Low-temperature magnetic transitions in Fe_2MnSi and Fe_2MnAl Heusler alloys prepared in bulk and ribbon form

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AIM:Characterization of Fe_2MnSi and Fe_2MnAl alloys prepared by traditional arc and induction melting techniques and non-traditional planar flow casting.

Samples preparation

Materials - Fe_2MnSi and **Fe_2MnAl** alloys prepared from high-purity elements (Fe - 99.95%, Mn - 99.9%, Si - 99.9%, and Al - 99.95%) at Institute of Physics, Slovak Academy of Sciences in Bratislava

Technological procedures - arc melting (AM) using a MAM-1 furnace, **induction melting (IM)**, and **planar flow casting (PFC)**

DAM and DIM samples

AM and **IM** used for production of button- and cylindrical-type ingots, melted four times to ensure good homogeneity, subsequently cut using spark erosion in deionized water into **discs 500 μm thick**, polished using Vibromet for 24h

Ribbon (R) samples

R samples 2 mm wide and 20 μm thick, side in contact with surrounding atmosphere denoted as **air side**, the opposite one as **wheel side**, high brittleness excluded any of surface treatment, higher surface roughness especially from wheel side

Experimental techniques

SEM (Scanning Electron Microscopy) - TESCAN LYRA 3XMU FEG/SEM, accelerating voltage 20 kV, Xmax80 Oxford Instruments detector for Energy Dispersive X-ray (EDX) analysis

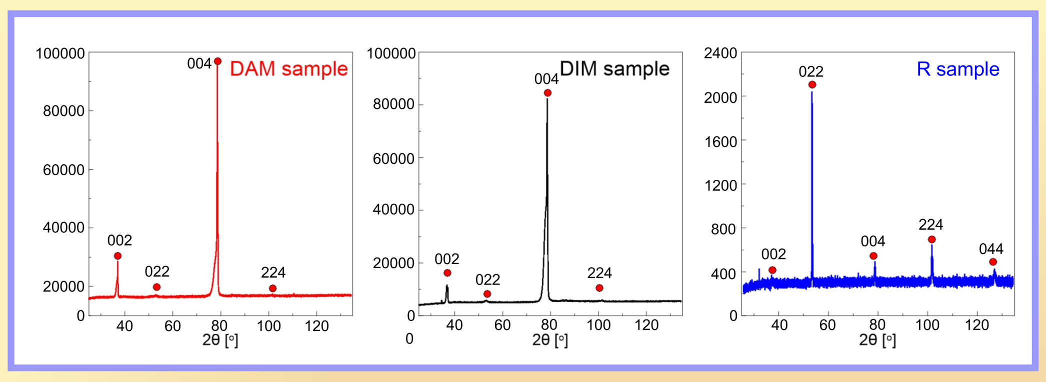
XRPD (X-Ray Powder Diffraction) - X'PERT PRO powder diffractometer, CoKα radiation (λ = 0.17902 nm), Bragg-Brentano geometry, 2θ = 25° - 135°, HighScore Plus software with Rietveld structure refinement method

MS (Mössbauer spectroscopy) - ^{57}Co(Rh) source, measurements at room temperature, Transmission MS applied for ribbons, Backscattering MS used for ingots, calibration of velocity scales with α-Fe, in the measured spectrum the crystalline components represented by singlet and doublets determined by discrete values of hyperfine parameters corresponding to paramagnetic phases: δ - isomer shift(s), Δ - quadrupole splitting(s)

Magnetic measurements - VSM Microsense EV9 magnetometer used for experiments at elevated temperatures (293 K - 573 K) with maximal field ±1600 kA/m (±2 T), PPMS Quantum Design Inc. applied for hysteresis loops at constant temteratures (293 K, 2 K) with maximal field ±400 kA/m (±5 T) and for FC-ZFC curves in magnetic field of 8 kA/m

Fe_2MnSi alloys

Phase and chemical composition, morphology



- XRD patterns analyzed by ICSD data sheet 659018 and results are shown in table below

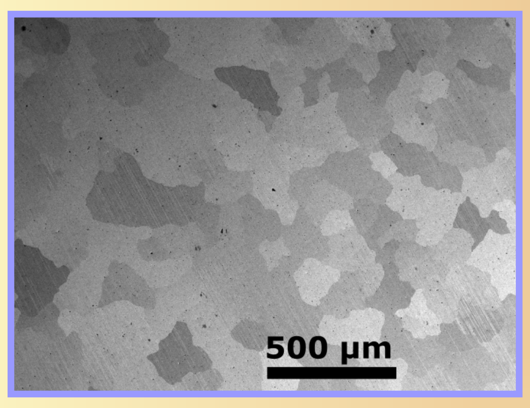
- the presence of cubic $L2_1$ phase confirmed

- diffractograms taken for different surfaces of the R sample were identical

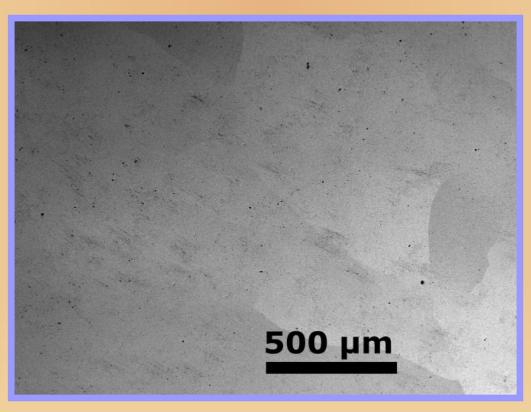
sample type		*a* (nm)	*E* (%)	*d* (nm)	Fe (at.%)	Mn (at.%)	Si (at.%)
DAM		0.569(3)	0.000	4.9	48.30±0.14	24.69±0.09	27.01±0.05
DIM		0.571(5)	0.000	3.7	48.08±0.25	24.69±0.17	27.23±0.18
R	air side	0.569(1)	0.051	86.5	47.10±0.19	27.62±0.52	25.28±0.36
	wheel side	0.560(2)	0.062	409.4	43.36±0.67	30.54±0.19	26.10±0.84

Lattice constant (a), microstrain (E), and diffracting domains size (d) estimated from XRD patterns; element concentration obtained by EDX analysis from areas about 1 mm^2.

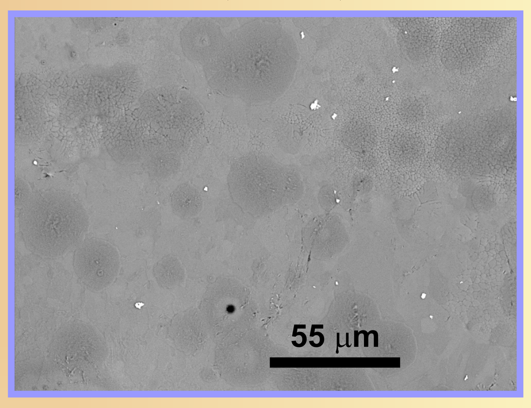
DAM



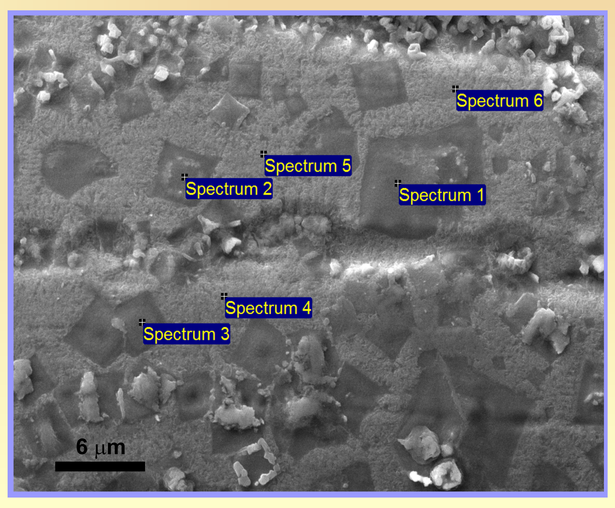
DIM



R (air side)

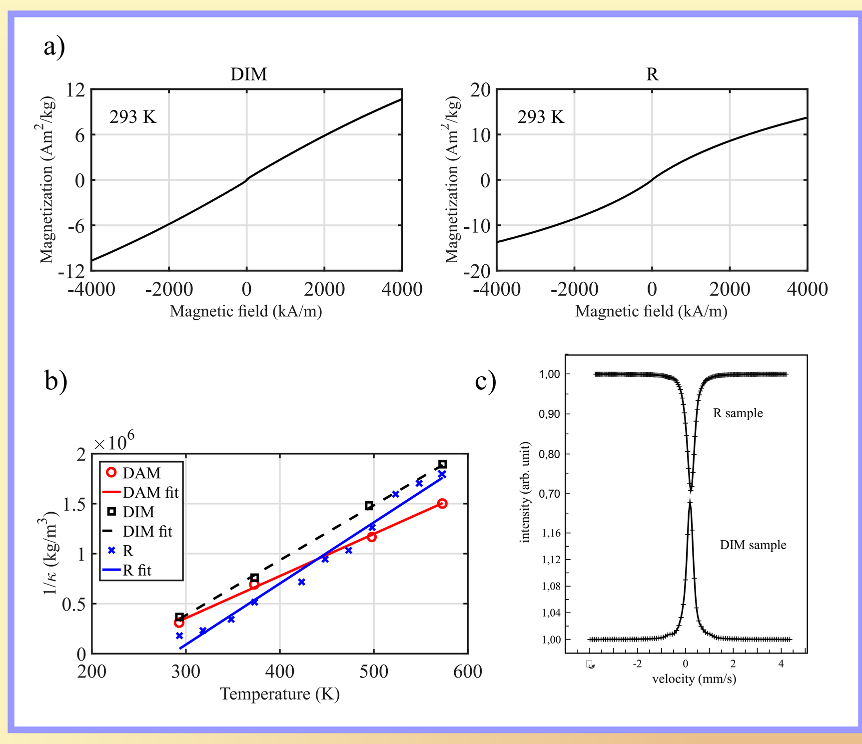


EDX point analysis from the wheel side of the R sample



Spectrum	Si (at. %)	Mn (at. %)	Fe (at. %)
1	28.19	32.07	39.73
2	27.59	35.25	37.16
3	27.49	34.49	38.02
4	27.00	27.47	45.53
5	27.69	26.86	45.46
6	27.38	27.58	45.04

Magnetic and Mössbauer results



- similar magnetic properties of DAM and DIM at room temperature (RT)

- RT hysteresis loops and Mössbauer spectra indicate paramagnetic behaviour of all samples

- Mössbauer spectra analyzed using dominant singlet and three doublets

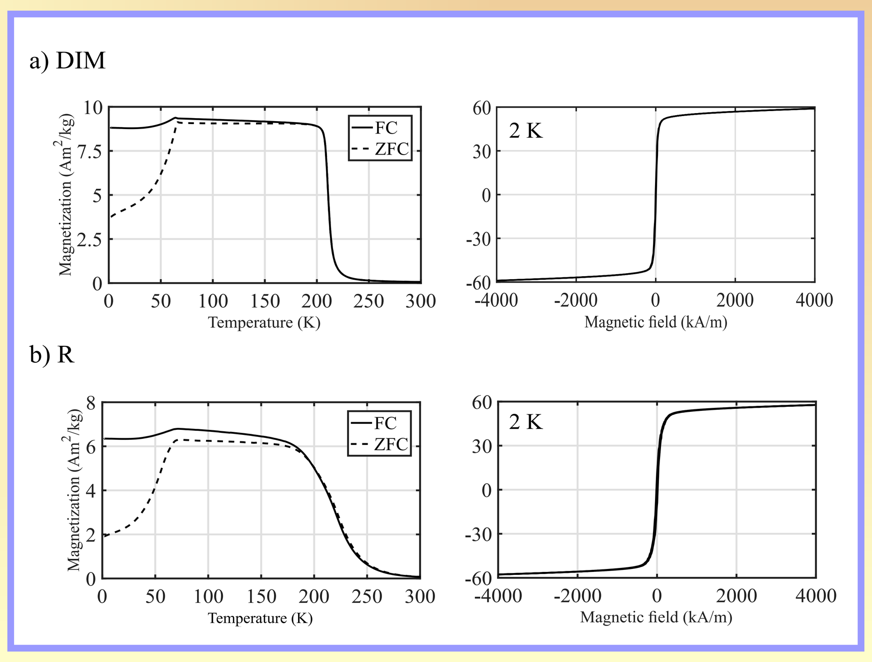
- precise identification of Curie temperature (T_c) done using the Curie-Weiss law:

$$1/\chi = (T - T_c)/C,$$

where χ is magnetic susceptibility of hysteresis loop, C describes the Curie constant, and T denotes applied temperatures in the range 293 K - 573 K

Fundamental magnetic parameters determined from hysteresis loops at 2 K and 293 K, FC-ZFC curves, and Curie-Weiss law; $M5$ - magnetization at 5T; M_r - remnant magnetization; H_c - corcive field; C - Curie constant; T_c - Curie temperature; T_R - temperature of maximum of ZFC curve.

sample type	T (K)	$M5$ (Am^2/kg)	M_r (Am^2/kg)	H_c (kA/m)	C (m^3K/kg)	T_c (K)	T_R (K)
DAM	293	10.55	0.02	8.84	$2.37\cdot10^{-4}$	215.78	68.58
	2	57.83	11.22	8.61			
DIM	293	10.65	0.04	10.14	$1.82\cdot10^{-4}$	230.04	66.42
	2	59.04	9.55	10.35			
R	293	13.55	0.01	1.13	$1.64\cdot10^{-4}$	271.52	69.02
	2	57.05	10.11	16.02			

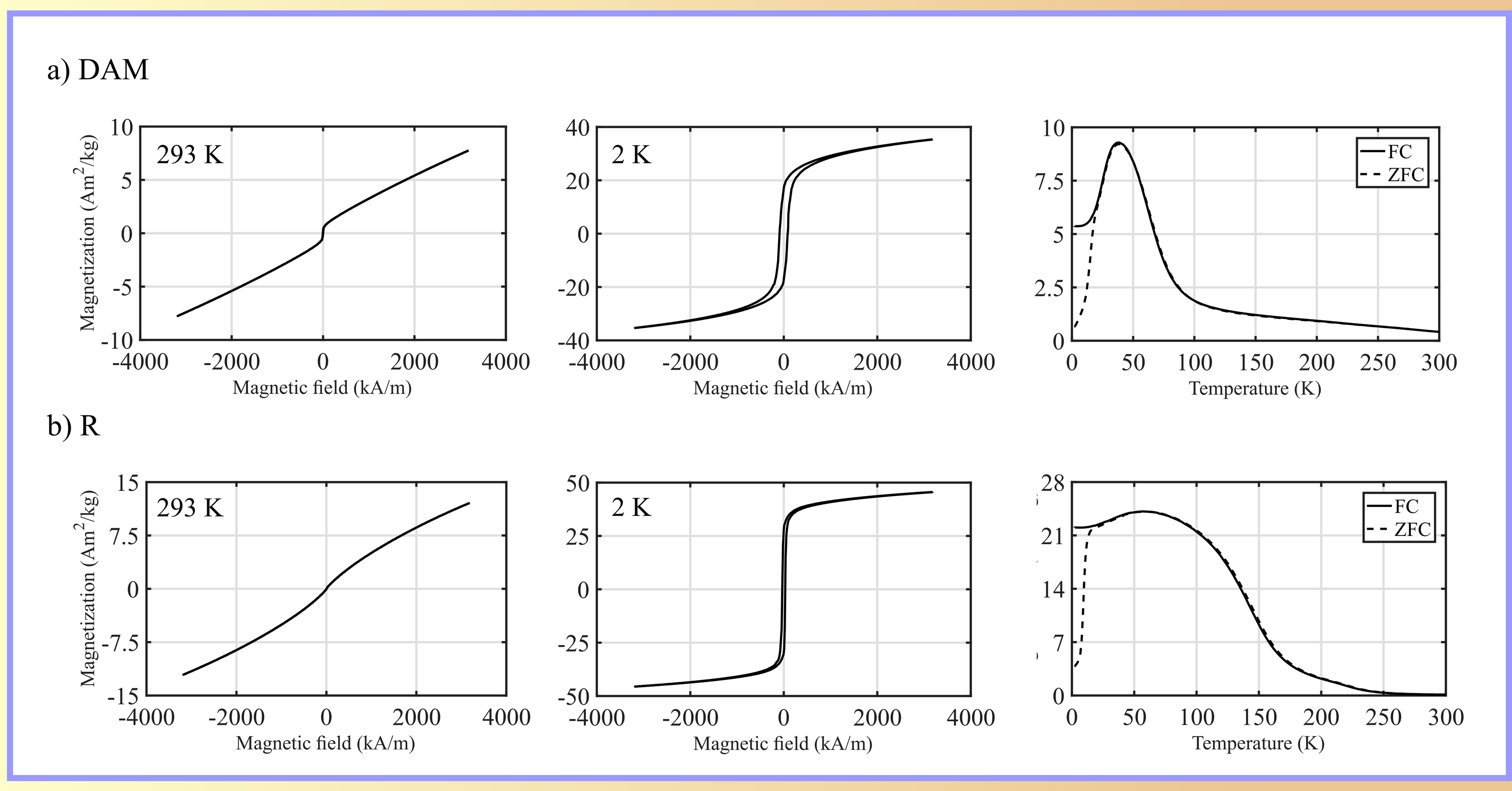


- position of the temperature of re-orientation (T_R ≈ 70 K) practically independent on the type of sample

- hysteresis loops measured below T_R consist of ferromagnetic part at lower magnetic fields followed by linear increase of magnetization indicating anti-ferromagnetic coupling of Mn atoms

- at 2 K hysteresis loops spontaneous magnetization was estimated to 53 Am^2/kg (1.85 μ_B) for all samples, while high field magnetic susceptibility changes slowly from $1.3\cdot10^{-6}$ m^3/kg at 50 K to $1.4\cdot10^{-6}$ m^3/kg at 2 K

Fe_2MnAl alloys



- XRD confirmed the presence of $L2_1$ cubic phase in all samples with lattice constant 0.582(1) nm practically independent on production technology

- conversely, stronger dependence of magnetic properties on preparation techniques than in the case of Fe_2MnSi alloys

- hysteresis loops at 293 K indicate paramagnetic behaviour of samples together with weak magnetization reversal more precisely seen for DAM

sample type	T (K)	$M5$ (Am^2/kg)	M_r (Am^2/kg)	H_c (kA/m)	C (m^3K/kg)	T_c (K)	T_R (K)
DAM	293	7.75	0.21	3.52	$2.82\cdot10^{-4}$	136.90	38.12
	2	35.34	16.93	87.16			
R	293	12.05	0.02	1.63	$1.60\cdot10^{-4}$	263.37	58.64
	2	45.66	28.56	32.12			

- high-temperature loops up to 573 K consisting of weak magnetization reversal at low magnetic fields and paramagnetic contribution at higher fields used for estimation of Curie temperature using the Curie-Weiss law

- both low-temperature magnetic transitions (T_c, T_R) markedly different for R and DAM samples, see table and FC-ZFC curves

- different magnetic behaviour of samples below T_R contrary to Fe_2MnSi

- spontaneous magnetization and high field magnetic susceptibility estimated from 2 K hysteresis loops are 28.1 Am^2/kg (0.97 μ_B) and $2.23\cdot10^{-6}$ m^3/kg for DAM and 39.5 Am^2/kg (1.37 μ_B) and $1.63\cdot10^{-6}$ m^3/kg for R

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