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Criterion for Ferromagnetism from Observations of Magnetic Isotherms*

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A criterion is proposed for determining the onset of ferromagnetism in a material as its temperature is lowered from a region in which the linearity of its magnetic moment *versus* field isotherm gives an indication of paramagnetism. Within the limits of validity of a molecular field treatment, the Curie temperature is shown to be in general indicated by the third power of the magnetization being proportional to the internal magnetic field. The method has been employed to redetermine the Curie point of nickel from the data of Weiss and Forrer, of Fe_3O_4 from the data of Smith and of some alloys from the data of Kaufmann and his collaborators and the author.

INTRODUCTION

By definition, a material is ferromagnetic if there can exist regions within the material in which a spontaneous magnetization exists; i.e., the magnetic moment per unit volume M is solely responsible for the magnetic induction B or, in Gaussian units, $B=4\pi M$ or, in rationalized mks units, $B=\mu_0 M$. Equivalently a material is ferromagnetic if it can possess regions of finite magnetization where the magnetic field intensity H is equal to zero within those regions. The temperature below which ferromagnetism occurs is called the Curie temperature and is a measure of the interaction energy associated with the ferromagnetism. The determination of the Curie temperature is not without ambiguity. Conceptually it appears easier to discuss the onset of ferromagnetism on decreasing temperature than the disappearance of ferromagnetism as the temperature is increased. This is because of the many complexities of the cooperative phenomena, in particular domain walls. We restrict the discussion to materials which when made in the form of an ellipsoid of revolution and placed in a uniform applied field, say from a solenoid, are uniformly magnetized when above their Curie temperatures. In such a case the field intensity inside the ellipsoid H_i will be given by (in Gaussian units)

$$H_i=H_0-4\pi DM,$$

where H_0 is the intensity of magnetization in the solenoid in the absence of the sample, D is the demagnetizing factor, and M the magnetic moment per unit volume. The susceptibility is given by

$$\chi=\frac{M}{H_i}=\frac{M}{H_0-4\pi DM},$$

which by the definition above must go to infinity as the temperature is lowered to the Curie temperature. To determine the Curie temperature it is necessary to measure H_0 and $4\pi DM$. If, however, H_0 is finite, $4\pi DM$ will equal H_0 only at some temperature below the Curie temperature, for it is only in the limit of $H_0=0$ that the true Curie point is found. There are two objections to obtaining a Curie point by going to zero field in this way. One is that, apart from the measurements of Smith[1] on Fe_3O_4 and some the author has made on alloys,[2] Curie point determinations have been carried out in finite fields which are in general greater than $H_0/4\pi M_0=\frac{1}{10}$, M_0 being the saturation magnetization at absolute zero. Thus, a true spontaneous magnetization is observed only at temperatures somewhat below the Curie point. The other objection is that the limit of zero field can be obscured by the presence of ferromagnetic impurities with a higher Curie point or by inhomogeneities in the case of alloys and compounds. It would be desirable, therefore, in magnetic analysis to have a criterion based on measurements at finite fields.

The criterion proposed here follows from considerations of the field dependence of the magnetization and is valid in the limits within which this dependence can be represented as

$$H_i=(1/\chi_0)M+\beta M^3+\gamma M^5+\cdots. \tag{1}$$

At the Curie point, by definition, $1/\chi_0=0$ so that in finite fields

$$H_i=\beta M^3+\gamma M^5+\cdots. \tag{2}$$

Inasmuch as the βM^3 term must dominate at low enough fields the criterion proposed is that at the Curie temperature the third power of the magnetization is proportional to the internal field. Though in this paper the demonstration is in terms of plots of M^3 *vs* H, in practice we plot H/M *vs* M^2 as this gives $1/\chi$ in the limit of zero field as the intercept on the H/M axis when extrapolated to $M^2=0$. Not only does the temperature at which $1/\chi$ goes to zero determine the Curie temperature but the slope of $1/\chi$ *vs* temperature gives a measure of the magnetic moment per atom of

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[1] D. O. Smith, Phys. Rev. **102**, 959 (1956); Rev. Sci. Instr. **27**, 261 (1956).

[2] A. Arrott and J. E. Goldman, Rev. Sci. Instr. **28**, 99, 1957; *Second Conference on Magnetism and Magnetic Materials* AIEE, 1957 (to be published), p. 285 and p. 516; A. Arrott, Ph.D. thesis, Carnegie Institute of Technology, 1954 (unpublished).

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the material. In addition, the coefficient of the M^3 term which is determined from the slope of either the M^3 *vs* H or M^2 *vs* H/M plot gives information as to what models of magnetization are most suitable for describing the results.

A. Validity of Criterion

The dependence of the internal field H on the third power of the magnetization will first be shown for the Weiss-Brillouin molecular field treatment and then shown to be true for a generalized molecular field treatment which includes collective electron models.

In the Weiss-Brillouin treatment the magnetization is given by

$$M=M_0\tanh\left(\frac{\mu(H+NM)}{KT}\right), \tag{3}$$

where M_0 is the spontaneous magnetization at absolute zero, μ is the moment per atom, and N is the molecular field constant. Rewriting gives

$$\frac{\mu H}{KT}+N\frac{\mu M}{KT}=\tanh^{-1}\left(\frac{M}{M_0}\right).$$

The right-hand side can be expanded in a power series for values of $M\ll M_0$ giving

$$\frac{\mu H}{KT}+N\frac{\mu M}{KT}=\frac{M}{M_0}+\frac{1}{3}\left(\frac{M}{M_0}\right)^3+\frac{1}{5}\left(\frac{M}{M_0}\right)^5+\cdots, \tag{4}$$

for which the inverse susceptibility, $1/\chi$, in the limit of zero field is given by

$$1/\chi=(KT/\mu M_0)-N \tag{5}$$

from which the Curie point $(1/\chi=0)$ is given by $T_c=(\mu N/K)M_0$. Hence at the Curie point

$$\frac{\mu H}{KT_c}=\frac{1}{3}\left(\frac{M}{M_0}\right)^3+\frac{1}{5}\left(\frac{M}{M_0}\right)^5+\cdots, \tag{6}$$

which shows the cubic nature of the relation between field and magnetization.

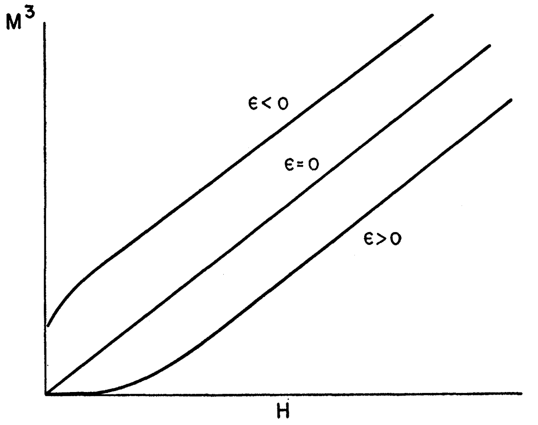

FIG. 1. Magnetization cubed *versus* field for temperatures just below the Curie point ($\epsilon<0$), at the Curie point ($\epsilon=0$), and just above the Curie point ($\epsilon>0$).

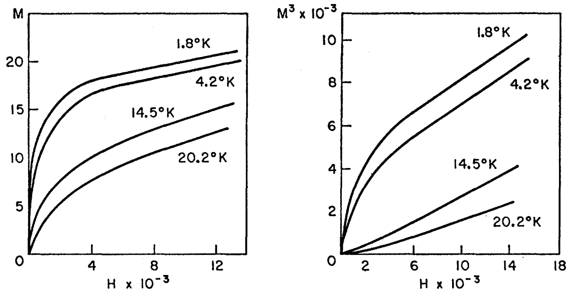

FIG. 2. Magnetization of 58% copper in nickel (data of Arrott and Goldman[2]).

Above or below the Curie point the magnetization depends on field as

$$\frac{\mu H}{KT}=\epsilon\frac{M}{M_0}+\frac{1}{3}\left(\frac{M}{M_0}\right)^3+\frac{1}{5}\left(\frac{M}{M_0}\right)^5+\cdots, \tag{7}$$

where

$$\epsilon=(1-T_c/T). \tag{8}$$

This relationship is illustrated in Fig. 1 which shows M^3 *vs* H for several values of ϵ.

The more general proof of this relationship depends on the existence of something mathematically equivalent to an effective field, H_{eff}. As long as the magnetization reverses without change in magnitude when H_{eff} is reversed, the magnetization is an odd function of H_{eff} and vice versa. Hence, for small values of M, we can write

$$\mu H_{\text{eff}}/KT=\alpha M+\beta M^3+\gamma M^5+\cdots. \tag{9}$$

H_{eff} is by definition $-\partial E/\partial M$, where E is the total energy of the material. The energy may be written as

$$E=E_0-MH+f(M), \tag{10}$$

where E_0 represents all the nonmagnetic contributions to the energy; MH is the energy of the material in the field; and $f(M)$ is the internal energy due to magnetization. As long as the internal energy due to magnetization does not change on reversal of magnetization, $f(M)$ is an even function of M. Hence

$$H_{\text{eff}}=-\partial E/\partial M=H+f'(M), \tag{11}$$

where $f'(M)$ is then an odd function of M, and can be written as

$$f'(M)=aM+bM^3+cM^5+\cdots, \tag{12}$$

combining Eqs. (9), (11), and (12) gives for the Curie point $T_c=\mu a/K\alpha$ and for the H near T_c

$$H=\epsilon M+AM^3+BM^5+\cdots, \tag{13}$$

where

$$\epsilon=[(T/T_c)-1]a.$$

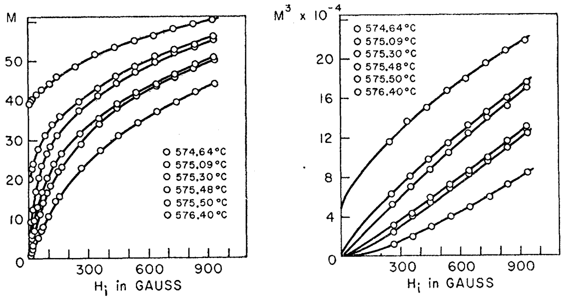

FIG. 3. Magnetization of Fe_3O_4 (data of Smith[1]).

B. Some Applications to Existing Data

The application of this criterion to some data of the author is shown in Fig. 2. In this case the copper-nickel alloy in question is in the critical range of the transition from ferromagnetism to nonferromagnetism as a function of composition. The strong curvature in the plots of M *vs* H is suggestive of ferromagnetism at all four temperatures shown. Measurements were carried out on these samples in applied fields as low as 20 oe which showed a magnetization did exist in zero internal field for 1.8°K and 4.2°K but not for 14.5°K and 20.2°K. The plot of M^3 *vs* H shows agreement that the Curie point lies between 4.2°K and 14.5°K. Using an M^2 *vs* H/M plot to obtain $1/\chi$ and then extrapolating $1/\chi$ *vs* T gives the Curie point near 11°K.

When applied to the data of Smith on Fe_3O_4, the comparison between the Curie temperature as determined by the M^3 *vs* H plot (see Fig. 3) with the value determined from measurements in very low field shows a disagreement of 0.1°C. This small disagreement is associated with slight deviations from expected behavior below 200 oe. Both criteria agree that the 575.09°C isotherm is ferromagnetic. The disagreement about the 575.30°C isotherm could be linked to the fact that the magnetization curves are not quite isotherms due to magnetocaloric effects.

An example of the usefulness of this criterion is provided by the data of Kaufmann *et al.*[3] on dilute alloys of iron in gold. The 3.2% Fe alloy, for instance, is assigned by Kaufmann *et al.* a Curie temperature of well over 100°K. The magnetization curves for 14°K, 20°K, and 63°K are shown in Fig. 4 together with a plot of σ^3 *vs* H. The Curie point of over 100°K was arrived at by extrapolating the high field data to zero field as shown by the dashed curve to obtain the "spontaneous magnetization" and then extrapolating the "spontaneous magnetization" *vs* temperature. From the σ^3 *vs* H plot it is obvious that 63°*K* is well above the Curie point. Uncertainty in the 5-kilogauss data leaves unclear whether or not there is even a small spontaneous magnetization at the lower temperatures. It is likely that this alloy reported to have a Curie temperature over 100°K is not ferromagnetic at all.

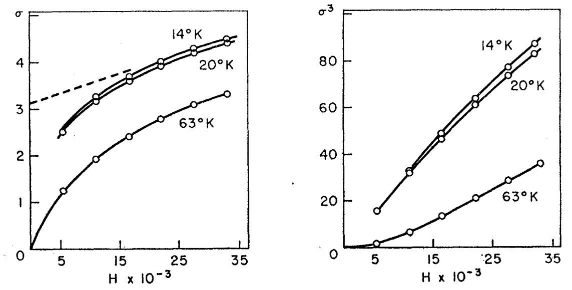

FIG. 4. Magnetization of 3.2% iron in gold (data of Kaufmann *et al.*[3]).

This discussion would not be complete without a consideration of the classic investigation of Weiss and Forrer on nickel.[4] As Weiss and Forrer arrive at their Curie point by use of the molecular field theory applied to their measurements above 10^4 oe, their value of the Curie point, 358°C, agrees exactly with the value found by applying the M^3 *vs* H criterion to the data in those high fields. They essentially ignore their lower field data in that they use extrapolation from high fields of magnetocaloric effect data and replot the magnetization data in the form of lines of constant M on a plot of H *vs* T. If the M^3 criterion is applied to the low-field data, the Curie point should be set at 354°C (see Fig. 5). One could conclude that either the Curie temperature is lower than set by Weiss and Forrer or else there is something very peculiar about the low-field data.

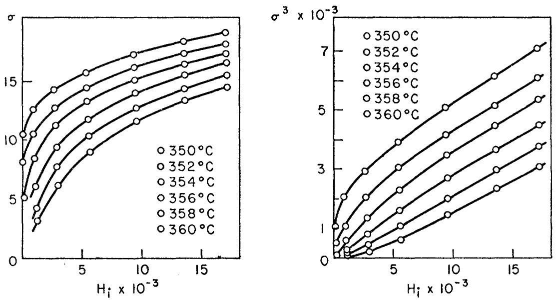

FIG. 5. Magnetization of nickel (data of Weiss and Forrer[4]).

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[3] Kaufmann, Pan, and Clark, Revs. Modern Phys. **17**, 87 (1945).

[4] P. Weiss and R. Forrer, Ann. phys. **5**, 153 (1926).