

## INVESTIGATION OF OXYGEN SUBLATTICE IN HIGH- $T_c$ CRYSTALS BY CHANNELING TECHNIQUE

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The features of the oxygen sublattice in a crystal  $\text{YBa}_2\text{Cu}_3\text{O}_7$  are investigated by channeling technique. The angular dependence of resonant nuclear reaction yield of ions  $\text{He}^+$  from oxygen nucleuses at energy of ions 3.055 MeV channeled along  $\langle 001 \rangle$  direction is measured. The best agreement of the calculated angular dependence of the reaction yield with experimental data are achieved at the assumption, that the oxygen sublattice are partly disordered (about 20 % of the oxygen atoms are random situated in the plane (110)) and the atoms of oxygen in oxygen chains are displaced from crystal sites at distance  $\sim 0.3$  C in the plane (110).

### INTRODUCTION

The channeling effect of the charged particles for research of crystals of complex chemical structure allows receiving the unique information on distribution of impurity, radiating defects and displacement of atoms from crystal sites in a crystal lattice [1]. A feature of a channeling technique for multicomponent crystals allows receiving the information on structure of crystals inaccessible by other methods. For example, the research of the crystal  $\text{YBa}_2\text{Cu}_3\text{O}_7$  by the channeling method in [2] has shows the abnormal change of static (and/or dynamic) displacement of atoms from crystal sites near the temperature of transition to the high- $T_c$  state. The shoulder method of the Rutherford backscattering angular dependences has enabled defining the model of structure  $\text{YBa}_2\text{Cu}_3\text{O}_7$ , containing static displacement of oxygen atoms [3].

Developed earlier in [4] the approach to the description the channeling effect of ions in multicomponent crystals allows calculating the basic channeling parameters for the crystal structure under the given experimental conditions. It has allowed by comparison of the calculated and experimental results for various crystallographic directions to receive the detailed information in [5—8] on structural features of a crystal lattice of  $\text{La}_2\text{CuO}_4$  and  $\text{Nd}_2\text{CuO}_4$  in a small depth (up to 0.1 mkm). In

the present work the research of features of channeling effect of  $\text{He}^+$  ions in a crystal  $\text{YBa}_2\text{Cu}_3\text{O}_7$  is carried out. On the basis of the theoretical analysis of experimental data on the angular dependence of resonant elastic scattering yield  $\text{He}^+$  from oxygen nucleuses the new data on structural features oxygen sublattice are obtained.

### THEORETICAL TREATMENT

The ion distribution, flux-peaking and angular yield curves for backscattering and for nuclear reaction have been calculated using the channeling model developed specially for single crystal of compounds in which the channels are formed by several different atomic rows [8]. The model is based on the continuum theory of channeling. The distribution  $g(\mathbf{p}_\perp, z)$  is obtained by solving the diffusion equation:

$$\frac{\partial g(\mathbf{p}_\perp, z)}{\partial z} = \text{div} \left[ D(\mathbf{p}_\perp) \text{grad}_{\mathbf{p}_\perp} g(\mathbf{p}_\perp, z) \right], \quad (1)$$

where  $\mathbf{p}_\perp, z$  are the transverse momentum of channeling ions and crystal depth, respectively,  $D(\mathbf{p}_\perp)$  is the diffusion coefficient due to scattering by electrons and lattice vibrations. The flux distribution in the transverse plane ( $x, y$ ) and crystal depth  $z$  is calculated by the equation:

$$F(x, y, z) = \int_{S_1} \frac{S_0}{S(\mathbf{p}_\perp)} g(\mathbf{p}_\perp, z) d\mathbf{p}_\perp + \\ + 1 - \int_{S_2} g(\mathbf{p}_\perp, z) d\mathbf{p}_\perp, \quad (2)$$

where  $S_1$  and  $S_2$  are the integration regions which can be determined by

$$U(x, y) - U_{\min} \leq \mathbf{p}_{\perp}^2 / 2M_1 \leq \mathbf{p}_c^2 / 2M_1,$$

and

$$\mathbf{p}_{\perp}^2 / 2M_1 \leq \mathbf{p}_c^2 / 2M_1,$$

respectively, where  $U(x, y)$  is the continuum potential,  $U_{\min}$  is the minimum value of the potential,  $M_1$  is the mass of incident ions,  $\mathbf{p}_c$  is the critical momentum,  $S(\mathbf{p}_{\perp})$  is the square of the accessible area in the transverse plane and  $S_0$  is the square of the transverse elementary cell. The resonant elastic scattering yield at the incident angle  $\psi_{in}$  can be expressed as follows:

$$\chi(z, \psi_{in}) = n_r + \sum_i n_i F(x_i, y_i, z), \quad (3)$$

where  $F(x_i, y_i, z)$  is the flux of ions in points  $(x_i, y_i)$  of the transverse plane,  $n_i$  is the relative concentration of oxygen atoms located in the  $i$ -position,  $n_r$  is the relative concentration of the random fraction of oxygen atoms.

### AXIAL CHANNELING OF IONS IN $\text{YBa}_2\text{Cu}_3\text{O}_7$ CRYSTAL

The elementary cell of a crystal  $\text{YBa}_2\text{Cu}_3\text{O}_7$  in the orthorhombic phase is shown in Fig. 1. Parameters of a lattice are:  $a = 3.82\text{C}$ ,  $b = 3.86\text{C}$ ,  $c = 11.67\text{C}$ . The channeling of ions along a direction  $\langle 001 \rangle$  is caused by interaction of

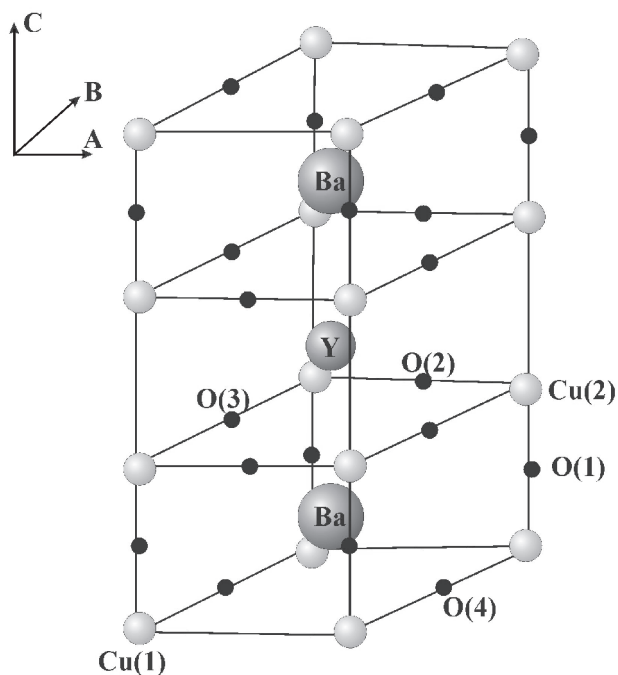


Fig. 1. Structure of the  $\text{YBa}_2\text{Cu}_3\text{O}_7$  crystal

ions with nuclear chains of four kinds: a) by chains consisting of atoms Y and Ba; b) by chains consisting of atoms Cu and O(1); c) by oxygen chains consisting of atoms O(3) and O(4); d) by oxygen chains consisting of atoms O(2).

The calculated diffusion coefficient  $D(\mathbf{p}_{\perp})$  for ions  $\text{He}^+$  with energy 3.055 MeV in channel  $\langle 001 \rangle$  is shown in Fig. 2. At small transverse momentum diffusion is defined by scattering with electrons and diffusion coefficient practically does not depend on a momentum. The increase of the diffusion coefficient at some value of a transverse momentum is corresponding to the contribution of scattering with atoms of different atomic chains. The first increase due to scattering with atoms of oxygen in a «weak» oxygen chain O(2)–O(2). At this momentum ions do not channeling along these oxygen chains and the accessible area cover the part of the channel, in which oxygen chains are located. So, the transverse momentum at the first step on the dependence  $D(\mathbf{p}_{\perp})$  is a critical for a «weak» oxygen chain. Second step corresponds to a critical momentum at which ions no more channeling along «strong» oxygen chains O(3)–O(4). The following features in behaviour of the diffusion coefficient are connected to scattering with mixed Cu–O(1) and Y–Ba atomic chains. At the transverse momentum larger than critical for Y–Ba atomic chains, all particles pass in a chaotic component of a beam. It is possible to say that this momentum is the critical for channeling along this crystallographic direction.

Experimental researches [3, 9] were carried out with the beam of ions  $\text{He}^+$  with energy

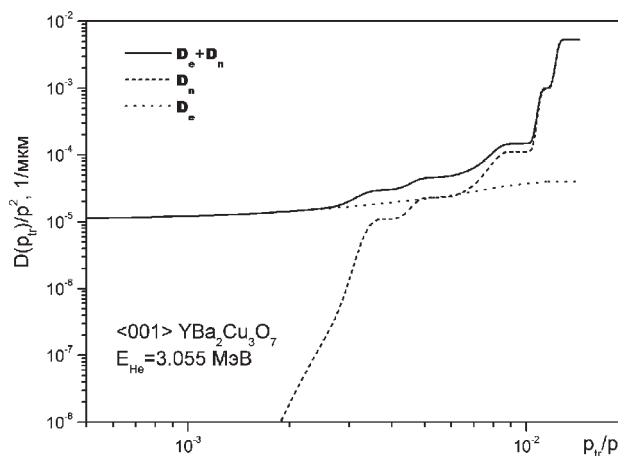


Fig. 2. Diffusion coefficients of 3.055 MeV ions  $\text{He}^+$  in the  $\langle 001 \rangle$  channel of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  crystal.

3.055 MeV chosen so that the elastic resonant scattering of ions with oxygen atoms (at  $E = 3.045$  MeV) were occurred near to a surface of a target. The crystal film  $\text{YBa}_2\text{Cu}_3\text{O}_7$  was put on a monocrystal substrate  $\text{Al}_2\text{O}_3$ . The thickness of a film on the data of the Rutherford backscattering was approximately 0.4 mkm. The value of the minimal yield of the Rutherford backscattering of ions by Ba atoms near to a surface did not exceed 0.06, that testified to sufficient perfection of crystal structure. The ions energy spectra were measured at room temperature at various orientation of a beam in respect to the axis  $\langle 001 \rangle$ . In the spectra the energy range of elastic resonant scattering by oxygen atoms was selected.

The angular dependence of the elastic resonant scattering yield of ions  $\text{He}^+$  from atoms of oxygen in respect to the axial direction  $\langle 001 \rangle$  has «steps» at the incident angle in a crystal  $\psi_{in} \approx \pm 0.4^\circ$  (Fig. 3). The similar form of angular dependence of the nuclear reaction yield of protons from atoms of oxygen in a crystal  $\text{La}_2\text{CuO}_4$  was discussed earlier in [6–8]. There was shown that position and width of the «step» are very sensitive to static or dynamic displacement of oxygen from crystal sites.

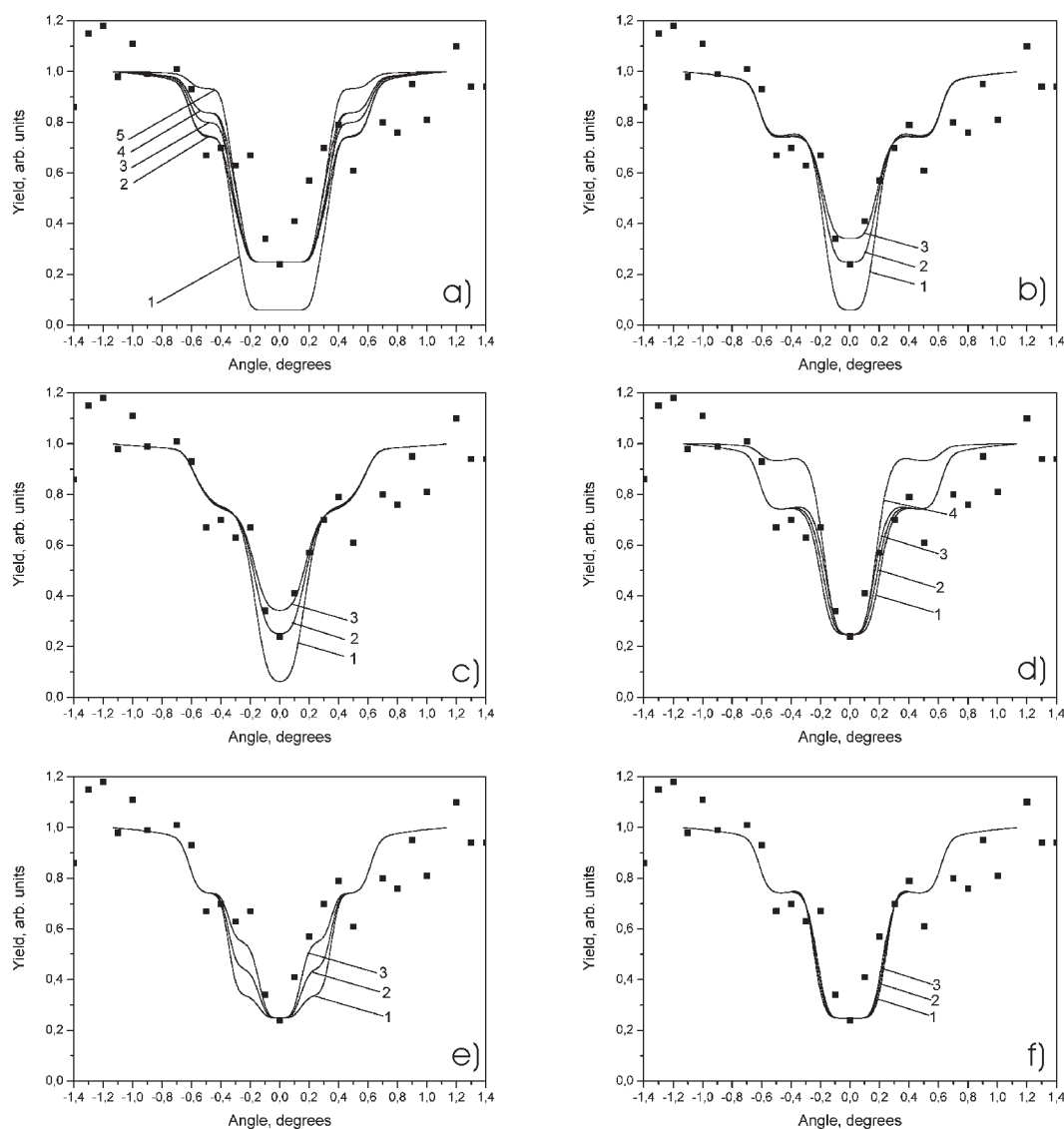
These «steps» in angular dependence of processes of close interactions can be explained as follows. At small incident angle atomic chains of all four types form the flux distributions of channeling ions. At increase of the incident angle the transverse energy becomes more than potential barriers of «weak» O(3)–O(4) and O(2)–O(2) oxygen chains and the flux distribution is formed by more «strong» Y–Ba and Cu–O(1) atomic chains. The channeling ions in respect to the chains of Y–Ba and Cu–O(1) have the greater probability of scattering with atomic chains of O(3)–O(4) and O(2)–O(2). As the atoms of oxygen are located in three various chains, the angular dependence of processes of close interaction with atoms of oxygen is a superposition of dependences caused by interaction of ions with atoms of oxygen in chains of three types: Cu–O(1), O(3)–O(4) and O(2)–O(2). Such superposition with the weight factors of atoms of oxygen in each chain from its total one in a crystal defines the observable form of angular dependence of a yield of processes of close interac-

tions. Width of angular dependence and position of «steps» are defined by average potential of oxygen chains, position of oxygen atoms in a lattice and relative concentration of oxygen in various chains. Comparing experimental angular dependence of the yield of process of close interaction on atoms of oxygen and calculated one for various amplitude thermal vibrations or static displacement from crystal sites it is possible to define their value.

## DISCUSSION

The calculated angular dependences of the yield of the close-impact processes along axial direction  $\langle 001 \rangle$  from the depth 0.01 mkm at the assumption that all atoms of oxygen are situated in the lattice sites are shown in Fig. 3a. The agreement of the calculated and experimental minimal yield is achieved at the assumption that some part (no more than 20 %) of oxygen atoms is situated in random positions in the plane (110). The calculated angular position and width of «steps» do not correspond to the experimental data. The increase of amplitude of thermal vibrations from 0.15C up to the abnormal large 0.40C reduces angular width of the dip, but it is not enough for the satisfactory consent with experimental data. It is reasonably to assume the existence of static displacements of oxygen atoms in the plane (110). The value of displacements strongly influences on the angular width of close-impact processes, and to estimate it need to achieve the best consent of the calculated and experimental data. In Figs. 3b, 3c the results of calculations at the assumption of such static displacement of atoms of oxygen from crystal sites are shown. The satisfactory agreement with the experimental data is achieved at the assumption that the atoms of oxygen from chains O(3)–O(4) and O(2)–O(2) are displaced from crystal sites on distance 0.3C in the (110) plane and the disordered part of oxygen is about 20 %. Thus the atoms of oxygen O(1) in chains Cu–O occupy equilibrium crystal sites.

The «steps» positions in the angular distribution correspond to experimental data (Fig. 3d) at the assumption that the random part of oxygen in a crystal lattice is formed only by atoms from chains O(3)–O(4) and O(2)–O(2). Let's note, that the assumption that displaced



*Fig. 3.* Calculated and experimental (dots) orientation dependence of the yield from oxygen atoms in the  $\langle 001 \rangle$  channel of  $\text{YBa}_2\text{Cu}_3\text{O}_7$ . a) Yield from the depth 0.01 mkm. All atoms of oxygen are in a crystal sites. 1 —  $n_r = 0$ ; 2 —  $n_r = 20\%$  in equal parts from chains O(3,4) and O(2); 3 —  $n_r = 20\%$  in equal parts from chains O(3,4), O(2) and Cu-O; 4 —  $n_r = 20\%$  in equal parts from chains O(3,4) and Cu-O or from chains O(2) and Cu-O; 5 —  $n_r = 20\%$  only from chains Cu-O. b) Yield from the depth 0.01 mkm. The atoms of oxygen from chains O(3,4) and O(2) are displaced from crystal sites in the (110) plane on distance 0.3C. 1 —  $n_r = 0$ ; 2 —  $n_r = 20\%$  in equal parts from chains O(3,4) and O(2); 3 —  $n_r = 30\%$  in equal parts from chains O(3,4) and O(2). c) Yield from the depth 0.05 mkm. The atoms of oxygen from chains O(3,4) and O(2) are displaced from crystal sites in the (110) plane on distance 0.3C. 1 —  $n_r = 0$ ; 2 —  $n_r = 20\%$  in equal parts from chains O(3,4) and O(2); 3 —  $n_r = 30\%$  in equal parts from chains O(3,4) and O(2). d) Yield from the depth 0.01 mkm. The atoms of oxygen from chains O(3,4) and O(2) are displaced from crystal sites in the (110) plane on distance 0.3C. 1 —  $n_r = 20\%$  only from chains O(3,4); 2 —  $n_r = 20\%$  in equal parts from chains O(3,4) and O(2); 3 —  $n_r = 20\%$  only from chains O(2); 4 —  $n_r = 20\%$  only from chains Cu-O. e) Yield from the depth 0.01 mkm. The atoms of oxygen only from chains O(2) are displaced from crystal sites in the (110) plane on distance 0.3C. 1 —  $n_r = 20\%$  only from chains O(2); 2 —  $n_r = 20\%$  in equal parts from chains O(3,4) and O(2); 3 —  $n_r = 20\%$  only from chains O(3,4). f) Yield from the depth 0.01 mkm. The atoms of oxygen only from chains O(3,4) are displaced from crystal sites in the (110) plane on distance 0.3C. 1 —  $n_r = 20\%$  only from chains O(2); 2 —  $n_r = 20\%$  in equal parts from chains O(3,4) and O(2); 3 —  $n_r = 20\%$  only from chains O(3,4)

atoms are only from chains O(3)–O(4) or only from chains O(2)–O(2) does not give such agreement with experimental results as the assumption of displacement of atoms from both chains (see Figs. 3e, 3f). The reduction of the value of displacement from 0.3C leads to increase of the width of the angular distribution, and increase leads to reduction one accordingly.

### CONCLUSION

From comparison of the measured and calculated yield of elastic resonant scattering of ions  $\text{He}^+$  with energy 3.055 MeV from atoms of oxygen in a crystal  $\text{YBa}_2\text{Cu}_3\text{O}_7$  along the axial direction  $\langle 001 \rangle$  it is appear that atoms of oxygen in chains O(3)–O(4) and O(2)–O(2) are displaced in the plane (110). The analysis of a feature of the angular distribution of the resonant elastic scattering yield allows to make a conclusion about presence of a random part of atoms of oxygen (i.e. occupying a chaotic sites in the plane (110)), which is formed of chains O(3)–O(4) and O(2)–O(2). The value of the disordered oxygen is no more than 20 %. Let's notice, that the similar estimation for the disordered oxygen subsystem was discussed for the crystal  $\text{La}_2\text{CuO}_4$ , which also was investigated by a channeling technique [5].

Earlier in [3, 9] by the analysis shoulder areas in angular dependence of the elastic resonant scattering yield of ions  $\text{He}^+$  from atoms of oxygen the assumption about displacement of oxygen atoms was made. Our results confirm and specify this conclusion. The maximal displacement is equal to 0.3C, and the vacancies arise in chains O(3)–O(4) and O(2)–O(2). The revealed properties of an oxygen subsystem of a crystal  $\text{YBa}_2\text{Cu}_3\text{O}_7$  are not a consequence of the chosen channeling model and are connected with the specificity of a crystal field of a lattice. It is possible, that the random located oxygen forms original structure of

carriers of a charge responsible for high- $T_c$  property of a crystal.

Use for measurements of one axial direction does not enable to determine a direction of displacement of atoms of oxygen in a projection to a plane (110). It is possible to take the assumption about "two by two" displacements in one direction, and the directions of displacement change on opposite for the following pairs along chains  $\langle 001 \rangle$ . The similar model of displacement of "goffered" type was discussed also in [3]. Such model of displacement of atoms of oxygen and value of displacement does not contradict to results of researches of structure  $\text{YBa}_2\text{Cu}_3\text{O}_7$  with the help of methods x-ray and neutron diffraction. These methods are rather tolerant to local distortions of a structure oxygen sublattice of a crystal and even the rather large static or dynamic displacement of atoms of oxygen from crystal sites difficultly to determine, if these displacement do not break symmetry of a lattice.

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