NUKLEONIKA 2003;48(Supplement 1):S65–S70

Crystal order and magnetic properties of $Fe_{2.4}V_{0.6}Al$ alloy studied by magnetostatic and Mössbauer methods

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Abstract Performed investigations showed that the magnetic properties of the $Fe_{2.4}V_{0.6}Al$ alloy markedly depend on the degree of atomic order of its cubic structure. The atomically disordered alloy with A2 (bcc) type structure exhibits ferromagnetic properties. Its Mössbauer spectra can be described by the hyperfine field distribution connected with various local environments of Fe atoms. Alloys with B2 (sc) and DO_3 (fcc) type structure do not exhibit magnetic transition above 4.2 K. High values of the magnetization and its strongly non-linear variation with magnetic field intensity in a wide temperature range suggest the presence of magnetic iron clusters in these alloys. Superparamagnetic relaxation times of the order of 10^{-9} s and 5×10^{-8} s correspond to the largest magnetic clusters with a magnetic moment of 4×10^3 μ_B in B2 and 10^4 μ_B in DO_3-type structure, respectively. Mössbauer spectra of these alloys confirm lack of the magnetic order and also suggest the presence of the Fe magnetic clusters with those relaxation times. It was shown that the increase of atomic order of the crystal structure causes formation of the Fe magnetic clusters and disappearing of the magnetic order.

Key words magnetic clusters • Mössbauer spectroscopy • relaxation time

Introduction

Recent investigations showed [3] that the $Fe_{3-x}V_xAl$ alloys crystallize in the cubic structure and, depending on the vanadium concentration and mechanical or thermal treatment, can exhibit a different atomic order which leads to bcc (A2), sc (B2) and fcc (DO_3) types of structure. It was also shown that magnetic properties of these alloys markedly depend on the degree of atomic order [3, 5, 6].

In this work we present the results of investigations of the influence of atomic order on the magnetic properties of the $Fe_{2.4}V_{0.6}Al$ alloy with cubic structure performed by magnetostatic and Mössbauer effect methods. From recent investigations [3, 5, 6] it follows that the $Fe_{2.4}V_{0.6}Al$ alloy can possess three different types of atomic order – A2 (bcc), B2 (sc) and DO_3 (fcc).

The $Fe_{3-x}V_xAl$ alloys with B2-type structure do not exhibit ferromagnetic properties for $x \geq 0.7$, while for $x \geq 1.0$ the B2-type structure cannot be obtained.

Hence, the $Fe_{2.4}V_{0.6}Al$ alloy is a very interesting object for investigating the influence of the atomic order and simultaneously of the local environment of the Fe atom on magnetic properties of the $Fe_{3-x}V_xAl$ alloys.

Experimental

The polycrystalline $Fe_{2.4}V_{0.6}Al$ alloy was obtained by arc melting of stoichiometric quantities of Fe, V and Al in an argon atmosphere. After melting the ingots were powdered to a particle size less than 30 μm and divided into three parts. Two parts of the sample were annealed in the

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Received: 17 July 2002, Accepted: 7 March 2003

temperature range 360÷500 K or 600÷850 K to obtain the B2 and DO_3-type structure, respectively. X-ray measurement of all samples was performed at room temperature using a Siemens X-ray diffractometer and Cu K_α radiation.

The magnetic measurements of the investigated samples were performed at temperatures up to 600 K in the magnetic field up to 12 kOe (1.2 T) using the Faraday method.

The Mössbauer measurement were performed in the temperature range from 20 to 400 K using a constant acceleration spectrometer with a ^{57}Co:Pd source and a closed cycle helium refrigerator. These Mössbauer spectra were calculated by means of a hyperfine magnetic field distribution using the Hesse-Rübartsch procedure [2], and this method of calculations gave an excellent description of the shapes of measured spectra. Some spectra (for B2 and DO_3-type structure alloys) were calculated by the hyperfine field fluctuation method of Blume and Tjon [4].

Results and discussion

X-ray measurement showed that all the powdered samples of the $Fe_{2.4}V_{0.6}Al$ alloy were single phase and their crystalline structure depended on thermal treatment after powdering. The powdered sample without any thermal treatment showed an atomically disordered bcc structure of A2-type with a lattice constant of 2.92 Å. It was also found that the A2-type structure of the powdered alloy is very unstable – annealing of this alloy at a temperature of about 420 K causes the phase transition of disorder–order type correlated with changes of its magnetic properties.

The powdered alloy annealed in the earlier mentioned temperature range exhibits an atomically ordered structure of B2 or DO_3-type with a lattice constant of 2.89 and 5.77 Å, respectively.

Magnetic studies showed that the alloy with the A2-type structure behaves as a typical ferromagnet with the Curie temperature about 420 K and the mean magnetic moment of the Fe atom $\mu_{Fe} = 1.42\ \mu_B$ at 80 K.

Alloys with the B2 and DO_3-type structure do not exhibit magnetic transition above 4.2 K but they are characterized by high values of magnetization and a strong nonlinear variation of magnetization with a magnetic field (Fig. 1) in the wide temperature range. This indicates the presence of Fe magnetic clusters with a large magnetic moment in these alloys. These clusters provide a superparamagnetic behavior of the investigated alloys.

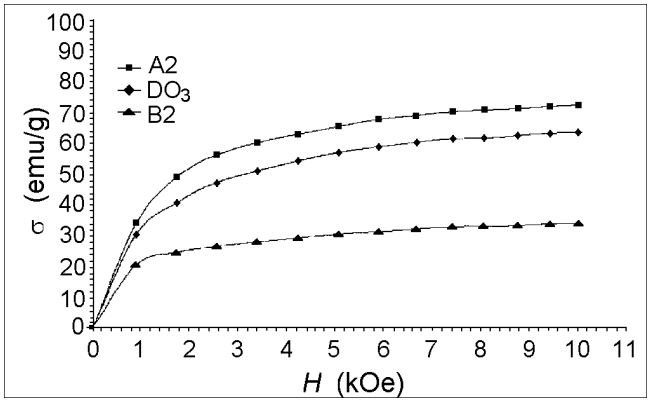

Fig. 1. The magnetizations isotherms for $Fe_{2.40}V_{0.60}Al$ alloy with the A2, B2 and DO_3-type structure at 80 K.

The magnetization σ of the investigated alloys was expressed as a function of the magnetic field H and temperature T by the Brillouin function $B_J(\alpha)$

$$\sigma(H, T) = \sigma_0 \cdot B_J(\alpha) \tag{1}$$

where: $\alpha = \mu H/kT$, μ is the magnetic moment of clusters; σ_0 is the saturation magnetization of the magnetic clusters.

From analysis of the magnetization isotherms the magnetic moment of the magnetic clusters was determined. It was found that the magnetic moment of the largest clusters is about $4 \times 10^3\ \mu_B$ and $10^4\ \mu_B$ in the alloy with B2 and DO_3-type structure, respectively. Assuming that the superparamagnetic relaxation time can be expressed by:

$$\tau = 10^{-9} \exp\left(\frac{KV}{kT}\right) \tag{2}$$

where K is the anisotropy constant for pure Fe and V is the volume of the magnetic cluster, the relaxation times $t = 10^{-9}$ s and $t = 5 \times 10^{-8}$ s at room temperature for the earlier mentioned magnetic clusters were estimated.

The mean magnetic moment of the Fe atom in an alloy with the B2 and DO_3-type structure $<\mu_{Fe}> = 0.8$ and $1.19\ \mu_B$, respectively, is significantly lower than for the alloy with the A2-type structure.

The Mössbauer absorption spectra and diagrams of hyperfine field distribution (H_{hf}) for an alloy with the A2, B2 and DO_3-type structure measured in the temperature range 20÷400 K are shown in Figs. 2–4. As can be seen from these figures, the Mössbauer spectra and diagrams of H_{hf} distributions markedly depend on the degree of atomic order of the crystal structure.

The shape of the Mössbauer spectra at room temperature for an alloy with the A2-type structure indicates the existence of magnetic order and a very small contribution of paramagnetic phase which is represented by a single line in the centre of the spectra. The magnetically ordered phase is revealed as superposition of several Zeeman sextets giving a wide hyperfine field distribution with the high mean hyperfine field $<H_{hf}>$.

According to [1] the local magnetic moment of the iron atom and hence also the hyperfine field on the ^{57}Fe nucleus in binary Fe-Al and ternary Fe-V-Al alloys depends on the number n of other iron atoms in the nearest neighbouring shell. Each local configuration of Fe atoms gives a single sextet which leads to a wide hyperfine field distribution. The wide hyperfine field distributions reflect atomic disorder and statistical distribution of the Fe, V and Al atoms in the crystal lattice. The paramagnetic phase is probably connected with Fe atoms having less than four Fe atoms as the nearest neighbours.

The Mössbauer spectra and hyperfine field distributions for the alloys with the B2 and DO_3-type structure proved the absence of long range magnetic order in these alloys. This confirms the results of magnetic measurements.

The Mössbauer spectrum at room temperature for the B2-type structure alloy (Fig. 3) shows a single broad line. It suggests the presence of superparamagnetic clusters in the alloy. Increasing width of this line with decreasing temperature indicates slowering of the magnetic moment fluctuations of these clusters.

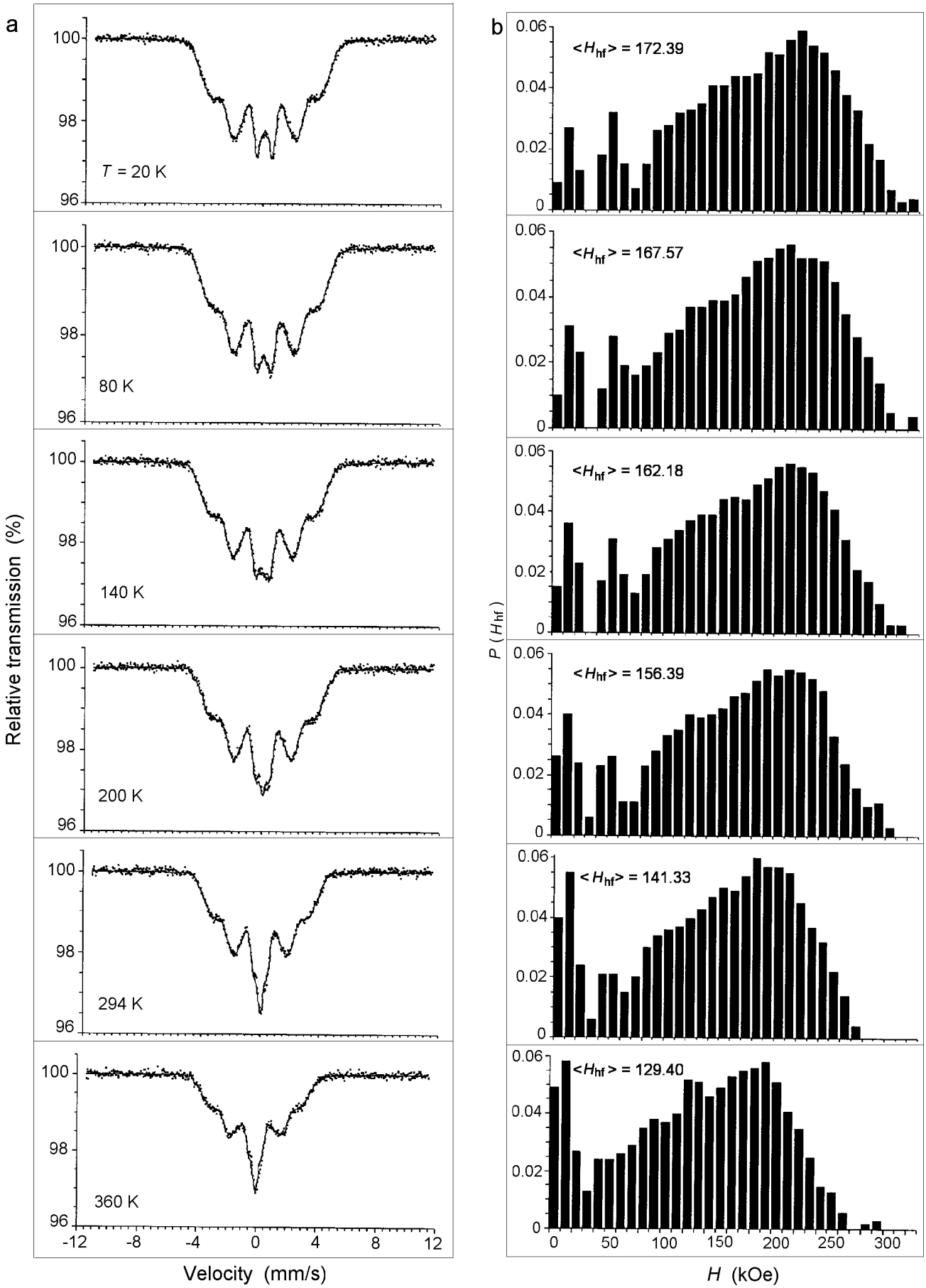

Fig. 2. a – The Mössbauer spectra; b – respective hyperfine field distributions for $Fe_{2.40}V_{0.60}Al$ alloy with the A2-type structure measured in the temperature range 20÷360 K.

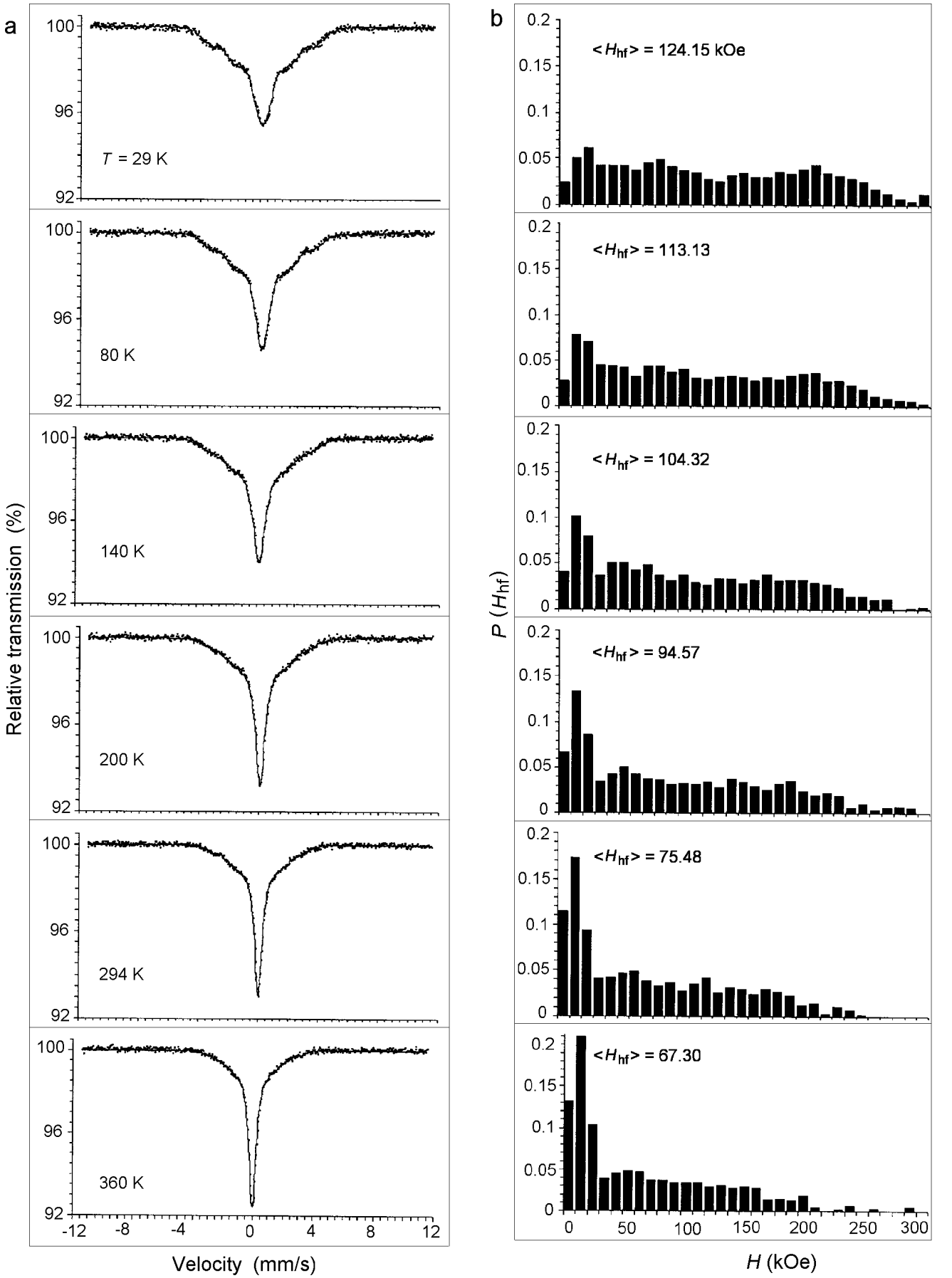

Fig. 3. a – The Mössbauer spectra; b – respective hyperfine field distributions for $Fe_{2.40}V_{0.60}Al$ alloy with the B2-type structure measured in the temperature range 20÷360 K.

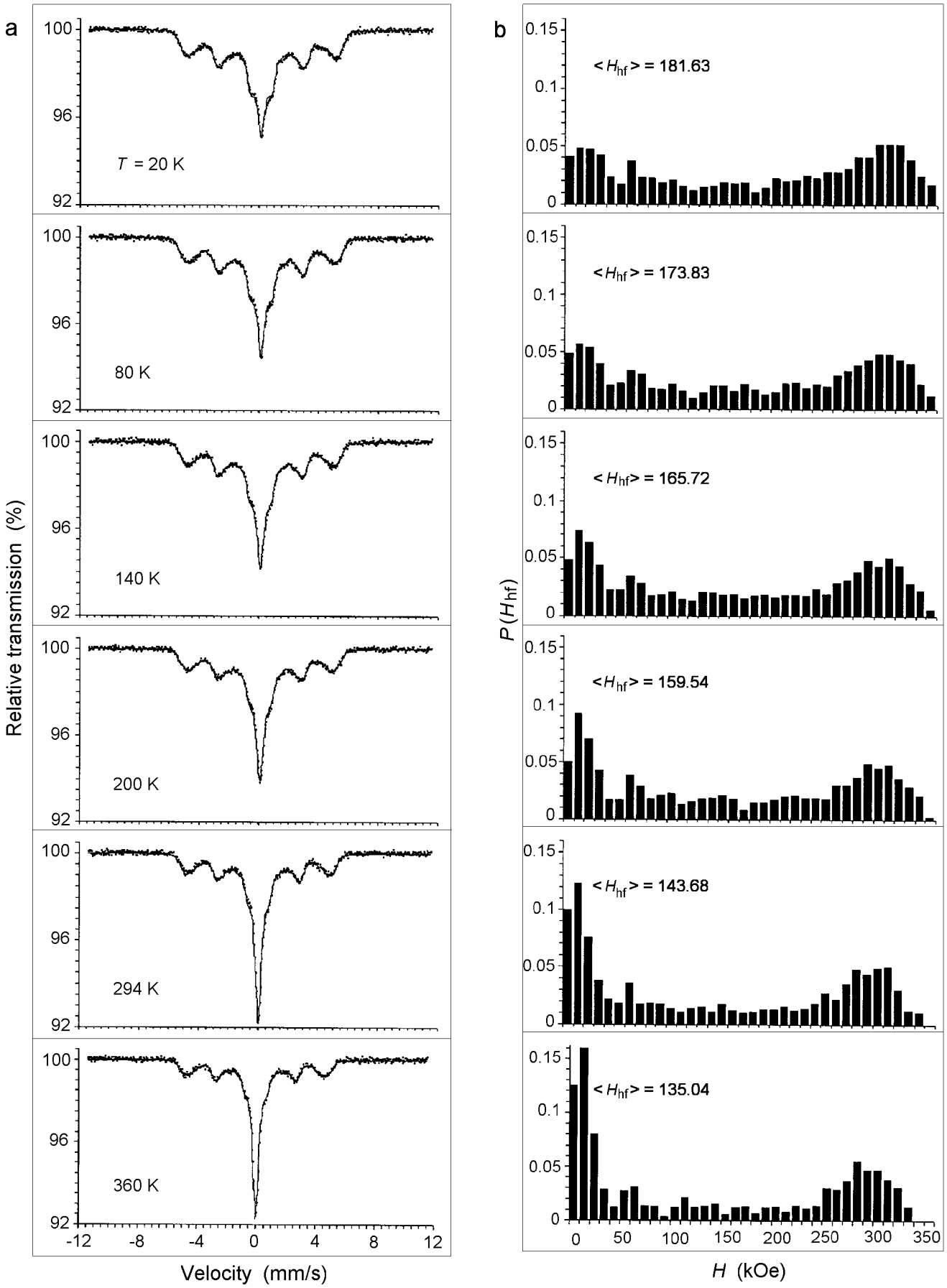

Fig. 4. a – The Mössbauer spectra; b – respective hyperfine field distributions for $Fe_{2.40}V_{0.60}Al$ alloy with the DO_3-type structure measured in the temperature range 20÷360 K.

In the $Fe_{2.4}V_{0.6}Al$ alloy with the B2-type structure, the Fe atom at the centre of cubic cell has 80% of Al and V atoms and 20% Fe atoms as the nearest neighbours. In this situation, the Fe atoms can possess only a low magnetic moment and the alloy can behave as paramagnet or a weak itinerant ferromagnet. However, magnetic investigations of this alloy suggest the superparamagnetic properties associated with the magnetic clusters of iron atoms. They are formed as a result of the local deviations from stoichiometry. It is supposed that the Fe atoms with more than four Fe atoms as the nearest ones and the next nearest neighbours can form magnetic clusters. They are separated by the regions in which the Fe atoms are surrounded only by V and Al atoms. An alloy containing separated magnetic clusters shows superparamagnetic properties. The diagram of the hyperfine field distribution reflects the continuous distribution of magnetic moments of clusters. Analysis of the Mössbauer spectra by the Blume-Tjon method showed the existence of two hyperfine fields ($H_{hf1} \approx 300$ kOe, $H_{hf_2} \approx 200$ kOe) with relaxation times of 5×10^{-9} s and 1.3×10^{-8} s, respectively for the spectrum at 30 K. These times became shorter with increasing temperature.

In the Fe_3Al alloy with the DO_3-type structure the Fe atoms occupy two non-equivalent positions. The Fe(I) atom with four Fe(II) atoms as neighbours has reduced the local magnetic moment (1.5 μ_B), whereas the Fe(II) atom with eight Fe(I) atoms as neighbours has a relatively large local magnetic moment (2.2 μ_B) [1]. According to Ref. [3], in the $Fe_{3-x}V_xAl$ alloys the vanadium atoms prefer the Fe(II) positions what reduces a number of iron atoms as the nearest neighbours of Fe(I) atoms and hence their magnetic moment. These atoms can be paramagnetic. The Fe(II) atoms have still eight iron atoms as the nearest neighbours and can support ferromagnetism in some regions or form ferromagnetic clusters with a large magnetic moment and a relatively long relaxation time. The Mössbauer spectra of DO_3-type alloys (Fig. 4) suggest this possibility. Zeeman sextets with the high hyperfine fields are visible on the Mössbauer spectra independently of temperature. Analysis of these spectra by the Blume-Tjon method gives two relaxation times 5×10^{-8} s and 5×10^{-9} s for $H_{hf} = 300$ kOe. The first value of time is comparable with that obtained from magnetic calculations for magnetic clusters with the magnetic moment of the order of 10^4 μ_B. The second one corresponds to smaller magnetic clusters.

Conclusion

Investigations performed in this work showed that the magnetic properties of the Fe-V-Al alloys strongly depend on the atomic order and the local environment of the Fe atoms. The increase of atomic order destroys the long range magnetic order and causes the formation of Fe magnetic clusters with various magnetic moments.

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