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Crystal electric field parameters determination for $R_2Fe_{14}B$ compounds based on Yamada-Kato model

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Abstract. Semi-empirical model developed by Yamada-Kato enables calculation of magnetic spin directions for $R_2Fe_{14}B$ compounds, based on minimization of free energy, and – in further steps – determination of spin reorientation temperatures for transitions from basal plane to axial easy magnetization direction. In our study, this model has been successfully used to determine crystal field and exchange field parameters for $Er_{2-x}Ce_xFe_{14}B$ compounds based on spin reorientation temperatures obtained experimentally from Mössbauer measurements.

Key words: crystal electric field parameters • model Yamada-Kato • Mössbauer effect • permanent magnet materials • spin reorientation

Introduction

$R_2Fe_{14}B$ (R – rare earth) intermetallic compounds belong to $Nd_2Fe_{14}B$ structure type and have a tetragonal crystal lattice with the $P4_2 = mnm$ space group [7]. They have 68 atoms in elementary cell. Fe atoms occupy six non-equivalent positions ($16k_1$, $16k_2$, $8j_1$, $8j_2$, $4e$, $4c$), rare earth two ($4f$, $4g$) and boron one position ($4g$). The high magnetocrystalline anisotropy of R- and $3d$-metal sublattice and their different behaviour with temperature are the reason for spin reorientation transitions. During these transitions, the tilting angle between the direction of the magnetization vector and c-crystallographic axis is changing. This process was observed, for example, by the Mössbauer method, for many compounds of $Nd_2Fe_{14}B$ type [1, 6, 8, 10–14, 16].

Theoretical description of this process was proposed by Yamada *et al*. [17].

Model description

The simplified Yamada-Kato model [4, 17] is based on free energy calculations and on separation of magnetic atoms into two coupled sublattices (R – rare earth sublattice and Fe-sublattice). Each sublattice is treated in a different way.

The R-sublattice is treated as an assembly of isolated atoms (R-R interaction is neglected) in the crystal electric field (CEF) and exchange field which describe the R-Fe interaction. The magnetic free energy can

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be calculated from the energy levels corresponding to the particular set of CEF parameters.

$$\widehat{H}_{\mathrm{CEF}}(i) = \sum_{n,m} B_n^m(i) O_n^m \tag{1}$$

where $B_n^m(i)$ are the crystal field parameters of the rare-earth atom at site i, O_n^m are the Stevens operators. The contributions from different R ions are assumed to be linear. In our considerations the same parameters for f and g sites were taken.

The Fe-sublattice is introduced in a phenomenological way. The free energy is approximated by anisotropy energy based on the temperature dependence of anisotropy constant $K_1(T)$ obtained experimentally [2] and scaled in temperatures to the Curie temperature of each compound. It is expected that the entropy term is negligible.

The two magnetic sublattices are coupled with an associated exchange energy of the molecular magnetic field, of the magnetization of the 3*d*-sublattice, acting on the magnetization of the rare-earth sublattice. The molecular field is related to the exchange fields, B_{ex}, acting between magnetic moments.

The total free energy is given by the formula [17]:

$$F(T,\theta) = -kT\sum_{i=1}^{4} \ln Z(i) + 28K_1(T)\sin^2\theta \tag{2}$$

where $Z(i)$ is the partition function

$$Z(i) = \sum_{j=1}^{2J+1} \exp\left[-\frac{E_j(i)}{kT}\right] \tag{3}$$

$E_j(i)$ are the energy levels of R-ion in i = 4*f*, 4*g* lattice sites.

The angle of spin orientation, for a chosen temperature T, was determined by minimizing the free energy function $F(\theta)$.

Application of the modified Yamada-Kato model to $Er_{2-x}Ce_xFe_{14}B$

Application of the model to $Er_{2-x}Ce_xFe_{14}B$ compounds with CEF and exchange field parameters taken from the literature (based on point charge approximation and derived from neutron studies [5, 15, 17]) reproduces in an excellent way the dependence of spin reorientation temperatures on composition for low Ce-content [11]. However, for higher Ce concentration substantial discrepancies occur between theoretical and experimental data [3, 9]. In addition, no evidence of conical spin arrangement was found – in contrast to predictions of the model. The absence of conical arrangement is qualitatively illustrated in Fig. 1 by observing the behaviour of the sixth line of $8j_2$ sublattice sextet. Separation of this line, as a result of reorientation process, is a consequence of a big change of angle between the direction of magnetic moment and electric-field gradient axis for this sublattice. This relates to big change of QS in $8j_2$ Zeeman sextet. The absence of intermediate arrangements and absence of "smearing" of the $8j_2$ line allows to conclude that there are no "intermediate" angles and thus – there is a sudden (flip-like) change with no intermediate (conical) arrangements of spins. Additionally, a gradual increase in amplitude of the separated line allows to conclude that not all magnetic moments reorient exactly at the same temperature – but in some range of temperatures.

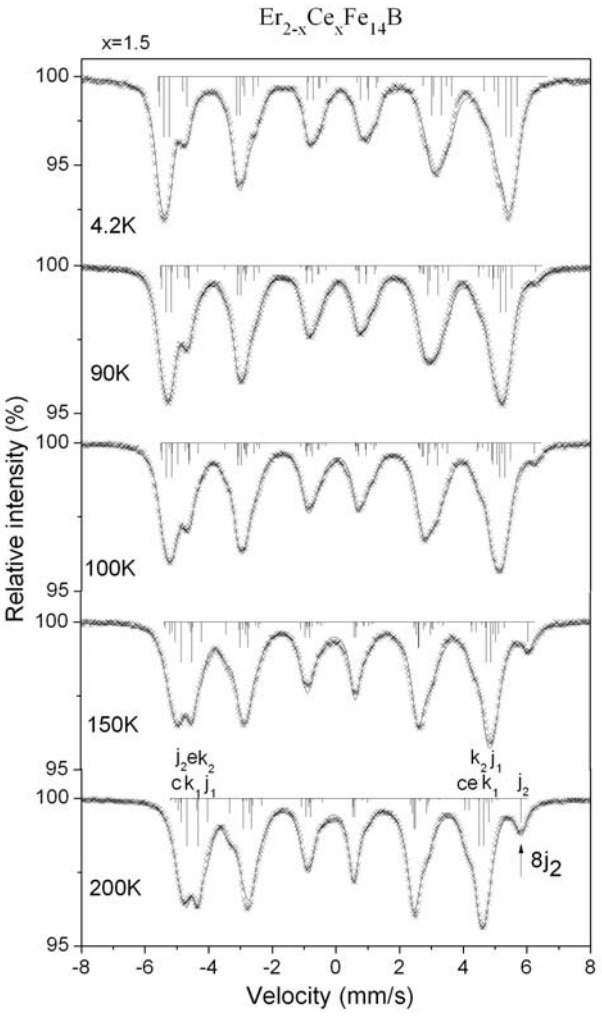

Fig. 1. Selected experimental ^{57}Fe Mossbauer spectra for x = 1.5 compound, $Er_{2-x}Ce_xFe_{14}B$. Solid lines are fits to the experimental data. The stick diagrams show the line positions and their relative intensities. The first and the sixth "high temperature" Zeeman sextet lines are marked by sublattice symbols on the 200 K spectrum.

The above observations gave the ground to the idea of trying to use the model in an "inversed sequence": to determine some of CEF parameters and exchange field parameters by fitting "theoretical" spin reorientation

Table 1. Crystal and exchange field parameters (in Kelvin) for $Re_2Fe_{14}B$ (Re = Er, Ce) obtained as a result of fitting the theoretical model to the experimental data

Re	B_2^0	B_2^2	B_4^0	B_6^0	$2(g_s-1)B_{ex}$
Er	0.562(26)	–0.417(62)	$-1.38(26)*10^{-3}$	$-0.79(15)*10^{-5}$	56.74(66)
Ce	–23.12(38)	40.2(1.1)	0.769(26)	0.0	32.5(3.0)

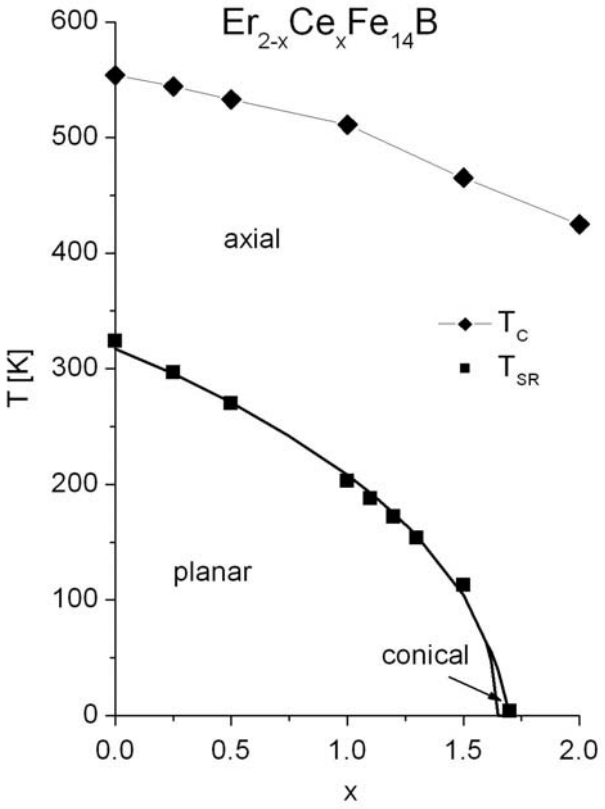

Fig. 2. Spin arrangement diagram for $Er_{2-x}Ce_xFe_{14}B$ system. T_C – Curie temperature, T_{SR} – spin reorientation temperature determined from Mössbauer measurements. The solid lines represent the limits of the range of the reorientation process obtained as a result of fit the theoretical model to the experimental data.

temperatures (obtained from the Yamada-Kato model) to experimental data obtained from our Mössbauer experiments. The result of our best attempt is shown in Table 1, containing the parameters obtained from fits. For such set of parameters, the fit is good (Fig. 2) and the conical configuration is almost negligible. A very strong influence of B_4^0 coefficient for Er on the width of conical region was observed.

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References

1. Abe M, Liou SH, Chien CL, Koon NC, Das BN, Callen E (1987) Mössbauer study of spin reorientation in $Y_{1.8}Er_{0.2}Fe_{14}B$. J Appl Phys 61:3568–3570
2. Andreev AV, Bartashevich MI, Goto T, Zadvorkin SM (1997) Magnetic properties of $(Y_{1-x}Th_x)_2Fe_{14}B$. J Alloys Compd 262/263:467–470
3. Bogacz BF, Pędziwiatr AT (2008) Mössbauer investigation of spin arrangements in $Er_{2-x}Ce_xFe_{14}B$. Acta Phys Pol A 114;6:1509–1516
4. Bogacz BF, Rubin J, Pędziwiatr AT, Bartolome J, Wielgosz R, Artigas M (1999) Calculation and experimental magnetic phase diagrams for $(Nd,Y)_2(FeCo)_{14}B$ systems. In: Proc of XXXIV Zakopane School of Physics, Zakopane 1:191–198
5. Coey JMD (1996) Rare-earth iron permanent magnet. Claredom Press, Oxford
6. Fujita A, Onodera H, Yamauchi H *et al*. (1987) Magnetic and ^{57}Fe Mössbauer studies of collinear spin rotation in $Ho_2Fe_{14}B$. J Magn Magn Mater 69:267–275
7. Herbst JF, Croat JJ, Pinkerton FE, Yelon WB (1984) Relationships between crystal structure and magnetic properties in $Nd_2Fe_{14}B$. Phys Rev B 29:4176–4178
8. Koon NC, Abe M, Callen E *et al*. (1986) Mössbauer evidence of spin reorientation in single crystal $Nd_2Fe_{14}B$. J Magn Magn Mater 54/57:593–594
9. Krawiec K, Pędziwiatr AT, Bogacz BF, Przewoźnik J, Żukrowski J, Kapusta Cz (2011) Magnetic structure studies of ternary borides $Er_{2-x}Ce_xFe_{14}B$. J Magn Magn Mater 323:2968–2972
10. Niarchos D, Simopoulos A (1986) Temperature variation of the spin reorientation in $Er_{2-x}Dy_xFe_{14}B$ alloys. Solid State Commun 59:669–672
11. Pędziwiatr AT, Bogacz BF, Gargula R (2003) Spin arrangement diagrams for $Er_{2-x}R_xFe_{14}B$ (R=Y,Ce) obtained with Mössbauer spectroscopy and phenomenological model. Nukleonika 48;Suppl 1:S59–S63
12. Pędziwiatr AT, Bogacz BF, Gargula R, Wróbel S (2002) Cerium influence on the spin reorientation in $Er_{2-x}Ce_xFe_{14}B$ evidenced by Mössbauer and DSC techniques. J Magn Magn Mater 248:19–25
13. Pędziwiatr AT, Wojciechowska A, Bogacz BF, Wróbel S (2005) Experimental studies of spin reorientations in $Er_{2-x}Th_xFe_{14}B$. J Phys: Condens Matter 17:6999–7007
14. Price DC, Day RK, Dunlop JB (1986) ^{57}Fe Mössbauer measurements of the spin reorientation in $Tm_2Fe_{14}B$. J Appl Phys 59:3585–3587
15. Radwański RJ, France JJ (1987) Rare-earth contribution to the magnetocrystalline anisotropy energy in $R_2Fe_{14}B$. Phys Rev B 36:8616–8621
16. Wojciechowska A, Pedziwiatr AT, Bogacz BF, Wróbel S (2007) Spin arrangement diagrams for $Er_{2-x}R_xFe_{14}B$ (R=Pr, Gd). J Alloys Compd 442:126–128
17. Yamada M, Kato H, Yamamoto H, Nakagawa Y (1988) Crystal-field analysis of the magnetization process in a series of $Nd_2Fe_{14}B$-type compounds. Phys Rev B 38:620–633