

Domain-Wall Pinning by Magnetic Inhomogeneities and Temperature Dependence of Coercivity

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A thesis submitted to the Faculty of Graduate
Studies of the University of Manitoba in the
partial fulfillment of the requirements for
the degree of Doctor of Philosophy.

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ISBN 0-315-63210-0

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ACKNOWLEDGEMENT

I am grateful to my supervisor, Dr. P. Gaunt for introducing me to the research area of domain wall pinning. He had actively been involved in this research and taught me the basic skills to do the experimental measurements.

I wish to thank Dr. G. Williams and Dr. A. H. Morrish, who acted as my acting supervisors and gave me valuable suggestions in the later stage of my Ph. D. program. Thanks are also given to Mrs. Gaunt for correcting the errors in English in my first paper published in Journal of Applied Physics. Special thanks are due to Dr. H. Kunkel for his very kind help, both in Physics and English, which has finally made my other two papers publishable.

A four-year University of Manitoba Graduate Fellowship is appreciated.

Finally, I would like to express my appreciation to the Department Office for the assistance with my financial support after my Graduate Fellowship ended.

ABSTRACT

The temperature dependence of coercivity, $H_c(T)$, of a ferromagnet may originate from two different sources. The first is the temperature dependencies of the intrinsic parameters, such as the anisotropy constants, the exchange constants and the spontaneous magnetizations in both the matrix and the pins. The total effect of these parameters can be described by the temperature dependence of the energy barriers which depend on these parameters. The second source is the thermally activated process of domain wall unpinning which is experimentally verified. In general, neither is negligible. Unfortunately the existing pinning models neglect either source with no reason, therefore systematic errors are inevitable in these models. The present work has as its aim the incorporation of these two contributions to the coercivity as a function of temperature in a single theory.

The experimental techniques used are magnetic hysteresis measurements, X-ray diffraction and transmission electron microscope (TEM) observations. These measurements are necessary to test the validity of the proposed models.

For $Pt_{0.7}Ni_{0.3}Fe$, it is suggested that $\{110\}$ lamellar junctions are the pinning sites and that the kinks of a Bloch wall at some lamellar junctions are the result of a reduction in the demagnetizing energy. An improved strong-pinning model is advanced to describe the temperature dependence of coercivity. The fitting is reasonably good. The value of the pin density from

the best fitting is in the range observed in TEM observations.

The grain boundaries are the dominant pinning sites in MnAlC magnets. The maximum pinning force between a domain wall and a planar pinning site is proportional to $K_1^{3/2}/A_1^{1/2}$ for a thin pinning site, where K_1 and A_1 are the uniaxial anisotropy and the exchange constants respectively in the matrix. The pin interaction range is proportional to $(A_2/K_2)^{1/2}$, where K_2 and A_2 are the anisotropy constant and the exchange constant respectively in pinning site. Gaunt's weak pinning model (*) is sufficient to describe the temperature dependence of coercivity of a MnAlC magnet provided H_0 , the critical field in the absence of thermal activation, is considered as a function of temperature. The pin density predicted by the weak-pinning model is verified by TEM observations.

The pin interaction potential proposed by Gaunt and Mylvaganam (**) is a good approximation only when $H_c(T)/H_0(T)$ is close to one. When $H_c(T)/H_0(T)$ is much less than one, as for $\text{Sm}(\text{CoNi})_{2.5}$, the error caused by this simplified potential is too large. Then both the strong-pinning and the weak-pinning models proposed by Gaunt (*) are not sufficient. A better form of the pin interaction potential is therefore necessary. A discrete spin model is used to calculate the pin interaction potential and then the activation energy to form a domain wall blister is calculated. It is found that the activation energy increases very quickly as $H_c(T)/H_0(T)$ decreases. The model successfully describes the temperature dependence of the coercivity of $\text{Sm}(\text{CoNi})_{2.5}$, showing that the main pinning sites are the magnetic inhomogeneities where

the Co atoms are replaced by Ni atoms. This conclusion is supported by the failure of TEM observations to find visible pinning sites. Finally, this model shows that the pin interaction range may have a significant effect on the shape of the $H_c(T)$ curve.

* P. Gaunt, Phil. Mag. B. 48, 261(1983).

** P. Gaunt and C. K. Mylvaganam, Phil. Mag. B. 39, 313(1979).

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Notation Guide

- A —mean wall area per pin.
- a —lattice constant
- $A_{1,2}$ —exchange constants in the matrix (1) or the pin (2).
- A_s —cross sectional area of a sensing coil.
- B —magnetic induction.
- b, b_0 —pin interaction range.
- $C_{i,j}$ —integral constants, $i=1,2,3$.
- D —demagnetizing factor.
- $d_{1,2}$ —dimensions of a lamellar junction.
- d_{hkl} —spacing of a lattice planes.
- D_p —distance of a diffracted beam spot from the center.
- E —activation energy.
- E_A —uniaxial anisotropy energy.
- E_D, E'_D —demagnetizing energy.
- E_e —exchange energy.
- E_H —extra field energy.
- E_i —total interaction energy.
- E_γ —extra wall energy.
- F —pinning force per unit area of domain wall.
- f, f_0 —maximum pinning force per pin.
- f' —maximum pinning force per unit area of a pin.

F_l —Lorentz force.
 H —effective magnetic field.
 $h, h_{1,2}$ —dimensionless magnetic field.
 H_a —applied magnetic field.
 H_c —magnetic coercivity.
 H_D —demagnetizing field.
 H_K —uniaxial anisotropy field.
 H_0 —critical field in the absence of thermal activation.
 h_0 —dimensionless critical field.
 J —exchange integral.
 k —Boltzmann's constant.
 $K_{1,2}$ —uniaxial anisotropy constant in the matrix (1) or pin (2).
 L —image correction factor.
 l_p —distance between two nearest pins.
 ld_2 —extra domain wall area.
 M —magnetization of the whole specimen.
 $M_{1,2}$ —spontaneous magnetization in the matrix (1) or pin (2).
 n —effective width of the Bloch wall.
 n_r —order of reflection.
 S —spin of an atom.
 S_p —mean area of a planar pin.
 T —absolute temperature.
 t —domain wall thickness.

t' —time.

$U(z), V(z)$ —interaction potential.

v —dimensionless wall displacement.

v_∞ —energy minimum position at $r \rightarrow \infty$.

v'_∞ —energy maximum position at $r \rightarrow \infty$.

W —planar pin width.

w —dimensionless planar pin width.

ΔX —mean dimension of a planar pin.

z, z_W —domain wall displacement.

Z, Z_W —dimensionless domain wall displacement.

$z'_{1,2}$ —pin boundaries.

γ, γ_0 —domain wall energy.

ε —dimensionless activation energy.

ε_i —electromotive force.

η —demagnetizing factor normalized by 4π .

θ —spin orientation.

$\theta_{1,2}$ —spin orientations at pin boundaries.

λ —wavelength.

$\pi\delta_{1,2}$ —wall thickness in the matrix (1) or pin (2).

ρ —pin density.

$\phi_{1,2}$ —magnetic flux.

ψ —incident angle of a beam.

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Chapter 1

Introduction

1.1 Domains and domain walls

Ferromagnets are defined as those materials in which atomic magnetic dipole moments are aligned parallel to each other. Such materials can therefore exhibit a magnetic moment even in absence of a magnetic field. Despite this property of spontaneous magnetization, a ferromagnet which has never been exposed to a magnetic field will not exhibit a magnetic moment. Upon the application of a small field however, a large moment may be seen.

To explain these phenomena, Weiss [1] predicted the existence of domains which are the small spontaneously magnetized regions within the material. These domains do have individual magnetic moments, but could be aligned in a random way to give a null resultant moment over the whole material. When a magnetic field is applied, they will realign themselves, giving a net resultant moment. Such a multi-domain configuration has a lower magnetostatic energy than a large single domain of uniform magnetization over the entire material. When the magnet is sufficiently small, the single domain configuration has a lower energy than the multi-domain configuration.

In 1932 Bloch [2] showed theoretically that the boundary between domains is not sharp on an atomic scale but is spread over a certain thickness wherein the direction of spins changes gradually from one domain to the next. This layer is usually called a domain wall and is sometimes referred to as a Bloch wall. The thickness of the domain wall is determined by the counterbalance of the exchange energy which tends to increase the thickness, and the anisotropy energy which tends to diminish it.

1.2 Hysteresis loop

Fig.1.1 shows a typical magnetization curve and a typical hysteresis loop of a ferromagnetic material. Starting from a demagnetized state, the magnetization, M , increases with an increase of the field along the curve OABC and finally reaches a value which is called the saturation magnetization. If the magnetic field is decreased from the saturated state C, the magnetization is gradually decreased along CD, not along CBAO, and at $H = 0$ it reaches the finite value which is called the remanence. Further increase of the magnetic field in a negative sense results in a continued decrease of the intensity of magnetization which finally falls to zero. The field at this point is called the coercive force or coercivity H_c . This portion, DE, of the magnetization curve is often referred to as a demagnetizing curve. Further increase of H in a negative sense results in an increase of the intensity of magnetization in a negative sense and finally leads to a negative saturation magnetization. If the field is then reversed to the positive sense, the magnetization will change along FGC. The closed loop CDEFGC is called the hysteresis loop.

1.3 Domain wall pinning and nucleation

The magnetization reversal may be achieved by domain-wall movement. In its movement, a domain wall will pass the energy barriers originating from magnetic inhomogeneities (pinning sites or repulsive sites). The pinning (or repulsive) sites are local regions with weaker (or stronger) exchange constants

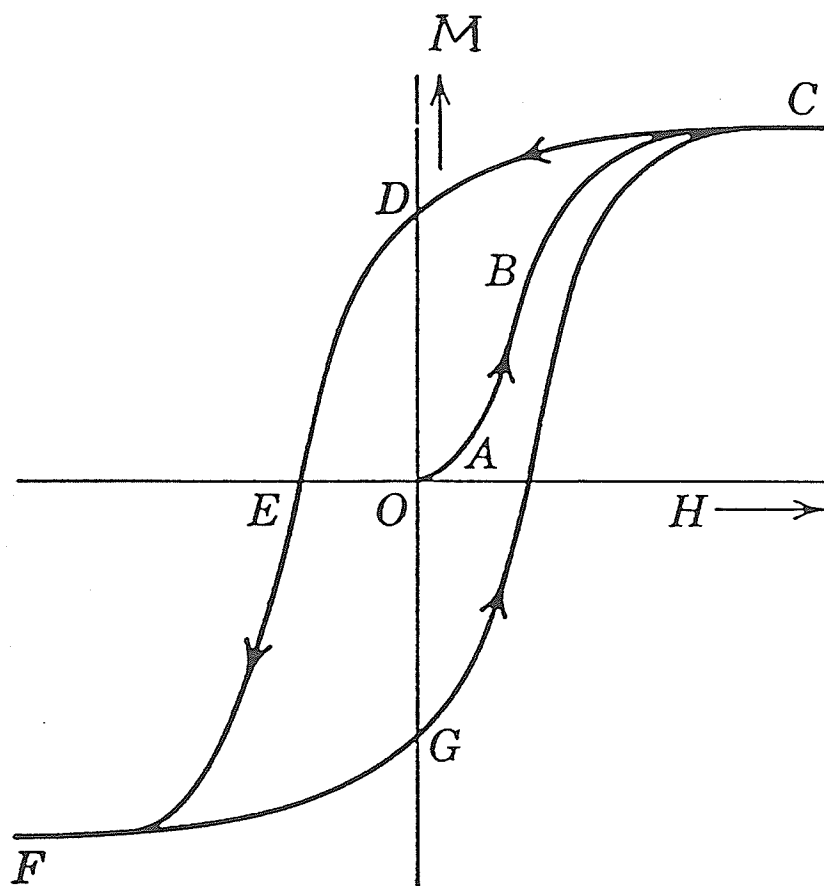


Figure 1.1: Hysteresis loop

and anisotropy constants as compared to the matrix. A moving wall has to pass energy barriers since pinning sites are invariably present. The domain walls may deviate from their energy minimum positions by thermal activation and/or by a driving field. In the former case alone, the walls will move in all directions with equal probabilities. On the average, the magnetization of the whole magnet remains unchanged. On the other hand, in the latter case the walls will move in a preferred direction, resulting in a change in the magnetization of the whole magnet. The coercivity is determined by the field required to propel a wall past the energy barriers.

Magnetization reversal could also be achieved by the nucleation process. In this process, the magnetization in some local regions with weak anisotropy energy can suddenly reverse its direction in a finite field, called the nucleation field. These nuclei may then expand in such a way that the magnetization of the whole material reduces to zero. Since the nuclei are themselves pinning sites, the condition for the expansion of nuclei is that the field required to unpin domain walls is less than the nucleation field. If this is not true, the coercivity will be determined by the domain wall unpinning process.

Both the nucleation model and the domain wall pinning model are not well established as yet, so that sometimes it is difficult to identify each mechanism with certainty. The present work is devoted only to the study of the domain-wall pinning mechanism.

1.4 Pinning models taking account of the thermal activation of domain walls

The moving walls have to pass the energy barriers in order to change magnetization of the whole magnet. Gaunt [3]-[8] had pointed out that domain walls may have thermal vibrational energy which can assist walls to pass the energy barriers. Strong evidence of this argument is found in the excellent experimental work by Siemers and Nembach [9]. They measured the domain wall velocity as a function of magnetic field and temperature for picture-frame type single crystal specimens of a ferromagnetic nickel-based alloy. The pinning sites are nonmagnetic particles. Temperature are selected between 1.7 and 5 K, which are far below the Curie temperature of 165 K. Therefore, all magnetic parameters, which govern the interaction between domain walls and pinning sites, are expected to be independent of temperature. But the measured domain-wall velocity is very sensitive to temperature. This phenomenon can only be explained by the thermally activated process of wall movement.

The main ideas in Gaunt's model can be seen in his recent work [7]. He considered the analogy between the coercivity of ferromagnetic materials containing magnetic inhomogeneities and the yield stress of two-phase alloys. Magnetic inhomogeneities may be any local regions with different magnetic properties from the matrix. In the magnetic case the domain walls may be pinned at magnetic inhomogeneities and in the mechanical problem dislocations are pinned at precipitates. The coercive field and the yield stress

are the critical field H_0 and the critical stress required to release the wall or dislocation from the pins. A fundamental approximation of the model developed to explain the unpinning is that the domain wall can be treated as a deformable membrane of surface energy γ per unit area pinned, like a buttoned cushion, at points on its surface. A further assumption is that the rather complex statistical problem of a random array of pins can be reduced to consideration of the forces required to move the wall over a unit step.

To solve the pinning problem for a particular material, it is therefore necessary to know the interaction potential $V(z)$. Gaunt [6] proposed a potential of the form

$$V = f[(z + b) - (z^3 + b^3)/3b^2], \quad (1.1)$$

where f is the maximum pinning force per pin and b is the pin interaction range. The minimum energy occurs at a point $z = -b$, and the maximum energy at $z = +b$ (Fig.1.2). Since the pinning sites always reduce the wall energy, the derivative of the potential should have positive slope throughout. Therefore when the pin width is much less than the wall thickness, t , the interaction range is related to the wall thickness in such a way that $4b = t$.

Suppose a segment of domain wall of area A is associated with a pinning site. The total energy associated with the segment is then the summation of the pin interaction potential and the magnetostatic energy, given by

$$U(z) = V(z) - 2HM_1Az, \quad (1.2)$$

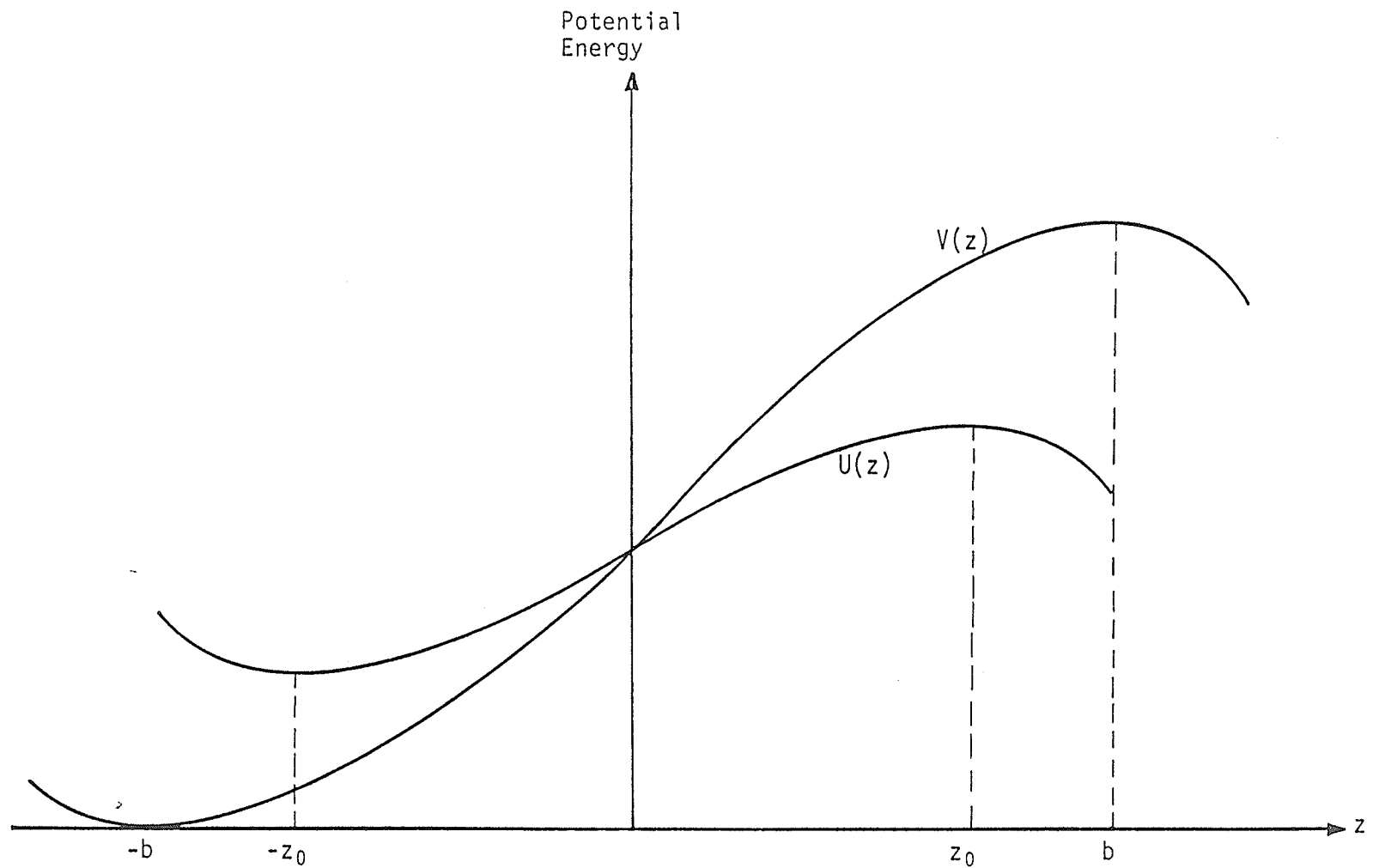


Figure 1.2: The variation with position of potential with and without the presence of a field.

where M_1 is the spontaneous magnetization in the matrix and H is the magnetic field. Differentiating Eq.(1.2) with respect to z gives the equilibrium wall position as

$$z_0 = \pm(1 - 2HM_1A/f)^{1/2}, \quad (1.3)$$

where the negative sign corresponds to an energy minimum and the positive to an energy maximum. The critical field in the absence of thermal activation, H_0 , is obtained by setting Eq.(1.3) to zero.

$$H_0 = f/2M_1A. \quad (1.4)$$

When $H = H_0$, the energy barriers a wall has to pass on breaking away from pins are essentially zero.

Gaunt [7] considered two different cases of domain wall pinning. When a domain wall leaves pins in a one-by-one process, the process is classified as strong pinning. When a wall leaves many pins simultaneously, the process is classified as weak pinning. The Friedel criterion, defined as the ratio of the mean wall area per pin in zero field to that in a finite field, is

$$\beta_0 = 3f(8\pi\gamma b)^{-1}, \quad (1.5)$$

which is used to distinguish strong pinning ($\beta_0 > 1$) from weak pinning ($\beta_0 < 1$). By taking account of the flexibility of domain walls, Gaunt found that the energy barrier in the strong pinning case is a function of the magnetic

field, given by

$$E = (4/3)bf[1 - (H/H_0)^{1/2}]^{3/2}. \quad (1.6)$$

This amount of energy can be supplied to the wall by the thermal vibration of the wall. Gaunt and Roy estimated the thermal vibrational energy to be in the order of $25 kT$, where k is the Boltzmann's constant and T is the temperature in degrees Kelvin. The coercivity, H_c , as a function of temperature is then obtained by setting Eq.(1.6) equal to $25 kT$, that is

$$(H_c/H_0)^{1/2} = 1 - [75kT/(4bf)]^{2/3}, \quad (1.7)$$

where

$$H_0 = 3\rho f^2/(4\pi M_1\gamma), \quad (1.8)$$

and ρ is the pin density. The activation energy in the weak pinning case is found to be

$$E = 2\pi N\gamma b^2(1 - 2HM_1/F), \quad (1.9)$$

where $2\pi N = 31.0$ is a constant and F is the pinning force per unit area of the domain wall. Gaunt thought that the pinning force can only come from statistical fluctuations of the pin distribution. He found that the maximum value of critical field can be approximated well by Eq.(1.8) in the weak pinning case, then the coercivity can be described by

$$H_c = H_0[1 - 25kT/(31\gamma b^2)]. \quad (1.10)$$

From Eqs. (1. 7) and (1. 10), Gaunt [11] concluded that in the strong-pinning case, $H_c^{1/2}$ should vary as $T^{2/3}$, whereas in the weak-pinning case, a linear relationship between H_c and T should be observed. In either case, H_0 is taken as a constant, sometimes it is named the absolute zero coercivity.

The above summary neglects the internal demagnetizing effects due to uncompensated spins on the surfaces of bowed walls. These effects modify the results somewhat so that, for example, H_0 for strong pinning depends on $f^{7/3}$ rather than f^2 . The corrections are discussed in detail in the original paper but, for the present purpose, will be neglected here.

Egami [12] studied the thermally activated process of domain wall unpinning in the case when the domain walls are extremely narrow. Then, the angles between adjacent layers of spins are not small so that the micromagnetic continuum approximation is not valid and a discrete spin model is necessary. Due to their atomic nature, these narrow Bloch walls have an intrinsic coercivity which is analogous to that of lattice dislocations, known as the Peierls-Nabarro force. Fig.1.3 shows two configurations of a domain wall. In configuration A the center of the wall lies on an atomic layer of spins which are oriented perpendicular to the easy axis. In configuration B the center of the wall lies between two atomic layers. It is calculated that the configuration A is energetically unfavorable. The fact that such configurations occur periodically when the wall moves through the crystal indicates that there is an energy barrier to the motion of domain walls even in perfect crystals. This energy barrier is called the Peierls Potential, and the related

domain wall pinning is sometimes named intrinsic pinning.

In his model Egami calculated the activation energy, in finite magnetic fields, required to form the nucleus on domain walls in the homogeneous nucleation process. The wall is subjected in a uniformly distributed array of Peierls potential assumed to have a sinusoidal or a parabolic form. He found that the nucleation energies calculated with the sinusoidal potential and with parabolic potential agree well, especially for small magnetic field; and that the activation energy for homogeneous thermal nucleation is dependent on the magnetic field and that there is inverse relationship between the activation energy and the magnetic field when the magnetic field is small compared to the critical field. Mathematically the activation energy [12] is given by

$$E_c = \langle S \rangle^2 J(16/n)(H_0/H)G_{Th}(H/H_0), \quad (1.11)$$

where S is the spin of an atom, J is an exchange integral, n is the effective width of the Bloch wall and G_{Th} is a function which is approximated by

$$G_{Th}(H/H_0) \approx 1 - (H/H_0)^2. \quad (1.12)$$

Only with the assistance of thermal vibrational energy is the domain wall unpinning possible. Although Egami described the thermally activated process as homogeneous nucleation, his model is essentially a domain-wall pinning model, not a nucleation model. Besides domain-wall pinning by magnetic inhomogeneities (extrinsic pinning) in the case when the domain walls are

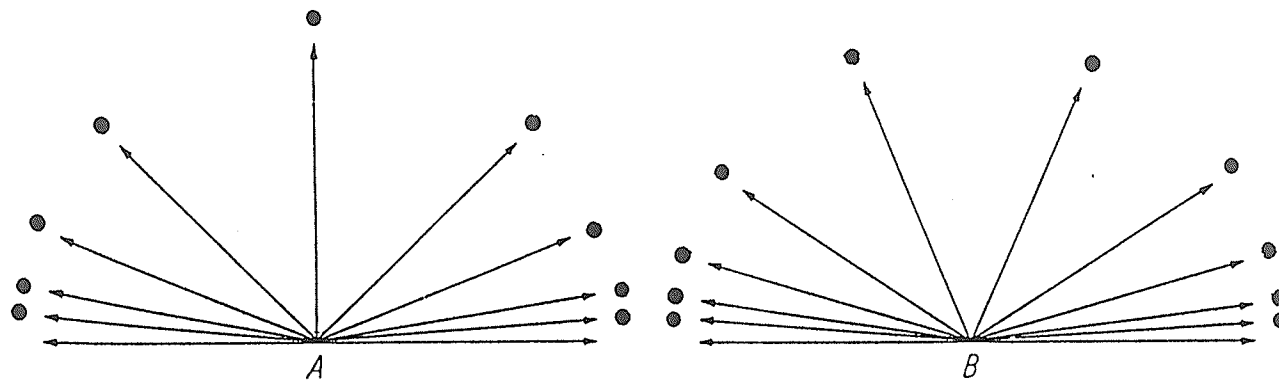


Figure 1.3: Schematic representation of the spin orientation in the atomic layers parallel to the domain wall for the symmetric positions A and B of the domain wall.

extremely narrow is not studied in his model.

1.5 Pinning models taking account of the temperature dependencies of magnetic parameters

Domain-wall pinning has been studied in a different way in which the thermal activation of domain walls is completely neglected and the temperature dependence of coercivity is described by the temperature dependences of the anisotropy constants $K_{1,2}$, the exchange constants $A_{1,2}$ and the spontaneous magnetizations $M_{1,2}$ (1—matrix, 2—pin site). Since the pin interaction potential is dependent on the magnetic parameters, the temperature dependences of these parameters enter into the energy barriers that the domain walls have to pass on breaking away from pinning sites. There are many papers [13]–[21] that study the coercivity in this way. Some typical works are described below.

Hilzinger [17] had studied the influence of planar defects on the coercivity of hard magnetic materials. In his model, it was assumed that the domain wall and the planar pinning site are infinitely extended and are parallel to each other. The domain wall is divided into three regions (Fig. 1.4, where W is the pin width). The pin boundaries are at $z = 0$ and $z = W$, the corresponding orientations of spins are θ_1 and θ_2 respectively. In the usual micromagnetic description, the wall energy per unit area can be expressed as a function of θ_1 , θ_2 , H and C , where C is an integral constant which can be

evaluated by an expression of θ_2 as a function of θ_1 , H and C . In order to avoid the problematical boundary condition for the derivative $d\theta/dz$, he determined θ_1 and θ_2 by minimizing the total wall energy with respect to these variables. The two equations thus obtained, together with that to determine C , were used to calculate θ_1 , θ_2 and C . The coercivity was then considered to be the critical value of the field that no longer yields a stable solution of these equations. In the case when the planar pinning site is much thinner than the wall thickness, the coercivity can be expressed by

$$H_c = \frac{1}{3\sqrt{3}} \frac{2K_1}{M_1} \frac{W}{\delta_1} \left(\frac{A_1}{A_2} - \frac{K_2}{K_1} \right), \quad (1.13)$$

where $\delta_1 = (A_1/K_1)^{1/2}$ is a measure of the domain-wall thickness.

Friedberg and Paul [18] obtained an expression equivalent to Eq. (1.13) in a similar way. The only difference is that unlike Hilzinger, Friedberg et al. used the boundary condition $A_i(\partial\theta/\partial z)_i = \text{const.}$ to calculate the integral constant C , leading to the same results.

Both models conclude that any barrier, no matter how small, is sufficient to pin the wall in the absence of a magnetic field, the wall is at the energy minimum position in this case.

Paul [20] used the theory of solitons to derive a rigorous solution to the problem of a ferromagnetic domain-wall in the presence of planar defects. There was no restriction on the thickness of the pinning site in his calculation. He found that Eq.(1.13) is a good approximation for the coercivity when

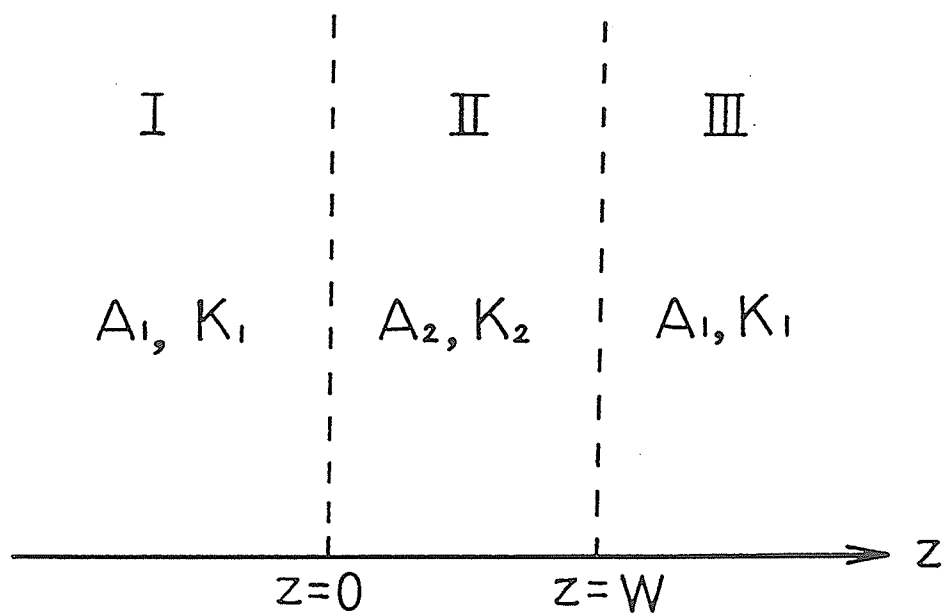


Figure 1.4: Geometry showing three regions.

the pin width is much less than wall thickness. However as the pin width increases, the coercivity deviates from the value predicted by Eq.(1.13). As is shown in Fig.1.5, the deviation is very large when the pin width is close to the wall thickness.

In ferromagnetic materials with large anisotropy constants ($K_1 > 10^8 \text{ erg/cm}^3$) the domain-wall width is in the order of magnitude of a few lattice constants. A discrete spin model is necessary to calculate the coercivity originated from the Peierls potential. Hilzinger and Kronmüller, by neglecting the thermal activation of the wall, calculated the coercivity to be

$$H_c = \text{const.}(A_1/D)\exp[-\pi\delta_1/D], \quad (1.14)$$

where D is the distance between two neighbouring atomic layers. This expression predicts the maximum coercivity originating from the Peierls potential to be about 100 Oe at 4.2 K for SmCo_5 alloy [23]. This value is much less than the experimental values which are of the order of 10 kOe. This means that the high coercivity of SmCo_5 hard magnet does not originate from the Peierls potential.

1.6 Discussion

Gaunt did notice the temperature dependence of the energy barriers when he studied the magnetic viscosity of a ferromagnet [24]. But unfortunately in his model describing the temperature dependence of coercivity, this effect

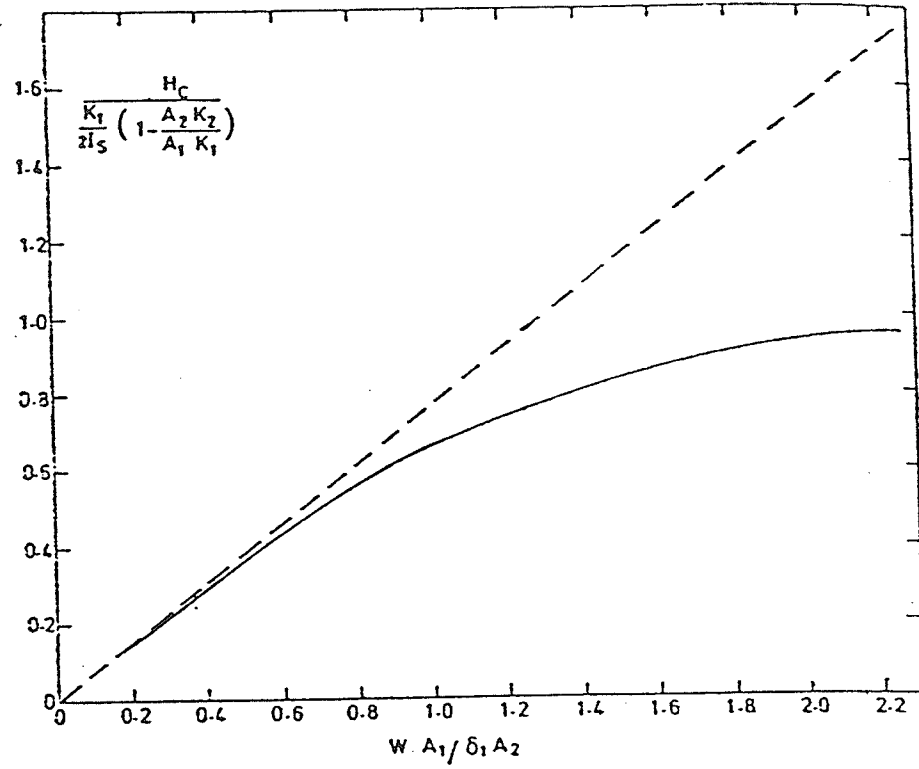


Figure 1.5: Graph of the reduced coercivity as a function of the ratio of pin width to wall half-width and the ratio of exchange constants in the matrix and at the pin. The solid line represents the rigorous solution. The dashed line is the approximate solution given by Eq.(1.13).

is completely neglected. The critical field H_0 is taken as a constant. Assuming that the pin density is independent of temperature, H_0 in Eq.(1.8) could be a constant only in two particular cases: (1) all f , M and γ are independent of temperature; (2) the numerator has the same temperature dependence as the denominator. In general H_0 may be strongly dependent on temperature. Therefore the simple relationships between coercivity and temperature predicted in Eqs. (1. 7) and (1. 10) are questionable.

As is described in section 1.5, Hilzinger, Kronmüller and Paul neglect the thermal activation of walls in their models, although in the special case, that is, domain-wall pinning originating from the Peierls potential, Kronmüller et al. [23] accept the idea of thermal activation of domain walls. As a result their models claim that the domain walls cannot leave pinning sites or at least, cannot leave the energy minimum positions in a null magnetic field. This is certainly not acceptable since experimental work [9] has shown evidence that wall unpinning can be assisted by thermal vibrational energy associated with domain walls. Therefore, these models are also questionable, although a lot of work has applied these models.

From above arguments, a reasonable model to describe the temperature dependence of coercivity should, in general, take account of both contributions to the drop of coercivity as a function of temperature. Systematic errors are inevitable in any model which neglects either contribution unless a reason can be given.

The author of this thesis has studied different kinds of pinning models

in some detail and tried to incorporate the two contributions into a single theory. It is found that Gaunt's model is suitable for this purpose. The theoretical calculations in the thesis are mainly devoted to expressions of maximum pinning force as functions of the anisotropy constants, the exchange constants and the spontaneous magnetizations in different situations. Further, the validity of the potential of the form in Eq.(1.1) is examined. Experimental work is necessary to confirm the proposed models. Coercivity measurements and transmission electron microscope (TEM) observations provide a way to test the new models.

Chapter 2

Experimental techniques and related theories

2.1 Magnetization measurement

2.1.1 Demagnetizing factor

The demagnetizing factor, D , which is determined by the geometry of the specimen, is defined by Bozorth [25] in the relationship

$$H_D = DM$$

where H_D is the demagnetizing field. The magnetization, M , of an ellipsoidal specimen is uniform in a uniform applied field, H_a . Therefore the effective field within the specimen is then

$$H = H_a - H_D \quad (2.1)$$

In a magnetization measurement for the coercivity only, the correction of field within the specimen by Eq. (2.1) is not necessary since when $H = H_c$, $M = 0$, $H_D = 0$ and then $H_a = H_c$.

2.1.2 Extraction method

When a magnetized specimen is placed in a sensing coil and then withdrawn, the resulting change in the magnetic flux through the coil generates an electromotive force in the coil. This force, when measured, enables us to calculate the magnetization of the specimen. This technique is called the extraction method; it is used in the coercivity measurement and is illustrated below.

Let the specimen with magnetization M have a cross sectional area A_s which, for simplicity, is equal to that of sensing coil. When the specimen is inside the sensing coil, the flux through A_s is

$$\phi_1 = (H + 4\pi M)A_s. \quad (2.2)$$

By making use of Eq. (2.1), ϕ_1 can be reexpressed as

$$\phi_1 = [H_a - (D - 4\pi)M]A_s. \quad (2.3)$$

When the specimen is withdrawn from the coil, the flux through the coil is now given by

$$\phi_2 = H_a A_s. \quad (2.4)$$

Therefore the change in flux is

$$\Delta\phi = (D - 4\pi)A_s M. \quad (2.5)$$

The e. m. f. generated in the coil due to a flux change is

$$\varepsilon_i \propto d\phi/dt'$$

where t' is the time.

$$\Delta\phi = \int (d\phi/dt') dt'$$

or

$$\Delta\phi \propto \int \varepsilon_i dt',$$

and

$$M \propto \int \varepsilon_i dt'. \quad (2.6)$$

Thus the integral with respect to time of the e.m.f. generated in the sensing coil is directly proportional to the magnetization of the specimen.

2.1.3 Electromagnet

A water-cooled Magnion electromagnet driven by a 25 A regulated power supply was used to obtain fields of magnitude up to 16 kOe. Polarity of the field could be reversed by a switch in the output of the power supply. A 9 mm hole was bored in the pole faces along the axis of the core to facilitate insertion and removal of the specimen. The field was set by a potentiometer in the power supply and it was calibrated by a flip coil flux meter.

Ideally the field should be uniform in the whole volume occupied by the specimen. In practice, however, it is not true. The fractional change in field in the longitudinal direction was determined, using a small sensing coil, to be

$$\Delta h_{d_l} = -0.00126 d_l^3$$

where d_l is in mm. In the radial direction the fractional change was

$$\Delta h_{d_r} = 0.00121 d_r^2$$

where d_r is in mm. Therefore the maximum fractional changes across the entire sample, taking $d_l = d_r = 1.5mm$, are

$$\Delta h_l(max) = -0.00425$$

and

$$\Delta h_r(max) = 0.00272,$$

which are negligible.

2.1.4 Image effect

Because the pole faces are made of material of high permeability, magnetic images are formed. The measurement made with an electromagnet has to be corrected for this image effect, since images interact with the specimen and increase the flux density around it (Fig. 2. 1). The image effect depends strongly on the field, causing the magnetic output for a constant moment sample to decrease by as much as 5 % at 16 kOe.

A small coil with a constant current to produce a constant moment was used to determine the correction factor L. The ratio of the real moment to the measured moment at a particular field gives the image correction factor at that field. The variation of this factor with field is shown in Fig. 2. 2.

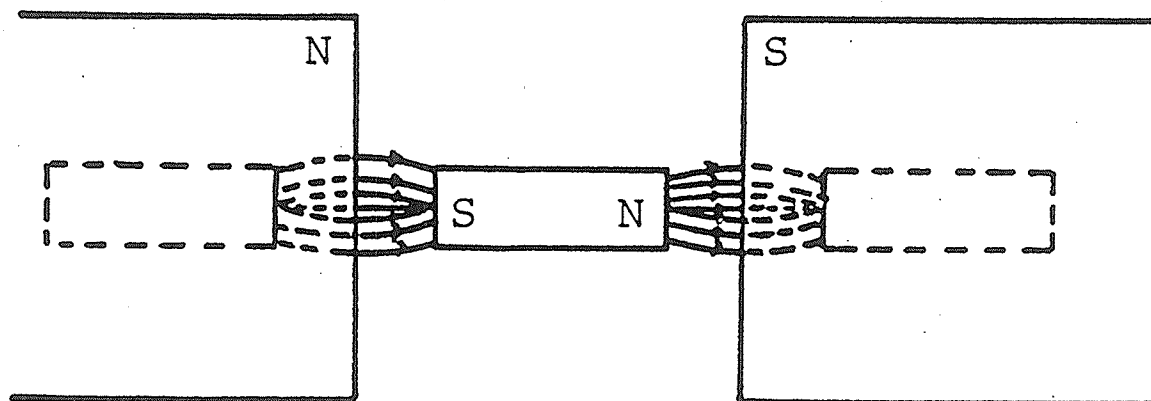


Figure 2.1: Image effect in the poles caused by the magnetized specimen.

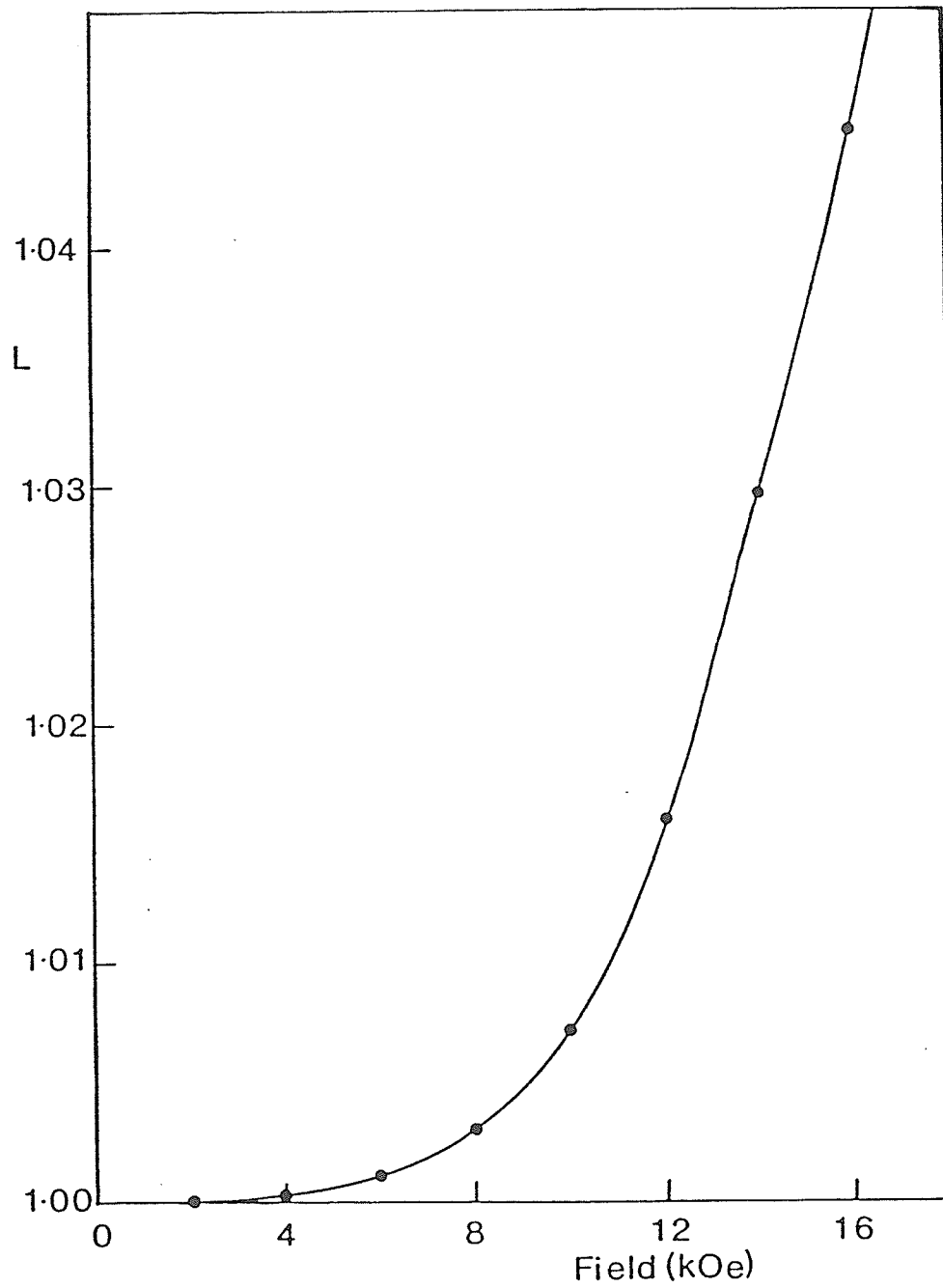


Figure 2.2: Dependence of the image correction factor, L , on the applied field.

2.1.5 Coil-Integrator system

A 15000 turn coil of B. S. 44 gauge copper wire was placed between the pole faces with axis along the field direction. This served as the pick up coil. When the sample was removed from the coil to the interior of the magnet core an e.m.f. was generated on the coil due to the flux change (Eq. 2.5). This e.m.f. was integrated using an electronic integrator (Fig. 2.3) based on a chopper stabilized operational amplifier. For static measurements the switch S_2 was open and the output voltage was displayed on a digital voltmeter and for magnetic viscosity measurement the switch S_2 was closed and the output was recorded on a chart recorder. The switch S_1 was normally closed. In a magnetic viscosity measurement, it was open only when the specimen was completely inserted into the coil. The whole system was calibrated using an annealed iron specimen. Taking 1707 cgs as the saturation magnetization moment of iron at room temperature [26], the calibration factor was determined to be 201 ± 1 erg/Oe per volt.

2.1.6 Cold finger

A cold finger was used to achieve a temperature range from 77 K to room temperature (300 K). This consisted of four coaxial stainless steel tubes as shown in Fig. 2.4. Liquid nitrogen flows down the inner tube which is isolated by a vacuum space. The two inner tubes are soldered to an oxygen-free high conductivity copper block. At the top of the block there is a hole for the escape of nitrogen gas through the space between the second and third tubes.

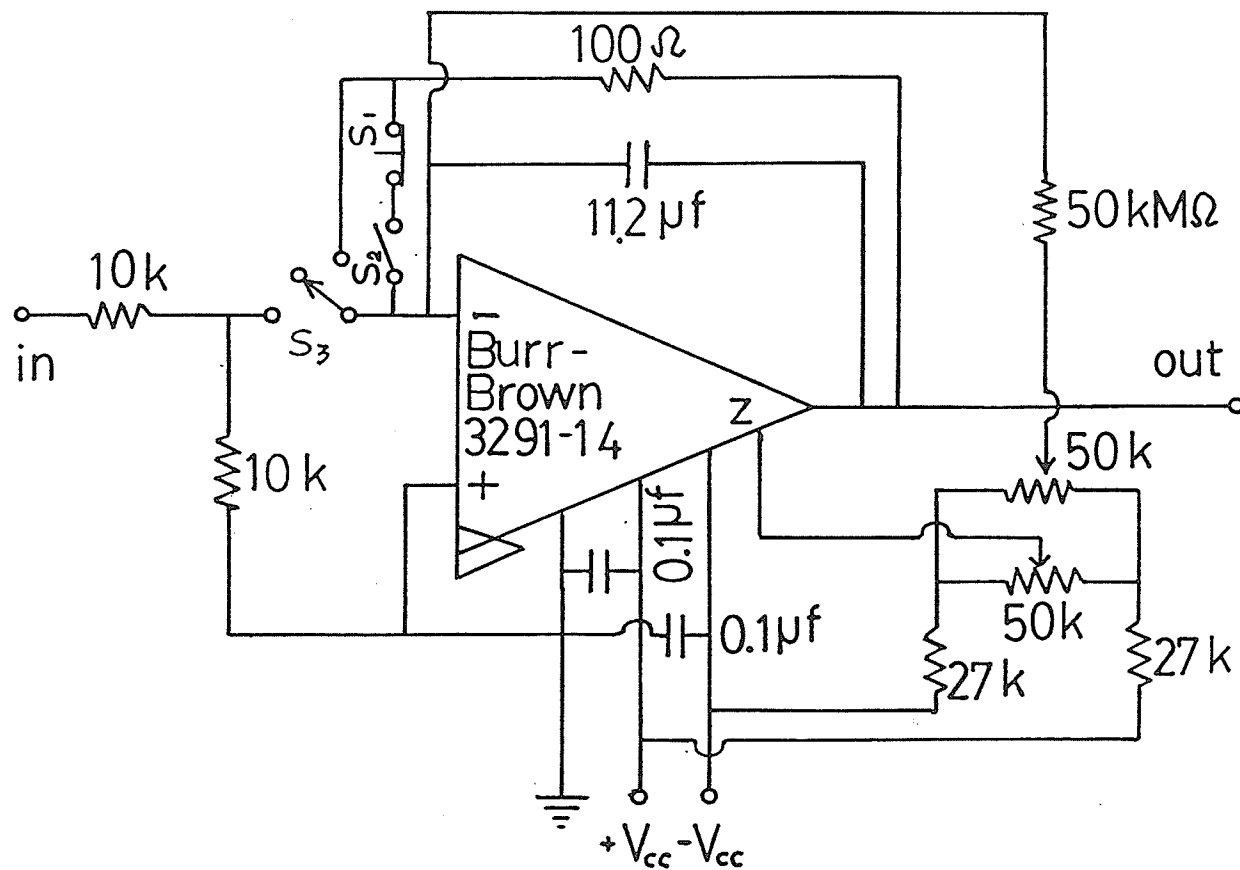


Figure 2.3: Schematic illustration of the electronic integrator.

Teflon spacers prevented the tubes from coming into contact with each other. Bellows reduced the possibility of damage caused by thermal expansion and contraction.

The copper block is used both as the heater and the sample holder. The heater's coil is made of nichrome wire having a resistance of 40 Ω . At the end of the heater, there is a Au 0.03 % at. Fe thermocouple used for the temperature controller. The sample holder is made of aluminum. At its end there is a copper-constantan thermocouple for measuring the temperature of the sample.

2.2 X-ray measurement

A crystal consists of a three-dimensional array of atoms. X-rays are scattered from these atoms in a manner similar to that in which light waves are scattered from the ruled lines of a grating. The condition for additive interference between the X-ray scattered from a set of lattice planes is the Bragg law given by

$$2d_{hkl}\sin\psi = n_r\lambda, \quad (2.7)$$

where d_{hkl} is the spacing of consecutive lattice planes, ψ is the angle the incident beams make with the planes, n_r is the order of the reflection and λ is the wavelength of X-rays. For a cubic lattice,

$$d_{hkl} = a/(h^2 + k^2 + l^2)^{1/2}, \quad (2.8)$$

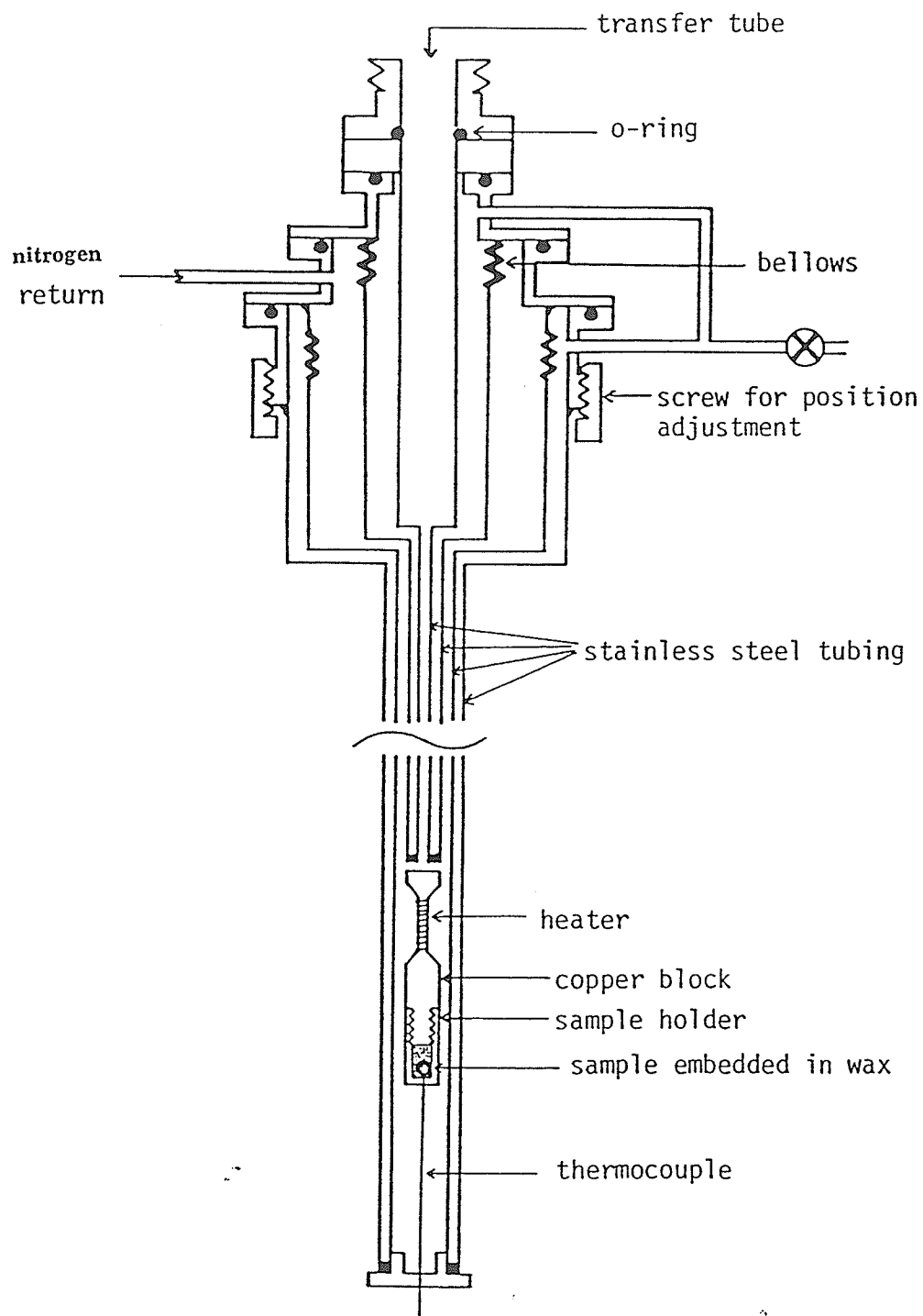


Figure 2.4: Diagram of cold finger.

where a is the lattice constant.

A fairly simple way to determine the lattice constants is to take powder photographs. For a given wave length, the deviation of a particular reflection is 2ψ , and therefore corresponding reflections from all crystals in the powder specimen must lie on a cone with its apex at the specimen, axis in the direction of the X-ray beam, and making a semi-vertical angle 2ψ (Fig. 2.5). The specimen is usually surrounded by a narrow strip of film which intercepts a great number of cones of reflection. Only a small part of each cone is recorded, but all reflections except those with ψ just greater than 0° or just less than 90° can reach the film (Fig. 2.6).

From the Bragg law, we find

$$\frac{\Delta d_{hkl}}{d_{hkl}} \propto \text{ctg}\psi. \quad (2.9)$$

If we plot d_{hkl} against $\text{ctg}\psi$ on a linear graph and then extrapolate to the point where $\text{ctg}\psi = 0$, we will get a very accurate value of d_{hkl} , according to Eq. (2.9). In reality however, the so-called N-R function is used instead of $\text{ctg}\psi$, which is given by

$$N - R = \frac{\cos^2\psi}{2} \left[\frac{1}{\sin\psi} + \frac{1}{\psi} \right]. \quad (2.10)$$

The current was 20 mA at 40 kV for a copper tube and 16 mA at 40 kV for a cobalt tube.

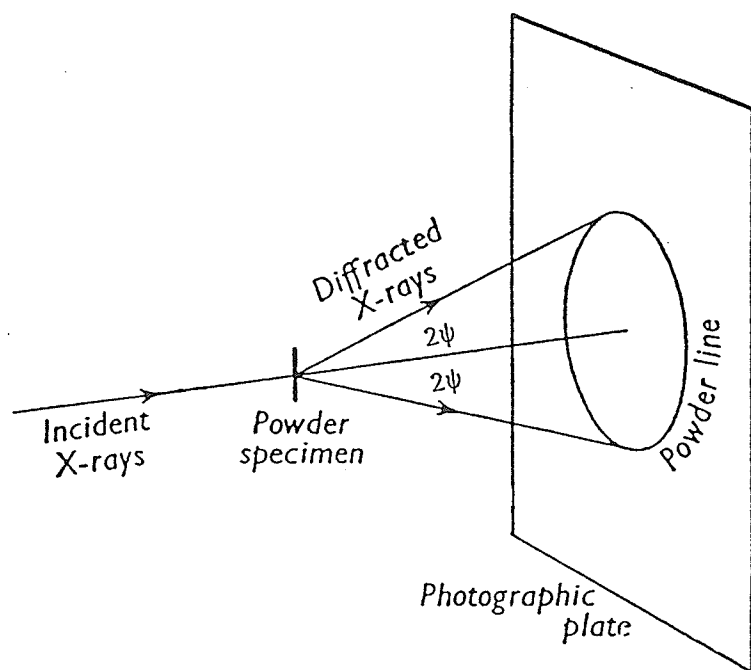


Figure 2.5: Diagram showing the reflection cones of a powder specimen.

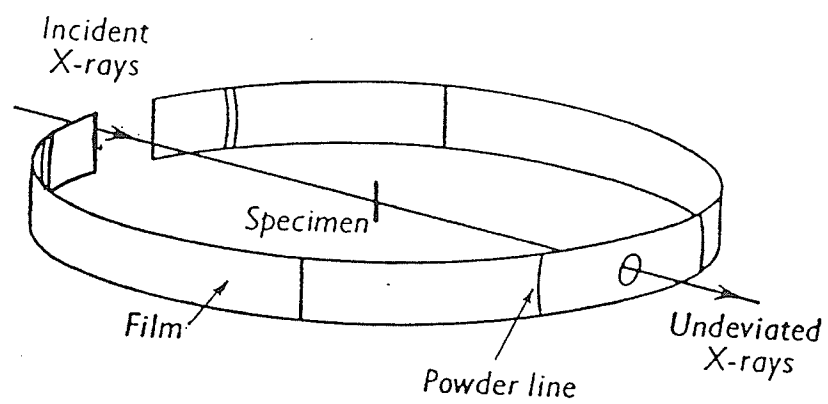


Figure 2.6: Diagram illustrating a powder photograph camera.

2.3 Electron microscopy

2.3.1 Specimen preparation

Good specimen preparation is the key to produce good electron micrographs. The specimens were disks of 3 mm in diameter, mechanically thinned to ~ 0.15 mm in thickness and then electropolished by a Fishione twin-jet electropolisher (Fig. 2.7). When the perforation happened at the center of the specimen the polishing was stopped automatically. Then the specimen was washed in methanol and finally was washed in water for at least 10 minutes to minimize contamination. A good specimen usually had shiny surfaces on both sides. The specimens were stored in vacuum to minimize oxidation.

2.3.2 Schematic illustration of TEM

If an electron is accelerated through a potential difference E_e , the wavelength of the electron is

$$\lambda = \frac{12.26}{E_e^{1/2}(1 + 0.9788 \times 10^{-6} E_e)^{1/2}} \quad (2.11)$$

where E_e is measured in volts and λ is in Å. For $E_e = 120$ kV, we find $\lambda = 0.0335$ Å. Such a wavelength makes the theoretical resolution of the TEM as high as 2 Å.

The transmission-electron microscope (TEM) used is the JEM120CXII TEM, manufactured by JOEL Limited. It consists of an electron gun and

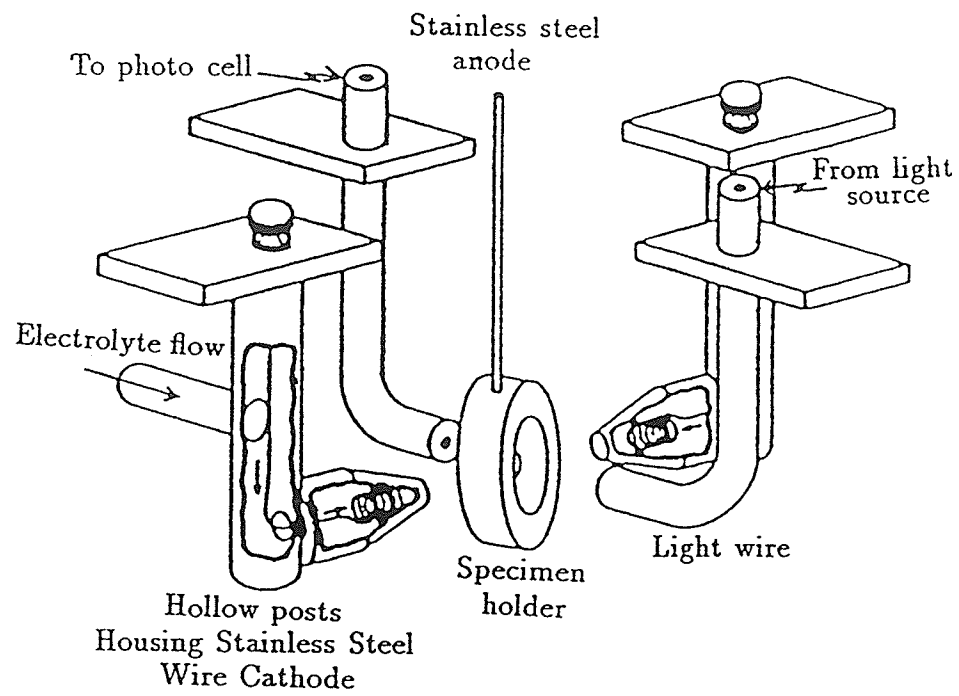


Figure 2.7: The essential components of a twin-jet electropolisher.

an assembly of lenses as shown in Fig. 2.8. The electron beam generated by the electron gun is accelerated by the electric potential difference between the filament and the anode. The lenses are usually magnetic ones because of their smaller optical aberrations and their freedom from the usual trouble associated with high voltages for electrostatic lenses. The total magnification is the product of the magnifications of the objective lens, the intermediate lens and the projector lens. The apertures are used to allow certain beam/beams to pass through and at the same time to block other unwanted beams. The vacuum inside the TEM is usually held at 10^{-7} Torr.

2.3.3 Selected area diffraction

When a selected-area aperture is inserted as shown in Fig. 2.8, only part of beam passing through the hole in the aperture can reach the final screen. The diffraction pattern of this area only is therefore observed. This technique is known as selected-area diffraction. It enables diffraction patterns to be taken from small areas of the specimen, so that the correlation between the features observed on the micrographs and the crystallograph of the specimen can be made. It is also very useful for identifying phases in an inhomogeneous specimen.

Since the wavelength of an electron is very short, it is relatively easy to index the diffraction pattern. Let's consider a simple electron-diffraction camera, with an electron beam striking a specimen and being diffracted to form a diffraction spot on the screen at a distance D_p from the center of the

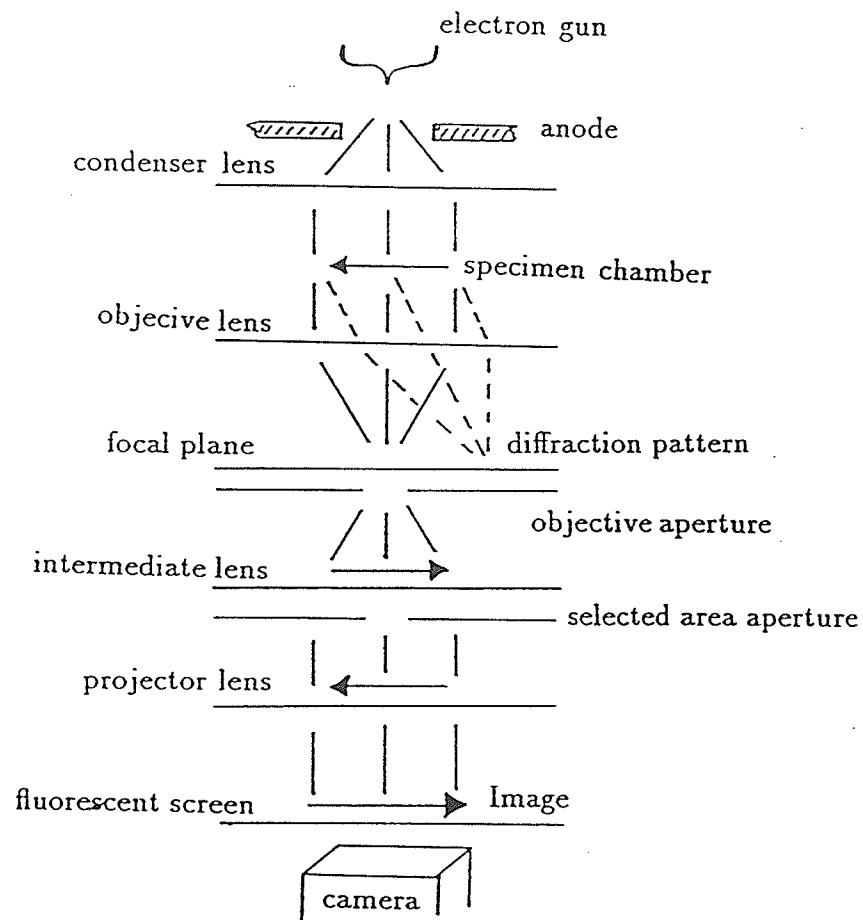


Figure 2.8: Schematic illustration of a TEM.

diffraction pattern as shown in Fig. 2.9. The camera length L and D are related by the expression

$$tg2\psi = D_p/L \quad (2.12)$$

where ψ is the incident angle. Since ψ is very small, we have from the Bragg law in Eq. (2.7), setting $n_r = 1$,

$$\psi \approx \lambda/2d_{hkl}. \quad (2.13)$$

Combining Eqs. (2.12) with (2.13), we obtain

$$D_p = L\lambda/d_{hkl}. \quad (2.14)$$

The product, $L\lambda$, is referred to as the camera constant. For a real TEM, L is a fictitious length since there are electron lenses after the specimen. To index the diffraction pattern, all we need to do is to use a ruler to measure a set of values of D_p for different diffraction spots, then by making use of Eq. (2.14), to calculate the different ratios of the d-spacing of sets of lattice planes, and finally from these ratios, to find the lattice structure and constants.

2.3.4 Bright field image and dark field image

Fig. 2.8 illustrates the formation of an image. The almost parallel illuminating beam of electrons is scattered by the specimen. In the case of crystalline materials this scattering takes the form of one or more Bragg diffracted beams travelling at small angles (~ 1 or 2 degrees) with the incident beam, which

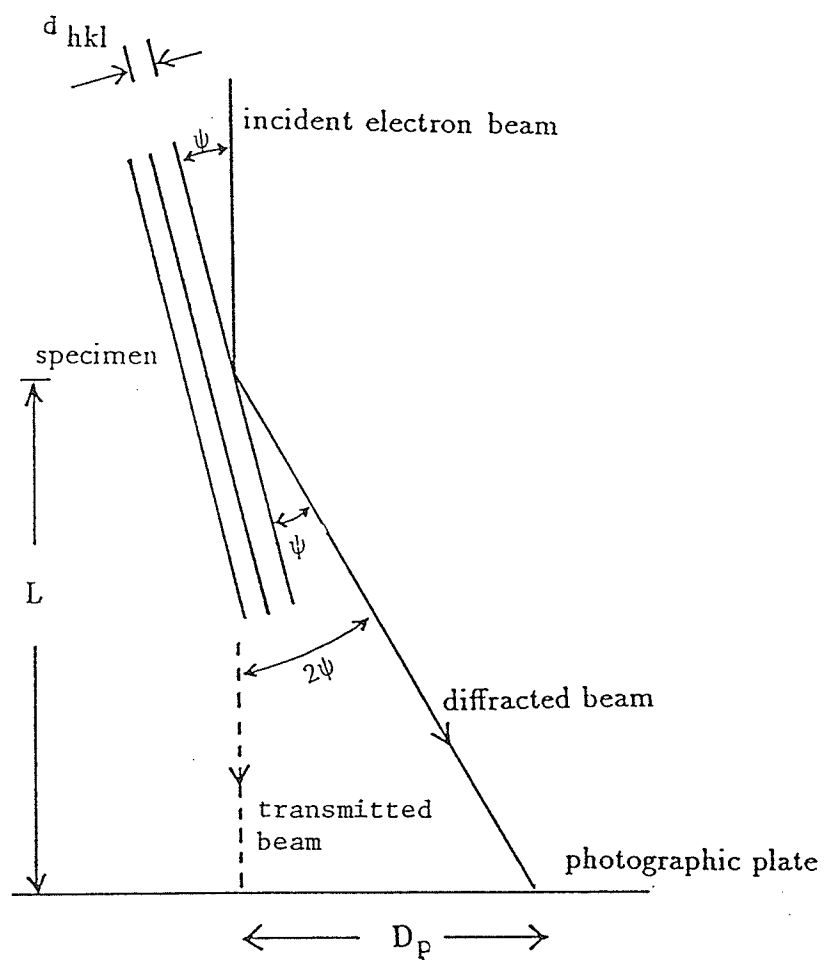


Figure 2.9: A simple electron diffraction camera.

are focused by the objective lens to form a transmission diffraction pattern in its back focal plane. An objective aperture is placed in this plane. If this aperture does not allow any Bragg deflection to pass through to the screen, the image is formed by the direct beam, as shown in Fig. 2.8. This image is called the bright-field image. Images can also be formed by any one diffracted beam by either displacing the aperture to receive this beam, or by tilting the illumination so that the required beam passes down the axis of the objective lens. The latter is preferable since it can largely reduce the effect of spherical aberration on the resolution of the image. The resulting image is known as a dark-field image which is very useful because it reveals which region of the specimen contributes to the diffracted beam.

2.3.5 Magnetic material observation device

It is not easy to observe the magnetic structures of ferromagnetic materials with a conventional transmission-electron microscope. The main reason is that the magnetic field of the objective lens interferes with the magnetic structure of the specimen and very often saturates the specimen. On the other hand, the field of the lens is often disturbed by the magnetic specimen. To avoid these problem, the current of the objective lens is turned off during the observation. But the resolution is greatly reduced in this way.

Therefore, a special type of objective lens (EM-AMG) was used to observe the magnetic materials. The pole-piece and its field profile are shown in Fig. 2.10. The field intensity at the specimen position is less than 5 Oe

along the axial direction and 0.3 Oe in the lateral direction. However it is essential to turn the objective lens current off when the specimen is exchanged. The maximum resolution of this special objective lens is about 12.6 Å.

2.3.6 Lorentz microscopy of magnetic domains

The traditional methods of observing domains and domain walls are the Bitter method and techniques based on the Kerr and Faraday effects. Lorentz microscopy, however, can reveal domains/domain walls and the pinning sites simultaneously. This makes the Lorentz microscopy very useful in the study of domain-wall pinning.

An electron in a magnetic field is subjected to a force, F_l , the Lorentz force, given by $\vec{F}_l = -(\vec{v}_e \times \vec{B})e/c_l$ where $\vec{v}_e$ is the velocity of the electron, c_l is the light speed in vacuum and $\vec{B}$ is the magnetic induction equal to $\vec{H} + 4\pi\vec{M}$. The external magnetic field on the specimen position in the TEM is negligible, so that the $\vec{B}$ is mainly determined by $\vec{M}$ of the specimen.

Consider a foil with 180 degree domains in which the magnetization is parallel to the plane of the foil and the domain walls, as well as in opposite directions in alternate domains (Fig. 2.11). The electron beam will be deflected in opposite directions in adjacent domains, leading to a deficiency of electrons at P_1 and an excess of electrons due to overlap at P_2 . If we therefore observe the foil out of focus in the plane P_1P_2 , the position of the domain walls will be revealed by black (excess) or white (deficiency) lines on the pho-

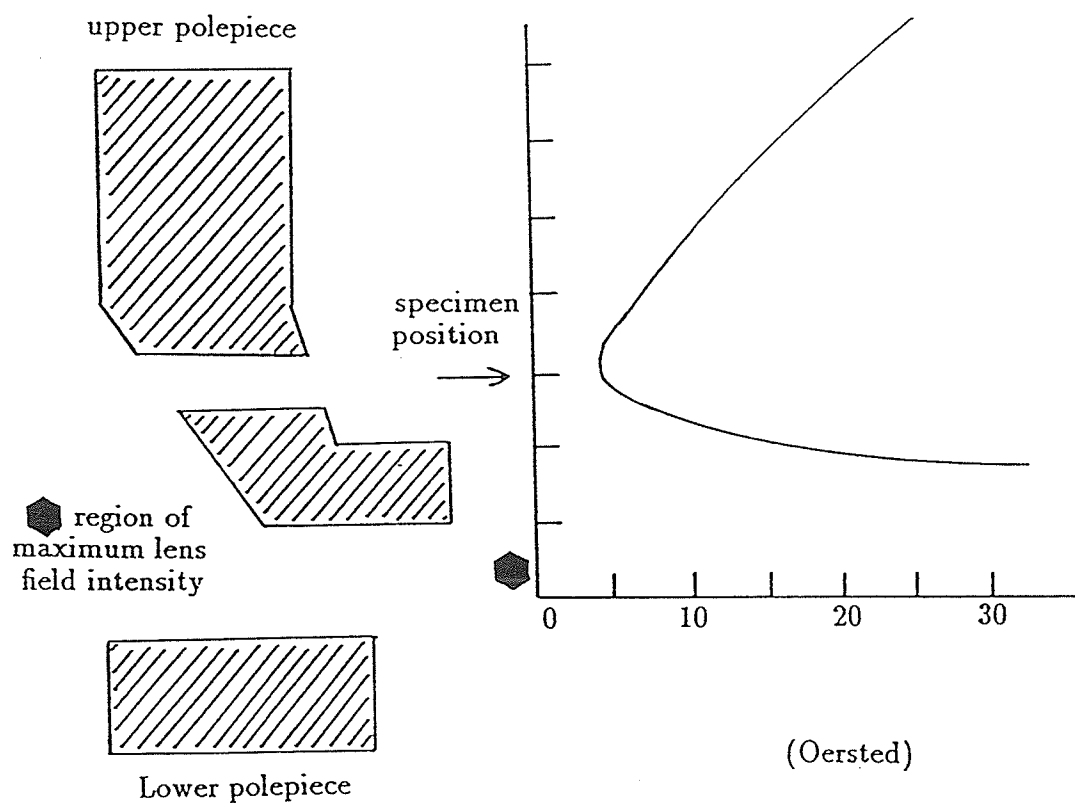


Figure 2.10: Diagram of a magnetic material observation device.

tographic plate. If the image is underfocused rather than overfocused (i.e. if the image is observed in plane P_3P_4), the white lines change into black lines and vice versa (Fig. 2.12). The technique is called the out-of-focus method.

Another technique to reveal magnetic contrast is the displaced-aperture method. Since the deflections caused by alternate domains in Fig. 2.11 are of opposite sign, the emergent electron beam is split into two parts travelling in two different directions. By displacing the objective aperture in the back focal plane it is possible to stop out one of the two divided beams. As a result alternate domains will now appear bright and dark in the image.

In the present work, only the out-of-focus method was used.

2.3.7 Calibration of the TEM

The calibrations of both the magnification and the camera length were not critical in the present work, because the diffraction pattern could be indexed without knowing the camera length L and a few percent of error in magnification has little effect on explaining the images. But the calibration for the rotation of the image with respect to the diffraction pattern is very important. This rotation depends on the total magnification of the final image and the camera length L .

The extent of image rotation is determined experimentally by using a molybdenum trioxide, MoO_3 , crystal supplied by the manufacture. This kind of crystal is known to have crystal edges perpendicular to the $[001]$ direction in the orthorhombic crystal lattice. A diffraction pattern and a micrograph

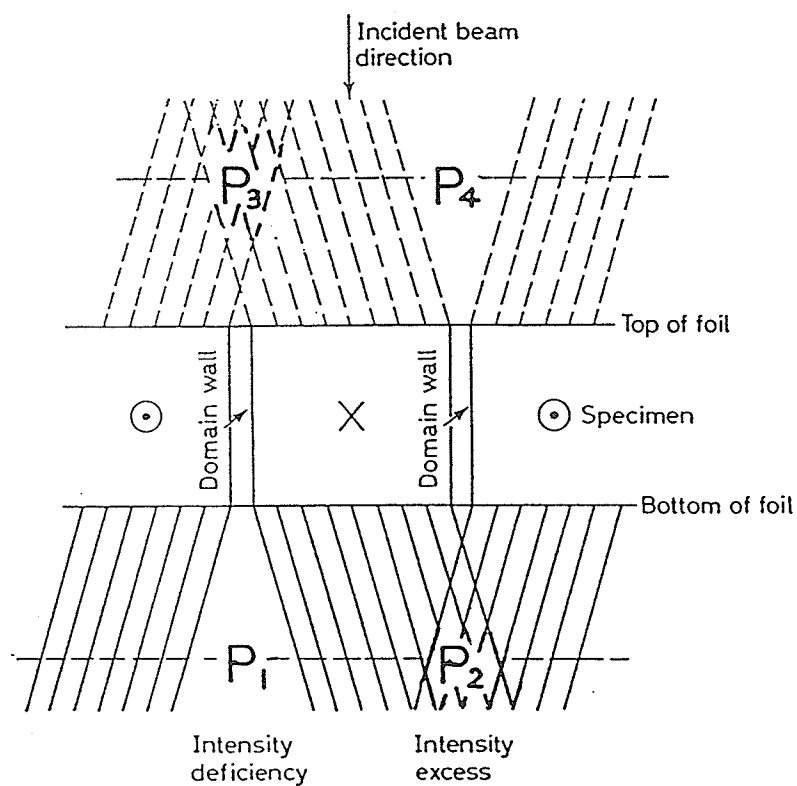


Figure 2.11: Observation of magnetic domains by the out-of-focus method. Electrons are deviated in opposite directions in adjacent 180 degree domains, leading to a deficiency of electrons at P₁, and an excess at P₂. The contrast of the virtual images at P₃ and P₄ is reversed.



Figure 2.12: The magnetic contrast reverses when the overfocusing changes to underfocusing.

Table 2.1: Angles of rotation at different operating conditions.

Voltage, Camera length	Mode	Angle (degree)
120 kV, 180 cm	MAG 100 k	58.5 c.c.w
	MAG 82 k	50.0 c.c.w
	MAG 60 k	38.5 c.c.w
	MAG 44 k	30.5 c.c.w
	MAG 31 k	23.0 c.c.w
	MAG 22 k	17.5 c.c.w
	SA MAG 31 k	69 c.w.
	SA MAG 22 k	82 c.w.
	SA MAG 15 k	85.5 c.c.w.
	SA MAG 11 k	82.5 c.c.w.
	SA MAG 8.9 k	77.5 c.c.w.
100 kV, 180 cm	MAG 100 k	57.0 c.c.w
	MAG 82 k	49.0 c.c.w
	MAG 60 k	37.0 c.c.w
	MAG 44 k	27.5 c.c.w
	MAG 31 k	21.0 c.c.w
	MAG 22 k	15.0 c.c.w

at the standard setting may now be taken using double exposures on the same photographic plate to eliminate errors in mounting the plates in the cassettes. The angles of rotation at different operating conditions are listed in Table 2.1.

Chapter 3

$\text{Pt}_{0.7}\text{Ni}_{0.3}\text{Fe}$ permanent magnet

3.1 Description of previous works

G. Hadjipanayis had done much work [27] on the $\text{Pt}_{0.7}\text{Ni}_{0.3}\text{Fe}$ ferromagnet. This compound has a disordered face-centered cubic structure after quenching from 1000°C . It forms a tetragonal ordered structure of the CuAu type at lower temperatures ($\sim 700^\circ\text{C}$) with Pt/Ni atoms at $[0\ 0\ 0]$ and $[1/2\ 1/2\ 0]$ and Fe atoms at $[0\ 1/2\ 1/2]$ and $[1/2\ 0\ 1/2]$, the value of c/a is ≈ 0.96 . The coercivity changes during this transformation. The disordered cubic phase has coercivities less than 50 Oe. The coercivity increases to a maximum and then decreases with aging time, and finally reaches a constant value of about 200 Oe. The highest coercivity corresponds to states in which the alloy consists predominantly of an ordered tetragonal phase. The Curie temperature was determined to be 462.5°C .

Gaunt [28] attempted to find the relationship between the crystal morphology and magnetic domain structure of the compound. He observed the specimen in the state of optimum hardness using a transmission electron microscope operating at 1 MV. $\{110\}$ lamellar traces were found, which are the local regions with tetragonal structure. The c axes of the tetragonal lamellae are at 90° to the c axis of the locally dominant tetragonal matrix. The domain walls showed a preference to stay at junctions between $\{110\}$ lamellae, and they were kinked somewhere [29]. Gaunt proposed that the lamellar junctions are pinning sites which result in a high coercivity for this compound.

3.2 Sample preparation

The specimens were quenched from 1000°C into ice water, then were aged at $700 \pm 2^\circ\text{C}$ for about 40 minutes to produce magnetic hardening. For TEM observation, the specimens were 3 mm diameter and ~ 0.1 mm thick after mechanically thinning. The electrolyte was of the following composition: 100 ml saturated solution of calcium chloride, 73 ml water, 45 ml hydrochloric acid, 10 ml perchloric acid and 90 ml glacial acetic acid. An ac current (60 Hz) with a density about $4\text{A}/\text{cm}^2$ was used in a twin-jet polisher. The temperature of the electrolyte was held at about -25°C during the electropolishing. The TEM was operated at 120 kV.

3.3 Coercivity

Measurements of the coercive field were made, using the appropriate inserts. The specimen was first magnetized to saturation and then gradually demagnetized. A demagnetizing curve was then obtained and the field at which magnetization disappeared, the coercive field, was thus determined. This procedure was followed for each specimen at different temperatures from 77 to 300 K.

The variation of the coercive field with temperature is shown in Table 3. 1.

Table 3.1: The coercivity of a $\text{Pt}_{0.7}\text{Ni}_{0.3}\text{Fe}$ magnet at different temperatures

T(K)	$H_c(\text{Oe})$
77.3	1680
96.0	1630
116	1580
128	1550
147	1510
163	1470
183	1450
200	1410
219	1350
239	1330
263	1280
296	1240

3.4 TEM observation

The specimens were prepared for TEM observation. The results confirm the previous work [28]. Fig. 3.1 is a in-focusing bright-field image of a (010) foil. It shows a smooth surface with no special feature. The diffraction pattern from this area is shown in Fig. 3.2. A dark-field image (Fig. 3.3) formed by a (100) superlattice reflection shows the ordered nuclei being aligned in certain directions which are identified with $[101]$ and $[\bar{1}01]$. Some of the nuclei have formed (101) and $(\bar{1}01)$ lamellae. Fig. 3.4 is an over-focusing image taken from the same area as in Fig. 3.1. Compared with Fig. 3.1, it is obvious that the new features originate from magnetic contrast. The comparatively long-range black and white contrast lines are identified with 180° domain walls. They are determined by the dominant phases, whose c

axis, and hence easy direction of magnetization is parallel to $[001]$. Their contrast changes when the focusing changes from over-focusing to under-focusing (Fig. 3.5). The 180° walls are kinked somewhere. Locally they do not always follow the $[001]$ direction because of the existence of pins. The short-range black and white contrast lines are identified with 90° domain walls associated with $\{110\}$ lamellar boundaries [28], since the easy direction of magnetization, the c-axis, rotates through 90° in crossing from matrix to lamella.

In reference [28], Gaunt claimed that when a lamellar trace crosses a 180° wall the black-white contrast of the lamella reverses. This phenomenon is, however, not always observable. A possible reason is that the width of the magnetic contrasts of 90° domain walls are the same order or even greater than that of the $\{110\}$ lamellae. Therefore the magnetic contrast of 90° walls may overlap each other, distorting the true images of 90° walls.

3.5 Pinning sites

Gaunt [28] proposed a model which considers the lamellar junctions to be pinning sites. Fig. 3.6 shows schematically how a wall can be pinned at lamellar junctions, where the arrows show the directions of the magnetization in each domain.

If a 180° wall crosses a lamellar junction it is not necessary to have a domain wall segment or uncompensated poles within the lamellae. If, however, the wall moves, an energy of interaction arises in the form of an extra

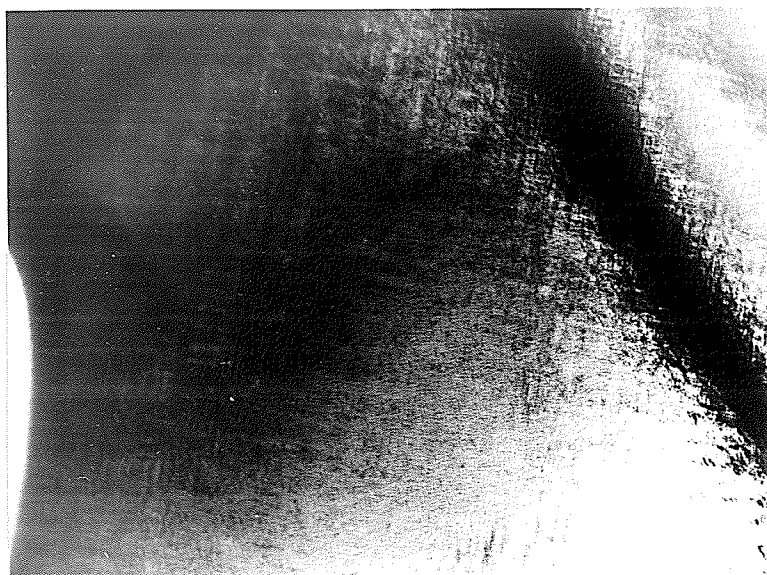


Figure 3.1: An in-focusing bright field image of a (010) foil of a $\text{Pt}_{0.7}\text{Ni}_{0.3}\text{Fe}$ magnet.

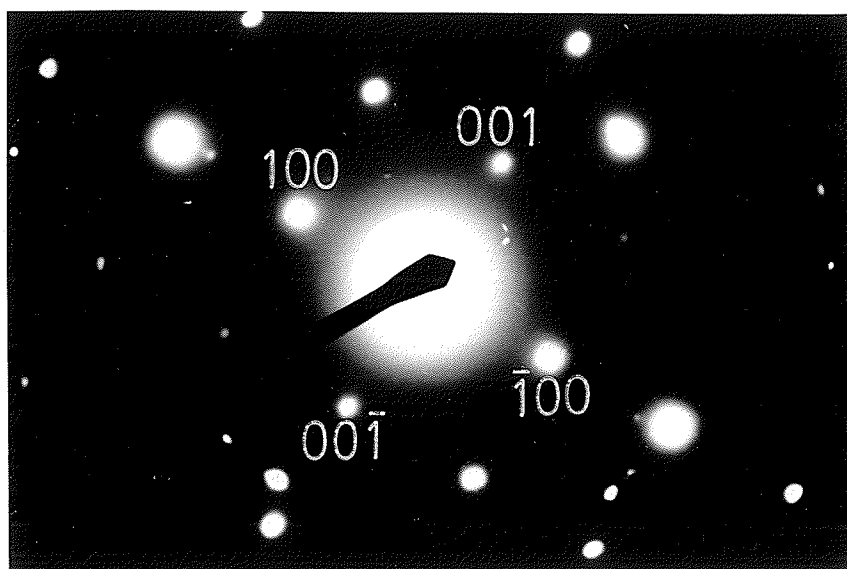


Figure 3.2: The diffraction pattern of the (010) foil for the area shown in Fig. 3.1.

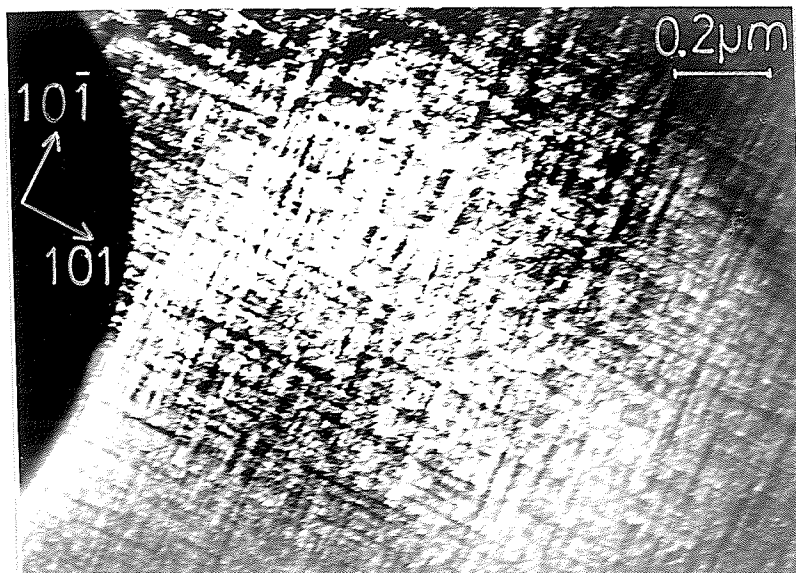


Figure 3.3: A dark field image formed by a (100) superlattice reflection, showing (101) and $(\bar{1}01)$ lamellae.

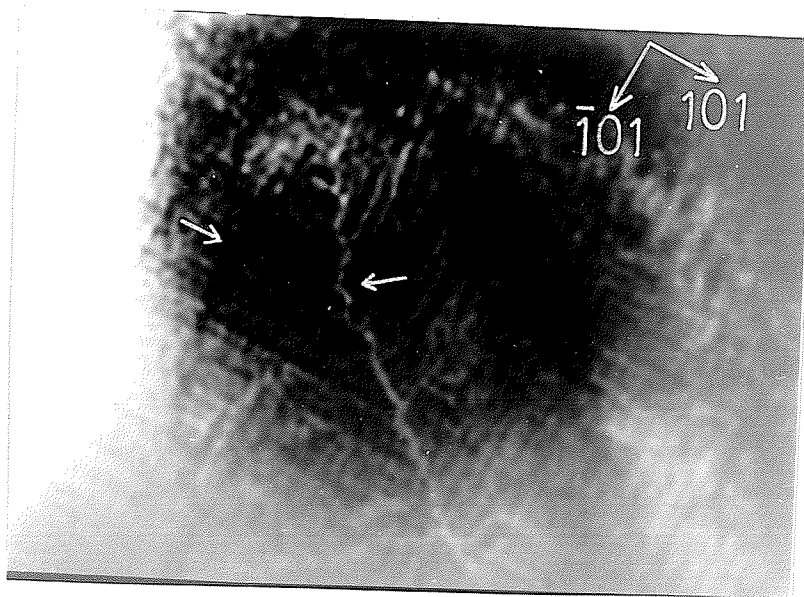


Figure 3.4: An over-focusing image taken from the same area as in Fig. 3.1.

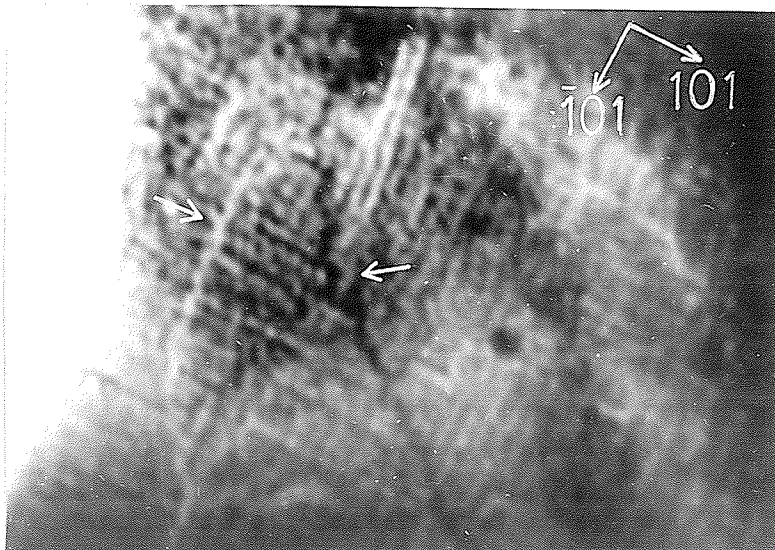


Figure 3.5: A under-focusing image compared with Fig. 3.4.

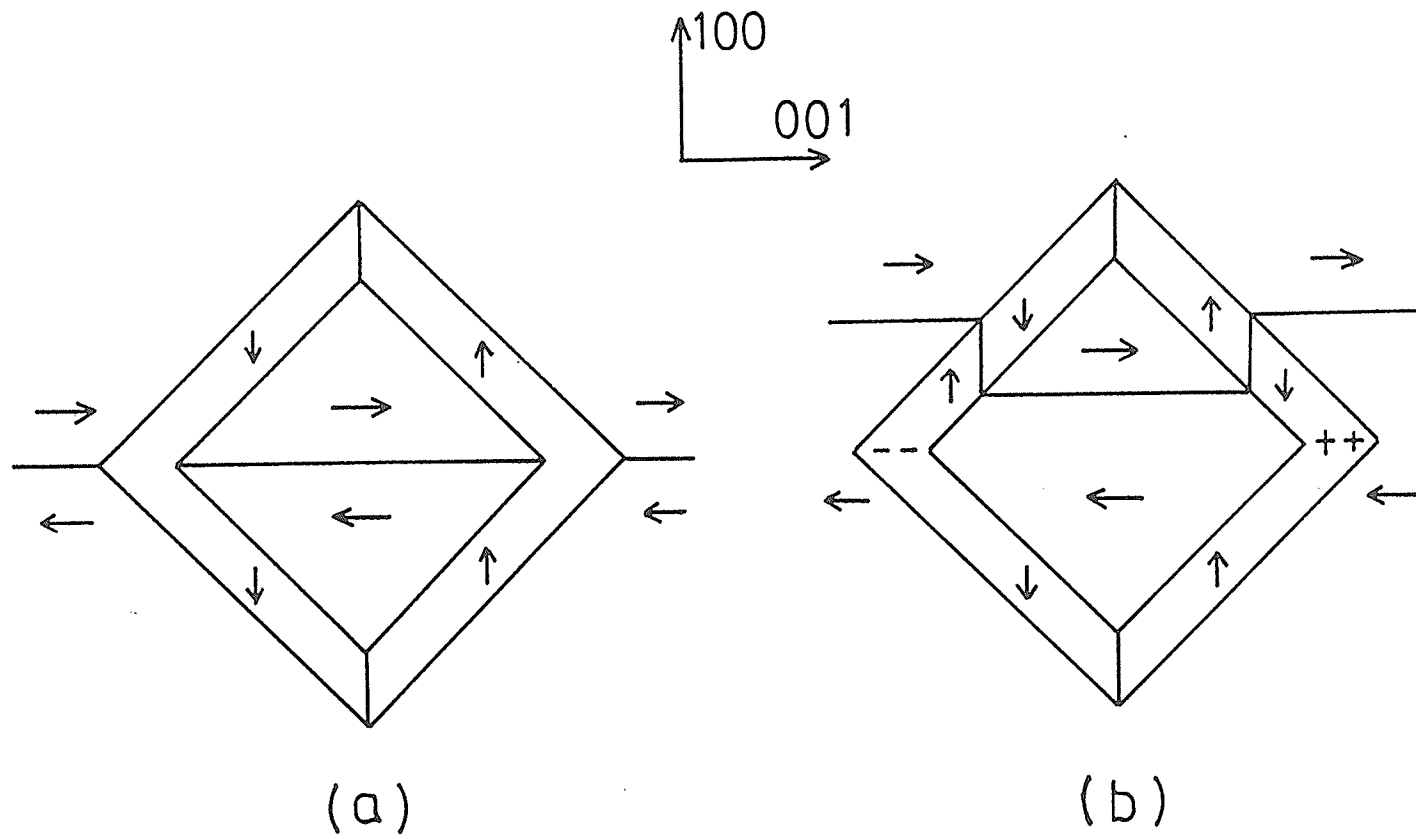


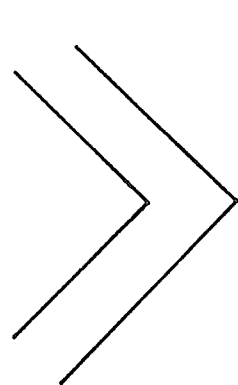
Figure 3.6: (a) 180° wall at lamellar junctions. (b) 180° wall leaving lamellar junctions.

segment of 180° wall and uncompensated poles within the lamellae. Without the assistance of external force (a driving field or thermal energy), the 180° wall will stay at lamellar junctions. Based on this model, Gaunt estimated the coercivity in the order of 1500 Oe, and in a similar way Hadjipanayis [27] got a value of 5000 Oe.

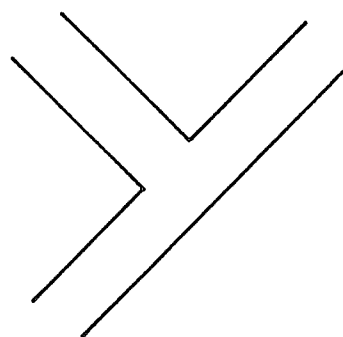
The present study extends Gaunt's idea to more general cases. In the optimum state (Fig. 3.3), the $\{110\}$ lamellae may intersect with each other, forming three different lamellar junctions, as shown in Fig. 3.7. All three types of lamellar junctions are pinning sites. Type I is the one analysed by Gaunt (Fig. 3.6). Fig. 3.8 shows how the type II lamellar junction pins the 180° wall.

Both Fig. 3.8 (a) and (b) show a 180° wall pinned at a T-shaped lamellar junction. But in (a) the wall is kinked, no magnetic pole appears; whereas in (b) a straight 180° wall will produce magnetic poles within the lamella, increasing the total energy. So state (b) is energetically unfavourable. This could be the reason why the 180° walls are observed kinked somewhere (Fig. 3.4). Fig. 3.8 (c) shows a 180° wall leaving the T-shaped lamellar junction. The extra energy arises from not only the extra wall area, but also the magnetic poles which come out inevitably within the lamella when the wall is leaving. This state is also energetically unfavourable. Only the state (a) is stable, corresponding to the minimum total energy.

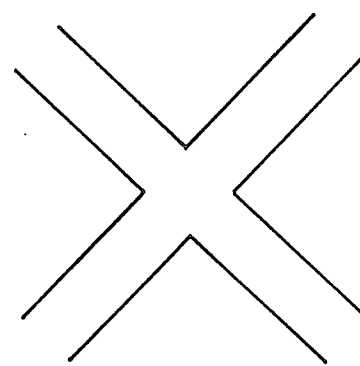
A cross lamellar junction can also pin the 180° wall. Fig. 3.9 (a) shows a 180° wall pinned at a cross lamellar junction. There is no magnetic pole



(I)



(II)



(III)

Figure 3.7: Three different lamellar junctions.

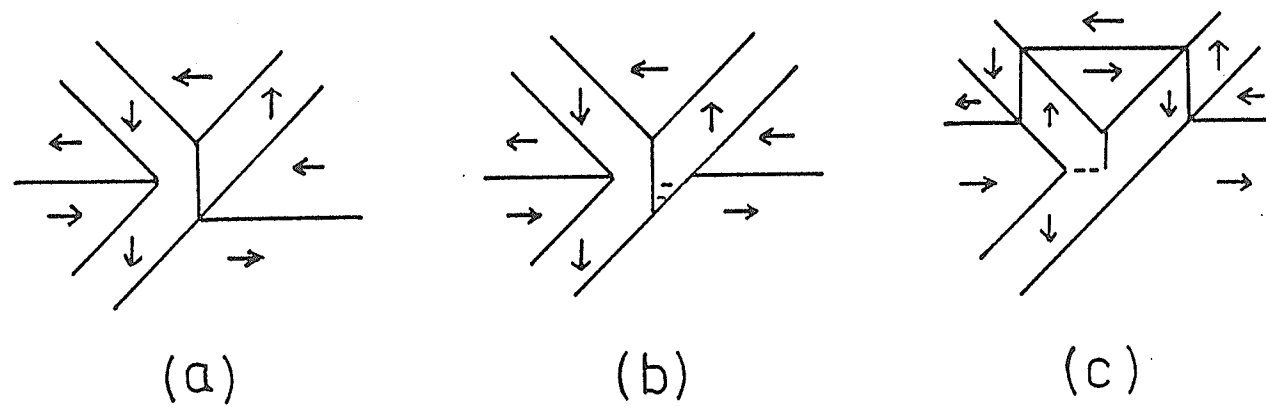


Figure 3.8: A T-shaped lamellar junction as a pinning site.

in the domains. When the wall is leaving the lamellar junction, the extra energy arises from both the extra wall area and the magnetic poles within the lamellae (Fig. 3.9 b) or the extra energy produced by the rotation of magnetization within the junction (Fig. 3.9 c). Both (b) and (c) are energetically unfavourable compared with (a) in Fig. 3.9. But (c) is even more unlikely since it needs higher energy than (b).

Experimentally it is not easy to find which kind of lamellar junction dominates the pinning process of the wall. But certainly, all three kinds of lamellar junctions exist within the specimen in the optimum magnetic state.

3.6 A model to describe the temperature dependence of coercivity

Only the type I lamellar junction in Fig. 3.7 is considered. The other two types of lamellar junctions are expected to have a pinning force of the same order as that of type I. In view of Eqs. (1.7), (1.8) and (1.10), it is essential to evaluate the maximum pinning force per pin. The model to do this is shown in Fig. 3.10.

On breaking away from the pinning sites, a domain wall has an extra domain-wall energy and an extra demagnetizing energy which are estimated as follows.

A. Evaluation of the extra domain wall energy

Let d_2 be the dimension of a lamellar junction in the direction which

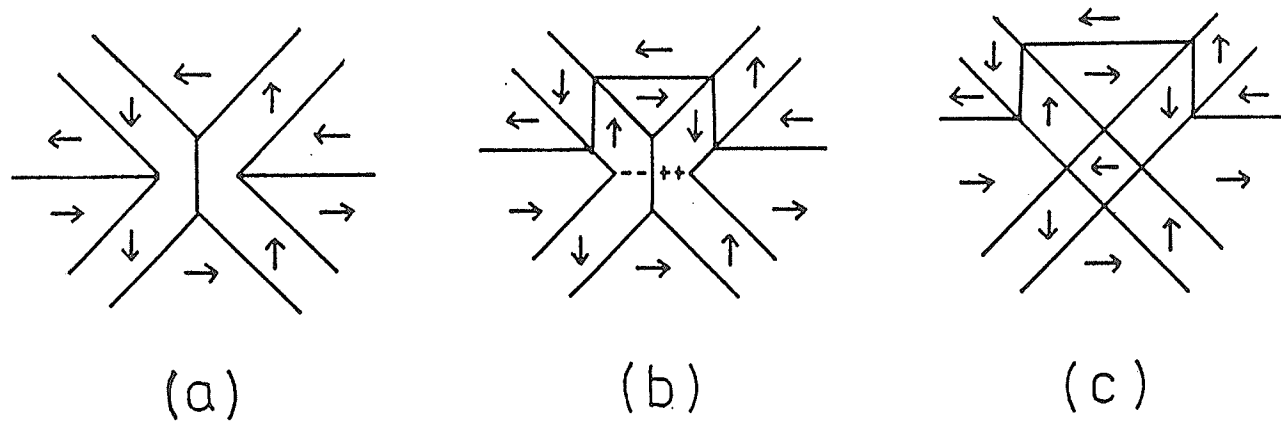


Figure 3.9: A cross lamellar junction as a pinning site.

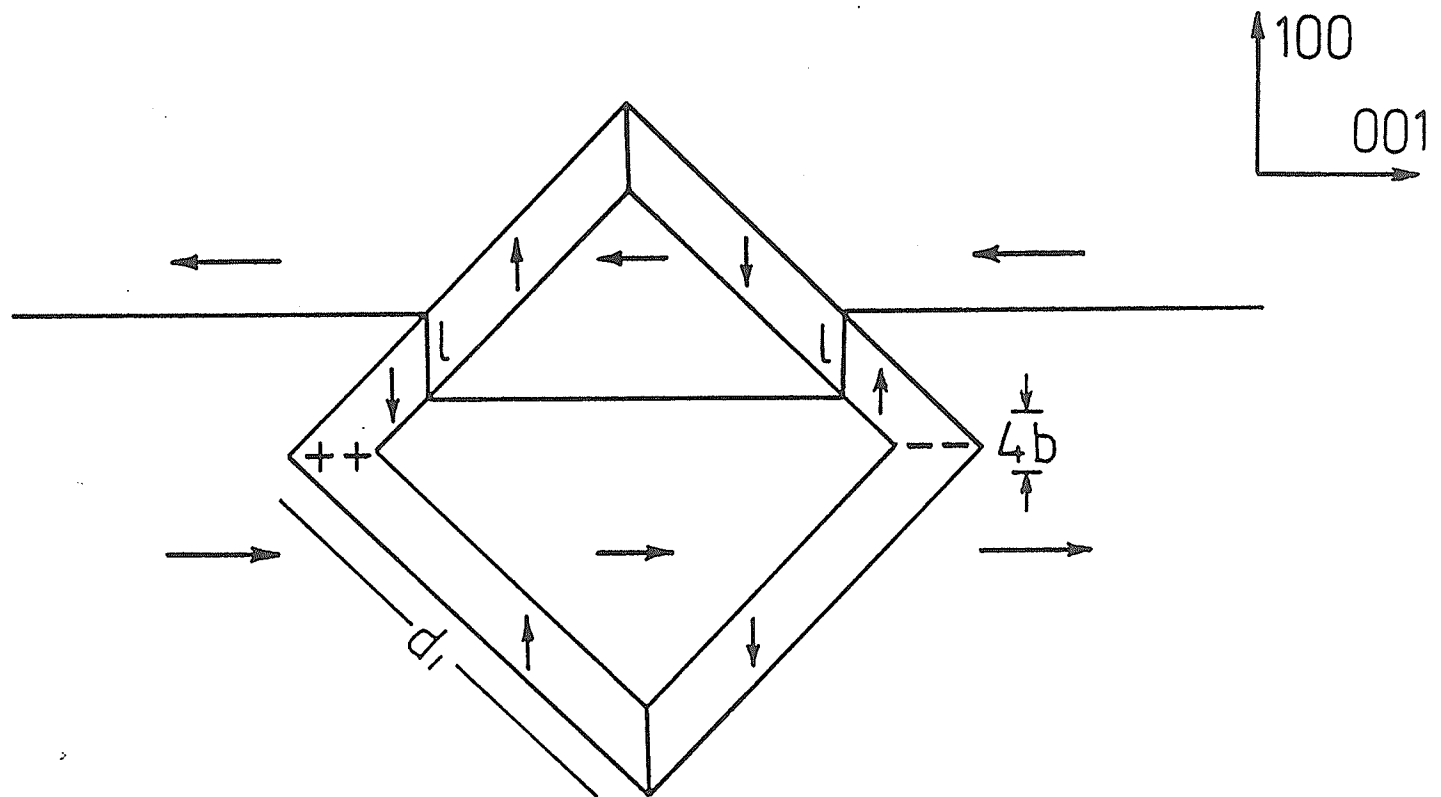


Figure 3.10: A model to calculate the maximum pinning force.

is perpendicular to the paper (Fig. 3.10). When a domain wall leaves a lamellar junction, it will have an extra area of ld_2 , corresponding to an extra wall energy of

$$E_\gamma = \gamma ld_2. \quad (3.1)$$

The contribution of E_γ to the maximum pinning force is then a constant given by

$$F_\gamma = \delta E_\gamma / \delta l = \gamma d_2. \quad (3.2)$$

B. Evaluation of the extra demagnetizing energy

Let t represent the thickness of the domain wall. When the domain wall is in its minimum energy state, its center coincides with the center of the pinning site (Fig. 3.6 a). When it moves $t/2$ away from the pinning site, the magnetic poles come out completely and the pin interaction potential reaches its maximum value. According to Gaunt's interaction potential (Eq. 1.1), the maximum and the minimum points of interaction potential are separated by a distance of $2b$, where b is the interaction range. In the present case, we therefore have

$$2b = t/2. \quad (3.3)$$

Then we obtain

$$b \propto (A_1/K_1)^{1/2}. \quad (3.4)$$

Eq. (3.4) gives the temperature dependence of b . Since the demagnetizing

energy reaches its maximum when the domain wall moves $\pm 2b$ away from its minimum energy position, it can be calculated as if the uncompensated poles are distributed uniformly on the two surfaces of a plate whose dimensions are $\sqrt{2}d_1$, d_2 and $4b$ along the three edges (Fig. 3.11). The demagnetizing energy associated with this plate is

$$E'_D = 2\pi M_1^2 V_p \eta, \quad (3.5)$$

where V_p is the volume of the plate, η is the demagnetizing factor normalized by 4π along the $\sqrt{2}d_1$ edge and $0 \leq \eta \leq 1$.

In view of the Fig. 3.10, we see that E'_D is associated with two lamellar junctions. The demagnetizing energy associated with one lamellar junction may be obtained by dividing E'_D by a factor of 2,

$$E_D = E'_D / 2 = 4\sqrt{2}\pi M_1^2 \eta d_1 d_2 b. \quad (3.6)$$

Since a distance $2b$ separates two points of energy maximum and minimum, the contribution of E_D to the maximum pinning force may be evaluated as

$$F_D = E_D / b = 4\sqrt{2}\pi M_1^2 \eta d_1 d_2. \quad (3.7)$$

The maximum pinning force f is then the summation of F_γ and F_D .

$$f = F_\gamma + F_D = \gamma d_2 + 4\sqrt{2}\pi M_1^2 \eta d_1 d_2. \quad (3.8)$$

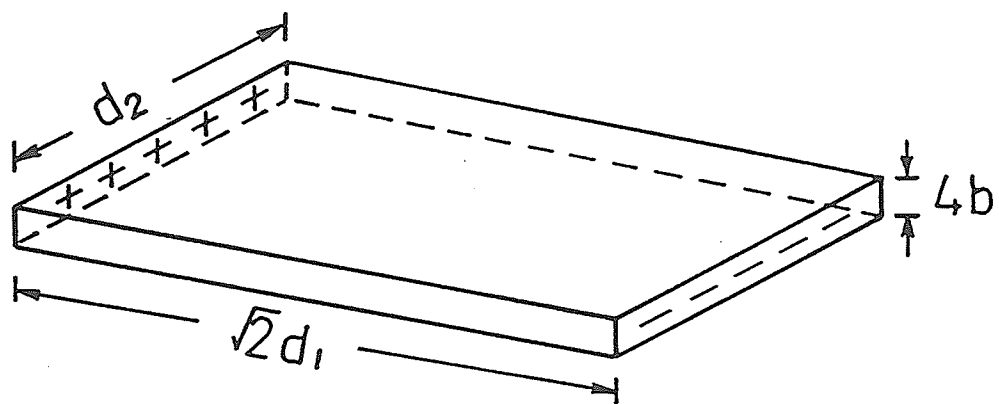


Figure 3.11: A fictitious plate to calculate the demagnetizing energy.

It is difficult to measure the value of d_2 . A good estimation for d_2 is that

$$d_2 = d_1. \quad (3.9)$$

Now the maximum pinning force per pin can be rewritten as

$$f = \gamma d_1 + 4\sqrt{2}\pi M_1^2 \eta d_1^2. \quad (3.10)$$

It is necessary to estimate the pin density. Suppose all pinning sites are distributed uniformly within the specimen and the distance between the two nearest pins is d_1 . A cubic unit cell is chosen in such a way that the length of each edge of the unit cell is $\sqrt{2}d_1$. It is shown in Fig. 3.12. There is one lamellar junction bar parallel to $[001]$ inside the cubic cell. There are 4 lamellar junction bars at edges in $[001]$ direction, but only 1/4 of them belonging to the cubic cell. Totally there are two lamellar junction bars which are parallel to $[001]$ direction in the cubic cell. There are four more lamellar junction bars in $[100]$ and $[010]$ directions. Altogether there are six lamellar junctions in the cubic cell. But one third of them are parallel to the easy axis of the specimen, which will not contribute to the pinning process. Therefore the net number of the lamellar junctions in the cube is four. The density of the pinning sites is then estimated as

$$\rho = \sqrt{2}/d_1^3. \quad (3.11)$$

It is necessary to know the temperature dependencies of the uniaxial anisotropy constant and of the spontaneous magnetization. G. Hadjipanayis [27] had measured these properties. From his results, it can be shown that

$$K_1 = 3.284 \times 10^7 [1 - (T/3885)] \text{ erg/cm}^3, \quad (3.12)$$

and

$$M_1 = 1224 [1 - (T/1800)^{3/2}] \text{ erg/Oe cm}^3. \quad (3.13)$$

The temperature dependence of A_1 may be obtained by making use of the relation [30]

$$A_1 \propto M_1^2. \quad (3.14)$$

Now the interaction range b and the wall energy per unit area γ can be rewritten as

$$b = b_0 (1 + T/7770) [1 - (T/1800)^{3/2}], \quad (3.15)$$

and

$$\gamma = \gamma_0 (1 - T/7770) [1 - (T/1800)^{3/2}], \quad (3.16)$$

where b_0 and γ_0 are the values of b and γ at absolute zero, respectively and $\gamma_0 = 14.5 \text{ erg/cm}^2$ [27].

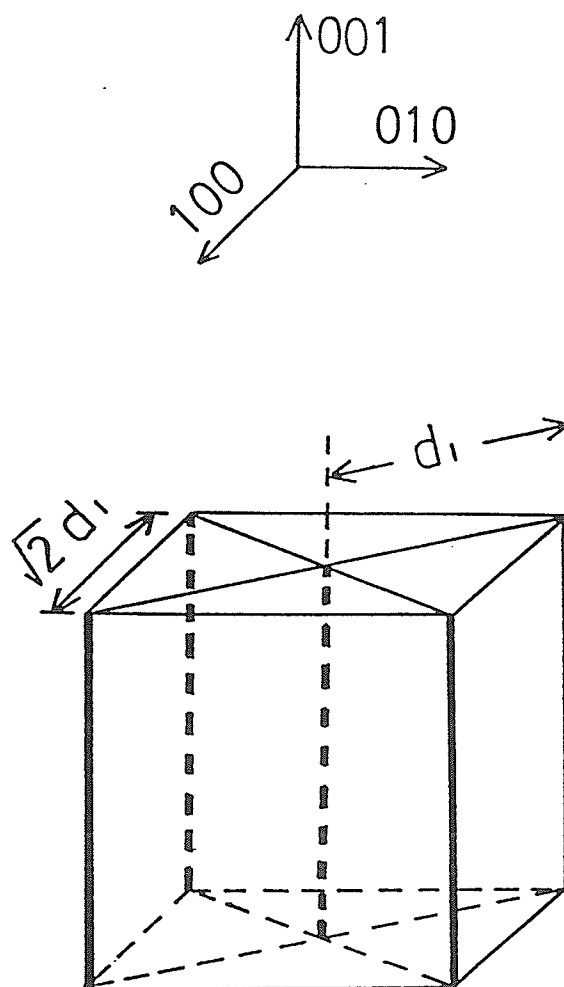


Figure 3.12: A unit cell to calculate the density of lamellar junctions.

A summation defined as

$$sum = \sum_{i=1}^{12} [H_{ci}(exp.) - H_{ci}(theo.)]^2 \quad (3.17)$$

is used in a least-square fitting. The experimental values of H_c are found to fit a strong-pinning model (Eqs. 1.7 and 1.8). The best fitting is obtained when $d_1 = 7.06 \times 10^{-6}$ cm, $b_0 = 6.73 \times 10^{-8}$ cm, and $\eta = 0.0595$. In view of Fig. 3.4, it is obvious that the value of d_1 is in the correct range. The best fitting gives the value of 1312 for the sum in Eq. (3.17), corresponding to an average deviation of 10 Oe for the predicted values of coercivity from the experimental values. This deviation is small compared with values about 1500 Oe. Fig. 3.13 compares the predicted values of coercivity with the experimental ones. Fig. 3.14 shows both H_c and H_0 as functions of temperature. It can be seen that H_0 drops only about 5 % in this temperature range whereas H_c drops about 26 % in the same temperature range. It seems that the height of the energy barriers is almost independent of temperature in this temperature range. The drop in the coercivity as a function of temperature is mainly due to thermally activated domain walls for this ferromagnet.

3.7 Discussion

The normalized demagnetizing factor η along the long edge of the plate (Fig. 3.11) may be estimated in another way. For a flat ellipsoid whose axes are $\sqrt{2}d_1$, d_2 and $4b$, with the condition that $4b \ll d_2$, $d_2 \approx \sqrt{2}d_1$, the value of

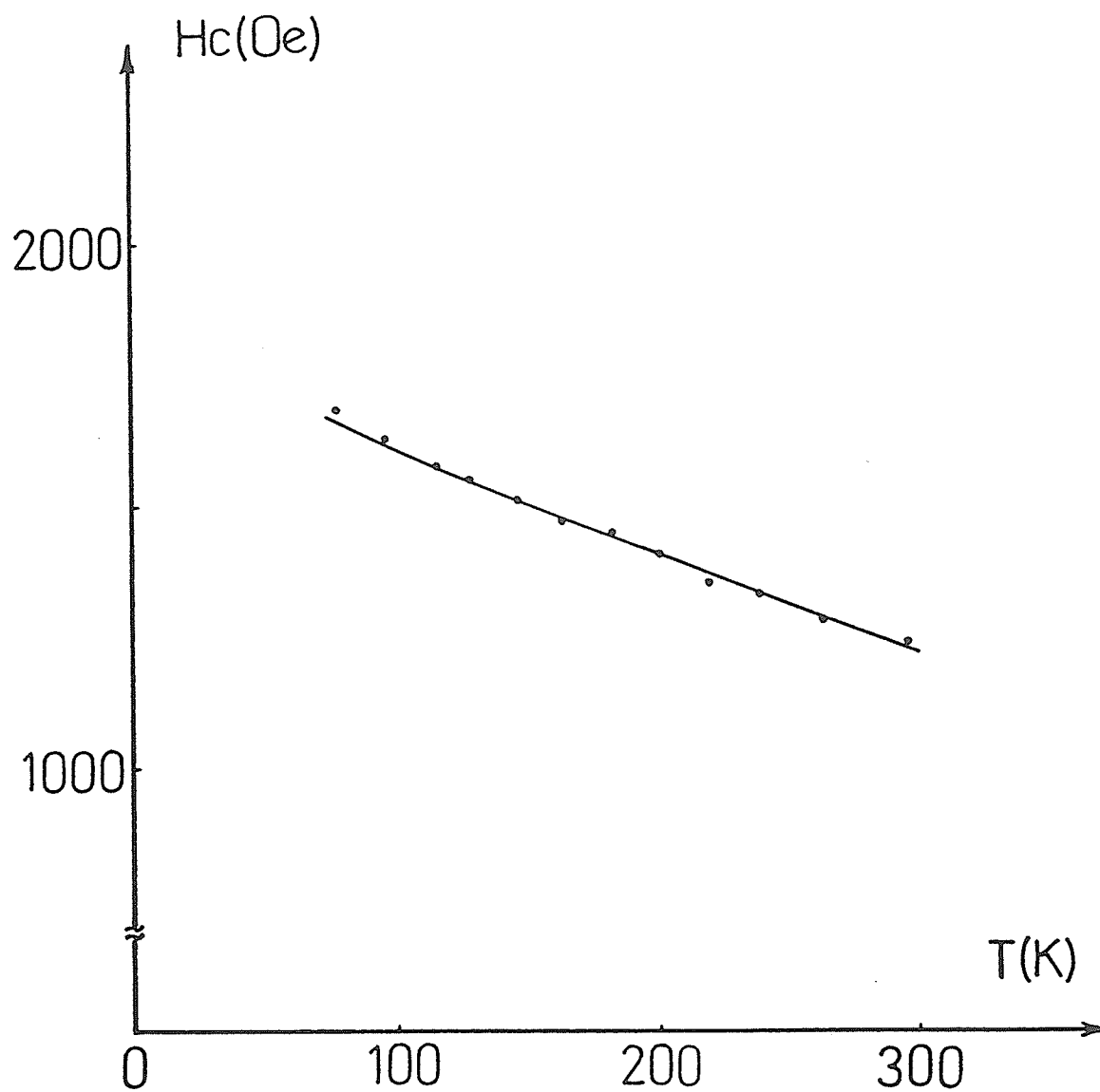


Figure 3.13: The experimental values (dots) of H_c fit a strong pinning model (solid line).

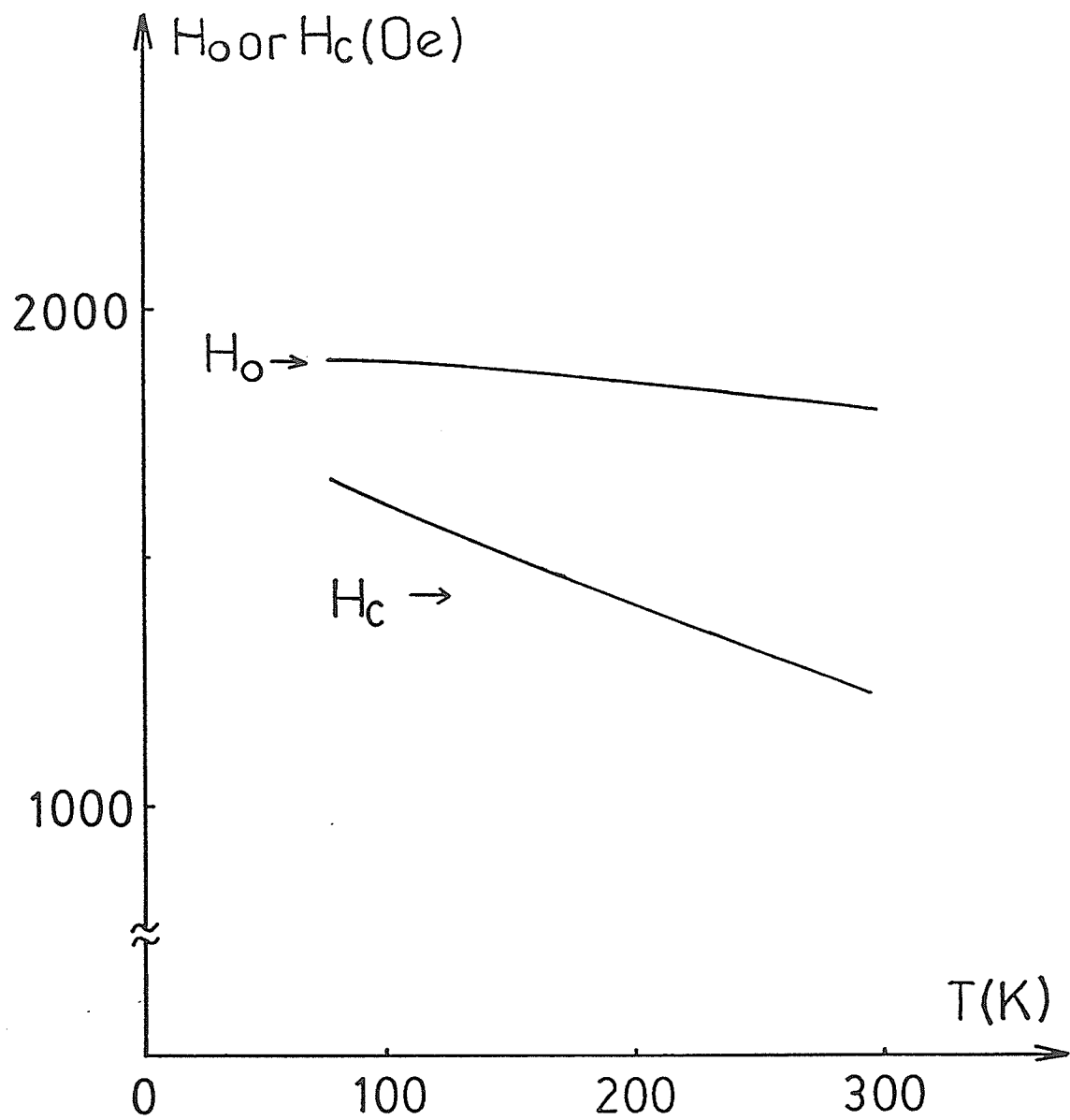


Figure 3.14: H_0 is almost independent of temperature, whereas H_c changes rapidly with temperature for a $\text{Pt}_{0.7}\text{Ni}_{0.3}\text{Fe}$.

η in the direction of the $\sqrt{2}d_1$ edge can be estimated by the relation [30]

$$\eta \approx \frac{\pi}{4} \frac{4b}{\sqrt{2}d_1} \approx 0.021. \quad (3.18)$$

In the present case, the value of η from the best fitting procedure is almost three times larger, at 0.0595. Since in the present model a plate rather than an ellipsoid is used and there are more than one nearest-neighbor interactions for each lamellar junction, such a difference is not surprising. The model, despite of its simplicity, gives a satisfactory description of the temperature dependence of coercivity and therefore confirms the pinning mechanism proposed by Gaunt [28] for a $\text{Pt}_{0.7}\text{Ni}_{0.3}\text{Fe}$ permanent magnet.

Chapter 4

Domain wall pinning by a thin
planar pin (continuous spin
model)

4.1 Introduction

In many important commercial permanent magnets, there are grain boundaries (the interfaces between grains in a polycrystalline material) which have different magnetic properties from the matrix. Such planar magnetic inhomogeneities can pin the domain walls if the wall energy is reduced by the existence of these inhomogeneities. Therefore it is very interesting to study the process of domain wall pinning by planar pinning sites.

Although many authors [17][18][20] have studied this particular pinning process, their models, as described in section 1.4, neglect thermal activation, and therefore are insufficient. Further work is necessary.

Ram et al. [31] proposed an idea of *localized pins* in which the pins are densely concentrated in some local regions. Transmission electron-microscope observations, however, do not confirm the existence of localized pins in MnAlC [31]. This means that localized pins, if they exist, are of small size, possibly of atomic dimensions. If many small pins are densely concentrated in a local planar region, it seems more reasonable to consider this region as one single planar pinning site, rather than a group of localized pins.

This chapter will treat the domain-wall pinning process in the case when the wall is not very thin. In this case the spins inside the wall rotate gradually and the spin rotation can be treated as continuous. For simplicity, all calculations in this chapter are aimed to the situation where the width of a planar pinning site is much thinner than the wall thickness. This assumption is satisfied in many cases.

4.2 Maximum pinning force

Fig. 4.1 shows the geometry of three regions. Region 1 and 3 are identical and represent the homogeneous material (matrix) whereas region 2 represents the planar pinning site characterized by an abrupt change in the magnetic properties. All three regions are assumed to have uniaxial anisotropy with easy axes along the $\vec{x}$ direction. The xyz' coordinates are fixed on the domain wall with the wall center at $z' = 0$. A magnetic field H is applied parallel to the magnetization on the left side of the domain wall. The domain moves in the $+\vec{z}'$ direction in finite field H . This is shown in Fig. 4.1 by artificially setting the wall boundaries at $\pm z'_0$. In the calculation z'_0 is taken as infinite as usual. The magnetization $\vec{M}$ within the wall makes an angle θ with respect to the $\vec{y}$ axis and is assumed to lie in the x-y plane. The angle θ thus starts from $-\pi/2$ and ends at $+\pi/2$. The boundaries of the planar pinning site are at z'_1 and z'_2 , the corresponding values of θ are θ_1 and θ_2 respectively. The pin width, W , is given by

$$W = z'_2 - z'_1. \quad (4.1)$$

The distance between the wall center and the pin center is z_W , that is

$$z_W = (z'_1 + z'_2)/2. \quad (4.2)$$

The domain wall will deviate from the point of minimum energy as a

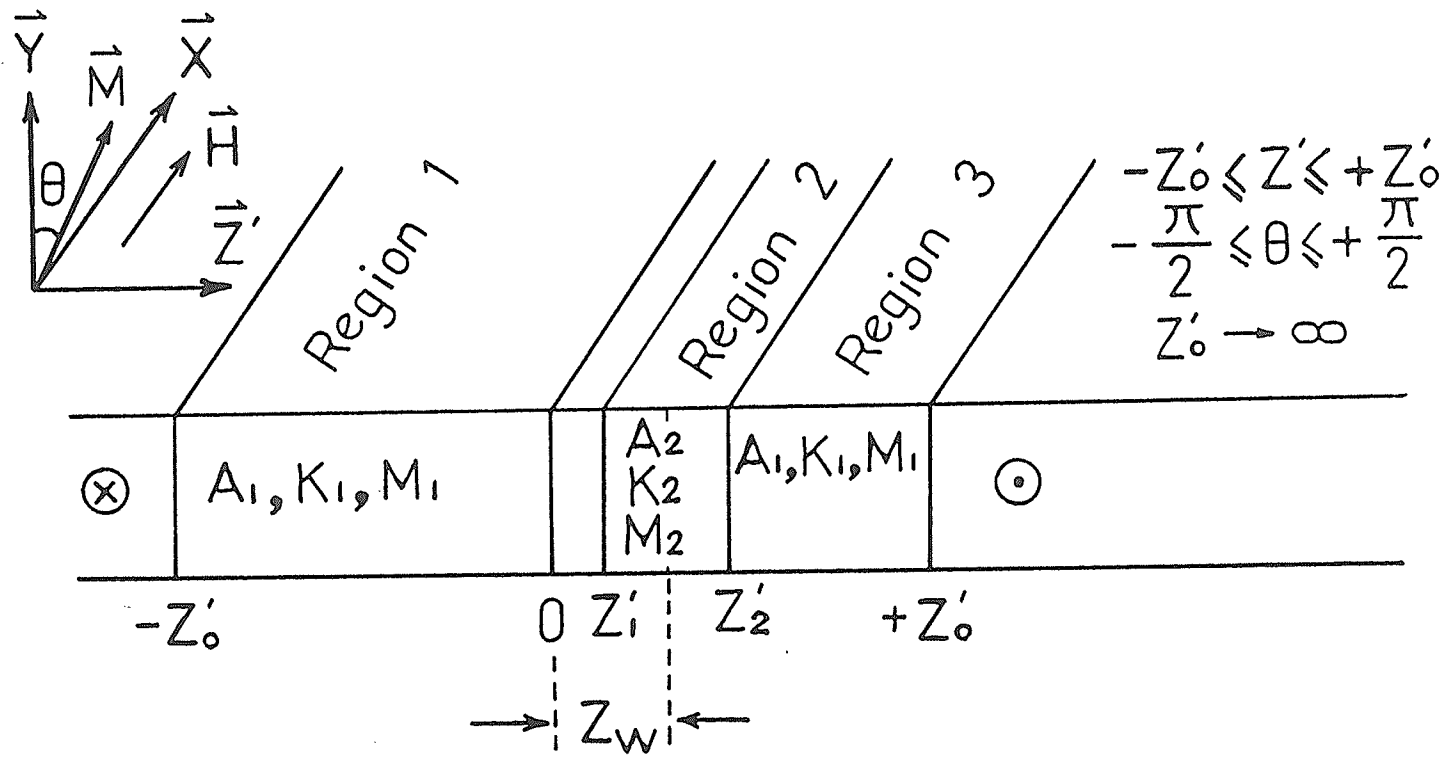


Figure 4.1: Geometry showing three regions.

result of either thermal activation or the application of a driving field. In the former case the pin interaction potential remains symmetric, the wall displacement $\pm z$ corresponding to the same energy increase; whereas a magnetic field H will destroy this symmetry. This distortion of the pin interaction potential is expected to be minimal as long as H is much smaller than the anisotropic field; thus as an approximation the effect of H will be neglected in the calculation of f .

The wall energy per unit area is

$$\gamma = \int [A_i (\frac{\partial \theta}{\partial z'})^2 + K_i \cos^2 \theta] dz', \quad (4.3)$$

where $i = 1, 2, 3$ represent three regions. The first term is the exchange energy and the second term is the uniaxial anisotropy energy.

Now the stable spin configuration can be obtained by minimizing the wall energy for a small variation of the spin arrangement inside the wall. When the angle θ of the spin at z' is changed by $\delta\theta$, the wall energy is changed by $\delta\gamma = (\partial\gamma/\partial\theta)\delta\theta$, which should vanish for the stable spin arrangement. This procedure leads to the Euler equations in three regions.

$$K_i \sin 2\theta + 2A_i (\frac{\partial^2 \theta}{\partial z'^2}) = 0. \quad (4.4)$$

Integrating the corresponding three Euler equations gives

$$K_i \cos^2 \theta - A_i (\frac{\partial \theta}{\partial z'})^2 = C_{1i}, \quad (4.5)$$

where C_{1i} are constants.

The boundary condition that $\partial\theta/\partial z' = 0$ at $\theta = \pm\pi/2$ gives $C_{11} = C_{13} = 0$. It is not easy to determine C_{12} because of the problematic boundary condition for the derivative ($\partial\theta/\partial z'$) at the pin boundaries. Friedberg and Paul [18] used the boundary condition that

$$A_i\left(\frac{\partial\theta}{\partial z'}\right) = \text{continuous}. \quad (4.6)$$

In their model, Friedberg et al. have completely neglected the thermally activated process of domain wall unpinning, therefore their model predicts that $|\theta_2| = |\theta_1|$ in zero magnetic field, which means that the domain wall is at the minimum pin interaction energy position in zero field. This model is certainly not acceptable if the thermally activated process of wall unpinning is considered, since the thermal energy can make a deviation of the wall from the position of the minimum energy even if the magnetic field is zero. The problem comes out of the application of the boundary condition of Eq. (4.6) since there are two pin boundaries and there is only one unknown constant. If the boundary condition of Eq. 4.6 is satisfied at one pin boundary $\theta = \theta_1$, it is usually not satisfied at another pin boundary $\theta = \theta_2$, because $|\theta_2| \neq |\theta_1|$ in general. A constant cannot have two values simultaneously. Therefore a new boundary condition is necessary. From Eq. (4.5), it is obvious that the only choice of C_{12} which is independent of θ is zero. A new boundary

condition for the derivative $(\partial\theta/\partial z')$ is thus supposed here.

$$\sqrt{\frac{A_i}{K_i}} \left(\frac{\partial\theta}{\partial z'} \right)_i = \text{cont.} \quad (4.7)$$

which yields an integral constant C_{12} of zero. This boundary condition shows that the thickness of a domain wall is determined by the counterbalance of the exchange energy and the anisotropy energy, as is well known.

Now the wall energy in regions 1 and 3 is calculated as

$$\begin{aligned} \gamma_1 + \gamma_3 &= 2\sqrt{A_1 K_1} \left[\int_{-\pi/2}^{\theta_1} \cos\theta \, d\theta + \int_{\theta_2}^{\pi/2} \cos\theta \, d\theta \right] \\ &= 2\sqrt{A_1 K_1} [2 + \sin\theta_1 - \sin\theta_2]. \end{aligned} \quad (4.8)$$

The wall energy in region 2 is calculated as

$$\begin{aligned} \gamma_2 &= 2\sqrt{A_2 K_2} \int_{\theta_1}^{\theta_2} \cos\theta \, d\theta \\ &= 2\sqrt{A_2 K_2} [\sin\theta_2 - \sin\theta_1]. \end{aligned} \quad (4.9)$$

It is required to estimate the maximum values of $|\theta_1|$ and $|\theta_2|$ when the domain wall is near a point of maximum pinning force. From Gaunt's potential of Eq. (1.1), the distance $2b$ separates two points where the pin interaction potential reaches its maximum and minimum respectively. The

maximum value of b for a thin planar pin is therefore given by

$$4b_{max.} = \pi\delta_1, \quad (4.10)$$

where $\pi\delta_1 = \pi(A_1/K_1)^{1/2}$ is the wall thickness. Eq. (4.10) then gives

$$b \leq \pi\delta_1/4. \quad (4.11)$$

From Eq. (4.11), one can expect that the pinning force reaches its maximum at $|\theta| = |\theta_{1,2}| \approx 5\pi/22 < 1$. Therefore the trigonometric functions, such as $\sin\theta_{1,2}$, etc., can be expanded in series to the fourth power of θ without significant error. Now the wall energy in three regions can be rewritten as

$$\gamma_1 + \gamma_3 = 2\sqrt{A_1K_1}[2 - (\theta_2 - \theta_1) + (\theta_2^3 - \theta_1^3)/6], \quad (4.12)$$

and

$$\gamma_2 = 2\sqrt{A_2K_2}[\theta_2 - \theta_1 - (\theta_2^3 - \theta_1^3)/6]. \quad (4.13)$$

The reason for this series expansion will be obvious later.

When a domain wall moves away from a pinning site, both θ_1 and θ_2 change; furthermore they are not independent of each other. This relationship is obtained below. From Eq. (4.5)

$$dz' = \delta_2(\cos\theta)^{-1} d\theta, \quad (4.14)$$

where $\delta_2 = (A_2/K_2)^{1/2}$. The pin width is then

$$W = \int_{z'_1}^{z'_2} dz' = \delta_2 \int_{\theta_1}^{\theta_2} (\cos \theta)^{-1} d\theta. \quad (4.15)$$

Since W is a constant, the expression for θ_2 as a function of θ_1 may be obtained from Eq. (4.15).

The following dimensionless parameters are introduced

$$Z = z/\delta_2, \quad w = W/\delta_2, \quad Z_W = z_W/\delta_2, \quad (4.16)$$

where z is defined as $z = -z_W$. Equation (4.15) can then be rewritten as

$$w = \int_{\theta_1}^{\theta_2} (\cos \theta)^{-1} d\theta, \quad (4.17)$$

that is

$$w = \ln \left[\frac{\operatorname{tg}(\pi/4 + \theta_2/2)}{\operatorname{tg}(\pi/4 + \theta_1/2)} \right]. \quad (4.18)$$

Because both $\theta_{1,2}$ are less than one, the series expansion gives

$$\ln \operatorname{tg} \left(\frac{\pi}{4} + \frac{\theta_{1,2}}{2} \right) \approx \theta_{1,2} + \frac{1}{6} \theta_{1,2}^3, \quad (4.19)$$

and

$$w = \theta_2 - \theta_1 + \frac{1}{6}(\theta_2^3 - \theta_1^3). \quad (4.20)$$

For a thin planar pinning site, $w \ll 1$; θ_2 is then solved from Eq. (4.20) by an iterative procedure to give, to the first power of w and fourth power of θ_1 ,

$$\theta_2 = w + \theta_1 - w\theta_1^2/2 + w\theta_1^4/4. \quad (4.21)$$

With Eqs. (4.12),(4.13) and (4.21), the domain-wall energy can be expressed as a function of θ_1 . That is

$$\begin{aligned} \gamma &= \gamma_1 + \gamma_2 + \gamma_3 \\ &= 4\sqrt{A_1 K_1} - 2(\sqrt{A_1 K_1} - \sqrt{A_2 K_2})w + (\sqrt{A_1 K_1} - \sqrt{A_2 K_2})(2\theta_1^2 - \theta_1^4)w. \end{aligned} \quad (4.22)$$

Next the expression of θ_1 is found as a function of the dimensionless wall displacement Z . The variation of θ_1 with respect to z' can then be obtained by integrating Eq. (4.5) in three regions. In region 1

$$z' = \delta_1 \ln \left[\operatorname{tg} \left(\frac{\pi}{4} + \frac{\theta}{2} \right) \right] + C_{21}, \quad (4.23)$$

in region 2

$$z' = \delta_2 \ln \left[\operatorname{tg} \left(\frac{\pi}{4} + \frac{\theta}{2} \right) \right] + C_{22}, \quad (4.24)$$

and in region 3

$$z' = \delta_1 \ln \left[\operatorname{tg} \left(\frac{\pi}{4} + \frac{\theta}{2} \right) \right] + C_{23}, \quad (4.25)$$

The boundary condition that θ is continuous at the pin boundaries gives

$$C_{21} = (\delta_2 - \delta_1) \ln \left[\operatorname{tg} \left(\frac{\pi}{4} + \frac{\theta_1}{2} \right) \right] + C_{22}, \quad (4.26)$$

and

$$C_{23} = (\delta_2 - \delta_1) \ln \left[\operatorname{tg} \left(\frac{\pi}{4} + \frac{\theta_2}{2} \right) \right] + C_{22}. \quad (4.27)$$

By making use of Eqs. (4.2) and (4.24), the wall displacement z_W can be rewritten in the form

$$\frac{z_W - C_{22}}{\delta_2} = \frac{1}{2}(\theta_1 + \theta_2) + \frac{1}{12}(\theta_1^3 + \theta_2^3). \quad (4.28)$$

Since θ_2 depends on θ_1 , Eq. (4.28) is an expression of θ_1 as a function of z_W . Again, application of an iterative procedure yields the expression that is, to the first power in w and fourth power in Z_c ,

$$\theta_1 = -\frac{1}{2}w - Z_c + \frac{1}{4}wZ_c^2 + \frac{1}{6}Z_c^3 - \frac{5}{24}wZ_c^4, \quad (4.29)$$

where $Z_c = (z_W - C_{22})/\delta_2$.

Now the wall energy in Eq. (4.22) can be rewritten as

$$\gamma = \gamma_0 + U(Z_c), \quad (4.30)$$

where

$$\gamma_0 = 4\sqrt{A_1 K_1} - 2(\sqrt{A_1 K_1} - \sqrt{A_2 K_2})w, \quad (4.31)$$

and

$$U(Z_c) = 2(\sqrt{A_1 K_1} - \sqrt{A_2 K_2})(Z_c^2 - \frac{5}{6}Z_c^4)w. \quad (4.32)$$

Since the pin interaction potential must be symmetric about the origin, the wall displacement $\pm z_W$ will cause the same amount of increase in energy. This condition requires the integral constant C_{22} to be zero. Actually, in most studies of domain-wall pinning, the coordinates are fixed at a pinning site, not on the wall. To be consistent with this, z_W is replaced by $z = -z_W$. Using the dimensionless parameters defined in Eq. (4.16), the pin interaction energy per unit area of a planar pinning site can finally be written as

$$U(Z) = 2(\sqrt{A_1 K_1} - \sqrt{A_2 K_2})(Z^2 - \frac{5}{6}Z^4)w. \quad (4.33)$$

The fact that Eq. (4.33) has one maximum inside the domain wall ($Z > 0$ region) requires some comment. Since a pinning site always reduces the wall energy, the potential should have a positive slope throughout. The problem arises simply because the high order terms (Z^6 and higher) are neglected in the calculation. However it is the maximum pinning force that is important in the present work, which is expected to occur at small Z . Then the Z^6 term is not important.

The maximum pinning force, f' , per unit area of a pinning site is obtained from the equation

$$\frac{d^2 U(Z)}{dZ^2} = 0, \quad (4.34)$$

which gives

$$f' = 1.2[\sqrt{A_1 K_1} - \sqrt{A_2 K_2}]w/\delta_2. \quad (4.35)$$

This occurs at

$$Z_0 = \pm\sqrt{0.2}. \quad (4.36)$$

In a magnetic field, a domain wall will move in the $+Z$ direction, therefore only $Z_0 = +\sqrt{0.2}$ is of interest. Thus the interaction range b is

$$b = \sqrt{0.2}\delta_2. \quad (4.37)$$

Eqs. (4.35) and (4.37) can now be used to obtain an expression for the pinning potential of the form given by Eq. (1.1) which is expected to be a good approximation to the real potential near a point of maximum pinning force.

Eq. (4.37) shows that b is proportional to δ_2 which is a measure of the magnetic properties of the pinning site, not of the magnet itself (matrix). Experimentally, it is not simple to measure δ_2 . As far as the temperature dependence of δ_2 is concerned, δ_2 may be assumed to have the same temperature dependence as δ_1 . Eq. (4.35) gives the dominant terms in f' . If A_2 and K_2 are assumed to have the same temperature dependence as A_1 and K_1 respectively, f' will linearly depend on $K_1^{3/2}/A_1^{1/2}$. This means that the temperature dependence of the maximum pinning force is determined by $K_1^{3/2}/A_1^{1/2}$, if the ratio of A_1/A_2 and K_1/K_2 are less sensitive to temperature, as compared with $A_{1,2}$ and $K_{1,2}$. This condition is usually satisfied. The di-

dimensionless pin width w is also sensitive to temperature and if the condition $w < 1$ is not satisfied, Eqs. (4.35) and (4.37) will not be valid. In that case, higher-order terms in w must be taken into consideration. However, in many cases the condition $w < 1$ is satisfied, and in these situations, the Eqs. (4.35) and (4.37) will be useful.

The effect of a magnetic field on the pin interaction potential and the maximum pinning force will be discussed in Appendix A.

4.3 Coercivity of a MnAlC permanent magnet

4.3.1 Description of previous works

The MnAlC ferromagnet has uniaxial anisotropy. Ram et al. [31] and Rothwarf et al. [32] have measured the coercivity of MnAlC ferromagnets. The coercivity was found to drop linearly as temperature increases. However, none of these authors has given a quantitative prediction of coercivity in their models. Houseman et al. [33] observed, using transmission-electron microscopy, that the grain boundaries are pinning sites. Ram et al. [31] tried to explain this phenomenon by so called *localized pins*. They proposed that the pins may concentrate in some local regions such as grain boundaries and the pins are very few in number elsewhere. From magnetic viscosity measurements, they predicted a maximum localized pin density of $\approx 5 \times 10^{15} \text{cm}^{-3}$ corresponding to a linear pin spacing $\approx 580 \text{ \AA}$, which should be easily observed by transmission-electron microscope. Unfortunately, electron micro-

scope observations have failed to confirm the existence of inhomogeneities separated by distances of this order that might be possible pinning sites. Although Ram et al. claimed that the Lorentz electron-microscopy domain contrast in such regions might be overwhelmed by strain-contrast effects, it is more likely that, as described in Chapter 1, this discrepancy occurs because their pinning models have obvious weaknesses. In this section, the results in section 4.2 will be applied to the MnAlC permanent magnet. For the reason described in section 4.1, a grain boundary will be treated as one single planar pinning site, rather than is a group of localized pins.

4.3.2 Sample preparation

The sample under investigation was a Matshushita Electrical Company MnAlC permanent magnet. The x-ray diffraction results showed that the compound has a tetragonal structure with the value of $c/a \approx 0.92$. Specimens were prepared for a transmission-electron microscopy examination. The electrolyte used to etch the sample was a solution of 25 % perchloric acid and 75 % glacial acetic acid. The current density was about 230 mA/mm². The specimens were then thermally demagnetized at 300°C for 5 minutes. It is not expected that thermal aging will significantly alter either the magnetic properties or the crystal structure of the specimen.

4.3.3 Transmission-electron microscopy

Fig. 4.2 shows an in-focus bright field image taken from a MnAlC specimen in a thermally demagnetized state. The grains and the grain boundaries can

Table 4.1: Coercive field tabulated for different temperatures.

T(K)	80	118	136	168	197	295
H_c (Oe)	4208	3970	3857	3656	3474	2860

be clearly seen. The white region is a hole. Fig. 4.3 shows an overfocused image in the same region as Fig. 4.2. The black-white lines are identified with domain walls since in the underfocused state, the magnetic contrast reverses (Fig. 4.4). Some walls are inside the grains, and others are attached to grain boundaries. After a field of 3.6 kOe ($> H_c$) is applied, the domain walls inside the grains disappear, whereas those attached to grain boundaries are still there. This is shown in Fig. 4.5. The transmission-electron microscopy therefore confirms the work by Houseman et al. [33]; the dominant pins are planar pinning sites at grain boundaries.

4.3.4 Temperature dependence of coercivity

Table 4.1 shows the coercivity as a function of temperature measured by Ram et al. [31].

Rothwarf et al. [32] have measured the temperature dependence of M_1 and K_1 . They find

$$M_1 = M_{10}(1 - T^{3/2}/50060) \quad (4.38)$$

and

$$K_1 = K_{10}(1 - T/2212 - T^{3/2}/50060), \quad (4.39)$$

where M_{10} and K_{10} are the values of M_1 and K_1 at absolute zero respectively.



Figure 4.2: An in-focus bright field image of MnAlC.



Figure 4.3: An overfocused image, compared with Fig. 4.2.



Figure 4.4: A under-focused image, compared with Fig. 4.3.

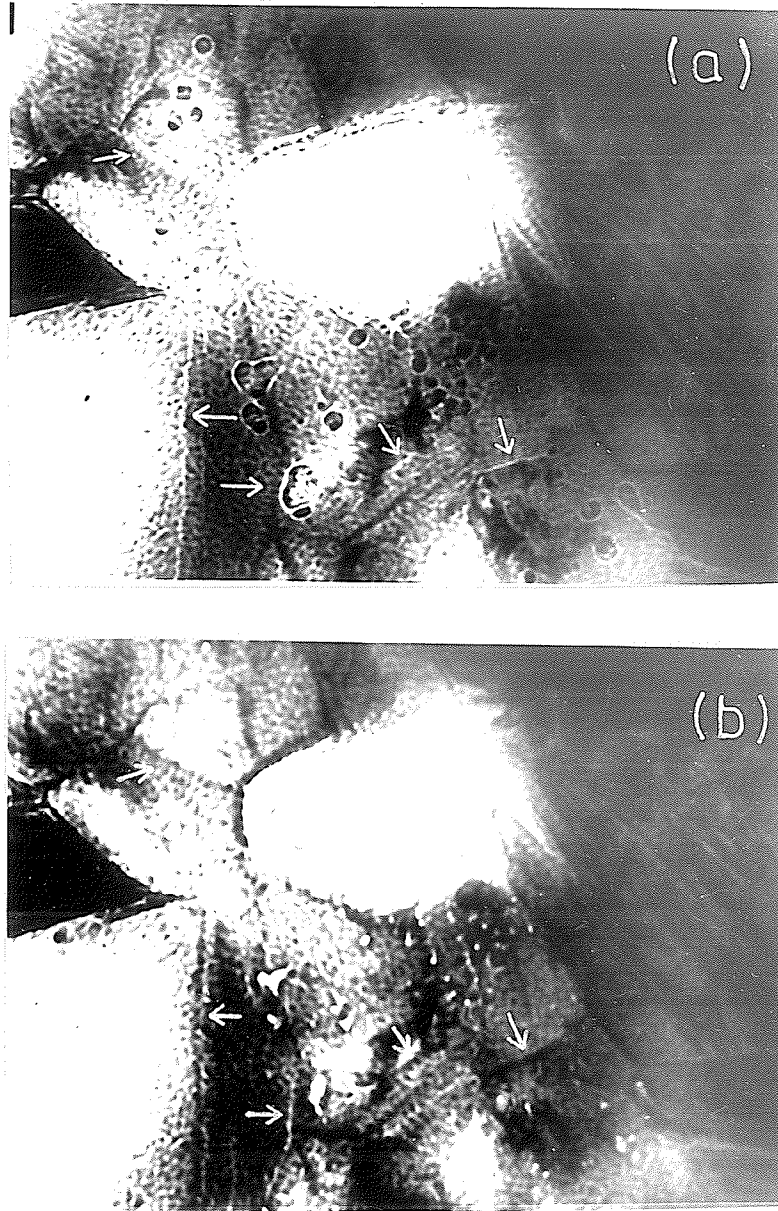


Figure 4.5: Domain walls are attached at grain boundaries, after a field of 3.6 kOe is applied, (a) under-focusing, (b) over-focusing.

The temperature dependence of the exchange constant is obtained by using relation [30]

$$A_1 \propto M_1^2. \quad (4.40)$$

Now the temperature dependencies of the wall energy γ , the interaction range b , and the maximum pinning force f can be written, by making use of Eqs. (4.38)-(4.40), as

$$\gamma = \gamma_0(1 - T/4424 - T^{3/2}/33373),$$

$$b = b_0(1 + T/4424 - T^{3/2}/100120),$$

and

$$f = f_0 \frac{1 - T/4424 - T^{3/2}/33373}{(1 + T/4424 - T^{3/2}/100120)^2},$$

where γ_0 , b_0 and f_0 are the values of γ , b and f at absolute zero respectively. The previous experimental results by Ram et al. [34] provided the following.

$$A_{10} = A_1(T = 0) = 3.35 \times 10^{-7} \text{ erg/cm},$$

$$M_{10} = 700 \text{ erg/cm}^3 \text{ Oe},$$

$$\sqrt{A_{10}K_{10}} = 2.45 \text{ erg/cm}^2. \quad (4.41)$$

The pin density is assumed to be independent of temperature. In order to estimate the pin density, cubic grains are assumed (Fig. 4.6). The average length of an edge of a grain is Δx and thus the volume of the grain is $(\Delta x)^3$.

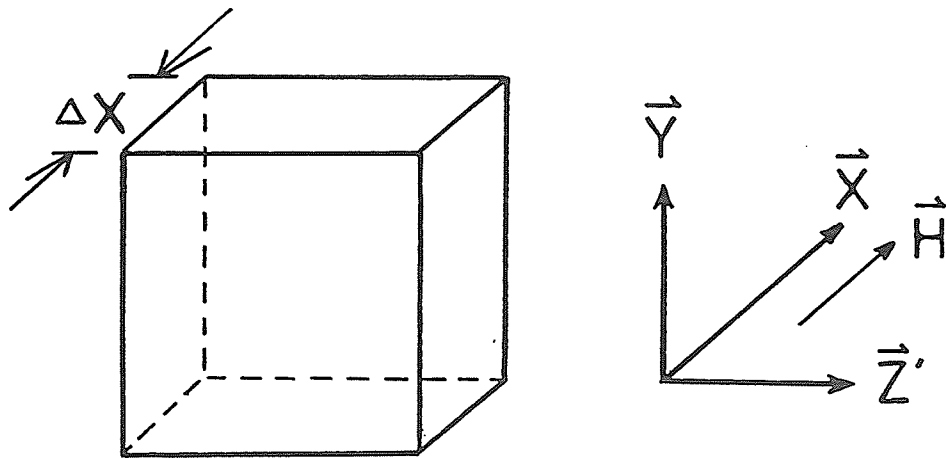


Figure 4.6: A cubic grain to estimate the pin density.

Each surface of the cubic grain is a grain boundary (pinning site) whose area is

$$S_p = (\Delta x)^2. \quad (4.42)$$

There are six grain boundaries only three of which belong to this cubic grain. One among three does not contribute to the pinning process because it is perpendicular to [001], the easy direction of the magnet. Therefore there are only two pinning sites in a volume of $(\Delta x)^3$. The pin density is then given by

$$\rho = 2/(\Delta x)^3. \quad (4.43)$$

By making use of eqs. (4.35) and (4.42), the maximum pinning force per planar pinning site can be written as

$$f = 1.2[\sqrt{A_1 K_1} - \sqrt{A_2 K_2}](\Delta x)^2 w / \delta_2. \quad (4.44)$$

The sum, S ,

$$S = \sum_{i=1}^6 [H_{ci}(exp.) - H_{ci}(theo.)]^2, \quad (4.45)$$

is now used in a least-square computer fit, where $H_{ci}(exp.)$ and $H_{ci}(theo.)$ are the experimental and the theoretical coercivity in Oe. The best fit is obtained with the weak pinning model which is rewritten here, as

$$H_c = H_0[1 - 25kT/31\gamma b^2], \quad (4.46)$$

where

$$H_0 = 3\rho f^2 / (4\pi\gamma M_1). \quad (4.47)$$

The procedure used for a least-square fit is as follows. Define

$$\alpha = A_2/A_1, \quad \beta = K_2/K_1, \quad \delta_{20} = \delta_2(T=0). \quad (4.48)$$

In the expression for H_0 , there is a product, $f_0^2\rho$, which is proportional to the combination of several unknown parameters,

$$(1 - \sqrt{\alpha\beta})^2 (W/\delta_{20}^2)^2 (\Delta x). \quad (4.49)$$

In the computer fitting, two variables, b_0 and $f_0^2\rho$ are adjusted to minimize the *sum* defined in Eq. (4.45). The minimum value obtained is 90 and the corresponding values of the parameters are

$$b_0 = 1.242 \times 10^{-7} \text{ cm}, \quad f_0^2\rho = 1.38 \times 10^8 \text{ erg}^2 \text{ cm}^{-5}.$$

Fig. 4.7 compares the experimental values of H_c (Table 4.1) with the predicted ones. The calculated H_c has an average deviation from the experimental value of $|\Delta H_c| \approx 4 \text{ Oe}$, which is negligible compared with coercive fields of a few kOe. The value of *sum* is very sensitive to b_0 ; a $\pm 1\%$ change in b_0 leads to a four-fold increase in *sum*. It is even more sensitive to $f_0^2\rho$; a $\pm 1\%$ changes in $f_0^2\rho$ produces a 65 fold increase in *sum*! An effort was made to fit $H_c(T)$ to the strong pinning model but failed to reduce the *sum* to a

Table 4.2: Calculated H_0 and H_c/H_0 at different temperatures

T(K)	80	118	136	168	197	295
$H_0(Oe)$	4480	4360	4308	4221	4147	3917
H_c/H_0	0.94	0.91	0.89	0.87	0.84	0.73

value less than 6000. The calculated values of H_0 and H_c/H_0 are tabulated in Table 4.2. It can be seen that H_0 drops about 12 % when the temperature increases from 80 K to 295 K, therefore the variation of the height of the energy barrier is not negligible in this temperature range. The ratio of H_c/H_0 is a measure of the thermally activated process of wall unpinning. The value of the ratio drops from 0.94 at 80 K to 0.73 at 295 K, indicating the important role of thermal activation of domain walls. Fig. 4.8 shows the critical field H_0 and the coercivity H_c at different temperatures. Both H_0 and H_c drop significantly as the temperature increases. The thermally activated process of wall unpinning causes the H_c to drop more rapidly than H_0 . Fig. 4.8 therefore shows again that both contributions are important to the temperature dependence of coercivity for a MnAlC permanent magnet.

4.3.5 Discussion

The observed value of Δx from transmission-electron microscope work is about 1 μm . A reasonable choice of W is 10^{-7} cm. The use of 10 $\text{\AA}$ as the pin width does not negate the continuum approximation of micromagnetics [19]. The calculated values of $f_0^2\rho$ and b_0 are used to predict the values of

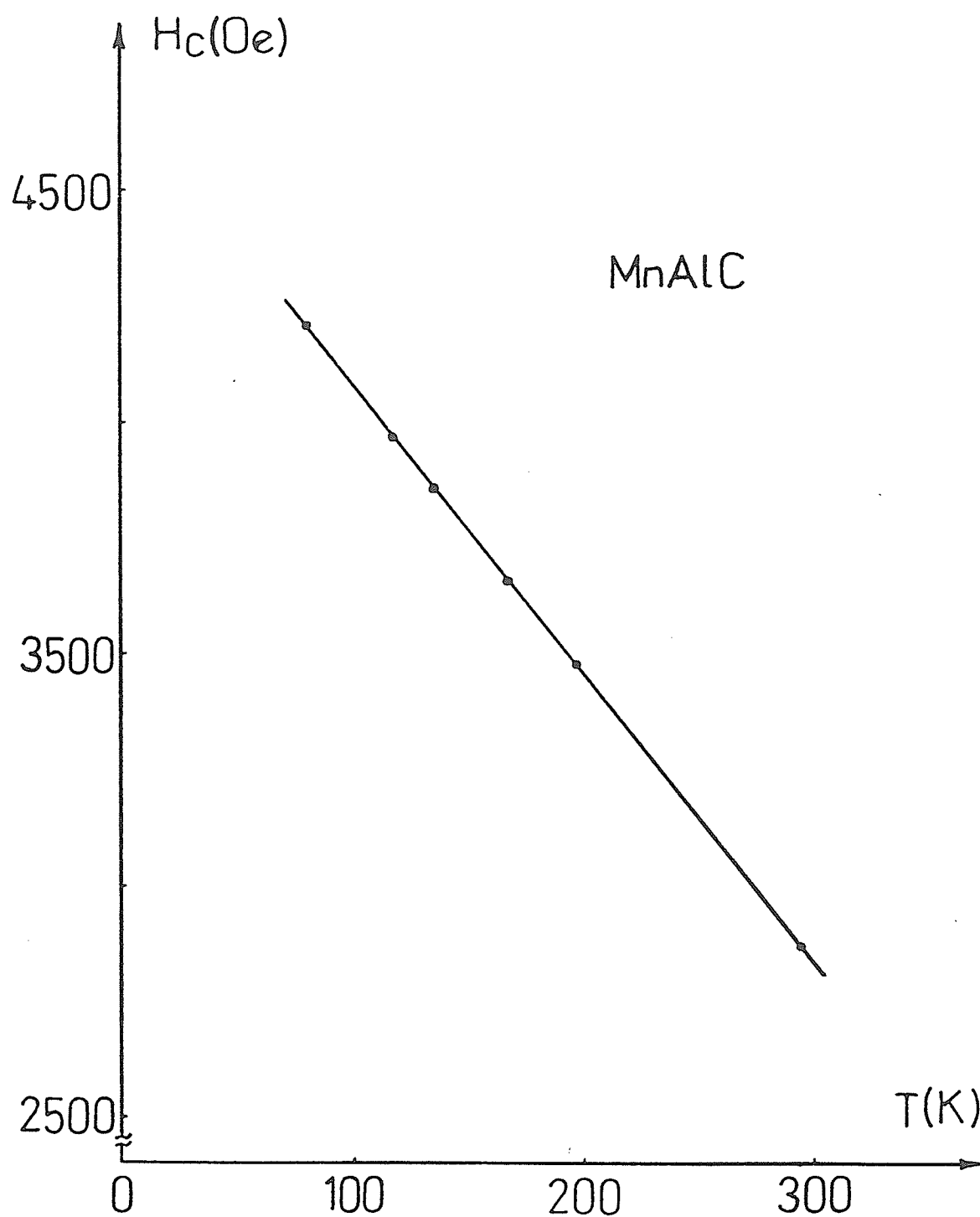


Figure 4.7: Coercivity at different temperatures for a MnAlC. The dots are experimental values and the solid line is predicted by a modified weak pinning model.

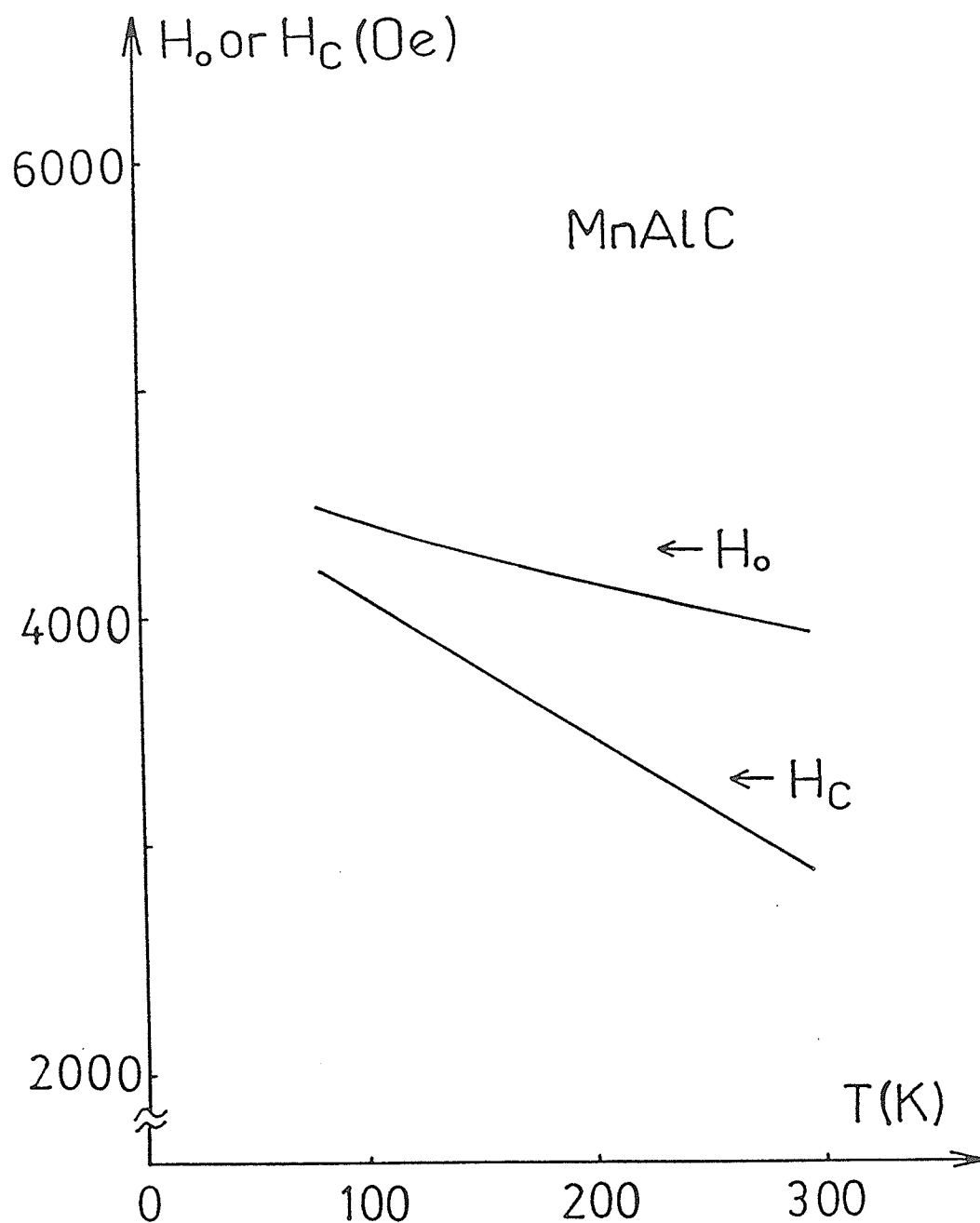


Figure 4.8: Comparison between H_0 and H_c as a function of temperature, showing that both the thermal activation of domain walls and the variation of energy barriers with temperature are important.

the magnetic properties of the pinning site. They are

$$\delta_{20} = 2.777 \times 10^{-7} \text{ cm}, \quad (4.50)$$

and

$$\alpha\beta = 0.6117. \quad (4.51)$$

From Eq. (4.41), we have

$$\delta_{10} = 1.367 \times 10^{-7} \text{ cm}. \quad (4.52)$$

Since

$$\delta_{20} = \sqrt{\frac{\alpha}{\beta}} \delta_{10}, \quad (4.53)$$

$$\frac{\alpha}{\beta} = 4.127. \quad (4.54)$$

Now α and β are solved from Eqs. (4.51) and (4.54); we obtain

$$\alpha = 1.589, \quad \beta = 0.385. \quad (4.55)$$

As a result, $\sqrt{A_2 K_2} = 0.782 \sqrt{A_1 K_1}$, which is reasonable because it indicates a reduced wall energy at the pinning site. Since there is large uncertainty in the determination of W and Δx , these values are acceptable only as nominal values. It is necessary to obtain more information about the properties of grain boundaries if the model is to be tested further. However the sensitivity

of *sum* on b_0 and $f_0^2\rho$ indicates that the model is quite reliable as far as the prediction of the coercivity is concerned.

Chapter 5

Domain-wall pinning by
pinning sites of atomic size
(discrete spin model)

5.1 Introduction

Domain walls are very thin, sometimes only a few atomic layers, in ferromagnets with extremely high anisotropy (for example, with $K_1 = 1.5 \times 10^8 \text{ erg/cm}^3$ for SmCo_5 , to be compared with $K_1 = 4.5 \times 10^6 \text{ erg/cm}^3$ for cobalt). As a consequence, the change in spin orientation of neighbouring spins inside the domain wall, especially at the center part of the wall, is too large to be treated as continuous. Therefore the continuum approximation of micromagnetics breaks down. The so-called discrete spin model is necessary to make any theoretical calculation related to the wall structure. In this model, a 180° domain wall is divided into three regions; a discrete region at the center of the domain wall where the angles between neighbouring spins are large and two continuous regions at the wall edges where the change in spin orientation between neighbouring spins is small enough to be treated as continuous (Fig. 5.1). This model has been used by Hilzinger et al. [22] and Broek et al. [35] to calculate the coercivity originating from the Peierls potential and by Craik et al. [36][37] to study domain-wall (thin walls) pinning by magnetic inhomogeneities in highly anisotropy materials. Unfortunately, the thermally activated process of domain wall unpinning is neglected in these works. In this chapter, these two contributions are incorporated into a single theory, with the special attention of the domain wall (thin wall) pinning by pinning sites of atomic size. An expression for the pin interaction potential is therefore necessary and will be given in the next section.

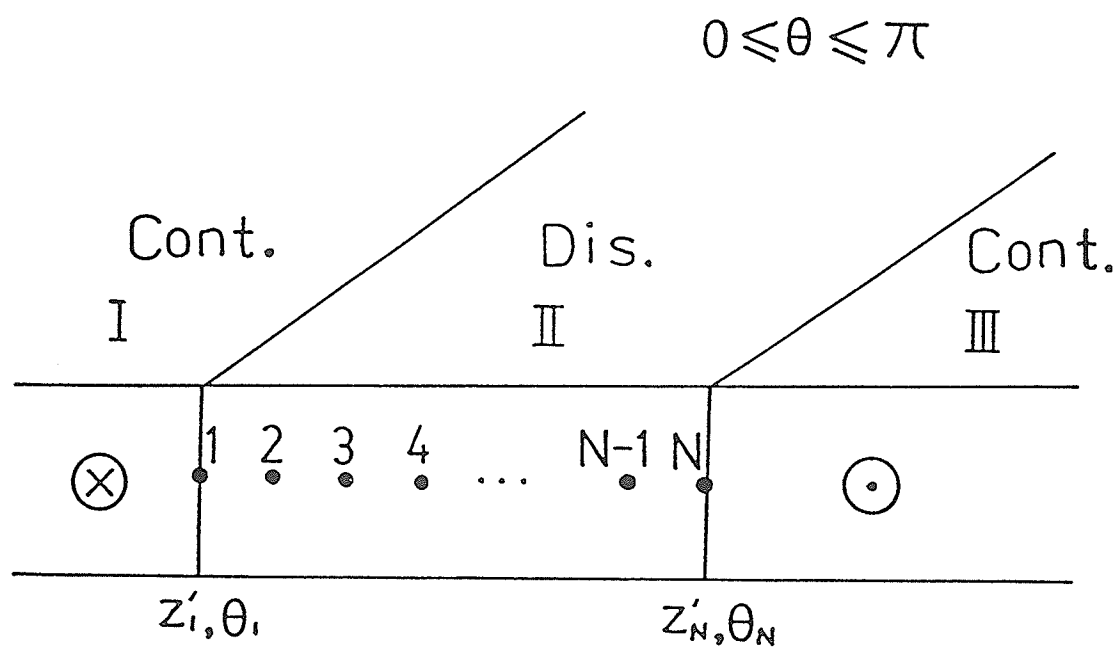


Figure 5.1: A 180° wall divided into one discrete region at the wall center and two continuous regions at the edges; the spin orientation is denoted by the angle θ .

5.2 Pinning potential

Consider a ferromagnet with uniaxial anisotropy and let us set up a wall energy function $\gamma(\theta_1, \theta_2, \dots, \theta_N)$, where θ_i is the angle made by the i th spin within the wall with respect to the easy axis; θ_1 and θ_N are the angles terminating the discrete region at either end and leading into the continuous regions, and $0 \leq \theta \leq \pi$ (Fig. 5.1). The wall energy is composed of the anisotropy and exchange energies. In continuous regions 1 and 3, the wall energy can be calculated by making use of the continuum approximation of micromagnetics.

$$\begin{aligned}
 \gamma_1 + \gamma_3 &= \int_{-\infty}^{z'_1} [K \sin^2 \theta + A(\frac{d\theta}{dz'})^2] dz' + \int_{z'_N}^{\infty} [K \sin^2 \theta + A(\frac{d\theta}{dz'})^2] dz' \\
 &= 2\sqrt{AK} [\int_0^{\theta_1} \sin \theta d\theta + \int_{\theta_N}^{\pi} \sin \theta d\theta] \\
 &= 2\sqrt{AK} (2 - \cos \theta_1 + \cos \theta_N), \tag{5.1}
 \end{aligned}$$

where A and K are the exchange constant and the anisotropy constant of the ferromagnet, respectively.

The wall energy in the discrete region contributes the most to the total wall energy. To simplify the calculation, the magnetic atoms within the magnet are assumed to form a simple cubic structure. The anisotropy

energy associated with the i th spin is

$$E_{A_i} = K_i a^3 \sin^2 \theta_i, \quad (5.2)$$

where a is the lattice constant. The exchange energy between the i, j th spins can be written as

$$E_{e_{i,j}} = 2A_{i,j}a[1 - \cos(\theta_i - \theta_j)], \quad (5.3)$$

where $A_{i,j}$ is the exchange constant between the atoms i and j ; and $A_{i,j} = A$, if no pinning site exists.

The spins in the same plane are assumed to have identical orientations and to lie in the plane. The wall energy, γ_2 , in the discrete region is then obtained by summing Eqs. (5.2) and (5.3) along the direction perpendicular to the wall surface, viz

$$\begin{aligned} \gamma_2 &= \frac{1}{a^2} \left\{ \sum_{i,j} 2A_{i,j}a[1 - \cos(\theta_i - \theta_j)] + \sum_i K_i a^3 \sin^2 \theta_i \right\} \\ &= \frac{1}{a^2} \left\{ \sum_{i=2}^{N-1} 2A_{i,i-1}a[1 - \cos(\theta_i - \theta_{i-1})] + \sum_{i=2}^{N-1} K_i a^3 \sin^2 \theta_i \right\} + \frac{2A_{N,N-1}}{a} [1 - \cos(\theta_N - \theta_{N-1})]. \end{aligned} \quad (5.4)$$

Only the nearest-neighbour interactions are considered in the summation.

The total domain-wall energy is then obtained by adding Eqs. (5.1) and (5.4) together.

$$\gamma = \gamma_1 + \gamma_2 + \gamma_3. \quad (5.5)$$

The effect of a pinning site on γ is introduced into the calculation in such a way that only one of the spin planes inside the discrete region lies at the pinning sites, it has a smaller exchange interaction with respect to the spins in neighbouring planes and/or a smaller anisotropy constant. This plane is located at different places whose distance to the wall center is denoted by a variable z , which is a measure of the domain-wall displacement with respect to the pinning site. For a given value of wall displacement z , the domain-wall energy defined by Eq. (5.5) is minimized to give the equilibrium values of the angles, θ_i .

$$\frac{\partial \gamma}{\partial \theta_i} = 0, \quad i = 1, 2, \dots, N. \quad (5.6)$$

The torque equations thus obtained are

$$\theta_2 = \theta_1 + \sin^{-1} \left[a \sqrt{\frac{AK}{A_{1,2}^2}} \sin \theta_1 \right], \quad (5.7)$$

$$\theta_{i+1} = \theta_i + \sin^{-1} \left[\frac{A_{i,i-1}}{A_{i,i+1}} \sin(\theta_i - \theta_{i-1}) + \frac{K_i}{A_{i,i+1}} \frac{a^2}{2} \sin 2\theta_i \right], \quad (5.8)$$

and

$$\sqrt{AK} \sin \theta_N - \frac{A_{N,N-1}}{a} \sin(\theta_N - \theta_{N-1}) = 0, \quad (5.9)$$

where $i = 2, 3, \dots, N - 1$.

This set of equations can be solved by a trial and error method [35] as follows. Given a trial value of θ_1 , the corresponding value of θ_2 can be found

from Eq. (5.7). Repeated application of Eq. (5.8) then gives the values of $\theta_3, \theta_4, \dots, \theta_N$. When the values of θ_{N-1} and θ_N satisfy Eq. (5.9) this indicates that the chosen value of θ_1 is correct; if not, the procedure has to be repeated with another values of θ_1 , etc. The equilibrium values of θ_i thus obtained are used to calculate the total domain-wall energy which is a function of the wall displacement z , $\gamma = \gamma(z)$. The pinning force is estimated with the relationship $[\gamma(z+a/2) - \gamma(z-a/2)]/a$. This procedure is done repeatedly for different combinations of A and K and for different values of the magnetic parameters in the pinning site. The numerical results show that the pinning potential, $V(z)$, may be described to a good approximation as

$$V(z) = 1.97fb[1 - 1/ch^2(0.66z/b)], \quad (5.10)$$

where f is the maximum pinning force given by

$$f = C_p(a/\delta)K[1 - \exp(-3\pi\delta/(10a))]. \quad (5.11)$$

C_p is a constant which depends on the ratios of A and K in the matrix compared to the corresponding parameters in the pinning site, δ is the measure of wall thickness,

$$\delta = (A/K)^{1/2}, \quad (5.12)$$

and b is the interaction range where the maximum pinning force occurs,

$$b \approx 0.5\delta. \quad (5.13)$$

We assume that Eqs. (5.10) and (5.11) are valid for extremely thin walls.

5.3 Activation energy to form a blister

In a finite magnetic field, bowing of the domain wall is possible but the bowed wall has a higher energy than the planar minimum energy wall. The bowed wall is usually called a blister. On neglecting demagnetizing effects, the activation energy required to form a blister consists of the extra wall energy, the interaction energy and the field energy. To simplify the calculation, a uniform distribution (simple cubic) of pins within the magnet is assumed. Certainly when b , the pin interaction range, is much less than l_p , the distance between the nearest neighbouring pins, a uniformly distributed pin array is expected to produce a noticeable pinning because the domain walls will not see this uniformity.

Consider a blister of cylindrical symmetry about the z axis; the plane perpendicular to the z axis is designated by the vector $\vec{x}$ (Fig. 5.2). The domain wall is parallel to this plane in zero field. The extra wall energy of a blister is

$$E_\gamma = \gamma \int (\sec\theta_x - 1) d\vec{x}. \quad (5.14)$$

But

$$\sec\theta_x = \sqrt{1 + (\nabla z)^2} \approx 1 + \frac{1}{2}(\nabla z)^2,$$

since $(\nabla z)^2 \ll 1$.

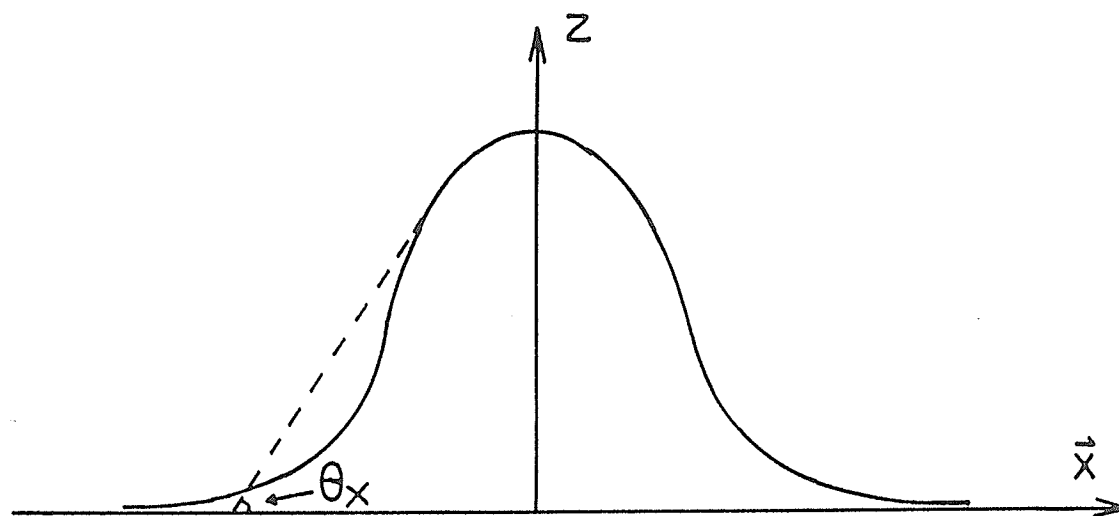


Figure 5.2: A blister of cylindrical symmetry.

Therefore the extra wall energy is

$$E_\gamma \approx \frac{\gamma}{2} \int (\nabla z)^2 d\vec{x}. \quad (5.15)$$

The interaction energy is

$$E_i = \int (V_t/l_p^2) d\vec{x}, \quad (5.16)$$

where V_t is the total pinning potential per single chain of pins (along the z axis) given by

$$V_t = \sum_{n=n_1}^{n_2} [V(z - nl_p) - V(z_\infty - nl_p)], \quad (5.17)$$

where z_∞ is the position of the energy minimum in a magnetic field H and n_1 and n_2 are integers. In principle, n_1 and n_2 should approach minus and plus infinity respectively. However they are actually limited by the underflow of the computer. The interaction potential of the form in Eq. (5.10) is used here, implying that the possible distortion of the pinning potential caused by the existence of other pins or by the magnetic field is neglected.

The field energy can be written as

$$E_H = -2HM \int (z - z_\infty) d\vec{x}. \quad (5.18)$$

The activation energy required to form a blister is then the summation

of three terms,

$$\begin{aligned}
E &= E_\gamma + E_i + E_H \\
&= \int [(\gamma/2)(\nabla z)^2 - 2HM(z - z_\infty) + V_i/l_p^2] d\vec{x}. \quad (5.19)
\end{aligned}$$

In order to obtain a dimensionless equation, the following transformations are performed.

$$\begin{aligned}
v &= z/b, & \vec{r} &= (1.97f/b\gamma)^{1/2} \vec{x}/l_p, \\
h &= 2H M l_p^2 / 1.97f, & \omega(v) &= 1 - 1/ch^2(0.66v), \quad (5.20)
\end{aligned}$$

so that

$$E = \gamma b^2 \varepsilon. \quad (5.21)$$

The dimensionless activation energy ε is

$$\varepsilon = \int \left[\frac{1}{2}(\nabla v)^2 - h(v - v_\infty) + \sum_{n=n_1}^{n_2} \{ \omega(v - n \frac{l_p}{b}) - \omega(v_\infty - n \frac{l_p}{b}) \} \right] d\vec{r}. \quad (5.22)$$

For a blister of cylindrical symmetry, $v(r)$ is independent of the polar angle, the resulting Euler equation is

$$\frac{d^2 v}{dr^2} + \frac{1}{r} \frac{dv}{dr} - 1.32 \sum_{n=n_1}^{n_2} \frac{th[0.66(v - n l_p/b)]}{ch^2[0.66(v - n l_p/b)]} + h = 0. \quad (5.23)$$

The boundary conditions at $r \rightarrow \infty$ are $dv/dr = d^2v/dr^2 = 0$ and at $r = 0$, $dv/dr = 0$. The values of v_∞ can be calculated by using the boundary condition at $r \rightarrow \infty$. Fig. 5.3 shows a typical curve of $v(r \rightarrow \infty)$ against h . For a finite h , there are two solutions; the v_∞ corresponding to the energy minimum, and v'_∞ to the energy maximum. The critical field in the absence of thermal activation, h_0 (or H_0), is defined in such a way that when $h = h_0$ (or $H = H_0$), $v_\infty = v'_\infty$. Fig. (5.4) shows the dimensionless critical field $H_0(2Ml_p^2/f)$ as a function of b/l_p . It can be seen that the critical field, H_0 , is essentially zero for a uniformly distributed pin array when $b/l_p = 0.9$ because the pinning forces completely cancel on both sides of the wall. Some typical values of h_0 are also listed in Table 5.1.

From Eq. (5.20),

$$H_0 \propto f/M. \quad (5.24)$$

As a result,

$$H_0 \propto (H_K/\delta)[1 - \exp(-3\pi\delta/10a)], \quad (5.25)$$

where H_K is the anisotropy field defined by $H_K = 2K/M$. Equation (5.25) shows that the temperature dependence of the critical field, H_0 , is determined by that of the anisotropy field either when the domain wall is very narrow or when the wall thickness is not sensitive to the temperature.

Eq. (5.23) is solved numerically by the Runge-Kutta method (Appendix B). At large r ($=20$), arbitrary values of v ($\approx v_\infty$) and dv/dr (≈ 0) are chosen. These values are finely tuned to obtain a non-oscillatory curve for $v(r)$ with

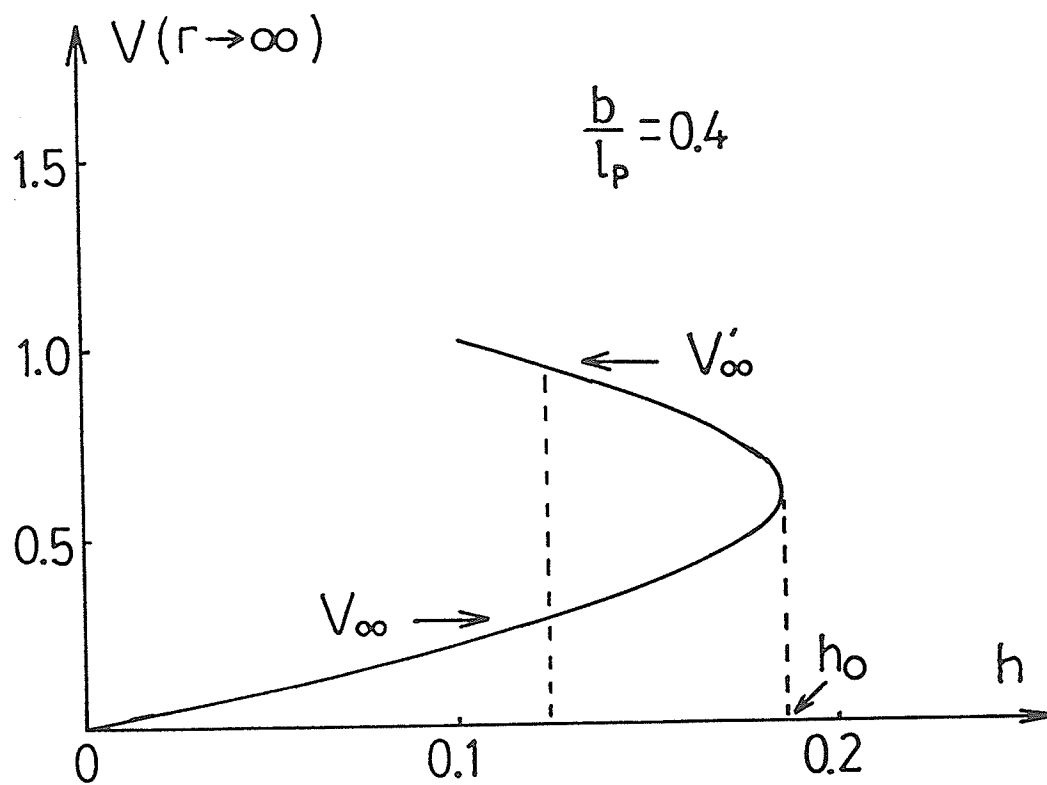


Figure 5.3: There are two values of v at $r \rightarrow \infty$ for a finite value of h . The h_0 is defined at a point where $v_\infty = v'_\infty$.

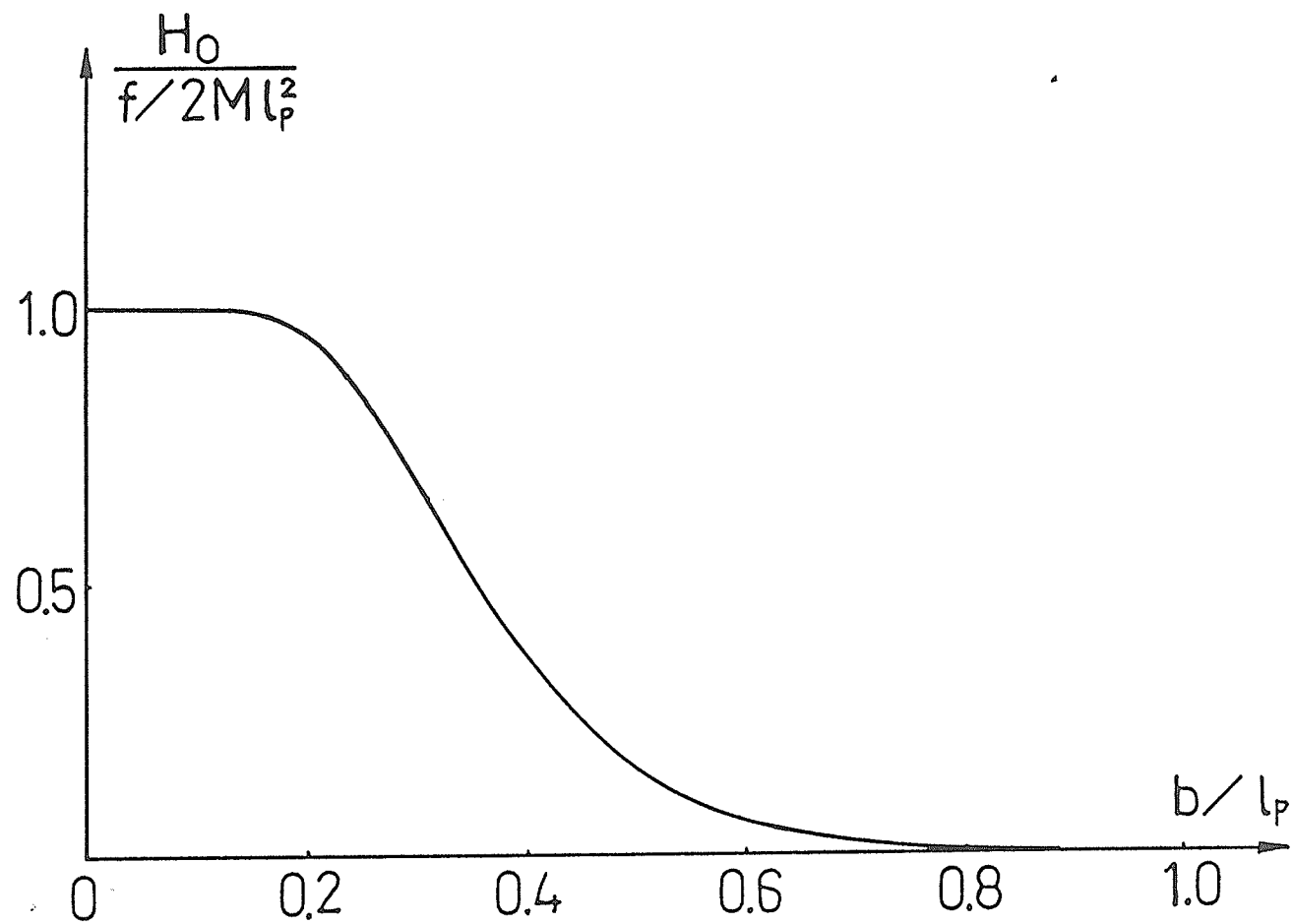


Figure 5.4: The dimensionless critical field as a function of the dimensionless pin interaction range.

the condition that $dv/dr \approx 0$ at $r \approx 0$. The choice of 0.1 as the intervals in r will cause errors in v and dv/dr of the order of 10^{-5} , which are negligible compared to the values of v of the order of one and values of dv/dr of the order of $10^{-1} - 10^{-3}$ in the part of the $v(r)$ curve that contributes the most to the activation energy.

It is found that dv/dr at $r \approx 0$ is very sensitive to the values of v and dv/dr at $r = 20$. However, when $|dv/dr|$ at $r \approx 0$ is less than 10^{-3} , the dimensionless activation energy, which is numerically calculated by using the Simpson method, is not sensitive to the values of v and dv/dr at $r = 20$. This gives evidence that the numerical results for ε are reasonably accurate.

Fig. (5.5) shows the profiles of blisters at two different magnetic fields. As expected, the displacement of the domain wall in the $+\vec{v}$ direction is much more for $h/h_0 = 0.99$ than for $h/h_0 = 0.5$, though locally that is not always true. Fig. (5.6) shows the dimensionless activation energy as a function of the pin interaction range b/l_p and magnetic field h/h_0 . It is obvious that the higher the value of b/l_p , the smaller the activation energy. The activation energy increases linearly on decreasing h/h_0 only when h/h_0 is close to one. A further decrease in h/h_0 will cause ε to deviate from this linear relationship and to increase rapidly. The dashed line in Fig. (5.6) is the prediction of the weak-pinning model proposed by Gaunt [7]. Gaunt's weak-pinning model studies the pinning process in the case when $b/l_p = 0$, although this was not explicitly pointed out in his paper. Since the pinning potential proposed by Gaunt (Eq. 1.1) is a good approximation to the real potential only near a

Table 5.1: A list of constants in the expression for dimensionless activation energy. The last column shows the dimensionless critical field h_0 .

b/l_p	$C_{\epsilon 1}$	$C_{\epsilon 2}$	$C_{\epsilon 3}$	$C_{\epsilon 4}$	h_0
0	577	14.6	665	2.83	0.508
0.1	139	11.5	639	2.77	0.508
0.2	90	31	130	1.27	0.484
0.3	30	16.9	42.5	0.87	0.347
0.4	17.3	14.8	20.9	0.76	0.184

point where the maximum pinning force occurs, it is not surprising to see large deviation between the predictions of the weak-pinning model and the $b/l_p = 0$ curve when h/h_0 is not close to one. This is reasonable because the condition that h/h_0 is close to one is equivalent to the condition of *being near a point where the maximum pinning force occurs*.

The dimensionless activation energy may be approximately described by the analytical expression.

$$\varepsilon = C_{\epsilon 1} \exp[-C_{\epsilon 2}(H/H_0)^2] + C_{\epsilon 3} \exp[-C_{\epsilon 4}(H/H_0)](1 - H/H_0), \quad (5.26)$$

where $C_{\epsilon i}$ ($i = 1, 2, 3, 4$) are constants which are listed in Table 5.1.

5.4 Application to a $\text{Sm}(\text{CoNi})_{2.5}$ permanent magnet

5.4.1 Description of previous work

A $\text{Sm}(\text{CoNi})_{2.5}$ permanent magnet has a hexagonal structure of the CaCu_5 type. It possesses a giant coercivity at low temperatures (~ 53 kOe at 4.2

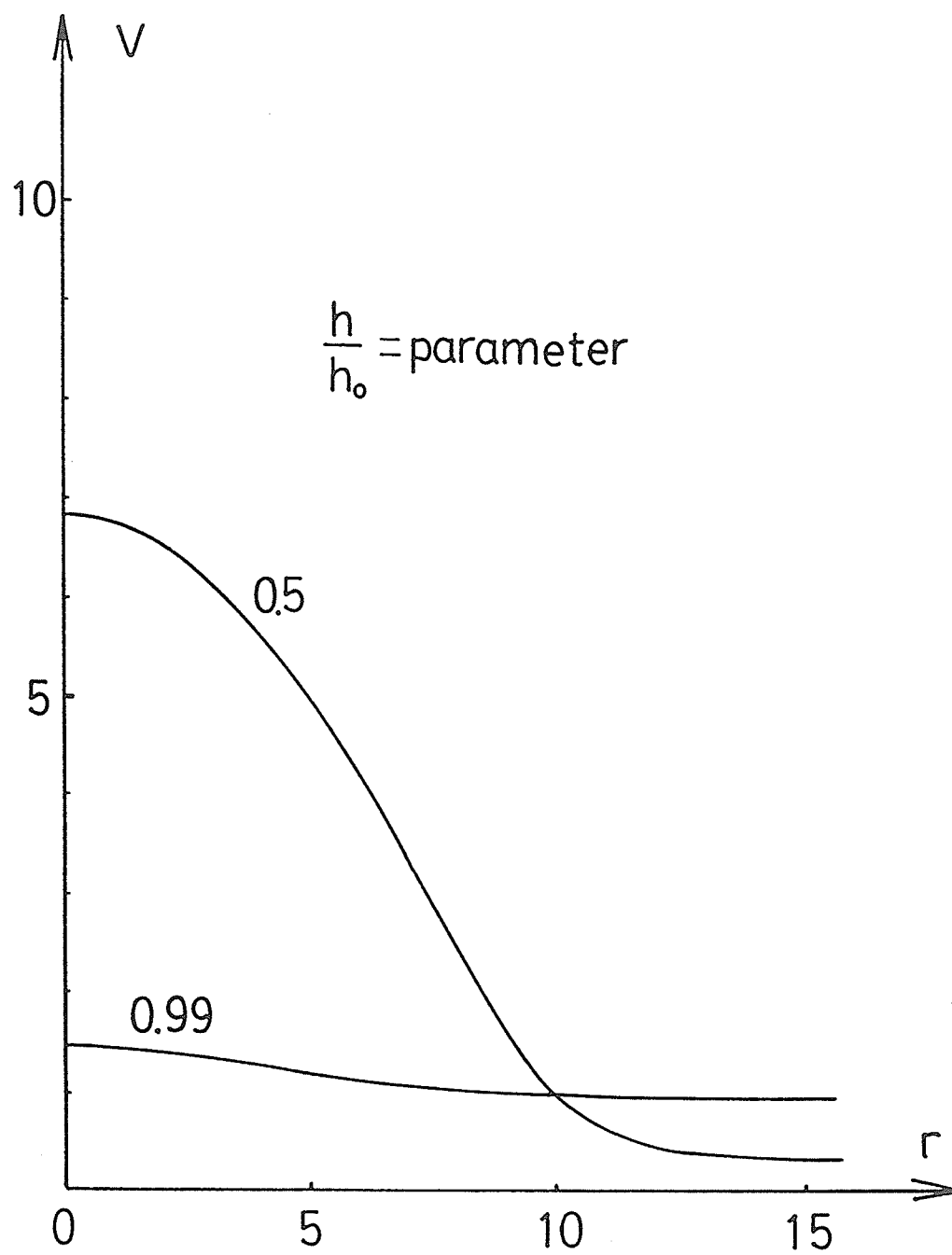


Figure 5.5: The profiles of blisters when $b/l_p = 0.1$.

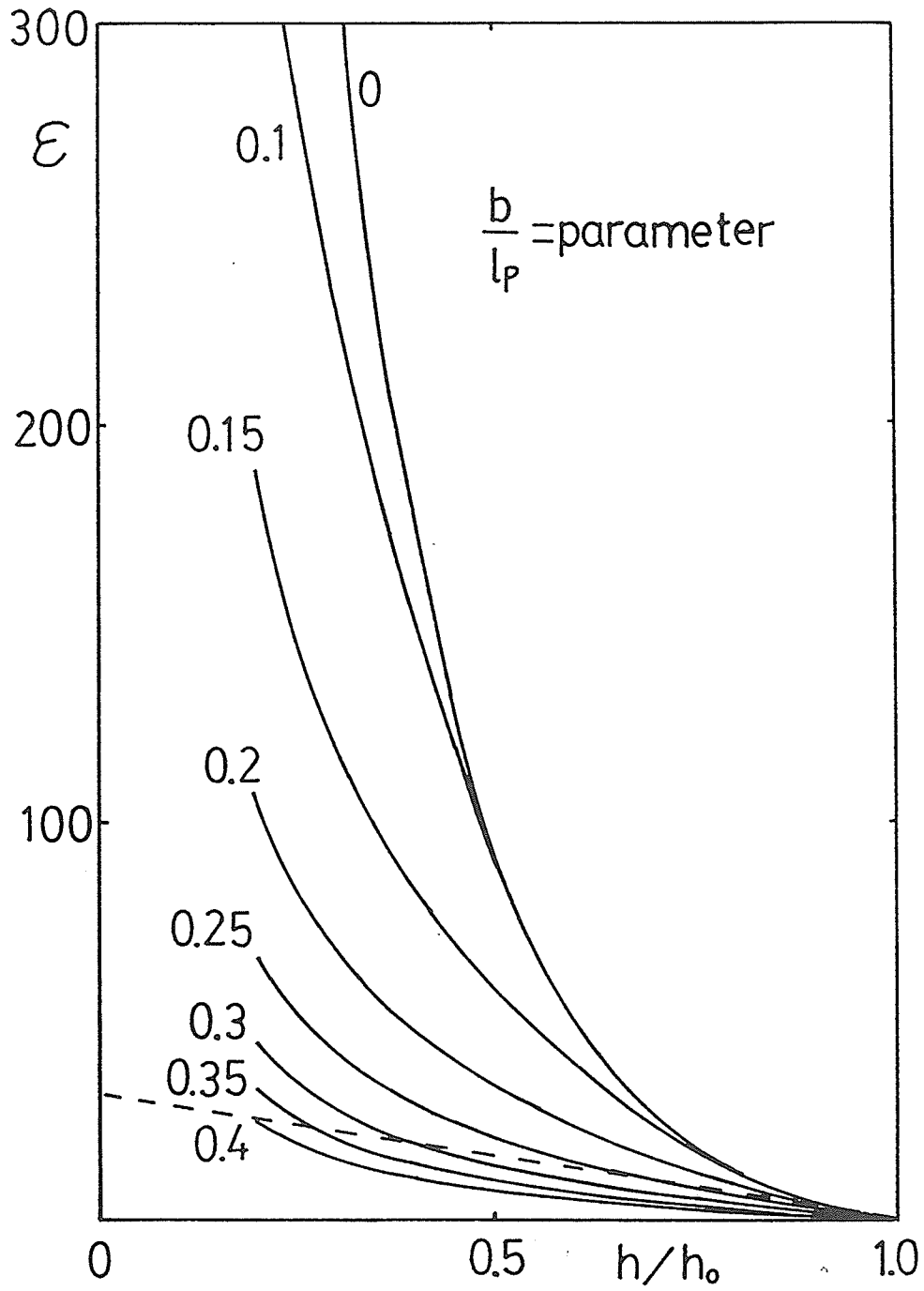


Figure 5.6: Dimensionless activation energy as a function of both h/h_0 and b/l_p ; the dashed line is the prediction from Gaunt's weak pinning model.

K). The coercivity drops rapidly as the temperature increases and has a value of ~ 6 kOe at 225 K. The Curie temperature is about 500 K. Parker and Oesterreicher [38] had tried to describe the temperature dependence of coercivity by a model proposed by Egami [12]. In this model the coercivity has its origin in the Peierls potential which exists even in perfect materials (see section 1.3). Such a pinning mechanism is sometimes called intrinsic pinning. In the case of SmCo_5 , the coercivity caused by intrinsic pinning is estimated by Kütterer et al. [23] to be about 50 to 100 Oe at 4.2 K. The compound $\text{Sm}(\text{CoNi})_{2.5}$ is a pseudobinary compound whose Peierls potential should not be much different from that for SmCo_5 because both magnets have the same structure. However the measured coercivity of $\text{Sm}(\text{CoNi})_{2.5}$ is 10^3 times higher than the prediction of Egami's model, indicating that it is necessary to consider the wall pinning by magnetic inhomogeneities occurring at those places where the Co atoms are replaced by Ni atoms.

5.4.2 Specimen preparation and TEM observation

The specimens of composition $\text{Sm}(\text{CoNi})_{\sim 2.5}$ were prepared for a transmission-electron microscope (TEM) study. The X-ray diffraction pattern shows that the compound has a hexagonal structure. The electrolyte used to polish the specimens was a solution of 15 % perchloric acid and 85 % glacial acetic acid. The current density was about 1.8 A/cm^2 . The specimen was held at 77 K during the TEM observation. No pinning site is visible from the transmission-electron micrograph, indicating that the pinning sites are of

small size ($< 14 \text{ \AA}$), perhaps of atomic size.

Since Parker et al. [38] had made measurement of coercivity of $\text{Sm}(\text{CoNi})_{2.5}$ over a temperature range from 4.2 K to 225 K, it is not necessary to repeat the same measurement again. Only a few points over 200 K were measured here. The results agree with Parker et al. reasonably well.

In the next section, the model presented in section 5.3 will be applied to $\text{Sm}(\text{CoNi})_{2.5}$, all the necessary experimental data are taken from reference [38].

5.4.3 Temperature dependence of coercivity

With Eqs. (5.11), (5.21), (5.25) and Table 5.1, and by setting $E = 25 kT$ [7], where k is the Boltzmann's constant, the temperature dependence of coercivity of a $\text{Sm}(\text{CoNi})_{2.5}$ magnet can now be described by a model which incorporates the two contributions to the decrease of coercivity as a function of temperature into a single theory. In addition there is no limitation on the value of H_c/H_0 .

Since $\text{Sm}(\text{CoNi})_{2.5}$ has a hexagonal structure, and the spacial distribution of the pinning sites is unknown, the physical significance of the parameter a in Eq. (5.11) is that it is the distance between two nearest atomic planes, which has the value of either $a = 4.3 \text{ \AA}$ for $\bar{1}010$ planes, or $a = 2.48 \text{ \AA}$ for $\bar{2}110$ planes, as measured by the X-ray diffraction technique.

The temperature dependencies of the anisotropy constant and the spon-

taneous magnetization are obtained from reference [38], viz

$$K = 3.06 \times 10^8 (1 - 1.42 \times 10^{-4} T^{3/2}) \text{ erg/cm}^3, \quad (5.27)$$

and

$$M = 534 (1 - 2.55 \times 10^{-5} T^{3/2}) \text{ erg/Oe cm}^3. \quad (5.28)$$

From these relations, the temperature dependence of δ can be written as

$$\delta = \delta_0 (1 - 2.55 \times 10^{-5} T^{3/2}) (1 - 1.42 \times 10^{-4} T^{3/2})^{-1/2}, \quad (5.29)$$

where the relation of Eq. (4.40) has been used.

The experimental values of the coercivity at different temperatures (Ref. 38) will be used to test the model by a least-square fitting. The procedure is as follows. For a combination of b/l_p and a , the two parameters, δ_0 and $C_p a/l_p^2$, are adjusted to get the best fitting. The results are shown in Fig. 5.7. The good fitting verifies the validity of the proposed model. The corresponding values are $b/l_p = 0.2$, $a = 4.3 \text{ \AA}$, $\delta_0 = 2.66 \text{ \AA}$ and $C_p a/l_p^2 = 1.29 \text{ \AA}$.

Since $b \approx 0.5\delta$, $b \approx 1.33 \text{ \AA}$, and $l_p \approx 6.64 \text{ \AA}$. This value is almost double the value estimated from the maximum pin density of 2.5 Ni atoms per unit cell. However, this is not too surprising since the model is a simplified one and also the calculated value of l_p represents an effective pin separation. The value $a = 4.3 \text{ \AA}$ gives the best agreement between the predicted values of

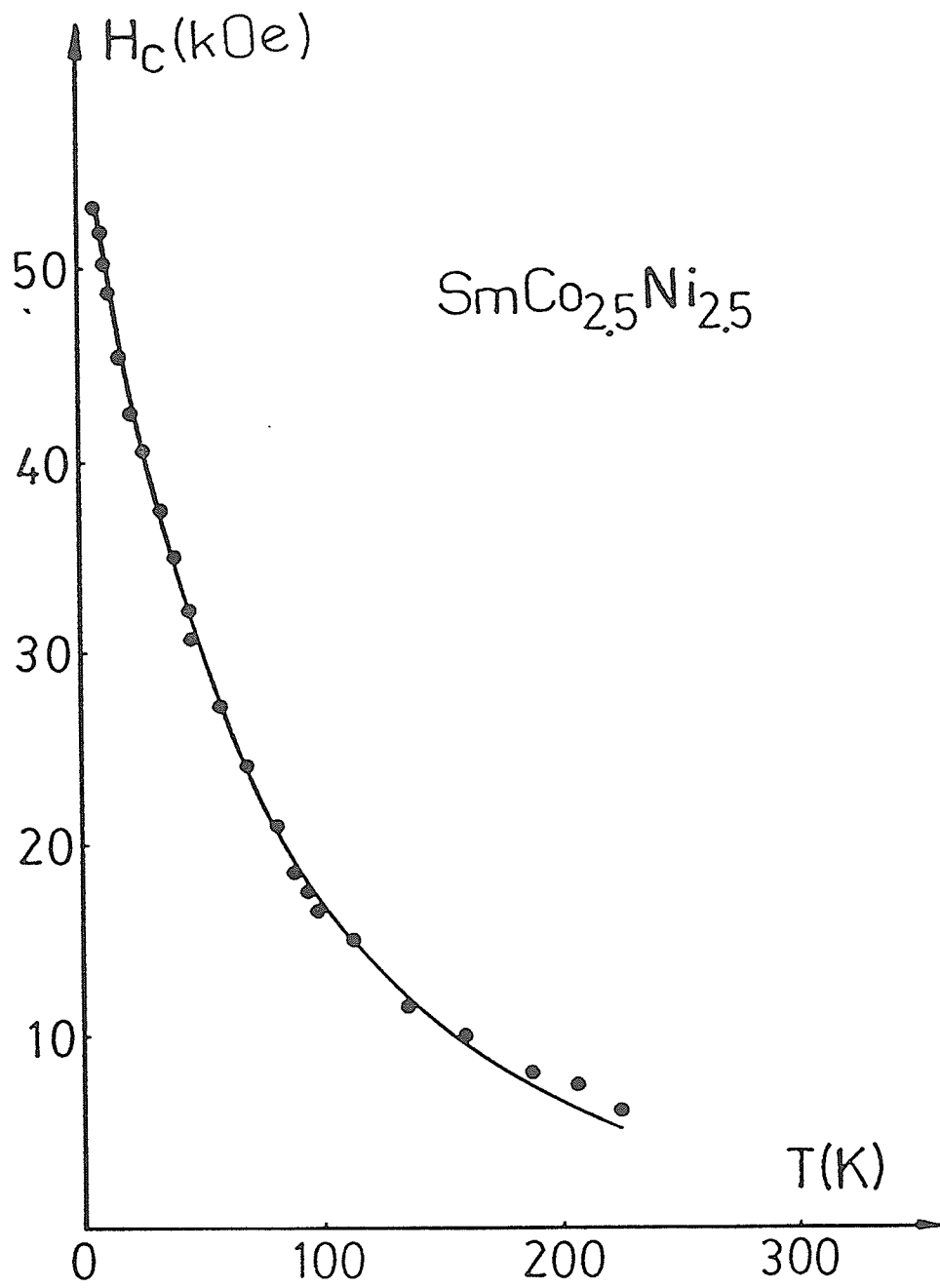


Figure 5.7: Temperature dependence of coercivity of a $\text{Sm}(\text{CoNi})_{2.5}$. The solid line is the prediction of the present model.

H_c and the experimental ones, indicating that the $\bar{1}010$ walls are dominant.

It is interesting to see how the critical field H_0 varies with temperature. Fig. (5.8) shows that H_0 drops by as much as 40 percent from 0 K to 225 K, indicating a significant variation in the energy barriers with temperature. Fig. (5.9) plots the ratio of $H_c(T)/H_0(T)$ against the temperature, showing that the thermal activation of domain walls is also very important. The dashed line is the prediction of Gaunt's weak pinning model. As is expected, the two curves agree well only near a point where H_c/H_0 is close to one.

5.4.4 Further predictions of the model

The model proposed in this chapter has been tested for a system with thin walls pinned by pins of atomic dimensions. For the first time it is shown that the giant coercivity of a $\text{Sm}(\text{CoNi})_{2.5}$ magnet originates from magnetic inhomogeneities within the magnet. Besides a satisfactory agreement between the predicted and the experimental values of coercivity, this model will predict other interesting things.

In Fig. (5.10), several $H_c(T)$ curves are plotted, where the critical field $H_0(T)$ is the same in all cases. But the parameters, γb^2 , are different, and $\gamma b^2(a) > \gamma b^2(b) > \gamma b^2(c)$. It can be seen that the larger the value of γb^2 , the more linear the $H_c(T)$ curve and the weaker the contribution of thermal activation of the wall to the temperature dependence of coercivity.

The curve c in Fig. (5.10) is sometimes observed experimentally [39]. Although Durst et al. attribute it to the wall-pinning originating from the

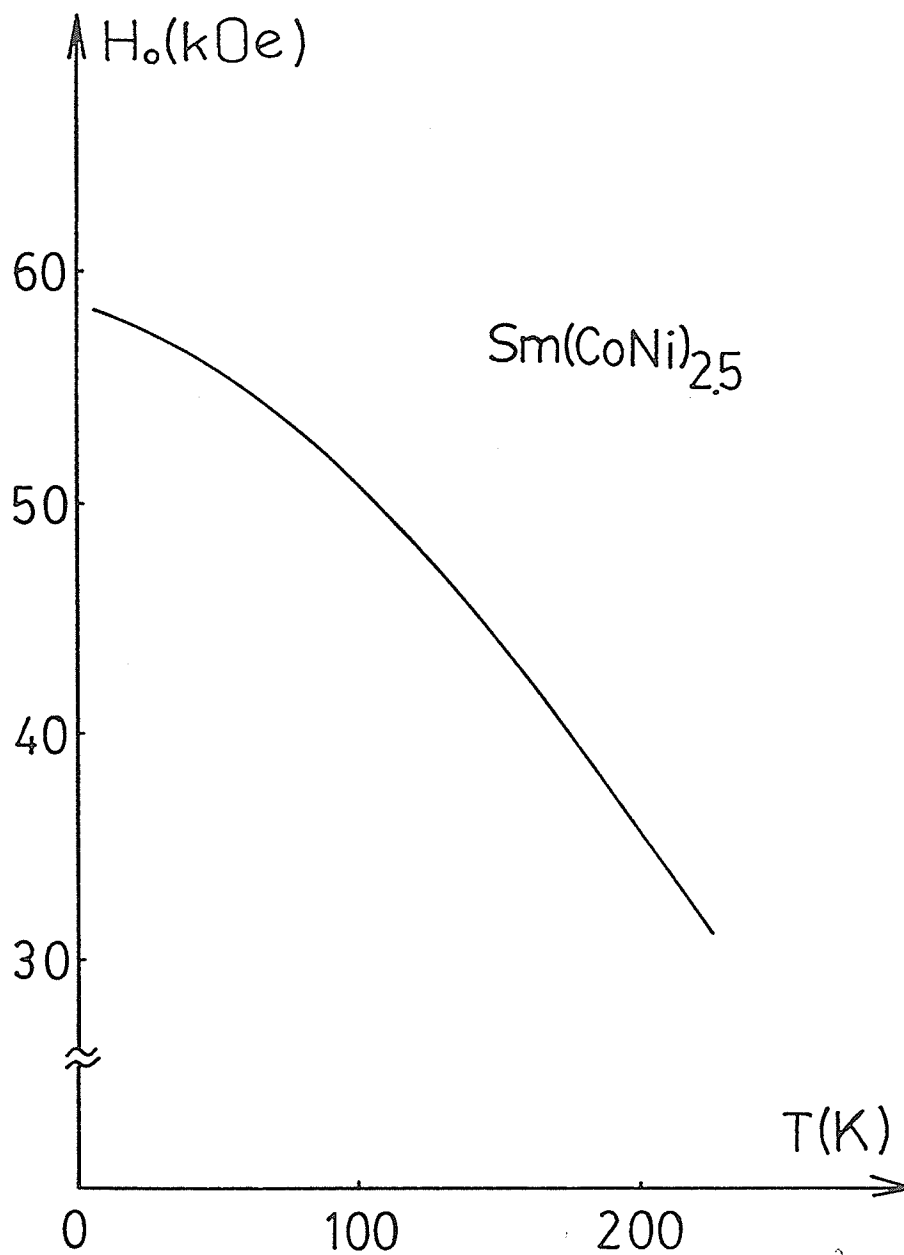


Figure 5.8: Critical field at different temperatures.

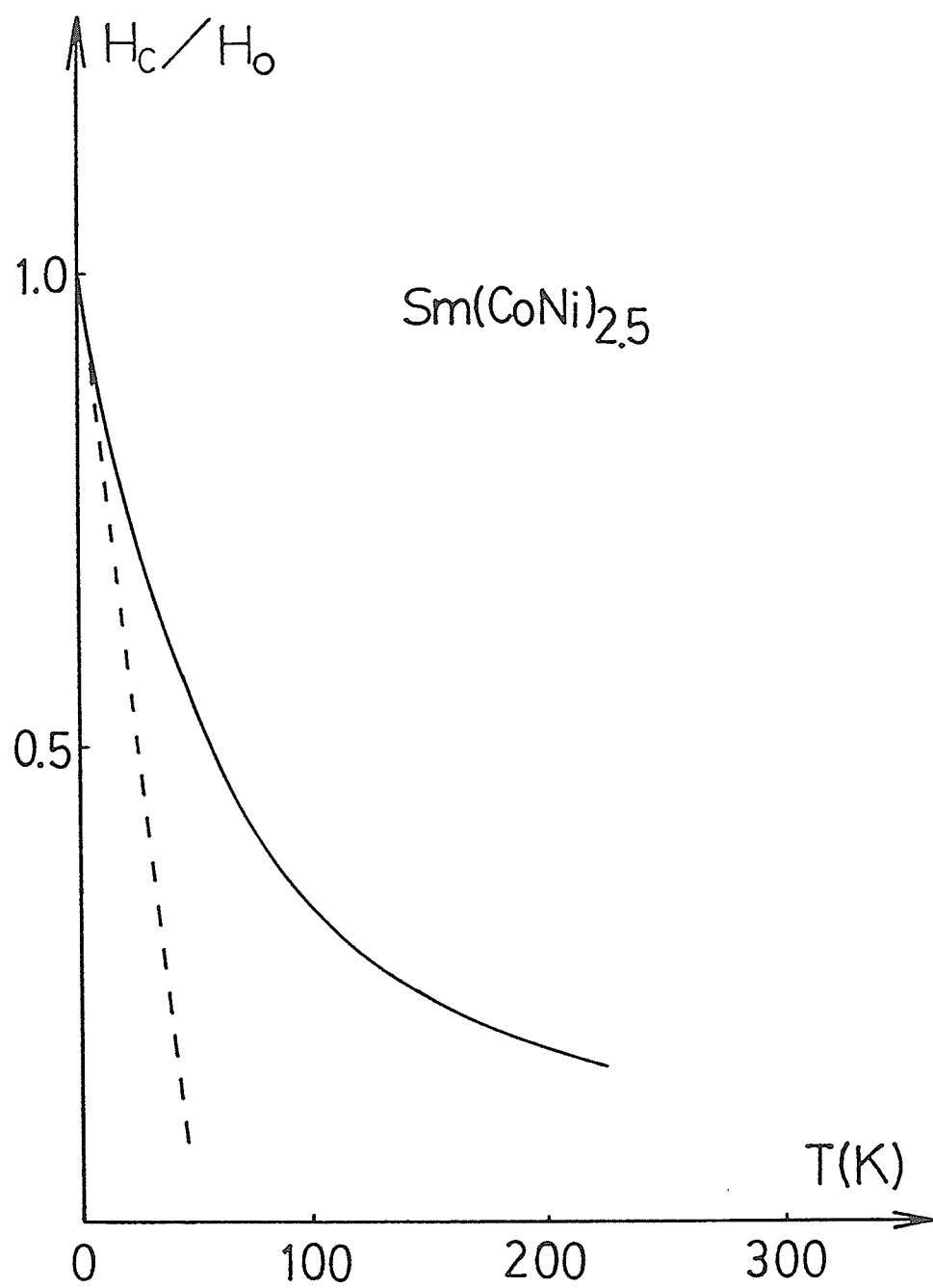


Figure 5.9: Thermal activation of walls are getting stronger on increasing temperature.

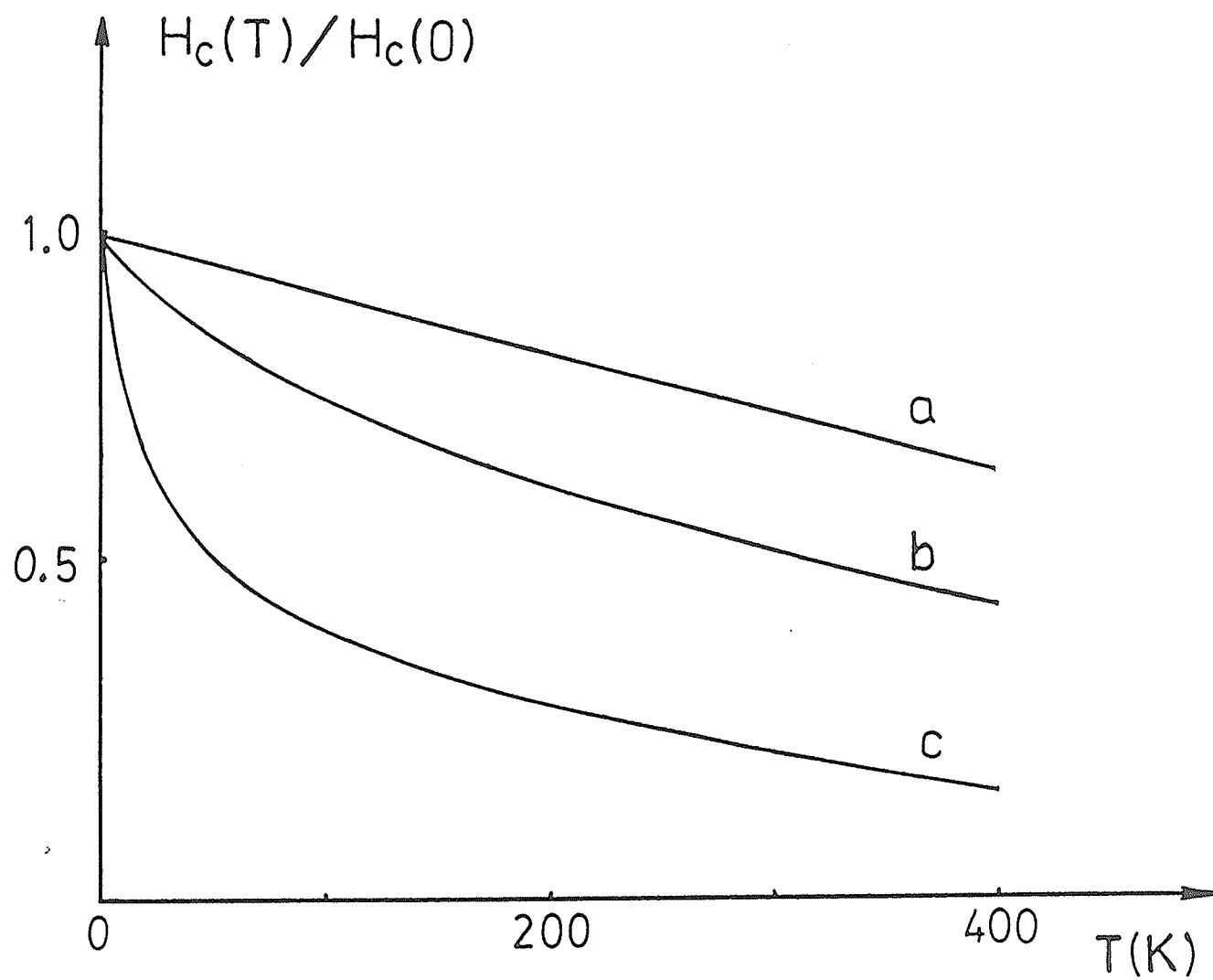


Figure 5.10: Illustration of the effect of γb^2 on the shape of $H_c(T)$ curve, where $\gamma b^2(a) > \gamma b^2(b) > \gamma b^2(c)$.

Peierls potential, even they themselves are in doubt about this claim, because the intrinsic pinning cannot produce magnetic hardness as large as ~ 30 kOe. The present model may give a reasonable explanation for this exponential decay-like curve. However further work will be necessary.

Chapter 6

Discussion

6.1 Summary

The present work is several successful attempts to try to describe the temperature dependence of coercivity on a reasonable physical basis. The models presented in Chapters 3, 4, 5 have the same idea, that is, the decrease of coercivity as a function of temperature originates from two factors. One is the temperature dependence of the intrinsic magnetic properties of the magnets and the other is the thermal activation of domain walls. The two factors are, in general, of equal importance, when the temperature dependence of coercivity is considered over a sufficiently wide temperature range. Any model that neglects either factor without good reason will be insufficient.

Recounting the features of the present study, the following conclusions may be drawn.

(a) The $\{110\}$ lamellar junctions in a $\text{Pt}_{0.7}\text{Ni}_{0.3}\text{Fe}$ alloy are the pinning sites which pin the domain walls, resulting in the magnetic hardness. The temperature dependence of coercivity of this alloy is mainly due to the thermally activated process of domain wall unpinning.

(b) The high coercivity of a MnAlC permanent magnet originates from the domain wall pinning at grain boundaries. Both the variation of the height of energy barriers with the temperature and the thermal activation of domain walls are important to the temperature dependence of coercivity of this magnet.

(c) The domain wall pinning by Ni atoms is the mechanism to account for the giant coercivity of a $\text{Sm}(\text{CoNi})_{2.5}$ ferromagnet. Similar to MnAlC,

both contributions are equally important to the temperature dependence of coercivity. The strong thermally activated process of domain wall unpinning results in the exponential decay-like $H_c(T)$ curve.

6.2 Further work

Chapter 4 has treated the pinning process when the planar pinning sites are thinner than the wall thickness and when the ratio of H_c/H_0 is close to one. Certainly, these limitations are not generally applicable. Further work to find general results will be useful, since the planar pinning sites are very often observed in many important commercially hard magnets such as SmCo_5 , the magnets based on Nd-B-Fe, etc. The general results are expected to be similar in form to Eqs. (5.25) and (5.26), since only the ratio of pin width to wall thickness appears in the expression for the maximum pinning force (Eq. 5.11) and no particular assumption is made in Chapter 5.

Schneider et al. [40] and Grössinger et al. [41] observed the coercivity of Nd-B-Fe based magnets have an exponential decay-like behaviour over a temperature range from 77 K to a point near the Curie temperature. In order to describe their experimental results, they assumed a mixture of different mechanisms for the high coercivity. However, in the light of the present work, this exponential decay-like curve could be simply due to the strong thermal activation of domain walls at high temperature, as illustrated in Fig. (5.10). It would probably be unnecessary to introduce the second pinning mechanism into the analysis.

Besides the planar pinning sites, the spherical pinning sites, either magnetic or non-magnetic, are also very often observed in real magnets. It will be very useful to study the pinning process of this mechanism based on the ideas presented in this thesis. A calculation of the pin interaction potential would be necessary.

Finally it is worthwhile to point out that the present work will certainly have some bearing on magnetic viscosity theories which based on domain-wall movement and on activation energies of domain-wall unpinning.

Appendix A

The influence of H on the maximum pinning force from a planar pinning site

When the influence of a magnetic field is considered, the domain-wall energy per unit area may be written as

$$\gamma = \int [A_i \left(\frac{\partial \theta}{\partial z'} \right)^2 + K_i \cos^2 \theta + M_i H \sin \theta] dz', \quad (\text{A.1})$$

where the last term in the integrand is the contribution of the magnetostatic energy (Fig. 4.1).

The stable wall structure is determined by the condition $\delta\gamma = 0$, which gives, after integrating the Euler equations in three different regions,

$$K_i \cos^2 \theta + M_i H \sin \theta - A_i \left(\frac{\partial \theta}{\partial z'} \right)^2 = C'_{1i}, \quad (\text{A.2})$$

where C'_{1i} are constants.

The boundary conditions that $\partial\theta/\partial z' = 0$ at $\theta = \pm\pi/2$ give $C'_{11} = -M_1H$ and $C'_{13} = M_1H$. The author is unable to find an expression for C'_{12} when H is nonzero. But the order of magnitude of C'_{12} may be found in the following way. The existence of a finite field H will impose an additional torque on the spins inside the wall. This additional torque, in the order of M_iH , is with or against the torque of order M_iH_K , related to uniaxial anisotropy energy and depends on the sign of θ ($-\pi/2 \leq \theta \leq +\pi/2$), where H_K is the uniaxial anisotropy field defined by $H_K = 2K_i/M_i$. Usually the condition $H \ll H_K$ and therefore $M_iH \ll M_iH_K$ is satisfied. This means that the magnetic field H will have a negligible effect on the orientation of the spins inside the domain wall. The spin configuration of the wall should not be altered significantly by the presence of H . Thus the value of C'_{12} should not deviate much from its value of zero in zero field. Specifically, the ratio of $|C'_{12}/K_2|$ should be much less than one, since K_2 is usually a large number of the order of at least 10^6 for hard magnets. This claim may be examined in another way. Suppose the planar-pinning site is located, by the thermal activation process, near the domain wall edges, $\theta \approx -\pi/2$ or $\theta \approx +\pi/2$. Then $\partial\theta/\partial z' \approx 0$ and from Eq. (A.2), $C'_{12} \approx -M_2H$ or $+M_2H$. This implies that C'_{12} is of the order of M_2H and that $|C'_{12}/K_2|$ is much less than one, since H is usually much less than the anisotropy field H_K .

The dimensionless parameters are defined as

$$w = W/\delta_2, \quad c_{12} = C'_{12}/K_2,$$

$$h_1 = HM_1/K_1, \quad h_2 = HM_2/K_2. \quad (\text{A.3})$$

Only the case where $w \ll 1$ will be studied. Therefore all dimensionless parameters defined in Eq. (A.3) are much less than one. Let ε_d represent any of the dimensionless parameters. When the domain-wall energy is calculated, the integrands will be, if necessary, expanded into power series up to the second power of ε_d . From the argument in section 4.2, the maximum values of $\theta_{1,2}$ at pin boundaries are about $5\pi/22$, which is less than one. Therefore the trigonometric functions can be expanded into series to the fourth power of θ without significant error. Now the wall energy in the three regions (Fig. 4.1) can be written as

$$\gamma_1 + \gamma_3 = 2\sqrt{A_1 K_1} \left[2 - \left(1 + \frac{h_1^2}{8}\right)(\theta_2 - \theta_1) - \frac{h_1}{4}(\theta_2^2 - \theta_1^2) + \left(\frac{1}{6} - \frac{h_1^2}{48}\right)(\theta_2^3 - \theta_1^3) - \frac{h_1}{24}(\theta_2^4 - \theta_1^4) \right], \quad (\text{A.4})$$

and

$$\begin{aligned} \gamma_2 = c_{12} K_2 w \delta_2 + 2\sqrt{A_2 K_2} \{ & [1 - \frac{c_{12}}{2} - \frac{c_{12}^2}{8}](\theta_2 - \theta_1) + h_2(\frac{1}{4} + \frac{c_{12}}{8})(\theta_2^2 - \theta_1^2) \\ & - (\frac{1}{6} + \frac{c_{12}}{12} + \frac{h_2^2}{24} + \frac{c_{12}^2}{16})(\theta_2^3 - \theta_1^3) + h_2(\frac{1}{24} + \frac{c_{12}}{12})(\theta_2^4 - \theta_1^4) \}. \end{aligned} \quad (\text{A.5})$$

The expression for θ_2 as a function of θ_1 may be obtained from the equation

$$w = \int_{\theta_1}^{\theta_2} (\cos^2 \theta + h_2 \sin \theta - c_{12})^{-1/2} d\theta. \quad (\text{A.6})$$

By using the same approximations mentioned before and an iterative procedure, θ_2 is found to be

$$\begin{aligned}\theta_2 = \theta_1 + (1 - \frac{c_{12}}{2})w + \frac{1}{2}(h_2 - w)w\theta_1 - (\frac{1}{2} + \frac{c_{12}}{4})w\theta_1^2 \\ + (\frac{5}{12}h_2 + \frac{3}{4}w)w\theta_1^3 + (\frac{1}{4} + \frac{10}{27}c_{12})w\theta_1^4.\end{aligned}\quad (\text{A.7})$$

By integrating Eq. (A.2) in three different regions, the relations between z' and θ can be obtained. There will be three integral constants, C'_{2i} , in the $z' = z'(\theta)$ expressions. The C'_{21} and C'_{23} are determined to be non-zero by the boundary condition that θ continuous at pin boundaries. The constant C'_{22} can be estimated in the following way. The orientations of spins at and near the center of the domain wall are, or are almost, perpendicular to the magnetic field and the easy axis of the magnet, these spins contribute the most to the wall energy. If a pinning site is at the wall center, it will primarily reduce the wall energy. In other words, the minimum pin interaction energy will happen at, or at least near a point of the wall center ($z' = 0$). This condition will set the constant C'_{22} to zero. In a similar way to that mentioned in section 4.2, the expression of θ_1 as a function of the dimensionless wall displacement $Z = z/\delta_2$ is calculated as

$$\theta_1 = \frac{1}{6}h_2 - \frac{1}{2}w - (1 - \frac{1}{2}c_{12})Z + (\frac{1}{4}w + \frac{1}{6}h_2)Z^2 + (\frac{1}{6} - \frac{1}{12}c_{12})Z^3 - (\frac{5}{24}w - \frac{1}{11}h_2)Z^4.\quad (\text{A.8})$$

With Eqs. (A.4), (A.5), (A.7) and (A.8) together, the wall energy per

unit area can finally be written as

$$\gamma = \gamma_0 + U(Z), \quad (\text{A.9})$$

where

$$\gamma_0 = 4\sqrt{A_1 K_1} - 2(\sqrt{A_1 K_1} - \sqrt{A_2 K_2})w + c_{12}K_2 w \delta_2 \left(\sqrt{\frac{A_1 K_1}{A_2 K_2}} - 1 \right), \quad (\text{A.10})$$

and

$$\begin{aligned} U(Z) = & 2\left[\sqrt{A_1 K_1}\left(\frac{1}{2}h_1 + \frac{1}{6}h_2\right) - \sqrt{A_2 K_2}\left(\frac{2}{3}h_2\right)\right]wZ \\ & + 2(\sqrt{A_1 K_1} - \sqrt{A_2 K_2})(1 - c_{12})wZ^2 \\ & - 2\left[\sqrt{A_1 K_1}\left(\frac{1}{6}h_1 - \frac{4}{29}h_2\right) - \sqrt{A_2 K_2}\left(\frac{5}{174}h_2\right)\right]wZ^3 \\ & - \frac{5}{3}(\sqrt{A_1 K_1} - \sqrt{A_2 K_2})(1 - c_{12})wZ^4. \end{aligned} \quad (\text{A.11})$$

The maximum pinning force, f' , per unit area of a planar pinning site is then obtained by the equation

$$\frac{d^2 U(Z)}{dZ^2} = 0. \quad (\text{A.12})$$

This happens at

$$Z \approx \sqrt{0.2}, \quad (\text{A.13})$$

with the expression

$$f' = 1.2(1 - c_{12})(\sqrt{A_1 K_1} - \sqrt{A_2 K_2})w/\delta_2 \\ + [(0.8h_1 + 0.5h_2)\sqrt{A_1 K_1} - 1.3h_2\sqrt{A_2 K_2}]w/\delta_2. \quad (\text{A.14})$$

For a pinning site, the second term in Eq. (A.14) is positive. This means that the maximum pinning force increases with a driving field. This is a source of the magnetic inertia of a domain wall. It is of interest to see what percentage of the total pinning force the field-related term is. Assume that $M_2 = 0.8M_1$, $K_2 = 0.8K_1$, $A_2 = 0.8A_1$ and $h_1 = 0.1$, then the second term in Eq. (A.14) is only about 10 % of the total pinning force. This is a typical maximum value of the percentage since the choice of $h_1 = 0.1$ is a typical maximum value of the driving field.

The conclusion is in favour of the assumption in Chapter 4 that the influence of a magnetic field on the pin interaction potential is not important, as long as the magnetic field is much less than the anisotropy field.

Appendix B

Runge-Kutta method

Suppose that the equation

$$\begin{cases} y'' &= g(x, y, y') \\ y(x_0) &= y_0, \quad y'(x_0) = y'_0 \end{cases}$$

has a unique solution.

Define $y' = z$, then the above equation can be rewritten as

$$\begin{cases} y' &= f(x, y, z) = z \\ z' &= g(x, y, z) \\ y(x_0) &= y_0, \quad z(x_0) = y'_0. \end{cases}$$

The numerical solutions are

$$\begin{cases} y_{n+1} &= y_n + \frac{1}{6}[k_1 + 2(k_2 + k_3) + k_4]; \\ z_{n+1} &= z_n + \frac{1}{6}[m_1 + 2(m_2 + m_3) + m_4], \end{cases}$$

where

$$k_1 = hf(x_n, y_n, z_n);$$

$$m_1 = hg(x_n, y_n, z_n);$$

$$k_2 = hf(x_n + h/2, y_n + k_1/2, z_n + m_1/2);$$

$$m_2 = hg(x_n + h/2, y_n + k_1/2, z_n + m_1/2);$$

$$k_3 = hf(x_n + h/2, y_n + k_2/2, z_n + m_2/2);$$

$$m_3 = hg(x_n + h/2, y_n + k_2/2, z_n + m_2/2);$$

$$k_4 = hf(x_n + h, y_n + k_3, z_n + m_3);$$

$$m_4 = hg(x_n + h, y_n + k_3, z_n + m_3).$$

$$y(x_{n+1}) \approx y_{n+1}, \quad z(x_{n+1}) \approx z_{n+1}, \quad (n = 0, 1, 2, \dots)$$

The parameter h is the length of the intervals in x ; the errors of final results are in the order of h^5 .

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