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Suppression of spin fluctuations by the magnetic field and Co concentration in amorphous $Fe_{90-x}Co_xZr_{10}$ alloys

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Abstract. Spin fluctuations manifest themselves in the temperature dependence of the spontaneous magnetization $M(T, H = 0)$ or the 'in-field' magnetization $M(T, H)$, through a contribution that varies with T as $[M(T, H)/M(0, H)] = 1 - A(H)T^2$ in the intermediate-temperature range and as $[M(T, H)/M(0, H)]^2 = 1 - A'(H)T^{4/3}$ for temperatures in the vicinity of T_c (Curie point) but still away from criticality. Suppression of spin fluctuations by either the Co concentration in a-$Fe_{90-x}Co_xZr_{10}$ alloys or the field H for a given composition in the a-$Fe_{90-x}Co_xZr_{10}$ and a-$Fe_{90+y}Zr_{10-y}$ alloy series is, for the first time, monitored through the decrease in the coefficients A and A' with increasing x for $H = 0$ or with H for fixed x or y. While $A(0)$ and $A'(0)$ *scale* with T_C^{-2} and $T_C^{-4/3}$, respectively, $A(H)$ and $A'(H)$ follow the relation $\mathcal{A}(H_{eff}) = \mathcal{A}(0)[1 - \mathcal{B}H_{eff}^{\psi}]$ where $\mathcal{A} = A$ or A', $\mathcal{B} = B$ or B', $\psi = n$ or n' and H_{eff} is the external field corrected for demagnetization. In contrast with the non-monotonic variation in B and n with x, B' varies with x as $B'(x) = B'(0)[1 - \mu x^{\phi}]$ and $n' = 0.50(2)$ is *independent* of x in the range $\leqslant x \leqslant 6$. The spin fluctuation model explains many of, but not all, these features.

1. Introduction

A recent bulk magnetization (BM) study [1–3] of amorphous (a−)$Fe_{90+y}Zr_{10-y}$ ($y = 0$, 1) and $Co_{90}Zr_{10}$ alloys permitted us to draw a number of conclusions concerning the nature of magnetism in these systems that include the following.

(i) Local spin-density fluctuations give a dominant contribution to the thermal demagnetization of the spontaneous magnetization $M(T, 0)$ in the former set of alloys over a wide range of intermediate temperatures and for temperatures close to T_C, while the Stoner single-particle excitations are mainly responsible for the decrease in $M(T, 0)$ with increasing temperature for $T \gtrsim 0.1T_C$ in a-$Co_{90}Zr_{10}$.

(ii) External magnetic fields $H \simeq 15$ kOe strongly suppress the spin fluctuations in a-$Fe_{90+y}Zr_{10-y}$ alloys but have almost no effect on the temperature dependence of magnetization in a-$Co_{90}Zr_{10}$.

The existence of spin-density fluctuations in a-$Fe_{90+y}Zr_{10-y}$ alloys has also been recently inferred from the electrical resistivity data [4]. The BM study, therefore, raises the following basic questions.

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(a) Does the absence of spin fluctuations in a-$Co_{90}Zr_{10}$ imply that, as more and more of the Fe atoms in a-$Fe_{90}Zr_{10}$ are substituted by Co atoms, spin fluctuations become progressively suppressed such that, when Co completely replaces Fe, spin fluctuations are totally suppressed or does it have some other interpretation?

(b) Why is the temperature dependence of magnetization unaffected by the external magnetic field in a-$Co_{90}Zr_{10}$?

(c) Can the suppression of spin fluctuations by the field be quantified?

In order to seek the answers to these questions, a detailed systematic investigation of magnetization as a function of temperature and external magnetic field in a-$Fe_{90-x}Co_xZr_{10}$ and a-$Fe_{90+y}Zr_{10-y}$ alloys was undertaken.

2. Experimental details

Magnetization M versus H isotherms in fields up to 15 kOe were measured for a-$Fe_{90-x}Co_xZr_{10}$ ($x = 0, 1, 2, 3, 4, 6$) and a-$Fe_{90+y}Zr_{10-y}$ ($y = 0, 1$) alloys at 1 K intervals in the temperature range 70 K $\leqslant T \lesssim T_C - 15$ K and at temperatures about 0.15 K apart in the range $T_C - 15$ K $\lesssim T \lesssim T_C$ using a vibrating-sample magnetometer (Princeton Applied Research VSM 4500 system). The magnetic moment was measured to a relative accuracy of 5×10^{-5} emu and the temperature stability was better than ± 25 mK (± 40 mK) for $T \leqslant 300$ K ($T > 300$ K). Details of sample preparation and characterization have been given in our earlier reports [1–3, 5]. M versus H isotherms are converted into a form that gives M as a function of T at fixed values of H, 0.5 kOe apart, in the interval 1.5 kOe$\leqslant H \leqslant$ 15 kOe. Such sets of data are referred to as the 'in-field' magnetization or $M(T, H)$ data in the subsequent text. A modified Arrott ($M^{1/\beta}$ versus $(H/M)^{1/\gamma}$) plot (MAP) is constructed out of the M versus H isotherms with the choice of spontaneous magnetization and initial susceptibility critical exponents β and γ that makes the MAP isotherms a set of parallel straight lines in the critical region. In this plot, H is the external field corrected for demagnetization.

3. Results and discussion

The values of spontaneous magnetization $M(T, 0)$ at different temperatures are computed from the intercepts on the ordinate ($M^{1/\beta}$ axis) obtained by extrapolating high-field linear portions of the MAP isotherms to $H = 0$. A detailed range-of-fit analysis (in which the values of free-fitting parameters and the quality of fits are continuously monitored as the temperature interval $T_{min} \leqslant T \leqslant T_{max}$ is progressively narrowed down by keeping $T_{min}(T_{max})$ fixed at a given value and lowering (raising) $T_{max}(T_{min})$ towards $T_{min}(T_{max})$ and whose details have been given elsewhere [1–3, 6]) of the $M(T, 0)$ and $M(T, H)$ data based on the expressions

$$m(H) \equiv [M(T, H)/M(0, H)] = 1 - A(H)T^2 \tag{1}$$

$$m^2(H) \equiv [M(T, H)/M(0, H)]^2 = 1 - A'(H)T^{4/3} \tag{2}$$

has been carried out to determine the field dependence of the coefficients A and A' of the T^2 and $T^{4/3}$ terms. In the event that the contribution to the thermal demagnetization of $M(T, 0)$ due to enhanced spin fluctuations *dominates* over that arising from the single particle excitations, the spin fluctuation model [7] predicts that $m(H = 0)$ varies with temperature in the *intermediate*-temperature range as T^2, i.e. equation (1) with

$$2A(H = 0) \equiv 2A(0) = T_0^{-2} \tag{3}$$

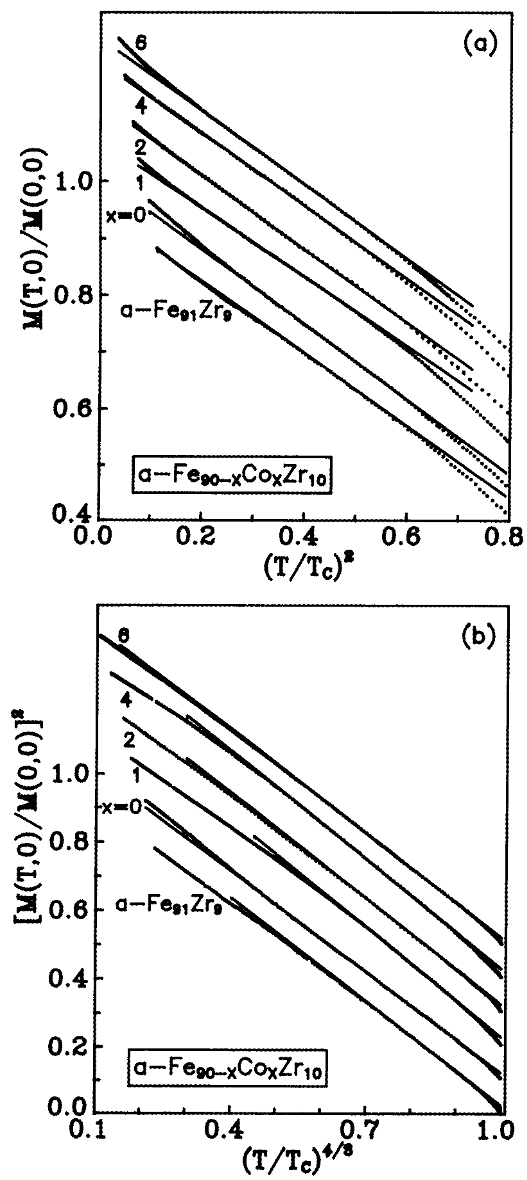

Figure 1. (a) $M(T,0)/M(0,0)$ versus $(T/T_C)^2$ and (b) $[M(T,0)/M(0,0)]^2$ versus $(T/T_C)^{4/3}$. Note that the zero on the ordinate scale for the alloys with $x = 0, 1, 2, 3, 4, 6$ is shifted upwards by 0.06, 0.12, 0.18, 0.24, 0.30 and 0.10, 0.20, 0.30, 0.40, 0.50 with respect to that for a-$Fe_{91}Zr_9$ in (a) and (b), respectively.

and for temperatures in the vicinity of T_C but still away from the critical point (i.e. for temperatures just outside the asymptotic critical region), $m^2(H = 0) \sim T^{4/3}$, i.e. equation (2) with

$$A'(H = 0) \equiv A'(0) = T_{SF}^{-4/3}. \tag{4}$$

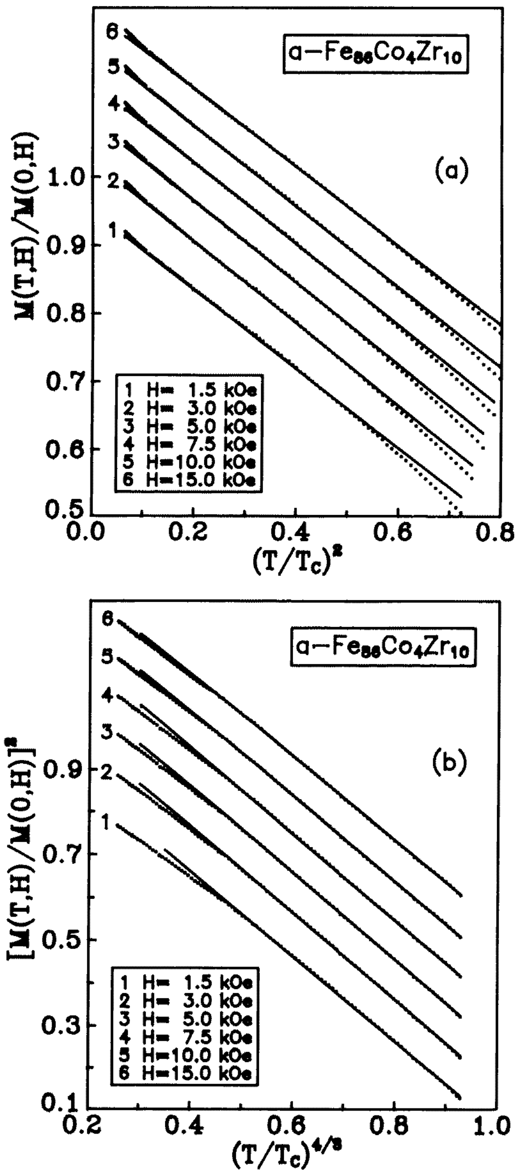

Figure 2. Plots of (a) $M(T, H)/M(0, H)$ versus $(T/T_C)^2$ and (b) $[M(T, H)/M(0, H)]^2$ versus $(T/T_C)^{4/3}$ at a few representative field values. Note that the zero of the ordinate scale for the data bearing the numbers 2, 3, 4, 5, 6 is shifted upwards by 0.05, 0.10, 0.15, 0.20, 0.25 and 0.08, 0.16, 0.24, 0.32, 0.40 with respect to data 1 in (a) and (b), respectively.

Equations (1) and (2) generalize the expressions predicted by the spin fluctuation model to include the effect of the external magnetic field on spin fluctuations. The main outcome of this analysis is the following.

(i) The temperature dependence of both ‘zero-field’ and ‘in-field’ magnetizations is best described by the expression that combines equations (1) and (3) in the intermediate-temperature range $T^* \leqslant T \leqslant T^{**}$, and by that which combines equations (2) and (4) for temperatures just outside the asymptotic critical region $T' \leqslant T \leqslant T''$ for the alloys with $x = 0, 1, 2, 4, 6$ and $y = 0, 1$, as is evident from figures 1 and 2; T^*, T^{**}, T' and T'', although *composition dependent*, possess typical values of $0.45T_C$, $0.74T_C$, $0.75T_C$ and $0.95T_C$, respectively.

(ii) When $H = 0$, the coefficients $A(0)$ and $A'(0)$ of the T^2 and $T^{4/3}$ terms in equations (1) and (2) decrease with increasing x or decreasing y (figure 3).

(iii) For a given composition, the coefficients $A(H)$ and $A'(H)$ appearing in equations (1) and (2) decrease with increasing applied field strength (figure 3).

It is well known that the Stoner model grossly overestimates T_C because this model holds a weak temperature dependence of the thermal density of states (DOS) (the one-electron DOS times the Fermi function) solely responsible for the T^2 decrease in $M(T, 0)$. By contrast, if local spin-density fluctuations dominantly contribute to the thermal demagnetization of $M(T, 0)$, one expects the spin fluctuation model [7] to predict correctly the value of T_C, i.e. $T_0 \approx T_C$ and $T_{SF} \simeq T_C$ in equation (3) and (4). Consistent with this expectation, the values of T_0 and T_{SF} calculated from equations (3) and (4) using the observed values of the coefficients $A(0)$ and $A'(0)$ demonstrate that $T_0/T_C = 0.86(1)$ and $T_{SF}/T_C = 1.02(1)$ regardless of alloy composition. Since these ratios are constant (within the uncertainty limits) over the composition range $x \leqslant 6$ and $y \leqslant 1$, it is not surprising that the coefficients $A(0)$ and $A'(0)$ *scale* with T_C^{-2} and $T_C^{-4/3}$ (figure 4) in accordance with equation (3) and (4). In view of the observation that T_C increases roughly linearly [5] with increasing x, the results $A(0) \propto T_C^{-2}$ and $A'(0) \propto T_C^{-4/3}$ offer a simple explanation for the decrease in $A(0)$ and $A'(0)$ (and hence for the suppression of spin fluctuations) with increasing x (figure 3). In order to make the underlying mechanism more transparent, we proceed as follows. The values of the Stoner parameter I have been calculated from the zero-field differential susceptibility $\chi(0, 0)$ at 0 K, the spontaneous magnetization $M(0, 0)$ at 0 K ($\chi(0, 0)$ and $M(0, 0)$ for each alloy have been computed from the slope and intercept on the ordinate of M^2 versus H/M plot isotherm taken at $T = 5$ K in fields up to 70 kOe) and the density $N(E_F)$ of single-particle states at the Fermi level E_F (estimated from the reported [8] values of the coefficient of the electronic specific heat after correcting them for the electron–phonon enhancement). Since both I and $M(0, 0)$ increase with increasing x, the exchange splitting of bands given by $\Delta E = IM(0, 0)/N\mu_B$, where N is the number of spins per unit volume, also increases with increasing x. A direct consequence of the increase in ΔE with increasing x is that the formation of correlated particle–hole pairs (local spin-density fluctuations) becomes increasingly difficult as x is increased. This leads to a progressive suppression of spin fluctuations by Co substitution. When the Co concentration is increased beyond $x = 6$, the spin fluctuation contribution to $M(T, 0)$ diminishes rapidly with the result that for compositions with x very close to 90 this contribution is negligibly small compared with that arising from single-particle excitations. Thus, a-$Co_{90}Zr_{10}$ represents an extreme situation in which the particle–hole pair excitations are very *weakly* correlated and the value of T_C is essentially determined by the single-particle excitations alone.

A rapid decrease in $A(H)$ and $A'(H)$ with increasing H (figure 3) is a clear indication of the suppression of spin fluctuations by the field. The effect of increasing H in the itinerant-electron picture is to increase the splitting between the spin-up and spin-down subbands and hence, by analogy to the influence of increasing ΔE by Co substitution on spin fluctuations discussed above, the field, like x, strongly suppresses the local spin-density fluctuations. It is also noticed from figure 3 that the rate at which the coefficient A or A' decreases with increasing H slows down considerably as x increases. In view of the observation that, even

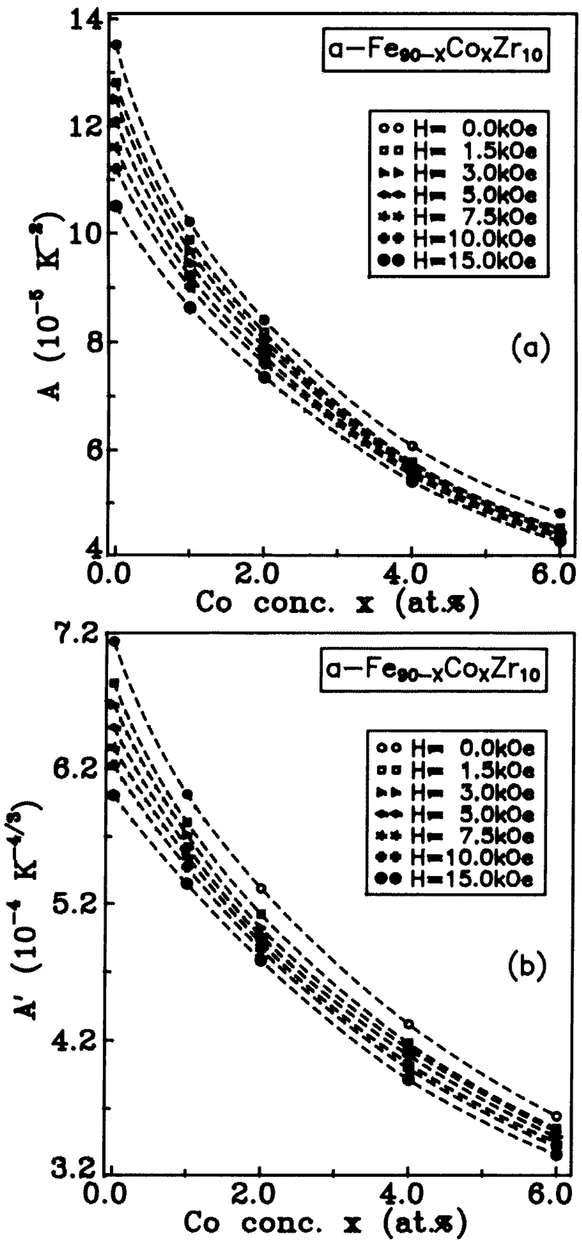

Figure 3. Concentration dependence of the coefficients (a) A of the T^2 term in equation (1) and (b) A' of the $T^{4/3}$ term in equation (2) for a few selected values of H.

in the absence of H, progressive replacement of Fe by Co leads to a strong suppression of spin fluctuations, the coefficients A and A' are far less *sensitive* to H for higher Co concentrations than for lower values of x because, at higher Co concentrations, the local spin-density fluctuations are already suppressed to a large extent even at $H = 0$ and the effect of H is reduced to a relatively insignificant level. An extreme situation arises when

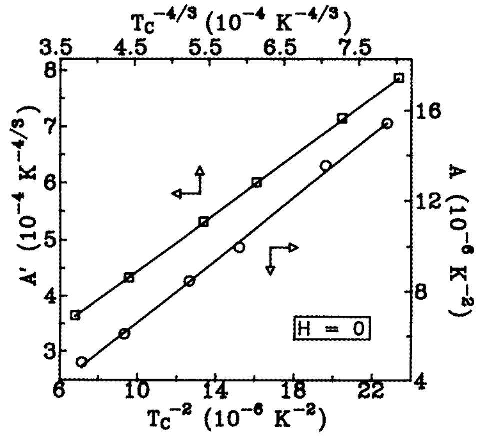

Figure 4. $A(H_{eff} = 0)$ versus T_C^{-2} and $A'(H_{eff} = 0)$ versus $T_C^{-4/3}$ plots.

the Co concentration approaches $x = 90$ in that spin fluctuations are completely suppressed even in the absence of H and no further suppression is possible by the external field. This inference conforms well with our earlier observation [3] that $M(T, 0)$ and $M(T, H)$ data even for fields as high as 15 kOe coincide with one another at all temperatures below 300 K in the case of a-$Co_{90}Zr_{10}$. Another important finding that merits attention is that the empirical relation

$$\mathcal{A}(H_{eff}) = \mathcal{A}(0)[1 - \mathcal{B}H_{eff}^{\psi}] \tag{5}$$

(where $\mathcal{A}$, $\mathcal{B}$ and ψ stand for A or A', B or B' and the exponent n or n', respectively, and H_{eff} is the effective field obtained after correcting H for demagnetization) closely reproduces the variation in A or A' with H_{eff} observed for $x \leqslant 6$ and $y = 0, 1$. While the parameters B, n and B' depend on the Co concentration, $n' = 0.50(2)$ is *independent* of x or y in the composition range covered in the present experiments. The $H_{eff}^{1/2}$ power-law dependent of A' on H_{eff} is clearly shown by the $A'(H_{eff})/A'(0)$ versus $H_{eff}^{1/2}$ plot in figure 5. However, the slope B' of the straight lines depicted in figure 5 decreases with increasing x in accordance with the empirical relation $B'(x) = B'(0)[1 - \mu x^{\phi}]$ in which $\mu = 4.15(5) \times 10^{-4}$ and $\phi = 0.25(2)$ while $B'(0) = 1.30(2) \times 10^{-3}$ is the experimentally determined value for the alloy with $x = 0$.

The theoretical attempts [9] made so far to quantify suppression of local spin-density fluctuations by the external magnetic field within the framework of the spin fluctuation model [7, 10] cannot be regarded as satisfactory because a *large number* of adjustable parameters (as against just two parameters $\mathcal{B}$ and ψ in equation (5)) and the *unrealistic* electron-gas model have been used to achieve quantitative agreement with the experimental $M(T, H)$ data obtained for Sc_3In. Moreover, while attempting a quantitative comparison between theory and experiment [9, 10], due consideration has not been given to the observation that different types of excitation are primarily responsible for the decay of $M(T, H)$ in different temperature ranges. Using the spin fluctuation model it is difficult to make specific predictions about the effect of the field on the local spin-density fluctuations because these fluctuations do not explicitly depend on H but, by virtue of their dependence on M, indirectly couple to H via M. However, for $T \sim T_C$, great simplification results from the fact that the Bose function $n(\omega) \simeq k_B T/\hbar\omega$ and the inverse longitudinal susceptibility

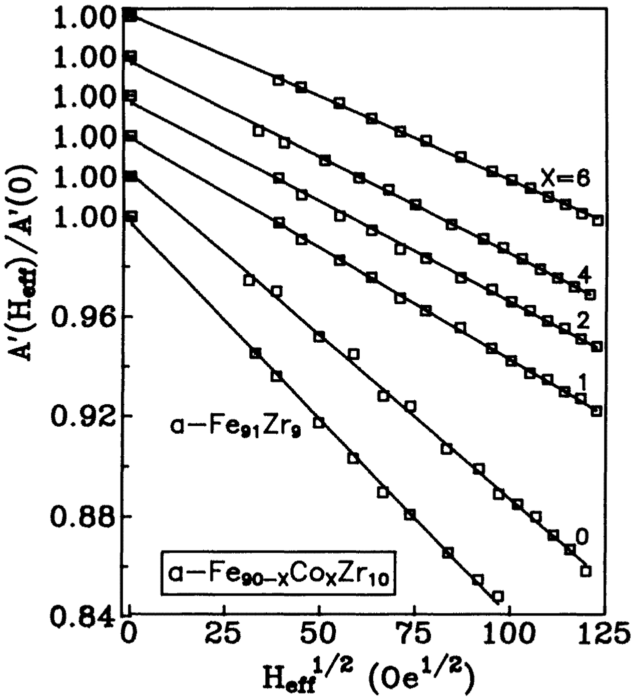

Figure 5. $A'(H_{eff})/A'(0)$ versus $H_{eff}^{1/2}$ plots for different compositions.

$\chi_{\parallel}^{-1} \simeq \chi_{\perp}^{-1}$ (inverse transverse susceptibility) and the 'in-field' magnetization can be put into the form [11]

$$\left[\frac{M(T,H)}{M(0,0)}\right]^2 = 1 - \left(\frac{T}{T_C}\right)^{4/3}\left[1 - \frac{\pi}{2q_c}\sqrt{\frac{g\mu_B}{D}}\sqrt{H}\right] \tag{6}$$

where q_c is the temperature-dependent cut-off wavevector [7]. In the light of equations (2) and (4), equation (6) has the same form as equation (5) with $B' = (\pi/2q_c)(g\mu_B/D)^{1/2}$. Thus, the spin fluctuation model correctly predicts the $\sqrt{H}$ dependence of the coefficient A'. Since the value of q_c depends on the band-structure details which are not available at present, we assume that $q_c \simeq 1$ Å^{-1} (*independent* of composition) and insert this value as well as the observed values of the spin-wave stiffness D [12] and splitting factor g [5] in the above expression with the result that B' possesses the values of 9.6×10^{-4} and 8.1×10^{-4} for the alloys with $x = 0$ and 6, respectively, as against the corresponding observed values of 13.0×10^{-4} and 6.6×10^{-4}. Considering that the composition dependence of q_c is not taken into account in the above calculation, agreement between theory and experiment is quite good. By contrast, such a comparison cannot be made at intermediate temperatures because no theoretical predictions, based on the spin fluctuation model, are available at present in this temperature range. (Note that, at such temperatures, the parameters $\mathcal{B}$ and ψ in equation (5) exhibit a *non-monotonic* variation with x.) Therefore, a theory based on the spin fluctuation model, which offers a quantitative explanation for the field dependence of the coefficient A observed in the intermediate-temperature range is called for.

4. Summary

A relation involving only two adjustable parameters has been proposed, for the first time, to quantify suppression of local spin-density fluctuations by an external magnetic field in itinerant ferromagnets. This relation is consistent with the predictions of the spin fluctuation model, particularly for temperatures in the vicinity of, but still away from, the Curie point. In addition, a simple but qualitative explanation is provided for the suppression of local spin-density fluctuations with Co concentration in a-$Fe_{90-x}Co_xZr_{10}$ alloys within the framework of the itinerant model.

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