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Absence of phase separation in the frustration-induced hole-interaction mechanism

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The pair-correlation functions for a hole-induced spin frustration model within a Bethe lattice approach is investigated. It is shown that the induced attraction between holes provokes the formation of small correlated clusters but it is not strong enough to promote a phase separation instability. We relate our results with the short correlation length of holes in high-T_c superconductors.

After the discovery of high-T_c superconductors,[1] there has been many suggested alternative pairing mechanisms to the traditional electron-phonon interactions,[2] but none is widely accepted. A large number of experimental studies have established that superconductivity arises after the doping of antiferromagnetic insulators.[3] This procedure generates holes in the CuO_2 sheets whose existence is the common structural feature to all high-T_c superconductors.[4] Among the suggested mechanisms in the literature, there exists a class whose origins lie on charge-transfer effects[5] and another class based on magnetic effects,[6,7] both associated with the hole degrees of freedom.

The most commonly studied model in the search for the pairing mechanism is the so-called *t-J* model.[6] However, a problem has been detected in such models connected with the existence of a phase separation, that is, the resultant effective attraction between holes does not ensure superconductivity but it may provoke a phase separation into two phases of different hole densities.[8] Hirsch *et al.*[9] have examined the stability of the ground state of CuO_2 finite systems with respect to a real phase separation of holes in the regime of attractive pair interaction. They have observed that both possibilities of stable or unstable situations may exist as a function of the Coulomb repulsion and kinetic energy with the unstable phase favored when larger clusters are considered. Experimental evidence on the existence of phase separation has been reported and its occurrence is attributed to both charge and spin degrees of freedom.[10] Recently, the investigation of the $U=\infty$ limit of the three-band Hubbard model,[11] exhibited a charge-transfer instability (CTI) in the ground state at a mean-field level just as in the weak coupling theory where a Hartree-Fock approach was employed.[12] It is found that associated with this CTI there is always a diverging compressibility leading to a phase separation. On the other hand, Moreo *et al.*[13] have studied a two-dimensional Hubbard model and have shown through the behavior of the density of particles as a function of the chemical potential, that it does not phase separate. Mean-field calculations based on an extended *t-J* model, namely, the Kondo-like spin-hole coupled, where competition between magnetic couplings is included,[14] exhibit phase separation and superconductivity depending on the relative values of the couplings. The results show that phase separation does not disrupt superconductivity and suggest that formation of pairs can stabilize the system against it, extending, in this way, the stable superconducting region.

Based on the idea that the holes are essentially localized at the O sites and induce a ferromagnetic interaction between neighboring Cu sites which were previously antiferromagnetically coupled, Aharony *et al.*[7] have shown that an effective attractive potential arises between mobile holes in order to minimize the frustration effects induced by those competing interactions and suggested that this pairing mechanism could lead to superconductivity with no reference to phase separation. Recently, we went beyond the mean field, evaluating exactly first neighbor hole-hole correlation within the hole-induced frustration model through a Bethe lattice approach.[15] Having fitted remarkably well the experimental data of $La_{2-y}Sr_yCuO_4$, we concluded that the superconducting transition takes place at an equicorrelation line in the temperature versus hole concentration plane.

In the present work we show that a phase with uniform density of holes is always stable for the hole-induced spin frustration model. We reach this conclusion through the behavior displayed by the various hole-hole correlations together with an analysis of the thermodynamic stability of system. Contrary to our previous paper[15] where we claimed the existence of pairing, we show that clusters of two holes are not obviously stable in this model. Actually the model leads to the formation of small correlated clusters with correlation length $\xi \leq 3$, in units of lattice spacing. We also discuss why phase separation does not occur in such a model in contrast with previous results by other approaches.

Let us begin by randomly distributing a mean concentration of holes in an antiferromagnet Ising background. The holes are localized at the O sites between two Cu ions. The net magnetic moment of a hole interacts with the Cu spins in such a way that it turns their coupling into an effective temperature-dependent coupling J_{eff} which at low temperatures will compete with the antiferromagnetic one J between the undoped pairs. The mobility of holes is considered by taking annealed averages over the hole distribution. It means that the holes are permitted to move through all the configurations that minimize globally the Helmholtz free energy. This constraint gives rise to correlations between holes so that their distribution is not random at all. the Hamiltonian that describes the magnetic interactions mentioned above can be written as

$$H=-\sum J(1-n_{ij})S_iS_j+J_{\rm eff}S_iS_jn_{ij}+\mu n_{ij}\ , \tag{1}$$

where the sum runs over all nearest-neighbor pairs and μ is a chemical potential per hole which controls the mean concentration of holes. Here, n_{ij} is the hole occupation number of the bond connecting the sites i and j, taking on values zero or one, for we do not allow double occupation. Besides being temperature dependent $J_{\rm eff}$ is a function of the αJ coupling between Cu spins and hole spins and also of the hole mediated coupling between the Cu ions γJ.

The hole concentration can be obtained through the evaluation of $\langle n_{ij}\rangle$, where $\langle \cdots \rangle$ means thermodynamic average over spin and hole configuration. The correlations between holes are defined as

$$C_R=\langle n_{ij}n_{k1}\rangle-\langle n_{ij}\rangle^2\ , \tag{2}$$

where R is the distance vector between the bonds i—j and k—1, so that C_1 is the first neighbor correlation, C_2 next-nearest neighbor correlation, and so on. An attraction tendency between holes implies that $C_R>0$. Likewise, if $C_R<0$ the holes tend to be apart. These correlation functions are exactly obtained when one represents Cu ions by Ising spins localized at the vertices of a Bethe lattice. This procedure has shown to work quite well in the range of parameters that is relevant to superconductivity.

Figure 1 illustrates some equicorrelation lines in the portion of the temperature versus hole concentration plane over which the transition to the superconductor state is supposed to occur for systems where the interaction between CuO_2 planes is neglected. Notice that the maximum value attained by the temperature at each equicorrelation line occurs around $x=0.15$ as is experimentally observed in a large variety of materials at the superconducting transition.[17] Also, the smaller the correlation required for the system to reach the superconductor state, the higher is the maximum in T_c. This means that, heavy-fermion systems where kinetic energies occur on a small scale compared with the energies involved by frustration, are naturally the best ones to look for higher T_c. Interplane coupling can also enhance the correlations and consequently increase T_c. The antiferromagnetic-paramagnetic line coincides in this approach with the line of zero correlation. This is due to the low degree of connectivity of the Bethe lattice and probably is not true for more realistic lattices.

Let us now discuss the stability of our system with respect to phase separation of holes. In the case of phase separation, at $T=0$ the system is composed of a hole-rich phase, with all holes condensed and a hole-poor phase where all bonds are hole free. In this way, one should have $C_1=x-x^2$. However that is not what happens. In Fig. 2 we plot the nearest-neighbor hole correlation versus hole concentration for a few temperatures. We choose coupling parameters in such a way that the system is frustrated at low temperatures. Notice that, at small hole concentrations the induced correlation increases as the temperature increases, which is a clear evidence that this correlation is induced by an energy minimization process due to frustration. For higher hole concentrations, this sort of behavior is changed indicating that thermal fluctuations are also important. However what is surprising in Fig. 2 is that at low temperatures the correlation does not reach the expected value for phase separation. It can reach at most half of that value for very small levels of concentration. The one-half factor can be easily understood if we think that for small concentrations the energy is minimized when clusters of four holes with a common neighbor Cu site are formed. In this way there is no frustration. The clusters are so apart that they are independent of one another. In this case, $\langle n_{ij}n_{kl}\rangle=x(3+3x)/6$, and so $C_1=(x-x^2)/2$. As hole concentration is increased the factor is even smaller

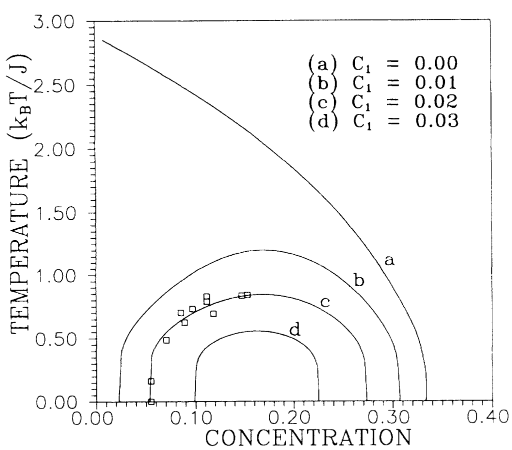

FIG. 1. Equicorrelation lines using the values $\alpha=1.9$ and $\gamma=0.69$ from *ab initio* calculation on finite clusters (Ref. 16). The data (squares) are from Shafer *et al.* (Ref. 18) for the superconducting transition in $La_{2-y}Sr_yCuO_4$.

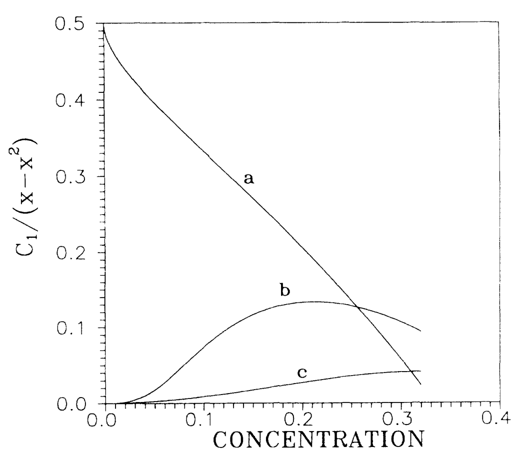

FIG. 2. First neighbor hole-hole correlations for $\alpha=1.5$ and $\gamma=1.0$, and (a) $k_BT/J=0.025$, (b) $k_BT/J=0.525$, (c) $k_BT/J=1.025$. Notice the crossover from frustration to fluctuation-induced pairing mechanism.

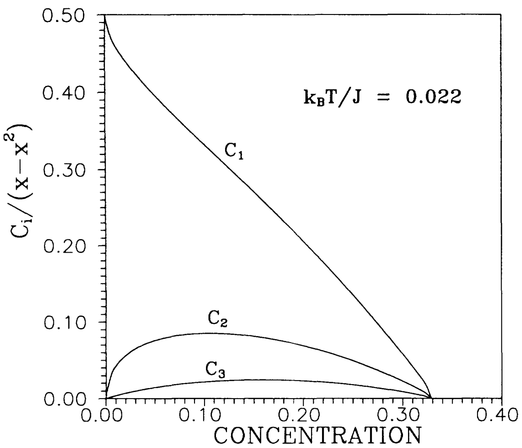

FIG. 3. First three hole-hole correlations for $\alpha=1.5$ and $\gamma=1.0$, at very low temperatures ($k_BT/J=0.022$).

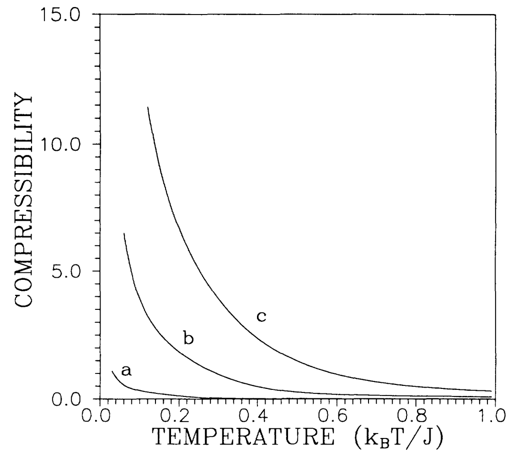

FIG. 5. Isothermal compressibility for $\alpha=1.5$ and $\gamma=1.0$ as a function of temperature for (a) $x=0.01$; (b) $x=0.10$; and (c) $x=0.30$.

than one-half. One can argue that this behavior is due to the breakdown of the antiferromagnetic background and that it may well prevent the system from a phase separation instability.

Having that in mind, we proceed to investigate the possibility that the system undergoes a phase separation. First we turn to Fig. 3 where we plot the hole-hole correlations for $R=1,2,3$ for very low temperatures. A general feature that we may observe is that for low concentrations C_2 and C_3 go to zero while C_1 goes to $(x-x^2)/2$, reinforcing the idea that independent clusters of four holes are formed. Note that for intermediate hole-concentration region, the function C_R for $R\geq 2$ increases a little and so by just looking at correlations between holes, one is not able to ensure that no infinite correlated clusters grows. The only undoubted way to study the stability of the system is to look for a diverging isothermal compressibility $\partial x/\partial\mu|_T$. Here, we compute it using the fluctuation-dissipation theorem,[19] summing up all hole-hole correlations:

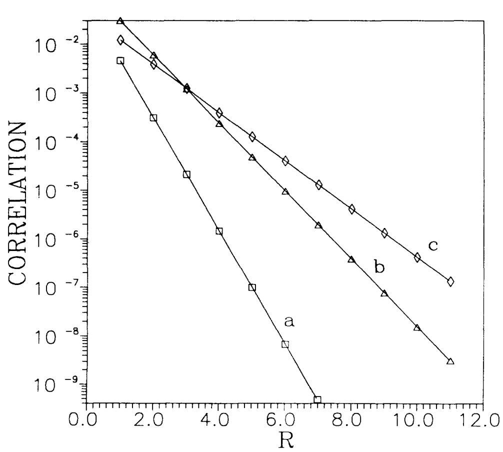

FIG. 4. Hole-hole correlations for $\alpha=1.5$ and $\gamma=1.0$ as a function of hole distance for (a) $x=0.01$; (b) $x=0.10$; and (c) $x=0.30$, and $k_BT/J=0.022$.

$$\left.\frac{\partial x}{\partial\mu}\right|_T=\frac{1}{k_BT}\sum_{R=0}^{\infty}N(R)C(R)\,, \tag{3}$$

where $N(R)$ is the number of neighbors at a lattice distance R, which for a Bethe lattice is exactly given by $N(R)=q(q-1)^R$, with q as the coordination number ($q=4$ in our approach). The correlations decay exponentially with R for all temperatures and hole concentration. In Fig. 4 we show the particular case of low temperatures, where the first-neighbor correlations are stronger in the regime of intermediate concentration. The series in Eq. (3) can then be easily summed up to give

$$\left.\frac{\partial x}{\partial\mu}\right|_T=\frac{1}{k_bT}\left[x-x^2+\frac{qC_1^2}{C_1-(q-1)C_2}\right], \tag{4}$$

where the self-correlation term is included. The compressibility is always well behaved as a function of temperature for all hole concentration (see Fig. 5). It diverges at $T=0$ indicating that a phase separation could take place only at the ground state. However, as the effective hole-hole interaction demonstrates when it vanishes as T goes to zero,[20] that is not the case. Actually, this divergence is similar to the one exhibited by noninteracting systems where $\chi\approx 1/T$. As it is depicted in Fig. 4, only clusters with finite correlation length are formed even at very low temperatures, that is, as $T\rightarrow 0$ $C_R\propto\exp(R-1)/(\xi-1)$, with $\xi\leq 3$. Thus the frustration-induced hole-attraction mechanism leads to a short correlation length between holes as it seems to be in the copper-oxide superconductors.[21]

In conclusion, we have investigated the correlation functions for the hole-induced frustration model through a random distribution of holes on an antiferromagnetic background represented by Ising spins placed on a Bethe lattice. By means of the fluctuation-dissipation theorem we obtained the isothermal compressibility which does not exhibit divergence at finite temperature. Our results show clearly that frustration effects are not strong enough to promote a phase separation and in this sense our model is closer to the one-band Hubbard model than to the t-J model. Furthermore, as we allow the holes to hop anywhere they can, according to the minimum condition on the Helmholtz free energy in our approach, our results agree with the large hopping amplitude of t-J-like,[8,14,22] and extended Hubbard models.[9,11,12] In this way, we may conjecture that, in addition to frustration effects, one must have a local charge transfer mechanism to stabilize a phase separation of holes. The frustration content is only strong enough to permit the formation of small correlated clusters, and we have found that near half-filling (zero doping) clusters of four holes are stable. The fact that clusters of more than two holes are stable, is a feature of the model that is not in agreement with experiments. We believe that this is due to the noninclusion of a kinetic energy term which would actually act as a cluster breaker. The effect of the kinetic energy on the clustering phenomenon displayed in the present work is being subject of current work.

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