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Magnetoelastic properties of $Nd_6Fe_{13}Cu$ intermetallic compound

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ABSTRACT

Magnetoelastic properties of $Nd_6Fe_{13}Cu$ intermetallic compound are reported. To study the magnetoelastic behaviour of this compound, the thermal expansion as well as the longitudinal (λ_l) and transverse (λ_t) magnetostriction were measured by using the strain gauge method in the selected temperature range of 80–500 K under applied magnetic fields up to 1.5 T. An anomaly and invar-type effects are observed in the linear thermal expansion and $\alpha(T)$ curves at the Néel temperature. The linear spontaneous magnetostriction decreases sharply by approaching the Néel temperature and also shows the short-range magnetic ordering effects when antiferromagnetic–paramagnetic transition occurs. In the low field region, the absolute values of the anisotropic magnetostriction are small and then start to increase with applied magnetic field. Each isofield curve of the anisotropic magnetostriction passes through a minimum and then approaches to zero with increasing temperature. This magnetostriction compensation arises from the difference in the magnetoelastic coupling constants of the sublattices in this compound.

1. Introduction

The ternary intermetallic $R_6T_{13}M$ compounds (R=light rare-earth, T=Fe or Co, M=Cu, Ag, Au, Si, Ga, etc.) are appeared as second phase in the M doped Nd–Fe–B permanent magnets and enhance their coercivity [1–4]. In addition, they absorb a large amount of hydrogen without any change in their symmetry, which accompany with drastic changes in the magnetic properties so that they are recognised as good catalysts [5,6]. Hence, they may provoke some interests in the technological field and fundamental investigations.

These compounds crystallise in the tetragonal $Nd_6Fe_{13}Si$ or $La_6Co_{11}Ga_3$ type structure with the I4/mcm space group. In this structure there are two rare-earth sites R_1 (8f) and R_2 (16l), four Fe sites Fe_1 (4d), Fe_2 (16k), Fe_3 ($16l_1$) and Fe_4 ($16l_2$), and one M site (4a) [2–8]. A complicated crystallographic structure and different competing interactions induce great debates about their spin configuration and magnetic behaviour [9–12].

In the literature, the $Nd_6Fe_{13}Cu$ compound was reported to be an antiferromagnetic (compensated ferrimagnetic) with the Néel temperature (T_N) of 419 K [10]. The Mössbauer study of $Nd_6Fe_{13}Cu$ compound between 85 and 295 K exhibits a basal magnetic anisotropy [13,14] and the opposite sign of the crystal field at two rare-earth sites [5]. Due to the large magnetocrystalline anisotropy of the rare-earth atom (Nd), crystal field and magnetic exchange interactions, it would be informative to study the magnetoelastic properties of this compound. So in this work, the magnetoelastic behaviours of this compound are investigated through thermal expansion and magnetostriction measurements.

2. Experimental

The $Nd_6Fe_{13}Cu$ compound was prepared by arc melting of the pure elements (99.9%) under the purified Ar atmosphere. The sample was remelted in a high-frequency induction furnace equipped with a water-cooled crucible. To assure the homogeneity the ingots were subsequently wrapped in a Ta foil, sealed in an evacuated quartz lump and annealed for 40 days at 550 °C [5].

The crystal structure of the sample was determined by X-ray diffraction using CuK_α radiation, the analysis of XRD pattern was carried out using Fullprof software.

The magnetisation measurements were performed on the compacted powder in presence of magnetic fields up to 7 T by using a commercial extracting sample magnetometer in the temperature ranging from 5 to 300 K.

Thermal expansion and magnetostriction measurements were performed on a disc shape sample with 6 mm diameter and 2 mm thickness using the strain gauge method in the temperature ranging from 80 to 500 K. No difference was observed between the strains measured in the plane or perpendicular to the plane (cooling direction after the melt) of the disc of

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the annealed sample, suggesting the absence of any preferred orientation effects. The linear thermal expansion (LTE) $\Delta l/l(T) = [l(T)-l(80\,\mathrm{K})]/l(80\,\mathrm{K})$ was deduced by measuring the relative change of length of the sample vs. temperature. The magnetostriction was measured (with an accuracy of 2×10^{-6}) parallel (longitudinal magnetostriction, λ_l) and perpendicular (transverse magnetostriction, λ_t) to the field direction, thus allowing to deduce the anisotropic magnetostriction $\Delta\lambda=\lambda_l-\lambda_t$, in magnetic fields up to 1.5 T.

3. Results and discussion

Analysis of the XRD pattern of $Nd_6Fe_{13}Cu$ compound shows the expected tetragonal structure (S.G. I4/mcm) with about 5 wt% Nd_2Fe_{17} impurity phase. From the analysis of the X-ray pattern, the lattice parameters a, c and volume of the unit cell are obtained as 8.096(2) Å, 22.279(1) Å and 1460.360 Å^3, respectively, which are comparable to the reported values [5].

The behaviour of magnetisation vs. field ($M(\mu_0H)$) of this compound is shown in Fig. 1. The small value of the magnetisation at 5 K and 7 T magnetic fields confirms that the magnetic structure consists of at least two magnetic sublattices in which the magnetic moments are somehow canted. The presence of Nd_2Fe_{17} phase increases the magnetisation value at low temperatures but it can be neglected [12].

The temperature dependence of the magnetisation ($M(T)$) of $Nd_6Fe_{13}Cu$ compound at selected fields is shown in Fig. 2. It can be seen that these magnetisation curves show a hill shape region in a temperature range depending on the strength of the magnetic field. The competition between the magnetic anisotropy and the thermal energy changes the canting angle between the magnetic moments and the applied magnetic field. The canting configuration of the moments of sublattices, which causes the enhancement of the resultant magnetisation appears in the magnetisation curves ($M(T)$) in the range from 30 to 150 K. This behaviour has been also reported for some other $R_6Fe_{13}M$ compounds [3,12]. Above 150 K, the magnetisation follows a natural decreasing behaviour due to the thermal fluctuations.

Fig. 3 shows the linear thermal expansion, $\Delta l/l(T)$ of $Nd_6Fe_{13}Cu$ compound, as well as the LTE coefficient, $\alpha(T)=d/dT[\Delta l/l(T)]$, deduced from the slope of the experimental curve of the linear thermal expansion at selected temperatures. The Néel temperature is indicated by the arrow. It can be seen that both $\Delta l/l(T)$ and

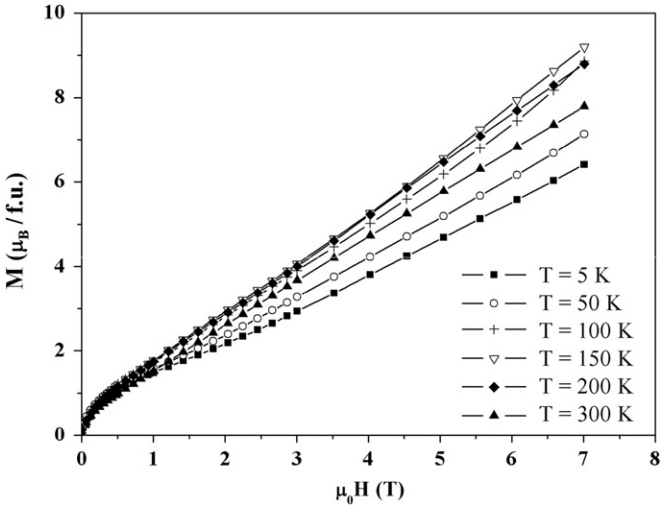

Fig. 1. Field dependence of the magnetisation of $Nd_6Fe_{13}Cu$ compound at different temperatures.

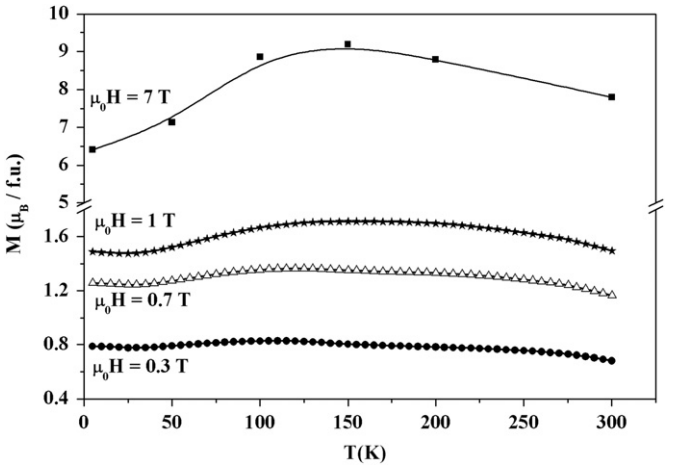

Fig. 2. Temperature dependence of the magnetisation of $Nd_6Fe_{13}Cu$ compound at different applied magnetic fields.

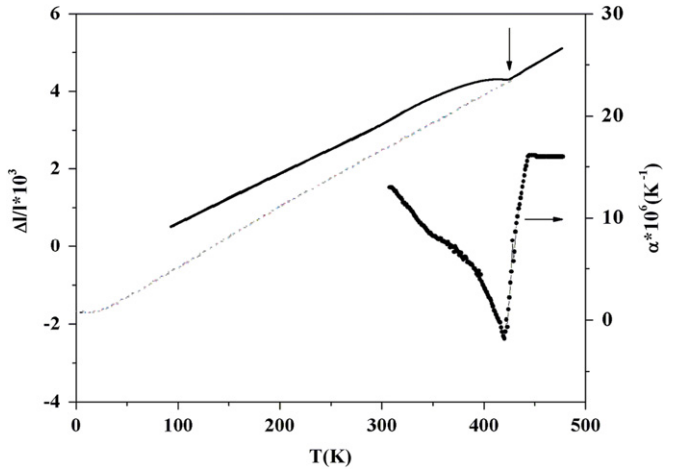

Fig. 3. Experimental thermal expansion $\Delta l/l(T)$ and linear thermal expansion coefficient $\alpha(T)$ curves of $Nd_6Fe_{13}Cu$ compound. Dash line exhibits calculated lattice contribution to the thermal expansion. Arrow indicates the Néel temperature.

$\alpha(T)$ curves show anomaly and invar effect by approaching T_N. The linear thermal expansion coefficient curve beyond the magnetic ordering temperature in the paramagnetic region is almost linear. The observed Néel temperature is approximately 421 K, which is in agreement with the reported result [10].

Below the magnetic ordering temperature the measured linear thermal expansion $(\Delta l/l(T))_{exp}$ of the magnetic materials is the combination of lattice $(\Delta l/l(T))_{latt}$ and magnetic $(\Delta l/l(T))_m$ contributions. The lattice contribution can be calculated using Grüneisen–Debye relation by fitting the experimental measurements in the paramagnetic region, with Debye temperature of $T_D=314$ K [14]. Therefore, the magnetic contribution or the so-called linear spontaneous magnetostriction is the difference between the experimental and the lattice thermal expansion $(\Delta l/l(T))_m=(\Delta l/l(T))_{exp}-(\Delta l/l(T))_{latt}$, which refers to the change of magnetic energy due to the temperature dependence of the crystallographic unit cell volume. The obtained linear spontaneous magnetostriction of $Nd_6Fe_{13}Cu$ compound is presented in Fig. 4. In the $R_6T_{13}M$ family of compounds the short-range magnetic order may originate from the Fe–Fe exchange interactions. These magnetic correlations usually are considered as primary step for expanding a macroscopically ordered magnetic phase over the volume of magnetic materials [15]. Considering the schematic representation of collinear antiferromagnetic

structure in Fig. 5, the short-range magnetic order between nearest neighbour iron atoms can be originated from n_{FeFe} intralayer ferromagnetic exchange interaction. However, the long-range magnetic order starts by activation of the Fe($16l_2$)–Nd(8f) interlayer planar ferromagnetic interaction (n_{RFe}), and will be completed by the antiferromagnetic arrangement of the blocks separated by the nonmagnetic M slabs. Therefore, the linear spontaneous magnetostriction of this compound below T_N=421 K can be attributed to the gradual occurrence of the long-range ordering of the magnetic moments. The small value of linear spontaneous magnetostriction beyond the magnetic ordering temperature may arise from the short-range magnetic ordering that has been observed in several R–T intermetallic compounds [16].

The anisotropic magnetostriction ($\Delta\lambda$) curves as a function of applied magnetic field are shown in Fig. 6. It can be seen that at first the absolute value of $\Delta\lambda$ is small and then it increases with applied field at temperatures below 145 K. Saturation behaviour starts from about 200 K. This trend may be attributed to the temperature dependence of the magnetostriction constants.

The temperature dependence of the anisotropic magnetostriction at selected fields is also represented in Fig. 7. All these curves have similar behaviour in the whole temperature range; the absolute value of $\Delta\lambda$ first increases with temperature up to about 145 K and then decreases to zero at magnetostriction compensation point that occurs above the room temperature.

Previous results on the $R_6Fe_{13}M$ compounds show the existence of two rare-earth sites with the opposite anisotropy. This

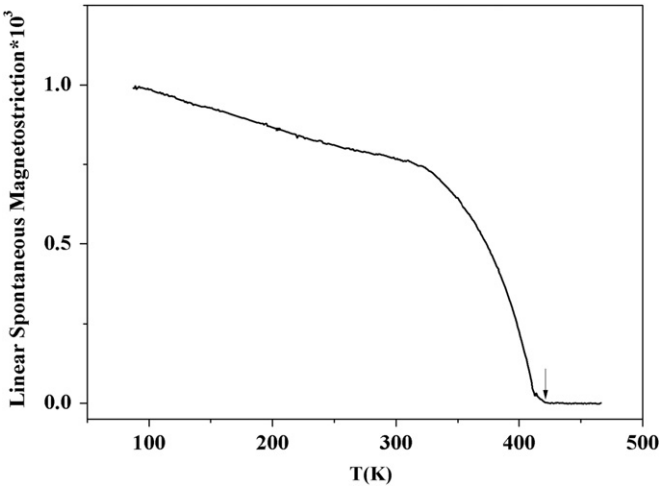

Fig. 4. Temperature dependence of the linear spontaneous magnetostriction of $Nd_6Fe_{13}Cu$ compound. Arrow indicates the beginning of paramagnetic behaviour.

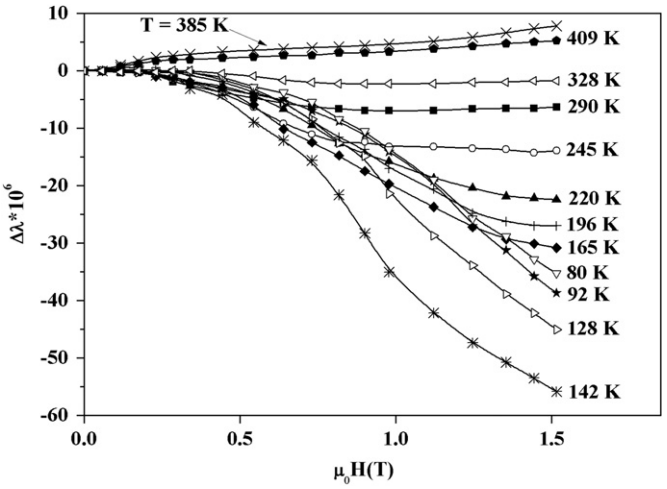

Fig. 6. Isothermal curves of the anisotropic magnetostriction of $Nd_6Fe_{13}Cu$ compound as a function of applied field at selected temperatures.

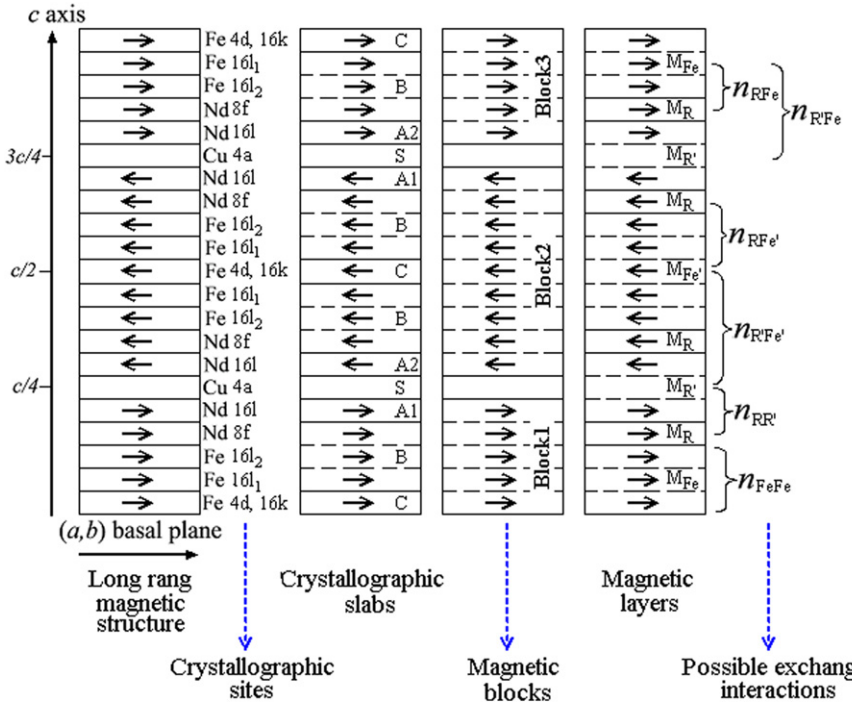

Fig. 5. Conventional concepts in description of the crystal and magnetic structure of $Nd_6Fe_{13}Cu$ compound based on Mössbauer spectral studies [13,14,15]. Estimation of the exchange field parameters shows that $|n_{RFe}|(|n_{R'Fe'}|) < |n_{RR'}| < |n_{FeFe'}| \ll |n_{RFe}| < |n_{FeFe}|$ and $n_{RFe} \approx n_{RR} \approx 0$. Consequently, the R(8f) moments are strongly ferromagnetically coupled to the Fe moments while R'(16l) ones coupled more weakly than R(8f) moments.

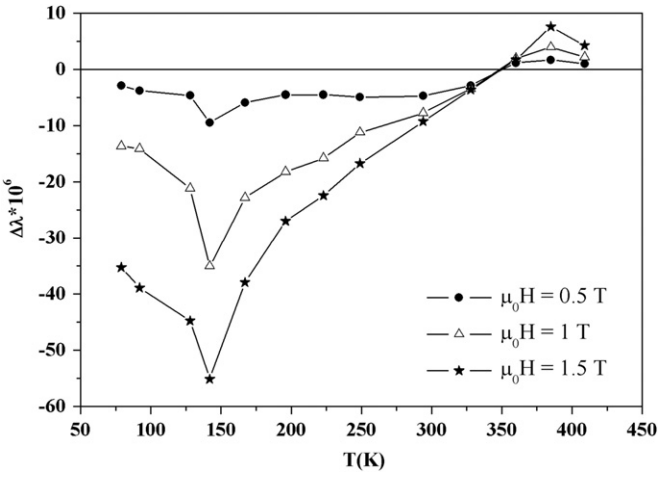

Fig. 7. Temperature dependence of the anisotropic magnetostriction of $Nd_6Fe_{13}Cu$ compound at selected applied fields.

means that these two sites have different easy directions (the 16l and 8f sites exhibit uniaxial and nearly planar anisotropy, respectively) [5,17]. So, the magnetostrictive strains with different sign can be expected at these two Nd sites. It is also found that the compensation point and spin reorientation transition coincide in $Nd_6Fe_{13}Si$ compound [18]. Below T_{SR}, the overall spin arrangement of the Nd(8f) and Fe atoms is planar. From these experimental facts we can deduce the magnetostrictive strain sign of the two Nd sites and the Fe sublattice. The value of $\Delta\lambda$ in $Nd_6Fe_{13}Si$ below T_{SR} is negative. Since the anisotropy of the rare-earth sublattice dominates at low temperatures, the Nd(8f) site with planar anisotropy creates negative strain. Above T_{SR} and at high temperatures, the Nd(16l) site and Fe sublattice with axial anisotropy have positive contribution to the $\Delta\lambda$ results.

By comparing the anisotropic magnetostriction results of $Nd_6Fe_{13}Si$ with that of the $Nd_6Fe_{13}Cu$ compound in Fig. 7, it can be concluded that by increasing the temperature the negative contribution of Nd(8f) site in $\Delta\lambda$ increases up to about 145 K, where $\Delta\lambda$ curves pass through minima. Then, as the temperature increases, the positive contribution of the Fe sublattice in magnetostriction compete more with negative contribution of Nd(8f) site. Finally, these two contributions compensate each other and the anisotropic magnetostriction goes to zero ($T_0 \approx 350$ K). Similar magnetostriction compensation behaviour has also been reported for $Pr_6Fe_{11}Ga_3$ [15]. After passing the compensation point the positive value of Fe planar sublattice to $\Delta\lambda$ increases and dominates that of the Nd(8f) site. By approaching ordering temperature $\Delta\lambda$ becomes zero.

4. Conclusion

The temperature dependence of magnetisation curves of $Nd_6Fe_{13}Cu$ compound shows a hill shape region and this may be attributed to the canting configuration of the magnetic moments of the sublattices. This canting configuration originates from the competition between the magnetic anisotropy and the thermal energy, which changes the canting angle between the magnetic moments and the applied magnetic field. Also in this compound the linear thermal expansion and magnetostriction behaviour were investigated. Well defined anomaly and invar-type effect are observed in the linear thermal expansion and $\alpha(T)$ curves at the Néel temperature. The linear spontaneous magnetostriction decreases sharply by approaching the Néel temperature (421 K), which is due to the short and long-range magnetic ordering when antiferromagnetic to paramagnetic transition occurs. Temperature dependence curves of anisotropic magnetostriction pass through minima at about 145 K and then go to zero at compensation temperature ($T_0 \approx 350$ K). This behaviour shows that the anisotropy of the Nd and Fe sublattices with different sign compete with each other.

References

[1] F. Weitzer, A. Leithe-Jasper, P. Rogl, K. Hiebl, A. Rainbacher, G. Wiesinger, W. Steiner, J. Fried, F.E. Wagner, J. Appl. Phys. 75 (1994) 7745.
[2] A. Leithe-Jasper, P. Rogl, G. Wiesinger, A. Rainbacher, R. Hatzl, M. Forsthuber, J. Magn. Magn. Mater. 170 (1997) 189.
[3] Q.F. Xiao, T. Zhao, Z.D. Zhang, M.H. Yu, X.G. Zhao, W. Liu, D.Y. Geng, X.K. Sun, F.R. de Boer, J. Magn. Magn. Mater. 184 (1998) 330.
[4] O. Isnard, G.J. Long, D. Hautot, K.H.J. Buschow, F. Grandjean, J. Phys. Condens. Matter 14 (2002) 12391.
[5] K.G. Knoch, A. Le Calvez, Q. Qi, A. Leithe-Jasper, J.M.D. Coey, J. Appl. Phys. 73 (1993) 5878.
[6] J.M.D. Coey, Qinian Qi, K.G. Knoch, A. Leithe-Jasper, P. Rogl, J. Magn. Magn. Mater. 129 (1994) 87.
[7] F. Wang, J. Wang, P. Zhang, B.G. Shen, Q. Yan, L. Zhang, Physica B 269 (1999) 17.
[8] P.S. Papamantellos, K.H.J. Buschow, C.H. de Groot, F.R. de Boer, C. Ritter, F. Fauth, Grit Boettger, J. Alloys Compd. 280 (1998) 44.
[9] S.J. Kennedy, E. Wu, F.W. Wang, P.L. Zhang, Q.W. Yan, Physica B 276–278 (2000) 622.
[10] C.H. de Groot, K.H.J. Buschow, F.R. Boer, Phys. Rev. B 57 (1998) 11472.
[11] P. Schobinger-Papamantellos, K.H.J. Buschow, C. Ritter, J. Alloys Compd. 359 (2003) 10.
[12] F. Grandjean, G.J. Lon, M. Guillo, O. Isnard, K.H.J. Buschow, Phys. Condens. Matter 16 (2004) 4347.
[13] D. Hautot, G.J. Long, F. Grandjean, C.H. de Groot, K.H.J. Buschow, J. Appl. Phys. 81 (1997) 5435.
[14] D. Hautot, G.J. Long, F. Grandjean, C.H. de Groot, K.H.J. Buschow, J. Appl. Phys. 83 (1998) 1554.
[15] M.R. Alinejad, N. Tajabor, F. Pourarian, J. Magn. Magn. Mater. 320 (2008) 2140.
[16] J.L. Wang, M.R. Ibarra, C. Marquina, B. García-Landa, O. Tegus, Q.F. Xiao, E. Brück, F.M. Yang, G.H. Wu, J. Appl. Phys. 91 (2002) 8216.
[17] Hong-Shuo Li, Bo-Ping Hu, J.M. Cadogan, J.P. Gavigan, J. Appl. Phys. 67 (1990) 4841.
[18] N. Tajabor, M.R. Alinejad, F. Pourarian, Physica B 321 (2002) 60.