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Spin-orbit scattering and pair breaking in a structurally disordered copper oxide layer

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To leading order in displacement size, the scattering of electrons in a Cu-O plane from O displacements perpendicular to that plane is due to spin-orbit coupling. This scattering is investigated with the following results: (1) As a consequence of time-reversal symmetry, spin fluctuations, which can strongly enhance scattering from a spin impurity, do not enhance spin-orbit scattering; and (2) for a superconductor with a $d_{x^2-y^2}$ gap function, pair breaking from spin-orbit scattering can be strong, particularly in a structurally disordered phase where locally CuO_6 octahedra tilt as in the orthorhombic phase of La_2CuO_4, but globally the average structure is tetragonal. These results are discussed in the context of the (La,Nd)-(Sr,Ba)-Cu-O system where certain structural transitions are observed to suppress superconductivity.

The microscopic origin of high transition temperatures in the cuprate superconductors is still unknown, even after six years of intense study. One school of thought holds that superconductivity in these materials arises from the exchange of nearly antiferromagnetic spin fluctuations. If this is the case then it is almost certain that Cooper pairs form with $d_{x^2-y^2}$ symmetry.[1]

There is now a great deal of experimental evidence which shows an intriguing interplay between small changes in lattice structure and superconductivity in the La-based cuprates.[2–4] This interplay was first observed in the $La_{2-x}Ba_xCuO_4$ system which, when $x \simeq 0.12$, undergoes two structural phase transitions.[2] The first transition is from an undistorted high-temperature tetragonal phase into a low-temperature orthorhombic (LTO) phase. In the LTO phase the CuO_6 octahedra making up each Cu-O layer tilt in a staggered fashion about the (110) axis. The second transition is from the LTO phase into a low-temperature tetragonal (LTT) phase in which, on average, the CuO_6 octahedra tilt first about the (100) and then the (010) axes in successive layers. In this new phase superconductivity appears to be completely destroyed,[2] and recent experiments on the $La_{2-x-y}Nd_ySr_xCuO_4$ system show a similar correlation between unusual low-temperature structural phases (i.e., the LTT phase, and another phase with space group *Pccn*, intermediate between the LTO and LTT phases) and suppression of superconductivity.[3,4]

One possible explanation for these experiments is that this suppression of superconductivity is due to pair breaking.[5,6] It is a well-known characteristic of unconventional pairing, such as *d*-wave, that the superconducting transition temperature, T_c, is sensitive to elastic impurity scattering.[7] Because the LTT and *Pccn* phases are stabilized by random substitution of Nd or Ba ions for La, it is likely that these phases contain more structural disorder than the LTO phase. If so, then elastic scattering of electrons from this disorder may be responsible for the observed suppression of superconductivity.

The tilting of a CuO_6 octahedron in a given Cu-O plane causes O ions to be displaced out of that plane. In what follows a "one-band" Hamiltonian is used to describe the coupling of electrons to these displacements:[11]

$$H_0 = -t \sum_{\substack{i,j\\ \alpha\beta}} c^\dagger_{i\alpha} \left[(1-\rho\theta_{ij}^2)\delta_{\alpha\beta} + i\nu\theta_{ij}\hat{\eta}_{ij}\cdot\sigma_{\alpha\beta} \right] c_{j\beta}. \quad (1)$$

The index i labels Cu sites on a two-dimensional square lattice with N sites, $c^\dagger_{i\alpha}$ is the creation operator for an electron with spin α at site i, and θ_{ij} is the angle between the Cu-O plane and the bond made by the Cu ion at site i and the O ion between sites i and j. Recent microscopic calculations have found that $\hat{\eta}_{i,i+\hat{x}} \simeq \hat{\mathbf{y}}$ and $\hat{\eta}_{i,i+\hat{y}} \simeq -\hat{\mathbf{x}}$.[8–10] Hamiltonian (1) describes two distinct electron-lattice couplings: (i) the spin-independent θ^2 coupling which arises from the quadratic modification of the Cu-O bond lengths in the presence of an O displacement; and (ii) the linear in θ coupling which occurs through the spin orbit.[11] At half filling (one electron per site) (ii) is responsible for the anisotropic Dzyaloshinki-Moriya corrections to the otherwise isotropic superexchange interaction between Cu spins.[12] The size of these corrections are known from experiment[13] and can be used to estimate ν.[8] The parameter values used here are $t \sim 400$ meV,[14] $\nu \sim 0.2$,[8,13] and ρ is expected to be of order 1.

For a coherent tilting distortion

$$\begin{aligned} \theta_{i,i+\hat{x}} &= \theta_0 \sin\chi \exp(i\mathbf{Q}\cdot\mathbf{r}_i), \\ \theta_{i,i+\hat{y}} &= \theta_0 \cos\chi \exp(i\mathbf{Q}\cdot\mathbf{r}_i), \end{aligned} \quad (2)$$

where $\mathbf{Q} \equiv (\pi,\pi)$, and where $\chi = \pi/4$ in the LTO phase, $\chi = 0$ in the LTT phase, and $0 < \chi < \pi/4$ in the *Pccn* phase. These coherent distortions cause Bragg scattering of electrons through the spin-orbit coupling term in (1). For Bragg scattering it is possible to rediagonalize (1) so that there is no scattering; however, a random component to θ_{ij} will give ergodic scattering.

Before proceeding it is useful to contrast spin-orbit scattering as described by (1) with spin-impurity scattering as described by the interaction Hamiltonian

$$H_{\rm spin} = J \sum_i \mathbf{S}_i \cdot c^\dagger_i \sigma c_i. \quad (3)$$

Electrons will scatter elastically from a random displacement field θ_{ij} as well as a random spin configuration $\mathbf{S}_i$ through the couplings in (1) and (3). Although both scattering processes involve spin, there is an important difference: Spin impurities are not time-reversal invariant perturbations ($\mathbf{S} \rightarrow -\mathbf{S}$ under time reversal) while spin-orbit impurities, i.e., O displacements, are ($\theta \rightarrow \theta$ under time reversal). One well-known consequence of this difference is that spin-impurity scattering is pair breaking for a conventional s-wave superconductor,[5] but spin-orbit scattering is not (Anderson's theorem).[15]

Another consequence of time-reversal symmetry appears when one considers the possible spin-fluctuation enhancement of the scattering vertex for spin-impurity and spin-orbit scattering. A spin impurity embedded in an electron fluid polarizes the spins which surround it. If this fluid is characterized by strong spin fluctuations, the polarized region can be quite large. Quasiparticles then scatter from the impurity spin together with its polarization cloud, and this leads to enhanced scattering at the characteristic spin-fluctuation wave vectors. Because both spin-orbit and spin-impurity scattering involve a spin flip, it is natural to ask if spin-orbit scattering can be similarly enhanced by spin fluctuations.

To answer this question, consider adding a Hubbard U interaction, ($H_{\text{Hub.}} = U \sum_i n_{i\uparrow} n_{i\downarrow}$), to (1) and (3). For both spin-orbit and spin-impurity scattering the renormalized scattering vertex can be written $\Gamma_{\alpha\beta}(\mathbf{k}, \mathbf{k}+\mathbf{q}) = \mathbf{\Lambda}_{\mathbf{k},\mathbf{k}+\mathbf{q}} \cdot \boldsymbol{\sigma}_{\alpha\beta}$. Figure 1 shows the diagrammatic equation for Γ where $H_{\text{Hub.}}$ is treated in the random-phase approximation (RPA). The corresponding self-consistent equation for $\mathbf{\Lambda}^{(\text{RPA})}$ is

$$\mathbf{\Lambda}^{(\text{RPA})}_{\mathbf{k},\mathbf{k}+\mathbf{q}} = \mathbf{\Lambda}^{(0)}_{\mathbf{k},\mathbf{k}+\mathbf{q}} + U \int \frac{d^2p}{(2\pi)^2} \frac{f_{\mathbf{p}+\mathbf{q}} - f_{\mathbf{p}}}{\epsilon_{\mathbf{p}+\mathbf{q}} - \epsilon_{\mathbf{p}}} \mathbf{\Lambda}^{(\text{RPA})}_{\mathbf{p},\mathbf{p}+\mathbf{q}}. \quad (4)$$

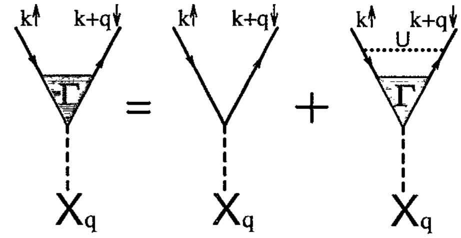

FIG. 1. Diagrammatic representation of the self-consistent equation for the RPA enhancement of a scattering vertex for a generic spin-flip scatterer (only the diagrams for the σ^- component are shown). For spin-impurity scattering ($X_{\mathbf{q}} = S_{\mathbf{q}}$) the solution to this equation gives a typical RPA enhancement factor of $[1-U\chi_0(\mathbf{q})]^{-1}$ while for spin-orbit scattering ($X_{\mathbf{q}} = \theta_{\mathbf{q}}$) there is no such enhancement.

Here $\epsilon_{\mathbf{q}} = -2t(\cos q_x + \cos q_y) - \mu$ where μ is the chemical potential, and $f_p \equiv f(\epsilon_p)$ is the Fermi function. Time-reversal symmetry requires that $\mathbf{\Lambda}_{\mathbf{k},\mathbf{k}'} = \pm\mathbf{\Lambda}_{-\mathbf{k},-\mathbf{k}'}$ with the + and − signs holding for spin-impurity and spin-orbit scattering, respectively. Because of this difference the solution to (4) is $\mathbf{\Lambda}^{(\text{RPA})}_{\mathbf{k},\mathbf{k}'} = [1 - U\chi_0(\mathbf{k} - \mathbf{k}')]^{-1}\mathbf{\Lambda}^{(0)}_{\mathbf{k},\mathbf{k}'}$ for spin-impurity scattering, where $\chi_0(\mathbf{q})$ is the static spin susceptibility for noninteracting electrons, and $\mathbf{\Lambda}^{(\text{RPA})}_{\mathbf{k},\mathbf{k}'} = \mathbf{\Lambda}^{(0)}_{\mathbf{k},\mathbf{k}'}$ for spin-orbit scattering. Thus, as a consequence of time-reversal symmetry, the ladder diagrams shown in Fig. 1, which enhance spin-impurity scattering when $1 - U\chi_0(\mathbf{k}-\mathbf{k}')$ is small, do not enhance spin-orbit scattering.

Next we proceed with the conventional pair-breaking analysis,[5] which begins with the linearized Gor'kov-Dyson equations in the Matsubara formalism

$$\Delta_{\mathbf{k},n} = -\pi T \sum_m^{|\omega_m|<\omega_{\text{SF}}} \int \frac{d\theta_{\mathbf{k}'}}{2\pi} N(\theta_{\mathbf{k}'}) \frac{\Delta_{\mathbf{k}',m}}{|Z_{\mathbf{k}',m}||\omega_m|} \left(V_{\mathbf{k},\mathbf{k}'} - \frac{1}{T}(|v^{(-)}_{\mathbf{k},\mathbf{k}'}|^2 + |w_{\mathbf{k},\mathbf{k}'}|^2)\delta_{m,n} \right), \quad (5)$$

$$\Sigma_{\mathbf{k},n} = i\omega_n(1 - Z_{\mathbf{k},n}) = -i\pi \text{sgn}(\omega_n) \int \frac{d\theta_{\mathbf{k}'}}{2\pi} N(\theta_{\mathbf{k}'})(|v^{(+)}_{\mathbf{k},\mathbf{k}'}|^2 + |w_{\mathbf{k},\mathbf{k}'}|^2), \quad (6)$$

where $\Delta_{\mathbf{k},n}$ and $\Sigma_{\mathbf{k},n}$ are the anomalous and normal self energies, $\omega_n = (2n+1)\pi T$ is the nth Matsubara frequency, the Fermi surface is parametrized by the angle $\theta_{\mathbf{k}}$, and $N(\theta_{\mathbf{k}})$ is the local density of states.

The phenomenological effective pairing interaction in (5) is taken to be

$$V_{\mathbf{k},\mathbf{k}'} = -\lambda\phi_d(\mathbf{k})\phi_d(\mathbf{k}') \quad (7)$$

where $\phi_d(\mathbf{k}) = A(\cos k_x - \cos k_y)$ with $A = [\int (d\theta_{\mathbf{k}}/2\pi) N(\theta_{\mathbf{k}})(\cos k_x - \cos k_y)^2]^{-1/2}$. For $\lambda > 0$ this interaction is attractive in the $d_{x^2-y^2}$ channel. The sum over Matsubara frequencies in (5) must be cut off for large frequencies. Within the spin-fluctuation model the cutoff ω_{SF} should be viewed as a characteristic spin-fluctuation frequency. The critical temperature T_c is determined by finding the temperature at which (5) and (6) have a nontrivial solution.

The functions $|v^{(\pm)}_{\mathbf{k},\mathbf{k}'}|^2$ and $|w_{\mathbf{k},\mathbf{k}'}|^2$ in (5) and (6) are the scattering matrix elements coming from the spin-orbit and spin-independent couplings in (1), respectively. To leading order in θ

$$|v^{(\pm)}_{\mathbf{k},\mathbf{k}'}|^2 = 4t^2\nu^2 \sum_{a,b\in\{x,y\}} C_{ab}(\mathbf{k} \pm \mathbf{k}') \sin\left(\frac{k_a \pm k'_a}{2}\right) \times \sin\left(\frac{k_b \pm k'_b}{2}\right) \quad (8)$$

and

$$|w_{\mathbf{k},\mathbf{k}'}|^2 = 4t^2\rho^2 \sum_{a,b\in\{x,y\}} F_{ab}(\mathbf{k} - \mathbf{k}') \cos\left(\frac{k_a - k'_a}{2}\right) \times \cos\left(\frac{k_b - k'_b}{2}\right) \quad (9)$$

with

$$C_{ab}(\mathbf{q}) = \langle \theta_{\mathbf{q},a}\theta_{-\mathbf{q},b}\rangle \ , \qquad (10)$$

$$F_{ab}(\mathbf{q}) = \int \frac{d^2p}{(2\pi)^2} \int \frac{d^2p'}{(2\pi)^2} \langle \theta_{\mathbf{q}+\mathbf{p},a}\theta_{-\mathbf{p},a}\theta_{-\mathbf{q}+\mathbf{p}',b}\theta_{-\mathbf{p}',b}\rangle \ , \qquad (11)$$

where $\langle\cdots\rangle$ denotes an average over disorder, and $\theta_{\mathbf{q},a} = 1/N\sum_i \exp(i\mathbf{q}\cdot\mathbf{r}_i)\theta_{i,i+\hat{\mathbf{a}}}$. Although spin-orbit scattering enters the equations for the anomalous and normal self energies differently because of the spin flip, for even-parity singlet pairing $\mathbf{k}'$ can be replaced by $-\mathbf{k}'$ in (5). Accordingly the ($\pm$) superscript is suppressed in what follows.

Assuming the gap function can be factorized as $\Delta_{\mathbf{k},m}/Z_{\mathbf{k},m} = \phi_d(k)\tilde{\Delta}_m$ then (5) and (6) can be combined to yield

$$\tilde{\Delta}_n = \pi T \sum_m^{|\omega_m|<\omega_{\rm SF}} \frac{\tilde{\Delta}_m}{|\omega_m|}\left(\lambda - \delta_{m,n}\frac{1}{\pi T\tau_{\rm pb}}\right). \qquad (12)$$

Here $1/\tau_{\rm pb} = 1/\tau_{\rm pb}^{\rm so} + 1/\tau_{\rm pb}^{\rm si}$ where $1/\tau_{\rm pb}^{\rm so}$ and $1/\tau_{\rm pb}^{\rm si}$ are the pair-breaking rates from spin-orbit and spin-independent scattering, respectively, and are given by

$$\frac{1}{\tau_{\rm pb}^{\rm so}} = \frac{\pi}{2}\int \frac{d\theta_{\mathbf{k}}}{2\pi}N(\theta_{\mathbf{k}})\int \frac{d\theta_{\mathbf{k}'}}{2\pi}N(\theta_{\mathbf{k}'})|v_{\mathbf{k},\mathbf{k}'}|^2 \times[\phi_d(\mathbf{k}) - \phi_d(\mathbf{k}')]^2 \qquad (13)$$

and a similar expression with $|v_{\mathbf{k},\mathbf{k}'}|^2$ replaced by $|w_{\mathbf{k},\mathbf{k}'}|^2$ for $1/\tau_{\rm pb}^{\rm si}$. Equation (12) is precisely the same as the equation for the suppressed T_c of a conventional s-wave superconductor in the presence of magnetic impurities.[6] The standard analysis then shows that T_c is reduced to zero when $1/\tau_{\rm pb} = \pi T_{c0}/2\gamma \simeq 0.88T_{c0}$, where T_{c0} is the transition temperature when $1/\tau_{\rm pb} = 0$ and the reduced transition temperature is T_c.[6]

To calculate $1/\tau_{\rm pb}$ it is necessary to know the correlation functions (10) and (11) which characterize the structural disorder. The LTT and *Pccn* phases of the (La,Nd)-(Ba,Sr)-Cu-O system are stabilized by randomly placed Nd or Ba ions at La sites. It is plausible that these randomly placed ions alter the local tilting environment so that the average structure is well defined, but locally the CuO_6 octahedra tilt about random axes. A simple model structure which may capture the essence of this type of disorder is one in which CuO_6 octahedra tilt coherently on length scales less than a structural coherence length, ξ_s, while on longer length scales the structure is completely disordered. In the presence of such disorder the function $C(\mathbf{q})$ is peaked at $\mathbf{q} = \mathbf{Q}$ and has a width $\Delta q \simeq 1/\xi_s$. For the calculations presented below we use $C_{ab}(\mathbf{q}) \propto \exp[-2\xi_s^2(\mathbf{q}-\mathbf{Q})^2]\delta_{a,b}$ where the normalization is fixed by the requirement that the integral of $C(\mathbf{q})$ over the Brillouin zone must equal the mean-square displacement angle θ_0^2. To allow a comparison of the relative importance of spin-orbit and spin-independent scattering it is further assumed that the disorder is Gaussian so that $F_{ab}(\mathbf{q}) = (2\pi)^{-2}\int d^2p C(\mathbf{q}+\mathbf{p})C(-\mathbf{p})\delta_{a,b}$.

First consider uncorrelated disorder ($\xi_s \to 0$). Performing the integral (13) for this case using a nearest-neighbor tight-binding band structure and taking a chemical potential of $\mu = -0.15t$ yields $1/\tau_{\rm pb}^{\rm so} \simeq 3.7t\nu^2\theta_0^2$ and $1/\tau_{\rm pb}^{\rm si} \simeq 1.3t\rho^2\theta_0^4$. These pair-breaking rates illustrate the importance of including spin-orbit coupling when treating electron scattering from structural disorder in a Cu-O layer. Spin-orbit scattering gives a pair-breaking rate which is quadratic in the root-mean-square displacement, while spin-independent scattering gives a rate which is quartic. However, the spin-orbit scattering rate also contains a factor of $\nu^2 \sim 4\times10^{-2}$ and so, for $\theta_0 \sim 0.1$, in the presence of uncorrelated disorder, spin-orbit and spin-independent scattering are roughly of equal strength.

When ξ_s is increased, pair-breaking from spin-orbit and spin-independent scattering are no longer comparable in magnitude. Figure 2 shows the "pair-breaking temperature" $T_{\rm pb} \equiv 2\gamma/\pi\tau_{\rm pb}$ due to spin-orbit and spin-independent scattering, plotted vs ξ_s (in units of the lattice spacing) for $\mu = -0.15t$, $\gamma = 0.2$, and $\rho = 3.4$ [this value of ρ is chosen for convenience so that $(1/\tau_{\rm pb})_{\rm so} = (1/\tau_{\rm pb})_{\rm si}$ when $\xi_s = 0$]. Any superconductor with a $d_{x^2-y^2}$ gap function which, in the absence of disorder, has a critical temperature $T_{c0} < T_{\rm pb}$ will have its T_c reduced to zero when the pair breaking lifetime is $\tau_{\rm pb}$. As ξ_s increases pair breaking from spin-independent scattering is suppressed and pair breaking from spin-orbit scattering is enhanced. Note that for some parameters the pair breaking from spin-orbit scattering can be strong enough to reduce to zero the T_c of a superconductor with $T_{c0} \sim 30$ K.

The reason for this enhancement is illustrated in Fig. 3. This figure shows the Fermi surface for a nearest-neighbor tight-binding band at 10% doping, a typical $\mathbf{k}$ point on that Fermi surface, and the region in momentum space containing those points $\mathbf{k}'$ for which the spin-orbit scat-

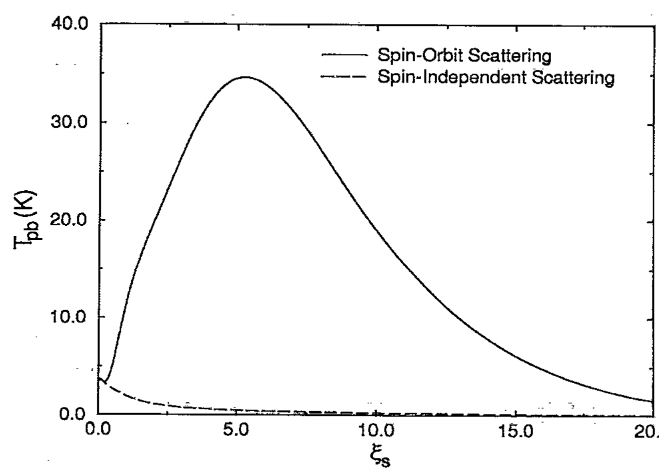

FIG. 2. Pair-breaking temperature, $T_{\rm pb} = 2\gamma/\pi\tau_{\rm pb}$, characterizing the pair-breaking effectiveness of spin-orbit scattering (solid line) and spin-independent scattering (dashed line) for a $d_{x^2-y^2}$ superconductor in a disordered tetragonal phase, plotted as a function of the structural coherence length of that phase, ξ_s. The parameter values used are $\mu = -0.15t$, $\theta_0 = 0.1$, $\nu = 0.2$, and $\rho \simeq 3.4$. The value of ρ has been chosen so that for uncorrelated disorder ($\xi_s = 0$) spin-orbit and spin-independent scattering are equally effective pair breakers. The enhancement of $T_{\rm pb}$ for spin-orbit scattering is due to the focused scattering across the Fermi surface shown in Fig. 3.

tering matrix element $|v_{\mathbf{k},\mathbf{k}'}|^2$ is large. This region is centered at $\mathbf{k}+\mathbf{Q}$ and has linear dimension ξ_s^{-1}. When ξ_s is large the region does not touch the Fermi surface and spin-orbit scattering is not an effective pair breaker. As ξ_s decreases the region grows, at some point touches the Fermi surface, and electrons begin to be strongly scattered. This "focused" large momentum scattering transfers electrons primarily between regions of the Fermi surface where a $d_{x^2-y^2}$ gap has different parities. As a result the anomalous and normal self-energy contributions to (13) add coherently rather than cancel as they do for a conventional s-wave superconductor.[6] This is what gives rise to strong pair breaking.

To summarize, the scattering of electrons in a Cu-O plane from O displacements perpendicular to that plane has been investigated. The leading order source of this scattering, in powers of displacement size, is spin-orbit coupling. Within the RPA, the spin-orbit scattering vertex is not enhanced by spin fluctuations, unlike scattering from an impurity spin. Also, for a superconductor with a $d_{x^2-y^2}$ gap function, spin-orbit scattering can be a strong pair breaker, particularly in a structurally disordered phase in which CuO_6 octahedra tilt coherently on small length scales, but are completely disordered on longer length scales. It is possible that the LTT and *Pccn* phases of the La-Nd-Sr-Cu-O and La-Ba-Cu-O systems have more structural disorder than the LTO phase, and that pair-breaking effects such as those discussed here are responsible for the observed suppression of superconductivity in these phases. If this is the case, then these observations are strongly suggestive of unconventional pairing and support the d-wave hypothesis of high-T_c superconductivity.

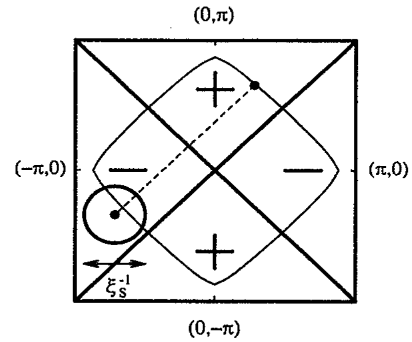

FIG. 3. Brillouin zone for a two-dimensional square lattice showing the tight-binding Fermi surface for 10% doping. The zone is divided into four quadrants marked + or − according to the parity of a $d_{x^2-y^2}$ gap. A typical point on the Fermi surface, $\mathbf{k}$ is marked with a black dot, as well as the point $\mathbf{k}+(\pi,\pi)$ where the initial electron would be Bragg scattered by a coherent staggered distortion. The circle surrounding the shifted point contains the region within which elastic spin-orbit scattering is strongest in a disordered tetragonal phase with a structural correlation length ξ_s. Because electrons are scattered most strongly across the Fermi surface from regions where the gap is positive to regions where it is negative this type of scattering is a particularly effective pair breaker for a $d_{x^2-y^2}$ superconductor.

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