PROBING THE CuO PLANES WITH POSITRONS IN HIGH $T_c$ CUPRATES:
THEORETICAL PREDICTIONS

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**ABSTRACT:** Positron annihilation spectroscopy is a useful tool to investigate the Fermi surface in
high $T_c$ superconductors. To study the physics of the copper-oxygen subsystem that forms the Cu-O
layers, it is important to provide theoretical predictions, on materials where there is a large overlap
between the positron and the interesting Cu-O planes. We have performed first-principle electronic
structure calculations obtained using the linear muffin-tin orbital and the full-potential linearized
augmented plane wave methods. The positron charge distributions and their sensitivity to different
potentials are calculated. Secondly, we have computed the annihilation rates and the electron-positron
momentum density in order to give predictions of the Fermi surface signals.

As superconductivity in copper oxides is thought to originate in the Cu-O planes, it is
interesting for positron annihilation to turn towards the study of materials where there is
a non negligible overlap between the positron density distribution and the interesting Cu-O
planes. Using the positron as a probe, one could test the validity of the traditional Fermi liquid
superconductivity theory in these compounds, by searching for the Fermi surface (FS) signals
in the two-photon momentum density (TPMD).

The determination of electron and positron states in solids is possible on the basis of the
Two-component Density Functional Theory (TDFT) [1, 2]. The ground state electron and
positron density $n^-$ and $n^+$ are calculated solving a set of one-particle Schrödinger equations
for electrons and positrons. Our calculations have been carried out using both the FLAPW
and the LMTO methods in the Local Density Approximation (LDA). The potential for the positron
is calculated by taking the electron screening into account as a lowering of the electrostatic
energy (correlation potential), which in a given point depends on the electron density at that
point only. We have used the LDA electron-positron correlation potential parametrized by
Boronski and Nieminen [1].

The TPMD $\rho^{p\gamma}(\mathbf{p})$, which is discontinuous when the electron bands cross the Fermi level,
is calculated using an enhancement factor $\gamma(n^-(\mathbf{r}))$, describing the local screening in a Local
Density Approximation (LDA) [2, 3]. The 2D-ACAR (2-Dimensional Angular Correlation of the
Annihilation Radiation) experiments measure the projection $N(p_x,p_y) = \int \rho^{p\gamma}(\mathbf{p}) dp_z$ of
the momentum density over a given direction, which in the present case is chosen to be the
c-axis of these materials.

In contrast with Y-Ba-Cu-O and Bi-Sr-Cu-O compounds where positron densities are small
in the Cu-O planes [4], Nd$_2$Cu$_2$O$_4$ [5] and Tl$_2$Ba$_2$CuO$_6$ [6, 7] are good candidates to
investigate the properties of the Cu-O planes by positron annihilation techniques.

In the case of La$_{2-x}$Sr$_x$CuO$_4$, calculations also show a non negligible overlap of the positron
with the Cu-O planes. Moreover, according to McMullen et al. [8], in La$_{2-x}$Sr$_x$CuO$_4$ all the
vacancies are only weak positron traps and therefore this material is also a good candidate for
2D-ACAR studies [9].

In the case of HgBa$_2$CuO$_4$ [10, 11], the positron density is largest in the void region located in
the Hg layer. A non-negligible overlap of the positron with the Cu-O planes is found only when the electron-positron correlation potential is included. Without electron-positron correlation potential, the localization of the positron in the Hg layer seems very drastic. The situation is similar to Bi$_2$Sr$_2$CaCu$_2$O$_{8+}$ [4], where the positron is a much better probe of Cu-O sheets when the correlation potential is included, though the weight between Bi-O layers remains substantial. In Y-Ba-Cu-O compounds and in Nd$_2$CeCuO$_4$, the effects of the correlation potential are small and the region sampled by the positron is essentially the same as when the positron-electron correlation potential is neglected. The correlation potential may give some exaggerated charge transfer if the void regions are not well described by the empty spheres of the LMTO methods.

In HgBa$_2$CuO$_4$, using the Jarborg-Singh enhancement factor [3, 12] for core and valence electrons, we obtain a calculated lifetime of 232 ps, when the positron feels only the electrostatic potential, and 200 ps, when the electron-positron correlation potential is included. Different forms of enhancement give smaller variations of the lifetime than the variation of having electron-positron potential included or not. For instance, by using the Boronski-Nieminen [1] enhancement, we obtain 187 ps instead of 200 ps. However, neglecting enhancement as in the independent particle model (IPM) gives a value far off, namely 730 ps. It is our experience [12] from other systems that despite a large difference in lifetimes, the strength of the FS steps in the 2D-ACAR are quite similar, when different enhancements or even the IPM are used. Different forms of enhancements are therefore not important, but the use of electron-positron potential can be important for the FS signals in some cases [4].

The theoretical 2D-ACAR HgBa$_2$CuO$_4$ spectrum along [100], calculated using LMTO, is shown in Fig. 1A. Without the empty spheres contribution the spectrum would be as the dashed line. Therefore the effect of the void regions is to produce a kind of dome feature in the 2D-ACAR. Some defects in real samples could also produce similar features in the experimental spectra.

In order to enhance small signals, we calculate the anisotropy of the 2D-ACAR distribution in the $p_x$, $p_y$ plane, obtained by subtraction of the cylindrical average of the distribution itself. The anisotropy in Fig. 1B is shown by the case of Tl$_2$Ba$_2$CuO$_4$, spectrum calculated using FLAPW: this anisotropy is similar to the La$_2$Sr$_2$CuO$_4$ case [5, 9] and the main features could be described by a linear combination of atomic orbitals-molecular orbital method [13], which considers only the 5 antibonding states formed between the 3d states of Cu and the 2s and 2p states of the surrounding oxygens for an octahedron cluster. Among these antibonding states we distinguish the $t_{2g}$ states from $d_{xz}$, $d_{yz}$, $d_{xy}$ atomic Cu orbitals, and $e_g$ states from $d_{x^2-y^2}$ and $d_{z^2}$. The anisotropy peaks in the diagonal directions shows a major $t_{2g}$ contribution, while the cross-like feature near the origin is due to the $d_{xz}$-$d_{yz}$ states. In this anisotropy, the chemical bonding information overshadows the smaller FS signals. The main difference with respect to YBa$_2$Cu$_3$O$_7$ is that here the wave function effects and FS pieces have the same four-fold symmetry, while the ridge FS in YBa$_2$Cu$_3$O$_7$ has a two-fold symmetry which distinguishes it from important four-fold symmetry wave function effect.

For Tl$_2$Ba$_2$CuO$_4$ there are two bands crossing the Fermi level, a Cu-O derived band centered at the corner X of the Brillouin zone enclosing holes, which produces the barrel FS, and a band with important Ti-O character enclosing electrons, which produces a spheroidal FS centered at $\Gamma$ [6]. The Ti-O band yields states considerably more extended than it would be if it had only Ti-O character: although their largest weight lies in the Ti-O planes, there is also an important weight on the O atom of the Ba layer and on the Cu atom. Thus the states of the Ti-O band overlap the positron fairly well. However the corresponding spheroidal FS is more difficult to resolve in 2D-ACAR spectra than if it were a 2D cylinder.

In order to enhance the FS related features, one applies the Lock-Crisp-West (LCW) folding
procedure to the 2D-ACAR spectrum. If the positron wave function is a constant, the resulting distribution provides a projection of the electron occupation number. In Fig. 2, one can see how the actual distributions differ from this ideal case. For Tl$_2$Ba$_2$CuO$_6$, the two bands crossing the Fermi level are clearly seen by the breaks of the LCW spectrum, however these breaks are strongly modulated by positron wave function effects. The amplitudes of the FS signals amount to 3% of the constant background and should allow for an experimental detection. The Tl-O and Cu-O FS breaks have amplitudes of the same order. In the calculated LCW for Nd$_2$CeCuO$_4$, the FS amplitudes were of the order of 5–4% and the wave function effects were much smaller.

In conclusion, the situation in oxides differs thus sharply from the one in simple metals: the main features are wave function effects with only weak FS signals, whereas in simple metals we see mainly FS effects. However, we predict that in La$_{2-x}$Sr$_x$CuO$_4$, Tl$_2$Ba$_2$CuO$_6$ and mainly in Nd$_{2-x}$Ce$_x$CuO$_4$ the FS signals should allow for experimental detection. In HgBa$_2$CuO$_4$ the FS amplitudes strongly depend on the electron-positron correlation potential. Our calculations describe the positron behavior in the crystal as long as no defects localize the light positive particle. Therefore it would be interesting to study the effects of vacancies [14] on the 2D-ACAR for future positron annihilation studies in high-$T_c$, where oxygen disorder and non stoichiometry is a general feature. The study of the defects itself could be a very interesting topic, as according to Raveu [15], they could play crucial roles in determining the superconducting properties of the copper oxides.

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References

Figure 2: The Tl₂Ba₂CuO₆ LCW spectrum calculated using FLAPW and the Nd₂₋ₓCeₓCuO₄ LCW spectrum calculated using LMTO. The x − y axis units are meV.


