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Photo-induced "high-temperature" superconductivity of multicomponent metal-oxide compounds

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The work is devoted to the problem of searching for substances with superconductivity at room temperature and atmospheric pressure. It develops ideas in the direction of studying the properties of multicomponent metal-oxide compounds. The main attention is paid to studies devoted to explanation of the role of various mechanisms of the formation of paired electrons in these compounds. Naturally, most of this kind of research is devoted to the study of the chemical structure and crystallographic structure of metal oxides, since they determine the properties of the electronic subsystem of these substances, which cause their transition to the superconducting state.

Analysis of the crystallographic structure of metal oxide $YBa_2Cu_3O_{7-x}$ leads to the conclusion that the main role in the formation of its superconducting state is played by the presence of planes CuO_2 , separated by solitary planes composed of chains CuO . The presence of chemically different states of like-named pairs of ions, characterized by different types of bonds (ionic and covalent), as well as the specific layered crystallographic structure of the metal-oxide give rise to the existence of clusters of negative U-centers capable of generating paired electrons realizing the superconductivity of metal-oxide substances.

This paper discusses the possibility of intensifying the transition of metal-oxide compounds to the superconducting state under conditions of irradiation with a photon flux. In this case, the formation of an energy spectrum that allows local pair transitions of electrons can be activated (the effect of internal photoionization) and, thus, the transition of metal oxides to the superconducting state can occur at higher temperatures than is observed under normal conditions.

Keywords: metal-oxide compounds, electron-phonon interaction, high-temperature superconductivity.

Фотоіндукована «високотемпературна» надпровідність полікомпонентних метал-оксидних з'єднань

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Робота присвячена проблемі пошуку речовин, що мають надпровідність при кімнатній температурі і атмосферному тиску. У ній розвиваються уявлення в напрямку вивчення властивостей полікомпонентних метал-оксидних сполук. Основна увага приділяється дослідженням, присвяченим з'ясуванню ролі різних механізмів утворення спарених електронів в цих сполуках. Природно, що велика частина такого роду досліджень присвячена вивченню хімічної будови і кристалографічної структури метал-оксидів, оскільки саме вони визначають властивості електронної підсистеми цих речовин, що зумовлюють їхній перехід до надпровідного стану.

Аналіз кристалографічної структури метал-оксиду $YBa_2Cu_3O_{7-x}$ призводить до висновку про те, що основну роль в процесі формування його надпровідного стану грає наявність площин CuO_2 , розділених відокремленими площинами, що складаються з ланцюжків CuO . Наявність хімічно різних станів однойменних пар іонів, що характеризуються різним типом зв'язку (іонним і ковалентним), а також специфічна шарувата кристалографічна структура метал-оксиду обумовлюють існування скупчень негативних U-центрів, здатних генерувати спарені електрони, що реалізують надпровідність метал-оксидних речовин.

У даній роботі обговорюється можливість інтенсифікації переходу метал-оксидних сполук до надпровідного стану в умовах опромінювання їх потоком фотонів. В цьому випадку процес формування енергетичного спектра, що допускає локальні парні переходи електронів, може бути активований (ефект внутрішньої фотоіонізації) і, таким чином, перехід метал-оксидів до надпровідного стану може здійснюватися при більш високих температурах, ніж це спостерігається за звичайних умов.

Ключові слова: метал-оксидні сполуки, фононна взаємодія, високотемпературна надпровідність.

Фотоиндуцированная «высокотемпературная» сверхпроводимость поликомпонентных металл-оксидных соединений

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Работа посвящена проблеме поиска веществ, обладающих сверхпроводимостью при комнатной температуре и атмосферном давлении. В ней развиваются представления в направлении изучения свойств поликомпонентных металл-оксидных соединений. Основное внимание уделяется исследованиям, посвященным выяснению роли различных механизмов образования спаренных электронов в этих соединениях. Естественно, что большая часть такого рода исследований посвящена изучению химического строения и кристаллографической структуры металл-оксидов, поскольку именно они определяют свойства электронной подсистемы этих веществ, обуславливающие переход их в сверхпроводящее состояние.

Анализ кристаллографической структуры металл-оксида $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ приводит к выводу о том, что основную роль в процессе формирования его сверхпроводящего состояния играет наличие плоскостей CuO_2 , разделенных уединенными плоскостями, состоящими из цепочек CuO . Наличие химически различных состояний одноименных пар ионов, характеризующихся разным типом связи (ионным и ковалентным), а также специфическая слоистая кристаллографическая структура металл-оксида обуславливают существование скоплений отрицательных U -центров, способных генерировать спаренные электроны, реализующие сверхпроводимость металл-оксидных веществ.

В данной работе обсуждается возможность интенсификации перехода металл-оксидных соединений в сверхпроводящее состояние в условиях облучения их потоком фотонов. В этом случае процесс формирования энергетического спектра, допускающего локальные парные переходы электронов, может быть активирован (эффект внутренней фотоионизации) и, таким образом, переход металл-оксидов в сверхпроводящее состояние может осуществляться при более высоких температурах, чем это наблюдается в обычных условиях.

Ключевые слова: металл-оксидные соединения, электрон-фононное взаимодействие, высокотемпературная сверхпроводимость.

1. Introduction

The discovery in 1986–1987 of "high-temperature" superconductivity (zero electrical resistance) of multicomponent metal-oxide compounds, aroused great interest in the study of the properties of this class of substances [1, 2]. These include a number of compounds described by the general chemical formula $\text{RBa}_2\text{Cu}_3\text{O}_{7-x}$, where $\text{R} = \text{Y}, \text{Nd}, \text{Sm}, \text{Eu}, \text{Ho}, \text{La}, \text{Pr}$ and etc. Of the indicated series of metal oxides, the compound $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ is most deeply and comprehensively studied. It is characterized by the transition temperature to the superconducting state $T_c \approx 90\text{K}$, which is an order of magnitude higher than the average transition temperature observed in the classical metal superconductors. It is this circumstance that was the reason that this group of substances was called "high-temperature" superconductors (HTSC). In addition to the above compounds, metal-oxide superconductors also include more complex compounds $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10-x}$, $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10-x}$ and $\text{Hg}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10-x}$, which have the highest transition temperature values among this class of substances: $T_c = 110, 125$ and 164K respectively. In further research, undertaken after the discovery of "high-temperature" superconductivity, the main efforts were directed to the discovery of new superconducting substances. At the same time, the strategic goal of all research was the discovery of substances with superconductivity at room temperature ($\approx 300\text{K}$) and atmospheric pressure. The requirement that this condition be satisfied is extremely important for the implementation of a wide and, most importantly,

economically profitable use of superconductors in practice. However, to date, the specified strategic goal of numerous studies on superconductivity remains unattained. One of the main reasons for this is the lack of clarity and unambiguity in understanding the microscopic mechanism of superconductivity of metal-oxide compounds. An attempt to explain the superconductivity of these substances using the BCS theory [3] (phonon pairing of electrons), which describes the "low-temperature" superconductivity of metals and metal alloys, turned out to be unacceptable [4]. In this case, however, a detailed study of the nature of elementary charge carriers in metal oxides showed that the electrical conductivity in them at temperatures $T \leq T_c$ is realized due to the transport of paired electrons (bosons), as in the case of "low-temperature" metal superconductors.

Currently, studies of the effect of superconductivity are carried out in two main directions. The first of them is the study of superconductivity of metal compounds with hydrogen (hydrides). These compounds, due to the low mass of hydrogen atoms, have an increased fraction of high-frequency phonons (optical mode). In accordance with the BCS theory, it is precisely this kind of compounds that should have the maximum electron - phonon interaction and, accordingly, be characterized by high temperatures T_c [5-7]. For example, it was found that the compound LaH_{10} turns into a superconductor at a temperature $\approx 250\text{K}$, which is very close to the desired temperature $\approx 300\text{K}$. However, this result did not fully solve the main problem formulated above, since

superconductivity in hydrides and their stable phase state are observed only under very high pressure $\approx (150 \div 200$ GPa). The creation of such a pressure level is a laborious and economically expensive technological task, which makes it practically impossible to use superconducting hydrides in practice.

Another modern direction of research on superconductivity is the continuation of study of the of multicomponent metal-oxide compounds properties. First of all, these are studies devoted to ascertain the role of various mechanisms of the paired electrons formation in these compounds. Naturally, most of this kind of research is devoted to the chemical and crystallographic structure study of metal oxides, since they determine the properties of the electronic subsystem of these substances.

This paper discusses the possibility of intensifying the transition of metal-oxide compounds to the superconducting state under conditions of irradiation with a photon flux. In this case, the formation of an energy spectrum that allows local pair transitions of electrons can be activated (the effect of internal photoionization) and, thus, the transition of metal oxides to the superconducting state can occur at higher temperatures than is observed under normal conditions.

2. Typical general properties of metal oxides and specific features of the electronic and phonon spectrum in this class of substances

1) First of all, it should be noted that all of the above-listed superconducting metal-oxide compounds contain a copper ion, as well as ions of some other metals, which have the ability to change their valence: (Cu^{1+} , Cu^{2+} , Cu^{3+}), (Bi^{3+} , Bi^{5+}). Accordingly, the presence of these ions makes it easy to change the oxidation state of this kind of oxides [8]. At the same time, there is a clear correlation between the value of the critical temperature T_c of these substances transition to the superconducting state and the concentration of oxygen ions [9 – 11].

2) The transition to the superconducting state of metal-oxide compounds occurs at relatively low values of the energy states of electrons density at the Fermi level: $N(E_F) \approx (10^{26} \div 10^{27})$ $1/\text{m}^3$. At the same time, the conduction bands of these substances are quite wide and are characterized by an interval of values $\approx (3 \div 10)$ eV.

3) All metal oxides have a specific phonon spectrum, characterized by the presence of a certain fraction of high-frequency phonons (optical mode) [12, 13].

4) All superconducting metal-oxide compounds are characterized by a temperature $T^* \geq T_c$, upon reaching which a deviation from the "normal" conductivity begins (the opening temperature of the so-called "pseudo-gap" in the energy spectrum of electrons) [14, 15]. In this case, the value of the temperature T^* depends on the value of the

parameter x (stoichiometry parameter). So, for example, in the connection $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ at a value of $x \approx 0.4$, the temperature T^* reaches the value ≈ 450 K, then, with decreasing x , it gradually decreases and at $x=0$ coincides with the critical temperature $T^* \approx T_c \approx 90$ K [15].

5) In the transition to the superconducting state at $T \leq T_c$ all metal oxides are characterized by the appearance in the energy spectrum of the electronic subsystem of a relatively narrow ($\approx 5 \cdot 10^{-3} \div 10^{-2}$ eV) energy gap near the Fermi level.

6) From the chemistry point of view, metal oxides are complex isodesmic substances with an ionic-covalent type of bond [16]. Specifically, in these compounds, the oxygen ion O (anion) forms chemical bonds with several different metal ions (cations). In this case, for the formation of a stable (thermodynamically equilibrium) structure of these compounds, it is necessary to fulfill the condition of electrical neutrality of the unit crystal cell. That is why some of the ions change their valence, and some ions may even be partially absent (for example, oxygen), violating the stoichiometry of the substance, while maintaining, however, the electroneutrality of the substance as a whole.

7) The features of the chemical composition of metal-oxide compounds indicated in items 1 and 6 lead to the fact that at the value of the parameter $x \approx 0$ in these substances a specific crystallographic structure of atoms is formed, described by an orthorhombic unit cell. A distinctive feature of this structure is that it is layered and contains two configurations of the oxygen environment of copper ions: a tetrahedral pyramid in the CuO_2 planes and a rhombus consisting of CuO chains in the basal plane (ab). Thus, a structure is formed, which consists of so-called "perovskite-like" blocks containing CuO_2 planes separated by solitary planes from CuO chains. It is this structure that is characteristic of superconducting metal oxides. With a large deficit of oxygen ions ($x > 0.5$), a different (tetragonal) crystal structure is realized, and superconductivity in this case disappears [17].

Thus, based on the above information on the general characteristic properties of metal-oxide superconducting compounds, we can draw the following conclusion. The main factors determining the formation of specific phonon and electronic spectra of metal-oxide compounds and, accordingly, causing their transition to the superconducting state, are the stoichiometric composition, as well as the crystallographic structure of these substances.

3. Possible mechanisms for the formation of localized electron pairs, the movement of which realizes the superconductivity of metal-oxide compounds

As mentioned above, in metal oxides, in addition to the electron - phonon mechanism of superconductivity, associated with the mutual attraction of electrons due to the

exchange of phonons, other mechanisms of the formation of electron pairs can also be realized. First of all, these can be mechanisms associated with the so-called exchange-correlation interaction of electrons. In this regard, many models have been discussed in attempts to explain the possible participation of various electronic excitations in the formation of paired charge carriers in HTSC: plasmons, excitons, spin fluctuations, etc [18, 19]. With all this, the contribution of the electron - phonon interaction during the formation of a superconducting state in these compounds is also not denied by anyone. In this regard, it is of interest to estimate the electron-phonon pairing constant λ , which appears in the standard formula of the BCS theory for T_c

$$T_c \approx \theta \cdot \exp [- (1+\lambda) / (\lambda - \mu)]. \quad (1)$$

Here θ is the Debye temperature, μ is the Coulomb pseudo-potential characterizing the repulsion of electrons (usually $\mu \approx 0,1 \div 0,15$). According to calculations performed in [20], in the phonon spectrum of metal oxides, there is a high-frequency mode associated with the possibility of displacement of oxygen ions located in the planes formed by CuO molecules. In this case, two oxygen ions located on opposite sides of the copper ion can move upward from the plane, and the other two can move downward. Calculation of the pairing constant value taking into account the role of the oxygen ions displacements, as well as the characteristic value $\theta \approx 300\text{K}$, showed that $\lambda \approx 0,2$. In accordance with relation (1), such a value of the pairing constant should determine the value $T_c \approx 10\text{K}$. This estimate indicates that the electron - phonon mechanism of electron pairing makes an appreciable, but not determinant contribution to the formation of the superconducting state in metal oxides. In this regard, the following conclusion can be drawn: until now, researchers have succeeded in quite deep progress in studying the properties of metal-oxide HTSCs. However, there are still many questions related to the explanation of their behavior, both in the "normal" and in the superconducting state. Until now, there is no unambiguous understanding of the nature and role of another, different from the electron-phonon, mechanism causing the appearance of electron pairs. Among the unresolved issues, it is also necessary to note the lack of understanding of the reasons leading to the appearance of a "pseudo-gap" in metal-oxide superconductors at a temperature T^* significantly exceeding the temperature of the superconducting transition T_c . The dependence of the T^* value on the doping degree of metal oxides with oxygen ions remains unexplained. The most acceptable is the physical concept, which explains almost all the features of the properties of metal-oxide HTSC compounds, proposed in the works [14, 21]. It is based on the idea of the existence in metal oxides

of special structural clusters of atoms, consisting of the so-called negative U-centers. In accordance with the chemical composition and crystallographic structure of metal oxides, the authors of the above works drew attention to the fact of the formation in these compounds of a special group of ions, consisting of two positive copper ions (cations) embedded in the CuO₂ plane and the surrounding negative oxygen ions (anions) (see Fig. 1).

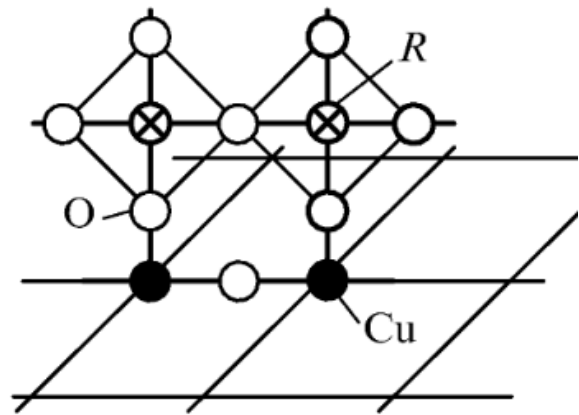


Fig. 1. Fragment of YBa₂Cu₃O_{7-x} crystal structure containing a Cu₂R₂O_n cluster, where Cu ions are "embedded" into the CuO₂ plane, and the R = Cu ions in the CuO₃ chains.

Such a structural element acquired the name "negative U-center", and its presence in metal-oxide compounds was confirmed experimentally. A detailed analysis of the forming energy spectrum of the electronic subsystem showed that the combination of a certain number of U centers (cluster formation) causes the appearance in the energy spectrum of levels that allow a local pair transition of electrons from an oxygen ion to two neighboring copper ions (Fig. 2).

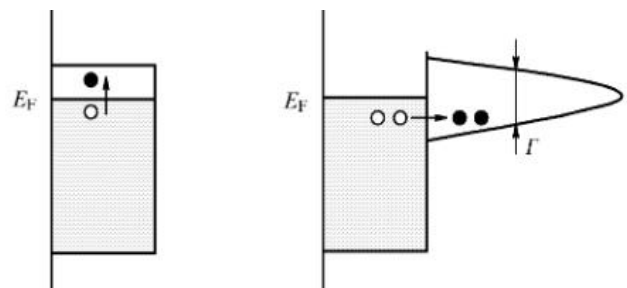


Fig. 2. Diagram of the band structure of the electronic energy spectrum: a) ordinary metal; b) metal-oxide compound [15]. Holes appear in the filled zone due to the transition of electron pairs to a pair level, the width of which $\Gamma \sim T$.

Wherein at the initial stage of the energy spectrum formation, the appearance of a small number of U-centers causes a slight "excess" electrical conductivity at the

temperature T^* (the opening temperature of the so-called pseudo-gap). The minimum cluster size, which makes the pair transition of electrons possible, is determined by the coherence length in the CuO_2 plane and is $\approx 10^{-9}$ m. As the concentration of oxygen ions in the metal oxide increases, ie, at $x \rightarrow 0$, the total length of the clusters containing U-centers increases, and the temperature T^* gradually decreases, approaching the temperature T_c . Thus, the critical temperature of the transition to the superconducting state is reached when the anionic sublattice of the metal-oxide compound is completely filled with oxygen ions and, accordingly, when the maximum concentration of negative U-centers is reached. It is important to pay attention to the following fact. As already mentioned, the compounds $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10-x}$, $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10-x}$ and $\text{Hg}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10-x}$ also belong to metal oxide superconductors. Moreover, they have the highest values of the critical temperature T_c . Following the above outlined logic, this can be explained by the fact that the structure of these compounds contains an additional number of CuO_2 planes, the presence of which leads to the formation of an increased number of U-centers in comparison with other metal-oxides. Accordingly, these compounds are characterized by higher values T_c . An interesting fact that evidence of reality of the above scheme is the result of the work [22], in which the process of increasing the total length of clusters from U-centers and, accordingly, the transition of the metal oxide to the superconducting state, was significantly activated by the action of external pressure.

Thus, we can conclude that the main role in the formation of the superconducting state in metal-oxide compounds is played by the presence of CuO_2 planes separated by solitary planes consisting of CuO chains. The presence of chemically different states of like-named pairs of ions, characterized by different types of bonds (ionic and covalent), as well as the specific layered crystallographic structure of metal-oxides determine the existence of negative U-centers clusters, capable of generating paired electrons, realizing the superconductivity of metal-oxide substances.

4. Possible increase in the transition temperature of metal oxides from the "normal" to the superconducting state under conditions of irradiation with photons

In accordance with the conclusion made in the previous paragraph concerning the nature of the mechanism of metal-oxide compounds transition to the superconducting state, it can be assumed that this transformation can occur at higher temperatures if these substances are irradiated with a flux of photons. Indeed, formation of the energy spectrum of negative U-centers clusters is associated with

the formation of ionic-covalent bonds between planes consisting of CuO_2 molecules and planes formed from CuO chains. This process can be intensified without disturbing the crystallographic structure of the compound under conditions of irradiation of metal oxides with a photon flux [23]. As already indicated, under normal conditions (without irradiation), the first signs of U-centers formation appear at the temperature T^* , which significantly exceeds the critical temperature T_c . In this regard, it is natural to assume that under conditions of photon irradiation, the formation of the maximum possible number of U-centers can be significantly accelerated. In this case, the thermal destruction of electron pairs will be compensated by the process of the creation of new pairs. Accordingly, the temperature T_c should approach the temperature T^* . Let us make quantitative estimates of some parameters of the photon flux, under the influence of which the described effect can be realized. First of all, it is obvious that the photon energy must satisfy the relation:

$$h \cdot \nu \approx W. \quad (2)$$

Here h is Planck's constant, W is the energy of the photoinduced chemical reaction causing the formation of a negative U-center, ν is the frequency ($\nu = c/L$, where c – the light speed, L – radiation wavelength). In the particular case under consideration, W is the energy required for the transfer of electrons from copper ions to oxygen ions. Obviously, quantitatively, the minimum value of W is equal to the thermal energy sufficient for the destruction of electron pairs in the discussed compounds: $W \approx kT_c$ (k – Boltzmann constant). Since for metal oxides $T_c \approx (10 \div 100)\text{K}$, then $W \approx (10^{-3} \div 10^{-2})\text{eV}$. Thus, it follows from relation (2) that $L \approx (10^{-5} \div 10^{-4})\text{m}$. Such a wavelength interval corresponds to infrared light radiation, sufficiently powerful fluxes of which can be easily achieved using laser technology [24]. To implement the described effect, the required photon flux density should approximately correspond to the atomic density of the irradiated substance: $n_{\text{phot}} \approx n_{\text{at}} \approx 10^{29} 1/\text{m}^3$. Such a photon flux density can be achieved using a laser with a power $\approx 10^{11} \text{W}/\text{m}^2$ and pulse duration $\approx 10^{-8} \text{s}$ [24]. It is important to take into account that infrared radiation penetrates into the volume of metal-oxide compounds to a depth $\approx 10^{-6} \text{m}$ [25]. Consequently, the described effect can be realized either in the near-surface layer of the irradiated sample, or in the volume of a film of the specified thickness. In addition, it is obvious that the described effect should show itself in full when using single-crystal samples, since grain boundaries in polycrystals, usually characterized by the size $\approx (10^{-5} \div 10^{-4})\text{m}$, are effective infrared scattering centers.

5. Conclusions

1) Under conditions of laser irradiation with a wavelength $L \leq 10^{-4}\text{m}$ and pulse duration $\approx 10^{-8}\text{s}$ when the power density is reached $\approx 10^{11}\text{W/m}^2$ In the energy spectrum of the electronic subsystem of metal-oxide HTSC compounds, a photoinduced chemical reaction can occur, accompanied by the transfer of electrons from copper atoms Cu to oxygen atoms O.

2) As a result of a photoinduced chemical reaction in metal oxides, special structural elements are formed – negative U-centers, the clusters of which are characterized by a specific energy spectrum that allows local pair transitions of electrons.

3) The appearance of pairs of electrons (bosons) in the discussed metal-oxide compounds can cause their electrical superconductivity. In this case, the critical temperature of these compounds transition to the superconducting state T_c under irradiation with a photons flux can increase and reach the value of the "pseudo-gap" opening temperature T^* (Fig. 3).

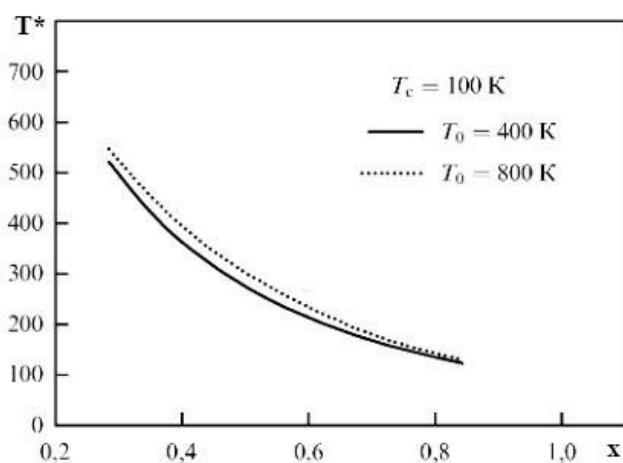


Fig. 3. Dependence of the "pseudo-gap" opening temperature T^* in the compound $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ on parameter x [15].

4) The described effect can be realized in near-surface layers with a thickness $\approx 10^{-6}\text{m}$ in bulk single-crystal samples or in thin films of the same thickness.

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