Journal of Alloys and Compounds 512 (2012) 128–131

Contents lists available at SciVerse ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Analysis on three-sublattice model of magnetic properties in rare-earth iron garnets under high magnetic fields

Wei Wang*, Ri Chen, Xin Qi

Department of Physics and Electronics, School of Science, Beijing University of Chemical Technology, Beijing 100029, China

ARTICLE INFO

Article history:
Received 30 June 2011
Received in revised form 8 September 2011
Accepted 8 September 2011
Available online 19 September 2011

Keywords:
Three-sublattice
Rare-earth iron garnet
Magnetic properties
Molecular field

ABSTRACT

In this paper, based on the molecular field theory, a new and improved three-sublattice model on studying the magnetic properties of ferrimagnetic rare-earth iron garnet in high magnetic fields is introduced. Here, the effective exchange field is described as $H_i = \lambda M = \lambda\chi H_e$, where λ is the coefficient associated with the molecular field, χ is the effective magnetic susceptibility, and H_e is external magnetic fields. As is known, the magnetic sublattices in rare-earth iron garnets can be classified three kinds labeled as *a*, *c* and *d*, in our calculations, whose magnetizations are defined as M_a, M_c and M_d, respectively. Then, using this model, the temperature and field dependences of the total magnetization in $Dy_3Fe_5O_{12}$ (DyIG) are discussed. Meanwhile, the magnetizations of the three kinds of magnetic sublattices are analyzed. Furthermore, our theory suggests that the coefficients α_i associated with λ and χ in DyIG show obvious anisotropic, temperature-dependence and field-dependence characteristics. And, the theoretical calculations exactly fit the experimental data.

1. Introduction

Due to a wide application in the field of magnetic storage, magnetic recording, magnetic field sensor and magneto-optical devices, lots of theoretical and experimental reports on magnetic properties of rare-earth (RE) iron garnet (REIG) have been presented over the past decades [1–3]. Meanwhile, it is noticeable that, up till now, due to the complicated magnetic structures of REIG, the physical mechanism of some magnetic behaviors in REIG, especially some heavy RE iron garnets, has not yet been entirely theoretically explored. In particular, recently, some new experimental phenomena on magnetic properties in REIG have been found under extreme conditions (low temperatures or high magnetic fields), such as magnetic anisotropy, nonlinear field-dependence characteristic, new magnetic phase diagrams and low temperature magnetic saturation [4,5]. Then, in recent years, great efforts have still been made to establish the related theoretical models for the study on the magnetic properties of the ferrimagnetic rare-earth iron garnet system [6].

As is known, to study the magnetic properties of REIG, many theoretical models have been presented, e.g., the Néel's model, the Yafet and Kittel's model and one suggested by Gilleo [7–9]. Nevertheless, it is also found that some models show little agreement with experimental observations. Here, a fact should be accepted, i.e., the molecular field theory of Néel has been shown very successful to explain the magnetic properties of rare-earth garnets or some other magnetic compounds [10,11]. Whereas, we also notice that, when using the molecular field theory, the determination of the molecular field coefficients is much more difficult, and shows great importance to the theoretical analyses on the magnetic properties of REIG [12]. Therefore, a large number of works have been devoted to the study of the molecular field coefficients. In fact, presently, as for most of the RE iron garnets, the values of the molecular field coefficients have been determined despite some debates on these values also exist in the literatures [13]. However, it is noticeable that, in some rare-earth iron garnets, few theoretical studies have been carried out to explain the magnetic behaviors under high magnetic fields by the molecular field theory.

In our previous papers, the molecular field theory is extended to analyze the exchange interaction in rare-earth garnets where the exchange field is expressed as $H_i = \lambda M = \lambda\chi H_e$, here, λ denotes the coefficient associated with the molecular field, χ represents the effective magnetic susceptibility, and H_e is the external magnetic fields. Then, a two-sublattice model of rare-earth gallium garnet is provided [14,15]. Furthermore, our theoretical calculations have indicated that the magnetic properties of some RE gallium and iron garnets, using this expression of the exchange field, can be successfully interpreted, especially in high magnetic fields [16]. Now, in this paper, this thought will continue to be used. And, in correspondence to REIG, a new and improved theoretical model of three-sublattice ferrimagnetic system is put forward. Meanwhile, based on this model, the field-dependence

* Corresponding author.
E-mail addresses: wangwei@mail.buct.edu.cn, wwyz20@sina.com (W. Wang).

0925-8388/$ – see front matter
doi:10.1016/j.jallcom.2011.09.041

and temperature-dependence properties of magnetization in DyIG are discussed under high magnetic fields. Moreover, the theoretical data are compared with the experimental results.

2. Outline of three-sublattice model

The general chemical structural formula for REIG can be written as $RE_3Fe_2Fe_3O_{12}$, with eight of these formula units per unit cell. With the overall symmetry being cubic, the space group of REIG is $Ia\bar{3}d$ (O_h^{10}), in which three special positions are occupied by magnetic ions. That is, two Fe^{3+} ions occupy the octahedral crystal site, expressed as 16a, three Fe^{3+} ions occupy tetrahedral crystal site (24d), and the RE ions occupy the dodecahedral crystal site (24c) with eightfold oxygen coordination. Therefore, the magnetization (M) of REIG originates from the contributions of these three kinds of magnetic ions whose magnetizations can be described as M_c, M_a and M_d, respectively. Thus, in this three-sublattice system, M can be written as

$$M = M_c + M_a + M_d, \tag{1}$$

where c, a and d represent the dodecahedral, octahedral and tetrahedral sites, respectively.

Referring to Langevin theory, we assume that the above three magnetizations are given by

$$M_i = M_{Si} \cdot B(y_i) = N_i J_i g_{Ji} \mu_B B(y_i), \tag{2}$$

here N_i ($i = a$, c and d) are the numbers of ions contribution to magnetization per unit volume, J_i denote the total quantum number, g_{Ji} represent the Lander factors, and the Brillouin functions are expressed as

$$B(y_i) = \left[(2J_i+1)/2J_i\right] \coth\left[((2J_i+1)/2J_i)\cdot y_i\right] - (1/2J_i)\coth\left[(1/2J_i)\cdot y_i\right]. \tag{3}$$

And,

$$y_c = \frac{J_c g_{Jc} \mu_B H_c}{k_B T},\ y_a = \frac{J_a g_{Ja} \mu_B H_a}{k_B T},\ y_d = \frac{J_d g_{Jd} \mu_B H_d}{k_B T}, \tag{4}$$

where H_c, H_a and H_d denote the total effective fields in c, a and d sublattices, respectively. In terms of the molecular field theory, considering external magnetic fields, the total effective fields can be described as

$$H_c = H_e + \lambda_{cd} M_d + \lambda_{cc} M_c + \lambda_{ca} M_a, \tag{5}$$

$$H_a = H_e + \lambda_{ad} M_d + \lambda_{ac} M_c + \lambda_{aa} M_a, \tag{6}$$

$$H_d = H_e + \lambda_{dd} M_d + \lambda_{dc} M_c + \lambda_{da} M_a. \tag{7}$$

Here λ_{ij} ($i,j = a$, c and d) are the corresponding exchange field coefficients between nearest-neighbor sublattices, and, according to our previous papers, it can be defined that $M_c = \chi_c H_e$, $M_a = \chi_a H_e$, and $M_d = \chi_d H_e$ where χ_c, χ_a and χ_d are the effective magnetic susceptibilities of c, a and d magnetic sublattices. Then, Eq. (4) can be rewritten as

$$y_c = \frac{J_c g_{Jc} \mu_B (1+\alpha_c) H_e}{k_B T},\ y_a = \frac{J_a g_{Ja} \mu_B (1+\alpha_a) H_e}{k_B T},$$

$$y_d = \frac{J_d g_{Jd} \mu_B (1+\alpha_d) H_e}{k_B T}, \tag{8}$$

where

$$\alpha_c = \lambda_{cd}\chi_d + \lambda_{cc}\chi_c + \lambda_{ca}\chi_a, \tag{9}$$

$$\alpha_a = \lambda_{ad}\chi_d + \lambda_{ac}\chi_c + \lambda_{aa}\chi_a, \tag{10}$$

$$\alpha_d = \lambda_{dd}\chi_d + \lambda_{dc}\chi_c + \lambda_{da}\chi_a. \tag{11}$$

On the other hand, in the light of the ferrimagnetic molecular field theory, we can assume that $\lambda_{ac} = \lambda_{ca}$, $\lambda_{ad} = \lambda_{da}$ and $\lambda_{cd} = \lambda_{dc}$.

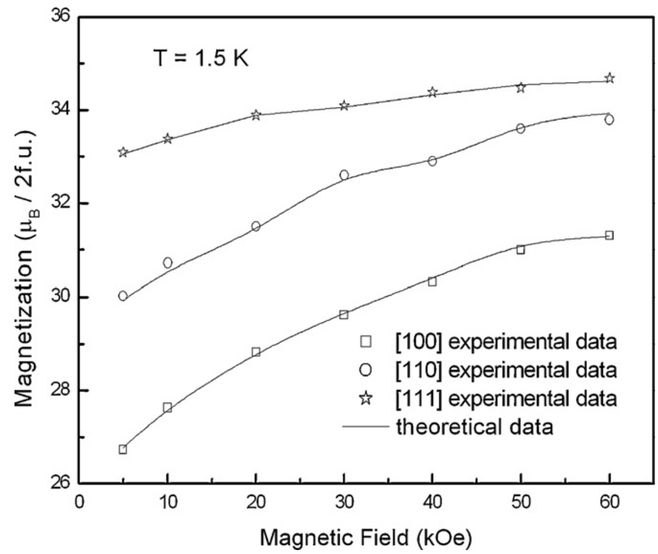

Fig. 1. The variation of magnetization with the external magnetic field along [1 0 0], [1 1 0] and [1 1 1] at 1.5 K (the experimental data obtained from Ref. [18]).

Thus, based on the above theory, according to Eq. (2), the values of M_a, M_c, and M_d in correspondence to these three magnetic sublattices can be calculated. Correspondingly, the variations of the total magnetization M with the temperature T and the external magnetic fields H_e can be discussed.

3. Results and discussion

It is known that, as to the magnetic properties of DyIG, many theoretical investigations have been carried out. Moreover, several sets of the molecular-field coefficients have been determined, and, the temperature-dependence of the molecular-field coefficients has been pointed out [17]. However, it is found that the recent experimental phenomena on magnetic properties in DyIG, especially under high magnetic fields and low temperatures, have not been theoretically explained [18,19]. So, now, the above three-sublattice model is applied to interpret the magnetic properties of DyIG in high magnetic fields.

As mentioned above, Dy^{3+} and Fe^{3+} ions are the magnetic ions contribution to the magnetic properties of DyIG, where three Dy^{3+} ions occupy c crystal site, two Fe^{3+} ions occupy a site, and other three Fe^{3+} ions occupy d site in a formula unit. In our calculations, as to Dy^{3+} ion, the ground term is $4f^9$, and the ground multiplet $^6H_{15/2}$ is considered, then, $J_c = 15/2$, and $g_{Jc} = 4/3$. And, as to Fe^{3+}, $J_a = J_d = 5/2$, and $g_{Ja} = g_{Jd} = 2$. Now, substituting the above parameters into Eqs. (2)–(8), we can obtain the magnetic moments of the three sublattices in DyIG under different conditions. Also, based on Eq. (1), the whole magnetic moment of DyIG can be presented by fitting the values of α_a, α_c and α_d in Eqs. (9)–(11). Figs. 1–3 give the theoretical fitting results of the magnetic curves in DyIG under high magnetic fields at low temperatures (T = 1.5 K, 4.2 K and 5 K), where the theoretical calculations are in good agreements with the experimental data. Certainly, detailed analyses on the magnetic moments of the three sublattices and the values of the coefficients α_a, α_c and α_d are carried out as follows.

Then, in our calculations, the magnetizations of the magnetic sublattices a, c and d are given, to further analyze the contribution of the above three magnetic sublattices to the magnetic properties in DyIG, at low temperatures along [1 0 0], [1 1 0] and [1 1 1] directions. Here, as is pointed out, the magnetic sublattice a is antiparallel to the magnetic sublattice d, and parallel to the magnetic sublattice c. So, in our fitting, it is found that $M_a > 0$, $M_c > 0$, and $M_d < 0$.

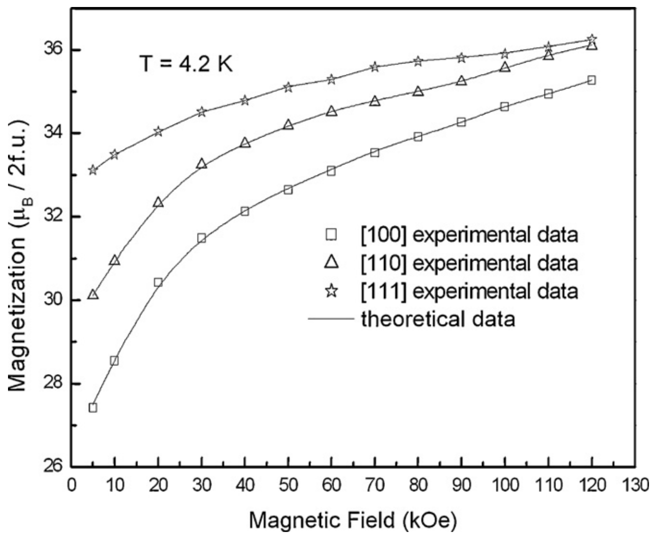

Fig. 2. The variation of magnetization with the external magnetic field along [1 0 0], [1 1 0] and [1 1 1] at 4.2 K (the experimental data obtained from Ref. [20]).

Figs. 4–6 give the field-dependence of three magnetic sublattices when the external magnetic field is parallel to [1 0 0], [1 1 0] and [1 1 1] directions at 1.5 K, 4.2 K and 5 K, respectively.

Theoretical calculations indicate that the moment of rare-earth ion in DyIG is larger than that of iron ion at low temperatures. Meanwhile, as to DyIG, the values of the magnetization of Dy^{3+} ion are nearly twice as many as those of Fe^{3+} ion occupying the magnetic sublattice *d*. Moreover, the moment of Fe^{3+} ion occupying magnetic sublattice *a* is weakest among these three magnetic sublattices. So, it can be demonstrated that, with the action of the external magnetic fields, the moment of rare-earth ion shows great importance to the magnetic properties of rare-earth iron garnets [12].

In addition, seen from Figs. 4–6, the magnetizations of the three magnetic sublattices show obvious anisotropy. And, our calculations indicate that the moment when H_e along to [1 1 1] direction is larger than those along [1 1 0] and [1 0 0] directions, which further proves that the [1 1 1] direction is the direction of easy axis. On the other hand, it is interesting that, under a low magnetic field, the values of M_a is bigger than M_d, while $M_d > M_a$ with the increase of the magnetic fields, which implies the change of the magnetic structures in DyIG under high magnetic fields. And, the transition magnetic fields at 1.5 K, 4.2 K and 5 K are about 15–20 kOe, 5–10 kOe and 10–15 kOe. Also, known from Fig. 5, with the increase of magnetic field, especially in higher magnetic fields, the magnetizations of the three magnetic sublattices change very little. And, it can be apparently found that the magnetization gradually tends to saturation when $H_e > 70$ kOe.

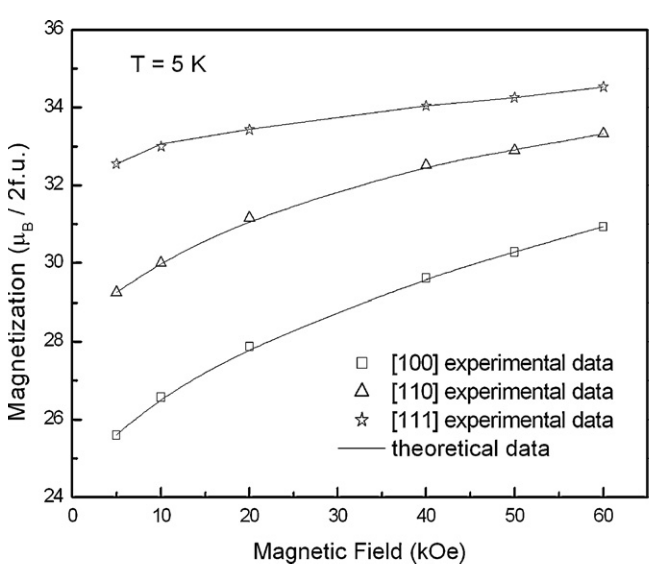

Fig. 3. The variation of magnetization with the external magnetic field along [1 0 0], [1 1 0] and [1 1 1] at 5 K (the experimental data obtained from Ref. [21]).

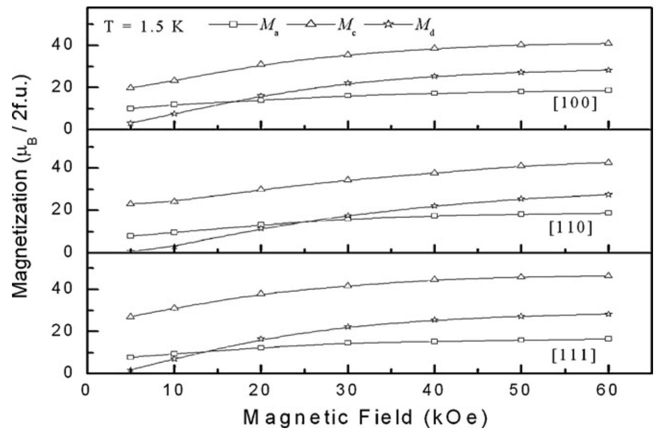

Fig. 4. The magnetic field dependence of the magnetizations in magnetic sublattices *a*, *c* and *d* along [1 0 0], [1 1 0] and [1 1 1] directions at 1.5 K.

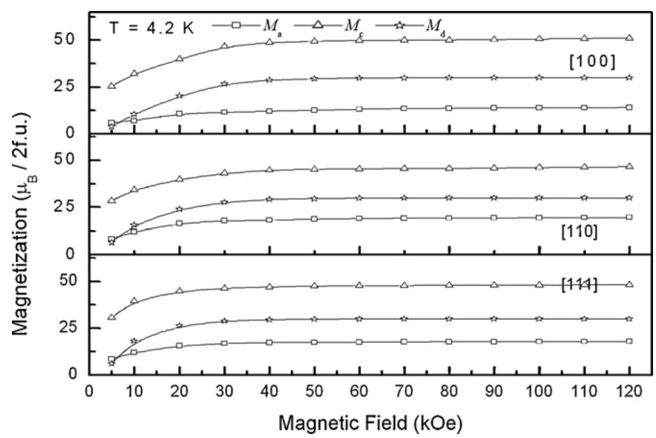

Fig. 5. The magnetic field dependence of the magnetizations in magnetic sublattices *a*, *c* and *d* along [1 0 0], [1 1 0] and [1 1 1] directions at 4.2 K.

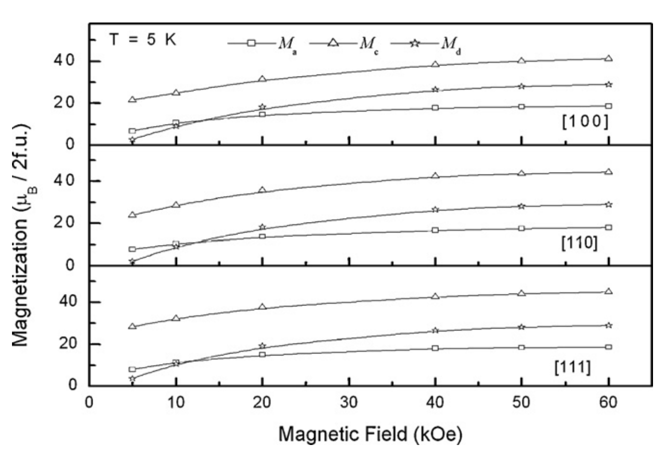

Fig. 6. The magnetic field dependence of the magnetizations in magnetic sublattices *a*, *c* and *d* along [1 0 0], [1 1 0] and [1 1 1] directions at 5 K.

Table 1
The values of the three parameters P_1, P_2 and P_3 of α_a, α_c and α_d along [1 0 0], [1 1 0] and [1 1 1] at 1. 5 K.

	[1 0 0]			[1 1 0]			[1 1 1]		
	α_a	α_c	α_d	α_a	α_c	α_d	α_a	α_c	α_d
P_1	0.28194	0.07616	−0.42135	0.33806	0.08226	−0.3529	0.17706	0.09624	−0.42073
P_2	3.92484	2.3472	2.52734	1.2653	1.97242	3.68657	3.69219	3.37323	2.35642
P_3	0.88935	−1.17737	−7.1429	5.47242	2.3205	−10.89428	−2.95805	−2.36245	−4.03343

Table 2
The values of the three parameters P_1, P_2 and P_3 of α_a, α_c and α_d along [1 0 0], [1 1 0] and [1 1 1] at 4.2 K.

	[1 0 0]			[1 1 0]			[1 1 1]		
	α_a	α_c	α_d	α_a	α_c	α_d	α_a	α_c	α_d
P_1	0.25534	0.36516	−2.5322	0.88931	0.16068	−2.4609	0.46252	0.10824	−2.94765
P_2	9.9125	10.00226	26.1108	18.0483	13.9569	14.0696	23.6405	20.5072	11.7467
P_3	−14.593	−14.862	−85.9008	−52.775	−23.6974	−39.4486	−67.763	−50.3814	−12.9519

Table 3
The values of the three parameters P_1, P_2 and P_3 of α_a, α_c and α_d along [1 0 0], [1 1 0] and [1 1 1] at 5 K.

	[1 0 0]			[1 1 0]			[1 1 1]		
	α_a	α_c	α_d	α_a	α_c	α_d	α_a	α_c	α_d
P_1	1.05894	0.22713	−1.665	0.7948	0.2634	−1.6659	1.02159	0.22778	−1.5919
P_2	13.3591	8.66946	8.8215	13.395	10.8646	8.4551	15.5133	13.06158	4.88468
P_3	−35.874	−4.1005	−17.683	−20.339	−11.4124	−13.505	−35.1435	−11.33238	−4.82273

Here, from Eqs. (9)–(11), it can be known that the coefficients α_a, α_c and α_d are associated with λ and χ, which implies that they are the functions of the external magnetic field H_e and the temperature T. Then, in our fitting, the field-dependence characteristics of the above three coefficients are described as follows.

$$\alpha_i = P_1 + P_2 \cdot H_e^{-1} + P_3 \cdot H_e^{-2}, \tag{12}$$

where $i = a$, c, d, and P_1, P_2 and P_3 are three parameters. In our calculations, the corresponding three parameters of α_a, α_c and α_d along [1 0 0], [1 1 0] and [1 1 1] at 1.5 K, 4.2 K and 5 K are listed in Tables 1–3, respectively. Therefore, the following conclusions can be drawn: (1) it is known that χ is the function of the external magnetic field, which leads to the complicated field-dependence properties of α_i; (2) seen from Tables 1–3, α_i show obvious anisotropy, which mainly originates from the anisotropic characteristic of λ; (3) meanwhile, it is worthy to point out that the different values of P_1, P_2 and P_3 at the above three temperatures also imply obvious temperature-dependence of α_i, in our opinion, which results from the temperature properties of λ and χ. However, owing to the lack of experimental data, the concrete relation of λ and χ with T cannot be given in this study. And, the further studies will be carried out in a forthcoming paper.

4. Conclusions

In this paper, to study the magnetic properties of rare-earth iron garnets under high magnetic fields, an improved three-sublattice model is provided. Here, the total effective field is regarded as the sum of the exchange field and the external magnetic field. And, using this model, the magnetic behaviors of DyIG under high magnetic fields are discussed. The wonderful fittings to the experiments confirm the correctness of our theoretical deductions. And, the anisotropy, temperature-dependence and field-dependence of magnetization in DyIG are explained. Meanwhile, the magnetizations of three-sublattice are calculated where it is pointed out that the magnetic properties of the rare-earth ions show great importance to the magnetic behaviors of rare-earth iron garnets. Moreover, as to the three-sublattice, the changes of the magnetizations with the external magnetic field at different temperatures are revealed. Also, theoretical calculations suggest that the coefficients α_i associated with λ and χ are the functions of the external magnetic fields and the temperatures.

Acknowledgements

This work is supported by the National Natural Science Foundation of China (grant nos. 11004005 and 60971019) and the program of excellent talents in Beijing of China (grant no. 2011D009016000001).

References

[1] S.H. Vajargah, H.R.M. Hosseini, Z.A. Nemati, J. Alloys Compd. 430 (2007) 339.
[2] M. Niyaifar, A. Beitollahi, N. Shiri, M. Mozaffari, J. Amighian, J. Magn. Magn. Mater. 322 (2010) 777.
[3] N. Tsidaeva, J. Alloys Compd. 374 (2004) 160;
N. Tsidaeva, V. Abaeva, J. Alloys Compd. 418 (2006) 145;
N. Tsidaeva, J. Alloys Compd. 408–412 (2006) 164.
[4] C.N. Chinnasamy, J.M. Greneche, M. Guillot, B. Latha, T. Sakai, C. Vittoria, V.G. Harris, J. Appl. Phys. 107 (2010) 09A512.
[5] A. Bouguerra, G. Fillion, S. de Brion, S. Khene, P. Wolfers, E.K. Hlil, J. Phys.: Condens. Matter 20 (2008) 295231.
[6] B. Vertruyen, R. Cloots, J.S. Abell, T.J. Jackson, R.C. da Silva, E. Popova, N. Keller, Phys. Rev. B 78 (2008) 094429.
[7] P.G. de Gennes, Phys. Rev. Lett. 3 (1959) 209.
[8] Y. Yafet, C. Kittel, Phys. Rev. 87 (1952) 290.
[9] M.A. Gilleo, J. Phys. Chem. Solids 13 (1960) 33.
[10] G.F. Dionne, J. Appl. Phys. 41 (1970) 4874;
G.F. Dionne, J. Appl. Phys. 42 (1971) 2142.
[11] S. Kervan, J. Alloys Compd. 368 (2004) 8;
A. Kılıç, S. Kervan, A. Gencer, J. Alloys Compd. 372 (2004) 25.
[12] M. Guillot, A. Marchand, P. Feldmann, H. Le Gall, J.M. Desvignes, J. Appl. Phys. 50 (1979) 2112.
[13] G.F. Dionne, J. Appl. Phys. 47 (1976) 4220.
[14] W. Wang, J. Appl. Phys. 102 (2007) 063905.
[15] W. Wang, Y. Yue, J. Alloys Compd. 488 (2009) 23.
[16] W. Wang, Y. Yue, D. Li, J. Appl. Phys. 105 (2009) 083909.
[17] M. Lahoubi, A. Bouguerra, A. Kihal, G. Fillion, J. Alloys Compd. 275–277 (1998) 598.
[18] M. Lahoubi, W. Younsi, M.L. Soltani, J. Voiron, D. Schmitt, J. Phys.: Conf. Ser. 150 (2009) 042108.
[19] M. Lahoubi, W. Younsi, M.L. Soltani, B. Ouladdiaf, J. Phys.: Conf. Ser. 200 (2010) 082018.
[20] A.E. Clark, E. Callen, J. Appl. Phys. 39 (1968) 5972.
[21] M. Lahoubi, A. Kihal, G. Fillion, Physical B 284–288 (2000) 1503.