Uniaxial Anisotropy of a Permalloy Crystal

SEIJI KAYA

Physics Department, Tokyo University, Tokyo, Japan

If Permalloy (76 percent Ni—24 percent Fe) which has been previously quenched from 600°C is annealed at 490°C, its energy content decreases with time of annealing, not monotonically but in two steps. By direct measurement of the time-rate of energy decrease, it was concluded that the first step is connected with formation of short-range order and the second with that of long-range order.

The effect of a magnetic field applied during the course of cooling on the maximum permeability is associated mainly with formation of short-range order. Taking into consideration the fact that the saturation of the ordered state is about 4 percent higher than that of the disordered one, one would expect the short-range order to develop in the form of prolate ellipsoids elongated in the direction of spontaneous magnetization. Torque curves, measured on a disk cut from a Permalloy crystal parallel to the (110) plane and heat treated in a magnetic field showed uniaxial anisotropy, superposed on the ordinary cubic one. This uniaxial anisotropy increases with the formation of short-range order, but the formation of long-range order, followed after that of short-range one, has a tendency to annihilate it. The constricted hysteresis loop, which is characteristic of the Perminvar property, becomes most eminent at the end of short-range order formation.

If we assume two neighboring uniaxial domains, whose axes intersect perpendicularly with each other, and whose boundary bisects the angle between them, we can easily calculate the hysteresis loop of constricted form. The main characteristic of this model is that the displacement of the boundary cannot take place even in a strong field.

I. EFFECTIVE TEMPERATURE RANGE FOR MAGNETIC FIELD COOLING

SINCE the discovery of Kelsal[1] in 1934, it has been well known that, in some nickel-iron alloys, application of a magnetic field during cooling from a high temperature strongly influences their ferromagnetic properties, e.g., a large increase in maximum permeability and a large decrease in saturation magnetostriction are observable in the direction of the previously applied field, and conversely for the perpendicular direction. This phenomenon indicates that magnetic domains within the specimen are held firmly parallel or antiparallel to the direction in which the field was applied during cooling. Later experiments[2] have shown that this tendency is closely related to the formation of superlattice of Ni_3Fe type, which is known to take place in the temperature range from 600°C to 400°C in the course of cooling.

In an experiment made to determine more definitely the effective temperature range for field cooling, Tomono[3] applied a magnetic field of 50 oersteds only in a limited temperature interval of 30°C while the specimen was cooling and measured the dependence of saturation magnetostriction on the temperature at which the field was applied. Figure 1 illustrates the results thus obtained for 76 Permalloy. As shown in the figure, the temperature range between 540°C and 510°C is most effective for field cooling.

When the specimen is cooled in this temperature range without the presence of an external magnetic field, magnetic domains, orientations of which are distributed at random, will show the tendency to retain the same randomness which characterized this distribution at high temperature. This is why slowly cooled Permalloy has a low permeability.

[1] G. A. Kelsal, Physics **5**, 169 (1934).
[2] S. Kaya, J. Faculty of Science, Hokkaido University **2**, 29 (1938).
[3] Y. Tomono, J. Phys. Soc. Japan **4**, 298 (1948).

II. ANNEALING AT THE CRITICAL TEMPERATURE

It has been reported[2] that, in the alloy corresponding to the composition Ni_3Fe, the critical temperature for the formation of long-range order on cooling is 490°C. Small amounts of manganese will result in increase of this temperature. For instance, in an alloy containing 1 percent manganese, this temperature is raised to 500°C. Recently Iida[4] has completed a series of very instructive experiments on the formation of short-range and long-range order. He measured the time rate of energy decrease while the specimen is kept at constant temperature, after cooling from a high temperature as rapidly as possible. This was carried out by measuring the necessary temperature difference between specimen and the surrounding container in order to maintain the temperature of the specimen constant. Figures 2 and 3 illustrate how the rate of energy decrease $-dU/dt$ varies with time of annealing, when specimen is kept at 510°C and 500°C, respectively, after having been cooled rapidly from 600°C. At 510°C,

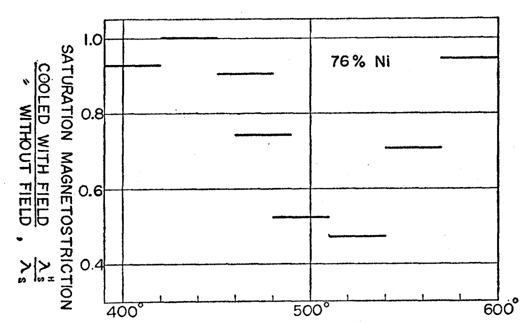

FIG. 1. Dependence of saturation magnetostriction of 76 Permalloy on the temperature at which an external magnetic field of 50 oersteds is applied during cooling. Magnetic field is kept on for a temperature interval of 30°C. Cooling velocity is about 1°C/min.

[4] S. Iida, J. Phys. Soc. Japan **7**, 373 (1952).

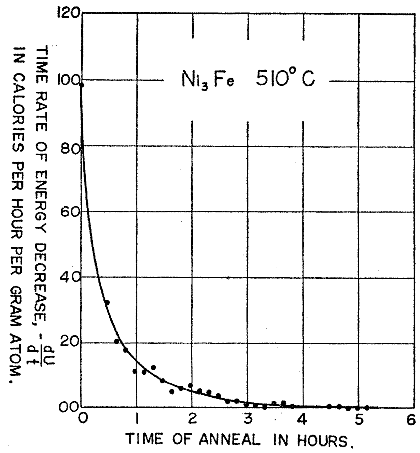

FIG. 2. Dependence of time rate of energy decrease $-(dU/dT)$ on the time of annealing, when 76 Permalloy is kept at 510°C, after cooling rapidly from high temperature.

$-dU/dt$ decreases monotonically and tends to zero after 3 or 4 hours. At 500°C quite some change takes place at the beginning, but after several hours it begins to increase, attains its maximum at 20 to 30 hours and again decreases to zero after about 50 hours.

These results differ somewhat in numerical details from those previously obtained[2] by measuring electric resistance changes; but this can probably be attributed to the difference in manganese content. It can be concluded that the first step of the energy decrease corresponds to the formation of short-range order and the second step to that of so-called long-range one. Special emphasis must be laid on the essential difference between the curve form of the first and second energy decrease. The first simple decrease can be associated with the formation of short-range order, or the increase of the number of pairs of near neighbors with unlike atoms, while the second decrease shows the characteristics of a cooperative rate process or the formation of long-range order. Figure 4 illustrates how the energy decreases with time of annealing, when the specimen is maintained at the respective temperature after cooling from 600°C.

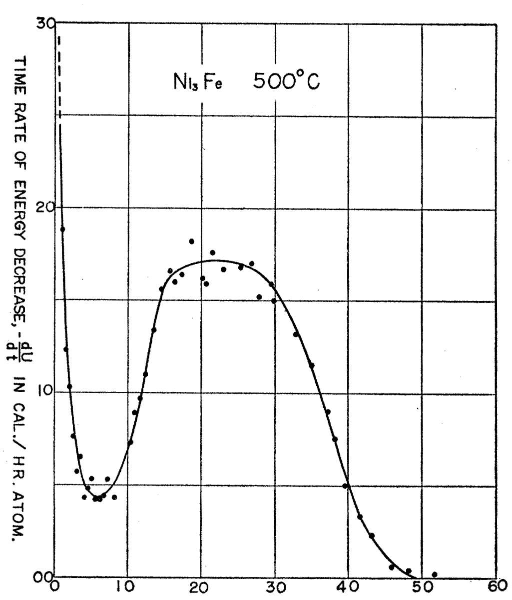

FIG. 3. Dependence of time rate of energy decrease $-(dU/dt)$ on the time of annealing, when 76 Permalloy is kept at 500°C, after cooled rapidly from high temperature.

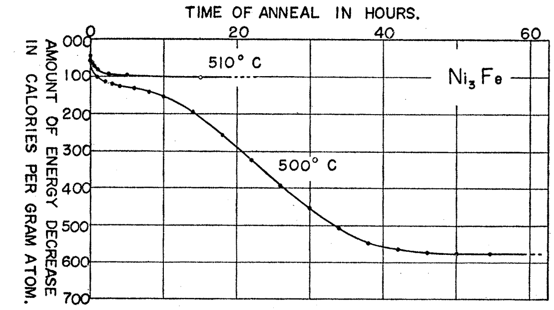

FIG. 4. Dependence of energy decrease on the annealing time. Annealing temperatures are 510°C and 500°C.

III. CHANGE OF MAGNETIC CHARACTERISTICS WITH TIME OF ANNEALING

As to the dependence of magnetic characteristics of 76 Permalloy on the time of annealing, Nagashima[5] has carried out a comprehensive study. The specimens of 0.3 mm in diameter and 140 cm in length were quenched from 600°C, and then were annealed at 490°C for a few hours and quenched to room temperature again. Figure 5 illustrates how the hysteresis loop of 76 Permalloy, thus heat treated, varies in its form with time of annealing. This shows that hysteresis loops of constricted form are absent in both the quenched and fully annealed states. In contrast to this, hysteresis loops of the specimen in which the composition deviates from Ni_3Fe (for instance, 81.5 Permalloy) show well-developed

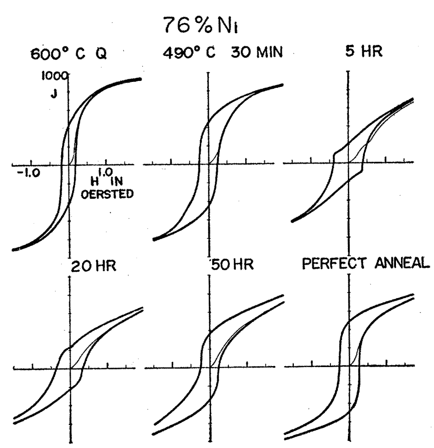

FIG. 5. Variation of the form of hysteresis loop for 76 Permalloy with time of annealing. (Annealing temperature=490°C.)

[5] T. Nagashima, J. Appl. Phys. Japan **19**, 53 (1950).

constricted form even in the fully annealed state, as will be seen in Fig. 6. Such a difference is to be attributed to the lack of perfectness of atomic ordering even in the fully annealed state. Figure 7 shows the dependence of coercive force and remanence of 76 Permalloy on the time of annealing.

If we examine the corresponding change shown in Fig. 2 and Fig. 7, it can be concluded that the properties of Perminvar or of constricted hysteresis loops are mainly related to the formation of short-range order, and further that the uniform formation of long-range order has a tendency to annihilate them.

IV. TORQUE CURVES TAKEN ON A DISK OF 76 PERMALLOY CRYSTAL

In order to obtain a better understanding of the mechanism of magnetic field cooling, Chikazumi[6] prepared a disk of 76 Permalloy crystal, which was cut parallel to the (110) plane, and measured torque curves

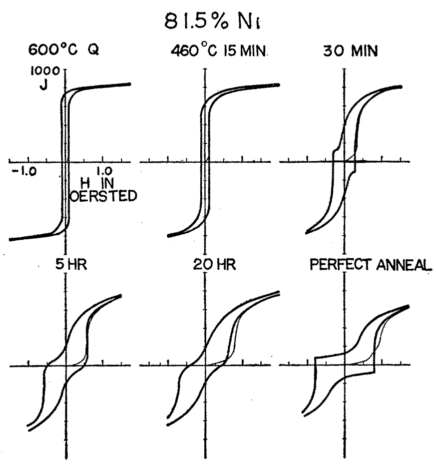

FIG. 6. Variation of the form of hysteresis loop for 81.5 Permalloy with time of annealing. (Annealing temperature = 490°C.)

for this crystal disk, after it was cooled in a magnetic field, applied parallel to [1$\bar{1}$1], [1$\bar{1}$0], and [001] directions, respectively. Since the torque is the negative rate of change of energy with crystal orientation, the energy necessary to bring the crystal to saturation can be obtained by integrating the torque curve with angle. Figure 8 illustrates the dependence of this energy, thus obtained, on the crystal orientation in the (110) plane. The energy E_t can be separated easily into two parts, E_c and E_u. E_c is the ordinary anisotropy energy with cubic symmetry, and E_u is an anisotropy energy with uniaxial symmetry. They can be expressed in most simplified form:

$$E_t = E_c + E_u = K_c(\alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2 + \alpha_1^2\alpha_2^2) + K_u(\alpha_1\beta_1 + \alpha_2\beta_2 + \alpha_3\beta_3)^2,$$

where α_i, β_i represent the direction cosines of spin and field, respectively, referred to the cubic axes.

[6] S. Chikazumi (unpublished data).

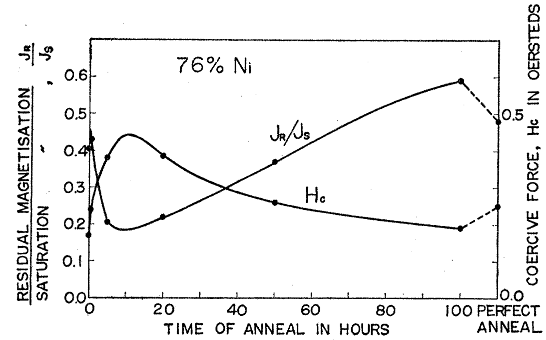

FIG. 7. Dependence of coercive force H_c and remanence J_r of 76 Permalloy on the time of annealing. (Annealing temperature = 490°C.)

As can be seen in Fig. 8, both K_c and K_u are negative in this experiment. The absolute magnitude of K_c has

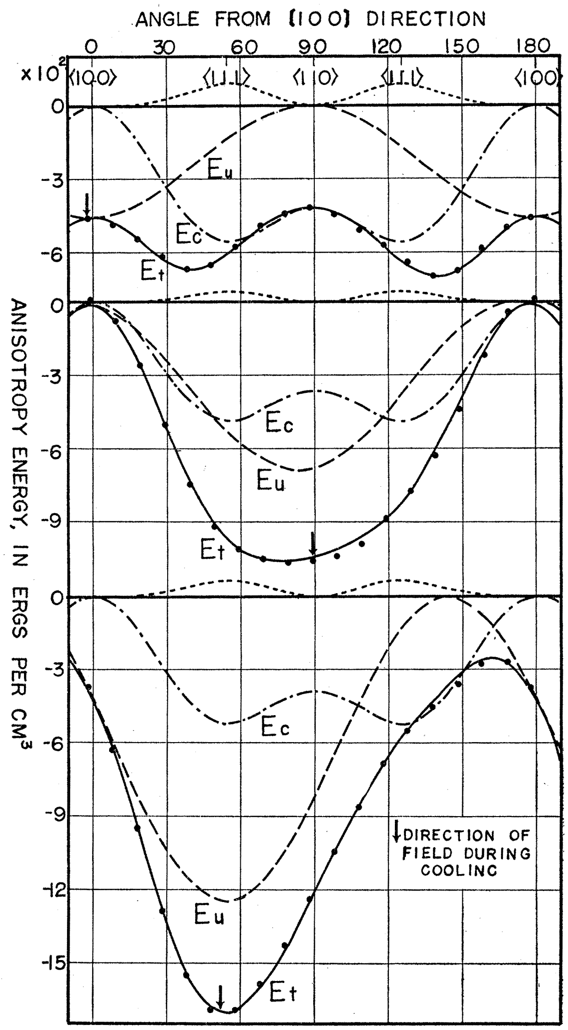

FIG. 8. Dependence of anisotropy energy of a 76 Permalloy crystal on the orientation in the (110) plane. Arrows indicate the orientations in which an external magnetic field is applied during cooling. (Cooling velocity = 22°C/min.) Total energy E_t is analyzed into E_c and E_u; the former has cubic symmetry and the latter uniaxial symmetry.

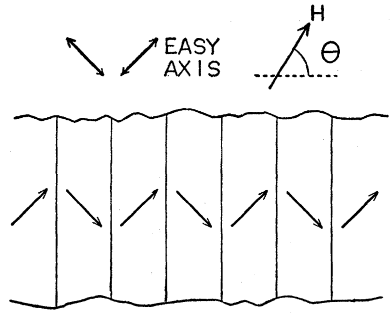

FIG. 9. A linear assemblage of magnetic domains. The uniaxial axes of neighboring domains intersect perpendicularly with each other, and their boundary bisects the angle between them.

been known to increase with the atomic ordering, whereas that of K_u is expected to increase rapidly with ordering, attain its maximum, and gradually decrease in accordance with the uniform formation of perfect order. The magnitude of K_u depends not only on atomic ordering, but also on the direction, to which the magnetic field is applied during the course of cooling. As shown in Fig. 8, the absolute magnitude of K_u is a maximum when the magnetic field is applied to [111] direction, and is a minimum when the field is parallel to the [001] direction. Hence, the dependence of K_u on the orientation of applied field during cooling can be expressed by

$$K_u = K_{u1} + K_{u2}(\beta_2^2\beta_3^2 + \beta_3^2\beta_1^2 + \beta_1^2\beta_2^2).$$

V. POSSIBLE ORIGIN OF UNIAXIAL ANISOTROPY

In order to answer the question as to the origin of the uniaxial anisotropy, an increase in saturation magnetization caused by atomic ordering will be suggested as a possible consideration. This increase is about 3 to 4 percent in 76 Permalloy, though it depends strongly upon the manganese content. If we assume nonuniform, locally developed atomic ordering, magnetostatic energy caused by free poles within the specimen can be expressed as follows:

$$E_m = \tfrac{1}{2}N(\Delta J_s)^2 V(1-V),$$

where N is the demagnetization factor of the ordered phase, ΔJ_s is the increased amount in saturation magnetization caused by atomic ordering, and V is the volume proportion of ordered phase. If atomic ordering develops in needle shape, N varies from zero to 2π, according to the orientation of magnetization.

If the values $\Delta J_s = 40$, $V = 0.5$ are substituted in this expression, we obtain an anisotropy energy of about 1000 erg/cc, which is in accord with the experimental data hitherto observed. The suggested mechanism of the origin of uniaxial anisotropy would become more comprehensible if attention is drawn to the fact that the Curie point is higher in the ordered phase than in disordered one.

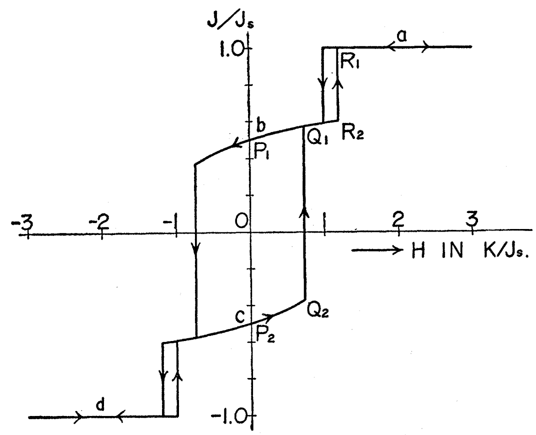

FIG. 11. Calculated hysteresis loop, when field is applied parallel to one of the uniaxial axes.

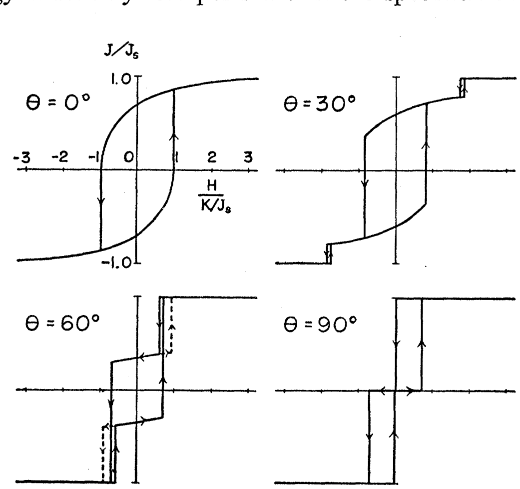

FIG. 10. Dependence of the form of hysteresis loop on the orientation of applied field.

VI. DEDUCTION OF CONSTRICTED HYSTERESIS LOOP FROM UNIAXIAL ANISOTROPY

Hitherto the constricted hysteresis loop and the properties of Perminvar were believed to be the consequence of strong interaction between two constituent phases, which show quite different magnetic characteristics with each other. Quite recently Iida[7] has succeeded in deducing mathematically the Perminvar properties using the simple model shown in Fig. 9. This model is, as shown in the figure, a linear assemblage of magnetic domains with uniaxial anisotropy only. The orientations of adjacent domains are at right angle to each other, and their boundary bisects the angle between them. (In the actual case the angle subtended between the adjacent domains is 70° or 110°.)

The action of the external field will be calculated in the ordinary way by minimizing the total energy, composed of the crystal anisotropy energy and the energy of interaction of the magnetization and the external field. It must be borne in mind, that in every case the condi-

[7] S. Iida (unpublished data).

tion of the continuity of normal component of magnetization at the boundary should be satisfied. Mathematical deduction of the hysteresis loop and of the initial curve can be carried out in quite an elementary way. Figure 10 shows the dependence of the shape of the hysteresis loop thus calculated on the orientation of applied field, where θ denotes the angle between the field and the wall normal. Explanation will be given here only in one typical case, where the magnetic field is applied parallel to one of the uniaxial axes ($\theta=\frac{1}{4}\pi$). As shown in Fig. 11, the descending branch of the loop is divided into four sections *a*, *b*, *c*, and *d*, and three abrupt decreases of magnetization (*ab*), (*bc*), and (*cd*) exist between them. The change of domains in each

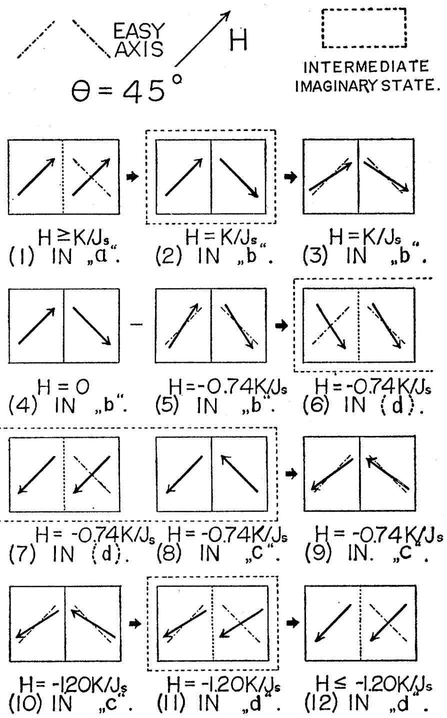

FIG. 12. Schematic diagram of the transitions of neighboring domains, when magnetization varies along the descending hysteresis curve.

stage is illustrated in Fig. 12. Following the decreasing applied field, an abrupt change (*ab*) will be initiated, if the energy of configuration (2) becomes smaller than that of configuration (1), and this will be carried out by the creation of a new boundary. Since the energy of configuration (3) is smaller than that of configuration (2), the transition (2) and (3) will be completed simultaneously.

At the abrupt change (*bc*), the transition is (5) to

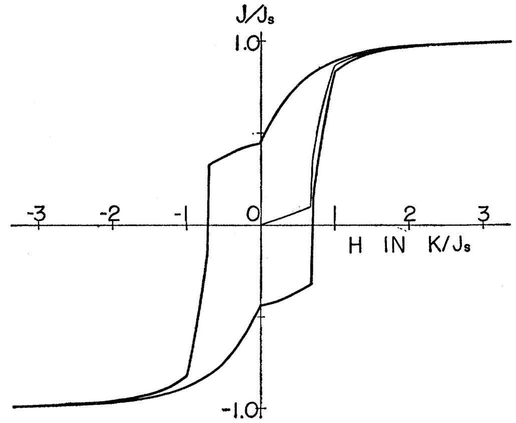

FIG. 13. Calculated hysteresis loop of polycrystalline material.

(9), and this will be analyzed into four intermediate transitions, of which (5) to (6) means annihilation of boundary and (7) to (8) means its creation. In the actual case of an abrupt change, creation or annihilation of a domain boundary would be accompanied with some difficulties associated with nucleation, but we have neglected this effect in this calculation. The field intensity, at which the abrupt change (*bc*) occurs, can be interpreted as the coercive force H_c and this amounts in this case to 0.73 K_u/J_s. If we substitute $K_u=1000$, H_c becomes 0.73 oersteds.

The initial curve can be easily obtained, by superposing the two branches of hysteresis loop, $P_1Q_1R_2R_1$ and $P_2Q_2Q_1R_2R_1$. Initial permeability μ_0 is expressed in this case:

$$\mu_0=1+\tfrac{1}{2}\pi(J_s^2/K_u),$$

and this furnishes us: $\mu_0=1500$, with $K_u=1000$, $J_s=1000$. Both H_c and μ_0 accord well with the experimental values.

It will be emphasized here, that, in some orientations of applied field, the initial curve extends out of the hysteresis loop. Figure 13 illustrates the initial and hysteresis curve of polycrystalline material, deduced by taking the average value, when the orientation of applied field varies from 0° to 90°. This agrees qualitatively with the experimental one shown in Fig. 5. Quantitatively some discrepancies exist between theories and experiment. For instance, the remanence J_r is 0.45 J_s, while the experimental value is much smaller. This discrepancy is probably due to the simplicity of the adopted model. In a more complete theory, a three-dimensional assemblage of domains, which, in the absence of an external field, would permit flux closure, should be adopted.

DISCUSSION

J. E. GOLDMAN, *Carnegie Institute of Technology, Pittsburgh, Pennsylvania:* There are two alternative possibilities in approaching an explanation of the effect of the magnetic anneal on the properties of Permalloy in certain specific composition ranges. One of these is to suppose that the certain characteristic properties of