Inhomogeneity-Induced Enhancement of the Pairing Interaction in Cuprate Superconductors

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Scanning tunneling spectroscopy has recently discovered a positive correlation between the magnitude of the superconducting gap and positions of dopant oxygen atoms in Bi-based cuprates. We propose a microscopic mechanism that could be responsible for this effect. In particular, we demonstrate that the dopant-induced spatial variation of the atomic levels always enhances the superexchange interaction.

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Scanning tunneling microscopy (STM) and scanning tunneling spectroscopy (STS) have recently confirmed that nanoscale electronic inhomogeneity is an inherent feature of many groups of high-temperature superconductors (HTSC). Being capable of direct probing the local density of states (LDOS), these methods revealed strong spatial modulation of the energy gap in Bi-based compounds [1]. This modulation occurs on a very short length scale, of the order of the coherence length. It has been shown that the system consists of regions of relatively small gap ($\Delta \approx 25$–35 meV) with high and sharp coherence peaks and regions of larger gap ($\Delta \approx 50$–75 meV) with small and broad peaks [2]. Since its discovery, the inhomogeneity was commonly attributed to a disorder introduced by poorly screened electrostatic potential of the out-of-plane oxygen dopant atoms. A lot of theoretical works exploited this idea, predicting modification of the energy gap in the vicinity of a dopant atom [3]. Very recently, STM experiments have shown a strong correlation between position of the dopant atoms and all manifestations of the nanoscale electronic disorder [4,5]. Thus, these experiments proved the impurities to be the source of the inhomogeneity. On the other hand, they revealed a very important feature: there is a *positive* correlation between the position of a dopant atom and the magnitude of a gap [4]. The sign of this correlation function contradicts the previous theoretical predictions, based on a direct reduction of the gap by the dopant's electrostatic potential [3]. Therefore, a new mechanism that is capable of gap enhancing close to impurities, is needed to explain the correlations. It was shown by Nunner *et al.* [6], that an assumption of an enhancement of the pairing potential by the presence of a nearby dopant atom leads to the correct correlation between the height of the coherence peaks and the magnitude of the gap as well as between the position of the dopant atom and the magnitude of the gap. The spectral properties could also be explained in a different way, assuming that the observed peaks at the edges of the gap arise from resonant bound states rather than they are the coherence peaks [7].

From the theoretical point of view, various parameters of the microscopic models of HTSC can be modified by the presence of the dopant atoms. These atoms are charged impurities and since cuprates are close to the insulating state, the number of carriers is too low to effectively screen their electrostatic potential. As a result, atomic levels in the CuO_2 plane are shifted in the vicinity of the dopants. The presence of the dopants also induces a local distortion of the lattice [8]. This, in turn, may modify the hopping matrix elements as well as the electron-phonon coupling. It has recently been shown that inhomogeneity itself can increase the superconducting transition temperature [9,10].

Here, we show that in strongly correlated systems, the position-dependent shift of the atomic levels alone is sufficient to enhance the pairing interaction, thereby leading to the correct sign of the dopant-gap correlation function. We assume that HTSC can be described by the t-J model, with the exchange interaction as the main pairing mechanism [11]. Although its basic version may be insufficient to describe all the properties of HTSC, it is the simplest model that captures the physics of strong correlations [12]. The physical picture behind our analysis is based on the same arguments, which allow one to derive the t-J model from the Hubbard one [13]. A virtual hopping of an electron to a neighboring site (occupied by an electron with the opposite spin) gives rise to an effective spin exchange interaction with potential $J = 4t^2/U$, where t is the hopping integral and U is the Coulomb potential. Obviously, local modification of the hopping matrix elements leads either to an enhancement [14] or to a reduction of the exchange interaction. Recent density functional investigations indicate that the dopant oxygen atoms are responsible for displacements of atoms in the CuO_2 plane [8]. Since these displacements are perpendicular to the plane, such a distortion increases the interatomic distances. Therefore, one might expect a reduction of the hopping integrals and a simultaneous decrease of J. However, the effective exchange interaction should depend also on the atomic levels at sites, which are involved in the virtual hopping. It comes from the fact, that the energy of the virtual doubly occupied state depends on these levels. The basic aim of this Letter is to show that the diagonal disorder always leads to an enhancement of the antiferromagnetic exchange interaction. As a result the superconducting gap increases in the vicinity of the dopant atoms.

We start with the two-dimensional one-band Hubbard model on a square lattice

$$H = -t\sum_{\langle i,j\rangle\sigma}(a^{\dagger}_{i\sigma}a_{j\sigma} + \text{H.c.}) + \sum_{i\sigma}(V_i - \mu)a^{\dagger}_{i\sigma}a_{i\sigma} + U\sum_i a^{\dagger}_{i\uparrow}a_{i\uparrow}a^{\dagger}_{i\downarrow}a_{i\downarrow}, \quad (1)$$

where $a^{\dagger}_{i\sigma}$ creates an electron with spin σ at site i, μ is the chemical potential and V_i is the atomic level at site i. The latter quantity accounts for the electrostatic potential of the dopant atoms.

In the following we argue that the enhancement of the pairing interaction is driven by a random distribution of V_i. However, there is a severe drawback of such assumption: strong enough dopant's potential should significantly affects the electron concentration in its neighborhood, resulting in a highly inhomogeneous charge distribution. On the other hand, the STS experiments show that the root mean square of the electron density in these systems are below 10% [4]. One can expect however, that strong Coulomb repulsion along with vicinity to half-filling, meaningfully reduce the dopant-induced inhomogeneity. Namely, the decrease of the local density of electrons close to the dopant atoms must result in its increase away from them. Such an increase is energetically highly unfavorable. In particular, at half-filling and for a strong Coulomb repulsion impurities cannot produce any charge inhomogeneities at all. In order to verify the effectiveness of this mechanism for a physically relevant doping, we have performed exact diagonalization study on a finite-size cluster. We have considered two impurities located $z = 1.5a$ above the cluster with a being the lattice constant. It is the distance between the dopant oxygen atom and the CuO_2 plane in Bi-based HTSC. These impurities act as a source of a screened electrostatic potential, that shifts the atomic levels $V_i = V_0\sum_m \exp(-R_{mi}/\lambda)/\tilde{R}_{mi}$, where the summation is carried out over all dopant atoms. Here, R_{mi} is a distance between site i and site m (above which the dopant atom is located) and $\tilde{R}_{mi} = \sqrt{R^2_{mi} + z^2}$. Such a form of V_i accounts for the fact that the potential is screened only by electrons moving in the CuO_2 plane. However, our final conclusions are independent of the form of V_i.

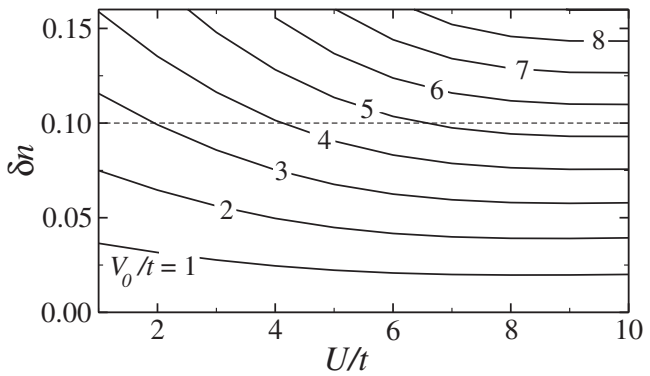

FIG. 1. Root mean square of the local electron density as a function of U for various values of V_0, as explicitly indicated in the figure. Results have been obtained for 10 electrons on a 12-site cluster with periodic boundary conditions. λ equals two lattice constants.

Figure 1 presents root mean square of the local electron concentration δn as a function of the Coulomb interaction U for various values of V_0. One can note a significant reduction of the charge inhomogeneity in a large-U regime in spite of the strong variation of the electrostatic potential V_i. In the uncorrelated case, inhomogeneity of the charge distribution depends on the ratio V_0/t. Contrary to this, in the strongly correlated nearly half-filled system inhomogeneity is determined predominantly by V_0/U. Therefore, small charge density variation observed in STS experiments does not require weak electrostatic potential of the dopant atoms.

We now turn to the main problem, i.e., the enhancement of the pairing interaction by the dopant atoms. For this sake, we make use of the Hubbard operators $X^{\alpha,\beta}_i = |\alpha\rangle_i\langle\beta|_i$ ($\alpha,\ \beta = 0, \uparrow, \downarrow, \uparrow\downarrow$) and carry out the canonical transformation $H \rightarrow \exp(-S)H\exp(S)$. The generator $S = -S^{\dagger}$ is chosen in such a way that the transformation eliminates the transfer of electrons between lower and upper Hubbard subbands. Straightforward calculations lead to the generator of the form

$$S = \sum_{\langle i,j\rangle,\sigma} s(\sigma)t\left[\frac{X^{\uparrow\downarrow,\bar{\sigma}}_i X^{0,\sigma}_j}{U + (V_i - V_j)} - \frac{X^{\sigma,0}_i X^{\bar{\sigma},\uparrow\downarrow}_j}{U - (V_i - V_j)}\right], \quad (2)$$

where $s(\uparrow) = 1$ and $s(\downarrow) = -1$.

Next, one can project out states with doubly occupied sites and obtain the effective Hamiltonian acting in the space spanned by states $|0\rangle_i$, $|\uparrow\rangle_i$ and $|\downarrow\rangle_i$. This is a generalized t-J Hamiltonian with a site dependent spin exchange interaction:

$$H_{t\text{-}J} = -t\sum_{\langle i,j\rangle\sigma}(\tilde{a}^{\dagger}_{i\sigma}\tilde{a}_{j\sigma} + \text{H.c.}) + \sum_{i\sigma}(V_i - \mu)\tilde{a}^{\dagger}_{i\sigma}\tilde{a}_{i\sigma} + \sum_{\langle i,j\rangle} J_{ij}\left(\vec{S}_i\cdot\vec{S}_j - \frac{1}{4}n_i n_j\right). \quad (3)$$

Here, $\tilde{a}^{\dagger}_{i\sigma}$ creates an electron at site i if this site previously had no electron. The interaction J_{ij} is given by

$$J_{ij} = \frac{4t^2}{U}(1 + \eta_{ij}), \quad (4)$$

where

$$\eta_{ij} = \frac{(V_i - V_j)^2}{U^2 - (V_i - V_j)^2} \geq 0. \quad (5)$$

In the homogeneous case ($V_i = \text{const}$), J_{ij} reduces itself to the standard form $J_0 = 4t^2/U$. Otherwise, it is always *larger* than J_0, independently of the distribution of the atomic levels and any particular form of V_i (provided $U > |V_i - V_j|$, what obviously holds true for HTSC). Moreover,

the strength of the coupling increases with the difference between the atomic levels at neighboring sites. Assuming J_{ij} to be the effective pairing interaction one comes to the conclusion that superconductivity can be enhanced by inhomogeneous distribution of the atomic levels. Since these levels directly affect the pairing interaction, spatial variation of the superconducting gap does not require strongly inhomogeneous charge distribution. The surprising enhancement can be easily understood from the analysis of virtual processes presented in Fig. 2. The resulting exchange interaction is proportional to the squared hopping matrix elements (t^2) and inversely proportional to the energy of the intermediate state with doubly occupied site. There are two second order processes that lead to the effective antiferromagnetic coupling of spins at sites i and j. The first of them [Fig. 2(a)] gives $J^a_{ij} = 2t^2/[U + (V_i - V_j)]$, whereas the second one [Fig. 2(b)] gives $J^b_{ij} = 2t^2/[U - (V_i - V_j)]$. Taking into account both these processes one obtains $J_{ij} = J^a_{ij} + J^b_{ij}$, what is the value given by Eq. (4).

Since the Coulomb repulsion is the dominating energy scale in cuprates, one can assume that $|V_i - V_j| \ll U$. Then, an approximate formula $\eta_{ij} \simeq (V_i - V_j)^2/U^2$ holds and one can see that η_{ij} decreases much faster than V_i when the distance from the dopant atom increases. As a result, a significant enhancement of the exchange interaction is expected only in the closest vicinity of the dopant. In Eq. (3) we have neglected the three-site term containing operators $X^{\sigma 0}_i X^{\bar{\sigma}\,\bar{\nu}}_j X^{0\nu}_k$. In the inhomogeneous case, in analogy to Eq. (4), the corresponding coupling constant can be written as $J_{ijk} = J_0(1 + \theta_{ijk})$, where $2\theta_{ijk} = (V_k - V_j)(U + V_j - V_k)^{-1} + (V_i - V_j)(U + V_j - V_i)^{-1}$. In contradistinction to η_{ij}, this quantity takes on both positive and negative values. However, when averaged over the whole system, η_{ij} and θ_{ijk} are equal.

It is known that an enhancement of the pairing potential close to impurities leads to the correct sign of the impurity-gap correlation function as it has been shown in Refs. [6,15]. However, in these Letters the spatial variation of the pairing potential is of phenomenological origin. Contrary to this, we have proposed a microscopic mechanism that gives an explicit form of this dependence. In the following, we investigate whether the proposed scenario quantitatively reproduces the STS data. In particular, it is necessary to check whether a significant enhancement of the superconducting gap can be obtained for the model parameters, which do not lead to strong fluctuations of the electron concentration.

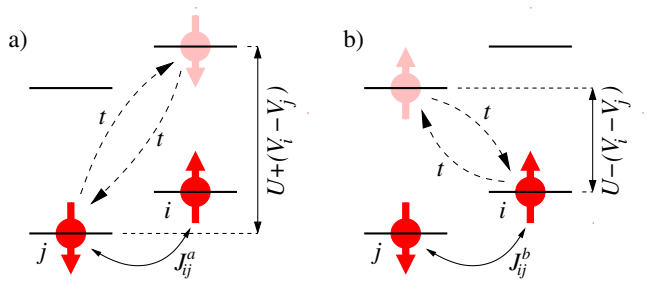

FIG. 2 (color online). Second order two-site processes contributing the effective coupling between spins at sites i and j.

In order to estimate the spatial variation of the superconducting order parameter, we have assumed the resonating valence bond scenario [11] and investigated the Hamiltonian (3) in the mean-field approximation with renormalized hopping integral $t \to \tilde{t} = (1 - n)t$ [11,16]. Within such an approach the constraint of no double occupancy is not fulfilled exactly. On the other hand, it is a method that allows one to investigate superconductivity in inhomogeneous systems by means of the Bogoliubov–de Gennes (BdG) equations. Following the experimental results we restrict further study to such values of the model parameters U and V_0, which give electron density fluctuations $\delta n < 0.1$ (see Fig. 1). Since the mean-field Hamiltonian does not include Coulomb correlations (apart from the bandwidth renormalization), taking into account V_i in the second term of the Hamiltonian (3) would expel electrons from the neighborhood of impurities almost completely. The influence of this term is unphysically enhanced due to the reduction of the kinetic energy ($t \to \tilde{t}$). On the other hand, as we have demonstrated in Fig. 1, this effect does not occur, when Coulomb repulsion is properly taken into account. In the t-J model, states with double occupancy are projected out completely and the charge inhomogeneity should be reduced even stronger than in the finite-U Hubbard model. Therefore, we neglect the term including V_i, keeping in mind that this approximation may overestimate Δ_i in the vicinity of impurities [3]. Then, the mean-field Hamiltonian takes on the form:

$$H_{t\text{-}J} = -\tilde{t}\sum_{\langle i,j\rangle\sigma}(a^\dagger_{i\sigma}a_{j\sigma} + \text{H.c.}) - \mu\sum_{i\sigma}a^\dagger_{i\sigma}a_{i\sigma} + \sum_{\langle i,j\rangle}[\Delta_{ij}(a^\dagger_{i\uparrow}a^\dagger_{j\downarrow} + a^\dagger_{j\uparrow}a^\dagger_{i\downarrow}) + \text{H.c.}], \quad (6)$$

where $\Delta_{ij} = -J_{ij}\langle a_{i\downarrow}a_{j\uparrow} + a_{j\downarrow}a_{i\uparrow}\rangle/2$. It can be diagonalized with the help of the transformation

$$c_{i\sigma} = \sum_n (u_{in}\gamma_{n\sigma} - s(\sigma)v^*_{in}\gamma^\dagger_{n\bar{\sigma}}), \quad (7)$$

where u_{in} and v_{in} fulfill the BdG equations:

$$\sum_j \begin{pmatrix} \mathcal{H}_{ij} & \Delta_{ij}\delta_{\langle ij\rangle} \\ \Delta^*_{ij}\delta_{\langle ij\rangle} & -\mathcal{H}_{ij} \end{pmatrix}\begin{pmatrix} u_{jn} \\ v_{jn} \end{pmatrix} = \varepsilon_n \begin{pmatrix} u_{in} \\ v_{in} \end{pmatrix}, \quad (8)$$

with $\mathcal{H}_{ij} = -\tilde{t}\delta_{\langle ij\rangle} - \mu\delta_{ij}$. Here, $\delta_{\langle ij\rangle} = 1$ for the neighboring sites i, j and 0 otherwise. The superconducting order parameter is determined self-consistently by:

$$\Delta_{ij} = \frac{J_{ij}}{2}\sum_n (u_{in}v^*_{jn} + u_{jn}v^*_{in})\tanh\frac{\varepsilon_n}{2k_BT}. \quad (9)$$

We have solved the BdG equations for the d-wave symmetry. The calculations have been carried out on a

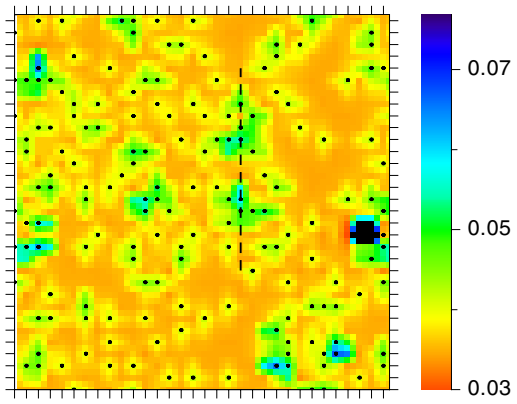

FIG. 3 (color online). Δ_i/t calculated for $U = 8t$, $V_0 = 5t$, $\lambda = 2a$ and $\bar{n} \simeq 0.83$. The positions of impurities are marked by dots. The dashed line shows the cross section, along which the LDOS presented in Fig. 4 was calculated.

32×32 cluster with 170 randomly distributed impurities, what gives concentration 16%. Assuming that each dopant introduces one hole into the CuO_2 plane, we have performed calculations for the same hole concentration. Figure 3 shows the spatial variation of the superconducting order parameter obtained for $U = 8t$, $V_0 = 5t$ and $\lambda = 2a$. For such model parameters exact diagonalization study gives the root mean square of the electron density below 0.1. (see Fig. 1), what is comparable to the experimentally observed fluctuations [4]. One can note a significant spatial variation of the superconducting gap. We have found that the root mean square of the order parameter equals 25% of its average value. As expected Δ_i increases in the vicinity of each impurity. However, the degree of this enhancement strongly depends on a configuration of impurities. It is significant in the regions, where several impurities are located close to each other. Otherwise it is much weaker. This result follows from the specific form of exchange interaction as given by Eq. (4). Because of the screening of the electrostatic potential, η_{ij} gives nonnegligible contribution to J_{ij} only in the closest neighborhood of an impurity. Consequently, the pairing interaction is enhanced in a region larger than the coherence length only provided this region contains several impurities. Figures 3 and 4 show that the mean-field approach correctly reproduces also the main qualitative features of the LDOS. In particular, increase of the superconducting gap in the vicinity of impurities is accompanied by a reduction of the height of the coherence peaks. The reduction is however much weaker than observed experimentally. This discrepancy can be attributed either to the limitations of the mean-field analysis or/and to fact, that we have considered only the increase of the pairing interaction and neglected other possible dopant-induced effects, e.g., modulation of the hopping integral.

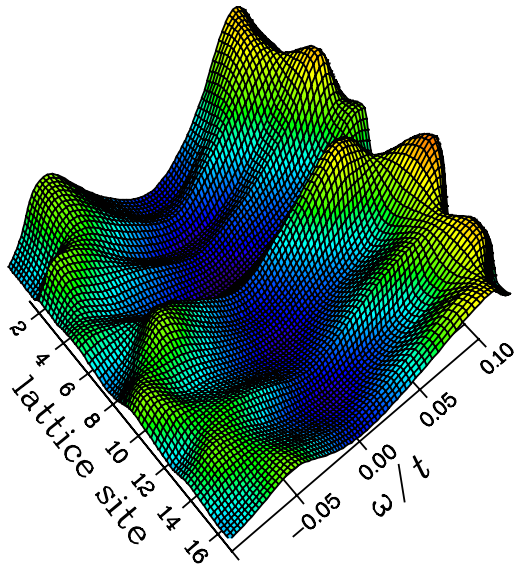

FIG. 4 (color online). LDOS calculated along the line marked in Fig. 3 for the same model parameters.

To conclude, we have derived a purely microscopic mechanism that can be responsible for the observed enhancement of the superconducting gap in the vicinity of impurities. One of the most attractive features of this approach concerns its simplicity. On the other hand, such a simple approach neglects other possible effects originating from the presence of the dopant oxygen atoms.

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