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MAGNETIC PROPERTIES OF $4f-3d$ INTERMETALLICS STUDIED BY ^{161}Dy AND ^{57}Fe MÖSSBAUER EFFECT

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Applications of the Mössbauer effect at the ^{161}Dy and ^{57}Fe nuclei to study heavy rare earth - transition metal compounds are presented. Magnetic hyperfine fields at dysprosium and iron nuclei observed in the $\text{Dy}_u(\text{M}-\text{M})_v$ intermetallics ($u/v = 2/17, 6/23, 1/3$, and $1/2$; $\text{M}-\text{M} = \text{Mn}-\text{Fe}$, $\text{Fe}-\text{Co}$, and $\text{Co}-\text{Ni}$), magnetic moments and magnetic ordering temperatures are discussed. The magnetic hyperfine field at iron nuclei and the field at dysprosium nuclei treated as functions of a number of $3d$ electrons form two different systems of the Slater-Pauling curves. The magnetic ordering temperatures treated as functions of the number of $3d$ electrons also form the u/v system of curves. Considering the magnetic data and the magnetic hyperfine fields it can be concluded that intermetallics can be treated as three-subsystem ferrimagnets. Consequently, a coexistence between the $4f$, $5d$, and $3d$ magnetism in the rare earth - transition metal compounds can be qualitatively explained.

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1. Introduction

The heavy rare earth - transition metal (R-M) intermetallics and the derivative R-M compounds with added boron, or carbon, hydrogen, nitrogen, silicon, are widely studied because of scientific and practical importance. The numerous magnetic properties and the hyperfine interactions of the R-M ferrimagnets were previously described in literature [1-4]. Actually, an influence of the R-sublattice on the magnetic properties and the hyperfine interactions in the R-M intermetallics is relatively well known. On the other hand, an influence of the M-sublattice on the mentioned properties is known only partially [1-7]. The ferrimagnetism of these compounds is related to the coexistence of $4f-3d$ magnetism, which is mediated mainly by the band-type $5d$ electrons [5, 8, 9]. Nevertheless, the electronic band structure of these intermetallics and in particular of their transition metal sublattice is complex and poorly understood yet.

The Mössbauer effect is an especially suitable nuclear method to study separately the R- or M-sublattice of the intermetallics [8, 9]. Often, for this purpose the Mössbauer effect at the ^{161}Dy and ^{57}Fe nuclei is used to test the rare earth and the transition metal sublattice, respectively [8–12].

Both the magnetic and the Mössbauer effect studies were performed previously for a number of the R_uM_v series [1–12]. In order to find the influence of the number of $3d$ electrons on the R and M magnetism, the pseudobinary $\text{R}_u(\text{M}-\text{M})_v$ compounds were studied, where $\text{M}-\text{M} = \text{Mn}-\text{Fe}, \text{Fe}-\text{Co}, \text{Co}-\text{Ni}, \text{etc.}$ [8–12]. The purpose of the paper is to present some literature data related to the $4f-3d$ magnetism of the $\text{R}_u(\text{M}-\text{M})_v$ intermetallics.

2. The Mössbauer effect

The Mössbauer effect (theory and experiment) is described in detail in a number of papers, for instance [13–18]. Especially widely described is the Mössbauer effect at the ^{57}Fe nuclei. Since the Mössbauer effect at the ^{161}Dy nuclei is less popular in the experimental practice it will be helpful to describe it shortly.

Absorbers in the form of discs of area 10 cm^2 containing about 8 mg/cm^2 of ^{161}Dy were prepared from the fine powder of the intermetallic and epoxy compounds [19].

The starting material for the Mössbauer source was about 30 mg of fluoride $^{160}\text{Gd}_{0.5}^{163}\text{Dy}_{0.5}\text{F}_3$ in which the isotopes of Gd and Dy were 90% enriched. The γ -source was ^{161}Tb obtained by a seven-day neutron irradiation (flux $2 \times 10^{14} \text{ n/(cm}^2 \text{ s)}$) of the starting fluoride [20]. The Mössbauer spectroscopy was performed on the ^{161}Dy nucleus whose nuclear transition from the $5/2^-$ excited level to the $5/2^+$ ground state emits a 25.65 keV γ -ray. During experiments the absorber was stationary and the source, at ambient temperature, was driven by a high velocity spectrometer (velocities up to 30 cm/s). The γ -quanta were detected by the krypton proportional counter. The counting gate was set over both 25.65 keV peak and the escape peak around 12 keV .

In the magnetically ordered solids the ^{161}Dy nuclei are under influence of the combined action of the magnetic and quadrupole hyperfine interactions. As a result, the sixfold degeneracy of both the excited and ground states is lifted and according to the electromagnetic selection rules the 16 electromagnetic transitions between the sublevels and then the 16 lines of the Mössbauer spectrum occur [14, 18]. The typical ^{161}Dy Mössbauer pattern can be found, for instance, in Refs. [21, 22]. The hyperfine interaction parameters, including the magnetic hyperfine field, were obtained from a numerical analysis of the pattern [14, 17, 18, 22].

3. R-sublattice and magnetism

Figure 1 (line 1) shows the exemplary linear dependence of the magnetic ordering temperature T_0 on the de Gennes factor G for the RFe_2 system [23]. This dependence is satisfactorily described by Taylor's formula [1]

$$T_0 = T_R + T_M, \quad (3.1)$$

where T_R, T_M is the contribution corresponding to the R-, M-sublattice, respectively. Moreover, $T_R = BG$, where B is a suitable constant and $G = (g-1)^2 J(J+1)$

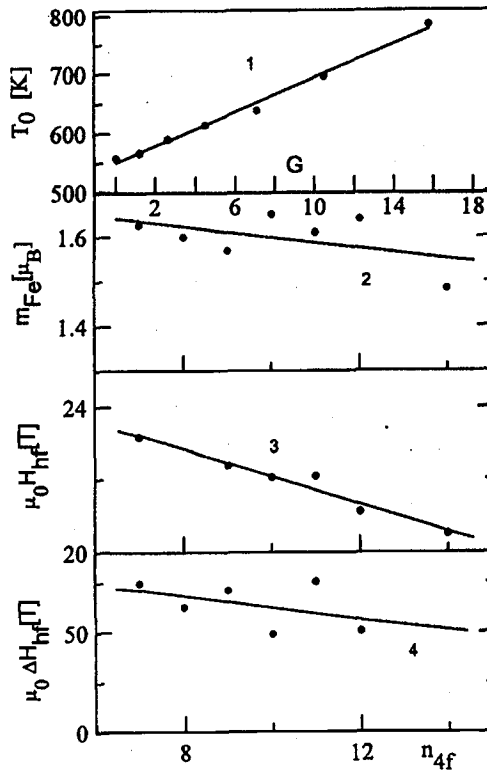


Fig. 1. The magnetic ordering temperature T_0 as a function of the de Gennes factor G (line 1), the magnetic moment m_{Fe} calculated per iron atom (line 2; 4.2 K), the magnetic hyperfine field $\mu_0 H_{\text{hf}}$ at ^{57}Fe nuclei (line 3; 4.2 K), the excess $\mu_0 \Delta H_{\text{hf}}$ of the magnetic hyperfine field at R nuclei (line 4; 4.2 K) for the RFe_2 compounds observed as functions of the number n_{4f} . The lines are least-squares fits.

is the factor related to the series of heavy rare earths: Gd ($G = 15.8$), Tb (10.5), Dy (7.1), Ho (4.5), Er (2.6), Tm (1.2), and Lu (0) [1]. This formula describes well the linear dependence $T_0(G)$ for all known R_uM_v intermetallics ($u/v = 2/17, 1/3$, and $1/2$) [23].

For the RFe_2 system $T_{\text{M}} = 549.9$ K and $B = 14.3$ K are fitted parameters. The T_{M} contribution is close to the magnetic ordering temperature of the LuFe_2 compound for which the R-sublattice is nonmagnetic [1, 2]. Additionally the $T_{\text{R}}(G)$ contribution observed across the heavy lanthanide series for RFe_2 is close to the magnetic ordering temperature for RAl_2 (nonmagnetic M-sublattice) observed across the lanthanide series [1, 2]. Analogous linear dependences corresponding, in general, to the R_uM_v systems, $\text{M} = \text{Fe}, \text{Co},$ and Ni , were also observed [1, 2, 23]. For all the R_uM_v systems a double inequality $T_{\text{M}}(\text{R}_u\text{Fe}_v) < T_{\text{M}}(\text{R}_u\text{Co}_v) > T_{\text{M}}(\text{R}_u\text{Ni}_v)$ is valid.

For the RFe_2 series of compounds Fig. 1 presents successively the magnetic moment m_{Fe} of iron atom (line 2), the magnetic hyperfine field $\mu_0 H_{\text{hf}}$ at iron

nuclei (line 3), and the magnetic hyperfine field excess $\mu_0\Delta H_{\text{hf}}$ at rare earth nuclei as functions of the number n_{4f} of electrons in the 4*f* shell of the R^{3+} ion [1, 2, 4, 12, 24, 25].

Magnetizations of the R_uM_v compounds ($u/v = 2/17, 6/23, 1/3,$ and $1/2$) were the subject of a number of investigations and the saturated magnetic moments per formula unit $m(R_uM_v)$ are relatively well known [1–4]. The magnetic moment m_{Fe} can be calculated by the following formula [1, 2, 26]:

$$m(R_uFe_v) = um_{4f} - vm_{\text{Fe}}, \quad (3.2)$$

where the magnetic moment of the rare earth ion $m_{4f} = gJ$, g is the Landé factor and J is the total angular momentum quantum number [1, 2, 27–29]. The moment m_{Fe} decreases slightly with n_{4f} (line 2). This decreasing tendency should be ascribed to a certain proportionality to the $(g - 1)J$ value [4]. The transition metal moments for the other R_uM_v compounds change analogously [1, 2, 26].

As a next step it is necessary to consider an influence of the 4*f* electrons on the ^{57}Fe magnetic hyperfine field (curve 3) observed in the transition metal sublattice. The magnetic hyperfine field at the iron nuclei can be approximated by the following formula [4]:

$$H_{\text{hf}} = H_{\text{cp}} + H_{\text{op}} + H_{\text{M}} + H_{\text{R}}, \quad (3.3)$$

where the terms H_{cp} , H_{op} , and H_{M} can be taken to be proportional to the magnetic moment of the transition metal atom and in the isostructural compounds the term H_{R} to be proportional to the $(g - 1)J$ value. The last term seems to be the main origin of the H_{hf} -field decreasing with n_{4f} . The contributions H_{cp} and H_{op} are originated by the core and own polarizations ascribed to the studied Fe atom. H_{M} and H_{R} are the transferred fields from the M- and R-sublattice, respectively.

It can be seen (lines 2, 3) that there is an approximate proportionality between the magnetic moment m_{Fe} and the observed magnetic hyperfine field H_{hf} [24, 26]

$$\mu_0 H_{\text{hf}} = Am_{\text{Fe}}, \quad (3.4)$$

where μ_0 is the magnetic permeability and constant $A = 1.5 \text{ T}/\mu_{\text{B}}$.

Subsequently, it would be interesting to consider an influence of the 4*f* electrons on the magnetic hyperfine fields at the rare earth nuclei. For the R-sublattice the magnetic hyperfine field is often expressed in a form [12, 21]

$$H_{\text{hf}} = H_{4f} + H_{\text{sp}} + H_{\text{tR}} + H_{\text{tM}}, \quad (3.5)$$

where H_{4f} is the field created by the 4*f* electrons localized on the rare earth ion and is composed of the orbit, spin, and core polarizations. The proportionality between the H_{4f} field and the number J is well established for the heavy rare earths [12]. The polarization of the neighbouring conduction electrons caused by the 4*f*-shell gives the self-polarization term H_{sp} and the contribution H_{tR} is the field transferred from the surrounding rare earth ions. The last H_{tM} term is characteristic of the R–M intermetallics. It is commonly accepted that this contribution is induced by the transition metal atoms [12, 21, 25]. In the case of the magnetic saturation, the terms: second and third are proportional to the $(g - 1)J$ value [12, 21, 25].

The last three terms in the formula (3.5) form the so-called hyperfine field excess, which can be calculated directly from the experimental data [12, 21, 25]

$$\Delta H_{\text{hf}} = H_{\text{hf}} - H_{4f}, \quad (3.6)$$

where here the H_{hf} term is the measured field. A way to appreciate the H_{4f} term is described elsewhere [12, 21, 25]. Since the H_{sp} and H_{tR} terms are rather small, thus in practice the field excess should be ascribed to the H_{tM} term [12, 21, 25]. A certain decreasing tendency of the $\mu_0\Delta H_{\text{hf}}$ excess (Fig. 1, curve 4) with the n_{4f} number is observed. This moderate decreasing should be attributed to the second and third terms in the formula which are proportional to the decreasing $(g-1)J$ value [1, 2, 27-29].

4. M-sublattice and magnetism

As a next step it is interesting to consider an influence of the M-sublattice on the magnetic properties and hyperfine interactions.

Figure 2 shows the known magnetic ordering temperatures T_0 for the transition metal series and the $\text{Dy}_u(\text{M}-\text{M})_v$ intermetallic system ($u/v = 2/17, 1/3,$ and $1/2$) [30-33]. The ordering temperatures for the $\text{Dy}(\text{Mn}-\text{Fe})_2$ series were determined considering the width of the ^{57}Fe Mössbauer line treated as function of temperature [32, 34]. Actually the data are not complete. Nevertheless, it can be deduced from the figure that the $T_0(n_{3d})$ curves form a system. The curves observed for intermetallics are shifted down as compared to the curve of transition metal series. The shift down can be related to the u/v composition parameter, but there is no direct proportionality. Moreover the position of the maximum of the curve is shifted to the left with the u/v parameter as compared to the curve of the transition metal series. The change of the magnetic ordering temperature with the n_{3d} number should be ascribed to the T_{M} term. This dependence is originated by the band structure of the M-sublattice. Since the band structure of the intermetallics is unknown up to date and there is no analytical formula for the contribution T_{M} thus a more detailed discussion is impossible at present.

In order to test the influence of the M-sublattice on the magnetism of both the sublattices, the systematic ^{161}Dy and ^{57}Fe Mössbauer effect studies were previously performed elsewhere for the exemplary $\text{Dy}(\text{Mn}-\text{Fe})_2$, $\text{Dy}(\text{Fe}-\text{Co})_2$, and $\text{Dy}(\text{Co}-\text{Ni})_2$ series [4, 8-10, 24]. Figure 3 shows the ^{161}Dy and ^{57}Fe magnetic hyperfine fields for these compounds. The magnetic hyperfine fields are plotted versus the average number n_{3d} of 3d electrons calculated per transition metal atom. Both fields appear to be strongly dependent on n_{3d} .

The ^{57}Fe field forms a typical Slater-Pauling dependence corresponding to the 3d band of the M-sublattice, with a maximum at $n_{3d} = 6.3$ electrons. The curve is slightly asymmetric and the linear parts of it can be approximated by the following simple phenomenological formula:

$$\mu_0 H_{\text{hf}}(n_{3d}) = \mu_0 H_{\text{hf}}(n_{3d} = 6.3) - h_i |6.3 - n_{3d}|, \quad (4.1)$$

where $i = 1$ for $n_{3d} < 6.3$ electrons and $i = 2$ for $n_{3d} > 6.3$ electrons and the parameters $\mu_0 H_{\text{hf}}(n_{3d} = 6.3)$, h_i are discussed elsewhere [8, 9]. This dependence can be qualitatively explained within the frame of the rigid 3d-band model.

A more unexpected result is that the ^{161}Dy magnetic hyperfine field and hence the magnetic hyperfine field excess (formulae (3.5), (3.6)) also form the Slater-Pauling curve [8]. In this case, however, a maximum of the field appears at about $n_{3d} = 5.9$ electrons. The last field should be ascribed to the rare earth

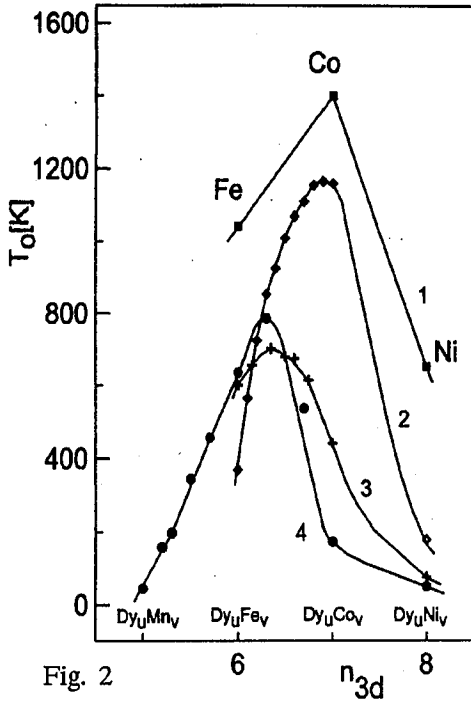


Fig. 2

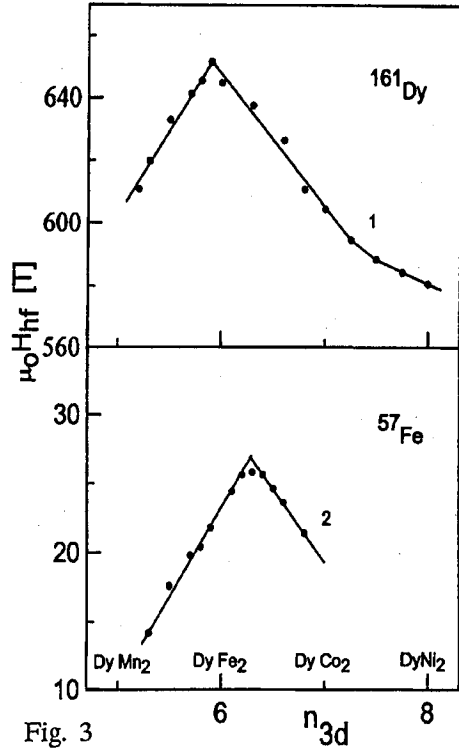


Fig. 3

Fig. 2. The magnetic ordering temperature T_0 as a function of the number n_{3d} of 3d electrons. The system of curves observed for: 3d metals (curve 1) and for the $Dy_u(M-M)_v$ compounds ($u/v = 2/17$ (2) [30], $1/3$ (3) [31], and $1/2$ (4) [32, 33]). The curves are added to follow the experimental points.

Fig. 3. The ^{161}Dy magnetic hyperfine field (1) and the ^{57}Fe magnetic hyperfine field (2) observed in the $Dy(M-M)_2$ compounds ($M-M = \text{Mn-Fe}$, Fe-Co , and Co-Ni) as functions of the number n_{3d} of 3d electrons.

sublattice and especially to the 5d-band electrons. The changes of the field related to the changes of the 5d band are enforced by the change of the number n_{3d} of 3d electrons [8, 9].

Considering (3.5, 3.6), formula (4.1) can be adjusted to describe approximately the linear regions of the ^{161}Dy Slater–Pauling dependence [8], namely

$$\mu_0 H_{\text{hf}}(n_{3d}) = \mu_0 H_{4f} + \mu_0 \Delta H_{\text{hf}}(n_{3d} = 5.9) - h_i |5.9 - n_{3d}|, \quad (4.2)$$

where h_i are suitable constants [8]. It should be noted that the maximum of the ^{161}Dy Slater–Pauling curve is shifted to the left by about 0.4 electrons as compared to the maximum of the ^{57}Fe curve. The existing shift suggests additionally an individuality of the ^{161}Dy Slater–Pauling curve.

Since there is no full analogy between the ^{57}Fe and the ^{161}Dy Slater–Pauling curves (they have different shapes, different slope rates, different asymmetries, different heights and, in particular, the maxima are shifted) a rather complex band-type mechanism to form the ^{161}Dy dependence is expected to exist [8].

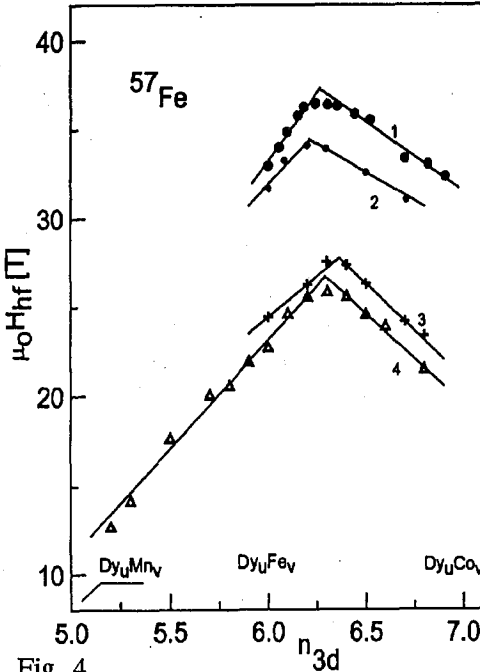


Fig. 4

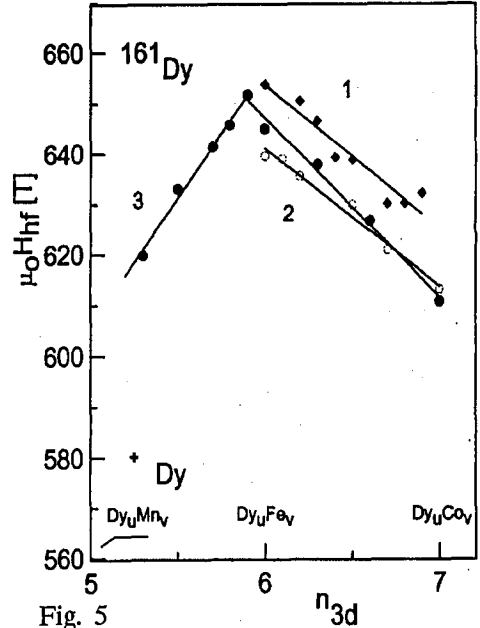


Fig. 5

Fig. 4. The system of the ^{57}Fe Slater-Pauling curves observed for the Fe-Co alloys (1) (300 K) [35] and the $\text{Dy}_u(\text{M}-\text{M})_v$ compounds ($\text{M}-\text{M} = \text{Mn}-\text{Fe}$ and $\text{Fe}-\text{Co}$) (4.2 K), $u/v = 2/17$ (2), $1/3$ (3) and $1/2$ (4) [8, 9]. The lines (excluding the data of the maximum) are least-squares fits.

Fig. 5. The system of the ^{161}Dy Slater-Pauling curves observed for the $\text{Dy}_u(\text{M}-\text{M})_v$ compounds (4.2 K), $u/v = 1/3$ (1), $2/17$ (2), and $1/2$ (3). The value for Dy metal is added. The lines are least-squares fits.

The system $H_{\text{hf}}(u/v, n_{3d})$ of the ^{57}Fe Slater-Pauling curves for the $\text{Fe}_{1-x}\text{Co}_x$ alloys and the $\text{Dy}_u(\text{Fe}_{1-x}\text{Co}_x)_v$; $u/v = 2/17$, $1/3$, and $1/2$, and Dy $(\text{Mn}-\text{Fe})_2$ compounds is presented in Fig. 4. In principle, the shapes of the Slater-Pauling curves are quite similar. The curves corresponding to the $\text{Dy}_u(\text{Fe}_{1-x}\text{Co}_x)_v$ series are shifted down as compared to the dependence of the $\text{Fe}_{1-x}\text{Co}_x$ alloys, which is described in detail elsewhere [35]. The value of the shift can be expressed by the following formula [9]:

$$\delta H_{\text{hf}}(u/v, x) = H_{\text{hf}}(\text{Fe}_{1-x}\text{Co}_x) - H_{\text{hf}}[\text{Dy}_u(\text{Fe}_{1-x}\text{Co}_x)_v]. \quad (4.3)$$

In general, δH_{hf} is a function of u/v and n_{3d} . For a given n_{3d} the value of the reduction δH_{hf} increases strongly, roughly linearly with u/v . On the other hand, following Fig. 4 it can be shown (not presented in the figure) that for a given u/v , $\delta H_{\text{hf}}(n_{3d})$ increases weakly versus n_{3d} (excluding the peak area). Nevertheless, it should be emphasized that there is no noticeable relation between the weak $\delta H_{\text{hf}}(n_{3d})$ dependence and the $H_{\text{hf}}(n_{3d})$ dependence which strongly changes by the Slater-Pauling manner. Considering this fact, it seems that the mechanism of the δH_{hf} shift-down is not directly originated by the 3d magnetisation.

Analogously, the system of the ^{161}Dy Slater–Pauling curves corresponding to the existing $\text{Dy}_u(\text{Mn}_{1-x}\text{Fe}_x)_v$, $\text{Dy}_u(\text{Fe}_{1-x}\text{Co}_x)_v$ series; $u/v = 2/17$ (1), $1/3$ (2), and $1/2$ (3) is presented in Fig. 4 [8, 36]. In this case, however, there is no considerable shift observed among curves. Data of all these curves are displaced relatively close to each other. This experimental result also should be related to the complex ferrimagnetism of intermetallics.

5. Summary

At first sight, the above presented experimental facts related to the ferrimagnetism of the R–M intermetallics seem to be mutually inconsistent. Additionally, there is no sound theory to describe these band-type ferrimagnets [1–5]. Nevertheless, taking into account the above discussed data, it seems that for a qualitative discussion the intermetallic ferrimagnets can be treated as being composed of three magnetic subsystems [13, 23]:

$$\text{R}_u\text{M}_v = (4f)_u^\uparrow [5d]_u^\uparrow \{3d\}_u^\downarrow \quad (5.1)$$

with the ferromagnetic $4f$ – $5d$ exchange coupling ($\uparrow\uparrow$) between the $4f$ -subsystem of the well-localized $4f$ electrons and the $5d$ -subsystem of the band electrons, and subsequently, with the antiferromagnetic $5d$ – $3d$ exchange coupling ($\uparrow\downarrow$) between the $5d$ -band subsystem and the $3d$ -band subsystem.

The $4f$ -subsystem creates the H_{4f} , H_{sp} and H_{tR} contributions to the magnetic hyperfine field at the ^{161}Dy nuclei. Subsequently, the $5d$ -subsystem forms the field H_{tM} (formula (3.5)), i.e. the main contribution to the excess [12, 21, 25]. It seems that this field is created by magnetization of the $5d$ -subsystem. Nevertheless, this magnetization is induced by the magnetization of the $3d$ -subsystem throughout the $5d$ – $3d$ exchange interactions [8, 9]. Thus, the induced magnetization of the $5d$ -subsystem changes with n_{3d} and as a result the ^{161}Dy Slater–Pauling curve is observed.

Assuming that the $5d$ -magnetization in the Dy_uFe_v system treated as a function of the u/v parameter is at the same level, the almost constant excess $\Delta H_{\text{hf}}(u/v)$ can be understood (Fig. 1, line 4) [8, 9]. For the Dy_uFe_v system, the number $n_{3d} = 6$. This situation is presumably repeated for any n_{3d} , and as a result the ^{161}Dy Slater–Pauling curves, for different u/v (Fig. 5), are situated in a close proximity [8].

Both the electronic structure and the ferrimagnetism of the R–M intermetallics are complex. It can be seen from the presented data that intermetallics can be treated approximately as being composed of three magnetic subsystems. The electronic structure, the magnetic properties, and the hyperfine interactions, mutually related, depend mainly on the number n_{3d} of $3d$ electrons which are present in the $3d$ -magnetic subsystem. The model considering three magnetic subsystems seems to be a good base to discuss qualitatively a number of the ferrimagnetic properties of the R–M intermetallics. The band-type properties can be qualitatively understood considering the Stoner-type rigid band model [36–38]. Nevertheless, a more exhaustive discussion of these properties would be possible after sound theoretical and numerical studies of the $5d$ -band and $3d$ -band electronic structures for the R–M compounds.

Acknowledgments

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