tained by Meissner,[26] that the melting point of crystals decreases with their thickness once they are thinner than $\sim 0.8\mu$. Finally, it has been observed,[27] that the magnetic susceptibility of hematite (a mineralogical species of α-FeOOH) decreases by more than one order of magnitude if the crystal diameter is reduced from 500μ to less than 10μ. A decrease in the mean susceptibility would certainly affect the magnetic anisotropy also. However, it is wise not to insist too much on this last argument, since there is a considerable controversy going on as to whether such variations in susceptibility with particle size are actually intrinsic effects or effects of adsorption and surface contaminations.

ACKNOWLEDGMENT

The experiments described in this paper and in others to follow have been carried out in the Laboratoire des recherches physiques à la Sorbonne, Paris, France. One of us (W. H.) recalls in gratitude the generosity of Professor Aimé Cotton, Head of the Department and President of the French Academy of Science, who made it possible to organize and to carry out a research project on colloid physical phenomena. The Faculté des Sciences of the Sorbonne are equally thanked for allocation of the annual receipts from the Vautier fund.

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[26] G. Tamann, Zeits. f. anorg. u. allgem. Chemie **110**, 166 (1920); F. Meissner; *ibid.*, **110**, 169 (1920). These authors explain their observation as an effect of surface energy. The concept of differences in crystal spacing is more recent.

[27] R. Chevalier and S. Mathieu, Comptes rendus **204**, 854 (1937).

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The Magnetic Structure of Iron Crystals

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A number of powder patterns found on demagnetized strain-free crystals of silicon-iron of known orientation are illustrated and discussed. The specimens were cut from strips of transformer steel in which fairly large crystals had been grown by the strain-anneal method. Smooth undamaged surfaces were prepared by electrolytic polishing. A magnetic field of only 10 oersteds applied normal to the specimens was sufficient to produce patterns; the intensity of the stray fields must thus be of this order of magnitude. On account of the large demagnetization factor (4π) of the specimens, the patterns are characteristic of the demagnetized state. All evidence points to a layer magnetization with alternate layers oppositely magnetized. The thickness of the layers in different specimens ranged from 10 to 50 microns. Superposed on this primary layer magnetization was a finer secondary structure, presumably localized near the crystal surface, and attributable to dendrite-like regions of reversed magnetization which reduce the magnetic energy associated with the surface stray fields.

PREVIOUS investigations of the ferromagnetic powder patterns of iron or silicon-iron crystals have failed to give a clear notion of the magnetic domain structure of these crystals. L. W. McKeehan and the writer[1,2] once studied in some detail the maze-like patterns found on mechanically polished silicon-iron crystals magnetized normal to their surface, but succeeding experiments[3] showed that the metallographic polishing used to prepare smooth surfaces played an important part in determining the scale and the geometrical form of the patterns. It is unlikely that these patterns furnish any clue to the primary domain structure of undamaged crystals. A number of patterns on iron specimens magnetized parallel to their surface have been

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[1] L. W. McKeehan and W. C. Elmore, Phys. Rev. **46**, 226 (1934).

[2] W. C. Elmore and L. W. McKeehan, Trans. A. I. M. E. *120*, 236 (1936).

[3] K. J. Sixtus, Phys. Rev. **51**, 870 (1937); W. C. Elmore, Phys. Rev. **51**, 982 (1937).

published by various authors,[4] but no one has yet attempted a detailed interpretation of the patterns in terms of local domain magnetization. To obtain more reliable information concerning the magnetic structure of demagnetized crystals it appears necessary to examine patterns on annealed specimens which have smooth surfaces free from polishing damage. Since suitable surfaces can now be prepared by the method of electrolytic polishing,[5] a further study of the colloid patterns on silicon-iron crystals has been undertaken.

APPARATUS, SPECIMENS, AND FERROMAGNETIC COLLOID

The microscope-magnet assembly used for producing and observing patterns is illustrated in Fig. 1. A flat specimen placed upon the large soft iron pole piece can be uniformly magnetized normal to its surface. The magnet carrying the specimen rests on a specially built microscope stage which can be moved in a horizontal plane by a pantograph arm. The arm magnifies the motion of the stage about thirty times and with it one can draw an outline of grain boundaries and record for future reference the position of areas photographed. Photomicrographs of patterns are taken on 35-mm film (Micropan or Microcopy) and subsequently enlarged to a standard magnification. Illumination is furnished by a Pointolite lamp together with a green filter.

In the present investigation the specimens were cut with a fine jeweler's saw from 14-mil silicon iron strip (about 3.5 percent Si) containing large crystals grown by the strain anneal method. Each specimen included several crystals which averaged about 5 mm in diameter and extended through the strip. Surfaces smooth enough for the purposes of the experiment were prepared by first cleaning (etching) the specimens with dilute nitric acid and then electrolytically polishing them in an orthophosphoric acid bath by the method which has been described elsewhere.[5] No mechanical grinding or polishing was used at any stage in the process.

The orientation of individual crystals was determined by the method of etch reflections.

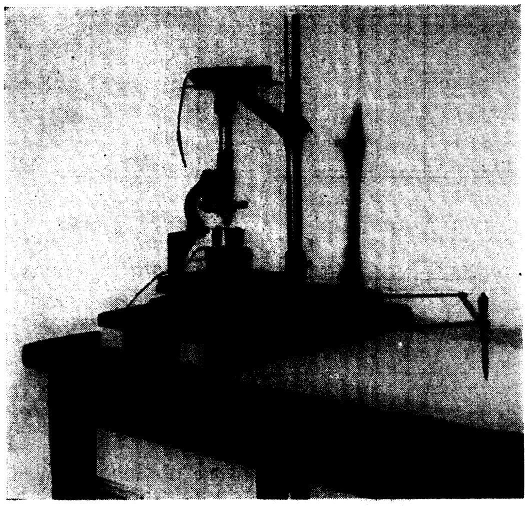

FIG. 1. Apparatus used for observing and photographing patterns.

Beautifully etched surfaces were obtained by temporarily removing the specimens from the electrolytic polishing bath prior to the completion of polishing. Usually angular coordinates for three {100} flashes were measured. When these were plotted on a stereographic projection the angles between pairs of points ordinarily differed from 90° by less than 1°. A slight tendency was noticed for the plane normals to lie too near the surface normal. A number of faint flashes were always observed, corresponding to other simple planes of high atom density. With a little practice there was no uncertainty in identifying the bright {100} flashes.

The technique of making patterns has been covered in previous papers so that only a few new observations will be recorded here. All the patterns to be described were formed with colloidal magnetite protected by soap.[6] It was noticed that the soap in the colloid formed a film on the specimen, helping to protect it from rust, and to prevent the colloid particles from adhering to its surface. This protective action made it unnecessary to coat the surface with a thin film of lacquer or grease as was done in earlier investigations. It has been found that colloidal magnetite oxidizes on standing; in a few weeks it gradually changes its color from dark red (almost black) to a lighter red and

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[4] For a brief review with references, see F. Bitter, *Introduction to Ferromagnetism* (McGraw-Hill, 1937), pp. 55 to 66. Also, see reference 3.

[5] W. C. Elmore, J. App. Phys. **10**, 724 (1939).

[6] W. C. Elmore, Phys. Rev. **54**, 1092 (1938).

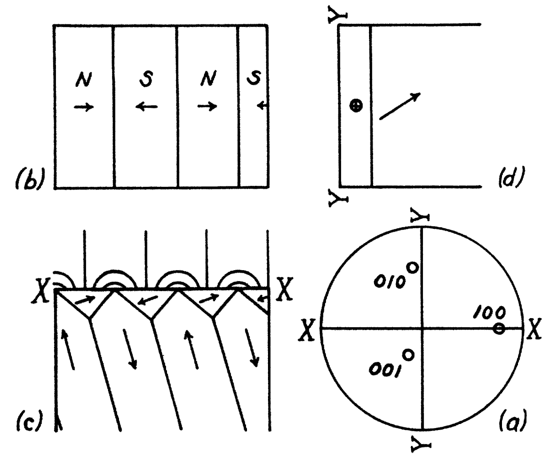

FIG. 2. Diagram illustrating one possible arrangement of magnetization at surface of crystal whose orientation is given by the stereographic projection (a). In (b), (c), and (d) are diagrams of magnetization at surface, and at representative sections $X-X$ and $Y-Y$ whose orientations are indicated in (a). The form of stray field to be expected from this structure is indicated above the $X-X$ section.

becomes less magnetic. Although the magnetic properties of the colloid can be partially restored by adding a small amount of photographic reducing agent, such as hydrochinon, together with a little alkali, patterns can be formed more readily with a freshly prepared colloid.

RESULTS AND DISCUSSION

Patterns were observed on a dozen or more crystals of known orientation. In general an applied normal field of only 10 oersteds was required to develop a pattern, although the pattern gained somewhat in sharpness when the field was moderately increased. On raising the field to 100 oersteds little or no further change was noticed. Therefore, the intensity of the surface stray fields must be of the order of magnitude of only 10 oersteds,[7] an important bit of information in deciding what sort of magnetic structure is responsible for the patterns. Thus the block structure once proposed[1] to account for the maze patterns on mechanically polished silicon-iron crystals need not be considered since it would result in much too intense stray fields. Before examining the new patterns in detail, let us consider in a general way what sort of magnetic structure an iron crystal may be expected to possess.

In an earlier paper devoted to patterns on cobalt[8] a review was made of the various factors which contribute to the formation of ferromagnetic domains.[9] For minimum energy a ferromagnetic crystal tends to be magnetized uniformly throughout its volume (minimum exchange interaction energy), to be magnetized only in certain directions (minimum magnetic anisotropy energy), and in the absence of an applied field to be magnetized so that there are no internal or superficial magnetic poles (minimum magnetic energy). The domain structure of a particular crystal represents a compromise state of magnetization, one which is consistent with minimum total energy insofar as energy losses accompanying its previous magnetic history permit. In unmagnetized ferromagnetic crystals free from strains (other than those of magnetostrictive origin) one expects to find the domain magnetization occurring principally in layers parallel to a direction of easy magnetization with adjacent layers ordinarily having opposite magnetization. Thus the layer magnetization in unstrained iron crystals will be parallel to $\langle 100\rangle$ axes.

The nature of the boundaries between ferromagnetic domains was discussed theoretically first by Bloch[10] and more recently by other authors.[11] At the so-called 180° boundaries separating oppositely magnetized layers minimum energy results when the electron spins responsible for ferromagnetism do not reverse their direction abruptly, but take one hundred or more atom spacings for the transition. In the transition region the magnetization vector is supposed to turn about the boundary normal, thereby making $\nabla\cdot\mathbf{I}=0$. Hence there is no contribution to the boundary energy of purely magnetic origin. In iron one ordinarily expects the 180° boundaries to be parallel to $\{100\}$ planes, since in the transition layer the changing direction of magnetization will then least offend the forces of magnetic anisotropy.[12]

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[7] W. C. Elmore, Phys. Rev. **58**, 640 (1940).

[8] W. C. Elmore, Phys. Rev. **53**, 757 (1938).

[9] For a recent discussion, see W. F. Brown, Jr., J. App. Phys. **11**, 160 (1940).

[10] F. Bloch, Zeits. f. Physik **74**, 295 (1932).

[11] L. Landau and E. Lifshitz, Physik. Zeits. Sowjetunion **8**, 153 (1935); R. Becker and W. Doring, *Ferromagnetismus* (Springer, 1939), p. 187.

[12] This conclusion appears obvious on viewing a model of the magnetic anisotropy surface of iron. Reference 4, p. 195.

Next let us consider the boundary between two regions each magnetized parallel to a different ⟨100⟩ axis. This 90° boundary may be expected to occur only if there result no internal magnetic poles. A simple analysis shows that the boundary must therefore occur on a plane having a zone axis of form ⟨110⟩. As with the 180° boundaries, the magnetization can again change its direction through the 90° boundary so that $\nabla\cdot\mathbf{I}=0$. To prove this, resolve the magnetization into two components, one normal and the other parallel to the boundary. The normal component does not depend on position; the other component can reverse its direction by turning about the boundary normal, so that neither component contributes to $\nabla\cdot\mathbf{I}$. Hence the energy of the 90° boundary need not contain a contribution of magnetic origin. However, widespread magnetostrictive stresses will always be set up by such a boundary, very likely making the boundary energy per unit area somewhat greater than that of the 180° variety where magnetostrictive stresses will be of minor importance since they must be primarily localized in the boundary itself. Hence it is not unreasonable that, in well-annealed iron crystals, the total area of 90° boundaries will be small, and that a 90° boundary on a {100} plane will occur but rarely. The energy per unit area of domain boundaries in iron crystals is not known accurately, but it is probably of the order of a few ergs per cm^2.

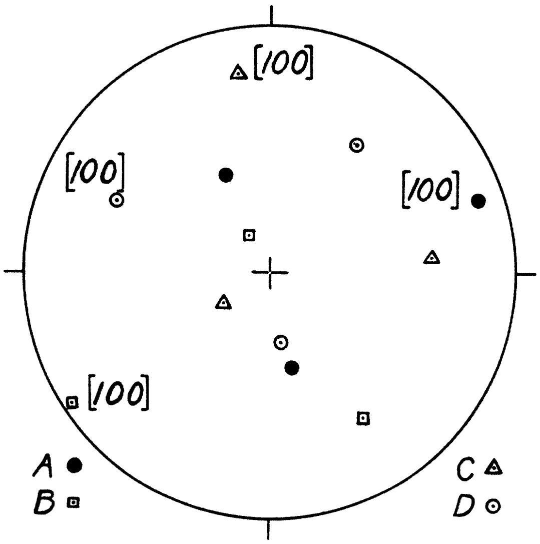

FIG. 3. Stereographic projection of the ⟨100⟩ axes of crystals A, B, C, and D. The north pole of the projection coincides with the surface normals, and the azimuth agrees with that of the various patterns illustrated in the text. The symbol [100] for each crystal has been assigned to the axis least inclined to the surface; symbols for the other two axes may be supplied by reference to the right-hand rule.

According to the previous discussion one expects an unstrained (ideal) iron crystal to possess for the greater part of its magnetic structure a layer magnetization, with boundaries separating oppositely magnetized layers on {100} planes, and with the layer magnetization parallel to one of the three ⟨100⟩ axes of the crystal. At a crystal surface which contains no direction of easy magnetization the layer structure will necessarily be modified in some way in order to avoid a large surface energy of magnetic origin. One possible modification of the layer structure is indicated in Fig. 2, where (a) gives the orientation of the crystal, (b) the magnetization at the surface, (c) and (d) the magnetization at representative sections $X-X$ and $Y-Y$ whose orientations are indicated in (a). For minimum energy the magnetization in the surface domains of triangular section will occupy a direction in between the ⟨100⟩ direction least inclined to the surface and the projection of the latter direction in the surface. The surface, therefore, will be covered with bands of north and south poles, giving the sort of stray field indicated above the $X-X$ section. Most of the lines of induction form closed circuits within the crystal.

FIG. 4. Pattern on crystal A near grain boundary; normal applied field of 22 oersteds. Magnification 100×.

A second possible modification of the simple layer structure which will minimize surface stray fields is suggested by the magnetic structure of cobalt. An analysis of colloid patterns on cobalt

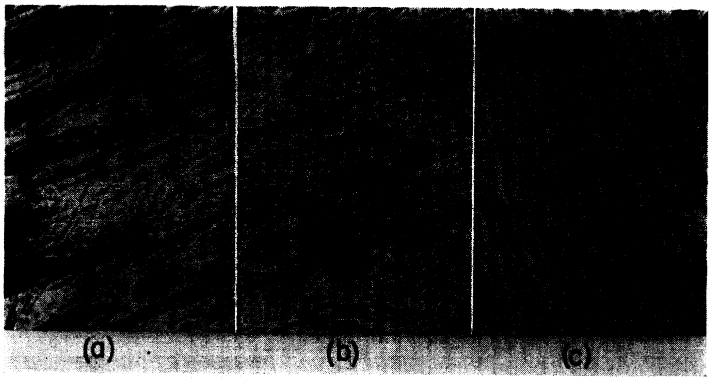

FIG. 5. Patterns on crystal A; normal applied field of 55 oersteds. The patterns (a) and (b) illustrate the development of a fine structure attributed to dendrite-like domains of reversed magnetization localized near the surface; (c) illustrates a second type of pattern found on crystal A. Magnification 100×.

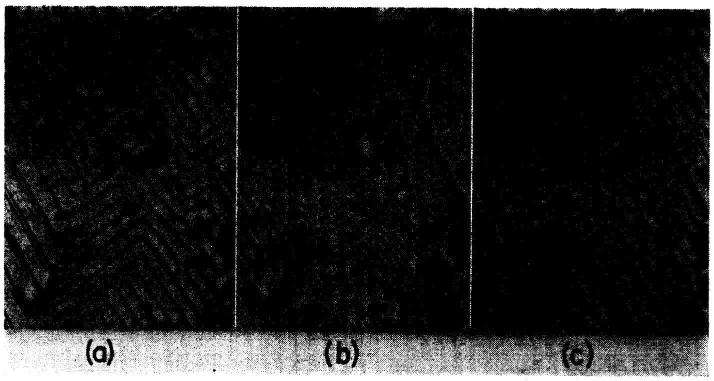

FIG. 6. Patterns on crystal B; (a) normal field of 95 oersteds; (b) no applied field; (c) same as (a) with direction of field reversed. Magnification 100×.

indicates that at surfaces considerably inclined to the hexagonal axis (the one direction of easy magnetization) there always occur dendrite-like regions of reversed magnetization extending a short distance into the primary layer magnetization. This surface fine structure evidently decreases the energy of magnetic origin at the cost of increasing, to a lesser extent, the energy tied up in the domain boundaries near the crystal surface. It is therefore reasonable to expect that the magnetic dendrites will occur whenever the magnitude of the surface stray field tends to exceed the value required for the formation of new nuclei of reversed magnetization, ordinarily a matter of a few oersteds. The dendrites constitute a fine structure superposed on the primary layer structure.

From the many patterns observed on different crystals it has been necessary to select for

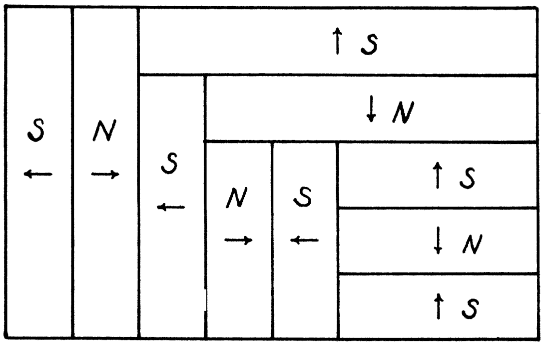

FIG. 7. Diagram of portion of crystal surface, suggesting arrangement of layer magnetization of Fig. 2 which will account for corners and other features in a maze pattern.

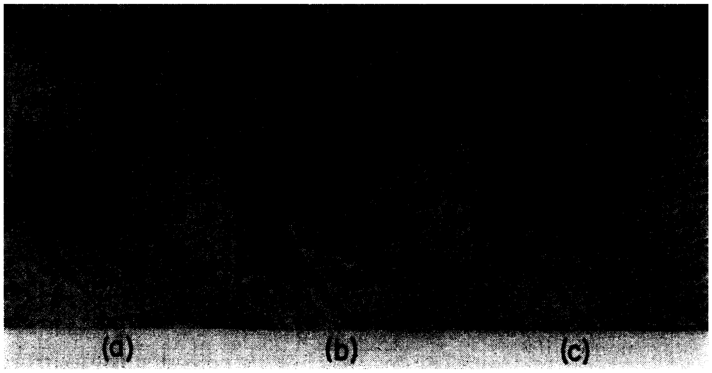

FIG. 8. Patterns on crystal C; (a), (b), and (c) as in Fig. 6. Magnification 100×.

reproduction only a few typical cases. These patterns, which occurred on four crystals designated by A, B, C, and D, show the variety in form which the magnetic structure can take. In addition some of the patterns reproduced here possess individual features which have been very helpful in deciding what arrangement of magnetization is responsible for a particular type of pattern. A stereographic projection, Fig. 3, shows the orientation of the ⟨100⟩ axes of the four crystals.

Let us first consider the pattern on crystal A, since this crystal has but one direction of easy magnetization, the [100] axis, approximately parallel to its surface. In Fig. 4 is shown the pattern near a grain boundary, produced by a normal field of 22 oersteds. This pattern, together with other patterns found with reversed field and with no field, is very similar to that found on prism planes of cobalt crystals. Hence there is little doubt that here we have a layer magnetization occurring in the [100] direction. Presumably the layers extend through to the opposite face of the crystal. The occurrence of the large magnetic dendrites at the grain boundary is especially to be noted and compared with the similar behavior exhibited by the cobalt patterns at a grain boundary.[13] In Figs. 5a and 5b are illustrated the changes in the pattern on crystal A occurring at successively greater distances from the grain boundary. The predominant layer structure can still be distinguished, but now the former bands are covered with many dendrite-like regions of reversed magnetization, presumably localized near the surface. The lack of fine structure near the grain boundary suggests that either strain, or a slight curvature of the surface (caused by the depression of the grain boundary by etching and by electrolytic polishing) has reduced the magnitude of the stray field by making the underlying magnetization more nearly parallel to the surface. The stray field in the vicinity of the grain boundary is thus too weak to initiate the fine structure. The pattern on crystal A differed from patterns on other crystals in that it completely changed its character near the diametrically opposite grain boundary. The pattern found there is illustrated in Fig. 5c. Although the heavy colloid deposits now run perpendicular to the [100] direction, the magnetic dendrites continue to be parallel to this direction. The evidence suggests that here we have a structure of the sort illustrated in Fig. 2, i.e., the primary layer magnetization occurs in either the [010] or in the [001] direction, with the principal domain boundaries on (100) planes.

In Fig. 6 are illustrated maze-like patterns found on crystal B, with no applied field, and with a positive or negative normal field of 95 oersteds. On relating the patterns to the crystal orientation the fact is noted that the fine structure occurs only with the lines perpendicular to the [010] axis (inclined about 20° to the surface) and not with the lines perpendicular to the [100] axis (inclined only a few degrees to the surface). It was further observed that the

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[13] Reference 8, Fig. 4.

FIG. 9. Pattern on crystal D; normal applied field of 22 oersteds. Magnification 100×.

pattern did not change its scale near grain boundaries, differing in this respect from the pattern on crystal A, Fig. 4. The location of the fine structure, the orientation of the principal pattern lines, and the behavior at grain boundaries all point to the magnetic structure of Fig. 2, at least as regards sets of parallel lines in the pattern. It remains to be demonstrated how the two-layer structures, parallel to the [010] and the [100] planes, can join together to form corners and other pattern details without producing excessive local magnetic fields. The only apparent way in which this can be done is suggested in Fig. 7. Each rectangular strip in this diagram represents the top of a domain of triangular cross section, as in Fig. 2, and is covered with magnetic poles of polarity indicated by *N* or *S*. A careful study of the colloid distribution in the three patterns of Fig. 6 tends to confirm this sort of arrangement. The narrowness of the pattern lines may be attributed to a slight non-uniformity in the distribution of surface poles on each domain of triangular cross section.

The similarity between the present patterns and the finer scaled maze patterns characteristic of mechanically polished crystals suggests that the two must have a similar explanation. Since the structure responsible for the earlier maze patterns is closely related to the damage left from polishing, it is reasonable to conclude that the fine-scaled layer magnetization (layers about 2 microns thick) is localized near the surface and that it gives way to a coarser layer magnetization within the crystal. Indeed evidence for such a structure was reported when the maze patterns were first described.[1] Kennard has already proposed[14] that a layer magnetization similar to Fig. 2 is responsible for the sets of parallel lines in maze patterns, but he attributes the surface stray fields to the "surface tension" of domain boundaries, rather than to the inclination of the direction of easy magnetization with respect to the surface, the view here adopted. It seems likely that the surface energy (tension) of the boundaries is too small to play an important part in producing surface stray fields.

Let us consider the patterns shown in Fig. 8, found near a grain boundary on crystal C. It is evident that the distribution of the colloid with no applied field, and with applied normal fields, the orientation of the bands, and the behavior of the pattern at the grain boundary all point again to the domain structure of Fig. 2, with boundaries between layers occurring on (100) planes. The bands, except for local imperfections, were observed to extend all the way across the crystal and to be covered everywhere with fine structure indicating the presence of local magnetic dendrites. It is interesting to compare the fine structure of Figs. 5c, 8, and 6 with one another, and to relate them to the angles between the surface and the ⟨100⟩ axes concerned. The three angles are, respectively, 6°, 12°, and 20° and the three patterns suggest, reasonably enough, that the dendrites point with increasing angles into the crystal surface.

Many of the patterns on other crystals were similar to those which have been discussed, indicating that layer magnetization, accompanied by a finer-scaled surface structure, is of common occurrence. The pattern of crystal D, Fig. 9, however, shows that the magnetic structure may be much more complicated in a crystal whose surface approaches the orientation of a {111} plane. The structure in Fig. 9 is very fine scaled and at first sight appears to bear no relation to the orientation of the crystal. Close inspection, however, reveals local groups of parallel lines whose orientation is for the most part along the traces of {100} planes in the surface. Although these lines suggest local regions of layer magnetization, it is likely that

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[14] E. H. Kennard, Phys. Rev. **55**, 312 (1938).

the pattern fails to reveal the nature of the domains lying deep in the crystal.

The scale of the various magnetic structures herein described deserves special mention. On the assumption that the interpretation of the patterns is substantially correct, then the thickness of the layer domains ranges from ten to fifty microns, varying from crystal to crystal, but remaining fairly uniform over considerable areas of one crystal. The structure of crystal D, of course, constitutes an exception. Ideally the scale of a magnetic structure is set when the energy associated with domain boundaries just equals that associated with the external surface of the crystal, since the former varies inversely and the latter directly with a length characterizing the structure.[8, 9] On account of the complexity of the magnetization in the surface domains, it appears impossible to make any accurate estimates of the energies involved. The variation in layer thickness for different crystals in the same specimen may be attributed partly to differences in crystal orientation, and partly to the effect of previous magnetic history. The relative importance of these two factors has not yet been looked into.

There are still to be investigated many interesting points regarding colloid patterns on iron crystals. It should be possible to follow in detail changes in magnetic structure accompanying the magnetization process, and to correlate abrupt structural changes with Barkhausen noise. The particular form taken by the magnetic structure is undoubtedly related to the previous thermal and magnetic history, grain size, the existence of strains, the hysteresis properties, initial permeability, etc., of the specimen. Definite information regarding these matters should lead to a better understanding of many aspects of ferromagnetism.

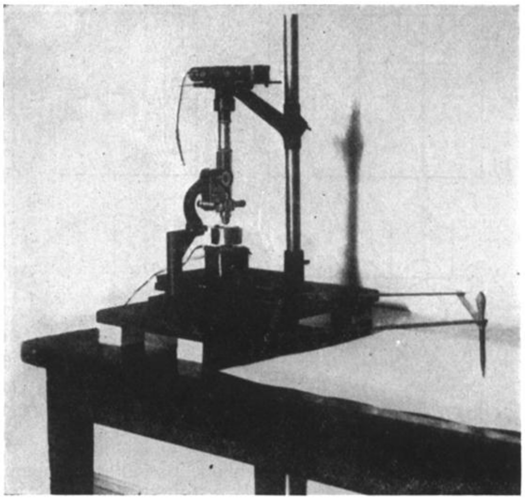
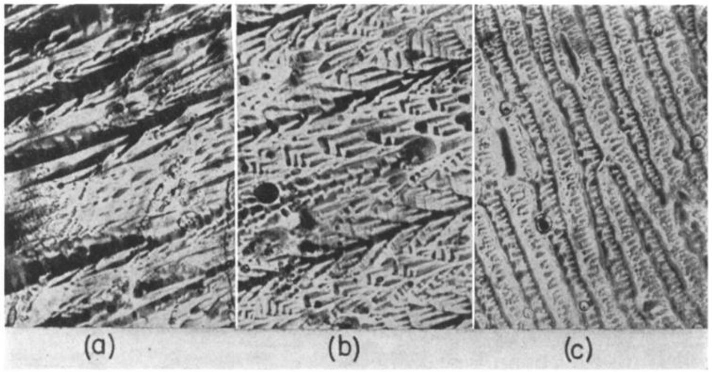


FIG. 1. Apparatus used for observing and photographing patterns.

FIG. 4. Pattern on crystal A near grain boundary; normal applied field of 22 oersteds. Magnification 100×.



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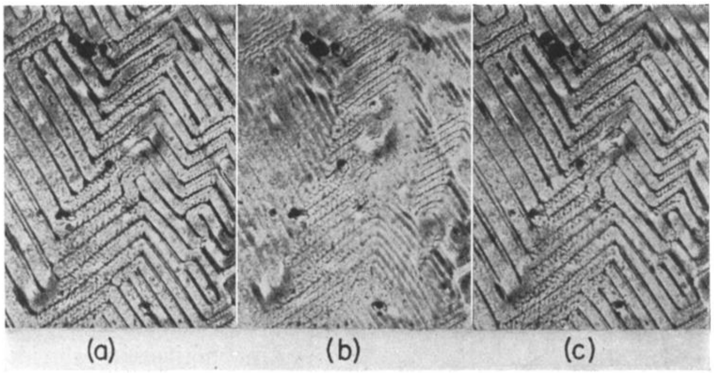

FIG. 6. Patterns on crystal B; (a) normal field of 95 oersteds; (b) no applied field; (c) same as (a) with direction of field reversed. Magnification 100×.

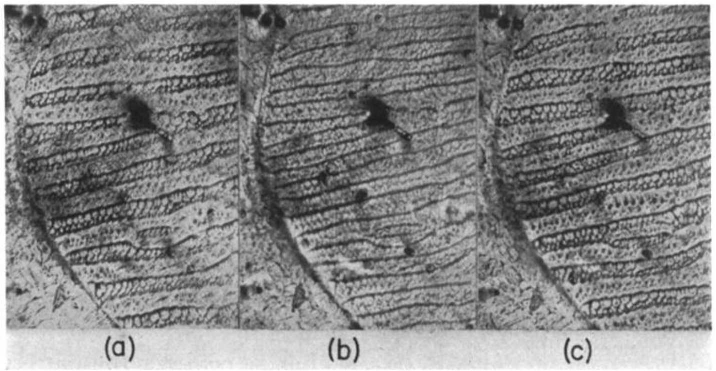

FIG. 8. Patterns on crystal C; (a), (b), and (c) as in Fig. 6. Magnification 100×.

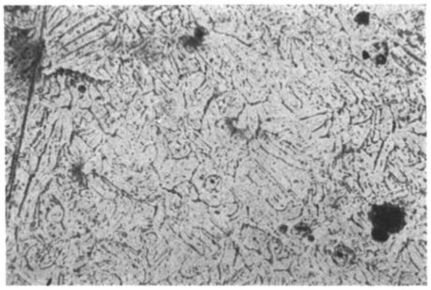

FIG. 9. Pattern on crystal D; normal applied field of 22 oersteds. Magnification 100×.