Mössbauer study of the Heusler-type Fe$_2$MAl compounds for M = V, Cr, Fe, Co, Ni

Eustachy S. Popiel,
Wiesława Zarek,
Marek Tuszyński

Abstract The crystal structure and magnetic properties of Fe$_2$MAl compounds for M = V, Cr, Fe, Co and Ni were investigated by X-ray, magnetostatic and Mössbauer effect methods. It was found that the investigated compounds possess not perfectly ordered fcc structure of the L2$_1$-type. With increasing number of $3d$ electrons of M component the transition from the paramagnetic to ferromagnetic behaviour follows. Mössbauer spectra were analysed by a hyperfine field distribution method. They are characteristic of the ferromagnetic phase for M = Fe, Co and Ni and indicated the early observed preference of these atoms for the Fe sites.

Key words Heusler alloys • Mössbauer spectroscopy • site preference • magnetostatic measurements

Introduction

Heusler-type alloys are cubic ternary compounds with the formula X$_2$YZ, where X and Y are two different transition metals and Z is one sp element.

They can crystallize in ordered fcc cubic structure of the L2$_1$-type. Three elements X, Y, Z occupy crystalographically non-equivalent positions [12]. Figure 1 shows the unit cell of such structure.

These alloys have been widely investigated because they exhibit very interesting magnetic and electric properties [1, 2, 7–11, 13, 16, 17]. Iron based transition metal alloys show a great sensitivity to effect of local environment [1, 6, 7, 11, 14, 15]. The tendency for partial disordering of the crystal structure is often observed [7, 8, 11, 14, 15].

In this work we present results of X-ray (XRD), magnetostatic and Mössbauer investigations of the Heusler-type compounds Fe$_2$MAl with M = V, Cr, Fe, Co and Ni.

Experimental

Polycrystalline Fe$_2$MAl compounds with M = V, Cr, Co and Ni were obtained by the Czochralski method from a levitated melt using high purity starting materials. After melting, the ingots were powdered to a particle size less than 30 μm and vacuum annealed to obtain the L2$_1$-type structure. Annealing procedure was similar to that presented in Ref. [2]. XRD measurements of all samples was performed at room temperature using a Siemens D5000 X-ray diffractometer and Cu K$_\alpha$ radiation.

The magnetic measurements of the investigated samples were performed at a temperature up to 800 K in

E. S. Popiel, W. Zarek, M. Tuszyński[✉]
Institute of Physics,
University of Silesia,
4 Uniwersytecka Str., 40-007 Katowice, Poland,
Tel.: +48 32/ 359 13 51, Fax: +48 32/ 258 84 31,
e-mail: tuszynsk@us.edu.pl
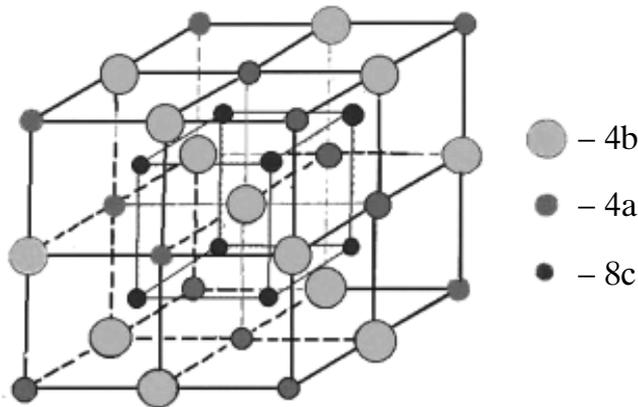
Fig. 1. The unit cell of the L2$_1$-type structure [12].

Fig. 2. Magnetization of the Fe$_2$FeAl, Fe$_2$CrAl and Fe$_2$VAl (insert) compounds *vs.* temperature measured in H = 0.05 T.

the magnetic field up to 12 kOe (1.2 T) using the Faraday method. The Curie temperature (T_C) for the investigated compounds was determined from measurements of the magnetization against the temperature in magnetic fields up to 0.1 T and also from Arrott plots. The saturation magnetization at a given temperature was estimated by extrapolation of the magnetization *vs.* reciprocal magnetic field intensity H to H^{-1} = 0.

The Mössbauer measurement were performed at room temperature using a constant acceleration spectrometer with a ^{57}Co:Pd source. These Mössbauer spectra were calculated by means of a discrete analysis (a single line and a few Zeeman sextets) and a hyperfine magnetic field distribution using the Hesse-Rübartsch procedure [3].

Results and discussion

XRD measurements showed that all investigated compounds were single phase and exhibited the fcc structure of the L2$_1$-type. Detailed analysis of intensity of the superstructure Bragg reflections showed partial atomic disorder of the crystal structure and a preference of the Co and Ni atoms to occupy the 8c sites. Results of crystallographic and magnetic investigations are presented in Table 1.

The temperature variations of magnetization for selected investigated compounds are presented in Fig. 2.

Magnetic studies showed that the Fe$_2$VAl compound above 4.2 K does not exhibit a magnetic transition. For the Fe$_2$CrAl compound, the magnetic transition extends over a wide temperature range. It was found that it disappears with increasing magnetic field intensity. The temperature variation of magnetization for the Fe$_2$FeAl compound is

typical for ferromagnets. Figure 3 shows magnetic isotherms at room temperature for the investigated compounds.

The Fe$_2$CrAl compound achieves the magnetic saturation at much lower magnetic field than the remaining compounds. This allows to suppose that this compound possesses a negligible magnetic anisotropy. The compounds with M = Fe, Co and Ni saturated in magnetic fields higher than 5 kOe (0.5 T). A detailed analysis of the magnetic isotherms showed that the approach to magnetic saturation is mainly determined by magnetic inhomogeneities connected with the atomic disorder of the crystal structure [5].

Figure 4 shows the Mössbauer absorption spectra and hyperfine field distributions for the investigated compounds.

The Fe$_2$VAl compound is a paramagnet at room temperature and its Mössbauer spectrum shows only a single broad absorption line. The Mössbauer spectrum of the Fe$_2$CrAl compound indicates the coexistence of paramagnetic and weak ordered ferromagnetic phases. Similar behaviour for that compound with the B2-type structure was also observed [7]. The hyperfine field distribution of this compound is broad and it indicates a partial atomic disorder of the crystal structure. A wide temperature range of the magnetic transition, observed for the Fe$_2$CrAl compound, (Fig. 2) and reaching the magnetic saturation in a weak magnetic field (Fig. 3) allowed to suppose the presence of magnetic clusters of Fe atoms with large magnetic moments (10^4 μ_B). From this reason, the
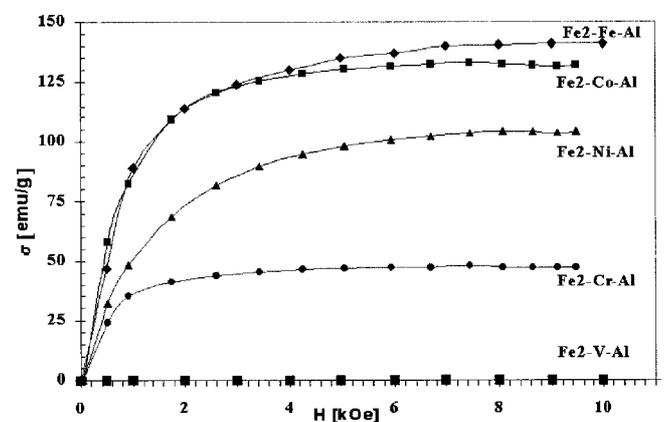
Table 1. Lattice constant a, magnetic moment μ at 300 K and the Curie temperature T_C for the investigated compounds.

Compounds	a [Å]	μ [μ_B/f.u.]	T_C [K]
Fe$_2$VAl	5.760 ± 0.005	Paramagnetic above 4.2 K	
Fe$_2$CrAl	5.791 ± 0.005	1.67 ± 0.01	330 ± 5
Fe$_2$FeAl	5.791 ± 0.005	5.00 ± 0.01	755 ± 2
Fe$_2$CoAl	5.766 ± 0.005	4.91 ± 0.01	>800
Fe$_2$NiAl	5.741 ± 0.005	4.00 ± 0.01	>800

Fig. 3. Magnetic isotherms at room temperature for the investigated compounds.
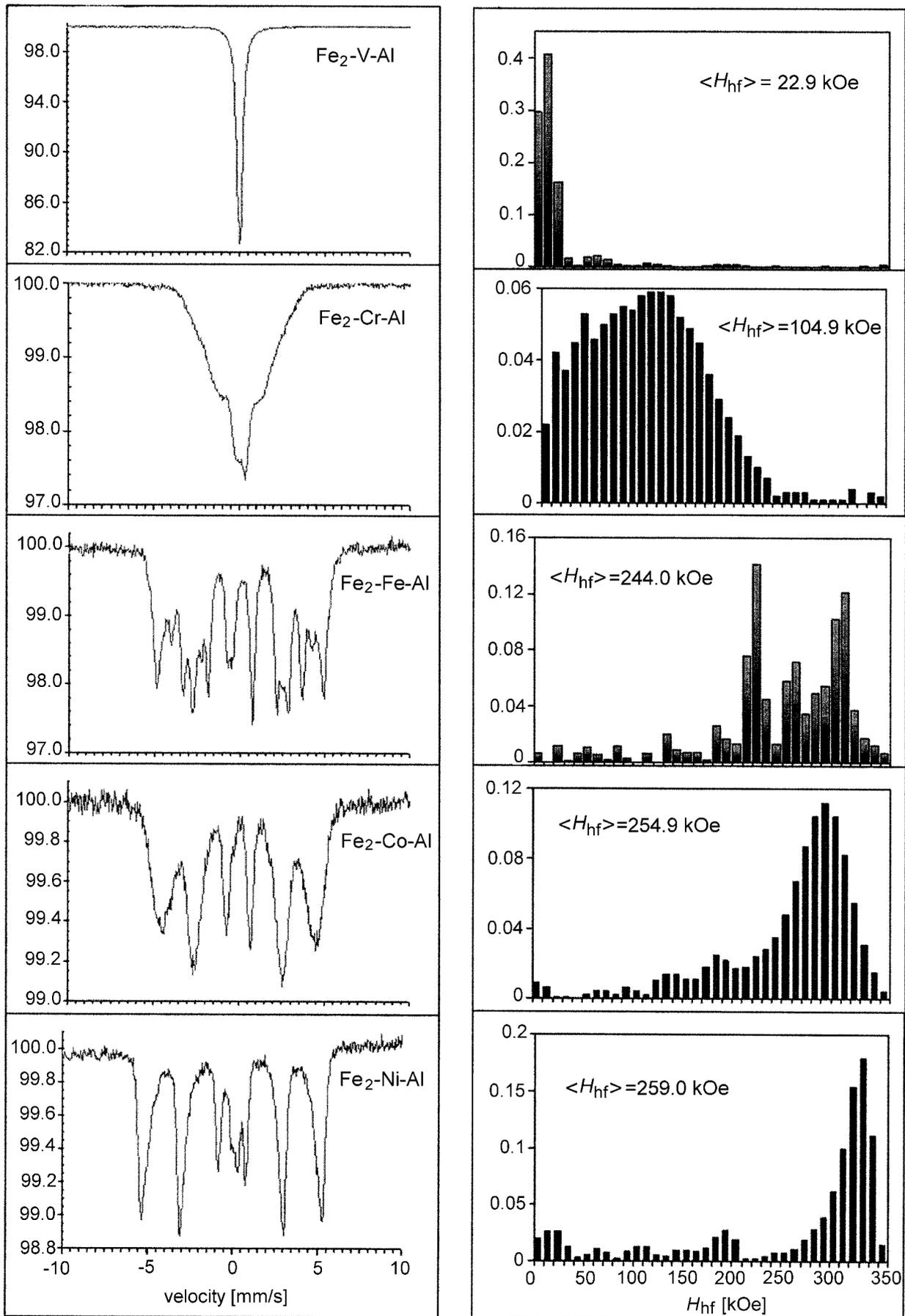
Fig. 4. Mössbauer spectra and hyperfine field distributions for the Fe$_2$MAl with M = V, Cr, Fe, Co and Ni.
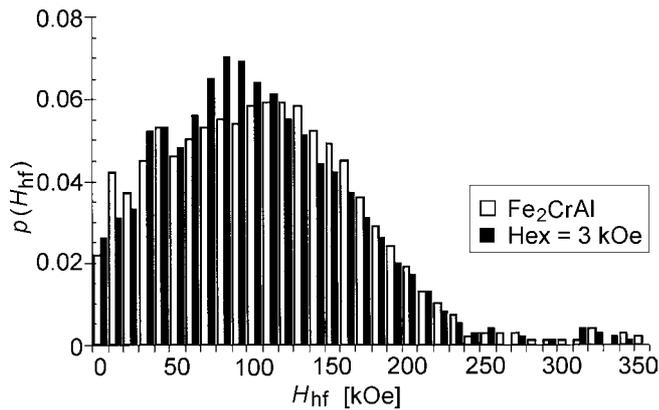
Fig. 5. The hyperfine field distributions for the Fe$_2$CrAl compound in the presence (open bars) and absence (full bars) of the external magnetic field.

Mössbauer measurement with an external magnetic field $H = 3$ kOe perpendicular to gamma rays was done at room temperature. Figure 5 shows a comparison of the hyperfine field distributions in the presence and absence of the external magnetic field for the Fe$_2$CrAl compound. As can be seen from this picture, the external magnetic field practically does not change the hyperfine field distribution. It means that the magnetic properties of this compound are not connected with Fe magnetic clusters.

The ferromagnetic properties of the compounds Fe$_2$MAl with M = Fe, Co and Ni suggested by magnetic measurements were confirmed by Mössbauer spectra (Fig. 4). Only for the Fe$_2$NiAl compound, a weak contribution of the paramagnetic phase – a single line – was observed. The hyperfine field distribution of the ferromagnetic compounds points to a partial atomic disorder of the crystal structure. In an ideal ordered L2$_1$-type structure, the Fe(I) atom at site 8c has 4 Al at sites 4a and 4 M atoms at sites 4b as near neighbours and it possesses a low magnetic moment. In this situation, only the Zeeman sextet with H_{hf} about 220 kOe should be observed. The exchange of an Fe atom with Al or M atoms causes that the Fe(II) atom at site 4a (4b site) is surrounded by 7, 8 Fe(I) atoms and possesses a high magnetic field ($H_{hf} = 315$–330 kOe).

In the Fe$_2$FeAl compound, the partial atomic disorder causes the hyperfine field distribution for Fe sites 4a and 8c (Fig. 4). As can be seen from Fig. 4 for the Fe$_2$NiAl and Fe$_2$CoAl compounds, the amount of Fe atoms at sites 8c decreases, and on the diagram of the hyperfine field distribution the contribution of a high H_{hf} component increases. It means that Co and Ni atoms prefer the site 8c. Such tendency was early observed for ternary Fe$_{3-x}$M$_x$Y alloys with Y = Al, Si and Ga, M = transition metals [4, 6–8, 14, 15].

Investigations of Heusler-like Fe$_2$MAl compounds with L2$_1$-type structure performed in this work show partial atomic disordering of the crystal structure and confirmed early observed site preference of the M atoms. With increasing number of 3d electrons of M component the transition from the paramagnetic to ferromagnetic behaviour follows.

References

1. Athanassiadis G, Le Caer G, Foct J, Rimlinger L (1964) Study of ternary ordered solid solutions derived from Fe$_3$Al by substitution. Phys Status Solidi A 40:425–435
2. Buschow KHJ, van Engen PG (1981) Magnetic and magneto-optical properties of Heusler alloy based on aluminium and gallium. J Magn Magn Mater 25:90–96
3. Frąckowiak J (1990) Mössbauer effect study of ordered and disordered Fe$_{(3-x)}$Al$_{(x)}$ (0 < x < 0.5) alloys. Hyperfine Interact 54:793–797
4. Fultz B, Gao Z-Q, Hamdeh HH, Oliver SA (1994) Local and nonlocal isomer shifts in bcc Fe-X alloys (X = Al, Si, Ga, Ge). Phys Rev B 49:6312–6315
5. Grössiger R (1981) A critical examination of the law of approach to saturation. Phys Status Solidi A 66:665–675
6. Grover AK, Pillay RG, Nagarajan V, Tandon PN (1980) Site preference and local environment effects in ferromagnetic ternary. J Magn Magn Mater 15/18:699–700
7. Lakshmi N, Venugopalan K, Varma J (2002) Mössbauer studies of hyperfine-field in disordered Fe$_2$CrAl. Pramana J Phys 59:531–537
8. Niculescu VA, Burch TJ, Budnick JJ (1983) A local environment description of hyperfine fields an atomic moments in Fe$_{3-x}$T$_x$Si alloys. J Magn Magn Mater 39:223–267
9. Nishino Y, Inoke S, Asano S, Kawaiya N (1993) Anomalous temperature dependence of the electrical resistivity in binary and pseudobinary alloys based on Fe$_3$Si. Phys Rev 48:13607–13613
10. Nishino Y, Kato M, Asano S, Soda K, Hayashi M, Mizutani K (1997) Semiconductor-like behaviour of electrical resistivity in Heusler-type Fe$_2$VAl compound. Phys Rev Lett 79:1909–1911
11. Okpalugo DE, Both JG, Faunce CA (1985) Onset of ferromagnetism in 3d-substituted FeAl alloys. J Phys F: Metal Phys 15:681–692
12. Pearson WB (1967) A handbook of lattice spacing and structure of metals and alloys. Vol 2. Pergamon, London
13. Popiel ES, Zarek W, Tuszyński M, Rendecki T (1989) Investigation of Fe$_{3-x}$V$_x$Al alloys with DO$_3$-type structure by X-ray, magnetostatic and Mössbauer effect methods. J Les-Comm Met 149:127–135
14. Satuła D, Dobrzyński L, Malinowski A, Szymański K, Waliszewski J (1995) Hyperfine fields, local environment effects and site preference in Fe$_{3-x}$Cr$_x$Al alloys. J Magn Magn Mater 151:211–220
15. Szymański K, Biernacka M, Dobrzyński L (2000) Mössbauer and magnetic studies of Fe$_{3-x}$Co$_x$Al. J Magn Magn Mater 210:150–162
16. Webster PJ, Ziebeck KRA (1983) The paramagnetic properties Heusler alloys containing iron. Phys Lett A 98:51–53
17. Webster PJ, Ziebeck KRA (1988) Magnetic properties of metals. Vol 19c. Springer, Berlin