# Shape of Temperature Dependence of Spontaneous Magnetization of Ferromagnets: Quantitative Analysis

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Analysis of available experimental data shows that there exists a limited variety of shapes of temperature dependence of spontaneous magnetization. For most metallic ferromagnets the shape (as opposed to scale) of the $M_s$ versus $T$ curve can be characterized by a single dimensionless parameter. A numerical description of the dependence $M_s(T)$ for a particular ferromagnetic material is thus reduced to evaluating three quantities: the saturation magnetization $M_0 \equiv M_s(0)$, the Curie point $T_C$, and the shape parameter $s$. It is demonstrated that classical spin ($S = \infty$) dynamics fails to describe correctly either of the finite-temperature characteristics, $T_C$ or $s$.

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Spontaneous magnetization $M_s$ is the most fundamental property of a ferromagnet. Not surprisingly a lot of effort has been spent over the last century on attempts to describe theoretically $M_s$ as a function of temperature, between zero Kelvin and the Curie point $T_C$, where $M_s$ vanishes. At present only the problem of evaluating saturation magnetization $M_0$, that is $M_s(T = 0)$, can be considered solved; calculations based upon the density functional theory (DFT) produce values of $M_0$ which are consistently in good agreement with experiment [1]. There have also appeared many reports of $T_C$ calculations (see, e.g., Ref. [2], also Ref. [3], and references therein) employing a combination of DFT and Langevin's spin dynamics, based on the classical Heisenberg model. The classical ($S = \infty$) approximation is examined at some length later and found inapplicable, in particular, to $T_C$ calculations.

However, the main subject of this Letter is the shape, rather than the scale, of temperature dependence of spontaneous magnetization. To study the shape of $M_s(T)$ in its pure form, it is convenient to introduce reduced spontaneous magnetization, $m \equiv M_s/M_0$, and reduced temperature, $\tau \equiv T/T_C$. Consider the following question: How various are the observed forms of $m(\tau)$? That there is no universal function $m(\tau)$ valid for all ferromagnets (the so-called Law of Corresponding States) was established experimentally over half a century ago [4,5]. If so, how many extra parameters are needed to fully describe the variety of existing shapes of $m(\tau)$?

An immediate answer is given by the molecular field theory: $m(\tau)$ depends on a single dimensionless parameter. In the localized, Weiss-Brillouin approach, this parameter is the relevant atomic quantum number, $S$ or $J$. In the itinerant version, due to Stoner, it is the ratio of the exchange to the Fermi energy. (An excellent description of both cases can be found in Morrish's textbook [6], whereas Aharoni's monograph [7] contains rather accurate explicit expressions for $m(\tau)$ obtained in the Weiss-Brillouin approach with $1/2 < S < 7/2$.) However, one cannot be fully satisfied with this answer since the molecular field theory does not describe the shape of $m(\tau)$ correctly.

There is no general analytical expression for $m(\tau)$ beyond the molecular field approximation, except in the two limiting cases, $\tau \rightarrow 0$ and $\tau \rightarrow 1$, although it has been recently demonstrated [8,9] that an accurate description of $m(\tau)$ in the entire interval $0 < \tau < 1$ is provided by a combination of two (in some cases, three) simple power laws, one for each of the temperature subintervals.

To advance the matters further, we propose to present the function $m(\tau)$ in the following form:

$$m(\tau) = [1 - s\tau^{3/2} - (1 - s)\tau^{p}]^{1/3}, \tag{1}$$

where $s$ and $p$ are parameters, $p > 3/2$, $s > 0$. Equation (1) is constructed to obey Bloch's 3/2 power law at low temperatures, $m \approx 1 - \frac{1}{3}s\tau^{3/2}$ as $\tau \rightarrow 0$, whereas in the critical region, $\tau \rightarrow 1$, $m$ is proportional to $(1 - \tau)^{1/3}$, as prompted by the critical behavior of the Heisenberg model [10].

TABLE I. Characteristics of temperature dependence of spontaneous magnetization of ferromagnets: saturation magnetization $M_0$, Curie temperature $T_C$, parameters entering Eq. (1), $p$ and $s$. The $T_C$ values are those used to normalize the data presented in Fig. 1; they do not necessarily coincide with the values given in the cited references.

| Compound | $M_0$ (emu/g) | $T_C$ (K) | $p$ | $s$ | Source |
|---|---|---|---|---|---|
| Fe | 222 | 1044 | 4 | 0.35 | Ref. [11] |
| Co, hcp | 164 | 1385 | 5/2 | 0.11 | Refs. [4,12] |
| Co, fcc | 166 | 1385 | 5/2 | 0.11 | Ref. [5] |
| Ni | 57.6, 58.6 | 628, 631 | 5/2 | 0.15 | Refs. [11,13] |
| Gd | 268 | 291 | 5/2 | 1.3 | Ref. [14] |
| $YCo_5$ | 124 | 930 | 5/2 | 0.7 | Ref. [15] |
| $Y_2Fe_{17}$ | 170 | 312 | 5/2 | 0.6 | Ref. [16] |
| GdZn | 186 | 270 | 5/2 | 1.9 | Ref. [17] |

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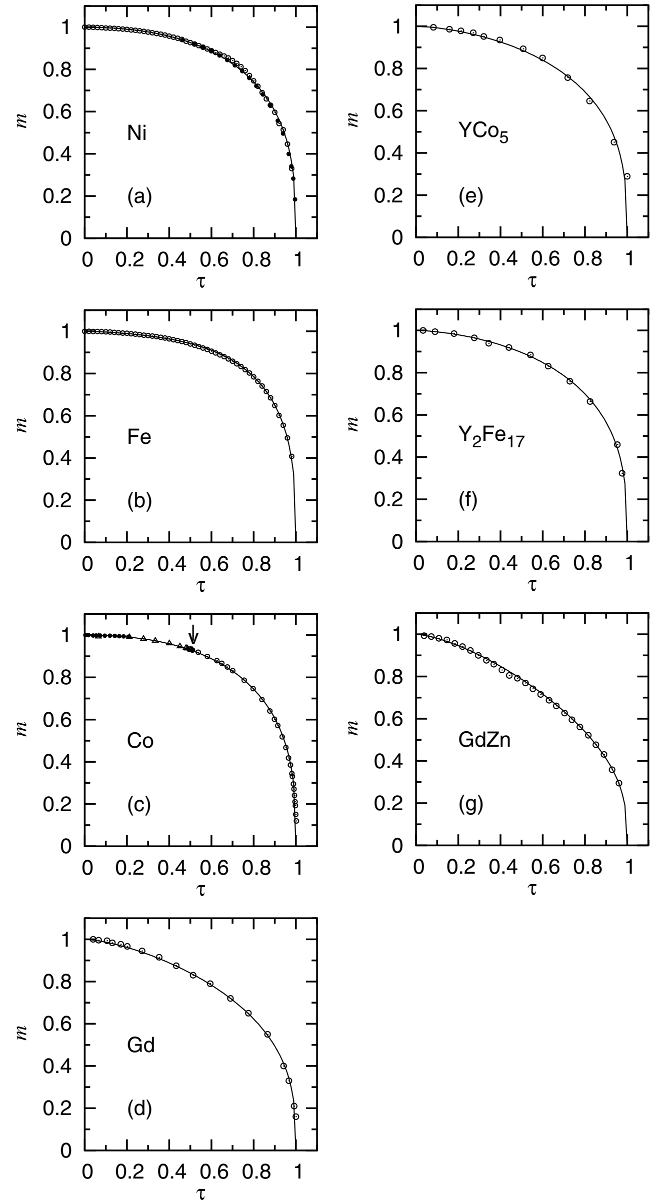

FIG. 1. Reduced spontaneous magnetization versus reduced temperature for several ferromagnets. The continuous lines were calculated using Eq. (1) with the $p$ and $s$ of Table I. The symbols are experimental data points from (a) Refs. [11] (○) and [13] (●); (b) Ref. [11]; (c) Refs. [5] (○), [12] (●), and [4] (△); (d) Ref. [14]; (e) Ref. [15]; (f) Ref. [16]; (g) Ref. [17]. The arrow marks the hcp-fcc transition point in cobalt.

Available experimental data were fitted to Eq. (1). The best-fit parameters are listed in Table I and the corresponding curves are shown in Fig. 1.

It turns out that all but one of the considered ferromagnets are described by Eq. (1) with $p = 5/2$. This result is not quite unexpected; it reminds us of Dyson's low-temperature expansion [18] for the quantity $m^3$, truncated after the third term. The only exception from this rule is bcc iron, which obeys Eq. (1) with $p = 4$. Setting aside this exception, one can say that the entire variety of observed $m(\tau)$ dependences can be characterized by a single parameter—the shape parameter $s$.

At this stage Eq. (1) should be regarded as an empirical expression; we are unable to strictly explain it or the fact that $p = 5/2$ in most cases.

We stress, however, that Eq. (1) pretends to describe the experimental $m(\tau)$ dependence as a whole, not just the asymptotics at $\tau \rightarrow 0$ or $\tau \rightarrow 1$. Small details of this dependence are often sample specific and sometimes can be traced back to impurities. A didactic example is Ni [Fig. 1(a)]. The more recent and more detailed data of Crangle and Goodman [11] (open circles) deviate from Eq. (1) (continuous line) around $\tau = 0.7$. However, the earlier data of Weiss and Forrer [13] (filled circles) show no such anomaly and comply with Eq. (1) everywhere. We therefore deliberately avoid considering any experimental data other than those obtained on stoichiometrically pure single crystals. It is interesting that the genuine anomaly associated with the hcp-fcc transition in cobalt is not visible on the scale of Fig. 1(c). Note that two slightly different values of $M_0$ were used to normalize the data above and below the transition point (indicated with an arrow) as could be reasonably expected from two structurally distinct phases.

Thus, the problem of describing temperature dependence of spontaneous magnetization is reduced to evaluating three quantities: two scale factors, $M_0$ and $T_C$, and one shape parameter $s$. Making use of the classical spin-wave theory [19], the latter can be expressed as follows:

$$s = \frac{3}{8}\pi^{-3/2}\zeta\left(\frac{3}{2}\right)\frac{\mu_B}{M_0}\left(\frac{kT_C}{D}\right)^{3/2} = 0.176\frac{\mu_B}{M_0}\left(\frac{kT_C}{D}\right)^{3/2}, \tag{2}$$

where $\zeta(x)$ stands for Riemann's $\zeta$ function, $\zeta(\frac{3}{2}) \approx 2.612$ [20], and $D$ is spin-wave stiffness (the coefficient in the parabolic magnon dispersion relation, $\hbar\omega = Dq^2$, valid in the limit $q \rightarrow 0$). Both $M_0$ and $D$ are ground state properties, and as such they should be described well by the DFT. This is particularly true about $M_0$ [1], the situation for $D$ being somewhat less satisfactory [2,3]. The discrepancies in the latter case could be accounted for, partially at least, by the errors made when deducing $D$ from both experimental and calculated dispersion curves. The fact that agreement with experiment is very good at larger $q$ [21] leaves room for optimism.

As regards Eq. (2) itself, it seems to relate $s$ and $D$ correctly, insofar as the available data allow us to judge; see Table II. Here, once again, iron is a notable exception, its $s_{\rm calc}$ being less than one-half of the corresponding "experimental" value from Table I.

Unlike $M_0$ and $D$, Curie temperature is not a ground state property, so no quantitative description of $T_C$ can be reasonably expected from DFT alone. As a first application of the obtained formula (1), let us demonstrate that DFT combined with classical spin dynamics, at present the most commonly used technique of evaluating $T_C$ [2,3], is grossly inadequate as an approach to describing $M_s(T)$ near $T_C$ [26]. To this end, setting $p = 5/2$ in Eq. (1), we write for the cube of the reduced magnetization the following asymptotic expression, which applies when $T \longrightarrow T_C - 0$:

$$m^3 = \left(\frac{5}{2} - s\right)\left(1 - \frac{T}{T_C}\right). \tag{3}$$

Since the prefactor in Eq. (3) must remain positive, the parameter $s$ has an upper bound of $5/2$. It has already been stated that $s$ must be positive, so that Bloch's $3/2$ power law is complied with as $\tau \longrightarrow 0$. Therefore,

$$0 < s < 5/2. \tag{4}$$

It now follows that the prefactor in Eq. (3) cannot be greater than $5/2$. Since there is no *a priori* physical reason limiting the slope of $m^3(\tau)$ near the Curie point, it is clear that there may exist exceptional cases not described by Eqs. (3) and (4), in which it was assumed that $p = 5/2$. A higher value of $p$ must be adopted then. Bcc iron apears to be such a case.

Now, if the classical spin dynamics was valid in the vicinity of the Curie point, it should not just yield the horizontal intercept of the linear $m^3$ versus $T$ dependence (3), i.e., the $T_C$. It must also reproduce correctly the slope of the line and hence the value of the shape parameter $s$. By virtue of Eq. (1) this would uniquely determine spontaneous magnetization as a function of temperature.

Figure 2 presents as $m^3$ versus $\tau$ the data points and fit of Fig. 1(a) for Ni. The points with error bars are the results of a Monte Carlo simulation [2] which employed the classical Heisenberg model. These points lie approximately on a straight line, as could be expected from the in principle exact treatment of the Heisenberg model. However, the slope of this line—the dotted line in Fig. 2—equals $-1.2$. According to Eq. (3) this corresponds to $s = 1.3$—a far cry from $s = 0.15$ obtained by fitting the experimental data. Taking into consideration that $s$ must be contained within the finite interval (4), one is forced to the following conclusion: the shape of $m(\tau)$ could not have been misrepresented more.

This should hardly come as a surprise; one may recollect that in the simple molecular field theory [6] the two extreme $m(\tau)$ curves are exactly those corresponding to $S = \infty$ (classical) and $S = 1/2$ (more relevant to Ni), their shapes being conspicuously distinct. As regards $T_C$, its dependence on $S$ for the Heisenberg model with nearest-neighbor exchange is known quite accurately from the high-temperature series. The relevant dimensionless quantity is $kT_C/JS^2 = \frac{55}{96}(Z - 1)(1 + 1/S - 1/11S^2)$, $Z$ being the lattice coordination number [27]. Here the difference between the $S = 1/2$ and the classical ($S = \infty$) cases is more than a factor of 2.6. There is little reason to expect that beyond the nearest-neighbor approximation this difference should become negligible. The only (if any) justification for using the classical approximation in finite-temperature calculations is that it allows one to avoid the awkward question of what should be taken for the atomic spin $S$ in itinerant ferromagnets. Of course, this convenience does not make the approximation itself quantitatively any better [28].

Talking about further possible uses of Eq. (1), one should mention a model description of $3d$-$4f$ intermetallic systems. An important component here is magnetization of the $3d$ sublattice, hard to determine experimentally. Note that the two representatives of this class of compounds in Table I, $YCo_5$ and $Y_2Fe_{17}$, have close shape parameters, $s = 0.7$ and 0.6, respectively. Such values of $s$ may be

TABLE II. Quantities used in conjunction with Eq. (2): spin-wave stiffness $D$ and saturation magnetization $M_0$. The latter data were taken from Table I and converted into the units of volume magnetization, so as to obtain dimensionless shape parameters. These are given in the last column and should be compared with the values of $s$ in Table I, obtained by fitting experimental data. The $T_C$ values used in the calculations were those given in Table I.

| Metal | $D$ (meV Å$^2$ | $M_0$ (kG) | $s_{\mathrm{calc}}$ |
|---|---|---|---|
| Ni | 400[a] | 0.51 | 0.16 |
| Fe | 307[b] | 1.75 | 0.15 |
| Co, hcp | 510[c] | 1.45 | 0.13 |
| Gd | ~20[d] | 2.12 | 1.1 |

[a] Reference [22].
[b] Reference [23].
[c] **q** ∥ [001], Ref. [24].
[d] Estimated from Fig. 1 of Ref. [25], **q** ∥ [001].

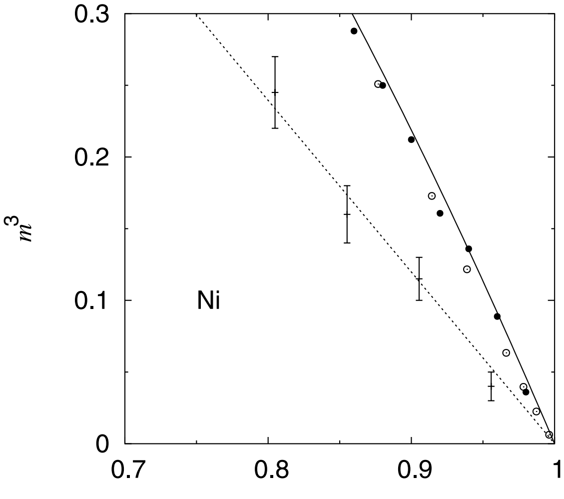

FIG. 2. Data points and fit of Fig. 1(a), presented as $m^3$ versus $\tau$. Shown as points with error bars are the Monte Carlo results of Ref. [2]. The dotted line corresponds to $m^3 = 1.2 \times (1 - \tau)$.

characteristic of iron- and cobalt-rich intermetallics in general [29]. The format of this Letter precludes us from proceeding any further in this direction.

To summarize, in an attempt to quantify the shape of temperature dependence of spontaneous magnetization, we have proposed for it a simple analytical representation, Eq. (1). All metallic ferromagnets for which appropriate experimental data are available obey this formula with the same exponent $p = 5/2$. The only exception is bcc iron, which also follows Eq. (1), but with $p = 4$, $s = 0.35$. With the aid of Eq. (1) it has been demonstrated that classical spin dynamics is inadequate as an approach to describing finite-temperature properties of ferromagnets. It would appear interesting to test the validity of Eq. (1) for a broader variety of ferromagnets, for which proper experimental data are presently not available.

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[26] It is of course also invalid at low $T$ where, in violation of the Third Law of Thermodynamics, it results in $M'(T = 0) \neq 0$; see Figs. 6–8 of Ref. [2].

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[28] The use of the classical approximation in the spin-wave theory is beyond reproach, since there it is limited to long wavelengths, $q \rightarrow 0$.

[29] For simplicity we regard $YCo_5$ and $Y_2Fe_{17}$ as one-sublattice ferromagnets, making no distinction between the Co or Fe atoms occupying crystallographically nonequivalent sites. Validity of this approximation is not immediately obvious and needs to be established experimentally. However, the fact that no visible anomaly of $m(\tau)$ accompanies the hcp-fcc transition in cobalt, Fig. 1(c), speaks in favor of such a simplification.