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Orientation relations and twinning in heterostructures $YBa_2Cu_3O_x$//$NdGaO_3$ and $YBa_2Cu_3O_x$//CeO_2//Al_2O_3

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Abstract

Epitaxial $YBa_2Cu_3O_{7-x}$ (YBCO) thin films on (110) $NdGaO_3$ and (100) CeO_2//$(1\bar{1}02)Al_2O_3$ substrates were studied with X-ray diffraction methods, orientation features of film twinning were determined. Films on both substrates were mainly *c*-oriented with $c = 11.67$ Å. Orthorhombic structure of $NdGaO_3$ results in increase of the angle between (110) and $(1\bar{1}0)$ twinning planes in YBCO films to 90.20° and in difference in volume of two twin domain systems. About 60% of YBCO film on CeO_2//Al_2O_3 substrates show no twinning. The lattice direction $\langle 1\bar{1}02\rangle$ of the Al_2O_3 substrate and $\langle 100\rangle$ of the CeO_2 film were inclined by 0.15°; this inclination can result from high lattice mismatch between CeO_2 film and Al_2O_3 substrate. The $\langle 001\rangle$ direction of the YBCO film was inclined to $\langle 1\bar{1}02\rangle$ Al_2O_3 direction only by 0.06°. The spread of in-substrate-plane misorientation in CeO_2 film of 1.65° was also higher than that of the YBCO film (1.22°).

Keywords: Thin films; Multilayes; Twinning; Domains; X-ray diffraction

1. Introduction

Modern technology of deposition of the superconductive $YBa_2Cu_3O_{7-x}$ (YBCO) thin films provides films of crystal structure close to the structure of single crystals. Most results on high-T_c superconducting thin films were obtained with YBCO grown with the *c*-axis normal to the surface of different substrates. Similar to single crystals, twinning in *c*-oriented films occurs on the $\{110\}/\langle 1\bar{1}0\rangle$ scheme with an angle of twinning about 1° [1,2]. The type of twinning structure of YBCO films is correlated with the structure of the substrate. In order to interpret the transport property such as the critical current density, the twin structure in the *a*–*b* plane is very important.

We present results of comparative studies of twinning structure in two *c*-oriented YBCO films on (110) $NdGaO_3$ and (100)CeO_2//$(1\bar{1}02)Al_2O_3$ substrates with similar property.

2. Experimental

YBCO films (1000–1500 Å thick) were deposited with the DC-sputtering of a stoichiometric YBCO target at high oxygen pressure [3] on (110) $NdGaO_3$ and r-cut sapphire substrates. Sapphire substrates were covered with a thin (300 Å) cerium oxide (CeO_2) buffer layer to prevent aluminium diffusion from the substrate into the film. The deposition

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conditions of CeO_2 provided formation of the buffer layer in (001) orientation. The film deposition parameters (substrate temperature, oxygen pressure, and discharge current density) were optimised to obtain best superconducting properties of the resulting film.

X-ray studies were performed using Siemens D500 and DRON-3M diffractometer systems. Both symmetric and asymmetric diffraction geometries were used. To study twinning in *c*-oriented YBCO films, it is necessary to observe reflections from crystallographic planes, tilted to (001) planes. The chosen (103) and (113) reflections are of the most intensive reflections in the YBCO structure.

Relative positions of diffraction peaks from corresponding planes provide the twinning angle value and the value of the angle between substrate and YBCO crystallographic planes. These angles were calculated using

$$\delta = \alpha\cos(\gamma) + \beta\sin(\gamma), \quad (1)$$

where δ — measured misorientation angle between film grains (twin domains), α and β — angles of misorientation between these grains in selected mutually perpendicular planes, γ — angle between planes corresponding to δ and α. α and β axes were chosen in substrate plane and normal to substrate surface.

Grain misorientation spread is present in two perpendicular directions: normal to the substrate sur-

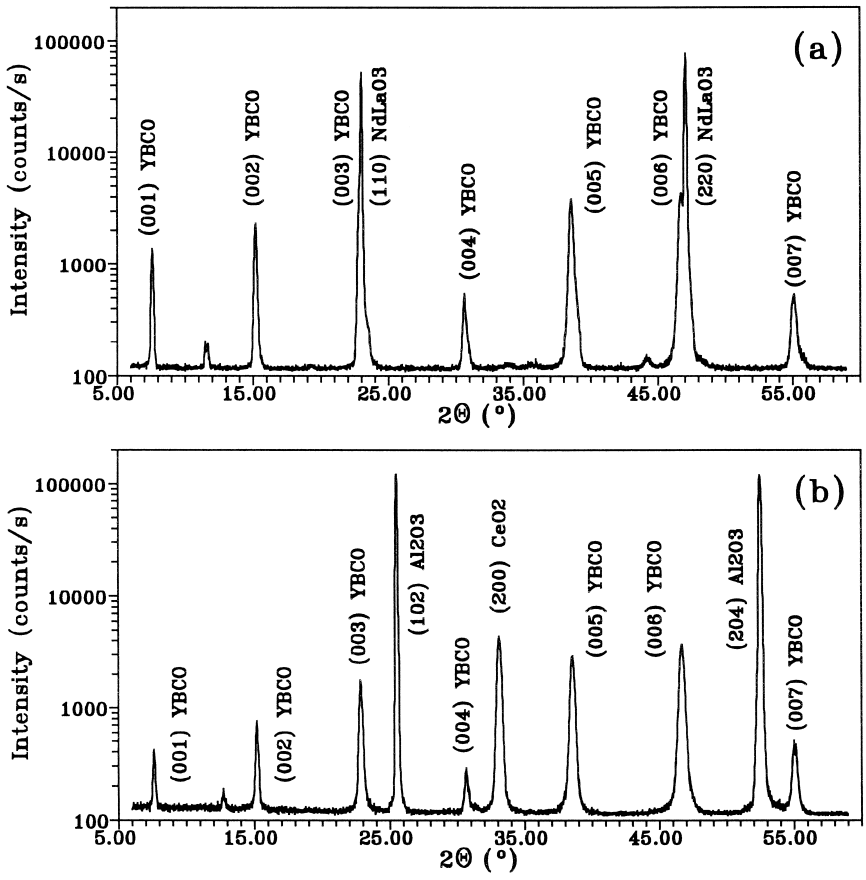

Fig. 1. X-ray diffraction θ–2θ scans of *c*-oriented YBCO films on (110) $NdGaO_3$ (a) and on (1102) Al_2O_3 with (100) CeO_2 buffer layer (b).

face and in the substrate plane. Values of this decomposition can be found as follows

$$\Delta\delta^2 = \Delta\alpha^2\cos^2(\chi) + \Delta\beta^2\sin^2(\chi), \quad (2)$$

where $\Delta\delta$ — angle of grain misorientation spread from the δ axis, $\Delta\alpha$ and $\Delta\beta$ — grain misorientation spread angles from the mutually perpendicular axes, χ — angle between axes δ and α.

3. Results and discussion

X-ray diffraction $\theta/2\theta$-scans of the YBCO films on (110) $NdGaO_3$ and on (001) CeO_2 //(1$\bar{1}$02) Al_2O_3 are presented on Fig. 1. The obtained films were epitaxial with the *c*-axis oriented normal to the substrate (*c*-oriented film).

Film lattice parameters were determined using (0 0 13), (3 0 10), and (0 3 10) reflections. Values $a = 3.827(1)$ Å, $b = 3.889(1)$ Å, $c = 11.674(2)$ Å were found for films on (110) $NdGaO_3$, while on CeO_2 //Al_2O_3, these were $a = 3.830(2)$ Å, $b = 3.880(2)$ Å, $c = 11.670(2)$ Å. Low *c* parameter level supposes high oxygen contents in both films [4,5].

3.1. YBCO film twinning on (110) $NdGaO_3$

θ-scan diffraction pattern in vicinity of (113) YBCO film reflection is shown on Fig. 2 and presents a distinctive picture of YBCO twinning. The obtained pattern can be arithmetically decomposed into four diffraction curves. *A* and *A*′ curves of this decomposition correspond to different twinning systems of (1$\bar{1}$0) plane, *B* curve results from other two twinning orientation on (110) plane, showing no splitting, and *C* curve corresponds to (020) $NdGaO_3$ planes. Relative peak positions reflect misorientation of corresponding planes. Diffraction patterns Fig. 2 allow determination of mutual orientation of $NdGaO_3$ and YBCO atomic planes (Fig. 3a) as well as mutual orientations of twinning parts. Twinning angle was calculated using Eq. (1), where γ is the angle between (113) and (110) planes in the YBCO lattice, $\gamma = 35°$. For the investigated YBCO film on $NdGaO_3$ we obtain $\beta = 0°$ and $\delta = 1.39°$ (angle between *A*

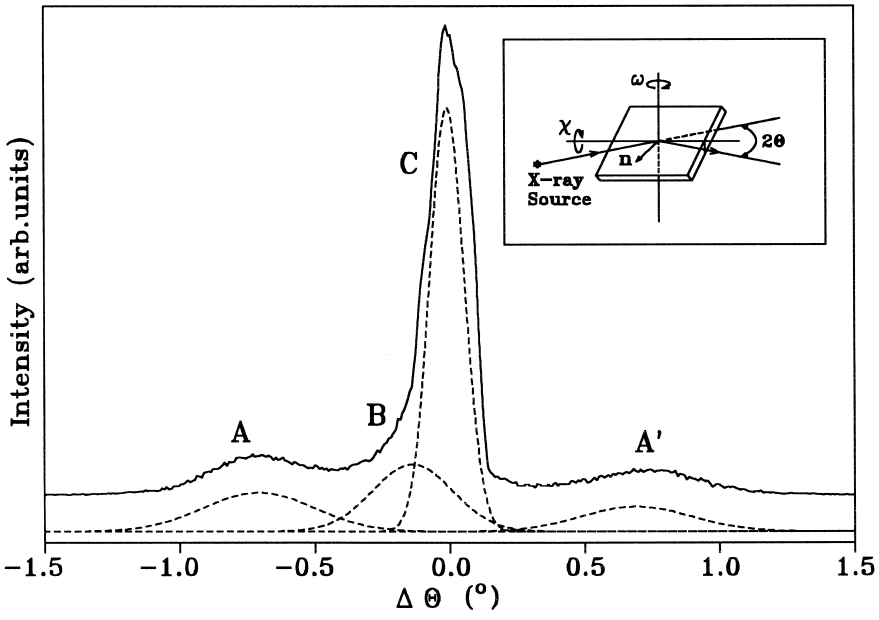

Fig. 2. X-ray diffraction θ-scans axis of (113) reflection of *c*-oriented YBCO films on (110) $NdGaO_3$. On the insert: scheme of the experiment.

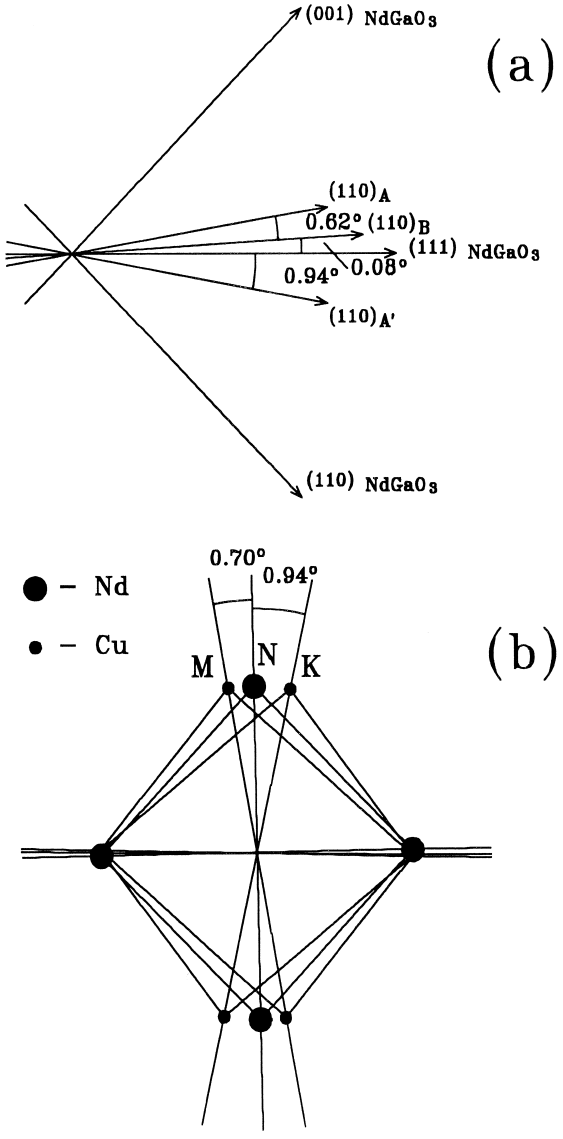

Fig. 3. (a) Scheme of atomic planes of YBCO film on (110) $NdGaO_3$ substrate. (b) Scheme of mutual location of atoms in (001) YBCO plane and in (110) $NdGaO_3$ planes.

and A' peaks and α — double twinning angle). This gives a twinning angle of 0.85°, corresponding to $a-b = 0.057$ Å. The $a-b$ value calculated from lattice parameters gives 0.061 Å in good agreement with the previous evaluation.

Application of Eq. (1) in a similar way allows evaluation of the angle between twinning YBCO planes from mutual positions of A, A', and B peaks, equal to 90.20°. Such discrepancy from 90° was also observed in Ref. [2] in YBCO film on $NdGaO_3$. The increase of the angle between twinning planes can be explained by symmetry of $NdGaO_3$ lattice. Lattice in (110) plane of $NdGaO_3$ is close to tetragonal with angle between (111) and (001) planes of 44.94° (measured $NdGaO_3$ lattice parameters were $a = 5.428(2)$ Å, $b = 5.499(2)$ Å, $c = 7.711(3)$ Å).

Knowledge of $NdGaO_3$ and YBCO lattice parameters allows determination of mutual atom positions in the (001) plane of the film and (110) plane of the substrate (Fig. 3b). Distance between the M atom of the A film twin orientation and N substrate atom corresponds to distance between the K atom of the A' film twin orientation and the same N substrate atom. These distances can be calculated and are $R_1 = 0.047$ Å and $R_2 = 0.063$ Å, correspondingly. With increase of the twin domain size, the discrepancy between film and substrate atom positions will increase also. This strain will increase faster for the A' twin domain, resulting in smaller size of the twin domain. This effect can be seen in Fig. 2: the ratio of A and A' peak amplitudes, corresponding to the volume of the different twin domain systems, is about 1.7. The evaluation of the volume ratio can be given as $(R_2/R_1)^2 = 1.8$, that is close to the experimentally observed value.

3.2. YBCO film twinning on (001) CeO_2//(1$\bar{1}$02) Al_2O_3

$\theta/2\theta$-scans of YBCO films on Al_2O_3 with CeO_2 buffer layer showed only (00l) peak family, supposing absence of a-oriented grains (Fig. 1). Presence of b-orientation can be checked by comparison of relative intensities of the (00l) reflections [6]. The (010) and (020) reflections will increase the (003) and (006) peak intensities from the theoretically calculated values for totally c-oriented film. This will result in change of the intensity ratios $I(005)/I(003)$ and $I(005)/I(006)$, compared with the theoretical values. In our case, the difference between observed intensity ratios and theoretical for $x = 0.1$ [7] is negligible, so probably b-orientation is also absent in the studied film.

Twinning in YBCO films on Al_2O_3 with CeO_2 buffer layer does not reveal in the distinctive splitting (Fig. 4b). Diffraction curves are more narrow

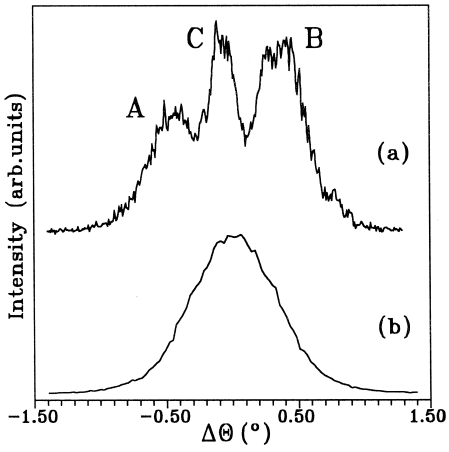

Fig. 4. (a) X-ray diffraction θ-scans of (103) reflection of YBCO c-oriented films on (110) $NdGaO_3$. A and B correspond to two twin orientations for different (110) and $(1\bar{1}0)$ twinning planes. C peak corresponds to diffraction from $(11\bar{2})$ $NdGaO_3$ planes. (b) X-ray diffraction θ-scans of (103) reflection of c-oriented YBCO films on $(1\bar{1}02)$ Al_2O_3 with (100) CeO_2 buffer layer.

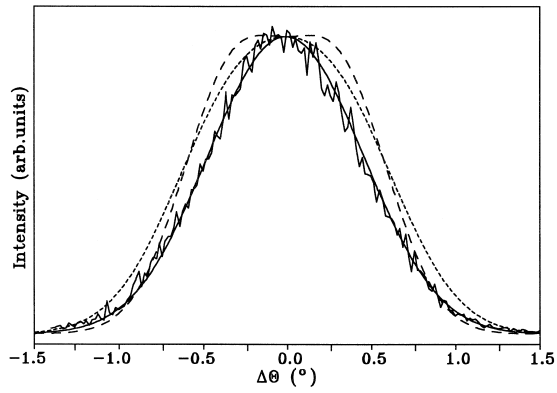

Fig. 5. X-ray diffraction θ-scans of YBCO c-oriented film on $(1\bar{1}02)$ Al_2O_3 with (100) CeO_2 buffer layer, (103) reflection. Solid line — theoretical calculation in supposition of 40% twinned film and 1.1° grain misorientation; dotted line — 100% and 1.1°; dashed line — 100% and 0.8°.

than for YBCO films on $NdGaO_3$, if we suppose merging of twin peaks into one broad peak. The $n(hkl)$ reflection width does not change with the increase of n, supposing broadening of the curve mainly due to grain misorientation, rather then size of domains or film defects [8].

The obtained X-ray diffraction curve was analysed in supposition of presence of twinned and not twinned YBCO phases in the studied film. Ratio of twinned and not twinned phase volumes, film grain misorientation angle, and parameter of orthorombicity have been varied. The best approximation was obtained with lattice parameters $a = 3.830$ Å, $b = 3.880$ Å, $c = 11.670$ Å, 1.1° misorientation angle and 40% part of the film twinned (Fig. 5). Similar calculation of twinned phase concentration was performed for films containing a-oriented inclusions. The a-oriented inclusions gave contribution into the not twinned phase reflections.

Presence of the not twinned phase in 100% c-oriented film can be explained by following possible reasons.

(a) The interface between twins can be broadened, and transformation from one twin orientation into another can form a phase with smaller twinning angle [9].

(b) The size of CeO_2 grain is smaller than size of a twin domain in the YBCO film (about 500 Å). In this case, the size of YBCO grains will be determined by the CeO_2 grain size and twinning in such small grains will be hardly probable.

X-ray diffraction θ-scans of (005) YBCO and (200) CeO_2 reflections are shown on Fig. 6. Deposi-

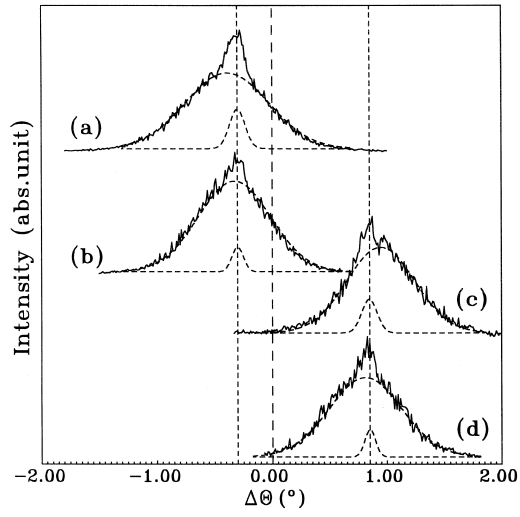

Fig. 6. X-ray diffraction θ-scans of (200) CeO_2 reflection (a,c) and (005) YBCO (b,d) in symmetric diffraction geometry. (a,b) and (c,d) scans were obtained after rotation of the sample 90° in substrate plane. CeO_2 deposition temperature 770°C. Dashed lines — position of normal to substrate (0.0°) and positions of $(1\bar{1}02)$ Al_2O_3.

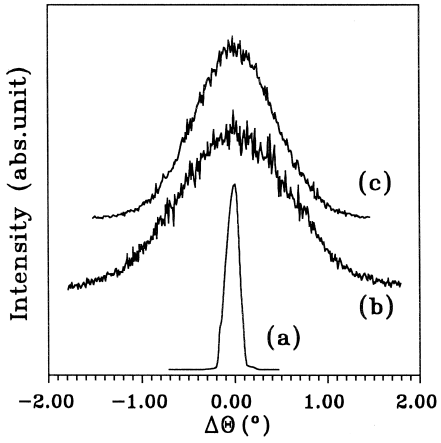

Fig. 7. X-ray diffraction θ-scans of YBCO//CeO_2 //Al_2O_3 film. (a) (11$\bar{2}$0) Al_2O_3 reflection; (b) (111) CeO_2 reflection, $\chi = 55°$; (c) (10$\bar{3}$) YBCO reflection, $\chi = 45°$.

tion temperature is 770°C for CeO_2 and 695°C for YBCO. Two diffraction scans in perpendicular directions are necessary to determine an unambiguous orientation and misorientation spread parameters of the film (Fig. 6a–d). Analysis of these diffraction patterns shows that (1$\bar{1}$02) Al_2O_3 direction is tilted from the substrate normal by 0.9°. The main (001) direction of YBCO is inclined by 0.06° and (100) CeO_2 direction is inclined by 0.15° to the (1$\bar{1}$02) Al_2O_3 direction. Position of normal to substrate corresponds to the dotted line on Fig. 6. Misorientation of grains can be evaluated as peak width on X-ray diffraction patterns Fig. 6a–d, and was equal both for YBCO film and for CeO_2 buffer layer in all scan directions (0.82–0.83°). Applying Eq. (2) for diffraction patterns Fig. 6a–c and Fig. 7 grain misorientation in substrate plane can be calculated, being 1.65° for CeO_2 buffer layer and 1.18–1.29° for YBCO film. Incomplete (40%) twinning of YBCO films results in uncertainty in the grain orientation spread calculation [10]. The YBCO grain orientation differs for different twin domain systems, increasing observed reflection width by 0.1°. Actual misorientation is smaller than calculated value of 1.18°–1.29°. The misorientation of YBCO film is sufficiently smaller than that of CeO_2 buffer layer; this effect can be explained if seeding of YBCO occurs only on CeO_2 grains of some certain orientation.

4. Conclusions

Twin structure and domain orientations of the YBCO films on (110) $NdGaO_3$ and (100) CeO_2 //(1$\bar{1}$02) Al_2O_3 were been investigated. Comparison of twinning structures of YBCO films with close crystallographic parameters, but on these different substrates shows particularities resulting from the nature of substrates. Twinning orientation features of the YBCO film on (110) $NdGaO_3$ correlated with symmetry substrate.

Presence of the not twinned phase in *c*-oriented (100) CeO_2 //(1$\bar{1}$02) Al_2O_3 film can be helpful to possibility of reducing twinning degrees and quantity of twinning borders in large part of film.

Study of mutual orientations of layers in YBCO//CeO_2 //Al_2O_3 heterostructures reveals inclinations of crystallographic planes between neighbour layers. Grain misorientation spread from the substrate normal is equal for all layers, but in substrate plane spread of grain misorientation is greater for the buffer CeO_2 layer. This effect proves significant influence of the sapphire substrate on the YBCO film. Further studies are necessary to find out the nature of this substrate–film interaction.

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References

[1] T. Scheme, P. Marienhoff, R. Herwig, M. Neuhaus, W. Jutzi, Physica C 197 (1992) 79.
[2] T. Steinborn, G. Miehe, J. Wiesner, E. Brecht, H. Fuess, G. Wirth, B. Schulte, M. Speckmann, H. Adrian, M. Maul, K. Petersen, W. Blau, M. McConnel, Physica C 220 (1994) 219.
[3] P.B. Mozhaev, G.A. Ovsyannikov, S.N. Polyakov, E.K. Kov'ev, N.P. Kukhta, Supercond. Phys. Chem. Technol. 9 (1997) 304, (in Russian).

[4] M.S. Osofsky, J.L. Cohn, E.F. Skelton, M.M. Miller, R.J. Soulen Jr., S.A. Wolf, T.O. Vanderah, Phys. Rev. B 45 (1992) 4916.
[5] J.D. Jorgensen, B.W. Veal, A.P. Paulikas, L.J. Nowicki, G.W. Crabtree, H. Claus, W.K. Kwok, Phys. Rev. B 41 (1990) 1863.
[6] C. Dubourdieu, J.P. Senateur, O. Thomas, F. Weiss, B.P. Thane, M. Brunel, Appl. Phys. Lett. 69 (1996) 1942.
[7] J. Ye, K. Nakamura, Phys. Rev. B 48 (1993) 7554.
[8] J.P. Gong, M. Kawasaki, K. Fujito, R. Tsuchiya, M. Yoshimoto, H. Koinuma, Phys. Rev. B 50 (1994) 3280.
[9] Y.A. Osip'yan, A.N. Afonikova, T.S. Parsamyan, V.S. Shehkman, I.M. Shmyt'ko, Pis'ma Zh. Eksp. Teor. Fiz. 47 (1988) 501, (in Russian).
[10] I.K. Bdikin, A.D. Mashtakov, P.B. Mozhaev, G.A. Ovsyannikov, Fiz. Tverd. Tela 40 (1998) 609, (in Russian).