

ANGULAR CORRELATION STUDIES IN NOBLE GASES

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ABSTRACT

There has been a recent revival of interest in the measurement of angular correlation of annihilation photons from the decay of positrons and positronium in gases. This revival has been stimulated by the possibility offered by the technique (a) to shed new light on the apparently low positronium formation fraction in the heavier noble gases; and (b) to provide information on positronium quenching processes in gases such as oxygen. There is also the potential for learning about positronium slowing down in gases.

This review will focus on experimental noble gas work conducted in the U.K. and Japan, and considers what new information has been, and may be, gained from these studies.

INTRODUCTION

Correct description of the angular correlation between gamma photons emitted upon the annihilation of positrons by atomic electrons, $I(\theta)$ has long been recognised as a stringent test of theories describing positron-atom interactions. For this reason many theoretical papers report calculations of scattering cross sections also include $I(\theta)$ and the annihilation cross sections resulting from the formalism employed.¹⁻⁵ Some results for $I(\theta)$ for the noble gases are depicted in figure 1.

However, experimental studies of $I(\theta)$ have been literally few and far between, although the technique has been widely used for condensed matter research. Page and coworkers published a short

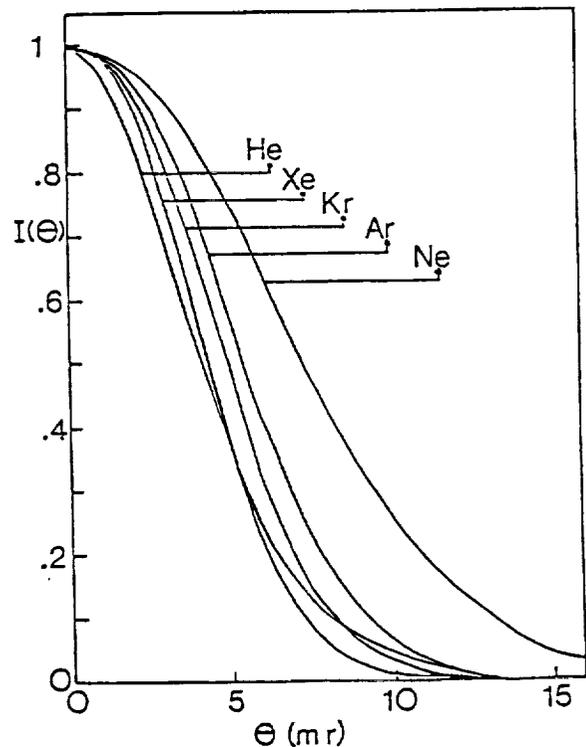


Figure 1

Theoretical angular correlation results from refs 1-5. Hummerston's model H5 is plotted for He; the calculations of Drachman and McEacran et al for He lie between the plots for Kr and Xe.

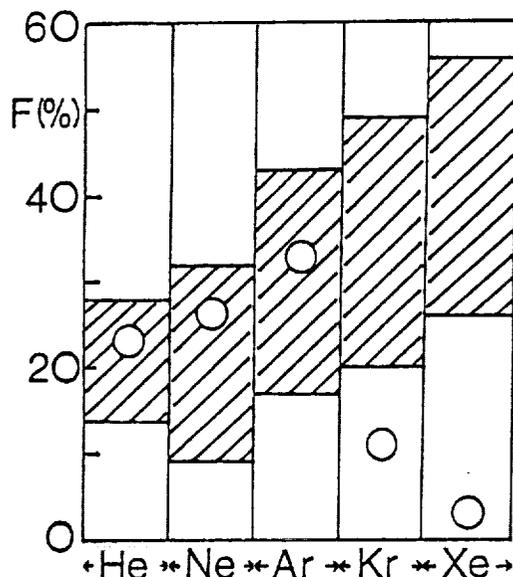


Figure 2
Measured Ps formation fractions for the noble gases (from lifetime measurements^{14,15}). Shaded areas depict the range of values predicted by the Ore model.

series of papers in the late 1950's in which the angular correlation technique was used to study positronium quenching and the effects of high electric fields on positronium formation^{6,7}, but their work in noble gases was restricted to argon. Until recently the only other work in gases has focussed on positronium chemistry in molecular gases⁸⁻¹², and measurements in liquid noble gases which were used in comparisons with the theoretical calculations referred to above.¹³

The recent, albeit small, revival in experimental work in this area has been in part stimulated by the intriguing results from positron lifetime measurements in the noble gases that the amount of ortho-positronium (o-Ps) formed in krypton and xenon appears to be much lower than is expected from the Ore model predictions. The results, summarised in figure 2, were originally reported by Coleman et al¹⁴ and Wright et al¹⁵, and have been discussed in review papers including those of Charlton¹⁶ and Griffiths.¹⁷ They were pursued further by Wright et al,¹⁸ who

reported the observation of fast lifetime components in spectra for both Kr and Xe. An example is shown in figure 3. Wright et al attributed the fast components to resonant capture of o-Ps into short-lived bound states by the Kr and Xe atoms, the measured fast lifetimes representing the capture rather than the annihilation rates. An alternative picture proposed by Jacobsen involves the spin conversion of fast o-Ps into p-Ps, whose decay is responsible for the fast components. Wright et al suggested that the models describing the mechanism for Ps formation and decay in the heavy noble gases could be tested by angular correlation measurements, and experiments were later performed on the two-dimensional angular correlation spectrometer at the University of East Anglia.¹⁹ The hope here was that the p-Ps component - difficult to identify

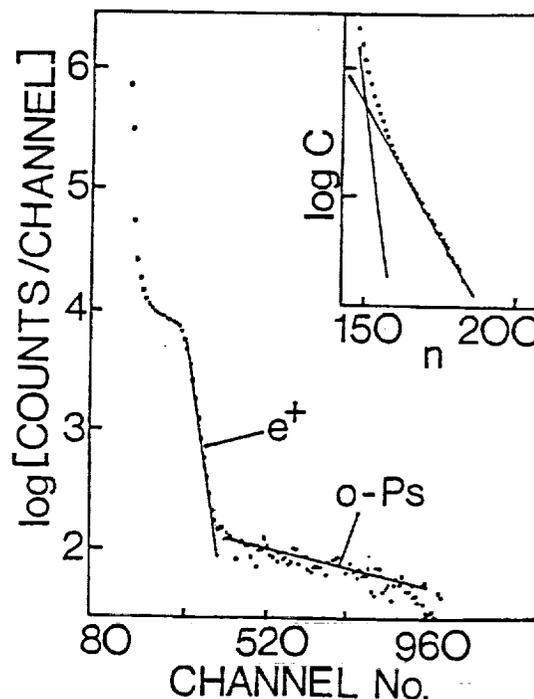


Figure 3
Lifetime spectrum for Xe at 9.54 amagat and 297K.¹⁸ The inset shows two fast components attributed by Wright et al to resonant capture of o-Ps into atomic bound states.

directly in lifetime spectra - could be seen in $I(\theta)$ measurements, and thus provide alternative information on the amount of positronium formed.

During the same period Hyodo and coworkers have also used the (one-dimensional) angular correlation technique to study positron-gas annihilation, using silica aerogel to stop enough positrons in a thin region to allow high-resolution angular correlation measurements to be made with good statistics, irrespective of gas pressure. In addition to a number of measurements of positronium quenching in molecular gases²⁰, these researchers have also attacked the problem of Ps formation in xenon^{21,22} and have obtained angular correlation data for He, Ne, Ar, Kr and Xe.²³

We shall now consider in more detail the experimental results for the noble gases. Their contributions to date to the understanding of positronium formation and slowing down in noble gases will be assessed, in addition to any new information they provide on the basic positron-atom annihilation process.

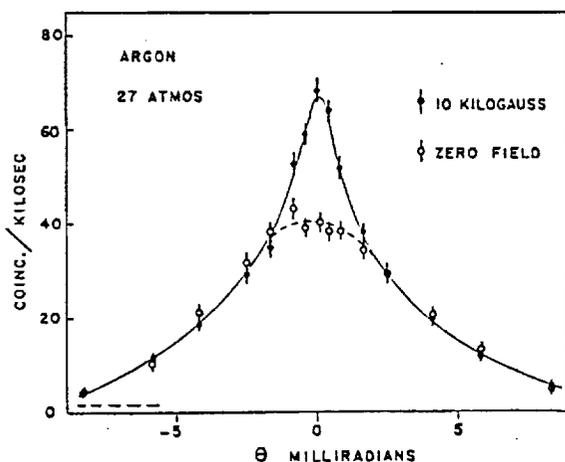


Figure 4
1D angular correlation spectra measured by Heinberg and Page.⁹

EARLY MEASUREMENTS

Figure 4 shows the result of Heinberg and Page⁹ for argon. The gas was at high pressure - 27 atmospheres - and measurements (one-dimensional angular correlation) were taken with and without a 1 T magnetic field present. The results are interesting because they show the appearance of a narrow component with the application of the magnetic field. This the authors correctly assigned to "thermalised" positronium, the relatively long-lived $m=0$ o-Ps/p-Ps mixed state. Before proceeding further, then, it may be useful to consider briefly the observables in the angular correlation measurements with which we are concerned here.

OBSERVABLES

1. Free positron-atom annihilation.

Here the positron is assumed to be thermalised; widths (FWHM) of $I(\theta)$ curves range from about 8 to 12 mrad, reflecting the mean momenta of electrons available for annihilation.

3. Mixed-state positronium decay.

Angular correlation measurements are often performed with the sample in a high magnetic field, usually used to transport positrons over a few cm from the radioactive source to the sample, so that the source is out of sight of the detectors placed at either side.

In a magnetic field B , the $m=0$ triplet substate of positronium mixes with the singlet state. The decay rates of the mixed states, λ_1 and λ_2 , are given by the expressions

$$\lambda_1 = (1 - x^2)\lambda_S + x^2\lambda_T$$

and

$$\lambda_2 = x^2\lambda_S + (1 - x^2)\lambda_T$$

where λ_S and λ_T are the annihilation rates for singlet and triplet decay, respectively (at low gas pressures - 3×10^9 and 7.14×10^6 s⁻¹, respectively) and $x = 2e\hbar\omega/mc^2\omega$.

At $B = 0.3T$, the field used in the UEA experiments described later, $x^2 = 0.012$ and $\lambda_1 = 7.9 \times 10^9 s^{-1}$ ($\approx \lambda_3$) and $\lambda_2^{-1} = 9.7 ns$. There is $\sim 91\%$ mixing at $0.3T$ and the states decay via two-gamma emission, so that the annihilation photons contribute to $I(\theta)$ spectra.

State 1 is almost identical to unmixed $p=Ps$. Let us assume that no positronium formed above the upper limit of the Ore Gap - the atomic ionisation threshold - i.e., having kinetic energy greater than its binding energy ($6.0eV$) - survives its next collision. Then we can say that at time zero we have a $p=Ps$ energy distribution ranging from 0 to $6.0eV$. Because the mean life of $p=Ps$ in the gas is $\sim 125ps$ significant slowing down prior to annihilation is unlikely, and component peaks of similar width ($\sim 10mrad$) should be present in spectra for all the noble gases, of intensity corresponding to one quarter of the total positronium decays.

State 2 is relatively very long-lived positronium, and as such can lose much of its energy in elastic collisions with gas atoms prior to annihilation. This results in a narrower component on the measured spectra, such as that of Heinberg and Page; the lighter the gas atoms the more efficient the slowing down and the narrower the component. (The gas density is so high in figure 4 that almost 100% thermalisation is likely.) One can arrive at a crude estimate the degree of slowing down by assuming that the mixed-state positronium atoms lose $2m/M$ of their energy on each elastic collision. Then, one can show that after one mean lifetime (say $10ns$) a $6.0eV$ Ps atom will have slowed to $[0.33 + 6.2/Z]^{-2} eV$ in one atmosphere of a noble gas of atomic number Z and elastic scattering cross section of $5 \times 10^{-16} cm^2$. For helium this yields $0.27eV$ (so thermalisation is almost certain), whereas in xenon the energy after $10ns$ is only $5.4eV$ - almost no slowing down at all.

Note that the three-gamma decay of $o=Ps$ atoms is not detected, as the technique

relies on the detection of two almost anticollinear gamma rays; therefore in a strong magnetic field only half of the positronium formed can contribute to an angular correlation spectrum (i.e., that in states 1 and 2 above).

SILICA AEROGEL MEASUREMENTS

Hyodo and coworkers have performed a series of experiments in noble and molecular gases with a high-resolution long-slit one-dimensional angular correlation apparatus described in reference 20. The annihilation signal rate was increased significantly, for gas pressures of one atmosphere or less,

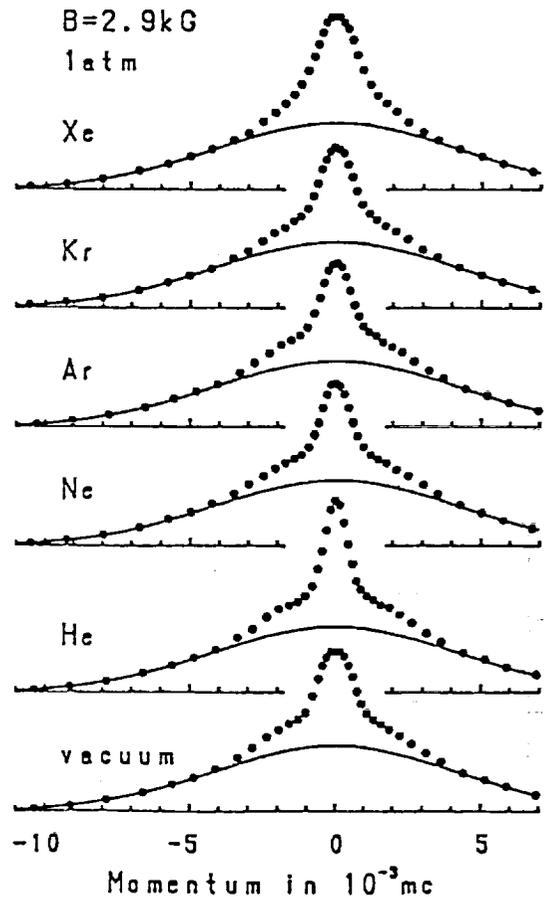


Figure 5
1D spectra of Kakimoto et al.²² for noble gases (with aerogel moderator), exhibiting narrow components corresponding to the decay of mixed-state Ps.

by stopping the positrons in silica aerogel (an aggregate of amorphous SiO_2 fine particle grains). This, naturally, gives rise to signal arising from interactions with the aerogel, which is measured separately and subtracted from the data.

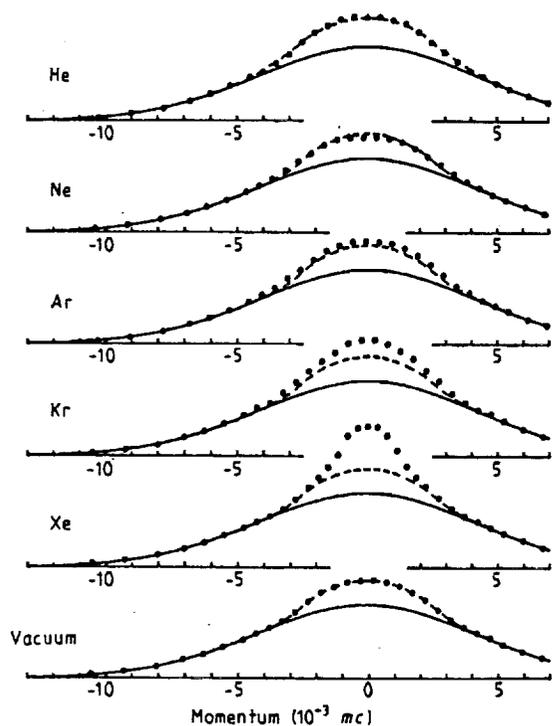


Figure 6

1D spectra of Kakimoto and Hyodo²¹ for noble gases, demonstrating the increasing Ps formation probabilities as Z increases, in contradiction with the results shown in Fig.2 from lifetime measurements.

Slowing down of positronium

Figure 5, taken from reference 22, exhibits narrow peaks attributed to the long-lived ("state 2") Ps referred to above. The broadening of the peak as Z increases is consistent with the decreasing slowing-down efficiencies discussed earlier. The peak in vacuum is due to Ps formed in the grains.

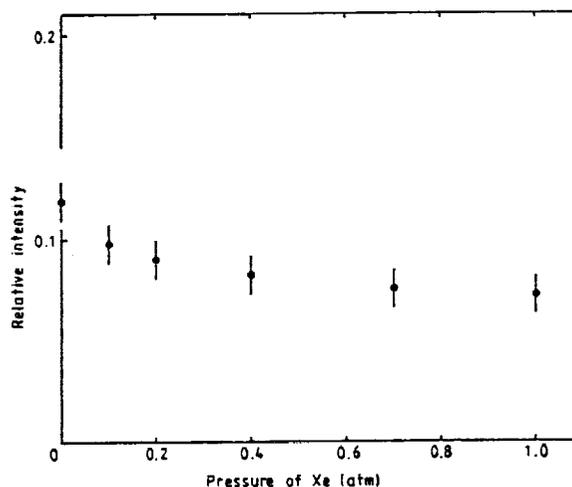


Figure 7

Relative intensity of mixed-state Ps in Xe as a function of gas pressure (from ref.22). The decrease in intensity is interpreted as evidence of strong o-Ps quenching in Xe.

Positronium formation in xenon

The same authors have concluded from their measurements in the noble gases and in xenon at different pressures and in a magnetic field (ref.21) that (a) there is substantial Ps formation in Kr and Xe (see, for example, figure 6), and that (b) part of the long-lived Ps is quenched by xenon (figure 7). This latter observation, say the authors, substantiates the model proposed by Wright et al based on the formation of Ps-xenon resonance states during Ps slowing down.

TWO-DIMENSIONAL ANGULAR CORRELATION MEASUREMENTS

Measurements of two-dimensional angular correlation spectra for positrons and positronium annihilating in pure He, Ne, Ar, Kr and Xe, and in He-Xe mixtures, have been performed using the Anger-camera based system developed and built at the University of East Anglia.²² This work was in collaboration with the positron group at University College London.

The spectrometer, used for many years to study electron momentum densities in metals and alloys, was adapted for the study of gases by (a) removing the existing sample holder, (b) installing a needle valve for the introduction of gases, and (c) using redesigned lead collimators which reduced the probability for detecting scattered gamma rays and defined well the viewed gas volume ($\approx 5\text{mm} \times 5\text{mm} \times 5\text{mm}$). No inner gas cell was used; it was found from early trials that premature annihilations from the cell walls and windows, especially of energetic Ps, could distort the measured spectra. Thus, only annihilation events in the gases were recorded; the probability of o-Ps reaching and be annihilated at the side walls of the entire sample chamber, within sight of the cameras, was found to be negligible. A constant magnetic field of 0.8T is used to transport positrons from the source to the viewed target volume, and so the mixed state Ps atoms discussed above are present.

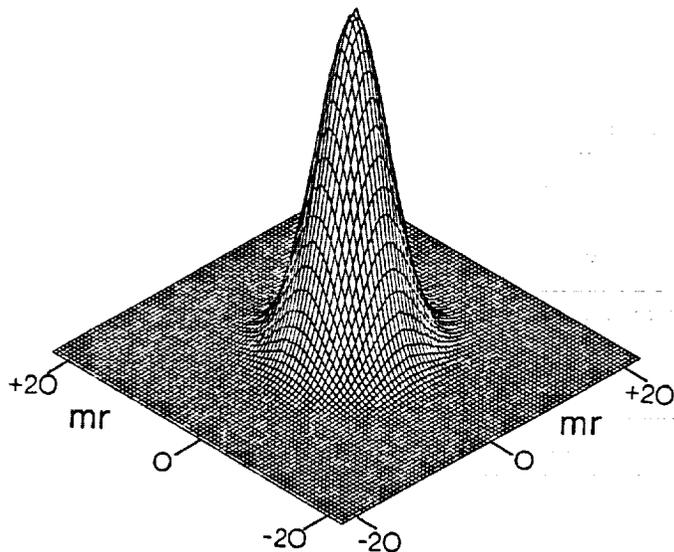


Figure 8

2D angular correlation result for argon at 1 atm, typical of those collected with the UEA spectrometer. The spectra are cylindrically symmetrical but a central cut (or angular average) allows better definition of component peaks than the equivalent 1D spectrum.

Unlike Hyodo et al, these measurements do not have the benefit of aerogel to stop positrons and increase statistics; by the same token, however, there is no aerogel background signal. Each run took several days (the lower the stopping power of the gas, the longer the run needed) and the cameras were moved in to 5m either side of the source, with an unavoidable loss of resolution (measured by recording spectra for a sample of quartz of suitable size: $\Delta\theta \approx 3.4\text{mr}$). A pressure of one atmosphere was maintained for each gas studied, this being the maximum allowable in the sample chamber. A typical two-dimensional result is shown in figure 8; as the spectra are cylindrically symmetrical there is strictly no need for two-(as opposed to one-) dimensional measurements. However, if a central cut through the 2D peak is taken - or, better still, a cylindrical average is derived - resolution of different components is more readily achieved than with a 1D spectrum. However, after extracting components from the peak, one then has to normalise intensities by first multiplying by the peak width. (For example, the ratio of the volume of revolution of a Gaussian distribution to its area is proportional to its standard deviation.)

Angularly-averaged results for helium and xenon are shown in figure 9. The most important difference is the presence of a separable narrow component in the He spectrum, again corresponding to "state 2" positronium reduced almost to thermal energies by collisions with the light He atoms. In xenon it is not possible to identify a narrow component, and - unfortunately - this means that without careful modelling the data cannot tell us directly whether there is a Ps component present or not. A possibility here is the interpretation of a series of He/Xe mixture results; as the Ore Gaps of the two gases do not overlap, and the scattering cross sections for Xe should swamp those for He, it is hoped that the He atoms act primarily as moderators for the Ps formed in the Xe. Indeed, a narrow component is seen in the mixture runs,

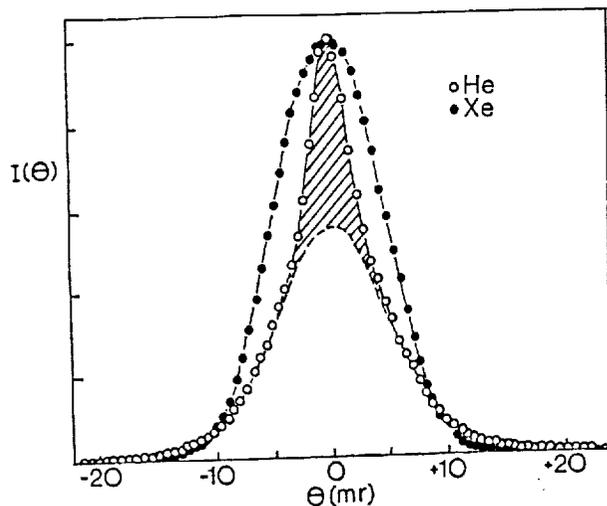


Figure 9

Central cuts through 2D spectra for He and Xe. The He spectrum is separated into positron/p-Ps and the narrower (shaded) "state 2" Ps components. The latter is of width close to the resolution of the spectrometer, and of intensity consistent with the Ps fraction shown in Fig.2. The width of the unshaded component is closer to the those calculated by Drachman and McEachran et al, Humberston's model DB, and the liquid He results of ref.13, than Humberston's H5 calculation (which is plotted in Fig.1).

and future analysis may yield more information on Ps formation in xenon.

Figure 10 shows a three-Gaussian fit to the angularly-averaged argon spectrum. The positron component is computed to be 80% of the spectrum (rotated about the vertical through its centre) and of width 11.5 mr, the state 1 p-Ps-like component 10% (width 10.2mr) and the state 2 long-lived Ps component 10% (width 5.7mr). Remembering that only half of the Ps formed can be observed on the spectrum, the positron result is consistent with 33% Ps formation in argon; its shape can be compared with the $I(\theta)$ calculation of McEachran et al³ and the liquid argon result of Briscoe et al.¹³ (see figure 11). Clearly, very

satisfactory agreement is obtained. Finally, the argon spectrum tells us that as the narrow component is still relatively wide, the state 2 Ps atoms are far from being thermalised through collisions with the argon atoms - perhaps still retaining, on average, -4eV at annihilation. This figure is reasonably consistent with the first-order calculations discussed in the preceding section.

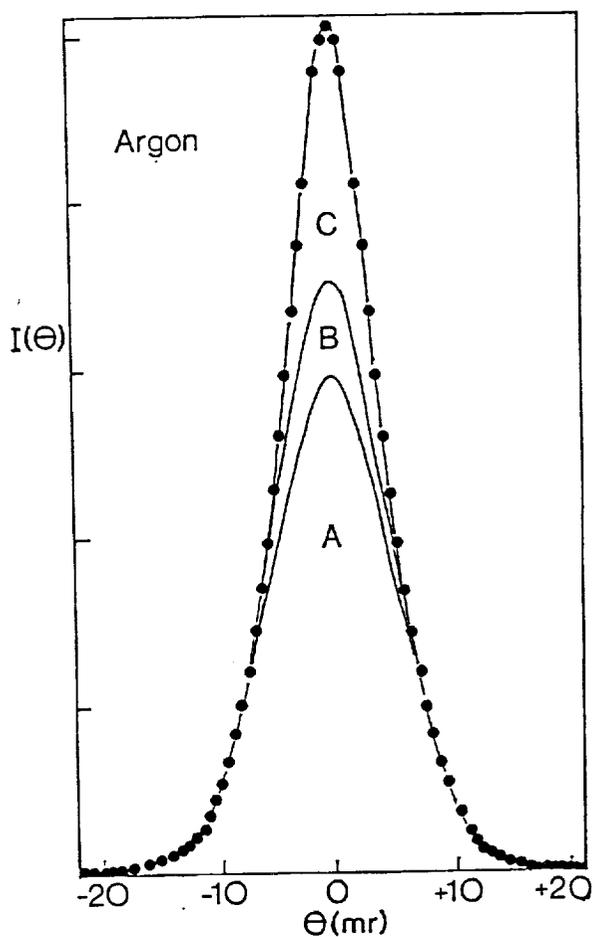


Figure 10

2D results for argon, cylindrically averaged. Gas pressure 1 atmosphere, $B = 0.8T$. Component A is due to free positron annihilation, B is p-Ps (state 1) and C is state 2 Ps.

CONCLUSIONS

Returning to the main motivations behind current activity in this field:

1. Positronium formation in xenon

Hyodo and coworkers claim that their results are consistent with the model of Wright et al, i.e. that positronium forms as expected (from Ore Gap considerations) but that o-Ps atoms are captured efficiently into an atomic bound state. Certainly positronium formation cross sections in Xe are large²⁵ and there is no reason to expect that positronium should not be formed in positron-xenon collisions. The 2D measurements at East Anglia, at the time of writing, do not provide us with clear evidence of Ps formation in Xe; hopes of unconstrained multicomponent fits to Xe spectra such as that illustrated in 9 must be considered to be remote if not impossible. It may be that measurements in pure Kr and Xe at much higher densities, such as those used by Heinberg and Page, would exhibit discernable features. However, there is some hope that the He-Xe mixture data may provide some relevant information, and we await further careful analyses of these data.

2. Positronium Slowing Down

Both Japanese and British groups appear to be able to provide information on positronium slowing down by elastic collisions with atoms, and with careful modelling one may even hope to gain some information on the order of magnitude of the Ps-atom scattering cross sections. The relative widths of the mixed-state Ps components should at least provide a comparison between those noble gases for which this component can be identified. Direct measurement of Ps-atom scattering cross sections is planned at University College London.

Angular correlation measurements in both ³He and ⁴He were performed as part of the UEA-UCL collaboration, in the hope that comparison of the widths of the state 2 positronium component would reflect only the mass difference between

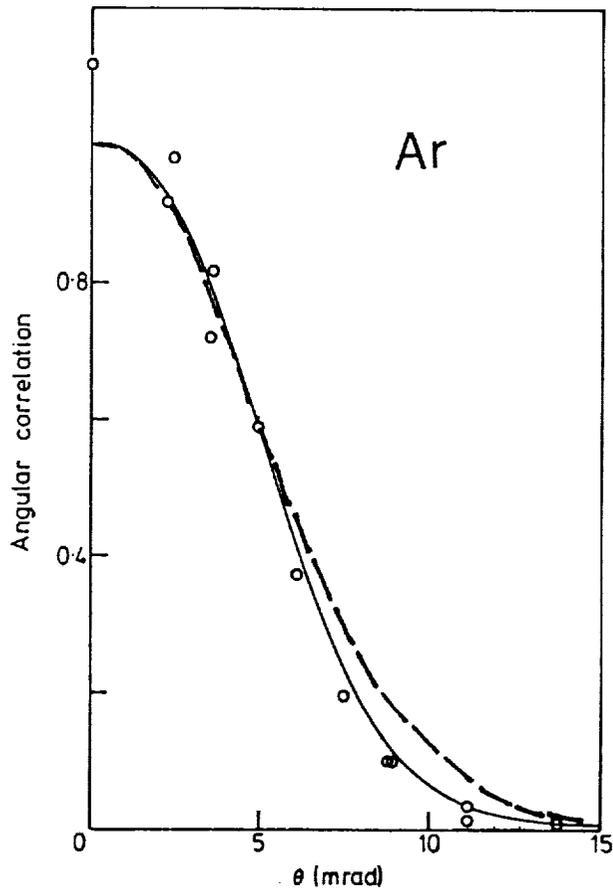


Figure 11

Calculated $I(\theta)$ from ref 4 (solid line) together with experimental results for liquid argon (ref 13) and the gaussian positron component from Fig. 10 (broken line).

the helium atoms; this would then allow assignment of a mean Ps-atom cross section in the few-eV region. Unfortunately, however, both ³He and ⁴He are efficient Ps moderators; the widths of both state 2 Ps peaks are very close to the system resolution, implying near-thermalisation of Ps in both gases.

3. $I(\theta)$ for Positron-Atom Annihilation

There is now hope that new experimental values for annihilation in the gaseous state will be available for direct comparison with theoretical calculations. The UEA-UCL results for He, Ne and Ar will certainly provide $I(\theta)$ for positron annihilation, as figure 11 illustrates.

work is continuing in Japan and further analysis of the 2D pure gas and gas mixture data is being pursued.

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