SUMMARY

High-precision bulk magnetization (BM) measurements on amorphous (a—) Fe_{90+y}Zr_{10-y} (y = 0, 1), Fe_{90-x}Co_{x}Zr_{10} (0 < x < 10; r = 90), (Fe_{p}Ni_{1-p})_{80}P_{14}B_{6} (0.1125 < p < 1.0) and (Fe_{p}N_{i_{1-p}})_{80}P_{14}B_{6} (0.0625 < p < 1.0) alloys over a wide range of temperatures (4 K < T < 300 K or 70 K < T < 400 K) and fields (0 < H < 15 kOe), ferromagnetic resonance (FMR) measurements on a - Fe_{90+y}Zr_{10-y} (y = 0, 1) and a - Fe_{90-x}Co_{x}Zr_{10} (0 < x < 10) alloys in the temperature range 77 K < T < 350 K [4 K < T < 600 K] have been performed with the following objectives in mind.

(i) To make an in-depth study of low-lying magnetic excitations in all the above-mentioned alloy systems.

(ii) To investigate the asymptotic critical behaviour of a—Fe_{90+y}Zr_{10-y} and a—Fe_{90-x}Co_{x}Zr_{10} alloys near the ferromagnetic (FM)-paramagnetic (PM) phase transition.

(iii) To study the effect of quenched randomness on the percolation critical behaviour of three-dimensional site-diluted ferromagnets.

(iv) To identify the scattering mechanisms responsible for p(T) in a - Fe_{90+y}Zr_{10-y} and a - Fe_{90-x}Co_{x}Zr_{10} alloys and to determine their relative strengths in different temperature ranges.

A brief summary of the most important findings, based on elaborate data analyses and a detailed discussion of the results in terms of the existing theoretical models, is given below.

In a - Fe_{90+y}Zr_{10-y} and a - Fe_{90-x}Co_{x}Zr_{10} alloys, magnetization (M) does not saturate even for external magnetic fields (H) as high as 70 kOe at 5 K for the alloys with y = 0, 1 and x ~ 6. The high-field differential susceptibility, \chi_{hf}(0), determined from the M vs. H isotherm taken at 5 K is extremely large in the alloys with y = 0, 1 and x = 0, 1 but decreases rapidly with increasing x for x ~ 4 so that it possesses values typical of crystalline counterparts for x > 6. Large values \chi_{hf}(0) have been observed previously in the alloys with p ~ 0.75 in the series \textit{a - (Fe_{p}Ni_{1-p})_{80}P_{14}B_{6}} well. Large \chi_{hf}(0) is a strong indication of the presence of competing interactions in the alloys mentioned above. Competing interactions progressively \textit{pick up} in strength as x \rightarrow 0, y \rightarrow 1 and p \rightarrow 1 in the corresponding alloy systems. This inference is further corroborated by the observations that (a) spontaneous magnetization as well as Curie temperature (Tc) fall steeply as these limiting concentration values (i.e., x = 0, y = 1 and p = 1) are approached, and (b) the spin-wave stiffness coefficient (D) to Tc ratio (i.e., D/Tc ratio) possesses a value \sim 0.14 that is characteristic of amorphous ferromagnets with
competing interactions. Competing interactions confine the direct exchange interactions to the nearest neighbours only and are primarily responsible for a non-collinear arrangement of spins in the ground state. Non-collinearity in the spin structure in a way ensures that the diffusons (diffusive modes associated with the longitudinal component of magnetization) contribute to the $T^{3/2}$ decrease of magnetization as significantly as the transverse spin fluctuations (spin waves) do. However, longitudinal spin fluctuations (diffusons) do not alter the 'spin-wave-only' value. $D_N$ of the spin wave stiffness because their coupling with the transverse spin fluctuations leads to propagating longitudinal excitations which peak at spin-wave energies in the inelastic neutron scattering (INS) spectra. Consequently, for the alloys with $x < 6$, $y = 0, 1$ and $p > 0.75$, $D_M$ (the value of $D$ deduced from the magnetization measurements) $< D_N$ (the value of $D$ determined from INS experiments). Significantly large longitudinal spin fluctuation contribution also gives rise to a $T$ temperature dependence of resistivity at low temperatures ($T < 10$ K) in $a - Fe_{90+y}Zr_{10-}$ ($y = 0, 1$) alloys.

Dominant contribution to the thermal demagnetization of spontaneous magnetization. $M(T, 0)$ in all the alloys in the series $a - Fe_{90+y}Zr_{10-y}$ and $a - Fe_{90-x}Co_xZr_{10}$ except for the one with $x = 90$, is due to spin-wave excitations at temperatures $T ~ 0.3T_C$ (at these temperatures, diffusons also give a contribution whose strength depends on composition) and enhanced fluctuations in the local magnetization over a wide temperature range extending from $0.4T_C$ to $0.95T_C$. In $a - Co_{90}Zr_{10}$ alloy, a dominant spin-wave contribution to both $M(T, 0)$ and $M(T, H)$ at low temperatures ($T ~ 0.17c$) is followed by an overwhelming contribution from Stoner single-particle excitations of weak-itinerant type at higher temperatures, implying thereby that the particle-hole pair excitations are very weakly correlated in this case. Notwithstanding the temperature ranges, which depend on composition, spin waves and local spin-density fluctuations do contribute to the thermal demagnetization in all the alloys in the series $a - (Fe_pN_{1+y})_{80}P_{14}B_{6}$ and $a - (Fe_pN_{1+y})_{80}P_{14}B_{6}$ with the exception of those with $p \sim 0.75$ in the latter series, in exactly the same way as in the alloys with $y = 0, 1$ and $T < 10$. For the compositions $p \sim 0.75$ in the alloy series $a - (Fe_pN_{1+y})_{80}P_{14}B_{6}$, a significant contribution from the Stoner single-particle excitations of strong-itinerant type accompanies a dominant spin-wave contribution at temperatures $< 300$ K. The above finding asserts that all the alloys in the series $a - Fe_{90+y}Zr_{10-y}$, $a - Fe_{90-x}Co_xZr_{10}$ and $a - (Fe_pN_{1+y})_{80}(B, Si)$ are weak itinerant ferromagnets while a transition from weak-itinerant to strong-itinerant ferromagnetism occurs at a concentration $p \sim 0.75$ in the $a - (Fe_pN_{1+y})_{80}P_{14}B_{6}$ by series. Despite an overwhelmingly large electron-phonon scattering (structural disorder scattering) contribution, spin wave excitations and local spin-density fluctuations make their presence felt through their contributions to resistivity in the temperature ranges similar to those observed in the $M(T, 0)$ data for the alloys with $y = 0, 1$ and $x < 10$. Contrasted with the suppression of spin fluctuations by the external magnetic field (H) and/or Co substitution in $a - Fe_{90-x}Co_xZr_{10}$ alloys, spin-wave stiffness does not depend on $H$ and spin-wave modes stiffen with increasing Co concentration. 
(i.e., \( D(0) \) increases with \( x \)). Sensitivity of spin fluctuations to field and Co concentration finds a straightforward explanation in terms of a spin fluctuation model.

Accurate values of the asymptotic critical exponents and universal amplitude ratios that characterize the FM-PM phase transition at \( T_C \), determined from the magnetization data taken in the critical region on \( a - Fe_{90+y}Zr_{10}(y = 0.1) \) and \( a - Fe_{90-x}Co_xZr_{10}(x = 0, 1, 2, 4, 6) \) alloys, are composition-independent and conform very well with the corresponding theoretical estimates for an ordered (pure) isotropic spin system with \( d - n = 3 \). This result vindicates the famous Harris criterion which states that the critical behaviour of a three-dimensional (3D) ordered spin system does not get altered in the presence of short-ranged quenched disorder if the specific heat critical exponent of the pure system is negative. Consistent with the result that the presently determined asymptotic critical exponents satisfy the Widom scaling equality to a very high degree of accuracy, the magnetization data obey the scaling equation of state (SF.S) valid for the second-order phase transition. SES analysis of the BM and FMR data taken in the critical region yields identical values for the effective critical exponents and amplitudes. This result, for the first time, projects the FMR technique as a powerful experimental tool to study critical behaviour of ferromagnetic systems. In accordance with the theoretical expectations, the present results demonstrate that the nonanalytic confluent corrections, arising from nonlinear irrelevant scaling fields, dominate over the analytic ones, originating from nonlinear relevant scaling fields, in the asymptotic critical region (ACR) but the reverse is true for temperatures outside the ACR. Another important finding is that the magnetic equation of state in linear variables forms an adequate description of the magnetization data for temperatures close to \( T_C \) whereas its counterpart in nonlinear variables properly accounts for the observed \( M(T, H) \) behaviour in a much wider temperature range around \( T_C \). Instead of exhibiting a Curie-Weiss-like behaviour, the initial susceptibility follows the generalized Curie-Weiss law for temperatures in the range \( T_C \) to \( \sim 1.5T_C \). This permits an unambiguous determination of the atomic moment in the paramagnetic state, \( q_c \). Consistent with the conclusions drawn from the \( M(T, H) \) data taken at temperatures below \( T_C \) regarding the nature of magnetism in these alloys, the observations that the \( q_c/q_s \) ratio is > 1 and scales with \( T_C^{-1} \) or \( T_C^{-1/3} \) assert that the alloys with \( y = 0, 1 \) and \( x < 6 \) are weak itinerant ferromagnets. An order of magnitude smaller value of the ratio \( \mu_0h_c/k_BT_C \) in the above-mentioned alloys than that theoretically predicted for a 3D Heisenberg ferromagnet is shown to imply that the fraction of spins actually participating in the FM-PM phase transition is as small as 5% for the alloy with \( x = 0 \) and increases to 12% at \( x = 6 \). This inference is in consonance with the absence of a pronounced anomaly in the temperature derivative of resistivity at \( T_C \) because it is impossible to detect a small amount of magnetic entropy released at \( T_C \) in the measurement of total resistivity which, besides this singular (magnetic) contribution, contains an overwhelmingly large non-singular contribution arising mainly from the scattering of conduction electrons from the disordered structure. The infinite ferromagnetic matrix plus finite (ferromagnetic) spin clusters model provides a simple but qualitative interpretation for all
the diverse aspects of the results mentioned so far.

The $T_c(p), M(T = 0,p)$ and $D(T = 0,p)$ data, obtained from the bulk magnetization measurements performed on a $-(Fe_pNi_{1-p})_{80}(B,Si)_{20}$ and $a-(Fe_pNi_{1-p})_{80}P_{14}B$ alloys, when analyzed with caution, yield accurate values for the 'percolation-to-thermal' crossover exponent, $\phi$, for Curie temperature and the percolation critical exponents, $\theta_p$, for spin wave stiffness at 0 K, and $\beta_p$ for spontaneous magnetization at 0 K. It is observed that the asymptotic critical region, where single power laws are valid, is wide for $T_c(p)$ and $D(T = 0,p)$ but extremely narrow for $M(T = 0,p)$; in the latter case, the leading 'correction-to-scaling' term had to be included in the expression for $M(T = 0,p)$ in order to arrive at the true asymptotic values of the critical exponent $\beta_p$ and amplitude $m_p$. Other percolation critical exponents such as $\nu_p$ for the correlation (connectedness) length and $\sigma$ for macroscopic conductivity are calculated from the exponent equalities using the presently determined values of the exponents $\beta_p$, $\theta_p$ and $\phi$.

Making use of the values of $\beta_p$, $\nu_p$ and $\sigma$ in the relations predicted by the self-similar fractal model for the structure of the percolating cluster at threshold ($p_c$), the fractal dimension $d$ and the fracton (spectral) dimensionality $d$ of the percolating cluster at $p_c$ are calculated. A close agreement between the percolation critical exponents for amorphous site-diluted ferromagnets, so determined, and those theoretically predicted for site- or bond-percolation on three-dimensional crystalline lattices asserts that the critical behaviour of percolation on a regular $d = 3$ lattice remains unaltered in the presence of quenched randomness if the specific heat critical exponent of the regular system is negative. In other words, the Harris criterion is valid for the percolation critical behaviour, as is the case for the thermal critical behaviour also. In accordance with the Alexander-Orbach conjecture, the fracton dimensionality $d$ has a value close to 4/3.

Moreover, the conductivity critical exponent $c$ obeys the Golden inequality $c < 2$. From the values of the critical concentrations for site percolation in the glassy alloy series in question, it is inferred that, particularly for concentrations near $p_c$, the range of exchange interactions in $a-(Fe_pNi_{1-p})_{80}P_{14}B$ alloys nearly equals the third nearest-neighbour (NN) distance whereas the exchange interactions in $a-(Fe_pNi_{1-p})_{80}(B,Si)_{20}$ alloys extend well beyond the third NN distance. The observation that the range of exchange interactions is widely different in the two series and yet the percolation critical exponents have the same values for both of them vindicates the universality hypothesis.

While the enhanced electron-electron interaction effects account for the $\sqrt{T}$ behaviour of $p(T)$ in the temperature range 10 K to 25 K, the weak-localization or quantum interference effects, electron-phonon scattering, electron-magnon scattering and scattering of conduction electrons from the spin fluctuations are the main mechanisms that contribute to the total resistivity for temperatures $T > 25$ K. Unambiguous identification of the dominant scattering processes in different temperature ranges and the determination of their relative magnitudes permits us to calculate the values of the diffusion constant, $D$, and the dephasing time (inelastic scattering time) $\tau_{1e}$. Consistent with the prediction of the spin fluctuation model, the coefficients of the
$7^2$ and $T^{5/3}$ terms (which reflect the spin fluctuation contribution to resistivity in different temperature ranges) decrease with increasing Co concentration and the coefficient of the $T^{5/3}$ term is larger for $T \sim T_c$ than for $T \sim 0$. Out of the inelastic scattering processes such as electron-phonon scattering, spin-orbit scattering and spin-flip scattering, that destroy phase coherence, electron-phonon scattering is the most effective dephasing mechanism. Dephasing due to inelastic electron-phonon scattering mechanism persists to temperatures well above $T_c$ and $\Theta_I$ for the alloys with $y = 0$ and 1.