

The Analysis of Magnetothermal Measurements

L. F. BATES AND A. J. PACEY

*University of Nottingham
England*

Following a brief review of the theory of magnetothermal effects, methods of identifying the principal magnetization processes in low fields are discussed. It is shown that under certain conditions, a constant, b_2 , may be estimated both from magnetothermal results and from coercivity data. Some values of b_2 are quoted which enable the principal low field magnetization processes in several metals to be identified with processes envisaged by Goodenough (1954).

1. Introduction

When the magnetic field acting upon a system changes rapidly by an increment ΔH , an adiabatic change of temperature, ΔT , takes place within the system. This temperature change can be regarded as a change in the thermal energy, $\Delta Q'$, of the system. The experimental study of magnetothermal changes consists of the measurement of $\Delta Q'$ corresponding to different field steps, ΔH , around the hysteresis cycle.

Attempts to explain the results of such experiments were not very successful up to 1949. In that year, Stoner and Rhodes¹⁾ derived an equation which explained the main features of the reversible magnetothermal effects. Denoting reversible quantities by the suffix R , this equation is

$$(\partial Q'_R / \partial H)_S = a[I + H(\partial I_R / \partial H)_T] + bH(\partial I_R / \partial H)_T. \quad (1)$$

The first term on the right hand side of the equation gives the contribution of the Weiss and Forrer magnetocaloric effect, the coefficient a being equal to $-(T/I_0)(dI_0/dT)$. The effect of rotational processes is represented by the other term, in which $b = (T/K)(dK/dT)$, K being the principal anisotropy constant.

In order to compare experimental results with the behaviour predicted by this equation, a quantity Q_R'' is obtained by subtracting the magnetocaloric effect from the measured thermal changes. Thus,

$$(\partial Q''_R / \partial H)_S = (\partial Q'_R / \partial H)_S - a[I + H(\partial I_R / \partial H)_T] \quad (2)$$

A coefficient, b'' is then calculated, as follows,

$$b'' = \frac{(\partial Q''_R / \partial H)_S}{H(\partial I_R / \partial H)_T} \quad (3)$$

It will be seen that if the Stoner and Rhodes equation is obeyed, b'' will be constant and equal to b . In practice, it is found that b'' is always constant in high fields and agrees well with values of b calculated from the anisotropy measurements. However, discrepancies occur in fields of the order of the coercivity, and below.

2. The Work of Teale and Rowlands

The weakness of the Stoner and Rhodes approach is that it assumes that rotations and the magnetocaloric effect are the only contributions to the magnetothermal effects, and it is not sufficiently flexible to take other effects into account. A great advance was made by Teale and Rowlands²⁾, who derived the Stoner and Rhodes equation by a different method, which introduced the free energy function, F , which was defined by Stoner³⁾

$$dF = -SdT + HdI. \quad (4)$$

When several magnetization processes are superimposed they can be represented by separate free energy functions which add simply, (e.g.), $F = F_1 + F_2 \dots F_N$. Teale and Rowlands assumed that for a particular magnetization process, the free energy would be a function of the relative magnetization only, that is,

$$F_n = A_n f(I/I_0). \quad (5)$$

Thus, for rotations, $F_1 = Kf(I/I_0)$.

The effect of assuming that N magnetization processes are superimposed is to modify the last term of the Stoner and Rhodes equation as follows,

$$(\partial Q'_R / \partial H)_S = a[I + H(\partial I_R / \partial H)_T] + \left(\sum_1^N b_n v_n \right) (\partial I_R / \partial H)_T. \quad (6)$$

Here, $b_n = (T/A_n)(dA_n/dT)$. Hence, where

Table I. Magnetization Processes with Expressions for b_n .

Rotations governed by Crystal Anisotropy	$(T/K)(dK/dT)$
Strain Anisotropy	$(T/\lambda_s)(d\lambda_s/dT)$
Shape Anisotropy	$(2T/I_0)(dI_0/dT)$
Domain Wall Movement Governed by Magnetostatic Energy	$(2T/I_0)(dI_0/dT)$
Kersten ⁴⁾	$(T/\gamma)(d\gamma/dT)$
Lilley ⁵⁾	$(2T/\lambda_s)(d\lambda_s/dT) - (T/K)(dK/dT)$
Strain Theory	$(T/I_0)(dI_0/dT) + (T/2\lambda_s)(d\lambda_s/dT)$

rotations occur, $b_1 = (T/K)(dK/dT)$, which is the same as b in the original Stoner and Rhodes equation. Other possible magnetization processes with the expressions for b_n which they are expected to give were set down by Teale and Rowlands, and are quoted in Table 1.

The function v_n in equation (6) will now be considered. Since $F = \sum_1^N F_n$, and, from equation (4) it follows that $(\partial F/\partial I)_T = H$, it is easily seen that

$$\sum_1^N v_n = H. \tag{7}$$

The difficulty about this approach is that there is no way of calculating v_n . But, it is clear that in high fields the equation must reduce to the original form of the Stoner and Rhodes equation, because the latter is found to agree with experiment in those regions. This means that, in high fields, $v_2, v_3 \dots v_N$ must become small, leaving $v_1 \approx H$ and $b'' = b_1$.

The first attempts to use the Teale and Rowlands theory were made in the hope that analogous behaviour would be found in low fields, with v_1 becoming small and $v_2 \approx H$, so that b'' approached b_2 .

However, b'' is found not to approach any constant value, but is always discontinuous in low fields. (In some cases, Bates and Clow⁶⁾ apparently found b'' to be approximately constant in low fields. However, the experimental points were very scattered, and a closer examination reveals discontinuity). The assumptions made in these early attempts to use the Teale and Rowlands theory resemble those made in recent work by Bates and Pacey⁷⁾. However, in the latter work, it is possible to estimate b_2 even when a discontinuity occurs.

3. The Discontinuity in b''

Before further advances could be made in the analysis of magnetothermal effects, it was necessary to understand how the discontinuity in b'' comes about. Saunders and Tebble⁸⁾ showed, (from equations 3 and 6), that

$$b''H = \sum_1^N b_n v_n. \tag{8}$$

Hence, applying equation (7),

$$(b'' - b_1)H = \sum_2^N (b_n - b_1)v_n. \tag{9}$$

They then argued that if the graph of b'' against H is shaped like a rectangular hyperbola, $\sum (b_n - b_1)v_n$ must be a constant. Accordingly, when $N=2$, v_2 is a constant. Saunders and Tebble then proceeded to search for a magnetization process for which v_2 would be constant.

Bates and Pacey took a different view, pointing out that where N is limited to 2, a plot of $(b'' - b_1)H$ vs. H reveals the shape of the function v_2 . Such a plot is shown in

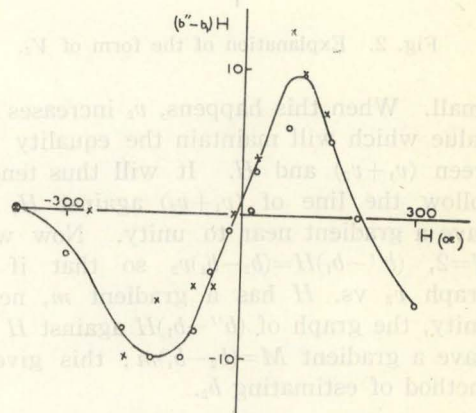


Fig. 1. Plot of $(b'' - b_1)H$ against H for cobalt
 ○ Tebble and Teale⁹⁾
 × Bates and Sherry¹⁰⁾

Fig. 1. Its shape is typical of several materials which have been investigated (e.g. Fig. 3), and shows that v_2 is not a constant, as suggested by the work of Saunders and Tebble. In very low fields, it can be approximately represented by a straight line of gradient M , when plotted against field. Thus, when $N=2$, v_2 is a straight line in low fields.

Bates and Pacey went on to explain how this could come about. When $N=2$, equations (7) and (8) become

$$v_1 + v_2 = H, \text{ and } b''H = b_1v_1 + b_2v_2.$$

Thus a graph of $(v_1 + v_2)$ against H (Fig. 2) is a straight line of unit gradient; v_1 is the derivative of the free energy due to rotations, and in high fields is much greater than v_2 . In lower fields, rotations become less important, and v_1 becomes relatively

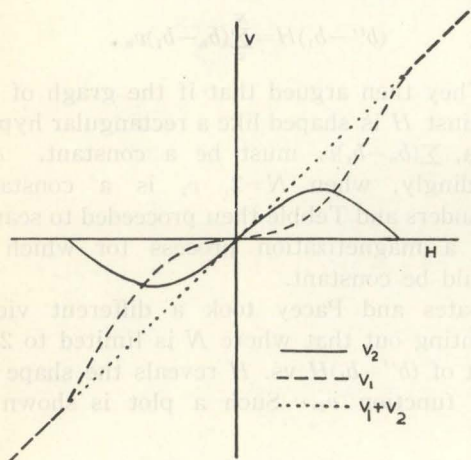


Fig. 2. Explanation of the form of V_2 .

small. When this happens, v_2 increases to a value which will maintain the equality between $(v_1 + v_2)$ and H . It will thus tend to follow the line of $(v_1 + v_2)$ against H , and have a gradient near to unity. Now when $N=2$, $(b'' - b_1)H = (b_2 - b_1)v_2$ so that if the graph v_2 vs. H has a gradient m , nearly unity, the graph of $(b'' - b_1)H$ against H will have a gradient $M = (b_2 - b_1)m$; this gives a method of estimating b_2 .

$$\text{We write } b_2 = (M/m) + b_1. \quad (10)$$

That this provides only an approximate value for b_2 is due not merely to the approxi-

mation involved in assuming that $m \approx 1$, but to the errors in b'' which are indicated by the scatter on Fig. 1. There is also some doubt of the validity of the Stoner and Rhodes equation in low fields, where irreversible processes are important. But, a more important objection to the reasoning as it stands is that it has not thus far accounted for the discontinuity in b'' . The argument is essentially the same as the earlier attempts to apply the Teale and Rowlands equation, in that it assumes that in low fields, $v_2 \approx H$. The advantage of the Bates and Pacey approach, however, is that the graphs of $(b'' - b_1)H$ against H give an indication of the form of v_2 , and an examination of Figs. 1 and 3 reveals that the curves do not pass

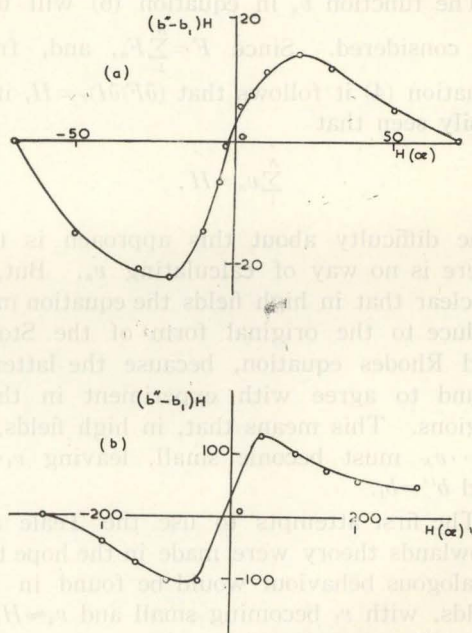


Fig. 3. $(b'' - b_1)H$ against H curves (a) Nickel (Tebble and Teale⁹); (b) Gadolinium at 277°K. (Note the change of scales)

through the origin. Thus, it is not sufficient to write $v_2 \approx H$ in low fields. An additional constant must be added, so that v_2 is represented by an equation $v_2 = L + mH$. The explanation of the shape of v_2 against H is not seriously affected thereby, since L is small, but the fact that v_2 is displaced from the origin does account for the discontinuity in b'' , as is illustrated in Fig. 4.

The discontinuity is also associated with

the fact that Q''_R is not symmetrical about zero field, as was pointed out by Bates, Christoffel, Clow and Davis¹¹. These workers examined a great many results, and found that for many materials, the Q''_R against H curves were symmetrical about the coercive point. If this result is accepted, it can be shown that L , the displacement of v_2 from the origin, is given by

$$L = (-b_2 H_c) / (b_2 - b_1). \quad (11)$$

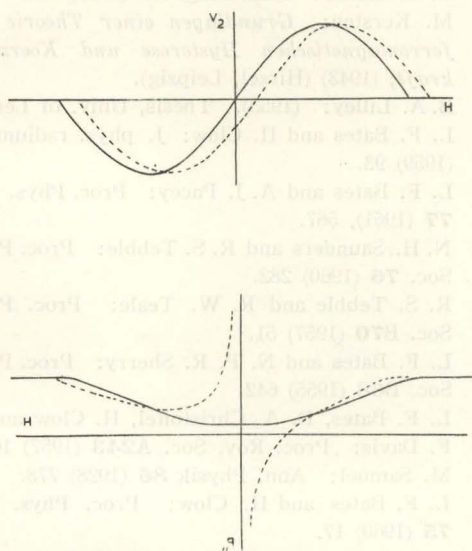


Fig. 4. Ideal curves for v_2 and b'' against H , showing the effects of displacing v_2 from the origin (dotted lines).

4. The Significance of b_2

A method for estimating b_2 has already been given (equation 10). It is clear that any low field magnetization process characterised by b_2 must make a major contribution to the coercivity. In order to relate the coefficient b_2 to the coercivity, the free energy function must be considered in more detail.

Since $H = \sum_{n=1}^N v_n$, (equation 7), it follows that

$$H_c = \left(\sum_{n=1}^N v_n \right)_{I=0}. \quad \text{From equation (5)}$$

$$v_n = (\partial F_n / \partial I)_T = (A_n / I_0) f_n'(I / I_0)$$

$$\therefore H_c = \sum_{n=1}^N (A_n f_n'(0) / I_0) \quad (12)$$

From this we obtain, since

$$b_n = (T / A_n) (dA_n / dT),$$

$$(T / H_c) (dH_c / dT) = - (T / I_0) (dI_0 / dT)$$

$$+ (1 / H_c) \sum_{n=1}^N b_n (A_n f_n'(0) / I_0). \quad (13)$$

If $N=2$, and in low fields $v_1 \ll v_2$, this equation reduces to

$$b_2 = (T / H_c) (dH_c / dT) + (T / I_0) (dI_0 / dT). \quad (14)$$

Equations (10) and (14) enable b_2 to be estimated from two different kinds of experimental data. They are both subject to the same conditions, viz., $N=2$ and $v_1 \ll v_2$. In view of the approximate nature of both methods, the agreement shown in Table II is reasonable. The values of b_2 indicated in this table have been compared with those

Table II. Values of b_2 Observed for Polycrystalline Specimens.

Material	b_2 (equation 10)	b_2 (equation 14)	Nearest Theoretical Expression
Silicon Iron			
0.288% Silicon	-0.13 ¹	—	$(T/\gamma)(d\gamma/dT) = -0.15$
4% Silicon	-0.27 ¹	—	$(T/\gamma)(d\gamma/dT) = -0.21$
Nickel			
Annealed, $H_c < 2$ Oe.	-2.7 ²	-1.2	$(T/\gamma)(d\gamma/dT) = -2.2$
Strained, $H_c \approx 25$ Oe.	—	-0.4	$(T/\lambda_s)(d\lambda_s/dT) = -0.3$
Cobalt			
Annealed, $H_c \approx 25$ Oe.	-0.9 ³	—	$(T/\gamma)(d\gamma/dT) = -0.55$
Annealed, $H_c = 11$ Oe.	—	-0.53 ⁴	$(T/\gamma)(d\gamma/dT) = -0.55$
Gadolinium			
(Measured at 233°K)	-1.1	-1.7	$(T/\gamma)(d\gamma/dT) = -1.0$
(Measured at 277°K)	-6.4	-7.0	$(T/\gamma)(d\gamma/dT) = -4.3$
Nickel Ferrite	-1.2 ⁵	-1.3 ⁵	$(2T/\lambda_s)(d\lambda_s/dT) - (T/K)(dK/dT) = -1.15$

Authors: 1. Saunders and Tebble⁸ 2. Tebble and Teale⁹ 3. Bates and Sherry¹⁰
4. Samuel¹² 5. Bates and Clow¹³.

predicted by the expressions given by Teale and Rowlands (Table I). It is rather striking that almost all the polycrystalline metal specimens give values of b_2 which are close to the values given by the expression $(T/\gamma)(d\gamma/dT)$. Teale and Rowlands identified this expression with Kersten's coercivity theory⁴⁾, but it can also be associated with a more recent theory due to Goodenough¹⁴⁾ which is not open to the same criticisms as Kersten's work (Néel¹³⁾), and is based on the growth of domains of reverse magnetization. It should be noted that the quantity $(T/\gamma)(d\gamma/dT)$ has been calculated on the assumption that $\gamma=(AK)^{\frac{1}{2}}$, where A is independent of temperature.

5. Conclusions

Recent work based on Teale and Rowlands derivation of the Stoner and Rhodes equation has been successful in explaining the magnetothermal behaviour of several materials of fairly low coercivity. It has been found possible to treat these materials on the assumption that only two magnetization processes are important in determining their behaviour. One of these processes, the rotation of magnetization vectors, is dominant in high fields, while the other magnetization process becomes important only in low fields. In several ferromagnetic metals, the low field process has been provisionally identified

as one of the effects discussed by Goodenough.

Acknowledgments

A. J. P. wishes to thank the Department of Scientific and Industrial Research for a maintenance grant.

References

1 E. C. Stoner and P. Rhodes: Phil. Mag. **40** (1949) 481.
2 R. W. Teale and G. R. Rowlands: Proc. Phys. Soc. **B70** (1957) 1123.
3 E. C. Stoner: Phil. Mag. **23** (1937) 833.
4 M. Kersten: *Grundlagen einer Theorie der ferromagnetischen Hysterese und Koerzitivkraft*, (1943) (Hirzel, Leipzig).
5 B. A. Lilley: (1953). Thesis, Univ. of Leeds.
6 L. F. Bates and H. Clow: J. phys. radium **20** (1959) 93.
7 L. F. Bates and A. J. Pacey: Proc. Phys. Soc. **77** (1961), 567.
8 N. H. Saunders and R. S. Tebble: Proc. Phys. Soc. **76** (1960) 282.
9 R. S. Tebble and R. W. Teale: Proc. Phys. Soc. **B70** (1957) 51.
10 L. F. Bates and N. P. R. Sherry: Proc. Phys. Soc. **B68** (1955) 642.
11 L. F. Bates, D. A. Christoffel, H. Clow and P. F. Davis: Proc. Roy. Soc. **A243** (1957) 160.
12 M. Samuel: Ann. Physik **86** (1928) 778.
13 L. F. Bates and H. Clow: Proc. Phys. Soc. **75** (1960) 17.
14 J. B. Goodenough: Phys. Rev. **95** (1954) 917.
15 L. Néel: Physica **15** (1949) 225.

Table II. Values of b_2 calculated for Polycrystalline Specimens

Material	Equation 10	Equation 11	Nearest Theoretical Expression
Nickel Ferrite	-1.2	-1.2	$(T/\gamma)(d\gamma/dT) = -1.2$
(Measured at 327°K)	-0.4	-1.0	$(T/\gamma)(d\gamma/dT) = -1.0$
Cadmium	-1.1	-1.2	$(T/\gamma)(d\gamma/dT) = -1.2$
(Measured at 325°K)	-	-	$(T/\gamma)(d\gamma/dT) = -0.5$
Amorphous Al-Ni-O	-	-0.3	$(T/\gamma)(d\gamma/dT) = -0.5$
Amorphous Al-Si-O	-0.9	-	$(T/\gamma)(d\gamma/dT) = -0.5$
Stainless Steel	-	-0.4	$(T/\gamma)(d\gamma/dT) = -0.5$
Amorphous Al-Si-O	-2.7	-1.2	$(T/\gamma)(d\gamma/dT) = -2.2$
Nickel	-0.3	-	$(T/\gamma)(d\gamma/dT) = -0.3$
1% Silicon	-0.13	-	$(T/\gamma)(d\gamma/dT) = -0.13$
0.25% Silicon	-	-	$(T/\gamma)(d\gamma/dT) = -0.13$
Silicon Iron	-	-	$(T/\gamma)(d\gamma/dT) = -0.13$