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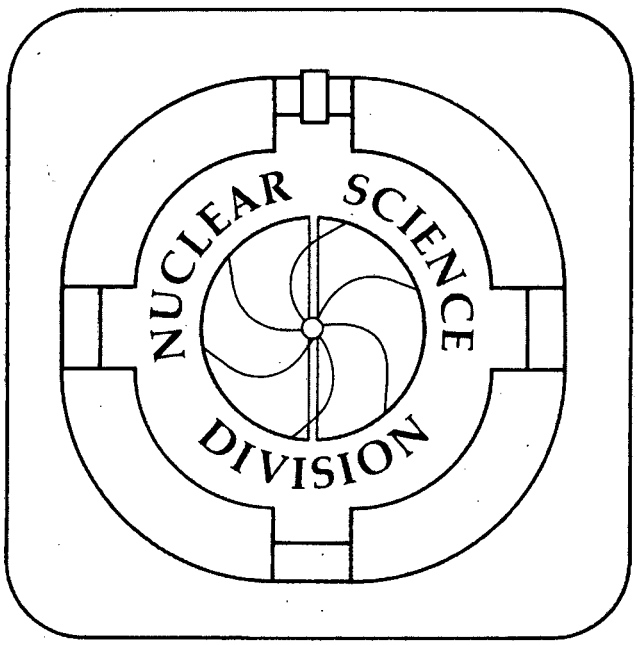

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THE EARLY DAYS OF ACCELERATOR MASS SPECTROMETRY

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Before 1978, there were just three papers that had reported work in high energy mass spectrometry—two in 1938, by me and my student Robert Cornog, and one in 1977, a preprint by another of my students, Richard Muller, and me. But a year later, Rich Muller published his classic paper on accelerator dating, and used a cyclotron to determine the age of a water sample from its Tritium content. This paper clearly outlined the techniques of ^{14}C dating by accelerator, and it was published before anyone else started to work on this now important field. Although he mentioned the usefulness of linear accelerators, he did not refer explicitly to the tandem accelerator, which is now the most widely used instrument. The fact that several tandem machines are now being dedicated to accelerator dating gives me an excuse to tell you of my involvement in the origin of charge exchange acceleration, an interesting story that is not part of the general lore of physics.

I have an intense dislike of pictures of the white-haired Albert Einsten because he was then no longer the wonderfully creative physicist who gave us special and general relativity. I have a picture of the real Einstein on my desk—a very young patent examiner who did marvelous things in his spare time. And although I am exceedingly interested in the history of physics, I have recently declined invitations to two conferences on the subject. That is because I would rather remember my friends who did pioneering work in nuclear physics as the vigorous young men they were, four and half decades ago, instead of the white-haired old men I would meet at such conferences and see in my shaving mirror. Emilio Segre brought this problem into focus for me at a talk he gave a few years ago on the life of Enrico Fermi. He told of Fermi's first visit to Gottingen, "where he met the boys who made quantum mechanics."

So I'll be telling you about what a couple of boys did in 1939, what a young man did in 1951, and what another young man and a now older man did in 1977, all of which was a prologue to this meeting. I should apologize for talking so much about my own work, even though that is what I was asked to do. In case you think I seem carried away by its importance, I'll ask you to remember that in 1977 I had stored away in various parts of my brain all the facts that Rich Muller put together to come up with the concept of

accelerator dating. If I can get you to ask the appropriate question, "How could anyone know all that Alvarez knew about the techniques, the dating problem, and its importance, and not arrive at the solution?", I will perhaps help you to put my remarks today in their proper perspective.

I'll make one more comment on my reasons for accepting this speaking assignment. I have always been unhappy about scientific history because, in my view, it so often skirts what I think is the most important element in a new observation or theory—how did this person happen to be doing this thing or thinking along these new lines? So, in a sense, I'm giving this talk to take you behind the scenes, to help you understand how I happened to be mixed up in three unusual projects.

I arrived in Berkeley, in May of 1936, with a brand new Ph.D. and essentially no knowledge at all of nuclear physics; I had worked with Arthur Compton in the field of cosmic rays. Very fortunately for me, the first of Hans Bethe's three monumental articles on nuclear physics appeared in the April 1936 issue of the Reviews of Modern Physics. I studied "Bethe's Bible" with great diligence, and very quickly I was "up to speed" in that I knew all kinds of important things about nuclear physics that even my new and experienced colleagues hadn't known the week before. Hans Bethe was making his transition from spectroscopist to nuclear physicist, and, in his characteristically thorough manner, he learned everything that had been done in his new field, added a lot of new ideas and calculations, and, most importantly for the rest of us, put it all down in very clear prose.

In addition to learning almost everything I knew about nuclear physics from those wonderful articles, I was most fascinated by several things that Hans said couldn't be done with the then present state of the art; and I worked on several of them. (Some of them became possible when reactors were built.) My most successful venture was discovering nuclear K-electron capture, a mode of decay I learned about from his article, which he said couldn't be directly observed because nothing was emitted except a neutrino. I corrected a minor error in one of his papers when I showed that internal conversion of gamma rays occurred in ^{67}Ga; he had said that such an effect could only be seen in the heavy "natural" radioactivities. The next of his "unobservables" that I tried to observe was the beta-decay of the free neutron, which I wouldn't have known about except for his article. He said, "The lifetime is too long to allow observation of the beta-decay of neutrons." The last pre-World War II experiment to be set up at the Berkeley 37-inch cyclotron was a search for that decay; my graduate student collaborator was Cornelius Tobias, and the experiment was terminated when the

cyclotron was reconfigured as a mass spectrometer to separate the uranium isotopes. Robert F. Mozley and I tried a different technique, using our Van de Graaff generator as a source, to observe the neutron decay after the war; but before it was successful, Art Snell of Oak Ridge and John Robson of Chalk River both did such good jobs using reactor neutrons that we were effectively "wiped out." (An interesting aspect of our unpublished work was the first observation that charged particles—electrons, in our case—could be stored in a "magnetic bottle" for long periods of time; this clue may help some of you to reinvent our experiment.) My last challenge of this kind involved Bethe's statement that "the probability of the disintegration of nuclei by neutrinos is so unobservably small--." Shortly after the war when all papers on nuclear physics were born "classified," Bruno Pontecorvo and I independently suggested that neutrino interactions could be observed in CCL_4, and if the Majorana theory was correct, anti-neutrinos from reactors could also be so observed. (My very long paper, which discussed the many sources of background counts, was declassified about ten years later.) As you all know, Ray Davis has spent about twenty years looking for solar neutrinos by this technique, and showing very importantly that the sun produces only about one third as many neutrinos as theory says it should.

One of the problems to which Bethe devoted a lot of attention involved Helium 3 and Hydrogen 3. He didn't question the universal belief that ^3He was heavier than the stable ^3H, and he quoted experimental results, from the D-D reaction energetics, to indicate that ^3He was about 0.2 Mev heavier than ^3H and should capture an electron to become ^3H with a "lifetime of about 5,000 years." ^3He and ^3H had of course been observed for the first time, as high speed reaction products from D+D collisions, by Oliphant, Harteck, and Rutherford in 1934. It was as though the alpha particle had been discovered but the Helium atom had not yet been seen—neither of the nuclei of mass three had been observed after they had come to rest, although several mass spectroscopists had incorrectly reported that they had seen stable ^3H in enriched water samples. The best way I know to show that everyone (and I don't even need to put quotation marks around that word) believed ^3H to be stable is to tell you of the very last paper Lord Rutherford published. It appeared in Nature, just 414 pages before the announcement of his death, and was entitled "Search for the Isotopes of Hydrogen and Helium of Mass 3." Rutherford made arrangements with the Norwegians to process nearly 50 kg. of 99.2% heavy water by further electrolysis down to a volume of 11 cubic centimeters. The overall reduction in water volume was about a factor of 10^9. Aston used his mass spectrometer to examine this sample, and reported that he could find "no trace of ^3H"; Rutherford was "very disappointed after the time and labor spent in

Norway in preparing this material." In the last three sentences he wrote during his fabulously successful life as an experimental physicist, he said,

> "A number of experiments have been made to detect the Helium isotope of mass 3 in ordinary Helium by direct spectroscopic methods, but with entirely negative results. This, however, is not surprising when we consider that terrestrial Helium is probably derived from alpha particles of mass 4 expelled from radioactive substances present on our earth. It is a striking fact that while in transmutation experiments using counter methods the D-D reaction is on a marked scale, giving rise to very large numbers of ^{3}H and ^{3}He particles, yet it does not seem feasible at the moment to obtain sufficient quantities of these two interesting isotopes to study their properties by ordinary physical chemical methods."

I'm terribly sorry that Lord Rutherford didn't live another two years, so that he could have learned how I showed in a very simple way that all three of the last sentences he wrote in his distinguished career suddenly were in need of revamping. I am confident that had he lived, one of my most prized possessions would be a letter from the grand old man himself, complimenting me on my work with Hydrogen and Helium of mass 3, which I shall describe.

But first let me relate an interesting incident that should convince anyone that Lord Rutherford never for an instant suspected that ^{3}H was radioactive. Bill Libby (the inventor of ^{14}C dating) told me that shortly after World War II, he was at the Cavendish Laboratory and asked the curator of the laboratory's museum if he could locate Lord Rutherford's old enriched water sample. When the sample was produced, Bill Libby had already found a portable Geiger counter, and he immediately put the water sample close to the detector tube. The result was a loud and rapid clicking of the loudspeaker, as the bremsstrahlung from the radioactivity of Tritium was detected by the Geiger counter. The fact that Lord Rutherford, with the great resources of the Cavendish Laboratory at his disposal, never made this simple test is what made me start this section with the observation that everybody at this time thought that Tritium was not radioactive, but rather a stable form of Hydrogen. And, as Hans Bethe had shown in his "Bible," that meant that ^{3}He was radioactive.

One night at home, almost two years after Lord Rutherford's article appeared in Nature, I was contemplating the ^{3}H-^{3}He problem, and I quickly calculated how much of both isotopes could be produced at the 37-inch cyclotron by a bombardment of Deuterium with deuterons. I was immediately impressed by the magnitude of the numbers, just as Rutherford had been, independently, two years earlier. My numbers showed that if I bombarded Deuterium in the 37-inch cyclotron for only an hour, introduced it into the ion source of the 60-inch cyclotron, and "tuned" the magnetic field so that the cyclotron

could accelerate ^{3}He ions, there should be a large enough beam current for the individual ^{3}He ions to be detected easily with my "thin ionization chamber." Since the 60-inch cyclotron had just been turned on, and since I had a thin ionization chamber plus pulse amplifier that I had used in many experiments, I decided to look for the radioacitvity of ^{3}He. I thought that if I tuned the 60-inch cyclotron for ^{3}He and fed it with bombarded Deuterium, I would certainly see the accelerated ^{3}He ions; and if I was lucky and the half-life of ^{3}He was not more than a year, I would be able to see the beam current slowly decay with its radioactive half-life, as I did the experiment on various occasions spaced out over the next few years. The only uncertain element in the experiment was a possible background of accelerated "junk ions," that might be produced in the cyclotron when it was tuned to accelerate the ^{3}He ions. As far as I knew, no one had ever put an ionization chamber able to detect individual accelerated ions in front of the beam window when the cyclotron was tuned through the range of magnetic fields where one would expect to see ^{3}He ions. If for some unsuspected reason there was a large background of ions in this region, then I couldn't do the experiment that I have just outlined. It was obvious to me that I should take a look myself to see if there were any background ions that would mask the effect that I would otherwise certainly see.

I therefore talked to one of the laboratory's uncommitted graduate students, Bob Cornog, and asked him if he would like to work with me on a search for the fate of the ^{3}H and ^{3}He ions that were produced in the D-D reaction. Bob was a friendly and energetic young man who hadn't yet done an experiment at the laboratory. He agreed with me that the first thing we had to do was check the background at the 60-inch cyclotron with the magnetic field set at three quarters of its normal value. Bob helped me push my amplifier cabinet from the old wooden 37-inch cyclotron laboratory to the new 60-inch laboratory just across the alleyway. I set the ionization chamber in front of the cyclotron's thin window and got the equipment running. Bob said that he had to throw the hammer in a track meet that afternoon, but he'd be back to work with me in the evening. But things started to work well in the middle of the afternoon, and I made arrangements with the cyclotron crew to operate the machine with its normal field—where I could observe both the deuterons and the ^{4}He ions being accelerated through the thin window and into my ionization chamber. Huge bursts of these ions paralyzed the amplifier, but it recovered quickly as soon as the oscillator was turned off. I then asked the crew to lower the magnetic field to three-quarters of its normal value, where ^{3}He, rather than ^{4}He, would be accelerated according to the cyclotron equation. They must have spent about half an hour adjusting the magnetic field to various values in this general range, while my eyes were glued to the cathode ray oscilloscope looking for

pulses. We had no intercom between the recently finished cyclotron target area and the control room, so most communications were by shouts. On occasion I would go into the control room to talk with my old friend Bill Farley about new search procedures. But it seemed pretty clear now that there were no "junk ions" being accelerated at any magnetic field values where the ^{3}He ions Bob and I were about to make with the 37-inch cyclotron should appear. This was naturally good news to me, since it meant that the "real experiment" could soon be undertaken, with almost a guarantee of success.

In my last conversation in the control room with Bill Farley and the other operators, I said that I was now convinced that there was no background in the ^{3}He range, but that I would like them to take the magnetic field once again up to full value so that I could make sure that my ionization chamber and amplifier were still working by watching the scope screen become paralyzed with enormous numbers of beam particles. I thought this simple act would terminate the exploratory experiment, but as we will now see, it led to one of the most important observations I ever made. If anyone were writing a scenario for this experiment, he would have ordered two things that fortunately didn't happen. In the first place, the operators should have turned off the radiofrequency oscillator that powered the "D's"--the cyclotron's accelerating electrodes. In keeping with good cyclotron practice, only after cutting the oscillator power would the operator have then turned off the magnetic field. But probably because the operators had been running the magnetic field up and down for the past half hour, always with the oscillator on, they cut the magnet power while the oscillator was left on. That was the first fortunate accident. And the second thing that would have been written into the scenario was that I would have walked away from my apparatus, knowing that the experiment was over. But for some reason I kept watching the oscilloscope screen after I had shouted "Cut." I was startled to see a burst of pulses on the oscilloscope that quickly appeared and then disappeared, as the magnetic field dropped through the "Helium three region." (The time constant of the magnet was many seconds because it wasn't constructed of laminated iron as a transformer is. So by Lenz' law, eddy currents in the iron kept the magnetic field from dropping rapidly to zero strength.) Soon thereafter there was a huge burst of pulses as the magnetic field went through one-half its normal value, where protons were accelerated. After I'd seen the protons, I ran to the control room and said, "Let's try that again." I probably also told them what had happened, so we repeated the experiment several times, and every time the magnetic field dropped rapidly throughout the Helium three region, I saw the pulses suddenly appear and then disappear. It was quickly obvious to me what was making them appear during the rapidly decreasing field although they hadn't shown up when the magnetic field had the same constant value. My

knowledge of the way eddy currents would behave, of the long "time constants for decay" of the cyclotron, and of the principles of the magnetic "shimming process" (the tailoring of the field shape to give proper vertical focusing) all combined quickly to tell me that the cyclotron was being fortuitously reshimmed by eddy currents in the pole pieces of the magnet caused by the rapid rate of change of the magnetic field downward. The long apprenticeship I had spent in the early design of the 60-inch cyclotron was now paying good dividends, as I quickly diagnosed the situation. (But that same apprenticeship had failed me earlier in the afternoon when I hadn't realized that the non-linear characteristics of the iron poletips of the magnet had destroyed the vertical focusing characteristics of the magnetic field when it was operating "far below saturation." So my observation that there was no "interfering background" at the ^{3}He magnet setting was now demonstrably wrong.)

The pulse height of the new particles was equal to that of the ^{4}He ions seen at full magnetic field. Since the ions had the same velocity, this meant that the new ones had a charge of 2. I now dashed over to my radioactivity laboratory where I had a box of carefully calibrated aluminum foils of various thicknesses that I had long used as absorbers of beta rays. I quickly calculated the range of ^{3}He particles in air, after such particles had been accelerated in the 60-inch cyclotron. (It was of course just 3/4 of the range of the ^{4}He ions.) I could therefore pick out just the proper amount of aluminum to keep these ^{3}He particles from entering my ionization chamber. After perhaps a score of "quick passages," with various thicknesses of aluminum foil interposed between cyclotron and ion chamber, I had confirmed the fact that the particles I was seeing on the oscilloscope were truly ^{3}He ions that had come from the Helium bottles that fed the cyclotron ion source. The Helium had in turn come from deep wells in Oklahoma where it had rested undisturbed for about one hundred million years. So ^{3}He wasn't radioactive, as everyone had so long believed—it was stable against beta decay; it was a natural constituent of ordinary gas-well Helium.

When Bob Cornog returned, after he had won the hammer throw, I had an exciting experience telling him everything that had happened that afternoon. It was obvious that the first thing we should do was shim the cyclotron with a set of circular pieces of iron, so that it would produce a steady beam of ^{3}He ions. Nothing further could be gained by observing isolated bursts of ^{3}He ions while the cyclotron was shimming itself through eddy currents. That seemed to be too much like Charles Lamb's famous story of the Chinese gentleman who accidentally discovered the joys of roast pork, and then had to burn down a house every time he wanted to experience again that delicious taste.

The importance of high energy mass spectrometry was demonstrated that afternoon for the first time. Instead of measuring a current of ions to a collector plate in a vacuum chamber, as the men who had incorrectly reported observing ^{3}He had done (when they were probably observing the molecular ion HD^+), I made the ions traverse an aluminum window which would split a molecular ion apart. I could then measure the true charge of the ions by pulse height and the true kinetic energy from the range. I also measured e/M from the cyclotron equation and velocity from the frequency and final orbit circumference, and all these numbers agreed among themselves to give both e and M. You are all familiar with these advantages of the accelerator technique, but I stumbled on them accidentally that Saturday afternoon. I had used a cyclotron in my experiment, not because of these advantages, but because it was the only mass spectrometer to which I had access!

In the next few days, and after a lot of trial and error, Bob Cornog put together a set of circular iron shims that would permit the 60-inch cyclotron to yield a constant beam of ^{3}He ions. Because of the work of Bob Wilson, the art of shimming had been replaced by a science of shimming, and this was of great assistance to Bob Cornog in his adjustment of the 60-inch cyclotron. So now we were ready to do some experiments with our newly discovered stable ^{3}He ions.

We did three principal experiments--a rough measurement of the relative abundance of ^{3}He and ^{4}He in a normal tank of gas-well Helium, and a fairly accurate measurement of the relative amounts of ^{3}He in gas-well Helium and in Helium derived from the atmosphere. And finally we used ^{3}He as a new kind of bombarding projectile to excite the reaction $^{28}Si(^3He,p)^{30}P$. We observed the famous 2.5 minute period of Phosphorous 30--the first artificially radioactive isotope ever made, which had the seen the light of day five years earlier in the laboratory of Fredrick Joliot and Irene Curie.

Bob Cornog and I wrote two very short Letters to the Editor of The Physical Review in the summer of '39. (Taken together, they occupied less than one page of the journal.) The first, dated July 31, was entitled "^{3}He in Helium" and it described the observations I made with the rapidly varying magnetic field. The second was dated August 29, and was entitled "Helium and Hydrogen of Mass 3." In this letter we described our experiments that were done after Bob had shimmed the cyclotron to produce the steady ^{3}He beam. We correctly observed the unexpected effect that ^{3}He was 10 times more abundant in atmospherically derived Helium than in gas-well Helium--we now know that the extra ^{3}He is produced by cosmic rays, just as ^{14}C is produced by cosmic rays in the atmosphere. We were off by a factor of 10 in our absolute abundance

of ^{3}He, but that was not an unreasonable error to make when the cyclotron was adjusted so differently to accelerate ^{3}He ions and ^{4}He ions. (We quoted 10^{-8} and 10^{-7} as the abundance of ^{3}He in gas-well and atmospheric Helium, whereas the present values are 10^{-7} and 10^{-6}.) Although we didn't mention it in our letter, we also obtained samples of Helium, from Professor Giauque's laboratory, which had been liquefied and evaporated many times. I thought that we might see the effect of fractional distillation, but there was no such effect. And we did report seeing ^{30}P.

In the second paragraph of our second letter we reported the first observations of the radioactivity of ^{3}He. I'm embarrassed to recall that it took me more than a week after the first observation of ^{3}He to realize that this meant that ^{3}H must be radioactive, and that we should search for that radioactivity. As soon as I told Bob Cornog what I had concluded about the radioactivity of ^{3}H, or Tritium as it is now universally called, he took charge of this part of the experiment and worked like a Trojan to build the equipment and make the observations. He bombarded a sample of Deuterium gas at the 37-inch cyclotron and showed that it was very radioactive with a lot of contamination activities. He then passed the gas through activated charcoal at liquid air temperature, which removed most of the contaminants, and then finally let the gas diffuse through hot Palladium. Hydrogen is the only gas known to diffuse through Palladium, so the gas which came out of the other side, and was used to fill an ionization chamber, was all heavy Hydrogen with no trace of impurities. We introduced this gas into a large ionization chamber with a volume of about one liter and observed its radioactivity on a vacuum tube electrometer. In our second letter we said:

> "The gas showed a definite activity of long half-life. The radiation emitted by this Hydrogen is of very short range as was shown by the almost linear form of the intensity vs. pressure curve when the gas was pumped out of the chamber. Once sufficient time has elapsed for us to make some statement regarding the half-life of this activity, we will submit the details of the work to this journal for publication."

The beta rays from Tritium were so low in energy, and of such a short range, that we could not get them through the thinnest aluminum foils into a more conventional radiation detector. Since we were unable to make a normal range measurement on the beta rays, I thought up the idea of looking at the activity vs. pressure curve. (For ordinary beta rays, the ionization would vary as the square of the pressure; if the pressure were cut in half, half the radioactive atoms would be lost, and each beta ray would make only half as many ion pairs in the lower density gas.) I doubt if anyone ever used this technique, either before or since, but it showed very well that the beta ray

energy was small indeed. (We now know that the beta rays have a maximum energy of 18.6 keV--about 1% of the typical beta ray energy one meets in nuclear physics.)

As far as I know, these were the last uses of an accelerator as a mass spectrometer until 1975 when I proposed a quark search to Rich Muller. I'll now describe how the tandem accelerator came into being.

In the Spring of 1975 I was eating breakfast before teaching an eight o'clock class in nuclear physics and reviewing what I would say about accelerators. I was prepared to tell my students that whenever they saw a singly charged particle beam produced in an acclerator that had an energy in electron volts that was greater, numerically, than the highest voltage seen in the machine, they would know that there was a changing magnetic field somewhere inside the box. I couldn't think of an exception to this general principle until I had finished my coffee and started to drive to the campus. But during the ten minute drive I realized that, if I made the accelerated ion change its charge from -1 to +1 inside an electrode, I could violate my newly postulated "law."

I published a one page article in the Review of Scientific Instruments in September 1951 entitled "Energy Doubling in dc Accelerators." This paper started with these sentences:

> "It is generally believed that charged particles cannot be accelerated from ground potential to ground potential unless they pass through a system which has associated with it a time varying magnetic field. Dc electric fields must satisfy the equation $\oint \mathbf{E} \cdot d\mathbf{s} = 0$, while the time varying fields used in radiofrequency accelerators and betatrons are freed from this restriction of scalar potential theory."

As soon as the paper appeared, I had a call from an old friend from wartime days, Dennis Robinson, who was then President of the High Voltage Engineering Corporation. He asked if he could visit with me the next week to talk about my recent invention, which he said he'd like to build and sell with a license from the Atomic Energy Commission (which I had thanked for supporting my work). I was of course pleased, and when he came to Berkeley we did talk about it for several days. One thing that concerned us was an unusual discharge mode that might be seen in a "straight through" charge exchange accelerator, where electrons could oscillate back and forth through the high voltage electrode. I thought for a while about converting my own 4 Mev Van de Graaff generator into a charge exchange accelerator, with a magnet in the high voltage terminal, but I ended up deciding to leave the whole business to Dennis Robinson. He invited me to be a consultant to his company, but I was actively consulting with Ernest

Lawrence's color television company at the time, and regretfully told Dennis that I couldn't spare the time. You are all familiar with the success his company has had in building and selling tandem accelerators, the possibility of which he first learned from my article.

I'll now transport you back thirty years to show that "everyone" at that time accepted the fact that the tandem was a brand new idea, which I had invented. This time, I'll have to put quotes around "everyone," because it turned out that there was a U.S. patent on such accelerators, but the accelerator fraternity didn't know of its existence. You have all heard of an identical situation, where the idea of strong focusing was first published in 1952 by Courant, Livingston, and Snyder, but then Nick Christophilos was found to have patented the idea two years earlier quite unbeknownst to the accelerator fraternity. The fact that Courant, Livingston, and Snyder are almost always credited with the discovery of strong focusing is probably somewhat due to to the fact that Christophilos wasn't "a member of the club"--he was an engineer who designed elevators in Athens and invented accelerators as a hobby. Contributing reasons may also be that strong focusing was the first major scientific accomplishment of the relatively new Brookhaven National Laboratory, but most importantly, in my view, was the fact that the independent Brookhaven discovery led immediately to the building of many strong focused accelerators, whereas Christopholos' work was buried in the patent literature, and had no influence on the development of accelerator technology.

The parallels between the tandem and strong focusing inventions are extraordinarily close, with one exception. The second inventors of the focusing scheme have received almost all the credit, whereas the man whose name is always associated with the tandem--Robert Van de Graaff--was not another independent inventor, but rather an excellent developmental engineer and Chief Scientist at the High Voltage Engineering Co. Neither Willard H. Bennett, who applied for a patent on the tandem in 1937, nor I, who first published it in 1951, are remembered as having anything to do with the matter. I really can't complain, since the accelerator fraternity has treated me well; for years they have used my name as an adjective to describe resonant cavity ionic linear accelerators. My personal satisfaction in the tandem matter is best illustrated by the following story that concerns the infinitely more important, and hotly contested, invention of medical anesthesia. Medical men almost universally credit that to W.T.G. Morton, who demonstrated it at the Massachusetts General Hospital in 1846. Later it was found that Crawford Long, a Georgian doctor, had performed operations under ether in 1842, but had then given up that practice without publishing his work. Long's statue is

in the U.S. Capitol's Statuary Hall as the "father of anesthesia." I remember reading a poignant statement by Morton's widow who defended her husband's claim as a public benefactor by saying something to the effect that although others may have been first, her husband's demonstration was followed immediately, worldwide, by the universal use of anesthesia. In an aside she wondered how those who claimed the credit could live with the knowledge that, because of their neglect to publish their work, countless people had suffered terrible tortures from which they could have been saved by a knowledge of anesthesia.

Compared to the invention of anesthesia, one of mankind's greatest boons, the tandem affair is of negligible importance. For that reason, I have never mentioned it either in print or in a talk in the past 30 years. But now that the occasion seems ripe for such a mention, I'll say that my satisfaction comes from two considerations: Number one is that I did it independently, and number two is that my publication led immediately to the worldwide use of a valuable technique that until then had been buried for more than a dozen years in the patent literature. I'll add in passing that, although everyone knows that all major accelerator improvements such as the cyclotron, betatron, and linear accelerator have been patented, the accelerator literature doesn't contain references to such patents, but only to articles in the open literature.

I'll conclude this vignette with an observation I once made when reading Livingston and Blewett's classic book on accelerators. In their brief section on charge exchange acceleration, they say that this idea was "proposed by W.H. Bennett[42] and L.W. Alvarez[43]." I expected the Bennett reference to be to his U.S. patent, because I was sure that he hadn't mentioned it in the open literature. But instead, I found three separate references to the 1936 Physical Review—one article and two abstracts. I immediately looked them up, and there was not a single word about charge exchange in any of them; they were all simply on the generation of negative hydrogen ions!

The experiment that revived Accelerator Mass Spectrometry was published in 1977, and came about in the following way. As a particle physicist I had been in conversations about quarks from the earliest days—in fact my bubble chamber group had published a good deal of the data that had led Murray Gell-Mann and, independently, George Zweig to propose the quark theory in 1964. Their quarks had electrical charges that were +2/3 and -1/3 times e, and many searches for such fractionally charged particles were soon underway. A year after the original quark papers appeared, Han and Nambu proposed a theory of integrally charged quarks. It was not taken very seriously at the time, because it required that there be nine kinds of such quarks, rather than the simpler original

theory that needed only three kinds of fractionally charged quarks. But as the years went by, it became apparent that the original three-quark theory needed to be modified by the addition of "color," and the number of quarks was therefore raised to nine. As more and more quark searches failed, I became interested in looking for integrally charged quarks because the two theories now seemed equally simple. I spoke to John Reynolds about the possibility of looking for Hydrogen ions of anomalous mass with one of his mass spectrometers. In fact, I suggested that we use two such devices in tandem, so scattering from slits, etc. could be eliminated as a source of incorrect mass measurements; and I further suggested that we use time of flight techniques as an additional verification of the mass of the Hydrogen-like quarks. John was anxious to collaborate, but we weren't able to fund the apparatus development, and we lost interest in the matter.

Several years later, when I was reading the performance specifications of our Laboratory's 88-inch cyclotron, I realized that it was ideally suited to look for positive singly charged stable quarks. It had been designed to give good magnetic focusing over a wide range of magnetic fields, and its frequency could also be changed quite easily. I explained to Rich Muller that if we looked for Hydrogen ions in the mass range from 0.3 AMU to 8 or 9, which was a practical limit for the 88-inch cyclotron, we would either find the quarks or set quite fantastic limits on their abundances in water. Not long after we started our search, I read a paper by Okun and Zeldovich in which they not only discussed the production of such quarks by cosmic ray bombardment—the mechanism I had calculated—but also suggested that such quarks could also be remnants of the big bang.

I once heard Rich Muller talk about his work in accelerator mass spectrometry, some of which greatly extended the work we did together on the quark search. He said, "Since the ideas all came from Luie, I felt that the only way I could earn the right to have my name on the paper was to do <u>all</u> the work." And he did do all the work except for running the cyclotron, which was done by Bill Holley and Ed Stevenson, whose names also appear on our paper. He borrowed all the solid state ionization detectors, counting equipment, etc., and I had the pleasure of sitting in the cyclotron control room, night after night, watching Rich make all the adjustments and request all the changes in operating conditions that I had personally made, by shouting, forty years earlier. It was Rich's first encounter with a cyclotron and he obviously enjoyed it.

Our paper gave limits on +1 charged quarks relative to protons as less than 2×10^{-19}, from mass 3 to mass 8.5 and about 10^{-14}, below mass 2. From mass 2 to 3, the limit varied from 10^{-18} to 2×10^{-19}. We said,

> "The advantage of the cyclotron over an ordinary mass spectrometer does not come from its high resolution—but from the high energy of the emerging beam: several Mev per nucleon. This high energy allowed us to send the beam into the particle identification detectors, and to get useful information on a particle-by-particle basis."

As far as I know, this statement in 1977 is the first overt mention of those features of acclerator mass spectrometry which you all appreciate. I had known them for some time, but they hadn't ever been stated so clearly until then.

That wraps up the early history of accelerator mass spectrometry; but I might say a few words about its application to "accelerator dating," a field in which I've had no direct involvement, even though I did contribute to its evolution in three ways. The first came from some conversations I had several years ago with Rich Muller when he was spending the summer trying to help our navy find Soviet submarines. He was studying the "wake radioactivity" left in the water behind any nuclear submarine whose reactor is not perfectly shielded. Seawater is the cheapest material with which to shield such a reactor and the question is then what is the easiest radioactive material to detect. Sodium 24 will certainly be made in quantity, and it emits two very high energy gamma rays in coincidence—a most unusual signature. But one's first guess is seldom the best, and I can't remember what Rich looked at next, but it had a higher production rate, and an uncomfortably long half-life, and therefore a smaller counting rate. I told Rich that some years earlier, I had seen a research proposal in which someone from SRI had suggested improving the accuracy of ^{14}C dating, by counting the atoms in a mass spectrometer, rather than waiting for them to decay, with their very long half life. I remembered that he had proposed using negative ions as a way of eliminating background. Rich and I agreed that the method probably hadn't worked, or we would have heard about it, but the idea of counting atoms rather than decays did appeal to him, and he tried to make use of it in the submarine detection business but with no success.

Then after we had made our quark search on the cyclotron, Rich put all the parts together and proposed to do accelerator dating, using ^{14}C, ^{10}Be, and ^{3}H, among other materials. So I soon found myself in my old role as spectator in the control room at the 88-inch cyclotron, as Rich performed the first accelerator dating experiment ever done—the measurement of the age of a water sample from its ^{3}H content. The sample was 24 years old and had been collected before the Tritium content had been raised by the thermonuclear bomb testing in the 1950's. Rich measured its age as 33 years.

So my first two contributions to accelerator dating, if they could be dignified by that description, were to introduce Rich Muller to the concept of accelerator mass spectrometry and to the fact that someone, unknown to me, had suggested that ^{14}C dating might be done by counting atoms, rather than decays. I did make one concrete suggestion to reduce background, after Rich had first proposed accelerator dating, but I'm sure he would have arrived at the same solution without my help. That was to separate isobars, for example ^{14}C from ^{14}N, by the "range method.

I've been pleased to see the rapid strides that have been made in a field that I apparently kicked off, quite unknowingly, and I've also enjoyed the opportunity to meet so many of the people who have contributed to the technology—people who were familiar to me by name, but not by face. And thank you for the opportunity to reminisce about events of long ago.

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