Comment on the Positron Surface-State Lifetime

Recently Lynn, Frieze, and Schultz[1] have succeeded in measuring for the first time the positron lifetime on a well-characterized metal surface. The measured lifetime for the Al(110) surface, 580 ± 10 psec, is clearly longer than the recent theoretical prediction, 400 psec, of Nieminen and Puska.[2] In this Comment we show how to resolve this discrepancy by removing an inconsistency from the theoretical treatment.

In an inhomogeneous electron system the positron lifetime and the electron-positron correlation energy are usually calculated by use of a local-density approximation (LDA). For the annihilation rate this means[3]

$$\lambda = \int d^3r\, |\psi_+(\vec{r})|^2 \lambda_0(n(\vec{r})), \qquad (1)$$

where ψ_+ is the positron wave function and $\lambda_0(n)$ the annihilation rate in a homogeneous electron gas of density n. This approximation leads to good results for positron lifetimes in vacancies and other small open-volume defects.[4] It is also supported by the model calculation of Inglesfield and Stott.[5] They showed explicitly that *inside* a model surface LDA works well as compared to the exact result for the random-phase approximation, both for the annihilation rate and the correlation energy. However, the low-density limit of $\lambda_0(n)$ is 2 nsec^{-1} which gives an upper limit of 500 psec for the lifetime calculated from Eq. (1). An experimental lifetime longer than that, in the absence of long-lived orthopositronium, provides clear evidence for the breakdown of LDA.

Inside a metal the positron-electron correlation energy, which arises from the screening, can be calculated by use of the LDA. Outside the metal surface, however, this cannot be done; in the vacuum far from the surface the positron correlation potential must approach the long-range image potential. The approximation of matching the local correlation energy to the asymptotic image potential was suggested by Hodges and Stott[6] who were the first to predict theoretically the existence of the positron surface state. As the positron far outside the surface feels the full image potential, the screening charge resides in the metal (forming the image), the positron is bare, and the local annihilation rate is zero. Thus the use of LDA for the annihilation rate ($\lambda \sim 2$ nsec^{-1}) far outside the surface is inconsistent with the use of the image potential for the correlation energy ($\lambda \sim 0$). This inconsistency appears in the previous local-density calculations.[2,7] We have now recalculated, using the three-dimensional program described in Ref. 2, the positron lifetimes by assuming that in the region where the image potential is used[2] the annihilation rate is equal to *zero*. The results exceed the local-density upper limit, in agreement with experiment. The calculated values are 630, 659, 602, and 552 psec for Al(110), Al(100), Cu(100), and Cu(110) surfaces, respectively. While the lifetimes are more sensitive than in LDA to the position of the effective image surface, the corrugated-mirror model of Ref. 2 not only gives accurate binding energies but can also predict qualitatively correct long (> 500 psec) lifetimes.

A nonlocal calculation for the positron correlation energy and annihilation rate at a surface has been carrried out by Nieminen and Hodges[8] using the plasmon approximation which gives the correct image potential at large distances. Their result for the surface state lifetime on Al, 540 psec, is again in fair agreement with the experimental result and much larger than the result of LDA (~ 300 psec) applied to the same positron and electron distributions.

In conclusion, when the positron correlation energy and annihilation rate are calculated in a consistent way, the theory explains, at least qualitatively, the lifetime of the positron surface state. A quantitative first-principles calculation, however, still constitutes a challenge to theorists.

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