Magnetic After-Effect in Iron due to Motion of Dislocations*

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The magnetic after-effect in iron at high temperatures due to motion of dislocations is investigated. The specimens, consisting of a few large crystals, are examined, some after a careful annealing and some after a small plastic deformation. The intensity of the after-effect is measured as the horizontal displacement (viscosity field) between two magnetization curves: one taken immediately after demagnetization, and the other taken a long time later.

On annealed Armco iron the magnetic viscosity begins to appear above 320°C, reaching 0.3 amp-turn/m at about 450°C. On the other hand, in cold worked specimens the viscosity field is appreciably larger and is already observable below 200°C. Similar results are obtained on high-purity electrolytic iron. Comparison with the relaxation of elastic modulus, occurring in the same temperature range, seems to confirm that the observed magnetic viscosity is due to dislocation motion.

An interpretation of the phenomenon, on the basis of the Vicena theory concerning the dependence of the coercive force on the dislocation density, is given.

INTRODUCTION

THE influence of time on some magnetic properties has been known since 1889,[1,2] but the physical basis of most of these effects has been clearly understood only recently. Apart from the after-effect due to thermal fluctuations[3,4] the magnetic viscosity is generally due to diffusion of solute atoms or lattice defects (diffusion viscosity). A comprehensive theory of the diffusion viscosity due to solute atoms has been given by Néel.[5–7] Experimentally, a great deal of work has been done on the viscosity due to interstitial C or N in iron by Snoek[8] and recently by others.[9,10] On the contrary very little attention has been given to the other possible sources of magnetic viscosity.

Since the observations of Weiss and Freudenreich[11] on time changes of permeability of an FeCo alloy, the only magnetic viscosities at high temperatures reported until now, are those of Fahlenbrach[12,13] on iron and on some iron alloys. The magnetic after-effect due to grain boundary relaxation has been considered by us in previous papers.[14,15]

The purpose of the present work is to study the magnetic viscosity due to dislocation motion. This source of after-effect has not been examined before, at least as far as we know. For this type of research pure iron has been chosen as the most suitable material. Nickel, although have a larger magnetostriction, has a too low Curie point. On the other hand magnetic alloys cannot be used for this type of investigation because the effects due to diffusion and reordering of solute atoms may entirely mask the effect of dislocations.

EXPERIMENTAL RESULTS

The specimens were long strips containing only two or three very large crystals and hence for our purposes can be considered as single crystals. The crystals have been grown by the standard technique of a critical cold working. After a preliminary cold rolling of about 80% they have been carefully annealed in dry hydrogen at about 750°C (650°C for the electrolytic iron), then they have been deformed again by rolling of 3% and kept at 870° for 100 hours. After this treatment crystals several centimeters long were obtained.

The technique for measuring the magnetic viscosity is the same as that described in previous papers.[14,15] At the temperature of measurement, the specimen is demagnetized by 50 cps current, in a few seconds. Then a 30 cps field H of the order of some amp-turn/m is applied by feeding a primary coil made of 1100 turns wound on a quartz tube 500 mm long, with inside diameter 80 mm. The secondary coil of 4000 turns is

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[1] I. A. Ewing, Proc. Roy. Soc. (London) **46**, 269 (1889).

[2] R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Company, New York, 1950).

[3] L. Néel, J. phys. radium **11**, 49 (1950).

[4] R. Street and J. C. Wolley, Proc. Phys. Soc. (London) **62**, 562 (1949).

[5] L. Néel, J. phys. radium **12**, 339 (1951).

[6] L. Néel, J. phys. radium **13**, 250 (1952).

[7] L. Néel, J. Appl. Phys. **30**, 3S (1959).

[8] J. L. Snoek, *New Developments in Ferromagnetic Materials* (Elsevier Publishing Company, Inc., Amsterdam, 1947).

[9] P. Brissoneau, Compt. rend. **244**, 868 (1957) and J. Phys. Chem. and Solids **7**, 22 (1958).

[10] G. Biorci, A. Ferro, and G. Montalenti, Atti accad. nazl. Lincei. Mem. Classe sci. fis. mat. e nat. Sez. VIII **24**, 542 (1958). See also Air Research and Development Command Technical Report No. 1331, 19 (unpublished). Instability of the Block Walls due to interstitial atoms in a ferromagnetic lattice.

[11] P. Weiss and J. de Freudenreich, Arch. Sci. Phys. Mat. **42**, 5 (1916) and **42**, 449 (1916).

[12] H. Fahlenbrach, Ann. Phys. (6) **2**, 355 (1948).

[13] H. Fahlenbrach and G. Sommerkorn, Tech. Mitt. Krupp **15**, 161 (1957).

[14] G. Biorci, A. Ferro, and G. Montalenti, *Congrès International sur la Physique de l'Etat Solide et ses applications à l'electronique et aux Telecommunications, Brussels, June 1958* (Academic Press, London, 1960).

[15] G. Biorci, A. Ferro, and G. Montalenti, Air Research and Development Command Technical Report No. 1131, March, 1958 (unpublished).

wound on a steatite spool. The dimensions of the spool are as follows: rectangular cross section 16×35 mm^2, length 150 mm. The voltage induced in the secondary coil is rectified and recorded.

From the curves of induced voltage vs time it is easy to deduce the magnetization curves $J_0(H)$ and $J_\infty(H)$ corresponding to $t=0$ (immediately after demagnetization), and to $t=\infty$. (taken 30 minutes after demagnetization, since thereafter the induced voltage remains practically constant).

According to Néel[5,6] the maximum value of the horizontal distance between J_0 and J_∞ is defined as the viscosity field, and it is taken as the measure of the intensity of the after-effect since it can be directly related to the cause of the viscosity. The viscosity field is nothing but the additional field to be applied in order to move the walls when they have become fixed in certain positions by diffusion of some type of defects. Hence the viscosity field is the physically most significant quantity for measuring a magnetic after-effect. Actually, the definition of Néel[5,6] refers to the curves $J_0(H)$ and $J_\infty(H)$ taken under static conditions, but, as shown previously,[15] the dynamic procedure gives essentially the same results.

The specimens were tested under different conditions; annealed, plastically bent with a strain of 0.16% and cold rolled to 1 and 2%. In the case of the deformed specimens, before starting the measurements, we partially recovered the cold work by keeping the specimens for some time at about 20°C above the maximum

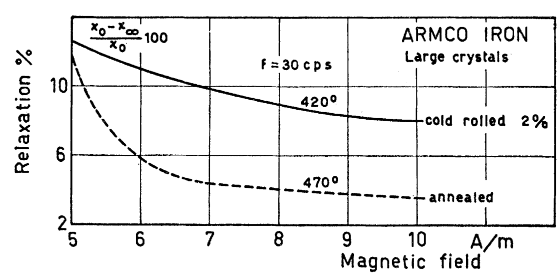

FIG. 1. Relative change of susceptibility (in percent) vs magnetic field, at various temperatures, of an Armco iron specimen. Broken lines: annealed; solid lines: cold rolled 2%. χ_0 taken 10 sec after demagnetization. χ_∞ 30 min later. Frequencies of the measuring field 30 cps.

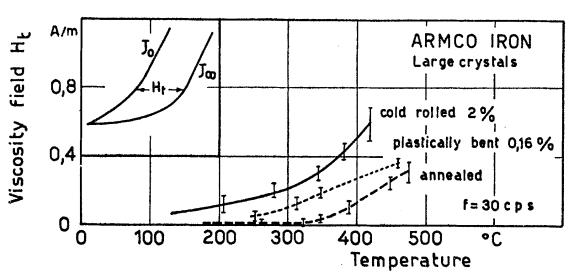

FIG. 2. Viscosity field vs temperature of an Armco iron specimen. Broken line: annealed; solid line: cold rolled 2%; dotted line: plastically bent 0.16%. Frequency of the measuring field: 30 cps.

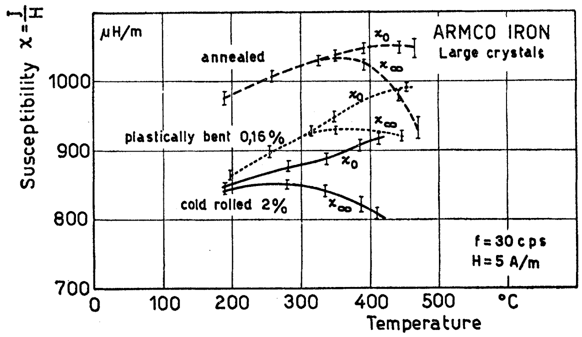

FIG. 3. Susceptibility measured 10 sec after demagnetization (χ_0) and 30 min after demagnetization (χ_∞), of an Armco iron specimen. Broken lines: annealed; solid lines: cold rolled 2%; dotted line: plastically bent 0.16%. Frequency of the measuring field: 30 cps.

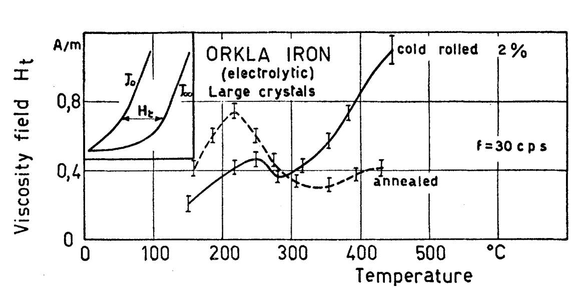

FIG. 4. Viscosity field vs temperature of an electrolytic Orkla iron specimen. Broken line: annealed; solid line: cold rolled 2%. Frequency of the measuring field: 30 cps.

temperature of operation. This temperature is slightly lower than the temperature of complete recrystallization. In this way the specimen becomes stabilized and effects of equal intensity can be observed many times at all temperatures below the maximum one. Moreover after having taken the measurements on a plastically deformed material, we annealed again the specimen. The curves of the viscosity field, in all cases, assumed again the original values of the annealed condition within the experimental errors.

The results obtained on Armco iron are given in Figs. 1, 2, and 3, and those obtained on the high-purity electrolytic iron in Figs. 4, 5, and 6. The measurements have been repeated several times and the data shown in the figures are average values. The internal friction and the elastic modulus of the same specimens were also measured at frequencies close to 1 cps. as a function of temperature (Fig. 7).

The results of the magnetic measurements can be summarized as follows. The viscosity field, in annealed single crystals of Armco iron, becomes appreciable at about 320°C and increases rapidly with temperature, reaching the value of about 0.3 amp-turn/m at 450°C (Fig. 2). Similar results are obtained on the electrolytic iron. In this case however an additional viscosity peak at 220°C is observed (Fig. 4). This peak has been found also on other samples of electrolytic iron produced in our laboratory.[14,15]

We have not yet been able to understand the origin of this peak: however if we subtract this peak from the curves of the viscosity field, we again get a curve, which increases with temperature and with the amount of cold working, in agreement with the measurements on the Armco specimens. In specimens plastically bent by 0.16% the effect of cold work on the value of the viscosity field is appreciable only in some specimens (Fig. 2), probably because the deformation is small.

INTERPRETATION OF THE RESULTS

It can easily be shown that the dislocations present in a ferromagnetic crystal exert an influence on the motion of the Bloch walls: in fact the magnetoelastic energy of the crystal changes when the walls move in the dislocations stress field.

These interactions have been investigated by Vicena[16] in his theory of the coercive force. In the simple case of a single edge dislocation parallel to a 180° Bloch wall, he has shown that the interaction energy is constant when the dislocation line is outside the wall thickness, but across the wall there is an energy step ΔW_1 pro-

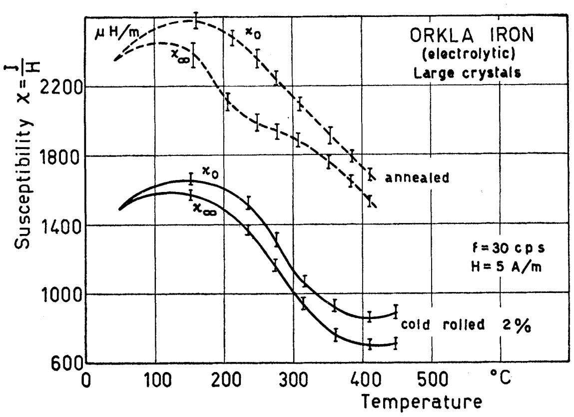

FIG. 5. Susceptibility measured 10 sec after demagnetization (χ_0) and 30 min after demagnetization (χ_∞), of an electroylytic Orkla iron specimen. Broken lines: annealed; solid lines: cold worked 2%. Frequency of the measuring field: 30 cps. Magnetic field 5 amp-turn/m.

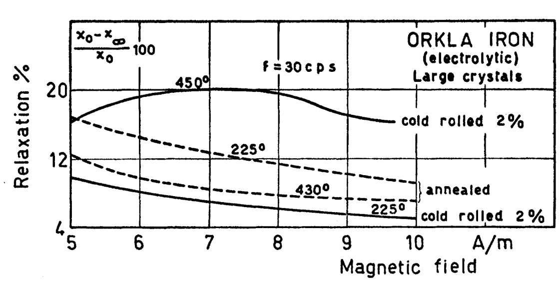

FIG. 6. Relative change of susceptibility (in percent) vs magnetic field, at various temperatures, of an Orkla iron specimen. Broken lines: annealed; solid lines: cold rolled 2%. χ_0 taken 10 sec. after demagnetization. χ_∞ 30 min later. Frequencies of the measuring field 30 cps.

[16] F. Vicena, Czechoslov. J. Phys. 5, 4 (1955).

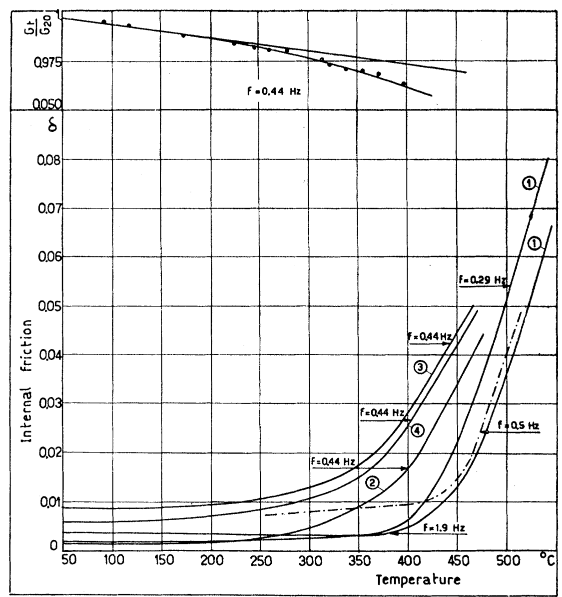

FIG. 7. Relative decrease of shear elastic modulus G with temperature for a specimen of electrolytic iron cold worked 0.5% (corresponding to the curve *n.* 4 of the internal friction). The relaxation to be considered is the difference from the linear decrease. Internal friction versus temperature: 1 Annealed Armco iron, large crystals. 2 Annealed electrolytic iron, large crystals (Orkla Met.). 3 Same as 2 but cold worked 0.5%. 4 Same as 3 after partial recovery at 430°C. dotted line: curve obtained from Kê (see reference 21).

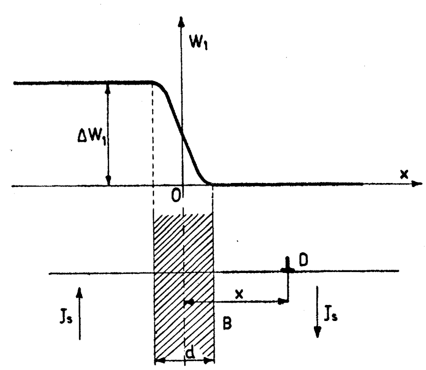

FIG. 8. Interaction energy W_1 of a dislocation D with a Bloch wall B as a function of the distance x between the dislocation line and the middle plane of the wall.

portional to λGbd, λ being the magnetostriction constant, G the shear modulus, b the Burgers vector and d the wall thickness (Fig. 8). ΔW_1 refers to one centimeter of wall, measured in the direction of the dislocation line. In other words the dislocations contained inside the Bloch walls undergo a pressure which tends to push them outside.

Therefore, if we demagnetize the specimen at a sufficiently high temperature, the dislocations which are inside the Bloch walls tend to move into the bodies of the domains. Hence, when we apply an external field, after having allowed the dislocations to displace them-

TABLE I. Values of H_{cr} and H_t.

Specimen	disl./cm^2	H_{cr} (theor.) amp-turn/m	H_t amp-turn/m $\Delta r/r=0{,}1$	$\Delta r/r=1$
Fe annealed	10^8	1.6	0.08	0.8
Fe	10^9	5	0.25	2.5
Fe cold worked 2%	10^{11}	50	2.5	25

selves by diffusion, the walls move into regions with a density of dislocation slightly higher than before. This additional hindrance to their motion is a viscosity field. A quantitative computation of the value of the viscosity field due to dislocations is obviously rather difficult. However an order of magnitude can be roughly estimated.

The theory of Vicena,[16] by proper use of simplifying assumptions, leads to the following expression for the coercive force:

$$H_{cr}=(\Delta W_1/2J_sL)(\ln L/d)^{\frac{1}{2}}(r)^{\frac{1}{2}}, \tag{1}$$

where L is the linear size of the domains, J_s the saturation magnetization, and r the dislocation density in disl./cm^2.

This formula has been experimentally verified by Vicena[16] and later by Malek[17]: the agreement with the experimental values turned out to be quite satisfactory.

By analogy with the results of Brissonneau,[18] on the hysteresis loops of iron loaded with carbon, we can assume that the viscosity field H_t is of the order of the variation of the coercive force [given by Eq. (1)] as the dislocation density increase by a small amount Δr. According to this assumption, putting $r+\Delta r$ in place of r in Eq. (1) and developing in power series, we obtain

$$H_t=(\Delta r/2r)H_{cr}. \tag{2}$$

The value of H_{cr}, in the case of iron, can be deduced from Eq. (1) by taking $\Delta W_1=0.35\times10^{-5}$ erg/cm (from Vicena[16]); $L=10^{-3}$ cm; $d=10^{-5}$ cm; $J_s=1710$ e.m.u. and for r the values given by Cottrell[19] on annealed and cold worked metals: i.e., respectively: about 10^8 disl/cm^2 for the annealed specimen and 10^{11} for the cold worked specimen.

As far as $\Delta r/r$ is concerned only a rough estimate is possible. The maximum value of $\Delta r/r$ is obviously 1. This corresponds to displacing all the dislocations contained inside the walls into adjacent regions of equal thickness. However, within the time interval of the experiment the dislocations do not move as far as a wall thickness (1000 A) since their movement is opposed by many hindrances (anchoring centers, interaction among dislocations and so on). Therefore at a temperature of about 400°C one can reasonably believe that $\Delta r/r$ is certainly much less than 1.

[17] Z. Malek, Z. angew. Phys. **9**, 279 (1957).

[18] P. Brissonneau, J. Appl. Phys. **29**, 249 (1958).

[19] A. H. Cottrell, *Theory of dislocations* (Oxford University Press, New York, 1958), pp. 102, 154.

Taking the values of 0.1 and 1 for $\Delta r/r$, one arrives at the values of H_t given in Table I. Another method for obtaining a rough estimate of the order of magnitude of the effect is given by a simple reasoning about the relaxation of the magnetostrictive stresses in the wall. It is known[20] that the magnetostrictive strain energy per unit surface of a 180° wall in iron of the order of:

$$(9/4)\lambda_{100}^2(c_{11}-c_{12})d/2, \tag{3}$$

where λ_{100} is the magnetostriction and c_{11}, c_{12} are the elastic constants of the single crystal.

If a certain fraction of dislocations is present within the wall and if it moves toward the outside, the magnetostrictive strain energy of the wall relaxes accordingly. At a given temperature the strain relaxation due to dislocation movement may be measured by the relaxation of elastic moduli $\Delta G/G$ observed in an internal friction measurement.

In fact, after the magnetostrictive stresses have relaxed in the region occupied by the wall, the wall itself will be in a position of minimum energy and a definite value of the field must be applied to pull it out of this position. This maximum value of the field is nothing else than the viscosity field.

Hence from Eq. (3) and taking into account the relaxation of the elastic moduli $\Delta G/G$, the maximum value of the viscosity field will be of the order of:

$$H_{t\max}=(1/2J_s)(9/4)(c_{11}-c_{12})\lambda_{100}^2\cdot\Delta G/G. \tag{4}$$

In fact this maximum value of the field will be reasonably reached for wall displacements of the order of $d/2$. In the case of iron this corresponds to a magnetization of about ten gauss as an order of magnitude. Above this value the viscosity field should remain rather constant for the reasons already given in the case of interstitial carbon in iron.[9,10]

By introducing in Eq. (4) the values of the magnetostriction of iron (20×10^{-6}) and a relaxation of elastic moduli of the order of 2%, as observed in a single crystal of iron at about 400°C (see Fig. 7), we obtain

$$H_{t\max}=0.8 \text{ amp-turn/m},$$

which is of the right order of magnitude as the experimental values.

COMPARISON WITH THE EXPERIMENTAL RESULTS

A careful examination of the curves given in Figs. 1 to 7 leads to the conclusion that the observed viscosity fields are due to dislocation movement according to the interpretation given in the preceding section. Firstly, the viscosity field in single crystals increases with the amount of cold work, and is of the same order as those tentatively computed above (Figs. 2 and 4). Secondly, the relaxation decreases with increasing field strengths (Figs. 1 and 6): this shows that the viscosity is due to

[20] C. Kittel, Revs. Modern Phys. **21**, 541 (1949).

a diffusion of defects contained within the domain walls,[6,10] as we have assumed in our interpretation of the effects.

Further proofs of the correctness of our interpretation are given by a comparison of the magnetic results with the internal friction curves. Our data on internal friction (Fig. 7) agree with those reported by others,[21,22] and correspond to other evidences of the dislocation movement. The shape of the curves of the viscosity field vs temperature appears to be analogous to that of internal friction. After a slight cold work the viscosity field increases, as the internal friction does, and becomes already appreciable at a low temperature ($\sim 200°C$).

Unfortunately the relaxation spectrum of the magnetic after-effect is very broad and hence it is impossible to determine the actual diffusion constants on which the after effect depends. However, at least in the case of annealed Armco iron (Fig. 2), we can obtain some rough information by comparing the temperature above which the magnetic viscosity increases rapidly with the temperature at which the internal friction increases rapidly too. From the internal friction measurements taken at two frequencies (Fig. 7), in agreement with other authors,[21,22] we obtain that in the region around 450°C the activation energy of the effect is about 80 000 cal/g atom, assuming that the relaxation times follow the simple law

$$\tau = \tau_0 \exp(Q/RT).$$

By using this value for Q and taking into account that the magnetic viscosity measurements have a relaxation time of the order of 3 min while the internal friction is measured dynamically at a frequency of the order of 1 cps, we obtain that the knee above which the internal friction increases corresponds to the knee in the magnetic viscosity curve as a function of temperature.

[21] T. S. Kê, Metals Technol. T. P. 2370, 448 June (1948).

[22] T. S. Kê, Trans. Am. Inst. Mining, Met. Petrol. Engrs. **188**, 575 (1950).

As far as the results on electrolytic iron are concerned, the higher values of viscosity field observed could be attributed to the higher mobility of dislocations in a very pure metal.

The decrease of the initial permeability at the temperature at which the viscosity field due to dislocations is observed, is a direct consequence of the presence of the viscosity effect itself (Figs. 3 and 5). In fact, the effect is identical to that observed in the case of interstitial solutions and already explained by Snoek.[8,23] However, since in this case the relaxation spectrum is broad, the decrease in permeability observed is generally more intense than that actually measurable in a relaxation experiment at a given temperature, because the time interval of measurement can never be very long.

However it can be seen that the dislocations, together with the grain boundaries and the atoms in solutions may be one of the causes of the anomalies in the initial permeability observed in the high-temperature range.

CONCLUSION

In the present paper we have made a first attempt to find a magnetic after-effect due to dislocation movement and to correlate this effect with the results of internal friction measurements.

The density of dislocations in single crystals of iron has been varied by cold working. A magnetic after-effect, increasing with the amount of cold work, has been observed. The temperatures at which the effect is observed are consistent with the measurements of internal friction.

A semiquantitative interpretation of the effects has been given: the observed magnetic after-effect can be explained as a relaxation of the magnetostrictive stresses due to the interaction among domain walls and dislocations.

[23] G. W. Rathenau, *Seminar on the Magnetic Properties of Metals and Alloys* (American Society for Metals, Cleveland, 1959), p. 168.