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A phase diagram of the thin films of $La_{2-x}Sr_xCuO_4$ cuprates

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Superconductivity of the thin films of the cuprate $La_{2-x}Sr_xCuO_4$, grown on $LaSrAlO_4$ and $SrTiO_3$ substrates, are considered within the extended Holstein-Hubbard model and bipolaronic theory of superconductivity. Within the framework the critical temperature of superconductivity, T_c, is associated with the temperature of Bose-Einstein condensation, T_{BEC}, of a gas of the intersite bipolarons. The Bose-Einstein condensation temperature, T_{BEC}, of the intersite bipolarons of $La_{2-x}Sr_xCuO_4$ thin films grown on $LaSrAlO_4$ and $SrTiO_3$ substrates is calculated as a function of doping level x. The obtained T_{BEC} versus x dependence has a dome-like shape which can be interpreted as a phase diagram of $La_{2-x}Sr_xCuO_4$ cuprates' thin films in (T_{BEC},x) plane.

Keywords: La-based cuprates, thin films, superconductivity, (bi)polarons, Bose-Einstein condensation, phase diagram

I. INTRODUCTION

For almost four decades now, the mechanism of high-T_c superconductivity (HTCS) of the cuprates remains unclear and there is no theoretical understanding of the phenomenon within the framework of a single concept [1]. The existing theoretical models are unable to explain the HTSC of $La_{2-x}Sr_xCuO_4$ (LSCO) cuprate which has the simplest crystal structure compared to other cuprates. The same can be said with respect to HTSC of the LSCO cuprate's thin films whose superconductivity differ from that of bulk counterparts [2].

Study of superconducting properties of the LSCO thin films has almost the three decades history and traces back to the work of Suzuki [3]. Since then, the experimental studies of superconductivity of the LSCO thin films show that the superconductivity of LSCO thin films depends on several factors. These factors are: the doping level (x) or concentration of Sr, the thickness of the thin film itself and the type of substrate on which the thin film is grown. All of the above factors affect the superconductivity of the thin films, determining, in particular, the value of its critical temperature of superconductivity (T_c). Early studies showed that T_c of LSCO thin films depends, at constant doping level, at constant film thickness and under the same thin film growth conditions, on the strains induces by lattice mismatch between the film and the substrate. It was established that for the (001) oriented thin film of LSCO the compressive strain of the *ab*-plane lattice constants enhances the values of T_c while the compressive strain of the lattice period along *c*- direction reduces the values of T_c [4]. For the LSCO thin film growth the two types of substrates, namely $LaSrAlO_4$ (LSAO) and $SrTiO_3$ (STO) with the lattice periods of 3.756 Å and 3.905 Å, respectively, are often used. Due to the fat that bulk LSCO cuprate has the lattice periods a =3.77357 Å and c =13.1973 Å it turns out that *ab*- plane periods of the LSCO thin film grown on LSAO (STO) substrate are compressed (elongated) while the period in the perpendicular *c*- direction is elongated (compressed). While, a research community understand that the

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reason of enhancement and/or reduction of T_c of the thin films originates from the structural changes of the lattice constants of the film at present there does not exist a theoretical model that qualitatively and quantitatively explain the discussing issue from a general point of view and taking into account a relevant features of the cuprates. Of course, there are the variety of qualitative approaches (theoretical models) to the strain(pressure)-induced high-T_c phenomena in the LSCO thin films [5–9] that pretend to the ability of explain the issue. However, the above works do not consider the prominent feature of cuprates that is a strong electron-phonon interaction (EPI) which leads to polaron/bipolaron formation in the strong coupling limit. In this work, we consider the HTSC of the LSCO thin films, taking into account the polaronic features of the phenomenon. For this, we use the extended Holstein-Hubbard (or Fröhlich-Coulomb) model of high-T_c superconductivity which assumes formation of intersite bipolarons and their Bose-Einstein condensation (BEC) giving rise the superconductivity. The main idea of our approach was given early in the works [10, 11]. In the work [10] the values of T_c of $La_{1.85}Sr_{0.15}CuO_4$ and $La_{1.9}Sr_{0.1}CuO_4$ thin films, grown on LSAO and STO substrates, were satisfactorily explained. The work [11] extends our approach to $RBa_2Cu_3O_{7-\delta}$ cuprates. In particular, we will present, here, 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). The dependence might be interpreted as a phase diagram of considering system in (T_{BEC},x) plane.

II. THE MODEL

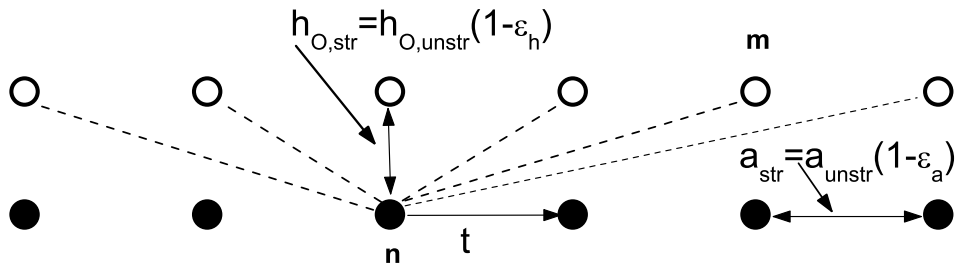


FIG. 1. The strained chain model lattice of cuprates. An electron performs hopping motion in a one-dimensional chain of the ions (lower chain - black circles) and interacts with all ions of an upper chain (open circles which is *apical* oxygen ions O(2)) via a density-displacement type force $f_{\mathbf{m}}(\mathbf{n})$ (dashed lines). The distance between the ions of lower chain a is set equal to CuO_2 in-plane lattice period of $La_{2-x}Sr_xCuO_4$. 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 $La_{2-x}Sr_xCuO_4$.

The cuprates are characterized by the strongly correlated electron (or hole) and the strongly coupled electron-phonon subsystems. Such the complex system can be described by means of an extended Holstein-Hubbard (or Fröhlich-Coulomb) model [12]. 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 mass of polaron and bipolaron have the same order. For the sake of convenience, we assume 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

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of the intersite bipolarons defines as [10, 11]

$$T_{BEC} = \frac{3.31\hbar^2 n^{2/3}}{2k_B m^*} e^{-g^2}, \tag{1}$$

where $\hbar$ (k_B) is Planck's (Boltzmann's) constant, $m^* = \hbar^2/2t(a)a^2$ 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. [13] in order to mimic an interaction of a hole on CuO_2 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 [14]). For this reason, here we consider only that component of the electron-lattice force which represents an interaction of a hole on CuO_2 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). Then, the mass renormalization factor g^2 can be expressed as:

$$g^2 = \gamma \frac{E_p}{\hbar\omega}, \tag{2}$$

where

$$\gamma = 1 - \frac{\sum_{\mathbf{m}} f_{\mathbf{m}}(\mathbf{n}) f_{\mathbf{m}}(\mathbf{n}+\mathbf{a})}{\sum_{\mathbf{m}} f^2_{\mathbf{m}}(\mathbf{n})} \tag{3}$$

is a numerical coefficient,

$$E_p = \frac{1}{2M\omega^2} \sum_{\mathbf{m}} f^2_{\mathbf{m}}(\mathbf{n}) \tag{4}$$

is a polaron energy (shift), M is *apical* oxygen ion's mass and $f_{\mathbf{m}}(\mathbf{n})$ is the *density-displacement* type EPI force defined by analytical formula

$$f_{\mathbf{m}}(\mathbf{n}) = \frac{\kappa h_O (1-\varepsilon_h)}{[\mid (\mathbf{n}-\mathbf{m})(1-\varepsilon_a) \mid^2 + (h_O(1-\varepsilon_h))^2]^{3/2}}. \tag{5}$$

Here κ is some coefficient, $\mid \mathbf{n}-\mathbf{m} \mid$ 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 *unstr* 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. [13] (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:

$$n = n_0(1+\varepsilon_a)(1+\varepsilon_b)(1+\varepsilon_c). \tag{6}$$

In obtaining Eq.(6), 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. $\sim \circ(\varepsilon^2)$. 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.

III. RESULTS AND DISCUSSION

In the previous section, we expressed the temperature of Bose-Einstein condensation of intersite bipolarons T_{BEC} (Eq.(1)) 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. Now, one can study the dependence of the temperature of Bose-Einstein condensation T_{BEC} on the lattices strains ε_a (or ε_b) and ε_c through n and g^2. The latter dependence, of course, originates from

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the polaronic effect. Here, we discuss the possibility of application of Eq.(1) to LSCO films with a thickness of 500 Å grown on two types of substrates, namely, on LSAO and STO substrates. In our formulas, (i) we put M=16 a.m.u. ($2.6565032{\cdot}10^{-26}$ kg); $\hbar\omega$=0.075 eV ($1.2016324237{\cdot}10^{-20}$ J); $n_0 = 1 \cdot 10^{21}$ cm^{-3}. The distance between the ions of lower chain a in Fig.1 we associate with the CuO_2-plane lattice period of tetragonal $La_{2-x}Sr_xCuO_4$. And the distance between the ions of lower chain and the ions of upper chain h_O in Fig.1 we associate with the Cu(1)-O(2) bond length of tetragonal $La_{2-x}Sr_xCuO_4$. The distances a and h_O for the tetragonal $La_{2-x}Sr_xCuO_4$ at different doping levels x (or Sr concentration) can be found in ref.[15]. As for the lattice constants as well as strains of the LSCO thin films grown on LSAO and STO substrates, we took them from ref.[2]. The calculated values of T_{BEC} of LSCO thin films, with different doping levels x, grown on LSAO and STO substrates are presented graphically 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, and 0.2. For each doping level, the strains are calculated using the experimental lattice parameters of LSCO both bulk and film. The experimental values of T_c of LSCO thin films grown on LSAO (STO) substrate are given by open circles (open squares) which connected with the blue line. The theoretically calculated values of T_{BEC} for the LSCO/LSAO and LSCO/STO systems are illustrated with the filled green circles and filled green squares (both connected with green line). For the purpose of comparison the experimental values of T_c of bulk LSCO are also presented with open stared red line.

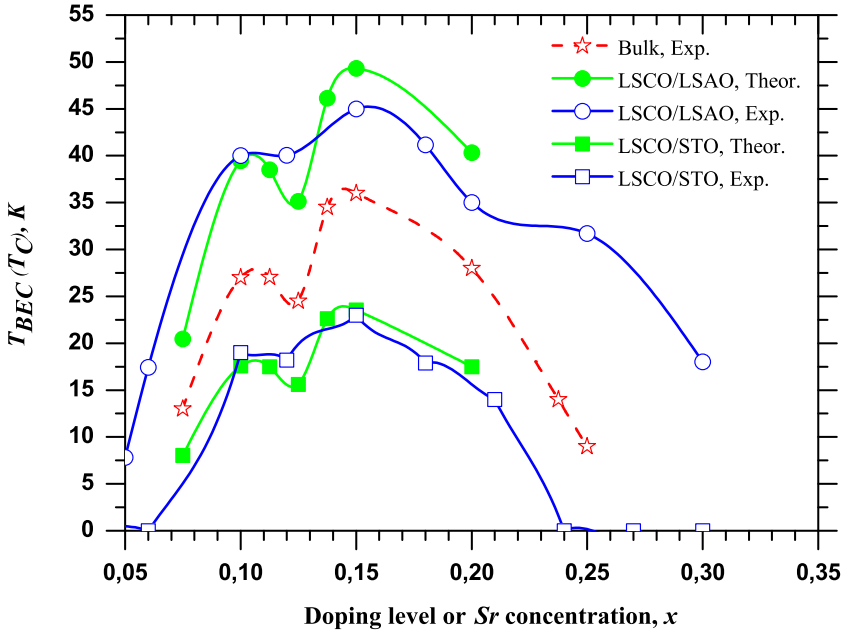


FIG. 2. A phase diagram of LSCO thin films with a thickness of 500 Å in (T_{BEC}, x) plane.

From Fig.2, one can see that there is a good agreement between theoretically calculated T_{BEC} of our model and the experimental values of T_c of LSCO thin films grown on both LSAO and STO substrates. 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 LCSO. In such circumstances, Fig.2 can be interpreted as the phase diagram of the strained LCSO thin films. It is useful also to note that the phase diagram of LCSO 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 cuprate. Obviously, this "1/8"-anomaly (property) of the phase diagram of LSCO thin films is originated from *polaronic-structural* effect. Thus, we have been able, within the proposed approach and the used model, reproduce realistic phase diagram for the strained LSCO thin films in (T_c,x) plane with "1/8"-anomaly feature.

IV. CONCLUSIONS

In conclusion, we studied the thin films of $La_{2-x}Sr_xCuO_4$ cuprate within the framework of the extended Holstein-Hubbard (or Fröhlich-Coulomb) model. Namely, we were interested with the values of T_c of LSCO thin films grown

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on LSAO and STO substrates. 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. The dependence of T_{BEC} on doping level x obtained in our model has a dome-like shape similar to the shape of T_c versus x dependence of bulk LSCO. The dependence has the "1/8"-anomaly feature and can be interpreted as a phase diagram of the strained $La_{2-x}Sr_xCuO_4$ cuprates' thin films.

COMPETING INTERESTS

The authors declare that they have no competing interests.

AUTHOR'S CONTRIBUTIONS

All authors contributed equally to the article. All authors have read and agreed to the published version of the manuscript.

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