. Only the portion near film edges is shown. Film thickness 922 Å Strip width about 2.0 mm

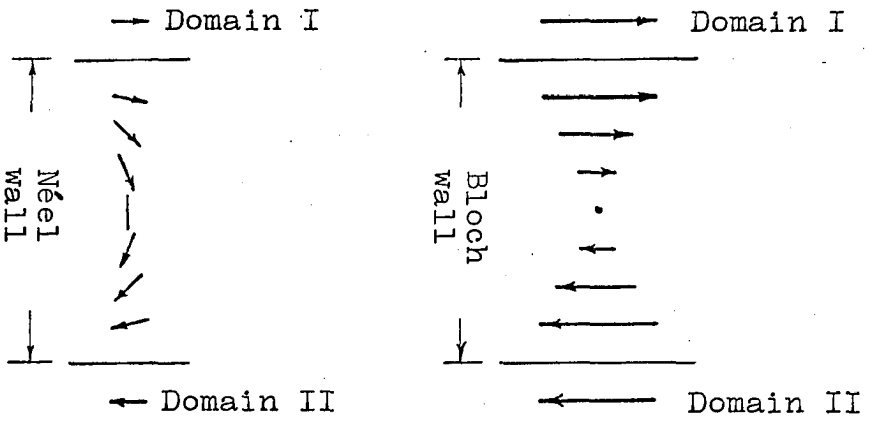


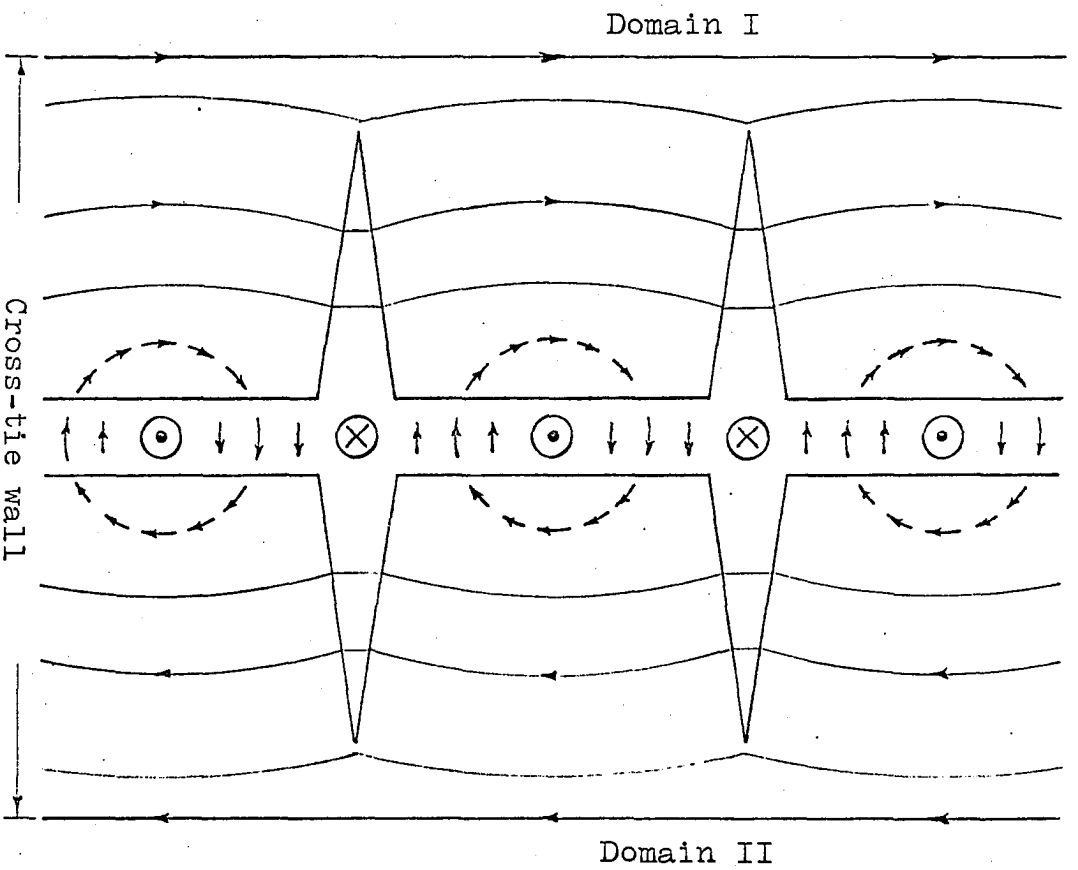
0 0.01 0.02 mm



0 0.1 mm

Figure 15. Schematic diagrams of different types of domain wall


Domain I
Néel wall
Domain II
Domain I
Bloch wall
Domain II


Domain I
Cross-tie wall
Domain II

III. SUMMARY AND CONCLUSIONS

A. Agreement Between Theory and This Investigation

The results presented in the previous sections tend to verify the theoretical approach taken by Middelhoek and Brown-LaBonte. The success of the present investigation lies primarily in the introduction of two factors, viz., the use of chemical etching and the use of an improved demagnetization technique.

The use of chemical etching resulted in the film strips having sharp edges without the introduction of a change in the easy axes directions near the edges of the strip. When evaporation onto a glass substrate through masking having slits is used to produce narrow strip films, the films thus formed have edges whose thickness, in general, is changed gradually rather than abruptly. In addition, the demagnetizing effect which builds up during the evaporation process introduces easy axis dispersion. These effects greatly influence the formation of the closure domains, particularly in the case of narrow strips. Due to the geometric shape of the film, easy axis dispersion tends to occur near the film edges in films deposited through a slit. At different parts of the film, then, the directions of the local easy axes can be different as illustrated in Figure 17(a).

If the film strip is formed by chemical etching, however, the local easy axes within the film will be essentially

parallel as illustrated in Figure 17(b). For film strips formed by evaporation through a slit, the directions of the local easy axes are different along the edge. Thus, when a triangular domain is formed, the anisotropy energies stored in each domain along the edge of the film strip are different. When the film strip is formed by the method of chemical etching, more uniform domains are produced making the anisotropy energies stored within each triangular domain the same. These sharper edges and the more uniform domain structure produce more reliable results.

The demagnetization method used in this study led to a more true minimum energy state of the film than previous methods. In Figure 16, a magnetization distribution in a closure domain configuration is proposed. Because of the coercive force of the film, the magnetization vectors in neighboring elongated domains might not be antiparallel to one another even after an ac or a dc field is applied in the hard direction as outlined earlier in the discussion of demagnetizing methods one and two. Instead, they may form small angles in relation to the average easy axis direction. Thus, a large anisotropy energy would be present even for a very small angular displacement δ. The situation created is entirely different, however, when an ac field is applied in the easy direction. This field helps to align the magnetization along the easy direction thereby reducing the angle δ which,

in turn, reduces the anisotropy energy. It is therefore logical to assume that when the demagnetizing field is applied in the easy direction rather than the hard direction, a nearly true minimum energy state of the film is reached.

The external applied field's contribution to the change in anisotropy energy of closure domains is rather small since small angle change in the closure domains will have little influence on the total anisotropy energy. This is particularly true for wide strips because the total area of the triangular domains is small compared to the total area of the elongated domains.

B. Explanation of the Deviation Between the Experimental and Theoretical Values

Examination of Figure 5 indicates that some deviation exists between the theoretical expectations and the experimental results. Possible reasons for this deviation are discussed under the following headings.

(1) Change in anisotropy energy of closure domains

For the narrow strips, the total area of triangular domains is comparable to the total area of elongated domains. Thus, the external field's contribution to the change in anisotropy energy of closure domains can not be entirely neglected. The meticulous examination and comparison of Table 1 and Figure 5 revealed that the domain wall energy density γ

calculations based on the wider film strips were closer to the theory than those which were calculated on the basis of the narrow strips. This was verified in all thicknesses of film studied (i.e., from 200 Å to 2700 Å).

(2) Easy axis and anisotropy dispersions

For a given sample with uniaxial anisotropy, the easy direction of magnetization is not uniform throughout the sample. This phenomenon is called the easy axis dispersion. Nor is the anisotropy field completely uniform at each point in the film, thus creating an anisotropy dispersion. Hence, the easy direction and the anisotropy field H_k for a given film are only average values.

Several investigators (1, 5, 15, 18) have studied the technical measurement of these dispersions. These dispersions are presumably the result of the microstructure of the film and the distribution of stress within it. Depending upon the perfection and the size of the film, the measured dispersion of the easy axis has been found to range from a very small angle to one of several degrees. For a perfect single crystal, this dispersion is negligibly small; within an actual polycrystalline film, however, dispersion can be quite large.

In 1964, Dove and Long (7) measured angular and magnitude dispersion within small areas (25 μ in diameter) of permalloy films using a magneto-optic technique. By studying the distribution of the loss curve obtained by this experiment, they

found that a spread of H_k values of $\pm$ 23% was typical of "good" films in the order of one centimeter in diameter. Equation 1 gives γ as being directly proportional to K. Since $H_k = \frac{2K}{M}$, this implies that γ is strongly dependent on anisotropy dispersion. From observation of the domain figures in the present study, it was determined that a 10% domain width variation was not uncommon. This was taken as evidence that anisotropy variation does indeed occur within a film.

(3) Coercive force of the domain wall

The coercive force of a domain wall is related to the energy required for a wall to overcome certain potential barriers in its motion through the film. The coercive force tends to prevent the film from reaching a true minimum energy state. If the wall energy density calculated from Equation 1 is to be correct, it is vitally important that the calculation be based upon measurements taken at a true state of minimum energy. Thus, it is possible to assume that deviation from the theory is at least partially a function of the domain wall coercive force due to film imperfections such as a surface roughness, finite crystallite size, and film pinholes.

(4) Approximate calculations used in the theoretical derivations

Domain wall energy consists of three energies: the stray field energy, the exchange energy, and the anisotropy energy.

The stray field energy is the magnetostatic energy associated with free magnetic poles. These poles in normal bulk materials are sufficiently far apart that, compared with the anisotropy and exchange energies, the stray field energy can be neglected. The poles in thin films, on the other hand, reside at the intersections of the walls with the surfaces, and being separated only by the film thickness are very close together, making it impossible to entirely neglect magnetostatic stray field energy.

A proper evaluation of the stray field energy requires an a priori knowledge of both the local stray field and the magnetization in the wall, thus making a knowledge of the shape of the wall necessary. To overcome the difficulty of not knowing the solution in advance, certain analytical simplifications must be used which at best only approximate the actual magnetization distribution in the wall.

The anisotropy energy of a domain wall is the result of the magnetization in the wall being turned out of the local easy direction. The exchange energy is the result of the exchange forces between neighboring magnetic dipoles of the wall. For 81% Ni - 19% Fe films, the stray field energy and the exchange energy are predominate over the anisotropy energy.

Middelhoek assumed that the direction of magnetization rotates linearly across the transition region from one domain

to the next, the width of the wall entering as a parameter. On the basis of this linear variation, the exchange and anisotropy energies were then calculated. The expression for the stray field, however, was only approximated by using a uniform mean magnetization in the transition region. To obtain a measure of the equilibrium wall thickness and energy, the resulting approximate expression for the energy was then minimized with respect to the transition width. Since all the terms in the expression for total energy are not calculated with the same magnetization distribution, the resulting minimum may be above, the same as, or below the actual energy for the wall.

Brown and LaBonte (6) pointed out that:

> ...the continuous one-dimensional magnetization of the 180° wall between two antiparallel domains is replaced by a discrete distribution for which the magnetostatic energy can be calculated exactly. Within the transition the magnitude of the magnetization is constrained to be the spontaneous moment M_S, but its direction is allowed to vary arbitrarily from point to point of a discrete grid of points. The conditions for minimizing the total energy of the discrete distribution are found by variational methods. The resulting equations are interpreted in terms of an 'effective field,' which is then used in digital-computer calculations to find the detailed structure of the equilibrium 180° magnetic transition and the corresponding wall energies.

The domain wall energy density computed by Brown and LaBonte was smaller than that calculated by Middelhoek. Both theoretically computed curves are plotted in Figure 5, together with the experimental curve measured in the course of the present investigation.

C. Discussion of the Domain Patterns Occurring on Wider Film Strips

For the entire range of film thicknesses studied in the present investigation (from 200 Å to 2700 Å), Lifshitz domain structures (Figures 11, 14) were observed whenever the strip width was greater than 0.5 mm. When the strip width was close to 0.5 mm, Lifshitz domain and 90° domains occurred alternatively. For strip widths greater than 0.5 mm, Lifshitz domains tended to appear between the 90° domains (Figure 14).

The existence of these edge domains can be attributed to the demagnetizing field near the edge of the film and to edge imperfections. Since no magnetic charge can exist freely in nature, the divergence of the magnetic flux density must be zero. The H field (the demagnetizing field) which occurs at the edge will consequently be equal in magnitude to the magnetization there, but opposite in direction. The presence of this field diametrically opposite to the magnetization at the edge tends to break up the single domain structure into a multi-domain structure.

The demagnetizing field along the longitudinal median of a narrow strip of film is comparable to the field at the edges. Thus, the field is allowed to break up the film into antiparallel, elongated domains. For wide strips, on the other hand, the demagnetizing field in the center is weaker than that at the edges and, although Lifshitz and 90° domains

are formed on the edges to reduce the magnetostatic energy, the longitudinal middle of the film tends to remain in a single domain state (Figure 8).

D. Conclusions

In conclusion, it is evident that there are many quasi-minimum energy states in a film possessing uniaxial anisotropy. Depending on the direction of external magnetic fields applied and the method of demagnetization, the film will tend to remain in one particular energy state. It is important in the calculation of domain wall energy density that the film be in an absolute minimum or at least a nearly absolute minimum energy state since the existence of this state is the basic assumption upon which the derivation is based.

In a study of the energy density of thin magnetic films made by varying the strip width and measuring the domain dimensions, extra care must be taken to assure that the easy axis will be uniform throughout the film. Chemical etching seems to provide such a solution.

During the present investigation, it became evident that film thickness can be determined if:

(1) the type of domain walls,

(2) the number of domains per unit length, and

(3) the strip width of the film are known.

This provides a convenient method of determining magnetic film thickness. Calculated from the experimental data collected in

Table 1, the curves of the thickness determined as a function of the number of domains per centimeter using the strip width as a parameter are drawn in Figure 18. The accuracy of thickness obtained from these curves will, of course, depend upon the uniformity of the domain width along the strip and the technique used in chemical etching.

E. Future Investigations

In the course of the present investigation, several related aspects of the problem area were brought to attention. These ideas, which may suggest future areas of examination and investigation, will be mentioned briefly.

(1) Domain width variations were obtained in the present experiment; that is, 10% domain width variation along the same strip was not uncommon. It is logical to assume that this is due to anisotropy variation of the film. By using the average value of γ, an investigation could be made to determine whether the anistropy constant K determined from Equation 1 at each median point of the strip and the anisotropy constant K measured by the magneto-optical method or rf-probe technique correspond for points at similar positions within the domain. Their values should be closely related.

(2) In order to bring magnetic films to their minimum energy state, the possible experimental results of other ac demagnetization methods present interesting possibilities. It

might be valuable to attempt an utilization of an ac rotation field for demagnetization.

(3) Besides using a magnetic field to demagnetize the film, other methods such as thermal demagnetization should be investigated. In this procedure, the film would be heated to its Curie temperature and cooled in the absence of a magnetic field. Hopefully, the experimental result would be similar to that of the present experiment.

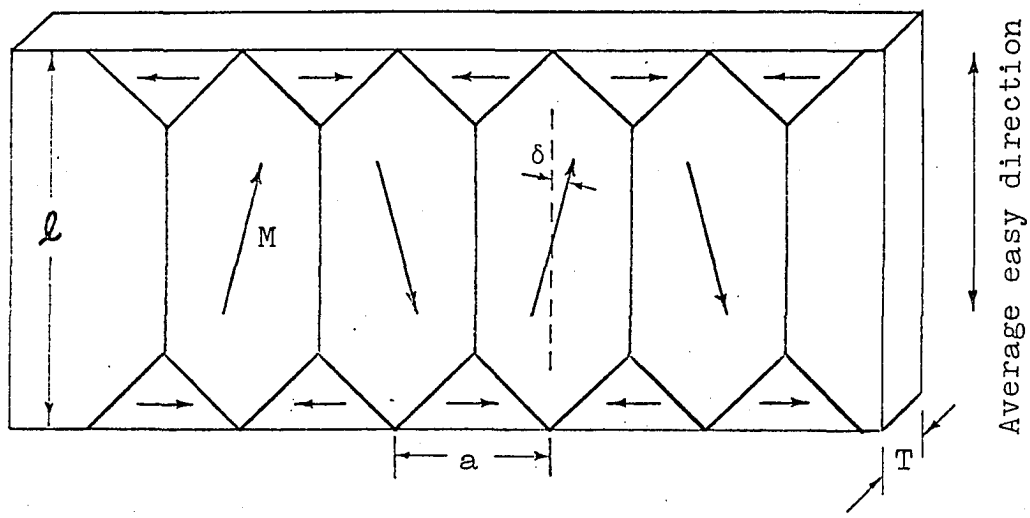
An extensive study of demagnetization methods would lead to a better understanding of the magnetization distributions and the minimum energy states of thin magnetic films.

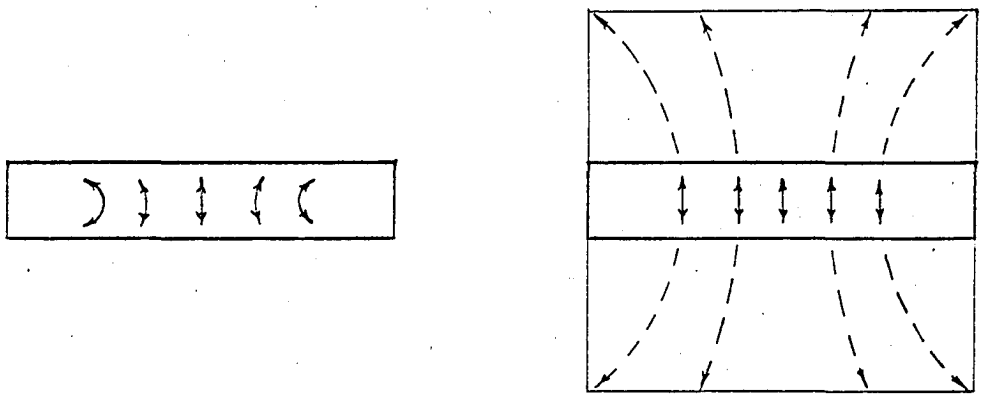
Figure 16. Possible magnetization distributions in the elongated domains for the closure domain model

Figure 17. Local easy axis distributions within a film strip

(a) Film strip is formed by evaporation through a slit

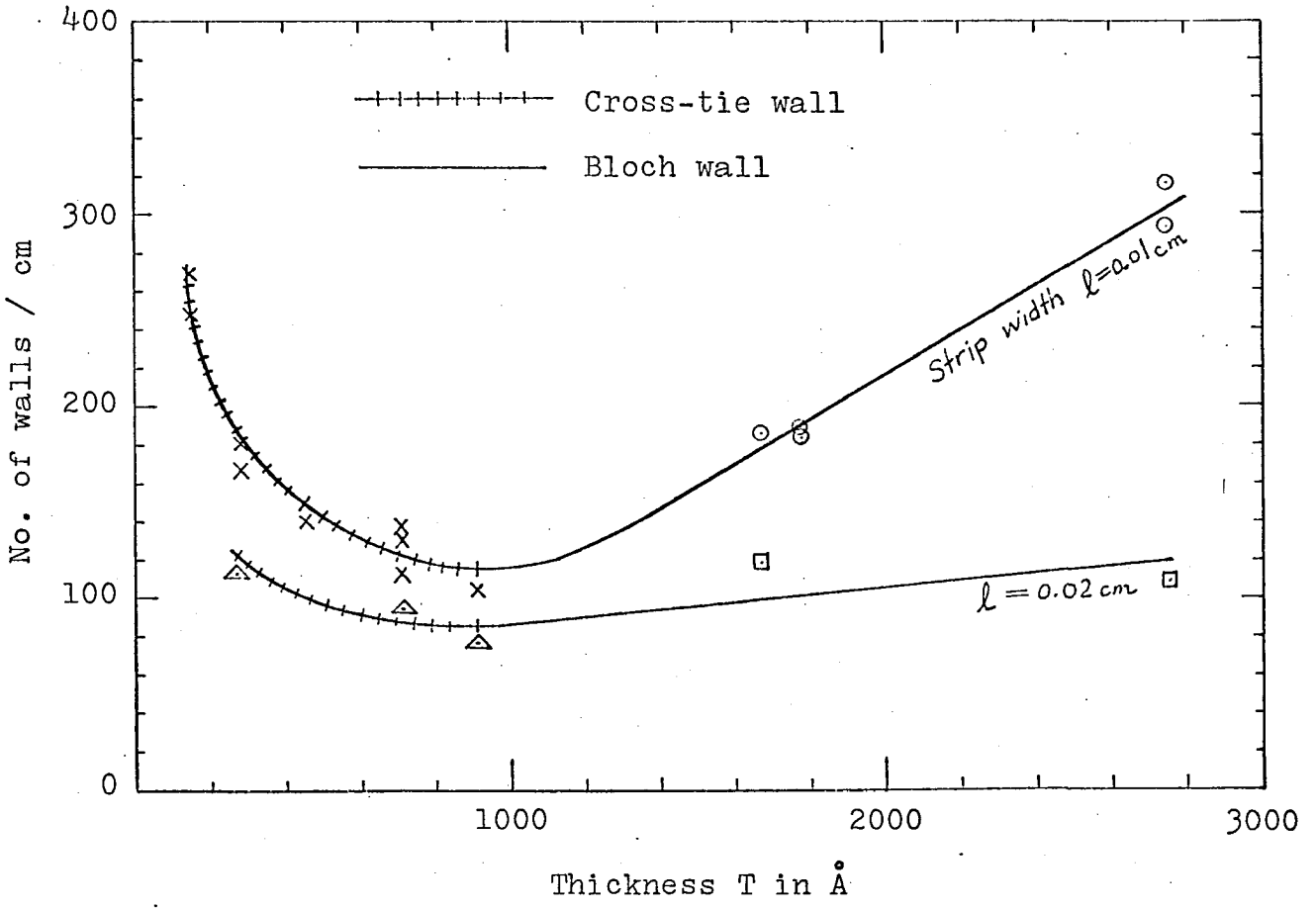
(b) Film strip is formed by chemical etching





(a) (b)

Figure 18. Relationship between number of walls per centimeter (obtained from closure domain observation) and thickness of film. Strip widths used as parameter. Data calculated from Table 1


400
300
200
100
0
No. of walls / cm
Cross-tie wall
Bloch wall
Strip width ℓ=0.01 cm
ℓ = 0.02 cm
1000
2000
3000
Thickness T in Å

IV. APPENDIX A

A. Table 1. Experimental Data of the Films

Film sample	Thick-ness (Å)	H_c (oe)	H_K (oe)	K (erg/cm)	a (cm)	ℓ (cm)	γ(calc.) (erg/cm^2)
7-25	142	3.18	5.08	2030	.00372	.009	1.56
					.00409	.009	1.89
7-15	284	3.33	4.45	1780	.00593	.009	3.49
					.00644	.009	4.10
					.00644	.009	4.10
					.00875	.023	2.97
7-19	461	2.54	2.86	1140	.00711	.010	2.88
					.00650	.0103	2.34
5-4	720	3.65	5.40	2160	.00755	.009	6.82
					.00867	.009	9.00
					.00735	.009	6.46
					.01030	.022	5.18
7-27	922	2.70	4.13	1650	.0096	.0095	8.00
					.0096	.0095	8.00
					.0135	.023	6.53
7-18	1670	3.33	4.44	1774	.00265	.011	0.566
					.00333	.011	0.894
					.00538	.011	2.34
					.00842	.023	2.74
					.01240	.048	2.84
7-29	1770	1.94	3.81	1520	.00530	.0105	2.03
					.00539	.0105	2.11
					.00539	.0105	2.11
7-20	2760	2.06	3.12	1248	.00337	.0103	0.69
					.00337	.0103	0.69
					.00315	.0103	0.602
					.00910	.0237	2.18

V. APPENDIX B

A. Coils Designed for Applying Demagnetization Fields

By referring to Figure 19, the magnetic field H produced at the center of a pair of coils due to a current i passing through the coils is:

$$H = \frac{4nia^2}{\pi\sqrt{\ell_1^2 + 2a_1^2}\ (\ell_1^2 + a_1^2)} \quad \text{amp-turns/meter}$$

or

$$H = \frac{1}{79.58}\ \frac{4nia^2}{\sqrt{\ell_1^2 + 2a_1^2}\ (\ell_1^2 + a_1^2)} \quad \text{oersteds}$$

$$= 4.62 \times 10^{-3}\ \frac{ni}{a} \quad \text{oersteds}$$

where

n = number of turns

i = current in amps.

a_1, ℓ_1 = length in meters.

Four pairs of coils were made using this formula. The oersted/ampere value was calculated and the value checked by a Gauss meter, a Hewlett-Packard 352A Gauss Meter Probe in conjunction with a Hewlett-Packard Model 428 (range 1000 ma) dc milliammeter. The earth field was shielded for the experiment.

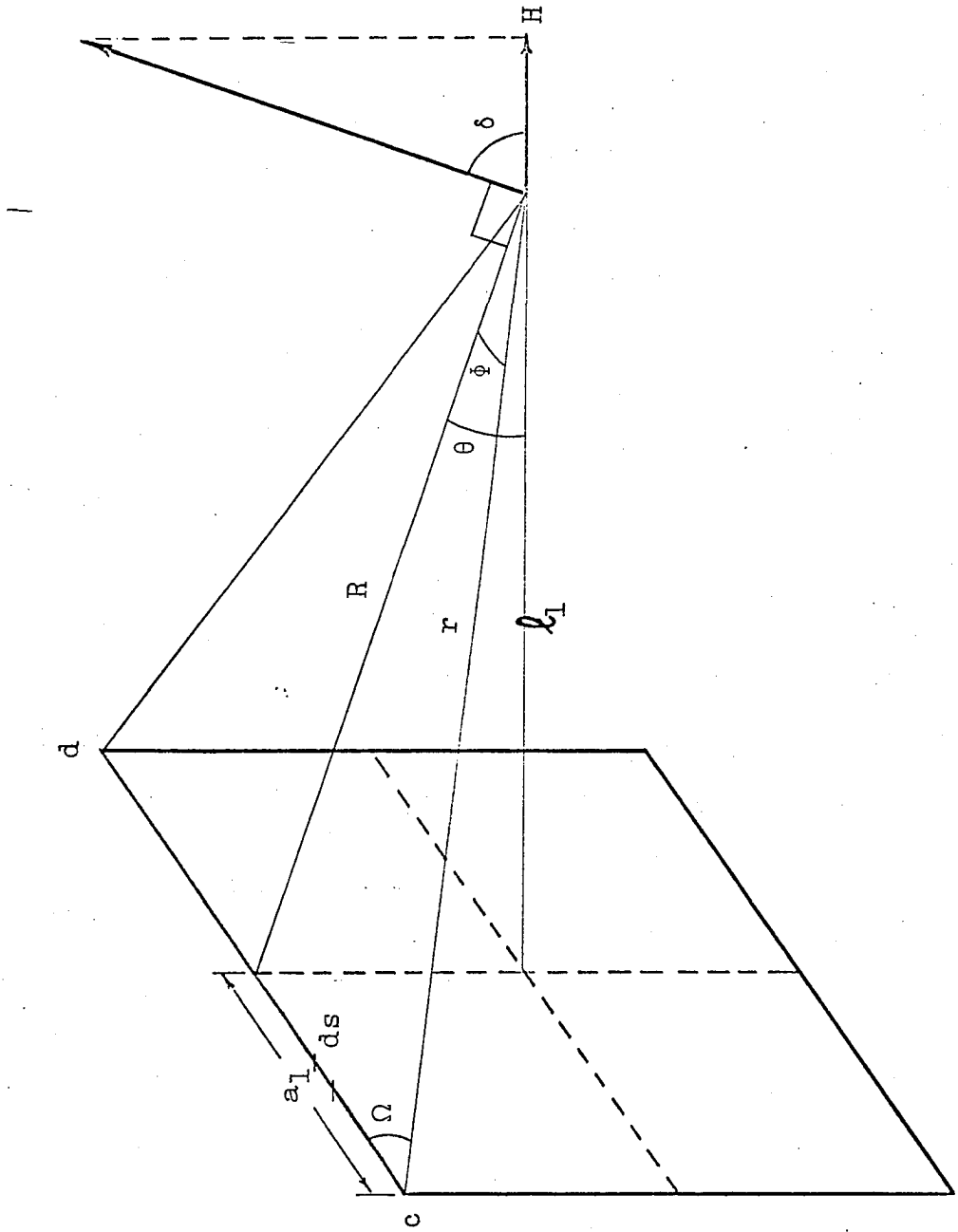
n = 100 turns	H/i(oer/amp)calc.	H/i(oer/amp)measured
L_1 $2\ell_1 = 2a_1 = 20.95$ cm	4.4	4.5
L_2 $2\ell_1 = 2a_1 = 16.20$ cm	5.7	5.95
L_3 $2\ell_1 = 2a_1 = 13.50$ cm	6.84	6.875
L_4 $2\ell_1 = 2a_1 = 11.10$ cm	8.32	8.62

Uniformity test:

A uniform magnetic field is required for ac demagnetization. Using the geometric center of the pair of coils as a reference, the percentage variation of the field was measured:

		Left				Right		
distance from center (cm)	0	1	2	3	0	1	2	3
L_1 H-field (arb.unit)	72	71.5	70.5	69	72	72.5	73	75
% variation	0	0.7	2.1	4.2	0	0.7	1.4	4.2
L_2 H-field (arb.unit)	66	67	70	74	66	65.5	65	63
% variation	0	1.5	6.1	12	0	.75	1.5	4.6
L_3 H-field (arb.unit)	80	78	75	72	80	82	86	92
% variation	0	2.5	6.3	10	0	2.5	7.5	15
L_4 H-field (arb.unit)	62	65	70	77	62	61.5	60	56
% variation	0	4.85	12.9	24.2	0	0.8	3.2	9.7

Figure 19. Schematic diagram for coil design


H
δ
Φ
θ
R
r
l_1
d
c
ds
a_1
Ω

VI. APPENDIX C

A. Relations of the Field Vectors to the Magnetic Polarization Vector in cgs and in MKS Systems of Units

To compare the results of the present experiment with those obtained by previous investigators, cgs units were used throughout the study. Since the MKS system of units is widely used in the engineering field, tables of the equation and conversion factors for both cgs and MKS systems of units are provided below as references.

Table 2. Relation of the field vectors, B and H, to the polarization vector M in both cgs and MKS units

Symbol	Quantity	Unrationalized system of cgs units $\vec{B} = \vec{H} + 4\pi\vec{M}$	Rationalized system of MKS units $\vec{B} = \mu_o \vec{H} + \vec{M}$
B	Magnetic flux density	gauss	Webers per square meter
H	Magnetic field intensity	oersted	Newtons per Weber or amperes per meter
M	Magnetization	gauss	Webers per square meter

Table 3. Conversion factors between cgs and MKS units

Symbol	Quantity	Multiplied number of	by	To obtain number of
Φ	Magnetic flux	Maxwells	10^{-8}	Webers
B	Magnetic flux density	Gauss	10^{-4}	Webers/meter^2
H	Magnetic field intensity	Oersteds	79.58	Ampere-turns/meter

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