

Mössbauer investigation of some bcc, Fe-based, multicomponent alloys

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Abstract. Mössbauer measurements of a Fe-based, bcc multicomponent alloys with concentration of non-ferrous element $x = (0.1, 0.3 \text{ and } 0.6)$ were performed. The effective thickness parameters and the second order Doppler shifts were determined as a function of temperature. The results indicate an apparent decrease of the recoilless fraction with x . The observed trends are consistent with theoretical results indicating modifications of phonon density of states by bond disorder.

Key words: Debye approximation • effective thickness • Mössbauer spectroscopy • second order Doppler shift

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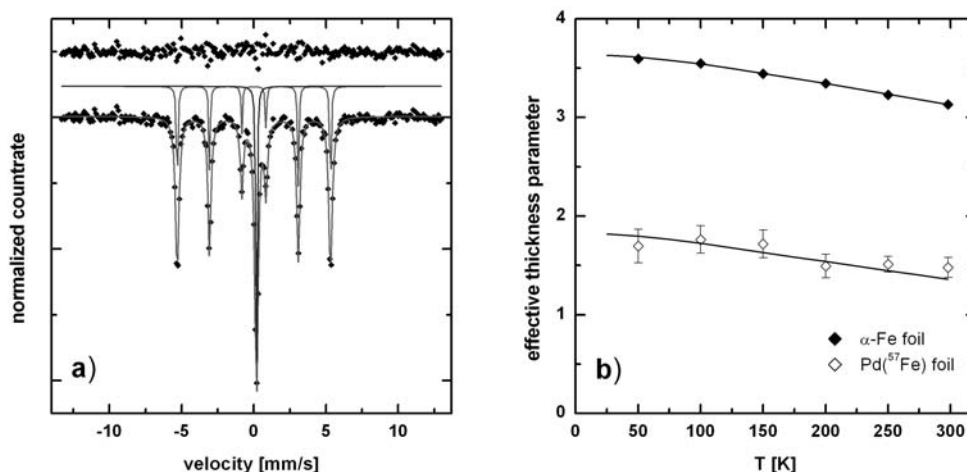
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Introduction

Recently, we have attempted to realize the idea of the so-called an average atom A in a single phase of $\text{Fe}_{1-x}\text{A}_x$ alloy. In an ideal case A is a combination of many elements with constant proportions and x is a control parameter. By changing x , the physical properties of the system could be systematically varied. However, the idea is not easy for realization because different crystallographic phases are usually formed. We have successfully synthesized $\text{Fe}_{1-x}\text{A}_x$, where A is combination of elements (Al, Si, V, Cr) [5, 10]. Performed investigations indicated the monotonic dependence of the hyperfine parameters [10] as well as a linear increase of the residual resistivity [5] as a function of x . These type of alloys served also as a model system with force-constant disorder, studied theoretically within coherent-potential approximation or exact diagonalization technique [7, 11, 14]. The force-constant disorder is a phenomenon important for understanding of dynamics of structurally disordered systems and origin of low frequency lattice excitations observed in these systems [2, and references therein]. The aim of this work was to study parameters related to lattice dynamics and hyperfine interactions by Mössbauer spectroscopy in some bcc multicomponent alloys.

Table 1. Nominal compositions of investigated alloys

| Average atom concentration x | Composition |
|--------------------------------|---|
| 0.1 | $\text{Fe}_{0.9000}\text{Al}_{0.0125}\text{Si}_{0.0125}\text{V}_{0.0125}\text{Cr}_{0.0125}\text{Co}_{0.0125}\text{Ni}_{0.0125}\text{Ga}_{0.0125}\text{Ge}_{0.0125}$ |
| 0.3 | $\text{Fe}_{0.7000}\text{Al}_{0.0375}\text{Si}_{0.0375}\text{V}_{0.0375}\text{Cr}_{0.0375}\text{Co}_{0.0375}\text{Ni}_{0.0375}\text{Ga}_{0.0375}\text{Ge}_{0.0375}$ |
| 0.6 | $\text{Fe}_{0.400}\text{Al}_{0.075}\text{Si}_{0.075}\text{V}_{0.075}\text{Cr}_{0.075}\text{Co}_{0.075}\text{Ni}_{0.075}\text{Ga}_{0.075}\text{Ge}_{0.075}$ |

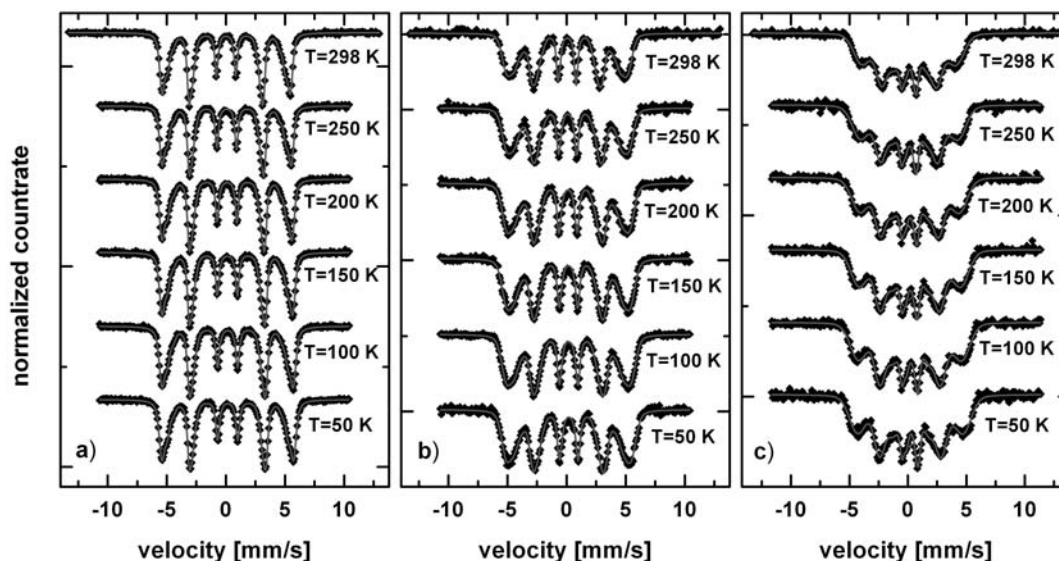
**Fig. 1.** (a) Mössbauer spectrum of joined α -Fe and ^{57}Fe enriched Pd foils measured at room temperature; (b) effective thickness parameter obtained for both foils (points). Solid lines are related with the Debye model approximation.

Experiment

The polycrystalline alloys $\text{Fe}_{1-x}\text{A}_x$, where A is the so-called average atom composed of a relatively large number of elements $A = \text{Al, Si, V, Cr, Co, Ni, Ga}$ and Ge and concentration $x = 0.1, 0.3$ and 0.6 were obtained. The nominal compositions of obtained samples are listed in Table 1. The elements were initially melted in an arc furnace to achieve homogeneous ingots. At the end of the preparation process, the rapid quenching technique was used to prevent phase separation. The structure of all obtained alloys, checked by X-ray spectroscopy, exhibits a single bcc phase.

Mössbauer spectra were collected at standard spectrometer working in constant acceleration mode. The closed cycle He refrigerator was used to perform low temperature measurements.

One aim of the studies was the investigation of temperature dependence of the effective thickness. The effective thickness was determined by the internal standard method [9]: the absorber is measured together with the reference standard of known thickness. Palladium foil with the alloyed ^{57}Fe isotope was chosen as the internal standard. Measurements of a natural α -Fe foil (of thickness about $8 \mu\text{m}$) and the palladium absorber at different temperatures were performed, see an example in Fig. 1a. By fitting a sextet with known temperature dependence of thickness [8], and a singlet in transmission integral approximation, thickness of the singlet was determined, see Fig. 1b. The solid curves shown in the graph represent temperature dependence of the thickness parameter calculated from the Debye approximation, with the Debye temperatures of $\Theta_D = 435 \text{ K}$ and 335 K for the iron and palladium foil, respec-

**Fig. 2.** Mössbauer measurements results for alloys with average atom concentration x equal to (a) 0.1, (b) 0.3 and (c) 0.6.

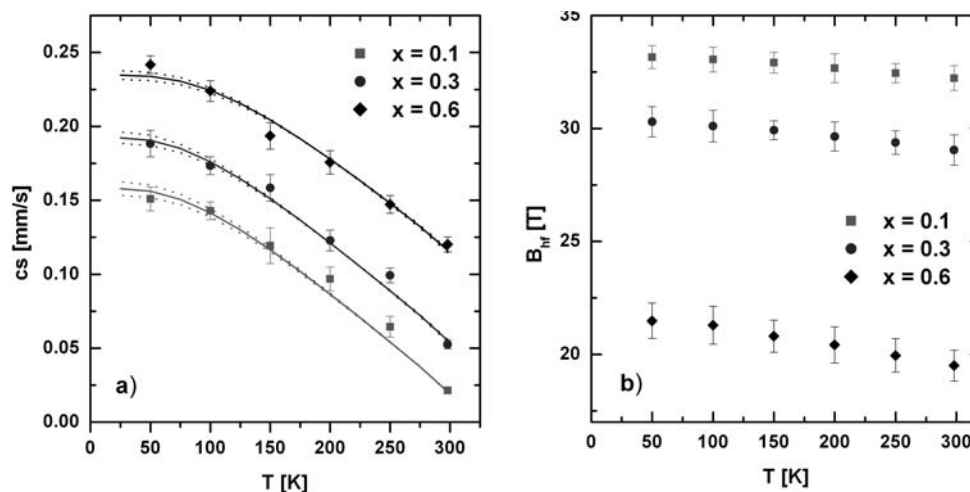


Fig. 3. Temperature dependencies of hyperfine parameters (a) centre shift and (b) hyperfine magnetic field, for investigated materials. Lines are related with the Debye model approximation: solid ones – theory with best fit value of Debye temperature Θ_{SOD} , dotted ones – denotes possible variation of the Debye temperature Θ_{SOD} for each sample.

tively. Obtained value is in good agreement with this reported in [4] for iron impurity in Pd.

The Mössbauer spectra of absorbers with average atom concentration $x = 0.1, 0.3$ and 0.6 , are shown in Fig. 2. The absorbers were also measured together with a palladium standard (spectra not shown).

Results

The shapes of the spectra with apparently broadened absorption lines were fitted by set components with Gaussian distribution of hyperfine magnetic field. A zero electric field gradient was assumed. The line width and the ratio of the intensities of the second and the third lines in sextets were kept constant for all temperatures. Results of the best fits to the spectra are presented in Fig. 2, while temperature dependence of the centre shift (cs) and the average hyperfine magnetic field B_{hf} are shown in Fig. 3. The distribution of the hyperfine fields determined for spectra shown in Fig. 2 were used in the analysis of the spectra measured with the internal standard. From these fits effective thicknesses were extracted and are shown in Fig. 4.

Temperature dependence of a centre shift is mainly due to the second order Doppler (SOD) shift, which is proportional to the average square of the velocity of thermal vibrations [6]. Assuming the Debye model, curves related to a SOD shift were fitted to the data, see the solid lines in Fig. 3. The Debye temperatures, derived from the fitted curves are $\Theta_{\text{SOD}} = (352 \pm 25)$ K, (354 ± 25) K and (453 ± 25) K for samples with average atom concentration $x = 0.1, 0.3$ and 0.6 , respectively. The Debye temperature for iron ($x = 0$) determined from the SOD shift, is 419 K [9]. The obtained data do not show clear dependence of Θ_{SOD} on the concentration x . The obtained large value of Θ_{SOD} value for a sample with $x = 0.6$ is not clear. We note that the SOD values have relatively large uncertainties because the variety of different elements caused spectra broadening.

Effective thickness is proportional to the recoilless fraction and this quantity is related to the average square of the amplitude of thermal vibration. The Debye approximation was used for the estimation of Debye temperatures resulting in $\Theta_f = (373 \pm 13)$ K, (336 ± 13) K and (316 ± 13) K for the samples with average atom concentration $x = 0.1, 0.3$ and 0.6 , respectively. The Θ_f for iron ($x = 0$) is 435 K [9]. A clear decrease of Θ_f with x is observed.

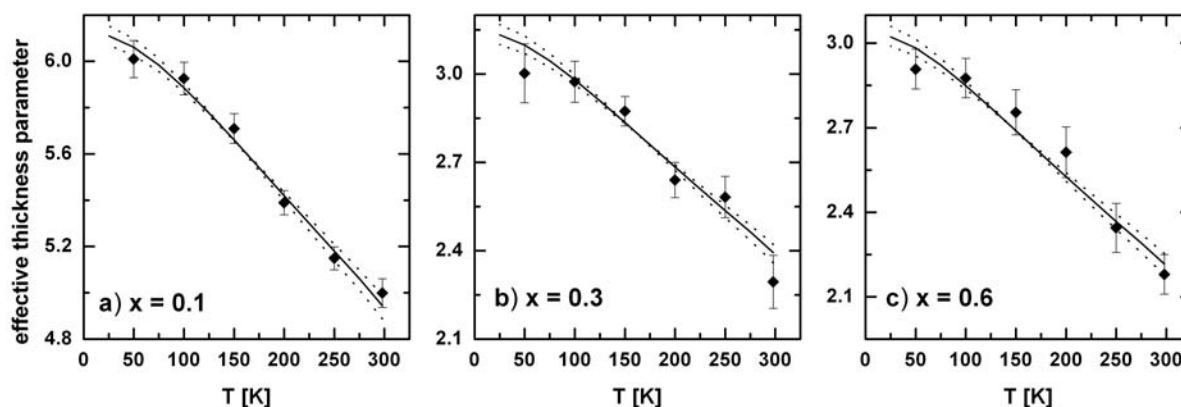


Fig. 4. Temperature dependence of effective thickness parameter (points) obtained for samples with (a) $x = 0.1$, (b) $x = 0.3$ and (c) $x = 0.6$. Lines are related with the Debye model approximation: solid ones – theory with best fit value of Debye temperature Θ_f , dotted ones – denotes possible variation of the Debye temperature Θ_f for each sample.

Discussion

One expects that in the bcc structure every Fe atom is surrounded by eight atoms, located at corners of the elemental cubic unit cell. Because of chemical disorder, hyperfine magnetic field distribution is observed as broad lines of the Zeeman sextets. The mean values of the hyperfine magnetic field decreases with increasing average atom concentration x , while the width of the distribution increases (not shown). This behaviour is consistent with increase of the average number of atoms with magnetic moment close to zero in the first coordination shell of Fe. In contrast to earlier studies, we do not observe features characteristic of short range order effects [9]. We think that this is the result of many elements forming the alloys and rapid cooling during the sample preparation.

Disorder in force constant modify the phonon density of states (DOS) $g(\omega)$. Observed discontinuity of the DOS function (so-called critical point or van Hove singularity) [13], with the extreme value of frequency ω , of the reference crystal (in our case iron) is shifted downwards in frequency due to level-repelling and hybridization effects [7, 11]. Average square of the amplitude of atomic displacement $\langle x^2 \rangle$ and velocity $\langle v^2 \rangle$ at a temperature of T in harmonic crystal approximation is [3, 12]:

$$(1) \quad \langle x^2 \rangle = \frac{\hbar}{2m} \int \frac{1}{\omega} \coth\left(\frac{\hbar\omega}{2k_B T}\right) g(\omega) d\omega$$

$$(2) \quad \langle v^2 \rangle = 3\langle v_x^2 \rangle = \frac{3\hbar}{2m} \int \omega \coth\left(\frac{\hbar\omega}{2k_B T}\right) g(\omega) d\omega$$

When the density of states $g(\omega)$ is modified and additional density of the states appear at low frequencies, one expects that $\langle x^2 \rangle$ is disturbed more than $\langle v^2 \rangle$ because $g(\omega)$ is weighted by $1/\omega$ in Eq. (1) while by ω in Eq. (2). Therefore, $\langle x^2 \rangle$ is more sensitive to the low energy vibrations than $\langle v^2 \rangle$. Our results seems to be consistent with this picture. Apparent decrease of the Debye temperature Θ_f with increasing average atom concentration x , is most probably related to the

influence of bond disorder on the modification of the phonon density of states in the low energy region.

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