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THE BUFFER-GAS POSITRON ACCUMULATOR AND RESONANCES IN POSITRON-MOLECULE INTERACTIONS

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ABSTRACT

This is a personal account of the development of our buffer-gas positron trap and the new generation of cold beams that these traps enabled. Dick Drachman provided much appreciated advice to us from the time we started the project. The physics underlying trap operation is related to resonances (or apparent resonances) in positronmolecule interactions. Amusingly, experiments enabled by the trap allowed us to understand these processes. The positron-resonance "box score" to date is one resounding "yes," namely vibrational Feshbach resonances in positron annihilation on hydrocarbons; a "probably" for positron-impact electronic excitation of CO and N₂; and a "maybe" for vibrational excitation of selected molecules. Two of these processes enabled the efficient operation of the trap, and one almost killed it in infancy. We conclude with a brief overview of further applications of the trapping technology discussed here, such as "massive" positron storage and beams with meV energy resolution.

IT ALL STARTED AT LUNCH WITH MARV

Since this paper is written in conjunction with the symposium marking the retirement of Dick Drachman and Aaron Temkin, I depart from third-person style to relate a personal view of the development of the buffer-gas trap and the physics that has come from it. I came to know Dick near the beginning of the trap project and because of it. I'd like to tell a bit here about the people involved in the development of the trap, including Dick, who helped us achieve important perspective and understanding of key physics issues that have arisen in the past two decades.

The story began in a lunchtime conversation with Marv Leventhal at AT&T Bell Laboratories in Murray Hill, New Jersey in 1983. Marv asked what one could do with positrons in a tokamak plasma. I responded by saying that there were interesting problems for sure, such as study of the turbulent transport of electrons out of the hot plasma. However, I didn't know how to get the positrons into the plasma in the first place (i.e., across the strong magnetic field). Marv said, "oh, I know how to get them in – that's easy – just convert them to positronium atoms; shoot them in; they'll ionize and you're in business." That started the whole thing off [1, 2]. Marv had done positron trapping earlier with Ben Brown, and so he was eager to pursue this kind of research. In a very nice experiment, they measured the Doppler linewidth of molecular hydrogen [3], to compare with Marv's balloon measurement of this line coming from the galactic center [4]. Regarding the proposed tokamak application, we did carry it some way forward, but

never actually did the experiment. However, this start enabled us to do many other things that turned out to be quite interesting and have enabled others to do more.

The trap was developed to accumulate positrons and store them efficiently. While the original goal was to provide an intense, pulsed positron source to study turbulence in tokamak fusion plasmas, we quickly realized that it might well have many other applications. By now, the trap has contributed to a wide range of scientific problems, including many aspects of positron interactions with atoms and molecules [5]; study of electron-positron plasmas [6]; commercial-prototype trap-based beams for materials characterization [7]; creation in the laboratory of the first low-energy antihydrogen [8]; and very recent new work to study the positronium (Ps) molecule, Ps₂ [9].

WHAT THE DESIGNERS DESIGNED AND WHO'S DICK DRACHMAN?

The way to make a nearly ideal "antimatter bottle" was known by the mid '80s, albeit not fully realized as such. It was called a Penning-Malmberg trap, adding John Malmberg's name to Penning's [10]. It was Malmberg and colleagues at the University of California, San Diego (UCSD) who developed the Penning trap to efficiently confine large quantities of single component plasma. Thus it was no accident that I later went to UCSD – in large part due to my interests in trapping positrons.

Malmberg and colleagues worked with electrons, but switching to positrons poses no problems from a plasma physics point of view. This nearly ideal "bottle" consists of a uniform magnetic field to confine the particles radially, with electrostatic potentials at the ends to confine their motion along the field. Confinement of plasmas of a single sign of charge in these devices is excellent. A theorem due to Tom O'Neil explains why [11]: the angular momentum of the plasma is dominated by the electromagnetic term, which is proportional to Σr_j^2 , where r_j is the radial position of plasma particle j. If you build a cylindrically symmetric device (i.e., about the magnetic field axis), there are no torques, and hence the plasma can't expand radially.

The trick, however, is getting the positrons into the trap efficiently without unnecessary losses, since positrons are expensive from the scientific point of view. Marv and I thought we might be able to do it using inelastic collisions. Early on, Marv had the idea to use vibrational excitation of molecules, since we wanted to avoid positronium formation that would be present if we used electronic excitation, for example. Good candidates seemed to be H_2 and N_2 , since they had relatively large vibrational energies (0.5 and 0.3 eV, respectively). This is where the connection to Dick Drachman began. I had no idea who "Drachman" was, but Marv would say, "*Drachman says*, ……" or "*we should ask Drachman about that*." This happened so many times that, when Marv said something to this effect standing in the hall outside his office, I made a note to find out who this guy was – he must really be somebody important!

While I had done a bit of atomic physics some 15 years earlier, the trap project provided me with the opportunity to become immersed in the field. I came to know Dick well and consulted him frequently. He provided that calm and reasoned voice to lead us

through many theoretical minefields (vats of snake oil too). He took the time to patiently teach me about theoretical atomic physics and to translate (in language even I could understand) various esoteric papers filled with surprising results (alas, not infrequently wrong). Dick was an enormous help in keeping us on track. He helped make the venture into positron-atomic physics both very productive and most enjoyable. Our first conversations related to vibrational excitation. Baille and Darewych had done a very nice calculation of vibrational excitation of H₂ by positron impact [12], and we wanted to understand what that would mean for us and how we might extrapolate to other targets.

Back at the trap project, we designed what was later to be called a three-stage, buffer-gas, positron accumulator, the principle of which is shown schematically in Fig. 1 [1]. The pressure in stage I is adjusted so that a positron would lose enough energy in one transit through the device to be trapped, then subsequent collisions would further lower the positron energy, so that it would end up trapped and cool in the lowest pressure region in stage III. We realized pretty quickly that, while stage II and III could operate on vibrational excitation, stage I required both a large cross section and a larger energy loss than vibrations could provide. So we planned that the first stage would operate on electronic excitation, even though this would carry with it some loss due to positronium formation.



Fig. 1. Schematic diagram of the three stage buffer-gas accumulator *circa* 1988. Top: electrode structure. Middle and bottom: pressure and electrical potential profiles. There is a uniform magnetic field ~ 0.9 kG in the *z* direction. Stages are labeled I, II and III. The trapping mechanism is inelastic collisions with the N₂ buffer gas, labeled A, B and B'. In 1989, A was electronic excitation, and B and B' were vibrational excitation of N₂ [13].

By 1985 we had made progress in planning for the trap and the positrons-intokamak experiment. It so happened that at that time, Allen Mills and Karl Canter were arranging a 60^{th} birthday celebration at Brandeis for Steve Berko, a giant in the field of



Fig. 2. (a) Photo of the attendees at Steve Berko's 60th birthday symposium at Brandeis in December 1984. It's virtually a who's who in the U. S. world of positron physics including Dick (upper left). Somehow I didn't hear the announcement about the picture. It is one if the few group photos at meetings that I'm sorry to have missed. Reprinted from Ref. [1] with permission from World Scientific Publishing Co. Pte. Ltd, Singapore.



Fig. 2. (b) Roster of names for the Berko symposium picture, December 1984. Reprinted from Ref. [1] with permission from World Scientific Publishing Co. Pte. Ltd, Singapore.

positron studies of solid-state systems, such as the Fermi surfaces in metals. Allen and Karl were planning the proceedings and Allen asked for a paper on our positron-trap project [1]. This first paper on the trap was also the first of only two papers that I've coauthored with Allen. Looking back, the paper was anything but modest in its promises: we "discussed the possibility" of accumulating positron plasmas containing 10¹⁰ particles and holding them for minutes. The latter turned out to be not too hard, given a few years, but the former has yet to be achieved some 20 years later (but we do hope to do it soon). As shown in Fig. 2, a host of big names attended the Berko symposium, including founders of the field, such as the discoverer of positronium, Martin Deutsch, and the future Nobel Prize winner, Steve Chu. It was really a great time for the positroners! Berko, whom I did not know very well before the meeting, was immensely gracious in accommodating the hoopla made over him. Best of all, I had an opportunity to meet *Drachman*, although I didn't realize at the time his strong connections to Brandeis.

Shortly after the conference, Fred Wysocki came from the Princeton Plasma Lab to Bell Labs as a post doc to spearhead the construction of the trap, and we were off to the "positron accumulator races." Figure 3 shows Marv, Fred, and Al Passner admiring the electrode structure of the first trap (gold plated no less). Al, who was an Associate Member of the Technical Staff at Bell Labs, had worked with me for a number of years before we began the trap project. He made a great contribution to its early success, similar to his contributions to many of our other experiments.



Fig. 3. Gold-plated copper electrode structure of the buffer-gas positron accumulator as assembled in 1986. From left to right, Al Passner, Fred Wysocki, and Marv Leventhal. The electrodes sit on laser tables where equipment for Allen Mills' Ps spectroscopy experiment [14] was to be set up.

MOTHER NATURE HAD OTHER IDEAS, THE THORN IN ALLEN'S SIDE, AND DICK'S SYMPOSIUM

Fred Wysocki designed a very nice device, and so it all looked great. However, when we turned the trap on, the results were terrible. We didn't get anything to speak of with vibrational excitation, but we could trap positrons when we increased the positron energy in the first stage to where we guessed we were exciting the molecules electronically. We tried a large number of molecules, and N₂ seemed to work best for reasons then unknown. Actually, we had wanted to avoid electronic excitation, since for almost all targets, the Ps formation channel is open there too, and the latter is a potent loss process. We had expected 10^6 positrons trapped and minute positron lifetimes, but we got about a factor of 100 less, as shown in Fig. 4. So we had not only an excitation problem, but also a lifetime problem as well. I was nevertheless pretty excited, somehow feeling that, if we weren't making a mistake, there was likely new physics there somewhere. *Lots and lots* of conversations with Dick followed these early results concerning the possible atomic physics processes involved in the trap operation.



Fig. 4. Data from a trapping experiment in August 1987. Above: number of positrons confined as a function of fill time, maximum number $\sim 1 \times 10^4$. Below: confinement as a function of time after the fill was turned off, indicating a 0.7 s confinement time. The confinement was limited by annihilation on a very low density of large, molecular impurities at pressures $< 10^{-9}$ torr.

Now there was another story unfolding here too. It was often said that, in those days, Bell Labs wasn't dollar limited, but was space limited. (Typically the opposite is true in universities.) When we started the trap project, we needed someplace to put it. I had a lab, but it was full of fluid convection apparatus, so no room there. Marv had only a small space, since he did mostly gamma-ray astronomy with groups elsewhere, so we had a problem. Fortunately, Allen (as in A. P. Mills, Jr. - the Allen, Allen) just happened to have a big, new-to-him lab in the basement that he hadn't moved into yet. So, we say "Allen" (imagine pleasant music in our voices), "errr, could we borrow your lab for just 18 months; we want to set up a trap and then move it to Princeton to study turbulence in tokamak plasmas?" "Well OK," (says the cooperative Dr. M., Jr.) "but remember - just 18 months!" Well, that was in '85, and as I recall, by '87, Allen was unhappy, since (see Fig. 5) there were tons of magnet, etc. sprawled all over his space and no signs of our departure in sight. So Allen began building his new Ps spectroscopy experiment [14] intertwined with ours like spaghetti in a bowl. Then when we first began trapping positrons, Allen was *really* an unhappy camper – well, not really, but he saw the problem coming even before we did. Allen was such a good scientist and believed so much in everybody's science that he couldn't throw us out if we were making progress. If our trap didn't work, we were out on the street (600 Mountain Avenue, Murray Hill NJ, to be precise). But it was working, and "so keep your elbows in," we're going to be very cozy for a while with two "full-bodied" experiments in one underground lab. Great guy, Allen; maybe not overjoyed at that moment, but he really did us a huge favor.



Fig. 5. Al Passner and me standing in front of the first buffer-gas trap in Allen Mills' lab *circa* 1987. The source is on the right behind the gas bottle; and Allen's Ps spectroscopy experiment (Helmholtz coils and rails), which was under construction, is "creeping in" at the lower left.

Back at the trap, we twiddled knobs and got the plasma lifetime to improve a bit at a time somehow. The trap had a continuous feed of tungsten-moderated positrons from a 22 Na source, so longer lifetimes meant more particles trapped. But we really didn't know what was going on. The seminal event occurred one afternoon. I came into the lab, and Al Passner was excited. *"Hey Cliff, look at this – I think it's impurities."* He had a small dewar of liquid nitrogen in one hand, and he was looking at the trapped particle signal on an oscilloscope as he poured nitrogen on the trap vacuum chamber. Liquid N₂ on – larger signal; take the LN₂ away – the signal decreased. This was a major turn of events. Sure enough, in the months that followed, we found that the better we made the vacuum, the better the trap worked.

The problem, it turned out, was large hydrocarbon molecules at the $\leq 10^{-9}$ torr level, for reasons no one knew at the time. Basically they had absolutely huge annihilation rates. It was good that Wysocki insisted on building such a good vacuum system in the first place, or the experiment would have never worked. So on the hardware side, we installed an *in situ* liquid nitrogen dewar in the vacuum system next to the final trapping stage to pump impurities, and we began switching out the turbopumps on the system (i.e., which required conventional oil mechanical pumps behind them) in favor of cryopumps that didn't require any backing.

On the physics side, the first major paper was a 1988 Phys. Rev. Letter (PRL) on (no surprise) annihilation on large molecules [15]. As shown in Fig. 6, we found that annihilation rates increased exponentially with molecular size. Plotted here is the conventional normalized annihilation rate, Z_{eff} , which is the measured annihilation rate, Γ , relative to that expected for positrons in a free electron gas of the same density, namely [16],

$$Z_{\rm eff} = \Gamma / \pi r_{\rm o}^2 c n_m, \tag{1}$$

In Eq. 1, r_0 is the classical electron radius, c is the speed of light, and n_m is the molecular number density. We had the intuition to point out in the PRL that the large Z_{eff} values were likely due to vibrational resonances and invoked the "RRKM" formalism from the chemistry literature to explain it. We were familiar with Heyland's work on small hydrocarbons (see Fig. 6) [17], but we were unaware of the seminal work of Paul and St. Pierre who studied annihilation in dense gases for hydrocarbons as large as butane (C₄H₁₀) [18]. We agreed quantitatively with the previous measurements and extended them to molecules as large as C₁₆H₃₄ finding annihilation rates orders of magnitude larger. More significant really was the ability to study positron interactions with molecules in a vacuum environment. There was now no question that the large rates were due to a two body effect, and we could also make independent measurements of the positron temperature [confirmed to be the electrode temperature of 300 K (i.e., 25 meV)].

We were on the road now. The next year we published another PRL announcing the first positron plasma in the laboratory [13]. It contained a modest 3 x 10^5 e^+ , with a Debye screening length, λ_D , which was a similarly modest 1/4 the plasma radius (i.e., a good measure of the plasma regime is the degree to which $\lambda_D \ll r_p$). Later, John

Malmberg and Hans Dehmelt told me that they were referees for the paper, exceedingly pleasing, because both were giants in the trap field. We realized that the annihilation on molecules might result in a spectrum of ions. At Allen Mills' suggestion, Al Passner looked and sure enough, there was a story to tell there too. We found that annihilation with a 300 K thermal distribution of positrons left a broad spectrum of ions [19]. I'm sorry to say that this is the second of only two papers I coauthored with Allen, during all our time at Bell Labs.



Fig. 6. Normalized annihilation rate Z_{eff}/Z for alkanes (C_nH_{2n+2}) as a function of the number of molecular electrons, Z: Open circles are from an experiment using atmospheric pressure test gas [17], and (•) are data taken with a low-pressure of the test gas in the positron trap. From Ref. [15].

Les Hulett and collaborators at Oak Ridge followed up on this positron-induced ionization effect and produced a series of interesting papers [20, 21]. Initially, there was a disagreement about the significance of our experiment, which resulted (amicably) in a joint publication [22]. Oakley Crawford from the Oak Ridge group wrote a nice theoretical paper that explained the basic phenomenon [23], saying that the incoming positrons annihilate with equal probability on any valence electron, not just the highest-lying molecular orbitals. Later we confirmed this prediction with Doppler broadening measurements [24, 25]; it was only then that I realized the full implications of Crawford's model.

At that point (Fall, '88), I moved from Allen's lab to UCSD in La Jolla, much to his relief. Sadly the collaboration with Marv and interactions with Allen tailed off as the miles separated us and other interests and obligations got in the way. In La Jolla, we continued the studies of annihilation in molecules and continue them still, many generations of experiments later. Dick's 1989 symposium at Goddard was, from my point of view, something of a coming-of-age party for the trap. We presented a paper on our annihilation results that was well received [26]. I was quite gratified that the trap-based results were embraced by the positron community as "mainstream," something that is frequently not easy coming into a new field with a different technique. The meeting was a great opportunity to meet people who would later play key roles in the science that the trap was to enable.

PHYSICS IN THE TRAP AND MORE CONVERSATIONS WITH DICK

More superb Princeton plasma talent came with Tom Murphy, who joined us at Bell Labs in time to disassemble the trap at Bell and move it to La Jolla. Even while a grad student at Princeton, Tom wrote a great paper on using a Ps beam to study transport in tokamaks [27]. Immediately upon our arrival in La Jolla, Gene Jerzewski joined the effort. His technical expertise and ability to teach students and post docs about hardware were invaluable to our efforts in the years to follow – wonderful contributions on a level with Al Passner's contributions at Bell.

In La Jolla, Tom Murphy focused on understanding *the* problem, namely annihilation in large molecules [28]. He also made great strides in understanding how the three-stage buffer-gas trap actually worked [29]. A key discovery was that the optimum trapping potential difference between stages was $\sim 9 - 10$ eV per stage for each of the three stages. As described below, this is the energy window in N₂ where the electronic excitation cross section is larger than that for Ps formation due to a resonance in the excitation channel not yet understood. The result is efficient trapping when all stages of the trap are tuned to operate in this regime.

On the annihilation front, Tom cleared up a long-standing ambiguity in the data for annihilation in xenon for a thermal distribution of positrons at 300 K, confirming $Z_{eff} \approx$ 400 [30], This value would be doubted by theorists for another decade, but is now the accepted number [5]. Tom also made new systematic studies of Z_{eff} for a range of compounds [28]. Along the way, there were innumerable conversations with Dick Drachman about low-energy positron interactions with atomic and molecular targets in our attempt to get some degree of theoretical understanding to match the results provided by our new experimental capabilities. He was kind enough to look over any paper that I sent him, including providing very useful comments and suggestions on our papers when we sent them to him in the draft stage.

In the early 90's, Shengzhang Tang joined us from Ken Roellig's shop, and Rod Greaves came from the plasma community. Koji Iwata did his thesis on annihilation ("*he did <u>all</u> this work for a thesis?*" one member of his doctoral committee asked!), and Mark Tinkle did his thesis on mode diagnostics of positron plasmas. Shenzhang took the last steps that we would take toward a Ps beam fusion diagnostic showing that one could make a pretty good, variable-energy Ps beam by charge exchange on H₂ [31]. We got a very important new tool from Shengzhang, namely our first Doppler broadening measurements [32], an example of which is shown in Fig. 7 [33]. Later this technique allowed us to determine the site of positron annihilation in large molecules [24, 25]; all

valence electrons seem to do the trick, as per Oakley Crawford's prediction a few years earlier. During that period, we also made quantitative studies of inner shell annihilation [34] and studied annihilation in polycyclic aromatic (PAH) molecules, which are important constituents of the interstellar medium [35, 36].



Fig. 7. (open circles) Doppler-broadened gamma-ray spectrum from positron annihilation on helium atoms and comparison with (-) theoretical predictions using a variational wavefunction, and (-) a Gaussian fit. From Ref. [33].



Fig. 8. Progress in trapping positrons with buffer-gas accumulators using ²²Na sources with strengths ($\sim 100 \text{ mCi}$), beginning with the first trap at Bell Labs. All except the last point (ATHENA collaboration [39]) are from the traps at Bell and UCSD.

Around '95, Rod Greaves pushed us to develop further the neon moderator for use with the trap [37]. Invented much earlier by Allen Mills [38], neon was not used as a moderator much, because the energy spread was considerably larger than that of tungsten (i.e., $\sim 1.5 \text{ eV}$, FWHM, as compared to $\sim 0.5 \text{ eV}$ for tungsten), even though neon is more efficient by an order of magnitude. Rod realized that this was not a disadvantage for the trap. The buffer-gas trap has an energy acceptance window $\sim 2 \text{ eV}$; and once trapped, the positrons cool to 25 meV, which is far superior to the energy spread of the conventional tungsten-moderated beam. Rod designed a compact system, which is now standard fare for traps and other applications.

In '97, we designed and built a second-generation three-stage positron trap that was somewhat more compact. By then we had improved the overall efficiency of the system by more than a factor of 10^4 as compared with the first results shown in Fig. 4. Figure 8 shows this progress and includes later results from the ATHENA antihydrogen collaboration at CERN. As discussed below, they used a buffer gas accumulator to accumulate positrons, then stacked them in a high-magnetic-field Penning-Malmberg trap.

THE TRAP-BASED BEAM AND THREE POSITRON-ATOMIC PHYSICS QUESTIONS

In '95, Chris Kurz came from MIT to join our group as a post doc, and Steven Gilbert joined to do a thesis and then a short post doc. With Chris, we made the first annihilation measurements as a function of positron temperature, heating them *in situ* in the trap with rf radiation [40]. This was a precursor to Rod Greaves, Chris and Steven realizing that we could make a cold beam by simply dumping the trap slowly [41]. The results, shown in Fig. 9, were spectacular and turned out to be a big advance for us. The



Fig. 9. Retarding potential curve of the cold trap-based positron beam indicating a parallel energy spread of 18 meV, FWHM at 1.7 eV [41]. The inset illustrates the beam-formation technique, whereby the potential of stage III of the positron trap is raised slowly to force the positrons out of the well.

beam was tunable from ~ 100 meV upwards and had a parallel energy spread of 18 meV. It was superior to conventional beams used for atomic physics studies by more than a factor of 10 in energy resolution.

Steven and Rod then decided to try a scattering experiment – vibrational excitation of CF_4 . It was supposed to be a preliminary experiment, but turned out so well we sent the results to PRL [42]. What we had done in just two years was to advance the state of the art of positron beams for positron-atomic physics by more than an order of magnitude in energy resolution. We had also developed a new scattering technique that advanced the state of the art, even in electron scattering, particularly for measuring integral inelastic scattering cross sections.

In '98, Rod Greaves moved on to First Point Scientific, Inc., in Agoura Hills CA. There he has developed commercial positron traps and compact neon moderator systems that have aided greatly in allowing people around the world to exploit positron trapping technology [7, 43]. On the science side, he continues to collaborate with our group, and as discussed below, more recently with Allen Mills, who went from Bell Labs to the University of California at Riverside in the late '90s.

Positron cooling is a crucial issue for both plasma and scattering experiments. Rod Greaves had measured gas-cooling times for positrons and found that CF_4 and SF_6 were the best for rapid cooling [44]. This was confirmed by Gilbert's direct measurement of the vibrational cross section for CF_4 . Giving the nod to its small annihilation cross section, we subsequently used small amounts of CF_4 in stage III of the trap for rapid cooling. This allowed us to cycle our pulsed positron beam rapidly for scattering and annihilation experiments.

Around that time, the results flowed in. Steven Gilbert made the first energyresolved measurements of positron annihilation on molecules, discovering huge Feshbach resonances and measuring the first positron-molecule binding energies [45]. Joan Marler did her thesis work under the tutelage of post doc James Sullivan who joined us from the Australian National University in Canberra, thoughtfully bringing his advisor Steve Buckman along for the second year. Steve Buckman and James gave our program an enormous boost, pushing us to measure every conceivable cross section and hunt for every resonance imaginable (at least that's what it seemed like to me) [46-50]. One of their "gifts" was the technique of unfolding electronic excitation cross sections in molecules using the known Franck Condon factors [47] – good physics and great fun.

Turning back to key features of the operation of the positron trap, there are three puzzles.

- Why is N₂ the best trapping gas?
- Why did CF₄ work so well for cooling positrons?
- What's the story with the large annihilation rates in molecules?



Fig. 10. Positron-impact cross section for the excitation of the $a^1\Pi$ state of N_2 [47, 51]. Shown for comparison (open circles) are electron data for the same cross section [52, 53]. Also shown by the lines are calculations by Marco Lima *et al.*, for the positron-impact process, using three different basis sets for the target. See Ref. [51] for details. The sharp rise at onset and relatively small near-threshold Ps formation cross section make N_2 the molecule of choice for buffer-gas trapping.

The cold beam told it all, more or less – at least all experimentally that is – the theory is about one and a half for three at this point. Shown in Fig. 10 is the positron-impact electronic excitation cross section for N_2 [47]. The unexpected, sharp rise at threshold (some kind of resonance?) opens up faster than the positronium formation cross section, which is a loss process in the positron trap. This resonance is a very efficient way for positrons to lose energy and drop into successive stages in the trap. In CO, there is a similar sharp resonance, but the Ps formation cross section is even larger, so CO is not as good as N_2 for trapping [51]. The case is closed for the experimentalists – the theorists still find this N_2 resonance difficult to explain,¹ as shown by the theory curves in Fig. 10.

The CF₄ vibrational excitation story has a similar ring to it. Gilbert's early measurement of the CF₄ cross section turned out to be inaccurate due to all the machinations we had to go through to measure it, so Joan Marler repeated the measurement, as shown in Fig. 11 [54]. We found a sharp rise at onset there too, and the largest vibrational cross section measured to date. Joan went on to make the same measurement for electron impact – the first *in situ* comparison of state-resolved electron and positron inelastic cross sections [54]. The fact that the cross sections were the same, both in magnitude and shape, provided the needed clue (kindly delivered to us by Gleb Gribakin) to compare the measurements with the predictions of the Born dipole model.

Indeed, the equality of the electron and positron scattering cross sections could be explained quantitatively by long range, electrostatic dipole coupling. Infrared absorption

¹ M. A. P. Lima, private communication, 2005.

measurements provide the strength of the dipole matrix element, and the theory fits quite well with no adjustable parameters. So in this case, the sharp rise in the cross section is not a resonance but arises naturally from the long-range electrostatic coupling. The remaining mystery is that all of the molecules and modes studied to date, except the one homopolar molecule, H_2 , have positron-impact vibrational cross sections with a very similar energy dependence, even though the magnitude of the Born dipole coupling is too small by as much as a factor of five for everything except CF₄ [55]. So there's more to be learned here.



Fig. 11. Positron- and electron-impact cross sections for excitation of the v_3 asymmetric stretch mode of CF₄. This is the largest positron-impact vibrational cross section measured to date. Also shown for comparison (–) are the predictions of the Born dipole model for this cross section, with no fitted parameters. From Ref. [54].

The final question led to the most spectacular result. As discussed above we, and others before us, had suspected that the large annihilation rates in molecules are due to vibrational resonances [15, 56], but it was hard to nail down experimentally with thermal distributions of positrons. Once we had the cold beam, Steven Gilbert and Levi Barnes (who had just joined the group as a Ph.D. student) took on the very ambitious project of studying annihilation rates in molecules as a function of positron energy. They built a wonderful apparatus such that they could cycle 10⁹ positrons with only one background count. This is needed because, while the resonances are quite large, the basic cross section [i.e., the Dirac cross section, c.f., Eq. (1)] is miniscule, and so without a very careful experiment, it is still difficult to distinguish the annihilation from extraneous effects.

The results, as advertised above and illustrated in Fig. 12, were spectacular indeed [45, 57]. Alkane molecules were a favorite target of ours for studying annihilation, because they are conveniently available in a variety of sizes and exhibit huge enhancements in annihilation rates. We found very large enhancements in alkanes with energy spectra that closely mimic the spectra of the molecular vibrational modes, with particularly large resonance associated with the C-H asymmetric stretch mode. The

added bonus was that the annihilation spectra are downshifted from the vibrational spectra. We interpret these data in the context of Gleb Gribakin's vibrational Feshbach resonance model [60] as evidence that positrons bind to alkanes. The downshift is a measure of the positron binding energy, which ranges from ~ 40 meV in butane (C₄H₁₀) to > 200 meV in C₁₂H₂₆. Levi Barnes and Jason Young have now carried these experiments further, most recently finding evidence for a second, *positronically excited* bound state in the very large alkanes C₁₂H₂₆ and C₁₄H₃₀ [57, 58].



Fig. 12. Z_{eff} for butane (•) as a function of positron energy: (a) 0 to 5 eV, and (b) 0 to 0.5 eV. From Refs. [57] [58]. The arrow on the abscissa in (a) is the threshold for positronium formation. Shown in (b) is the vibrational-mode spectrum of butane (--, arbitrary vertical scale), with each mode broadened by 25 meV [59]. The downshift, $\varepsilon_{\rm b}$, represents the positron-molecule binding energy which is ~ 40 meV for butane. Arrows on the ordinate indicate values of $Z_{\rm eff}$ for a 300 K Maxwellian distribution of positrons. In (a), $Z_{\rm eff}$ at energies ≥ 0.5 eV is ~ 100, comparable to the value of Z = 34 for this molecule, which is expected in the absence of the vibrational resonances.

MORE PHYSICS FROM THE TRAP AND WHAT THE FUTURE MIGHT HAVE IN STORE

I've carried the positron trapping saga and positron-atomic physics story along from the mid 80's to the present. There is a parallel story for positron plasmas that is too lengthy to tell here in any detail [6]. Mark Tinkle did a thesis on the development of mode diagnostics for positron plasmas [61, 62]. Rod Greaves and I did the first electronpositron plasma experiment, studying the instability generated when an electron beam is passed through a positron plasma [63, 64]. Greaves used a rotating electric field to compress positron plasmas radially (the so-called "rotating wall" effect) [44, 65], a tool recently developed further by James Danielson [66, 67], that has enormous potential for tailoring positron plasmas and beams. James is now continuing this research and developing new methods to trap more positrons (i.e., a "multicell trap" [68]), and to produce much colder (the goal 1 meV, FWHM) positron beams [6].

At First Point Scientific Inc., Rod Greaves developed a commercial prototype buffer gas trap for materials characterization and positron research [43]. The basic buffer-gas trap design was used by the ATHENA collaboration to make the first low-energy antihydrogen [8]. The trap gave them a significant advantage *vis a vis* their competitors in the ATRAP collaboration, who also created low-energy antihydrogen very close to the same time in a set of complementary experiments [8, 69]. James Sullivan and Steve Buckman have now built a trap in Canberra for positron atomic physics research. Mike Charlton has one at Swansea [70], as does Igor Meshkov in Dubna [71], who got the design from Mike and is making a new Ps beam facility for fundamental physics studies.

Very recently, Allen Mills, David Cassidy, Rod Greaves, and collaborators used Rod's version of the buffer gas trap to observe effects they attribute to creation of the first Ps_2 molecules in the laboratory [9]. This line of experiments has enormous promise, and is likely to lead to the creation and study of BEC Ps, which is a long term goal of Allen and Phil Platzman [72, 73].

So it's been a very productive and rewarding time. The Drachman-Temkin symposium is a great place to tell the story. Dick's guidance and counsel was much appreciated by all of us involved in the positron trapping effort. As I finished writing this, one thought occurred to me: What if Marv and I had sat at different lunch tables that day at Bell? I may well have not had the opportunity to work with positrons or meet Dick – it was truly good luck the way it turned out!

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References

- Surko, C. M., Leventhal, M., Crane, W. S., *et al.*, The Positron Trap A New Tool for Plasma Physics, in *Positron Studies of Solids, Surfaces, and Atoms; a Symposium to Celebrate Stephan Berko's 60th Birthday*, edited by J. A. P. Mills, W. S. Crane and K. F. Canter (World Scientific, 1984), p. 222.
- 2. Surko, C. M., Leventhal, M., Crane, W. S., *et al.*, Use of Positrons to Study Transport in Tokamak Plasmas, *Rev. of Sci. Instrum.* **57**, 1862, 1986.
- 3. Brown, B. L. and Leventhal, M., Laboratory Simulation of Direct Positron Annihilation in a Neutral-Hydrogen Galactic Environment, *Phys. Rev. Lett.* **57**, 1651, 1986.

- 4. Leventhal, M., Callum, C. J. M., and Stang, P. D., Detection of 511 Kev Positron-Annihilation Radiation from the Galactic-Center Direction, *Astrophys. J.*, L11, 1978.
- 5. Surko, C. M., Gribakin, G. F., and Buckman, S. J., Low-Energy Positron Interactions with Atoms and Molecules, *J. Phys. B: At. Mol. Opt. Phys.* **38**, R57 2005.
- 6. Surko, C. M. and Greaves, R. G., Emerging Science and Technology of Antimatter Plasmas and Trap-Based Beams, *Phys. Plasmas* **11**, 2333, 2004.
- 7. Greaves, R. G. and Moxom, J. M., Recent Results on Trap-Based Positron Beams, *Mat. Sci. Forum* **445-446**, 419, 2004.
- 8. Amoretti, M., Amsler, C., Bonomi, G., *et al.*, Production and Detection of Cold Antihydrogen Atoms, *Nature* **419**, 456, 2002.
- 9. Cassidy, D. B., Deng, S. H. M., Greaves, R. G., *et al.*, Experiments with a High-Density Positronium Gas, *Phys. Rev. Lett.* **95**, 195006, 2005.
- 10. Malmberg, J. H. and deGrassie, J. S., Properties of Nonneutral Plasma, *Phys. Rev. Lett.* **35**, 577, 1975.
- 11. O'Neil, T. M., A Confinement Theorem for Nonneutral Plasmas, *Phys. of Fluids* **23**, 2216, 1980.
- 12. Baille, P. and Darewych, J. W., Collisions Entre Un Positron Et Une Molecule D'hydrogene Avec Excitation De Vibrations, *J. de Phys. Lett.* **35**, 243, 1974.
- 13. Surko, C. M., Leventhal, M., and Passner, A., Positron Plasma in the Laboratory, *Phys. Rev. Lett.* **62**, 901, 1989.
- Fee, M. S., Chu, S., A. P. Mills, Jr., *et al.*, Measurement of the Positronium 13S1 -23S1 Interval by Continuous-Wave Two-Photon Excitation, *Phys. Rev Lett.* 70, 1397, 1993.
- 15. Surko, C. M., Passner, A., Leventhal, M., *et al.*, Bound States of Positrons and Large Molecules, *Phys. Rev. Lett.* **61**, 1831, 1988.
- 16. Fraser, P. A., Positrons and Positronium in Gases, *Adv. in At. and Mol. Phys.* **4**, 63, 1968.
- 17. Heyland, G. R., Charlton, M., Griffith, T. C., *et al.*, Positron Lifetime Spectra for Gases, *Can. J. Phys.* **60**, 503, 1982.
- 18. Paul, D. A. L. and Saint-Pierre, L., Rapid Annihilation of Positrons in Polyatomic Gases, *Phys. Rev. Lett.* **11**, 493, 1963.
- 19. Passner, A., Surko, C. M., Leventhal, M., *et al.*, Ion Production by Positron-Molecule Resonances, *Phys. Rev. A* **39**, 3706, 1989.
- Hulett, L. D., Donohue, D. L., Xu, J., *et al.*, Mass Spectrometry Studies of the Ionization of Organic Molecules by Low-Energy Positrons, *Chem. Phys. Lett.* 216, 236, 1993.
- 21. McLuckey, S. A., Hulett, L. D., Xu, J., *et al.*, Gas-Phase Ionization of Polyatomic Molecules Via Interactions with Positrons, *Rap. Comm. Mass Sp.* **10**, 269, 1996.
- 22. McLuckey, S. A., Glish, G. L., Donohue, D. L., *et al.*, Positron Ionization Mass Spectrometry: Ionization by Fast Positrons, *Int'l. J. of Mass Spectrometry and Ion Processes* **97**, 237, 1990.
- 23. Crawford, O. H., Mechanism for Fragmentation of Molecules by Positron Annihilation, *Phys. Rev. A* **49**, R3147, 1994.

- 24. Iwata, K., Greaves, R. G., and Surko, C. M., Gamma-Ray Spectra from Positron Annihilation on Atoms and Molecules, *Phys. Rev. A* **55**, 3586, 1997.
- 25. Iwata, K., Gribakin, G. F., Greaves, R. G., *et al.*, Positron Annihilation on Large Molecules, *Phys. Rev. A* **61**, 022719, 2000.
- Leventhal, M., Passner, A., and Surko, C. M., Positron-molecule Bound States and Positive Ion Production, in *Annihilation in Gases and Galaxies*, edited by R. J. Drachman (National Aeronautics and Space Administration, Washington, DC, 1990), p. 272.
- 27. Murphy, T. J., Positron Deposition in Plasmas by Positronium Beam Ionization and Transport of Positrons in Tokamak Plasmas, *Plasma Phys. and Controlled Fusion* **29**, 549, 1987.
- 28. Murphy, T. J. and Surko, C. M., Annihilation of Positrons on Organic Molecules, *Phys. Rev. Lett.* **67**, 2954, 1991.
- 29. Murphy, T. J. and Surko, C. M., Positron Trapping in an Electrostatic Well by Inelastic Collisions with Nitrogen Molecules, *Phys. Rev. A* **46**, 5696, 1992.
- 30. Murphy, T. J. and Surko, C. M., Annihilation of Positrons in Xenon Gas, *J. Phys. B: At. Mol. Opt. Phys.* **23**, L727, 1990.
- 31. Tang, S. and Surko, C. M., Angular Dependence of Positronium Formation in Molecular Hydrogen, *Phys. Rev. A* **47**, 743, 1993.
- 32. Tang, S., Tinkle, M. D., Greaves, R. G., *et al.*, Annihilation Gamma-Ray Spectra from Positron-Molecule Interactions, *Phys. Rev. Lett.* **68**, 3793, 1992.
- 33. VanReeth, P., Humberston, J. W., Iwata, K., *et al.*, Annihilation in Low-Energy Positron-Helium Scattering, *J. Phys. B: At. Mol. Opt. Phys.* **29**, L465, 1996.
- 34. Iwata, K., Gribakin, G., Greaves, R. G., *et al.*, Positron Annihilation with Inner-Shell Electrons in Noble Gas Atoms, *Phys. Rev. Lett.* **79**, 39, 1997.
- 35. Surko, C. M., Greaves, R. G., and Leventhal, M., Use of Traps to Study Positron Annihilation in Astrophysically Relevant Media, *Hyperfine Interactions* **81**, 239, 1993.
- 36. Iwata, K., Greaves, R. G., and Surko, C. M., Positron Annihilation in a Simulated Interstellar Medium, *Can. J. Phys.* **51**, 407, 1996.
- 37. Greaves, R. G. and Surko, C. M., Solid Neon Moderator for Positron Trapping Experiments, *Can. J. Phys.* **51**, 445, 1996.
- 38. Mills, A. P., Jr. and Gullikson, E. M., Solid Neon Moderator for Producing Slow Positrons, *Appl. Phys. Lett.* **49**, 1121, 1986.
- 39. Jorgensen, L. V., Amoretti, M., Bonomi, G., *et al.*, New Source of Dense, Cryogenic Positron Plasma, *Phys. Rev. Lett.* **95**, 025002, 2005.
- 40. Kurz, C., Greaves, R. G., and Surko, C. M., Temperature Dependence of Positron Annihilation Rates in Noble Gases, *Phys. Rev. Lett.* **77**, 2929, 1996.
- 41. Gilbert, S. J., Kurz, C., Greaves, R. G., *et al.*, Creation of a Monoenergetic Pulsed Positron Beam, *Appl. Phys. Lett.* **70**, 1944, 1997.
- 42. Gilbert, S. J., Greaves, R. G., and Surko, C. M., Positron Scattering from Atoms and Molecules at Low Energies, *Phys. Rev. Lett.* **82**, 5032, 1999.
- Greaves, R. G. and Moxom, J., Design and Performance of a Trap-based Positron Beam Source, in *Non-Neutral Plasma Physics V*, edited by M. Schauer, T. Mitchell and R. Nebel (American Institute of Physics, 2003), p. 140.

- 44. Greaves, R. G. and Surko, C. M., Inward Transport and Compression of a Positron Plasma by a Rotating Electric Field, *Phys. Rev. Lett.* **85**, 1883, 2000.
- 45. Gilbert, S. J., Barnes, L. D., Sullivan, J. P., *et al.*, Vibrational Resonance Enhancement of Positron Annihilation in Molecules, *Phys. Rev. Lett.* **88**, 0443201, 2002.
- 46. Sullivan, J., Gilbert, S. J., and Surko, C. M., Excitation of Molecular Vibrations by Positron Impact, *Phys. Rev. Lett.* **86**, 1494, 2001.
- 47. Sullivan, J. P., Marler, J. P., Gilbert, S. J., *et al.*, Excitation of Electronic States of Ar, H₂, and N₂ by Positron Impact, *Phys. Rev. Lett.* **87**, 073201, 2001.
- 48. Sullivan, J. P., Gilbert, S. J., Buckman, S. J., *et al.*, Search for Resonances in the Scattering of Low-Energy Positrons from Atoms and Molecules, *J. Phys. B: At. Mol. Opt. Phys.* **34**, L467, 2001.
- 49. Sullivan, J. P., Gilbert, S. J., Marler, J. P., *et al.*, Positron Scattering from Atoms and Molecules Using a Magnetized Beam, *Phys. Rev. A* **66**, 042708, 2002.
- 50. Marler, J. P., Sullivan, J. P., and Surko, C. M., Ionization and Positronium Formation in Noble Gases, *Phys. Rev. A* **71**, 022701, 2005.
- 51. Marler, J. P. and Surko, C. M., Positron-Impact Ionization, Positronium Formation and Electronic Excitation Cross Sections for Diatomic Molecules, *Phys. Rev.* A72, 062713, 2005.
- 52. Campbell, L., Brunger, M. J., Nolan, A. M., *et al.*, Integral Cross Sections for Electron Impact Excitation of Electronic State of N₂, *J. Phys. B: At. Mol. Opt. Phys.* **34**, 1185, 2001.
- 53. Mason, N. J. and Newell, W. R., Electron Impact Total Excitation Cross Section of the A¹ Π_g State of N₂, *J. Phys. B* **20**, 3913, 1987.
- Marler, J. P. and Surko, C. M., Systematic Comparison of Positron and Electron Impact Excitation of the N₃ Vibrational Mode of Cf₄, *Phys. Rev. A* 72, 062702, 2005.
- 55. Marler, J. P., Gribakin, G., and Surko, C. M., Comparison of Positron-Impact Vibrational Excitation Cross Sections with the Born-Dipole Model, *Nucl. Instrum Methods A*, in press, 2006.
- 56. Smith, P. M. and Paul, D. A. L., Positron Annihilation in Methane Gas, *Can. J. Phys.* **48**, 2984, 1970.
- 57. Barnes, L. D., Gilbert, S. J., and Surko, C. M., Energy-Resolved Positron Annihilation for Molecules, *Phys. Rev. A* 67, 032706, 2003.
- 58. Barnes, L. D., Young, J. A., and Surko, C. M., Studies of Energy-Resolved Positron Annihilation Rates for Molecules, *Phys. Rev. A*, submitted, 2006.
- 59. Gribakin, G. F. and Gill, P. M. W., The Role of Vibrational Doorway States in Positron Annihilation with Large Molecules, *Nucl. Instrum. Methods B* **221**, 30, 2004.
- 60. Gribakin, G. F., Mechanisms of Positron Annihilation on Molecules, *Phys. Rev. A* **61**, 22720, 2000.
- 61. Tinkle, M. D., Greaves, R. G., Surko, C. M., *et al.*, Low-Order Modes as Diagnostics of Spheroidal Non-Neutral Plasmas, *Phys. Rev. Lett.* **72**, 352, 1994.
- 62. Tinkle, M. D., Greaves, R. G., and Surko, C. M., Modes of Spheroidal Ion Plasmas at the Brillouin Limit, *AIP Conference Proceedings* **331**, 229, 1995.

- 63. Greaves, R. G. and Surko, C. M., An Electron-Positron Beam-Plasma Experiment, *Phys. Rev. Lett.* **75**, 3846, 1995.
- 64. Gilbert, S. J., Dubin, D. H. E., Greaves, R. G., *et al.*, An Electron-Positron Beam-Plasma Instability, *Phys. of Plasmas* **8**, 4982, 2001.
- 65. Greaves, R. G. and Surko, C. M., Radial Compression and Inward Transport of Positron Plasmas Using a Rotating Electric Field, *Phys. Plasmas* **8**, 1879, 2001.
- 66. Danielson, J. R. and Surko, C. M., Torque-Balanced High-Density Steady States of Single Component Plasmas, *Phys. Rev. Lett.* **95**, 035001, 2005.
- 67. Danielson, J. R. and Surko, C. M., Radial Compression and Torque Balanced Steady States of Single-Component Plasmas in Penning-Malmberg Traps, *Phys. Rev. A*, in press, 2006.
- 68. Surko, C. M. and Greaves, R. G., A Multi-Cell Trap to Confine Large Numbers of Positrons, *Rad. Chem. and Phys.* **68**, 419, 2003.
- Gabrielse, G., Bowden, N., Oxley, P., *et al.*, Background-Free Observation of Cold Antihydrogen with Field-Ionization Analysis of Its States, *Phys. Rev. Lett.* 89, 213401, 2002.
- 70. Clarke, J., vanderWerf, D. P., Charlton, M., et al., Developments in the Trapping and Accumulation of Slow Positrons Using the Buffer Gas Technique, *Nonneutral Plasma Physics V*, edited by M. Schauer, T. Mitchell and R. Nebel (American Institute of Physics, 2003), p. 178.
- 71. Meshkov, I. N., Lepta Project: Generation and Study of Positronium in Directed Fluxes, *Nucl. Instrum Methods B* **221**, 168 2004.
- 72. Platzman, P. M. and Mills, A. P., Jr., Possibilities for Bose Condensation of Positronium, *Phys. Rev. B* **49**, 454, 1994.
- 73. Mills, A. P., Jr., Positronium Molecule Formation, Bose-Einstein Condensation and Stimulated Annihilation, *Nucl. Instrum. Methods B* **192**, 107, 2002.