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Doped Mott Insulators within the Strong Coupling Regime

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Abstract: We have studied the thermodynamic properties of electron doped Niodium Cellenium Copper Oxide ($Nd_{2-x}Ce_xCuO_4$ -NCCO) and Praseodymium Cellenium Copper Oxide ($Pr_{2-x}Ce_xCuO_4$-PCCO) and hole doped Yitrium Barium Copper Oxide (YBa_2CuO_7-YBCO) and Lanthanum Strontium Copper Oxide ($La_{2-x}Sr_xCuO_4$-LSCO) within the framework of modified Bose-Fermi-Hubbard model in the strong coupling limit where U/t=12. The total energy of the system increases exponentially with the temperature which is attributed to increased thermal fluctuations thus enhancing the conduction electrons. At lower temperatures, entropy increases exponentially and corresponds to ~3.1 $\times 10^{-3}$eV/K^2 for YBCO and ~3.2 $\times 10^{-3}$eV/K^2 for LSCO occurring respectively at 297 K and 239 K is noted. At 200 K, NCCO and PCCO have numerical entropies of ~3.097 $\times 10^{-3}$eV/K^2 and ~2.663 $\times 10^{-3}$eV/K^2 respectively. Apparently, the maximum entropy for electron-doped system is smaller than that of the hole-doped ones indicating that electron-doped PCCO would be preferred for high-T_C superconductivity since SC process is a high order process that requires low entropy

Keywords: thermodynamic properties, strong coupling limit, Bose-Fermi-Hubbard model, entropy.

1. Introduction

High temperature cuprate superconductors were discovered in 1986 [1], who cooled down $LaBaCuO_4$ and discovered its transition temperature at 30 K. Many other high-T_C cuprates were discovered in the subsequent years. The highest recorded T_C is 166 K, observed in a Hg-based cuprate under pressure [2]. Recently, the appearance of superconductivity in H_2S at high pressures [3], [4] beat down the record of higher T_C held by the cuprates. The high-temperature cuprate superconductors may be categorized as hole- and electron-doped materials. The hole-doped materials constitute the majority of the cuprate superconductors, while electron doping is observed in a relatively small number of materials, mainly the $(Nd, Pr)_{2-x}Ce_xCuO_4$ systems and the infinite-layer (Sr, L) CuO_2 (L=La, Sm, Nd, Gd) materials [5]. The phase diagrams of the hole- and electron-doped materials have some similarities [6] with the parent materials being antiferro-magnetic (AFM) insulators in both cases. However, the electron-doped materials are also noticeably different in that the AFM region extends to a much higher doping with an almost non-existent pseudogap region [7]. The crystal structure of the electron-doped compounds is characterized by a lack of apical oxygens, resulting in a compound that is a Slater insulator material in which the insulating behavior is the result of the presence of magnetic long-range order. This is in sharp contrast with the hole-doped materials, which are insulating due to the strong electronic correlations but not owing to magnetism.

The biggest motivation for research in high temperature superconductivity is the determination of the conduction mechanism. The discovery of superconductivity in the cuprate oxides, especially above the liquid Nitrogen temperature, openly challenged the BCS theory on many fronts such as isotope shift, short coherence length, high transition temperatures, electric and magnetic anisotropies etc which were notably different from conventional superconductors. Spin fluctuation mechanism (where pairs are bound because of magnetic interactions between the electrons'spins) has been tried with little success. The nature of the electron-boson interaction or coupling in high-T_C cuprate superconductors remains a debated issue [8] due to incomplete comprehension of the electronic correlation that leads to the pseudogap formation [9]. Hence studying the coupling between electrons and bosonic excitations that mediate the formation of Cooper pairs besides boson-boson and electron-electron interaction is significant to understand superconductivity. There was spirited theoretical and experimental effort to explain the nature of their microscopic behavior as they are not completely described by the BCS theory [10]. However, very little theoretical work has been dedicated to thermodynamic properties [11].

Several theoretical models have been advanced to explain high temperature superconductivity. The Hubbard model is exactly solvable in one dimension, where the ground state at half filling is a Mott insulator for any repulsive non-zero interaction [12]. Despite the simplicity of this model, the physics arising in multiple

dimensions casts uncertainties and needs to be explored. Many-body approximations have been applied along the years in different lattice geometries and coupling regimes (U/t). Among them, is the Quantum and Variational Monte Carlo calculation, Dynamical Mean-Field Theory, Density Matrix Renormalization Group and later, Density Matrix Embedding Theory [13] which have shown their limitations to describe the strongly correlated regime of cuprates. The strong finite size hinders extraction of low-energy scales, which are important to capture the competition between different ground states often present in correlated electron systems [14]. However, all these approaches have shown their inadequacies to describe the strongly correlated regime of the Hubbard model ($U/t \gg 1$) [12]. Cuprate superconductors belong in the strong coupling regime where U/t~12 to 16 [15]. In this study, we focused on the strong coupling regime where U/t=12. The two-dimensional t-J model has been studied [16] for many years as a model for the copper oxide planes found in high temperature superconductors. Despite considerable effort, the basic thermodynamic properties of this model are still not well understood and that its basic feature is still controversial [17], [18]. Irrespective of intensive and extensive efforts, it is still difficult to solve exactly the two-dimensional t-J model, and thus the mechanism of the high-temperature superconductivity still remains puzzling up to now [19].

In this work we deduce the internal energy, E and the entropy, S(T) of selected hole and electron doped cuprates. The electron-doped compounds Nd_{2-x} Ce_xCuO_4 (NCCO) and Pr_{2-x} Ce_xCuO_4 are studied and compared with YBa_2CuO_7 (Y123) and La_{2-x} Sr_xCuO_4 (LSCO)-hole-doped materials by using Bogoliubov-Valatin transformation technique.

2. Model and Approach

Doping introduces free electrons at the Fermi level of the superconductor. The free electrons interact among themselves and so to the formed cooper pairs within the crystal. The free electrons (fermions) could well be described by Fermi-Hubbard model while the interacting cooper pairs (bosons) are described by the Bose-Hubbard model. In our Hamiltonian an additional term accounting for the interaction between the bosons and fermions is introduced. Considering a doped high-T_C system where particles are interacting, the Hamiltonian can be decomposed as;

$$H_T = H_b + H_f + H_{b-f} \tag{3.1}$$

Where H_b represents interacting bosons (described by the Bose-Hubbard model), H_f accounts for the interacting fermions (described by Fermi-Hubbard model) and H_{b-f} represents the interaction/ coupling between fermions and bosons.The effective Hamiltonian, for this system in a strong coupling regime is given by;

$$H = -J\sum_{\langle k,-k\rangle}(b_k^\dagger b_k + b_{-k}^\dagger b_{-k}) - \mu\sum_k b_k^\dagger b_k + \frac{U}{2}\sum_k b_k^\dagger b_k (b_k^\dagger b_k - 1) \tag{3.2}$$
$$-t\sum_k c_k^\dagger c_k - U\sum_{k,-k} c_{k\uparrow}^\dagger c_{-k}^\dagger c_{-k\downarrow} c_{k\uparrow} - \mu\sum_k c_k^\dagger c_k - U\sum_k c_k^\dagger c_k b_k^\dagger b_k$$

This Hamiltonian is diagonalized by both fermionic and bosonic canonical transformations resulting into ground state energy of the system.

$$E_0 = -4t - 2\sqrt{5}\,\Delta - 8\mu + 8J + 6U \tag{3.3}$$

This is based on the fact that, at high temperatures most of the electrons are thermally excited to higher states and thus, all the occupation numbers are equal to zero. Besides, the coefficients of the off diagonal terms gives; $v_k = 2$; $u_k = \sqrt{5}$ [20].

In order to introduce temperature dependence and project the properties of the system, equation (3.3) is multiplied by a thermal activation term $e^{\frac{-E_0}{kT}}$ giving rise to the system energy;

$$E_T = \left(-4t - 2\sqrt{5}\Delta - 8\mu - 8J + 3U\right)e^{-\frac{\left(-4t-2\sqrt{5}\Delta-8\mu-8J+3U\right)}{100kT}} \tag{3.4}$$

The system entropy is determined to be;

$$S = \left(K + \frac{\left(-4t-2\sqrt{5}\Delta-8\mu-8J+3U\right)}{T}\right) \tag{3.5}$$

The critical temperature is obtained by taking the partial derivative of specific heat, C_V [20].

$$T_C = \frac{\left(-4t-2\sqrt{5}\Delta-8\mu-8J+3U\right)}{200k} \tag{3.6}$$

3. Results and Discussion

3.1 Internal Energy

The results obtained for hole-doped cuprates are shown in figure (1.1). Qualitatively, the curves for YBCO and LSCO, show a similar behavior which is in consonance with other theoretical findings. The Curves are analogous to the ones obtained by [21] while studying the thermodynamics of the two-dimensional Hubbard

model based on the exact two-body s-matrix. Similar curves (inset) were obtained by [22] while working on the phase diagram and thermodynamics of the three-dimensional Bose-Hubbard model. Quantitatively however, LSCO has higher energy than YBCO up to a temperature of ≈290 K where both have same energy (0.8935 eV). Perhaps, this energy could be an indicator of the transition point from the superconducting state to the normal state. However, at temperatures approaching room temperature (T>290 K), the trend is reversed with YBCO showing a lower rate of change of system energy. The transfer energy (t) corresponding to YBCO and LSCO are 0.8067 eV and 0.7530 eV respectively (Table 1.1). Hence the displayed results are a manifestation that superconductors with higher transfer energy undergo a high temperature rate of change of energy and the converse is true for those with lower transfer energy. Based on these findings, one can argue that YBCO would be a better candidate for the construction of a room-temperature superconductor compared to LSCO since high-T_C superconductivity is a low-energy process which demands that the system energy should be kept as low as possible.

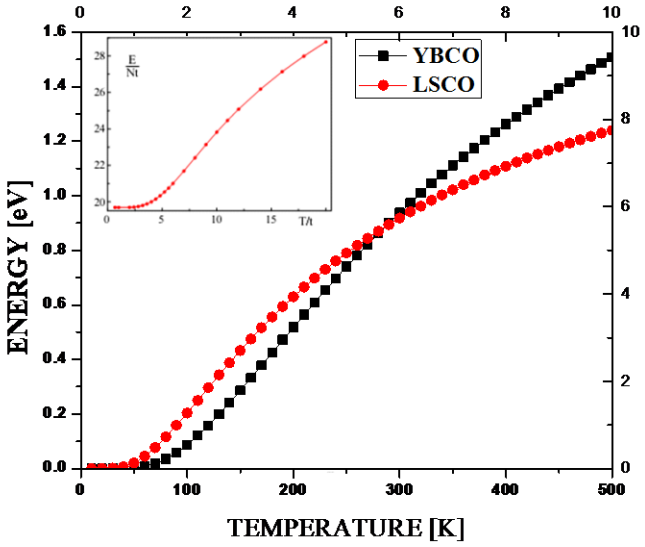

Figure 1.1: Variation of system energy with temperature for YBCO and LSCO. Experimental parameters: t=0.44eV, U=12t, J = 0.4t, μ=U/2 and Δ=1.6 eV for YBCO and t=0.43 eV, U=12t, J = 0.4t, μ=U/2 and Δ=1.8 eV for LSCO

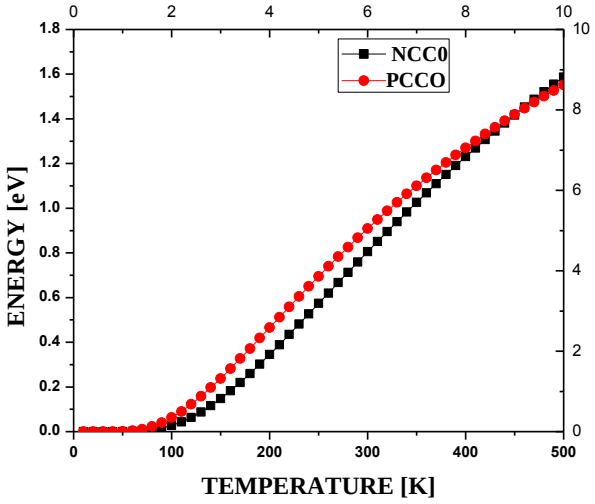

Figure 1.2: Variation of system energy with temperature for NCCO and PCCO. Experimental parameters: t=0.42 eV, U=12t, J = 0.4t, μ=U/2 and Δ=1.2 eV for NCCO and t=0.38 eV, U=12t, J = 0.4t, μ=U/2 and Δ=1.2eV.

Figure (1.2) shows the energy curves obtained for the electron doped cuprates. The behavior of the curves is qualitatively the same as for the hole-doped cuprates and concurs with the findings by [22], [21] and [23]. At 300 K, NCCO and PCCO have energies 0.805 eV and 0.909 eV respectively. The two cuprates have the same energy of 1.362 eV at T=430 K. Conversely, the hole doped cuprates gave 0.918 eV energy at 300 K and registers lower energies thereafter. Both the hole doped and electron doped cuprates have comparable energies at 300 K. However, NCCO is projected to have the least energy at room temperature and would therefore be a preferred material for room temperature superconductivity.

At T=0, the energy of either system is zero in conformity with equation (3.4) and consistent with the nature of super-fluids. This scenario implies that the states above the superconducting gap are empty while those below are completely filled. The system energy increases with temperature and this is attributed to increased thermal fluctuations which enhances the conduction electrons. As a result, the cooper pairs break up and the superconducting gap is suppressed. The reasonable correspondences of the energy magnitudes and temperature dependencies suggest that the electron-doped materials are approximately as strongly correlated as the hole-doped ones. The numerical values for transfer energies, t of the four cuprates is determined, using equation (3.6) and considering their experimental T_C values to be; YBCO-0.8067 eV, LSCO-0.7530 eV, NCCO-1.4123 eV and PCCO-1.1493 eV. The room temperature superconductivity corresponds to a $T_C \approx 300$ K. This value can thus be obtained if the transfer energies are raised to: 2.6023 eV for YBCO, 5.9447 eV for LSCO, 17.2935 eV for NCCO and 15.3924 eV for PCCO. From, these results, one gathers that for both categories of cuprates, the higher the T_C value the higher the transfer energy. Hence based on our model, YBCO requires the least amount of energy transfer for it to strike room temperature superconductivity. The transfer energy can be increased by increasing the on-site coulomb repulsion, U and exchange energy, J from the relation given by [24].

3.2 TEMPERATURE DEPENDANCE OF SUPERCONDUCTING ENTROPY

The qualitative features of the observed cuprate entropy are reproduced using our model for both hole and electron doped cuprates [16], [18], [25], [26], [27], and [28]. An exponential growth of entropy for both cases is noted at lower temperatures as in figures (1.3) and (1.4). The entropy curves concurs with the ones obtained by [29] while studying low temperature statistical thermodynamics of binary Bose-Fermi system. The maximum entropy for YBCO and LSCO is approximately 3.1×10^{-3} eV/K and 3.2×10^{-3} eV/K respectively. Clearly, from figure (1.3), the rate of increase of entropy with temperature of the system is higher for LSCO compared to that of YBCO and hence YBCO is a better candidate material for High- T_C superconductivity due to its lower entropy value. The transition temperature corresponding to the peak entropy for YBCO is $T_C = 297$ K and that of LSCO is $T_C = 239$ K. From a thermodynamic point of view, the superconducting (SC) phase appears below the T_C because the free energy of the SC phase becomes less than the free energy of the normal phase for all temperatures below T_C [30]. The state of disorder diminishes with decrease in the internal energy of the system (i.e particles settle and interact less as the system liberates energy) [31] which mirrors our results in figure (1.3) and (1.4). Our results point out that the entropy in the SC state is less than that in the normal state for all temperatures below T_C. This implies that the SC is more ordered than the normal state i.e the fraction of electrons that are thermally excited in the normal state becomes ordered in the SC state. Our value for entropy is in faithful conformity with other theoretical studies as reported by [23], [26] and [28].

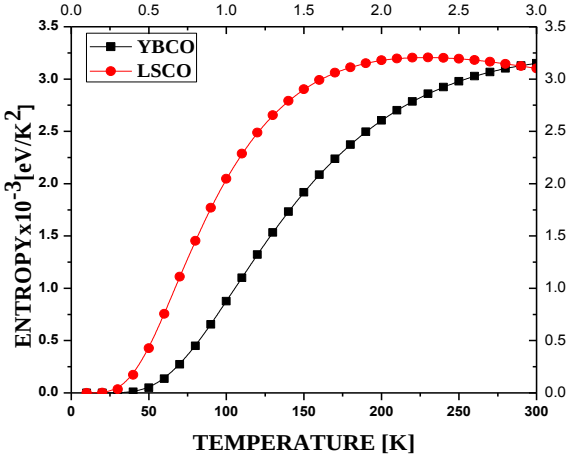

Figure 1.3: The thermodynamic entropy in superconducting state for the hole-doped cuprates

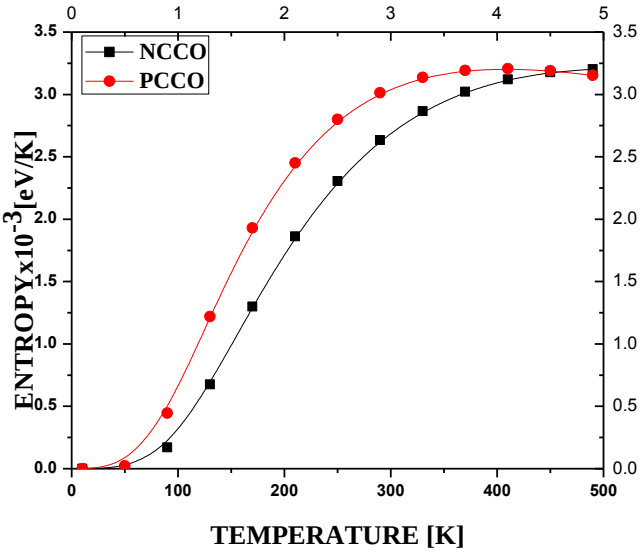

Figure 1.4: The thermodynamic entropy in superconducting state for the electro-doped cuprates.

Figure (1.4) shows the variation of entropy with temperature for electron doped cuprates (NCCO and PCCO. The curves obtained are sigmoidal and entropy increases exponentially with temperature with NCCO having higher value than PCCO. For, NCCO, the maximum entropy is 3.097×10^{-3} eV/K at $T_C = 200$ K while for PCCO is 2.663×10^{-3} eV/K at $T_C = 200$ K. Comparing the two electron doped cuprates, PCCO is thus a better choice for high-T_C superconductivity. The values of this theoretical study are thus in close proximity to the range of values from other research findings. Our results clearly shows that the entropy of the electron-doped system is smaller than that of the hole-doped ones and this is attributed to the strong AF short-range correlation that survives longer in the electron-doped system. The fact that the entropy heads towards zero as $T\rightarrow0$, is important as it asserts the thermodynamic consistency of our data. Entropy is a state of molecular disorder and particles' internal energy reduces as the system cools culminating into reduced particle motion [32]. This observation is consistent with our results for both hole and electron doped cuprates as typified by the graphs in figure (1.3) and (1.4).

Conclusion

We have been able to modify and diagonalize the Bose-Fermi-Hubbard model using BVT. We further examined the internal energy and hence entropy from the diagonalized Hamiltonian within the strong coupling limit. We deduce that superconductivity is a low energy process and hence, YBCO is projected to be the likely suitable material for the construction of room temperature superconductors. Our results point out that the entropy in the SC state is less than that in the normal state for all temperatures below T_C. This implies that the SC state is more ordered than the normal state i.e the fraction of electrons that are thermally excited in the normal state becomes ordered in the SC state. The lowest entropy of ~$3.1.0\times10^{-3}$ eVK^{-1} (YBCO) and ~2.6×10^{-3} eVK^{-1} (PCCO) is obtained from our model and we deduce that superconductivity in the electron-doped cuprates is highly ordered compared to the hole- doped ones.

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