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SEMIEMPIRICAL CORRELATION OF β^+ ANNIHILATION RATES IN METALS

James A. McHugh

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ABSTRACT

It is shown that the annihilation rates of positrons in metals can be correlated to a good degree by a simple model of the annihilation process based on an assumption that the fractional number of electrons available for annihilation varies as the inverse of the atomic volume of the metal.

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The annihilation rates of positrons in metals has been shown in the work of Bisi et al. to be dependent on electron density.[1] However, theoretical correlations of the annihilation rate with the electron density have not been completely successful in fitting the data.[2,3]

In this paper, a semiempirical relationship between the annihilation rate and the electron density in metals is reported.

The mechanism assumed applicable to the annihilation process in metals is based on a model suggested by Gerholm, in which the formation of positronium and its subsequent interaction with the free electrons of the metal is the basis.[2] The annihilation process, in view of this model, can be thought of as consisting of two competing processes: annihilation with the positronium electron (partial rate λ_o), and annihilation with the free electrons (partial rate λ_r). The total rate λ is then merely the sum of two terms:

$$\lambda = \lambda_o + \lambda_r \, . \tag{1}$$

The rate associated with the positronium electron (λ_o) should be approximately equal to the spin-averaged rate in the limit of rapid singlet-triplet conversion,[2] $\lambda_s/4$, where λ_s is for the unperturbed singlet rate.[3] It is essential to note that λ_o is a constant, independent of electron density, and equal in this approximation for all metals.

The rate associated with the free electrons, using positronium as a reference state, is then equal to $\lambda_s n/4n_s$, where n is the effective electron density encountered by the positron, and n_s is the density of the positronium 1S state electron at the positron. The density n must now be evaluated in terms of some parameters that characterize the various metals.

The total number of electrons per unit volume in a given metal is $NZ\rho/A$, where N, Z, ρ and A are Avogadro's number, atomic number, density, and atomic weight, respectively. The total number is not available for annihilation because the positron is excluded from the vicinity of the inner electron shells by the nuclear field. Therefore, only a fraction F of the total can annihilate. If one observes that the fraction F will in general be small, and that the atomic binding in metals is a measure of the electron density in the interstices and varies in a crude manner as the inverse of the atomic volume V_a, one can, as a first approximation, set $F = C/V_a$ or $C\rho/A$. In order to be dimensionally correct, the proportionality constant C could be associated with a quantity $V-V_c$, the total volume minus a core volume. The core represents the bound electrons or electrons unavailable for annihilation. The partial rate associated with the free electrons can now be written as $(\lambda_s/4n_s)(NCZ\rho^2/A^2)$, or by grouping the constants together as K in the expression for λ_r and substituting into Eq. (1), the total rate becomes

$$\lambda = \lambda_o + KZ\rho^2/A^2.$$

In plotting annihilation rate versus $Z\rho^2/A^2$, a straight line snould result. Fig. 1 shows such a plot.

The intercept λ_o is not equal to the spin-averaged rate[3] (2×10^9 sec^{-1}) as was expected. This suggests, providing the model is correct, that the actual rate in the limit of complete triplet quenching is of the order 3.6×10^9 sec^{-1}.

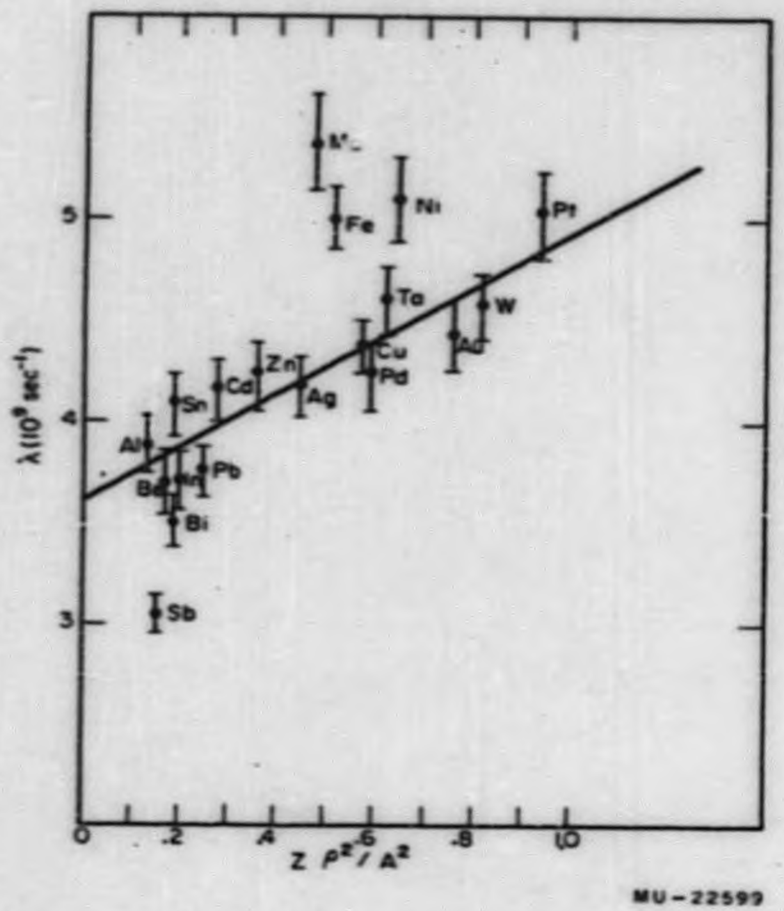

Fig. 1. Positron annihilation rate in metals as a function of $Z \rho^2/A^2$. (Rate data of A. Bisi, et al.[1])

The annihilation rates of Mo, Fe, Ni, and Sb show deviations from the expected rates. It should be noted that Fe and Ni are ferromagnetic and Sb is approaching nonmetallic character. These properties might tend to have a great effect on the rate.

The interesting point of this development is that the rate is proportional to $Z\,\rho^2/A^2$, which is not subject to a parameter defining a fixed number of free electrons, such as the valence. The major assumption open to question is the one of the fraction F being truly proportional to $1/V_a$. In view of the previous arguments and the experimental agreement with the majority of the metals, it seems to be a reasonable assumption. However, the final result should be viewed as semiempirical, in light of our inability to further justify, on a rigorous basis, the interpretation of the electron density.

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