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The Curie temperature of the ferromagnetic transition metals and their compounds

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Abstract. The Curie temperatures of Fe, Co and Ni as well as of their compounds with Y (including $\text{Y}_2\text{Fe}_{14}\text{B}$) are calculated by including the effects of spin fluctuations in the long wavelength limit via a renormalisation of Landau coefficients. The numerical values depend on the use of recent calculations of correlation effects and of the band structure. As a result, good agreement is found with the experimental values of T_C of the materials covered.

1. Introduction

The calculation of the Curie temperature of the ferromagnetic 3d metals and their compounds is a non-trivial problem as at least two types of elementary excitations are involved: namely single-particle (Stoner) excitations, and spin fluctuations or paramagnons, as well as spin waves.

The Stoner model of itinerant electron magnetism usually gives a rather good description of the magnetic ground state at $T=0$ K. Regarding its applicability for finite temperatures, however, it has been reported frequently during the last 10 years that the Curie temperatures, determined from the Stoner model and from the ground state properties and the fine structure of the density of states at the Fermi energy, are much too high. Gunnarsson (1976) reported values of the Stoner Curie temperature T_C^S as being equal to 4400–6300 K for Fe and 2900 K for Ni. The reason for these unrealistically high values is not entirely due to the defects of the Stoner theory. Gunnarsson derived his values from band-structure calculations where the effects of exchange and correlation are treated within the local spin density (LSD) functional formalism. Employing this treatment always leads to overestimates of the exchange splitting ΔE which is calculated to be the expectation value of the difference of the spin up and spin down exchange potentials. As the value of T_C^S is closely related to the value of ΔE this systematic error also leads to an overestimation of the Curie temperature. Oleś and Stollhoff (1986) and Stollhoff (1987) improved these results by treating the missing correlation effects using their local approach (LA). Their values for the exchange splitting are not only significantly lower but they also agree with the experimentally determined band splitting.

Table 1 shows the values of ΔE from LSD and LA and the associated values of the Stoner model Curie temperature T_C^S . As these values of T_C^S are still higher than the experimental ones, the additional reduction must arise from spin fluctuation phenomena.

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Table 1. A comparison between the exchange splitting ΔE (in eV) and the resulting Stoner Curie temperature T_C^S (in K) calculated from pure local spin density approximation[†] (LSD) for exchange and correlation, and the same quantities calculated using the local ansatz (LA)[‡] to improve the treatment of correlation effects within LSD. T_F is the Fermi degeneracy temperature defined by $IN_p - 1 = T_C^S/T_F^2$. N_p is the density of states at the Fermi energy in the paramagnetic state; I is the Stoner parameter.

	ΔE^\dagger	IN_p^\dagger	$T_C^S^\dagger$	T_F	ΔE^\ddagger	IN_p^\ddagger	$T_C^S^\ddagger$
Fe	2.00	1.6	5300	6840	1.41	1.14	2560
Co	1.54	1.7	4000	4780	1.11	1.22	2240
Ni	0.58	2.1	2900	2765	0.38	1.42	1790

[†] Values given by Gunnarsson (1976).

[‡] Values given by Oleś and Stollhoff (1986).

2. The treatment of fluctuations in itinerant systems

We write the inverse susceptibility in the form given by Lonzarich and Taillefer (1985),

$$\frac{H}{M} = A + BM^2 + B(3\langle m_{\parallel}^2 \rangle + 2\langle m_{\perp}^2 \rangle) \quad (1)$$

where $\langle m_{\parallel}^2 \rangle$ and $\langle m_{\perp}^2 \rangle$ are the mean square values of the parallel and transverse local fluctuating moments. Also, in equation (1), A and B are the usual Landau coefficients as they appear in the analogous Stoner theory, being defined by

$$A = -\frac{1}{2\chi_0} \left(1 - \frac{T^2}{T_C^S} \right) \quad B = \frac{1}{2\chi_0 M_0^2}. \quad (2)$$

Here M_0^2 is the square of the equilibrium magnetic moment at $T=0$, given by $M_0^2 = |A/B|_{T=0}$ and χ_0 the exchange enhanced susceptibility at equilibrium given by Wohlfarth (1962) and Gersdorf (1962) as

$$\chi_0^{-1} = (4\mu_B^2)^{-1} (N^+(\epsilon_F)^{-1} + N^-(\epsilon_F)^{-1}) - I/2\mu_B^2 \quad (3)$$

where I is the Stoner parameter. Also T_C^S is the Stoner Curie temperature discussed in § 1.

Wagner and Wohlfarth (1986) introduced a significant step towards the treatment of spin fluctuations in itinerant systems. Noting that the fluctuating magnetic moments must decrease as the static magnetisation M increases, they expanded the fluctuation terms in powers of M ,

$$B(3\langle m_{\parallel}^2 \rangle + 2\langle m_{\perp}^2 \rangle) = a_1 - a_2 M^2 + a_3 M^4 \quad (4)$$

($a_1, a_2, a_3 > 0$). Mohn *et al* (1987) showed that this treatment leads to a renormalisation of the Landau coefficients A and B as used in equation (1) and $a_1(T)$ was shown to be given by

$$a_1(T) = 2KCP_C(1 - t_C^2) \frac{T}{T_C}. \quad (5)$$

Here K is the compressibility, C the magnetoelastic coupling constant and P_C the critical pressure for the disappearance of ferromagnetism. Also t_C is defined by the ratio T_C/T_C^S , where T_C is the experimental Curie temperature. Rewriting equation (5) we define as a

physical quantity $\chi_{\text{SF}}(T)$, the spin fluctuation susceptibility, with a_1 given by

$$a_1(T) = (1 - t_C^2) \frac{1}{\chi_{\text{SF}}(T)}. \quad (6)$$

The susceptibility χ_{SF} for $T \geq T_C$ may be obtained from equation (1). On this basis we find at $T = T_C$ that

$$\chi_{\text{SF}} = \frac{\langle m^2 \rangle}{3k_B T_C} \quad \text{where } \langle m^2 \rangle = \frac{1}{3}(3\langle m_{\parallel}^2 \rangle + 2\langle m_{\perp}^2 \rangle). \quad (7)$$

At the Curie temperature T_C the amplitude of the fluctuating magnetic moment can be expressed in terms of the static equilibrium magnetisation M_0 at $T=0$ by equation (4) of Mohn *et al* (1987) and a_1 then becomes

$$a_1(T_C) = \frac{5k_B T_C}{M_0^2}. \quad (8)$$

The renormalised Landau coefficient $A'(T_C) = A(T_C) + a_1(T_C)$ now determines the Curie temperature in terms of ground state properties related to the band structure. The treatment is also compatible with the Curie–Weiss constant.

At $T = T_C$ the bulk magnetisation M vanishes, giving

$$A' = A + a_1 = 0 = -\frac{1}{2\chi_0} \left(1 - \frac{T_C^2}{T_C^{\text{S}^2}} \right) + \frac{5k_B T_C}{M_0^2}, \quad (9)$$

so that the Curie temperature T_C is given by

$$\frac{T_C^2}{T_C^{\text{S}^2}} + \frac{T_C}{T_{\text{SF}}} - 1 = 0 \quad (10)$$

with T_{SF} a characteristic temperature describing the influence of spin fluctuations, given by

$$T_{\text{SF}} = \frac{M_0^2}{10k_B \chi_0}. \quad (11)$$

Equation (10) combines two extreme theories. These are, on the one hand, a pure spin fluctuation dependent description of magnetic properties where the Stoner Curie temperature is always assumed to be infinite (Moriya 1985). In this case the quadratic term in (10) vanishes and T_C is then equal to T_{SF} . Such a model may be applicable if T_C^{S} exceeds T_C by a factor of three or more, in which case the influence of the quadratic term is lower than $\sim 10\%$. At the other extreme, there is the Stoner theory, taking into account single-particle excitations only, a description which may be valid for systems where T_{SF} is large compared with T_C .

The result given in equation (10) is very similar to that of Lonzarich and Taillefer (1985) who found, on a completely different basis, that

$$\frac{T_C^2}{T_C^{\text{S}^2}} + \frac{T_C^{4/3}}{T_1^{4/3}} - 1 = 0. \quad (12)$$

The difference in the exponents in (10) and (12) is due to the influence of a finite cut-off wavevector in their theory, which is assumed to show a temperature dependence proportional to $T^{1/3}$ (see appendix 1).

The results from equation (10) can be illustrated by writing it as

$$\frac{T_C^2}{T_{SF}^2} + \frac{T_C}{T_{SF}} \frac{T_C^{S^2}}{T_{SF}^2} - \frac{T_C^{S^2}}{T_{SF}^2} = 0. \quad (13)$$

Plotting T_C/T_{SF} as a function of T_C^S/T_{SF} gives a universal curve for all systems (figure 1). This figure displays the excellent agreement between the experimental results (open circles) which were calculated using the experimental values of T_C and the theoretically predicted behaviour given by the solutions of equation (13).

3. Fe, Co and Ni

As the theory was developed for monoatomic solids, we now discuss the findings for Fe, Co and Ni. Table 2 shows the numerical results for these systems. The calculated values for T_C agree very well with the experimental ones, within an error of about 3–5%, but the actual behaviour of these three metals is different. For Fe and Ni the Stoner Curie temperature T_C^S is rather high compared with T_C , a fact which can be seen from the values of $t_C = T_C/T_C^S$ being 0.41 and 0.35, respectively. For these two systems the Curie temperature is mainly determined by the value of the spin fluctuation temperature T_{SF} . (The influence of the single-particle excitations is only 15–20%). For Co both the Stoner Curie temperature T_C^S and the spin fluctuation temperature T_{SF} have almost the same value. To obtain T_C , T_{SF} is reduced by 70% due to the influence of single-particle excitations. This result is rather surprising as it was thought that the influence of single-particle excitations should increase from Fe→Ni. The explanation can be found through the physical interpretation of equation (11). It contains two competing terms, the square of the equilibrium magnetisation M_0 , which tends to raise T_{SF} (therefore favouring single-particle behaviour), and the high-field susceptibility χ_0 which lowers T_{SF} . High values of M_0^2 will suppress the onset of spin fluctuations because the high value of the molecular field associated with it causes the fluctuating moments to keep in line with the direction of M_0 rather than fluctuating. The influence of χ_0 works in the opposite direction. High values of χ_0 mean that a reasonable amplitude of the fluctuating magnetic moment can be observed even at low temperatures.

Table 2. Comparison between the experimental and calculated values of the Curie temperature. T_C^S is the Stoner Curie temperature, taken from table 1. T_{SF} is the spin fluctuation temperature calculated from equation (11). $\langle m^2 \rangle^{1/2}$ is the fluctuating magnetic moment at T_C (calc) which may be compared with values given by other spin fluctuation theories; t_C is the ratio between the experimental Curie temperature and the Stoner Curie temperature as defined by Mohn *et al* (1987). In order to achieve a consistent description on the basis of the results from table 1, we performed self-consistent band calculations, employing the ASW method of Williams *et al* (1979) for the paramagnetic state, and calculated the high-field susceptibility from a rigid band shift with the reduced values for ΔE from table 1.

	T_C^S	T_{SF}	T_C (calc)	T_C (exp)	$\langle m^2 \rangle_{T_C}^{1/2}$	t_C
Fe	2560	1293	1068	1043	1.52	0.41
Co	2240	2439	1436	1388	0.93	0.62
Ni	1790	759	656	631	0.42	0.35

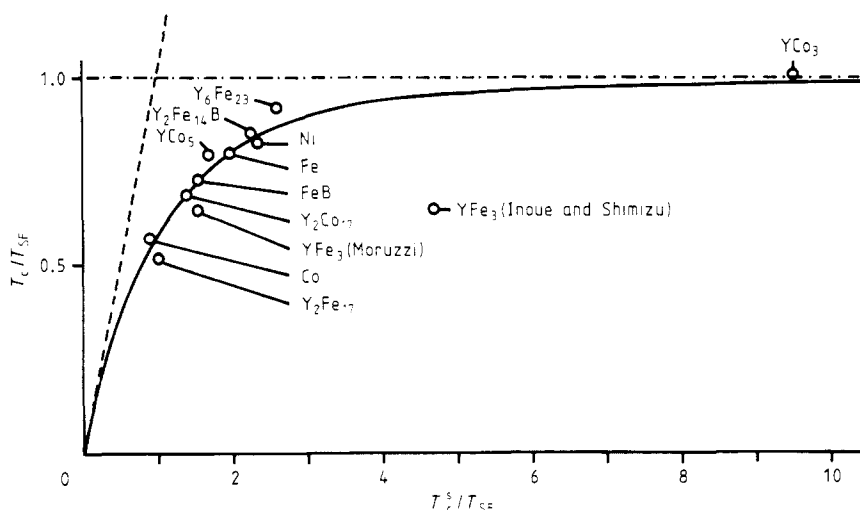


Figure 1. The solutions of equation (14) (full curve) and the actual values for 12 different systems (open circles). The broken line is the limit for pure Stoner behaviour ($t_C = 1$) whereas the asymptotic limit for pure fluctuation behaviour is given by the chain line. The values of T_C^S and T_{SF} are obtained as described in the captions of tables 1 and 2. The explanation for the two different points for YFe_3 is given in the caption to table 3.

For Fe the square of the magnetic moment would certainly suppress spin fluctuations. As iron is a weak ferromagnet its high-field susceptibility is large, so that spin fluctuations take over again. Ni is the opposite extreme but leading to the same result. As M_0^2 is a factor 15 times smaller than for Fe one would expect Ni to be entirely governed by spin fluctuations. The small susceptibility (Ni is a strong ferromagnet), however, makes it behave very much like iron again. Nevertheless the Curie temperatures of both metals are mainly determined by spin fluctuations so that we want to call such systems fluctuation systems.

Co behaves differently. Its relatively high magnetic moment M_0 suppresses spin fluctuations, but the small value for χ_0 (Co is a strong ferromagnet as is Ni) makes T_{SF} very large (even larger than T_C^S), so that the Curie temperature of Co is mainly determined by single-particle excitations. We want to call systems like this Stoner systems. The value of $t_C = T_C/T_C^S$ can be used to characterise the type of system involved. Stoner systems can be found for $t_C > 0.5$, fluctuation systems for $t_C < 0.5$. This also means in terms of the quantity T_C^S/T_{SF} that Stoner systems belong to values $0 \leq T_C^S/T_{SF} \leq 1.5$ and fluctuation systems cover the range from 1.5 to infinity.

This behaviour is shown in figure 1 where Fe, Co and Ni are found to be close to the ideal curve. The Y-Fe and Y-Co compounds also shown in figure 1 are discussed in the next section. The plotted curve is found to lie between two straight lines, one the solution for pure fluctuation behaviour, given by

$$T_C/T_{SF} - 1 = 0 \quad (14)$$

which is the asymptotic solution of equation (10), and the other the tangent to the curve at the origin, given by

$$T_C^2/T_C^{S^2} - 1 = 0 \quad (15)$$

which is the solution of equation (10) for pure single-particle behaviour. Equation (15) corresponds to $t_C = T_C/T_C^S = 1$. Co, with its strong single-particle influence ($t_C = 0.62$), is consequently found to be near the left end of the curve, close to the tangent $t_C = 1$. Fe and Ni show similar behaviour and are found to be further to the right, as also described by the lower values of $t_C = 0.41$ and $t_C = 0.35$, respectively.

4. The application to intermetallic compounds

On the basis of a theory for the Curie temperature for monoatomic solids we now describe an application to intermetallic compounds and alloys. As the spin fluctuations involved are assumed to exist in the long wavelength limit they are treated as uniform fluctuations of the magnetisation regardless of wavevector space. As the wavelength of these fluctuations is certainly larger than the interatomic distances an averaging process is involved. The spin fluctuation temperature may therefore depend only on the average magnetic moment and on the average susceptibility of the magnetic constituents in a compound or alloy. A second argument for this random phase-like approach comes from the fact that any fluctuations appear to be of extremely long wavelength in the vicinity of a critical point. However it may well be that this assumption for the spin fluctuation temperature is not generally valid. Applying these assumptions to the Y-Fe and Y-Co intermetallic compounds shows excellent values for T_C compared with the experimental ones (table 3).

Table 3. A comparison between experimental and calculated values for T_C . The band-structure data are taken from Inoue and Shimizu (1985, 1986). Experimental data for T_C are taken from Buschow (1989) using the mean values within the temperature ranges state. All susceptibilities used to determine T_{SF} were calculated using the values of Stollhoff (1987) for the Stoner parameter I , being equal to 0.65 eV for iron and 0.68 eV for Co. The amplitude of the fluctuating magnetic moment $\langle m^2 \rangle^{1/2}$ at T_C is given in units of Bohr magnetons.

	T_C^S	T_{SF}	T_C (calc)	T_C (exp)	$\langle m^2 \rangle^{1/2}$	t_C
Y_2Fe_{17}	612	615	379	317	1.02	0.52
Y_6Fe_{23}	1312	523	458	481	1.46	0.37
YFe_3	3721 (1223)†	787	754 (598)†	514	1.02	0.14 (0.42)†
$Y_2Fe_{14}B$	1508	674	575	573	1.66	0.38
$FeB $	1223	792	600	582	0.74	0.48
$Y_2Co_{17}‡$	2390	1706	1243	1185	1.04	0.50
YCo_5	2081	1235	968	987	1.02	0.47
Y_2Co_7	1218	899	646	640	0.88	0.60
YCo_3	2827	210 (298)§	208 (295)§	300	0.60	0.11

† The quantities in parentheses refer to a paramagnetic self-consistent band-structure calculation by Moruzzi *et al* (1983).

‡ Inoue and Shimizu (1985) deal only with d bands, which leads to an unrealistically low susceptibility for Y_2Co_{17} . To correct for this defect of their model we assumed N^+ to have the same value as for pure Co including s and p bands.

§ Szpunar and Kozarzewski (1977) also performed calculations of the Y-Co system employing a CPA method. Their values for the magnetic moment of the Co site agree with the data by Inoue and Shimizu (1985). Only for YCo_3 do they find $1.0 \mu_B$ instead of $0.84 \mu_B$. The values in parentheses refer to their calculation.

|| Band-structure data taken from Mohn and Pettifor (1987), experimental value of T_C taken from Cadeville (1965).

The values of the Stoner Curie temperature T_C^S used here were derived using the relation given in the caption to table 1. For the Fermi degeneracy temperature we used the same values as for the pure metals. As the Stoner exchange energy I is known to be rather constant regardless of the local environment we calculated T_C^S using the band-structure data as stated in table 3.

The actual value of T_C^S is, however, not very critical because the influence of the spin fluctuation temperature T_{SF} usually is much stronger than the influence of T_C^S so that possible uncertainties in T_C^S should not affect the results greatly. Figures 2(a) and (b) display the experimental and theoretical Curie temperatures for $Y_{1-x}Co_x$ and $Y_{1-x}Fe_x$, respectively. The full circles which denote the value of the spin fluctuation temperature T_{SF} show that, for the Y-Co system, the influence of single-particle excitations systematically increases as one approaches pure Co. However, for the Y-Fe compounds no such systematic behaviour occurs. The reason for this can be found in the fact that for the weakly ferromagnetic iron compounds, the high-field susceptibility shows large variations and subsequently the character of the T_C behaviour changes. For the strongly ferromagnetic Co systems, however, the high-field susceptibility has an almost constant value. It is certainly worthwhile to note another difference between Y-Co and Y-Fe compounds. Whereas for the Y-Co compounds HCP Co fits nicely into the general trend towards the full curve, this is not the case for the Y-Fe compounds and BCC Fe. This difference can be attributed to a change in structure as most of the Y-Fe compounds can

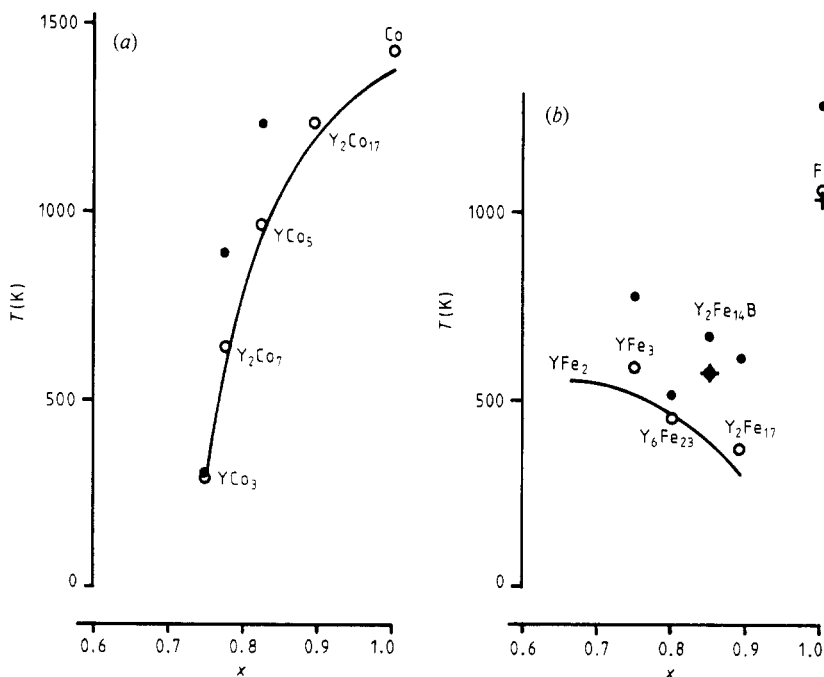


Figure 2. Experimental Curie temperatures $T_C^{(exp)}$ (full curve), theoretical T_C (open circles) and associated values of the spin fluctuation temperature T_{SF} (full circles) for (a) $Y_{1-x}Co_x$ and (b) $Y_{1-x}Fe_x$. (T_{SF} for HCP Co is not plotted because of its value being too high.) In (b) where two of the displayed systems ($Y_2Fe_{14}B$ and BCC Fe) are found to be off the general trend for $T_C^{(exp)}$, the values for $T_C^{(exp)}$ are given by crosses.

be derived from 'FCC'-like structures, which show higher densities of states at the Fermi energy and subsequently a lower spin fluctuation temperature T_{SF} . The crystallographic change to BCC for pure Fe lowers χ_0 by a factor of about three compared with Y_2Fe_{17} and hence leads to $T_{\text{C}} = 1043$ K.

The compound $\text{Nd}_2\text{Fe}_{14}\text{B}$ has been found to be an excellent permanent magnet with a large energy product (Sagawa *et al* 1984). Because the d-band structure in $\text{Nd}_2\text{Fe}_{14}\text{B}$ is similar to that in $\text{Y}_2\text{Fe}_{14}\text{B}$, a study of the band-structure of the latter compound may give an answer to one of the remaining problems relating to this compound, namely the relatively low Curie temperature which makes the industrial applications somewhat problematic.

From the understanding of the mechanisms determining the Curie temperature of such systems we may be able to improve this technologically very important feature of the modern permanent magnets.

Raising the Curie temperature of $\text{Nd}_2\text{Fe}_{14}\text{B}$ can be done in several ways. The most unlikely one is by raising M_0 , because the average magnetic moment is already found to be $2.32 \mu_{\text{B}}$. A proper strategy may be to raise T_{C}^{S} or lower χ_0 which seems to be the most promising approach. This can be done by replacing some of the 16 Fe atoms in the crystallographic k_2 position, e.g. by another magnetic 3d atom. Replacing one of these sites by Co should raise T_{C} by ~ 30 K, because Co carries a magnetic moment of about $1.6 \mu_{\text{B}}$ which will lower the average magnetic moment to $2.2 \mu_{\text{B}}/3\text{d atom}$. Co, a strong ferromagnet, will also lower the spin up density of states and therefore the high-field susceptibility. In general, Fe atoms should be replaced by atoms that show a similar electronic structure, in which case they easily hybridise with the d bands of the host lattice. Using largely different atoms may lead to a drastic decrease of T_{C} , because they not only carry no magnetic moment but may also form an impurity state which usually leads to a Lorentzian shaped high density of states around the Fermi energy.

High and narrow peaks in the density of states usually occur due to flat bands in the Brillouin zone or from degeneracies of bands with similar symmetry. In the latter case a distortion of the lattice can break the symmetry and subsequently lift the degeneracy. Flat bands near the top of the d band are very common in d metals (e.g. HCP Co, FCC Pd) where these effects may be overcome by implanting atoms on interstitial sites to remove long-range translational symmetry.

The results of table 3 are concerned with materials with relatively high T_{C} values, ≥ 300 K. For those with lower Curie points the analysis based on relation (7) breaks down and must be replaced by one involving saturating moments, replacing the linear dependence on T . A discussion of this effect, with application to weak itinerant ferromagnets such as ZrZn_2 ($T_{\text{C}} = 28$ K), will be published elsewhere.

5. Conclusion

The theory presented in this paper yields a satisfactory description of the Curie temperatures of 3d metals, alloys and compounds in a temperature range not too close to zero. It was shown that for most systems the influence of spin fluctuations clearly dominates the description of T_{C} ; thus for most of the cases described the spin fluctuation temperature T_{SF} , defined by equation (11), gives at least a very good estimate for the experimental value of T_{C} . As the calculation of T_{SF} does not require any free parameters, but contains only quantities which are given by a band-structure calculation or by experiment, it is very easy to apply the theory to many different materials. The quantities

M_0 and χ_0 are directly related to the electronic properties of a solid and therefore provide a ready understanding of the forces destroying magnetic order in a solid. The coefficient $t_C = T_C/T_C^S$ gives a measure of the relative importance of single-particle excitations ($t_C = 1$) and spin fluctuations ($t_C = 0$) in this sense, and tables 2 and 3 give values for a variety of materials. As the present model was developed to describe the Curie temperature of itinerant systems we would expect it to work also for other itinerant electron materials like the actinides and their compounds. A successful treatment of systems like rare earths with localised electrons seems to be very unlikely on this basis. We have here achieved the aim of an earlier discussion (Wohlfarth 1980): '... it seems clear that both Stoner excitations and spin fluctuation effects determine together the physical behaviour of ferromagnetics ...; we hope to have shown that (the) data must be analysed very carefully and without prejudice'.

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Appendix 1

From equations (7)–(9) we derive an expression for the mean square of the fluctuating magnetic moment at T_C ,

$$\langle m^2 \rangle = 6k_B T_C \chi_0 = \frac{3k_B T_C}{|A|}. \quad (\text{A.1})$$

In (A.1) we made use of the definition of the Landau coefficient A (equation (2)) for small values of T_C/T_C^S . This result can now be formally compared with the formula given by Lonzarich and Taillefer (1985):

$$\langle m^2 \rangle = 3k_B T_C \sum_{q < q_C} (\tilde{A} + cq^2)^{-1}. \quad (\text{A.2})$$

Here A and \tilde{A} must not be confused as \tilde{A} is defined to vanish at T_C . What remains at T_C is a sum over all q vectors for a fixed spin wave stiffness constant c . This point illustrates the different approaches of the two theories, as the present model was formulated in the long wavelength limit ($q \rightarrow 0$) throughout and therefore does not contain any wavevector-dependent quantities. Both formulae, however, lead to a spin fluctuation temperature showing striking similarities:

$$T_{\text{SF}} = \frac{M_0^2}{10k_B \chi_0}$$

(present work)

$$T_1 = \frac{2.387(\hbar\gamma)^{1/4} M_0^{3/2} c}{k_B}$$

(Lonzarich and Taillefer 1985) where γ is related to the relaxation frequency of an isotropic band and c is proportional to χ_0^{-1} .

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