# Positron annihilation from nearly localized Fermi liquids: A probe of pairing

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Based on a theory of positron annihilation from systems with strongly correlated electrons, it is predicted that positron-annihilation spectroscopy (PAS) can be an extremely useful probe of pairing and superconductivity in such systems. Specifically, it is shown that if the nearly localized electrons form pairs which are small, then the annihilation characteristics are changed significantly from those in the unpaired state, and the changes reflect both the amplitude and the *symmetry* of the order parameter. This has obvious consequences for the high-temperature superconductors which have small coherence lengths and where, it has been argued, the Hubbard-model description may be appropriate.

Positron-annihilation spectroscopy (PAS) has long been recognized as a powerful tool for studying the electronic properties of metals.[1,2] In recent years, PAS has been applied to the study of the $CuO_2$-based superconductors.[3,4] These are systems which are believed to fall within the general category of *nearly localized* Fermi liquids.[5,6]

PAS studies of the new superconductors yielded a few surprises. It was observed[3] that, unlike in earlier superconductors, the positron lifetime changes on going through the superconducting transition. The nature of this change was found to depend on the purity of the samples, but the lifetime in the normal and superconducting states were always found to differ significantly. Theoretical studies of positron annihilation have been mostly focused on an electron gas.[7–9] One of the predictions of these theories is the insensitivity of PAS to the BCS (Ref. 10) superconducting transition.[11] This could lead one to speculate that the origin of effects observed in the oxide superconductors lie in the intrinsically different response of positrons to pairing in a system of nearly localized fermions as compared to a weakly interacting Fermi liquid.

It is not difficult to envisage a simple physical system in which positrons may respond to pairing. One could imagine a solid made up of two different types of sites with a small concentration of one type ("hole" sites) less attractive to the positron than the other, and distributed randomly among the others. The electrons that the positron annihilate with are imagined to be tightly bound to these two types of sites. Because the hole sites are less attractive to the positron, the overlap with the electrons around "hole" sites is less than that for the filled sites. As one increases the concentration of holes, therefore, the total electron-positron overlap decreases. Keeping the hole concentration fixed, one could change the nature of the *distribution* from a random distribution of *single* holes to *pairs* of holes. The electron-positron overlap is still affected, because the positron would now be kept out of a larger region containing more tightly bound electrons. This is due to the difference between the repulsive potential of a *pair of holes* as compared to two isolated holes.[12] The situation is very similar to the problem of a divacancy versus a vacancy. Further, the pair can have different shapes and the overlap would reflect the shape of this pair. The electron-positron overlap would then be a sensitive probe of the *symmetry* and *amplitude* of the pairing of these holes.

Approximately, this qualitative picture emerges from a model of "strongly correlated" electrons, and, in this work, the positron-annihilation characteristics of this model are calculated and shown to have essentially the same features as discussed above.

The most important prediction of the present study is that PAS would provide detailed information about the nature of pairing in the high-temperature superconductors (cf. Fig. 1) if indeed, they fall within the category of strongly correlated systems; if not, then PAS should not exhibit any dramatic changes on going through the superconducting transition. Current experiments[3,4] indicate that changes do occur but more detailed experimental studies have to be carried out before PAS can be used to make any definite statements regarding the nature of the oxide superconductors.

A positron interacts strongly with the electrons of the system being studied. It is, therefore, crucial to take into account the effects of electron-positron correlation on the annihilation characteristics. There is an extensive literature on the subject of electron-positron interaction in a homogeneous *electron gas* and its effects on PAS.[7–9] These studies have also been extended to inhomogeneous systems within a local-density-functional (LDF) formalism.[13,14] In an electron gas, one is concerned with the interaction of a *delocalized* positron with a *delocalized* electron. In contrast, in the nearly localized electron systems, one is closer to the atomic limit and the electron-positron interaction is better described as a short-ranged real-space interaction. The treatment of this problem is beyond the realm of applicability of the LDF formalism.

In the present work, the $t$-$J$ model[5,6] is adopted for describing the dynamics of the strongly correlated valence electrons. These electrons make up only a small

 

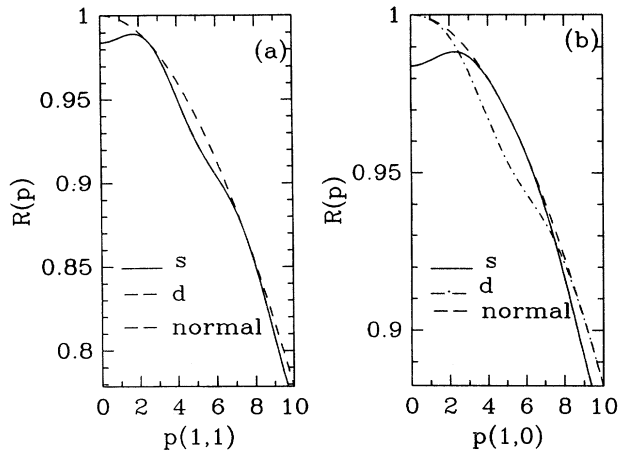


FIG. 1. Plots of $R(\mathbf{p})$ [coming from *all* the core electrons (cf. text) which dominate the electron-positron overlap] in the superconducting state ($s$- or $d$-wave pairing) for a square lattice. The parameters entering the calculation are chosen to be typical of the $CuO_2$-based superconductors; $\Delta=0.10$, $p_F$ (Fermi momentum of holes)$=\pi/5a$, where $a$ is the lattice constant, $\delta=0.10$. (a) depicts $R(\mathbf{p})$ along the (1,1) direction and $p$ is in units of $p_F$. (b) $R(\mathbf{p})$ along (1,0). $R_N(p)$ is taken to be Gaussian with half-width equal to $2/a$.

fraction of the total number of electrons is systems like the $CuO_2$-based superconductors. The remaining electrons are relevant for positron annihilation since they dominate the total electron-positron overlap. In the present model, it is assumed that the relevant time scale of motion of these electrons is much longer than the positron or the valence electrons, and are treated as frozen core-like electrons. The interaction of the positron with these core electrons can be incorporated into a periodic potential. The interaction of the positron with the Hubbard electrons is modeled by a short-ranged potential. The nature of the pairing interaction between the electrons is left largely unspecified in this model. Various mechanisms for pairing within the context of a Hubbard model have been suggested[15] and different scenarios will be considered.

There have been a number of studies of various aspects of the $t$-$J$ model[16] and only the features relevant to the study of PAS will be noted here. The $t$-$J$ Hamiltonian acts on a subspace of states with no doubly occupied sites. Close to the half-filled limit, the $t$-$J$ model describes a few *holes* or *empty sites* propagating in the presence of a lattice of spins. The description of holes or empty sites as particles is a valid concept only in the nearly localized regime where the no-double-occupancy constraint is enforced. It does not carry over to a noninteracting gas of electrons.[5] In the slave-fermion representations,[17–20] the spins and holes are represented by bosons and fermions, respectively, via a formal transformation of the electron operators: $c_{i\sigma}^{\dagger}=f_i b_{i\sigma}^{\dagger}$. Here $f_i^{\dagger}$ keeps tracks of the holes, and $b_{i\sigma}^{\dagger}$ keeps track of the spins, and there is a no-double-occupancy constraint enforced through

$$f_i^{\dagger}f_i+\sum_{\sigma} b_{i\sigma}^{\dagger}b_{i\sigma}=1 \ .$$

In the half-filled limit, there are no holes and the positron moves in a periodic potential. The effects of the interaction of the positron with the "core" electrons is incorporated into this potential. The residual interaction to be taken into account away from half-filling is that between the *positron* and the *holes* or empty sites. This is a *repulsive* interaction which has the effect of keeping the positrons away from the empty sites and leads to an "anticorrelation" between the holes and the positron. It should be stressed that this is a transformation, dictated by convenience, of the original electron-positron interaction problem to one that is more natural in the nearly localized regime.

In considering positron annihilation, the spins can be ignored except for their role in renormalizing the hole motion. In mean-field treatments,[17,20] the effective hopping amplitude $\bar{t}$ is found to be proportional to the hole concentration for small doping. A model Hamiltonian describing the holes and the positron can be written as

$$H=\bar{t}\sum_{\langle ij\rangle} f_i^{\dagger}f_j+\sum_{i,j} T_{ij}d_i^{\dagger}d_j+V\sum_i N_i n_i \ . \tag{1}$$

Here $n_i=f_i^{\dagger}f_i$ and $N_i=d_i^{\dagger}d_i$ are the hole and positron density, respectively. Wannier orbitals have been used as the single-particle basis set for holes and positrons, with the caveat that the positron Wannier orbitals are centered, not at an atomic site, but at an interstitial site. The label $i$ can therefore be thought of as labeling a unit cell centered on an atomic site. The last term in Eq. (1), describing the "on-site" repulsion between the holes and the positrons, is the interaction term that can give rise to interesting effects in PAS.

The central quantity in PAS is the electron-positron pair momentum distribution function, $R(\mathbf{p})$:

$$R(\mathbf{p})=\left\langle \int d\mathbf{r}\int d\mathbf{r}'\exp[-i\mathbf{p}\cdot(\mathbf{r}-\mathbf{r}')] \times\Phi_1^{\dagger}(\mathbf{r}')\Phi_2^{\dagger}(\mathbf{r}')\Phi_1(\mathbf{r})\Phi_2(\mathbf{r})\right\rangle , \tag{2}$$

where $\Phi_1$ and $\Phi_2$ are the electron and positron field operators, respectively. The function $R(\mathbf{p})$ measures the probability of finding an electron-positron pair with a momentum $\mathbf{p}$. The integral of this over all momenta gives the total overlap, the inverse of which determines the positron lifetime.[1,21]

The overlap with the core electrons, i.e., all the electrons except the ones in the $t$-$J$ model, dominates the total overlap since these are the *majority* of electrons. From the physical picture presented earlier, this is expected to be significantly affected by the interaction with the holes, and it is, therefore, sufficient to consider only this part of the overlap in analyzing the effects of pairing on $R(\mathbf{p})$. The calculations presented below will *a posteriori* justify this assumption.

The repulsive term in the Hamiltonian has the effect of keeping a hole and a positron from occupying the same site. This does not lead to any localization effects because the concentration of holes is small, and there is only one positron in the system at a time. Taking a cue from the

variational treatments of the Hubbard model, the calculation of $R(\mathbf{p})$ will be based on a Gutzwiller-type wave function[22] for the hole-positron ground state:

$$\Psi=\Pi_i(1-gn_iN_i)\Psi_0 \ . \tag{3}$$

The state $\Psi_0$ is the ground state in the *absence* of any positron-hole interaction. The correlation factor $g$ is the variational parameter, and serves to reduce the weights of those configurations where the hole and the positron are in the same unit cell. The value of the variational parameter $g$ lies between 0 and 1. The effects due to the pairing of holes can be investigated by allowing for a nonzero pairing order parameter in $\Psi_0$.[23] The evaluation of the core contribution to $R(\mathbf{p})$ [cf. Eq. (2)] involves calculating this function for a *given* configuration of holes, and then averaging over all possible hole configurations.[21] This quenched average is difficult to calculate in general. However, because of the short-ranged nature of the interaction, the possible hole configurations can be divided into two categories, one with no holes occupying the same unity cell as the positron, and the other with a hole and positron in the same unit cell. Then $R(\mathbf{p})$ can be evaluated by considering the two classes separately and adding up their contributions. The projection into the two classes can be conveniently carried out by using the operator $f_i^\dagger f_i$ since

$$f_i^\dagger f_i+\sum_\sigma b_{i\sigma}^\dagger b_{i\sigma}=1 \ .$$

This leads to

$$R(\mathbf{p})=\sum_{i,j}\left\langle \left[f_i^\dagger f_i+\sum_\sigma b_{i\sigma}^\dagger b_{i\sigma}\right]\left[f_j^\dagger f_j+\sum_\sigma b_{j\sigma}^\dagger b_{j\sigma}\right]d_i^\dagger d_j\right\rangle \sum_{\alpha,\beta}\int d\mathbf{r}\int d\mathbf{r}'\exp[-i\mathbf{p}\cdot(\mathbf{r}-\mathbf{r}')]\psi_{i\alpha}(\mathbf{r})\phi_i(\mathbf{r})\psi_{j\beta}^*(\mathbf{r}')\phi_j^*(\mathbf{r}') \ . \tag{4}$$

Here $\phi_i$ and $\psi_{i\alpha}$ are the $e^+$ Wannier state and the one-electron core states, respectively. It has been assumed that only electron and positron wave functions centered in the same unit cell overlap. In the absence of any interaction, the expectation value reduces to $\langle d_i^\dagger d_j\rangle$.

To calculate $R(\mathbf{p})$, the Gutzwiller-type variational wave function [Eq. (3)] is used.[22] This task is further simplified by noting that there are two *small parameters* in the problem, the hole concentration $\delta$ and the positron density $n_0$. Keeping leading terms of the expansion in $\delta$ and $n_0$, the function

$$F(\mathbf{q})=\sum_{i,j}\exp[-i\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j)]\times\left\langle\left[f_i^\dagger f_i+\sum_\sigma b_{i\sigma}^\dagger b_{i\sigma}\right]\left[f_j^\dagger f_j+\sum_\sigma b_{j\sigma}^\dagger b_{j\sigma}\right]d_i^\dagger d_j\right\rangle \tag{5}$$

can be rewritten as

$$\begin{aligned}F(\mathbf{q})&=G^0(\mathbf{q})-\sum_{\mathbf{q}'}\chi(\mathbf{q}')[\Pi^0(\mathbf{q}+\mathbf{q}')-\Pi(\mathbf{q}+\mathbf{q}')]\\&\simeq G^0(\mathbf{q})-\delta(\Pi^0(\mathbf{q}+\mathbf{p}_0)-\Pi(\mathbf{q}+\mathbf{p}_0)) \ .\end{aligned} \tag{6}$$

The functions appearing in Eq. (6), $G^0$, $\chi$, $\Pi^0$, and $\Pi$, are the discrete Fourier transforms of $\langle d_i^\dagger d_j\rangle_0$, $\langle f_i^\dagger f_j\rangle_0$, $\langle f_i^\dagger f_j\rangle_0\langle d_i^\dagger d_j\rangle_0$, and $\langle f_i^\dagger f_j d_i^\dagger d_j\rangle$, respectively.

In writing down the last step of Eq. (6), it has been assumed that the Fermi momentum of the holes, which determines the length scale of $\chi(\mathbf{q})$, is small compared to the scale over which $\Pi(\mathbf{q})-\Pi^0(\mathbf{q})$ varies significantly. Since we are interested in studying the effects of pairing on $\Pi-\Pi^0$, this implies that we are restricting ourselves to pair sizes which are *small* compared to the interhole separation. This is the opposite limit of Cooper pairs where the pairs are large compared to the interparticle separation.

In the normal state, $\Psi_0$ contains no condensate fraction, and therefore there are no anomalous expectation values like $\langle f_i^\dagger f_j^\dagger\rangle_0$. Keeping only the leading terms in an expansion in $\delta$ and $n_0$, $\Pi(\mathbf{q})$ can be evaluated to be

$$\Pi(\mathbf{q})=\frac{\Pi^0(\mathbf{q})}{1+g\Pi^0(\mathbf{q})} \ . \tag{7}$$

It is the approximation of small $\delta$ and $n_0$ that makes it possible to write down the closed-form expression for $\Pi(\mathbf{q})$ and is not possible in general.[22]

The superconducting ground state is characterized by a nonzero expectation value $\langle c_{i\sigma}^\dagger c_{j\sigma'}^\dagger\rangle_0$. In the slave-fermion representation, this would imply nonzero values for both $\langle f_i f_j\rangle$ and $\langle b_{i\sigma}^\dagger b_{j\sigma'}^\dagger\rangle$. The latter is nonzero as long as the ground state has some singlet correlations, and the interesting question to ask is what effect, if any, the pairing of the holes has on the electron-positron pair momentum distribution. Allowing for the appearance of the anomalous expectation values in Eq. (7), and retaining only leading terms in $\delta$, $n_0$, and $\langle f_i f_j\rangle$, leads to

$$\begin{aligned}&[1+g\Pi^0(\mathbf{q})]\Pi(\mathbf{q})=\Pi^0(\mathbf{q})-gS(\mathbf{q})+O(g^2) \ ,\\&S(\mathbf{q})=\sum_{\mathbf{q}'}|\Delta(\mathbf{q}')|^2\,|G^0(\mathbf{q}-\mathbf{q}')|^2\\&\qquad\simeq|\Delta(\mathbf{q})|^2 \ .\end{aligned} \tag{8}$$

Here $\Delta(\mathbf{q})$ is the discrete Fourier transform of $\langle f_i f_j\rangle$, the pairing wave function on a lattice.[10] This modifies the electron-positron pair momentum distribution to

$$R_S(\mathbf{p})=R_N(\mathbf{p})-\delta\left[\frac{gS(\mathbf{p}+\mathbf{p}_0)}{1+g\Pi^0(\mathbf{p}+\mathbf{p}_0)}\right]R_{\text{core}}(\mathbf{p})\ . \tag{9}$$

Here $R_N$ is the distribution in the normal state, $R_S$ in the superconducting state, and

$$R_{\text{core}}(\mathbf{p})=\left|\sum_\alpha \int d\mathbf{r}\exp(-i\mathbf{p}\cdot\mathbf{r})\psi_\alpha(\mathbf{r})\phi(\mathbf{r})\right|^2 , \tag{10}$$

where the forms $\psi_{i\alpha}=\psi_\alpha(\mathbf{r}-\mathbf{R}_i)$ and $\phi_i=\phi(\mathbf{r}-\mathbf{R}_i)$, have been used, and $\phi(\mathbf{r})$ is the wave function of the orbital making up the lowest-lying positron band.

It is clear from the above equation that the pair momentum distribution coming from the overlap of the positron with the tightly bound electrons, which is the *dominant* part of the overlap, is modified by the *real-space pairing of holes,* and that it reflects the symmetry of the pairing. Since this is the main conclusion of this work, it is worth reviewing the assumptions which went into deriving it. The main assumption was that the pairs are small. This implies that $|\Delta(q)|^2$ has to vary over length scales large compared to the Fermi momentum of the holes, since, otherwise, the convolution in Eq. (6) would smear out the effect. One therefore needs both (a) strong correlations under which the electron-positron interaction can be transformed to a hole position interaction, and (b) short-ranged pairs. The extreme short-ranged pairing limit is nearest-neighbor pairing on the lattice, and results for $R(\mathbf{p})$, calculated for two different symmetries of pairing, are presented in Fig. 1. A square lattice is assumed, which would apply to $CuO_2$ systems if $R(\mathbf{p})$ along the $C$ axis is a constant. The results have been derived within a Gutzwiller approximation but could have been derived using other perturbative techniques. The actual expansion parameter in Eq. (8) is $g\delta$ and the terms left out are of order $g^2\Delta^2\delta$ or smaller. Even for $g\sim 1$, these terms are small compared to the leading ones.

As shown in Fig. 1, a nearest-neighbor pairing with extended $s$-wave symmetry on a square lattice with a lattice constant $a$, which leads to

$$\Delta(\mathbf{p})=\Delta[\cos(p_x a)+\cos(p_y a)]\ ,$$

manifests itself in angular correlation experiments as modulating the isotropic distribution $R_{\text{core}}(\mathbf{p})$. Figure 1(a) shows $R(\mathbf{p})$ along the (1,1) direction; along this same direction, there would be no effect due to pairing if the symmetry of the pairing was extended $d$ wave $[(\cos(p_x a)-\cos(p_y a)]$. Along the (1,0) direction [Fig. 1(b)] both symmetries would modify $R(\mathbf{p})$. This clearly demonstrates that monitoring the change in the angular correlation function as one goes through the superconducting transition temperature would lead to a direct measurement of the symmetry of the order parameter.

The effects occur over large regions in momentum (large compared to $p_F$) and are not expected to be washed out by resolution effects, since typical resolutions are comparable to $p_F$.[24] It should be remarked that any effect expected in a BCS superconductor would occur near $p\sim p_F$.[11]

The positron lifetime measures the inverse of the total overlap and, integrating $R(\mathbf{p})$, it can be easily shown that the temperature dependence of the lifetime measures $\Delta^2(T)$. However, the valence contribution has been totally neglected in this analysis and has to be added to the core contribution before comparing to experiments. The temperature dependence of the lifetime had been obtained previously using qualitative arguments.[12] A different approach has been used in Ref. 25 to understand the origin of this temperature dependence.

It should be stressed that throughout this paper it has been assumed that the positron samples the $CuO_2$ planes. If this is not true, then the application of the present analysis is dubious, since the core electrons probably do not dominate the overlap when the positron is in the chain. There are indications that the positron preferentially samples the chains in $YBa_2Cu_3O_7$ but samples the planes in $La_2CuO_4$-based superconductors. The latter would therefore be the ideal system to investigate.

The amplitude of the effect on $R(\mathbf{p})$ depends on the hole concentration, the amplitude of the pairing wave function $\Delta$, and the variational parameter $g$. In the limit of small $\delta$ and large $V$, and $\bar{t}\sim\delta$, $g=1-T\delta/V$. Here $T$ is the nearest-neighbor hopping amplitude for the positron. Making the assumption that $T$ and $V$ are comparable would lead to $g=1-\delta$, which is not a small number. It is expected that the electron-hole repulsion has to be comparable to the positron bandwidth before any interesting effects occur. The amplitude of the pairing wave function is roughly given by the ratio of the energy gap to the Fermi energy.[10] Since $T_c$ is large and the Fermi energy is small in the oxide superconductors, this is much larger than in conventional superconductors, and is of the order of 0.1. This is the value used in Fig. 1.

In conclusion, it has been shown that, in a model of strongly correlated systems, the positron can be a sensitive probe of electron pairing. The model was constructed to be on the opposite end of the spectrum from a delocalized electron gas to stress the difference between a strongly correlated Hubbard-like system and a system of weakly interacting delocalized electrons. One of the most interesting unresolved issues in the high-temperature superconductors is the nature of pairing and condensation. The present work opens up the possibility of using PAS as probe of the nature of pairing in these systems.

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[1]See articles in *Positron Annihilation,* edited by L. Doriken-VanPratt, M. Doriken, and D. Segers (World Scientific, Singapore, 1988); R. N. West, Adv. Phys. **22**, 263 (1974).

[2]For reviews see, *Positron Solid State Physics,* edited by W. Brandt and A. Dupasquier (North-Holland, Amsterdam, 1983).

[3]E. C. Von Stetten *et al.*, Phys. Rev. Lett. **60**, 2198 (1988); D. R. Harshman *et al.*, Phys. Rev. B **38**, 848 (1988); Y. C. Jean *et al.*, J. Phys. Condensed Mat. **1**, 2989 (1989).

[4]L. C. Smedskjaer *et al.*, Physica C (Amsterdam) **156**, 269

(1988).
[5]C. Gross, R. Joynt, and T. M. Rice, Phys. Rev. B **36**, 381 (1987).
[6]P. W. Anderson, Science **235**, 1196 (1987).
[7]J. P. Carbotte and S. Kahana, Phys. Rev. **139**, A213 (1965); J. P. Carbotte, *Positron Solid State Physics,* Ref. 2.
[8]S. Kahana, Phys. Rev. **129**, 1622 (1963).
[9]J. Arponen and E. Pajanne, Ann. Phys. (N.Y.) **121**, 343 (1979).
[10]J. R. Schrieffer, *Theory of Superconductivity* (Benjamin, Reading, MA, 1964).
[11]R. Benedek and H.-B. Schüttler, Phys. Rev. B **41**, 1789 (1990).
[12]Bulbul Chakraborty, Phys. Rev. B **39**, 215 (1989).
[13]Bulbul Chakraborty, Phys. Rev. B **24**, 7423 (1982).
[14]E. Boronski and R. M. Nieminen, Phys. Rev. B **34**, 3820 (1986).
[15]See, for example, P. A. Lee, Phys. Rev. Lett. **63**, 680 (1989).
[16]See, for example, G. Baskaran, Z. Zou, and P. W. Anderson, Solid State Commun. **63**, 973 (1987).
[17]C. L. Kane, P. A. Lee, T. K. Ng, B. Chakraborty, and N. Read, Phys. Rev. B **41**, 2653 (1990).
[18]D. Yoshioka, J. Phys. Soc. Jpn. **58**, 32 (1989).
[19]K. Flensberg, P. Hedegård, and M. B. Pedersen, Phys. Rev. B **40**, 850 (1989).
[20]C. Jayaprakash, H. R. Krishnamurthy, and S. Sarkar, Phys. Rev. B **40**, 2610 (1989).
[21]In most treatments of $R(\mathbf{p})$, the order of averaging and integration is not important and the averaging is done before the integration. In the present case the order has to be preserved since there can be significant differences between the time scales associated with the dynamics of the holes and the positron.
[22]W. Metzner and D. Vollhardt, Phys. Rev. Lett. **59**, 121 (1987).
[23]One might wonder about the fate of the $t$-$J$-model correlation and the pairing correlation in the state $\Psi$. It can be argued that these remain essentially the same as in $\Psi_0$, provided that the positron kinetic energy $T$ is small compared to $V$ and the Hubbard $U$. If $T$ becomes the largest energy scale in the system then the whole model breaks down.
[24]Typical experimental angular resolution is $\sim 0.5$ mrad$\times 0.5$ mrad. The $p_F$ chosen corresponds to 0.6 mrad.
[25]T. McMullen, Phys. Rev. B **41**, 877 (1990).