TEMPERATURE HYSTERESIS OF DOMAIN STRUCTURE IN SILICON IRON CRYSTALS

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The domain structure of silicon iron crystals before and after heating from room temperature up to 550°C was studied by the powder pattern technique. Irreversible changes of the domain structure were detected following a temperature cycle. The size, shape and number of closure domains change and the boundaries between basic domains are displaced; temperature hysteresis of the domain structure is observed. This behavior is explained using the domain theory of ferromagnetic structure.

I. INTRODUCTION

THE domain structure of a ferromagnetic substance is known to depend on its basic properties (magnetic saturation, the anisotropy and magnetostriction constants), its shape and size, and different kinds of lattice defects (residual strains, impurities, discontinuities, etc.). Since the basic properties are temperature-dependent we may expect the domain structure to vary with temperature. We may also expect that the magnetic structure will not return to its original form following a temperature cycle, since both reversible and irreversible changes may accompany temperature variations. Temperature hysteresis of the domain structure should therefore be observed.

The present work was performed to detect the temperature hysteresis of domain structure in silicon iron crystals and to establish the most general laws of this effect. We are not aware of any previous special investigation of this type.

II. DESCRIPTION OF SAMPLES AND EXPERIMENTAL TECHNIQUE

The magnetic structure was observed by means of the powder-pattern technique.[1] Both single crystals and polycrystalline samples of silicon iron (3.5% Si) 15 mm in diameter and 0.3—0.7 mm thick were used. The investigated surface of the single crystals was approximately parallel to the (011) plane. In the case of polycrystalline samples, which consisted of large grains 0.5—3.0 mm in diameter, the domain structure was observed on single grains having surfaces close to (001) or (011). The samples were etched from sheet silicon iron, and after mechanical grinding and polishing were vacuum annealed at 1250°C. The magnetic structure of some samples was studied immediately following this high-temperature annealing, while other samples were subjected to supplementary electrolytic polishing. For the purpose of studying the effect of a temperature cycle on the form of the magnetic structure, the samples were heated from room temperature to different temperatures up to 550°C and were then cooled to the initial temperature. The samples were heated in a special device placed within a vacuum chamber (Fig. 1) that was fastened to the microscope stage. A copper core 2 bearing a sample 3 on its upper end was inserted into a double-wound electric furnace 1. A copper cover 4 was provided for temperature equalization. A thermocouple 5 was fitted into the copper core. The furnace was fastened by means of the supports 6 to a heavy water-cooled iron plate 7. For the purpose of preventing oxidation during heating the furnace together with the sample was placed inside a vacuum chamber consisting of a heavy iron dome 8 and base 7, which at the same time provided magnetic screening for the sample. Thus the samples were both heated and cooled in a vacuum which was free of external magnetic fields.
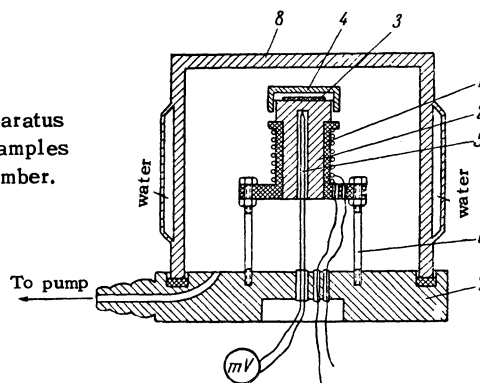

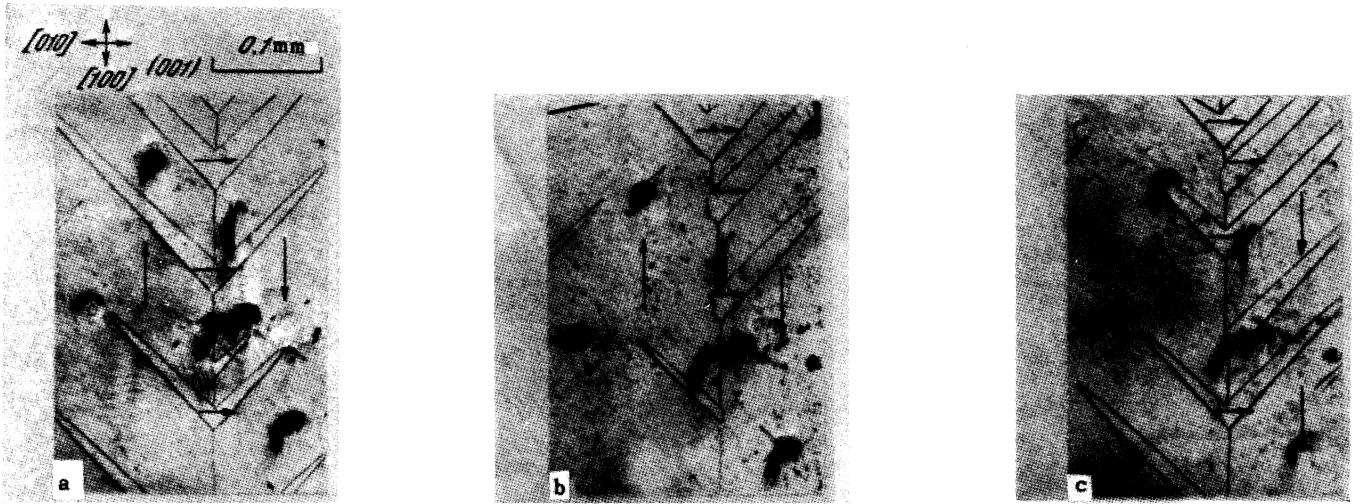FIG. 1. Apparatus for heating of samples and vacuum chamber.

FIG. 2. Powder pattern on a crystal surface approximately parallel to the (001) plane (type A structure); a — after demagnetization, b — after heating to 400°C, c — after re-demagnetization.

Prior to observation of the domain structure each sample was demagnetized in a solenoid by means of a field which varied smoothly from 600 oersteds to zero. The sample was then placed upon the heating core 2. A magnetic colloidal suspension was applied to the surface of the sample; the powder pattern was then observed and photographed. After the suspension had been carefully wiped off the sample was covered with the copper cap 4, and the dome 8 was fastened to the plate 7. After a vacuum had been established the sample was gradually heated to the required temperature, at which it was maintained during 30 min. When the sample and furnace were subsequently cooled to room temperature the magnetic suspension was again applied to the surface of the sample and the powder pattern was photographed in exactly the same area as before heating.

The sample was thereupon demagnetized by a variable field and the powder pattern on its surface was again photographed. In the case of monocrystalline disks the powder pattern was photographed along a diameter perpendicular to the direction of easy magnetization lying in the plane of the sample. Repeated observations of the powder patterns following successive alternations of careful wiping and applying of the suspension showed that this procedure does not affect the magnetic structure.

For the purpose of detecting temperature hysteresis of the domain structure we compared the powder-pattern photographs of the same area a) after the first demagnetization, b) after heating and cooling and c) after the second demagnetization process.

III. EXPERIMENTAL RESULTS

Heating from 20 to 200°C followed by cooling to 20°C produced no essential changes in the domain structure. With heating to higher temperatures irreversible movements of boundaries between the basic domains became clearly distinguishable, accompanied by readjustment of the surface domains of closure. Reorganization of the domain structure was studied carefully following heating up to 400— 550°C; the existence of temperature hysteresis was definitely established. Although subsequent demagnetization in a variable magnetic field restores the original type of domain structure, the boundaries of the basic domains and the domains of closure do not occupy exactly the same positions that are observed following the first demagnetization process prior to heating. This irreproducibility follows consistently; after each re-demagnetization at room temperature the magnetic structure is, as a rule, not entirely reproduced in all details.

The photographs of powder patterns in Figs. 2, 3, 5, and 6 show how the domain structure is modified in some of the simplest cases following a temperature cycle. Figure 2 shows powder patterns on a crystal having its surface approximately parallel to the (001) plane. In the demagnetized state (Fig. 2a) two basic domains are visible, separated by a 180° boundary, and the surface closure domains exhibit a "Christmas tree" structure.[1] The arrows in the photographs indicate the direction of magnetization I_s in the domains. Following heating to 400°C and subsequent cooling to the original room temperature the 180° boundary is observed to move irreversibly to the right (Fig.

FIG. 3. Powder pattern on a crystal surface approximately parallel to the (001) plane (type B structure): a — after demagnetization, b — after heating to 400°C, c — after re-demagnetization.

2b) and the number of domains of closure (tree branches) is reduced. Re-demagnetization (Fig. 2c) returns the 180° boundary to its original position, while the number of closure domains increases and their original appearance is approximated.
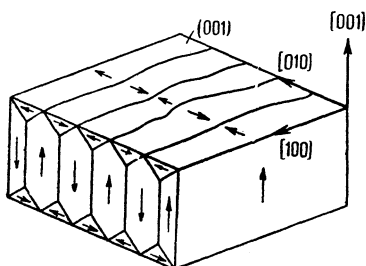

FIG. 4. Diagram of type B structure.

The powder patterns in Fig. 3 also belong to a crystal with its surface approximately parallel to (001), but differ from the preceding case by exhibiting type B structure (Fig. 3a) in the initial demagnetized state.[2] This means that the basic domains are located inside the crystal, while the observed surface reveals the bases of triangular-prism closure domains separated by zigzag boundaries. Type B structure is illustrated schematically in Fig. 4. In addition, smaller "comb-like" domains of closure are observed within the prismatic domains of closure.[2] Heating to 450° followed by cooling to room temperature considerably modifies the closure domain structure (Fig. 3b); the "combs" increase in size but decrease in
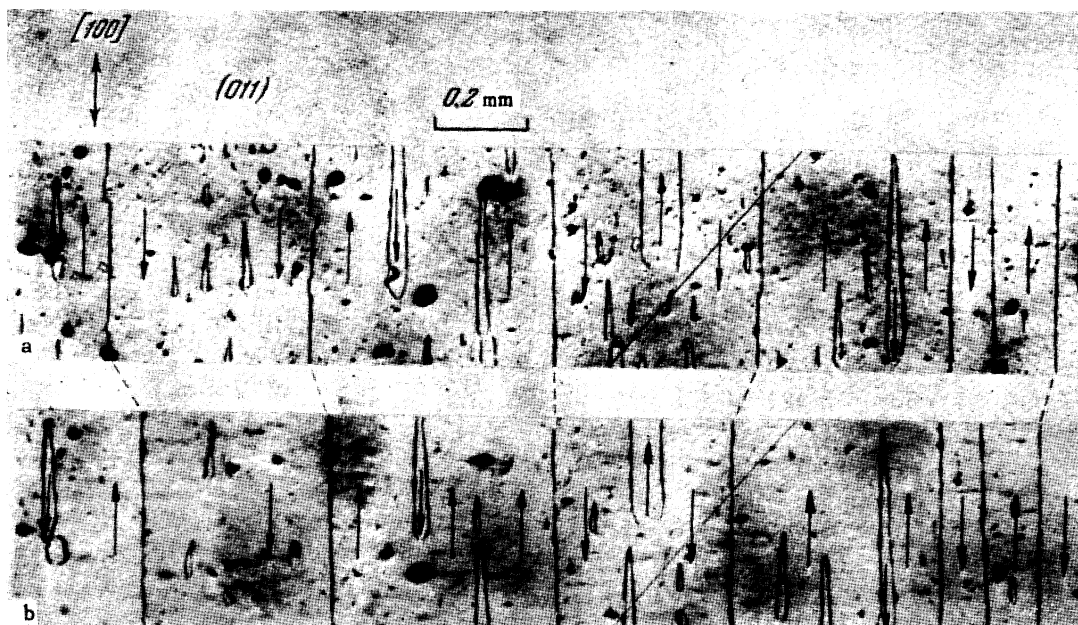

FIG. 5. Powder pattern on a crystal surface approximately parallel to the (011) plane: a — after demagnetization, b — after heating to 550°C.
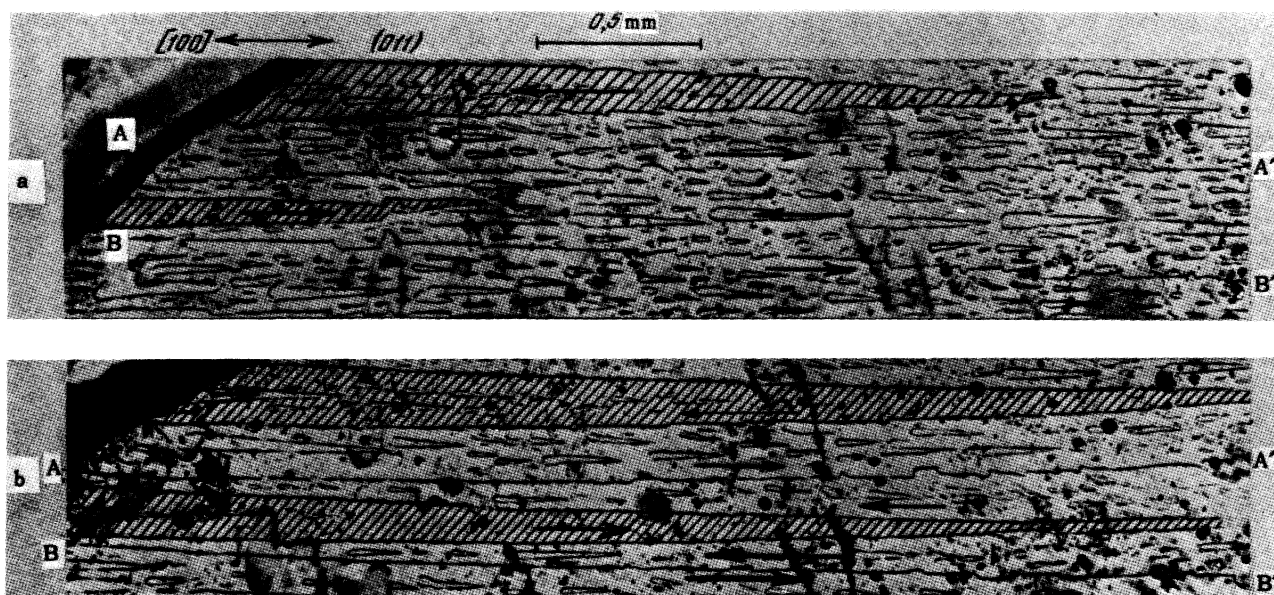
FIG. 6. Powder pattern on a crystal surface approximately parallel to the (011) plane: a — after demagnetization, b — after heating to 550°C.

number. Following the re-demagnetization, however, the original appearance of the domain structure is restored (Fig. 3c).

Figure 5 shows photographs of powder patterns on a single-crystal surface approximately parallel to (011). The plane of the sample contains one direction of easy magnetization; in the initial demagnetized state (Fig. 5a) the crystal is divided into oppositely magnetized domains separated by 180° boundaries. The surface also reveals a small number of closure domains shaped like drops of a liquid.[2] Heating to 550°C followed by cooling to room temperature induces an irreversible change of the magnetic structure (Fig. 5b) in which the boundaries of the basic domains are shifted while the drop-shaped regions change in size and decrease in number. The dashed lines connecting boundaries of the basic domains before and after heating show the extent to which the boundaries are shifted as a result of the temperature cycle.

Figure 6 also shows the powder pattern on the edge of a monocrystalline disk the surface of which is also approximately parallel to the (011) plane. Here the easy direction of magnetization closest to the crystal surface forms a larger angle with the latter than in the preceding case. In the demagnetized state (Fig. 6a) the sample is also divided into plane-parallel domains separated by 180°C boundaries that exhibit jogs (AA' denotes the upper boundary and BB' denotes the lower boundary). The surface reveals a large number of drop-shaped closure domains, as well as relatively large (hatched) dagger-shaped regions starting at the edge of the crystal. Figure 6b shows the domain structure after heating to 550°C.

A comparison of Figs. 6a and 6b shows that the temperature cycle does not induce an appreciable shift of the boundaries between the basic domains. These boundaries become more distinct and straighter. At the same time the closure domains are considerably reorganized; the drop-shaped domains decrease in number but some of them increase in size. The greatest change is exhibited by the dagger-shaped domains of closure located at the edge of the crystal. These domains grow much larger as a result of the temperature cycle and new dagger-shaped domains appear. Within the expanded upper dagger-shaped domain in Fig. 6b a new but smaller domain of the same type appears with antiparallel magnetization.

The experimental results thus show that a temperature cycle between 20 and 400—500°C induces considerable irreversible change of magnetic structure in monocrystalline and polycrystalline silicon iron. These changes can be summarized as follows: The surface domains of closure change in size and shape, and some of them disappear; the dagger-shaped domains of closure at the edge of the crystal increase in size; the boundaries between the basic regions are shifted.

IV. ANALYSIS OF RESULTS

We shall now attempt to account for the observed irreversible changes of the domain structure. We shall first determine what changes of magnetic structure could be expected when the temperature of silicon iron crystals is elevated. At room tem-
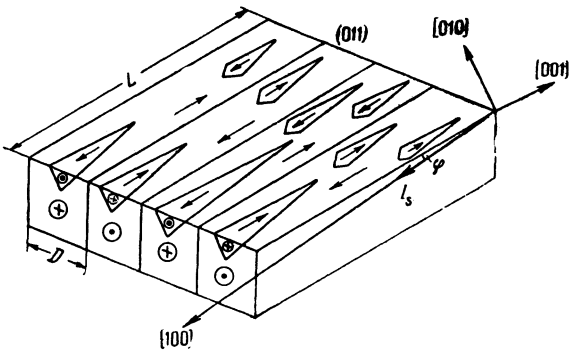
FIG. 7. Diagram illustrating type A structure. φ — angle between the [100] direction and the crystal surface.

perature a crystal having its surface approximately parallel to (011) is usually divided into basic domains separated by 180° boundaries; magnetization of these regions is oriented along the [100] tetragonal axis closest to the surface of the sample and forming an angle φ with the latter. This type of structure is represented schematically in Fig. 7. On the surface and edges of this crystal stray magnetic fields appear, whose energy is reduced through the formation of closure domains (drop-shaped domains on the surface and dagger-shaped domains on the edge). Energy is expended for the formation of the closure domains because boundaries are developed between these domains and the basic domains within which the former appear. Thus the sizes and shapes of closure domains are determined by the relationship between the stray-field energy and the wall energy. With weakening of the stray fields these domains should be reduced in size, but their size should increase when the wall energy is reduced.

The energy of the stray fields on the crystal surface that is approximately parallel to the (011) plane is, according to Kittel,[3]

$$F_1 = 0.85\, I_s^2 D \sin^2 \varphi / (1 + \mu^*),\qquad(1)$$

where D is the width of the basic domains (Fig. 7) and $\mu^* = 1 + 2\pi I_s^2/K$. Through study of the domain structure by means of powder patterns it has been found that in silicon iron crystals having surfaces approximately parallel to (011), the magnetization I_s is parallel to the [001] axis in both the basic and closure domains. D can therefore be calculated by means of the formula derived for a uniaxial magnetic crystal:[3]

$$D = (\gamma L / 1.7 I_s^2)^{1/2},\qquad(2)$$

where L is the length of the basic domains. We thus obtain

$$F_1 = \frac{0.33 I_s \sin^2 \varphi \sqrt{L\gamma}}{1 + \pi I_s^2 / K}.\qquad(3)$$

$\gamma \sim \sqrt{aK + b\lambda_s^2 E}$ is the density of the wall energy;[4] here a and b are constants of the order of unity, λ_s is the saturation value of the magnetostriction and E is the elastic modulus. In our samples of silicon iron $K \gg \lambda_s^2 E$ (since $K \approx 3 \times 10^5$ erg/cm³, (reference 5), $\lambda_{[100]} \approx 2 \times 10^{-5}$ (reference 6) and $E \approx 2 \times 10^{12}$ dyne/cm²). We shall therefore hereinafter assume $\gamma \sim \sqrt{K}$. Equation (3) shows that F_1 depends on a number of temperature-sensitive ferromagnetic parameters: I_s, K, γ.

Our samples were heated from room temperature up to 400—550°C. By this process the saturation magnetization of silicon iron is not greatly (10—20%) reduced, but the anisotropy constant K is reduced by a factor of 3—6.[5] For silicon iron $I_s = 1600$; therefore $\pi I_s^2/K \gg 1$. It follows that $F_1 \sim K^{5/4}/I_s$. In view of the fact that with increasing temperature the decrease of I_s is insignificant compared with that of K, we take

$$F_1(T) \sim K^{5/4}(T).\qquad(4)$$

The temperature dependence of the wall energy density is

$$\gamma(T) \sim K^{1/2}(T).\qquad(5)$$

It follows from (4) and (5) that since F_1 decreases more strongly than γ, the surface drop-shaped closure domains must diminish in size, so that some of them may even disappear.

Both reversible and irreversible changes of the domain structure may occur when the crystals are again brought down to room temperature. The irreversible changes should partially conserve all characteristics of the domain structure reorganization which result from the higher temperature. In the present specific case we may expect the irreversibility to be manifested by a different size of the closure domains compared with the initial state prior to heating. In addition, some of the disappearing closure domains may not reappear because nuclei of new regions are formed with difficulty. Figures 5 and 6 contain the experimental evidence for these conclusions.

We shall now consider how a temperature rise should affect the appearance of the dagger-shaped closure domains at the crystal edge (Figs. 6 and 7). The stray-field energy on the lateral crystal surface is

$$F_2 = 0.85\, I_s^2 D \cos^2 \varphi.\qquad(6)$$

Equation (6) does not contain μ^* because this quantity may be neglected for large angles between I_s and the crystal surface in question.[1] Substituting D from (2) into (6) and considering that in our case I_s is only slightly temperature-dependent, we have

$$F_2(T) \sim K^{1/4}(T). \tag{7}$$

A comparison of (7) and (5) indicates that with rising temperature F_2 will decrease more slowly than the wall energy density. As a result the dagger-shaped closure domains on the edge should grow; they may also increase in number. Assuming that with a reduction to room temperature the structure may not return to its original condition (before heating), we may expect a growth of the dagger-shaped regions as a result of a temperature cycle. This is confirmed experimentally by the powder patterns given in Fig. 6, where it is shown how the temperature cycle brings about a considerable growth and multiplication of the dagger-shaped domains.

We may similarly consider how a higher temperature affects the structure of closure domains on a crystal surface approximately parallel to the (001) plane. When the surface is accurately parallel to (001) the magnetic domain structure predicted by Landau and Lifshitz appears.[7] This structure is shown schematically on the end surface in Fig. 4, where the basic domains and the triangular closure domains on the edges are represented. When the crystal surface is slightly inclined to (001) stray fields appear on the surface, and the energy of these fields may be reduced through the formation of surface closure domains (trees, tree trunks, etc.). For this structure, according to Kittel,[3] the width of the basic domains is calculated taking the magnetoelastic energy into account:

$$D = (4\gamma L / \lambda_{[100]}^2 c_{11})^{1/2}, \tag{8}$$

where c_{11} is the elastic modulus. As the temperature of a silicon iron crystal increases to 500°C, $\lambda_{[100]}$ can only increase[6] while c_{11} decreases slightly. D will therefore diminish with rising temperature because of the growth of $\lambda_{[100]}$ as well as the reduction of K. From (1) and (8) we obtain $F_1(T) \sim K^{5/4}(T)$. We should therefore observe a reduction of closure domain size when a crystal having its surface approximately parallel to (001) is heated. This reconstruction of the closure domains may also follow a temperature cycle, as can be seen from the powder patterns in Fig. 2.

A temperature rise can also affect the appearance of the basic domains. The boundaries between these regions tend to assume the positions for minimum wall energy. This remains true if the total energy of the crystal is not enhanced through increases in other forms of energy. With rising temperature the regions of minimum γ are rearranged; the boundaries may thus be shifted. The boundaries of closure domains may be shifted for the same reason.

It follows from (2) and (8) that a temperature rise will also change the equilibrium width D of the basic domains. For example, in the case of the domain structure represented in Fig. 7 we may assume $D \sim \gamma^{1/2} \sim K^{1/4}$ according to (2), since in our experiments I_S changes very little with temperature. Higher temperatures may therefore break down the basic domains; in the present instance this may be brought about through growth of the dagger-shaped closure domains and through their transformation into basic domains.*

It follows from our analysis that the magnetic structure of silicon iron crystals should change when their temperature is elevated.† It may be expected that when the original temperature of these crystals is restored some of the changes are conserved. The existing theory is unable to predict the extent of this irreversibility of the magnetic structure. Only experiments such as those described in the present paper can supply pertinent information. We have shown that a temperature cycle is accompanied by a boundary displacement of the basic domains.

It is reasonable to expect that the domain structure of a heated sample will be metastable after being restored to its original temperature. Therefore if a heated sample is demagnetized by a variable field the magnetic structure should on the whole return to the original form exhibited by the demagnetized sample before heating. This conclusion is confirmed by the photographs in Figs. 2 and 3.

The temperature hysteresis of domain structure which we have described in the case of silicon iron crystals should also occur in all other ferromagnetic materials possessing a multidomain magnetic structure.

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*Strictly speaking, we cannot consider changes of the closure domains and basic domains independently, since all the domains of a crystal are interrelated. In polycrystalline samples this interrelation also exists between domains located in different grains, especially between those which are in contact. Our treatment is adequate, however, for the discovery of qualitative laws.

†Kirenskiĭ and Degtyarev[8] used the Kerr effect to observe the magnetic structure on the oxidized (011) plane of a silicon iron crystal which was heated from 20 to 700° C. No changes of the magnetic structure were detected. Since these observations were made under very low magnification, small heat-induced changes of the magnetic structure may have been overlooked.

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Translated by I. Emin
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