Alec T. Stewart (1925-2014) and Positronium

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Abstract. In this article I highlight the positronium-related work of the late Alec Stewart whose valuable contributions helped to establish the field of positron annihilation studies.

It is a great loss to us, both personally and professionally, that Professor Alec T. Stewart of Queen’s University, Kingston, Canada, passed away on July 28th, 2014. He began his research life with neutron scattering experiments, working with Professor Bertram Brockhouse at the Chalk River Laboratories in Canada [1-3]. He changed his research field to that of positron annihilation and subsequently established a method of angular correlation of annihilation radiation (ACAR) measurement to investigate positron and positronium (Ps) interactions with matter.

The one dimensional angular correlation of annihilation radiation (1D-ACAR) curve is a projection of the photon-pair momentum density, \( \rho_{2\gamma}(\vec{p}) \), onto one direction. For, say the \( x \)-direction, the curve, \( N(p_x) \), can be expressed as

\[
N(p_x) \propto \int \rho_{2\gamma}(\vec{p}) d\rho_y d\rho_z.
\]

Due to momentum conservation during the \( 2\gamma \) annihilation, \( \rho_{2\gamma}(\vec{p}) \) is, at the same time, the momentum density of the fully-interacting, annihilating spin-singlet electron-positron pair. Stewart studied the momentum dependent enhancement of the annihilation rate resulting in the bulge of the parabolic 1D-ACAR curves for alkali metals, and the effective mass of the positron due to the positron-electron and positron-phonon interactions which can be deduced from the smearing of the Fermi cut-off of the 1D-ACAR. I will not go into detail about this aspect of his work here, but rather concentrate on his interest in Ps.

The ACAR data for the annihilation of a valence electron of the medium with a positron without forming Ps (free positron annihilation) or with a positron bound in Ps (pickoff annihilation) is approximated by the sum of the momenta of the annihilating electron and positron pair. In the case of the self-annihilation of para-positronium (\( p \)-Ps), on the other hand, the relative momenta of the electron and the positron cancel and the ACAR data directly represents the translational total momentum, or the centre of mass momentum, of the Ps.

Ps may exist in either a non-localized or a localized state, depending on its interactions with the medium. When the Ps is in a localized state, \( \Phi_L(\vec{r}_e, \vec{r}_p) \), with \( \vec{r}_e \) and \( \vec{r}_p \) being the coordinates of the electron and the positron in the Ps, the momentum density is determined by the Fourier transform of the electron-positron contact amplitude of the wave function, \( \Phi_L(\vec{r}, \vec{r}) \), such that

\[
\rho_{2\gamma}(\vec{p}) \propto \left| \int \Phi_L(\vec{r}, \vec{r}) e^{-\vec{p}\cdot \vec{r}} d^3r \right|^2.
\]
The greater the localization, the wider the ACAR curve. It was suggested by Ferrell [4] in 1957 that Ps in liquid He was localized in a cavity which Ps itself created because of a strong repulsive exchange force between Ps and a He molecule. Later, Roellig and Kelly [5] estimated the size of the bubble in high density gaseous He from the pickoff annihilation rate of o-Ps. Soon after this, Stewart, with Briscoe and Choi performed a direct measurement of the bubble size in liquid He and other liquids using 1D-ACAR curves [6]. The curves had two components. The broad component was attributed to annihilation of the electrons of the liquid’s atoms and molecules and reflected their momentum distributions. The narrow component was attributed to the self-annihilation of p-Ps localized in a bubble. Stewart, Briscoe and Steimbacher later added further data which aided in forming a more comprehensive account of the observation [7].

If it is assumed that Ps is localized in a spherical square well of radius $R$, the total energy of the bubble state, $E_{\text{tot}}$, is written as

$$E_{\text{tot}} = E_{\text{cm}}(R) + \frac{4}{3}\pi R^3 p + 4\pi R^2 \sigma,$$

where $E_{\text{cm}}(R)$ is the centre of mass zero-point energy of the localized Ps, the second term corresponds to the work required to remove atoms from the bubble volume in a liquid of pressure $p$, and the last term is the bubble surface energy with specific surface energy $\sigma$. If we assume an infinite well depth, the electron-positron contact amplitude of the Ps is

$$\phi_L(\vec{r}, \vec{r}_p) = \frac{A \sin \sqrt{ \frac{2mE_{\text{cm}}}{\hbar^2} r}}{r}$$

where

$$E_{\text{cm}} = \frac{1}{2m} \left( \frac{\hbar \pi}{R} \right)^2.$$

It is a straightforward procedure to calculate the 1D-ACAR curve $N(p_x, p_z)$ with $\rho_{2\gamma}(\vec{p})$ for this contact amplitude. The full-width-at-half-maximum $\theta_{\text{FWHM}}$ in milliradians has a relation with the radius of the cavity as

$$R = 1.659 \frac{\theta_{\text{FWHM}}}{\text{nm}}.$$

The experimentally obtained value was $\theta_{\text{FWHM}} = 0.86$ milliradians and thus $R = 1.93 \pm 0.13$ nm. The equilibrium bubble radius estimated by minimizing $E_{\text{tot}}$ with respect to the bubble radius gives an equation for $R$, solving which, with a surface tension of 0.096 dyn cm$^{-1}$, gives the value $R = 1.88$ nm. (The numbers cited here are from ref. [7] rather than [6].) The good agreement indicates that the method used for measuring the bubble size is feasible.

The analysis of the long positron lifetime [5] verified the formation of bubbles by long-lived o-Ps. The ACAR study showed more directly that even a short-lived p-Ps atom creates a bubble around it before annihilation. This study also showed that the formation of the bubble is completed in a time shorter than 100ps.

When the Ps is in a delocalized, quasi free state, its wave function, $\Phi_{\tilde{k}}(\vec{r}, \vec{r}_p)$, is characterized by a translational wave vector $\tilde{k}$. If we neglect a periodic structure of the medium as in a crystal, the electron-positron contact amplitude is a plane wave so that its Fourier transform is a delta function $\delta(h \tilde{k} - \vec{p})$ and thus

$$\rho_{2\gamma}(\vec{p}) \propto \sum_{\tilde{k}} n_{\tilde{k}} \left| \int \Phi_{\tilde{k}}(\vec{r}, \vec{r}) e^{-i\vec{p} \cdot \vec{r}} d^3r \right|^2 = \sum_{\tilde{k}} n_{\tilde{k}} \delta(hk - p) = n_{\tilde{p}/\hbar}.$$

Since the thermalized Ps atoms follow the Boltzmann distribution, $n_{\tilde{p}/\hbar} \propto \exp \left( \frac{-p^2}{2m^* k_B T} \right)$, where $m^*$ is the effective mass of the Ps and $k_B$ is the Boltzmann constant. Thus, the width of the component is not
determined by the width of the electron-positron contact amplitude, but the Boltzmann distribution of the states and hence temperature dependence is expected. When the medium is a crystal, Ps is actually in a Bloch state. The wave function is modulated by the periodicity of the crystal field that the momentum density has a non-zero component at the momenta corresponding to the reciprocal lattice vectors

$$\Phi_{\vec{k}}(\vec{r}_1, \vec{r}_2) \propto \sum_{\vec{G}} a_{\vec{G}}(\vec{k}) e^{i(\vec{k} + \vec{G}) \cdot \vec{r}},$$

where $\vec{G}$ is the reciprocal vectors and $a_{\vec{G}}(\vec{k})$ is the amplitude of the higher momentum components. Then the momentum density is written as

$$\rho_{2y}(\vec{p}) \propto \sum_{\vec{G}} n\left(\frac{\vec{p}}{\hbar} - \vec{G}\right) \left| a_{\vec{G}}(\vec{p}/\hbar - \vec{G})\right|^2.$$

Here $\vec{G}$ is the reciprocal lattice vector which projects $\vec{p}/\hbar - \vec{G}$ into the first Brillouin zone and again $n\left(\frac{\vec{p}}{\hbar} - \vec{G}\right)$ is the Boltzmann distribution. The resulting 1D-ACAR curve is a series of Gaussian curves centred at the projection of $\vec{G}$ onto the x direction as experimentally shown by Brandt et al. [8] and Greenberger et al. [9]. Stewart and co-workers [10] investigated the Ps components in crystalline quartz and showed that the orthogonality of the electron part of the Ps wave function and the wave function of the valence electrons in the crystal, or the indistinguishability of the electrons explains the magnitude of the higher momentum components.

Following this, Kubica and Stewart measured the 1D-ACAR for quartz at 4.2K [11]. The width was quite small, corresponding to a distribution at 10K if the effective mass was $2m_e$, where $m_e$ is the mass of the electron in vacuum. The effective mass is known to be greater than $2m_e$ [10,12,13] by analysing the temperature dependence of the width. Therefore, it is safe to say that the Ps in quartz at 4.2K is thermalized within the experimental uncertainty. Thus also, in a solid, thermalization time for Ps is shorter than the lifetime of $p$-Ps.

I worked with Alec Stewart for two years (1980-82) at the Queen’s University, Canada, where I studied positron-electron and positron-phonon interactions in potassium [14]. Several years before this, I had found that Ps exists in alkali halides in delocalized Bloch states at low temperatures and self-trapped at higher temperatures including room temperature [15]. This explained why the 1D-ACAR curves for alkali halides systematically measured by Stewart and Pope at room temperature did not show distinct narrow peaks. If we look at the curves carefully, they are more triangular in form than typical 1D-ACAR curves for metals and insulators. The broadened narrow component due to the $p$-Ps being localized in a much smaller volume than that in liquid inert gases gives rise to the characteristic shapes. Alec was interested in this phenomenon and we conducted measurements on the temperature dependence of the positron lifetime in NaF and NaCl [16]. The results were consistent with the picture that Ps which were free at low temperatures underwent transition to a self-trapped state at higher temperatures, and that the accompanying deformation was of a dilation type.

After returning to Japan, I conducted some joint research with Alec and others on the temperature dependence of the Ps momentum density in SiO$_2$, ice and BaF$_2$ [17], as well as 2D-ACAR studies of alkali metals [18] and a Compton profile study on Li [19].

I also collaborated with Alec Stewart on the ACAR study of the thermalisation of Ps in gases, which evolved from my experience in Kingston. On considering the relationship between the momentum density and the shape of 1D-ACAR curve, I noticed that an isotropic momentum density always gave a convexed 1D-ACAR curve and the extreme case was a distribution only on a thin spherical shell giving a rectangular curve. Thus, when I measured the 1D-ACAR curve of silica aerogel and found that it had a rather flat top, I reasoned that it must consist of a Gaussian-like broad component from the valence electrons of amorphous silica and a more or less rectangular component from $p$-Ps emitted isotropically in all directions from the grain with an energy determined by the negative work function [20]. These initial findings were then followed by our collaboration with Alec...
on the thermalisation of positronium via collisions with the silica aerogel grain surfaces and a variety of gas molecules [21].

Prof. Alec Stewart’s enthusiasm for physics was undeniable and his approach to understanding physical phenomena was to seek the most straightforward and essential models. Doing research with him was immensely pleasurable and I learned much from him during our conversations together.

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