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SUBJECT: **A FREE ENERGY MODEL FOR THE HYSTERESIS LOOP**

To: David R. Brown

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Abstract: The free energy of magnetization is expressed as a fourth degree polynomial in the magnetization with a linear term due to an external field and quadratic and quartic terms due to Fermi, exchange, magnetostatic, etc., energies as well as to entropy effects.

Below the Curie temperature the free energy function has three real extrema, namely two minima and a maximum, if the absolute magnitude of the external field lies below a critical value. If the absolute magnitude of the external field exceeds this critical value, there is only one extremum, namely a minimum. When the absolute value of the field equals the critical value, there is a minimum and a point of inflection.

It is assumed that the magnetization of a system is always such as to minimize the free energy, but that the system is not always capable of settling in the lowest relative minimum of the free energy. On the basis of this model, the critical field and the corresponding magnetization are calculated as functions of parameters peculiar to the material and temperature, and experimental data obtained from hysteresis loops at various temperatures are presented.

Calculation of magnetization as a function of the external field results in a universal expression for the hysteresis loop, which is compared with some experimentally observed loops.

Introduction

It can be shown[1] that the internal energy of magnetization in the absence of an external field can be expanded in an even series in the magnetization

$$E = (A - B + C)\,M^2 + DM^4 + \ldots.$$

1. Slater: "Quantum Theory of Matter," McGraw-Hill Book Co., Inc. 1st Ed. (1951) Appendix 22, p. 515.

where: AM^2 is a term due to the Fermi energy; $A > 0$
BM^2 is a term due to the exchange energy; $B > 0$
CM^2 is a term due to the magnetostrictive, magnetostatic, etc. energies
DM^4 includes higher order terms in all these energies.

Similarly, the entropy can be expanded as follows:

$$S = -PM^2 - QM^4 - \ldots.; \quad P > 0, \; Q > 0$$

The free energy is then given by:

$$F = E - TS = (A - B + C + PT)M^2 + (D + QT)M^4 \qquad \text{(Equ. 1)}$$

which has extrema for the values

$$M = 0, \pm \sqrt{\frac{-A + B - C - PT}{2\,(D + QT)}}$$

When $T > \frac{A - B + C}{P}$ there is only one real minimum and the material is paramagnetic.

When $T < \frac{A - B + C}{P}$ there are two real minima separated by a maximum at $M = 0$. The system is in this case equally likely to assume a positive as a negative magnetization. In fact some regions may become positively magnetized, others negatively, so that the total magnetization is zero. Reversing the magnetization of a region requires the energy needed to pass over the maximum at $M = 0$. Since such reversal is improbable, the magnetization of anyone region remains constant during an observation, so that domain patterns are obtained for ferromagnetic, but not for paramagnetic materials. For the latter, there is no energy barrier; therefore, anyone region is equally likely to have a positive as a negative magnetization with reversal of magnetization occurring all the time. During the period of an observation anyone region in a paramagnetic material has therefore an average magnetization zero.

The transition from ferro- to paramagnetism occurs at a temperature $T = T_c$ given by:

$$T_c = \frac{A - B + C}{P}$$

This is by definition the Curie temperature. Substitution into equation 1 produces:

$$F = -P\,(T_c - T)M^2 + (D + QT)M^4 \qquad \text{(Equ. 2)}$$

When an external field H is applied to the system, the free energy is decreased by an amount HM, so that

$$F = -HM - P(T_c - T)M^2 + (D + QT)M^4 \qquad \text{(Equ. 3)}$$

Figures 1a, 1b, 1c, 1d, 1e, 1f, and 1g show the free energy plotted vs. magnetization for various values of the field strength.

In previously unmagnetized materials, the total volume of the domains with positive magnetization equals that of the domains with negative magnetization, the magnitude of each magnetization being given by the position of the minima in Figure 1d. When a positive field is imposed, the free energy changes as indicated by Figures 1c, 1b, and 1a. The positions of the minima are changed by the application of a field; a positive field causes both minima to shift in the positive direction, so that the volume of positively magnetized domains now exceeds that of negatively magnetized domains. Some motion of domain walls is thus required to maintain the system at minimum free energy. Figure 1b shows the free energy curve for the critical free strength $H = H_c$ where there is only one minimum as well as a point of inflection. When $H \geq H_c$, there is no energy barrier keeping the system from settling in the lower (now only) relative minimum. Therefore when the field strength increases from zero, a cataclysmic reversal of the domains with magnetization opposite in direction to the external field occurs when the field strength equals H_c. Further increase in field strength simply shifts the position of the minimum to the right, so that the average magnetization increases toward saturation.

A subsequent decrease in field strength decreases the average magnetization. When $H = H_c$ is reached, the free-energy curve again has the form shown in Figure 1b, and further decrease in field strength produces successively the curves in Figures 1c, 1d, and 1e. The magnetization of the system is now given by the position of the right-hand minimum, which is the lower of the two relative minima when $H > 0$, and the higher when $H < 0$. When $H < 0$ there is, therefore, a tendency for the system to reverse its magnetization, which is prevented by the energy barrier. As shown in Figure 1f, this barrier vanishes when $H = -H_c$, so that another cataclysmic reversal then takes place. Alternating the magnetic field with an amplitude greater than $|H_c|$ therefore produces a nonlinear variation in magnetization such as that described by the hysteresis loop. The upper branch of the loop corresponds to the right-hand minimum, hence to a decreasing field; the lower branch corresponds to the left-hand minimum and increasing field. When $|H|>|H_c|$, the magnetization curve is single valued because the free energy has only a single minimum.

Computation of the Critical Field and the Corresponding Magnetization

According to what has preceeded, the hysteresis curve can be calculated on the basis of the assumption that the magnetization has a value that minimizes the free energy. The values of magnetization that minimize the free energy are obtained by differentiating the free energy as given in equation 3 with respect to the magnetization and equating the result to zero:

$$M^3 - \frac{P(T_c - T)}{2(D + QT)} M - \frac{H}{4(D + QT)} = 0 \qquad \text{(Equ. 4)}$$

The discriminant[2] of this equation is:

$$\Delta = \frac{1}{2}\left[\frac{P(T_c - T)}{D + QT}\right]^3 - \frac{27}{16}\left[\frac{H}{D + QT}\right]^2$$

A double root, i.e. a point of inflection in the free energy curve, as shown in Figures 1b and 1f, occurs when this discriminant equals zero. Therefore the critical value of the field strength, H_c, is found by equating the discriminant to zero.

$$\therefore \quad H_c = \frac{\left[(2/3)\,P(T_c - T)\right]^{3/2}}{(D + QT)^{1/2}} \qquad \text{(Equ. 5)}$$

Substituting this value for H in equation 4 and solving the latter equation for M produces the value of the magnetization just after the cataclysmic reversal has occurred, which is called M_c:

$$M_c^{\,3} = \frac{H_c}{D + QT} = \left[\frac{2}{3}\,\frac{P(T_c - T)}{D + QT}\right]^{3/2} \qquad \text{(Equ. 6)}$$

From equations 5 and 6 can be derived the following two relations:

$$\frac{H_c}{M_c} = (2/3)\,P(T_c - T) \qquad \text{(Equ. 7)}$$

and

$$\frac{H_c}{M_c^{\,3}} = D + QT \qquad \text{(Equ. 8)}$$

By plotting experimentally obtained values of H_c/M_c and $H_c/M_c^{\,3}$ vs. T, one can calculate the coefficients P, D, and Q (See "Results and Comparison with Experimental Data).

2. "New First Course in the Theory of Equations" L. E. Dickson, J. Wiley & Sons, p. 46, 1939.

Computation of the Magnetization Curve

To find the magnetization for any value of the field strength, it is necessary to solve equation 4 for all values of H. Equation 4 can be rewritten by substituting for $P(T_c-T)$ and $(D + QT)$ the values given by equations 7 and 8:

$$M^3 - \frac{3}{4} M_c^2 M - \frac{1}{4} \frac{M_c^3}{H_c} H = 0$$

$$\left(\frac{M}{M_c}\right)^3 - \frac{3}{4}\left(\frac{M}{M_c}\right) - \frac{1}{4}\left(\frac{H}{H_c}\right) = 0 \qquad \text{(Equ. 9)}$$

The variables M/M_c and H/H_c express the magnetization and critical field strength as dimensionless quantities in units of the critical values M_c and H_c. This is just what is done experimentally when the oscilloscope picture is normalized. Equation 9 can therefore be said to represent a normalized hysteresis loop; the normalizing coefficients H_c and M_c depend on the temperature and the nature of the medium as expressed by equations 5, 6, 7, and 8, but equation 9 is universal, i.e. independent of the medium and of the temperature.

Equation 9 must be solved separately for the case $|H| \leq |H_c|$ and for the case $|H| \geq |H_c|$. For the former case, set $H/H_c = \cos 3A$. Then the three real solutions are:*

$$\frac{M}{M_c} = \cos A,\ \cos(A + 120^\circ),\ \cos(A + 240^\circ), \qquad \text{(Equ. 10)}$$

where the first two solutions correspond to free energy minima, the latter to the energy maximum. Since equation 9 contains no quadratic term, the sum of its three solutions equals zero, so that the energy maximum occurs at a value of the magnetization equal to the difference in absolute values of the magnetization corresponding to the energy minima.

When $|H| > |H_c|$, M/M_c is given by equation 11.

$$\frac{M}{M_c} = \frac{1}{2}\sqrt[3]{\frac{H}{H_c}}\left[\sqrt[3]{1 + \sqrt{1-\left(\frac{H_c}{H}\right)^2}} + \sqrt[3]{1 - \sqrt{1-\left(\frac{H_c}{H}\right)^2}}\right] \qquad \text{(Equ. 11)}$$

and hence approaches a cubic parabola asymptotically. When $H = \pm H_c$, $M = \pm M_c$ according to both equations 10 and 11.

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* This can be proven by direct substitution into equation 9, remembering that $\cos^3 x = (1/4)(\cos 3x + 3\cos x)$.

Results and Comparisons with Experiments

Figure 2 shows a plot of H_c/M_c vs. (T_c-T) using experimental values reported by Channing Morrison for a Mn-Mg ferrite (MF-1118, F-259).[3] Near the Curie point the horizontal and vertical gain are very large, so that failure to reach saturation is overstressed in this region. This results in too large values of H_c/M_c and accounts for the tendency of experimental points to veer away from the origin.

From Figure 2 it is found that $P = 8.34 \times 10^{-6} \frac{\text{erg}/\text{cm}^3/\text{degree}}{\text{gauss}^2}$. It is hoped that during the summer of 1953 sufficiently accurate measurements will be made so that H_c/M_c^3 can also be plotted in order to botain D and Q. It will be interesting to compare results of temperature dependent measurements for various materials.

In Figure 3 is drawn the hysteresis loop obtained by solving equation 9, and for comparison Figure 4 shows a loop obtained experimentally with molybdenum permalloy.

Conclusions

It has been shown that simple fundamental models for magnetism lead directly to a hysteresis loop which resembles many experimentally observed hysteresis loops. The calculated loop has the characteristic "Sloping shoulders," which have heretofore been explained by a rotation of domains. The present model neglects such rotation, and yet the sloping shoulder is apparent.

The very simple model employed has led to the universal equation 9 in much the same way as a simple kinetic gas theory led to the universal Boyle's law. Deviations from the universal law depend on the individual materials concerned. Some of the causes of deviations from the universal hysteresis loop will now be discussed. In the first place, the experimentally observed loop represents $d\phi/dt$ vs. I, and the finite size of the sample causes the sides of the loop to slope. Secondly, it has been assumed that reversal of magnetization occurs only when the energy barrier vanishes. Actually a "tunnel effect" allows reversal through the barrier, the speed (hence amount) of reversal being proportional to the negative magnitude of the barrier expressed in units of kT. Thus the sides of the loop do slope somewhat because the reversal can start somewhat below the critical field strength. Minor loops are obtained when the amplitude of the alternating field strength

3. Channing Morrison, E-491, "Hysteresis Loop Characteristics of MF-1118 for Different Temperatures," Digital Computer Laboratory, MIT.

is slightly less than the critical value; partial reversal then takes place through the barrier, but a good deal of the material remains captured behind the barrier, so that the resulting magnetization is less than on the saturation loop.

Figure 6 shows a "DC" loop. When the system is reversing its magnetization ($|H| \leq |H_c|$) the magnitude field strength is suddenly decreased in magnitude, so that an energy barrier is reintroduced. The material that had not yet reversed its magnetization is then caught again, but the material that was already reversed is now in the lower relative minimum, and hence will not reverse again. Therefore the magnetization remains essentially constant until the magnitude of the field strength is once more increased to the critical value, when more reversal can take place.

Thirdly, only two directions of magnetization have been assumed, namely positive and negative. Actually a range of 360° with respect to the direction of the external field is possible, and Figure 1 should be three dimensional with a free energy surface analogous to Eyring surfaces in reaction kinetics. The system might reverse its magnetization by going around the energy barrier rather than over the top; this would amount to rotation of the magnetic vector. For directions of easy magnetization, the free energy surface would have a pit surrounded by barriers; when the material is isotropic, Figure 1d would only need to be rotated about its axis of symmetry to produce the free energy surface. Thus the hysteresis loop could be determined by the motion of a particle in a complicated system of mountains, crevices and passes, with deviations from a straight path representing rotation. The possibility of rotation would make the loop less square than the one shown in Figure 3. Further it has been assumed that the only force opposing reversal of magnetization is that due to the free energy barrier, and that the reversal occurs instantaneously. Frictional forces, i.e., forces that are not simply derived from a potential as assumed in equation 1, have not been taken into account.

Finally, it is assumed that the system can always be in a state of lowest free energy. Actually "supersaturation" may occur, for the mechansim of reversal of magnetization requires formation of domains of reverse magnetization just as precipitation form solution requires nuclei of condensation to keep the solution from being supersaturated.[4] The

4. J. B. Goodenough, E-532, "Nucleation of Domains of Reverse Magnetization & Switching Characteristics of Magnetic Materials," Digital Computer Laboratory, MIT.

third and fourth factors tend to make the loop more square than that shown in Figure 3. Figure 5 shows an experimentally observed loop with "double shoulders." The shoulder can be explained as follows: The observed hysteresis loop is a combination of two loops. The outer loop represents one in which reversal of magnetization is inhibited by the lack of domains of reverse magnetization. The inner loop represents the free energy or equilibrium hysteresis loop of Figure 3. When the absolute value of the field reaches a critical value, domains of reverse magnetization are formed, so that the magnetization drops down to the equilibrium loop.

Signed Arthur L. Loeb
Arthur L. Loeb

Approved DRB
David R. Brown

ALL/jk

Drawings attached:

Figure 1	B-55551
Figure 2	A-54663
Figure 3	A-54665
Figure 4	"
Figure 5	A-55564
Figure 6	A-55557

cc: Group 63 Staff

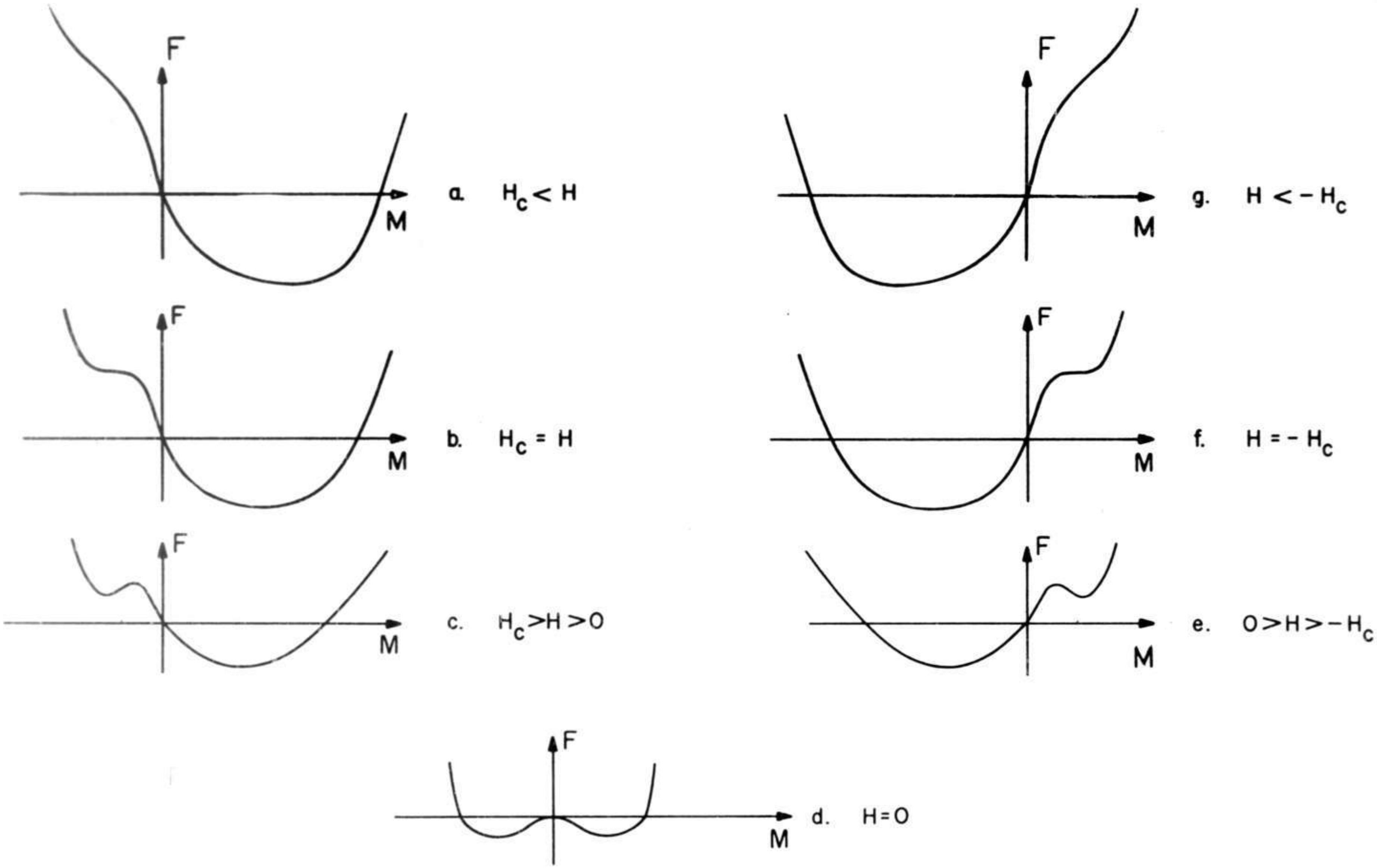
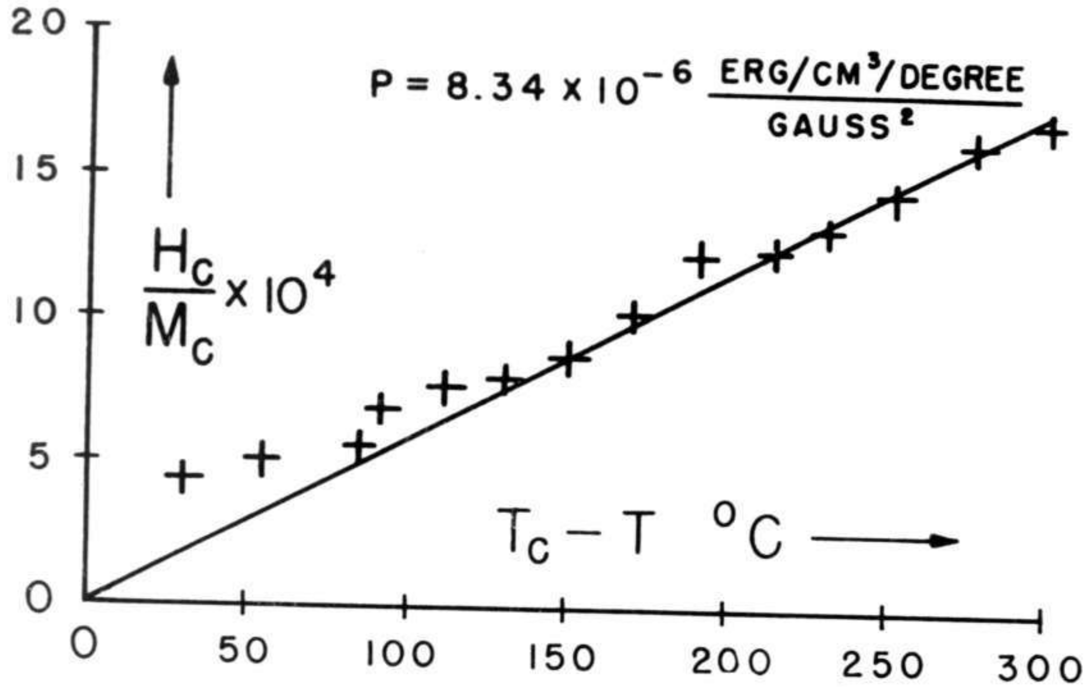


FIG. 1
FREE ENERGY OF MAGNETIZATION IN AN EXTERNAL FIELD

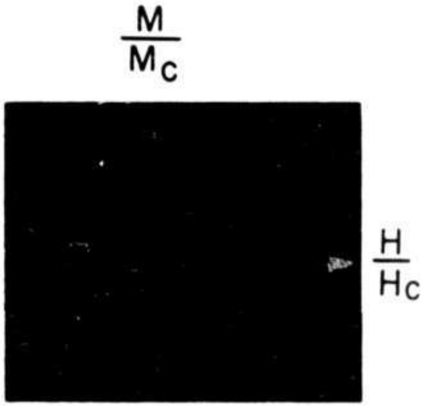
A-55551

A-54663
F-1896
SN-547



EXPERIMENTAL VERIFICATION AND DETERMINATION OF ENTROPY COEFFICIENT

FIG. 2



CALCULATED

FIG. 3

OBSERVED

FIG. 4

REDUCED HYSTERESIS LOOP

A-54665
F-1898
SN-549

A-55564

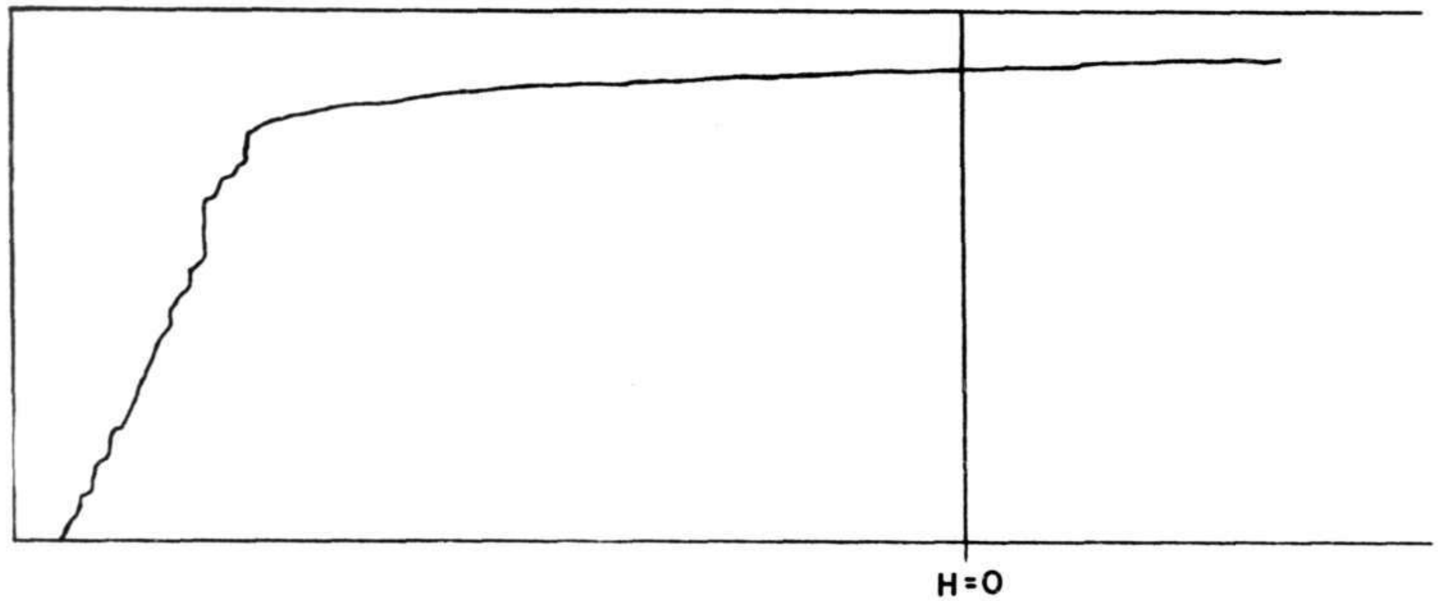


FIG. 5

PORTION OF A DC HYSTERESIS LOOP WITH MULTIPLE SHOULDER

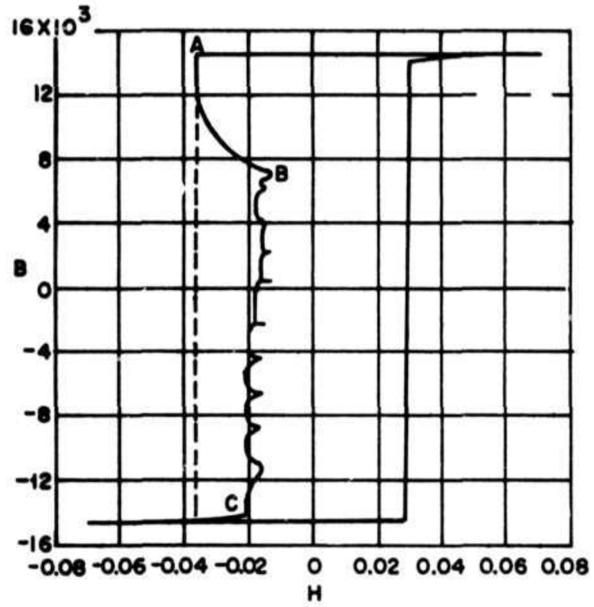


FIG. 6

HYSTERESIS LOOP OF PERMINVAR

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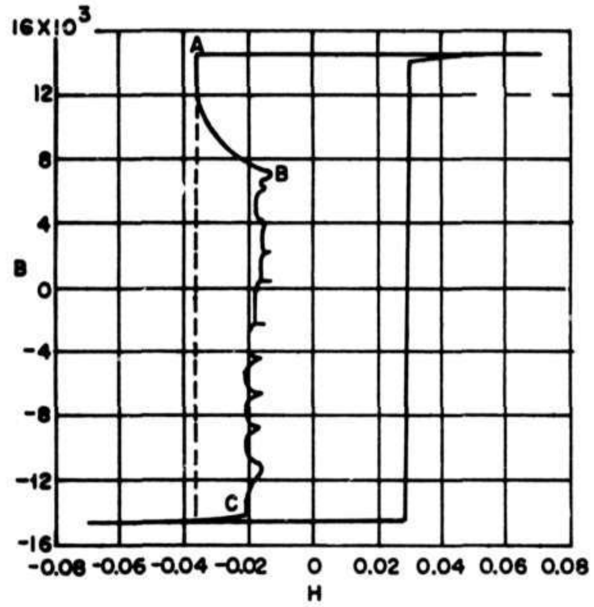

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HYSTERESIS LOOP OF PERMINVAR

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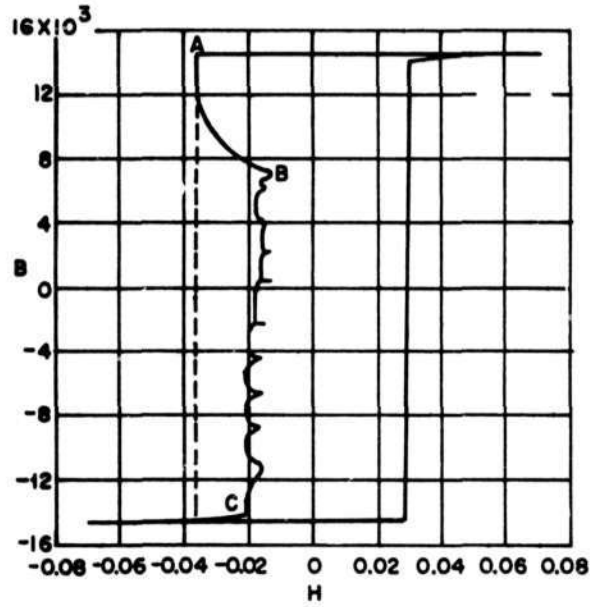

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