What Can Positrons Contribute to High-T_c Superconductivity? *

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The positron annihilation technique has made well-known contributions to the study of Fermi surfaces in "classical" superconductors, including A15 phases where the definition of the Fermi surface has been questioned on the grounds of Anderson localization. In the case of the superconducting oxides, even more far out models were proposed, which made the clear imaging of the Fermi surface by positrons desirable. The difficulties due to the predicted weakness of the signal, and the large possibility for trapping have now been surmounted and the Fermi surface has been seen; what more can we learn from positrons?

After presenting the actual situation with experiment, we will comment on enhancement and correlation and their effect on ACAR and lifetime studies. Then we explain the picture of Jarlborg and Singh of enhancement, with its recent tests for many substances. We conclude by asking the question of sensitivity of positrons to many-body effects. Ferromagnetism, antiferromagnetism, possibly charge density waves have been seen – superconductors, heavy fermions and spinons-holons would pose a problem.

Stephan Berko (1924–1991) was interested in these problems and knew that better machines and better detectors would open new possibilities.

Key words: Positron annihilation; Angular correlation of annihilation radiation (ACAR); Electron momentum density; High-T_c superconductivity.

2 D-ACAR and Lifetime Effects in High-T_c Superconductors

The work of the Geneva group since ICPA-8 was devoted to an important part to the study of High-T_c Superconductivity. At ICPA-8 we thought we had found the Fermi surface (FS) in $YBa_2Cu_3O_7$ [1]. However, the study of nonmetallic crystals of the same material showed us structures quite similar to the ones observed in the metallic substance, which means that, unlike in ordinary metals, other effects than FS-breaks contribute prominently to the two-particle momentum density (TPMD).

Predictions from band theory (for instance [2], [3], [4]) made it clear that the Fermi breaks would be quite weak compared to statistics. We had, however, big enough crystals and high enough counting rates that we should have seen the FS signal. While our counters were slowly producing gigacounts of ACAR data, we were busy trying to grow better crystals, learning more on the (im)perfection of our crystals from lifetime data and trying to understand the implications of some of the wilder theoretical schemes on the ACAR spectra and Fermi breaks.

The recent Argonne conference on Fermiology brought a turning point. West et al. [5], [6] and Smedskjaer et al. [7] showed their data from untwinned crystals and convinced us that they showed structure related to the chains that break the tetragonal symmetry in metallic $YBa_2Cu_3O_7$. That conference also gave us a good insight into the remarkable potential of photoemession spectroscopy, with a momentum-resolved signal of the discontinuity at the FS and its evolution with temperature upon formation of a superconducting gap.

Since that conference we have made progress also in Geneva. First, we are now also getting evidence for FS-related structure from ACAR in (still very small) untwinned single crystals. For a report on this recent work see [8] and [9].

Second, turning back to our high-statistics data on twinned crystals, we have been able to isolate a signal related to the FS ridge (Figs. 1 D and 1 E). At room temperature (Fig. 1 E), the crossing of two equivalent
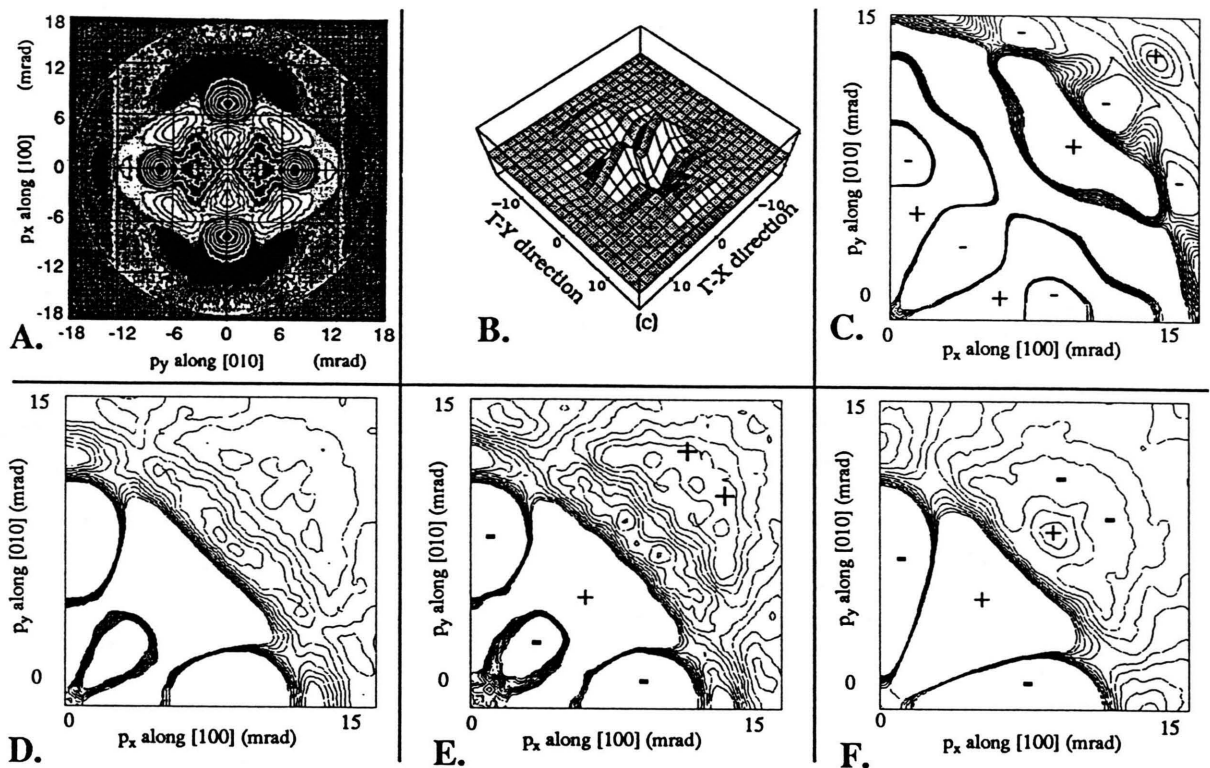
Fig. 1. Anisotropic part of 2D-ACAR in the [001] plane of YBaCuO. A: Haghighi et al. [5], [6], untwinned (UT) and superconducting (SC), B: Smedskajer et al. [7], UT & SC; C: LMTO calculation, twinned (TW) & SC; D: Geneva TW & SC measured at 40 K; E: Geneva TW & SC measured at 300 K; F: Geneva insulating phase measured at 40 K.

ridges is clearly seen as straight lines in the contour plot around $p_x = p_y = 13$ mrad, crossing with a sharp edge. This signature is also present in the low-temperature data (Fig. 1 D), but with less clarity. It should be pointed out that these signals are extremely small: the spacing of the contours is $2 \cdot 10^{-4}$ of the peak intensity. The proof that these signals are due to the FS ridge is given by the insulating single crystal (Fig. 1 F): these data show a minimum instead of the maximum observed for the metallic phase. Another confirmation is given by LMTO calculations (Fig. 1 C): the ridges induce here clear deviations of the contour lines, even if the crossing is not observed, a fact due to the effect of deformation induced by the procedure used to extract the anisotropy of the data, as shown by the model calculation illustrated in Fig. 2: in the case of a single ridge (Fig. 2 A), as in untwinned crystals, a circular shadow is projected over the whole distribution and interferes with the ridges themselves. For two perpendicular ridges (Fig. 2 B), the effect of the pro-

jected shadows is more complex and disturbs the ridges more significantly. In real situations, shadows induced by structures other, and possibly larger, than the ridges, may distort these more severely, making their observation more difficult. In such cases, analyzing the data on the ground of anisotropies may be not suitable, suggesting other methods as derivation or difference between two data sets.

It should be pointed out that the proof of FS signals in data from twinned crystals, we make here, is of considerable importance for three reasons: 1) it proves that not all the positrons are trapped by twin boundaries, 2) having the same symmetry, it makes possible the comparison with data from the insulating phase and 3) it offers the possibility to work with much larger and abundant single crystals.

Theoretically the TPMD spectra can be calculated in the Independent-Particle Model (IPM), where the positron-electron pair wavefunction is just the product of the electron wavefunction times the thermalized
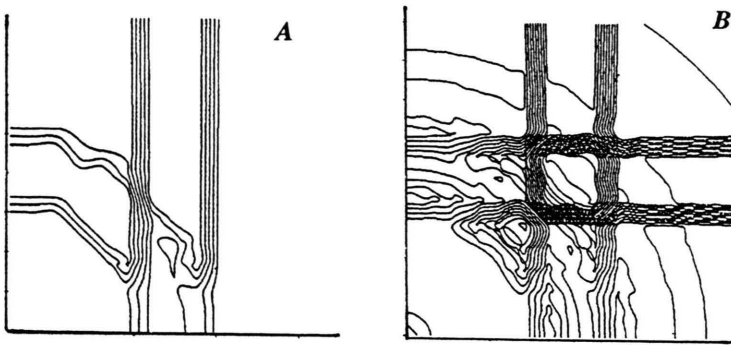
Fig. 2. Effect of taking the anisotropic part of a model distribution by subtracting the circular average at each radius. A: the model distribution consists in one ridge, like in untwinned crystals of $YBa_2Cu_3O_7$; B: model of two ridges as in twinned crystals.

positron wavefunction. Wavefunctions and energies of these particles are calculated using the density functional formalism.

Beyond the IPM approximation, the pair wavefunction (for $r_p = r_e$) may be expressed by means of the electron and positron wavefunctions times an enhancement factor that includes the polarization cloud around the positron. The enhancement is not very important in the LCW space but gives sizable effects in ACAR distributions and is very important for positron lifetimes.

For a more quantitative discussion it is good to remember the role of the two-particle density, expressed in terms of annihilation operators ψ:

$$P(1,2) = \langle |\psi_+^*(1)\psi_-^*(2)\psi_-(2)\psi_+(1)| \rangle. \quad (1)$$

The use that is made of this function by different authors in the analysis of correlations in positron annihilation has been recently reviewed by Barbiellini [10], and we follow his notation.

The part of $P(1,2)$ due to correlations, $C(1,2)$, is put in evidence by

$$P(1,2) = n^+(1)n^-(2)(1 + C(1,2))$$
$$= n^+(1)n^-(2)g(1,2). \quad (2)$$

From there is defined the $e^+ - e^-$ correlation potential energy

$$V_c(r_1) = -\int_0^1 dz \int \frac{e^2 n(r_2) C(r_1, r_2; z)}{2|r_1 - r_2|} dr_2, \quad (3)$$

where z is the coupling parameter, and the Feynman-Hellmann theorem has been used (see [11]). In order to calculate the wavefunction of the positron, one uses the same potential as was used for the electrons, except for the opposite sign.

The annihilation rate for the two-gamma process becomes

$$\lambda = \pi r_e^2 c \int P_+(r, r) dr. \quad (4)$$

The two-particle density is parametrized by $\gamma(r) = g(r, r)$, and this enhancement function can take quite large values.

In addition to the total annihilation rate, we measure the momentum density   (5)

$$\varrho(p) = \delta(p - k - G) \sum_i |\int \sqrt{\gamma(i, r)} \psi_+(r) \psi_{-,i}(r) e^{ipr} dr|^2,$$

and this time it is expressed by the positron wavefunction $\psi_+(r)$ and the enhancement $\gamma(i, r)$ of the density of individual electronic states $\psi_{-,i}(r)$ on the positron site.

Recently several calculations of positron lifetimes in many solids have been performed. The results obtained by Barbiellini et al. [12] are compatible with the calculations of Jensen [13], Puska [14] and of Sterne and Kaiser [15]. Jensen, Puska as well as Sterne and Kaiser use the local density enhancement factor derived from Fermi liquid theory parametrized by Boronski and Nieminen [16]. Barbiellini et al. [12] based their calculations on the local-density approach of Jarlborg and Singh [17]. In this quite intuitive method, the enhancement factor is obtained by solving an electron-positron two-body problem within a correlation cell. The enhancement factor at a given point depends only on the electronic density at that point. The resulting enhancement factor resembles the one of Boronski and Nieminen [16], and therefore the results of lifetimes are close to the ones obtained by the other authors and, as seen in Table 1, also from experiment. The local density enhancement works well when the screening cloud around the positron is short-range.

Jarlborg and Singh [17] reduce the many-body problem into a two-body problem. If we consider a

positron at r_p, the complete electron solution comes from a Schrödinger equation like

$$[-\nabla^2 + V_{\text{ext}}(r) + V_H(r) + V_{xc}(r) + 1/(r - r_p)]\,\Psi(r)$$
$$= E\,\Psi(r). \quad (6)$$

This is now separated into two solutions depending on the positron Coulomb potential. One $\Psi_1(r)$ is valid for $|r - r_p| \geqq r_s$ and one $\Psi_2(r)$ is valid for $|r - r_p| \leqq r_s$, where r_s is essentially the electron-gas parameter,

$$[-\nabla^2 + V_{\text{ext}}(r) + V_H(r) + V_{xc}(r)]\,\Psi_1(r) = E_1\,\Psi_1(r), \quad (7)$$

$$[-\nabla^2 + 1/(r - r_p)]\,\Psi_2(r) = E_2\,\Psi_2(r). \quad (8)$$

The first of these two equations is the normal LDA band problem for the electron without positron, whereas the second could equally well describe a positron around a fixed electron. For a reduced mass $1/2$ the solution $\Psi_2(r)$ gives a cusp for $r = r_p$ that is the enhancement factor. It can also be used to derive a correlation potential for the positron state.

This incomplete separation into two independent problems works best for high electron densities, since r_s is then small compared to the size of an atom. For low-density systems, different methods have to introduce some coupling of the two equations in order to approach the original one, or to use a perturbational approach. Via the former original equation one should obtain the modification of bands and electron states owing to a localized positron, and in turn lattice relaxations appear in the modified $V_{\text{ext}}(r)$. Such calculations are not extensively used, while different perturbative approaches using lattice Bloch functions have been tried, for example: Sormann and Puff [18], Boronski [19] and Jarlborg et al. [20]. However, smooth Bloch functions are not practical functions to describe a localized cusp, and convergence problems may occur in high-density regions. The result of a calculation based on this method can be expressed as an enhancement factor γ and compared to the results based on correlations in the electron gas. For a reduced mass $\mu = 1/2$ there is good agreement, as seen in Figure 3.

When comparing calculations with experiment, some authors introduce an enhancement factor with an energy- or momentum-dependence. (Sormann and Puff [18], Daniuk, Kontrym-Sznajd, Rubaszek, Stachowiak, Mayers, Walters, and West [21] and Jarlborg et al. [20]). Genoud [22] has determined energy-dependent enhancement factors for a number of transition metals.
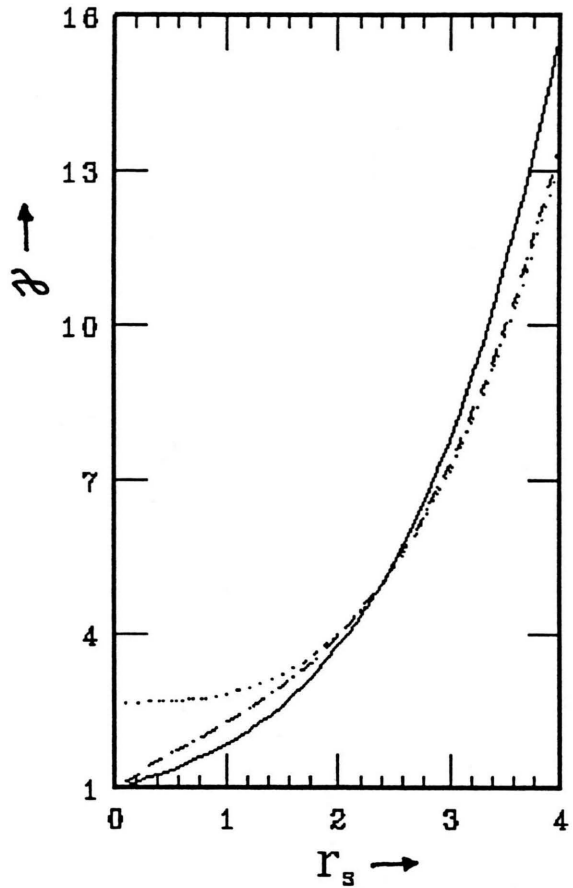
Fig. 3. Enhancement factors. Short-dashed line: Brandt-Reinheimer [38], dashed line: Boronski-Niemenen [16], solid line: Jarlborg-Singh [17].

Table 1. Positron life-times (in ps) calculated by different methods.

Material	Cs	V	Pd	Pt	γ-Ce
IPM	5,466	359	301	273	861
Brandt-Reinheimer (1971)	401	106	92	85	196
Boronski-Nieminen (1985)	394	115	102	94	200
Jarlborg-Singh (1987)	246	128	115	107	208
Jarlborg-Singh + V_{corr}	245	126	113	105	204
Experiment	418	130	96	99	235

The method of Jarlborg and Singh [17] has also been used to provide enhancement factors for lifetime and ACAR in high-T_c superconductors, particularly YBaCuO. The advent of high-T_c has set in motion a search for a new, more exotic description of what was traditionally called the conduction electrons. It was

felt that strong correlative forces must be at work and that under the influence of these forces not only would there occur the high superconducting transition temperature, but that even in the "normal" state the charge carriers would in fact be complicated systems moving in highly correlated ways. While searching for the FS – which proved (and in part still proves) to be difficult to see – it was, and still is natural to wonder what the influence of strong correlations would be on the EMD. The relative mastery in calculating correlation and enhancement effects in ACAR and lifetime with the methods outlined above encourages us to study with the conventional density functional formalism the nature of the electronic structure of the superconducting oxides.

Effect of Magnetic Field

The effect of a high magnetic field on a 2 D electron gas has been well studied, and we all remember the drawing where the Fermi sphere surrounds the cylinders which represent the cyclotron orbits in momentum space. Barnes [23] has pointed out that it would be misleading to use this picture to visualize the effect of magnetic field on the EMD. Instead we may start from the free-electron Hamiltonian in Landau gauge,

$$H = \frac{1}{2m}[p^2 + \hbar^2 k_y^2 - 2eB x \hbar k_y + (eB)^2 x^2], \qquad (9)$$

which leads to the wave functions

$$\phi_{n, k_y}(x, y) = e^{(i k_y y)} H_n(x - A) \qquad (10)$$

with offset $A = R_c^2 k_y$, where $R_c^2 = \hbar k/eB$ is the cyclotron radius square and $H_n(x)$ are the harmonic oscillator functions.

Since the harmonic oscillator functions $H_n(x)$ are eigenfunctions of the Fourier Transformation (FT), we get easily to momentum space, but are disappointed to find an EMD with no cylindrical symmetry and no gauge invariance. The problem of symmetrical gauges in the hydrogen atom was recently treated by Lena, Delande and Gay [24]. As Barnes [23] has shown, the problem does not exist for the TPMD, which is the physically observable quantity.

The TPMD is given by

$$\varrho(p_x, p_y) \cong |\int e^{i p_x x} H_n(x) H_0(x - R_c^2 p_y) \, dx|^2, \qquad (11)$$

and therein the positron function of lowest energy is

$$\varphi_{+, 0, k_y} = e^{(-2eB x^2)/\hbar}. \qquad (12)$$

We notice that, while the EMD was dependent on p_y, the TPMD depends on the product of two wavefunctions with a relative displacement proportional to p_y. This displacement restores cylindrical symmetry (the same effect occurs in the Compton effect).

In the end, the cylindrical surface gives way to a density distribution of thickness d_{k_y}, which leads to an angle $\delta \alpha$,

$$\delta \alpha = \lambda_{\text{Compton}}/R_c. \qquad (13)$$

For instance, at 20 Tesla, the spread in angles of annihilation radiation will be of the order of 0.2 mrad, which corresponds to our present experimental resolution at low temperature.

The spread is wide enough so that the different n-states under the Fermi sphere or in the Fermi cylinder are no longer separated clearly. The drop at the Fermi energy will be smeared over $d_{k_y} = 1/R_c$. As we have seen with presently available fields, this smearing stays in the limits of present resolution – with special care, it might be rendered visible.

Bigger fields can be generated by pulsed techniques, but they are unlikely to be practical for observation by positrons. Kittel once called the molecular field "the poor man's high field", and also showed that such fields would act on localized magnetic moments, but not on the motion of conduction electrons. Lately, flux states have been discussed which might produce large effective internal fields, and their observation by positrons might be a welcome addition to the presently used optical methods in the study of symmetry breaking by anyons in high-T_c superconductors. Barnes [23] has discussed the effect of various spinon/holon states on the positronic TPMD. He points out that superconduction ordering would smear out a sharp Fermi surface in this case.

The effect of the exchange splitting has been seen with positron studies in ferromagnetic Fe and Ni. These experiments were first undertaken by Hanna and Preston ([25], followed by Berko [26], Mijnarends [27] and by the Geneva group, which produced a chart of spin polarization versus electron momentum for Ni from 4.4 K up to T_{Curie} [28]).

The spin polarization can be measured, since ^{22}Na emits polarized positrons (parity violation by the neutrino). Since the positron polarization remains essentially intact during the slowdown of the positron in the sample (as recently studied in Fe by Blank, Schimmele and Seeger [29]), and since two-gamma annihilation occurs only from the s-states, the difference be-
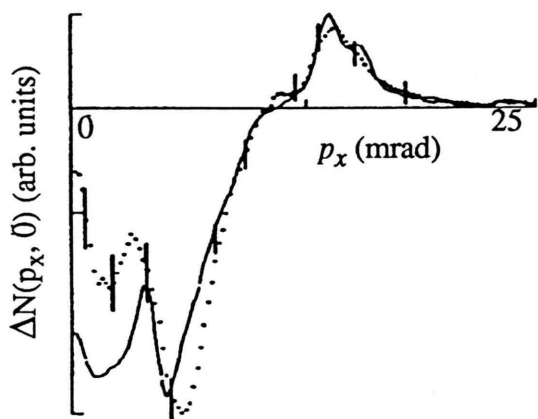
Fig. 4. Relative spin density of Ni along [1 1 0]: experiment (dotted line) and calculation by LMTO method (full line) including parametric enhancement [22].
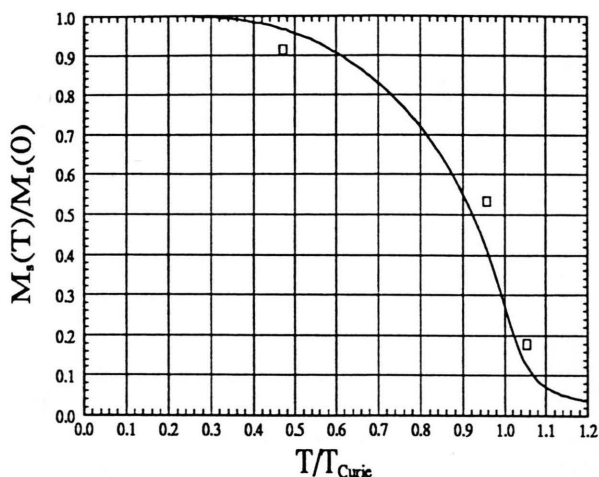

Fig. 6. Schematics of the Photo Emission Spectroscopy (PES) and Positron ACAR methods for a nearly-free-electron model.

Fig. 5. Saturation magnetization of Ni for $J = 1/2$ and $H = 4\,\mathrm{T}$. The squares are relative spin densities [28].

tween spin-up and spin-down can be monitored from reversal of the external applied magnetic field. Genoud et al. [28], see Figs. 4 and 5, found that the polarization varies like the Brillouin function with spin 1/2, for an applied field of 4 Tesla. The shape of the polarization remains stable below T_{Curie}, but at 660 K there is a remarkable change: the remaining polarization seems to be due to the conduction electrons. Is it due to a transition from localized to band magnetism? If so, then positrons would detect an important change of correlations at T_{Curie}.

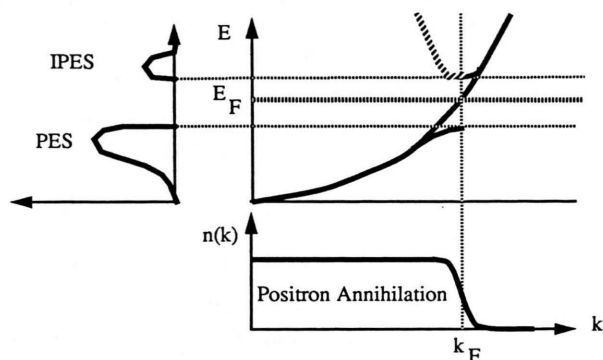ACAR distributions from selfconsistent, spin-polarized LMTO calculations reproduce well the spin-projected momentum densities that are measured at low temperature [22], [28]. The separation between localized and itinerant spin densities is clear and depends only weakly on different enhancement models. It is therefore worth noting that the usual problem of a too large exchange splitting that Local Spin Density (LSD) gives for Ni, does not show up in ACAR calculations, since ACAR is sensitive to densities rather than the splitting. In addition, the delocalized positron makes the negatively polarized itinerant density evident. LMTO results cannot be used directly for comparison with high-temperature measurements, since fluctuating moments are not taken into account. However, rather equilibrated reduction of localized and de-localized density, with increased temperature, seems to support a Stoner picture without fluctuations, at least for not too large T. This appears not to be the situation in Fe, where much larger moments stay localized on each site, even near the Curie temperature [30].

The results on ferromagnetism give us a good indication of the considerable potential of positrons for experimental study of the effects of correlations. But of more immediate interest in high-T_c superconductors are the phenomena of antiferromagnetism, spin- and charge-density waves, and of course superconductivity. So what is the effect of these phenomena on positron annihilation?

The effect of antiferromagnetic ordering has been observed in Cr by Singh, Manuel and Walker [31]. Antiferromagnetic ordering lowers the symmetry of the fcc lattice of Cr into simple cubic symmetry, with the effect that certain levels which are accidentally

degenerated in the paramagnetic state become split by the antiferromagnetic field at low temperature. If these energies lie close to E_F and to the X point, then the transition to paramagnetism might open pockets which are observable by positrons. These effects have been observed in chromium – we do not know yet whether they might be observable in LaCuO, for instance.

The possible effect of charge and spin density waves in high-T_c superconductors has been discussed by Friedel and Peter [32]. The idea, going back already to Berko and Plaskett [33], is that such oscillations might have a wave vector of $2 k_F$, in which case they might open an energy gap at the FS. Such a gap might make the difference between a conductor and an insulator. Also, it could be visible in photoemission experiments. Nevertheless, it might escape detection by ACAR experiment. The point is that detection by photoemission depends on the width of the gap and energy resolution of the spectrometer, whereas detection by ACAR depends on momentum resolution and on the spread in momentum of the wave functions near the induced gap (see Figure 6). This smearing is determined by the perturbing potential E_g. Instead of the sharp Fermi break, we get a break of angular width d_a of order

$$d_a = E_g/(m c v_F), \qquad (14)$$

which might well escape detection ($E_g = 0.1$ eV and $m c v_F = 500$ eV gives $d_a = 0.2$ mrad, close to our present experimental resolution of ACAR at low temperature). On the other hand, E_g is well above spectroscopic resolution. For the case of antiferromagnetic Cr mentioned above, the estimation is very similar; detection is, however, easier since E_g is of the order of 1 eV.

The case of positrons in "classical" superconductors was discussed by Barnes and Peter [34]. This time, we obtain, for the angular resolution

$$d_a = \Delta/(m c v_F), \qquad (15)$$

and hence, for high T_c, numerically similar values as above. The estimate depends on the size of the gap,

which is still subject to debate. Information obtained by positrons would be highly momentum-resolved.

In a recent study, Maniv et al. [35] have solved the BCS-equation in a strong magnetic field and found strong quantum oscillations for the energy gap. They have, however, not included the effect of the magnetic field on the electronic spins.

With these examples we hope to have made clear that positrons are in principle sensitive to effects of correlation. In ferro- and antiferromagnetics quite detailed effects have been seen. In high-T_c superconductors we have only just identified the FS, but now the way is open for the search of specific effects such as the smearing of the FS due to a k-dependent energy gap.

A final remark concerns the possibility of improving positron annihilation apparatus, and the consequences of such improvements for the study of many-body effects. In a recent workshop in the Paul Scherrer Institute at Villigen [36], it was proposed that positron flux might be increased by several orders of magnitude over our present laboratory ^{22}Na source, which was 50 mCi when it was new, and which theoretically produced 89% of positrons, $\sim 10^7 \, e^+/s$, resulting for optimal crystal sizes in a coincidence rate of 500/s. This is in contrast, for instance, to the source strength discussed by Mills [37] of about $10^{10} \, e^+/s$, which would give a much higher counting rate, but which could still be handled by present detectors. Part of this improvement would certainly be used to shorten the measuring time: present times are uncomfortably long with respect to expected sample stability. However, part of it could be traded for resolution – in the case of 2D compounds linear resolution. Obviously, focussing in energy and space will open the possibility of using smaller (and often better) samples and of using depth profiling, but in the context of learning about many-body effects angular resolution is paramount. We have seen that for several properties we are with present technology just at the threshold of detectability. Therefore, and given the fact that our sources are not optimized, we should hope for speedy progress in the technology of our experiments.

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