

La_{2-x}Sr_xCuO₄ yupqa pardalarida T_c ning legirlash va parda qalinligiga bog'liqligi haqida

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Annotatsiya. Kuprat La_{2-x}Sr_xCuO₄ yupqa pardalar kengaytirilgan Holstein-Hubbard modeli va o'ta o'tkazuvchanlikning bipolaron nazariyasi doirasida ko'rib chiqiladi. Qatlamlararo bipolaronlar ideal gazining Bose-Eynshteyn kondensatsiyasi (T_{BEC}) harorati va yupqa pardalaridagi tarangliklar o'rtasidagi bog'liqlik o'rnatiladi. LaSrAlO₄ va SrTiO₃ substratlarida o'stirilgan La_{2-x}Sr_xCuO₄ yupqa pardalar kupratidagi bipolaronlar T_{BEC} ning hisoblangan qiymatlari yuqori tizimlardagi o'ta o'tkazuvchanlikning (T_c) kritik haroratining eksperimental qiymatlari bilan mos kelishi ko'rsatilgan. ($T_{BEC,x}$) tekislikdagi La_{2-x}Sr_xCuO₄ yupqa pardalarning gumbaz shaklidagi fazali diagrammasi olinadi, bu La_{2-x}Sr_xCuO₄ yupqa pardalarning x fazali diagrammasiga nisbatan tanish gumbaz shaklidagi T_c turiga o'xshaydi. T_c ning yupqa parda qalinligiga bog'liqligini o'rganish uchun muqobil (bipolaronik) yondashuv taklif etiladi. Ushbu yangi yondashuvga asoslangan dastlabki natijalar La_{2-x}Sr_xCuO₄ yupqa pardalarning yuqori T_c o'ta o'tkazuvchanligi va ko'rib chiqilayotgan tizimdagi qatlamlararo bipolaronlar Bose-Eynshteyn kondensatsiyasi o'rtasida chuqur bog'liqlik mavjudligini aniq ko'rsatmoqda.

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Kalit so'zlar: kupratlar, La_{2-x}Sr_xCuO₄, o'ta o'tkazuvchanlik, Bose-Eynshteyn kondensatsiyasi, bipolaron, bipolaron, parda qalinligi.

О легировании и зависимости T_c тонких пленок La от толщины.

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Аннотация. Тонкие пленки купрата La_{2-x}Sr_xCuO₄ рассмотрены в рамках расширенной модели Гольштейна-Хаббарда и биполярной теории сверхпроводимости. Установлена связь между температурой бозе-эйнштейновской конденсации (T_{BEC}) идеального газа межузельных биполяронов и деформациями в тонких пленках. Показано, что расчетные значения T_{BEC} биполяронов в тонких пленках купрата La_{2-x}Sr_xCuO₄, выращенных на подложках LaSrAlO₄ и SrTiO₃, коррелируют с экспериментальными значениями критической температуры сверхпроводимости (T_c) указанных систем. Получена фазовая диаграмма куполообразного типа тонких пленок La_{2-x}Sr_xCuO₄ в плоскости (T_{BEC}, X), которая аналогична известной фазовой диаграмме куполообразного типа T_c в зависимости от x тонких пленок La_{2-x}Sr_xCuO₄. Предложен альтернативный (биполярный) подход к изучению зависимости T_c от толщины пленки. Предварительные результаты, основанные на этом новом подходе, ясно указывают на существование глубокой взаимосвязи между высокотемпературной сверхпроводимостью тонких пленок La_{2-x}Sr_xCuO₄ и Бозе-Эйнштейновской конденсацией межузельных биполяронов в рассматриваемой системе.

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Ключевые слова: купраты, La_{2-x}Sr_xCuO₄, сверхпроводимость, Бозе-Эйнштейновская конденсация, полярон, биполярон, тонких пленок.

On Doping and Thickness Dependence of T_c of La_{2-x}Sr_xCuO₄ Thin Films

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Abstract. The cuprate $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ thin films are considered within the extended Holstein-Hubbard model and bipolaronic theory of superconductivity. A relation between the temperature of Bose-Einstein condensation (T_{BEC}) of an ideal gas of the intersite bipolarons and the strains in the thin films is established. It is shown that the calculated values of T_{BEC} of the bipolarons in the cuprate $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ thin films grown on LaSrAlO_4 and SrTiO_3 substrates correlate with the experimental values of the critical temperature of superconductivity (T_c) of the above systems. A dome-shape type phase diagram of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ thin films in the (T_{BEC}, x) plane is obtained which is similar to the familiar dome-shape type T_c versus x phase diagram of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ thin films. An alternative (bipolaronic) approach to study a dependence of T_c on a film's thickness is proposed. Preliminary results based on this new approach clearly indicate that there exist a profound relationship between high- T_c superconductivity of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ thin films and Bose-Einstein condensation of the intersite bipolarons in the considering system.

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Key words: cuprates, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, superconductivity, Bose-Einstein condensation, polaron, bipolaron, Thickness.

Introduction

Superconductivity (SC) of the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) thin films remains not entirely clear. It was established that SC properties of LSCO thin films differ from that of bulk counterparts depend on several factors such as doping level (x), the thickness of the thin film and the type of substrate for the thin film [1]. All of the above factors affect the SC properties of the thin films, determining, in particular, the value of its critical temperature of superconductivity (T_c). There is contradicting reports on SC of LSCO thin films [2,3]. There are different theoretical models to the strain(pressure)-induced high- T_c phenomena in the LSCO thin films [4, 5, 6, 7, 8]. Though in the above models the authors in some way interpret the numerical values of T_c of the LSCO thin films with different thickness and at different doping levels they do that ignoring the role of the electron-phonon interaction (EPI), in particular the role apex oxygen atoms whose vibrations has a profound effect on the value of T_c of LSCO thin films [9]. It is well established that in cuprates EPI is strong enough for favouring (bi)polaron formation [10] and it manifests itself as polaronic nature of charge carriers in the cuprates [11]. The results of many experiments are also in satisfactory agreement with a (bi)polaronic approach to cuprate superconductivity [12, 13, 14]. In this report we present the results of our study on interpreting SC of LSCO thin films from bipolaronic SC point of view. In doing this, we rely on the extended Holstein-Hubbard (or Fröhlich-Coulomb) model of high- T_c SC which assumes formation of intersite bipolarons and their Bose-Einstein condensation (BEC). The main idea of our approach was given early in the works [15, 16]. In the work [15], the values of T_c of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ and $\text{La}_{1.9}\text{Sr}_{0.1}\text{CuO}_4$ thin films, grown on LaSrAlO_4 (LSAO) and SrTiO_3 (STO) substrates, were satisfactorily explained. The work [16] extends our approach to $\text{RBa}_2\text{Cu}_3\text{O}_{7-\delta}$ cuprates. We will try to explain the dependence of the value of T_c of LSCO thin films, grown on LSAO and (STO) substrates, on doping level x (or Sr concentration) and on the film thickness.

Bipolaron model for SC of LSCO thin films

The cuprates are characterized as a strongly correlated electron (or hole) and strongly coupled electron-phonon system. Such the system can be described by means of an extended Holstein-Hubbard (or Fröhlich-Coulomb) model [17]. It was shown that within the model, at strong EPI regime and nonadiabatic approximation, intersite bipolaron tunnel in the first order of polaron tunneling and its mass has the same order as polaron mass. For the sake of simplicity, we suppose that the intersite bipolarons form an ideal gas of charged carriers and mass of bipolaron is twice of

polaron's mass, i.e. $m_{bp}=2m_p$. Then, the temperature of Bose-Einstein condensation of the intersite bipolarons defines as [15, 16]

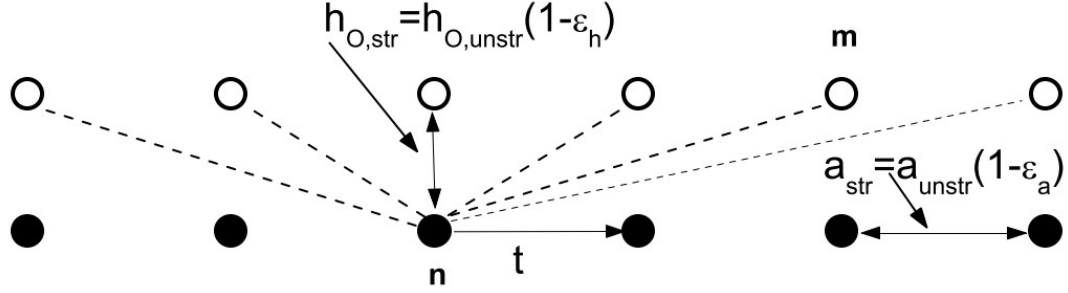


Figure 1: The strained chain model lattice of cuprates. An electron performs hopping motion in a one-dimensional chain of the ions (lower chain - black circles) and interacts with all ions of an upper chain (open circles which is *apical* oxygen ions O(2)) via a density-displacement type force $f_m(n)$ (dashed lines). The distance between the ions of lower chain a is set equal to CuO₂ in-plane lattice period of La_{2-x}Sr_xCuO₄. The distance between the ions of lower chain and the ions of upper chain are assumed equal to h_o which is Cu(1)-O(2) bond length of La_{2-x}Sr_xCuO₄.

$$T_{BEC} = \frac{3.31 \hbar^2 n^{2/3}}{2 k_B m^{\dot{c}}} e^{-g^2}, \quad (1)$$

where $\hbar$ (k_B) is Planck's (Boltzmann's) constant, $m^{\dot{c}} = \hbar^2 / 2t(a)a^2$ is the bare band mass, n is density of the intersite bipolarons and g^2 is the mass renormalization factor of polaronic system. In order to get expression for g^2 , one uses a chain model lattice (Fig.1) that was introduced by Alexandrov and Kornilovitch in ref. [18] in order to mimic an interaction of a hole on CuO₂ plane with the vibrations of *apical* oxygen ions in the cuprates. Convincing evidence for a such coupling of in-plane holes with the c -axis polarized vibrations of apical oxygen ions comes from many experiments (for example [19]). Therefore, here we consider only that component of the electron-lattice force which represents an interaction of a hole on CuO₂ plane with the c -axis polarized apical oxygen vibrations. In addition, for the sake of simplicity, it is assumed that *apical* oxygen ions are dispersionless Einstein oscillators with the vibration frequency ω , and we estimate the mass renormalization factor g^2 within extended Holstein model (which is consistent with the ideality of Bose gas of intersite bipolarons). In that case, the mass renormalization factor g^2 for the given model lattice can be expressed as:

$$g^2 = \gamma \frac{E_p}{\hbar \omega}, \quad (2)$$

where

$$\gamma = 1 - \frac{\sum_m f_m(n) f_m(n+a)}{\sum_m f_m^2(n)} \quad (3)$$

is a numerical coefficient,

$$E_p = \frac{1}{2M\omega^2} \sum_m f_m^2(n) \quad (4)$$

is a polaron shift, M is apical oxygen ion's mass and $f_m(n)$ is the *density-displacement* type EPI force defined by analytical formula

$$f_m(n) = \frac{\kappa h_o(1-\varepsilon_h)}{\sqrt[n-m]{a}} \quad (5)$$

Here κ is some coefficient, $\sqrt[n-m]{a}$ is the distance measured in units of the lattice constant a , h_o is Cu(1)-O(2) bond length, ε_a is the strain of the a -axis lattice period, and ε_h is the strain of h_o . The lattice strains are defined as $\varepsilon_a = (a_{unst} - a_{str})/a_{unst}$ and $\varepsilon_h = (h_{o,unst} - h_{o,str})/h_{o,unst}$, where subscripts *unst* and *str* stand for unstrained and strained, respectively. Thus, the positive (negative) value of the variable ε_i ($i=a, h$) corresponds to the compressive (tensile) strain. Equation(5) is a generalization of the force considered in ref. [18] (see Eq.(9) there). Such an explicit choice of the analytical formula for the force of EPI allows one to interrelate the temperature of Bose-Einstein condensation of the intersite bipolarons with the lattice strains through the mass of intersite bipolaron. The strains contribute to T_{BEC} not only through g^2 but also through bipolaron's concentration n as upon deformation an initial volume V_0 of the sample changes and becomes $V(\varepsilon_a, \varepsilon_b, \varepsilon_c) = V_0(1-\varepsilon_a)(1-\varepsilon_b)(1-\varepsilon_c)$. As a result, the concentration of intersite bipolarons becomes the function of the strains too:

$$n = n_0(1+\varepsilon_a)(1+\varepsilon_b)(1+\varepsilon_c). \quad (6)$$

In obtaining the last expression, we put $n_0 = N/V_0$ (N is the total number of intersite bipolarons in the sample) and neglected the terms of the quadratic order of smallness, i.e. $\sim \varepsilon^2$. The above equations are the main analytical results of the model, according to which discussion of the experimental data for LSCO thin films will be done in the next section.

The dependencies of T_{BEC}

In our study Bose-Einstein condensation temperature of an ideal gas of the intersite bipolarons T_{BEC} (Eq.(1)) expressed through two basic parameters of a system: (i) the density of intersite bipolarons n and (ii) the exponent g^2 of the polaron mass enhancement. Both of n and g^2 depend on the lattices strains ε_a and ε_c . In turn, T_{BEC} depends on the lattice strains. It is well known that the strains in the lattice of LSCO thin films originated from mismatch of the lattice periods of the film and substrate. This mismatch originated strain is also depends on the film's thickness as [20]

$$\varepsilon_a = \frac{2\mu}{4\pi(1-\nu)\bar{B}} \frac{\ln(h_f/\bar{a})}{(h_f/\bar{a})}, \quad (7)$$

where ν is Poisson's ratio, μ is the shear modulus of the film and substrate and is considered to be the same for both of them, $\bar{B}$ is the biaxial elastic modulus of the film, $\bar{a}$ is the mean value of the lattices periods of film and substrate and h_f is thin film's thickness. Below we present the results of our study on the doping dependence of T_{BEC} of intersite bipolarons of LSCO thin films, grown on LSAO and (STO) substrates, on doping level x (or Sr concentration) and on the film thickness h_f .

The doping dependence of T_{BEC}

We calculated the values of T_{BEC} of intersite bipolarons of LSCO thin films, with different doping levels x , grown on LSAO and STO substrates. The results are graphically illustrated in Fig.2.

These values of T_{BEC} are calculated for the doping levels $x=0.075, 0.1, 0.1125, 0.1250, 0.1375, 0.15, 0.2$ and 0.25 . For each doping level, the strains are calculated using the experimental lattice parameters of LSCO both bulk and film. For more details of the calculation we refer reader to our recent work [21]. It is remarkable that T_{BEC} versus x dependence obtained in our model has a dome-like shape similar to the shape of T_c versus x dependence of bulk LSCO, i.e. the phase diagram of bulk LSCO. In such circumstances, Fig.2 can be interpreted as the phase diagram of the strained LSCO thin films. It is interesting that the phase diagram of LSCO thin films exhibits a feature, near doping level $x=1/8$, similar to the observed "1/8"-anomaly in the phase diagram of bulk LSCO. Of course, this "1/8"-anomaly (feature) of the phase diagram of LSCO thin films is originated from *polaronic-structural* effect. And it is one of the conclusions of our model that gives realistic phase diagram for the strained LSCO thin films in (T_c, x) plane with "1/8"-anomaly feature. As one can see from the Fig.2 the calculated values of T_{BEC} of the ideal gas of intersite bipolarons in LSCO thin film grown on LSAO and STO substrates are in satisfactory agreement with the values of the temperature of superconductivity T_c of the considering thin films.

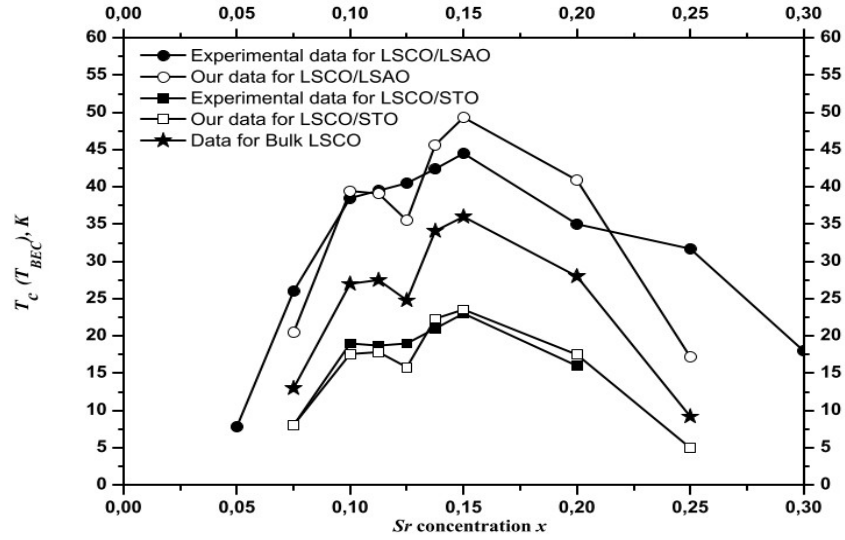


Figure 2: The phase diagram of LSCO films with a thickness of 500 Å in (T_{BEC}, x) plane. The experimental values of T_c of LSCO thin films grown on LSAO (STO) substrate are given by filled circles (filled squares). The calculated values of T_{BEC} of intersite bipolarons of LSCO/LSAO (LSCO/STO) system are given by open circles (open squares). The values of T_{BEC} in both systems are calculated using the lattice parameters of the film and the substrate that correspond to the temperature 295 K. The dashed line presents the experimental values of T_c of bulk LSCO. The lines are a guide to the eye.

The thickness dependence of T_{BEC}

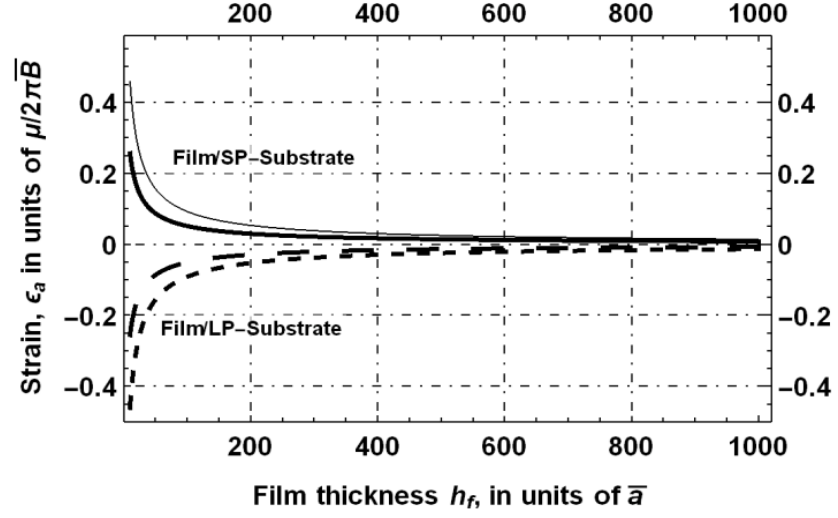


Figure 3: Dependence of the misfit strain ε_a on the thickness h_f of LSCO thin films. The misfit strain ε_a and the film thickness are measured in units of $\mu/2\pi\bar{B}$ and $\bar{a}$, respectively. The solid and long dashed (thin and short dashed) lines are calculated at the value of Poisson coefficient $\nu=0.1$ ($\nu=0.5$)

As we saw above the misfit strain depends on the film's thickness (according to Eq.7). The sign of the misfit strain may be positive or negative depending on the relation between lattice periods of film and substrate. A sign of the misfit strain defined positively and negatively if a film is deposited on a small period (SP) substrate and large period (LP) substrate, respectively. Graphical dependencies of ε_a on the film's thickness, for the lattice of Fig.1 with periods $a=h_o=1$, are given in Fig.3 at the values of Poisson's coefficient $\nu=0.1$ and $\nu=0.5$. It is seen that as the film

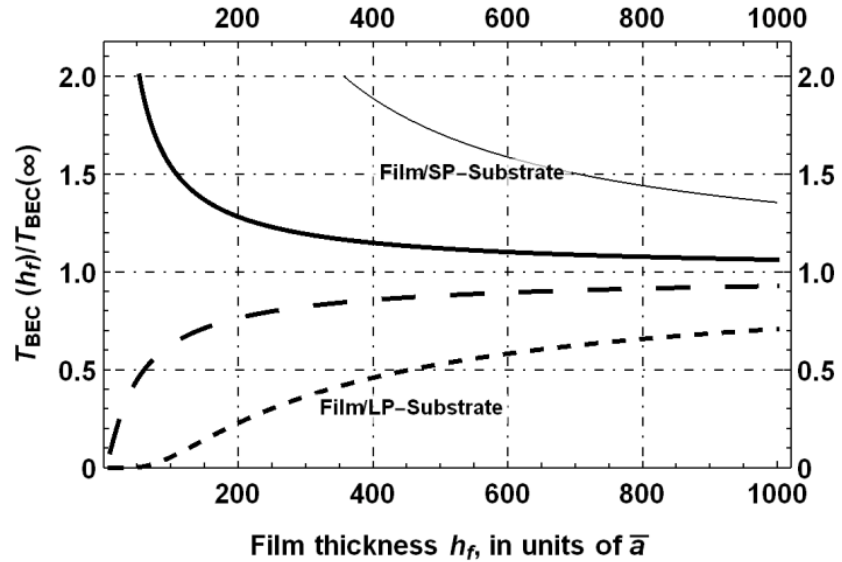


Figure 4: Dependence of normalized $T_{BEC}(h_f)/T_{BEC}(\infty)$ of intersite bipolarons on the thickness of LSCO thin films. The film thickness is measured in units of $\bar{a}$. The solid and long dashed (thin and short dashed) lines are calculated at the value of Poisson coefficient $\nu=0.1$ ($\nu=0.5$)

thickness diminishes the absolute value of the misfit strain increases. In such circumstances, it is expected, according to our model, that when the film is deposited on SP (LP) substrate T_{BEC} should decrease (increase) with the film thickness. Our model convincingly predicts the existence of such a type correlation between T_{BEC} and h_f . In order to show this explicitly we calculated the values of $T_{BEC}(h_f)/T_{BEC}(\infty)$ of intersite bipolarons of the systems LSCO/LSAO and LSCO/STO as a function of the film's thickness. Here, $T_{BEC}(h_f)$ ($T_{BEC}(\infty)$) is the value of T_{BEC} of LSCO film with the thickness of h_f (∞). The thickness $h_f = \infty$ corresponds to the bulk state of LSCO cuprate. The value of $T_{BEC}(\infty)$ is set equal to 36 K which is equivalent the choosing of the value E_p equal to 0,5667 eV. The calculated values of $T_{BEC}(h_f)/T_{BEC}(\infty)$ as a function of the film's thickness h_f are shown graphically in Fig.4 for the same values of Poisson's coefficient $\nu=0.1$ and $\nu=0.5$. Interestingly that the critical temperature of SC T_c of LSCO thin films and Bose-Einstein condensation temperature of the intersite bipolarons of LSCO thin films depend on the film's thickness in the same way. This finding clearly indicates an existence of profound relationship between SC of LSCO thin films and Bose-Einstein condensation of the intersite bipolarons in the considering systems. To the knowledge of authors there are few works that study the dependence of T_c on h_f of LSCO films [6, 7]. The present work opens an alternative path of investigation for study of considering issue. Thus, detailed numerical works that take into account a more realistic crystal structures of cuprates should be done in a future. The latter work is in progress.

Conclusion

We studied the thin films of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ cuprate within the framework of the extended Holstein-Hubbard (or Fröhlich-Coulomb) model. We accepted the bipolaronic mechanism of superconductivity for cuprates in which T_c is associated with the Bose-Einstein condensation temperature T_{BEC} of the ideal gas of the intersite bipolarons. We found satisfactory agreement between the calculated values of Bose-Einstein condensation temperature T_{BEC} of the ideal gas of intersite bipolarons in LSCO thin films grown on LSAO and STO substrates and the temperature of superconductivity T_c of the considering thin films. T_{BEC} versus x dependence obtained in our model has a dome-like shape similar to the shape of T_c versus x dependence of bulk LSCO. This dependence has the "1/8"-anomaly feature and can be interpreted as a phase diagram of strained LSCO thin films. We studied the dependence of T_{BEC} on the film's thickness. And we found that the critical temperature of SC T_c of LSCO thin films and Bose-Einstein condensation temperature of the intersite bipolarons of LSCO thin films depend on the film's thickness in the same way. Our findings clearly indicate the existence of profound relationship between SC of LSCO thin films and Bose-Einstein condensation of the intersite bipolarons of LSCO thin films.

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