Identification of the Curie point in FINEMET based alloys using the in-situ X-ray diffraction

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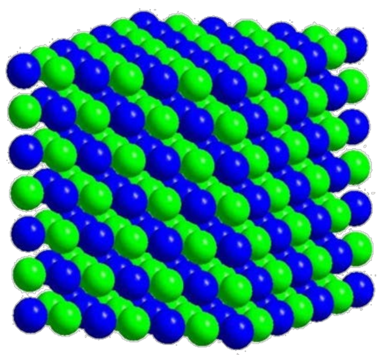
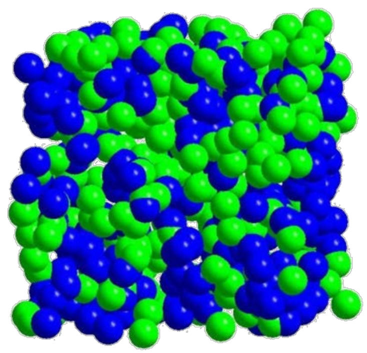
Motivation

- to find correlation between structural changes occurring at the atomic level and macroscopic characteristics of the investigated materials such as metallic glasses (MGs)

Challenges





The atomic arrangement in amorphous alloys is not well understood compared with the well-defined long-range order in crystalline materials.

Methods for structural characterization of MGs

- x-ray and neutron diffraction (XRD, ND)
- transmission electron microscopy (TEM)
- small angle scattering (SXAS, SNAS)
- x-ray absorption spectroscopy (EXAFS, XANES)
- Mossbauer spectroscopy (MS)
- others

Methods for macroscopic magnetic characterization of MGs

- thermomagnetic measurements
- hysteresis loops measurements
- susceptibility and permeability measurements

NS and MS are well recognized for allowing direct correlation of the magnetic state of a material with its structure.

Motivation

Thermal expansion measured by the XRD

A.R. Yavari et al. *Acta Materialia* **53** (2005) 1611-1619

$$\left\{\frac{Q_{\max}(T_0)}{Q_{\max}(T)}\right\}^3 = \left\{\frac{V(T)}{V(T_0)}\right\} = 1 + \alpha_{th}(T - T_0)$$

- the relaxation T_r and glass T_g temperature and thermal expansion coefficients were identified using XRD
- irreversible processes were revealed on the base of reciprocal space analysis

Strain distribution measured by the XRD

H. Poulsen et al. *Nat. Mater.* **4** (2005) 33

$$\varepsilon_i(\varphi_i, \sigma) = \frac{Q_{\max}(\varphi_i, \sigma_0) - Q_{\max}(\varphi_i, \sigma)}{Q_{\max}(\varphi_i, \sigma)}$$

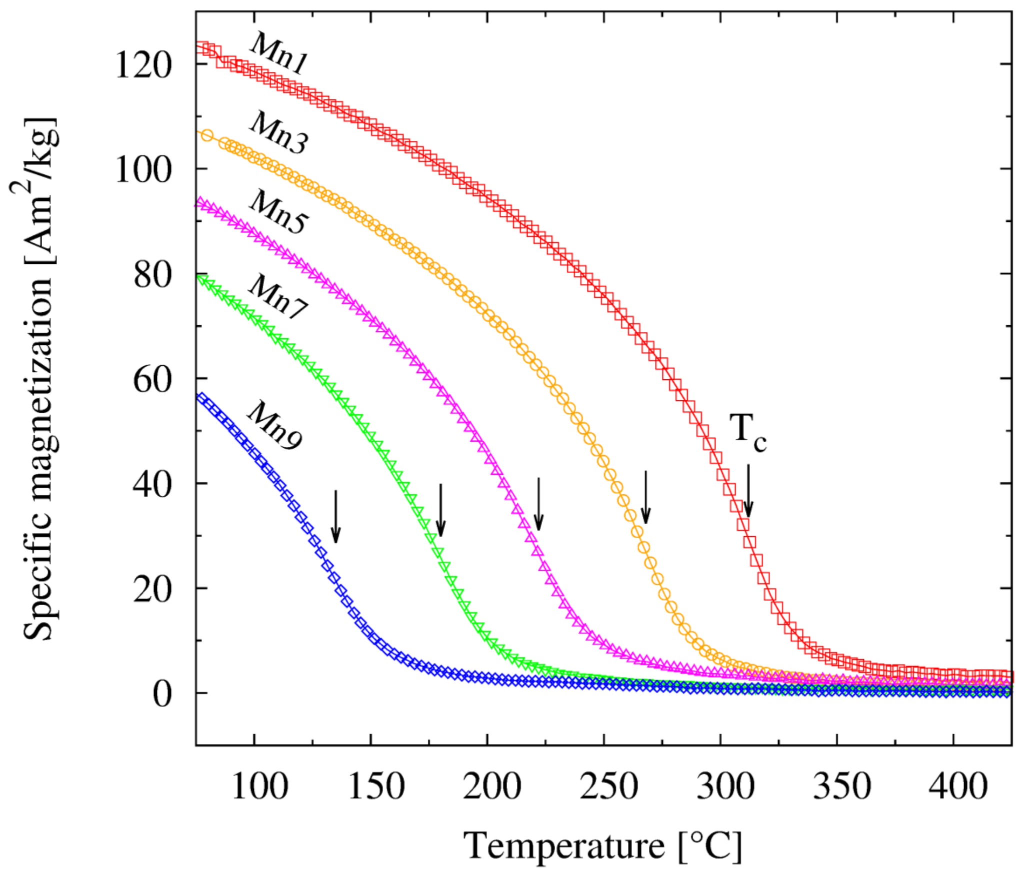
- transversal and tensile coefficient of deformation tensors were estimated using XRD

- *Could the XRD be used to detect the transformation from the fero-magnetic to paramagnetic state in the case of MGs???*

Selection of a suitable MG system

- we are looking for a MG system in which the gradual change of the chemical composition influences the Curie temperature

$Fe_{(73.5-x)}Mn_xSi_{13.5}Cu_1Nb_3B_9$ for x = 1, 3, 5,7 and 9--> **FINEMET based MGs**

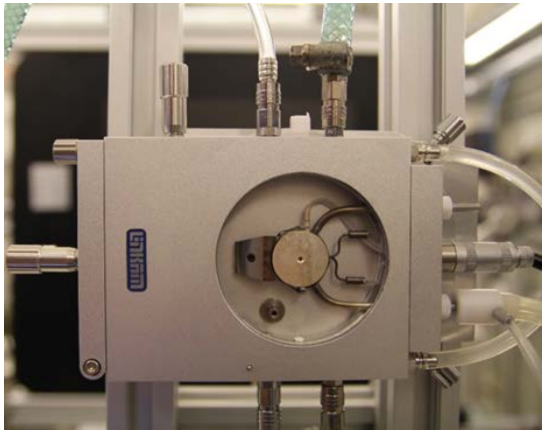


- the Curie temperature T_C of the amorphous phase linearly decreases with increasing amount of Mn in the investigated samples

In-situ XRD measurements

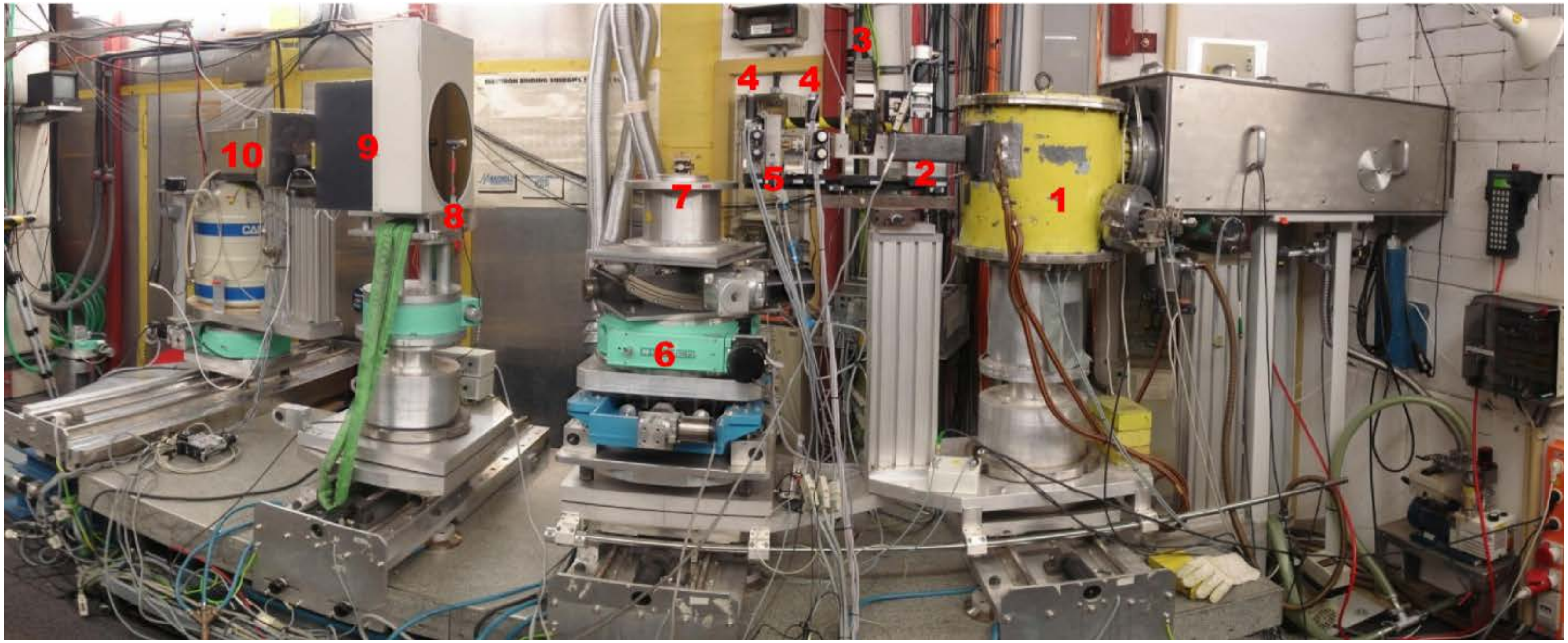
- were performed at the wiggler beamline BW5 at HASYLAB/DESY (Hamburg, Germany)
  - $\lambda = 0.1128$ Å, beam size 0.5×0.5 mm^2
  - illumination time 20 sec
  - diffraction patterns were collected continuously during heating

Linkam THMS600 hot stage

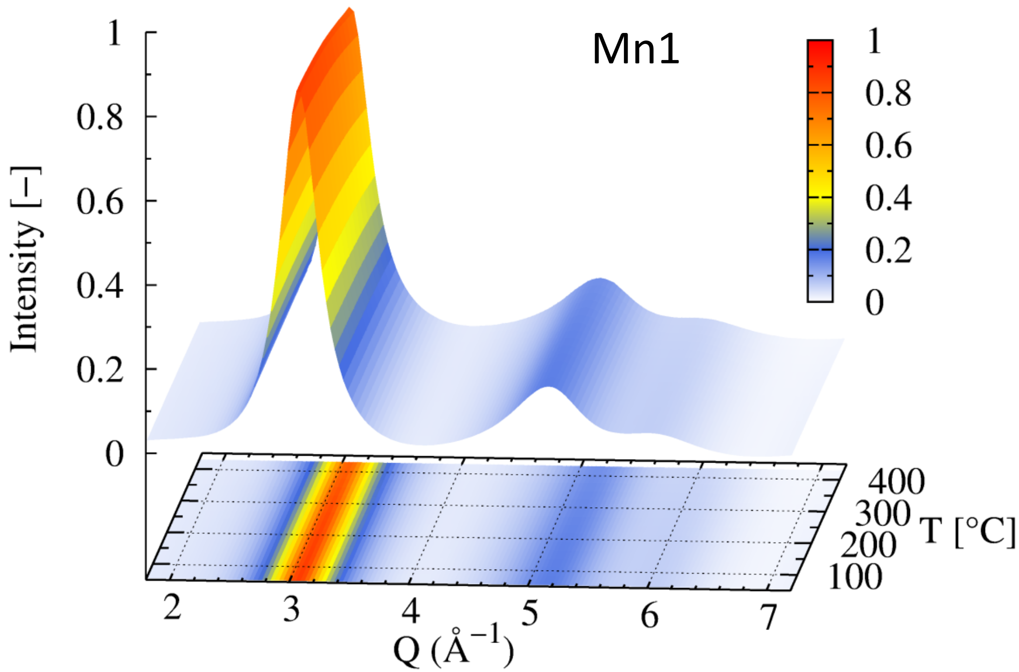


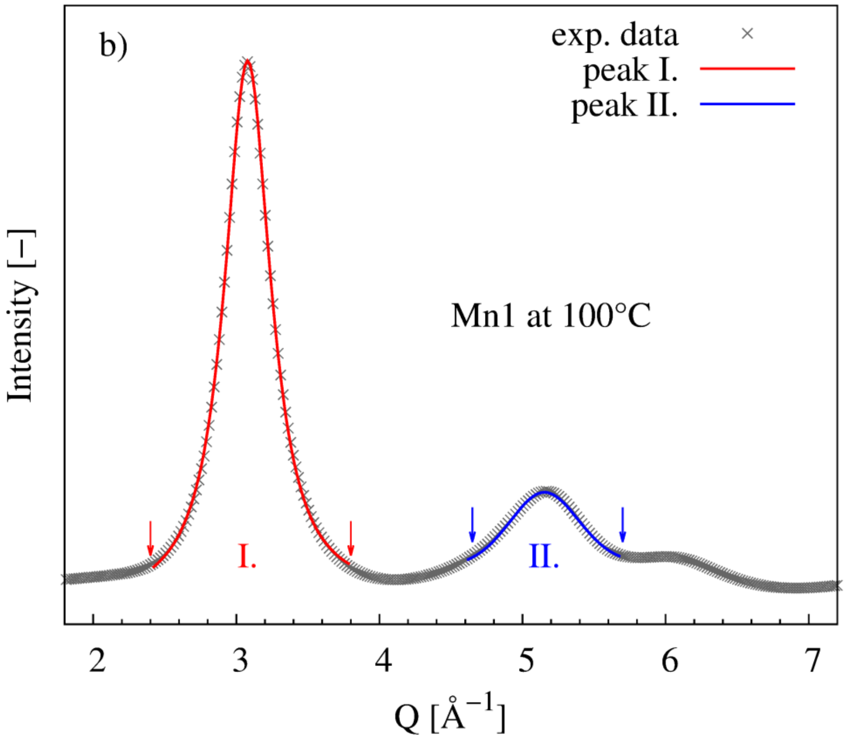
Sample treatment

25°C —(10°C/min)→ 450°C



Results in reciprocal space

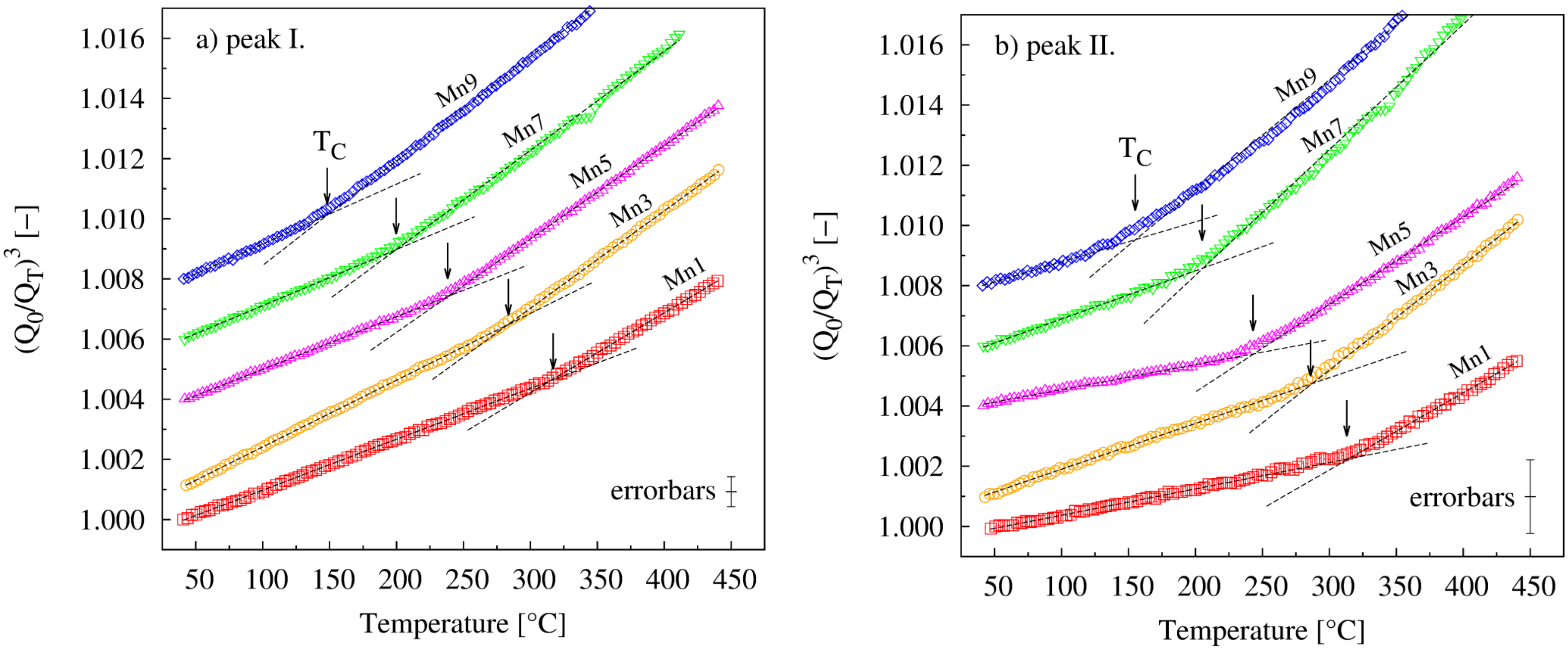




- the samples remain in an amorphous state during the whole heating
- detailed analysis shows a continuous shift of the first diffraction maxima

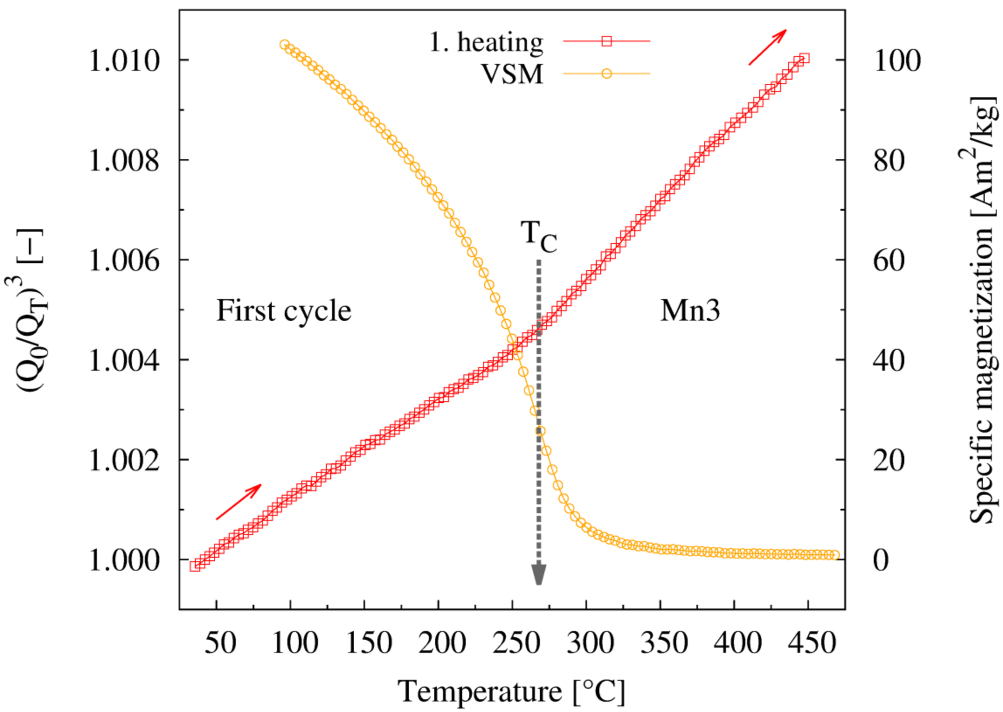
- the first and second peak are fitted with the pseudo-Voight function

Results in reciprocal space



- two distinct temperature regions with different slopes of the curves can be recognized
- the thermal expansion coefficient is increased about 1.5 to 1.8 times about the critical temperature

Results in reciprocal space



sample	T_C^{VSM} [°C]	T_C^{XRD} [°C]
Mn1	312±5	313±15
Mn3	268±5	286±15
Mn5	222±5	243±15
Mn7	180±5	205±15
Mn9	135±5	155±15

S.Michalik et al., J.Phys. D: Appl. Phys. (2012) 455302

- the $(Q_0/Q_T)^3$ ratio changes the slope around the Curie temperature determined as an inflection point of the corresponding thermomagnetic curve.

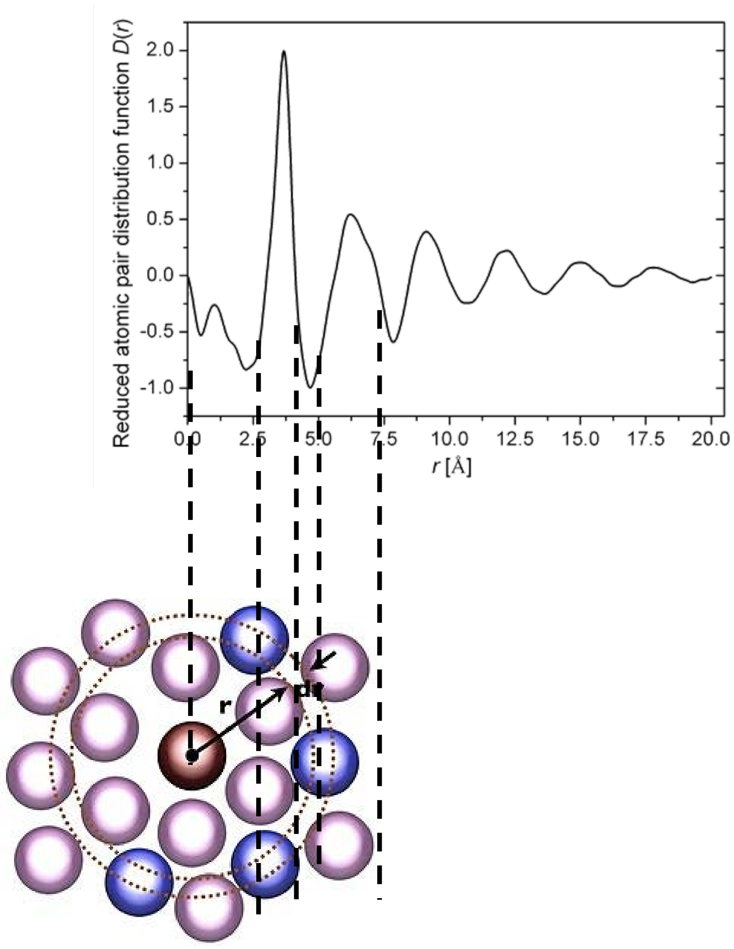
In-situ XRD provide enough sensitivity to detect the transformation from the ferromagnetic to the paramagnetic state in the case of Fe-based metallic glasses.

Results in real space
Atomic pair distribution functions

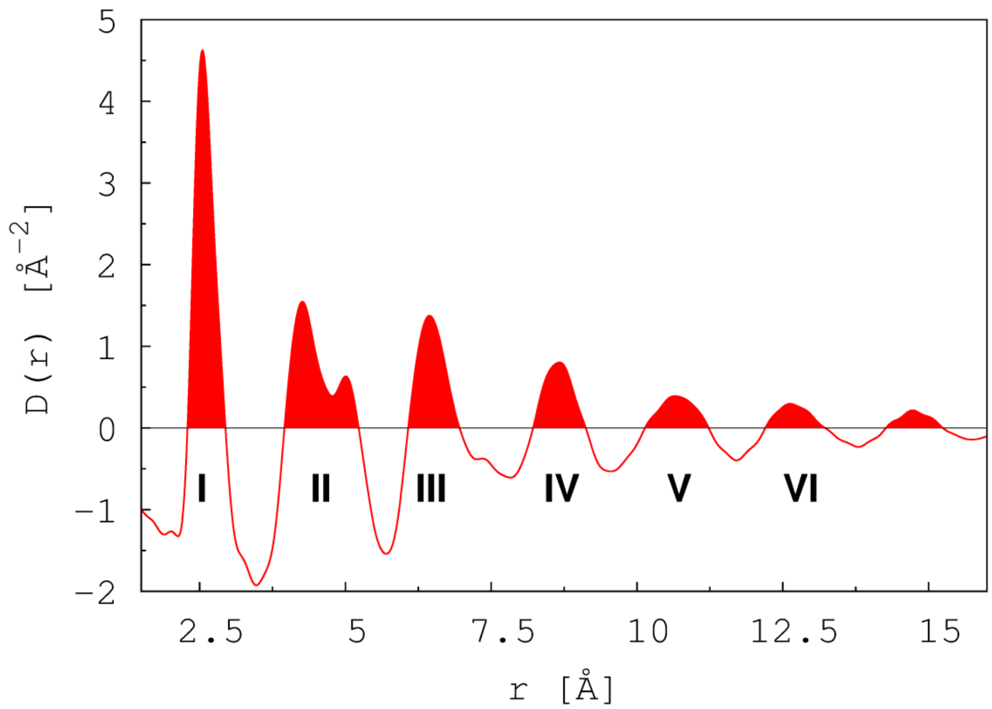
- in order to get some additional information, reduced atomic pair distribution functions were calculated

$$S(Q) = \frac{I_e(Q) - \left\langle f^2(Q) \right\rangle}{\left\langle f(Q) \right\rangle^2}$$

$$D(r) = \frac{2}{\pi} \int_{Q_{\min}}^{Q_{\max}} Q[S(Q)-1]\sin(rQ)dQ$$



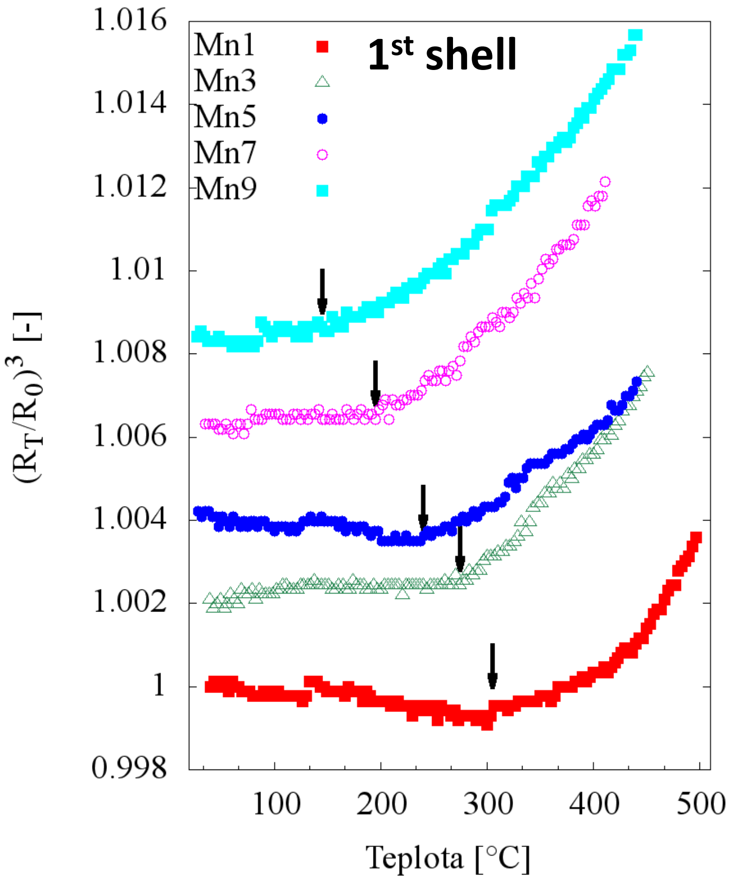
Results in real space

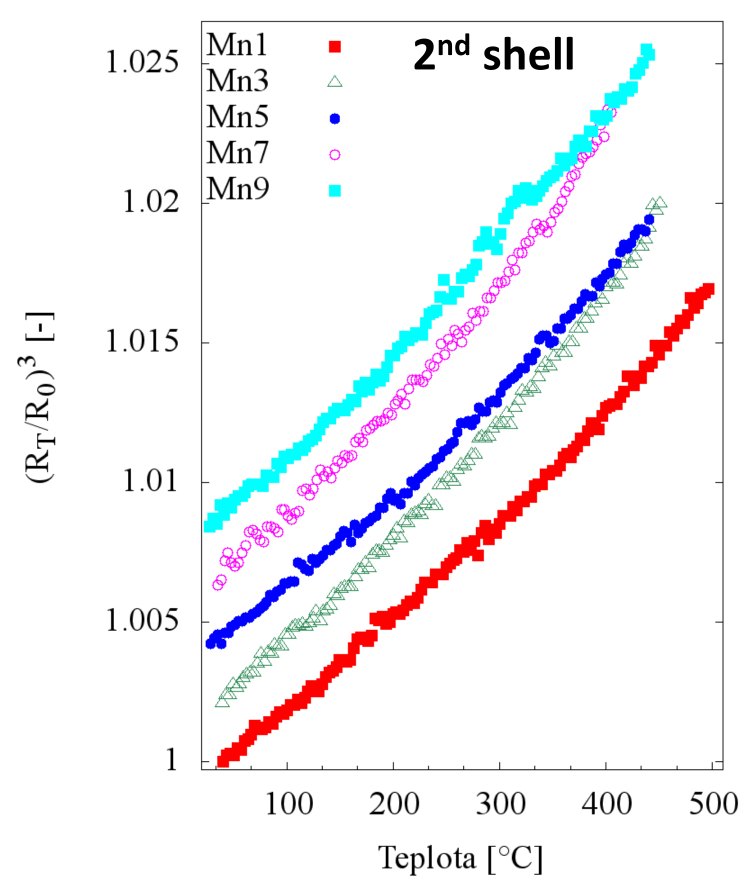


- the interpretation is ambitious due to the multicomponent character of the investigated samples
- atomic scattering factors and atomic radii of Fe and Mn atoms are practically the same (r_{Fe}= 1.241 Å and r_{Mn} = 1.240 Å)

- we decided to follow relative changes of the center of the mass of coordination shells as a function of temperature

Results in real space





- the first maximum of the reduced distribution function $D(r)$ is sensitive to the Curie temperature

- coordination shells of $D(r)$ at higher r-values are without any indication of the feromagnetic phase transformation

Conclusions

- the thermal expansion behaviour of $Fe_{(73.5-x)}Mn_xSi_{13.5}Cu_1Nb_3B_9$ for x = 1, 3, 5, 7 and 9 was followed on the atomic scale using the in-situ XRD
- thermal expansion behaviour reflects the transition from the ferromagnetic to the paramagnetic state
- the temperatures at which the expansion curves changes their slopes are in good agreement with values of the Curie temperatures determined from thermomagnetic curves

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Our results suggest that in-situ XRD measurements provide enough sensitivity to detect the transition from the ferromagnetic to the paramagnetic state in the case of Fe-based metallic glasses.

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