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Observation of Long Positron Lifetimes in Crystals with a Weak Magnetic Field

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Abstract
Lifetime distributions of time-coincident, 180o γ-rays from KCl, KBr, KI and C_8H_{10} (naphthalene) in 12 and 95 gauss magnetic fields have been measured. The samples were exposed to low frequency γ-ray sources with energies above the electron/positron pair-formation threshold. According to theory, in these fields slow positrons may bind to anions and form positronium with lifetimes of tens to hundreds of minutes. At the low rates gas-filled radiation detectors worked satisfactorily. Lifetimes agree with classical electric dipole decay that is expected of long-lived positronium formed in crossed motional electric and magnetic fields. This work fills an important gap in knowledge that has existed since the discovery of the positron in 1932. The results reported herein establish the highest energy pure $_\gamma$-ray emitter and longest lived excited atomic state known at this time.

Introduction
Since the discovery of the positron over ninety years ago it has been axiomatic that its lifetime against annihilation in solids depends only on the atomic electron density and square of the positron wave function [1,2]. The wave function depends on the atomic structure and its calculation can be challenging. Twenty-two years later it was shown that the lifetime of the electron-positron bound state called positronium (Ps) is quenched by external magnetic fields [3]. Moving forward forty-two years, following invention of pseudo-momentum that allowed separation of collective and internal motions, it was predicted that crossed electric and magnetic fields form long-lived Ps, with lifetimes of order one year [4-7]. Recently it was proposed that long-lived Ps exists in extra-galactic jets [8]. Still, it is eight decades since the invention of quantum mechanics and the existence of long-lived positrons in solids is not settled.

To challenge the previously untested crossed field theory, we devised an experiment to form and detect in solids what traditionally would be called 'metastable Ps'. Several issues had to be addressed to stage an experiment with some hope of success. First, to avoid self-annihilation the positron and electron must be separated by a large distance. According to the theory of a motional electric field and an applied magnetic field, by choosing the Ps pseudo-momentum just above unity (atomic units) one can simultaneously avoid unstable coulomb states and arrive at the greatest separation when the magnetic field is smallest [5,7]. Readily available fields of less than 100 gauss (G) give lifetimes of tens to hundreds of minutes.

Second, solids must be found where the positron survives amidst many electrons. Crystalline materials are a good choice based on the abundance of experimental and theoretical understanding of positrons in these media. In particular, studies in alkali halides show that the positron binds to the anion and behaves like a free particle plane wave not localized to any one anion [9-13]. If the positron in metastable Ps would behave similarly and symmetrically among all electrons within its range of migration, its predicted electric dipole oscillator structure would provide a unique time profile of annihilations of 'doublehelix' Ps, named for the electron-positron entanglement in the magnetic field [14]. Similar circumstances may also exist for other crystalline materials such as the hydrocarbon C_8H_{10} (naphthalene) [15,16].

Third, the apparatus should match physics requirements. Following the discovery of the positron in 1932, gas-filled Geiger counters were successfully used in low rate coincidence experiments to enrich positrons in cosmic rays and those made by natural $_\gamma$-ray sources in cloud chamber photographs [17,18]. They were later used to discover positron-emitting radionuclides and measure lifetimes of tens of minutes, as well as measure angular correlations of γ-ray pairs from positron annihilation [19,20].

Geiger counters gave way to delayed coincidence methods with fast scintillation counters that were used to discover and measure extremely short Ps lifetimes and study magnetic quenching of Ps in alkali halides [21-25]. However, this method is not reliable for long lifetimes, and because of the focus on short lifetimes may explain why metastable Ps was not detected in the last six decades. To fill this void test unfulfilled predictions, we assembled an experiment comprised of weak $_{\gamma}$-ray sources, alkali halide and hydrocarbon targets, permanent magnets, two Geiger counters and a time coincidence circuit to measure 180o 2γ annihilations over long time baselines. To the best of our knowledge no similar experiment has been reported in the history of positron physics.

Experimental Setup

Two Aware Electronics RM-80 pancake type Geiger counters viewed 210 g of salt-substitute KCl ($_{\rho}$ = 1.52 g/cm^3) (Figure 1). The counters had 45 mm diameter thin mica windows and a measured efficiency of 22% for 511 keV γ-rays. They were oriented at 180o to detect annihilation γ-ray pairs in coincidence (Cbox). Each counter was 150 mm from the center of a 117 mm long x 38 mm diameter thin walled cylinder of KCl. To test the system the KCl was replaced with a ^{22}Na positron source with 20 mm thick lead cylindrical collimators having 9 mm diameter center holes in front of each counter. The coincidence to singles ratio was (8.3±1.7) •10-4 that compares favorably to (10±1) •10-4 from a similar experiment [20].

A magnetic field was created by a Helmholtz pair of 17 mm diameter x 3 mm thick NdFeB disc magnets with 12,300 G residual field. Each was centered 30 mm from the ends of the inside surface of the 75 mm long return ends of a semi-rectangular, 35 mm wide x 2 mm thick x 150 mm long iron yoke. Using onaxis magnetometer measurements of 10 and 20 G at the center and 45 mm from a pole face, respectively, a fit to an analytic form gave an average field of 12 G over ±30 mm from the center. Perpendicular to the central axis, the field at the center fell to 10 G at one cylinder radius and 4 G at two cylinder radii. The weakly diamagnetic KCl should have no measureable effect on the magnetic field.

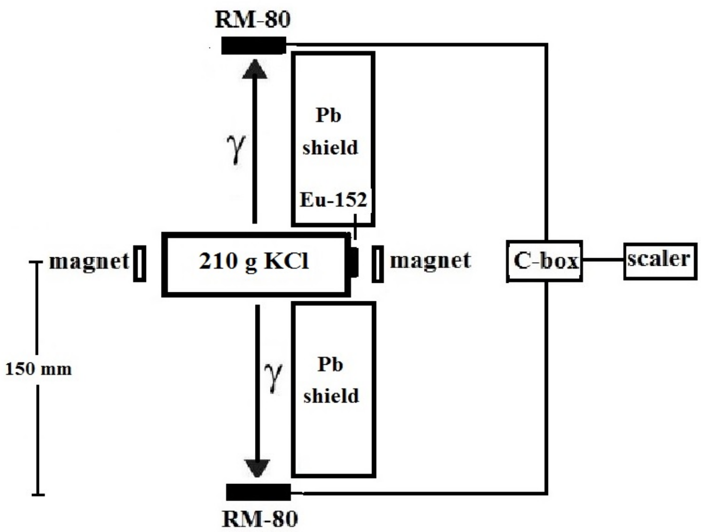

Figure 1: Experimental Setup: Plan view; 210 g KCl Cylinder with 2 NdFeB disc Magnets, 12 G Central Field at 150 mm disc Separation, two RM-80 Geiger Counters, Lead Shields, Coincidence Circuit (C-box), Scaler; Magnet Iron yoke not shown.

The major source of γ-rays above 1020 keV was ^{40}K that releases a 1461 keV line in natural KCl at the rate of 99 γpm/g•210 g = 2.1•10^4 γpm. A modest increase in γ-ray flux was made by attaching a 1 μCi ^{152}Eu source to one end of the KCl cylinder. Including 72% for γ-ray branching, 51% for energies above 1020 keV and 50% for solid angle, it injected 0.7•10^4 γpm into the KCl. Finally, the cylinder was bathed in ground level atmospheric radiation, including important γ-ray emitters ^{214}Bi in radon gas and groundsourced ^{40}K. From models of γ-ray emission in radon ^{214}Bi injected 0.05 γ/(cm^2•s) • 60 s/min • 216 cm^2 • 0.5 (above 1020 keV) = 0.03•10^4 $_{\gamma p}$m into the KCl. Measured relative strengths of the 1461 keV and ^{214} Bi 1765 keV lines give 0.15 • 10^4 $_{\gamma p}$m for ^{40}K for a total of 0.18 • 10^4 $_{\gamma pm}$ from ground level atmospheric radiation. In summary, the grand total with energy above 1020 keV was 3.0•10^4 γpm, with 70% (^{40}K), 23% (^{152}Eu) and 7% (atmospheric radon and ground-sourced ^{40}K) [26,27].

Results and Discussion

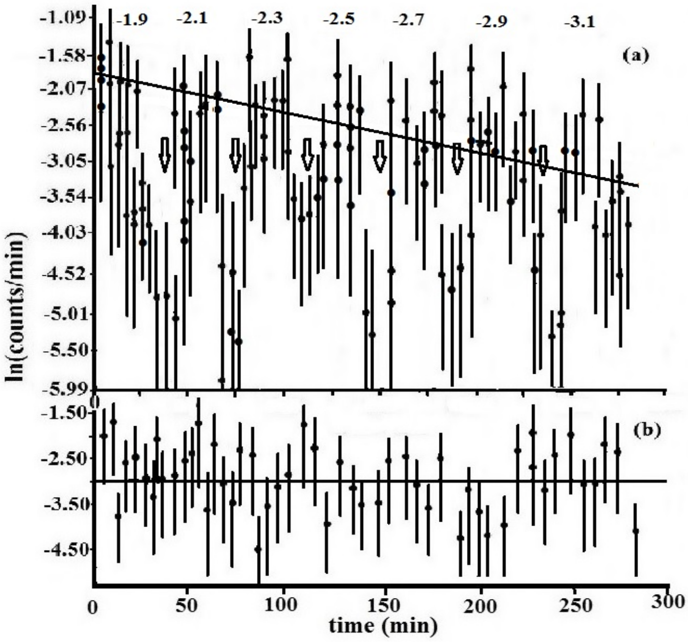

Figure 2: Logarithmic Counting Rate: (a) 12 G; KCl (210 g); Top, Peak Rates (cpm); Curve, 233±40 Min Mean Lifetime; Arrows, Minima, 40±12 Min Period (b) 0 Gauss: Curve, 0.045±0.015 cpm: Statistical Errors.

KCl (12 G) Lifetime Distribution: Rates were measured in 280 min runs with a few seconds for magnet insertion. Forty-five runs were combined to give the lifetime distribution in Figure 2(a) that displays a prominent modulated exponential decay. A mean lifetime of 233±40 min (solid line) was found by fitting to the eight largest points in each of the seven maxima (t = 20, 60, ••). The data can be expressed in the form (0.16±0.03) • exp (-t/233±40) • $\sin^2$ ($_{\pi}$t/40±8) cpm. Integration over 280 min gives an average rate f = 0.046±0.014 cpm. Figure 2(b) shows no evidence for modulation or decay with the magnets removed.

Expected Rates and Background: Taking into account the pair formation cross section, the rate for positron formation in the 60 mm of unshielded KCl is $3.0 \cdot 10^4$ $_{\gamma pm}$ • [1-exp ($-1.6 \cdot 10^{-5}$ mm^{-1}•60 mm)] = 29 pairs pm [28]. Multiplying by counter efficiencies of 0.025 (pair solid angle) and $(0.22)^2$ (counter efficiencies) gives 0.035 cpm, in agreement with the 0.046±0.014 cpm average in Figure 2(a). In an ideal background free environment the modulation minima in Figure 2(a) should be zero. Therefore, the average of the six minima at t = 40, 80, ••, or 0.006±0.002 cpm, is a direct measure of the accidentals rate. In summary, accidental counts are too small to account for the features of Figure 2(a).

Other Pair Formation Experiments: Three measurements of the $^{40}{}_{K\rightarrow\beta^+}$ energy spectrum using coincident NaI counters without a magnetic field have been reported [29-31]. The 1461 keV γ-ray pair produces an annihilation energy peak at 441 keV near the β+ spectrum endpoint. From Table II the measured pair/β+ ratio is 1.34. Multiplying by known β+/β- and β-/γ ratios of $1.12 \cdot 10^{-5}$ and 8.52, respectively, gives $1.3 \cdot 10^{-4}$ pairs/γ [30]. Further multiplying by 3.0•104 γpm and known ratios of 60 mm/11.3mm (Table III [30]) for relative γ-ray absorption lengths and $(18/15)^2$ [30, Table I] for pair Z dependence, as well as efficiencies, gives 0.04 cpm that agrees with Figure 2(a).

Theoretical Overview: To facilitate the following discussion, we briefly review theory [5]. In an electric field created by Ps moving on the y-axis in a magnetic field B on the z-axis, the potential (au) is V(x, 0, 0) = $B^2x^2/4 + BKx/2 + K^2/4 - 1/$[abs(x)], where K is the Ps pseudo-momentum. For B/K << 1 an outer well exists at x_o = - K/B [4]. Large positron-electron separation requires small B with the choice of K being just above unity to avoid unstable coulomb states [8]. To illustrate, Figure 3 shows V(x, 0, 0) for B = 7 gauss ($2.9787234 \cdot 10^{-9}$ au) and K = 1.01 au. The well is centered at x_o = $-3.39 \cdot 10^{8}$au, or the electronpositron separation is 18 mm, with a depth V_o = -B/K = $-2.95 \cdot 10^{-9}$ au ($-8.0 \cdot 10^{-8}$ eV). Figure 3 shows the ground state at $1.48 \cdot 10^{-9}$ au ($4.0 \cdot 10^{-8}$ eV) below the ionization level at I = B = $2.98 \cdot 10^{-9}$ au ($8.1 \cdot 10^{-8}$ eV).

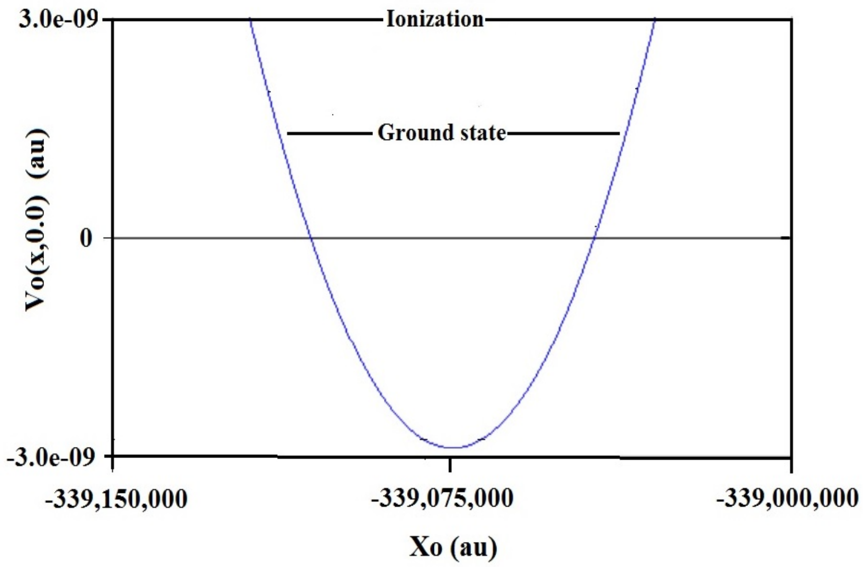

Figure 3: Effective Potential: B = 2.98•10-9 au (7 gauss), K = 1.01 au.

Interpretation of KCl (12 G) Data: As discussed earlier the positron may form a Wheeler compound $(Cle^{-})e^{+}$ [2]. In a weak magnetic field the weakly bound $e^{-}e^{+}$ pair would constitute an anisotropic electric dipole oscillator with a dipole moment $p_x = 1.6 \bullet 10^{-19}$ C • 0.011 m = $1.7 \bullet 10^{-21}$ C-m and angular frequency ωx = $(B^2 + 4/x_o^3)1/2$ = 204 MHz [5]. With $k = \omega_x/c = 0.68\ m^{-1}$ and the counter at R = 0.15 m, then kR << 1 and therefore the oscillator emits into the far field. The power is $1.73 \bullet 10^{-13} \bullet {}_{v}^{4} \bullet p_{x}^{2}{}_{/\kappa}$ = $1.15 \bullet 10^{-25}$ watts, where κ = 4.85 is the KCl static dielectric constant [32]. The dipole also rotates in the x-y plane at the same (cyclotron) frequency and radiates along both the x- and y-axes for a total power of $2.3 \bullet 10^{-25}$ watts.

For a thermal metastable Ps the lifetime 1/Γ is $4.0 \bullet 10^{-21}$J/$2.3 \bullet 10^{-25}$ watts = $1.7 \bullet 10^{4}$ s, or 290 min, in fair agreement with the 233±40 min. The dipole collapses as it radiates, triggering annihilations at the same rate. Ref. asserts that at low magnetic fields near K = 1 the probability density, or square of the wave function at the origin, falls over sixteen orders of magnitude [5]. Multiplication by the 142 ns oPs lifetime gives a lifetime of over one-half year. It must be pointed out that the claimed lifetime mechanism, quantum barrier tunneling, is not to be confused with the much faster radiation mechanism.

In analogy to atomic fluorescence, when γ-rays of frequency f and width Θ satisfy Θ/ω_L, << 1 and Γ/f << 1, modulation should be strong [33]. With Γ = 0.0043±0.007 min^{-1} and γ-ray polarities of E2 (^{40}K), E1 and E2 (^{152}Eu) and M1 and E2 (^{214}Bi) at 1000-2000 keV energy giving $_{\Theta\,\approx}10^{-11}$s, $_{\Theta}/_{\omega L}$ = $4.9 \bullet 10^{-20}$ and Γ/f = 0.09 [34]. Therefore, strong modulation is expected and observed with a peak/valley intensity ratio of ≈ 20 and period of $(0.5 \bullet f)^{-1}$ = 40±12 min, where 0.5 accounts for two counters in the circular rotation. f. No effects in KCl lifetime experiments in strong fields? Delayed coincidence lifetime experiments have not observed metastable Ps in magnetic quenching experiments in KCl [22-25]. Examination of the dipole power formula reveals that the metastable Ps lifetime scales as B-2. For example, at 10 kG the lifetime is 233 min•$[12/(10{,}000)]^2$ = 335 $_{\mu}$s. With start-stop rates of $10^3\ s^{-1}$, the minimum accidentals rate is 2 • $(10^3\ s^{-1})^2$ • $(335 \bullet 10^{-6}$ s) = 0.7 • 10 $^{3}\ s^{-1}$, or 70% of the singles rates that would obscure metastable Ps signals.

Magnetic Field Scaling in Alkali Halides: The magnet yoke was shortened and a 73 mm long x 40 mm diameter KCl cylinder (124 g) was inserted between the pole faces. Axial fields at the center and 25 mm from a pole face were measured at 50 and 150 G, respectively. Integration of an analytic form between ±30 mm from the center gives an average of 95 G. Scaling from 12 to 95 G predicts a 233 • $(12/95)^2$ = 3.7 min lifetime, in agreement with the 4.1±0.4 min lifetime in Figure 4(a). With $_{\Gamma}$ = 0.24 ±0.02 min^{-1}, Γ/f >> 1 and modulation is critically damped and not observed. Magnetic field scaling is confirmed [33].

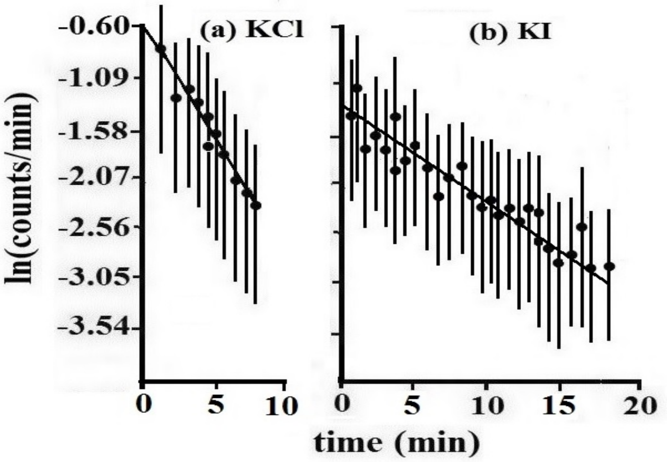

Figure 4: Logarithmic Counting Rate Versus Time at 95 G: (a) KCl (124 g); Curve, 4.1±0.4 Min Mean Lifetime; (b) KI (100 g); curve, 10±1 Min Mean Lifetime: Statistical Errors.

h. Anion density scaling in alkali halides: 100 g cylinders of KI powder (ρ=2.0 g/cm^3) and granulated KBr (ρ=1.7 g/cm^3) were inserted between the magnet pole faces so that they experienced the 95 G field.

Ps lifetimes in alkali halides vary inversely with anion density n*, where $n^* = n/[1- 4\pi n R_+^3/3]$, $n = \rho N_A/A$, N_A is Avagadro's number, A is the molecular weight and R_+ is the K^+ ion radius, taken as 1.96 • 10^{-10} m [13,35]. Substitution for n gives n*(KCl)/n*(KI) = 2.13 and n*(KCl)/n*(KBr) = 1.90. Multiplying by 4.1±0.4 min gives KI and KBr lifetimes of 8.7±0.9 and 7.8±0.8 min, respectively. A straight line fit gives lifetimes of 10±1 min (Fig. 4(b)) for KI and 8.5±1.3 min for KBr (not shown). Both have Γ/f >> 1 and no modulation is expected. Metastable Ps lifetimes in alkali halides scale with the inverse anion density. i) Lifetime in a crystalline hydrocarbon: To explore other crystalline materials, the lifetime of C_8H_{10}, or naphthalene, a polycyclic aromatic hydrocarbon compound, was measured. The 95 G magnet was used, with the ^{152}Eu source sandwiched at the magnet center between two 40 mm diameter x 40 mm long, 40 g cylinders of finely granulated naphthalene (ρ = 0.93 g/cm^3, κ = 2.50). Figure 5 shows the lifetime distribution with a modulated exponential decay given by (5.5±01.5) • exp (-t/9.8±2.0) • $\sin^2$ (4.5±1) maxima and minima that are in agreement within errors. Scaling the 290 min electric dipole lifetime for KCl by the ratio of dielectric constants, 4.85/2.50, and square of magnetic fields, $(12/95)^2$, gives 9.0 min, in good agreement with the naphthalene lifetime of 9.8±2.0 min. Finally, $\Theta/\omega_L \approx 6.2 \cdot 10^{-21}$ s^{-1}, but Γ/f = 0.52±0.15. Therefore, weak modulation is expected and observed (peak/valley intensity ratio ≈ 3).

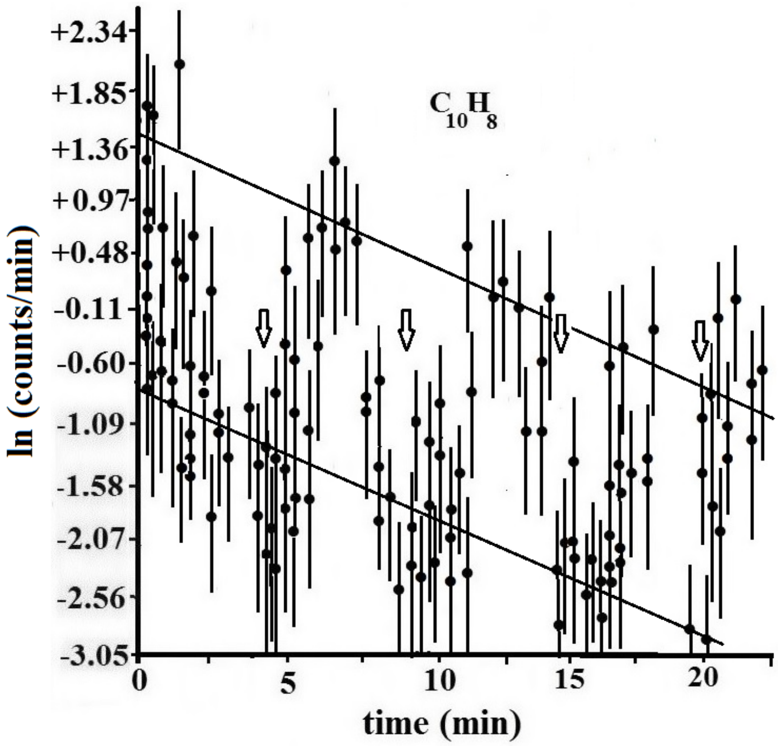

Figure 5: Logarithmic Counting for Naphthalene at 95 Gauss: Solid Lines; Mean Lifetime of 9.8±2 min from Fits to Minima and Maxima; Arrows, Locations Of Minima, 9±2 Min Period: Statistical Errors.

Lifetime quenching of naphthalene has been studied at 4-20 kG and 2.1-15.8 kG [15,16]. Quenching is much stronger than can be explained by Halpern theory modified to account for positron contact with its material surroundings. The existence of two Ps-like centers is suggested, one with normal contact density and the other 'anomalous'. Scaling by B^{-2} from 9.8 min at 95 G predicts positron lifetimes of 1.2 s at 2.1 kG and 0.013 s at 20 kG. As pointed out previously, these lifetimes would not be detected by the delayed coincidence method used in both experiments. Such losses could be interpreted as excessive quenching.

Conclusions

The 2γ lifetime distribution of crystalline KCl exposed to γ-rays above the pair-formation threshold at 12 G shows strongly modulated decay with an exponential lifetime and modulation period of 233±40 min and 40±5 min, respectively. The calculated lifetime for electric dipole radiation is 290 min and the modulation period is consistent with $_\gamma$-rays as predicted by theory. Measured lifetimes at 95 G in KCl, KBr and KI of 4.1±0.4, 8.5±1.3 min and 10.0±1.0 min, respectively, are consistent with B-2 and anion density scaling. Measurements at 95 G in the crystalline hydrocarbon compound naphthalene give a weakly modulated exponential distribution with a lifetime and modulation period of 9.8±2.0 min and 4.5±1 min, respectively. Lifetimes between KCl and naphthalene scale with dielectric constant and magnetic field. To the best of our knowledge these are the first observations of long-lived positrons in solids. Metastable Ps is the highest energy pure γ-ray emitter and longest lived excited atom known.

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