Experiments on the Nature of Ferromagnetism

By FRANCIS BITTER
Westinghouse Research Laboratories, East Pittsburgh, Pennsylvania

(Received May 31, 1932)

Irregularities in the magnetization of ferromagnetic crystals were studied by means of magnetic Fe_2O_3 particles about 1μ in diameter suspended in ethyl acetate. On iron crystals the particles settle along parallel lines spaced about 0.1 mm apart and more or less at right angles to the applied field. On nickel crystals the pattern is similar, but the lines split up into more complicated structures as the applied field is increased. On cobalt crystals straight lines are obtained on some crystals, and spotty patterns on others. The spots arrange themselves in rows when the crystals are magnetized. The patterns can be destroyed by relatively slight surface strains. There is some evidence that in the straight line patterns the direction of the lines is related to the direction of magnetization. An explanation of these effects is not known.

THE object of the experiments described below was to find any irregularities or inhomogeneities that exist in the magnetization of ferromagnetic crystals.

APPARATUS AND MATERIALS

The method used was the old magnetic powder method. In order to reveal small details, the particles must be small and have as large a magnetic moment as possible without causing them to form chains in the direction of the magnetic field. They must further be suspended in some medium that will allow them to settle according to the pattern of the field. Particles of red Fe_2O_3 having diameters in the neighborhood of 1μ were found very satisfactory.[1] These were suspended in ethyl acetate, which, because of its low viscosity, allows the particles to settle fairly rapidly and without undue disturbance from currents in the liquid. Observations were made by magnetizing a flat sample in the direction of its length and placing a drop of the suspension on the surface of the crystal to be investigated. In order to be certain that the patterns obtained were determined by the sample investigated rather than by the interaction of the Fe_2O_3 particles, observations were made with particles of different size, and of different substances suspended in various liquids, and with no liquid at all. In all cases the same patterns were observed.

A few samples of iron were prepared by elongating small annealed slabs about 3 percent and annealing in hydrogen just below the $\alpha-\gamma$ transformation point. This material was very pure, having a total impurity before the above treatment of about 0.01 percent, chiefly Si. Some samples were ground flat and annealed. All other samples were made by melting in alundum troughs in an electrically-heated alundum tube furnace through which purified dry hydrogen flowed continuously. In order to observe the effects re-

[1] F. Bitter, Phys. Rev. **38**, 1903 (1931).

ported below, it is desirable to have a shiny, smooth, unstrained surface. Hydrogen was used to prevent surface tarnish due to oxidation. Considerable hydrogen was absorbed while the samples were molten, and given off during solidification, producing cavities in the ingots and locally imperfect surfaces. In most cases, however, at least a part of the sample was satisfactory.

I should like to express my indebtedness to various members of the Laboratories for their assistance—to Mr. L. McCulloch for preparing the Fe_2O_3 powder; to Dr. V. Hicks for making an x-ray examination of this powder; to Miss M. Ferguson for taking the best of the photographs reproduced below; to Mr. P. G. McVetty for stretching the strain-annealed samples of iron; and especially to Mr. A. A. Frey, who built the furnaces and prepared most of the samples investigated.

Results

Iron

In a sample of iron having small grains, whether annealed or not, irregular patterns are found. The deposits do not follow any obvious structural features, such as grain boundaries. If, however, the grains are somewhat larger, and unstrained, deposits like that shown in Fig. 1 are formed, which, for larger fields, degenerate into irregular patterns. In Fig. 1 some of the

Fig. 1. Deposit on a sample of 4 percent Fe-Si with large grains. Field horizontal. Approximate magnification ×2. Deposits are black in all photographs.

grains are crossed by systems of parallel lines more or less at right angles to the direction of the applied field. The lines are not spaced perfectly evenly, but are of the order of 0.1 mm apart. Fig. 2 shows the deposit obtained near a grain boundary on an Fe-Si sample, and Fig. 3 shows the slip lines obtained on a similar sample after a slight reduction by cold rolling. The similarity is rather striking. In both cases there is a tendency for the lines to continue in a changed direction after crossing a grain boundary.

In general, the deposits on iron are indefinite, and it often occurs that no regular patterns are observed at all. Better results were obtained with an alloy containing 4 percent Si than with pure iron. An attempt was made to determine how the pattern changed as the direction of magnetization was varied. As a rule the pattern was obscured, but on several crystals three sets of lines were observed all told. Further observation is required to establish the generality of this result.

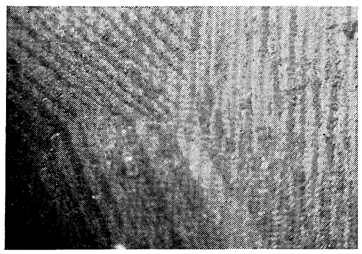


Fig. 2.

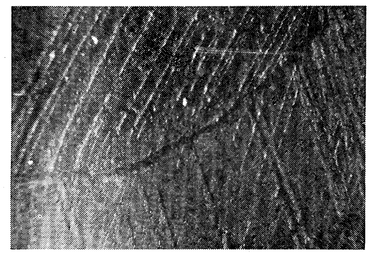


Fig. 3.

Fig. 2. Deposit on a 4 percent Fe-Si sample near a grain boundary. Field horizontal. Approximate magnification $\times 10$.

Fig. 3. Slip lines in a 4 percent Fe-Si sample. Approximate magnification $\times 10$.

Cobalt

Typical deposits on cobalt are shown in Figs. 4–6. These illustrate the state of affairs before a magnetic field is applied. In Fig. 4 the spots vary a good deal in size. Fig. 5 shows grain boundaries, and the tendency for the

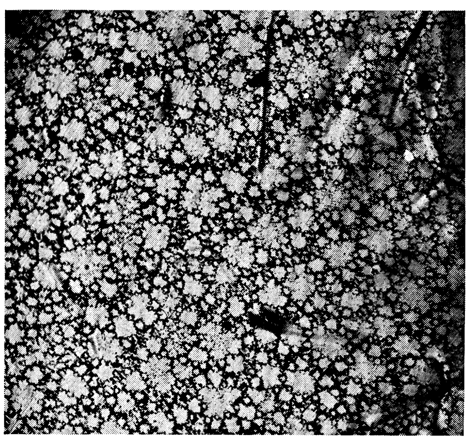


Fig. 4.

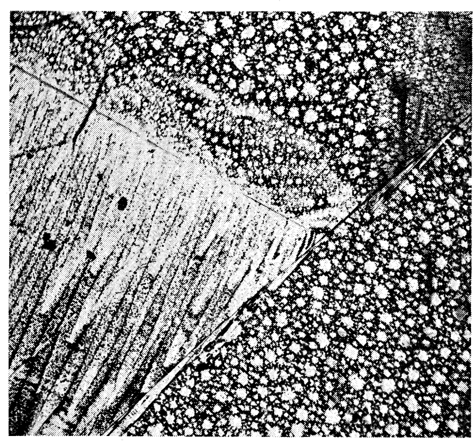


Fig. 5.

spots to form a hexagonal array. Also a grain having a straight line pattern is shown. Fig. 6 shows transition patterns between the spotty and the straight line types. Just what difference exists between samples showing different patterns is not known. One sample containing 20 percent of iron showed no pattern whatsoever. Until further evidence is obtained, we may make the reasonable assumption that the irregularities are produced by strains, impurities, or other imperfections. In Figs. 7–9 the effect of magnetization on

Fig. 6.

Fig. 4–6. Deposits on unmagnetized cobalt crystals. Magnification approximately $\times$**70.**

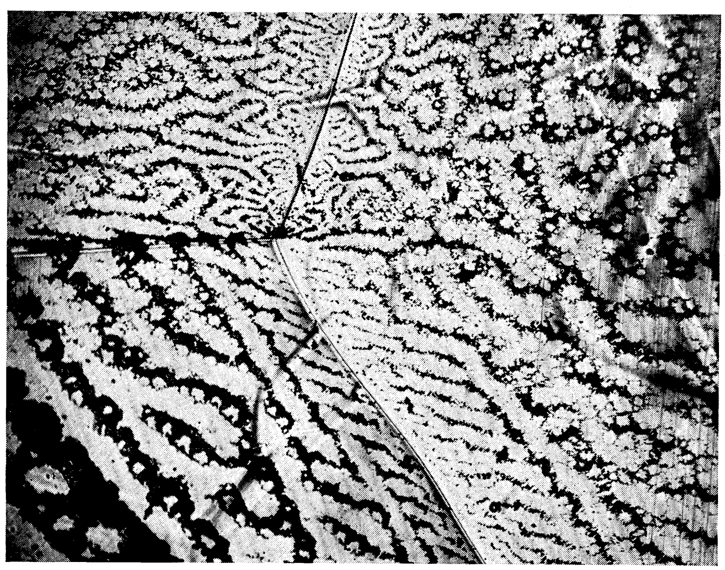


Fig. 7. Deposit on magnetized cobalt near a grain boundary. Approximate magnification $\times$**80.**

the spotty type of deposit is shown, the magnetization being stronger in Fig. 9 than in Fig. 8. Fig. 7 shows the distorting effect of the grain boundary. The line pattern illustrated in Fig. 5 is affected little if at all by magnetization, and has in no case been observed to change its direction on the crystal surface as the direction of the applied field was varied.

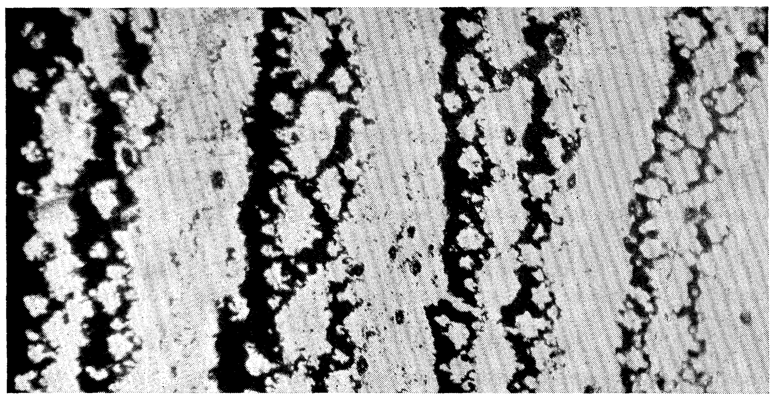


Fig. 8.

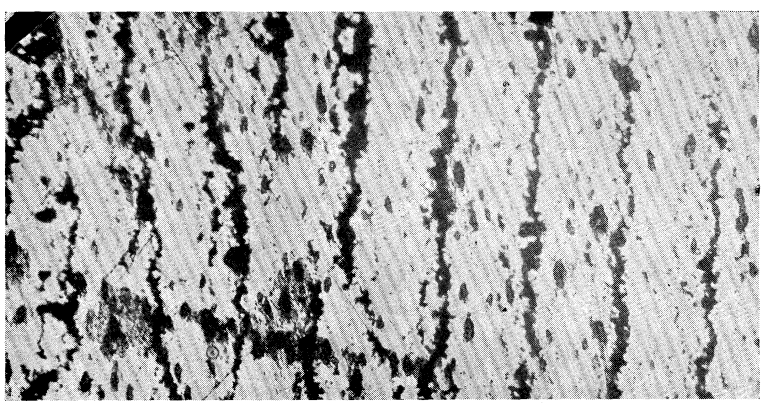


Fig. 9.

Fig. 8 and 9. Deposits on a magnetized cobalt crystal. Approximate magnification ×160.

Nickel

A typical deposit on nickel before being exposed to any magnetic field other than that of the earth is shown in Fig. 10. On magnetization the patterns become more definite, as shown in Figs. 11a and 11b. Sometimes the lines are curved, especially near grain boundaries, and sometimes they show a break as is the case for one of the lines in Fig. 10. It quite often happens that the spacing of the lines changes progressively from one side of a crystal to the other, as can be seen in Fig. 11a. These lines are not associated with fixed places in a crystal. This is illustrated in Fig. 12 in which photographs of three different deposits on the same region are superposed. It seems quite generally true that the position of the lines changes after demagnetization.

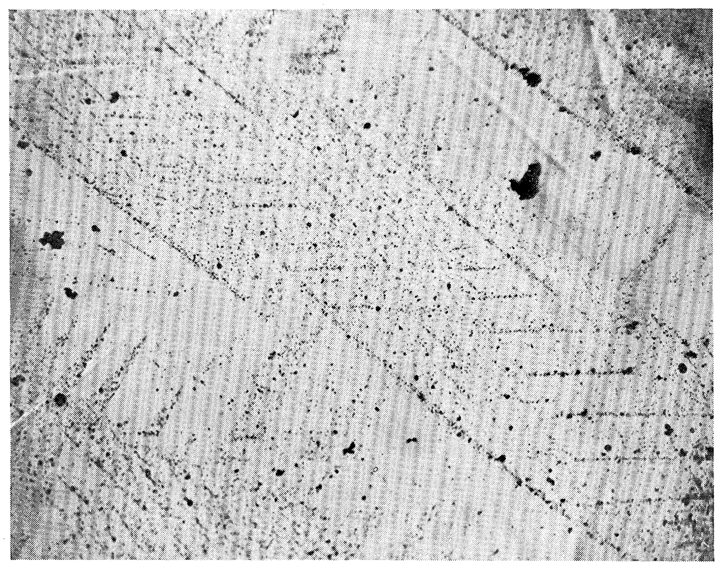
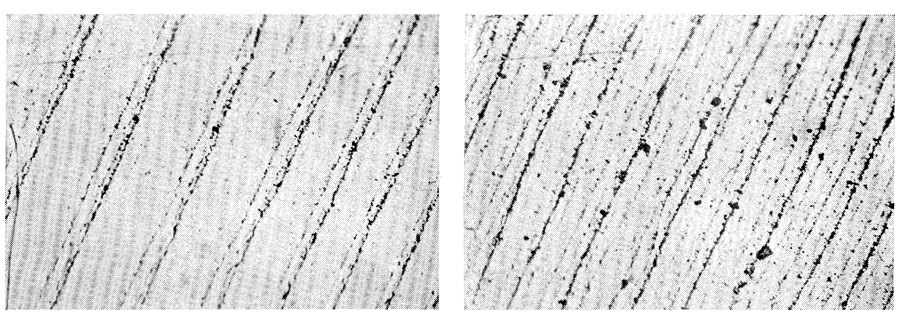


Fig. 10. Deposit on a nickel crystal before magnetization. Approximate magnification $\times 80$.

Some samples in the shape of disks were prepared, and magnetized in eight directions at 45° intervals. The deposits were photographed and the total number of directions in which lines appeared was determined for five crystals.



a b

Figs. 11a and 11b. Typical deposits on nickel with the applied field acting in a horizontal direction. Approximate magnification $\times 65$.

In each case the lines appeared in four directions all told. In those positions in which two such directions made nearly equal angles with the applied field chevron patterns like that shown in Fig. 13 appeared.

If successive patterns are obtained in the same region in gradually increasing fields without demagnetization between observations, the phenomenon shown in Fig. 14 is found. The wavy marks on the photographs are surface irregularities that are of no interest here except insofar as they may be used as a reference system fixed in the crystal. In the twelve photographs shown the applied field increases progressively, being smallest in 1 and largest in 12. The position of the lines as shown in the first photograph does not change radically. The lines, however, split up as the magnetizing field is increased, first into two, then four, and on the original photograph, No. 12, each of the original lines is seen to consist of eight components. In addition a new line appears between each of the main groups in photograph No. 7 and splits into two components also. The smallest splitting here shown is deter-

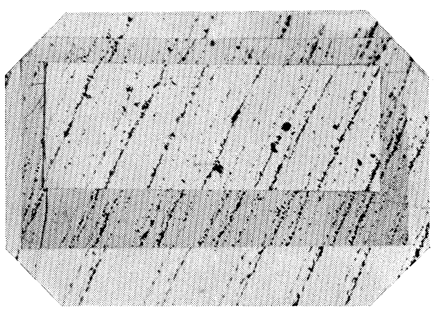


Fig. 12. Superposed photographs of deposits obtained on the same region at different times, the sample being demagnetized after each observation.

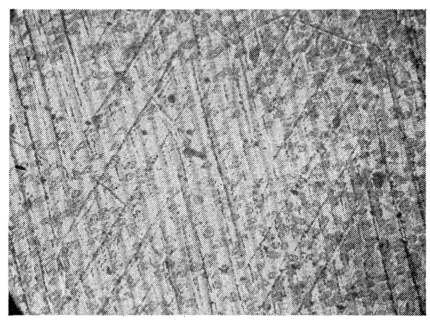


Fig. 13. Peculiar pattern obtained on a nickel crystal.

mined by the method, and it seems very probable that much more detail exists than is here indicated. To obtain these results it is essential not to demagnetize the sample between observations. The sample consisted of crystals having linear dimensions of from 1 to 4 cm imbedded in a large slab, 27 cm$\times$4 cm$\times$1 cm. It was consequently possible to measure only the intensity of magnetization of the aggregate. This was done with a fluxmeter and the value of $I=350$ or about 75 percent of saturation was obtained for the conditions under which deposit No. 12 was produced. Magnetization of this intensity is confined to the directions of easiest magnetization within the crystal.

Alloys containing 10 percent, 20 percent, and 30 percent of iron were prepared by melting and slow cooling. That containing 10 percent of iron showed patterns typical of nickel, but less distinctly than usual. No patterns whatsoever could be found on the other samples.

The effects are exceedingly sensitive to strain. Some samples that were lightly pressed by a C-clamp no longer gave any patterns, nor was anything to be found on polished samples until they had been annealed.

Discussion

If it is quite generally true, as has been found in a few cases, that the line patterns can appear in three directions in iron, in four directions in nickel, and in only one direction in cobalt, it seems reasonable to assume that they are somehow related to the three (100) axes in iron, the four (111) axes in nickel, and the single (0001) axis in cobalt. This assumption is supported by

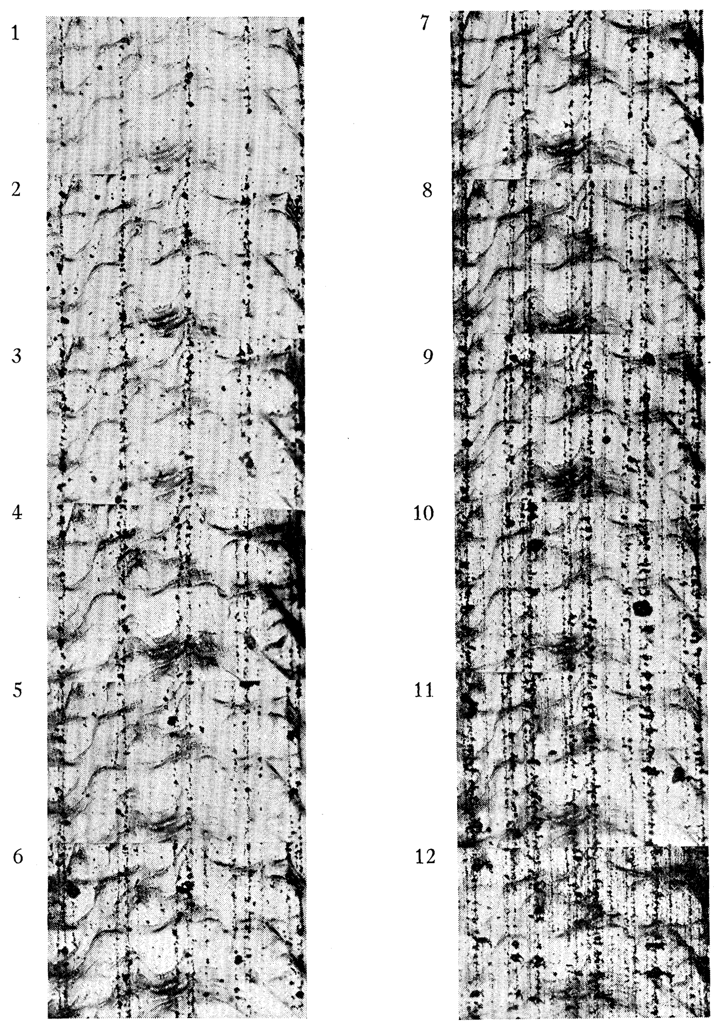


Fig. 14. Twelve photographs of the same region, the applied field being gradually increased between observations.

the fact that in each case the axis mentioned has a special significance magnetically—it is the direction of easiest magnetization within the crystal. Taking into consideration the above material, and the fact that the lines are sometimes curved or broken, we suggest the hypothesis that inhomogeneities occur along planes perpendicular to the direction of magnetization, and that the lines observed are parallel to the intersection of such planes with the surface under observation. In the fields used the direction of magnetization probably differed in no case appreciably from the direction of easiest magnetization. The above hypothesis fits into the observation that in nickel and cobalt alloying tends to destroy the pattern, for the effect of a foreign substance must, as a rule, be to introduce local fluctuations in the direction and intensity of magnetization. This reasoning can be applied to the observations on iron if we suppose that it is exceptionally difficult to produce a satisfactorily uniform crystal of iron, and that the addition of silicon tends to remove certain imperfections. The above discussion does not take into account the spotty pattern obtained on certain cobalt crystals. It seems likely that this is more complicated in its origin, and occurs whenever the hexagonal axis is normal to the surface of the crystal.

From the observations so far made, it is not possible to determine the nature of the inhomogeneities revealed. They may be changes in direction or intensity of magnetization, or both. Bearing in mind that in this discussion a change in direction, to be detected as such, must cover a region large compared to 1μ, it is probable that we have to do with changes in direction in cases like that shown in Fig. 13, but that in patterns like that shown in Fig. 11 there is only a change in intensity. It is most important to settle definitely the nature of the inhomogeneities by further and more detailed observations.

The origin of the patterns obtained is of course even more obscure. The conception is current that ferromagnetic crystals are made up of spontaneously magnetized regions whose directions of magnetization are distributed among the various energetically possible orientations. Magnetization is then supposed to result partly from a growth of some of these regions at the expense of their neighbors,[2] partly from changes in the direction of magnetization of each region. It seems a little difficult to fit this picture into the above photographs unless the regions are smaller than 1μ. The secondary structures suggested by Zwicky[3] offer a different attack on the problem. Finally, we must remember that a crystal consists of atoms in different excited states, that the various periodic motions which its constituent atoms and electrons perform most probably are coupled together in a variety of ways, and that in addition there must be a certain amount of electromagnetic radiation present. Any of these factors may have an influence in determining the equilibrium configuration of a crystal, and it is perhaps the most important aspect of experiments such as the above to furnish evidence from which conclusions on these points may eventually be drawn.

[2] F. Bloch, Zeits. f. Physik. **74**, 295 (1932).
[3] F. Zwicky, Phys. Rev. **38**, 1772 (1932).

Fig. 1. Deposit on a sample of 4 percent Fe-Si with large grains. Field horizontal. Approximate magnification $\times 2$. Deposits are black in all photographs.

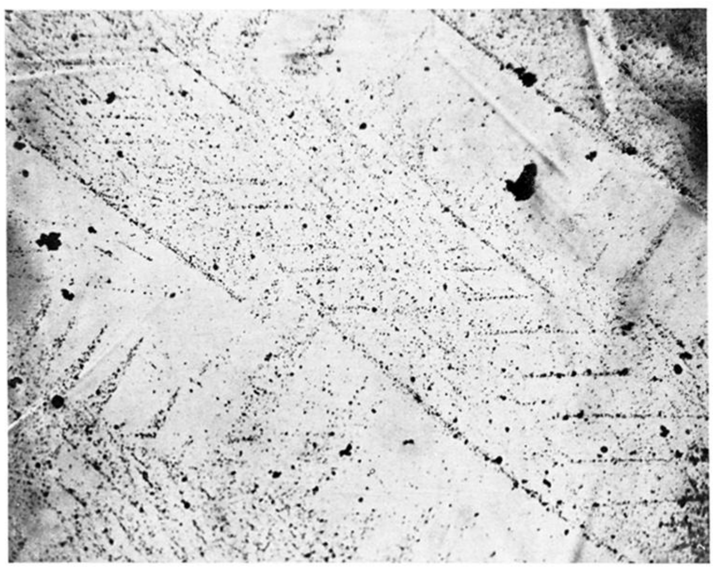


Fig. 10. Deposit on a nickel crystal before magnetization. Approximate magnification $\times 80$.

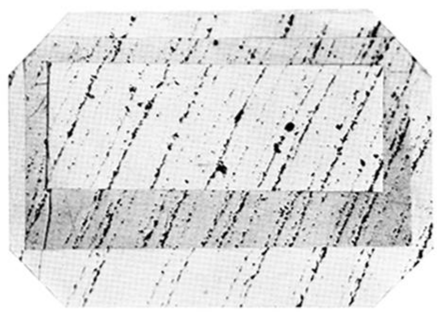


Fig. 12. Superposed photographs of deposits obtained on the same region at different times, the sample being demagnetized after each observation.

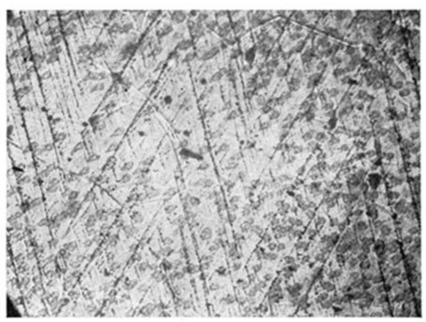


Fig. 13. Peculiar pattern obtained on
a nickel crystal.

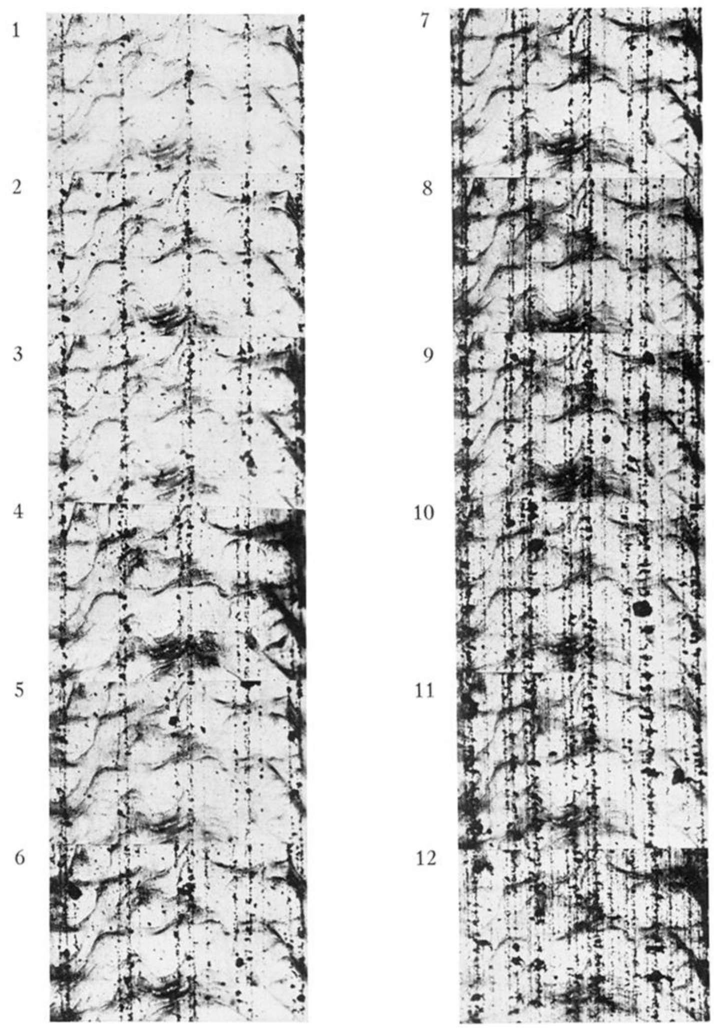


Fig. 14. Twelve photographs of the same region, the applied field being gradually increased between observations.

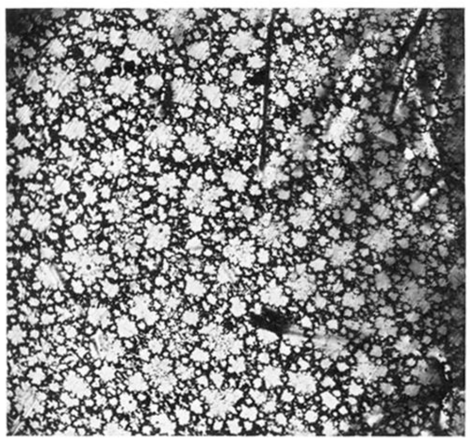


Fig. 4.

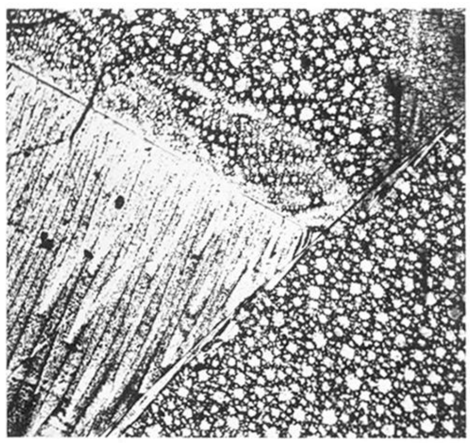


Fig. 5.

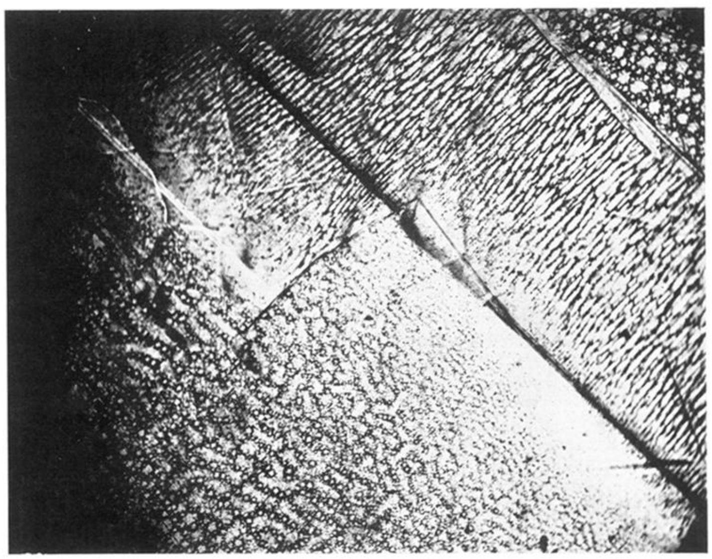


Fig. 6.

Fig. 4–6. Deposits on unmagnetized cobalt crystals. Magnification approximately $\times$**70.**

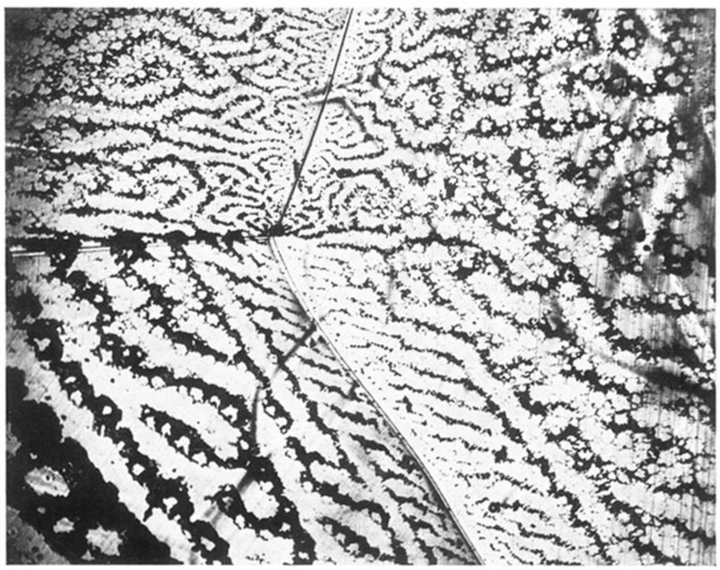
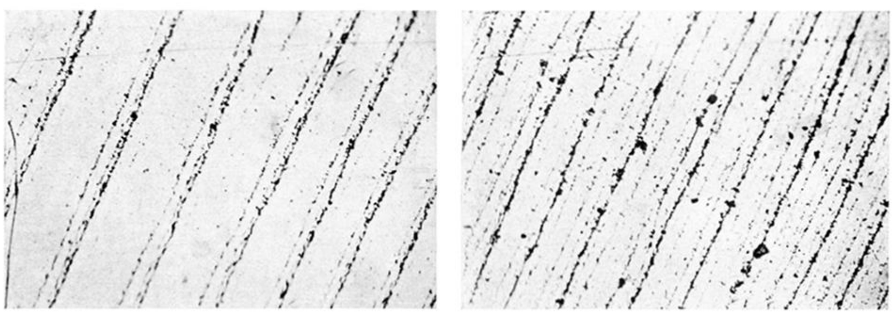


Fig. 7. Deposit on magnetized cobalt near a grain boundary. Approximate magnification $\times 80$.



a b

Figs. 11a and 11b. Typical deposits on nickel with the applied field acting in a horizontal direction. Approximate magnification $\times 65$.

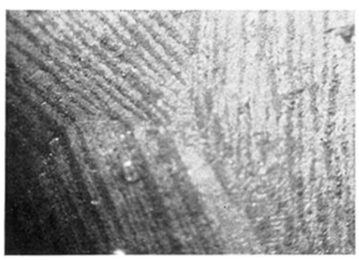


Fig. 2.

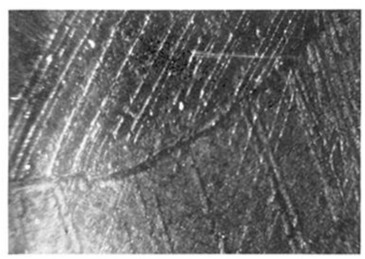


Fig. 3.

Fig. 2. Deposit on a 4 percent Fe-Si sample near a grain boundary. Field horizontal. Approximate magnification $\times 10$.

Fig. 3. Slip lines in a 4 percent Fe-Si sample. Approximate magnification $\times 10$.

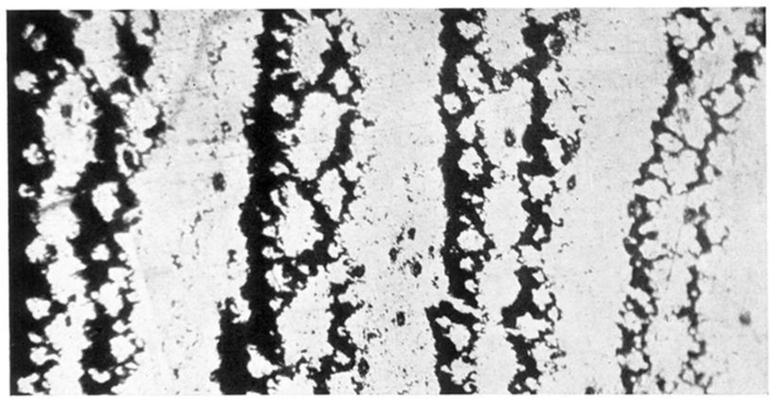


Fig. 8.

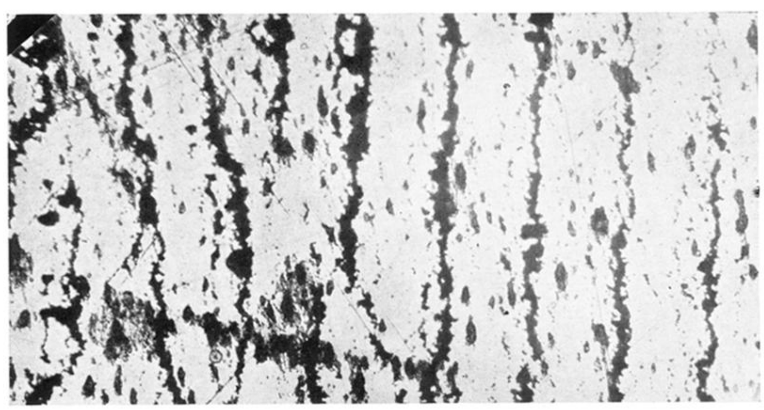


Fig. 9.

Fig. 8 and 9. Deposits on a magnetized cobalt crystal. Approximate magnification $\times 160$.