Pseudogap Induced Antiferromagnetic Spin Correlation in High-Temperature Superconductors

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The pseudogap phenomena observed on cuprate high temperature superconductors are investigated based on the exact diagonalization method on the finite cluster $t$-$J$ model. The results show the presence of the gap-like behavior in the temperature dependence of various magnetic properties; the NMR relaxation rate, the neutron scattering intensity and the static susceptibility. The calculated spin correlation function indicates that the pseudogap behavior arises associated with the development of the antiferromagnetic spin correlation with decreasing the temperature. The numerical results are presented to clarify the model parameter dependence, that covers the realistic experimental situation. The effect of the next-nearest neighbor hopping $t'$ is also studied.

I. INTRODUCTION

The high-temperature superconductors exhibit various anomalous metallic properties in their normal phases. A gap-like behavior, first discovered by the NMR relaxation rate measurement [1] as one of these properties has attracted a lot of current interest. It is characteristic to the underdoped cuprates and observed as a broad peak in the $1/T_1 T$-$T$ curve at a slightly higher temperature than the superconducting transition point $T_c$. The possible relation of the pseudogap phenomena with the origin of the superconductivity underlies the reason of such interest. The behavior has now been detected in the temperature dependence of various other physical quantities; the neutron scattering intensity [2,3], the magnetic susceptibility [4], the resistivity [5], the Hall coefficient [6] and the angle-resolved photo-emission spectrum (ARPES) [7].

A lot of theories have been proposed to explain the phenomena. Since they were observed only for compounds with double CuO$_2$ layers like YBa$_2$Cu$_4$O$_8$ for the first time, the interlayer coupling was supposed to play the predominant role on the gap formation. [8-10] However, after the discovery of the phenomena on the mono-layer oxide HgBa$_2$CuO$_4$ by the NMR measurement [11], most theories are based on the model with a single CuO$_2$ plane, such as the square-lattice $t$-$J$ model which has been extensively studied theoretically in the description of the cuprate superconductivity. [12] Based on the concept of the spin-charge separation. [13] Tanamoto et al. [14] showed that the $t$-$J$ model exhibits a gap-like behavior in its spin excitation spectrum as the result of the spinon condensation by applying the mean field approximation to the resonating valence bond basis. Onoda et al. [15] also derived the occurrence of the gap-like behavior in the temperature dependence of the resistivity within the same model using the slave-boson mean field approximation. These results, however, crucially depend on the nature of the approximation, the validity of which is not so well established.

Miyake and Narikiyo, on the other hand, suggested that the pseudogap rather originates from the enhancement of the antiferromagnetic spin fluctuation paying attention on the nesting effect based on the phenomenological itinerant-localized duality theory. [16] In our previous study, we also proposed a simple explanation based on the well-known property of the Heisenberg magnets. [17] We were successful in demonstrating the presence of the gap-like behavior in the temperature dependence of the NMR relaxation rate by numerically diagonalizing the finite cluster $t$-$J$ model for sufficiently large $J$. Our result shows it is interpreted as a crossover from the paramagnetic to the magnetically well correlated state induced by the rapid development of the antiferromagnetic spin correlation.

Lately lots of interest have been paid on the role of the superconducting fluctuations. For instance, Koikegami and Yamada [18], based on the $d$-$p$ model, indicated that superconducting fluctuations will survive at higher temperature than the transition temperature $T_c$ based on the fluctuation exchange approximation. This means that the gap-like behavior is regarded as a precursor of the gap formation of the strong coupling superconductivity as was predicted by the self-consistent $t$-matrix approximation by Yanase and Yamada [19]. With the use of the $t$-matrix approximation Kobayashi et al. [20] showed that the gap-like behavior is induced by the appearance of the superconducting fluctuations while the antiferromagnetic spin fluctuation is suppressed on the contrary. Onoda and Imada [21] claim that the competition between the superconducting fluctuations and antiferromagnetic spin fluctuations is necessary for deriving the gap behavior.

In spite of lots of theoretical proposals, the final answer is still far from being established. Most theories, however, seem to have an agreement on the important role of anisotropic spin correlation on the pseudogap formation. In the present paper, in order to confirm the validity of our mechanism we extend our numerical investigation and derive the temperature dependence of various physical properties, including $1/T_1 T$, in a wider parameter region that covers realistic situations for high-$T_c$ cuprates. The effect of the next-nearest neighbor hole hopping with a coupling $t'$ is also studied. The effect of...
the antiferromagnetic spin correlation on the pseudogap formation is then clarified without resorting to any approximation. If we take into account the lack of reliable approximation scheme in treating the strongly correlated systems, the numerical analysis of the model has its own significance in order to check the validity of various explanations.

II. MECHANISM OF PSEUDOGAP

According to our proposal, the local spin excitations are suppressed around a characteristic temperature, of the same magnitude as the antiferromagnetic coupling J, associated with the growth of the short-range antiferromagnetic correlation as we decrease the temperature. As a result various magnetic quantities will show their particular T-dependence in reflecting the reduced local excitations. In fact the high temperature series expansion for the square-lattice antiferromagnetic Heisenberg model shows a broad peak in the temperature dependence of the NMR relaxation rate 1/T1T around T ∼ J. [22] Since the hole motion acts to destroy the short-range antiferromagnetic order, the hole doping will reduce the crossover temperature in agreement with the observed hole doping dependence of the pseudogap temperature. Thus the pseudogap behavior is simply realized as the manifestation of the growth of the short-range antiferromagnetic correlation. The above picture has been supported by our numerical exact diagonalization study on the two-dimensional t-J model with 10 lattice sites. [17] We found that for a small hole concentration (δ = 0.1) the T dependence of 1/T1T does show a broad peak around the temperature where the nearest neighbor antiferromagnetic spin correlation shows rapid growing, for a sufficiently large antiferromagnetic coupling (J/t = 0.6). In the next section we perform the same calculation to investigate how our previous results are modified by using realistic parameters for the cuprate superconductors and also to examine the presence of possible gap-like behaviors for various magnetic properties.

III. NUMERICAL ANALYSIS OF T-J MODEL

Let us first consider the standard square-lattice t-J model Hamiltonian defined by

\begin{equation}
H = -t \sum_{<i,j>,\sigma} (c_i^\dagger \sigma c_{j\sigma} + c_{i\sigma}^\dagger c_{j\sigma}) + J \sum_{<i,j>} (S_i \cdot S_j - \frac{1}{4} n_i n_j),
\end{equation}

where t is the nearest neighbor electron hopping integral and J is the antiferromagnetic Heisenberg exchange constant between spins on adjacent lattice sites. Throughout the paper, all the energies are measured in units of t. With the use of the calculated eigenvalues and eigenvectors, we evaluated the temperature dependence of the imaginary part of the dynamical spin susceptibility of conduction electrons Imχ(q, ω) and the static spin correlation function \((S_i^z S_j^z)\). In actual numerical estimations of Imχ(q, ω) the δ-function is approximated by the Lorentzian distribution with some small width. Since we need all the eigenvalues and eigenvectors, the cluster size of the model was limited by the available disk space of the computer. We show in the following the results for the \(\sqrt{10} \times \sqrt{10}\) cluster under the periodic boundary condition.

A. NMR relaxation rate

In order to obtain the temperature dependence of the NMR relaxation rate, we evaluate 1/T1T by the following formula:

\begin{equation}
\frac{1}{T_1T} = \lim_{\omega \to 0} \frac{1}{\omega} \sum_q \text{Im} \chi(q, \omega).
\end{equation}

The effect of the q-dependence of the hyperfine form factor is neglected, for simplicity. To clarify the role of antiferromagnetic spin correlation, we have also evaluated the T dependence of the Q = (π, π) component of the spin correlation function,

\begin{equation}
S(Q) = \frac{1}{N} \sum_{i,j} (-1)^{(j-i)(\hat{x}+\hat{y})} \langle S_i^z S_j^z \rangle.
\end{equation}

In our previous study [17] the following nearest-neighbor spin correlation function was used instead for the purpose,

\begin{equation}
C_1 = \frac{1}{N} \sum_{i} \frac{1}{\pi} \sum_{\rho=\pm \hat{x}, \pm \hat{y}} \langle S_i^z S_{i+\rho}^z \rangle.
\end{equation}

We are using S(Q) here to emphasize the continuity that there exists the antiferromagnetic order in the limit of no doping. Note that the terms, magnetically singlet and antiferromagnetic, are used in nearly the same meaning. We are only concerned with the short range magnetic correlation of the system of finite clusters. According to the above definitions there is no reason to make definite distinction between S(Q) and C1. Actually both of S(Q) and C1 show almost no essential difference in their temperature dependence [23].

We show in Fig.1 results of T dependence of (a) 1/T1T and (b) S(Q) for the undoped case (δ = 0) for J=0.6, 0.5, and 0.4. Only for J=0.4, the temperature dependence of S(Q) has already been reported by Tohyama et al. [24] For cuprate superconductors the value J = 0.4 is estimated experimentally. All the calculated T dependence of 1/T1T exhibits a broad peak around T ∼ J where S(Q) shows the significant increase. The above behavior of 1/T1T for δ = 0 is, for instance, consistent with the high temperature series expansion for the Heisenberg model. [22] The quantum Monte Carlo simulation
study by Sandvik and Scalapino \[26\], however, showed no peak behavior in the $1/T_1T$ curve. No peak is also observed by the NMR measurement for the undoped La$_2$CuO$_4$. \[27\] The reason is the temperature in these studies is not so high enough to cover the crossover region of the order of $J$.

The same results are shown in Figs.2(a) and 2(b) for the one-hole case ($\delta = 0.1$). Peaks in the $T$ dependence of $1/T_1T$ for $J=0.5$ and $0.6$ are shifted to lower temperature where the rapid growth of $S(Q)$ is observed, in agreement with our mechanism. Although no peak appears for the case $J=0.4$, a slight hump still exists around the same temperature. Such a behavior will be detected as a deviation from the Curie-Weiss like temperature dependence of $1/T_1T$, rather than a peak as was actually observed for La$_{2-x}$Sr$_x$CuO$_4$. \[28\] The same calculation for the hole-doped $4\times4$ $t$-$J$ cluster \[29\] does not show any gap-like anomalies. This is probably because the temperature range didn’t cover the region where the pseudogap is expected to appear. The present results indicate that the gap-like behavior appears at much higher temperature than the calculated lowest excitation gap $\Delta$ of the cluster. For example, the broad peak appears around $T \sim 0.4$, while the estimated $\Delta$ is 0.146, for $J = 0.6$ and $\delta = 0.1$. \[3\] Therefore it excludes the possibility that the pseudogap is caused by the discreteness of energy levels of finite systems. The same results for the two-hole system ($\delta = 0.2$) are shown in Figs.3(a) and 3(b). Although no gap-like behavior is observed, they are also consistent with our view because of the absence of any significant enhancement of the antiferromagnetic spin correlation.

### B. Neutron scattering intensity

The measurement of the neutron scattering intensity of YBa$_2$Cu$_3$O$_{6.6}$ by Sternlieb et al. \[3\] indicated the presence of the pseudogap behavior in the temperature dependence of the $q$-integrated dynamical susceptibility

$$\text{Im}\chi(\omega) \equiv \int dq \text{Im}\chi(q, \omega). \quad (5)$$

We show, in Figs.4, 5 and 6, the temperature dependence of (a) $\text{Im}\chi(Q, \omega)$ and (b) $\text{Im}\chi(\omega)$ for a small $\omega$ ($=0.01t$) for $\delta = 0$, 0.1 and 0.2. Figs.4(a), 5(a) and 6(a) show the monotonic increase of $\text{Im}\chi(Q, \omega)$ for all the values of $J$ with decreasing $T$, independent of the doping $\delta$. On the other hand, $q$-integrated local susceptibility in Figs.4(b), 5(b) and 6(b) reveals a gap-like behavior around the same crossover temperature as that of $1/T_1T$ for $\delta \leq 0.1$. These results are in good agreement with the
above neutron scattering experiment. As for the former, the low-frequency limit of \( \text{Im}\chi(Q,\omega) \) is given by

\[
\text{Im}\chi(Q,\omega) \to \frac{\chi(Q,0)}{\Gamma_Q} \propto \chi^2(Q,0), \quad (\omega \to 0),
\]

where \( \Gamma_Q \) is the Lorentzian distribution width of the frequency spectrum. The monotonic increase of \( \text{Im}\chi(Q,\omega) \) with decreasing the temperature is therefore consistent with the absence of the pseudogap behavior in the \( T \)-dependence of \( 1/T_{2G}^2 \), i.e. \( T_{2G}^2 \propto \chi^{-1}(Q,0) \), by the NMR measurement. [30]

\[J=0.4\] \[J=0.5\] \[J=0.6\]

**FIG. 4.** Temperature dependence of (a) \( \text{Im}\chi(Q,\omega) \) and (b) \( \text{Im}\chi(\omega) \) for \( \delta = 0.0 \).

\[J=0.4\] \[J=0.5\] \[J=0.6\]

**FIG. 5.** Temperature dependence of (a) \( \text{Im}\chi(Q,\omega) \) and (b) \( \text{Im}\chi(\omega) \) for \( \delta = 0.1 \).

**C. Magnetic susceptibility**

The pseudogap behavior has also been observed in the temperature dependence of the static magnetic susceptibility \( \chi \). [4] The calculated results of \( \chi \) for \( \delta = 0, 0.1 \) and \( 0.2 \) are shown in Figs. [6], [7] and [8], respectively. In all the figures we can see the crossover behaviors around the same temperature as those of \( 1/T_{1T} \) and \( \text{Im}\chi(\omega) \) in the underdoped case (\( \delta \leq 0.1 \)), even for the realistic value \( J = 0.4 \). As shown in Fig. 9, peaks appear at lower temperature for \( \delta = 0.2 \). However they result from the different origin. If there are two holes in a finite cluster model, its ground state always becomes singlet and the total energy has some small excitation gap. Therefore the susceptibility shows a peak around the temperature corresponding to the lowest excitation gap of the system that comes from these finite size effects. The behavior has nothing to do with our mechanism. (The same calculation of \( \chi \) for \( J = 0.4 \) by Tohyama et al. [24] yielded a peak even for \( \delta = 0.3 \).)

\[J=0.4\] \[J=0.5\] \[J=0.6\]

**FIG. 6.** Temperature dependence of (a) \( \text{Im}\chi(Q,\omega) \) and (b) \( \text{Im}\chi(\omega) \) for \( \delta = 0.2 \).

**FIG. 7.** Temperature dependence of \( \chi \) for \( \delta = 0.0 \).
including the next-nearest neighbor hole hopping, another possible reason, let us discuss below the effect of compared with that of \( J \) for mon crossover temperature. The characteristic temperature due to the finite size effect. As \( J \) increases, the crossover temperature is lowered by the effect. The narrowing of the peak width takes place at the same time. As the result the slight anomaly in Fig. 2(a) for \( J = 0.4 \), shifted to lower temperature, now becomes evident if we assume \( t' = -0.3 \) as shown Fig. 9(b). Therefore it is possible to predict the crossover temperature around a realistic temperature range \( \sim J/4 \) by including the effect of the next-nearest neighbor hopping.

As we increases the value of \( t' \), there appears, however, another difficulty. Because the peak temperature is lowered and becomes comparable with the order of the lowest excitation gap of the cluster, it is very difficult to distinguish the peak behavior from the one that comes from the discreteness of the energy levels of the system. Further calculations on larger size clusters will be necessary to clarify the situation and to obtain reliable conclusions.

To conclude, all the results presented above support our view that the pseudogap behavior observed for various magnetic properties are induced by the growth of the antiferromagnetic spin correlation.

IV. NEXT-NEAREST NEIGHBOR HOPPING \( t' \)

In the previous section we showed that the antiferromagnetic spin correlation produces a gap-like behavior in the \( T \)-dependence of \( 1/T_1 T \), \( \text{Im} \chi(\omega) \) and \( \chi \) around a common crossover temperature. The characteristic temperature for \( \delta = 0.1 \) is roughly estimated as \( \sim J/2 \), as shown in Figs. 2(a), 3(b) and 3. It is, however, much higher than observed ones for real cuprates. This is partly because the effect of the hole hopping \( t \) is overestimated compared with that of \( J \) due to the finite size effect. As another possible reason, let us discuss below the effect of including the next-nearest neighbor hole hopping,

\[
H' = -t' \sum_{<i,j>_{\mathbf{1},\sigma}} (c_{i,s}^\dagger c_{j,s} + c_{i,s} c_{j,s}), \tag{6}
\]

into the \( t-J \) Hamiltonian (4), where \( \sum_{<i,j>_{\mathbf{1}}} \) means the summation over all the next-nearest sites. The term is originally introduced to explain the dynamics of the correlated motion of spin and hole-hopping degrees of freedom in some cuprates. The value \( t' \sim -0.3 \) is estimated to be suitable for \( \text{Sr}_2\text{CuO}_2\text{Cl}_2 \) by the exact diagonalization study. [11] To clarify the effect of \( t' \) on the pseudogap formation, we show the \( T \)-dependence of \( 1/T_1 T \) for various values of \( t' \) in Figs. 10(a) for \( J = 0.6 \) and 10(b) for \( J = 0.4 \) (\( \delta = 0.1 \)). From Fig. 10(a) we can see that the peak temperatures are lowered by the effect. The narrowing of the peak width takes place at the same time.

As the result the slight anomaly in Fig. 3(a) for \( J = 0.4 \), shifted to lower temperature, now becomes evident if we assume \( t' = -0.3 \) as shown Fig. 10(b). Therefore it is possible to predict the crossover temperature around a realistic temperature range \( \sim J/4 \) by including the effect of the next-nearest neighbor hopping.

V. DISCUSSION

The results of the present study clearly showed the presence of gap-like behaviors in the temperature dependence of various magnetic properties around a single crossover temperature due to the development of antiferromagnetic spin correlation. Experimentally detailed gap-like behaviors differ slightly with each other depending on materials. In the case of \( \text{La}_{2-\delta}\text{Sr}_\delta\text{CuO}_4 \), for instance, the crossover temperature of \( \chi (T_\chi) \) is slightly higher than that of \( 1/T_1 T (T_{\text{NMR}}) \). The recent NMR
Knight shift measurement for Bi$_2$Sr$_2$CaCu$_2$O$_{8+x}$ also indicates the presence of two different crossover temperatures. Stimulated by these discoveries, it is argued that the higher crossover temperature (the large pseudogap) will be associated with the growth of the antiferromagnetic spin correlation. On the other hand, the lower one (the small pseudogap) will be due to the singlet (or superconducting pair) formation, i.e. $T_{c}^{*}$ will be assigned to the former, while $T_{NMR}$ the latter. From our approach based on the finite cluster model, it will be difficult to predict the presence of double pseudogap phenomena.

Our model Hamiltonian employed here is quite simplified one. In order to deal with actual systems we have to include various additional interactions specific to each system. In the present paper we are particularly interested in properties that are common to all the cuprate superconductors. Therefore in order to compare with experiments, we have to be aware that the observed properties are intrinsic to all of them or not. It will also be important to confirm whether the presence of two crossover temperatures is common to all the cuprates systems.

As was pointed out in the Introduction, spin-gap phenomena is characteristic to the underdoped cuprates where the superconducting transition temperature $T_c$ decreases proportional to the hole doping concentration. On the other hand the pseudogap temperature increases. Because of this opposite doping dependence, it seems to be difficult to associate the pseudo-gap phenomena with the superconducting pairing fluctuations. The antiferromagnetic correlation, therefore, plays the dominant role on reducing the low energy excitations of the system as far as the magnetic freedoms are concerned. Only around the critical temperature $T_c$, they will be further modified by the presence of the superconducting transition. These effect are limited to the very low energy regions. The present mechanism is based on the well-known properties of low-dimensional Heisenberg magnets. The overall doping dependence of the crossover temperature is also in agreements with experiments. It decreases with increasing hole doping, whereas the transition temperature $T_c$ increases towards the optimum condition. Because of these reasons we suppose that our mechanism is one of most probable candidates for the phenomena.

Because the model treated here is a single-hole system ($\delta = 0.1$ for the 10 site cluster), its ground state is doublet. Even in this doublet case we can derive the gap-like behavior in the temperature dependence of $1/T_1 T$. If the same calculation is done on a two-hole system of a larger cluster with the same doping $\delta$, a slightly different properties will be obtained because its ground state now becomes singlet. We expect that the gap-like behavior will then be more evident. The difference becomes less conspicuous, if we increase the size of clusters, for it results from the size effect. Though it is very difficult because of various limitations concerning the available resources of computer systems, it will be interesting to extend our present numerical diagonalization study to the two-hole system with larger cluster size to clarified the situation.

VI. SUMMARY

We have shown that the pseudogap behaviors observed in the high-temperature superconductors originate from the growth of the antiferromagnetic spin correlation based on the exact diagonalization study on the finite cluster $t$-$J$ model in a realistic parameter region for the real cuprates. We also found that the pseudogap temperature is lowered by including the next-nearest neighbor hopping effect.

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