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Temperature dependence of the spontaneous magnetization of body-centred-cubic bulk iron, amorphous iron and thin iron films

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Abstract

It is shown that the thermal decrease of the spontaneous magnetization of iron differs characteristically depending on the preparation conditions. In all cases, the deviations from saturation at absolute zero can be accurately described by a single temperature power term T^ε which holds up to several hundreds of kelvins. The empirical exponents are $\varepsilon = 2$ for crystalline bulk iron, $\varepsilon = 3/2$ for amorphous iron as well as for isotropic (crystalline) two-dimensional iron films, but $\varepsilon = 5/2$ for inhomogeneous (pseudomorphic) monatomic layers. In crystalline films which are sufficiently thick (~ 100 – 300 nm) to allow the excitation of standing spin waves, the T^2 bulk law is confirmed for the uniform precession mode, but for states with a periodically modulated magnetization due to spin wave resonance, a $T^{3/2}$ dependence holds. For ultrathin iron films with more than three atomic layers, a thermodynamic crossover from $T^{3/2}$ to T^2 is observed at higher temperatures. This is indicative of a dimensionality change of the relevant interactions from 2 to 3. Crystalline and amorphous bulk iron also exhibit a crossover, but only for the amplitude and not for the exponent of the T^ε law. This is assumed to be caused by the thermal variation of the magnetic interaction strength. All observed empirical exponents ε fit into a recently proposed scheme of universality classes for the low-temperature behaviour of the order parameter in materials with half-integer spin quantum number and three-, two- and one-dimensional interactions. It can therefore be concluded that the thermodynamics of iron is like that of an insulating material with $s = 1/2$.

1. Introduction

In 1929 Weiss and Forrer observed that in the temperature range $100 \text{ K} < T < 290 \text{ K}$ the thermal decrease of the spontaneous magnetization of crystalline bulk iron can be accurately described by a single T^2 power law [1]. In this temperature interval the deviations from the normalized saturation value change from 0.002 to 0.018.

Table 1. Empirical power laws describing the deviations of the magnetic order parameter from saturation at absolute zero for 3D, 2D and 1D interactions and for integer and half-integer spin quantum numbers. For anisotropic 3D interactions the same exponent as for isotropic 2D interactions applies. Similarly, systems with anisotropic 2D interactions seem to belong to the same universality class as 1D systems.

		integer spin	half - integer spin
exchange interactions	3D	$T^{2/3}$	T^2
	2D 3D anisotropic	T^2	$T^{3/2}$
	1D 2D anisotropic	T^3	$T^{5/2}$

The empirical T^2 law of [1] was reported one year before the first spin wave calculation was performed by Bloch (in 1930) [2]. As is well known, Bloch's theory predicts that the spontaneous magnetization of the Heisenberg ferromagnet with bilinear nearest-neighbour interactions decreases according to a $T^{3/2}$ term in a first approximation. Later on, Dyson [3] showed that the subsequent terms in this power series expansion are $T^{5/2}, T^{7/2}, \dots$

Since the development of the Bloch–Dyson spin wave theory, most experimentalists have described their results for the temperature dependence of the spontaneous magnetization [4] and for the experimental magnon dispersion curves [5] using the power series development of the spin wave theory. As with all power series, not only is the convergence of this series a considerable mathematical problem (the inclusion of the next higher term usually changes all coefficients of the lower terms) but also the number of free fitting parameters increases unacceptably on the inclusion of higher-order terms. A good agreement between theory and experiment is then neither surprising nor convincing. In particular, if there is no independent control for the fitted exchange parameters, no conclusive test of the theory is achieved.

As we have demonstrated with many experimental examples [6–8], a fit with a single T^ε power term using an appropriate exponent from table 1 allows one to describe the temperature dependence of the order parameter excellently over a much larger temperature range than the multiparameter fit of the Bloch–Dyson power series. Moreover, the empirical T^2 law first observed for iron was frequently reported to hold over a large temperature range also for ferromagnets with localized moments such as CrO_2 [9, 10] and GdMg [11, 12] and also for antiferromagnets such as KCuF_3 [13] and EuTe [14, 15] and for the metamagnet EuSe [16]. On the other hand, $T^{3/2}$ laws are firmly established for amorphous metals such as glassy iron–nickel alloys [17–19], amorphous Co [20, 21] and amorphous Fe [22]. Also, for magnets having the Th_3P_4 defect structure [23] and for the layered material K_2CuF_4 with dominating two-dimensional interactions, $T^{3/2}$ laws are observed [24]. More systematic investigations of many Heisenberg systems, i.e., materials with pure spin magnetism, finally led to the setting up of six universality classes for the low-temperature thermodynamics of the order parameter [8]. The six classes are distinguished by whether the spin quantum number is integer or half-integer and by whether the relevant exchange interactions are 3D, 2D or 1D. For the convenience of the reader we reproduce in table 1 the universality scheme for the exponent ε .

It becomes obvious from the unusual exponent values of table 1 that the thermodynamics of real Heisenberg magnets differs considerably from the predictions of classical spin wave

theories. Most surprising is the fact that the exponents ε are independent of the spin order type and that they are different for integer and half-integer spin quantum numbers.

As a reason for the discrepancy with the classical spin wave theories, fourth-order interactions were proposed [25, 26]. These are identified as generally strong [27], but were never properly implemented in the spin wave calculations.

Although very detailed theories exist for the itinerant ferromagnetism of iron, nickel and cobalt [28–30], the accuracy of the band calculations seems not to be sufficient to justify a direct comparison with the experimental spontaneous magnetization data which have an accuracy of better than 10^{-3} . We therefore prefer to follow the empirical mode of analysis according to [1] and table 1 using a single temperature power term. The aim of the present communication is to demonstrate that this analysis works excellently not only for insulators with localized magnetic moments but, surprisingly, also for the band ferromagnets. We present no new experimental results, but new evaluations of already published data.

The empirical T^2 law for bcc iron is also confirmed for fcc nickel [8]. In view of this experimental fact, the more complicated spin wave description using more fitting parameters appears hardly justified [31, 32]. Moreover, the spontaneous magnetization of hcp cobalt exhibits a $T^{3/2}$ law with a crossover to a $T^{5/2}$ law for $T \rightarrow 0$ [8]. All of these exponents conform to the scheme of table 1 if one assumes a half-integer spin of $s = 1/2$ and isotropic 3D interactions for the cubic ferromagnets Fe and Ni but anisotropic interactions for hcp cobalt. Here we will show that the three exponents 2, $3/2$ and $5/2$ can all be verified for iron if suitably prepared. This further supports the conclusion that the universality scheme of table 1 holds, curiously, also for the itinerant ferromagnets.

2. Bcc bulk iron

Since the first investigation of [1], the spontaneous magnetization of crystalline bulk iron has been repeatedly measured. These investigations arrived at different conclusions as regards the exponent of the leading power term for asymptotically small temperatures. One experimental problem with macroscopic magnetization measurements is that a finite field for demagnetization must be applied. Therefore, paramagnetic impurities in the sample as well as in the sample support can add a perturbing $(1/T)$ -like magnetization contribution, in particular if extremely small deviations from saturation are to be measured for $T \rightarrow 0$. Zero-field methods such as Mössbauer effect (ME) spectroscopy and nuclear magnetic resonance (NMR) are nearly unaffected by paramagnetic impurities, but a specific problem with NMR is that of distinguishing between signals coming from the magnetic domains and those coming from the domain walls [33]. The two resonance signals can have different temperature dependences.

The experimental T^2 dependence of [1] is excellently confirmed and extended to the temperature range $77 \text{ K} < T < 398 \text{ K}$ by the macroscopic magnetization data of Butler *et al* [33]. In order to demonstrate the precision as well as the agreement of the two data sets, we have plotted in figure 1 the two normalized spontaneous magnetization values versus T^2 (upper straight line). The normalized zero-field ME and NMR data of [33] also agree with each other and also exhibit a T^2 dependence but with a definitely larger slope of $0.278 \times 10^{-6} \text{ K}^{-2}$ compared to $0.225 \times 10^{-6} \text{ K}^{-2}$ for the magnetization data. We propose to ascribe this difference not necessarily to an implicit temperature dependence of the hyperfine coupling constant but mainly to the fact that magnetic domains are present in the zero-field ME and NMR experiments. For the inhomogeneous state of magnetization in the domain walls, the magnetic interactions seem to be weaker compared to those for the homogeneous state in the volume of the domains. This leads to a reduced average interaction strength and, as a consequence, to a steeper slope for the zero-field ME and NMR data in figure 1. We will come

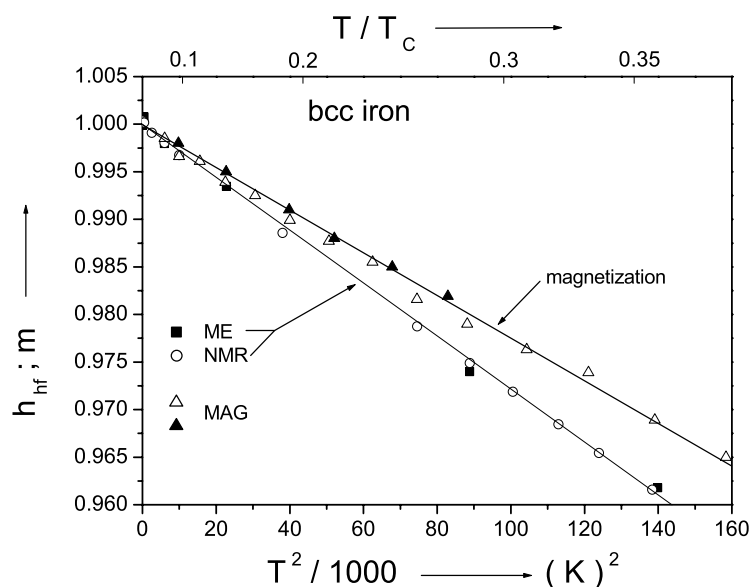


Figure 1. Upper straight line: normalized spontaneous magnetization data for bcc bulk iron from [1] (filled triangles) and from [33] (open triangles) plotted versus T^2 . Lower straight line: normalized hyperfine field data from ME and NMR experiments [33]. For these zero-field methods a steeper T^2 dependence results (see the discussion in the text). The T^2 dependence is characteristic for half-integer spin and isotropic 3D interactions.

back to this point in more detail in the section on standing spin wave resonance (SSWR) in film samples. In the films the magnetic interactions are clearly weaker for the state in which the magnetization is inhomogeneous, i.e., periodically modulated due to the excitation of standing spin waves (SSW) by radio-frequency irradiation.

The NMR and ME measurements of [33] have been conducted to a lowest temperature of 1.5 and 4.2 K, respectively. Since these low-temperature data also do not deviate from the T^2 fit, we can be sure that the T^2 law is correct down to asymptotically small temperatures.

In contrast to the results of figure 1, the magnetization data of Pauthenet [34] and Argyle *et al* [31] deflect clearly from a T^2 dependence towards larger values for $T \rightarrow 0$. These deviations can largely be described by a $1/T$ term, as is typical for paramagnetic impurities. Since those asymptotic deviations from the T^2 dependence are not reproduced by the other methods and authors, they can be excluded from further consideration. It should be noted that small T^{-1} -like contributions to the magnetization have the general tendency to reduce the effective exponent below the value of $\varepsilon = 2$. A pure power law is not observed in the presence of paramagnetic impurities and a fitting procedure treating the exponent ε of the leading T^ε power term as a free fitting parameter runs into difficulties. Because of this problem, the leading exponent was fixed to 3/2 in [31]. The fit range had accordingly to be reduced to $T < 90$ K and the next higher $T^{5/2}$ term could, of course, not be identified with certainty.

The most complete data set for the spontaneous magnetization of bcc bulk iron is that of Crangle and Goodman [35]. These data also show no definite deviation from a T^2 dependence within the experimental errors down to the lowest temperature. In the upper diagram of figure 2 we have plotted the tabulated normalized spontaneous magnetization data of [35] versus the squared ratio of the absolute temperature divided by the Curie temperature of $T_C = 1044$ K. In this representation a good linear fit is possible up to a temperature of ~ 467 K, which

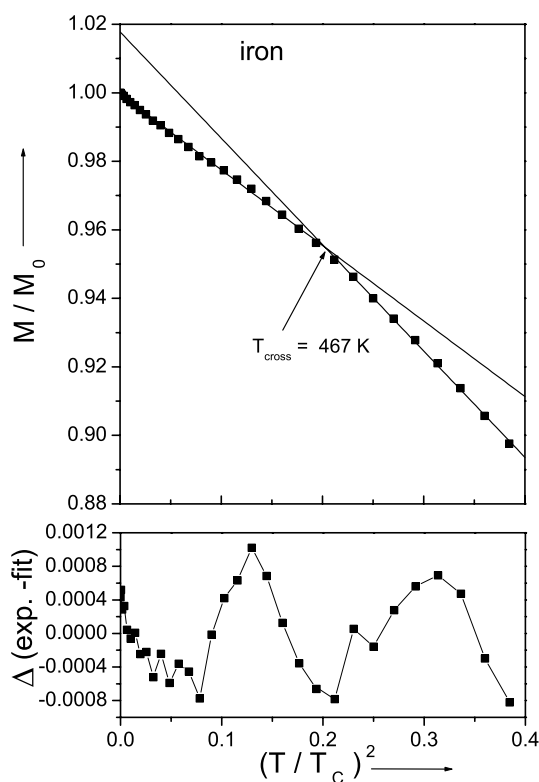


Figure 2. Upper diagram: normalized spontaneous magnetization data for $T/T_C < 0.62$ for iron taken from [35] as a function of squared absolute temperature divided by the Curie temperature ($T_C = 1044 \text{ K}$). Crossover between two T^2 functions (amplitude crossover) is observed at 467 K ($T/T_C = 0.45$). The smaller slope for $T < 467 \text{ K}$ indicates larger magnetic interactions compared to the case for $T > 467 \text{ K}$. Lower diagram: differences between experimental data and the two respective T^2 fit functions.

corresponds to $T/T_C = 0.45$. In the temperature interval $470 < T < 650 \text{ K}$ another T^2 fit with a steeper slope is possible. To show the quality of the two fits, the lower diagram of figure 2 displays the differences between the experimental data and the respective T^2 fit functions. It can be seen that the average error is of the order of $\pm 5 \times 10^{-4}$ only. This conforms to the usual experimental limits of accuracy in measurements of the relative magnetization and justifies our fitting procedure with two T^2 functions as physically reasonable (though other interpretations can perhaps not completely be excluded).

We interpret the change from one T^2 function to another T^2 function as a thermodynamic crossover phenomenon. One, historically first, indication for a crossover between two $T^{3/2}$ functions can be noted from figure 2 of [36]. In this work the spontaneous magnetization of an Invar alloy with composition $\text{Ni}_{0.348}\text{Fe}_{0.652}$ was measured (see the section below about FMR). Crossover between different power functions for the order parameter must be considered as a discrete thermodynamic reaction caused by a continuous variation of the interaction strength as a function of temperature. This is a common experimental observation for metallic ferromagnets and was found for GdZn , GdAl_2 [37] and also the hcp ferromagnets gadolinium and cobalt [8]. For the cubic ferromagnets Fe, Ni and GdZn the crossover is between two T^2 functions, meaning that the interactions remain isotropic and only change their absolute

strength as a function of temperature (amplitude crossover). On the other hand, for materials with an anisotropic magnetostriction which is normally associated with a lattice distortion, the anisotropy or dimensionality of the relevant interactions can change. This is the case for the hexagonal ferromagnets gadolinium and cobalt which show a crossover between a high-temperature $T^{3/2}$ function and a low-temperature $T^{5/2}$ function (exponent crossover). According to table 1, this indicates a change of the relevant interactions from anisotropic 3D to 1D.

The observed smaller slope of the low-temperature T^2 function in bcc iron (see figure 2) means that the interactions become stronger with decreasing temperature. A comparison between the two fitted T^2 coefficients allows one to estimate that the interactions have increased by 18%. A similar crossover phenomenon to that for iron is revealed also in the spontaneous magnetization data of nickel, tabulated also in [35].

In metallic ferromagnets the conduction bands are involved in the magnetic interactions. These bands are known to split and/or shift under the action of the strong internal polarization fields. As a consequence, the interaction strength will change as a function of temperature (or as a function of the spontaneous magnetization). It can be assumed that those changes are—like the magnetization—continuous. If these have exceeded some threshold value, a thermodynamic crossover, i.e. an analytical change to another T^e function, occurs.

In antiferromagnets the internal polarization fields are much weaker and, as a consequence, the conduction bands will split and/or shift much less. The thermal variation of the magnetic interaction strength is therefore comparatively weak and crossover is less likely to occur. In contrast to the case for the metallic ferromagnets Fe, Ni, Gd, Co, GdZn and GdAl₂, no crossover is observed for the metallic antiferromagnet GdAg [7, 37]. Also, in insulators the exchange interactions change comparatively weakly as a function of temperature and an amplitude crossover occurs only in cubic materials which exhibit a very strong isotropic magnetostriction. An exponent crossover can be expected only in non-cubic materials with a strong anisotropic exchange striction (lattice distortion).

3. Amorphous bulk iron

It is a well established experimental fact that the thermal decrease of the spontaneous magnetization of amorphous Fe, Co and Ni alloys follows a $T^{3/2}$ power law [17–22]. This has been verified using Mössbauer spectroscopic measurements [17, 18] and macroscopic magnetization measurements [19–22]. The amorphous state is usually stabilized by addition of phosphorus and boron. For more details, we refer the reader to the review article of Hansen [38].

The most complete set of spontaneous magnetization data for an amorphous ferromagnet seems to be that of Kaul obtained for Fe₄₀Ni₄₀P₁₄B₆, which is known as Metglas[®]2826 [19]. In figure 3 we have plotted Kaul's data versus $T^{3/2}$ as far as this law holds. In agreement with all other investigations of amorphous alloys, figure 3 clearly demonstrates that a single $T^{3/2}$ power term is sufficient to describe the thermal decrease of the spontaneous magnetization accurately over a sizable temperature range. This common behaviour of amorphous alloys was erroneously considered as experimental proof of Bloch's spin wave theory [2]. The agreement with Bloch's exponent of 3/2 must, however, be considered fortuitous because, first, the classical spin wave theory requires a translational symmetry of the magnetic lattice which is not given in the amorphous state and, second, the unusually large validity range of the experimental $T^{3/2}$ function is inconsistent with the approximation made in Bloch's calculation. Moreover, the large validity range of the $T^{3/2}$ term suggests that this law is exact and not only the first-order term for $T \rightarrow 0$. At least a possible $T^{5/2}$ term is far too small compared with the Bloch–Dyson spin wave theory.

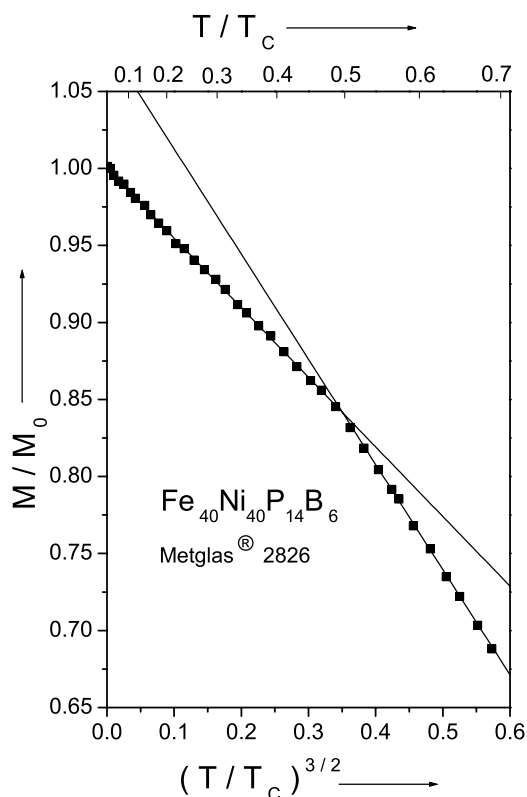


Figure 3. Normalized spontaneous magnetization of a glassy Fe–Ni alloy (Metglas[®] 2826) versus $T^{3/2}$ taken from [19]. A very similar behaviour to that for bcc iron (see figure 2) results for the amorphous state if the spontaneous magnetization is plotted on a $T^{3/2}$ scale instead on a T^2 scale. Crossover to a $T^{3/2}$ function with a smaller slope (low-temperature section) indicates that the magnetic interactions increase with decreasing temperature.

As we now know, the exponent of $3/2$ is typical for half-integer spin values (see table 1). This exponent applies not only for amorphous alloys but also for crystalline materials with either anisotropic three-dimensional interactions or isotropic two-dimensional interactions [8]. The polycrystalline Ni–Fe alloys of the Invar composition range are further examples showing the $T^{3/2}$ behaviour (see the discussion of figure 9). Examples showing anisotropic 3D interactions include the hcp ferromagnets gadolinium and cobalt [8]. In contrast to the glassy Ni–Fe alloys, the hcp ferromagnets Gd and Co finally exhibit a crossover to a $T^{5/2}$ law, as is characteristic for dominating 1D interactions. Such an exponent crossover is not to be expected for an amorphous material. In fact, a similar amplitude crossover to that for crystalline iron (see figure 2) is revealed from figure 3. This phenomenon we consider again as a singular thermodynamic reaction caused by a strong continuous thermal variation of the exchange interactions without a change of their symmetry. It is very surprising that very similar diagrams are obtained if the magnetization data for crystalline iron (figure 2) and for the amorphous iron–nickel alloy (figure 3) are plotted versus T^2 and versus $T^{3/2}$, respectively.

Also, for Gd_4Bi_3 and Gd_4Sb_3 , having the inverse bcc Th_3P_4 defect structure, a $T^{3/2}$ law was found to hold up to 0.8 of the Curie temperature [23]. In these materials the local symmetry at the Gd site is very low and, apparently, the spin dynamics is as in the amorphous state.

4. Crossover from 2D to 3D interactions in ultrathin Fe films

Observation of a long-range magnetic order in real Heisenberg materials with 2D interactions ($\varepsilon = 2$ or $3/2$) is still a puzzle. Assuming only bilinear exchange interactions, classical spin wave theories arrive at the conclusion that no long-range magnetic order is possible for Heisenberg systems with 2D interactions [2, 39, 40]. Only due to dipolar and/or magnetocrystalline anisotropy is a long-range magnetic order stabilized [41, 42]. On the other hand, table 1 suggests that in real systems a long-range magnetic order exists also for 2D interactions, since a single temperature power term with a particular exponent ε holds for 2D systems as well. For more details on the properties of ultrathin magnetic films, we refer the reader to the article of Gradmann [43].

Very sophisticated Mössbauer spectroscopic measurements on epitaxial Fe films grown on W(110) and covered by Ag have been performed by Korecki *et al* [44]. Near to the Ag interface the saturation hyperfine field, i.e. the magnetic moment per Fe, is not perfectly constant but oscillates slightly from layer to layer [45]. This is a complication for our purposes here. Implanting probe layers of the Mössbauer-active ^{57}Fe isotope at different positions in a ^{56}Fe film, the properties of the individual layers next to the surface have been investigated separately. This method allowed it to be proved that for all layers next to the surface a $T^{3/2}$ temperature dependence results for the hyperfine field, at least for the silver-covered (110) Fe film [45]. The saturation hyperfine field and the coefficient of the $T^{3/2}$ term approach well defined values for the layers which are more distant from the Fe/Ag interface. Also, in [46–50] a $T^{3/2}$ dependence was reported for silver-covered (110) iron films. The $T^{3/2}$ law is, hence, well established experimentally for ultrathin Fe films. According to table 1, the $T^{3/2}$ dependence is characteristic for the isotropic Heisenberg system with 2D interactions and half-integer spin quantum number.

The question now is: how does the thermodynamics, i.e., the dimensionality of the relevant exchange interactions, change from 2D to 3D as a function of an increasing film thickness? This change must be associated with a crossover from a $T^{3/2}$ dependence to a T^2 dependence of the hyperfine field (which is assumed to be proportional to the order parameter).

The most complete set of hyperfine field values was presented in [44]. In that work, epitaxial $^{57}\text{Fe}(110)$ films with thickness values of 3.4, 5.3, 8.6 and 20.5 atomic layers were investigated. The observed Mössbauer spectra of these films each consist of a superposition of one low-field spectrum to be attributed to the first atomic layer on the tungsten substrate and one high-field spectrum which is characteristic for all Fe layers on top of the first one. In the following, we consider only the high-field spectral component, which can be considered as representative for the isotropic 2D material.

In agreement with [46], it was observed also in [44] that the hyperfine field of the high-field component obeys a $T^{3/2}$ temperature dependence up to $T \sim 300$ K for all four thickness values. In figure 4 we show the normalized hyperfine field data of [44] plotted versus $T^{3/2}$. The different slopes are essentially due to the Curie temperatures decreasing with decreasing film thickness. It is very surprising that the exponent of $3/2$ which is characteristic for isotropic 2D interactions can be observed for a large span of thickness values, i.e., for samples with considerably different Curie temperatures. This shows that the conditions for the occurrence of a particular exponent ε are not very critical. Connected with this is the fact that a limited number of stable exponents (universality classes) is sufficient to characterize all Heisenberg magnets.

The $T^{3/2}$ dependence of the ultrathin Fe films is well confirmed also by numerous other investigations [43–50]. Our interest here is in the hyperfine field data beyond the validity range of the $T^{3/2}$ dependence, i.e. for temperatures larger than those shown in figure 4. These data

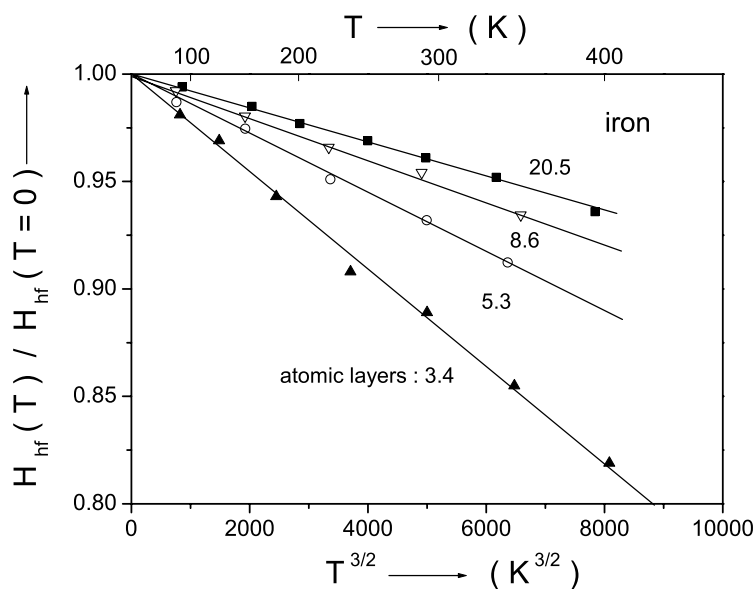


Figure 4. Normalized hyperfine field data for isotropic iron films with 3.4, 5.3, 8.6 and 20.5 atomic layers as a function of $T^{3/2}$ obtained with ME spectroscopy. Shown are just the low-temperature data of [44] which exhibit the $T^{3/2}$ dependence. The exponent of $3/2$ is typical for isotropic 2D interactions and half-integer spin values (see table 1).

can consistently be interpreted by a crossover to a T^2 function. Figure 5 shows the normalized hyperfine field values for an extended temperature range but plotted versus T^2 . Using a T^2 scale in figure 5, the normalized bulk magnetization data of Weiss and Forrer [1] as well as those of Butler *et al* [33] fall on a straight line which reaches unity for $T \rightarrow 0$. The outcome is different for the high-temperature T^2 functions of the films, for which the extrapolated ordinate values decrease continuously with decreasing film thickness. The vertical shift of the fitted straight lines in figure 5 shows that the bulk T^2 function is steadily depressed with decreasing number of atomic layers. This downward shift increases the ordinate range for the $T^{3/2}$ function which follows in the low-temperature regime. Note that for the T^2 scale used in figure 5, the $T^{3/2}$ functions of figure 4 are curved with an increasing slope for $T \rightarrow 0$.

We must admit that the difference between the exponents $3/2$ and 2 is not very large. This implies that the crossover between the two analytical functions is relatively flat. Additionally, the hyperfine field data for the ultrathin films have, necessarily, a limited accuracy. This, of course, limits the precision of our analysis.

In order to demonstrate those difficulties we have plotted all hyperfine field data for the sample with 8.6 atomic layers of iron versus $T^{3/2}$ in figure 6. It can be seen that for $T > 400$ K the difference between the $T^{3/2}$ and the T^2 fit function is marked. On the other hand, the high-temperature T^2 fit is up to a temperature where, normally, description by a single T^ϵ function is possible. As many experimental examples including figures 2 and 3 show, a single power law usually holds at least up to $\sim 2/3$ of T_C . Empirically, it appears therefore justified to interpret the temperature dependence of the hyperfine field data as a crossover from a $T^{3/2}$ function for $T \leq 300$ K to a T^2 function for $T \geq 300$ K. A theoretical justification for this interpretation constitutes, of course, a big challenge for the future. The empirical analysis just given can at least be considered as supplying a possible solution to the long-standing problem of how the crossover from the 2D to the 3D thermodynamics proceeds as a function of an increasing film thickness.

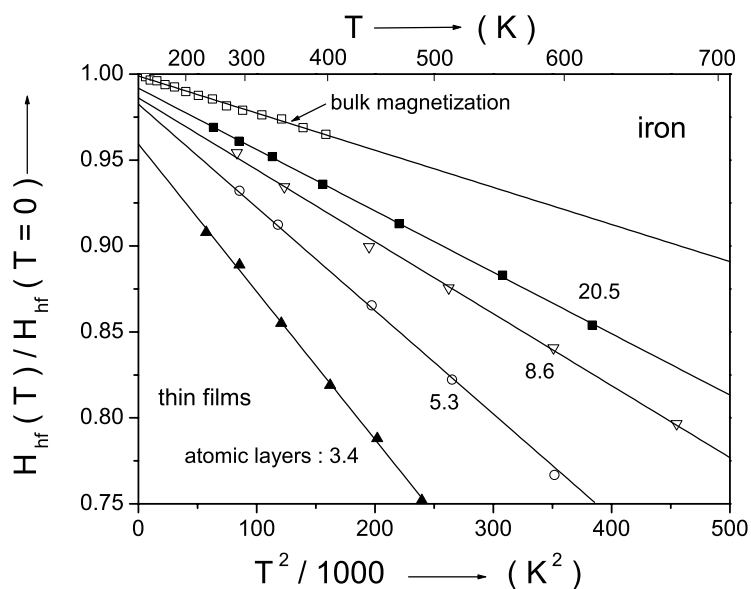


Figure 5. Normalized hyperfine field data of [44] for an extended temperature range but plotted versus T^2 . The T^2 dependence is characteristic for isotropic 3D interactions and conforms to the behaviour of bulk iron (open squares taken from [33]).

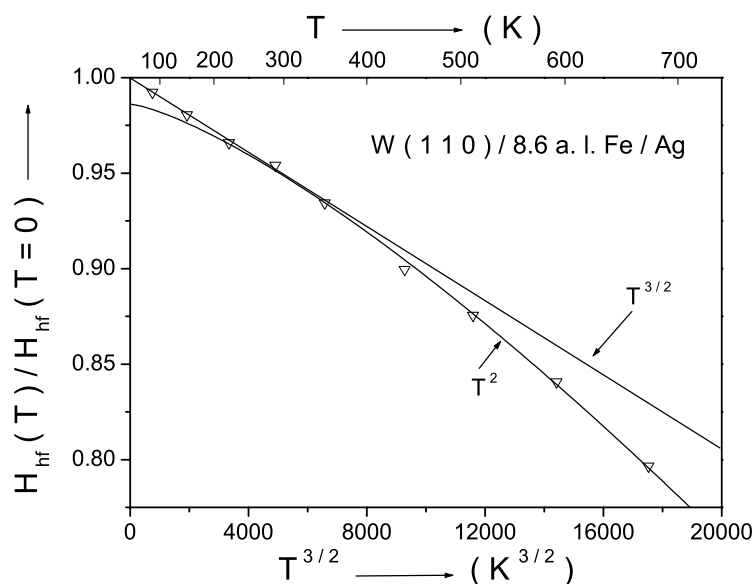


Figure 6. Normalized hyperfine field data for the iron film with 8.6 atomic layers as a function of $T^{3/2}$. The crossover from $T^{3/2}$ to T^2 is rather flat, but definite differences between the two functions result for larger temperatures. The high-temperature T^2 fit is for a temperature range where description by a single power term normally holds ($T < 0.75T_C$). From [44].

It is very surprising that even the sample with only 3.4 atomic layers of iron exhibits a crossover to the 3D thermodynamics (to a T^2 dependence) though with a relatively small amplitude of the T^2 term only. This seems to be connected with the fact that the saturation

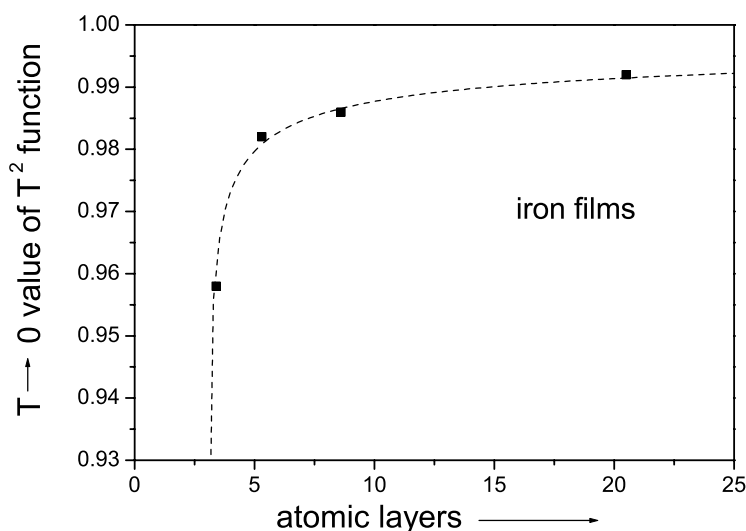


Figure 7. The value of the T^2 fit function (see figure 5) extrapolated to $T \rightarrow 0$ as a function of the number of atomic layers. The dashed curve is a guide to the eye only. For all Fe films with more than ~ 3 atomic layers this amplitude is finite and crossover to 3D behaviour occurs. A pure 3D thermodynamics is realized for an amplitude value of unity, which seems to be reached only for an infinite number of layers.

hyperfine field of this sample is still very similar to the bulk value (even somewhat larger) [45]. A quantitative investigation of the extrapolated ordinate values of the T^2 functions (see figure 7) allows one to estimate that the 3D properties have disappeared completely only for samples with less than three atomic layers. Of course, three layers are the absolute minimum for the realization of spin waves perpendicular to the film plane. We must, hence, conclude that the typical 3D thermodynamics (T^2 dependence) can be realized with spin waves having very short wavelengths only. On the other hand, figure 7 also shows that the amplitude of the T^2 function seems to approach unity only for asymptotically large thickness values. The crossover from a 2D to a 3D behaviour is therefore not only related to the film thickness but also related to temperature. This crossover seems to be observable in the range between three atomic layers and—in principle—up to an infinite number of layers. Also the crossover temperatures decrease with increasing number of layers and seem to vanish only for an infinite thickness value.

In other words, the 2D behaviour ($T^{3/2}$ function) holds at low temperatures even for very thick films, but it describes very small deviations from saturation only and can, practically, not be distinguished from the dominating T^2 law. As is well known, for $T \rightarrow 0$ only spin waves with a wavelength increasing to infinity are left. If the relevant spin waves have assumed a wavelength which is larger than the thickness of the film, a crossover to 2D behaviour occurs.

5. The monatomic Fe layer on W(110)

Przybylski and Gradmann [49] have investigated the pseudomorphic Fe monolayer grown on W(110) and covered by silver, using Mössbauer spectroscopy. Monolayers cannot be grown without structural imperfections or anisotropies [50]. It is difficult to specify the type of anisotropy on the basis of the observed exponent ε since we expect the same T^ε power law with $\varepsilon = 5/2$ for anisotropic 2D systems and for amorphous 2D systems including 2D systems

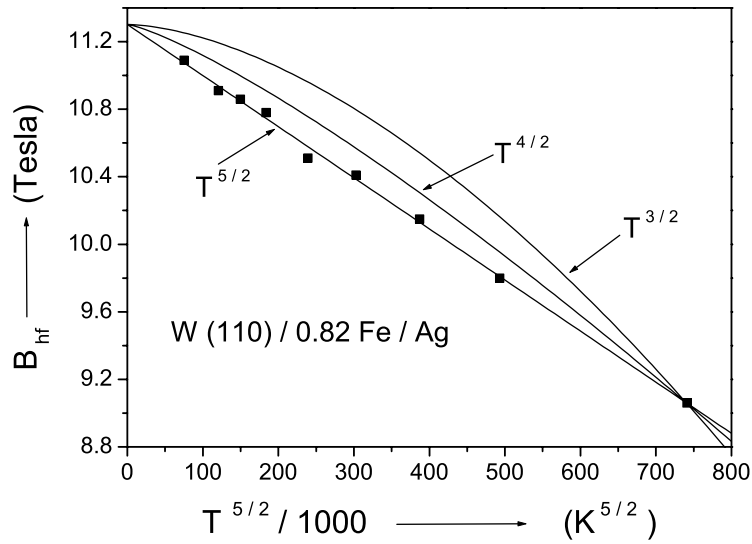


Figure 8. ^{57}Fe hyperfine field data of [49] for the monatomic Fe layer (0.82 bulk layers) grown on W(110) and covered by a protective silver film as a function of $T^{5/2}$. In alternative plots for $T^{3/2}$ and $T^{4/2}$, the fitted line in the $T^{5/2}$ representation would be bent, as is indicated by the curves labelled by $T^{3/2}$ and $T^{4/2}$. The saturation hyperfine field is very nearly 1/3 of the bulk value. The exponent of 5/2 is characteristic for an inhomogeneous 2D system with half-integer spin value.

in which the volume of the domain walls compares with the volume of the domains. Moreover, $\varepsilon = 5/2$ is characteristic for 1D systems according to table 1.

In contrast to the cases for many other monolayer systems, the magnetization is in the film plane for the pseudomorphic Fe monolayer under consideration. The observed non-linear temperature dependence of the spontaneous magnetization seems to be connected with this [43].

Figure 8 shows the experimental hyperfine field data of [49] versus $T^{5/2}$. The resulting linear dependence in this representation shows that the exponent of 5/2 fits excellently. For comparison, the two curves labelled by $T^{3/2}$ and $T^{4/2}$ indicate how the fitted straight line in the $T^{5/2}$ representation would be bent if the data were plotted for $T^{3/2}$ or $T^{4/2}$, respectively. The strong curvature of these two alternative temperature functions demonstrates the precision with which the exponent of 5/2 can be identified. It is interesting to note that for the monolayer considered, not only is the critical temperature about 1/3 of the bulk value but also the saturation hyperfine field is nicely 1/3 of the bulk value. Both features are consistent with a magnetic dimensionality of unity for this anisotropic monolayer.

The exponent of 5/2 is well known from systems with half-integer spin in which the spin dynamics is confined to one crystallographic axis. The most prominent example is MnF_2 which exhibits the typical thermodynamics of a 1D Heisenberg system at least in the ordered state [51]. In this material with the rutile structure and a pure spin of $S = 5/2$, the magnetic interactions are much stronger along the crystallographic c -axis than in the basal plane. As a consequence, these interactions prevail and confine the spin dynamics to the c -axis. This is most clearly seen from the fact that only the correlation length parallel to the c -axis diverges at the Néel temperature [52, 53]. Observation of an energy gap in the magnon excitation spectrum in this material without any single-ion anisotropy further demonstrates the geometrically confined spin dynamics. Further examples exhibiting the $T^{5/2}$ law are the hcp ferromagnets cobalt

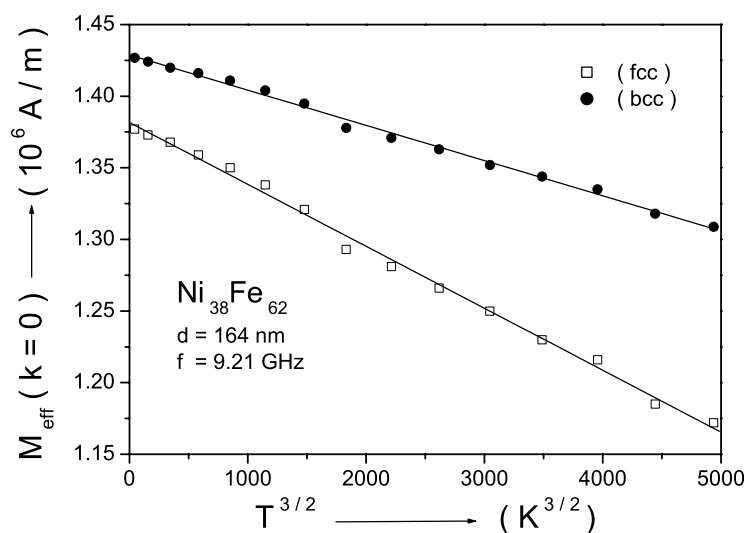


Figure 9. Spontaneous magnetization of a $\text{Ni}_{38}\text{Fe}_{62}$ film with a thickness of 164 nm evaluated from FMR (uniform precession mode: $k = 0$) as a function of $T^{3/2}$. In the polycrystalline Invar alloy of composition $\text{Ni}_{38}\text{Fe}_{62}$, the bcc phase (iron) and the fcc phase (nickel) coexist. Due to the different resonance conditions, FMR resolves the two crystallographic phases. The $T^{3/2}$ dependence is typical for a microscopically inhomogeneous 3D system with half-integral spin value (see figure 3). Data are reproduced from [54].

and gadolinium, but these show the $T^{5/2}$ law only for small temperatures where the relevant interactions have changed from anisotropic 3D to 1D.

It is very surprising that materials which are as different as bulk MnF_2 and the anisotropic monatomic Fe layer can belong to the same universality class. Not only do the geometrical dimensions of the sample seem to be unimportant but so also is the fact that the monatomic Fe layer is an itinerant (metallic) ferromagnet while MnF_2 is an insulator with localized magnetic moments. We must assume that in MnF_2 there are also finite but weak interactions transverse to the symmetry axis, but these seem to be not relevant for the thermodynamics. Since the proposed universality scheme of table 1 covers all these physically different examples, it must originate from very general thermodynamic grounds.

6. FMR and standing spin waves in Ni–Fe alloy films

The ferromagnetic resonance (FMR) technique is a very important method for studying the spontaneous magnetization of thin films. Normally, FMR experiments are performed on films with thickness values of several hundred nanometres. For such thickness values, bulk magnetic properties can be anticipated.

Most FMR experiments have been performed on films of the alloy system of fcc nickel and bcc iron. Depending on the composition, these Ni–Fe alloys provide very interesting structural and magnetic properties. In the permalloy region near compositions $\text{Ni}_{80}\text{Fe}_{20}$ (fcc structure), very high values of the magnetic permeability and very low values of the magnetostriction are observed. In the Invar region near compositions $\text{Ni}_{35}\text{Fe}_{65}$, lattice instabilities occur which are responsible for a number of anomalies such as a strong magnetostriction and very low values of the spontaneous magnetization. The Invar alloys are polycrystalline mixtures of the bcc and the fcc phase. As a consequence of the microscopically inhomogeneous structure, the

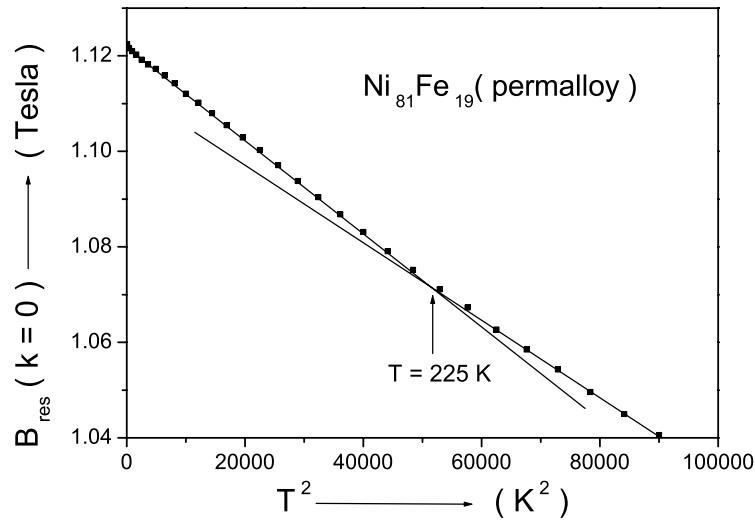


Figure 10. The field for resonance of a permalloy film with a composition of $\text{Ni}_{81}\text{Fe}_{19}$ and a thickness of 504 nm evaluated from FMR (uniform precession mode: $k = 0$) as a function of T^2 . For this film with a homogeneous fcc structure the same temperature dependence ($\sim T^2$) as for bulk iron or nickel is observed (see figures 1 and 2). From the crossover at $T \cong 225$ K it can be concluded that the magnetic interactions increase with increasing temperature (smaller slope for $T > 225$ K). Data are reproduced from [56].

uniform precession mode (spontaneous magnetization) follows a $T^{3/2}$ law (see figure 9) like for the amorphous Ni–Fe bulk materials (compare figure 3). Since the magnetic densities are different for the two crystallographic phases, FMR allows one to resolve the two phases on account of the different fields for resonance. In this way, Kordecki was able to establish that the two FMR resonance frequencies exhibit the same $T^{3/2}$ dependence [54]. In figure 9 we have reproduced Kordecki's data for the temperature dependence of the FMR frequencies of the two crystallographic components of a $\text{Ni}_{38}\text{Fe}_{62}$ Invar film with a thickness of 164 nm. Using a $T^{3/2}$ scale a quite linear behaviour results for both FMR signals. The given magnetization values are obtained by converting the measured resonance frequencies of the uniform precession mode into magnetization values using appropriate g -factors.

Permalloy films with compositions near $\text{Ni}_{80}\text{Fe}_{20}$, on the other hand, have a homogeneous fcc structure. In fact, for these films the FMR frequency (or field for resonance at constant frequency) of the uniform precession mode, i.e., the spontaneous magnetization, decreases according to a T^2 law (see figure 10) like for the crystalline Fe and Ni bulk samples. This analytically simple temperature dependence has often not been recognized and more complicated fitting procedures using a $T^{5/2}$ term in addition to the common $T^{3/2}$ term have been tried [55]. Even using a sum of these two power terms, the agreement between fit and experimental data is limited to $T < 150$ K while the empirical T^2 description in figure 10 fits up to 225 K. Figure 10 reproduces the FMR data of Weber and Tannenwald [56] obtained on a $\text{Ni}_{81}\text{Fe}_{19}$ permalloy film with a thickness of 504 nm on a T^2 scale. These data reveal a clear amplitude crossover to a second T^2 dependence at ~ 225 K, surprisingly with a smaller slope for $T > 225$ K which is in contrast to the situation for the bulk Fe (see figure 2) and Ni crystals.

In the uniform precession mode (quantum number of $k = 0$) the magnetization precesses as a whole. The outcome is different if SSW with $k \neq 0$ are excited by radio-frequency irradiation. In these resonance states the magnetization is inhomogeneous, i.e. spatially and

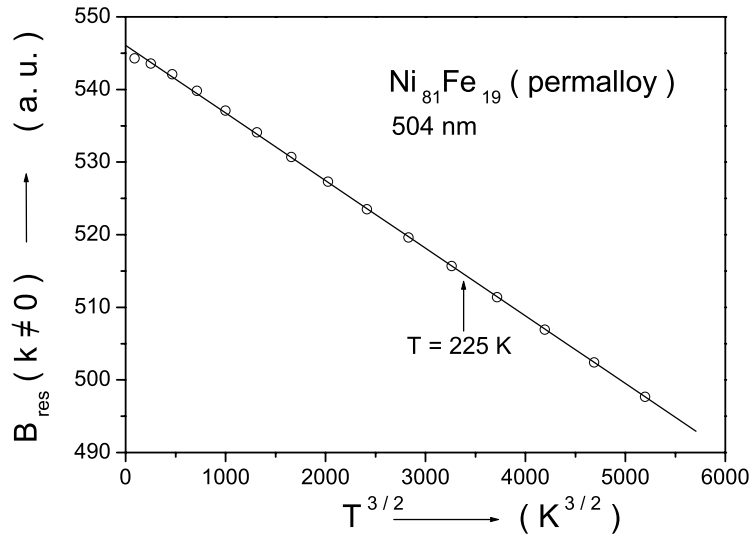


Figure 11. The field for resonance of the same permalloy film as in figure 10 but for the state with excited SSW versus $T^{3/2}$. The crossover at $T = 225$ K seen in the uniform precession mode (see figure 10) is missing. This and the $T^{3/2}$ dependence of $B_{res}(T)$ show that the magnetic interactions are different for the state in which SSW are excited compared to the uniform precession mode. Data are reproduced from [56].

temporally periodically modulated. The different modes are distinguished by the quantum number k which counts the number of half-waves transverse to the film. The quantum number k does not, however, have the character of a wavevector, as it is used to characterize propagating spin waves. Instead, k must be considered as a scalar quantum number which provides a measure of the ‘inhomogeneity’ of the magnetization of the individual resonance modes. The resonance condition $B_{res} \sim k^2$ is, of course, the same for the Invar films ($T^{3/2}$ dependence of the spontaneous magnetization) as well as for the permalloy films (T^2 dependence of spontaneous magnetization).

For the homogeneous permalloy films as well as for the pure iron [57], nickel [58] and cobalt films [59], the resonance frequency of the SSW modes has been reported to follow a $T^{3/2}$ dependence over a large temperature range. To demonstrate this, we have plotted in figure 11 the resonance frequency of the SSW modes (the coefficient of the k^2 dependence) of [56] for the same 504 nm Ni₈₁Fe₁₉ permalloy film as in figure 10 versus $T^{3/2}$. On this temperature scale a linear dependence results up to 300 K except perhaps for the temperature range below 30 K. These asymptotic deviations could indicate a crossover for $T \rightarrow 0$, but since the numerical values differ extremely little from saturation, no definite proof of this is possible.

It is very surprising that the crossover at ~ 225 K observed in the uniform precession mode (see figure 10) is missing for the SSW modes in figure 11. Since the crossover from one T^2 function to another T^2 function in the $M(T)$ data of figure 10 indicates a change in the strength of the magnetic interactions, a similar crossover should occur also for the SSW modes. From the fact that this is evidently not the case, we conclude that the magnetic interactions are not dramatically but are definitely different in the state in which the magnetization is twisted, i.e. periodically modulated by the excitation of SSW.

In conclusion, for all homogeneous films the temperature dependence $\sim T^{3/2}$ of the SSW modes can be considered as a firmly established experimental fact. The exponent of $3/2$

was never understood on the basis of the conventional theory of the spin wave resonance experiments [60]. According to this theory, the SSW modes sample the spin wave stiffness constant $D(T)$ for which the classical theory predicts $D \sim T^{5/2}$ [3].

In order to resolve this discrepancy, we want to propose an alternative interpretation of the spin wave resonance experiments. In contrast to the thermal spin waves which are evidently acoustic (and propagating) modes, the SSW induced by the resonant irradiation of an rf field are discrete and coherent eigenstates of a square-well potential. These artificial states are, clearly, never realized thermally and must be considered as inhomogeneous states of magnetization similar to the static situation in the domain walls. Since the magnetization is periodically twisted, i.e. inhomogeneous, we must assume that the exchange interactions are similarly inhomogeneous. Apparently, this particular type of inhomogeneity induces an exponent crossover from T^2 to the same $T^{3/2}$ temperature dependence as applies for the magnetization of the amorphous bulk materials (figure 3) and for the polycrystalline Invar films (figure 9). On the other hand, the presence of magnetic domains in the zero-field NMR and ME measurements (see figure 1) seems to induce only an amplitude crossover with a larger slope of the T^2 function for the zero-field situation compared to the macroscopic spontaneous magnetization obtained for magnetically saturated samples with no magnetic domains.

There are still problems with this interpretation. First, the definition of the order parameter is not quite obvious if there is no constant and homogeneous magnetization. Second, since we assume that the thermal decrease of the state with $k = 0$ is according to a T^2 dependence but for the excited states with $k \neq 0$ it is according to a $T^{3/2}$ dependence, we must assume that saturation was achieved by the irradiation of the resonance frequency. In other words, in order to observe a pure $T^{3/2}$ law it is necessary that the state with $k = 0$ is completely suppressed, which requires an intense rf field such that saturation or a population inversion is achieved. Third, for the SSW modes with very small k -values, i.e. with a very long wavelength, the modulation of the magnetization is only weak. For these modes a crossover to a T^2 dependence can be expected. It is therefore necessary to measure the temperature dependence of these modes individually and not to average over all modes (k^2 dependence) as is normally done.

In order to show more clearly that the magnetic interactions are weaker for the SSW modes ($k \neq 0$) with a modulated magnetization compared to that of the uniform precession mode ($k = 0$), we have plotted the normalized fields for resonance from figures 10 and 11 versus the linear temperature in figure 12. It can be seen that the SSW mode decreases clearly more strongly with temperature than the $k = 0$ mode despite a smaller exponent ($\varepsilon = 3/2$).

In a similar way, one can explain the steeper slope for the zero-field ME and NMR data compared to the spontaneous magnetization data in figure 1 by the weaker interactions for the inhomogeneous state of magnetization due to the presence of magnetic domains. In contrast to the SSW modes in the films, the small volume fraction of the domain walls in bulk iron is not sufficient for an exponent crossover from T^2 to $T^{3/2}$ and only gives rise to an amplitude crossover.

In some spin wave resonance experiments it was observed that the $T^{3/2}$ dependence of the SSW modes was not realized perfectly. An unrecognized crossover is frequently a reason for this. In the iron films of [57], for instance, the $T^{3/2}$ dependence was established only for higher temperatures. For $T \rightarrow 0$ a rather undefined analytical behaviour was observed. Replotting these data for $T^{3/2}$ (see figure 13) reveals that in the low-temperature regime the SSW frequency can well be interpreted as a crossover to a second $T^{3/2}$ function with a smaller slope. It should be noted that these crossover phenomena depend somewhat on the composition of the films and are not such reproducible as in high-quality single crystals. Also, in figure 11 a similar crossover to that in figure 13 is likely to occur but at a much smaller temperature and therefore this crossover is not definitely resolved.

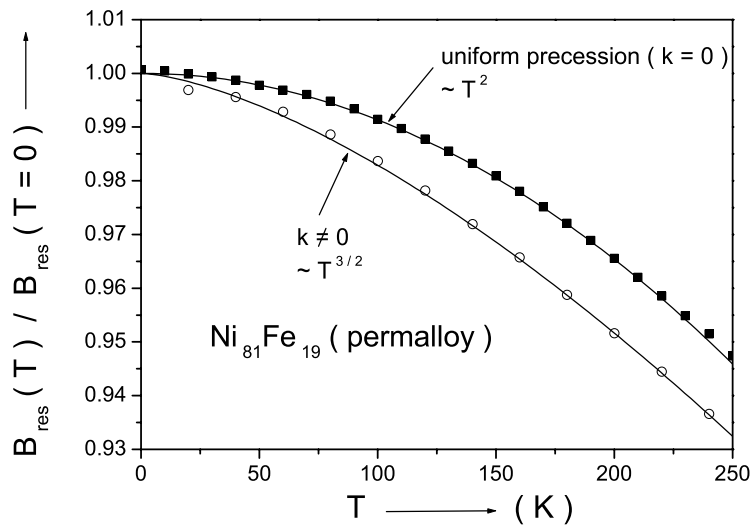


Figure 12. Normalized resonance fields for the uniform precession mode ($k = 0$) and the SSW modes ($k \neq 0$) as a function of absolute temperature. These data are identical to those of figures 10 and 11. The steeper decrease of the resonance field for the SSW mode ($k \neq 0$) indicates weaker magnetic interactions for the periodically modulated state of magnetization compared to the uniform state.

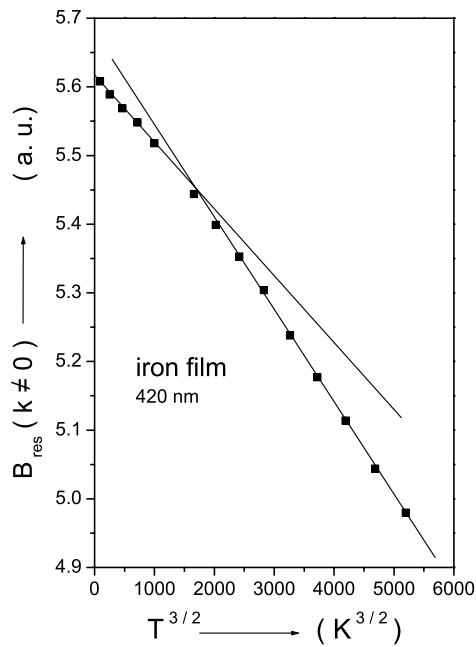


Figure 13. The field for resonance of a pure iron film (taken from [57]) as a function of $T^{3/2}$ revealing amplitude crossover between two $T^{3/2}$ regions. Crossover was often not recognized in the past and this led to wrong conclusions regarding the exponent if the fit was made over a temperature range including a crossover.

7. Conclusions

We have reconsidered published data for the spontaneous magnetization of iron samples with different morphologies. Weiss and Forrer were the first to observe that the thermal decrease of the spontaneous magnetization of bulk iron follows a T^2 dependence up to 300 K [1]. This historical result is confirmed to hold at least to a lowest temperature of 1.5 K by the NMR data of Butler *et al* [33]. The most complete set of magnetization data, that of Crangle and Goodman [35], shows that the T^2 dependence continues up to 467 K and is followed by a crossover to a second T^2 dependence with a larger amplitude. It should be noted that in two other magnetization studies of bulk iron, a fit with the anticipated exponent of $3/2$ has been applied to the spontaneous magnetization for $T \rightarrow 0$ [31, 34]. This fit worked approximately only up to 90 K because of a $(1/T)$ -like contamination due to paramagnetic impurities.

For the ultrathin and isotropic iron layers, a $T^{3/2}$ law is firmly established by means of ME spectroscopy [43–50]. This temperature dependence is well known for Heisenberg systems with half-integer spin and isotropic 2D interactions according to table 1. Here we observed that the ME data beyond the validity region of the $T^{3/2}$ law can be interpreted as a crossover to a T^2 function. This crossover is characteristic for a dimensionality change of the relevant interactions from 2 to 3 and occurs for all Fe films with more than three atomic layers. With this analysis we offer an answer to the long-standing problem of how the change from a 2D to a 3D thermodynamics proceeds in thin films as a function of an increasing film thickness. The proposed solution to this problem implies that a critical thickness value does not exist. Crossover from 2D thermodynamics at low temperatures to 3D thermodynamics at high temperatures occurs for all films except ones with fewer than three atomic layers.

For the inhomogeneous (pseudomorphic) monatomic Fe layer the exponent of $5/2$ was found to hold with a high precision. The exponent of $5/2$ is well known from materials with dominating axial interaction such as hcp gadolinium and cobalt [8] and for MnF_2 [51]. We must, however, assume that the interactions are 2D but anisotropic in the pseudomorphic monatomic Fe layer and that the exponent of $5/2$ is common to crystalline materials with 1D interactions and to anisotropic or amorphous materials with 2D interactions. In other words, amorphous materials have an exponent which is characteristic for a dimensionality reduced by one compared to the same material in the crystalline state. Crystalline and amorphous bulk iron confirm this rule for 3D interactions.

Up to now it was not understood why in the FMR experiments on films, instead of the anticipated $T^{5/2}$ law [60] a $T^{3/2}$ law is observed for the state in which SSW are excited ($k \neq 0$). As a solution to this problem we propose that these experiments measure not the spin wave dispersion constant $D(T)$, but the magnetization of an artificial state with a periodically modulated anisotropic magnetization. In this state the interactions seem to be also anisotropic, such that instead of the T^2 law for the homogeneous magnetization ($k = 0$), the same $T^{3/2}$ law results as for the spontaneous magnetization of amorphous or polycrystalline samples. Moreover, the exchange interactions are definitely decreased for the inhomogeneous state of magnetization.

In conclusion, all three exponents (2, $3/2$ and $5/2$) pertinent to half-integer spin quantum numbers and 3D, 2D and 1D interactions can be observed for iron if it is suitably prepared. Iron therefore fits the universality scheme of table 1 which, originally, was set up for insulators with localized spin moments. Nickel and cobalt fit this scheme equally well. Although it is reasonable to assume that in contrast to the localized 3d and 4f electrons the band electrons are not coupled to larger moment units and that the spin value is $1/2$ in the itinerant ferromagnets [61], the common thermodynamic behaviour of insulators and itinerant ferromagnets is a puzzle.

A long-range magnetic order in 2D and 1D Heisenberg systems contradicts Bloch's theoretical conclusion [2] which was later on generalized by Mermin and Wagner [39] and by Bruno [40]. In these classical theories only bilinear (second-order) exchange interactions are considered. As we have shown in numerous experimental investigations, fourth-order exchange interactions, i.e. biquadratic, three-spin and four-spin interactions, are generally strong and cannot be neglected [25–27]. It is tempting to assume that these interactions stabilize the long-range magnetic order in Heisenberg materials with 2D and 1D interactions.

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