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Research on processes of texture formation in "NiW substrate and TiN coating" system and creation of the new type textured paramagnetic substrates for HTS based on $YBa_2Cu_3O_7$

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The paper investigates the processes of texture formation in thin-film two-component system based on paramagnetic Ni — 9.5 at. % W substrate with TiN coating. It is studied the influence of deposition time and pressure of nitrogen on the cubic texture formation in both components of "NiW/TiN" system by using XRD methods. It is observed the effect of re-orientation of crystal planes in the Ni — 9.5 at. % W tape exposed the influence of the coating that leads to a substantial enhancing the cubic texture of the paramagnetic substrate. It is also established that optimization of the conditions of thin TiN layer deposition makes it possible to obtain quasi single crystalline TiN coatings with cubic texture. These textured Ni — 9.5 at. % W/TiN substrates admit the epitaxial growth of high quality HTS films with the high current carrying capacity.

Keywords: 2G HTS; texture; Ni — 9.5 at. % W alloy; TiN buffer layer; paramagnetic substrate.

Изучены процессы текстурообразования в тонкопленочной двухкомпонентной системе на основе металлической подложки из парамагнитного сплава Ni — 9.5 at. % W с буферным покрытием TiN. Методами рентгеноструктурного анализа исследовано влияние времени осаждения и давления паров азота на процесс формирования кубической текстуры в обеих компонентах системы "NiW/TiN". Обнаружен эффект переориентации кристаллических плоскостей в ленте сплава Ni — 9.5 at.% W под воздействием покрытия, ведущий к существенному усилению кубической текстуры подложки. Показано, что при оптимизации условий нанесения тонкослойного покрытия в системе "Ni — 9.5 at.% W/TiN" имеет место формирование квазимонокристаллических слоев TiN с кубической текстурой.

Дослідження процесів формування текстури в системі "NiW підложка та TiN покриття" і створення сарамагнітних сідложек сового типу для високотемпературних надпровідників на основі YBa2Cu3O7. *М.С.Сунгуров, С.О.Леонов, Т.В.Сухарєва, В.В.Дерев'янко, В.О.Фінкель, Ю.М.Шахов.*

Вивчено процеси формування текстури у тонкошаровій двокомпонентній системі на основі металевої підкладки з парамагнітного сплаву Ni — 9,5 ат. % W із буферним покриттям TiN. Методами рентгеноструктурного аналізу досліджено вплив часу осадження і тиску пари азоту на процес формування кубічної текстури в обох компонентах системи "NiW/TiN". Виявлено ефект переорієнтації кристалічних площин у стрічці сплаву Ni — 9.5 ат.% W під впливом покриття, що призводить до істотного посилення кубічної текстури підкладки. Показано, що при оптимізації умов нанесення тонкошарового покриття у системі "Ni — 9.5 ат.% W/TiN" має місце формування квазімонокристалічнх шарів TiN з кубічної текстурою.

1. Introduction

Research in the field of high-temperature superconductors of the second generation (2G HTS) based on HTS YBCO textured films is of particular interest because it opens up the new perspectives for creating the conductors, which could work at strong magnetic fields and temperature of liquid nitrogen boiling point (77.4 K) [1–4].

Architecture of 2G HTS with crucial current density j_c ~ 10^6 A/cm^2 at the liquid nitrogen temperature consists of:

— substrate (a thin tape mostly of Ni–W alloys with different compositions);

— buffer layer/layers (oxides, nitrides, in particular TiN);

— a single crystalline superconducting $YBa_2Cu_3O_{7-\delta}$ with T_c ~ 92 K.

Elements of such superconducting system for effective functioning should avoid ferromagnetic hysteresis losses in alternating current applications and should be textured according to the scheme $\{100\}_{NiW}\|\{100\}_{TiN}\|\{100\}_{YBCO}$. This scheme provides the formation of biaxial cube texture in the film of HTS.

Therefore one of the main challenges for the coated conductors is to produce paramagnetic alloy substrates with the high mechanical characteristics and sharp cubic texture. The paramagnetism property is provided by increasing the concentration of tungsten in Ni–W alloy up to 9 at. % W and higher. However, in this case the problem occurs with formation of the sharp cube texture, which is hindered by low energy of stacking faults E_{sf}. The E_{sf} value decreases with increasing the content of alloying element in the alloy [5]. Thus the carrying out a fundamental research of texture formation processes in the system "metallic substrate — buffer coating" is essential for achieving the significant progress in the field of effective coated superconductors creation.

Earlier the new effects of texture formation in both components of the system of "Ni–W/TiN" were found in our investigations [6–8]. The main purpose of the current work is to investigate the nature of the texture formation processes in the "Ni — 9.5 at. % W/TiN" system.

To achieve the work goal it is necessary to develop the technology of the substrate construction in the following areas:

— creating the high-quality metal tape based on Ni — 9.5 at.% W paramagnetic alloy;

— optimization of conditions for TiN coating deposition.

2. Experimental

Preparation of substrates was carried out according to scheme which includes the following steps: 1) synthesis of Ni–W alloys with different concentration of tungsten (0–9.5 at. % W); 2) thin-layer tape production; 3) high-temperature treatment of Ni–W tapes; 4) deposition of TiN coating; 5) XRD analysis.

Initial materials for obtaining the Ni–W alloys were Ni and W powders with 99.98–99.99 % purity (by metallic impurities). The following methods were used for the purification from gaseous impurities (the main impurity is oxygen represented as nickel and tungsten oxides): 1) the heat treatment at temperatures ~ 850°C for purification of Ni powder; 2) for the refinement of W powder it was applied the high-temperature treatment (1000–1200°C) in reducing Ar + 4 % H_2 gaseous mixture flow [9]. The paramagnetic alloy was synthesized by means of powder metallurgy in deep vacuum (p ~ 10^{-6} Torr) at T = 1200°C during t = 4 h. Obtained ingots were rolled up to 50–100 μm at the room temperatures to perform the metallic tapes. The total degree of cold-rolling deformation was about 95 %. Resulting operation during the tape production was high-temperature annealing at T = 1150°C during t = 2 h.

Thin layers of titanium nitride (TiN) on the surface of Ni — 9.5 at. % W tapes were obtained by method of ion-plasma deposition [9]. Specific parameters of the TiN buffer layer deposition varied within the following ranges: negative substrate potential U = 50–300 V; arc current I = 80 A; temperature of substrate t_s ~ 450°C; nitrogen pressure in chamber $p(N_2)$ = 1.2–6.2·10^{-2} Torr; deposition time t_{TiN} = 60–900 s.

XRD analysis (Cu Kα radiation) was used to solve the following tasks: determination

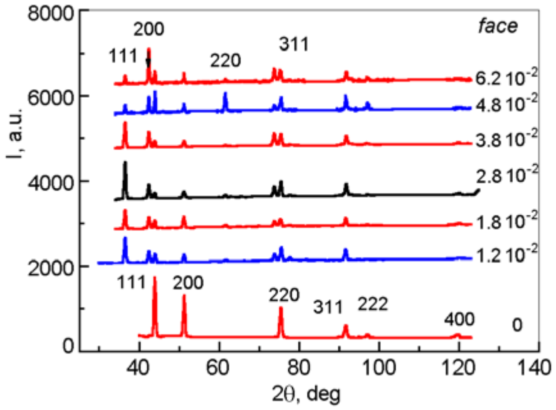

Fig. 1. XRD patterns of FRONT side of Ni — 9.5 at. % W substrate with TiN coating at different pressures of nitrogen $p(N_2)$ = 1.2–6.2·10^{-2} Torr.

of the phase composition of the system components of the substrate + coating; determination of lattice parameters of Ni–W and TiN; analysis of the texture of the substrate and coating according to the method developed in our paper [8]; determination of the TiN coating thickness. The method of determination the TiN-layer thickness is based on X-ray absorption. Intensity of the beam reflected from crystal plane (*hkl*) of the sample with the coating thickness *h* is as follows:

$$I_{hkl}(h) = I_{hkl}(0) \cdot \exp(-2h \cdot \mu_{TiN}/\cos(\theta)), \quad (1)$$

$I_{hkl}(h)$ — intensity of the beam reflected from the substrate with coating;

$I_{hkl}(0)$ — intensity of the beam reflected from the substrate without coating;

h — coating layer thickness;

μ_{TiN} — the linear absorption coefficient of the coating material;

θ — the Bragg angle.

Equation (1) and mathematical modeling of the relative intensity of some X-ray interferences, in our case — the cubic plane ($h00$) of the Ni–W substrate, makes it possible to determine the coating thickness h_{TiN} within ~ 10 %.

3. Results and discussion

Main results were obtained by the studying of Ni — 9.5 at. % W tapes with thin TiN coating. In accordance with the work purpose the research program implemented in the present study had two directions:

1. The TiN coating deposition at a constant time t_{TiN} ~ 600 sec in a wide range of pressures of nitrogen $p(N_2)$ = 1.2–6.2·10^{-2} Torr for optimization.

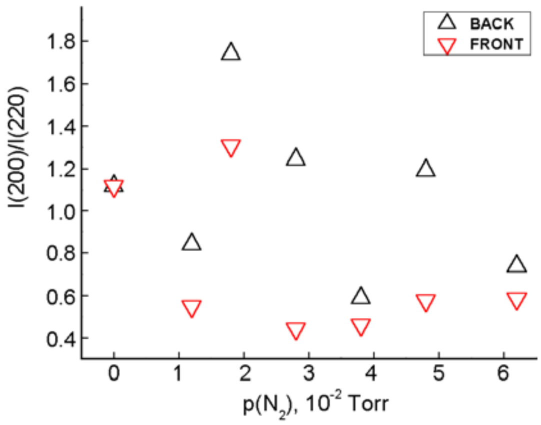

Fig. 2. Values of the ratio I_{200}/I_{220} for FRONT and BACK sides of Ni — 9.5 at. % W substrate after TiN deposition.

2. The TiN coating deposition at a constant (optimal) pressure of nitrogen in a range of times t_{TiN} = 60–900 sec.

It should be noted that the TiN coating could be deposited on front and back side of the Ni — 9.5 at. % W substrate. Fig. 1 presents a set of diffraction patterns for the experimental series at different pressures values for the front side of the paramagnetic Ni–W substrate. There two systems of diffraction lines, which belong to the FCC lattice of Ni — 9.5 at. % W alloy and TiN lattice of NaCl-type. As the pressure of nitrogen $p(N_2)$ growths, texture of the substrate exhibits variations: redistribution of intensity in the diffraction lines is observed. With the nitrogen pressure growth there were not found significant changes in the distribution of the TiN intensities. Determination of regularities of the texture formation on the substrate back side requires further study and it is not presented in this paper.

The intensity of the cubic diffraction lines can be considered as a measure of the cubic texture development in the first approximation. In the present study as a "texture parameter" was chosen the ratio of the cubic (200) and diagonal (220) diffraction line intensity of Ni — 9.5 at. % W. Values of the "texture parameter" $I_{200}/I_{220}(p_N)$ based on the XRD data for the substrate front side are plotted in Fig. 2. It is found that the maximum value of the "texture parameter" is observed at $p(N_2)$ = 1.8·10^{-2} Torr for the front side of Ni — 9.5 at. % W substrate, thus it can be argued that this vapor pressure of nitrogen in a vacuum chamber can be considered as the optimal

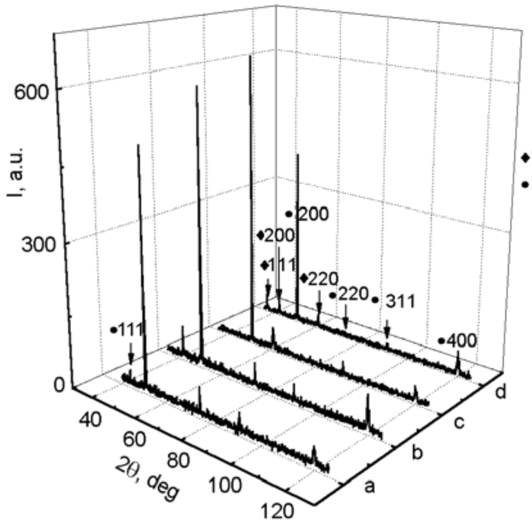

Fig. 3. XRD patterns of Ni — 9.5 at. % W alloy tapes with TiN coating (FRONT side of the substrate) at $p(N_2) = 1.8 \cdot 10^{-2}$ Torr in the range of deposition times: a) t_{TiN} = 60 sec; b) t_{TiN} = 120 sec; c) t_{TiN} = 420 sec; d) t_{TiN} = 900 sec.

for the most suitable development of a the cubic texture in the Ni — 9.5 at. % W tape.

Evolution of diffraction pattern depending on the TiN deposition time at constant pressure of nitrogen is plotted in Fig. 3. As the deposition time t_{TiN} increases, in other words as the TiN h_{TiN} thickness growths, the relative intensity of (h00) type diffraction lines increases, while the intensities of reflections from the diagonal (hh0) and other FCC lattice planes decreases. Such character of the diffraction pattern evolution indicates qualitatively the substantial intensification of the cubic texture degree in the Ni 9.5 % W paramagnetic alloy tape. This tendency continues up to t_{TiN} = 420 sec (h_{TiN} ~ 2 μm). The processes of the cubic texture formation are also observed in the coating layers of TiN. This is proved by the presence of only (200) reflex, which belongs to cubic plane of the TiN lattice. It should be mentioned that other diffraction lines of TiN phase appear on the XRD pattern at the coating thickness $h_{TiN} \geq 2$ μm.

In order to determine fine variations of texture in the "Ni — 9.5 at. % W/TiN" system the algorithm of planar texture characterization was used as a supplementation to the classical ways. This method is based on construction and analysis of the "circular" diagrams of the angular distribution of intensity from the crystallographic planes. The validity of the hypothesis of the perfect cubic texture realization was verified by using the well-known χ^2 method [10]. The χ^2 value was calculated for n = 24 degrees of freedom. The ideal texture should correspond to $\chi^2 = 0$ at a correlation coefficient of $R^2 = 1$. In the present work, it was the most important to investigate the cubic (h00) planes of the metallic Ni–W substrate and TiN coating. Fig. 4 shows a set of the "circular" diagrams corresponding to uncoated Ni — 9.5 at. % W substrate (a), substrate after deposition (b) and TiN layer (c). The diagrams refer to the front side and t_{TiN} = 120 sec (h_{TiN} ~ 1 μm). At this thickness the cube texture of TiN layer with the maximum sharpness (in terms of the mini-

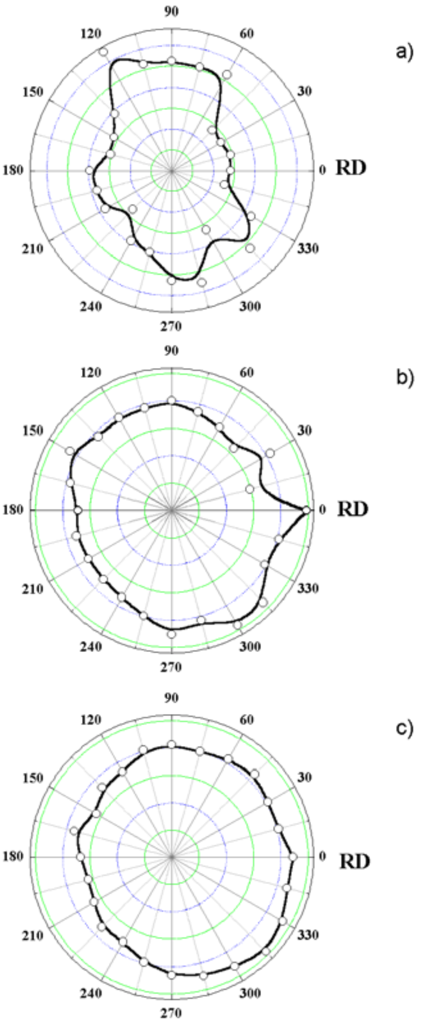

Fig. 4. Circular diagrams of (200) planes related to formation of sharp cubic texture at $p(N_2) = 1.8 \cdot 10^{-2}$ Torr and h_{TiN} ~ 1 μm: a) Ni — 9.5 at. % W without coating ($\chi^2 = 0.082$); b) Ni — 9.5 at. % W after TiN deposition ($\chi^2 = 0.022$); c) TiN layer ($\chi^2 = 0.015$).

Table. Scenario of texture formation in Ni — 9.5 at. % W/TiN system

Deposition time t_{TiN}, sec.	State of structure	
	TiN	Ni — 9.5 at. % W
0		Weak cubic texture
60	Diffraction lines for TiN phase are absent	The beginning of the texture enhancement process
120	Cubic texture is formed	The cube texture continues to evolve
420	Degree of texture is reduced	The maximum degree of cubic texture
900	Degree of texture is reduced	Degree of texture is reduced

mal value of χ^2 parameter) is formed. The cubic texture in Ni — 9.5 at. % W is rather strong, but its maximum is observed at t_{TiN} = 420 sec (h_{TiN} ~ 2 μm). Note that the Ni–W texture achieved at h_{TiN} = 1 μm is more perfect compared to that in original sample before coating.

The subject of discussion in this paper is a set of observed effects related to peculiarities of the cubic texture formation in the thin-film system "Ni — 9.5 at. % W/TiN". Based on the experimental data the texture formation scenario in the system "Ni — 9.5 at. % W/TiN" can be reconstructed (see Table). It can be assumed that this scenario results from the existence of the previously unknown effect "contra epitaxy": the anomalous enhancement of the cubic texture of the metal substrate under the influence of thin coating. The anomalous effect of enhancing the cubic texture of the metal substrate based on Ni — 9.5 at. % W under influence of the TiN coating may be associated with the increase in the stacking fault energy E_{sf} as a result of influence of short-range compressive stresses that arise along the phase boundary "Ni — 9.5 at. % W/TiN".

The formation of quasi single crystalline structure (h00) in the layers of titanium nitride can also be linked to the emergence of the strains in the system "Ni — 9.5 at. % W/TiN". Growth of the mono crystalline structure TiN has a purely epitaxial character.

4. Conclusion

The current work is devoted to solving two major problems: 1) investigation of nature of the processes of texture formation in two component systems, intended for deposition of quasi monocrystalline films of superconductor; 2) development of research-based principles to improve the architecture of 2G HTS with the high current carrying capacity. We believe that the main results of the study are 1) detection of the fundamental effect of "contra epitaxy" — the formation of a the strong cube texture in substrate under the effect of buffer coating; 2) development of the strategy for obtaining the substrates based on paramagnetic alloy tape of Ni — 9.5 at. % W with TiN coating having the cube texture for coated superconductors applications.

References

1. D.Larbalestier, A.Gurevich, D.M.Feldmann, A.Polyanskii, *Nature*, **414**, 368 (2001).
2. M.W.Rupich, X.Li, C.Thieme et al., *Supercond.Sci..Technol.*, **23**, 014015 (2009).
3. A.Goyal, D.F.Lee, F.A.List et al., *Physica C*, **357**, 903 (2001).
4. J.Eickemeyer, R.Huuhne, A.Guuth et al., *Supercond.Sci.Technol.*, **23**, 085012 (2010).
5. F.A.Mohamed, T.G.Langdon, *Acta Metall.*, **22**, 6 (1974).
6. V.A.Finkel, A.M.Bovda, V.V.Derevyanko et al., *Functional Materials*, **19**, 109 (2012).
7. V.A.Finkel, V.V.Derevyanko, M.S.Sungurov et al., *Funct.Mater.*, **20**, 103 (2013).
8. M.S.Sunhurov, V.V.Derevyanko, S.A.Leonov et al., *Techn.Phys.Lett.*, **40**, 797 (2014).
9. I.I.Axenov, V.M.Khoroshikh, N.S.Lomino et al., *IEEE Trans.Plasma Science*, **27**, 1026 (1999).
10. D.Hudson, Statistics. Lectures on Elementary Statistics and Probability, 2nd ed. Mir, Moscow (1970), p. 186 [In Russian].