MUONIUM-THE EARLY EXPERIMENTS

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During the period from 1957 to 1964 a series of experiments led by Vernon Hughes was conducted at the Columbia University Nevis Cyclotron. The formation of the μ^+e^- atom, muonium, in purified argon was established through the observation of the Larmor precession frequency characteristic of muonium. The ground state hyperfine splitting was first roughly measured with a depolarization quenching experiment and subsequently with a precision microwave resonance experiment. At the end of this period the hyperfine splitting had been measured to 4 ppm establishing that the ground state hyperfine splitting of muonium ($\Delta\nu$) could be calculated with high accuracy from the quantum-electrodynamic bound-state two body equation. Any breakdown of this basic assumption, such as unknown couplings to the muon or electron fields, would alter the theoretical value of $\Delta\nu$.

1. Introduction

An INSPEC search on the topic "muonium" back to 1969 will report 4597 papers with 124 of these in 2003-2004 alone. The current muonium literature includes papers not only on precision measurements of the ground state hyperfine splitting where the current precision is 12 parts per billion, but on muonium applications in Solid State and Chemistry. For Vernon Hughes, muonium represented 47 years of continuous involvement from 1957 to 2003. This article will review how this came about and describe the early muonium experiments. To do this we will have to turn the clock back to 1957.

2. Early Background and History

After the discovery of parity non-conservation in the weak interactions and the realization that stopping muon beams could be highly polarized, Chicago and Columbia groups studied muon depolarization in various materials. It was discovered that the stopping muon polarization could range from 0 to a value consistent with 100% polarization depending on the stopping material. While the μ^- largely form mu-mesic atoms and disappear by muon capture, the μ^+ disappear by free decay and the resultant positron asymmetry with respect to the muon spin direction can be used to determine the muon polarization at the time of decay. The Chicago group under Val Telegdi noted in one of their 1957 papers that *"The formation of a μ^+e^- atom **"muonium"** can be the possible cause for the reduced asymmetry of $\mu - e$ decay in certain materials.* The history of the early muonium searches and various remarks on formation are partly in Bulletin of the American Physical Society abstracts for short talks presented at Society meetings.The formation of muonium and its depolarizing effects was specifically discussed in several papers.[1] A subsequent paper by Breit and Hughes pointed out that microwave induced transitions between muonium magnetic substates could be used to test assumptions regarding muonium formation and that the microwave frequencies at which the transitions are induced depend upon the muonium hyperfine splitting.[2]

3. Early Experiments

There followed a number of experiments at Chicago and Columbia to detect muonium directly. In the Columbia experiment It was suggested that muonium is formed when positive muons are stopped in nitrous oxide gas.[3] Hughes, together with student D. McColm and A. Lurio subsequently searched for muonium in N_2O and Ar by looking for a change in muon polarization upon the application of low frequency RF at the muonium Zeeman frequency.[4] No muon polarization change was observed, but the authors pointed out that gas impurities, particularly O_2 and NO could inhibit the formation of free muonium.

I joined the small Yale Hughes group, then consisting of K. Ziock and Yale student D. McColm, in 1958 at the suggestion of L. Lederman who was my Columbia advisor. The group was systematically studying muon depolarization in gases, searching for muonium formation candidates. The idea was to start with gases known to support the formation of positronium. The gases studied were O_2, N_2O, SF_6, and Ar. At the same time an argon purification system was under construction due to the oxygen contamination concern. On the basis of the depolarization measurements we decided that purified argon was the best candidate for muonium formation. Once the argon purification system was ready, we set up a low transverse field experiment to search for the direct precession of the muonium triplet state.

The triplet state would be expected to precess at one half the free positron precession frequency since the g factor is one compared to the positron g factor of two. The procedure was to observe the positron rate at fixed angle in about a 4 gauss transverse magnetic field. Since the muonium precession frequency is about 1.4 MHz/gauss, more than 10 precession periods would be observed in a 4 gauss field in a muon lifetime. The observed effect would then be a sinusoidal modulation of the decay rate as a function of time.

4. First Observation of Muonium Precession

After several runs in late 1959 ruling out all gas targets except purified argon, the muonium precession signal was observed in the purified argon target early in 1960.[5] The target fully depolarized the muons in a transverse field of 3-5 gauss and the time dependence showed the precession modulation at the muonium frequency. The stainless steel target contained argon at 50 atm. The gas was circulated through an in-line titanium getter heated to 500°. The experimental arrangement is shown in Fig.1 A photograph of

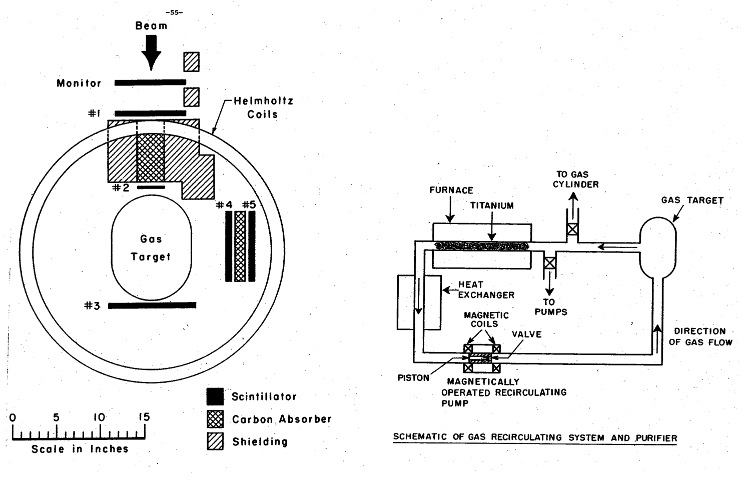

Figure 1. Experimental arrangement. Left:target; Right:purifier

the beam line showing the target, helmholtz coils and the brick enclosed purifier is shown in Fig. 2.

The electron decay signals were fit to an exponential modulated by a

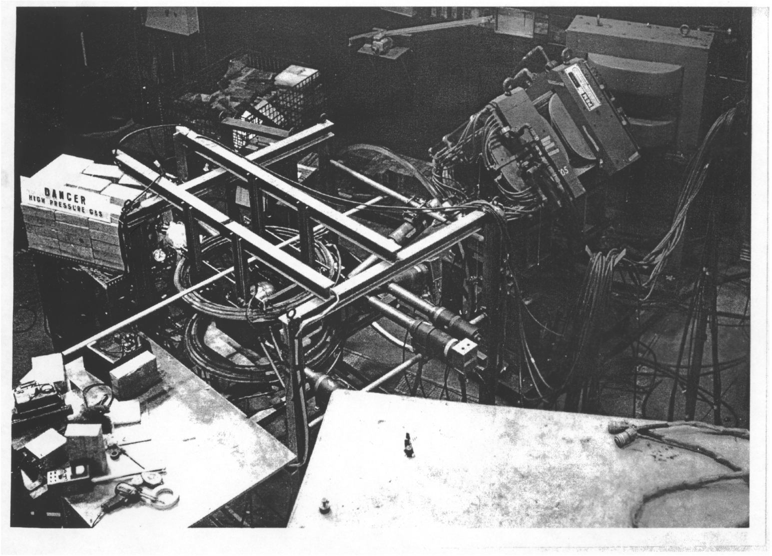

Figure 2. Beam-line photo showing target and purifier.

muonium Larmor precession term as:

$$y_i = e^{-t_i/\tau}\{C + Ae^{-t_i/\tau'}\sin[2\pi f(t_i + t_0)]\}$$

Here τ is the muon lifetime, τ' is a parameter introduced to allow for line broadening and f is the trial value for the precession frequency of the magnetic moment of muonium. A typical positron decay distribution is shown in Fig. 3. The Larmor precession wiggles are not apparent by inspection due to the relatively poor statistics, but the Fourier analyses always showed a strong peak at the larmor frequency.

Results of the frequency analysis of the decay data for several different values of the magnetic field are shown in Fig. 4. The solid curves were obtained from the analysis of the data and represent the percent amplitude of A compared to the total counting rate. The error bars correspond to an error of one sigma in the percent amplitude. The dashed curves are are theoretical line shapes centered in each case at the muonium precession frequency predicted from the measured value of the magnetic field. Resonances are clearly seen at the frequencies which are predicted for muonium precession on the basis of the magnetic field measurements. The observed amplitudes of the resonances are 4 to 5 standard deviations. Additional runs with unpolarized muons showed no resonance signal. These data are

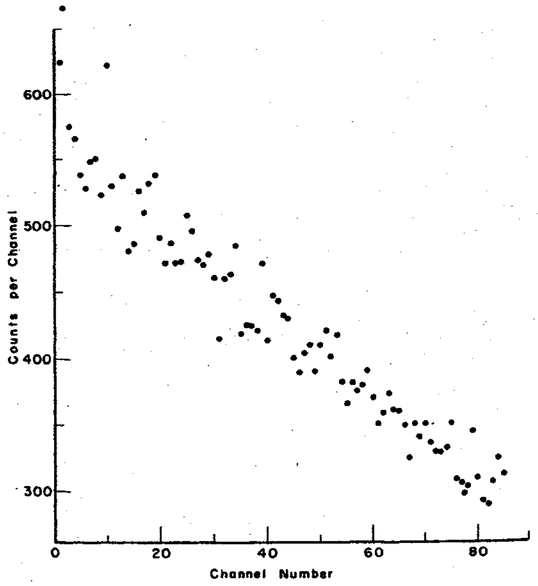

Figure 3. A typical set of positron decay data from the time to pulse height converter, obtained with H = 4.96 G.

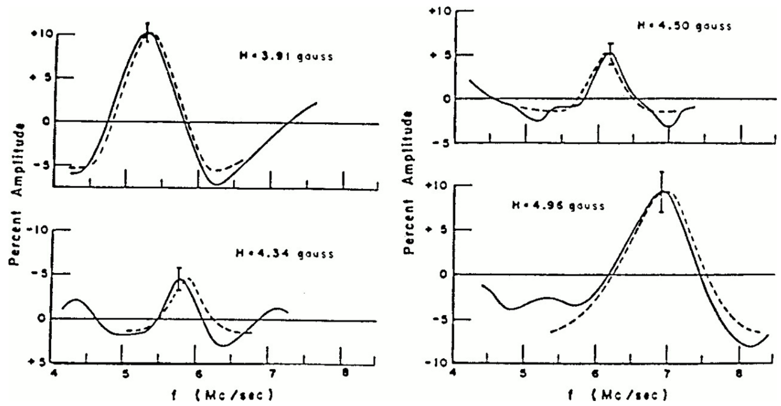

Figure 4. Frequency analysis of the positron decay signal

consistent with close to 100% muonium formation in purified argon.[5 6]

5. First Measurement of $\Delta\nu$

The discovery of muonium made it possible to continue with the original goal of measuring the ground state hyperfine splitting. The decision was made to design a high field microwave experiment. Under the assumption that the muon is a Dirac particle, the lowest order theoretical value for the the hyperfine splitting $\Delta\nu$ is:

$$\Delta\nu = \left(\frac{16}{3}\alpha^2 c R_\infty \frac{\mu_\mu}{\mu_0}\right)\left(1+\frac{m}{M}\right)^{-3}\left(1+\frac{\alpha}{\pi}\right)$$

where α = fine structure constant, c = velocity of light, R_∞ = Rydberg constant for infinite mass, m = electron mass, M = muon mass, μ_0 = Bohr magneton, and μ_μ = muon magneton. Known values of the constants gives $\Delta\nu$ = 4460.0 MHz. The magnetic field dependence of the magnetic substates is given by the Breit-Rabi diagram shown below.

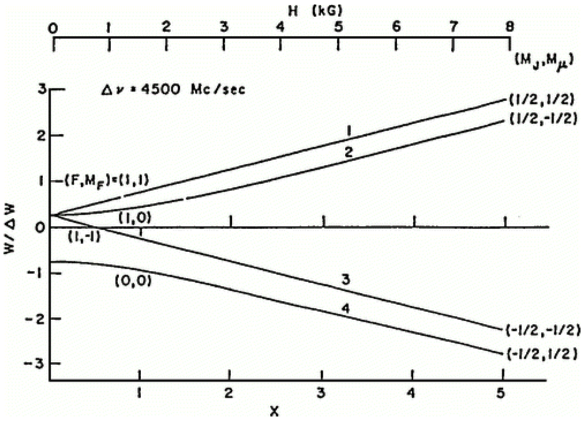

Figure 5. Muonium Breit-Rabi diagram

In the figure $x = (g_J - g_\mu)\mu_0 H/\Delta W$ where $\Delta W/h = \Delta\nu$ and H is the external magnetic field. As a preliminary to the microwave experiment, in order to obtain a rough measurement of $\Delta\nu$ and to confirm the expected behavior, a static field repolarization experiment was planned. If a strong magnetic field along the direction of the original muon polarization is applied, the muon and electron m_μ and m_J are good quantum numbers. In this limit the muon will tend to retain its initial polarization according to:

$$P = \frac{1}{2} + \frac{1}{2}\left(\frac{x^2}{1+x^2}\right).$$

The experimental procedure then is to have forward and backward counters and measure the forward-backward asymmetry as a function of magnetic field.

For this experiment an old cloud chamber magnet, capable of operating to 7 kg, was used in a longitudinal field configuration together with the same stainless steel 50 atm gas target used in the Larmor precession experiment. The experimental arrangement is shown is Fig. 6. The results of the

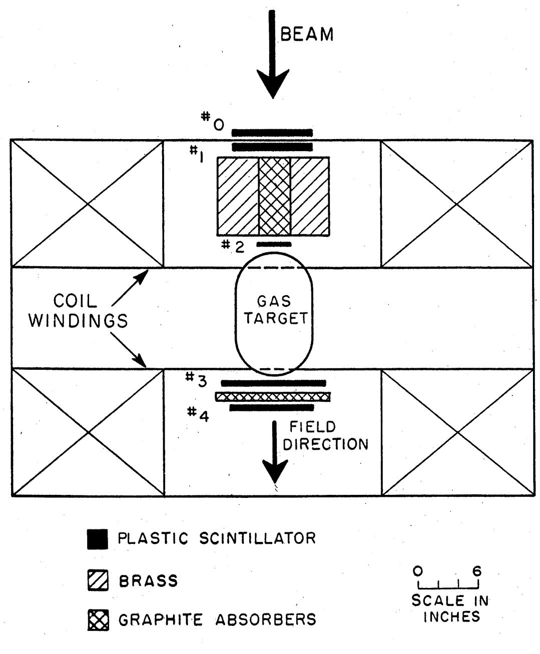

Figure 6. Experimental arrangement

experiment are shown in Fig. 7.[6,7] The quantity R is the ratio of the positron counting rate for a dummy target to the positron counting rate for the gas target with both rates normalized to 1 at H = 0.

The data points are indicated together with their error bars. The solid curve is the theoretical curve which corresponds to the expected theoretical

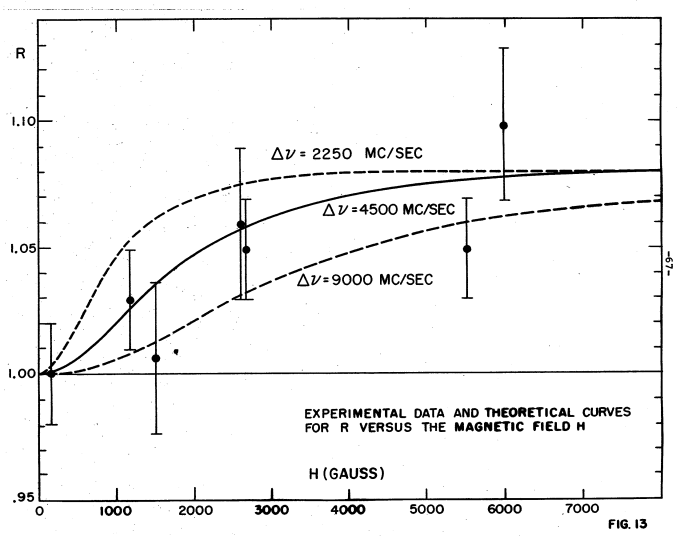

Figure 7. Experimental data and the theoretical curves for the quantity R vs the magnetic field H.

value of $\Delta\nu = 4500$ MHz, whereas the two dashed curves correspond to $\Delta\nu = 2250$ MHz and $\Delta\nu = 9000$ MHz. The theory curves are drawn on the assumption that all the muons stopped in the argon gas form muonium, which is supported by evidence from the earlier experiment. The data are consistent with the assumptions that $\Delta\nu = 4500$ MHz, that all muons form muonium, and that $\Delta\nu$ lies between 2250 MHz and 9000 MHz.

6. The First Microwave Experiment

While the high field repolarization experiment was in progress in 1961, J. Bailey, a Yale Post-Doc, and Yale student W. F. Cleland joined the group. A major effort was underway to design and build the high field microwave experiment. It was now necessary to have a more accurate prediction of the hyperfine splitting, so additional corrections from the literature[8] resulted in an improved calculation:

$$\Delta\nu(theory) = \left(\tfrac{16}{3}\alpha^2 cR_\infty \tfrac{\mu_\mu}{\mu_0}\right)\left(1+\tfrac{m_e}{m_\mu}\right)^{-3}\left(1+\tfrac{3}{2}\alpha^2\right)\left(1+\tfrac{\alpha}{2\pi}-0.328\tfrac{\alpha^2}{\pi^2}\right)$$
$$\times\left(1+\tfrac{\alpha}{2\pi}+0.75\tfrac{\alpha^2}{\pi^2}\right)\left(1-1.81\alpha^2\right)\left(1-\tfrac{3\alpha m_e}{\pi m_\mu}\ln\tfrac{m_\mu}{m_e}\right).$$

Use of the fundamental constants gives:

$$\Delta\nu(theory) = 4463.13 \pm 0.10 \text{ MHz}.$$

The experiment was designed to induce microwave transitions between two hfs magnetic substates of muonium designated by their strong-field quantum numbers $(m_J, m_\mu) = (\frac{1}{2}, \frac{1}{2}) \longleftrightarrow (\frac{1}{2}, -\frac{1}{2})$. The transition is observed under approximate strong field conditions for which the transition frequency is roughly $\Delta\nu/2$. The Breit-Rabi formula allows an exact calculation of $\Delta\nu$ from the observed resonance condition of the microwaves and the static magnetic field.

The microwave source was a Varian 802B 1 kW klystron driving a thin aluminium cavity operating in the TM_{110} mode. The unloaded Q was about 26,000. The aluminium RF cavity was inside a 55 atm stainless steel pressure tank. The trigger counters defined a stopping muon $(12\bar{3})$ and the decay positron was a forward direction decay defined by $(34\bar{2})$. The magnet was the same one used in the depolarization quenching experiment described above. The ratio R was defined as events(microwaves on) divided by events(microwaves off). At resonance the angular distribution of the decay positrons changes and with a resultant increase in R at resonance. In practice, the microwave frequency was fixed and the static magnetic field was varied. For the early runs the hyperfine transition was driven in power broadened modes resulting in linewidths ranging from 44 to 120 gauss. The experimental arrangement is shown in Fig. 8.

One of the the first resonance curves obtained, plotted during the run and taken directly from the log book, is shown is Fig. 9.

Additional resonance curves taken at different combinations of microwave power and gas pressures are shown in Fig. 10.

The result from the combination of all the early microwave runs is:

$$\Delta\nu_{expt} = 4461.3 \pm 2.0 \text{ MHz}.$$

in excellent agreement with the theoretical value to within the experimental error of 1 part in 2200.[9,10] The pressure shift extrapolated from hydrogen values was found to be -0.81 MHz at 50 atm and considered as part of the error.

7. The Final Microwave Experiments at Nevis

In the period from 1962 to 1964 the high field microwave measurements were substantially improved. The group grew in size when Yale Post-Doc M. Eckhause, Yale student R. M. Mobley, and Columbia student J. E. Rothberg joined the group. The measurement precision was greatly increased by (1) obtaining much narrower resonance lines through use of a more homogeneous magnetic field and low microwave power, (2) improving counting

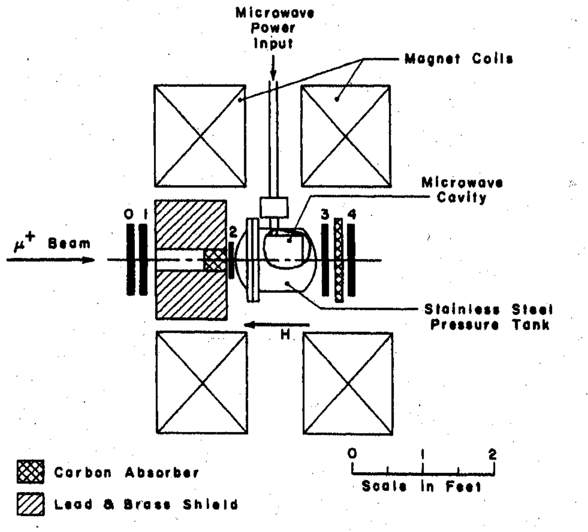

Figure 8. Experimental arrangement. 0, 1, 2, 3, and 4 are scintillation counters.

statistics with longer runs and detecting both forward and backward decay positrons, (3) improving reliability and stability of all the components in the experiment. The argon pressure shift was measured directly showing a linear dependence with the quadratic term consistent with zero. The argon pressure shift data are shown in Fig. 11.

The final Nevis high-field measurement for $\Delta\nu$ gave:[11]

$$\Delta\nu(expt) = 4463.24 \pm 0.12 \text{ MHz (27ppm)}$$

The newest theoretical value at this time was:

$$\Delta\nu(theory) = 4463.326 \pm 0.019 \text{ MHz (4.2ppm)}$$

The excellent agreement provided a confirmation of the theoretical formula for $\Delta\nu$ to the order $\alpha^3 R_\infty$.

After this time, the group activities moved to Los Alamos. The Nevis era had ended. A beautiful series of Los Alamos weak field experiments that the Yale group conducted with new collaborators resulted in the precision today of 12 ppb.

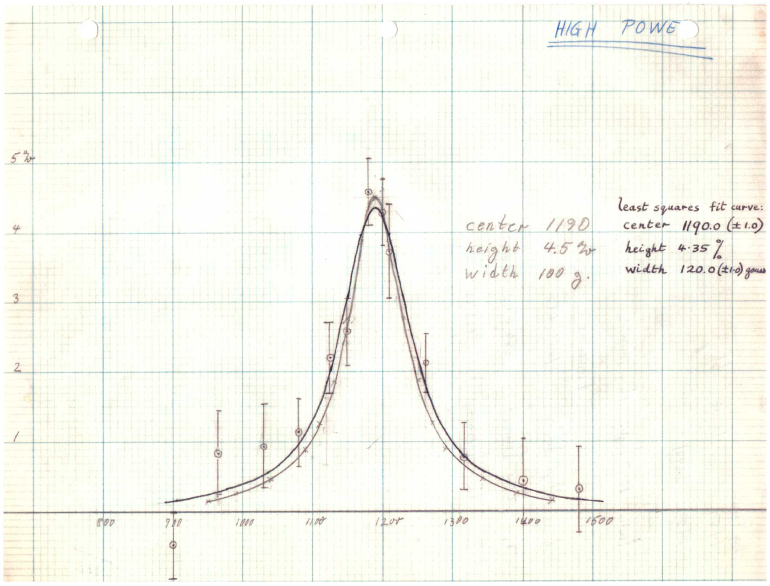

Figure 9. First muonium hyperfine resonance obtained. The microwave frequency is 1850.08 MHz with an Ar pressure of 55 atm. The line center is is at $H_{center} = 5725$ g with a line width of 120 g.

8. Muonium in Semiconductors

During the period 1959-1960, George Feher was a Visiting Associate Professor at Columbia University from Bell Labs. While at Bell Labs Feher originated and developed the Electron Nuclear Double Resonance (ENDOR) technique and was an expert in paramagnetic impurities in solids. At Columbia he became aware of the muonium work and suggested that muonium would form in solids as shallow doner hydrogen-like states and the system so formed would be well suited as a structural probe in less-well-understood materials. Allan Sachs and I offered to do the experiment and Feher supplied a large number of silicon and germanium ingots from Bell Labs. To my knowledge, Feher was the first person to suggest that muonium would be of interest to the semiconductor community.

The apparatus which was used for the muonium depolarization quenching studies was suitable, and we proceeded to build a cryogenic target for the Si and Ge target samples. This work was decoupled from the Yale group, and since I was a Columbia student I was free to work on this experiment. Vernon was not terribly excited about this project but never said

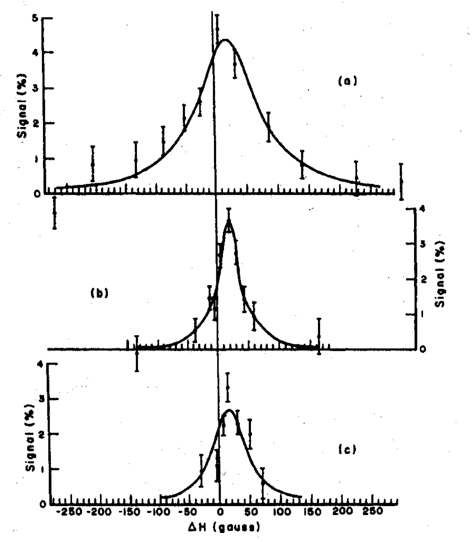

Figure 10. Three resonance curves taken under the following conditions: (a) 50 atm, 800 W; (b) 50 atm, 240 W; (c) 35 atm, 400 W. The zero of the horizontal axis is the expected line center.

anything negative. The samples ranged from p-type Si consisting of five samples with doping ranges between 10^{13} and 10^{19} holes/cm^3 to n-type Si with a comparable doping range. The samples provide a continuous range of electron concentration from about 10^2 to 10^{18} electrons/cm^3. These samples were run at room temperature and in addition there was an n-type Ge sample with 10^{15} electrons/cm^3 run at both room and liquid nitrogen temperature.

The results for these samples in shown in Fig. 12. The results show that with increasing electron concentration there is increasing depolarization consistent with the formation of muonium. After about 10^{14} electrons/cm^3 the polarization starts to rise with increasing electron concentration as the semiconductor shows a behavior similar to positive muons in a metal. The Ge sample at liquid nitrogen temperature showed almost complete depolarization. A preliminary depolarization quenching measurement of sample 11 found that a field of 1000 gauss quenched more than half of the muon depolarization.[15]

Feher left after the first experiment, taking a position at UC, San Diego,

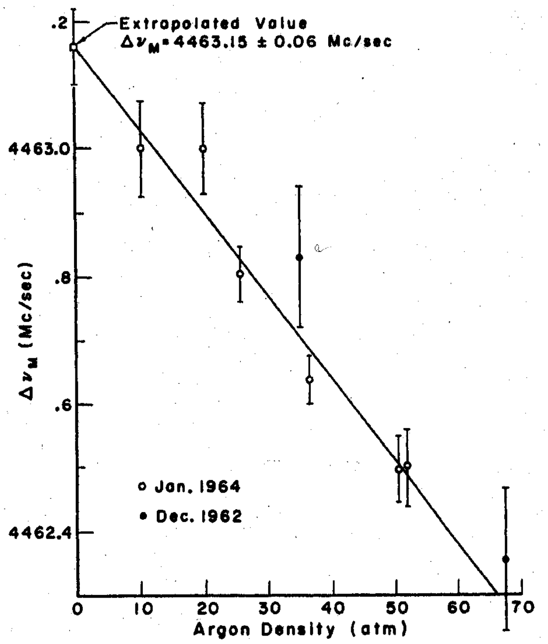

Figure 11. $\Delta\nu$ as a function of argon gas density assuming a perfect gas law. The density is given in units of equivalent pressure at 0° C.

where he is to this day. We continued the program with a cryogenic target capable of going to liquid helium temperatures. The work became the thesis of Bob Eisenstein, a student of Allan Sachs. The targets were expanded from Si and Ge to include LiF, MgO, Red P, and Black P. Depolarization quenching measurements were made on all the sample at room, liquid nitrogen, and liquid helium temperatures. The data were fit to a model which allowed for multiple muonium formation. Some of the low temperature samples quenched the more than half the depolarization with as little as 100 gauss. The results showed that some of the samples at the low temperatures were consistent with a small number of muonium formations.[16] After Eisenstein finished his thesis we had all moved on to other things and the program was never continued at Nevis. Subsequently, muonium in semiconductors became a full-fledged field with considerable experimental and theoretical work.[17]

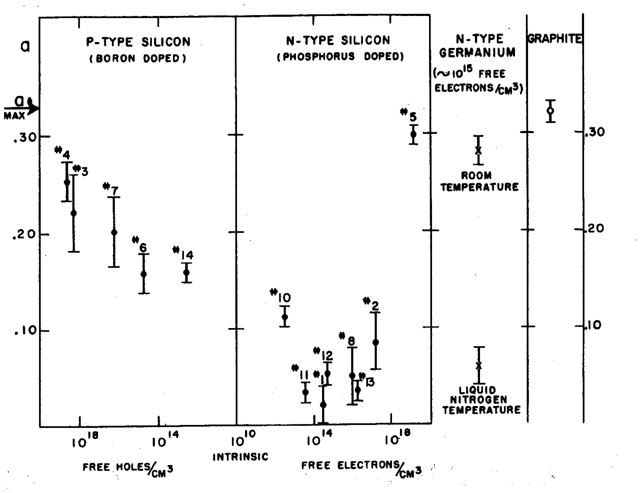

Figure 12. Experimental value of the asymmetry parameter vs. free electron concentration in n-type Si and free hole concentration in in p-type Si at room temperature, and the asymmetry parameter in one Ge sample at room and liquid nitrogen temperature.

9. The naming of muonium

Vernon always claimed that the μ^+e^- atom was first called muonium by Pontecorvo.[12] However, the term used in Pontcorvo's paper is not *muonium* but *mesonium*. The paper in the original russian uses the same term. The student lore was that the term muonium was coined by Rabi, but I do not recall that anyone ever asked him. The first mention of muonium in the literature that I know of is a paper by Friedman and Telegdi, looking for parity non-conservation of muon decay in nuclear emulsion.[13] Both Telegdi and Hughes had positronium experiments to their credit, so it seems quite sensible that it should have been named *muonium*.

However, critics abound. One common complaint was that *muonium* should be reserved for the $\mu^+\mu^-$ bound state akin to positronium and that the μ^+e^- bound state should have been called *muium*. Why did this not happen? The excuses were listed as follows:

- No one could pronounce *muium*.
- The name became popular before conventions were formally estab-

lished, and you can't change all the literature retroactively.

- No one has ever observed $\mu^+\mu^-$, so there is no practical conflict.

Finally everything was set straight by the International Union of Pure and Applied Chemistry (IUPAC) when in 2001 recommendations were made for naming conventions of muonium and muonium compounds. They state "Although chemical reactions of muonium reactions have been studied for two decades, the nomenclature of muoniumm and related species has not been addressed by the IUPAC." They formally endorse the name *muonium* for the μ^+e^- bound state with the symbol "Mu".[14] As for the annoyance of naming $\mu^+\mu^-$, they recommended "muonic muonium". So after 44 years, *muonium* is official.

10. Acknowledgements

The time during which these experiments were done was a very exciting period, following so closely the discovery of parity non-conservation in the weak interactions. We worked very closely together in an era where experiments were done by just a few people. I would like to thank J. Bailey, W. Cleland, D. McColm, and K. Ziock for their friendship and collaboration. Vernon provided the inspiration for all of us. He will be sorely missed.

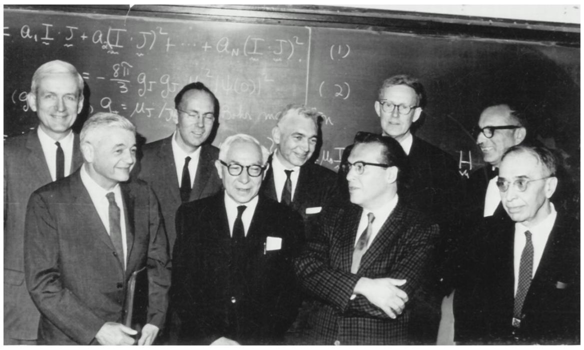

Figure 13. Vernon, at about the time (1967) of the Nevis period. From the left: N. Ramsey, G. Zacharias, C. Townes, I.I. Rabi, VWH, J. Schwinger, E. Purcell, W. Nierenberg, and G. Breit

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