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The Analysis of Magnetization Curves

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By an analysis of a magnetization curve is meant the determination of the character and extent of the various elementary changes occurring over any part of the curve. Even if the quantitative characteristics of the various elementary processes are known, an analysis cannot in general be made simply on the basis of the information provided by a given magnetization curve, owing to the varied and complicated way in which the different processes are superposed with an ordinary polycrystalline specimen. Further information is obtainable from measurements such as those of Bates on the thermal changes accompanying adiabatic magnetization, but little progress can be made in the analysis of curves in the hysteresis range unless the magnetic and thermal effects due to the reversible and irreversible changes separately can be unambiguously determined.

A broad survey of the general problem is given with particular reference to recent theoretical and experimental work at Leeds. The theoretical approach and the methods of analysis are along the same lines as in the treatment of the magnetocaloric effect by Stoner and Rhodes (1949). The experimental work reviewed includes studies of the reversible susceptibility, the Barkhausen effect, and the reversible change of magnetization with temperature at constant field (Tebble, with Corner, Newhouse, Skidmore, Wood, and others, 1948 onwards).

Through this work, it is now possible to determine not only the relative contributions from reversible and irreversible processes to the change of magnetization over any range but also the thermal changes associated with each. The part of the reversible change due to change in intrinsic magnetization can be determined, and in many cases analysis will show whether the remainder of the reversible change is mainly due to rotation of the magnetization against the crystalline and strain anisotropy forces, or to other processes such as boundary movements. Much detailed information about irreversible processes is obtained from determinations of the number-size relations of Barkhausen discontinuities, which in general correspond not to spontaneous movements of primary domain walls as wholes, but to smaller scale sectional irregularities in the motion. The thermal effect associated with these irreversible processes may be a heating or a cooling.

Partial analyses are presented for annealed nickel, hard-drawn iron, and other materials. These show clearly the potentiality of the general method for obtaining detailed quantitative information about the elementary processes contributing to the change of magnetization along a magnetization curve.

I. INTRODUCTION

THE magnetization curves of ordinary polycrystalline ferromagnetic materials present a complicated problem, or group of problems, and it is still far from possible to give a complete detailed explanation of their forms and quantitative characteristics, although during the past twenty-five years great progress has been made. On the one hand, the essential characteristics of ferromagnetic materials have received a satisfying explanation within the general framework provided by quantum-mechanical principles. On the other, the various secondary effects which determine the form of a magnetization curve have been intensively studied, and a better knowledge, if not always understanding, has been gained of the various types of elementary process by which change of magnetization may occur.

Three main types of elementary process contribute to the change in apparent magnetization: change of intrinsic magnetization, that is, of the orientational distribution of the elementary carriers of the magnetic moment, within each uni-directionally magnetized region, or domain; rotation of the magnetization vector in such a region as a whole against anisotropy forces; and movement of the boundaries of these regions. Elementary changes of the last two types may be reversible or irreversible.

With the wealth of information now available about elementary processes it might seem that a careful consideration of the quantitative characteristics of a given magnetization curve of a particular material could result in fairly definite conclusions about the nature, sequence, and relative proportions of the elementary changes contributing to the observed changes in magnetization. In fact, except for very restricted ranges of a magnetization curve, little more than a rough guess can be made as to the relative contributions from the different types of elementary process. This paper is concerned with the general problem of the "analysis" of magnetization curves with a view to obtaining more definite and more complete information about the elementary processes taking place. Without further information than is provided by the magnetization curve itself (together with a knowledge of the essential magnetic constants of the material) it is not possible to determine with any certainty which elementary processes are mainly contributory to the observed changes in magnetization. The subject matter is therefore in part experimental, in that a brief account is given of some of the recent and current experimental investigations which provide additional material which can contribute to such analyses; and, in part, in a broad sense, theoretical, in that the methods by which the experimental material may be utilized in analyses are indicated, and the thermodynamic and other arguments involved are outlined.

It will be obvious that a very wide range of work is potentially relevant to the general theme. In a contribution to a symposium, however, it is proper that the writer should present primarily an account of work with which he has been immediately associated, and this paper is accordingly largely concerned with recent work at Leeds, published and unpublished, to which specific references will be restricted. This, however, is not intended to ignore other work, much of which has been fully surveyed in a recent comprehensive review article[1] on magnetization curves. That article makes it unnecessary to refer, except incidentally, to the general background or the wider bearing of the work here considered.

The information additional to that given directly by magnetic measurements which is thought to be of the greatest value in analyzing magnetization curves is that which can be provided by studies of the thermal changes accompanying magnetization. Hysteresis losses and the magnetocaloric effect in high fields are special examples of these thermal effects, but they have usually been considered in isolation. The determination of the temperature changes accompanying adiabatic magnetization in low and moderate fields presents a difficult experimental problem which has been successfully attacked only in fairly recent years, notably by Bates and his collaborators at Nottingham. The extraction of information in the most convenient form from the experimental results, and the planning of these and related experiments, call for careful consideration of the thermodynamics of magnetization, which, as the general problem is approached here, is of central importance in the analysis of magnetization curves.

After this introduction, a few of the more important of the relevant thermodynamic relations are stated, with a short commentary (II), and a summary is given of the characteristics of the main elementary magnetization processes (III). Some recent experimental investigations on magnetization curves are then outlined, first the more purely magnetic (IV), and next the magnetothermal investigations (V). The combination of the results of different experiments in analyzing magnetization curves is considered, and some illustrative results are presented (VI). Finally, the requirements, possibilities, and limitations of the general method are considered in a summarizing discussion (VII).

II. THE THERMODYNAMICS OF MAGNETIZATION

The thermodynamics of magnetization has been developed in a number of papers[2] in a form which is convenient when the "system of interest" is a particular magnetizable body in an external field. The most useful relations for the present purpose are given in a recent paper[3] on magnetothermal effects in ferromagnetics. Here only a few essential equations will be given with a brief commentary.

A starting point is the equation for the increase of total energy E_t associated with a magnetizable body, with increase in the external field H_e, namely,

$$dE_t = dQ - IdH_e, \tag{1}$$

where I is the volume intensity of magnetization, dQ the heat transferred to the body, and E and Q refer to unit volume. The total energy includes the potential energy of the body in the external field $-IH_e$, and also the demagnetizing field energy $+\frac{1}{2}N_D I^2$, where N_D is the demagnetization coefficient. When these are subtracted from E_t, remembering that the demagnetizing field H_i is equal to $-N_D I$ and that the "corrected" field H is equal to $H_e + H_i$, the following defining equation is obtained for an internal energy function E:

$$dE = dQ + HdI. \tag{2}$$

This equation is in a form convenient for further development. It has the obvious advantage that the energy E and the I, H relations are characteristic of the material, and are not, like the I, H_e relations, dependent on the shape of the specimen. (It may be noted that it is by no means always possible to allow for shape effects by a simple "correction" of the external field, but it is not necessary to consider this particular complication here.)

Equation (2) is applicable whether or not the changes are reversible. A change of energy ΔE may be divided into two parts, a "thermal" part ΔE_T associated with change of temperature, and a magnetic part ΔE_M, associated with change of magnetization. For an adiabatic change ($\Delta Q = 0$), it follows that

$$\Delta E = \Delta E_T + \Delta E_M = \int HdI. \tag{3}$$

It is often convenient to consider, instead of the change of temperature ΔT, with adiabatic change of field, the corresponding "heat developed," $\Delta Q'$ (not to be confused with ΔQ), related to ΔT by

$$\Delta Q' = C\rho\Delta T, \tag{4}$$

where C is the specific heat and ρ the density. Thus, in an adiabatic change,

$$\Delta E_M = \int HdI - \Delta Q'. \tag{5}$$

The change of internal energy associated with a change of magnetization along a magnetization curve can therefore be determined if measurements are also made of the thermal effect accompanying adiabatic magnetization.

For reversible changes, (2) may be written as

$$dE = TdS + HdI, \tag{6}$$

where S is the entropy. Over a reversible range the state is determined uniquely by T or S and H or I, and three other thermodynamic functions may be formed in the usual way. One of these, $E' = E - HI$, is of the energy type, and two, $F = E - TS$ and $F' = E' - TS$, are of the free energy type. From (6) and the corresponding equations for E', F, and F', four reciprocal relations of the

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[1] E. C. Stoner, Repts. Prog. Phys. **13**, 83 (1950).

[2] E. C. Stoner, Phil. Mag. **19**, 565 (1935); **23**, 833 (1937); Trans. Roy. Soc. (London) **A235**, 165 (1936).

[3] E. C. Stoner and P. Rhodes, Phil. Mag. **40**, 481 (1949).

Maxwell type are at once obtainable. Among the many relations which may be derived, the following are the most useful in the present connection:

$$\left(\frac{\partial T}{\partial H}\right)_S=-\left(\frac{\partial I}{\partial S}\right)_H=-\frac{T}{\rho C_H}\left(\frac{\partial I}{\partial T}\right)_H=-\frac{T}{\rho C_H}\left(\frac{\partial S}{\partial H}\right)_T; \quad (7)$$

$$\left(\frac{\partial T}{\partial I}\right)_S=\left(\frac{\partial H}{\partial S}\right)_I=\frac{T}{\rho C_I}\left(\frac{\partial H}{\partial T}\right)_I=-\frac{T}{\rho C_I}\left(\frac{\partial S}{\partial I}\right)_T. \quad (8)$$

In the applications to be made, the specific heats at constant field C_H and at constant magnetization C_I may both be taken, with negligible error, as equal to the specific heat C, as ordinarily measured in zero field.

The value of these relations will only become apparent in subsequent sections, but it will be clear at once that over ranges in which the changes of magnetization are reversible, there is full scope for the application of thermodynamic reasoning. Over ranges in which irreversible changes occur, that is, for ferromagnetics, in low and moderate fields, the information which can be derived from thermodynamic considerations is much more limited. Broadly speaking, conclusions can be drawn about energy changes but not about entropy changes.

The difficulties in the application of thermodynamics to the magnetic behavior of ferromagnetics do not arise simply because of the occurrence of irreversible processes. For many systems which pass irreversibly from an initial to a final state (e.g., a gas expanding into a vacuum) it is possible to find the change of entropy by devising a reversible path for the passage from one state to the other. For ferromagnetics the alternative reversible path may indeed be difficult to devise, but this is connected with an even more fundamental difficulty. This is that the state of the system is not uniquely specified by the parameters, say H and I, giving the position of the representative point on the magnetization curve. The state so specified is not in general an equilibrium state, nor is it a metastable state of a simple kind, and it is compatible with a vast number of different small scale "patterns." The macroscopic state of the system (a particular ferromagnetic specimen) corresponds to the aggregate of the stable and metastable states of the constituent groups of units, that is, domains large and small, which form a pattern ever changing as the magnetization changes. The scale of this pattern falls in an awkward range. If it were larger, the methods of thermodynamics would be more readily applicable; if it were smaller, those of statistical mechanics would apply.

III. ELEMENTARY MAGNETIZATION PROCESSES

A ferromagnetic may be regarded as made up of domains $1, 2, \cdots j, \cdots$, of volumes $v_1, v_2 \cdots v_j \cdots$, each of which, at temperatures well below the Curie point θ, has an intrinsic magnetization I_m approximating closely to the quasi-saturation magnetization. The value of I_m varies with temperature and field. It may be well to note at once that though the change with field may be small (e.g., for nickel at room temperature the difference between I_m for $H=10^4$, and I_m for $H=0$ is less than 1 in 200) the associated energy change may be of predominant importance. The apparent magnetization, as ordinarily measured, may be written

$$I=\sum_j v_j(I_m)_j\cos\phi_j, \quad (9)$$

where ϕ_j is the angle between $(I_m)_j$ and I, and the summation is over the domains in unit volume ($\sum v_j=1$). Change of I may therefore be expressed as a resultant effect of changes in I_m, ϕ, and v:

$$dI=\sum_j\{v_j\cos\phi_j d(I_m)_j-v_j(I_m)_j\sin\phi_j d\phi_j+(I_m)_j\cos\phi_j dv_j\}. \quad (10)$$

The three terms correspond to the following three elementary processes: (a) change of intrinsic magnetization, (b) rotation of the domain magnetization vector, and (c) boundary movement. The relative magnitudes of the contributions to the change of I in passing from the demagnetized state to quasi saturation are ordinarily in the order c, b, a, while the associated energetic changes are in the order a, b, c. The effective utilization of the results of magnetothermal measurements in the analysis of magnetization curves is largely dependent on the extent to which this qualitative statement can be made quantitative and precise. A full discussion of the various elementary processes would require a book, and the points mentioned here are limited to those which have a direct bearing on the problem of analysis. It will be sufficient for the present purpose to make the statements in a form applicable to ferromagnetics well below the Curie point (e.g., iron, nickel, or cobalt at room temperature).

Change in Intrinsic Magnetization

It is not commonly appreciated that there is no precise knowledge of the variation of intrinsic magnetization with field even in high fields. In the quasi-saturation range the variation of the apparent magnetization may be represented empirically by

$$I=I_0\left(1-\frac{a}{H}-\frac{b}{H^2}\right)+cH, \quad (11)$$

but it is fair to say that there are at present no experimental results from which the value of c in the term in H, corresponding to increase in intrinsic magnetization, can be determined with any certainty. Indirect methods are therefore necessary to determine the effects associated with changes in intrinsic magnetization.

The most promising procedure is to make use of (7), which in conjunction with (4) gives for the heat developed in a reversible adiabatic magnetization process

$$\left(\frac{\partial Q'}{\partial H}\right)_S=-T\left(\frac{\partial I}{\partial T}\right)_H. \tag{12}$$

As the variation of the intrinsic magnetization I_m over the field range from 0 to several thousand oersteds is small, the variation with temperature is given nearly enough over the whole range by $(\partial I_0/\partial T)$, where I_0 is the spontaneous magnetization in zero field. This quantity is known, or determinable, with considerable accuracy. The sorting out of the contribution $(\Delta Q')_i$ from the change in intrinsic magnetization to the aggregate heat developed, $\Delta Q'$, along a section of a magnetization curve involves a rather tricky argument. Except in high fields the local directions of I_m are not in general parallel to H, but are distributed in a manner which gives the resultant bulk magnetization I; further, there are changes of the local I_m accompanying the changes of local directions with H. The final result of the argument[3] is, however, simple. The contribution is given by

$$(\Delta Q')_i=a\int d(IH), \tag{13}$$

where $a=-(T/I_0)(\partial I_0/\partial T)$. The relation (13) is believed to hold generally, whether or not the changes along the magnetization curve are reversible; for the changes in intrinsic magnetization themselves are, unlike rotational changes and boundary movements, always reversible. Since $(\partial I_0/\partial T)$ is negative, increase of intrinsic magnetization in an adiabatic change of field is accompanied by heating. In terms of entropy, this arises from the decrease in magnetic entropy.

The situation is somewhat peculiar in that although only the roughest estimate may be made of the change of intrinsic magnetization with field, the corresponding heat developed in an adiabatic change of field may be accurately determined. It has long been known that the magnetocaloric effect in high fields is almost entirely due to change in intrinsic magnetization. From a quantitative application of (13) it turns out that even in low fields $(\Delta Q')_i$ may account for the greater part of the observed $\Delta Q'$. The relation (13) is therefore of central importance in the effective utilization of magnetothermal results in the analysis of magnetization curves.

Rotational Processes

Change of magnetization due to rotation of the domain magnetization vectors may occur to some extent over any part of a magnetization curve, but it is usually predominant between the knee of the curve and technical saturation (or between saturation and remanence). As the field increases rotation takes place from "easy" directions determined by the local anisotropy towards the direction of the field. Extensive experimental and theoretical investigations, fully reviewed elsewhere,[1] have been made of natural crystalline anisotropy, of anisotropy due to strain and, with particular reference to single-domain particles, of anisotropy due to shape. To avoid complication of detail in dealing with principles it will be convenient to fix attention on crystalline anisotropy, and to suppose that the main anisotropy characteristics are determined by a single coefficient whose modulus is symbolized by k (e.g., for nickel, $k=|K_4|$, $K_4\simeq-4.9\times10^4$ erg cm^{-3}). When k is constant throughout a specimen, the magnetization curve is, in principle, calculable if the orientational distribution of the constituent crystals is known. Whether it is or not, however, a straightforward argument[3] gives as an expression for the heat developed from rotational changes $(\Delta Q')_k$ in an adiabatic change of field over a reversible rotational range,

$$(\Delta Q')_k=b\int HdI, \tag{14}$$

where $b=(T/k)(dk/dT)$. Since (dk/dT) is usually, if not invariably, negative, increase of magnetization by rotations in an adiabatic change of field is accompanied by cooling. This corresponds to an increase in the magnetic entropy. The range of validity of (14) is much more restricted than that of (13). It holds only over a range in which the change of magnetization is reversible, and due entirely (apart from the small change in intrinsic magnetization) to rotations of the local magnetization vectors. The value of b which can be calculated from the single crystal data (when these are available) can be expected to apply closely to a polycrystalline specimen only if this is made as free as possible from internal strains by a thorough annealing treatment.

In general the value of k and of dk/dT will be modified by local strains and possibly other irregularities. Owing to the complexity of the internal stress distribution problem, and also, incidentally, to the paucity of fundamental data on crystal magnetostriction coefficients and their temperature variation, it is virtually impossible at present to make even rough *a priori* estimates of the effective value of b (and of its possible variation with field) for unannealed specimens.

Under certain conditions rotational changes may take place irreversibly. Irreversible rotational changes have been very fully studied in relation to the interpretation of the properties of certain types of high coercivity material in terms of the behavior of single domain particles.[4] It is interesting to note that precise thermodynamical calculations for a complex irreversible magnetization process can be made, when, as here, the mechanism of the process is known.[5] In "ordinary"

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[4] E. C. Stoner and E. P. Wohlfarth, Trans. Roy. Soc. (London) **A240**, 599 (1948).

[5] P. Rhodes, Proc. Leeds Phil. Soc. **5**, 116 (1948).

materials, however, irreversible processes of this kind are unlikely to be important, and they will not therefore be discussed in this paper.

Boundary Movement

There is now no doubt that in ordinary unstrained polycrystalline material nearly the whole of the change of magnetization in the lower field range may be attributed to reversible or irreversible movements of domain boundaries. In recent years extensive investigations have been made on domain structure by the powder pattern method (Williams, Bozorth, Bates, and others), and the general character of the beautiful patterns obtained is now well known. In one simple (though experimentally very difficult) case, with a "picture-frame" crystal, the change of magnetization has been quantitatively correlated with the observed boundary movement (Williams and Shockley). On the theoretical side, boundary energies and widths have been studied (Néel, and more recently Lilley[6]) and also the dynamics of the boundary movements (Néel, Kittel, and others). Most of this work has been fully reviewed elsewhere.[1] Although so much more is now known than, say, ten years ago, there are still many gaps and uncertainties. For example, fundamental difficulties are encountered in attempting to make any but the roughest theoretical estimates of the absolute value of boundary energies, and experimental estimates are indirect and uncertain; and even for simple domain patterns, whose form is in accordance with theoretical calculations, the spacings as measured often differ by a factor of two or more from those given by the unavoidably somewhat idealized theory.

The complexity in detail of the powder patterns obtained when the conditions are other than the very simplest (i.e., with single-crystal specimens and with the surfaces examined close to well-defined crystallographic planes containing at least one easy axis) suggests that theoretical treatments of ordinary magnetization curves, in the boundary movement ranges, must at present, to be useful, be of a somewhat general type rather than of a type which takes cognizance, explicitly, of the details of the elementary processes involved.

Usually, over the initial part of a magnetization curve, the magnetization is reversible over a limited range, and

$$I=\kappa_0 H.$$

It follows at once from (4) and (7) that over this range

$$\Delta Q'=a'\int d(IH)=b'\int H dI, \qquad (15)$$

where $a'=-\frac{1}{2}(T/\kappa_0)(d\kappa_0/dT)$, $b'=2a'$. Since $(d\kappa_0/dT)$ is usually positive, adiabatic increase of field is accompanied by cooling, as for rotational changes. In low fields boundary movement results in a change of direction of magnetization vectors from one easy direction to another, more favorably orientated with respect to the field. The effects indicated by (15) may be in part attributed to slight differences in the energy associated with orientations along different ideally equivalent easy axes (so that the atomic interpretation of the effects would be the same as for ordinary rotational changes), and in part to the hindrances to boundary movement due to local irregularities. There is no generally applicable method of making *a priori* calculations of the coefficients in (15).

Beyond the very limited initial reversible range, reversible and irreversible boundary movements are superposed. Recent experimental work on the determination of the reversible susceptibility over the whole of the magnetization curve will be described in the next section. From such a determination, in conjunction with the magnetization curve, it is possible to find the irreversible contribution to the change of magnetization, that is, in ordinary materials, the contribution from irreversible boundary movements. Information about the character of the elementary irreversible changes can be obtained from experiments on the Barkhausen effect, which are also described. It will be convenient, therefore, to give further consideration to reversible and irreversible boundary movements in the next section rather than here.

IV. EXPERIMENTAL INVESTIGATIONS: MAGNETIC

With a view to illustrating the kind of "supplementary" magnetic investigations which are of value in the analysis of magnetization curves, a brief outline will be given in this section of some recent experimental work at Leeds by Tebble and others on reversible susceptibility and the Barkhausen effect. When it became possible to include experimental work on ferromagnetism in the research program of the Physics Department at the University of Leeds some few years ago, the study of the Barkhausen effect was an almost obvious choice, for little work was being done on it elsewhere, and indeed comparatively little had been done on the ordinary effect since the classical investigations of Bozorth and Dillinger (1929–32), which left many questions unanswered. In connection with this work, it became necessary to measure reversible susceptibilities, for which improved methods were developed. For the kind of analysis in view here, reversible susceptibility measurements are of more direct importance than those on the Barkhausen effect, and they will therefore be considered first.

Reversible Susceptibility

It is well known that the differential suceptibility, as measured by the slope of the magnetization curve at a particular point, may be separated into reversible

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[6] B. A. Lilley, Phil. Mag. 41, 792 (1950).

and irreversible components such that

$$\kappa_d = \kappa_r + \kappa_{\mathrm{irr}}. \tag{16}$$

The conventional definition of κ_r corresponds to

$$\kappa_r = \lim_{\Delta H \to 0} (\Delta I / \Delta H), \tag{17}$$

where the field increment ΔH is in the *opposite* direction to the last increment of the field bringing the specimen to the point (I, H). Experimental methods for measuring κ_r based directly on (17) are tedious and inaccurate. It is usually supposed that the same reversible susceptibility is measured by the use of alternating fields. The methods developed by Tebble and his collaborators (1950 onwards) involve essentially the measurement of the mutual inductance of two coaxial coils with and without the specimen under test as a core. Measurements were made with various amplitudes of the alternating field. Over most of the steady field range the limiting value of κ_r for the materials examined did not differ significantly from that for an amplitude of the order 10^{-3} oersted, which was ordinarily used in the work.

In the first method the mutual inductance was measured by an inductometer bridge arrangement, which, with amplification of the out-of-balance current, gave an over-all accuracy in measurements of κ_r of about 1 in 1000. Some typical results are shown in Fig. 1. With results of this kind, the fractional part of the total change which is due to reversible processes may readily be found. An incidental result of the experiments was a demonstration that κ_r is not a unique function of the magnetization I, as has sometimes been suggested. This is shown by Figs. 1(*a*) and 1(*b*), and that the suggested relation is not even a rough approximation was shown conclusively by measurements of κ_r at points within the main hysteresis loop; κ_r is, in fact, not a unique function of I except in the higher field range where I is a unique function of H.

The method of measuring κ_r which has been outlined is capable of high accuracy, but the time required for a complete set of measurements is considerable. For many purposes a somewhat lower accuracy is adequate, and in making measurements at temperatures other than room temperature a method of recording (κ_r, H) curves rapidly while the temperature of the specimen is held constant and measured accurately is particularly desirable. In a semi-automatic method which has been developed,[7,8] a κ_r, H curve is traced out on an oscilloscope screen, and is recorded photographically. After applying the usual corrections, the measurements give κ_r values to an over-all accuracy of about 1 percent. For measurements above and below room temperature suitable heated and cooled liquids are pumped through the specimen tube.

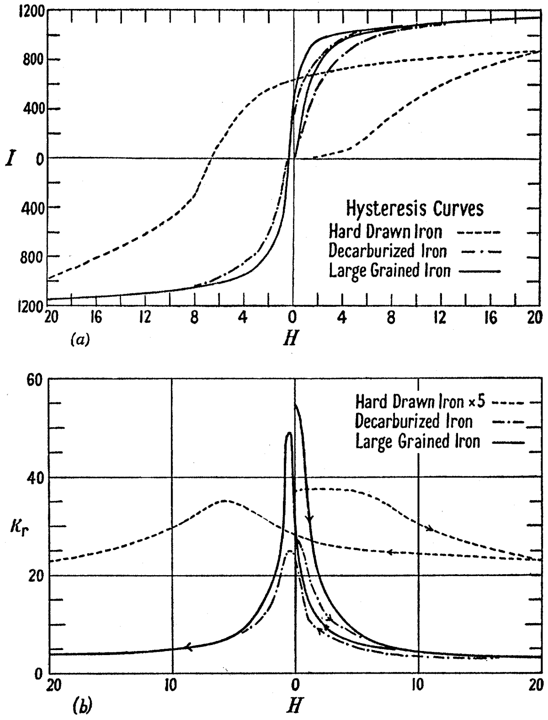

FIG. 1. (*a*) Hysteresis curves for hard-drawn iron, decarburized iron, and large grained iron. (*b*) Variation of reversible susceptibility κ_r with polarizing field H.

The forms of the curves obtained photographically by this arrangement are the same as those in Fig. 1(b). Measurements have been made over a temperature range from −50°C to +200°C for annealed nickel and nickel under tension, with a maximum field of 100 oersteds. The details of the κ_r, H and also the κ_r, I curves at different temperatures present many interesting features, which are of obvious importance in connection with the question of the contributions to the reversible changes from rotational and boundary movement processes. These details cannot, however, be usefully discussed in this paper.

The Barkhausen Effect

In the experimental work started by Bush and Tebble[9] and carried further by Tebble and others, the aim has been to develop a method for determining the number, size distribution of Barkhausen discontinuities over the whole, or any part of a magnetization curve between specified field limits. A slowly changing field is applied to the specimen, and the discontinuous changes in magnetization are detected as voltage pulses induced in a search coil wound round the specimen. These pulses are amplified and can be displayed on an oscillo-

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[7] R. S. Tebble and W. D. Corner, Proc. Phys. Soc. (London) **B53**, 1005 (1950).

[8] Tebble, Corner, and Wood, Proc. Phys. Soc. (London) **B54**, 753 (1951).

[9] H. D. Bush and R. S. Tebble, Proc. Phys. Soc. (London) **60**, 370 (1948).

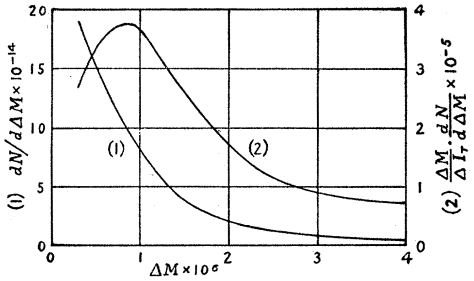

FIG. 2. Distribution and contribution curves for Barkhausen discontinuities in hard-drawn iron. ΔM, change in magnetic moment corresponding to discontinuity. N, number of discontinuities per cm^3. ΔI_T, total change in magnetization.

scope for visual observation or photographic recording, or passed through an amplitude discriminator to a counting unit. For an account of the many subsidiary experiments and calculations carried out in connection with the detailed design of the later form of the apparatus in attempting to insure reliability of counts over as wide a size range as possible, reference must be made to the original paper.[10] It is necessary that the dimensions of the search coil in relation to those of the specimen should be such that equal elementary changes of magnetic moment in any part of the specimen should induce equal voltage pulses in the coil. In connection with this (and also with the process of calibration by the use of a small current-carrying coil placed axially in the search coil) a rigorous theoretical investigation has been made of the flux distribution of a magnetic dipole in a cylindrical specimen.[11]

In the experiments under review the minimum discontinuous changes of magnetic moment which could be detected with reasonable certainty above the noise level were about 0.3×10^{-6} emu (corresponding to about 0.74×10^{13} atoms or a volume of 0.87×10^{-10} cm^3 in iron, and to 2.6×10^{13} atoms or 3.1×10^{-10} cm^3 in nickel). It should be mentioned that the time required for anything approaching a complete investigation of a single specimen may be several months. This is because the number of countable discontinuities in a half-cycle is very large (of the order of 10^7 for the specimens of iron used), and to minimize overlap effects the rate of change of field must be very small (of the order of 10^{-4} oersted sec^{-1} in these experiments).

The primary results give the number of discontinuities over the chosen H range above a minimum size fixed by the discriminator setting. From the results for different settings, corresponding to different minimum values of ΔM, the change in magnetic moment, curves giving the number, size distribution ($dN/d\Delta M$ against ΔM) may be obtained; multiplication of the ordinates of these curves by ΔM then gives contribution, size curves (i.e., curves giving the contribution to the discontinuous change of magnetization per unit ΔM range against ΔM). Examples of these curves are shown in Fig. 2. The distribution curves are all quasi-exponential in character, and their general form appears to be much the same for particular sections of the magnetization curve as for the whole range examined. The noise level sets a lower limit to the value of ΔM for which the counts may be regarded as reliable. With a contribution curve such as that for hard-drawn iron, a plausible extrapolation may be made to zero ΔM, but for most of the materials examined the contribution curves, like the distribution curves, are quasi-exponential over the experimental range, and there is no soundly based method for extrapolating the curves to lower values of ΔM. It is, however, a simple matter to estimate the contribution $(dI/dH)_B$ from Barkhausen discontinuities above the minimum ΔM for which reliable counts may be made to the total change of magnetization with field $(dI/dH)_T$. As an illustrative example, the results obtained for hard-drawn iron (for which other curves are given in Figs. 1 and 2) are shown in Fig. 3, and some numerical details for this material and for annealed nickel are given in Table I.

Curves such as those of Fig. 3 provide a very searching test of the experimental methods. Too much attention should not be paid to the $(dI/dH)_B$ curve lying in part above the $(dI/dH)_T$ curve; this obvious but not serious discrepancy is attributed in the main to the relatively large errors which may arise in the differentiation of experimental curves even when the errors in the experimental points are relatively small. The figure is believed to give a fair impression of the order of magnitude of the incidental uncertainties which arise in comparisons of this kind. The numbers in Table I indicate that for hard-drawn iron nearly the whole of the directly measured change of magnetization is accounted for as the sum of the reversible changes (as determined from the reversible susceptibility) and the Barkhausen discontinuities actually counted (with

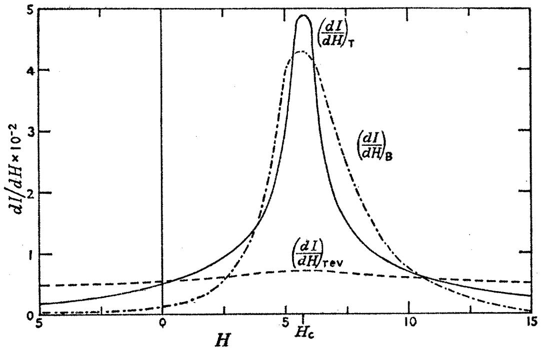

FIG. 3. dI/dH curves for hard-drawn iron: ——— $(dI/dH)_T$; — — — — — — $(dI/dH)_B$; — — — $(dI/dH)_{rev}$. The curves for $(dI/dH)_B$ and $(dI/dH)_{rev}$ give the contributions from the Barkhausen discontinuities and from the reversible changes, respectively, to the total change of magnetization with field $(dI/dH)_T$.

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[10] Tebble, Skidmore, and Corner, Proc. Phys. Soc. (London) **A63**, 739 (1950).

[11] E. W. Elcock, Proc. Leeds Phil. Soc. **6**, 1 (1952).

$\Delta M > 0.3 \times 10^{-6}$). The deficit of some 6 percent may plausibly be attributed to the smaller discontinuities.

For annealed nickel (see Table I) the difference of nearly 40 percent between ΔI_T and the sum of the actually "observed" ΔI_B and ΔI_{rev} is much greater than for hard-drawn iron, and for a single crystal iron specimen, subsequently investigated,[12] a still greater difference has been found. At least part of the difference may be accounted for by the contribution from the uncounted small discontinuities, but in addition to this errors may arise in the counts for some of the materials owing to the occurrence of discontinuities in "bursts." One discontinuous change may trigger a number of others, so that a succession of discontinuities occurs in a very short time interval which is independent of the rate of change of field. In the design of the apparatus particular attention is given to the optimum relations between the time constants of the coil, amplifier, and discriminator systems and the decay time of the eddy currents in the specimen associated with a discontinuity, and the arrangement is such that reliable counts can be made at a rate of the order of 1000 per sec for regular or randomly distributed single discontinuities. It seems inherently impossible to obtain accurate counts of the individual components in a burst. Fortunately the occurrence of bursts is manifested visually and audibly in the scaling unit and recorder with slow rates of field change, and is also obvious on oscillographic records. It was, for example, not apparent for hard-drawn iron, but very noticeable for large-grained and single-crystal iron. For the first material, the curves derived from the experimental counts show quantitatively that practically the whole of the difference between the total and reversible changes in magnetization, ΔI_T and ΔI_{rev}, is due to the irreversible changes associated with Barkhausen discontinuities. There can be little doubt that this is so in all cases, but that in materials for which the burst phenomena are prominent the actual number of discontinuities may be significantly greater than the number counted in the experiments. Even then the distribution and contribution curves are believed to be substantially accurate for the larger values of ΔM (say, $\Delta M > 10^{-6}$), for in a burst it will be mainly the smaller discontinuous changes initiated by a larger one which are missed.

TABLE I. Contribution of Barkhausen discontinuities and reversible processes ($\triangle I_B$ and $\triangle I_{\text{rev}}$) to the total change in magnetization.

Material	H range	Limits of ΔM (10^{-6} emu)	ΔI_T	Contributions (and percentage of ΔI_T) ΔI_B	ΔI_{rev}	$\Delta I_B + \Delta I_{\text{rev}}$
Hard-drawn iron	±18.6	0.3 – 35	2150	1844 (86)	180 (8.4)	2024 (94)
Annealed nickel	±14.9	0.3 – 12	700	326 (47)	115 (16.4)	441 (63)

The mean, or weighted average value of ΔM can perhaps best be taken as the value above which the discontinuities account for half the total discontinuous magnetization (i.e., $\frac{1}{2}(\Delta I_T - \Delta I_{\text{rev}})$). For hard-drawn iron this mean value is $\Delta M = 1.6 \times 10^{-6}$ (corresponding to a minimum of 6.5×10^{11} atoms or a volume of 4.6×10^{-10} cm^3). For large-grained iron the mean value is about half as great, for annealed nickel about a quarter (through the corresponding volume here is about the same as for hard-drawn iron). The volumes corresponding to these discontinuities in magnetization are very much smaller (by a factor of the order of 10^5) than the volumes of the primary domains, as revealed by powder patterns, in single-crystal and large-grained material, though they are comparable in size with the smaller of the secondary domains (of the closure type, or associated with inclusions). Since the Barkhausen disconcontinuities may account for the greater part of the change of magnetization where boundary movement is predominant, they cannot be due solely to changes in the secondary domains, but must be associated mainly with the movement of the primary domain boundaries, in particular of the 180° boundaries. The usual minimum energy conditions do not preclude wrinkles or corrugations in these boundaries (as distinct from 90° boundaries), provided that the corrugations run parallel to the direction of magnetization.[12] The state of minimum energy may therefore be attained by small scale variations of curvature conditioned by the localized strains, inclusions, or irregularities of composition and the associated disperse fields without seriously affecting a large-scale planeness. It is suggested that the Barkhausen discontinuities arise mainly from the continuous localized readjustments or jerks accompanying (and indeed constituting) the movement of the 180° boundary as a whole. The details of the individual processes involved have been worked out in certain idealized cases, and the usual formal theory of irreversible boundary movements can be taken over at once, though the detailed picture is different.

With small-grained materials the Barkhausen volumes may be comparable in size with the individual grains, and considerable caution would be necessary in extending the interpretation just outlined to these materials. A discussion of this particular aspect of the general problem cannot, however, usefully be taken up here.

Whether or not the general type of interpretation suggested is correct, these studies of the Barkhausen effect have provided quantitative knowledge about elementary irreversible processes which will be invaluable as a background to constructive thinking about the individual processes, and also about the aggregate effects as manifested in the over-all irreversible changes along a magnetization curve.

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[12] V. L. Newhouse, thesis, Leeds, 1952. A paper on this work is in preparation.

V. EXPERIMENTAL INVESTIGATIONS: MAGNETOTHERMAL

Change of Temperature with Change of Field

The importance of studies of the magnetocaloric effect (the change of temperature accompanying adiabatic change of field) in low and moderate as well as in high fields has already been indicated. The experimental work, which has been surveyed elsewhere,[1,3] will not be considered here, as it will be dealt with in the paper contributed by Bates, who is responsible for most of the recent work and whose papers may be consulted for full details. It will be sufficient to recall that, if accurate estimates of dT/dH are to be made from the observed values of $\Delta T/\Delta H$ in the lower field ranges, where I and dI/dH may change rapidly with H, the increments ΔH must be small; a lower limit is set by the many difficulties in measuring small temperature changes. It seems unlikely that there will be any substantial and useful increase over the highest sensitivity which has been attained, of 1 scale division for a temperature change of about 5×10^{-6} degree (corresponding to a heat developed of about 200 erg cm^{-3}). This in itself is a remarkable achievement. The minimum field change necessary to give a reliably measurable temperature change varies with the material and field range, but inspection of the curves given in the next section shows that it is seldom likely to be less than 5 or 10 oersteds. The Q' curves in Figs. 5–8 show the general character of the results that have been obtained.

Change of Magnetization with Temperature

The magnetocaloric measurements give the temperature change (or the heat developed) in all the superposed processes contributing to the change of magnetization, and one of the main obstacles to a more detailed interpretation of the results in the lower field range is the difficulty of estimating separately the contributions from reversible and irreversible processes. This was emphasized in the course of a discussion of magnetothermal effects,[3] where it was shown that in the extensive series of magnetocaloric measurements of Okamura (1936) the method used for estimating separately the reversible and irreversible heat developed was unsound, and that it could lead at best to only a very rough qualitative indication of the general trend of the separate heat changes. It was suggested that the required information might be obtainable from measurements of κ_r, H curves over a range of temperatures, but further consideration has shown that this argument too was defective. A solution of the problem has been found, in principle, in the direct application of the thermodynamic relation (7), or (12), namely,

$$\left(\frac{\partial Q'}{\partial H}\right)_S = C_p\left(\frac{\partial T}{\partial H}\right)_S = -T\left(\frac{\partial I}{\partial T}\right)_H. \tag{18}$$

This equation relates the reversible part of the heat change accompanying adiabatic change of field with the reversible change of magnetization with temperature at constant field. Thus if $(\Delta I/\Delta T)_H$ can be determined with increments of T sufficiently small that the change is reversible, the corresponding value of $(\partial Q'/\partial H)_{\text{rev}}$ can be derived. By measurements at different values of H, the value of $(\Delta Q')_r$ over any part of the range can be obtained, and if magnetocaloric measurements of $\Delta Q'$ have been made, the reversible and irreversible contributions in

$$\Delta Q' = \Delta Q_r' + \Delta Q'_{\text{irr}} \tag{19}$$

are separately determined.

Suitable experimental methods for making the necessary measurements have recently been developed and applied in a detailed investigation of annealed nickel.[13] There is, of course, no difficulty in principle in determining $(\partial I/\partial T)_H$ in the higher fields (i.e., well above the hysteresis range), as it is merely necessary to determine the ordinary magnetization curve with sufficient accuracy at a number of different temperatures. In the lower fields, where I is not a unique function of H and T, this procedure does not give the required results. It is necessary to start at the point on a magnetization curve at which $(\partial I/\partial T)_H$ is required, and to measure the change of I as the temperature is changed. A magnetometer method is clearly indicated. Some curves obtained with the standard type of arrangement are shown in Fig. 4. With the change of temperature of the order of 20°C at a field of 1.40, well above the coercive field for this particular specimen (about 0.2) it is apparent from the large temperature hysteresis that most of the change of magnetization is irreversible. Experiments on the Barkhausen effect accompanying change of temperature have given similar indications,[14] even qualitative observations enabling a useful estimate to be made of the relative degrees of irreversibility in different changes. From the work on reversible

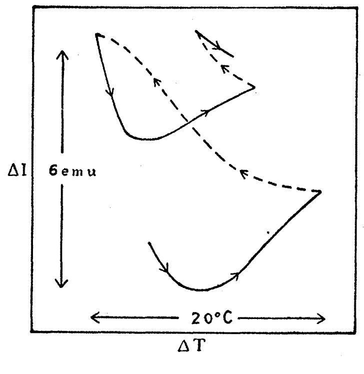

FIG 4. Typical results of magnetometer experiments showing variation of intensity of magnetization with temperature. Annealed nickel, $H=1.40$ oersteds. $(\Delta T)_{\max}=20°\text{C}$.

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[13] Tebble, Wood, and Florentin, Proc. Phys. Soc. (London), to be published (1952).

[14] V. L. Newhouse, Proc. Phys. Soc. (London) **A65**, 325 (1952).

susceptibility,[7] the magnitude of ΔI within which the change is effectively reversible can be found; from this and a rough knowledge of $(\partial I/\partial T)_H$ the corresponding magnitude of ΔT can be estimated. In the range of special interest it is of the order of 1°C, and the experimental problem is therefore that of devising a method of measuring the corresponding small changes of intensity. With the standard arrangement used in obtaining the curves of Fig. 4, the over-all sensitivity was 1-mm magnetometer scale deflection for a change ΔI of 0.38 emu. An increase in sensitivity by about 20 is required to enable reliable measurements to be made of $(\partial I/\partial T)$ in the range 0.4 to 0.04.

The essentials of the arrangement finally adopted are as follows. Two "identical" specimens were mounted within identical magnetizing solenoids, connected in series opposition, and placed equidistant from the magnetometer. The temperature of one specimen was kept constant by a flow of water, that of the other raised or lowered by a controlled flow of butyl alcohol, heated or cooled. The specimens, about 40 cm long, were accurately prolate spheroidal in shape, with an equatorial diameter of about 0.4 cm. (This has the double advantage that the intensity is uniform, and that, as the field at an external point can be exactly calculated, the magnetometer may be close to the specimen.) The magnetometer was an astatic modification of the Sucksmith horizontal suspension type. An over-all sensitivity of up to 1-mm scale deflection for a change ΔI of 0.020 emu could be used. With the specimen at the same I, H point as in Fig. 4, the change of I with T was found to be reversible and linear over the maximum range of 1.3°C, with $(\Delta I)_{\max}$ about 0.16, and reliable readings could be taken at intervals of about a tenth of this range.

The above details are very incomplete, but they are given to show that while it is quite practicable to make measurements of $(\partial I/\partial T)_{\text{rev}}$ over the low field range of a magnetization curve, considerable care is necessary if reliable results are to be obtained. A presentation and discussion of some of these results will be included in the next section.

VI. THE ANALYSIS OF MAGNETIZATION CURVES: ILLUSTRATIVE RESULTS

The low field magnetic properties of ferromagnetics are strongly structure sensitive. Ideally, therefore, if the results of different types of experiment are to be utilized in the analysis of the magnetization curves of a particular material, the experiments should all be made with the same specimen. This is not entirely practicable, as specimens differing in dimensions are required for the different investigations, but with cooperation between the different experimentalists, it can be sufficiently nearly attained by the use of the same material subjected to the same thermal and mechanical treatments; the most sensitive criterion of similarity being the magnetization curve itself. Most of the work so far has been carried out without having clearly in mind the requirements to be fulfilled if the different results are to be satisfactorily linked together, and usually only one or two sets of data are available for each particular material. For this reason it is not possible to illustrate the full scope of the type of analysis envisaged.

In the different investigations, however, the materials used have often been sufficiently similar magnetically (sometimes by deliberate choice) that useful cross comparisons can be made. Following the historical course of development of the general scheme, an account will first be given of the partial analysis which can be effected by utilizing magnetization curve and magnetocaloric data, and the manner in which this analysis can be made more complete by utilizing other data will then be outlined.

Magnetization Curves and Magnetocaloric Data

In the method of analysis introduced by Stoner and Rhodes,[3] the heat developed in an adiabatic change of field from an appropriate initial value H_0 to H is expressed by

$$Q'=a\int d(IH)+\int b''HdI, \tag{20}$$

where the lower and upper limits of the integrals correspond to H_0 and H. The first term corresponds to change in intrinsic magnetization (see Eq. (13)), and the coefficient a can be calculated from I_0 and dI_0/dT (see Eqs. (12) and (13)). Since a is known, or determinable, with considerable accuracy, it is convenient to subtract this part of the contribution to the experimental Q' and to consider the remainder, Q'', where

$$Q''=Q'-a\int d(IH)=\int b''HdI. \tag{21}$$

The coefficient b'' is essentially experimental and is to be determined from the I, H and Q'', H curves from the relation

$$b''=(I/H)(dQ''/dI)\simeq\Delta Q''/\Delta\int HdI, \tag{22}$$

using increments as small as is consistent with the avoidance of nonsignificant irregularities due to the inevitable scatter of the experimental points. If the change in magnetization corresponding to Q'' were entirely due to reversible rotations in the natural anisotropy fields of the crystals, b'' would be constant and equal to b as calculated from the crystal anisotropy constants (see Eq. (14)). In general, b'' will differ from b owing to internal strains and other irregularities, and also it will vary with H, particularly over ranges where irreversible changes are predominant. A comparison of b'' with b may then give some indication of the state of the material and the nature of the elementary mag-

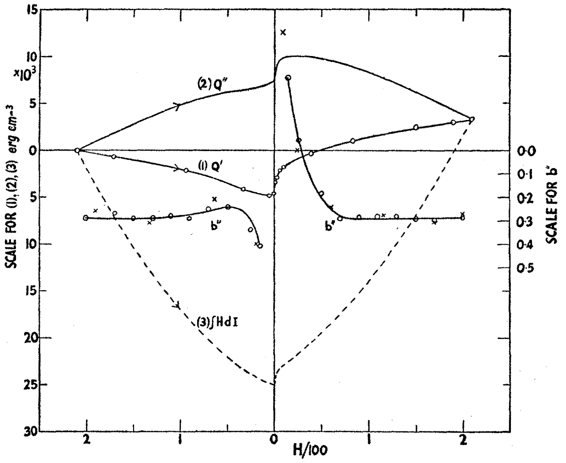


FIG. 5. Magnetocaloric effect in annealed Armco iron. Data for curves (1) and (3) from Hardy and Quimby (1938). Q', heat developed. Q'', heat developed less that due to change in intrinsic magnetization. $b''=\Delta Q''/\Delta\int HdI$. Circles, values derived from smoothed curves. Crosses, values from experimental points.

netization processes occurring. The "ideal" values of a and b and some other data for the ferromagnetic elements are given in Table II, and the results of representative analyses are shown graphically in Figs. 5–8.

As the curves of Figs. 5, 6, and 7 have been fully discussed elsewhere,[3] mention will be made here of only a few of the main points. It becomes evident on considering the curves that the greater part of the adiabatic temperature change accompanying magnetization can be accounted for by a superposition of two effects: the reversible heating (on increase of field) associated with increase of intrinsic magnetization, and the reversible cooling (on increase of field) associated with rotations of the domain magnetization vectors in the local anisotropic field. (It should be noted that over a half-cycle, starting from $|H_{\max}|$ as H_0, a "cooling" effect gives a positive Q', or Q'', in the low field region and conversely.) In cobalt the rotational effect is predominant. The other extreme, in which the intrinsic effect is predominant is represented by certain alloys, in which, particularly when the Curie point is not far above the temperature of the experiments, relatively enormous heating effects have been obtained (e.g., for Monel metal, 70 Ni, 30 Cu in a half-cycle with $|H_{\max}|$ about 360, $-Q'$ exceeds 100×10^3). For iron and nickel the component heating and cooling effects are of the same order of magnitude, the resultant effects are small (and hence difficult to measure), and the experimental Q' curves show an extraordinary diversity for closely similar materials, and also for the same material taken over half-cycles with different limits.

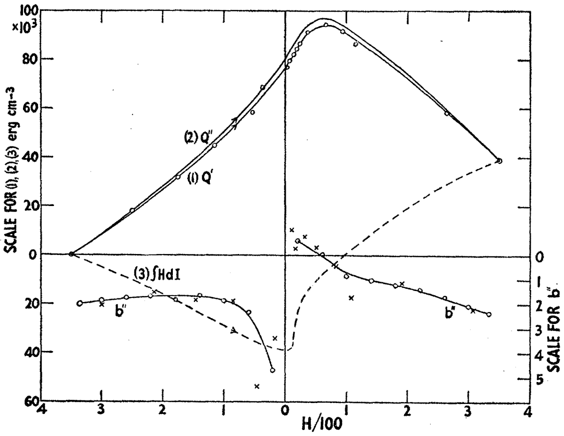


FIG. 6. Magnetocaloric effect in annealed cobalt. Data for curves (1) and (3) from Bates and Edmondson (1947).

The values of b'' obtained from the experimental results are in many cases reasonably constant over a field range in which the change of magnetization would ordinarily be assumed to be due to reversible and primarily rotational processes. The mean values of $-b''$ from the initial $I_{\max}$ to I_{rem} for Fe, Co, and Ni (Figs. 5–7) are about 0.30, 2.0, and 3.7 as compared with calculated values of b (at the temperatures of the experiments) of 0.63, 1.3, and 5.9. (It should be noted that the temperature of the specimens was about 41°C in the experiments of Hardy and Quimby, and about 20°C in those of Bates.) Over the constant b'' range between about 50 and 200 oersteds in Fig. 8, the value of b'' of about 4.7 agrees almost exactly with the calculated value of 4.5 for b. When it is borne in mind that by the method of treatment of the results experimental uncertainties are all concentrated in b'', and that owing to the uncertainties in the values of the crystal constants[1] the calculated values of b may be in error by 20 percent or more, the agreement as to order of magnitude of b'' and b must be considered satisfactory. The differences, however, even in the quasi-constant ranges, are probably significant and may be tentatively attributed to residual strains even in annealed materials. It has been shown[3] that the effect of such strains is to reduce the effective numerical value of b in nickel, but in general not even a rough estimate of the effect can be made owing to the absence of reliable data on the crystal magnetostriction coefficients.

TABLE II. Data for ferromagnetic elements at 290°K. $a=-(T/I_0)(dI_0/dT)$. $b=(T/k)(dk/dT)$. $\Delta Q'$, energy change, erg cm⁻³, corresponding to temperature change ΔT.

	$I_0\times10^{-3}$	$a\times10^2$	$-b$	$(\Delta T/\Delta Q')\times10^8$
Fe	1.72	3.4	0.58	2.8_3
Ni	0.48_5	14.3	4.5	2.5_6
Co	1.42	0.8_8	1.3	2.6_2

In low fields the striking variations of b'' may be taken as an indication of the occurrence of irreversible processes, and little progress can be made in analyzing this part of the magnetization curve unless the reversible and irreversible heating effects can be separated (see below). The decrease in the numerical value of b'' in high fields, apparent in Figs. 7 and 8 for nickel, occurs in

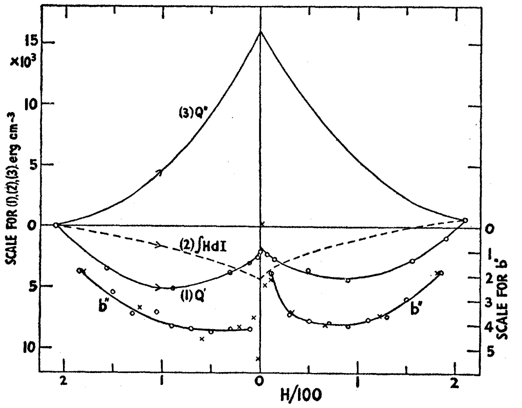


FIG. 7. Magnetocaloric effect in annealed nickel. Data for curves (1) and (3) from Hardy and Quimby (1938).

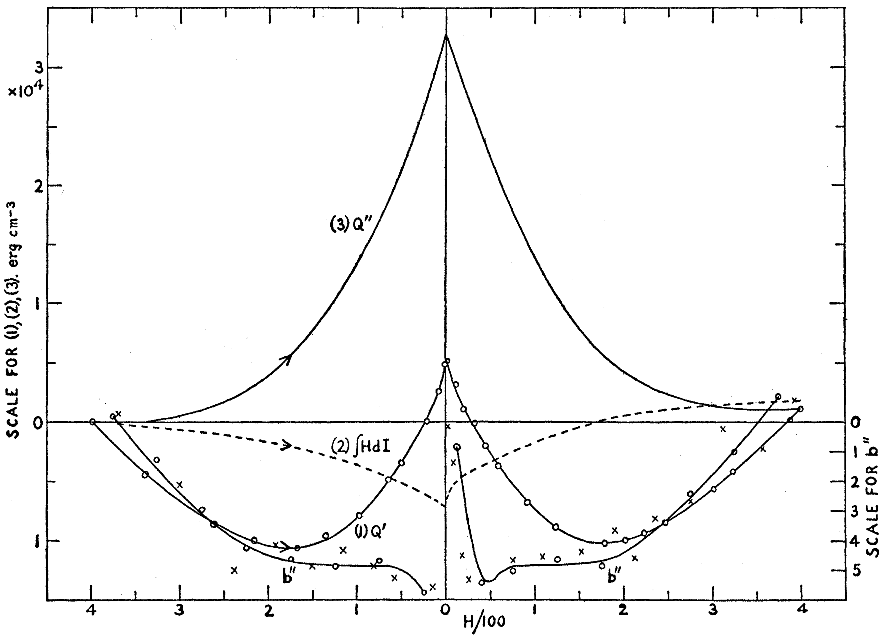
a reversible range, and raises an entirely new problem. Although accurate estimates of b'' cannot be made in the higher fields without precision measurements of the magnetization curves, the systematic decrease in $|b''|$ cannot be attributed to uncertainties arising for this reason. In connection with Fig. 7, it was suggested[3] that the decrease might be due to a relative increase in higher fields in the contribution from crystal grains with higher strain anisotropy and numerically smaller b. It does not seem possible, however, to account for very small values of b'' in this way (an estimated lower limit[15] to $-b''$ is 0.27), and it is still less possible to account for a change of sign, such as is shown in the highest fields in Fig. 8. The basic fact is that in the higher fields the rate at which heat is developed with change of field dQ'/dH approaches and eventually exceeds the calculated rate due to change in intrinsic magnetization dQ_i'/dH. In other words, unless the calculated dQ_i'/dH is much in error, which there is no reason to suppose, there is an additional heating effect other than those so far considered which may exceed, numerically, the coiling effect associated with rotational processes. Two possible processes have been suggested. One is the quasi-paramagnetic re-orientation of the moments of "small domains," which provides a possible explanation of an otherwise unexplained excess in the value of dI/dH in very high fields.[16] The other is the gradual "fading-out" of domain boundaries as the directions of magnetization on either side become more nearly parallel. Either of these processes would lead to an increase in the effective value of the coefficient a, which preliminary calculations show could be of the required order of magnitude over the relevant ranges of magnetization curves. Much more theoretical and experimental work will, however, be required before definite conclusions can be reached.

Variation of Magnetization with Temperature

From suitable measurements on the change of magnetization with temperature in constant field it is possible, as described in Sec. V, to derive values of $(\partial Q'/\partial H)_S$ [see Eq. (18)], so that in principle the thermal changes associated with reversible and irreversible processes can be separated. Some results obtained by Tebble *et al.*[13] for annealed nickel are shown in Fig. 9.

That the curves 1a and 1b do not join smoothly is probably attributable mainly to incomplete reversi-

FIG. 8. Magnetocaloric effect in annealed nickel. Data for curves (1) and (3) from Bates and Davis (1950).



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[15] P. Rhodes, unpublished.

[16] K. L. Hunt (to be published, 1952).

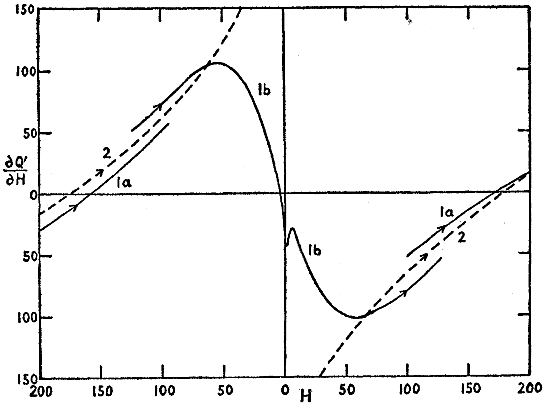

FIG. 9. Reversible and total magnetocaloric effect in annealed nickel. $(\partial Q'/\partial H)$, erg cm^{-3} oersted^{-1}. 1. $(\partial Q'/\partial H)_S$ (isentropic) derived from $(I/T)_H$ for nickel 2. 1a. $(T)_{\max}=20°\text{C}$. 1b. $(T)_{\max}=1.3°\text{C}$. Data from Tebble, Wood, and Florentin (1952). 2. $(\partial Q'/\partial H)_Q$ (adiabatic) from direct measurements for nickel 1. Data from Bates and Davis (1950) (for I, H curves see Fig. 10).

bility of the change of I with T with a maximum value of ΔT of 20°C even in fields greater than 100 oersteds. The "annealed nickel" of Tebble was sufficiently similar (see Fig. 10) to that of Bates, for which the magnetocaloric results are shown in Fig. 8, for a comparison of the two sets of results to be significant at fields well above the coercive fields (about 0.4 and 2.0, respectively). The close agreement between the curves (1) and (2), obtained by totally different methods, for fields greater than about 60 oersteds, where the changes are predominantly reversible, confirms the general theoretical treatment, and indicates, in a striking way, the general reliability of the experimental results. The divergence of the curves below 60 oersteds is to be associated with the rapidly increasing effect of irreversible processes.

The heat developed reversibly over the range from $H=-50$ to $H=+50$ is shown by the Q'_{rev} curve in Fig. 10. The total heat developed, Q', over this range (taken from Fig. 8) is also shown. The difference between the two curves gives approximately the heat developed in irreversible processes.

By allowing for the contribution to $(\partial Q'/\partial H)_{\text{rev}}$ from the changes in intrinsic magnetization, a value of b''_{rev} may be obtained in which the effect of irreversible changes is eliminated. When κ_r is known, a convenient formula is

$$b''_{\text{rev}}=\frac{(dQ'_{\text{rev}}/dH)-a(I+\kappa_r H)}{\kappa_r H}. \tag{23}$$

The so calculated values of b''_{rev}, which should correspond only to reversible rotations and boundary movements, are shown in Fig. 10. Approximate constancy of this "reversible" b'' is maintained down to about $H=5$, rather than to about $H=70$ as for b'' in Fig. 9. The actual value of b''_{rev} is very close to the ideal value of b calculated from the crystal anisotropy (Table I) over the whole of this range, suggesting that reversible movements of a domain boundary are thermally equivalent to reversible rotations of the magnetization vector in a domain. Owing to the rapid variation of I with H for fields between -2 and $+2$ oersteds for this material ($H_c \simeq 0.36$), the values of b''_{rev} in this range are very uncertain, and too much weight should not be attached to the curious variations indicated by the curve.

Although the ascending and descending branches of the hysteresis curve practically coincide for $|H|>20$, it is apparent from Fig. 10 that thermal effects associated with irreversible processes are spread over a considerably wider range. Along the ascending branch of the curve, the irreversible changes result in heating (positive gradient of the Q'_{irrev} curve) up to the region of the coercive field, and thereafter in cooling. (The net irreversible effect over a complete ascending curve is a heating corresponding to half the "hysteresis loss," but for this particular material the maximum value of Q' greatly exceeds the net heating over the half-cycle.) There is, of course, no general thermodynamic reason for expecting an irreversible change to be accompanied by heating rather than cooling, and these experiments show quite clearly that the thermal changes associated with irreversible magnetization processes are neither necessarily positive nor wholly irreversible. In fact, the major part of the thermal change associated with irreversible processes for annealed nickel is reversible, and the sign and magnitude of the effect indicate that the associated change of internal magnetic energy [see Eq. (5)] is mainly due to the change of direction of the magnetization vectors in the local anisotropic fields.

The above short discussion shows that potentially by combining the results of magnetic and magnetothermal investigations much information can be obtained about the details of the elementary processes occurring along the course of a magnetization curve. A fully quantitative analysis of the lower field regions cannot, however, be usefully undertaken until the various experiments are made with essentially the "same" material.

VII. SUMMARIZING DISCUSSION

The theme of this paper has been magnetic and magnetothermal investigations, the results of which can be linked together to give information about the processes contributing to the change of magnetization with field along the magnetization curves of ferromagnetic materials. The basic question being asked is, "What are the elementary processes occurring, and with what relative and absolute magnetic and thermal effects, at each stage in the course of the change of the bulk magnetization?" An inquiry of this kind could not be undertaken unless much was already known about the main elementary processes from more direct experimental investigations and theoretical studies; for a quantitative treatment, moreover, the spontaneous magnetization I_0 and the crystal anisotropy and magnetostriction coefficients must be known, and also the variation with temperature of each of these.

Reference has been made to five types of experimental investigation, the results of which have been utilized in such partial analyses as have been presented:

(1) The variation of I with H at constant temperature, i.e., the ordinary magnetization curves.

(2) The reversible susceptibility κ_r.

(3) The magnetocaloric effect, that is, the change of temperature accompanying adiabatic magnetization. The experimental results give effectively $(\partial T/\partial H)_Q$, or the corresponding heat developed $(\partial Q'/\partial H)_Q$.

(4) The reversible change of magnetization with temperature, $(\partial I/\partial T)_H$, which gives the reversible change of temperature, or the heat developed reversibly with change of field, that is, $(\partial Q'/\partial H)_S$.

(5) The Barkhausen effect. The experiments give the size distribution of the discontinuities in magnetization above a certain minimum size, and the contribution of these essentially irreversible changes to the total change of magnetization over the whole or any part of the field range examined.

In connection with (1) and the frequent use of the phrase "the magnetization curve" in this paper, it may be prudent to state explicitly that there is not a unique magnetization curve for any material showing hysteresis effects, but that it has not seemed necessary to enter here into either the subtleties of the defining conditions or the difficulties of accurate measurement. The curves mainly considered in illustration have been the ascending branches of hysteresis loops taken after the establishment of cyclic conditions by the standard methods; but the general scheme of analysis would apply equally well to any reproducible type of curve.

Measurements of κ_r (2) in conjunction with (1) will give the reversible component of the change of magnetization over any part of the field range.

The magnetocaloric measurements (3) give the thermal effects associated with the total change of magnetization. Over reversible ranges conclusions can be drawn, from the sign and magnitude of the effect, about the contributions from the change in intrinsic magnetization and from other reversible changes, by far the most important of these, in the cases so far examined, being the rotations of the domain magnetization vectors. The thermal effects associated with change in magnetization by change in intrinsic magnetization and by rotational change are of opposite sign, and the remarkable diversity in the magnetocaloric curves for different materials is largely due to the different magnitudes and relative proportions of these two effects. The presence of strain is qualitatively indicated by a deviation in the values of a characteristic coefficient b'', calculable from the experimental results, from the value of a coefficient b, which may be derived from the crystal anisotropy data. The possibility of giving a more quantitative interpretation of the experimental results is dependent on further, or more accurate data becoming available for crystal anisotropy and magnetostriction.

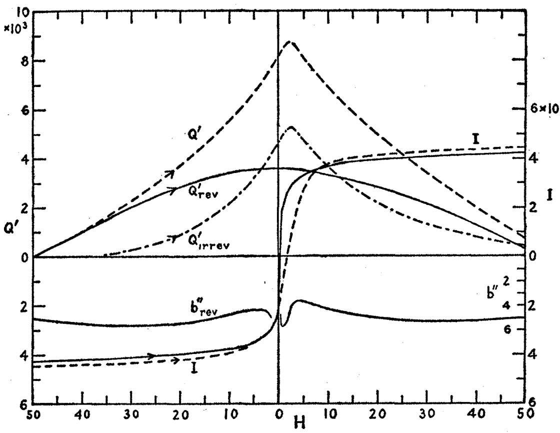

FIG. 10. Magnetocaloric effect in annealed nickel. Q', total heat developed from magneto-caloric measurements for nickel 1 (I, H curve broken) (Bates and Davis). Q'_{rev}, heat developed in reversible changes, from measurements of $(\partial I/\partial T)_H$ for nickel 2 (I, H curve full) (Tebble, Wood, and Florentin). $Q'_{\text{irrev}} = Q' - Q'_{\text{rev}}$. b''_{rev}, effective thermal change coefficient, derived from Q'_{rev}.

Little progress can be made in interpreting the magnetocaloric results in the lower field ranges unless the thermal effects associated with reversible and irreversible processes can be separately estimated. This is made possible by measurements on the reversible change of magnetization with temperature (4), from which the thermal contribution from reversible processes to the magnetocaloric effect can be derived, and hence also, if measurements of type (3) and (4) are made on the same material, that from irreversible processes. The reversible thermal effects even well into the hysteresis region are mainly associated with rotation of the domain magnetization vectors. The thermal effects associated with irreversible processes may be of either sign and are not wholly irreversible. In the material which has been most carefully examined, annealed nickel, the main part of the thermal change, is that associated with the change in state, corresponding to the change in direction of the domain magnetization vectors or the magnitude of the intrinsic magnetization, and the strictly irreversible heating effect associated with the irreversible processes by which the change of state is attained is comparatively small. It is this residual effect which contributes to the net heating over a complete cycle, or half-cycle.

The work on the Barkhausen effect (5) has shown that the discontinuities in magnetization are not in general due to spontaneous movements of primary domain boundaries as wholes, but to irregularities on a smaller scale in the movement, particularly of 180° boundaries. It seems probable that as the field changes the boundary adjusts itself to its changing quasi-equilibrium position and maintains an approximate planeness by a sequence of sudden localized jumps of sections of the boundary, of magnitudes conditioned by localized irregularities. It seems certain that almost the whole of the irreversible change of magnetization is due to these Barkhausen jumps but that in all materials part of the irreversible contribution must be attributed to discontinuities below the minimum size for which reliable counts can be made, and in some materials the discontinuities occur in part in rapid bursts and cannot be individually recorded.

It can hardly be suggested that a thorough investigation of the Barkhausen effect should be made for every ferromagnetic material. This is, indeed, unnecessary, for, although there are many special problems in connection with it which call for fuller examination, the recent work which has been outlined has gone far towards showing the character of the processes to which irreversible changes in magnetization may be attributed. The over-all irreversible change can be most readily obtained by the more direct determinations of the I, H curves (1) and the κ_r, H curves (2). The two types of magnetothermal investigation ((3) and (4)) are, however, indispensable if an adequate analysis is to be made. As investigations of this kind make considerable demands on experimental skill and on time, it is particularly desirable that there should be the closest cooperation between those engaged in such work in the choice of materials for examination.

The information that can be obtained by combining the results of different investigations has been exemplified in Sec. VI. The analyses are, however, partial, and it will not be possible to assess fully the value of the general method until more complete sets of results are available for one and the same material. It would be unwise to base general conclusions on the partial analyses for a limited number of materials which have so far been made. These partial analyses do, however, show that by making measurements on the macroscopic behavior of ferromagnetic materials in two or more types of experiment, and combining the results in some such manner as that outlined, extensive information may be obtained about the microscopic processes taking place. An analysis of this kind does not, of course, provide a theoretical "explanation" of the form and quantitative characteristics of a given magnetization curve; but the prospects of a satisfying explanation will become much brighter when what is happening becomes clearer—when it is known, in short, what it is that has to be explained.

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