CHAPTER 3
LOW-LYING MAGNETIC EXCITATIONS

In this chapter, the magnetization data taken over a wide temperature range extending from 4.2 K to \( T_C \) on amorphous \( Fe - Zr, Fe - Co - Zr \) and \((Fe,Ni)\)-metalloid alloy systems are presented, analyzed and discussed in the light of existing theoretical models.

A. \( a-Fe_{90+y}Zr_{10-y} \) and \( a-Fe_{90-x}Co_xZr_{10} \) alloys

3.1. Results and Analysis

3.1.1. 'In-field' Magnetization

The reduced 'in-field' magnetization, \( \frac{M(T,H)}{M(0,H)} \), for \( a-Fe_{90+y}Zr_{10-y} \) \((y = 0, 1)\) and \( a-Fe_{90-x}Co_xZr_{10} \)(\(z = 0, 1, 2, 4, 6, 8\) and 10) alloys measured at different temperatures in a constant magnetic field of 10 kOe by VSM (vibrating sample magnetometer) and SQUID magnetometer is plotted as a function of temperature in Fig. 3.1. A remarkably good agreement between the SQUID and VSM data in the overlapping temperature range is noticed from this figure. An elaborate analysis of the \( M(T,H) \) data, whose details are given below, is attempted based on the following expressions, predicted by the conventional spin-wave (SW) theory [1-3], Stoner model [3,4] and theoretical treatments [5] that include local spin density fluctuations (LSF) in addition to spin waves and single-particle (SP) excitations, for various contributions to the thermal demagnetization of either 'in-field' magnetization, i.e., \( \Delta m(T,H) = [M(0,H) - M(T,H)]/M(0,H) \), or spontaneous magnetization, i.e., \( \Delta m(T,0) = [M(0,0) - M(T,0)]/M(0,0) \) (for details see Chapter 1),

\[
\Delta m_{SW}(T,H) = \frac{g\mu B}{M(0,H)} \left\{ Z \left( \frac{3}{2}, t_H \right) \left( \frac{k_BT}{4\pi D(T)} \right)^{3/2} + 15\pi \beta Z \left( \frac{5}{2}, t_H \right) \left( \frac{k_BT}{4\pi D(T)} \right)^{5/2} \right\}
\]  

(3.1a)

with the spin wave stiffness coefficient \( D(T) \) given by

\[
D(T) = D(0)(1 - D_2T^2)
\]  

(3.16)

or

\[
D(T) = D(0)(1 - D_{5/2}T^{5/2})
\]  

(3.1c)
Fig. 3.1 Comparison of the SQUID and VSM magnetization data taken at $H = 10$ kOe.
\[ \Delta m_{SP}(T, H) = A(H) T^2 \quad \text{for weak itinerant ferromagnets} \]  
\[ \Delta m_{SF}(T, H) = A(H) T^2 \quad \text{for strong itinerant ferromagnets} \]  
\[ [M(T, 0)/M(0, 0)]_{SP + LSF}^2 \left[ 1 - Am(T, 0) \right]^2 = 1 - \left( \frac{T}{T_C} \right)^2 \]  
and
\[ [M(T, 0)/M(0, 0)]_{SF}^2 = \left[ 1 - Am(T, 0) \right]^2 = 1 - \left( \frac{T}{T_C} \right)^{4/3} \]  
Since the spin fluctuation theories [5-7], based on the itinerant-electron model, predict that both single-particle excitations and local spin density fluctuations get strongly suppressed in the presence of an external magnetic field, Eqs.(3.3a) and (3.36) can be generalized to include the effect of external field as
\[ [M(T, H)/M(0, H)]_{SP}^2 \left[ 1 - Am(T, H) \right]^2 = C'' - A''(H) T^2 \]  
and
\[ [M(T, H)/M(0, H)]_{SF}^2 \left[ 1 - Am(T, H) \right]^2 = C' - A'(H) T^{4/3} \]  
so that in zero-field Eqs.(3.3c) and (3.3d) reduce to Eqs.(3.3a) and (3.36), respectively. In order to test the validity of the above expressions (i.e., Eqs.(3.1), (3.26), (3.3c) and (3.3c/)), the reduced magnetization \( m(T, H) = M(T, H)/M(0, H) \) and its square are plotted against various powers of \( T \), as shown in figures 3.2(a)-(d) for the \( M(T, H) \) data taken on the amorphous alloys in question using SQUID magnetometer. These plots clearly indicate that the power laws \( m(T, H) \sim 1 - BT^{3/2}, m(T, H) \sim C - AT^2 \) and \( m^2(T, H) \sim C - A'T^{4/3} \) describe very well the observed temperature dependence of \( M(T, H) \) at low temperatures, over intermediate temperatures and for temperatures close to \( T_C \), respectively, for the alloys with \( y = 0 \) and \( x \leq 10 \) with the exception of \( x = 8 \) and \( 10 \) alloys for which the expression \( m^2(T, H) \sim C'' - A''T^2 \) reproduces the \( M(T,H) \) data more closely than the one, \( m(T, H) \sim C - AT^2 \), in the intermediate temperature range. Similar plots constructed for the alloy with \( x = 90 \) (the \( M(T,H) \) data for this alloy were taken in the temperature range from 4.2 K to 300 K at \( H = 9 \) kOe using Faraday balance) and shown in Figs. 3.3(a) and 3.3(b) indicate that the \( T^{3/2} \) law alone seems to describe \( M(T, H) \) data at low temperatures \((T < 175 \text{ K})\), as is the case for other compositions in the alloy series \( \text{Fe}_{90+y} \text{Zr}_{10-y} \) and \( \text{Fe}_{80-z} \text{Co}_z \text{Zr}_{10} \). However, for temperatures above 175 K, this alloy exhibits the same behaviour as that of the alloys with \( x = 8 \) and \( 10 \) in that the expression \( m^2(T, H) \sim C'' - A''T^2 \) fits the \( M(T,H) \) data better than the relation \( m(T, H) \sim C - AT^2 \).

Having roughly identified the temperature ranges over which different types of excitations dominantly contribute to the thermal demagnetization of 'in-field' magnetization for all the
Fig. 3.2 Reduced magnetization versus (a) \((T/T_c)^{3/2}\) and (b) \((T/T_c)^2\). Solid lines denote the least-squares fits to the data.
Fig. 3.2 $[M(T, H)/M(0, H)]^2$ as function of (c) $(T/T_C)^{4/3}$ and (d) $(T/T_C)^2$. Solid lines denote the least squares fits to the data.
Fig. 3.3 (a) $M(T, H)/M(0, H)$ versus $T^{3/2}$ and $T^2$, (b) $[M(T, H)/M(0, H)]^2$ versus $T^{4/3}$ and $T^2$ for a $Co_{90}Zr_{10}$ at $H = 0$ and 9 kOe. Solid lines denote theoretical fits to the data.
alloys under investigation, a detailed range-of-fit analysis of the magnetization data based on
the expressions predicted by the existing theoretical models is performed to find out their relative
importance in different temperature ranges. The details of this range-of-fit analysis are given
below.

Using the value of demagnetizing factor N, obtained by the method described in chapter 2,
and the estimates of splitting factor g (~ 2.07 ± 0.02) and anisotropy field, \( H_A \), deduced from
the FMR measurements [8-10], theoretical fits to \( Am(T, H) \) data have been attempted based on
the expression

\[
\Delta m(T, H) = \Delta m_{SW}(T, H) + \Delta m_{SP}(T, H)
\]  

(3.4)

(where \( \Delta m_{SW}(T, H) \) and \( \Delta m_{SP}(T, H) \) are given by Eqs.(3.1a) and (3.2)) with \( D(T) \) given by
either Eq.(3.1b) or Eq.(3.1c). A nonlinear least-squares (LS) fit computer program, developed
for this purpose, was employed to arrive at the best LS fits. Such fits involve all the possible
combinations based on Eqs.(3.1)-(3.2), i.e., the cases (i) \( \beta = \alpha = A = 0 \) or (ii) \( \beta \neq 0, A = \alpha = 0 \)
or (iii) \( \beta = A = 0, A \neq 0 \) or (iv) \( \beta = A = 0, A \neq 0 \) or (v) \( \Delta m_{SW} - A - 0, A \neq 0 \) or (vi)
\( \Delta m_{SW} = A = 0, A \neq 0 \) corresponding to the fits based on (i) the \( T^{3/2} \) term alone or (ii) the
\( T^{3/2} + T^{5/2} \) terms or (iii) the \( T^{3/2} + T^2 \) terms or (iv) the \( T^{3/2} + T^{3/2} \exp(-\Delta/k_B T) \) or (v)
Eq.(3.2a) alone or (vi) Eq.(3.2b) alone, respectively. Moreover, in each one of the cases (i)-(iv),
three possible expressions for \( D(T) \), i.e., \( D(T) = D(0) + D(T) = D(0)(1 - D_T^2) \) or \( D(T) = D(0)(1 - D_5/2 T^{5/2}) \), are
considered. In addition to these fits, theoretical fits based on Eq.(3.3c) and Eq.(3.3d) are also attempted. In other words, sixteen different types of fits to \( Am(T, H) \)
data have been attempted in total. In the range-of-fit analysis of the above-mentioned theoretical
fits, the values of free fitting parameters and the quality of fits are monitored continuously as the
temperature interval \( T_{min} < T < T_{max} \) is progressively narrowed down either by keeping \( T_{min} \)
fixed at a given value and lowering \( T_{max} \) in steps of 2-3 K towards \( T_{min} \) or by keeping \( T_{max} \) fixed at a
certain value and raising \( T_{min} \) towards \( T_{max} \). Typical results of this type of analysis when \( T_{max} \)
is varied are shown in Figs. 3.4(a)-(e) for the alloy with \( x = 90 \). These plots are representative
of all other alloy compositions. Similar plots are obtained when \( T_{min} \) is varied. In these figures,
a reduced chi square, \( \chi^2 = \sum (x^2) \) defined as the sum of deviation squares (\( x^2 \)) for the \( M(T, H) \) data in
a given temperature interval divided by the total number of data points (N) in that temperature
interval minus the number of free fitting parameters \( (N_{para}) \), i.e., \( \chi^2 = \chi^2/(N - N_{para}) \). Such a
choice enables one to make an unambiguous assessment of the quality of not only a given type of
fit as a function of \( T_{max} \) or \( T_{min} \) but also of different type of fits in the same temperature interval.
Judging by the value of \( \chi^2 \) and by the stability of fitting parameters against a wide variation of
\( T_{max} \) or \( T_{min} \) (cf. Figs. 3.4(a)-(e)), the following observations can be made. (i) Out of all the fits
attempted, the one, based on the theoretical expression Eq.(3.4), that combines Eq.(3.1a) and
Eq.(3.1b) and sets \( \Delta m_{SP} - \beta \neq 0 \) provides the best fit to \( Am(T, H) \) data for temperatures below
a certain temperature \( T^* \). For temperatures \( T > T^* \), even this combination does not describe
Fig. 3.4 (a)-(d) Variation of free fitting parameters for $a - C_{090} Z_{j10}$ with the upper limit of the range $T_{\text{min}} < T < T_{\text{max}}$ when $T_{\text{min}}$ is fixed at 4.2 K for the LS fits to the $M(T, H = 9kOe$ data based on Eqs.(3.1)-(3.4). (e) Variation of $\chi$ with $T_{\text{min}}$ ($T_{\text{max}}$) when $T_{\text{max}}$ ($T_{\text{min}}$) is kept fixed.
the temperature variation of $\Delta m(T, H)$ properly as indicated by the unphysical increase in $\chi^2_{\text{fit}}$ as in most of the fitting parameters with increasing $T_{\text{max}}$. (ii) For $T < T^*$, regardless of temperature range chosen, addition of single-particle term ($\Delta m_{\text{SP}}$) or higher-order spin wave term ($T^{5/2}$) to the $T^{3/2}$ term does not improve the quality of fit and leaves the parameters of the $T^{3/2}$ fit practically unaltered. (iii) The LS fits to the $\Delta m(T, H)$ data for $T < T^*$ based on theoretical expressions which set $D(T) = D(0)(1 - D_2 T^2)$ give lower $\chi^2_{\text{fit}}$ than those for which $D(T) = D(0)(1 - D_3 T^{5/2})$, whereas the quality of fits based on the expressions that consider $D(T)$ to be temperature-independent, i.e., $D(T) = D(0)$, are much worse compared to the ones that allow for the temperature dependence of $D$. (iv) Eq. (3.26) provides an excellent fit to the data in the intermediate temperature range whereas Eq. (3.2a) does not describe the $Am(T, H)$ data in any temperature range even with unphysical values for the parameters $A$ and $A'$. For the alloy with $x = 90$, the situation in the intermediate temperature range is similar to that of the alloys with $x = 8$ and 10. As seen from the Figs. 3.4(d) and 3.4(e) for the alloy with $x = 90$, the expression $m^2(T, H) C'' - A'' T^2$ yields a slightly lower $\chi^2_{\text{fit}}$ in a larger temperature range of 210 K to 300 K compared to that $m(T, H) C - A T^2$ which fits in the range 220 K to 300 K. (v) For temperatures close to $T_C$, the best description to $M(T, H)$ data is provided by the expression $m^2(T, H) C''' - A' T^{4/3}$ for the alloys with $y = 0$ & 1 and $x < 6$. The analysis of the type described above has also been performed on the $M(T, H)$ data taken at $H = 10$ kOe using VSM in the temperature range from 70 K to $T_C$ on the alloys with $y = 0$ & 1 and $x < 6$ with the same results as those yielded by the SQUID data.

With a view to study in detail the effect of external magnetic field on the single-particle excitations and local spin density fluctuations (LSF), the $M(T, H)$ data at several constant external magnetic field values in steps of 1 kOe in the interval 1.5 kOe < $H$ < 15 kOe are obtained from the M vs. H isotherms measured using VSM in the temperature range from 70 K to $T_C$ on the alloys with $y = 0$ & 1 and $x < 6$. A detailed range-of-fit analysis based on Eqs. (3.2b) and Eq. (3.3d), as described earlier, has been carried out to determine the field dependence of the coefficients $A$ and $A'$ of the $T^2$ and $T^{4/3}$ terms, respectively. The best LS fits for the alloy with $x = 4$ are shown in Figs. 3.5(a) and 3.5(b) as continuous straight lines through the data points at a few representative field values. Such plots for other alloy concentrations are identical to those depicted in Figs. 3.5(a) and 3.5(b). As the applied field is increased, the temperature range over which the $T^2$ term fits broadens slightly and shifts to higher temperatures whereas the range of the $T^{4/3}$ fit seems to shift to lower temperatures. For a given concentration, considerable overlap of temperature ranges of $T^2$ and $T^{4/3}$ fits occurs as the field is increased, so much so that for certain Co concentrations the overlap is nearly complete at fields $H \sim 10$ kOe. However, if the temperature range is narrowed down by lowering $T_{\text{max}}$ towards $T_{\text{min}}$, the $T^2$ term (Eq. (3.2b)) provides a better fit to the data than the $T^{4/3}$ term whereas the reverse is true when the temperature range is narrowed down by raising $T_{\text{min}}$ towards $T_C$.

The final outcome of such an elaborate data analysis can be summarized as follows.
Fig. 3.5 $M(T, H)/M(0, H)$ vs. $(T/T_C)^2$ and $[M(T, H)/M(0, H)]^2$ vs. $(T/T_C)^{4/3}$ plots at a few selected field values. Continuous curves denote LS fits to the data based on Eqs.(3.5b) and (3.3d), respectively. Note that the data denoted by the numbers 2, 3, 4, 5 are shifted up by the amounts 0.05, 0.10, 0.15, 0.20 and 0.08, 0.16, 0.24, 0.32 with respect to data} in (a) and (b) respectivey.
(i) For all the alloys including the one with \( x = 90 \), the best fit to \( \Delta m(T, H) \) data for temperatures \( T < T^* \) is provided by the expression

\[
\Delta m(T, H) = \frac{g \mu_B}{M(0, H)} Z^{\frac{3}{2}} T \left[ \frac{k_B T}{4 \pi D(0)(1 - D_2 T^2)} \right]^{3/2}
\]

with the parameter values and temperature ranges for different compositions given in Table 3.1, which also includes the corresponding values for the VSM data taken at \( H = 10 \) kOe within the square brackets. (ii) For the alloys with \( y = 0 \& 1 \) and \( x < 6 \), \( M(T, H) \) data are best described by Eq.(3.26) or its more general form given by

\[
M(T, H)/M(0, H) = C - A(H) T^2
\]

over the intermediate temperature range \( T^* < T < T'' \) while Eq.(3.3d) yields the best fit to the \( M(T, H) \) data for temperatures close to \( T_c \) in the interval \( V < T < T'' \); the values of different parameters and the temperature ranges for such fits are listed in Table 3.1. These fits are represented by the continuous straight lines in Figs. 3.2(a)-(d). By contrast, for the alloys with \( x = 8, 10 \& 90 \), Eq.(3.3c) reproduces the observed temperature variation of magnetization more closely in the range \( T^1 < T < T^4 \) than Eq.(3.2b) (Figs. 3.2(d), 3.3(a) and 3.3(b)). (iii) The VSM and SQUID data taken at \( H = 10 \) kOe yield identical results (Table 3.1). In this context, it is gratifying to note that even though the VSM data for temperatures below 70 K, which are crucial to an accurate determination of the spin-wave parameters such as \( D(0), D_2 \), etc., are not presently available, the \( T^3/2 \) fits to VSM and SQUID data, based on Eq.(3.5a), give the same (within the uncertainty limits) values for these parameters. (iv) The coefficients \( A \) and \( A' \) of the \( T^2 \) and \( T^4/3 \) terms in Eq.(3.56) and Eq.(3.3d), respectively, decrease with increasing applied magnetic field for a given composition and with increasing Co concentration for a given field value (Figs. 3.6(a) and 3.6(b)). This observation implies that single-particle excitations and LSF get suppressed by both magnetic field and Co substitution. These results are discussed in section 3.2.

3.1.2. Spontaneous Magnetization

**Temperature dependence:**

In order to determine spontaneous magnetization at different temperatures, the \( M \) vs. \( H \) isotherms in external fields up to 15 kOe have been measured by means of VSM on a - \( Fe_{90+y}Zr_{10-y} \) (\( y = 0 \& 1 \)) and a - \( Fe_{90-x}Co_xZr_4 \) (\( x = 0, 1, 2, 4, 6 \)) alloys in steps of 1 K from 70 K to \( T_c \) (4.2-300 K for \( x = 90 \)). Typical plots of \( M^2 \) versus \( H/M \) (Arrott plots) at a few specified temperatures, constructed out of these \( M \) vs. \( H \) isotherms with external field corrected for the demagnetizing field, are displayed in figures 3.7(a)-(c). The customary approach of determining spontaneous magnetization at different \textit{temperatures}, \( M(T, 0) \), from
Table 3.1(a): The spin-wave parameters for a - Fe$_{91}$Zr$_9$, and a - Fe$_{90-x}$Co$_x$Zr$_{10}$ alloys deduced from the SQUID magnetization data at $H = 10$ kOe. The corresponding parameter values obtained from the VSM magnetization data at $H = 10$ kOe are displayed within the square brackets. The numbers in parentheses denote the uncertainty in the least significant figure. *Estimated from the value of coefficient $A''$ in Eq.(3.3a).

<table>
<thead>
<tr>
<th>Alloy/Conc. $x$</th>
<th>$t^<em>$ = $T^</em>/T_C$</th>
<th>M(0,H) (G)</th>
<th>D(0) (meV Å$^2$)</th>
<th>$D_2$ ($10^{-6}$ K$^{-2}$)</th>
<th>$T_C$ (K)</th>
<th>$D(0)/T_C$ (meV Å$^2$/K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe$_{91}$Zr$_9$</td>
<td>0.50</td>
<td>969(25) [930(30)]</td>
<td>29(2) [29(3)]</td>
<td>2.0(10) [1.9(10)]</td>
<td>209.66(5)</td>
<td>0.138(10)</td>
</tr>
<tr>
<td>0</td>
<td>0.50</td>
<td>1052(20) [1015(30)]</td>
<td>31(2) [32(3)]</td>
<td>1.5(10) [1.4(10)]</td>
<td>225.00(5)</td>
<td>0.137(10)</td>
</tr>
<tr>
<td>1</td>
<td>0.45</td>
<td>1120(25) [1100(30)]</td>
<td>35(2) [34(3)]</td>
<td>2.0(5) [2.0(5)]</td>
<td>256.66(5)</td>
<td>0.136(12)</td>
</tr>
<tr>
<td>2</td>
<td>0.45</td>
<td>1165(25) [1150(35)]</td>
<td>38(3) [36(3)]</td>
<td>2.5(5) [2.6(5)]</td>
<td>281.60(5)</td>
<td>0.135(14)</td>
</tr>
<tr>
<td>4</td>
<td>0.45</td>
<td>1305(20) [1280(25)]</td>
<td>45(3) [41(4)]</td>
<td>2.0(5) [2.2(5)]</td>
<td>327.95(5)</td>
<td>0.137(13)</td>
</tr>
<tr>
<td>6</td>
<td>0.40</td>
<td>1315(25) [1290(30)]</td>
<td>52(3) [48(5)]</td>
<td>3.5(10) [3.2(10)]</td>
<td>374.75(5)</td>
<td>0.142(14)</td>
</tr>
<tr>
<td>8</td>
<td>0.40</td>
<td>1343(25)</td>
<td>62(3)</td>
<td>3.5(10)</td>
<td>419.50(10)</td>
<td>0.148(8)</td>
</tr>
<tr>
<td>10</td>
<td>0.40</td>
<td>1360(25)</td>
<td>70(2)</td>
<td>3.0(10)</td>
<td>462.50(10)</td>
<td>0.152(5)</td>
</tr>
<tr>
<td>90</td>
<td>0.10</td>
<td>1034(20)</td>
<td>386(4)</td>
<td>0.3(1)</td>
<td>1600(80)*</td>
<td></td>
</tr>
</tbody>
</table>
Table 3.1(b): Parameter values and temperature ranges for the fit to the SQUID [VSM] $M(T,H)$ data at $H = 10$ kOe based on Eqs.(3.26), (3.3c) and (3.3d) of the text and $t = T/T_C$.

<table>
<thead>
<tr>
<th>Alloy/ Conc. x</th>
<th>fit range $t'' - t^{***}$</th>
<th>$A$ ($10^{-6}$ K$^{-2}$)</th>
<th>fit range $t^\dagger - t^{\dagger\dagger}$</th>
<th>$A''$ ($10^{-6}$ K$^{-2}$)</th>
<th>fit range $t' - t''$</th>
<th>$A'$ ($10^{-4}$ K$^{-4/3}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Fe_{91}Zr_9$</td>
<td>0.58-0.98 [0.65-0.98]</td>
<td>12.0(2) [12.2(2)]</td>
<td></td>
<td>0.53-0.87 [0.66-0.83]</td>
<td></td>
<td>6.60(5) [6.65(5)]</td>
</tr>
<tr>
<td></td>
<td>0.55-0.91 [0.58-0.98]</td>
<td>11.0(3) [11.2(2)]</td>
<td></td>
<td>0.62-0.84 [0.62-0.84]</td>
<td>6.12(8) [6.22(8)]</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.53-0.92 [0.55-0.95]</td>
<td>9.2(1) [9.2(2)]</td>
<td></td>
<td>0.50-0.82 [0.50-0.85]</td>
<td>5.50(5) [5.46(4)]</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.49-0.84 [0.45-0.89]</td>
<td>7.8(3) [7.6(2)]</td>
<td></td>
<td>0.46-0.79 [0.50-0.80]</td>
<td>4.94(8) [4.85(5)]</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.40-0.69 [0.40-0.75]</td>
<td>5.4(2) [5.5(1)]</td>
<td></td>
<td>0.56-0.93 [0.56-0.93]</td>
<td>3.90(5) [3.98(4)]</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.44-0.79 [0.45-0.78]</td>
<td>4.5(2) [4.4(1)]</td>
<td></td>
<td>0.70-0.90 [0.55-0.87]</td>
<td>3.44(4) [3.41(3)]</td>
<td></td>
</tr>
<tr>
<td>8</td>
<td>0.74-0.83</td>
<td></td>
<td></td>
<td></td>
<td>5.45(5)</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>0.53-0.75</td>
<td></td>
<td></td>
<td></td>
<td>4.72(5)</td>
<td></td>
</tr>
<tr>
<td>90</td>
<td>0.14-0.20</td>
<td>0.39(1)</td>
<td></td>
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<td></td>
<td></td>
</tr>
</tbody>
</table>
Fig. 3.6 Concentration dependence of the coefficients (a) $A$ in Eq.(3.2b) or (3.5b) and (b) $A'$ in Eq.(3.3d) for a few selected values of $H$. 

\begin{align*}
\text{(a)}
\end{align*}

\begin{align*}
\text{(b)}
\end{align*}
Fig. 3.7 (a) Arrott plot isotherms for \( a\text{-}Fe_{91}Zr_{9} \) at a few selected temperatures.
Fig. 3.7 $M^2$ vs. $H/M$ isotherms at a few representative temperatures for (b) a $Fe_{90}Zr_{10}$ and (c) a $Co_{90}Zr_{10}$. 
the intercepts on the ordinate that the linear extrapolation of the high-field portions of \( M^2 \) vs. \( H/M \) isotherms to \( H = 0 \) yields has not been adopted in the present case (except for the alloy with \( z = 90 \)) for the following reason. Slight but finite curvature even in the high-field portion of the \( M^2 \) vs. \( H/M \) isotherms makes an accurate determination of \( M(T,0) \) impossible because such an extrapolation cannot be carried out unambiguously. Since such a problem has not been encountered in the case of \( a\text{-Co}_{90}\text{Zr}_{10} \), this procedure has been followed for that alloy only. For the remaining alloys, this problem has been effectively tackled by constructing the modified Arrott plot (MAP) \( (M^{1/\beta} \text{ vs. } (H/M)^{\gamma} \) plot) with the choice of spontaneous magnetization and initial susceptibility critical exponents \( \beta \) and \( \gamma \), respectively, that makes MAP isotherms a set of parallel straight lines in the critical region. This procedure is discussed in detail in Chapter 4, which deals with the thermal critical behaviour of these alloys. The modified Arrott plot for \( a\text{-Fe}_{81}\text{Zr}_{9} \), which is representative of all the remaining alloy compositions in the above-mentioned series, is shown in Fig. 4.10 of Chapter 4. The values of spontaneous magnetization at different temperatures are then computed from the intercepts on the ordinate (\( M^1 \) axis) obtained by extrapolating high-field linear portions of the MAP isotherms to \( H = 0 \). The spontaneous magnetization, \( M(T,0) \), data, so obtained, for the alloys with \( y = 0, 1 \) and \( x < 6 \) are shown in Fig. 3.8. The \( M(T,0) \) data have been analyzed in exactly the same way as the 'in-field' magnetization data. The data presented in figures 3.9(a) and 3.9(b) clearly demonstrate that the functional dependence of \( M(T,0) \) on \( T \) is adequately described by Eq.(3.56) or Eq.(3.2b) with \( M(T,H)/M(0,H) \) placed by \( M(T,0)/M(0,0), (\equiv m(T,0)) \) in the intermediate temperature range and by Eq.(3.36) for temperatures close to \( T_C \). The departure of the \( m(T,0) \) data from fits based on Eq.(3.56) on the low-temperature side is mainly caused by spin wave excitations which dominate at low temperatures. For the alloy with \( x = 90 \), \( M(T,0) \) data almost coincides with \( M(T,H) \) data (cf. Figs. 3.3(a) and 3.3(b)) and hence the functional dependence of \( M(T,0) \) on \( T \) is exactly the same as that of \( M(T,H) \). The range-of-fit analysis of \( M(T,0) \) data (for \( x = 90 \)), based on Eqs.(3.3a) and suitably modified versions of Eqs.(3.1), (3.2) and (3.5), carried out in the same way as described for the \( M(T,H) \) data, reveals that the best fits to the \( Af(T,0) \) data for \( T < T^* \) are provided by Eq.(3.5a) with the quantities \( Z(3/2, t_H) \) and \( M(0, H) \) in this equation replaced by \( \xi(3/2) \) and \( M(0,0) \), and with the parameter values given in Table 3.2 while Eq.(3.3a) reproduces the observed temperature variation of \( M(T,0) \) over the intermediate temperature range \( (T^* < T < T^{***}) \) more closely than the modified zero-field version of Eq.(3.5b) does. The temperature ranges and values of free fitting parameters for the above-mentioned alloys are given in Table 3.2 and the best LS fits are represented by solid straight lines through data points in Figs. 3.9(a) and 3.9(b).

*The values at 0 K:*

The magnetization, \( M(T,H) \), data taken at \( T = 5 \) K in applied fields up to 70 kOe on
Table 3.2: The parameter values and temperature ranges for the fits to the \( M(H=0,T) \) data based on Eqs.(3.1)-(3.3) of the text for the \( a - Fe_{91}Zr_9 \) and \( a - Fe_{90-x}Co_xZr_9 \) alloys. The values within the square brackets are the \( M(0,0) \) values obtained directly from \( M \) vs. \( H \) isotherm at \( T = 5 \) K. The numbers in parentheses denote the uncertainty in the least significant figure. *Extrapolated values of the coefficient \( A''(0) \) in Eq.(3.3c) obtained from \( A''(H) \) at \( H = 10 \) kOe. **Observed value of \( A''(0) \).

<table>
<thead>
<tr>
<th>Alloy/Conc. x</th>
<th>( t^* )</th>
<th>( M(0,0) ) (G)</th>
<th>D(0) (meV Å²)</th>
<th>( D_2 ) ( \times 10^{-6} ) K⁻²</th>
<th>fit range ( t^{<strong>} - t^{</strong>*} )</th>
<th>( t'' ) ( \times 10^{-6} ) K⁻²</th>
<th>fit range ( t'-t'' )</th>
<th>( A' ) ( \times 10^{-4} ) K⁻⁴/³</th>
</tr>
</thead>
<tbody>
<tr>
<td>( Fe_{91}Zr_9 )</td>
<td>0.50</td>
<td>945(35) [965(30)]</td>
<td>28(3)</td>
<td>1.8(10)</td>
<td>0.53-0.77</td>
<td>15.4(2)</td>
<td>0.74-0.95</td>
<td>7.85(5)</td>
</tr>
<tr>
<td>0</td>
<td>0.50</td>
<td>1026(30) [1041(30)]</td>
<td>32(3)</td>
<td>1.7(10)</td>
<td>0.51-0.78</td>
<td>13.5(4)</td>
<td>0.67-0.97</td>
<td>7.13(8)</td>
</tr>
<tr>
<td>1</td>
<td>0.45</td>
<td>1075(35) [1100(30)]</td>
<td>34(3)</td>
<td>2.2(5)</td>
<td>0.44-0.71</td>
<td>9.7(1)</td>
<td>0.74-0.94</td>
<td>5.99(5)</td>
</tr>
<tr>
<td>2</td>
<td>0.45</td>
<td>1145(35) [1170(30)]</td>
<td>37(3)</td>
<td>2.5(5)</td>
<td>0.45-0.69</td>
<td>8.4(2)</td>
<td>0.67-0.95</td>
<td>5.30(8)</td>
</tr>
<tr>
<td>4</td>
<td>0.45</td>
<td>1275(35) [1296(30)]</td>
<td>44(4)</td>
<td>2.2(5)</td>
<td>0.44-0.73</td>
<td>6.1(1)</td>
<td>0.64-0.93</td>
<td>4.31(5)</td>
</tr>
<tr>
<td>6</td>
<td>0.40</td>
<td>1290(30) [1305(30)]</td>
<td>51(5)</td>
<td>3.0(10)</td>
<td>0.45-0.75</td>
<td>4.8(2)</td>
<td>0.64-0.96</td>
<td>3.63(4)</td>
</tr>
<tr>
<td>8</td>
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<td>1344(30)</td>
<td></td>
<td></td>
<td>{7.6(2)}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>10</td>
<td></td>
<td>1359(30)</td>
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<td></td>
<td>{6.3(2)}</td>
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</tr>
<tr>
<td>90</td>
<td>0.10</td>
<td>1031(20)</td>
<td>386(4)</td>
<td>0.3(1)</td>
<td>{0.20(1)}</td>
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</tr>
</tbody>
</table>
Fig. 3.8 Reduced spontaneous magnetization versus reduced temperature.
Fig. 3.9 $M(T, 0)/M(0, 0)$ vs. $(T/T_C)^2$ and $[M(T, 0)/M(0, 0)]^2$ vs. $(T/T_C)^{4/3}$ plots constructed using VSM M(T,0) data. Note that the data for $x = 0, 1, 2, 4, 6$ are shifted up by the amounts 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 of $\alpha - Fe_{91}Zr_9$ in (a) and (b), respectively. Solid lines denote LS fits to the data based on Eqs.(3.5b) and (3.3d), in (a) and (b), respectively.
$a$ - Fe$_{90-x}$Co$_x$Zr$_9$ and $a$ - Fe$_{90-x}$Co$_x$Zr$_{10}$ (0 < $x$ < 10) alloys using SQUID magnetometer are plotted in the form of $M(0, H)/M(0, 0)$ vs. $H$ in figure 3.10. Note that no distinction has been made between the value of magnetization at 5 K and 0 K in this figure and in the figures 3.9(a) and 3.9(b). The differential susceptibility, $\chi(0, H)$, as a function of field, obtained by numerical differentiation of these $M$ vs. $H$ curves, is shown in Fig. 3.11. A close examination of these figures reveals that for the alloys with zero or low Co concentration, magnetization does not saturate even in fields as high as 70 kOe and that $\chi(0, H)$ gradually decreases with increasing $H$ and approaches a constant value at high fields. This limiting value is nothing but the high-field susceptibility, $\chi_{hf}$. The inset of Fig. 3.11 shows $\chi_{hf}$ plotted against Co concentration, $x$. As the Co concentration is increased, a plateau in the $\chi(0, H)$ vs. $H$ curve is reached at a lower field and correspondingly the field at which technical saturation in magnetization occurs also shifts to lower values (inset of Fig. 3.10) while $\chi_{hf}$ decreases rapidly for $x$ < 4 and the decreasing trend slows down considerably for $x$ > 6 (inset of Fig. 3.11). The value of spontaneous magnetization at 0 K, $M(0, 0)$, is then obtained by subtracting $\chi_{hf} H$ from $M(T, H)$ at $H$ = 70 kOe. The values of $M(0, 0)$ and $\chi_{hf}$, so obtained, are listed in Tables 3.2 and 3.3, respectively.

3.2. Discussion

Before applying the theoretical models briefly introduced in Chapter 1 to the systems under consideration, relevant details about the itinerant-electron models that bring out clearly the role of spin density fluctuations and Stoner single-particle excitations in determining the magnetic properties of weak itinerant ferromagnets are given below. The main reason for giving these details is to explore the possibility of attempting a quantitative comparison between experiment and theory.

3.2.1. Spin fluctuation model: A unified approach

In this model, it is assumed that following the Ginzburg-Landau formalism, the local free energy density can be expanded in terms of a small and slowly varying classical order parameter (local magnetization) $M + \vec{m}(r)$ as $[5]$

$$f(r) = f_0 + \frac{a}{2} |\vec{M} + \vec{m}(r)|^2 + \frac{b}{4} |\vec{M} + \vec{m}(r)|^4 + \frac{c}{2} \sum_\nu |\nabla m_\nu(r)|^2 + \cdots$$

where

$$\vec{m}(r) = \frac{1}{\sqrt{q}} \sum_q \vec{m}(q) \exp(i \vec{q} \cdot \vec{r}), \quad \vec{m}(q) = \begin{cases} \vec{m}(q) & q < q_c \\ 0 & \text{otherwise} \end{cases}$$
Table 3.3: Band and exchange parameters. \(^a\) Ref.[40,41].

<table>
<thead>
<tr>
<th>Alloy/Conc. (x)</th>
<th>(\mu_0) ((\mu_B))</th>
<th>(\chi_{hf}) (10(^{-4}))</th>
<th>(\chi(0,0)) (10(^{-4}))</th>
<th>(S)</th>
<th>(\text{IN}(E_F))</th>
<th>(\text{N}(E_F)^a) (eV(^{-1}))</th>
<th>(I) (eV)</th>
<th>(v) [TF] [K]</th>
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</thead>
<tbody>
<tr>
<td>(Fe_9Zr_9)</td>
<td>1.34(2)</td>
<td>12.32(10)</td>
<td>14.00(20)</td>
<td>83(1)</td>
<td>1.012(1)</td>
<td>4.00</td>
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<td>8.86(20)</td>
<td>10.37(20)</td>
<td>66(1)</td>
<td>1.015(1)</td>
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<td>1.52(2)</td>
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<td>7.78(10)</td>
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<td>1.019(1)</td>
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<td>0.29(1)</td>
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<tr>
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<td>1.62(2)</td>
<td>4.47(15)</td>
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<td>1.029(1)</td>
<td>3.30</td>
<td>0.31(1)</td>
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<tr>
<td>4</td>
<td>1.79(2)</td>
<td>2.61(10)</td>
<td>2.97(10)</td>
<td>24(1)</td>
<td>1.042(1)</td>
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<td>1.81(2)</td>
<td>1.38(10)</td>
<td>1.50(10)</td>
<td>13(1)</td>
<td>1.079(4)</td>
<td>2.82</td>
<td>0.38(1)</td>
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</tr>
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<td>1.00(15)</td>
<td>1.34(10)</td>
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<td>1.084(5)</td>
<td>2.70</td>
<td>0.40(1)</td>
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<tr>
<td>10</td>
<td>1.84(2)</td>
<td>0.60(20)</td>
<td>0.64(10)</td>
<td>6(1)</td>
<td>1.170(23)</td>
<td>2.61</td>
<td>0.45(1)</td>
<td></td>
</tr>
<tr>
<td>90</td>
<td>1.52(2)</td>
<td>3.50(10)</td>
<td>3.07(5)</td>
<td>36(1)</td>
<td>1.028(10)</td>
<td>1.94</td>
<td>0.53(1)</td>
<td></td>
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</tbody>
</table>
Fig. 3.10 $M$ vs. $H$ curves taken at $T = 5$ K; for the sake of clarity, such curves for $x = 8$, 6, 4, 2, 1, 0 and $\alpha-\text{Fe}_{91}\text{Zr}_9$ are shifted up by the amounts 0.1375, 0.275, 0.4125, 0.5152, 0.6875, 0.825 and 0.9625 with respect to the one for $x = 10$, respectively.

Fig. 3.11 Differential susceptibility as a function of $H$. Inset shows the variation of $\chi_{hf}$ with $z$. Note that the error bars for $\chi_{hf}$ are smaller than the size of the symbols and smooth curves (dashed curve in the inset) serve as a guide to the eye.
\( f_0 \) is independent of \( M + \vec{m}(\vec{r}) \) and \( \nu \) is the polarization index. The coefficients \( a, b \) and \( c \), in general, depend on the cut-off wavevector \( q_c \) which is itself temperature dependent. The total free energy is obtained from the above expression in a straightforward manner with the result [5,11]

\[
F(M) = F(0) + \frac{1}{2} [a + b(3 < m^2_\parallel > + 2 < m^2_\perp >)] M^2 + \frac{b}{4} M^4
\]

with

\[
F(0) = F_0 + \frac{1}{2} \left( a + c q^2 \right) ( < m^2_\parallel > + 2 < m^2_\perp > ) + \frac{b}{4} \left( < m^2_\parallel >^2 + 4 < m^2_\perp >^2 + 4 < m^2_\perp > < m^2_\parallel > \right)
\]

\[
<m^2_\nu> = 4\hbar \int \frac{d^2q}{(2\pi)^2} \int_0^\infty \frac{d\omega}{2\pi} n(\omega) \text{Im} \chi_\nu(q,\omega)
\]

\[
n(\omega) = [\exp(\hbar\omega/k_BT) - 1]^{-1}
\]

In Eqs. (3.6)-(3.8), \( < m^2_\parallel > \) and \( < m^2_\perp > \) are the thermal variances of the local magnetization parallel (\( || \)) and perpendicular (\( _\perp \)) to the average magnetization \( M \), respectively. \( \nu = \|, \perp, n(\omega) \) is the Bose function and \( \chi_\nu(q,\omega) \) is the dynamical wavevector dependent susceptibility.

The magnetic equation of state, which describes the dependence of \( M \) on \( T \) and \( H \), is obtained from Eq. (3.6) by differentiating with respect to \( M \), i.e.,

\[
\frac{1}{M} \frac{\partial F}{\partial M} = \frac{H}{M} = a + b(3 < m^2_\parallel > + 2 < m^2_\perp >) + b M^2
\]

where \( a \) and \( b \), the Landau coefficients for the Stoner theory, are defined by [3,4,12]

\[
a(T) = -[2\chi(0,0)]^{-1} [1 - (T/T_c^S)^2 - BST^4]
\]

\[
b(T) = [2\chi(0,0) M^2(0,0)]^{-1} [1 + CT^2]
\]

with

\[
\chi(0,0) = N\mu_B^2 N(E_F)(T_F/T_c^S)^2 = N\mu_B^2 N(E_F) S
\]

\[
M^2(0,0) = (N\mu_B\mu_0)^2 = (S\gamma)^{-1}
\]

\[
T_F^2 = (\pi^2k_B^2/6)\nu'
\]

\[
\nu' = \left[ N'(E_F)/N(E_F) \right] - \left[ N''(E_F)/N(E_F) \right]
\]
In above equations, $\chi(0,0)$ and $\mu_o$ are the zero-field differential susceptibility and moment per alloy atom at 0 K, respectively, $S$ is the Stoner enhancement factor, $\lambda$ is a measure of the exchange splitting of bands, $N$ is the number of atoms per unit volume, $N(E_F)$ is the density of single-particle states at the Fermi level $E_F$ and $N'(E_F), N''(E_F)$ and $N'''(E_F)$ are its first, second, third and fourth energy derivatives, and $T_C$ is the Stoner Curie temperature. In the event that no external magnetic field is applied ($H = 0$), Eqs.(3.7)-(3.19) permit calculation of the temperature dependence of spontaneous magnetization for temperatures below $T_C$ as follows. When $H = 0$, Eq.(3.9) reduces to

$$M^2(T, 0) = -\frac{a(T)}{b(T)} - (3 < m_{||}^2 > + 2 < m_{\perp}^2 >).$$

Generally, the coefficient $C$ in Eq.(3.11) is negligibly small so that the term $CT^2$ in Eq.(3.11) can be dropped altogether at all temperatures (even for $T > T_C$) and Eqs.(3.20), (3.10) and (3.11) can be combined to yield

$$M^2(T, 0) = M^2(0, 0)[1 - (T/T_C^S)^2 - BST^4] - (3 < m_{||}^2 > + 2 < m_{\perp}^2 >).$$

The actual functional form of $M(T, 0)$ in different temperature ranges below $T_C$ can be derived from Eq.(3.21) with the aid of Eq.(3.7) provided due consideration is given to the fact that $< m_{\perp}^2 >$ consists of two parts: spin wave (SW) part, which is dominant at low temperatures, and spin fluctuation (SF) part which dominates at intermediate temperatures and for temperatures close to $T_C$. The contributions of the $< m_{\perp}^2 >_{SW}$ and $< m_{\perp}^2 >_{SF}$ parts to thermal demagnetization are obtained from Eq.(3.7) by inserting the following expressions [5] for $\text{Im} \chi_{\nu}(q, \omega)$ in this equation and then evaluating the integrals.

$$[\text{Im} \chi_{\perp}(q, \omega)]_{SW} = \frac{\pi}{2} \omega \chi_{\perp}(q) [\delta(\omega - \omega(q)) + \delta(\omega + \omega(q))].$$

(3.22)
respectively, in the random phase approximation (RPA) at low \(q\) and for a cubic lattice. In Eqs. (3.24) and (3.25), \(\chi_{v}(\mathbf{q}, \omega = 0)\) is a component of the static susceptibility, \(\chi_{v} = \chi_{v}(\mathbf{q}, \omega = 0)\), \(g\) is the spectroscopic splitting factor while the parameters \(\tau' = (4/\pi)S^{-1}\chi(0,0)v_{F} + \cdots \) (to zeroth order in \(<m^{2}>\), where \(v_{F}\) is the Fermi velocity) and \(\alpha\) (the coefficient of the gradient term in the Ginzburg-Landau expansion) are independent of \(v\) for small \(M\).

At low temperatures \((0 \sim T \sim 0.37c)\), \(T < T_{C}^{S}\) and the term \(B S T^4\) in Eq.(3.21) is negligibly small so that Eq.(3.21) can be approximated by

\[
M(T, 0) \approx M(0, 0) \left[ 1 - \frac{<m_{z}^2>}{M^2(0,0)} - \frac{3}{2} \frac{<m_{z}^2>}{M^2(0,0)} \right]
\]

Thermal demagnetization of \(M(T, 0)\) in elemental (homogeneous) crystalline ferromagnets such as \(Fe, Co, Ni\) at low temperatures is mainly due to spin wave excitations (transverse spin fluctuations), i.e., the second term in Eq.(3.26) is exceedingly large compared to the third. The expression for \([Im \chi_{\nu}(\mathbf{q}, \omega)]_{SW}\), Eq.(3.22), when substituted in Eq.(3.7) and the result inserted in Eq.(3.26), yields the well-known Bloch \(T^{3/2}\) power law

\[
\frac{M(T, 0)}{M(0, 0)} = 1 - \frac{g \mu_B}{M(0, 0)} \xi(3/2) \left[ \frac{k_B T}{4\pi D(T)} \right]^{3/2}
\]

Considering that \(\chi_{\nu}^{-1} = \partial H_{\nu} / \partial M = H / M (x_{\nu}^{-1} = \partial H_{\nu} / \partial M) = dH/dM\), Eq.(3.24) relates the spin wave stiffness coefficient, \(D(T)\), with the spontaneous magnetization, \(M(T, 0)\), through the expression \(D(T) = g \mu_B \epsilon_{\nu} M(T, 0)\), note that in the presence of the external magnetic field, \(H\), the same equation yields the spin-wave energy gap equal to \(g \mu_B M (\chi_{\nu}^{-1} = g \mu_B H)\). In the case of crystalline or amorphous ferromagnets with competing interactions and/or Invar systems, the contribution to \(M(T, 0)\) arising from longitudinal spin fluctuations, i.e., from the third term in Eq.(3.26), cannot be ignored even at low temperatures since the orientation of a given magnetic
moment in such systems is, in general, not parallel to the direction of bulk magnetization. As a consequence, the displacements of the longitudinal component of magnetization from the local equilibrium value are of the same order of magnitude as the transverse displacements which give rise to spin waves. Thus, the diffusive modes ("diffusons") associated with the longitudinal component of magnetization (which are of hydrodynamic origin) contribute to the $T^{3/2}$ decrease of magnetization as significantly as the transverse fluctuations (spin waves) do. In sharp contrast with spin waves (undamped modes), diffusons represent overdamped modes that are described by Eq.(3.7) with $\text{Im} \chi_{\parallel}(\vec{q}, \omega)$ given by the version of Eq.(3.23) in which $\Gamma_{\parallel}(\vec{q}) = D_i q^2$ [13] and $D_i$ is the diffusion constant. The contribution due to diffusons to the decline of $M(T, 0)$ with $T$, computed from Eq.(3.7) and the modified version of Eq.(3.23), is given by [13]

$$<m_{||}^2> = \frac{A}{2} \xi(3/2) \left( \frac{k_B T}{2\pi D_i} \right)^{3/2}$$

(3.28)

where $A$ is a constant. According to Eqs.(3.26)-(3.28), thermal demagnetization of spontaneous magnetization is faster in spin systems in which diffusons do contribute, besides spin waves, to the $T^{3/2}$ dependence of $M(T, 0)$. However, unlike magnons, diffusons show up as a broad central (elastic) peak in the (inelastic) neutron scattering intensity versus neutron energy isotherms taken at constant values of $q$ [13]. An immediate consequence of this prediction is that the value of spin wave stiffness coefficient deduced from the magnetization data, $D_M$, should be substantially lower [13] than that measured in the inelastic neutron scattering experiments, $D_N$, in such systems. Such a discrepancy between the values of $D$, i.e., $D_N \gg D_M$, has indeed been found in a number of Invar systems.

In the intermediate range of temperatures (typically, $0.4T_C \sim T \sim 0.8T_C$), spin-wave contribution to $M(T, 0)$ is completely swamped by the spin fluctuation contribution, which, in this temperature range, varies with temperature as [5]

$$\frac{<m_{||}^2>_{SF}}{M^2(0, 0)} = \frac{3<m_{||}^2> + 2<m_1^2>}{M^2(0, 0)} = \left( \frac{T}{T_0} \right)^2$$

(3.29)

The $T^2$ dependence of $<m_{||}^2>_{SF}$ over a wide range of intermediate temperatures is basically dictated by the dominant longitudinal spin fluctuation contribution. Combining Eqs.(3.21) and (3.29), one obtains

$$\left[ \frac{M(T, 0)}{M(0, 0)} \right]^2 = 1 - \left[ \left( \frac{1}{T_C^8} \right)^2 + \left( \frac{1}{T_0^8} \right)^2 \right] T^2 - BST^4 = 1 - \left( \frac{T}{T_C^8} \right)^2 + B'T^4$$

(3.30)

where $(1/T_C^8)^2 = (1/T_C^8)^2$ and $B' = -B$. The last term in Eq.(3.30) is normally too small to merit consideration because the coefficient $B$ usually has a value close to zero (note that depending upon the structure of the density of states curve and hence on the relative
magnitude of various terms in Eq.(3.18), $B$ can be either positive or negative or zero) but it can be significant for systems in which the Stoner enhancement factor $5$ has an extremely large value, i.e., for very weak itinerant ferromagnets in which $\Delta N(E_F)\rightarrow 1$ and hence $5 \rightarrow \infty$. From the coefficient of the $T^2$ term in Eq.(3.30), it is evident that local spin density fluctuations (Stoner single-particle excitations) dominantly contribute to $M(T,0)$ if $T_0 \ll T_C^S (T_C^S < T_0)$. However, if $T_0 \approx T_C^S$, the contributions due to LSF and SP excitations are comparable in magnitude.

For temperature close to $T_C (0.87c \sim T \sim 0.957c)$, $\chi^{-1}_1$ is essentially approximated by $\chi^{-1}_l$ and the longitudinal and transverse fluctuations are thus treated on the same footing with the result that the temperature variation of LSF contribution in this temperature region is accurately given by [5]

$$\frac{<m^2>_SF}{M_2(0,0)} = \left(\frac{3 <m^2>_l + 2 <m^2>_T}{M_2(0,0)}\right)^{4/3}$$

(3.31)

where $T_1 = 2.387\, D(0)\sqrt{M(0,0)}(h\gamma')^{1/4}/g\mu_B$ where the $T^{4/3}$ dependence of $<m^2>_SF$ in the above-mentioned temperature range is basically due to transverse spin fluctuations [7]; longitudinal fluctuations lead to a change in the magnitude of $T_C$ but do not affect the $T^{4/3}$ dependence. Moreover, at such temperatures, the spin-splitting of bands is approaching zero, $N(E_F)$ and its derivatives are undergoing substantial changes especially for weak itinerant ferromagnets (for which the $T^4$ term in Eq.(3.21) is significant at intermediate temperatures) and consequently, the coefficient $B$ assumes a considerably reduced value compared to that in the intermediate temperature range. Substituting the result, Eq.(3.31), for the SF term and completely dropping the $T^4$ term in Eq.(3.21) gives

$$\left[\frac{M(T,0)}{M(0,0)}\right]^2 = 1 - \left(\frac{T}{T_C^S}\right)^2 - \left(\frac{T}{T_1}\right)^{4/3}.$$  

(3.32)

The Curie temperature $T_C$ can be determined from Eq.(3.32) by the condition $M(T,0) = 0$ at $T = T_C$ provided the values of $T_C^S$ and $T_1$ are known. Alternatively, at $T = T_C$, Eq.(3.32) reduces to

$$1 - \left(\frac{T_C}{T_C^S}\right)^2 - \left(\frac{T_C}{T_1}\right)^{4/3} = 0.$$  

(3.33)

From Eq.(3.33), it follows that $T_C = T_C^S$ if $T_C^S < T_1$ and $T_C = T_1$ if $T_1 < T_C^S$. In these two limits, single-particle excitations and enhanced local spin-density fluctuations are respectively predominant. Specifically in the latter limit, Eq.(3.32) assumes the form (cf. Eq.(3.36) and Eq.(1.22) of Chapter 1)

$$\left[\frac{M(T,0)}{M(0,0)}\right]^2 = 1 - \left(\frac{T}{T_C}\right)^{4/3}$$

(3.34)
In the paramagnetic regime \((T > T_c)\), \(\chi_{\perp}^{-1} = \chi_{\parallel}^{-1} = \chi^{-1}\) and as such \(<m^2> = <m^2>^x = \frac{1}{3}\), and for reasons stated above, the terms \(T^4\) in Eq.(3.10) and \(T^2\) in Eq.(3.11) can be completely ignored. With this input, the magnetic equation of state, Eq.(3.9), takes the form

\[
\chi^{-1}(T) = a(T) + \frac{5b}{3} <m^2> + b, M^2(T, H) \tag{3.35}
\]

with

\[
a(T) = -[2\chi(0, 0)]^{-1}[1 - (T/T_C^S)^2] \tag{3.36a}
\]

and

\[
b = [2\chi(0, 0)M^2(0, 0)]^{-1} \tag{3.36b}
\]

Mohn and Wohlfarth [14] have calculated the temperature dependence of \(<m^2>\) by approximating the fluctuating moment by a three-dimensional harmonic oscillator whose maximum amplitude equals the saturation moment squared. For temperatures above \(T_C\) but well below \(T_C^S\), i.e., \(T \sim T_C\), such calculations yield [14]

\[
<m^2> = 6 k_B \chi(0, 0) T \tag{3.37}
\]

Now that the inverse initial susceptibility vanishes at \(T_c\) and so does the spontaneous magnetization \(M(T, 0)\), Eqs.(3.35)-(3.37) permit an estimation of \(T_C\) through the relation

\[
1 - \left(\frac{T_C}{T_S^C}\right)^2 = \frac{T_C}{T_S^C} = 0 \tag{3.38}
\]

where

\[
T_S^C = M^2(0, 0)/1k_B \chi(0, 0) \tag{3.39}
\]

Following this approach, Mohn and Wohlfarth [14] have been able to calculate the Curie temperatures for Fe, Co and Ni as well as for their compounds with \(Y\) with a fair degree of accuracy. In the event that \(T_C^S > T_S^C\), Eq.(3.38) asserts that \(T_S^C = T_C\).

In the following text, the present findings are discussed in terms of the existing theoretical models including the one described above. Since both 'in-field' and 'zero-field' (spontaneous) magnetizations exhibit roughly the same temperature in a given temperature range but completely different temperature variations in the low-temperature region, the intermediate temperature region and the region close to \(T_C\), it is imperative to discuss the results obtained in these regions separately.
3.2.2. Low-temperature or spin-wave region

The main observations of the present study in the low temperature region can be summarized as follows. (i) In \( a\text{-Fe}_{90+y}\text{Zr}_{10-y} (y = 0, 1) \) and \( a\text{-Fe}_{90-x}\text{Co}_x\text{Zr}_{10} \) \((x = 0, 1, 2, 4, 6, 8, 10 \& 90)\) alloys, the spin wave excitations give dominant contribution to the thermal demagnetization of both spontaneous as well as 'in-field' magnetization for temperatures below \( T^* (T^* \sim 175 K \text{ or } 0.1T_C) \) which ranges between 0.57\( T_C \) and 0.47\( T_C \) for the alloys with \( y = 1, 0 < x < 10 \) \( (x = 90) \). The values of \( M(0,H) \) and \( M(0,0) \) obtained from the best \( LS \) fits are in good agreement (within the uncertainty limits) with those actually measured at \( T \sim 5 K \). Such a close agreement particularly in the case of \( M(T,0) \) data is gratifying considering the fact that the spontaneous magnetization for all the alloys (except for \( I = 90 \)) in the present study was determined for \( T > 70 K \) only. (ii) The present results clearly demonstrate that the temperature dependence of spin wave stiffness \( D \) cannot be ignored and \( D(T) \) renormalizes with temperature in accordance with the expression, \( D(T) = D(0)(1- \frac{D^2T}{2}) \), predicted by the itinerant-electron model for all the alloys under investigation. (iii) The spin wave stiffness at \( 0K, D(0), \) does not depend on the external magnetic field. This finding refutes the earlier claim \([15,16]\) that \( D(0) \) is field-dependent, (iv) The \( D(0)/(T_C) \) ratio for the alloys with \( y = 0, 1, x < 6 \) possesses a value close to 0.14, which is typical of amorphous alloys with competing interactions, while for the alloys with \( x = 8 \) and 10, \( D(0)/T_C \approx 0.14 \). On the other hand, this ratio for \( a\text{-Co}_{90}\text{Zr}_{10} \) has a value that is typical of the Co-based alloys. (v) It is evident from the concentration dependence of \( T_C, D(0), \) and \( M(0,0) \) shown in Fig. 3.12 that \( T_C \) and \( D(0) \) increase more or less linearly with \( x \) while the steep rise in \( M(0,0) \) observed for \( x \sim 4 \) slows down considerably beyond \( x = 6 \) so much so that \( M(0,0) \) remains nearly unaltered for higher Co concentrations.

The existence of well-defined spin wave excitations at low temperatures (observation (i)) in amorphous alloys under investigation can be understood in terms of both localized \([\text{Eq.(3.1a)}]\) as well as itinerant-electron \([\text{Eq.(3.27)}]\) models. However, the observation (ii) provides evidence for itinerant behaviour in these alloys and indicates that the magnon-single-particle interactions dominate over magnon-magnon interactions. The values of spin wave stiffness \( D(0), \) obtained from spontaneous and in-field magnetization (i.e., \( M(T,0) \) and \( M(T,H) \)) data agree very well with one another. However, at low temperatures, the spontaneous magnetization data yield a lower value of \( D(0) \) than that obtained from the \( M(T,H) \) data for the alloys with \( y = 0, 1, x \sim 4 \), as already reported \([17]\) for \( a\text{-Fe}_{90+y}\text{Zr}_{10+y} \) alloys. This discrepancy in the values of \( D(0) \) should not be taken to imply that \( D(0) \) is field-dependent but the reduced value of \( D(0) \) should be viewed as signaling the softening of spin wave modes \([9,17,18]\) in the reentrant state (in which long-range ferromagnetic order coexists with cluster spin glass order) which comes into existence at temperatures \( T < T_{RE}, \) well below \( T_C \), in these alloys \([19,20]\). In the present work, one does not observe the softening of spin wave modes for the alloys with \( y = 0, 1 \) and \( x < 4 \) because the spontaneous magnetization data in the present case are available.
Fig. 3.12 Variation of $T_C$, $D(0)$ and $M(0,0)$ with Co concentration.

Fig. 3.13 $D(0)$ vs. $T_C$ for $\alpha$-Fe$_{91}$Zr$_9$ and $\alpha$-Fe$_{90}$-$x$Co$_x$Zr$_{10}$ alloys. Similar data on other systems available in the literature are also included for comparison.
only for $T > 70$ K, a temperature well above the reentrant temperature, $T_{RE}$. For $x \sim 6$, such a behaviour is not expected since the reentrant behaviour is completely suppressed when $x \sim 4$ \cite{9,21}. Thus, it is not surprising that no such complication occurs in the case of $a$-Co$_{90}$Zr$_{10}$ alloy \cite{22} which behaves like a conventional ferromagnet \cite{19} down to the lowest temperature and for which $M(T, 0)$ data have been taken for temperatures extending from 300 K down to 4.2 K in the present work. In view of the field-independent value of spin wave stiffness, $D(0)$, deduced from the present study, the dependence of $D(0)$ on $H$ reported \cite{15,16} earlier could be an artifact of the analysis which attributes the observed thermal demagnetization to either spin-wave or single-particle contribution alone and neither takes into account the temperature renormalization of $D$ nor corrects for the gap in the spin wave spectrum arising from the applied field. The plot of $D(0)$ vs. $T_C$ for the presently investigated alloys shown in Fig. 3.13 also includes the $D(0)$ data for several 3d transition metal-metalloid amorphous alloys available in the literature. According to the theoretical prediction \cite{23}, based on the Heisenberg model, the $D(0)$ values for amorphous ferromagnetic alloys, when plotted against $T_C$, should fall on a straight line represented by

$$D(0) = D_o + m T_C$$

(3.40)

where $m = 0.144$ meVÅ$^2$K$^{-1}$ and $D_o$ is either finite or zero depending on whether the exchange interactions extend beyond the nearest neighbour (NN) distance or not. It is noticed from the Fig. 3.13 that the $D(0)$ values for $a$-(Fe, M)-B alloys ($M$ - CT, Mn, W) \cite{24-26}, in which competing interactions are known to be present, fall on a straight line with slope $m = 0.144$ meVÅ$^2$K$^{-1}$ and passing through the origin whereas the $D(0)$ values for $a$-(Fe,Ni)-M(M - P, B, Si, Al) alloys \cite{23,27,28} fall on a different straight line parallel to the earlier one but with finite intercept $D_o = 24 \pm 3$ meVÅ$^2$. This implies that the competing interactions present in the former set of amorphous alloys restrict the range of exchange interactions to the nearest neighbours only whereas the direct exchange interactions extend to the next nearest neighbours in the latter set. Following these arguments, the $D(0)$ values for the alloys with $y = 1, 0 \leq x \leq 6$ fall on the straight line with $m = 0.144$ meVÅ$^2$K$^{-1}$ and $D_o = 0$ indicating thereby that the competing interactions present in these alloys confine the direct exchange interactions to nearest neighbours only. With increasing Co concentration, the competing interactions are gradually suppressed and for the alloys with $x > 6$, $D(0)$ values depart from this straight line and approach the line with finite intercept. This observation implies that exchange interactions in these alloys extend beyond the NN distance.

A linear relation between $D(0)$ and $T_C$ of the type $D(0) = m T_C$ is also predicted by the theory, based on the itinerant-electron model, due to Katsuki and Wohlfarth \cite{29}. With the assumption that the Curie temperature $T_C$ is determined by the Stoner single-particle excitations alone, Katsuki and Wohlfarth \cite{29} have derived for weak itinerant ferromagnets the following relation between $D(0)$ and $T_C$.
where $o$ is the nearest-neighbour (NN) distance and $f(n)$ is a function of the number of electrons per atom determined by the band structure. When the effective-mass approximation is used, Eq.(3.41) reduces to [29]

$$D(0) = \frac{k_B}{6 \sqrt{2} k_F^2} T_C$$

(3.42)

where $k_F$ is the Fermi wavevector. The typical value of $k_F = 1.5 \AA^{-1}$ when inserted into Eq.(3.42), yields the value of slope ($m$) as 0.014 meV $\AA^2 K^{-1}$. This slope value is exactly one order of magnitude smaller than the observed one. Such a large discrepancy between theory and experiment is not surprising in view of the fact that the assumptions, namely, (i) $T_C$ is determined by single-particle excitations alone and (ii) the effective-mass approximation, on which the above theory rests, are not valid in the present case. The description of different theoretical models given in Chapter 1 and section 3.2.1 asserts that even for weak itinerant ferromagnets the Stoner theory, which regards the single-particle excitations as the sole cause of thermal demagnetization in such systems, invariably overestimates $T_C$ and that this theory has to be modified to include the effect of local spin-density fluctuations on the thermal demagnetization if a correct estimate of $T_C$ and a proper description of Curie-Weiss behaviour of magnetic susceptibility for $T > T_C$ is sought. Thus, there is a dire necessity of reviewing a relation of the type Eq.(3.41) or (3.42) within the framework of a theory which includes local spin density fluctuations, besides the spin wave and Stoner single-particle excitations, i.e., in the light of the spin fluctuation model described in section 3.2.1.

The results of the present investigation that provide a strong evidence for the existence of well-defined spin wave excitations in the amorphous alloys in question are in direct contradiction with the earlier claim [30], based on inelastic neutron scattering experiments, that no propagating features, indicative of spin waves, are noticed in the constant-$q$ scans at any temperature below $T_C$ in the wavevector transfer range $0.05 \AA^{-1} < q < 0.12 \AA^{-1}$ in $Fe_{90}Zr_{10}$ alloys. In order to resolve this apparent contradiction, recourse is taken to the infinite three-dimensional (3D) ferromagnetic (FM) matrix plus finite FM spin clusters model proposed by Kaul for amorphous ferromagnets [17,18,31-33]. In this phenomenological model, it is postulated that (a) the spin system for $T < T_C$ consists of an infinite 3D FM matrix and finite spin clusters (composed of a set of ferromagnetically coupled spins), which are embedded in, but ‘isolated’ from, the FM matrix by zones of frustrated spins surrounding the finite clusters (Fig. 3.14(a)), (b) a wide distribution in the size of spin clusters exists, and (c) the spin clusters are not completely isolated in that the long-range RKKY interactions provide a weak coupling between the finite clusters and the FM matrix and also between the clusters themselves. The mechanism that can lead to such a spin structure can be understood as follows. In view of the fact that the cooling rate is not uniform throughout the melt during the melt-quenching process, the nearest-neighbour
Fig. 3.14 Schematic depiction of the infinite ferromagnetic matrix plus finite spin clusters model and the spin-wave dispersion relation that it predicts for (a) $T < T_c$ and (b) $T > T_c$. 
(NN) distance between the atoms varies erratically from one portion of the amorphous alloy to the other so much so that the average NN distance is appreciably greater in certain microscopic regions than in the remaining bulk. Considerable mismatch in the NN interatomic spacings is, therefore, expected to occur within the zones that separate these microscopic regions from the bulk. As a consequence of this mismatch, large 'quenched-in' local stresses exist in these zones. For a magnetic alloy system in which the majority of the atoms carry a magnetic moment, the microscopic regions and the bulk can be identified as the finite spin clusters and infinite matrix, respectively. If the average NN distance \( r_{NN} \) between the spins in the matrix just exceeds the critical distance, \( r_c \), at which the exchange integral changes sign in the Bethe-Slater curve, ferromagnetic coupling exists not only between the spins in the infinite matrix but also between those within the finite spin clusters (ferromagnetic coupling being stronger in the finite clusters than in the bulk); whereas the spins contained in the zones surrounding the clusters get 'frustrated' due to both a sizable magnetostrictive coupling (since the amorphous alloys in question exhibit Invar behaviour [34]) between the spins and the 'quenched-in' local stresses and competing interactions between the spins arising due to the fluctuation in the NN distance around \( r_c \) within these zones. Within the framework of this model, the following explanation can be offered for the absence of spin-wave-like features in the inelastic neutron scattering (INS) spectra [30] taken in a certain wavevector transfer range. Though spin waves, whose stiffness is controlled by the strength of exchange interaction between spins in the FM matrix, are excited for all wavevectors in the infinite ferromagnetic matrix at temperatures \( T < T_c \), all of them do not propagate through the matrix unhindered for the following reasons. The spin waves for which \( q \) falls within the range \( q_{c1} < q < q_{c2} \) (Fig. 3.14(a)), where \( q_{c1} \) and \( q_{c2} \) are the caliper dimensions of the smallest (largest) and the largest (smallest) spin cluster in the wave vector (direct) space, get severely damped due to coupling to, and intense scattering from, the finite spin clusters. Therefore, if the INS measurements are performed in the wavevector range \( q_{c1} < q < q_{c2} \), only a broad "diffusive-like" spectrum with no propagating features would be observed at any temperature below \( T_C \). By contrast, constant-\( q \) scans recorded at the wavevector values that lie outside this \( q \) range should exhibit well-defined spin-wave peaks for all temperatures below \( T_C \) but the nature and origin of these spin waves now depend on whether \( q < q_{c1} \) or \( q > q_{c2} \). In the long-wavelength limit (i.e., when \( q < q_{c1} \)), well-defined spin waves can be excited in the FM matrix only and that too at temperatures well below \( T_C \) because of the low energy-cost involved, and such spin waves propagate through the matrix without any significant damping. On the other hand, in the short-wavelength limit (i.e., when \( q > q_{c2} \)) spin waves can be excited in the FM matrix as well as in the finite clusters; in the latter case, either at very high incident neutron energies when the temperature is low or at high temperatures for the range of incident neutron energies conventionally used. However, but in this case (i.e., when \( q > q_{c2} \)) the spin waves in FM matrix are expected to get damped due to strong exchange fluctuations (caused by the fluctuation in the NN distance between spins) as contrasted with the intra-cluster spin waves which should be relatively well-defined because the exchange coupling
between the spins within the clusters is much stronger and has a much narrower distribution. Thus, the INS spectra should consist of reasonably sharp spin wave excitations superposed on very broad "diffusive-like" structure arising from the overdamped FM spin waves. The magnon dispersion relation for all \( q \) and \( T < T_c \), schematically depicted in Fig. 3.14(a), indicates that the slope of ftw, versus \( q^2 \) straight line, i.e., the spin wave stiffness coefficient, \( D \), is larger for \( q > q_{c2} \) than for \( q < q_{c1} \) because the exchange coupling between the spins in the clusters is much stronger than that in the FM matrix. As the temperature is increased through \( T_c \), exchange coupling between spins in the FM matrix weakens, the frustration zones start 'melting' away and the finite clusters grow in size by polarizing spins originally belonging to the frustration zones (as well as some spins of the FM matrix) and interact with one another through individual spins of the matrix. For \( T > T_c \), the long-wavelength spin waves characteristic of the FM matrix are completely absent and well-defined intra-cluster spin waves can be excited only for \( q > q_c \) (Fig. 3.14(b)) but now \( q_c \ll q_{c2} \). In view of the earlier finding [33] that in \( a - \text{Fe}_{0.02} \text{Zr}_{10} \) alloys the average cluster size for \( T \sim T_c \) is \( - 25 \AA \), the range of \( q \) values (\( 0.05 \AA^{-1} < q < 0.12 \AA^{-1} \)) covered in the INS experiments [30] falls well within the range \( q_{c1} < q < q_{c2} \) in figure 3.14(a) and hence no resolvable spin-wave peaks are found in the INS spectra. In view of the foregoing arguments, the INS measurements for \( T < T_c \) need to be extended to \( q \) values low enough (\( q < 0.05 \AA^{-1} \)) to observe well-defined spin waves, characteristic of the 3D FM matrix.

In Co containing alloys, the Fe-Co and Co-Co exchange interactions are ferromagnetic and much stronger than the Fe-Fe exchange coupling with the result that a progressive replacement of Fe by Co in \( a - \text{Fe}_{0.02} \text{Co}_{x} \text{Zr}_{10} \) alloys gradually suppresses the competing interactions in the frustration zones. This, in turn, leads to breaking up of finite spin clusters into smaller ones and merging of some of them with the infinite FM matrix. Hence, as the Co concentration is increased, the number of spins within the FM matrix increases at the expense of those forming finite clusters, finite clusters shrink in size and decrease in number, and cluster size distribution narrows down while the average distance between the FM matrix spins increases and the spins within the FM matrix tend towards a collinear configuration. As a consequence, the exchange coupling increases in strength (hence both \( D(0) \) and \( T_c \) also assume higher values; in conformity with the observation (v) mentioned above) and the spin system becomes more and more homogeneous with increasing Co concentration. This implies that the shaded region (Fig. 3.14(a)) in the magnon dispersion curve between \( q_{c1} \) and \( q_{c2} \) gradually shrinks and shifts to higher \( q \) values with increasing Co concentration so as to finally disappear at some value of \( x \). These arguments assert that, if the inelastic neutron scattering (INS) experiments in the same range of \( q \) values (i.e., \( 0.05 \AA^{-1} < q < 0.12 \AA^{-1} \)) as used earlier [30], are performed on \( a - \text{Fe}_{0.02} \text{Co}_{x} \text{Zr}_{10} \) alloys, the constant-\( q \) scans exhibiting broad diffusive-like structure at zero or even low Co concentrations should gradually acquire the propagating features corresponding to well-defined long wavelength spin wave excitations even in this wavevector transfer range at
higher Co concentrations. Such a behaviour has indeed been observed recently [35] in the INS spectra taken in the above-mentioned $q$ range on the homologous alloy series $a$-$Fe_{60-x}Co_xZr_{10}$. The competing interactions present in the alloys with $x \sim 6$ give rise to slight canting of the spins in the FM matrix at low temperatures, $T \sim T_{RE}$, due to large local random anisotropy fields that come into play when spin clusters freeze in random orientations [17]. The canted spin arrangement, in turn, reduces net exchange coupling between the FM matrix spins, leading to softening of FM spin wave modes, and makes the saturation in magnetization extremely difficult to achieve even at fields as high as 150 kOe [15,16,36] and temperatures as low as 4.2 K. As the Co concentration is increased, the competing interactions (and hence the reentrant behaviour at low temperatures) get progressively suppressed with the result that the canting angle between the spins decreases and the spin arrangement becomes increasingly collinear. This explains the sharp rise in spontaneous magnetization at 0 K, $M_S = M(0,0)$, for compositions $x \sim 4$ and slower increase or even saturation in $M_S(x)$ curve at higher Co concentrations (Fig. 3.11). The above arguments also provide a straightforward interpretation for the observation (Fig. 3.10 and inset of Fig. 3.11) that at low temperatures, saturation in magnetization is achieved at lower fields for compositions $x > 4$ than for $x \sim 4$ and that $\chi H$ decreases rapidly for $x < 6$ but remains essentially constant for $x > 6$.

Another consequence of the non-collinear ground state spin arrangement in amorphous $Fe_{90+y}Zr_{10-y}$ and $a$-$Fe_{60-x}Co_xZr_{10}$ alloys with $y \sim 2$ and $x \sim 4$ is that the diffusive modes associated with the longitudinal component of magnetization ("diffusons") do contribute, besides magnons, to the $T^{3/2}$ decrease of magnetization, Eq.(3.28), and thereby lower spin wave stiffness coefficient, $D_M(0)$, in magnitude compared to the value that $D_N(0)$ possesses when such a contribution (due to diffusons) is totally absent, as happens to be the case in collinear ferromagnets. Considering that diffusons give rise to an elastic peak [13] in the INS spectra, the value of spin wave stiffness determined from such measurements, i.e., $D_N(0)$, should greatly exceed $D_M(0)$ in these systems. Such a disparity between $D_N(0)$ and $D_M(0)$ is expected to reduce continuously with decreasing $y$ or increasing $x$ as the spin orientations tend towards a collinear configuration. This process continues till a value of $x$ is reduced beyond which $D_N(0)$ equals $D_M(0)$ because by then non-collinearity is fully suppressed. This prediction needs to be verified by INS measurements performed at very low $q$ ($q \ll 0.05 \AA^{-1}$) on $a$-$Fe_{90-x}Co_xZr_{10}$ alloys. It should be emphasized at this stage that a progressive suppression of Invar effect and reentrant behaviour with Co substitution is accompanied by an increase of $D_M(0)$ towards $D_N(0)$ and an enhancement of both $D_M(0)$ and $D_N(0)$ such that the difference $D_N(0) - D_M(0)$ diminishes in magnitude as $x$ increases. Yet another interesting aspect of spin waves in weak itinerant ferromagnets is that in the absence of any spin-dependent (spin-orbit or magnetic) impurity scattering process, spin diffusion provides the sole intrinsic mechanism for relaxation of the long-wavelength, low-frequency modes of spin-density fluctuations. This diffusive relaxation causes
a damping of spin waves (proportional to the spin-diffusion constant) which is temperature-independent and varies with $q$ as $q^4$ [37]. Consistent with this theoretical result, recent inelastic neutron scattering (INS) experiments [35] on the spin dynamics of amorphous $Fe_{90-x}Ni_xZr_{10}$ alloys have revealed a temperature-independent spin-wave linewidth which exhibits a $q^4$ or $q^5$ dependence. Temperature-independent spin-wave damping proportional to $q^5$ is also predicted by the random Heisenberg model of localized spins [38,39] in which damping of spin waves arises from the scattering of magnons from fluctuations in the exchange interaction but this damping mechanism yields a value for magnon linewidth which is an order of magnitude smaller than the observed values.

### 3.2.3. Intermediate temperature region

In the intermediate temperature range ($T'' < T < T''''$), the main observations are as follows. (i) The temperature dependence of both 'in-field' and spontaneous magnetization is best described by the appropriate versions of Eqs.(3.56) (with $C \approx 1$) and (3.3c) (with $C'' \sim 1$) or (3.3a) for the alloys with $y = 0, 1$, $x = 0, 1, 2, 4, 6$ and $z = 8, 10, 90$, respectively, (Figs. 3.2(b), 3.2(d), 3.5(a) and 3.9(a)). (ii) In the absence of the external magnetic field, the coefficient of the $T^2$ term in Eq.(3.56) (with $C \sim 1$ and $H = 0$) or Eq.(3.3c)(with $C'' \approx 1$, $H = 0$) decreases with decreasing $y$ or increasing $x$ (Fig. 3.6(a)). (iii) For a given composition, the coefficient of the $T^2$ term, i.e., $A$ in Eq.(3.5b) and $A''$ in Eq.(3.3c), decreases with increasing applied field (Figs. 3.6(a) and 3.15). Note that (a) the field dependence of $A''$ is not shown in figures 3.6(a) and 3.15 since the values of $A''$ at $H = 0$ and $H = 10$ kOe only have been determined in the present work and (b) $H_{\text{eff}}$ in Fig. 3.15 (and those subsequent figures in which it appears) denote the effective field defined as $H_{\text{eff}} = H - 4 \pi N \langle M \rangle$, where $N$ is the demagnetizing factor and $\langle M \rangle$ is the average value of magnetization over the temperature interval ($T'' < T < T''''$) of the fit based on Eq.(3.5b) or Eq.(3.3c). The error bars for $H_{\text{eff}}$ (less than or equal to the symbol size in the above-mentioned figures) are computed taking into account both the uncertainty in $H$ and deviations of the extreme values of $M$ from its average value, i.e., $M(T''')$— $\langle M \rangle$ and $\langle M \rangle$.

Now that the least-squares fits to the $M(T,H)$ or $M(T,0)$ data based on Eqs.(3.56) and (3.3c) yield values for the parameters $C$ and $C''$ that are close to unity and independent of the field $H$ (Tables 3.1 and 3.2), Eq.(3.5b) can be rewritten in the form

$$\left[\frac{M(T,H)}{M(0,H)}\right]^2 = 1 - 2A(H)T^2A^2(H)T^4$$

(3.43)

where $H$ is either finite or zero. Eq.(3.43) obviously reduces to

$$\left[\frac{M(T,H)}{M(0,H)}\right]^2 = 1 - 2A(H)T^2$$

(3.44)
Fig. 3.15 Variation of the coefficient $A(H_{eff})$ with $H_{eff}$ for different Co concentrations and $a$ - $Fe_{91}Zr_{9}$. 

Fig. 3.16 $A(H_{eff}=0)$ vs. $T_{c}^{-2}$ and $A'(H_{eff}=0)$ vs. $T_{c}^{-4/3}$ plots
i.e., Eq.(3.3c) with $C'' m 1$ and $A''(H) = 2A(H)$ or Eq.(3.3a) with

$$T_C^2 = 2A(0),$$

(3.45)

when the $T^4$ term in Eq.(3.43) is negligibly small compared to the $T^2$ term.

In view of Eq.(3.30), the finding that the expressions (3.43) and (3.44) reproduce closely the observed variation of $M(T, 0)$ and $M(T, H)$ with temperature in the alloys with $y \leq 1, 0 \leq z \leq 6$ and $x = 8, 10$ and 90, respectively, (observation (i) mentioned above) implies that the $T^4$ term in Eq.(3.30) makes a significant contribution to $M(T, 0)$ and $M(T, H)$ only for the first set of alloys; presumably due to a sizable value of the Stoner enhancement factor $S$. To verify this, values of $\chi(0, 0)$ and $M(0, 0)$ (and hence $\mu_0$) for each alloy are computed respectively from the slope and intercept (on the ordinate) of the Arrott ($M^2$ vs. $H/M$) plot isotherm at $T = 5$ K, constructed out the $M(T, H)$ data taken at 5 K (Fig. 3.10). From the values of these quantities so obtained and listed in Table 3.3, one can, in principle, calculate various band and exchange parameters using Eqs.(3.12) - (3.19) provided the actual shape of the density of states (DOS) curve for each composition is known. In the absence of any such information, values of $S$ displayed in Table 3.3 are deduced from Eq.(3.12) by making use of the presently determined values of $\chi(0, 0)$ and those of $N(E_F)$ estimated from the coefficient $\gamma = (v^2 k_B^2/3)N(E_F)$ of the electronic specific heat, reported in the literature [40,41] for the glassy alloys in question, after making corrections [41] for the electron-phonon enhancement. Note that the values of $N(E_F)$ for Co concentrations other than $x = 0,4,10$ and 90 are the interpolated values obtained by passing a cubic spline through the data points in the $N(E_F)$ vs. $x$ plot [40] over the concentration range $0 < x < 30$. The data presented in Table 3.3 clearly demonstrates that (a) $S$ indeed possesses large values for $x \sim 6$ and decreases rapidly with increasing $x$ and (b) Stoner parameter $S$ and hence the exchange splitting of bands $AE = [M(0,0)/N\mu_B]$ decreases with $x$ while $N(E_F)$ decreases with $x$ such that the Stoner criterion for ferromagnetism, i.e., $N(E_F) > 1$, is satisfied for all the compositions. A direct consequence of the increase in $AE$ with $x$ is that the excitation of single particles and more so 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 with Co substitution (Fig. 3.6(a)).

Considering the well-known fact that by holding the weak temperature dependence of the thermal density of states (the one-electron density of states times the Fermi function) solely responsible for the $T^2$ decrease of $M(T, 0)$, the Stoner model grossly overestimates $T_C, T_C^2 > T_o$ in Eq.(3.30) and as such $T_o \approx T_C$. Alternatively, local spin-density fluctuations dominantly contribute to the thermal demagnetization of $M(T, 0)$ in the intermediate temperature range. This inference is vindicated by the observation that the values of $T_C^*$ calculated from the relation $T_C^* = [2A(0)]^{-1/2}$ [cf. Eqs.(3.30) and (3.43)] using the presently determined values of $A(0)$ are consistently lower than the actual $T_c$ values (Table 3.4), i.e., $T_C^*/T_o$ ratio ranges between 0.85
Table 3.4: Comparision of the spin fluctuation temperatures with the Curie temperature. *Values obtained from the estimates of $A^x(0)$ given in Table 3.2.

<table>
<thead>
<tr>
<th>conc. $y/x$</th>
<th>$T_c$ (K)</th>
<th>$T'_c$ (K)</th>
<th>$T'_c/T_c$</th>
<th>$r_c$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$V = l$</td>
<td>209.66(5)</td>
<td>180(1)</td>
<td>0.86(1)</td>
<td>213(1)</td>
</tr>
<tr>
<td>$x = y = 0$</td>
<td>225.00(5)</td>
<td>193(3)</td>
<td>0.86(1)</td>
<td>229(2)</td>
</tr>
<tr>
<td>$x = 1$</td>
<td>256.66(5)</td>
<td>227(1)</td>
<td>0.88(1)</td>
<td>261(2)</td>
</tr>
<tr>
<td>$x = 2$</td>
<td>281.60(5)</td>
<td>244(3)</td>
<td>0.87(1)</td>
<td>286(3)</td>
</tr>
<tr>
<td>$x = 4$</td>
<td>327.95(5)</td>
<td>286(2)</td>
<td>0.87(1)</td>
<td>334(3)</td>
</tr>
<tr>
<td>$x = 6$</td>
<td>374.75(5)</td>
<td>323(6)</td>
<td>0.86(2)</td>
<td>380(3)</td>
</tr>
<tr>
<td>$x = 8$</td>
<td>419.50(10)</td>
<td>$[362(5)]^a$</td>
<td>$[0.86(1)]^a$</td>
<td>1.016(5)</td>
</tr>
<tr>
<td>$x = 10$</td>
<td>462.50(10)</td>
<td>$[398(6)]^a$</td>
<td>$[0.86(1)]^b$</td>
<td>1.018(10)</td>
</tr>
<tr>
<td>$x = 90$</td>
<td>$[1600(80)]$</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
and 0.87 for the alloys with \( y = 0, 1 \) and \( x < 6 \). If \( T_C^* \) for the alloys with \( x = 8 \) and 10 is computed from the relation \( T_C^* = [A(0)]^{-1/2} \) (i.e., ignoring the \( T^4 \) term in Eq.(3.30) and comparing Eq.(3.30) with the zero-field \( (H = 0) \) version of Eq.(3.44)), \( T_C^*/T_C \) ratio for these samples too comes out to be 0.86. Since this ratio is nearly constant (~ 0.86) over this composition range, it is not surprising that the coefficient \( A(0) \) scales with \( T_C^* \) (Fig. 3.16) in accordance with Eq.(3.45). The result \( A(0) \propto T_C^* \) together with the finding that \( T_C \) increases with \( x \) (Fig. 3.12) is the manifestation of the fact that the spin fluctuations get progressively suppressed as the Co concentration increases. When Co concentration is increased beyond \( x = 6 \), the contribution to \( M(T, 0) \) arising from spin fluctuations diminishes at a rapid rate with the result that it is reduced to an insignificant level for compositions in the vicinity of \( x = 90 \). In other words, the decline of \( M(T, 0) \) with \( T \) in the intermediate temperature range in \( a-Co_{90}Zr_{10} \) is mainly due to Stoner single-particle excitations. This deduction is further supported by the 'in-field' magnetization data, as shown below. Thus, \( a-Co_{90}Zr_{10} \) represents the extreme situation in which the particle-hole pair excitations are weakly correlated and \( T_C^* \approx T_C \) In such a case, Eq.(3.30) reduces to the expression \( [M(T, 0)/M(0, 0)]^2 = 1 - (T/T_C)^2 \), which forms an adequate description of the observed variation of \( M(T, 0) \) with \( T \) in this alloy (Fig. 3.3(b)).

A close scrutiny of the Co concentration dependence of \( A(H) \) for a few representative values of \( H \) depicted in Fig. 3.6(a) reveals that the \( A(H) \) vs. \( x \) curves for finite fields are similar in shape to the one for \( H = 0 \) but are shifted down by an increasing amount with respect to it for higher field values. For a given concentration, the field dependence of \( A(H_{eff}) \)s clearly brought out by the data presented in figure 3.15. A rapid decline in the magnitude of \( A(H_{eff}) \) with increasing \( H_{eff} \) is a clear indication of the suppression of spin fluctuations with field. The effect of increasing magnetic field in the itinerant-electron picture is to increase the splitting between spin-up and spin-down sub-bands and hence, in analogy with the influence of increasing \( AE \) by Co substitution on spin fluctuations discussed above, field, like \( x \), strongly suppresses local spin-density fluctuations. The theoretical attempts [7,42] made so far to quantify suppression of spin fluctuations by external magnetic field within the framework of the spin fluctuation model cannot be regarded as satisfactory because a large number of adjustable parameters and the unrealistic electron-gas model have been used to achieve quantitative agreement with the experimental \( M(T, H) \) data. Moreover, the same \( M(T, H) \) data on \( Sc_3In \) have found qualitative explanation in terms of a band model [43] which does not take into account the local spin-density fluctuations and differs from the Stoner model in that, in addition to the Stoner exchange interaction parameter \( I \), it has another interaction parameter (a nearest-neighbour ferromagnetic exchange interaction \( J \)) that gives rise to a temperature- and magnetization-dependent band narrowing. However, even this qualitative agreement between the experimental data and the variation predicted by the latter theoretical treatment [43] cannot be taken seriously because this model fails to produce the experimental variations, e.g., the \( T^{4/3} \) dependence of \( M(T, 0) \) and \( T^{5/3} \) dependence of resistivity in certain temperature ranges, which the spin fluctuation model
successfully docs. The limitation of the spin fluctuation model to make specific predictions about the effect of field on spin fluctuations stems from the fact that spin fluctuations do not explicitly depend on the external magnetic field $H$ but, by virtue of their dependence on $M$, indirectly couple to $H$ via magnetization. Another point that merits attention at this stage is that while attempting a quantitative comparison between theory and experiment due consideration has not been given to the observation that different types of excitations are primarily responsible for the decay of magnetization in different temperature ranges. In order to arrive at the exact functional dependence of the coefficient $A$ in Eq. (3.5b) or Eq. (3.43) on $H$, a number of functional forms that could reproduce the observed variation of $A$ with $H$ were tried with the result that the empirical relation

$$A(H_{\text{eff}}) = A(0)[1 - B H_{\text{eff}}^n]$$

(3.46)

with the choice of the parameters $B$ and $n$ given in Fig. 3.17 describes the $A(H)$ data the best, as is evidenced from the quality of fits (continuous curves), based on Eq. (3.46), in figure 3.15. It is also noticed from Fig. 3.15 that the rate at which the coefficient $A$ decreases with $H_{\text{eff}}$ slows down considerably as $x$ increases, e.g., $<dA/dH_{\text{eff}}>_x=6 \simeq (1/7) <dA/dH_{\text{eff}}>_x=0$, where $<>$ denotes the average value in the range $1.5 \text{ kOe} < H_{\text{eff}} < 15 \text{ kOe}$. Considering that in the absence of $H$, progressive substitution of Fe by Co leads to a strong suppression of fluctuations, it is not surprising that the coefficient $A$ is far less sensitive to $H$ for higher Co concentrations than it is for lower Co concentrations. An extreme situation arises when Co concentration approaches $x = 90$ in that the 'zero-field' and 'in-field' magnetization data even for fields as high as 15 kOe coincide with one another at all temperatures below 300 K (Fig. 3.3). This observation implies that spin fluctuations are completely suppressed as this concentration ($Co_{90}Zr_{10}$) is reached and no further suppression is possible with the external field. Thus the Stoner single-particle excitations are solely responsible for thermal demagnetization in this temperature range in $a = Co_{90}Zr_{10}$. Note that the exchange field (proportional to spontaneous magnetization) when $H = 0$ plays the same role as the external field so far as their influence on the spin fluctuations is concerned.

### 3.2.4. Temperatures close to $T_c$

In this temperature range ($T' < T < T''$), the main observations can be summarized as follows. (i) Appropriate versions of Eqs. (3.3d) (with $C' \simeq 1$) and (3.36) yield the best description for the temperature dependence of both 'in-field' and 'zero-field' magnetization in the temperature interval $V < T < T''$ for the alloys with $y = 0, 1$ and $x < 6$ (Figs. 3.2(c), 3.5(b) and 3.9(b)). (ii) When $H = 0$, the coefficient of the $T^4/3$ term in Eq. (3.3d) (with $C \sim 1$ and $H = 0$) or Eq. (3.36) decreases rapidly with decreasing $y$ or increasing $x$ (Fig. 3.6(b)). (iii) For
Fig. 3.17 Fit parameters for the LS fits to the $A(H_{\text{eff}})$ and $A'(H_{\text{eff}})$ data based on Eqs. (3.46) and (3.47), respectively.

Fig. 3.18 Variation of $A'(H_{\text{eff}})$ with $H_{\text{eff}}$ for different Co concentrations and $\alpha = \text{Fe}_{90-\chi}\text{Co}_{\chi}\text{Zr}_{10}$. 
In an attempt to find out the exact functional dependence of the coefficient $A'$ of the $T^{4/3}$ term in Eq. (3.3d) on $H$, several expressions representing different functional forms were fitted to the $A'(H)$ data. Out of these expressions, the one that yielded the best least-squares (LS) fit to the $A'(H)$ data has the form

$$A'(H_{\text{eff}}) = A'(0)[1 - B'H_{\text{eff}}^{n'}]$$

Such LS fits with the choice of parameter $B'$ given in Fig. 3.17 and $n' = 0.50 \pm 0.02$ are represented by continuous curves in Fig. 3.18. An excellent agreement between the best LS fits based on Eq. (3.47) and $A'(H_{\text{eff}})$ data is evident from this figure. The $H^{1/2}$ power law dependence of $A'$ on $H_{\text{eff}}$ for the compositions $y = 0$, 1 and $x < 6$ is clearly brought out by the $A'(H_{\text{eff}})/A'(0)$ versus $H_{\text{eff}}^{1/2}$ plot shown in Fig. 3.19. Note that a similar study on the alloys with $x = 8$, 10 and 90 for temperatures close to $T_C$ was not undertaken for the following reason. These alloys have high Curie temperatures ($T_C > 300$ K) so that an appreciable interference of the data from structural relaxation-induced specious effects is expected at such temperatures. Since the data cannot be unambiguously corrected for such effects, the studies of this type are bound to be inconclusive. While the exponent $n'$ is independent of composition in the range $0 < x < 6$, the slope $B'$ of the $A'(H_{\text{eff}})/A'(0)$. $H_{\text{eff}}^{1/2}$ straight lines (the IS fits) in Fig. 3.19 decreases with $x$ in accordance with the empirical relation $B'(x) = B'(0)[1 - \alpha x^r]$ in which $B'(0) = 1.30 \pm 0.02$, $a = (4.15 \pm 0.05)x 10^{-4}$ and $\beta = 0.25 \pm 0.02$. The continuous curve through the $B'(x)$ data in Fig. 3.17 denotes the LS fit based on the above expression with the choice of parameters just stated. The spin fluctuation model, described in section 3.2.1, provides an adequate theoretical justification for the observed $H_{\text{eff}}^{1/2}$ dependence of $A'(H_{\text{eff}})$ as is evidenced below. In the temperature range under consideration, the Bose function $n(\omega)$ and $\chi_1^{-1}$ can be approximated by $kBT/\hbar\omega$ and $\chi_1^{-1}$, respectively. Eqs. (3.7), (3.8), (3.23) and (3.25), when
Fig. 3.19 $A'(H_{eff})/A'(0)$ vs. $H_{eff}^{1/2}$ plots for different compositions.
combined and solved for $<m^2>$ with these assumptions, yield [15]

$$<m^2>_v = \frac{1}{2\pi^2} g \mu_B M(T, H) \left( \frac{k_B T}{D} \right) q_c \left[ 1 - \frac{\pi}{2q_c} (\frac{g \mu_B}{D})^{1/2} \sqrt{H} \right].$$  \hspace{1cm} (3.48)

where the temperature-dependent cut-off wavevector $q_c$ is given by [5]

$$q_c \simeq (k_B T / h \gamma'_\nu c_v)^{1/3}. \hspace{1cm} (3.49)$$

Substituting for $D$ and $q_c$ in the prefactor, Eq.(3.48) can be cast into the form

$$<m^2>_v = \frac{1}{2\pi^2 h^{1/3} \gamma'_\nu c_v^{1/3}} \left( \frac{k_B}{c_v} \right)^{4/3} T^{4/3} \left[ 1 - \frac{\pi}{2q_c} (\frac{g \mu_B}{D})^{1/2} \sqrt{H} \right].$$  \hspace{1cm} (3.50)

Use of Eq.(3.50) in the following version [45] of the magnetic equation of state, Eq.(3.9),

$$\left[ \frac{M(T, H)}{M(0, H)} \right]^2 = \left[ \frac{M(0, 0)}{M(0, H)} \right]^2 \left[ 1 - \frac{3 <m^2>_v}{M^2(0, 0)} - \frac{2 <m^2>_v}{M^2(0, 0)} \right]$$

results in the expression

$$\left[ \frac{M(T, H)}{M(0, 0)} \right]^2 = 1 - \left( \frac{T}{T_C} \right)^{4/3} \left[ 1 - \frac{\pi}{2q_c} (\frac{g \mu_B}{D})^{1/2} \sqrt{H} \right].$$  \hspace{1cm} (3.52)

Considering that $M(0, H) \sim M(0, 0)$, Eq.(3.52) clearly demonstrates that the spin fluctuation model correctly predicts the $\sqrt{H}$ dependence of $A'(H)$. Using the typical values $q_c \sim 1 \AA^{-1}$, $g \sim 2$ and $D \sim 100$ meV $\AA^2$, the coefficient of the $\sqrt{H}$ term turns out to be $\sim 5 \times 10^{-4}$. This estimate is of the same order of magnitude as the observed values. If for a moment $q_c \sim (- 1 \AA^{-1})$ is taken to be independent of composition and the observed values of stiffness coefficient $D$ and splitting factor $g (- 2.07 \pm 0.02$, value determined from FMR measurements) are inserted into Eq.(3.52), the coefficient $B'$ of the $\sqrt{H}$ term possesses the values $9.6 \times 10^{-4}$ and $8.1 \times 10^{-4}$ for the alloys with $x = 0$ and 6, respectively, as against the observed values $13.0 \times 10^{-4}$ and $6.6 \times 10^{-4}$. From this comparison, one may be tempted to conclude that the spin fluctuation model predicts a much slower decrease of the coefficient $B'$ with $x$ than the observed one. But when it is realized that both the quantities $c_v$ (the coefficient of the gradient term in the Ginzburg-Landau expansion) and $\gamma'_v$, appearing in the expression, Eq.(3.49), of $q_c$, decrease rapidly with $x$ because these alloys become more and more homogeneous magnetically and $N(E_F)$ falls steeply as the Co concentration in them is increased, a much closer agreement between the theoretical and experimental variation of $B'$ with $x$ is expected. For a quantitative comparison between theory and experiment, values of $c_v$ and $\gamma'_v$ for different compositions are needed. Such data are not available at present.
3.3. Conclusions

Based on an elaborate analysis of highly accurate magnetization data taken over a wide range of temperatures and external magnetic fields on amorphous Fe_{90+y}Zr_{10-y} (y = 0, 1) and Fe_{90-x}Co_xZr_{10} (x = 0, 1, 2, 4, 6, 8, 10 and 90) alloys and a detailed discussion of the results, following conclusions can be drawn.

(i) Magnetization at 5 K does not saturate even for fields as high as 70 kOe for the alloys with y = 0, 1 and x ~ 6. The high-field differential susceptibility, \( \chi_{hf}(0) \), is extremely large in the alloys with y = 0, 1 and x = 0, 1 and decreases rapidly with increasing x for x ~ 4 so that it possess values typical of crystalline counterparts for x > 6. Large \( \chi_{hf}(0) \) strongly indicate presence of competing interactions in the alloys with y = 0, 1 and x ~ 6.

(ii) While spin-wave excitations are mainly responsible for thermal demagnetization at low temperatures \( (T \sim 0.37c) \), enhanced fluctuations in the local magnetization give a dominant contribution to \( M(T, 0) \) over a wide range of intermediate temperatures \( (0.47b \sim T \sim 0.87b) \) and for temperatures close to \( T_c \) \( (0.87b \sim T \sim 0.957b) \) in all the alloys except for the one with z = 90. In this alloy, dominant spin-wave contribution to both \( M(T, 0) \) and \( M(T,H) \) at low temperatures \( (T \sim 0.17b) \) is followed by an overwhelming contribution from Stoner single-particle excitations at higher temperatures, implying thereby that the particle-hole pair excitations are very weakly correlated in this case.

(iii) The spin-wave stiffness coefficient \( D \) is independent of the external field for all the compositions while the \( D/T_c \) ratio possesses a value \( \sim 0.14 \) in the alloys with y = 0, 1 and x < 6 which is characteristic of amorphous ferromagnets with competing interactions. In these alloys, competing interactions confine the direct exchange interactions to the nearest-neighbours only. The range of direct exchange interactions extends beyond the first nearest-neighbour distance in the alloys with x > 6.

(iv) The observed temperature renormalization of \( D \) augurs well with the temperature variation predicted by the itinerant-electron model and asserts that the magnon - single-particle interactions are more important in these systems than the magnon-magnon interactions.

(v) For compositions y = 0, 1 and z \( \sim 6 \), the value of \( D \) directly measured by inelastic neutron scattering (INS) experiments, \( D_{N} \), is expected to greatly exceed that deduced from the magnetic measurements, \( D_{M} \). This is so because in these alloys, "diffusons" (longitudinal spin fluctuations) contribute to the \( T^{3/2} \) decrease of magnetization as significantly as the transverse spin fluctuations (spin waves) do but they (diffusons) show up as an elastic peak in the INS spectra.

(vi) The infinite three-dimensional ferromagnetic (FM) matrix plus finite FM clusters model offers a straightforward explanation not only for the absence of spin-wave peaks in the INS
spectra taker in the wavevector transfer range of $0.05 \AA^{-1} < q < 0.12 \AA^{-1}$ but also for the composition dependence of $D(0)$, $T_C$, $M(0,0)$ and $\chi_{hf}(0)$.

(vii) In accordance with the predictions of the spin fluctuation model, spin fluctuations get strongly suppressed by Co concentration and external magnetic field. This model provides a consistent theoretical basis for the observed temperature dependence of spontaneous and 'in-field' magnetization over the entire temperature range $0 \sim T \sim T_C$.

(viii) The Stoner criterion $IN(E_F) > 1$ for the occurrence of ferromagnetism is satisfied. All the alloys studied in this work are weak itinerant ferromagnets.

Before concluding this section, certain issues relating to the application of the spin fluctuation (SF) model to the type of spin systems under consideration need to be addressed. From a puristic point of view, this model is strictly valid for nearly ferromagnetic metals or ferromagnetic metals with unsaturated moments only. Therefore, a direct application of the SF model to a concentrated system with 90 at.% or more 3d transition metal content would seem to be far fetched unless due consideration is given to the fact that the ferromagnetic ground state in $\text{o-Fe}_{100-\text{p}}\text{Zr}_\text{p}$ alloys becomes unstable when $p = p_c \sim 7$ at.% [36,46] and the alloys with $y = 0, 1$ and $x = 0, 1$, in particular, have a composition close to $p = p_c$ where the ferromagnetic instability occurs. Considering that the SF model adequately describes the magnetic behaviour of the alloy systems in question, it is not immediately obvious as to why some of its predictions such as the field dependence of $D$ and the Curie-Weiss behaviour of magnetic susceptibility for $T > T_C$ (chapter 4) are not borne out by the present results. One way to reconcile with this situation is to invoke the possibility that the field dependence of $D$ in the investigated alloys is so small as to escape detection in the measurements that cover a relatively narrow field range ($0 \sim H \sim 15$ kOe). Magnetic measurements over a much wider field range (e.g., $0 \sim H \sim 150$ kOe) should help in clarifying this issue. The deviation from the Curie-Weiss behaviour in the present case is due to giant FM clusters which exist even at temperatures well above $T_C$ and interact with one another through individual spins, as inferred previously from magnetization [17,33], Mossbauer [18,47], ac susceptibility [33,48], ferromagnetic resonance [8-10,22,49] and small-angle neutron scattering [50] studies. The spin fluctuation model, however, fails completely in the critical region because this theory is nothing more than the first fluctuation correction to Landau theory. Therefore, the SF theory represents a first step towards a crossover theory between the Landau region and the critical region. A correct description of the magnetic and thermodynamic behaviour in the critical region is provided by the renormalization group calculations, as is evident from the data presented in the following chapter. For this reason, one full chapter (Chapter 4) is devoted to the critical behaviour of the alloy systems of interest in this work.
B.: \(a-(Fe_pNi_{1-p})_{80}(B,Si)_{20}\) and \(a-(Fe_pNi_{1-p})_{80}P_{14}B_6\) alloys

3.4. Results and Analysis

Fig. 3.20 shows the magnetic phase diagrams for \(a-(Fe_pNi_{1-p})_{80}(B,Si)_{20}\) (series I) and \(a-(Fe_pNi_{1-p})_{80}P_{14}B_6\) (series II) alloys constructed using the values of \(T_C\) (Curie temperature), \(T_{SG}\) (spin glass freezing temperature) and \(T_{RE}\) (the reentrant transition temperature), accurately determined from ac susceptibility, electrical resistivity and bulk magnetization measurements [19,23,31,32,51-56]. Figure 3.20 indicates that the critical concentration \(p = p_c\), at which the ferromagnetic (FM)-paramagnetic (PM), PM-spin glass (SG) and FM-reentrant (RE) phase transition lines meet, is different for the two series; \(p_c \sim 0.03\) for series I and \(p_c \sim 0.10\) for series II. Since the low-lying magnetic excitations in the compositions exhibiting ferromagnetism are of main interest in this chapter, henceforth attention is focussed on the FM phase only.

The magnetization, \(M(T,H)\) data taken in the temperature range from 4.2 K to 300 K (or \(T_C\), which over is lower) at an external magnetic field of \(H = 9\) Koe on \(a-(Fe_pNi_{1-p})_{80}B_{19}Si_{1}\) \((0.0625 < p < 0.20)\), \(a-(Fe_pNi_{1-p})_{80}B_{20}\) \((0.25 < p < 1.0)\) and \(a-(Fe_pNi_{1-p})_{80}P_{14}B_6\) \((0.1125 < p < 1.0)\) alloys using the Faraday technique were analyzed in the same way as in the case of \(a-Fe_{80-x}Co_xZr_{10}\) alloys. In order to identify the temperature ranges over which the functional dependence of \(M(T,H)\) on \(T\) is governed by a dominant contribution arising from one or more types of low-lying magnetic excitations such as spin waves, single-particle excitations and local spin-density fluctuations, the reduced magnetization squared, \([M(T,H)/M(0,H)]^2\), is plotted against \(T^2\) and \(T^2\). Such plots for \(a-Fe_{80}B_{20}\) are shown in Figs. 3.21(a) and 3.21(b), also serve as an illustration of the behaviour observed in the remaining compositions. These figures indicate that for the \(a-Fe_{80}B_{20}\) alloy, the \(M(T,H) = 9\) Koe) data are better described by a \(T^{3/2}\) law for \(T \sim 140\) K and by a \(T^2\) law for \(140 < T < 300\) K while the other power laws (Fig. 3.21(b)) do not fit the data in any temperature range. Having completed such an exercise on the \(M(T,H)\) data of all the compositions in both the alloy series, a detailed 'range-of-fit' analysis of the type described in Sec.3.1.1 has been carried out to ascertain the relative importance of different contributions to \(Am(T,H) = [M(0,H) - M(T,H)]/M(0,H)\). A representative example of the variation of various fitting parameters with the upper limit \(T_{max}\) of the temperature range \(T_{min} < T < T_{max}\) used for different types of fits is shown in figure 3.22. For the least-squares fits to \(M(T,H)\) data based on Eqs.(3.1)-(3.3), use has been made of the values of demagnetizing factor \(N\) determined from low-field magnetization measurements and those of the splitting factor \(g\) as well as anisotropy field \(H_A\) reported in the literature [19,23,57,58]. Such an exhaustive analysis of the magnetization data taken on these alloys is necessitated by the fact that most of the previous determinations [19,23,24,26-28,53,59,78] of the spin wave stiffness, \(D\), do not take into account either the single-particle contribution or the effect of external magnetic field or the temperature renormalization of \(D\) eventhough inelastic neutron scattering (INS) data [79-82] clearly demonstrate that an appreciable reduction
Fig. 3.20 Magnetic Phase diagrams for \( a - (Fe_p Ni_{1-p})_{80}(B, Si)_{20} \) and \( a - (Fe_p Ni_{1-p})_{80}P_{14}B_6 \) alloys.
Fig. 3.21 (a) $M(T, H)/M(0, H) vs. T^{3/2}$ and $T^2$, and (b) $[M(T, t f)/M(0, V f)]^2$ vs. $T^{4/3}$ and $T^2$ plots for $a\text{-Fe}_{80}B_{20}$ at $H = 9$ kOe. Dashed curves represent the LS fits to the data.
Fig. 3.22 (a)-(d) Variation of free fitting parameters with the upper limit ($T_{max}$) of the range $T_{min} < T < T_{max}$ when $T_{min}$ is fixed at 4.2 K for the LS fits to $M(T, H=9kOe)$ data taken on a $Fe_{80}B_{20}$ based on (a) Eqs. (3.1), (3.2) and (3.4).
in $D$ occurs with increasing temperature. The main observations based on the above analysis are as follows. (i) For temperatures $t = (T/T_c) < t^*(p)$, the best fit to $\Delta m(T, H)$ data is provided by Eq.(3.1a) with $\theta = 0$ and $D(T) = D(0)(1 - D_{5/2}T^{5/2})$ for the alloys with $p < 0.625$ and $D(T) = D(0)(1 - D_2T^2)$ for those with $p > 0.75$ in series I. For $t > t^*(p)$, the Stoner single-particle excitations of weak-itinerant (WI) type, i.e., Eq.(3.26), and local spin-density fluctuations (LSF), i.e., Eq.(3.30), are responsible for the thermal demagnetization for all the compositions in series I. In series II, the observed temperature variation of $\Delta m(T, H)$ is closely reproduced by the first spin-wave term in Eq.(3.1a) alone, i.e., by the expression represented by Eq.(3.1a) with $\theta = 0$ and $D(T) = D(0)(1 - D_{5/2}T^{5/2})$ for temperatures $t < 0.9 (T < 300 \text{ K})$ and concentrations $p < 0.25 (0.375 < p < 0.625)$. However, for the alloys with $p > 0.75$, the $T^{3/2}$ spin-wave term in Eq.(3.1a) with $D(T) = D(0)(1 - D_2T^2)$ plus the single-particle contribution of strong-itinerant (SI) type described by Eq.(3.2a) yield the best fit to the $\Delta m(T, H)$ data in the entire temperature range ($0 \sim T < 300 \text{ K}$). (ii) In alloy series I, the single-particle contribution of WI type (Eq.(3.26)) as well as the LSF contribution (Eq.(3.30)), though present at all temperatures, are completely overshadowed by the spin-wave contribution for $t \sim t^*(p)$ but they completely account for the observed temperature dependence of $\Delta m(T, H)$ for $t > t^*(p)$. By comparison, single-particle excitations of the SI type (Eq.(3.2a)) contribute significantly, besides a dominant spin-wave contribution, to $\Delta m(T, H)$ in the entire temperature range $T < 300 \text{ K}$ only for Fe concentrations $p > 0.75$ in alloy series II whereas for $p < 0.625$ in this alloy series, the spin wave contribution is so large as to make a reliable estimation of $\Delta m_{SP}(T, H)$ and $\Delta m_{LSF}(T, H)$ virtually impossible. The latter inference can be drawn from the fact that the addition of single-particle and LSF contributions of the form given by Eq.(3.26) or Eq.(3.30) to the spin-wave term marginally improves the quality of fit for $p < 0.625$ while the addition of single-particle contribution of the SI type, Eq.(3.2a), brings forth a marked improvement in the quality of fit for concentrations $p > 0.75$ compared to that when only the $T^{3/2}$ term is present. (iii) The quality of LS fits based on the theoretical expressions that set $D(T) = D(0)$ is much worse compared to the ones that allow $D(T)$ to vary as $D(T) \sim T^{5/2}$ or $T^2$, indicating thereby that the temperature renormalization of D is important. (iv) The LS fits are able to clearly distinguish between the different functional dependences of $D(T)$ on $T$ and they reveal that $D(T) \sim T^{5/2}$ for $p < 0.625$ whereas $D(T) \sim T^2$ for $p > 0.75$ in both the alloy series.

Figures 3.23 and 3.24 show the reduced magnetization data plotted against $T^{3/2}$ for series I and II, respectively. The continuous curves through the data points represent the best LS fits to the $\Delta m(T, H)$ data with the parameter values for series I and II given in Tables 3.5 and 3.6, respectively. The presently determined parameters, $M(0,0)$, $D(0)$, $T_c$, $D_{5/2}$, $D_2$ and $A$ are plotted as functions of Fe concentration for both the alloy series in figures 3.25 and 3.26 together with the values of these parameters for crystalline $(c-)$Fe$_x$Ni$_{100-x}$ alloys [83,84]. Note that in these figures, $p$, which denotes the Fe concentration in the alloy series I and II, has been changed to $x = 80p$ in order to facilitate a direct comparison between the present results and
Fig. 3.23 Modified Bloch law behaviour of the $M(T,H)$ data taken at $H = 9$ kOe for a few representative compositions in the $a - (Fe_pNi_{1-p})_{80}(BSi)_{20}$ alloy series. The continuous curves through the data points represent the LS fits to the data.
Fig. 3.24 Modified Bloch law behaviour of the $M(T,H)$ data taken at $H = 9$ kOe for a few representative compositions in the $a-(Fe_pNi_{1-p})_{80}P_{14}B_6$ alloy series. The continuous curves through the data points represent the LS fits to the data.
Fig. 3.25 (a) Variation of $T_C$, $D(0)$ and $M(0,0)$ with $x$ in $a - Fe_xNi_{80-x}(B, Si)$ (open squares), $a - Fe_xNi_{80-x}P_{14}B_6$ (open circles) and fcc $Fe_xNi_{100-x}$ (open triangles, data taken from Ref.[83,84]) alloys.

Fig. 3.26 The parameters $A$, $D_2$, $D_{5/2}$ and $t^*$ as function of $x$ for $a - Fe_xNi_{80-x}(B, Si)_{2}$ $a - Fe_xNi_{80-x}P_{14}B_6$ and fcc $Fe_xNi_{100-x}$ (data taken from Ref. [83,84]) alloys.
those previously reported [83,84] for crystalline (c-) Fe$_x$Ni$_{100-x}$ alloys. In addition, the values of mean-square range of exchange interaction, $<r^2>$, for different compositions in series I and II, computed from the relations [19,23,53,85]

$$D_{5/2} = \pi \xi(5/2) \left( \frac{g\mu_B}{M(0,0)} \right) \left( \frac{k_B}{4\pi D(0)} \right)^{5/2} <r^2>$$

(3.53)

and

$$\frac{D(0)}{T_C} = \frac{(S/3) \int r^2 J(r) G(r) dr}{[25(S+1)/3k_B] \int J(r) G(r) dr} = \frac{k_B}{2(S+1)} <r^2>$$

(3.54)

using the presently determined values of $M(0,0)$, $D(0)$, $T_C$ and $D_{5/2}$ and by setting $\xi = 1$, are included in Tables 3.5 and 3.6 for comparison. The observations (i) - (iii) mentioned above assert that the temperature renormalization of $D$ cannot be neglected for the alloys in question and the property $D_N$ (stiffness value determined from inelastic neutron scattering data) $>$ $D_M$ (stiffness deduced from the magnetization data) is inherent to Invar systems (e.g., a - Fe$_{80}B_{20}$ in Table 3.5). This result refutes the earlier claims [77,86] that the wide disparity between the values of $D_M$ and $D_N$ usually found in Fe-based amorphous (and crystalline) Invar alloys completely disappears when contribution to $\Delta m(T, H)$ arising from both spin-wave and single-particle excitations (of either WI type [77] or SI type [86]) are taken into account without considering the temperature renormalization of spin wave stiffness $D$.

3.5. Discussion

In order to facilitate a direct comparison between the present results and those reported on c—Fe$_x$Ni$_{100-x}$ alloys, the Fe concentration in the amorphous alloys in question will be referred to as $x$ (=80$p$) instead of $p$ in this section. Before proceeding with the discussion, the main findings of this work are summarized below.

(i) In a — Fe$_x$Ni$_{80-x}(B,Si)_{20}$ alloys (series I), the spin wave excitations alone seem to account for the observed thermal demagnetization below a certain (reduced) temperature $t''$, which decreases with increasing $x$ (Fig. 3.26), but the existence of single particle (SP) excitations of weak-itinerant (WI) type and local spin-density fluctuations (LSF) cannot be ruled out completely. For $t > t''$, SP excitations of WI type (Eq.(3.2b)) and LSF (Eqs.(3.30) and (3.43)) completely account for the decrease of magnetization with increasing temperature. As in a — Fe$_x$Ni$_{80-x}(B,Si)_{20}$ alloys for $t < t''$, the spin-wave (SW) contribution completely masks the WI-type SP plus the LSF contributions in the temperature range $0 \sim T \sim 0.97b$ ($0 \sim T < 300$ K) for Fe concentrations $x < 20$ ($30 < x < 50$) in the a - Fe$_x$Ni$_{80-x}P_{14}B_{6}$ alloy series (series II); however, SP excitations of strong-itinerant (SI) type (Eq.(3.2a)) contribute significantly
Table 3.5: Splitting factor $g$, density $\rho$, spin-wave and other relevant magnetic parameters, and mean-square range of the exchange interaction $\langle r^2 \rangle$ for $\alpha$-(Fe$_p$Ni$_{1-p}$)$_{60}$(B,Si)$_{20}$ alloys. Numbers in parentheses denote the estimated uncertainty in the least significant figure while those within the square brackets are the values of $\langle r^2 \rangle$ expressed in terms of the average nearest-neighbor distance $\xi = 2.55 \, \text{Å}.$

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*Value of $D(0)$ determined by Brillouin scattering (Ref.[60]).
Table 3.6: Splitting factor $g$, density $\rho$, spin-wave and other relevant magnetic parameters, and mean-square range of the exchange interaction ($r^2$) for α-(Fe$_2$Ni$_{1-x}$)$_x$P$_{11.3}$ alloys. Numbers in parentheses denote the estimated uncertainty in the least significant figure while those within the square brackets are the values of ($r^2$) expressed in terms of the average nearest-neighbor distance $b = 2.55$ Å.

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$^a$Value of $D(0)$ and $D_{1/2}$ evaluated from the Fig. 5 of Ref.[103].
to the thermal demagnetization, in addition to the dominant SW contribution, in the entire temperature range $0 \sim T \sim 300$ K only for $x \sim 60$ at.% in series II. In c - $Fe_xNi_{100-x}$ alloys, a SP contribution of WI type accompanies a dominant SW contribution throughout the temperature range covered in the experiments (note that the SP and SW contributions become comparable only for $x \sim 60$ at.%) as is also the case with $a\cdot (Fe_xNi_{100-x})_{80}(Si)_{20}$ alloys in the entire composition range and $a\cdot Fe_xNi_{100-x}P_{14}B_6$ alloys for $x<50$ at.%. The only difference between the crystalline and amorphous alloys is that in the former case, the SP contribution (i.e., coefficient A in Fig. 3.26) could be estimated because of its larger magnitude whereas in the amorphous case, such an estimation is only possible for all the concentrations in series I at $t>t'$. (ii) In both the amorphous alloy series, $D$ varies with temperature as $D(T) \sim T^{5/2}$ for $x<50$ at.%, in accordance with the expression (Eq.(3.1c)) predicted by the localized-electron (Heisenberg) model, whereas consistent with the behaviour of $D(T)$ (Eq.(3.1b)) predicted by the itinerant-electron model it, renormalizes with temperature as $D(T) \sim T^2$ for $Ft$ concentrations in the range $60<x<80$. A crossover from the $T^{5/2}$ to $T^2$ dependence of spin wave stiffness at $x \sim 60$ at.% in the presently studied amorphous alloys has also been previously observed [83,84] in $c\cdot Fe_xNi_{100-x}$ alloys (Fig. 3.26). The concentration dependence of the coefficient $D_{5/2}$ of the $T^{5/2}$ term in Eq.(3.1r) for the amorphous alloys investigated is similar to that observed in $c\cdot Fe_xNi_{100-x}$ alloys. (iii) The values of $<r^2>$ for different compositions in series I and II computed from Eq.(3.53) (Tables 3.5 and 3.6) suggest that the range of exchange interaction increases rapidly with $x$ whereas Eq.(3.54) predicts that $<r^2>$ decreases with increasing $x$. (iv) The spontaneous magnetization, $M(T=0,x)$, versus $x$ curves in the range $0<x<60$ for series I and II are systematically shifted down by ~20 and ~40 emu/g, respectively, with respect to that for $c\cdot Fe_xNi_{100-x}$ alloys (Fig. 3.25). However, a sudden drop in $M(0,x)$ for $x>60$ at.% in the $c\cdot Fe_xNi_{100-x}$ series is not observed in the investigated amorphous alloys for which $M(0,x)$ increases with $x$ but with a progressively slower rate (Fig. 3.25). (v) The spin-wave stiffness at 0 K, $D(T=0,x)$ and Curie temperature, $T_c(x)$, as functions of $x$ go through a broad peak at $x \sim 60$ at.% ($x \sim 70$ at.%) for $a\cdot Fe_xNi_{100-x}(B,Si)(a\cdot Fe_xNi_{100-x}P_{14}B_6)$ alloys (Fig. 3.25). As $x$ is increased beyond $x \sim 60$ at.% , both $D(0,x)$ and $T_c(x)$ decrease at rate which is very steep in $c\cdot Fe_xNi_{100-x}$ alloys but slows down considerably in the amorphous alloys studied so much so that the drop in these quantities is barely discernible in $a\cdot Fe_xNi_{100-x}P_{14}B_6$ alloys.

Within the framework of the band model, the effect of increasing the $Fe$ concentration $x$ is to increase the exchange splitting of spin-up ($d^*$) and spin-down ($d^*$) d subbands (since $T_c$ increases with $x$, Figs. 3.20 and 3.25) and to shift the Fermi level $E_F$ to higher energies (as the spontaneous magnetization $M(0,x)$ increases with $x$, Fig. 3.25). The observation (i) mentioned above implies that $E_F$ lies within the $d_{\uparrow}$ and $d_{\downarrow}$ subbands in the entire composition range for the alloy series I (weak itinerant ferromagnetism) whereas $E_F$ in series II shifts up with $x$ to such an extent that it lies just above the top of $d_{\downarrow}$ subband at $x \sim 60$ at.%, where a transition from
weak ($x < 60 \text{ at.\%}$) to strong ($x > 60 \text{ at.\%}$) itinerant ferromagnetism occurs. This deduction is consistent with the results of spontaneous resistivity anisotropy [87,88], high-field susceptibility [88], spin-polarized photoemission [89] and Compton scattering [90] experiments on the same or similar Fe — Ni amorphous alloys as the present ones. In addition, the observation that $\tau''$ decreases with $x$ is consistent with the result [91] that the temperature range over which $T^{3/2}$ variation of spontaneous magnetization in crystalline Fe dominates is confined to temperatures $T < 0.157c$ or simply to $T^* < 0.15$ only. In view of the arguments presented in subsection 3.2.3., one expects a significant reduction in the magnitude of coefficient $A$ of the $T^2$ term in Eq.(3.2b) or Eq.(3.5b) in the presence of an external magnetic field as large as 9 kOe compared to its value in zero field because of the suppression of LSF by external field. Nevertheless, if we ignore this reduction in $A$ for the moment, i.e., set $A(H) = A(0)$, and calculate $T_C^*$ from the relation $T_C^* = (2A(0))^{-1/2}$ using the present $A$ values, the numerical estimates of $T_C^*$, so obtained, consistently exceed the actual $T_C$ values by about 10% for a - Fe$_{x}$Ni$_{80-x}$(Si)$_{20}$ alloys. Since $A(0)$ is actually larger than $A(H)$, the values of $T_C^*$ should be very close to those of $T_C$. Considering that the Stoner theory overestimates $T_C$ by at least an order of magnitude, this result strongly suggests that local spin-density fluctuations dominantly contribute to thermal demagnetization in these alloys.

The observation (ii) that a transition from $T^{5/2}$ variation of $D(T)$ to $T^2$ occurs at $x \sim 60$ at.% should not be interpreted as a transition from localized- to itinerant-electron behaviour but as an indication of the fact that the magnon-magnon interactions, within the framework of itinerant-electron model (Eq.(1.21), Chapter 1), weaken with increasing $x$ so much so that the magnon-SP interactions show up with ease for $x > 60$ at.%. It is now well-known [19,23,53,92] that the nearest-neighbour (NN) configuration of atoms (short-range order) in the Fe — Ni amorphous alloys in question is similar to that found in their crystalline counterparts. In view of this result, the finding that the concentration dependence of $D_{5/2}$ as the same (within the error limits) regardless of whether Fe - Ni alloys are in the crystalline state or in the amorphous state and irrespective of whether the metalloids are present or not suggests that the observed functional dependence of $D_{5/2}$ on $x$ (and hence the magnon-magnon interactions) is primarily dictated by the alteration in NN coordination (both type and number) brought about by the change in Fe concentration. Considering that even the slight change in NN coordination has a marked influence on the creation or annihilation of short-wavelength magnons, the magnon-magnon interactions involving the short-wavelength magnons play a decisive role in the temperature renormalization of spin-wave energies. In view of this argument and the observation that $M(0,0)$ and $D(0)$ have much lower values in the amorphous state compared to those in the crystalline state, it is not surprising that the values of $< r^2 >$ calculated from Eq.(3.53) (Tables 3.5 and 3.6) underestimate the range of exchange interactions and hence tend to give the misleading impression that such interactions for the glassy alloys with $x$ in the range $0 < x < 60$ at.% are confined to the nearest neighbours only. By contrast, the values of $< r^2 >$ computed
from the $D(0)/T_c$ ratio using Eq.(3.54) (Tables 3.5 and 3.6) provide a better handle on the range of exchange interactions since both $D(0)$ and $T_c$ are directly related to the $\chi$ exchange integral through the expression given by the numerator and denominator in Eq.(3.54), respectively.

A downward shift in the $M(T = 0, x)$ vs. $x$ curves (Fig. 3.25) for amorphous alloys in question, i.e., observation (iv) stated above, can be understood [52] in terms of the simple but crude rigid band model by assuming that P donates roughly twice as many electrons per atom to the transition metal d bands as B does. The sharp drop in $M(T = 0, x)$, $D(T = 0, x)$ and $T_c(x)$ for $x > 60$ at.% in co$^{1+}$Fe$_x$Ni$_{100-x}$ alloys is a manifestation of the Invar effect, i.e., the coexistence of antiferromagnetically coupled (low-spin state) and ferromagnetically coupled (high-spin state) spin pairs. Alternatively, the antiferromagnetic (AF) interactions build up at the expense of the ferromagnetic (FM) interactions in these alloys as $x$ is increased beyond 60 at.% resulting in a sharp drop in these quantities. In view of these arguments, progressive slowing down of the rate of increase in $M(T = 0, x)$ and drop in $D(T = 0, x)$, $T_c(x)$ for $x > 60$ at.% in the presently studied alloys indicate that the competing (AF+FM) interactions, more pronounced in co$^{1+}$Fe$_x$B$_{20}$ than in co$^{1+}$Fe$_x$P$_{14}$B$_6$, are present in the alloys with $x$ in the range 60 $\sim$ $x$ $\leq$ 80. This inference is consistent not only with the finding that the ratio $D(0)/T_c$ for co$^{1+}$Fe$_80$B$_{20}$ possesses a value (= 0.14 meV $A^2/K$, Table 3.5) characteristic [9,10,17,26] of amorphous ferromagnets with competing interactions but also with the fact that co$^{1+}$Fe$_80$B$_{20}$ does exhibit [93] Invar behaviour. Competing interactions, in turn, give rise to canted spin arrangement in such ferromagnets. Mossbauer [94] and spin-polarized neutron scattering [95] experiments performed on amorphous Fe-rich (Fe-Ni)-metalloid and/or Fe-metalloid alloys do support the existence of non-collinear (canted) spin structure in the alloys with Fe concentration greater than 60 at.%.

As already mentioned earlier, the present results unambiguously demonstrate that $D_N \gg D_M$ is a characteristic property of the amorphous ferromagnets with competing interactions and/or of amorphous ferromagnetic alloys that exhibit Invar effect (e.g., co$^{1+}$Fe$_80$B$_{20}$ in Table 3.5). Several arguments [13,79-82,85,96-98] have been put forward to explain this discrepancy between $D_M$ and $D_N$ in amorphous ferromagnetic alloys. According to Continentino and Rivier [13], the diffusive modes (“diffusons”) originating from the longitudinal spin fluctuations, contribute as significantly to the $T^{3/2}$ decrease of magnetization in amorphous canted ferromagnets (i.e., the glassy ferromagnets with a noncollinear ground-state) as the transverse spin fluctuations (magnons) do, but these diffusons do not give rise to any propagating features in the constant-$q$ INS intensity versus energy scans. The recent triple axis polarized INS data [99] on co$^{1+}$Fe$_{86}$B$_{14}$ (Invar system) and co$^{1+}$Fe$_{40}$Ni$_{40}$P$_{14}$B$_6$ (non-Invar system) alloys provide strong evidence for the existence of longitudinal spin fluctuations, which, far from being nonpropagating modes (as envisaged by Continentino and Rivier [13]), appear as propagating excitations at energies close to the spin wave peaks in constant-$q$ scans for temperatures well below $T_C$ in both the alloy systems studied. Moreover, these longitudinal propagating excitations, like
magnons, follow the dispersion relation $E(T) = D(T)q^2$ and hence give rise to an additional $T^{3/2}$ contribution to thermal demagnetization while leaving $D_N$ unaltered from its 'spin-wave-only' value. The possibility of a coupling between transverse (spin-wave) and longitudinal spin fluctuations that leads to propagating longitudinal excitations which peak at spin-wave energies was earlier suggested by various workers [100-102]. Another important observation made by Lynn et al. [99], based on the triple-axis polarized INS experiments mentioned above, is that the intensity of the longitudinal mode peaks is very weak in a - $Fe_{40}Ni_{40}P_{14}B_{6}$ compared to that in a - $Fe_{86}B_{14}$. This result is in agreement with the present work (Tables 3.5 and 3.6) that the discrepancy between $D_M$ and $D_N$ is more pronounced for a — $Fe_{80}B_{20}(D_N/D_M = 1.85(32))$ than for a - $Fe_{40}Ni_{40}P_{14}B(D_N/D_M = 1.13(2))$. However, none of the theoretical models proposed hitherto [13,100-102] explains why the peaks in the INS spectra belonging to the longitudinal spin fluctuations are more intense in Invar alloys than in non-Invar systems. These results find a simple interpretation in light of the infinite three-dimensional (3D) ferromagnetic (FM) matrix plus finite FM spin clusters model [17,18,31,32,51] proposed earlier for amorphous ferromagnetic alloys and already described in detail in section 3.2.2. In this model, wild fluctuations in the NN distance within the frustration zones surrounding the finite clusters around the critical distance ($r_c$) at which the exchange integral changes sign in the Bethe-Slater curve gives rise to competing interactions, which, in turn, result in noncollinear arrangement of spins within the infinite 3D FM matrix in amorphous alloys. In Invar systems, an additional contribution to competing interactions arises from strong magnetostrictive coupling between the local ‘quenched-in’ stresses and spins in the buffer zones surrounding the finite spin clusters. Thus, the longitudinal spin fluctuations are more pronounced in Invar systems than in non-Invar ones. As the concentration of the moment bearing (Fe) atoms in the amorphous alloys under investigation increases, the total density decreases (Tables 3.5 and 3.6) such that the average NN distance ($r_{NN}$) between spins in both the FM matrix and the finite spin clusters increases progressively beyond $r_c$ for $x < 60$ at.%, whereas for $x > 60$ at.% the finite spin clusters grow at the expense of the bulk with the result that $r_{NN}$ between spins in the finite clusters is much larger than $r_c$ but $r_{NN}$ is lowered towards $r_c$ for spins in the FM matrix. This process leads to an increase in the average NN (positive) exchange coupling, $J_{NN}$ between spins in the FM matrix (and hence in $D(T,0,x)$ and $Tc(x)$) for $x$ up to 60 at.% but as $x$ is increased beyond 60 at.%, $J_{NN}$ decreases and so do $D(T = 0,x)$ and $T_c(x)$.

3.6. Conclusions

From the detailed analysis and discussion of highly precise magnetization data taken on amorphous ($a-$)$(Fe_pNi_{1-p})_{80}(B,Si)_{20}$ (series I) and $(Fe_pNi_{1-p})_{80}P_{14}B_{6}$ (series II) alloys, the following conclusions can be drawn.
(i) In series I, the spin wave contribution to thermal demagnetization completely dominates over that due to single-particle (SP) excitations of the ‘weak-itinerant-type’ plus spin-density fluctuations (LSF) in the temperature range $0 \sim \tau (=T/T_c) \sim \tau^*(p)$ for all $p$ but the reverse is true for all compositions at temperatures $\tau > \tau^*(p)$. By contrast, in series II, the SW contribution masks the feeble contribution from SP excitations of ‘weak-itinerant-type’ and LSF in the range of temperatures $3.8 \text{K} < T < 0.97\text{c}$ ($3.8 \text{K} < T < 300\text{K}$) for $p < 0.25$ ($0.375 < p < 0.625$), but for $p \sim 0.75$ and $T < 300\text{K}$, a significant contribution from SP excitations of strong-itinerant type accompanies a dominant spin-wave contribution. This result indicates that all the compositions in the $a-(Fe_pNi_{1-p})_{80}(Si)_{20}$ alloy series behave as weak itinerant ferromagnets while a transition from weak-itinerant to strong-itinerant ferromagnetism occurs at a concentration $p \sim 0.75$.

(ii) In both the alloy series, the spin-wave stiffness coefficient renormalizes with $T$ as $D(T) \sim T^{5/2}$ for Fe concentrations up to $p \sim 0.625$ whereas $D(T) \sim T^{3}$ for $0.75 < p < 1.0$. Such a crossover in the temperature dependence of $D$ from $T^{5/2}$ to $T^{3}$ at $p \sim 0.75$ is also observed in $c - Fe_xNi_{100-x}$ ($x = 80\text{p}$) alloys. Within the framework of the itinerant-electron model, this observation implies that the magnon-magnon interactions (especially the ones involving short-wavelength magnons) weaken with increasing $p$ so much so that magnon-SP interactions show up with ease for $p \sim 0.75$.

(iii) The present results unambiguously demonstrate that $D_N$ (the value of $D$ determined from either inelastic neutron scattering (INS) or Brillouin scattering experiments) $>$ $D_M$ (the value of $D$ deduced from the magnetization measurements) is a characteristic property of the amorphous ferromagnets with competing interactions and/or of amorphous ferromagnetic alloys that exhibit Invar effect. Based on the observed $Fe$ concentration dependence of $M(T = 0,p)$, $D(T = 0,p)$ and $T_c(p)$ in the investigated amorphous alloys, it is argued that the competing interactions present in the alloys with $p$ in the range $0.75 \sim p \sim 1.0$ give rise to noncollinear (canted) arrangement of spins in the ground state. This canted spin structure for $p \sim 0.75$, in turn, gives rise to longitudinal spin fluctuations which contribute as significantly to the $T^{3/2}$ decrease of magnetization as the transverse spin fluctuations (spin waves) do but leave $D_N$ unaltered from its ‘spin-wave-only’ value. This explains as to why $D_N > D_M$ for the alloys with $p \sim 0.75$.

(iv) The direct exchange interactions extend beyond the second nearest-neighbor distance for compositions close to, but above, the critical concentration for the appearance of long-range ferromagnetic order whereas the competing interactions in the amorphous alloys with $p > 0.75$ confine the direct d-d exchange interactions to the nearest neighbors only.

(v) The concentration dependence of $D$ and $T_c$ observed in the present work as well as the recent finding that the peaks in the INS spectra due to the longitudinal spin fluctuations are
more intense in Invar alloys than in non Invar systems find a straightforward explanation in terms of the infinite 3D ferromagnetic matrix (FM) plus finite FM spin clusters model.
References


