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Superconductivity and Structure of the Multilayer Cu/Nb/Nb–Ti Nanocomposites

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Abstract—The multilayer composite tapes containing layers of superconducting alloy of niobium with 31 or 50 wt % of Ti separated by niobium layers were investigated. The thickness of a layer is from ~140 to ~10 nm. Composites were produced by three-step rolling of multilayer packages. Effective fixation of the superconducting vortex filaments occurred at the interlayer boundaries of Nb–NbTi, as evidenced by the large anisotropy of the critical current density. The second critical magnetic field was found also dependent on the orientation of the plane of rolling of the composite relative to the external magnetic field, and decreases with decreasing thickness of the layers.

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Mixed state of the second kind superconductors is characterized by the penetration the sample with the magnetic field H in the form of quantum vortex filaments. Each filament has a normal core with the radius of the order of the coherence length ξ, elongated along the H direction, around which the undamped superconducting current flows oriented so that it creates a magnetic field directed along the normal core and coinciding with the direction of the external field. The vortex current covers the area with the radius of the order of the penetration depth of the magnetic field λ, which can significantly exceed ξ.

When electric current is passed through the superconductor the Lorentz force acts on the filaments. In superconductors without structural defects with a very low Lorentz force the vortex filaments would begin to move that leads to heat generation and destruction of superconductivity. In the real situation, the superconductors, like any other metal, contain various structural defects which can fix the filaments. Now for the motion of the vortex filaments a finite current is required, which would create a Lorentz force sufficient for the separation the vortex filaments from the defects. This current is defined as critical current I_c, and its density, as the critical current density j_c. Note that not all defects are able to interact effectively with the vortex filaments. Vacancies, single impurity atoms, and similar "minor" defects are ineffective, as their size typically is much smaller than the characteristic size of the filament, the coherence length ξ. On the contrary, the structural defects with the size of the order ξ and more are very effective as the centers of fixing (pinning) the vortex filaments and they can provide the large critical current density j_c in the superconductor [1]. The defects in the normal state are more effective than the superconducting defects.

In [1] the interaction of vortex filaments with a flat surface of a superconductor of the second kind was theoretically examined, and it was shown that even a defect-free superconducting wafer of the thickness $d >> \lambda$ in the mixed state can bear substantial superconducting current ~10^5 A cm^{-2}. Imagine that a thick solid plate is replaced by a set of thin superconducting plates, artificially separated by a layer of insulator or normal metal. Now the current can flow through each of these plates, and the whole current flowing through the entire cross section of the composite superconductor will be large.

Such a situation was realized in the multilayer Cu/Nb composites where the layers of niobium being a superconductor of the second kind are separated by the layers of copper. It was found that the ratio of j_c values at the magnetic field parallel and perpendicular to the tape plane was equal to 410 in the fields 0.5–0.6 T [2].

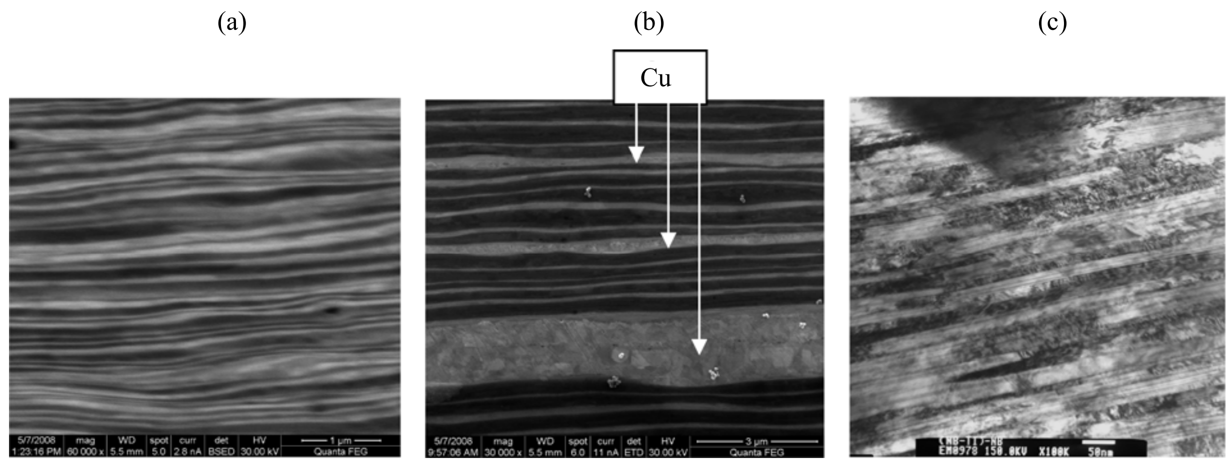

Fig. 1. Microstructure of the cross-section of composite films: (a) Cu/Nb/Nb31Ti; (b) the Cu/Nb/Nb50Ti composite with internal copper layers, and (c) the same after annealing at 360°C for 3 h. All the tapes after 3rd stage of rolling.

Such a large anisotropy of the critical current density indicates the fixation of the vortices on the long copper–niobium planar boundaries.

The aim of this work is to study a multilayer composite tape with the superconducting layers of Nb–(30, 31, and 50 wt %)Ti alloy capable to deformation, and normal niobium layers. Niobium, being a superconductor of the second kind, here plays a role of a normal metal, as the critical current measurements were performed in magnetic fields much higher than its second critical field.

The composites contained from ~1200 to ~29000 layers of niobium–titanium alloy. At the tape thickness 0.3 mm the thickness of the layers varied depending on the number of the layers from ~140 to ~10 nm.

In the Cu/Nb/Nb50Ti composites the groups containing equal number of the alloy and the niobium layers were separated by the layers of copper. Their number, including the two outer layers, was 16. The thickness of the Cu layers was ~9.7 μm. The thickness of the niobium and alloy layers was the same. The Cu/Nb/Nb31Ti composites differed slightly: they had only two outer Cu-layer of ~9 μm thickness and the thickness of each niobium layer was 1.5 times less than the thickness of the alloy. For each alloy five versions of composites was produced differing in the number and thickness of the niobium and alloy layers.

The composite tapes were obtained by rolling in several stages [3]. Each stage included assembling a multilayer package, a hot rolling and a rolling of the package at room temperature. At the first stage a package was assembled from the foils of niobium and the alloy. To obtain a set of composite tapes with different thickness of individual layers, the initial numbers of the foils of niobium and the alloy were taken different. For example, for a Cu/Nb/Nb50Ti composite with a minimum number of layers (675 of niobium and 540 of alloy), and, hence, with their maximum thickness, the number of Nb foils was equal to 5, the number of the alloy foils was 4. For the tapes with a maximum number of layers (7440 of Nb and 6975 of alloy) and minimum thickness, the initial number of Nb foils was 16, the number of the alloy foils was 15. At the second stage were assembled packages of 9 or 31 multilayer foils of thickness 0.3 mm obtained in the first stage, and at the third stage were assembled 15 multilayer foils obtained in the second stage, and 16 Cu foils. Copper was introduced as a stabilizer, by analogy with the traditional multicore superconducting cable. The Cu/Nb/Nb31Ti composite with a maximum number of layers was rolled additionally to a thickness of 0.15 and 0.075 mm in order to bring the thickness of the layers of niobium and alloy to ~5 and ~2.5 nm, respectively.

Figure 1a shows the microstructure of the cross-section of one of the described composites, which contains 2730 of niobium layers and 2340 of the Nb–31%Ti alloy layers. The section is located along the direction of rolling. According to the laws of the scanning electron microscopy, the niobium layers look as bright bands, the alloy layers are dark. Figure 1b shows the microstructure of the cross-section of the composite tape with internal copper layers after the

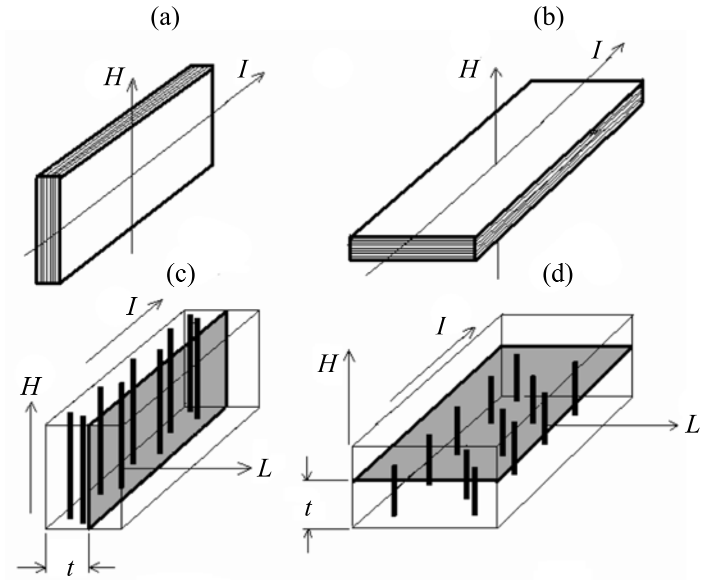

Fig. 2. Schematic representation of the sample orientation relative to the direction of magnetic field *H* in the superconducting solenoid and the location of vortex filaments in the Nb–Ti layer of the thickness *t*: (a, c) $H_{||}$ plane and (b, d) $H\perp$ plane of the composite.

2nd and 3rd stages of rolling. Initially, the package contained 6 foils of niobium and 5 foils of the Nb–50% Ti alloy. It contains 605 of alloy layers, 726 of niobium layers and 121 of copper layers, of which 11 layers are thicker than others.

In the composites with internal Cu layers, the Nb layers in addition to the formation of Nb/Nb–Ti interlayer boundaries served as the diffusion barriers against the interaction of copper with the titanium contained in the alloy.

The above data illustrating the structures were obtained by scanning electron microscopy. Figure 1c shows the microstructure of one of the composites, the image was obtained using transmission electron microscopy. It is evident that the mixing of the layers due to cold deformation and low-temperature annealing does not occur. Estimated thickness of a layer does not exceed 100 nm, and in most cases is 50–60 nm, which is well consistent with the calculated values of the thickness of the layers of niobium and the alloy.

Measurements of the critical current I_c were carried out at a temperature of 4.2 K in external magnetic field *H* up to 7 T induced with a superconducting solenoid. The values of I_c were determined from the Volt-Ampere characteristics. The critical current density j_c was calculated over the whole section of a sample. The plane of rolling the composite tape was oriented either parallel (Fig. 2a) or perpendicular (Fig. 2b) to the direction of *H*, and the flow of the transport current was perpendicular to the *H* in both cases.

As mentioned above, at passing transport current *I* through the conductor, the Lorentz force *L* acts on the vortex filaments. In the case of parallel orientation of the magnetic field and the plane of the composite rolling (Fig. 2c) they can be fixed on the Nb–NbTi borders (in Fig. 2 the vortices are depicted as vertical bars, the Nb–NbTi boundary is gray.) At the perpendicular orientation (Fig. 2d) the filaments cannot be fixed at the interlayer boundaries. But in a real situation the conductor always has other structural defects, which can fix the filaments, and therefore the critical current is not equal to zero in the second case as well. The question is: how efficient is such a fixation?

Figure 3 shows experimental plots of the critical current density j_c vs. the magnetic field *H* for the samples of multilayer Nb/Nb30Ti composite of thickness 0.15 mm, after various thermal-mechanical processings. The composite contained ~15300 Nb layers of thickness ~3.5 nm and ~14400 alloy layers of thickness ~5.5 nm. The upper group of the j_c–(*H*) plots relates to the samples in which the critical current was measured when the plane of the composite and the

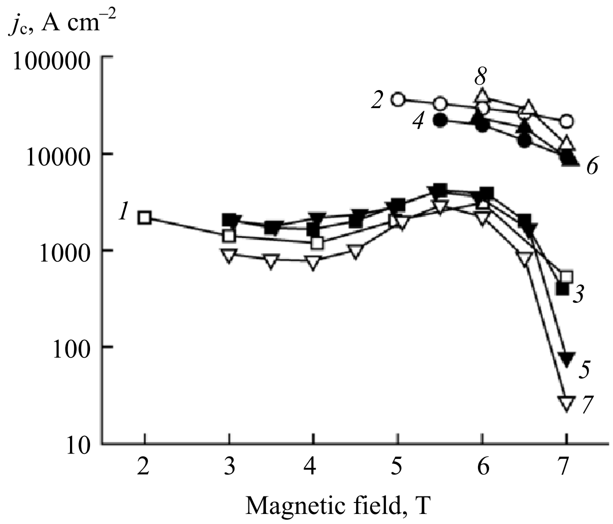

Fig. 3. Plots of j_c vs. H for the Nb/Nb30Ti composite: (*1, 2*) after the cold deformation, (*3, 4*) at the cold deformation + annealing at 600°C, 5 h, (*5, 6*) at the cold deformation + 600°C, 5 h + 250°C, 295 h, (*7, 8*) at the cold deformation + 250°C, 295 h. Upper group of curves is for $H_{\|}$ to the rolling plane, lower group of curves for $H\perp$ to the rolling plane.

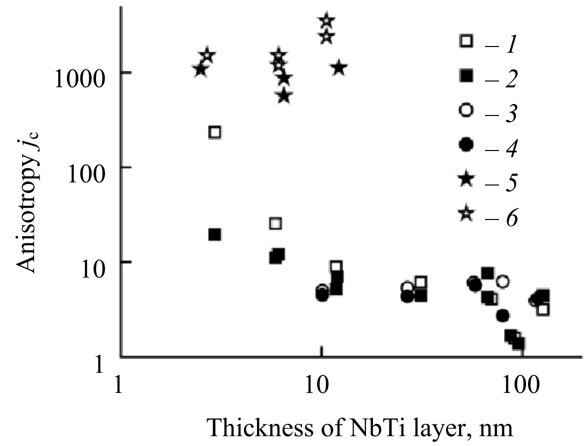

Fig. 4. Dependence of $j_{c\|}/jc\perp$ on the thickness of the layer of niobium alloy containing (*1, 2, 6*) 31 and (*3–5*) 50% of Ti at H = 6 T: (*1, 3*) no annealing, (*2*) annealing at 360°C for 3 h, and (*4*) annealing at 400°C for 3 h.

direction of the magnetic field were parallel, the lower group relates to the perpendicular orientation of the magnetic field and the composite plane. It is seen that the values of j_c corresponding to these groups differ by an order of magnitude in the fields 5–6.5 T, and even more than by an order of magnitude in the field 7 T.

An evidence of the effective fixation of the vortices at the boundaries between layers of niobium and the alloy is the anisotropy of the critical current density j_c, equal to the ratio of the critical current density measured at the parallel and perpendicular orientations of the plane of the composite and the field, $j_{c\|}/j_{c}\perp$. Dependence of the anisotropy on the thickness of (Nb–Ti)-layer t is shown in Fig. 4. At the decrease in t the anisotropy increased from 3.5 for the composites with the layers of 138 nm thickness, up to 11–20 for the composite films with a layer thickness of about 5.5 nm. The largest anisotropy for this family of composites equal to 235 was achieved in the tapes of 0.075 mm thickness, with t ~3 nm. In some experiments this ratio $j_{c\|}/j_{c}\perp$ exceeded 2000 (see Fig. 4, dots *5* and *6*).

It can be assumed that the qualitative change in the fixation of the superconducting vortices at the interlayer boundaries of Nb–NbTi occurs at the approximate thickness of layers 10 nm.

Comparing the dependence of critical current density on magnetic field $j_c(H)$ in the composite films with the layers of various thicknesses one can see that j_c value of the composites with the layers of alloy ≤12 nm thick in the magnetic field above 6 T drops down sharply. In many cases, it became even less than j_c of the composites with the layers of 80–120 nm thickness, despite the fact that in the fields above 6 T the ratio of critical current densities is reverse. Such a course of dependence $j_c(H)$ is typical when the H value is close to the second critical magnetic field of the examined sample. Therefore, experiments were undertaken to measure the H_{c2} of the composites Cu/Nb/Nb30Ti and Cu/Nb/Nb31Ti containing the minimum and maximum number of layers.

The second critical magnetic field H_{c2} was determined from measurements of H_{c2} near the critical temperature [4]. We constructed the plots $H_{c2}(T)$, which near T_c are straight lines, and calculated initially the slope $(-dH_{c2}/dT)_{T=Tc}$, and then $H_{c2}(0\ K) = 0.69\cdot T_c(-dH_{c2}/dT)_{T=Tc}$ and H_{c2}(4.2 K) which was ~95% of the H_{c2}(0 K). By analogy with the measurements of the critical current, the H_{c2} was measured at parallel and perpendicular orientations of the rolling plane of the composite to the direction to the magnetic field.

The final results of measurements of H_{c2} are summarized in the table. For comparison, the table contains also the results of similar measurements of H_{c2} for a common (single layer) tape of the Nb–31%Ti alloy, as

Results of meeasuring H_{c2} in the region near T_c

Run no.	Description of sample	$(-dH_{c2}/dT)_{T=Tc}$, T K^{-1}		H_{c2}(4.2 K), T	
		$H\perp$ rolling plane	$H_{\parallel}$ rolling plane	$H\perp$ rolling plane	$H_{\parallel}$ rolling plane
1	9100/91/138 nm, Cu/Nb/Nb31Ti 360°C, 2 h	2.17	2.21	13.8	14.2
2	9400/8/12 nm, Cu/Nb/Nb31Ti without annealing	1.50	1.82	9.7	11.8
3	The same 360°C, 3 h	1.46	1.78	9.4	11.5
4	Tape of Nb–31%Ti alloy, 0.3 mm thickness without annealing	2.34	2.27	15.2	14.8
5	~27800 layers 9.6 nm, Cu/Nb31Ti/Nb31Ti, 300°C, 2 h annealing	2.39	2.36	15.4	15.3
6	8600/3.6/5.4 nm, Cu/Nb/Nb30Ti without annealing	1.60	1.88	10.0	11.8
7	The same 250°C, 295 h	1.54	1.87	9.6	11.8
8	The same 600°C, 5 h	1.57	1.68	9.9	10.7
9	The same 600°C, 5 h + 250°C, 295 h	1.57	1.71	9.9	10.9
10	The same 350°C, 285 h	1.44	1.75	9.0	11.0
11	8600/7/10.5 nm, Cu/Nb/Nb30Ti 350°C, 285 h annealing	1.51	1.75	9.4	10.7

well as for the Cu/Nb31Ti/Nb31Ti composite, in which all layers are of 9.6 nm thickness and made of the Nb–31%Ti alloy, but the number of layers is 27791, that roughly corresponds to the total number of the layers of niobium and the alloy in the Cu/Nb/NbTi composites.

For the composite tape containing the largest number of layers, at the measurements with the rolling plane parallel to H, the value of H_{c2} is significantly lower (11.8 T, see the table, run no. *2*) than for the monolayer tape of the Nb–31%Ti alloy (14.8 T, run no. *4*). Annealing at 360°C, which resulted in the formation of the α-phase in the layers virtually not affected the H_{c2} (11.5 T, run no. *3*). The value of H_{c2} (4.2 K) of the composite with the layers thickness by an order of magnitude greater, at the same orientation of the rolling plane to the H was 14.2 T, which practically coincides with H_{c2} of common (Nb–Ti)-tape (see the table, run no. *1*). A greater reduction of the H_{c2} was observed in the case of perpendicular orientation of the rolling plane and H: up to 13.8 and 9.7 T for the composites with the smallest and the largest number of layers, respectively.

In the Cu/Nb31Ti/Nb31Ti composite the vortex filaments at the parallel orientation of the rolling plane and H were fixed at the interlayer “superconductor–superconductor” boundaries. This tape had parallel $H_{c2}(T)$ lines at both orientations of the rolling plane and H and, hence, the same slope values $(-dH_{c2}/dT)_{T=Tc}$ (2.36–2.39 T K^{-1}) and H_{c2}(4.2 K) (15.3–5.4 T), close to the values of the same parameters in the single-layer (Nb–Ti)-tape (see the table, run no. *5*).

The behavior of the composites in an external magnetic field differs from that of the bulk superconductors. This can be explained by the effect of proximity according to which the Nb-layers reduce the H_{c2} of Nb–Ti layers by the value of the order of the coherence length $\xi(T) = (\Phi_0/2\pi H_{c2})^{1/2}$ [1], where Φ_0 is a magnetic flux quantum. At H_{c2} = 11 T, $\xi(T)$ ~5 nm. It is just the layer thickness whose further decrease should not lead to the decrease in H_{c2}. Decrease of H_{c2} explains the sharp drop in j_c in the fields over 6.5 T although its normal level is high, and in the field 5 T it equals 10^5 A cm^{-2} at the calucalting j_c over the superconductor section.

Thus, by the method of 3-stage deformation by rolling the prototypes of multilayer composite films were obtained containing alternating layers of nanometer thickness of superconducting niobium–titanium alloy and niobium.

Measurements of the critical current in an external magnetic field with parallel and perpendicular orientation of the tape rolling plane and the magnetic field showed that the effective fixation of superconducting vortex filaments occurs at the Nb/NbTi interlayer boundary. This was evidenced by the high values of the anisotropy of the critical current density in the alloy of thickness 10 nm or less.

The second critical magnetic field of the multilayer composite films is significantly lower than in the usual single-layer tape with the alloy of the same composition as the layers in the composite, and it decreased with increasing number of layers. Furthermore, H_{c2} as well as the critical current density depended on the relative orientation of the plane of rolling of the composite and the direction of the external magnetic field.

REFERENCES

1. Schmidt, V.V., *Vvedenie v fiziku sverkhprovodnikov* (Introduction to the Physics of Superconductors), Moscow: Nauka, 1982, p. 240.
2. Karpov, M.I., Korzhov V.P., Vnukov V.I., Volkov, K.G., and Medved’, N.V., *Materialovedenie*, 2005, no. 1, p. 43.
3. Karpov, M.I., Korzhov, V.P., Vnukov, V.I., Terekhova, I.S., Abrosimova, G.E., and Khodos, I.I., *Deformation i Razrushenie Materialov*, 2008, no. 6, p. 18.
4. Hake, R.R., *Appl. Phys. Lett*., 1967, vol. 10, no. 6, p. 189.