Pick-off annihilation of delocalized positronium in BaF_2 at elevated temperatures

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Abstract. Positron lifetime components and associated intensities in the temperature range 300 – 800 K were measured using standard fast-fast coincidence technique. Two lifetime components were resolved after background and source corrections. The long lifetime component decreases in the temperature range from 500 ps at 300 K to 402 ps at 711 K. This corresponds to a fractional increase in the annihilation rate of 22% in the temperature range 300 K to 693 K. The de-trapping of positronium from the Bloch states followed by annihilation through the 'pick-off' process appears to be one of the dominant processes in the long lifetime components in the temperature range. Annihilation rates from positron annihilations with valence and core electrons of the individual atoms of the sample are also calculated using density functional theory in the framework of generalized gradient approximation.

1. Introduction

Positronium which is a hydrogen-like bound state, has two configurations that depend on the relative spins of the positron and electron. The singlet state (S=0, M=0) is referred to as para-positronium (p-Ps), where S is the total spin and M is the *z*-component quantum number. The triplet states (S=1, M=0, ±1) are referred to as ortho-positronium (o-Ps). Para-positronium emits two gamma photons while ortho-positronium emits three gamma photons. Experimental measurements of positronium annihilations use the two lowest states. These two lowest states are both in ground states (n=1 and L=0) namely $^1S_0$ (p-Ps) and $^3S_1$ (o-Ps). The decay rate of p-Ps ground state, 1^1S_0, is calculated as a series of α given by [1]

$$\Gamma(p-Ps\rightarrow\gamma\gamma)=\frac{\alpha^5 m_e}{2}[1-\left(5-\frac{\pi^2}{4}\right)\frac{\alpha}{\pi}+2\alpha^2 ln\frac{1}{\alpha}+1.73\left(\frac{\alpha}{\pi}\right)^2-3\frac{\alpha^3}{2\pi}ln^2\frac{1}{\alpha}+O(\alpha^3 ln\frac{1}{\alpha})] \quad (1)$$

where $\alpha \approx 1/137$

Para-positronium mean lifetime calculated from equation (1) is about 125 ps and more calculations [2-5] are in agreement. The ground state decay rate of the ortho-positronium, 1^1S_1, decays into an odd
number of photons and is given by [6-7]

$$\Gamma(o-Ps\rightarrow\gamma\gamma\gamma)=\frac{2(\pi^2-9)\alpha^6 m_e}{9\pi}[1-10.28661\frac{\alpha}{\pi}-\frac{\alpha^2}{3}ln\frac{1}{\alpha}+ B_0\left(\frac{\alpha}{\pi}\right)^2-\frac{3\alpha^3}{2\pi}\ln\left(\frac{1}{\alpha}\right)+O(\alpha^3 ln\alpha) \quad (2)$$

where B_0 is a two-loop coefficient needed to bring theory into an agreement with experiment. The effects of higher order term $O(\alpha^6 m_e)$ are fully discussed elsewhere [8-12]

The mean lifetime of ortho-positronium is about 142 ns [13-14]. The lifetime of ortho-positronium is very long such that it is possible that some processes can take place and disturb the ortho-positronium system before it annihilates. One of the processes known as "pick-off" annihilation involves the positron in the ortho-positronium system colliding and annihilating with one of the electron of the host material but not with the electron that is part of the positronium system [15]. The "pick-off" process is carried out in accordance with the reaction

$$Ps+e^-\rightarrow 2\gamma+e^- \quad (3)$$

where e^- is the free electron of conduction band or valence electron of lattice atom

The annihilation rate of Ps in a solid can be written as

$$\Gamma=\kappa\Gamma_o+\Gamma_{pick-off} \quad (4)$$

where $\kappa\Gamma_o$ is self-annihilation rate and $\Gamma_{pick-off}$ is the pick-off annihilation rate. Pick-off annihilation rate can be obtained from a standard Monte Carlo sampling as

$$\Gamma_{pick-off}=\pi r_e^2 c\int n_-(r)n_+\gamma[n_-(r)]d^3r \quad (5)$$

where $n\_(r)$, $n_+(r)$ are the electron and positron densities respectively and γ is the enhancement factor.

The other process is the conversion of ortho-positronium into para-positronium due to spin-orbit interaction [15]. As soon as the conversion takes place, para-positronium annihilates quickly. Hoydo et al [16-17] suggested that in alkali halide crystals, positronium exist in a free Bloch state at very low temperatures. A simple model proposed by Hoydo et al [23] which include free and trapped states for positrons and positronium is used. The positron lifetimes and their corresponding intensities in the framework of this model are given by

$$I_1\tau_1=I_f\tau_f+I_{pPs}\tau_{pPs}+I_{foPs}\tau_{foPs} \quad (6)$$

$$I_2\tau_2=I_{t+}\tau_{t+}+I_{toPs}\tau_{toPs} \quad (7)$$

$$I_1=I_f+I_{pPs}+I_{foPs} \quad (8)$$

$$I_2=I_{t+}+I_{toPs} \quad (9)$$

where the subscripts *f*, *t+*, *pPs*, *foPs* and *toPs* refer to the free positron, trapped positron, free para-positronium, free ortho-positronium and trapped ortho-positronium respectively.

More studies on KCl and NaF [18-20] reported a localization of positronium as the temperature rises in the temperature range 9 – 400 K. Alkaline earth halides have a structure close to that of alkali halides. Elefteriades et al [21] measured positron lifetimes in barium fluoride in the temperature range 10 – 293 K. They found an increase in the intensity of a long lifetime component between 12 and 80 K followed by a decrease of intensity from about 63% to 37% in the temperature

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range 80 – 293 K. An increase in the positronium formation probability, associated with temperature range 10 – 80 K, was viewed as localization of positronium and they also suggested that a decrease in the second lifetime component intensity from about 63% to 37% was probable due to the de-trapping of localized positronium at temperatures higher than 80 K.

In this work we explore the temperature dependence of annihilation parameters in ionic barium fluoride in the higher temperature range 300 – 800 K. We further study the annihilation rates due to positrons annihilating with valence and core electrons of the individual atoms in barium fluoride in the temperature range using density functional theory in the framework of generalized gradient approximation. We also understand that in alkali earth halide, only anionic vacancies, which are positive, are thermally created and the long positron lifetime measurements would be difficult to attribute to annihilations at defects in the temperature range 300 – 800 K. We have not yet found any literature reporting in this temperature interval using positron annihilation technique.

2. Experiment

Two barium fluoride samples each of surface area 8 mm × 10 mm and thickness of 2 mm and of cleavage [111] were obtained from Goodfellows, Britain. A positron source of activity 15 μCi of ^{22}NaCl was sealed between two electron-welded nickel foils (density of 8.9 g/cm^{-3}) each of thickness 7 μm. The radioactive area was about 16 mm^2. Sealed positron source was sandwiched between two equal barium fluoride samples in a standard sandwich arrangement**.** The source-sample-heater arrangement is shown in figure 1

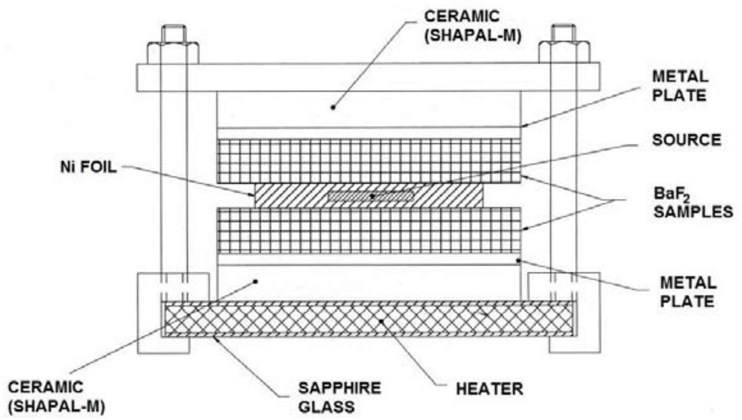

Figure 1. Sample holder system showing the source-sample-heater arrangement.

Annihilation rate measurements wfere obtained by utilizing a fast-fast coincidence system with two collinear photomultipliers (XP2020) coupled to barium fluoride scintillators. Positron lifetime spectrum in ZnO was accumulated for the calibration of the system. The full width at half maximum (FWHM) of the time resolution was 220 ps of the Gaussian resolution function.

Lifetime spectra were analysed using LIFSPECFIT [22]. A two-component fit was resolved following source correction and background subtraction. Throughout the experiment, the reduced chi-square (χ^2) for most of set data was fairly close to unity.

3. Results and discussions

Barium fluoride is an alkali earth halide of cubic structure of space group Fm3m. Figures 2 and 3 show the long and short positron lifetime components and the corresponding intensities respectively at different temperatures. The long positron lifetime component, as shown in figure 2, decreases from 500 ps at 300 K to 402 ps at 711 K with a minimum value of 392 ps at approximately 640 K. This corresponds to 22% fractional increase of annihilation rate in the temperature range 300 – 640 K. The measurements were not repeated but the trend seems to be consistent with the measurements carried out by Elefteriades et al [21] although their range is 30 – 300 K.

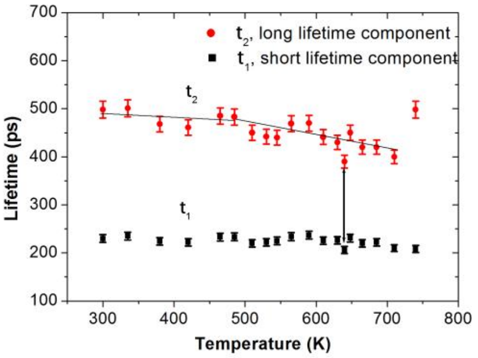

Figure 2. Short and long positron lifetime components in the temperature range 300 – 800 K

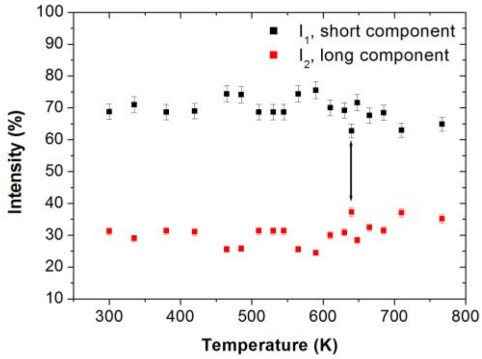

Figure 3. Measured intensities for long and short lifetime components

The "pick-off" annihilations usually take place where the positronium is in a quasistable state [16]. Therefore, in our case in which the temperature range 300 – 711 K is considered, delocalization and the "pick-off" annihilation process is considered. Pick-off process is one of the key processes which explain the observed fractional increase of about 22% in the annihilation rate (or a decrease in long lifetime component) in the temperature range 300 – 640 K. The o-Ps to 2γ radiation is strongly influenced by a high number of electrons more especially, the unpaired electrons of anionic interstitials, which strongly influence or enhance the pick-off process as described in equation 4. When the pick-off process takes place, the delocalized ortho-positronium lifetime is shortened as can be seen from the data in figure 2. It is clear that as the number of anion interstitials increases with temperature, the annihilation rate increases (corresponding to a decrease in the long lifetime component). On the other hand, a large percentage of positrons annihilate in the bulk. It is worth mentioning that although anionic vacancy can find it difficult to trap a positron, there is a probability that it can readily trap an electron (forming an F-center) [24] because of its positive potential which eventually attracts and annihilates with a positron. Barium is an electron-rich atom with 5p and 6s orbitals dominating the fluorine 2p and 2s electrons in terms of high electron density. Using annihilation equations 6-9, we deduce the following annihilation intensities shown in Table 1. The singlet state, p-Ps, which has a very short lifetime of about 125 ps, has 6% intensity whilst the triplet state o-Ps in general has a slightly higher intensity than p-Ps. The lifetime of o-Ps is greatly reduced by the pick-off process with one of the host atom electrons. The two positroniums are different in terms of relative spins between positron and electron within the positronium.

Table 1. Deduced intensities of positrons and positronium in barium fluoride

BaF2		Intensities
Para-positronium	pPs	0.06 ± 0.01
Ortho-positronium	oPs	0.07 ± 0.01
Free positrons	e^+_f	0.56 ± 0.01
Trapped positrons	e^+_t	0.31 ± 0.01

The suggestion is that in the temperature range 300 – 640 K the dominant process is due to the detrapping of previously localized positronium. The temperature range 640 – 711 K shows a slight increase in the annihilation rate corresponding to a decrease slight decrease in the longer lifetime component. Other contributions toward the positron short lifetime component emanate from the positrons annihilating with valence and core electrons of the individual atoms of the sample. This is achieved through solving Schrodinger equation separately for electrons and positron wavefunctions to obtain positron and electron densities. The annihilation rates are then calculated from

$$\lambda_j = \pi r_o^2 c \int dr \gamma^{GGA}(\boldsymbol{r}) n_+(\boldsymbol{r}) n_j(\boldsymbol{r}) \tag{10}$$

where r_0, c, γ^{GGA}, n_+ and n_j are classical radius of electron, speed of light, enhancement factor in the framework of generalized gradient approximation, positron density and electron density in state j respectively

Using the generalized gradient approximation, the positron wavefunction peaks around the barium atom and in the process increasing the annihilation rate probability as shown in figure 4 while positron density around the fluorine atom remains relatively low as shown in figure 5. The pick-off annihilation with valence electrons in barium increases while the annihilation with fluorine valence electrons decreases with temperature as shown in Table 2.

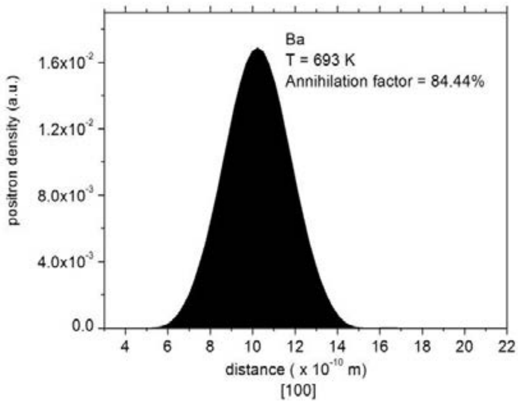

Figure 4. Positron wavefunction density peaks around the barium atom

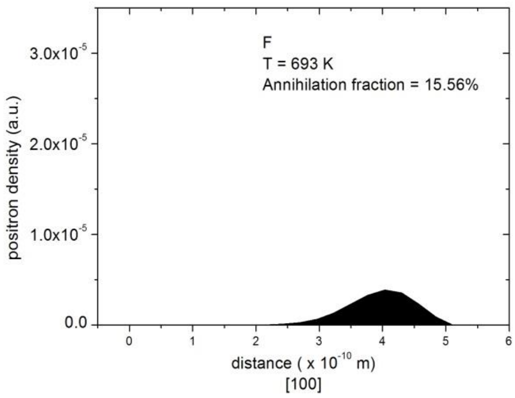

Figure 5. Positron wavefunction density around the fluorine atom

Table 2. Annihilation fractions and rates are calculated in the framework of a generalized gradient approximation.

BaF_2	λ_{core} (per ns)	λ_{5s} (per ns)	λ_{5p} (per ns)	λ_{6s} (per ns)	Annih. fraction (%)
F-vacancy 300 K					
Ba	0.0053	0.0721	0.6189	3.4563	84.36
F	0.7688				15.64
F-vacancy 580 K					
Ba	0.0051	0.0698	0.6019	3.4257	84.41
F	0.7578				15.59
F-vacancy 693 K					
Ba	0.0049	0.0684	0.5921	3.4084	84.44
F	0.7508				15.56
F-divacancy 693 K					
Ba	0.0052	0.0709	0.6138	3.486	86.57
F	0.6481				13.43

4. Conclusion

We find that the annihilation rate via pick-off annihilation in the temperature range 300 – 750 K primarily due annihilation of ortho-positronium positron with one of the valence electrons of the host atoms, is one of the dominant processes in barium fluoride and as a result the lifetome of o-Ps is highly reduced. The intensity of the long lifetime component is indeed attributed to the annihilation of a delocalized o-Ps through pick-off process. We are also of the view that more positron annihilation data at elevated temperatures greater than 750 K is needed in order to have a complete picture regarding the existence of positronium in barium fluoride since not much is covered of this material in the literature using positron annihilation spectroscopy in the temperatures greater than 600 K. The new proposed region is also of interest since it covers the excess specific heat anomaly in barium fluoride.

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