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ON POSITRON ANNIHILATION IN CONCENTRATED RANDOM ALLOYS AND SUPERCONDUCTING CUPRATES

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ABSTRACT

We discuss an application of a generalisation of the Lock-Crisp- West theorem to concentrated random alloys. Using a theory developed for binary random alloys we explore a possibility of positron localisation in the new high temperature superconductors.

CONCENTRATED RANDOM ALLOYS

As the temperature is lowered binary alloys either order or phase separate. A particularly interesting class of ordering processes are those which result in long period (several unit cells), often incommensurate, modulations of the composition. Well known examples of this phenomenon are Cu_cPd_{1-c} and Cu_cAu_{1-c} (c is a concentration), where the ordering transitions are driven by the parallel sheets of the Fermi Surface (FS) /1/.

The most suitable experimental technique for studying such phenomena is the measurement of the angular correlation of positron annihilation radiation (ACAR) using two-dimensional (2D) position sensitive γ-ray detectors /2/. What can be measured in such an experiment is the electron-positron momentum distribution $\rho^{ep}(\vec{p})$, integrated over one variable of $\vec{p}$, the total momentum of annihilating electron-positron pair. A set of breaks occurring in $\rho^{ep}(\vec{p})$ at $\vec{p} = \vec{p}_F$ ($\vec{p}_F$ is a Fermi momentum) defines the FS topology /3/.

Lock-Crisp-West (LCW) theorem /4/ has been extremely useful in recovering the FS information from the 2D ACAR data. It superimposes the data from all over an extended momentum space onto the first Brillouin zone, thus emphasising important features of the FS topology /5/. This theorem is definitely valid for a constant positron wavefunction, as in this case the folded data will be proportional to the occupation number of electronic states. In the case of a realistic positron wavefunction, however, it is valid only approximately, although it still remains a most powerful tool of interpreting the 2D ACAR data, if supported by the first principles calculations of the electron-positron Bloch momentum distributions

$$\rho_B^{ep}(\vec{k}) = \sum_{\vec{G}} \rho^{ep}(\vec{p}) = \sum_{\vec{G}} \rho^{ep}(\vec{k} + \vec{G}),$$

where $\vec{G}$ is a reciprocal lattice vector, and $\vec{k}$ is restricted to the first Brillouin zone.

A formalism for calculating the configurationally averaged electron and electron-positron momentum densities, $\bar{\rho}^{e}(\vec{p})$ and $\bar{\rho}^{ep}(\vec{p})$, for binary random alloys within the Korringa-Kohn-Rostoker coherent-potential-approximation (KKR-CPA) band theory method /6/ and the independent particle model has been developed in /3/. Using the main results of /3/, the formulae for evaluating the configurationally averaged electron and electron-positron Bloch momentum densities, $\bar{\rho}_B^{e}(\vec{k})$ and $\bar{\rho}_B^{ep}(\vec{k})$, have been derived in /5/. In contrast to momentum distributions, Bloch momentum distributions could be decomposed according to angular momentum, so one can study s, p and d electron contributions separately. In particular, the effect of the positron wavefunction, modulating those contributions

in various ways can be investigated in detail. As an example, we show in Fig. 1 the configurationally averaged electron and electron-positron Bloch momentum densities in $Cu_{60}Ni_{40}$ random alloy, along the (001) direction.

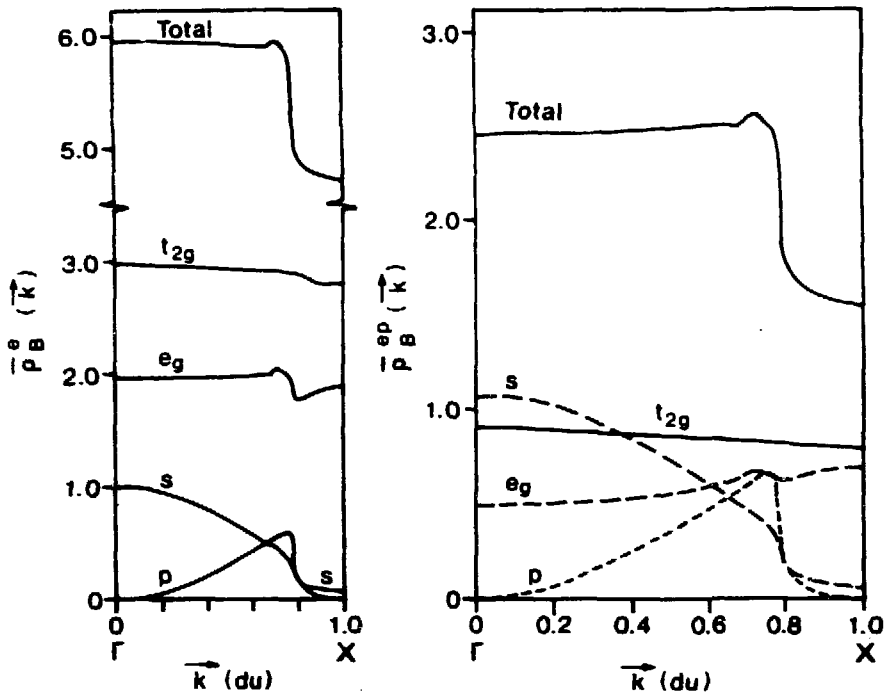

Fig. 1. The distributions $\bar{\rho}_B^e(\vec{k})$ and $\bar{\rho}_B^{ep}(\vec{k})$ along the (001) direction in $Cu_{60}Ni_{40}$ and their decomposition into s(ℓ = 0, m = 0; where ℓ and m are orbital and magnetic quantum numbers, respectively), p(ℓ = 1, m = -1, 0, +1), t_{2g}(ℓ = 2, m = -2, -1, 1), e_g(ℓ = 2, m = 0, 2) contributions.

We see from Fig. 1 that the break in $\bar{\rho}_B^e(\vec{k})$ at $|\vec{k}|$ = 0.78 du (dimensionless units) is coming mostly from the p electrons, but is also reflected in the s and e_g channels. In $\bar{\rho}_B^{ep}(\vec{k})$ the break is reduced due to a substantial decrease of the d(t_{2g} and e_g) contributions. The break in the e_g channel has almost disappeared, whilst the p contribution to the break has slightly increased. In general, the effect of the positron wavefunction is to enhance s and p contributions and to reduce the d contributions. Moreover, one can see that the reduction in the t_{2g} channel is much larger than in the e_g channel. Also, the t_{2g} curves show much less structure than the e_g ones. Especially, the hump around the Fermi momentum is very well reflected in the e_g channel, and is associated with the peculiar behaviour of 'Δ_2^{Ni} band' in the $Cu_{60}Ni_{40}$ system.

In conclusion, we would like to stress that apart from containing more physical information, $\rho_B^{ep}(\vec{k})$ is much easier to evaluate than $\bar{\rho}^{ep}(\vec{p})$ due to the matrix elements which do not depend explicitly on momentum. Owing to that it is now feasible to produce contour plots of the theoretical 2D ACAR surfaces for disordered alloys and to stimulate more interest in studying these systems using 2D ACAR.

SUPERCONDUCTING CUPRATES

There is considerable current interest in probing the electronic structure of the new high temperature superconductors by positron annihilation studies. Potentially, there are two types of questions which positron annihilation experiments are uniquely well placed to explore. The first concerns the number of oxygen vacancies under a variety of equilibrium and non-equilibrium conditions. This quantity appears to be one of the principle factors which must be monitored during the preparation of these new superconductors, as well as playing a decisive role in determining the number of carriers involved in the superconductivity. The second type of questions, to which positron annihilation experiments may be alone in being able to provide an answer, are those related to the FS. Evidently, whether the FS exists and, if it does, what its shape is must be regarded as central to the nature of the new superconductors.

Clearly, the answer to the questions we pose is strongly dependent on the possibility of a positron localisation by the oxygen vacancy. No trapping is good news for the FS studies, but it means that oxygen vacancies are difficult to detect. On the other hand, if positrons are easily localised, and hence the vacancies can be readily detected, the FS work becomes problematic. Given this situation, our principle aim is to investigate whether or not the positron can get localised by the oxygen vacancy.

The question of positron localisation by oxygen vacancies is not as straightforward as that for vacancies in metals. It has to do with the oxygen being a negative, O^{2-}, ion which for the positron at a distance is strongly attractive. Thus, while the lack of a positive nuclei at the centre of a vacancy is attractive for a positron, the competing attraction of the more numerous negative ions elsewhere may prevent the vacancy

binding the positron. To explore the validity of this observation we shall regard the oxygens and the vacancies in our material as two components of a more or less random alloy and use the principles governing the localisation of positrons in binary alloys. A theory of the conditions necessary for positron trapping in such systems, based on the CPA /6/, has been developed by Szotek et al in /3/, and we shall use their main results as the basis for our discussion.

To state these results succinctly, consider a lattice of A and B sites and assume that the positron hops from site to site with an amplitude W_p, the band width, and that it has energy E_A at an A site and E_B at a B site. As was stressed in /3/, the implication of the above general prediction of the CPA for a positron is that for $E_A - E_B >> W_p$ it will be more or less trapped at an A site and will annihilate preferentially with the electrons which form the electron density at the A sites.

The next step in our argument is to identify the A and B sites of the above discussion with oxygens and vacancies in the $YBa_2Cu_3O_{6+x}$ compounds (x changes between 0 and 1). We pick a chain oxygen, O^{III}, in $YBa_2Cu_3O_7$ compound to be A, and its absence, V^{III}, to be B (see /7/ for details). Thus $YBa_2Cu_3O_6$ is a pure 'vacancy' compound, and $YBa_2Cu_3O_7$ is the pure 'oxygen' system. Hence, the compounds for intermediate values of x may be regarded as alloys of oxygens and vacancies on the sublattice composed of III-sites.

Given the above model we can determine the site energies $E_A = E_{O^{III}}$ and $E_B = E_{V^{III}}$ by calculating the low lying energy eigenstates of a positron in the two pure compounds, $YBa_2Cu_3O_6$ and $YBa_2Cu_3O_7$, and locating the mid points of the lowest bands. For this we have performed the separate LMTO-ASA bandstructure calculations for $YBa_2Cu_3O_6$ and $YBa_2Cu_3O_7$ compounds, with the positron potentials constructed on the basis of the self-consistent electronic charge distributions obtained using the LMTO-ASA method /7/. Surprisingly, the $YBa_2Cu_3O_6$ and $YBa_2Cu_3O_7$ positron bands are very close. Taking $\Delta E = E_{O^{III}} - E_{V^{III}}$ to be the difference between the bottoms of the bands at the Γ point we find $\Delta E = 0.242$ eV. As we have argued above, this is to be compared to the band width W_p. We find the bands to be very anisotropic, however the band widths for various

possible symmetry directions of the simple tetragonal structure are very much the same for both compounds. In the language of tight binding Hamiltonians this means that there is no off-diagonal randomness. Consequently, our arguments apply without modification. Thus comparing ΔE to the band widths in directions Γ-X, Γ-M (basal plane) and Γ-Z (c axis) we conclude that while the positron may not be able to move in the Γ-Z direction ($\Delta E >> W_p^{\Gamma-Z} \approx 0.1$ eV), it will not be trapped by vacancies in the plane ($\Delta E << W_p^{\Gamma-X} \approx 2.0$ eV and $\Delta E << W_p^{\Gamma-M} \approx 3.5$ eV).

In short, the realistic calculations of the positron band structures in $YBa_2Cu_3O_7$ and $YBa_2Cu_3O_6$ strongly suggest that oxygen vacancies do not localise positrons in these compounds. This may be considered to be a consequence of the fact that the overall repulsion of positrons by the nuclei and attraction by the electrons of negative ions effectively cancel.

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