Dopant-Modulated Pair Interaction in Cuprate Superconductors

Tamara S. Nunner, Brian M. Andersen, Ashot Melikyan, and P. J. Hirschfeld
Department of Physics, University of Florida, Gainesville, Florida 32611, USA
(Received 27 April 2005; published 19 October 2005)

A comparison of recent experimental STM data with single-impurity and many-impurity Bogoliubov–de Gennes calculations strongly suggests that random out-of-plane dopant atoms in cuprates modulate the pair interaction locally. This type of disorder is crucial to understanding the nanoscale electronic inhomogeneity observed in BSCCO-2212, and can reproduce observed correlations between the positions of impurity atoms and various aspects of the local density of states such as the gap magnitude and the height of the coherence peaks. Our results imply that each dopant atom modulates the pair interaction on a length scale of order one lattice constant.

DOI: 10.1103/PhysRevLett.95.177003 PACS numbers: 74.72.−h, 74.20.Fg, 74.25.Jb

The discovery of nanoscale inhomogeneity in the cuprates has recently generated intense interest. In particular, the spectral gap in the local density of states (LDOS), as observed by scanning tunneling microscopy (STM) [1–4] in $Bi_2Sr_2CaCu_2O_{8+x}$ (BSCCO), varies by a factor of 2 over distances of 20–30 Å. This unusual behavior may help reveal how the cuprates evolve from the Mott insulating state at half filling to the superconducting state at finite doping. The hole concentration in the CuO_2 planes of BSCCO is proportional to the number of out-of plane dopant atoms, which also introduce disorder. This has led to the proposition that poorly screened electrostatic potentials of the dopant atoms generate a variation in the local doping concentration and thus give rise to the gap modulations observed in STM [5–7]. Poor screening has also been argued to result in enhanced forward scattering [8], which appears to be compatible with photoemission [9,10] and transport measurements [11] in the superconducting state of BSCCO. An alternate perspective is explored in several works which associate inhomogeneous electronic structure with a competing order parameter, such as antiferromagnetism [12–14]. Only very recently has it been possible to measure correlations between the inhomogeneities observed in STM and positions of dopant atoms [15], thus providing a clue to the relation between disorder and doping in this compound, as well as a means to examine the above proposals.

In this Letter, we assume that the electronic inhomogeneity observed by STM, at least in the optimally to overdoped samples, can be understood within the framework of BCS theory in the presence of disorder. We show that the conventional modeling of disorder as a set of random potential scatterers fails to reproduce the most prominent features of the STM experiments: (i) the subgap spectra are spatially extremely homogeneous [3], (ii) the coherence peaks in regions with larger gap tend to be much broader and reduced in height, (iii) the ''coherence peak'' positions are symmetric about zero bias, (iv) the dopants are found to correlate positively with large gap regions [15], and (v) charge modulations are considered to be smaller than 7% [15]. We propose that the dopant atoms modulate the local pair potential; i.e., the local attractive coupling g between electrons is spatially dependent. In conventional superconductors, such effects are difficult to observe because atomic-scale modulations in g produce LDOS modulations only on the scale of the coherence length ξ_0. In the cuprates, however, the situation is different due to the short coherence length. We demonstrate that a model in which dopant atoms modulate the pair interaction gives excellent agreement with respect to the above mentioned key characteristics of the STM data. A modulated pair interaction could arise from local lattice distortions surrounding the dopant atoms, which modify the electron-phonon interaction in their vicinity. Another possibility is that the lattice distortions modulate the superexchange interaction locally, which is supported by the fact that the superexchange in these materials varies strongly under pressure [16]. Even in conventional superconductors, modulated pair interactions have been suggested [17].

Model.—We consider the following mean-field Hamiltonian for a singlet d-wave superconductor

$$\hat{H} = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}} \hat{c}^{\dagger}_{\mathbf{k}\sigma} \hat{c}_{\mathbf{k}\sigma} + \sum_{i\sigma} V_i \hat{c}^{\dagger}_{i\sigma} \hat{c}_{i\sigma} + \sum_{\langle ij \rangle} (\Delta_{ij} \hat{c}^{\dagger}_{i\uparrow} \hat{c}^{\dagger}_{j\downarrow} + \text{H.c.}), \tag{1}$$

where $\epsilon_{\mathbf{k}} = -2t(\cos k_x + \cos k_y) - 4t' \cos k_x \cos k_y - \mu$ and $\sum_{\langle ij \rangle}$ denotes summation over neighboring lattice sites i and j. In the remainder of the Letter we will set $t'/t = -0.3$ and adjust μ to model the Fermi surface of BSCCO near optimal doping (for the homogeneous system, $\mu/t = -1.0$). In order to account for disorder in the out-of-plane dopants, which are separated from the CuO_2 plane by a distance z, we include an impurity potential modeled by $V_i = V_0 f_i$, where $f_i \equiv \sum_s \exp(-r_{is}/\lambda)/r_{is}$, and r_{is} is the distance from a dopant atom s to the lattice site i in the plane. Distances are measured in units of $\sqrt{2}a$, where a is the Cu-Cu distance. The nearest-neighbor d-wave order parameter $\Delta_{ij} = g_{ij} \langle \hat{c}_{i\uparrow} \hat{c}_{j\downarrow} - \hat{c}_{j\downarrow} \hat{c}_{i\uparrow} \rangle$ is determined self-consistently using (1) with $g_{ij} = g + \delta g(f_i + f_j)/2$ with modulation δg; this form is assumed purely for convenience in order to introduce a range λ. In traditional BCS

theory, $g_{ij} = g$ is spatially uniform, and Δ_{ij} is only modulated in the vicinity of potential scatterers [18,19]. We will argue that this approach is unable to reproduce observations (i)–(v) outlined above, and that g_{ij} is strongly modified near the dopant atoms.

Smooth potential.—If the potential caused by the out-of-plane dopant atoms were very smooth on the scale of ξ_0, the local properties of the inhomogeneous system would be determined by the local value of the disorder potential and the local value of the pairing interaction. Therefore, one would expect an LDOS which is locally similar to a clean superconductor with renormalized chemical potential $\mu - V_i$ in the case of a smooth potential V_i, or with renormalized bond order parameter $\Delta_{ij} + \delta\Delta_{ij}$ for a smooth off-diagonal (OD) potential. In the case of a conventional diagonal potential, a gap size modulation will be induced because the gap is a relatively sensitive function of the local chemical potential; see Fig. 1(a). On the other hand, modulations of this type will inevitably have coherence peak weight-position correlations opposite to experiment, since large gap values in the homogeneous system imply (within BCS theory) that spectral weight removed from low energies is transferred into the coherence peaks [Fig. 1(b)]. In a tight binding model, this effect can be further enhanced by the presence of a nearby van Hove singularity ($\omega_{vH} = \sqrt{(4t' + \mu)^2 + (4\Delta_0)^2}$) especially at $\mu/t = -1.2$ where it coincides with the gap edge. Here Δ_0 is the bond order parameter in the homogeneous system. A similar although less pronounced positive correlation between coherence peak weight and position arises also for the smooth OD case. Note that throughout this work we neglect inelastic scattering that would broaden the tunneling conductance peaks at large bias but would not change their weight, thus leaving our conclusions unaffected.

Single-impurity scattering.—Since a smooth disorder potential cannot reproduce the experimentally observed relation between the weight of the coherence peak and the gap magnitude, we now address the opposite limit, i.e., a very spiky potential caused by a dopant potential with short range on the scale of ξ_0. Some insight into this situation can be obtained by analyzing single-impurity scattering processes, which should be dominant for sufficiently short ranged and weak scattering potentials, where interference effects are negligible. For simplicity, we assume a constant order parameter in the following T-matrix analysis and postpone the fully self-consistent treatment to the many-impurity case.

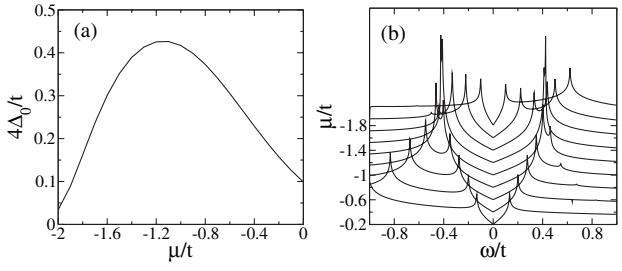

FIG. 1. (a) Bulk order parameter Δ_0 for $t'/t = -0.3$ and a constant nearest-neighbor attraction of $g/t = 1.16$ as a function of chemical potential μ. (b) LDOS for different μ using the order parameter displayed in (a).

A weak to intermediate strength impurity $V \lesssim t$ does not lead to well-defined resonant states inside the gap, as shown in Fig. 2(a). The positions of the coherence peaks are hardly shifted at all, and while the spectral weight of the coherence peaks is modified, this occurs in a distinctly particle-hole asymmetric fashion. This is in striking contrast to the STM spectra, where inhomogeneous but particle-hole symmetric coherence peak modulations are observed. In addition, there is no distinct feature in experiment corresponding to the van Hove features present, as, e.g., in Figs. 1(b) and 2(a).

These shortcomings of the conventional potential scattering model can be overcome by considering OD scattering instead. For the sake of clarity, in this paragraph we neglect the diagonal component of the potential. Figure 2(b) and 2(c) show the LDOS at the impurity site for a "pointlike" OD scatterer with d-wave symmetry on the four bonds emanating from site $i = 0$, $\delta\Delta_{0,\pm\hat{x}} = -\delta\Delta_{0,\pm\hat{y}} \equiv \delta\Delta$, and a more extended OD scatterer with $\delta\Delta_{ij} = \pm\delta\Delta(f_i + f_j)/2$, where f_i is defined below Eq. (1), and the negative sign applies to bonds oriented along the $\hat{y}$ direction. Scattering off an order parameter enhancement [see Fig. 2(b)] strongly suppresses the coherence peaks for large values of $\delta\Delta$ or more extended OD scatterers. For scattering by a local order parameter suppression [see Fig. 2(c)], exactly the opposite happens: an Andreev resonance forms just below the gap edge, similar to the case where the order parameter is suppressed near surfaces [20,21] or on finite patches [22]. For large negative values of $\delta\Delta$, or more extended OD scatterers, the Andreev resonance moves to smaller energies, and its peak height increases. It draws most of its spectral weight from the van Hove singularity at $(\pi, 0)$, which is close to the part of the Fermi surface with the largest d-wave gap, i.e., the

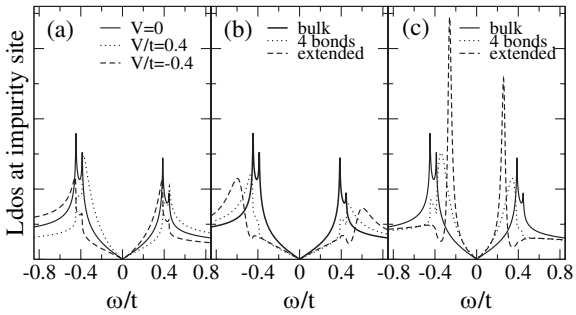

FIG. 2. On-site LDOS for different single-impurity models with $t'/t = -0.3$, $\mu/t = -1$, and $\Delta_0/t = 0.1$. (a) Weak pointlike potential scatterer. (b) Dotted line: attractive pointlike OD scatterer with $\delta\Delta = \Delta_0$ on the four bonds surrounding the impurity site. Dashed line: extended attractive OD scatterer with $\lambda = z = 1$. (c) Same as (b), with $\delta\Delta = -\Delta_0$.

part which is most affected by order parameter modulations. Although this indicates that the weight of the resonance depends on band structure, we find that the phenomenon is very robust over a wide range of t' and μ.

Many-impurity results.—We now address the effects of self-consistency and interference between many impurities by solving the Bogoliubov–de Gennes (BdG) equations resulting from Eq. (1), on a 80×80 lattice rotated by $\pi/4$ compared to the Cu-O bond direction (as in experimental STM maps); i.e., our system contains 2×80^2 lattice sites in total. We assume that the dopant atoms are interstitial oxygens, which each add two holes to the CuO_2 plane. Thus we consider a random distribution of 7.5% dopant atoms for optimal doping.

In the limit of a smooth potential [Fig. 3(a)], the many-impurity results agree well with the local μ picture discussed above. The correlation between the dopant positions and the gap amplitude depends strongly on the magnitude of the potential due to the nonmonotonic dependence of Δ_0 on the local μ, as shown in Fig. 1(a). The spatial variation of the gap, however, is not rapid enough to reproduce the grainy gap maps seen experimentally with gap ''patches'' of typical size 20–30 Å [3]; one is therefore forced to consider ''spikier'' potentials [Fig. 3(b)]. In the weak limit $V \lesssim t$, one recovers the results of the single-impurity case; i.e., the coherence peaks are modulated in a particle-hole asymmetric way. For the stronger spiky potentials required to reproduce the magnitude of the gap modulations observed in STM, subgap states start to form in contradiction with experiment [see Fig. 3(b)]. Further discrepancies between Figs. 3(a) and 3(b) and the experimental spectra are: (i) the LDOS clearly does not exhibit the inverse relation between gap size and coherence peak height; (ii) the spectra are quite particle-hole asymmetric [see Fig. 3(b) and high energy regions in Fig. 3(a)]; and (iii) the sizable potential required to induce gap modulations inevitably leads to large [$\mathcal{O}$(50%–100%)] local charge modulations. The latter point puts strong constraints on any potential scattering model, since the primary role of the impurity potential is to couple to the density.

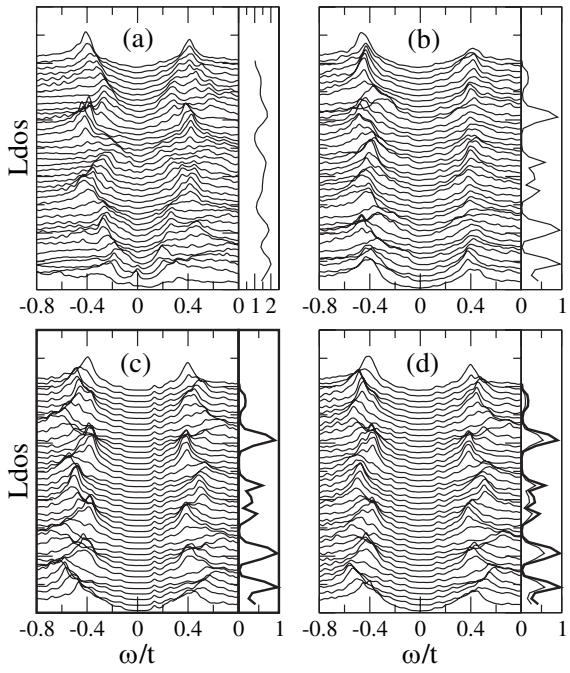

FIG. 3. LDOS from self-consistent solution of BdG equations, along a straight line for (a) conventional potential with $z = 2$, $\lambda = 2$, $V_0 = 1.5t$; (b) same as (a), but with $z = 0.57$, $\lambda = 0.5$, $V_0 = t$; (c) OD potential with $z = 0.57$, $\lambda = 0.5$, $\delta g = t$; and (d), combination of OD potential shown in (c) with conventional potential as in (b) but with $V_0 = 0.6t$. Conventional (OD) potentials are depicted to right of each panel as a thin (thick) line in units of t.

A typical LDOS line scan for a many OD impurity calculation is shown in Fig. 3(c). Note that, by construction, this model has homogeneous low-energy LDOS as well as strong correlations between the dopant positions and the local gap values. As in the single-impurity case, the line shape of the LDOS near the gap edge is determined primarily by Andreev scattering. Because the form of the LDOS near the gap edge is reminiscent of a coherence peak, we will simply adopt this terminology, as used in experiment. Although the dopant atoms inevitably give rise to a conventional potential as well, the qualitative features of OD scattering just described are not affected by a moderate admixture of conventional scattering [see Fig. 3(d)]. Comparing Figs. 3(a)–3(d), it is evident that the OD LDOS spectra are far more particle-hole symmetric than those with potential disorder, and display the inverse relation between gap magnitude and coherence peak height, as expected from the single-impurity discussion (see Fig. 2). In Fig. 4 we show the associated gap map (a), the coherence peak height map (b), and the charge modulation map (c) for parameters corresponding to Fig. 3(c). Figure 4(d) displays the correlation functions between the gap map and the dopants, and the gap map and the peak height map [23]. The local pairing modulation shown in Fig. 4 reproduces qualitatively the correct negative correlation between the gap amplitude and the coherence peak height [3,24], the positive correlation between dopant atom locations and large gap values, and the relatively small charge modulations observed in experiment [15]. In addition, the spectra exhibit the same remarkable particle-hole symmetric modulations of the coherence peaks observed in experiment [3,4]. This symmetry should manifest itself in Fourier transform quasiparticle interference patterns as well.

In the OD scattering model, short-distance correlations between the dopant atoms and the gap size are nearly perfect, as seen in Fig. 4(d); indeed, they are considerably stronger than reported in experiment [15]. This might be due to the difficulty of identifying all dopant positions experimentally, to the presence of additional cation disorder in BSCCO [25], or to the finite experimental resolution of the dopant resonances. The dopant-gap correlations are quite robust against inclusion of a conventional scattering

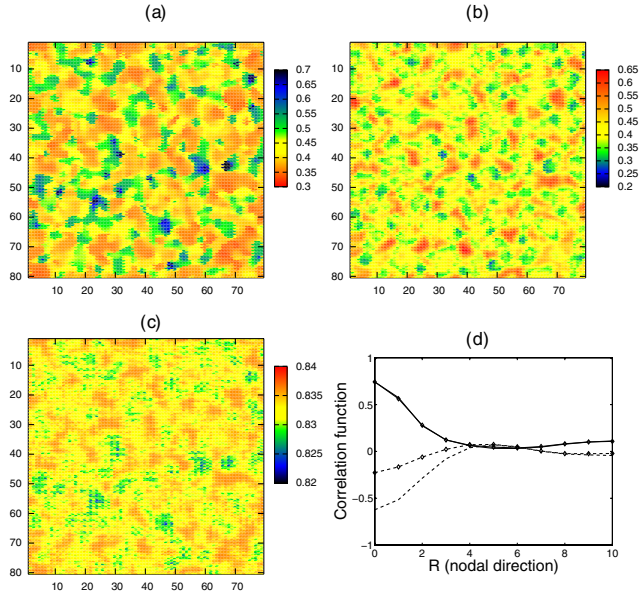

FIG. 4 (color online). Many OD impurity model for parameters of Fig. 3(c): (a) 2D real-space map of the local coherence peak position (gap) in units of t; (b) coherence peak height [note the inverse color scale with respect to (a)]; (c) total charge (note the small scale); and (d) the correlation function between the gap map and the dopant atoms (solid line), and the gap map and the peak height map (dashed line). Lines without (with) symbols correspond to the parameters used in Fig. 3(c) [Fig. 3(d)].

component [the two solid curves in Fig. 4(d) coincide], but the gap-peak height correlations are rapidly suppressed, as seen in Fig. 4(d).

A natural question is the extent to which these correlations are robust against different choices of parameters. We find that the local spectral properties in the spiky regime of the OD model are insensitive to parameters, provided the amplitude of the gap modulation $\delta\Delta$ is comparable to or larger than the splitting of the van Hove and coherence peaks in the pure system. In that case the weight of the van Hove peak is absorbed into the coherence peak [Fig. 2(c)].

While we assert the primacy of the OD channel of scattering for the modulation of the states near the antinode, we emphasize that nodal quasiparticles are very weakly scattered by this potential, and so microwave and thermal transport are probably only minimally influenced by the effects discussed here [11]. This further implies that the elastic contribution to the ARPES spectral peaks near the antinodal and nodal points are determined by completely different scattering processes.

In the calculations reported here we have focused on optimal doping where it is generally believed that a mean-field BCS treatment is appropriate. In order to address the doping dependence of the inhomogeneities the presence of strong correlations in the underdoped regime has to be taken into account.

Conclusions.—We have offered strong evidence that the inhomogeneity in the coherence peak position as observed in STM experiments is driven by dopant atoms, located away from the CuO_2 plane, whose primary effect on one-particle properties is to modulate the local pair interaction. This ansatz allowed us to reproduce most of the correlations observed in recent STM experiments: nanoscale inhomogeneity of the coherence peak position [1–4], homogeneity of the LDOS at low bias [3], low charge disorder [15], negative peak-height to gap value correlation [3,24], and positive gap -dopant position correlation [15]. We believe that our results represent an important step towards understanding the microscopic nature of the pair interaction.

The authors are grateful to Yu. S. Barash, W. Chen, J. C. Davis, J. Lee, K. McElroy, J. Slezak, and L. Zhu for useful discussions. Work was supported by the A. v. Humboldt Foundation (T. S. N.) and ONR Grant No. N00014-04-0060 (P. J. H., B. M. A.).

[1] T. Cren *et al.*, Europhys. Lett. **54**, 84 (2001).
[2] S. H. Pan *et al.*, Nature (London) **413**, 282 (2001).
[3] K. M. Lang *et al.*, Nature (London) **415**, 412 (2002).
[4] C. Howald, P. Fournier, and A. Kapitulnik, Phys. Rev. B **64**, 100504(R) (2001).
[5] Z. Wang *et al.*, Phys. Rev. B **65**, 064509(R) (2002).
[6] Q.-H. Wang, J. H. Han, and D.-H. Lee, Phys. Rev. B **65**, 054501 (2002).
[7] I. Martin and A. V. Balatsky, Physica C (Amsterdam) **357-360**, 46 (2001).
[8] E. Abrahams and C. M. Varma, Proc. Natl. Acad. Sci. U.S.A. **97**, 5714 (2000).
[9] D. J. Scalapino, T. S. Nunner, and P. J. Hirschfeld, cond-mat/0409204.
[10] L. Zhu, P. J. Hirschfeld, and D. J. Scalapino, Phys. Rev. B **70**, 214503 (2004).
[11] T. S. Nunner and P. J. Hirschfeld, Phys. Rev. B **72**, 014514 (2005).
[12] S. A. Kivelson *et al.*, Rev. Mod. Phys. **75**, 1201 (2003).
[13] W. A. Atkinson, Phys. Rev. B **71**, 024516 (2005).
[14] G. Alvarez *et al.*, Phys. Rev. B **71**, 014514 (2005).
[15] K. McElroy *et al.*, Science **309**, 1048 (2005).
[16] M. C. Aronson *et al.*, Phys. Rev. B **44**, 4657 (1991).
[17] I. N. Khlyustikov and A. I. Buzdin, Adv. Phys. **36**, 271 (1987); A. I. Larkin, Sov. Phys. JETP **31**, 784 (1970).
[18] M. H. Hettler and P. J. Hirschfeld, Phys. Rev. B **59**, 9606 (1999).
[19] A. Shnirman *et al.*, Phys. Rev. B **60**, 7517 (1999).
[20] L. J. Buchholtz *et al.*, J. Low Temp. Phys. **101**, 1099 (1995).
[21] Yu. S. Barash, A. A. Svidzinsky, and H. Burkhardt, Phys. Rev. B **55**, 15 282 (1997).
[22] A. Fang *et al.*, cond-mat/0508253.
[23] The correlation function between f and g is defined as $C_{f,g}(\mathbf{R}) \equiv \int d^2\mathbf{r}[f(\mathbf{r}) - \langle f\rangle][g(\mathbf{r}+\mathbf{R}) - \langle g\rangle]/(\sqrt{A_f A_g})$, $A_f \equiv \int d^2\mathbf{r}[f(\mathbf{r}) - \langle f\rangle]^2$ and $\langle f\rangle$ is the average of f.
[24] A. Fang *et al.*, Phys. Rev. B **70**, 214514 (2004).
[25] H. Eisaki *et al.*, Phys. Rev. B **69**, 064512 (2004).