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Positron Diffusion in Solids[*]

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Theoretical positron-lifetime spectra are derived for targets with structures such that positrons can diffuse into interstices where they remain trapped until, after a distinct lifetime, they annihilate with an electron into γ quanta. Exact expressions and convenient approximate formulas for different structural geometries are presented. The contribution of trapped positrons to the angular correlation between the two annihilation γ quanta is derived. The propagation of errors in determining positron-diffusion constants or, conversely, parameters of the target microstructure from positron-annihilation characteristics is discussed.

In a previous paper,[1] a strong dependence of positron-lifetime spectra on the grain size in powders was interpreted in terms of the diffusion of positronium (Ps) atoms from the solid material into the interstices between the grains. For example, in a series of SiO_2 powders of varying grain size (40–300 Å) one observes three lifetime components of intensities I_m, $m = 1, 2, 3$, normalized such that $\sum_m I_m = 1$, with lifetimes τ_m or annihilation rates $\gamma_m \equiv \tau_m^{-1}$, respectively.[2] The intensity I_3 of the component with the longest lifetime ($\tau_3 \simeq 140$ nsec) grows with decreasing grain size at the expense of the intensity I_2 of the component with the intermediate lifetime ($\tau_2 \simeq 2$ nsec). The intensity I_1 of the component with the shortest lifetime ($\tau_1 \simeq 0.4$ nsec) remains essentially constant. I_2 is attributed to the electron pickoff annihilation of orthopositronium (o-Ps: positron and electron spins parallel) in the solid with pickoff rate $\gamma_p \simeq \tau_2^{-1}$. The third component, of intensity I_3, decays with the self-annihilation rate of o-Ps. We attribute I_3 to o-Ps atoms that have diffused, before they could annihilate in the grains by electron pickoff, into the voids where they annihilate at a rate $\gamma_3 < \gamma_p$.[1] I_1 comprises all other annihilations, viz., the annihilation with target electrons of positrons that have not formed Ps and the self-annihilation of parapositronium atoms (p-Ps: positron and electron spins antiparallel) that form concurrently with o-Ps in a proportion given by spin statistics.

Interesting applications of these ideas have been made in various contexts.[3-6] The notion of diffusion and trapping in voids, of course, need not be restricted to Ps. In fact, certain positron-annihilation characteristics in solids can be interpreted in terms of the diffusion of positrons to defects acting as positron traps.[7] Recent experimental studies support this interpretation.[8-10]

We return here to the problem of positron diffusion to derive exact theoretical spectra to be expected under diffusion conditions, and to present the results for different geometries of the target microstructure. In applications to crystal defects the approximate spectrum derived earlier[7] is shown to be adequate for the analysis of positron-lifetime spectra because in such cases the uncertainties in the data cannot distinguish between the exact and the approximate expressions. Optimum experimental conditions can be cited for determining positron or positronium-diffusion constants D or, alternatively, given D, for determining structural parameters of the target material.

Consider a target consisting of small solids (subscript s) interspersed by voids (subscript v). An injected positron is stopped at time $t = 0$ at some point $\vec{r}$ with uniform probability density $\hat{p}(\vec{r}, t = 0)$ throughout the solid matrix, and with zero probability in the voids. If positrons can diffuse in the solid in which they were thermalized, some reach the surface and can escape into the voids where they annihilate with a lifetime τ_v which is long compared to the lifetime τ_s in the solid. In time, $\hat{p}(\vec{r}, t)$ changes as

$$\frac{\partial \hat{p}}{\partial t} = -\gamma_s \hat{p} + D\nabla^2 \hat{p} \ , \tag{1}$$

subject to the condition $\hat{p}(\vec{R}, t) = 0$, where $\vec{R}$ describes the surface $S(\vec{R})$ of the solid of volume $V(\vec{R})$, acting as an absorbing boundary. The fraction of positrons in the solid, $n_s(t)$, is then

$$n_s(t) = \int \hat{p}(\vec{r}, t)\, dV(\vec{R}) \,. \tag{2}$$

With the substitution $\hat{p}(\vec{r}, t) = p(\vec{r}, t) e^{-\gamma_s t}$, Eq. (1) reduces to the usual diffusion equation for stable particles ($\gamma_s = 0$). In terms of the remaining fraction of stable particles,

$$M(t) = \int p(\vec{r}, t)\, dV(\vec{R}) \,, \tag{3}$$

Eq. (2) can be written as $n_s(t) = M(t)e^{-\gamma_s t}$. The rate equation governing $n_s(t)$ becomes

$$\frac{dn_s(t)}{dt} = -\gamma_s n_s(t) + \left(\frac{dM(t)}{dt}\right) e^{-\gamma_s t} \,. \tag{4}$$

The rate equation governing the positron fraction in the voids, $n_v(t)$, annihilating there with rate γ_v, becomes

$$\frac{dn_v(t)}{dt} = -\gamma_v n_v(t) - \frac{dM(t)}{dt} e^{-\gamma_s t} \,, \tag{5}$$

where by Eqs. (1) and (3), in terms of a mean positron-diffusion constant D,

$$\frac{dM(t)}{dt} = D \int \nabla p(\vec{R}, t) \cdot d\vec{S}(\vec{R}) \,, \tag{6}$$

by Gauss's theorem.

The solutions of Eqs. (4) and (5), subject to the initial conditions $n_s(t=0) = 1$ and $n_v(0) = 0$, can be combined into $n(t) = n_s(t) + n_v(t)$ to give the lifetime spectrum

$$n(t) = M(t)\, e^{-\gamma_s t} - \int_0^t dt' \frac{dM(t')}{dt'} e^{-\gamma_s t'} e^{-\gamma_v (t - t')} \,. \tag{7}$$

The function $M(t)$ is known for different geometries of the solids.[11] For foils (dimension $\mu = 1$) of thickness $2R$, long fibers ($\mu = 2$) of radius R, and spheres ($\mu = 3$) of radius R, it is given by

$$M_\mu(t) = 2\mu \sum_{\nu=1}^{\infty} \lambda_{\mu\nu}^{-2} \exp\left[-\left(\frac{D}{R^2}\right) \lambda_{\mu\nu}^2 t\right] , \tag{8}$$

where, depending on the geometry of the solid μ, the constants $\lambda_{\mu\nu}$ take the values

$$\mu = \quad 1\,, \quad 2\,, \quad 3\,,$$

$$\lambda_{\mu\nu} = \pi(\nu - \tfrac{1}{2})\,, \quad j_{0\nu}\,, \quad \pi\nu \,.$$

They fulfill the normalization condition

$$2\mu \sum_{\nu=1}^{\infty} \lambda_{\mu\nu}^{-2} = 1 \,.$$

The constants $j_{0\nu}$ are the solutions of $J_0(x) = 0$, where $J_0(x)$ is the Bessel function of the first kind of order zero; they are tabulated.[12]

Equation (7) with Eq. (8) yields the lifetime spectrum for a target composed of solids of type μ,

$$n_\mu(t) = 2\mu \sum_{\nu=1}^{\infty} \left(\frac{1}{\lambda_{\mu\nu}^2} - \frac{\beta^2}{\lambda_{\mu\nu}^2 \beta^2 + g^2}\right) \times \exp\left[-(\lambda_{\mu\nu}^2 \beta^2 + 1)\gamma_s t\right] + \Phi_\mu(\beta, g) \exp(-\gamma_v t) \,, \tag{9}$$

with the abbreviations

$$\beta^2 \equiv D/\gamma_s R^2 \,, \tag{10}$$

$$g^2 \equiv 1 - \gamma_v/\gamma_s \,. \tag{11}$$

The intensity Φ_μ of the diffusion-controlled lifetime component is given by

$$\Phi_\mu(\beta, g) = 2\mu \sum_{\nu=1}^{\infty} \frac{\beta^2}{\lambda_{\mu\nu}^2 \beta^2 + g^2} \equiv \mu \frac{\beta}{g} \frac{I_{\mu/2}(g/\beta)}{I_{\mu/2-1}(g/\beta)} \ , \tag{12}$$

where $I_n(x)$ is the modified Bessel function of order n. Specifically,

$$\Phi_1(\beta/g) = (\beta/g) \tanh(g/\beta) \quad \text{(foil)}, \tag{13}$$

$$\Phi_2(\beta/g) = \frac{2(\beta/g) I_1(g/\beta)}{I_0(g/\beta)} \quad \text{(fiber)}, \tag{14}$$

$$\Phi_3(\beta/g) = 3(\beta/g)\left[\coth(g/\beta) - (\beta/g)\right] \quad \text{(sphere)}. \tag{15}$$

For $\mu = 3$ and $g = 1$, we retrieve the expression derived by Flügge and Zimens[13] for the escape of emanation from solid spheres in the limit of zero

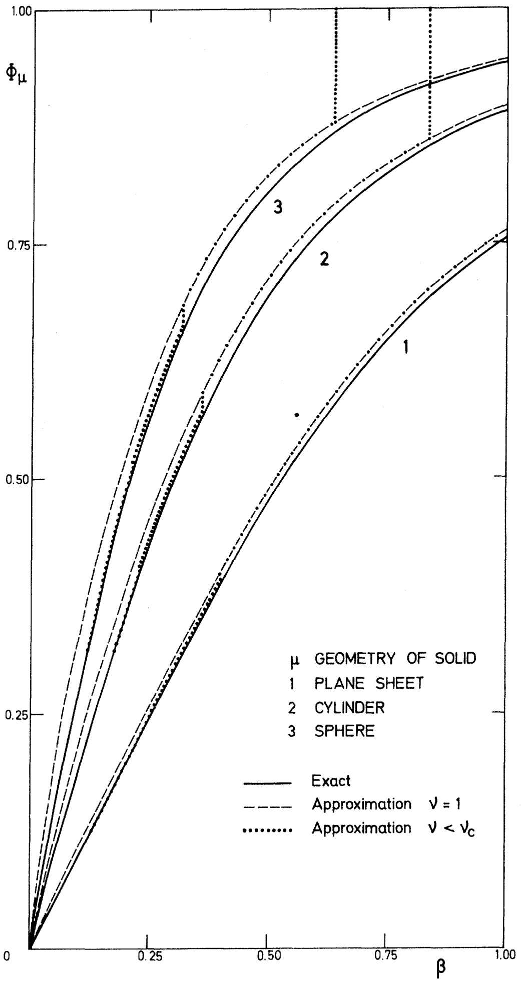

FIG. 1. Dependence of $\Phi_\mu(\beta, g=1)$ on β for different geometries. The approximations are discussed in the text.

recoil depletion. The exact result Eq. (15) supercedes Eq. (3) of Ref. 1, which does not exclude all random walks crossing the surface more than once[14] and does not exhibit, through g, the dependence on γ_v. The solid curves in Fig. 1 show $\Phi_\mu(\beta, g=1)$ for the three geometries $\mu = 1, 2, 3$.

We note that the first term of Eq. (9) consists of a sum of infinitely many components ν of rapidly descending importance. This apparently poses difficulties for the decomposition of experimental lifetime spectra. However, the terms $\nu > 1$ normally decay so rapidly that they submerge in a shortest lifetime component or in the prompt spectrum of the instrument. We demonstrate this by plotting, as dashed curves for $g = 1$ in Fig. 1, the apparent $\Phi_\mu^{\mathrm{app}}(\beta/g)$ that will be extracted from experimental data if only the leading term $\nu = 1$ in Eq. (9) is identified as a short lifetime component,

$$\Phi_\mu^{\mathrm{app}}\left(\frac{\beta}{g}\right) = \Phi_\mu\left(\frac{\beta}{g}\right) \Big/ \left[2\mu\left(\frac{1}{\lambda_{\mu 1}{}^2} - \frac{\beta^2}{\lambda_{\mu 1}{}^2\beta^2 + g^2}\right) + \Phi_\mu\left(\frac{\beta}{g}\right)\right]. \quad (16)$$

Another estimate of the error incurred by treating the sum in Eq. (9) as a single lifetime component results if one retains terms ν only up to a ν_c for which, say, $(\lambda_{\mu\nu_c}{}^2\beta^2 + 1) < 5$ corresponding to a time resolution $\lesssim 0.2\tau_s$,

$$\Phi_\mu^{\mathrm{app}}\left(\frac{\beta}{g}\right) = \Phi_\mu\left(\frac{\beta}{g}\right) \Big/ \left[2\mu \sum_{\nu=1}^{\nu_c}\left(\frac{1}{\lambda_{\mu\nu}{}^2} - \frac{\beta^2}{\lambda_{\mu\nu}{}^2\beta^2 + g^2}\right) + \Phi_\mu\left(\frac{\beta}{g}\right)\right]. \quad (17)$$

The dotted curves in Fig. 1 display Eq. (17) for $g = 1$ with discontinuities where, in our example, successive terms are cut off sharply by the condition $\lambda_{\mu\nu}{}^2\beta^2 + 1 = 5$.

These approximate results follow the exact curves closely. We conclude that, in general, only errors small compared to the usual uncertainties in lifetime decompositions are introduced by treating the sum in Eq. (9) as representing a single lifetime component.[15]

A reevaluation of the data reported in Ref. 2 according to Eq. (16) for $\mu = 3$ with Eq. (15) yields the Ps diffusion constants

$$D(\mathrm{SiO_2}) = (1.45 \pm 0.15) \times 10^{-5}\,\mathrm{cm^2 sec^{-1}},$$

$$D(\mathrm{Al_2O_3}) = (5.5 \pm 1) \times 10^{-5}\,\mathrm{cm^2 sec^{-1}},$$

and

$$D(\mathrm{MgO}) = (25 \pm 15) \times 10^{-5}\,\mathrm{cm^2 sec^{-1}}.$$

A simple, transparent, and yet in many instances adequate, formulation of the β dependence of Φ_μ obtains if one retains only the first term $\nu = 1$ in both the sums of Eqs. (9) and (12) renormalized by dividing with $2\mu/\lambda_{\mu 1}{}^2$. If we denote the effective escape rate from the solid as

$$\kappa_\mu \equiv \lambda_{\mu 1}{}^2\beta^2\gamma_s \quad (18)$$

and omit the subscript μ for brevity, we retrieve the lifetime spectrum derived earlier,[7]

$$n(t) = \left(1 - \frac{\kappa}{\kappa + \gamma_s - \gamma_v}\right) e^{-(\kappa+\gamma_s)t} + \frac{\kappa}{\kappa + \gamma_s - \gamma_v}\, e^{-\gamma_v t}. \quad (19)$$

The escape rates κ for various geometries are tabulated in Table I.[16] Equation (19) is an excellent approximation for internal microscopic sinks and has been applied to the trapping of positrons in crystal defects.[7-10] More precisely, for spherical voids of radius r present in a concentration $N = 3/4\pi r_1^3$ and, hence, a relative volume concentration $c = (r/r_1)^3$, the fraction of particles trapped according to the

TABLE I. Escape rates from solids and trapping rates into voids.

Structure	κ
Foil (thickness $2R$)	$\pi^2 D/4R^2$
Fiber (radius R)	$5.78D/R^2$
Sphere (radius R)	$\pi^2 D/R^2$
Rectangular slab (dimension $X\times Y\times Z$)	$\pi^2 D(X^{-2}+Y^{-2}+Z^{-2})$
Cylindrical pores ($\sim$dislocations) [radius r, concentration N (pores/area) $=(\pi r_1^2)^{-1}$]	$\dfrac{4\pi ND}{\ln(1/\pi N r^2)}$
Spherical voids ($\sim$vacancies) [radius r, concentration N (voids/volume) $=3/4\pi r_1^3$]	$4\pi r ND$

first-term decay mode retained in Eq. (19) is $1-\frac{4}{7}(r/r_1)^2+O(r^4/r_1^4)$.[17] That is, Eq. (19) is exact to $O(c^{2/3})$. Even at trap concentrations as high as $c\sim 10^3$ ppm, Eq. (19) describes the trapping process correctly for more than 99% of all positrons. If propagation as wave packets dominates, κ must be expressed in terms of quantum-mechanical trapping cross sections.[10]

In macroscopic solids, a "spurious" low-intensity component (1–3% of the total spectrum) with a long lifetime is often observed with an external positron source which, in some cases, can be traced to "surface effects." Indeed, such components may be a common phenomenon, of intensity $\frac{1}{2}\Phi_1\simeq\beta/2g$, Eq. (13), where the effective R is some fraction of the width of the positron range profile in the solid, if the surface conditions provide states that act as an absorbing barrier with positron lifetime longer than the bulk of the solid.

On their diffusion path to the surface of a solid, the positrons may be trapped at a rate κ_d in defects (last two entries in Table I) where they annihilate with a distinct rate γ_d such that $\gamma_v<\gamma_d<\gamma_s$. This changes Eqs. (4) and (5) in that γ_s is replaced by $\gamma_s+\kappa_d$. When solved in combination with the rate equation describing A-center formation by positron trapping on defects,

$$\frac{dn_d(t)}{dt}=-\gamma_d n_d(t)+\kappa_d n_s(t)\,, \tag{20}$$

where $n_d(0)=0$, a component of intermediate lifetime $\gamma_d^{-1}\equiv\tau_d$, appears in the positron-lifetime spectrum with an intensity proportional to κ_d:

$$n(t)=2\mu\sum_{\nu=1}^{\infty}\left[\left(\frac{1}{\lambda_{\mu\nu}{}^2}-\frac{\beta^2}{\lambda_{\mu\nu}{}^2\beta^2+g_v{}^2}\right)-\frac{\kappa_d}{\gamma_s g_d^2}\left(\frac{1}{\lambda_{\mu\nu}{}^2}-\frac{\beta^2}{\lambda_{\mu\nu}{}^2\beta^2+g_d^2}\right)\right]\exp[-(\lambda_{\mu\nu}{}^2\beta^2+1)\gamma_s t-\kappa_d t]$$
$$+\frac{\kappa_d}{\gamma_s g_d{}^2}\left[1-\Phi_\mu\!\left(\frac{\beta}{g_d}\right)\right]e^{-\gamma_d t}+\Phi_\mu\!\left(\frac{\beta}{g_v}\right)e^{-\gamma_v t}\,, \tag{21}$$

where

$$g_v^2\equiv g^2+(\kappa_d/\gamma_s)\,, \tag{22}$$

$$g_d^2\equiv g^2+(\kappa_d+\gamma_v-\gamma_d)/\gamma_s\,, \tag{23}$$

and g^2 is defined in Eq. (11).

If data are analyzed in terms of Eq. (19), the intensity $\Phi_\mu(\beta/g_v)$ of the longest-lived component due to annihilations in the voids depends, then, on the argument $\beta/g_v=(\beta/g)[1+(\kappa_d/\gamma_s g^2)]^{-1/2}$, resulting in an effective diffusion constant

$$D^{\mathrm{eff}}=D[1+(\kappa_d/\gamma_s g^2)]^{-1}\,. \tag{24}$$

One estimates that $D^{\mathrm{eff}}\simeq D$ for all defect concentrations at which defect trapping remains a distinct process ($c<10^3$ ppm).[15] In particular, the interestingly large variations noted earlier[1] between the measured positronium-diffusion constants quoted above for different solids could not be accounted for by differences in defect concentrations. When the voids are of atomic dimensions, one has $g<1$ and retrieves, in the approximation of Eq. (19), the lifetime spectrum of positrons annihilating in a solid with two types of noninteracting traps.[18]

Practically all positrons in solids annihilate with electrons into two γ quanta. The γ rays emerge from the site of the annihilation in opposite directions with small deviations from 180°, of order milliradians, which for thermalized positrons reflect the distribution in momentum of the electrons in the domain of overlap between the positron wave function and the electron wave function of the solid. If positrons become trapped in voids, their wave functions overlap at the boundary with electrons of lower density and lower momenta than in the solid. Therefore, trapped positrons give rise not only to a long-lived component in the lifetime spectrum but also to a component with narrower angular correlation than positrons annihilating in the solid. Such interrelations between lifetime and angular correlation with changing defect concentration are observed experimentally.[19] If positrons are trapped as Ps, the narrow component attributed to the self-annihilation of p-Ps changes: Confinement in microscopic voids widens the narrow component by an amount that corresponds to the zero-point energy of the p-Ps atom in the confining potential well.[20] If o-Ps diffuses from small solids into macroscopic voids, pickoff annihilation from the solid surface can normally be neglected.[1] The contribution of this component to the angular correlation is merely the small fraction of the 3γ self-annihilation spectrum of o-Ps in the void that can be registered by the 2γ coincidence apparatus.

It is of interest, then, to exhibit the relation between the diffusion-controlled lifetime component Φ_μ and the fraction F_μ of annihilations in voids which changes the angular correlation. The number of positrons in the solid at time t is

$$n_s(t) = 2\mu \sum_{\nu=1}^{\infty} \lambda_{\mu\nu}^{-2} \exp[-(\beta^2\lambda_{\mu\nu}^2 + 1)\gamma_s t], \quad (25)$$

so that

$$F_\mu(\beta) = 1 - \gamma_s \int_0^\infty n_s(t)\,dt = \Phi_\mu(\beta, g=1) \ . \quad (26)$$

It follows that only in powders with large voids, where $\gamma_v \ll \gamma_s$ and thus $g \simeq 1$, is the intensity Φ_μ equal to the fraction F_μ of positrons annihilating in voids. In dense materials with microscopic voids such as vacancies, g is typically of order 0.5, and thus $F < \Phi$. Specifically in the approximation of Eq. (19),

$$\Phi = \frac{F}{g^2 + (1-g^2)F} \ . \quad (27)$$

Normally only changes in F and Φ are of interest as observed, for example, if the defect concentration in a material is varied. By Eq. (27) the two observations are proportional to first order,

$$\Delta\Phi/\Phi^2 = g^2(\Delta F/F^2) \ . \quad (28)$$

Thus even when ΔF varies significantly, $\Delta\Phi$ can remain too small to be resolved, if g is small, and the presence of defects will manifest itself only through an increase of the mean lifetime.[21,22] The rate of capture by small internal positron traps in solids or liquids, κ, can be determined in the approximation Eq. (19) directly from either Φ or from F, or any characteristics of the angular correlation curve that is proportional to F, such as the change in counting rate at the maximum or the change in the width of the angular-correlation curve, because

$$\kappa = (\gamma_s - \gamma_v)\Phi/(1-\Phi) = \gamma_s F/(1-F) \ . \quad (29)$$

Otherwise, the escape rate D/R^2 must be determined with the aid of Eqs. (12) and (26) by a fitting procedure.

If the material contains large voids that are filled with a gas dense enough to stop positrons to form initially a significant fraction of Ps, n_{0v}, in the voids so that $n_s(t=0) \equiv n_{0s} = 1 - n_{0s}$ but not so dense as to reduce significantly the energy that Ps atoms can gain by diffusing into the voids, the intensity of the long-lived component becomes

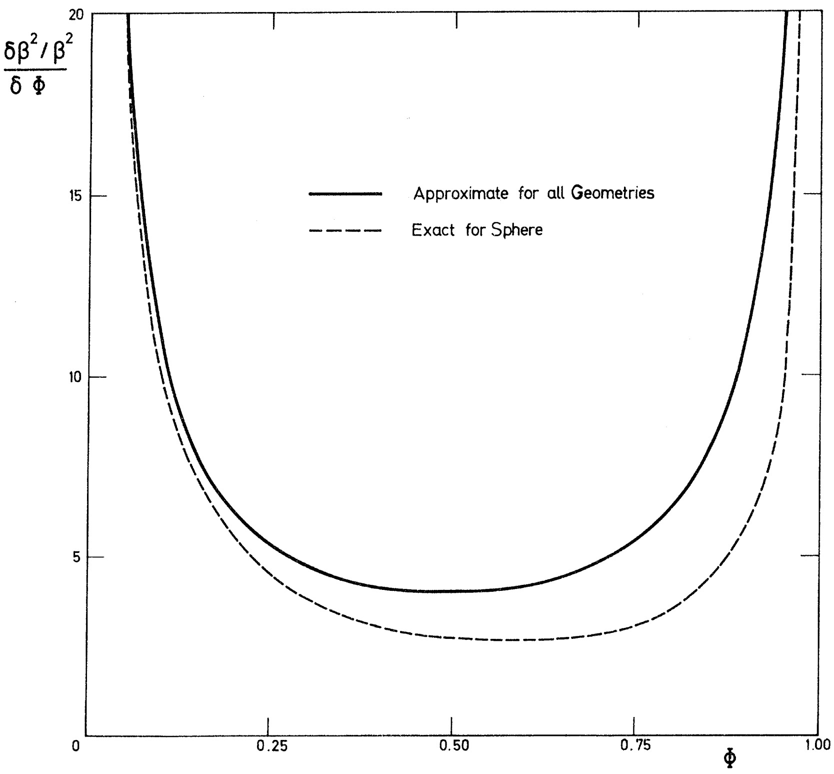

FIG. 2. Relative error in β^2 determination from Φ in units of its uncertainty $\delta\Phi$.

$$\Phi'_\mu(\beta, g) = n_{0v} + n_{0s}\,\Phi_\mu(\beta, g)\ . \tag{30}$$

The fraction annihilating in the voids is

$$F'_\mu = \Phi'_\mu(\beta, g=1) \leqslant \Phi'_\mu(\beta, g)\ . \tag{31}$$

These interrelations apply in many instances, for example, to the study of molecular sieves[5] and semicrystalline polymers.[23] Moreover, important aspects of positron chemistry, in particular relations between lifetime spectra and angular-correlation data in solutions,[24] can be understood through these formulas in terms of positron diffusion and reaction rates.

We conclude with a brief comment about the principal uncertainties encountered in determining β^2 from a diffusion-controlled component in positron-lifetime spectra. Since β^2 depends on D, R, and γ_s, any two of these parameters must be known from independent observations to determine the third. The relative uncertainties propagate as

$$\delta\beta^2/\beta^2 = \delta D/D + \delta\tau_s/\tau_s + \delta R^2/R^2\ ,$$

where in the approximation Eq. (19)

$$\delta\beta^2/\beta^2 = \delta\Phi/\Phi(1-\Phi)\ . \tag{32}$$

Figure 2 shows Eq. (32) together with the exact curve for spheres calculated from Eq. (15), for fixed $\delta\Phi$. There is a broad optimal range of Φ_μ values centered around $\Phi_\mu \simeq 0.5$, where the error in extracting β^2 from lifetime measurements is the smallest and dependent only on $\delta\Phi_\mu$. For $\Phi_\mu < 0.2$ the uncertainty becomes large because then $\delta\Phi_\mu \sim \Phi_\mu$; for $\Phi_\mu > 0.8$ only a lower limit for β can be estimated from the experimental data.

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