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13. ABSTRACT (Maximum 200 words) <p>The objective of this research was to study the interaction of positrons with molecules at energies below the threshold for positronium atom formation. While there is evidence that positrons bind, or form long-lived resonances with large atoms and molecules, this phenomenon is not understood. This phenomenon has a number of potentially important scientific and technological applications, including aspects of atomic and molecular physics, the formation of positive ions for mass spectrometry, and gamma ray astronomy. Many facets of the interaction of low-energy positrons with molecules were illuminated, including the scaling of positron annihilation rate with molecular ionization potential the identification of other trends as a function of chemical structure. Spectra of the gamma ray radiation resulting when a positron annihilates on a molecule were also measured. The measurements indicate that the site of positron binding is on the C-H bond in hydrocarbon molecules and on the fluorine atoms in fluorine compounds. The measured annihilation rates on large aromatics indicate that these molecules are likely to be an important source of the annihilation radiation observed from the interstellar medium.</p>			
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**Final Technical Report****ONR Grant #N00014-89-J-1769****Positron-Molecule Interactions****October 1, 1989 to December 31, 1993**

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Grant Officer: Peter Reynolds

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*I. Overview of the research program* This research exploited the techniques we have developed to accumulate, store and cool low energy positrons. The objective of the work was to study the interaction of positrons with molecules at energies below the threshold for positronium atom formation. There is evidence that positrons bind, or form long-lived resonances with large atoms and molecules. This phenomenon is not understood, yet it has a number of scientific and technological applications for atomic and molecular physics and for gamma ray astronomy. Other potential applications include the formation of positive ions for mass spectrometry and the study of atomic and molecular clusters, both in the laboratory and for astrophysical applications.

*II. Operation of the positron trap.* The work began with studies of the interaction of positrons with molecular nitrogen. The goal of this work was to understand these interactions sufficiently well to optimize the operation of our positron trapping scheme. This scheme uses inelastic scattering from nitrogen as a mechanism to trap and cool the positrons. The positron trap employs three stages of this process, so any inefficiency enters as the third power of the loss rate.

Through a series of improvements, we were able to find that the optimum efficiency was obtained when the incident positron energy was sufficient to excite the  $N_2$  electronically. This discovery was counter to the conventional wisdom, which indicated that, at these energies, the loss due to positronium atom formation would be deleterious. It had been thought that the optimum

strategy would be to work, where possible, at lower energies where vibrational modes of the nitrogen could be excited, thereby avoiding the loss due to positronium formation. In contrast, we found that, at about 8 to 9 eV incident positron energy, there is a sizable cross section for electronic excitation of the nitrogen, while the cross section for positronium atom formation at these energies is much smaller. We were eventually able to tune all three stages of the trap to operate in this manner, thereby obtaining an overall trapping efficiency of positrons of 40% for slow positrons. This was a factor of 7 improvement from the maximum efficiency at the beginning of the Grant, and approximately 50% greater than the largest value that we thought was possible under ideal circumstances using the nitrogen buffer gas scheme. The trap holds the record for slow positron trapping efficiency.

*III. Annihilation rates in xenon.* An early project in the Grant was to study the interaction of positrons with atomic Xe. The annihilation rate of positrons in Xe had been studied previously in dense gases. The previous measurements differed by a factor of from 4 to 6 in the annihilation rate,  $\Gamma$ . We were able to measure  $\Gamma$  in the tenuous gas density of the positron trap. The difficulty with the previous measurements, was that in dense gases, it was thought that atomic clusters might be forming around the positron, thereby qualitatively altering the physics. We were able to make a definitive measurement and clear up this issue.

*IV. Annihilation rate studies for large molecules.* One of the main thrusts of our work was a systematic study of positron interactions with various chemical species. While this study is still continuing, much progress has been made, and the results have generated a number of important insights. We extended our previous measurements of alkane molecules and began work on substituted alkanes, such as chlorocarbons, fluorocarbons, and bromocarbons. As a result of this work, we discovered that, while the annihilation rate,  $\Gamma$ , did not fit on a universal curve when plotted as a function of traditional quantities such as molecular charge or molecular polarizability, the data did fit a universal curve when plotted as a function of the ionization potential,  $I$ , of the molecule. In particular, we discovered that the  $\ln(\Gamma)$  is a linear function of  $(I - 6.8 \text{ eV})^{-1}$ . [The quantity 6.8 eV is the binding energy of a positronium atom.] This dependence is not yet understood theoretically, but the problem is actively being pursued. The observed dependence, might indicate a physical picture in which a positronium atom moves in the field of a positive ion.

*V. Studies of aromatic molecules and implications for astrophysics.* We began systematic studies of annihilation rates for aromatic molecules. One of the first observations was that  $\Gamma$  in benzene is smaller by a factor of 6 than that observed in hexane. (See below for further studies of this effect.) However, as more rings are added to make larger aromatic molecules,  $\Gamma$  increases by a factor of 10 with each ring. Thus, while benzene has  $\Gamma = 18,000$ , anthracene corresponds to  $\Gamma = 4,400,000$ . Here, we quote  $\Gamma$  in units of  $Z_{\text{eff}}$ , which is the measured annihilation rate, relative to that for a gas of uncorrelated electrons with the average density corresponding to the total number of electrons in the system (i.e., as if there were no correlation of the spatial positions of the electrons).

We quickly realized that the observation of the large annihilation rates of the larger aromatic molecules has important implications for gamma ray astronomy. Positron annihilation provides the strongest astrophysical source of gamma-ray line radiation. Furthermore, it has the added utility that the attenuation of these gamma rays by matter is much smaller than other forms of electromagnetic radiation, such as infrared and visible photons and x rays. Consequently, the annihilation radiation provides an important source of information concerning astrophysical phenomena.

The interstellar medium (ISM) contains polycyclic aromatic hydrocarbons (PAH's), such as anthracene and pyrene (the latter a four-ring aromatic). We estimate that, when one takes into account the relative abundances of atomic hydrogen and the PAH's in the ISM, annihilation on the PAH's is approximately as likely as annihilation on H. Thus, the gamma radiation from the ISM will contain information concerning the PAH's, and neglect of these effects is likely to lead to incorrect interpretations of the annihilation radiation from the ISM. As discussed below, we later began the first studies of the energy spectra of annihilation radiation from aromatic molecules. It is likely that this same enhancement in annihilation rate will be observed for atomic clusters and dust grains as we have observed for large molecules. Thus, the annihilation radiation can provide information about these materials in the ISM potentially also.

*VI. Effects of double bonds on annihilation rates.* Motivated by the benzene measurements, which indicated that the annihilation rates in hexane and benzene were qualitatively different, we began a study of alkene molecules -- i.e., alkane-like structures containing double bonds. This work and the studies of aromatics resulted in the discovery that the scaling of  $\Gamma$  with

$(I - 6.8 \text{ eV})^{-1}$  does not work for molecules with double bonds, when a value of  $I$  is assumed that corresponds to the lowest ionization potential. However, this scaling is obeyed, at least approximately, if the ionization potential of a sigma bond in the molecule is used.

*VII. The first gamma ray experiments.* The next phase of the work entailed making the first systematic studies of the energy spectra of annihilation gamma rays resulting from positron annihilation on molecules. Prior to our work, only one molecule had been studied -- molecular hydrogen was studied in a *tour-de-force* experiment, motivated by its importance for astrophysics.

We borrowed a gamma ray detector from the gamma ray astronomy group at UCSD to make these measurements. The spectra give information about the momentum distribution of the electrons with which the positron annihilates. This momentum distribution, in turn, provides information about the electron orbital itself, and thus the gamma ray spectra can provide microscopic information about the location of the positron on the molecule. While the borrowed detector did not have the optimum energy resolution, we were able to do a "proof of principle" experiment. The results also provided the important result that the positron appears to be located on the C-H bond in hydrocarbons, including the alkanes and benzene and on the fluorine atoms in fluorine compounds such as fluorocarbons and  $\text{SF}_6$ . These results will be immensely helpful to theoretical efforts to understand positron binding, in they limit enormously the uncertainty in a physical picture of the possible sites of positron attachment.

*VIII. New evidence for the importance of vibrational modes in the binding.* Recently, we returned to the benzene problem. We first guessed that the low annihilation rate for benzene might be due to the symmetry of the molecule. Thus, we measured cyclohexane to find that it also had a similar low annihilation rate. Thus, symmetry is important. The next issue was whether the low annihilation rates for symmetric rings are due to the electronic structure or to the vibrational structure of the molecules. Recently, we studied deuterated benzene to find that the presence of even one deuteron on a benzene increased  $Z_{\text{eff}}$  by more than a factor of two. Thus, the details of the vibrational modes do have a significant effect on the annihilation rates. The observed enhancement is qualitatively consistent with the so-called "RRKM" theory of electron (in our case positron) attachment. We had previously considered this theory, but tended to discard it on the basis of deuteration experiments in hexane which indicated little effect of

deuteration. We plan to repeat the hexane experiments and conduct other deuteration experiments as well.

*IX. Molecular ionization by positrons.* During the Grant, we began a dialogue with the positron group at Oak Ridge National Laboratory, directed by Lester Hulet. This resulted in a collaborative effort to try to understand more thoroughly the results of the first study of molecular ionization by low-energy positrons, which was conducted by our group in 1988. We were recently able to show that the N<sub>2</sub> buffer gas played a role in the observed ionization at early times, but had a negligible effect at times longer than approximately the ion confinement time of the nitrogen. These results validated the claim that the previous experiment was the first observation of molecular ionization by positrons, and it gives us insight into how to conduct more precise ionization studies in the future, using our positron trapping techniques. This work was written up in a joint paper, now in press.

*X.I Capabilities for new positron experiments.* Throughout the course of the Grant, our abilities to accumulate, store, and manipulate positron gases and plasmas were greatly enhanced. This was aided immeasurably by complementary funding for positron plasma studies through a separate ONR Grant supervised by C.W. Roberson. We now have the ability to vary the energy of the positrons in the trap, in a controlled way, by applying rf noise. We can also measure the temperature of the positrons quantitatively and non-destructively, by measuring the modes of oscillation of the positron gas. We have also developed the ability to transfer positron gases and plasmas into a separate electrode structure in a different vacuum environment. This will allow us to conduct experiments on low-vapor-pressure materials in a UHV environment. Finally, efforts to develop *in situ* ion-cyclotron-resonance mass spectrometry are in progress.

*X. Summary.* This Grant allowed us to build an important research program in the area of study of the interaction of low-energy positrons with molecules and to potentially study similar species, such as clusters and dust grains. Many new facets of the interaction of low-energy positrons with molecules have been illuminated, including the scaling of annihilation rate with ionization potential of the molecules and the identification of trends as a function of other features of chemical structure. The work led naturally to studies of the resulting gamma ray annihilation radiation, which promises both to be an important tool in understanding the fundamental physics of positron-molecule interactions, and it also has important applications to gamma ray astronomy. Due to the progress made during this work and the cooperation of the ONR Grant officer in providing close-out funds in FY's

'93 and '94, we were able to secure funds from NSF to continue this work. During 1993, a new high-resolution gamma-ray detector was purchased with NSF funds, and a new generation of high-resolution gamma ray measurements is in progress.

February 14, 1994



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10. "Ion Production by Positron-Molecule Resonances," G. L. Glish, R. G. Greaves, S. A. McLucky, L. D. Hulet, C. M. Surko, J. Xu and D. L. Donohue, *Phys. Rev. A*, in press.

**Papers in preparation:**

1. "Measurements of Positron Annihilation Rates on Molecules," K. Iwata, R. G. Greaves, T. J. Murphy, M. D. Tinkle, and C. M. Surko, to be submitted to *Phys. Rev.*