Pair breaking by impurities in the two-dimensional $t-J$ model

J. Riera$^1$, S. Koval$^1$, D. Poilblanc$^2$ and F. Pantigny$^2$

$^1$Instituto de Física Rosario, Consejo Nacional de Investigaciones Científicas y Técnicas and Departamento de Física, Universidad Nacional de Rosario, Blvd. 27 de Febrero 210bis, 2000-Rosario, Argentina

$^2$Laboratoire de Physique Quantique, Université Paul Sabatier, 31062 Toulouse, France

(October 95)

Pair breaking mechanisms by impurities are investigated in the two-dimensional $t-J$ model by exact diagonalization techniques. Analysis of binding energies, pairing correlations, dynamical spin and pair susceptibilities shows that non-magnetic impurities are more effective in suppressing pairing than magnetic ones in agreement with experimental studies of Zn- and Ni-substituted High-Tc superconductors.

PACS Numbers: 74.20.Mn, 74.62.Dh, 71.55.-i, 75.40.Mg

Many recent experiments of hole-doped cuprate superconductors showing a large anisotropy consistent with gap nodes on the Fermi surface have been interpreted in terms of an unconventional $d$-wave pairing state. This possibility of $d$-wave superconductivity has in turn renewed the interest in studying the effects of impurities on the superconducting state. It is well-known that magnetic impurities are strong pair breakers for singlet superconductors while nonmagnetic impurities have a pair breaking effect only for higher orbital momentum states such as a $d$-wave pairing state. In fact, in several recent studies it was suggested the possibility of using the response of these materials to different types of impurities to distinguish between an unconventional $d$-wave pairing and alternative scenarios such as a more conventional anisotropic $s$-wave pairing. Experimental studies have shown that divalent Zn and Ni ions go to the planar Cu(2) sites with the Zn impurity having a $S = 0$ configuration and the Ni impurity a $S = 1$ configuration. The important experimental fact is that the nominally nonmagnetic impurity Zn is more effective than the magnetic impurity Ni in destroying superconductivity. In conventional superconductors, with the usual $s$-wave superconductivity, a magnetic impurity is most destructive due to the magnetic pair-breaking effect. Although the observed behavior could be characteristic of a $d$-wave pairing, it has been also suggested that a Zn impurity may not simply behave as a vacancy because it could induce a magnetic moment in the CuO$_2$ planes due to the strong correlations present. However, other experimental studies indicate that the estimated impurity moment-carrier exchange in Zn doped YBa$_2$Cu$_3$O$_7$ is too small to account for the Tc suppression by a magnetic pair-breaking mechanism.

From a theoretical point of view it is of extreme importance to explain at least qualitatively the effects of both magnetic and non-magnetic impurities on the superconducting properties of the cuprates as seen experimentally. These effects have not been clearly explained by existing analytical macroscopical theories. An alternative approach is to analyze them in the context of microscopic models using numerical techniques. This program has been recently initiated by Poilblanc, Scalapino and Hanke by studying the two-dimensional (2D) $t-J$ model in the presence of a single impurity. The two-dimensional $t-J$ model has been extensively studied in the context of high-Tc superconductivity since it contains the essential low-energy physics of the CuO$_2$ planes present in the cuprates. The $S = 0$ Zn impurity was modelled by an inert site, while the $S = 1$ Ni impurity was approximated by a static spin-1/2 (see Hamiltonian below). The main results obtained by these authors is that a hole binds to the impurity with different spatial symmetries.

The purpose of this Letter is to study the effect of magnetic and nonmagnetic impurities on several quantities related to pairing of holes in the 2D $t-J$ model near half-filling. This study at zero temperature will be performed using exact diagonalization techniques on finite clusters. Our starting point is the fact that numerical studies of the pure $t-J$ model on the square lattice have shown that a two-hole bound state is formed for $J > J_c$ $\sim 0.3$ $t$ in the bulk limit and with $d_{x^2-y^2}$ internal symmetry. These studies have also given strong unbiased indications of $d_{x^2-y^2}$ superconductivity at quarter filling ($n = 0.5$) in the vicinity of phase separation, although they are less conclusive near half-filling. Our objective is to elucidate which is the pair-breaking mechanism specially for the case of non-magnetic impurities.

The Hamiltonian of the $t-J$ model in the presence of impurities is:

$$H = -t \sum_{\langle i,j \rangle, i,j \neq i_0 \sigma} (\tilde{c}^\dagger_{i\sigma} \tilde{c}_{j\sigma} + \tilde{c}^\dagger_{j\sigma} \tilde{c}_{i\sigma}) + J \sum_{\langle i,j \rangle, i,j \neq i_0} \langle S_i \cdot S_j - \frac{1}{4} n_i n_j \rangle + J' \sum_{\langle \mathbf{i}, \mathbf{j} \rangle} \langle (S_{i_0} \cdot S_{j} - \frac{1}{4} n_{i_0} n_j) \rangle$$

(1)

where $\tilde{c}^\dagger_{i\sigma}$ is an electron creation operator at site $i$ with
spin $\sigma$ with the constraint of no double occupancy, $n_i = n_{i,\uparrow} + n_{i,\downarrow}$ is the electron number operator. The impurities are located at the sites $\{i_0\}$. Here $t$ is the hopping parameter, which we choose as the scale of energies, $J$ is the AF exchange interaction and $J'$ is the magnetic coupling between the impurity spin and the electrons in nearest neighbor (NN) sites. The special case $J' = 0$ corresponds to an inert site describing a Zn impurity. \[19\]

We adopt periodic boundary conditions throughout.

As a preliminary study, it is interesting to investigate the effect of the magnitude of the impurity spin $S_{i_0}$. Following previous work, we define the hole-impurity binding energy \[14\]

$$E_{B,1} = e(1h, 1i) - e(0h, 1i) - e(1h, 0i), \quad (2)$$

where $e(Nh, Mi) = E(Nh, Mi) - E(0h, 0i)$. $E(Nh, Mi)$ is the ground state energy of the system with $N$ holes and $M$ impurities. For the case of a spin 1 impurity embedded in small clusters, bound states are found (i.e. $E_{B,1} < 0$) in all symmetry channels provided $|J'/J| > \text{ does not exceed small critical values. This is qualitatively similar to the spin 1/2 impurity \[17\] as clearly seen in Fig. 1 showing a comparison of $E_{B,1}$ calculated on a 20-site cluster in the d-wave channel for spin 1/2 and spin 1 impurities. Note that the coupling $J'$ is more effective to destroy the bound-state for larger impurity spin. This can be simply understood from a simple argument relating the binding energy with the number of broken bonds in the AF background \[18\] (this also applies to pairing of holes \[17\]). Indeed, while the average magnetic energy per bond for the bonds not connected to the impurity \[22\] depends weakly on $J'$, the magnetic energy for the bonds connected to the impurity essentially scales like $JS_{i_0}$ so that the effective short range attractive potential for holes weakens for increasing $J'$ or $S_{i_0}$. In the following, we shall restrict ourself to the lowest energy sector. Moreover, since from Fig. 1 the results for $S = 1/2$ and $S = 1$ are very similar for the region of interest $J' > 0$, we shall consider only the case of spin-1/2 impurities.

In order to determine the binding of holes in pairs in the vicinity of impurities let us start examining the following combinations of ground state energies:

$$E_{B,2} = e(2h, 0i) - 2e(1h, 0i)$$
$$E'_{B,2} = e(2h, 0i) + e(0h, 1i) - e(1h, 1i) - e(1h, 0i) \quad (3)$$
$$E_{B,3} = e(2h, 1i) - e(0h, 1i) - e(2h, 0i)$$

The physical meaning of these quantities is quite obvious. $E_{B,2}$ is the usual binding energy for the pure system. \[18\] $E_{B,3}$ corresponds to compare the state where the two holes are trapped to the impurity with the state where the holes move away from the impurity. $E_{B,2}$ is the energy difference between the state with a hole pair away from the impurity and the state with one hole bound to the impurity. Thus, for the holes forming a bound pair not trapped by the impurity the following inequalities should hold: $E_{B,2} < 0$, $E_{B,2} < 0$, and $E_{B,3} > 0$.

The results for $E_{B,2}$, $E'_{B,2}$ and $E_{B,3}$ obtained for the $4 \times 4$ lattice and for the case of a nonmagnetic impurity $J' = 0$ are shown in Fig. 2(a). Very similar results are also obtained for the 18 sites tilted cluster. It can be seen that the conditions for the existence of a hole pair not bound to the impurity are satisfied in the range $0.2 \leq J/t \leq 0.5$. For a better understanding of the physical situation that emerges from the study of $E_{B,2}$, $E'_{B,2}$ and $E_{B,3}$, it is important to examine also the hole-impurity binding energy $E_{B,1}$ as well as

$$E_{B,4} = e(2h, 2i) - 2e(1h, 1i). \quad (4)$$

Notice that $E_{B,4}$ as $E'_{B,2}$ reduces to $E_{B,2}$ in the absence of impurities. $E_{B,4} > 0$ and $E_{B,1} < 0$ means pair-breaking effect, each hole been trapped by an impurity. Both quantities have been added to Fig. 2(a). $E_{B,1}$ is negative for $J \geq 0.2$ implying that there is a hole-impurity bound state. It is interesting to note that taking into account the results for $E_{B,2}$, $E'_{B,2}$ and $E_{B,3}$ discussed above this hole-impurity bound state only appears for $J \geq 0.5$ when a second hole is added. However, as we shall show below, the tendency to form a hole-impurity bound state is the main source of suppression of pairing. Also interesting is the result of $E_{B,4}$ indicating that two impurities split the hole pair for $J \leq 0.3$.

The most important results for their connection to the experimental results observed in the cuprates are shown in Fig. 2(b). In this figure, we show $E'_{B,2}$ and $E_{B,3}$ obtained for the $4 \times 4$ cluster for several values of $J'/J$. We also add $E_{B,2}$ for comparison. It can be seen that $E'_{B,2}$ is suppressed as $J'/J$ decreases from 1, corresponding to the pure case except for the condition of exclusion of holes at the impurity sites, to 0 which corresponds to the nonmagnetic case. This behavior of the binding energy in the presence of impurities is consistent with the experimentally observed one. $E_{B,3}$ also decreases in such a way that the interval of $J/t$ where the bound state of holes exists and is not trapped to the impurity narrows down as $J'/J$ decreases.

Additional evidence of the strongest pair-breaker effect of nonmagnetic impurities in the $t-J$ model comes from the study of the quasiparticle weight of pairs and pairing correlations with $d_{x^2-y^2}$ symmetry. The quasiparticle weight of pairs $Z_{2h}$ is defined as

$$Z_{2h} = \frac{|\langle \Psi_0^{2h} | \Delta | \Psi_0^{0h} \rangle|}{|\langle \Psi_0^{0h} | \Delta | \Psi_0^{0h} \rangle|^{1/2}} \quad (5)$$

where $\Delta = \frac{1}{N} \sum_{i \neq i_0} \Delta_i$. The pairing operator at site $i \neq i_0$ is

$$\Delta_i = \sum_{\mu} g_{\mu} c_{i \mu} c_{i+4 \mu}, \quad (6)$$

where the sum extends over the NN of site $i$ and $g_{\mu}$ are the form factors that determine the pairing symmetry. Note that when $i$ corresponds to one of the four NN sites of $i_0$ the sum over $\mu$ is restricted to only $i + \mu \neq i_0$. 

2
The rest of the notation is standard.\textsuperscript{[24,25]} The results obtained in the $4 \times 4$ lattice for $d_{x^2-y^2}$ symmetry as a function of $J/t$ and $J'/J = 0.5$ and 1.0. are shown in Fig. 3(a). It can be seen that the largest reduction of $Z_{2K}$ corresponds to the $J'/J = 0$ case. Notice from the result for $J'/J = 0.5$ that this reduction is not linear in $J'/J$. The pairing correlations are defined as

$$C_{\Delta}(r) = \frac{1}{N} \sum_{i} <\Delta_{i}\Delta_{i+r}>,$$

(7)

where, in the presence of an impurity, the sum is restricted to $i$ and $i + r \neq i_{0}$. It is well-known that close to half-filling the pairing correlations at large distances are quite suppressed.\textsuperscript{[21]} For this reason, in Fig. 3(b) we show $C_{\Delta}(r)$ for $r \geq 1$ on the $4 \times 4$ lattice at quarter filling with $J/t = 3$ where they are maximum in the pure system.\textsuperscript{[26]} The tail in $C_{\Delta}(r)$ is mostly suppressed for $J'/J = 0$. The results for $J'/J = 0.5$ indicate again a nonlinear behavior of pairing as a function of $J'/J$.

In order to understand the origin of the pair breaking mechanism let us examine the quantity $W_{p}$ defined as the probability of finding the pair of holes in NN sites. In Fig. 4(a) we show $W_{p}$ as a function of $J/t$ for the pure system (full circles) and in the presence of a single impurity with $J'/J = 0$ (full squares). It is also shown for various $J'/J$ the quantity $W_{p}$ computed with the constraint that the holes can not occupy the NN sites of the impurity. For the case of $J'/J = 0$, by comparing the total $W_{p}$ with the restricted one it can be concluded that there is a large contribution to $W_{p}$ coming from the hole pair bound to the impurity. This contribution becomes smaller as $J'/J$ is increased from zero.

To complete the characterization of the effects of impurities on the ground state of the $t-J$ model we briefly discuss their effect on the magnetic order\textsuperscript{[21]} and on the dynamical magnetic fluctuations. The dynamical spin structure factor at the AF wavevector ($\pi, \pi$) shown in Fig. 4(b) exhibits, in comparison to the pure case\textsuperscript{[18,22]}, new structures at low energies. Spectral weight is transferred from the peak at $\omega \sim 0.8J$, characteristic of the pure two-hole doped $4 \times 4$ system, to lower energies of order $J'$. This new resonance can be interpreted as the singlet-triplet excitation energy of the singlet impurity-hole bound-state. These results can be related to recent neutron scattering experiments which have reported that the magnetic pseudogap at the AF wavevector disappears with Zn doping.\textsuperscript{[22]}

In summary, we have studied using exact diagonalization of the 2D $t-J$ model in the presence of magnetic ($S = 1/2$) and nonmagnetic ($S = 0$) impurities. We have shown through an analysis of binding energies, pairing correlations and dynamical pair susceptibility that the nonmagnetic impurity has a stronger pair breaker effect than the magnetic impurity. These results are in agreement with experimental findings in Zn ($S = 0$) and Ni ($S = 1$) doped cuprates. This agreement reaffirms the ability of microscopic strongly correlated electronic models to describe the physics of CuO$_2$ planes in the cuprates. We have also given some indications of the possible essential role of the binding of holes to impurities in suppressing superconductivity in these systems.

D.P. thanks D.J. Scalapino and W. Hanke for many insightful discussions. J.R. thanks E. Dagotto for useful comments. Part of the numerical calculations were done at IDRIS (France).
J. Scalapino, and E. Y. Loh, Phys. Rev. Lett. 62, 2192 (1989).

[25] D. Poilblanc, J. Riera and E. Dagotto, Phys. Rev. B 49, 12318 (1994); for details see also D. Poilblanc, Phys. Rev. B 48, 3368 (1993).

[26] Y. Hasegawa and D. Poilblanc, Phys. Rev. B 41, 9555 (1990).

[27] K. Kakurai et al., Phys. Rev. B 48, 3485 (1993).

FIG. 1. $E_{B,1}$ vs. $J'/J$ for d-wave orbital symmetry, for a spin 1/2 impurity (solid squares) and a spin 1 impurity (open squares).

FIG. 2. a) $E_{B,2}$ (open squares), $E'_{B,2}$ (open diamonds), $E_{B,3}$ (solid circles), $E_{B,1}$ (solid squares) and $E_{B,4}$ (solid triangles), defined in the text, as a function of $J/t$ and for $J'=0$. b) $E_{B,3}$ (open symbols) and $E'_{B,2}$ (solid symbols) vs. $J/t$. The results correspond to $J'=0$ (squares), $J'=J/3$ (circles), $J'=2J/3$ (triangles) and $J'=J$ (diamonds). We also show for comparison the result of $E_{B,2}$ (crosses).

FIG. 3. a) The quasiparticle spectral weight $Z_{2h}$ as a function of $J/t$ for different values of $J'/J$. b) The pairing correlation $C_\Delta(r)$ at quarter filling for different values of $J'/J$. The results for the model with no impurities are shown with solid circles.

FIG. 4. a) The probability $W_p$ vs. $J/t$. The results for the pure system are shown with solid circles while with solid squares we show the results with one impurity in the system and $J'=0$. The results for $W_p$ with the exclusion of the NN sites of the impurity are shown for various values of $J'/J$ with open symbols as indicated on the plot. b) Dynamical spin structure factor (arbitrary units) in the two hole doped system for $J=1.0$, and $J'=0$ (solid line), 0.25 (dashed line), 0.5 (dotted line) and 1.0 (thick solid line).
$N=20$

$J=0.5$

$E_{B,1}$ vs $J'/J$ for $S=1/2$ and $S=1$
Figure (a) shows the variation of $Z_{2h}$ with $J/t$ for different values of $J'/J$. Figure (b) illustrates the behavior of $C_v(r)$ for $r$ ranging from 1.0 to 3.0, with various $J'/J$ values distinguished by distinct symbols and styles.
