DOE/ER/40314-8

PROGRESS REPORT

ON

RESEARCH IN NUCLEAR PHYSICS

June 1, 1993 - July 31, 1994

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August, 1994

PREPARED FOR THE U. S. DEPARTMENT OF ENERGY
UNDER GRANT NUMBER DE-FG05-87ER40314

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PREFACE

The progress on Grant No. DE-FG05-87ER40314 from June 1, 1993 to July 31, 1994, is summarized in this report. The main emphases of the program are studies of the structure of neutron-rich nuclei and rare electron capture processes.

Most of our activities for the past year were focussed on the rare electron capture component of our program, using experimental facilities at Tennessee Technological University (TTU) and Montana State University (M_TSU). In addition, work of astrophysical significance was carried out at Lawrence Berkeley Laboratories (LBL). We are still waiting for all of the equipment needed for our research on neutron-rich nuclei to be developed at the Argonne Tandem-Linac Accelerator System (ATLAS).

Three undergraduate students, all majoring in physics, are currently working on the project. They are Alex Altgilbers, Brian Faircloth, and Predrag Miočinović. All are making significant progress, both in developing their research skills and in making positive contributions to the project. Danny Bardayan, a former member of our group, recently finished his first year of graduate study at Yale University.

Gloria Julian has continued as the secretary, accountant, and general bureaucratic trouble-shooter for the project. As always, we appreciate her efficient, accurate service.

We would like to thank the many scientists and support personnel at other institutions and laboratories who have assisted us with this work. We are particularly grateful for the hospitality and support we have received during visits to LBL and M_TSU.

Finally, we would like to remind the reader that the unpublished work described in this report is preliminary and should not be quoted in the literature without prior permission.

R. L. Kozub

M. M. Hindi

Search for a 17 keV Neutrino in the Internal Bremsstrahlung Spectrum of ^{125}I

M. M. Hindi, R. L. Kozub, and S. J. Robinson

In last year's progress report[1] we reported preliminary results on the search for 17-keV neutrinos in the internal bremsstrahlung spectrum of ^{125}I. We have since reanalyzed our data using recently obtained[2] relativistic theoretical IB shapes. We set an upper limit of 0.4% for the admixture of a 17 keV neutrino, at the 90% confidence level, and exclude a 0.8% admixture at the 99.6% confidence level. We also find that the relativistic calculations of Surić *et al.* reproduce the shape and relative intensity of IB partial spectra within a few percent. We have published our results in the Physical Review C. A reprint of the paper is included as an Appendix in this report.

[1] R.L. Kozub and M.M. Hindi, *Progress Report on Research in Nuclear Physics, 1992-1993*, (DOE/ER/40314-7), 6 (1993).

[2] T. Surić, R. Horvat, and K. Pisk, Phys. Rev. C47, 47 (1993).

Absolute Intensity of Internal Bremsstrahlung from the EC Decay of ^{125}I

M. M. Hindi, R. L. Kozub, and S. J. Robinson

There are very few accurate measurements of the absolute intensity of the internal bremsstrahlung (IB) spectrum which accompanies electron capture decay. This is especially true for decays with low Q value, where the IB spectrum is dominated by capture from p orbitals. During the course of our experiment to search for 17 keV neutrinos in the IB spectrum of ^{125}I[1] we obtained absolutely calibrated IB spectra; however, because of the large difference in the x-ray count rate from the ^{125}I source ($\sim$100 mCi) and from available calibration sources ($\sim$ a few μCi), we were unable to (easily) calibrate the ^{125}I source strength and hence to give the absolute intensity of the IB spectrum. Several half-lives have now elapsed since the completion of the IB measurement and the source strength has become sufficiently low to allow an absolute calibration.

The yield of Te K x rays and of the 35.5-keV γ ray from the decay of the ^{125}Te^m isomer were measured using a planar Ge detector without any absorbers and with the source at a distance of 16.5 cm from the detector. The absolute efficiency of the detector as a function of energy was obtained from calibrated ^{57}Co, ^{109}Cd, and ^{133}Ba point sources. The resulting activity of the source, at the time of calibration (11-18-1993), was thus found to be 125 ± 8 μCi.

Figure 1 shows the raw ^{125}I IB spectrum used for determining the absolute IB yield. The spectrum was obtained during our experiment to search for 17 keV neutrinos.[1] The spectrum was collected over a period of 12 hours. The source strength at the time of collection (5-15-1992) was 78.4 ± 5.0 mCi. A precision 60.0 Hz pulser was used to correct for dead time and pileup losses. The relative efficiency of the detector as a function of energy was determined with a ^{182}Ta source. The absolute efficiency at 122.034 keV was determined from a calibrated ^{57}Co source. The contributions of pileup, background, and contaminants were subtracted in the same manner as described previously in Ref. 1. Figure 2 shows the absolutely normalized net IB spectrum together with a fit using Surić *et al.*'s calculation[2] (convoluted with the response function of the detector). The theoretical calculation was multiplied by a fitted normalization parameter. The fitted value was 0.857 ± 0.003, where the error is statistical only. We estimate the uncertainty in the absolute efficiency calibration of the IB spectrum to be $\pm$10%, arising mainly from the uncertainty in the position of the ^{125}I source relative to the position of the calibration source. We combine this uncertainty in quadrature with the 6.4% uncertainty in the ^{125}I source strength to obtain 0.86 ± 0.10 for the normalization parameter.

[1] M. M. Hindi, R. L. Kozub, and S. J. Robinson, Phys. Rev. C49, 3289 (1994). See Appendix for reprint.

[2] T. Surić, R. Horvat, and K. Pisk, Phys. Rev. C47, 47 (1993).

The above normalization factor indicates that the Surić calculation essentially reproduces the p IB spectrum for ^{125}I. This is in contrast to the result for ^{193}Pt, where the Surić calculation[2] underestimates the experiment by about a factor of two. Because of the paucity of accurate absolute p IB measurements, it is not clear whether the agreement with our data is fortuitous, or whether there is a systematic divergence between theory and experiment as a function of the atomic number. Clearly more systematics are needed before a firm conclusion can be reached.

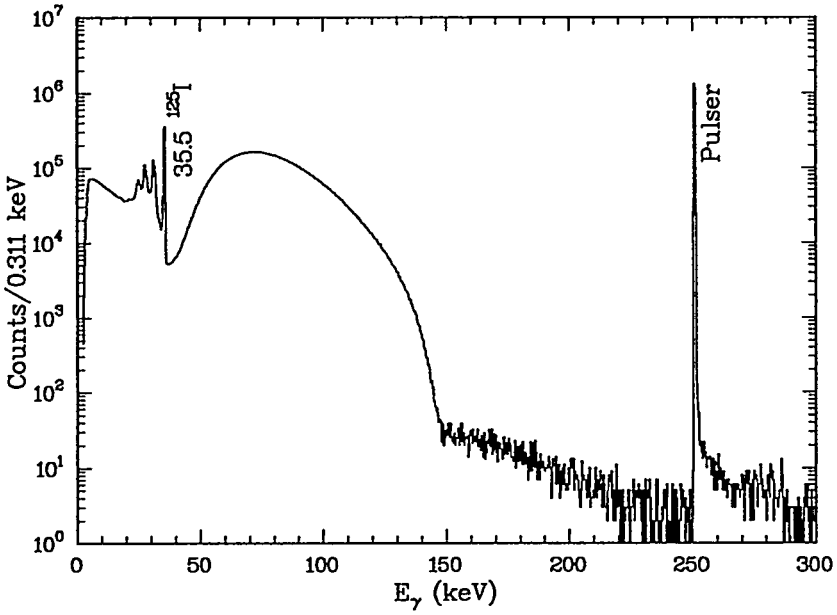

Figure 1: A 12 hour raw spectrum of the ^{125}I source recorded in a planar Ge detector.

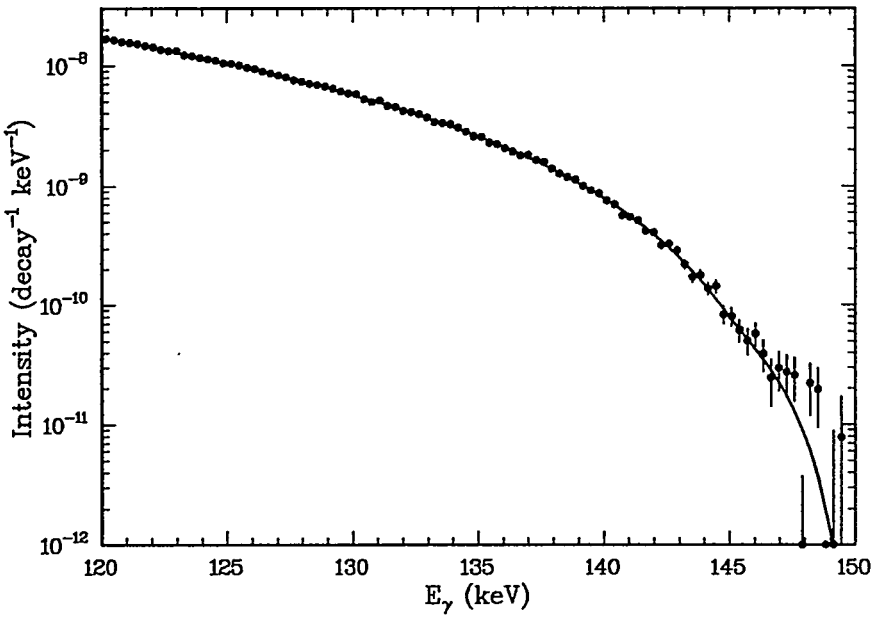

Figure 2: Net IB spectrum of ^{125}I above the $1s$ endpoint. The solid line is a normalized fit using the Surić *et al.* calculation. The (multiplicative) normalization factor for the theoretical calculation is 0.86 ± 0.10.

Internal Bremsstrahlung Endpoint of ^{109}Cd

M. M. Hindi and R. L. Kozub

There is a discrepancy in the measured Q_{EC} value of ^{109}Cd; the value obtained from the internal bremsstrahlung (IB) endpoint [1] ($Q_{EC} = 182 \pm 2$ keV) is in sharp disagreement with the value deduced from the L to K capture ratio[2] ($Q_{EC} = 201 \pm 3$ keV), which in turn is in disagreement with the value extracted from the $L + M + \cdots$ to K capture ratio[2] ($Q_{EC} = 220 \pm 3$ keV). We have attempted (but so far failed) to resolve this discrepancy by remeasuring the IB endpoint.

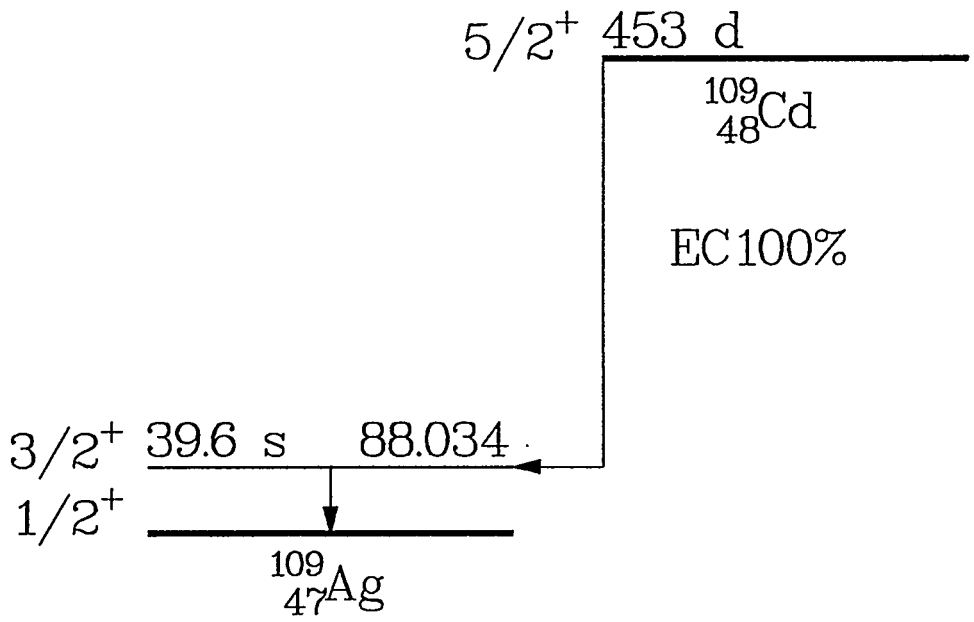

Figure 1: Decay scheme of ^{109}Cd. Level energy is in keV.

Figure 1 shows the decay scheme of ^{109}Cd. The decay proceeds exclusively to the ^{109}Ag isomer at 88 keV. The difficulty in the measurement is that the IB endpoint would be only 6 keV (for $Q_{EC} = 182$ keV, 38 keV for $Q_{EC} = 220$ keV) above the 88-keV γ-ray line which would dominate the spectrum. Singles spectra from a ≈ 5 μCi ^{109}Cd standard calibration source were recorded using a planar Ge detector (FWHM = 510 eV at 88 keV) and an amplifier equipped with pileup rejection circuitry. Copper absorbers in front of the detector attenuated the intense ^{109}Ag x rays (which follow the EC decay and the internal conversion of the 88-keV state) to negligible levels. Spectra were collected at six

[1] K. P. Gopinathan and W. Rubinson, B.A.P.S. **13**, 1452 (1968).
[2] W. Goedbloed *et al.*, Nucl. Instrum. Methods **88**, 197 (1970).

different count rates, ranging from 92 cps to 337 cps. The variation in count rate was achieved by varying the distance between the source and the detector from $\approx$ 1 cm to a few cm. The spectrum was collected for a 10-day period at each position. Background spectra were collected before and after the measurement for a total period of 22.5 days.

The pileup rejection circuit of the amplifier rejects consecutive pulses as long as the separation between them is $\geq$ 350 ns. We attempted to resolve the contribution of IB above the 88-keV γ-ray peak from the residual pileup by performing a linear fit to the intensity (at each γ-ray energy) as a function of count rate and extrapolating to zero count rate. This method requires that the response of the detector (i.e., the spectral shape) remain the same at the different count rates. As figure 2 shows, changing the position of the source also changed the ratio of Compton-scattered events to full energy events. Since the pileup in the IB region results mainly from the summing of a full energy event with a Compton event, changing that ratio changes the amount of pileup regardless of the change in the count rate. Therefore it was not possible to extract the correct pileup by the above method.

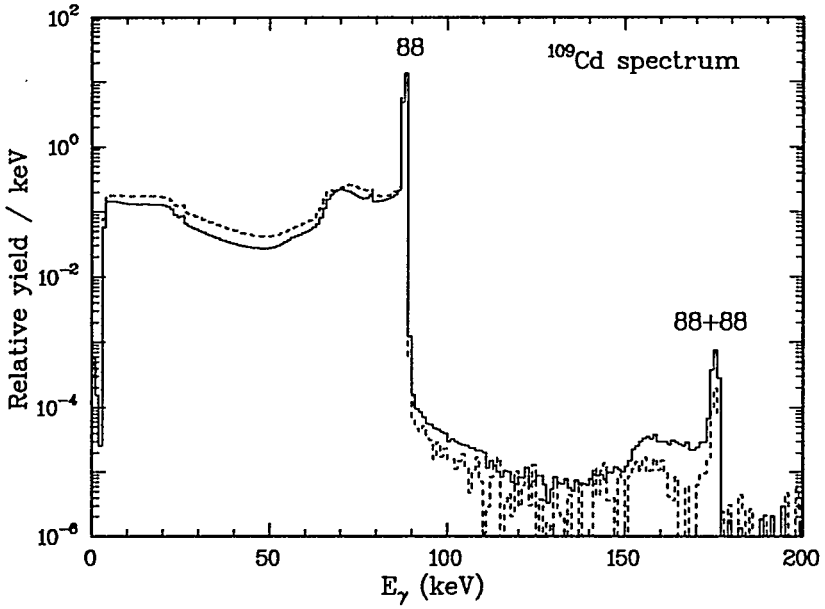

Figure 2: ^{109}Cd spectra collected at different source positions, normalized to the 88-keV full energy yield. The solid line is with the source at 1 cm from the detector (count rate=337 cps); the dashed line is with the source at a few cm (count rate=92 cps).

We also attempted to extract the pileup by calculating the expected pileup spectrum using a Monte Carlo simulation. The pulse shape from the amplifier was digitized. Two pulses with heights randomly selected from the measured spectral distribution and separated by a random time interval between 0 and 350 ns were added together and the

resulting peak found numerically. Figure 3(a) shows the resulting Monte Carlo-generated pileup spectrum together with the background-subtracted measured spectrum. The pileup spectrum was normalized to the measured spectrum in the interval 142–172 keV. Although the Monte Carlo pileup spectrum agrees very well with the measured spectrum in the fitted interval, we were not sure that the rejection efficiency (or the resolving time) of the amplifier's pileup rejection circuitry is completely independent of the sizes of the summed pulses. Such dependence, if it did exist, would probably be for large pulses followed by small pulses, which is precisely the origin of pileup in the IB energy region. To test whether the calculated spectrum accounts fully for pileup in the energy range 93–143 keV (i.e., 4–55 keV above the 88-keV energy) we also compared the Monte Carlo spectrum to the measured spectrum in the energy range 348–427 keV – a region where only pileup with the pulser peak (at 343 keV) is expected. As figure 3(b) shows, the Monte Carlo spectrum agrees reasonably well with the measured spectrum in the region 398–427 keV (i.e., for energies from 55–84 keV above the pulser energy), but grossly underestimates the pileup in the region 347–397 keV (i.e., for energies from 4–54 keV above the pulser energy). From this we conclude that, indeed, the rejection efficiency decreases for small pulses following large pulses and, hence, that an IB spectrum obtained by subtracting the Monte-Carlo-generated pileup spectrum would still contain residual pileup. This is confirmed by Table I, where the results of fits to such extracted spectra are displayed for each of the count rates. It is seen that the normalization factor (the ratio of the measured spectrum to the theoretical spectrum of Surić *et al.*[3]) increases with count rate and is much larger than the expected normalization factor of 0.86 (the normalization factor we obtained for the ^{125}I IB spectrum, presented elsewhere in this report).

Table 1: Fit to IB extracted from spectra at different count rates.

Count rate (cps)	Normalization	Q_{EC} (keV)
92.02	1.3 ± 1.2	211.6 ± 9.5
100.76	2.0 ± 1.5	206.0 ± 6.0
153.07	2.5 ± 1.5	206.6 ± 4.9
225.99	2.8 ± 1.4	204.6 ± 3.8
286.49	2.9 ± 0.7	205.3 ± 1.7
336.95	4.3 ± 0.8	201.8 ± 1.3

We are currently examining whether it is possible to extract the pileup by making

[3]T. Surić, R. Horvat, and K. Pisk, Phys. Rev. C47, 47 (1993).

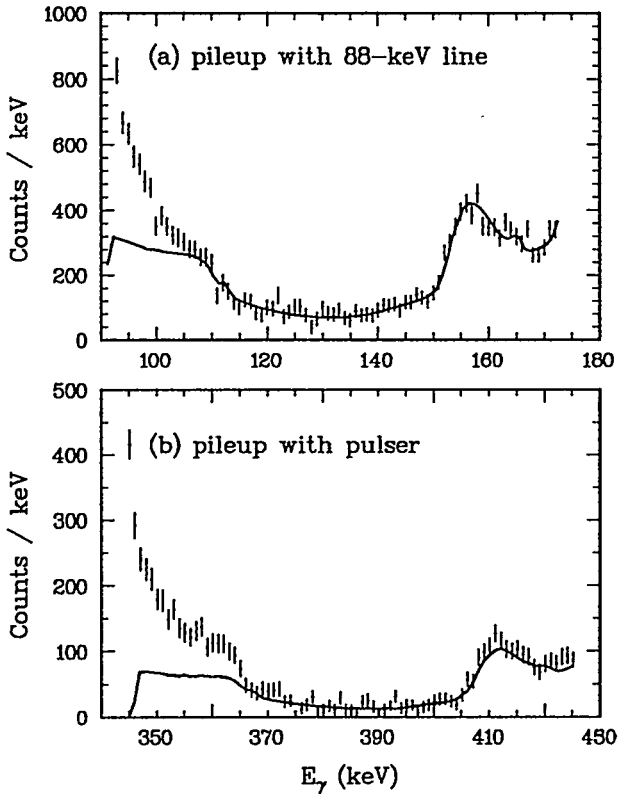

Figure 3: (a) The raw ^{109}Cd spectrum (data points) and Monte Carlo simulated pileup spectrum (solid line). The simulated spectrum is normalized to the data by a fit in the energy region 142–172 keV. (b) Same as (a), for the energy region above the pulser peak.

use of both the Monte Carlo-generated spectrum and the rate dependence of the pileup. We are also investigating the possibility of online radiochemical separation of the 40-s ^{109}Ag^m from the ^{109}Cd parent. If this were feasible, the 88-keV γ ray contribution to the spectrum would be diminished substantially and the problem with pileup would be much less severe. Such an experiment would be conducted at Lawrence Berkeley Laboratory, in collaboration with Dr. Eric Norman.

Search for Massive Neutrinos in the Recoil Spectrum of ^{37}Cl following Electron Capture Decay of ^{37}Ar

M. M. Hindi, R. L. Kozub, S. J. Robinson, A. Altgilbers,[†] B. Faircloth,[†] R. Avci,[‡] Lin Zhu,[‡] and G. J. Lapeyre[‡]

We are developing an experiment to measure the spectrum of recoil velocities of ^{37}Cl ions following the electron capture (EC) decay of a monolayer of ^{37}Ar adsorbed on a cold surface. One of the aims of this experiment is to search for massive neutrinos which might be admixed with the electron neutrino in the decay. The mass and mixing fraction of such heavy neutrinos would be reflected as an extra peak in the otherwise monoenergetic recoil spectrum; the energy shift would be related to the mass of the heavy neutrino by $\Delta E/E = (m_\nu/Q)^2$, where $Q = 814keV$ is the EC Q vlaue, and the area of the extra peak would be related to the mixing fraction. In last year's Progress Report[1] we described the experimental arrangement and gave indications that we are observing, in singles, the ^{37}Cl recoils and Auger electrons following the EC decay. Since that time we have measured the spectrum of recoil velocities of the ^{37}Cl's via a time-of-flight measurement in coincidence with the Auger electrons, and measured the fraction of desorbing ions and their charge distribution. Our findings, which are summarized below, have been accepted for publication.[2]

We find that the ionic desorbing fraction is 10%. The charge distribution of the recoiling ions differs substantially from that expected for isolated ions. The energy disribution of the recoiling ions in coincidence with Auger electrons has the expected central energy of 9 eV, but has a FWHM of $\sim$ 30%, which is larger than that expected in coincidence with LMM Augers (20%) and with KLL Augers (14%). We have tentatively attributed these findings to inelastic charge exchange of the multiply-ionized recoiling Cl atom with its ^{36}Ar neighbours and possibly with the Au substrate. Since we do not yet have a full *quantitative* understanding of the factors contributing to the energy broadening, and since we are currently working on several improvements and modifications (described below) which we hope will reduce the broadening, we refrain from quoting limits on neutrino mixing from these preliminary results.

The improvements we are currently working on are the following:

1. We will produce the ^{37}Ar via the ^{40}Ca$(n,\alpha)^{37}$Ar reaction. This will allow much higher activities at smaller coverages. In our previous experiments the ^{37}Ar was

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[1]R. L. Kozub and M. M. Hindi, *Progress Report on Research in Nuclear Physics, 1992-1993*, (DOE/ER/40314-7), 6 (1993).

[2]L. Zhu, *et al.*, J. Vac. Sci. Technol. A **12**, 2037 (1994); see reprint in the Appendix.

produced from the $^{36}\mathrm{Ar}(n,\gamma)^{37}\mathrm{Ar}$ reaction. Since we could not separate the ^{36}Ar from the ^{37}Ar, one monolayer had only about 0.01% radioactive ^{37}Ar atoms, with the rest being ^{36}Ar. If we produce the ^{37}Ar via the $^{40}\mathrm{Ca}(n,\alpha)^{37}\mathrm{Ar}$ reaction, we will be able to adsorb only ^{37}Ar on the surface and we should be able to produce much higher activites *and* use a smaller area.

2. We will use a position sensitive microchannel plate (PSMCP) to detect the ions. This should allow us to bring the detector closer to the source, thus increasing the solid angle and shortening the flight path while maintaining knowledge of the flight distance.

3. We will be conducting the time of flight measurement in coincidence with x rays, instead of with Auger electrons. There are several advantages to detecting the recoils in coincidence with K x rays. Detecting a K x ray will insure that the resulting Cl ion will end up with charge $+1e$; in that case even if charge transfer does happen, the Cl atom will become neutral and hence would not be detected by our setup. To see why detecting a K x ray (more specifically, a $K\alpha$ x ray) selects atoms that end up with charge 1, recall that the EC capture process leaves the Cl atom *neutral*, but with a hole in the K shell. The K hole is normally (90.3% of the time) filled with a $KL_{2,3}L_{2,3}$ Auger transition which leaves the Cl with a charge $+1e$ and with two holes in the $L_{2,3}$ shells. Each of these L holes in turn decays by an LMM transition, which ultimately leaves the Cl ion triply charged. When the initial K hole is filled by a radiative transition, however (the remaining 9.7% of the time), the hole is transferred to the $L_{2,3}$ shells and the atom *remains neutral* and now has only *one hole* in the L shell. This L hole again decays by an LMM transition leaving the Cl ion with a charge of $+1e$. Another advantage of performing the coincidence with x rays is that the recoil velocity given the atom when the x ray is emitted (0.32% of the neutrino-induced recoil velocity) is much smaller than that given the atom when a KLL Auger is emitted (6.4% of the neutrino-induced recoil velocity); this would allow the use of large area x ray detectors without having to have position sensitivity, which would compensate somewhat for the smaller x ray emission probability. Yet another advantage of detecting x rays is that it would leave only one LMM Auger undetected, where as coincidences with a KLL Auger would leave *two* LMM Augers undetected; each LMM Auger imparts a recoil that is 1.8% of the neutrino-induced recoil.

   We have not yet settled on a suitable x ray detector. An ultrahigh vaccuum-compatible detector with a reasonable efficiency and good energy and timing resolutions is needed.

4. We are using a graphite substrate to adsorb the Ar on. Because of the saturated surface bonds and the lower surface electron density, the charge transfer probability to the Cl ion should hopefully be lower than it is for the gold backing which we have

used so far. In addition, because of its lower Z, the backscattered fraction of Auger electrons should be much lower for graphite than it is for gold.

The above improvements should enable us to set limits of $\leq 0.1\%$ on the admixture of heavy neutrinos with the electron neutrino. If there were no further reduction in the broadening of the Cl velocity distibution (from what we have already measured), our measurements would be sensitive to heavy neutrino masses in the range 600-700 keV. Any reduction in the width of the velocity distribution would then allow us to probe lower masses. The mixing limits which we hope to achieve should be a factor of $\approx 5 - 10$ lower than the current limits on the admixture of the electron neutrino with a tau neutrino in that mass range.[3]

[3]See, for example, F. Boehm and P. Vogel, *Physics of Massive Neutrinos* (Cambridge University Press, London, 1992).

The Auger Relaxation of ^{37}Cl following Electron Capture Decay of ^{37}Ar

Lin Zhu,[†] R. Avci,[†] G. J. Lapeyre[†] M. M. Hindi, R. L. Kozub and S. J. Robinson

The experimental system which we have developed to measure the spectrum of recoil velocities of ^{37}Cl ions following the electron capture (EC) decay of ^{37}Ar also allows the study of the Auger relaxation process following the EC decay. The EC decay creates a unique initial state where 90% of the time a single hole is created in the K shell while the rest of the shells are essentially left intact. The detailed processes associated with the decay of such a deep hole are hard to study using the conventional techniques of Auger and photo electron spectroscopy with normal (non-radioactive) atoms. For example, when Ar is bombarded with 5 keV electrons the fraction of K holes created is over two orders of magnitudes less than that of L holes. It might be possible to improve the ratio with a photon beam, but multiple ionization associated with photo-absorption might obscure the process of multiple ionization associated with the subsequent decay of the K hole. Thus the EC process allows direct observation of some very interesting relaxation processes which are amenable to theoretical calculations, but which would be difficult to get at experimentally in a clean way using conventional techniques. For example, two phenomena which have been predicted theoretically, but not observed *directly* (to the best of our knowledge) are (1) the decay of a K hole by double Auger (DA) emission and (2) the shift in energy of an LMM Auger line from a multiply ionized L shell. We report here, for the first time, the direct observation of these two phenomena as an example of our technique.

Some details of our experimental setup are described in Ref. [1]. [1] Briefly, a mixture of one part ^{36}Ar and $\sim 5\times10^{-5}$ parts ^{37}Ar is physisorbed on a gold plated Si wafer kept at 16 K under ultrahigh vaccuum conditions ($\sim10^{-10}$ torr). Two microchannel plates face one side of the sample and a cylindrical mirror analyzer (CMA) faces the other side. The two microchannel plates (MCP) allow us to record time-coincidences between the desorbed ^{37}Cl ion and one of the Auger electrons emitted during the relaxation process, or electron-electron (e-e) coincidences associated with the K-hole decay, with sub-nanosecond time resolution. Each of the MCP's has a retarding screen in front of it which allows the vetoing of electrons (or ions) below a preselected energy. Thus the system allows a measurement of the time of flight of the desorbed ^{37}Cl or the energy distribution of correlated electrons in an Auger cascade.

In the double Auger decay of a K hole the energy released in the filling of the hole is shared by two electrons. To study this process we record coincidences between two electrons both of which have energy above the LMM energy ($\approx$ 180 eV). Fig. 1 shows the measured energy dependence of the probability distribution for one of the two coincident

[†]Physics Department, Montana State University.

[1] L. Zhu, *et al.*, J. Vac. Sci. Technol. A **12**, 2037 (1994); see reprint in the Appendix.

electrons. The kinetic energy of the other electron was kept above 1300 eV by placing a retarding voltage of 1300 V to the screen at the entrance of the MCP. The inset shows the theoretical prediction of the energy dependence for a Neon atom. [2] Notice that there is a qualitative agreement between our observations and the theoretical predictions (use the part of the curve in the inset between 5 to 25 Ry for qualitative comparison). This is the first time (to the best of our knowledge) such a measurement has been performed.

Because the DA process depends strongly on the energy partitioning between the two electrons and on their relative angle it is difficult to obtain the probability of the process relative to the normal Auger process from a measurement conducted over a limited energy range and at one angle. The preferred energy configuration of the double Auger emission is for one of the electrons to take most of the energy with the second receiving the remaining (small) balance. The angular correlations between the two electrons is predicted[2] to be forward peaked for the preferred energy configuration and backward peaked when the energies are close to each other. In our setup the MCP detectors were 45° from each other. Our preliminary measurements indicate that in this geometry the double Auger cross section is less than 5% of the normal Auger process. We are in the process of constructing and testing a position sensitive detector to study these effects.

Fig. 2 shows three superimposed Auger spectra in the LLM and LMM region of Cl and Ar. The full line is the Auger spectrum associated with the ^{37}Ar EC decay. The spectrum is taken with the double pass CMA. The peaks labeled LLM and LMM are those associated with the known Cl Auger emission lines as the spectrum taken from CsCl salt, also shown in the figure, indicates. The CsCl spectrum is taken by a Phi-595 scanning Auger system and is background-subtracted and normalized to fit the scale of the ^{37}Cl Auger spectrum. We also show, for comparison, the LMM Auger peak of natural Ar gas physisorbed on our substrate and bombarded by a 1000-eV electron beam. It is worth noting that the ^{37}Cl spectrum is taken with only 5×10^{-5} monolayer, while the natural Ar spectrum is taken with at least one monolayer coverage.

We tentatively identify the ^{37}Cl Auger peak labeled LMM^H as a *hypersatellite* This is the first time, to the best of our knowledge, a peak with such a large shift (about 22 eV higher energy than the normal LMM line) is reported. Notice that LMM^H peak is not visible in the normal Cl Auger spectra. A high resolution study [3] on Ar gas does not report such peak.

Our hypothesis at this time is that the peak results from an LMM transition following the DA process. The double Auger transition takes place by filling the K hole with an electron from the L shell and the resultant energy (~2.6 keV) is shared by *two* ejected electrons. When the two electrons are ejected from the L shell, *three* holes are produced in the L shell, in contrast to the normal KLL Auger transition, which leaves only two

[2]M.Ya. Amusia, I.S. Lee and V.A. Kilin, Phys. Rev. A 45, 4576 (1992).
[3]L.O. Werme, T. Bergmark, and K. Siegbahn, Physica Scripta 8, 149 (1973).

holes in the L shell. The extra hole in the L shell alters (increases) the separation between the L and M levels causing a ~22 eV increase in the kinetic energy of the subsequent LMM emission. We have confirmed that the LMM^H peak is associated with the EC by recording coincidences between it and electrons from the DA transition. This hypothesis is also bolstered by the observation that the area of the LMM^H peak in the singles (CMA) spectrum is about 14% of the normal LMM peak, while the fraction in coincidence with the DA electrons (energy > 400 eV) is 25% of the normal LMM electrons in coincidence with the KLL electrons. Because, according to our hypothesis, the LMM^H electron follows a DA decay, it is approximately twice as likely to be in coincidence with an electron (with energy > 400 eV) as a normal LMM electron is. This hypothesis also suggests that the DA process (integrated over all electron energies and angles) could be as high as 14% of the normal Auger process.

The reason the LMM^H peak would not have been observed in LMM Auger spectra produced by electron bombardment (such as the work in Ref. [3], for example, or our CsCl spectrum) is that a 5 keV electron beam ionizes the L shell much more efficiently than the K shell, as indicated by the ratio of the sensitivity factors S_L:S_K=~120:1 for the LMM and KLL Auger emission from Cl, respectively. Thus, if our hypothesis is correct, the LMM^H peak produced in such experiments would be about 120/0.14=860 times smaller than the normal LMM peak and would be effectively buried in the background.

In conclusion, we presented some very interesting measurements probing some fundamental questions regarding the relaxation process of a K hole created as a result of the EC decay process. We report the first observation of a hypersatellite LMM peak which we interpret as arising from the extra hole created in the L shell as a result of double Auger emission. We are in the process of building position sensitive microchannel plates which will improve our energy and position resolution. We are also trying to increase the density of the active Ar atoms on the surface.

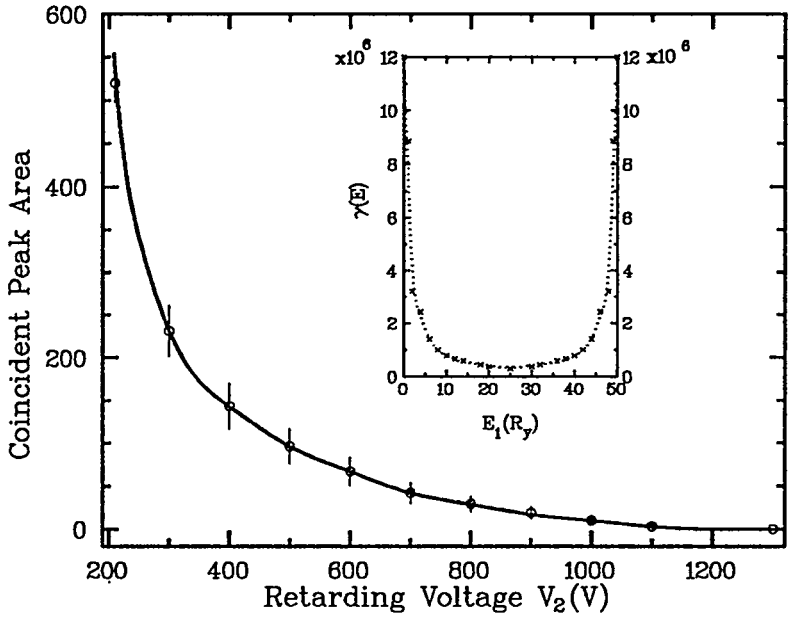

Figure 1: Energy dependence of the double Auger (DA) emission. The inset shows the theoretical prediction by Ref [2] of the energy dependence of the DA emission for Ne. Qualitative comparison should be made between 5 and 25 Rydberg.

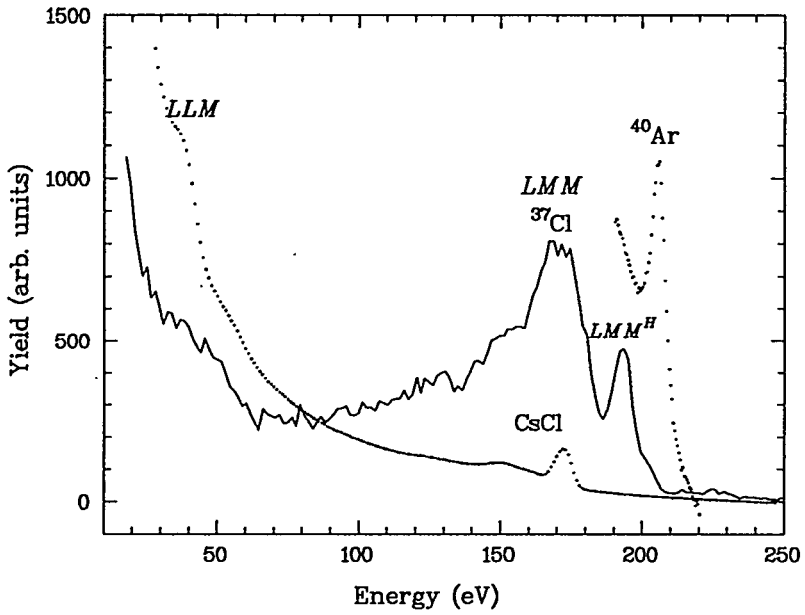

Figure 2: Comparison of various Auger spectra. The full line corresponds to the ^{37}Cl following the EC decay. Note that above 210 eV kinetic energy the background is nearly zero. The dotted lines correspond to the regular Auger spectrum of CsCl and ^{40}Ar exited by electron bombardment. The very high background of the latter cases have been subtracted for comparison. The extra peak labeled as LMM^H is referred to as a hypersatellite LMM peak and is explained in the text.

Neutralization of Ions After Neutrino-Induced Desorption

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As described elsewhere,[1] we are developing an experiment to search for evidence of massive neutrinos in the electron capture (EC) decay of ^{37}Ar. In principle, neutrinos having masses as large as the EC Q-value of 814 keV could be emitted in this decay. During our first series of experimental runs, a monolayer of ^{36,37}Ar was frozen on a gold surface. The spectrum of recoil velocities of positively charged ^{37}Cl ions, desorbed via neutrino emission in the EC process, was deduced from time-of-flight measurements. (The Cl atoms become charged almost immediately after the EC by emitting Auger electrons as the orbital electron vacancies are filled.) The distribution of recoil velocities would be affected if there were some mixing of the electron neutrino with a massive neutrino of another flavor. For example, if mixing should occur with a neutrino having a mass at the current upper limit for the muon neutrino ($<$250 keV), the ion kinetic energies would be reduced by about 10% for those events. In order to search for neutrinos over the widest possible mass range, it is important to minimize the spread in velocities due to other effects. Secondly, in order to set a meaningful limit on the neutrino mixing fraction, it is necessary to know how the fraction of recoils detected would change with recoil velocity. The primary purpose of this paper is to address this second issue.

The peak in our time-of-flight spectrum was broadened more than expected, an effect which we have attributed primarily to charge exchange between multiply charged ^{37}Cl ions and neighboring Ar atoms. This produces two charged atoms very near one another and subjects both to acceleration. On the other hand, a ^{37}Cl ion which becomes only *singly* charged through Auger emission would either retain this charge and possibly be detected in our microchannel plate detector (MCP), or gain electrons and not be detected. (The MCP was screened to allow only positive ions to be detected.) Selecting these singly charged events would thus remove the Coulomb-accelerated events from the data and thereby improve the overall resolution significantly. Singly charged ions occur in about 10% of the decays, when the initial hole in the K shell is filled by ($\approx$2.7 keV) x-ray emission and a single Auger electron is emitted upon the filling of the subsequent hole in the L shell. Thus, the observation of such events can be enhanced by requiring an x-ray-ion coincidence, and that is one of the major improvements we hope to make in the experiment in the near future (as described elsewhere in this report).

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[1]L. Zhu, et al., Jour. Vac. Sci. Tech. (in press, 1994); see preprint in the Appendix and other articles in this report.

The ^{37}Cl ions may also gain electrons from the surface upon which the Ar is frozen, and even singly charged ions may become neutralized in this way. (In our next experimental attempt, we plan to use a graphite surface instead of gold.) In general, both resonant and direct Auger mechanisms may contribute to the neutralization. However, the resonant process is essentially forbidden for Cl ions near a graphite surface, since the workfunction of the surface (5 eV) is greater than the ionization energy of the lowest excited state of the neutralized ion ($\approx$4 eV). The direct Auger process, on the other hand, can occur if the ion has a vacancy at an energy which is lower than the Fermi energy of the surface. This process involves the ejection of an electron from the surface at the same time that the ion is neutralized. Hagstrum[2] has deduced that essentially all neutralization occurs by this mechanism for energies <10 eV. In our case, a desorbed Cl ion has a kinetic energy of 9.54 eV for a zero-mass neutrino. For these reasons, our discussion here will be confined to the direct Auger neutralization process. The extent to which neutralization occurs, and how it depends on ion velocity, will directly affect both the fraction of recoils detected in our experiment (i. e., the counting rate corresponding to zero-mass neutrino emissions) and our sensitivity to neutrino mixing.

Theoretical and experimental work has been done to find the survival rates of singly charged ions which approach a metal surface.[2,3,4] In such cases, the metal is modelled as "jellium," i. e., as an electron gas embedded in a constant positive background. Apparently, very little work has been concerned with the survival rates of ions near semiconducting or graphite surfaces. For this reason, we will treat our graphite substrate as a jellium-like surface. Because graphite has saturated surface bonds and a lower surface electron density than metals, this should result in the worst-case scenario for ion neutralization in our experiment.

According to Hagstrum[2] and Snowdon et al.,[4] a good approximation of the probability density for electron transfer from the metal surface to an ion on an incident trajectory is given by the phenomenological relation,

$$P_t(S,|v|) = \left(\frac{A}{|v|}\right) \exp\left[-\left(\frac{A}{a|v|}\right) e^{-aS} - aS\right] \theta(S-S_0)\,, \qquad (1)$$

where S is the distance of the ion from the surface, S_0, A, and a are constants determined by fitting experimental data, v is the velocity component of the ion perpendicular to the

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[2]H.D. Hagstrum, Phys. Rev. **96**, 325 (1954); *ibid.*, p. 336.

[3]R. Hentschke, K.J. Snowdon, P. Hertel and W. Heiland, Surf. Sci. **173**, 565 (1986).

[4]K.J. Snowdon, R. Hentschke, A. Närmann and W. Heiland, Surf. Sci. **173**, 581 (1986) and references therein.

surface and $\theta(x)$ is the unit step function. Using Cobas and Lamb[5] as a guide, we derive a similar expression for ions on an outgoing trajectory:

$$P_t(S,|v|) = \left(\frac{A}{|v|}\right) \exp\left[\frac{A}{a|v|}\left(e^{-aS}-e^{-aS_\tau}\right) - aS\right] \theta(S-S_0) \qquad (2)$$

where S_τ is the mean distance at which the atoms become charged. This is given simply by

$$S_\tau(v) = v\tau + R \qquad (3)$$

where τ is the mean lifetime of the excited ^{37}Cl atom and R is the radius of the ^{37}Ar atom prior to decay. The velocity of a 9.54-eV ^{37}Cl ion is 0.071 Å/fs, and the mean lifetime is taken to be 9 fs, the sum of the meanlives for filling the holes in the K shell ($\approx$1 fs) and the L shell ($\approx$8 fs) sequentially. Taking the radius of an Ar atom on the surface to be 1.86 Å,[6] one obtains $S_\tau \approx 2.5$ Å. This is much greater than typical values obtained for S_0,[4] so it is appropriate to set $S_0 = S_\tau$ in Equation 2.

Experimental data for the neutralization of Cl^+ ions by a graphite substrate do not exist, so we will refer to Hagstrum[2] for values of A and a. He finds that a good approximation is $a = 2\lambda$, where

$$\lambda = \sqrt{\frac{2m_e\eta}{\hbar^2}} . \qquad (4)$$

As described by Hagstrum,[2] η depends on the atomic and metal wave functions. For our application, a reasonable choice is $\eta = \varphi$, the work function of graphite (5.0 eV), which yields a value for a of 2.29 Å^{-1}. Hagstrum found the values of A for the singly charged noble gas ions by fitting experimental data on Auger electron emissions from the surface. He gave the value of A=210 fs^{-1} for 40-eV Xe^+ ions, which have a velocity and ionization energy similar to that of a 10 eV Cl^+ ion. Plots of P_t *vs* S for these values are shown in Figure 1 for "fast" (maximum speed or 9.54 eV) and "slow" (one-half maximum speed or 0.25x9.54 eV) $^{37}Cl^+$ ions, which would correspond to neutrino masses of zero and 705 keV, respectively.

The probability of Cl^+ ions being neutralized at a distance S from the surface is found by integrating Equation 2:

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[5]A. Cobas and W. E. Lamb, Jr., Phys. Rev. **65**, 327 (1944).

[6]H.H. Claassen, *The Noble Gases* (D.C.Heath & Co., 1966), pp. 17, 23.

$$P_n(S,|v|) = \int_R^S P_t(S',|v|)\,dS'. \quad (5)$$

The probability of survival of Cl^+ ions at the distance S is then

$$P_s(S,|v|) = 1 - P_n(S,|v|)\,. \quad (6)$$

Figure 2 shows the probability of survival of "fast" and "slow" $^{37}Cl^+$ ions. The quantity of interest for our experiment is the ratio of survival probabilities of "fast" and "slow" ions at infinite distance from the surface, i. e.,

$$ratio = \frac{P_s(\infty,|v_f|)}{P_s(\infty,|v_s|)}, \quad (7)$$

where v_f and v_s are the velocities of the "fast" and the "slow" $^{37}Cl^+$ ions, respectively.

As can be seen from Figure 2, the survival probability of the Cl^+ ions beyond ≈ 4 Å from a metal surface is predicted to be very small. Our proposed x-ray-ion coincidence experiment would be impossible under such conditions, and may be a problem even with graphite. One way to lower the probability of transition, i. e., to increase the number of ions that reach the detector, is to increase S_τ. This could be achieved by placing several monolayers of stable Ar (e. g., ^{40}Ar) between the graphite substrate and the ^{37}Ar. The ionization energy of argon in the solid state is 13.8 eV,[7] which is greater than the maximum ionization energy of chlorine (12.97 eV);[8] therefore Auger neutralization is not possible from argon. The effect of using such a "buffer zone" has been observed by Dujardin, et al.,[7] who used 100 eV photons to induce desorption of Ar^+ ions from Ar condensed on a platinum substrate. The yield of Ar^+ ions increased rapidly with the addition of every monolayer of Ar between the second and fifth monolayers, but reached a saturation for thicker layers.

If a stable Ar "bed" is used in our experiment, Equation 2 should be rewritten as

$$S_\tau(v) = v\tau + R + Mt_{Ar}, \quad (8)$$

where M is the number of stable monolayers, and t_{Ar} is the effective thickness of one monolayer (≈ 2.63 Å, assuming a face-centered cubic structure for solid Ar[6]). The Cl

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[7]G. Dujardin, L. Hellner, L. Philippe, M.-J. Besnard-Ramage and P. Cirkel, Phys. Rev. B **48**, 14529 (1993) and references therein.

[8]The ionization energy is smaller when the ion is closer to the surface because of image charge effects. See Ref. 3 for further details.

atoms will now be much farther from the substrate when they become charged, and this will change P_n and P_s significantly. Using the numerical values and simple relationships given above, we obtain $P_s \approx 1$ for both "fast" and "slow" ions if $M \geq 3$.

This might seem to solve the problem of ion survival, but only for our selected values of A and a; the uncertainties in these parameters have yet to be addressed. As stated earlier, A and a are normally found by fitting experimental data, and such data do not exist. Fortunately, the addition of three monolayers of argon to the substrate solves this problem, too. In Figures 3 and 4, the fast-slow survival ratio (Equation 7) is plotted versus A and a, respectively, for two monolayers (dashed line) and for three monolayers (full line). The ranges of values for A and a are taken from Hagstrum.[2] It would appear from Figures 3 and 4 that this ratio will be very close to unity for three or more monolayers of Ar, independently of the values of A and a. Of course, more layers may be needed to prevent neutralization of a high percentage of the Cl^+ ions, as many of them will become charged in less than the average time of 9 fs. However, the corresponding average flight distance (≈ 0.6 Å) is quite small compared to the total thickness of three Ar monolayers, so this should not be a serious problem.

Finally, Dujardin, et al.[7] observed that the effective work function of Pt was lowered from 5.65 eV to 4.3(1) eV with the addition of Ar layers, and Ishi and Ohno[9] observed a lowering of the work function for most substrates when covered with Xe. Thus, a similar effect should be expected when Ar is added to a graphite substrate, and this could induce some resonant neutralization. The effect should be small, however, owing to the large (tunneling) distance between the ion and the surface under such conditions.

In summary, it would appear that a few monolayers of stable Ar over the substrate should prevent neutralization of the singly charged Cl ions. This conclusion is reached by using typical parameters for metallic surfaces, but it is presumed that such neutralization would be even less likely for a graphite surface under similar conditions. Thus, it should be possible to set up the experiment such that no corrections for ion velocity will be necessary in determining neutrino mixing fractions. Measurements of the x-ray-ion coincidence yield as a function of the number of monolayers of Ar "bedding" should prove interesting.

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[9]S. Ishi and Y. Ohno, Surf. Sci. **159**, L401 (1985).

Figure Captions

Figure 1: Transition probability density vs. distance from the surface (Eq. 2) for "fast" (solid line) and "slow" (dashed line) Cl^+ ions.

Figure 2: Probability of survival vs. distance from a metal surface (Eq. 2) for "fast" (solid line) and "slow" (dashed line) Cl^+ ions.

Figure 3: Fast-to-slow ratio of survival probabilities vs. parameter A (Eq. 7) for two monolayers (dashed line) and three monolayers (solid line).

Figure 4: Fast-to-slow ratio of survival probabilities vs. parameter a (Eq. 7) for two monolayers (dashed line) and three monolayers (solid line).

Figure 1

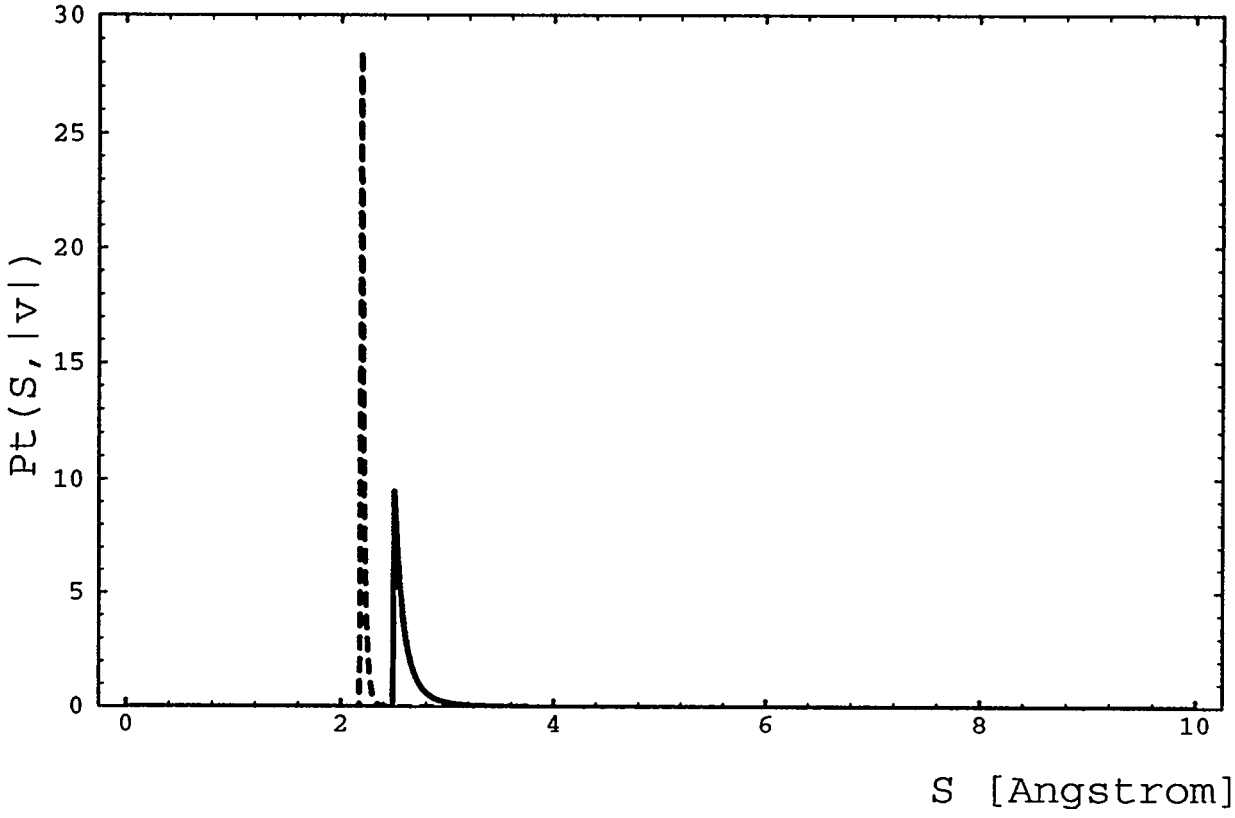

Figure 2

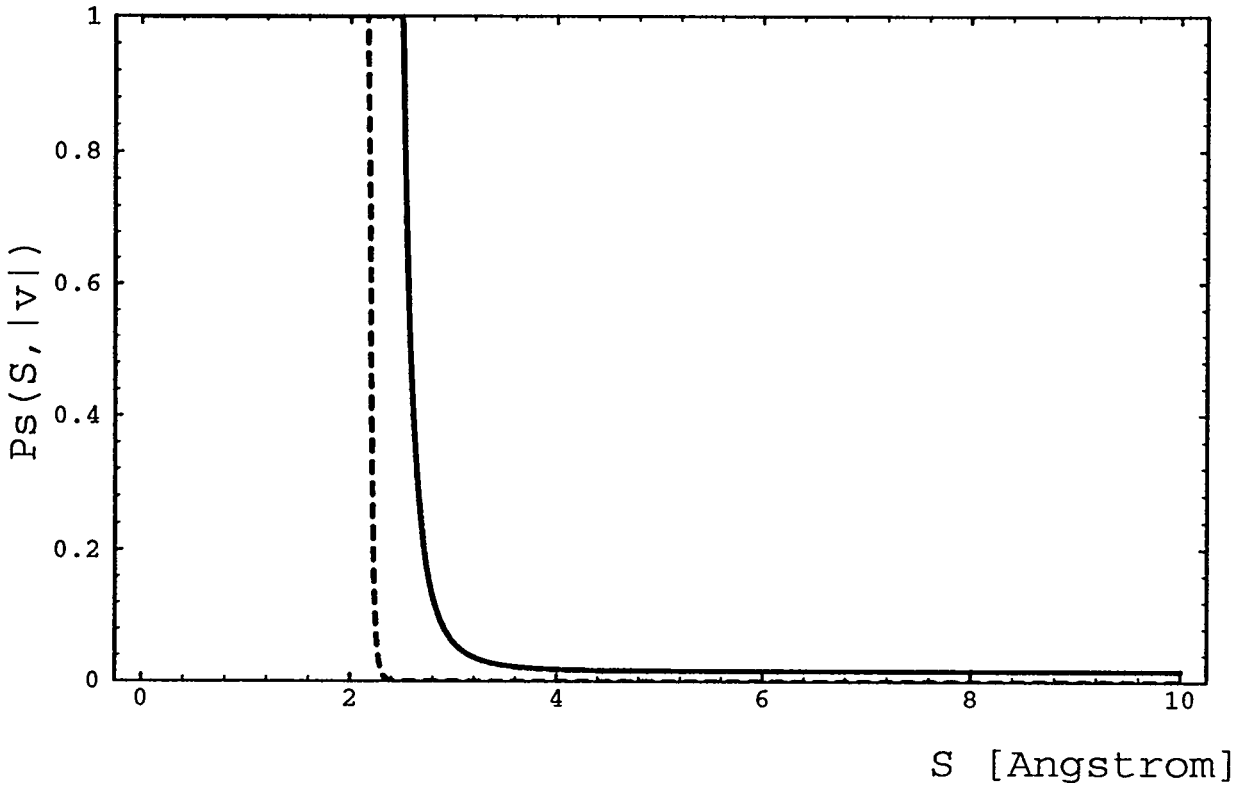

Figure 3

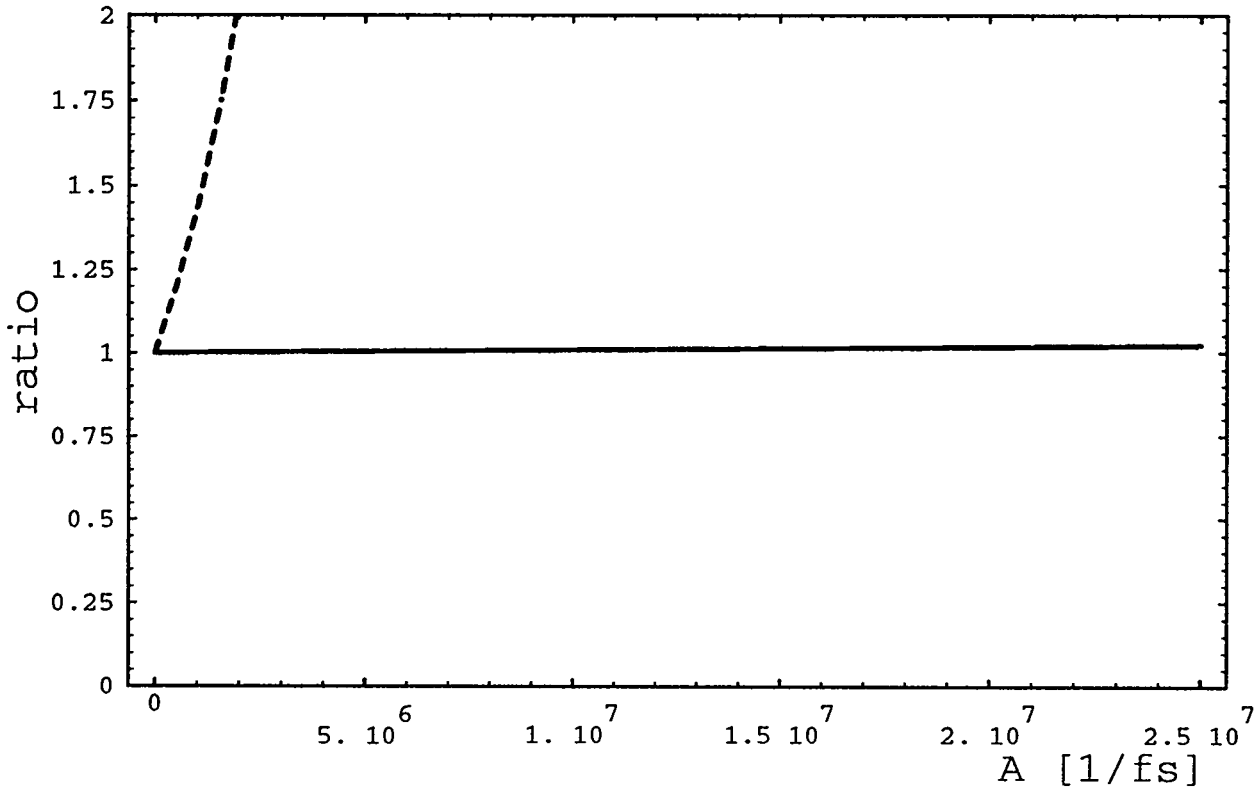
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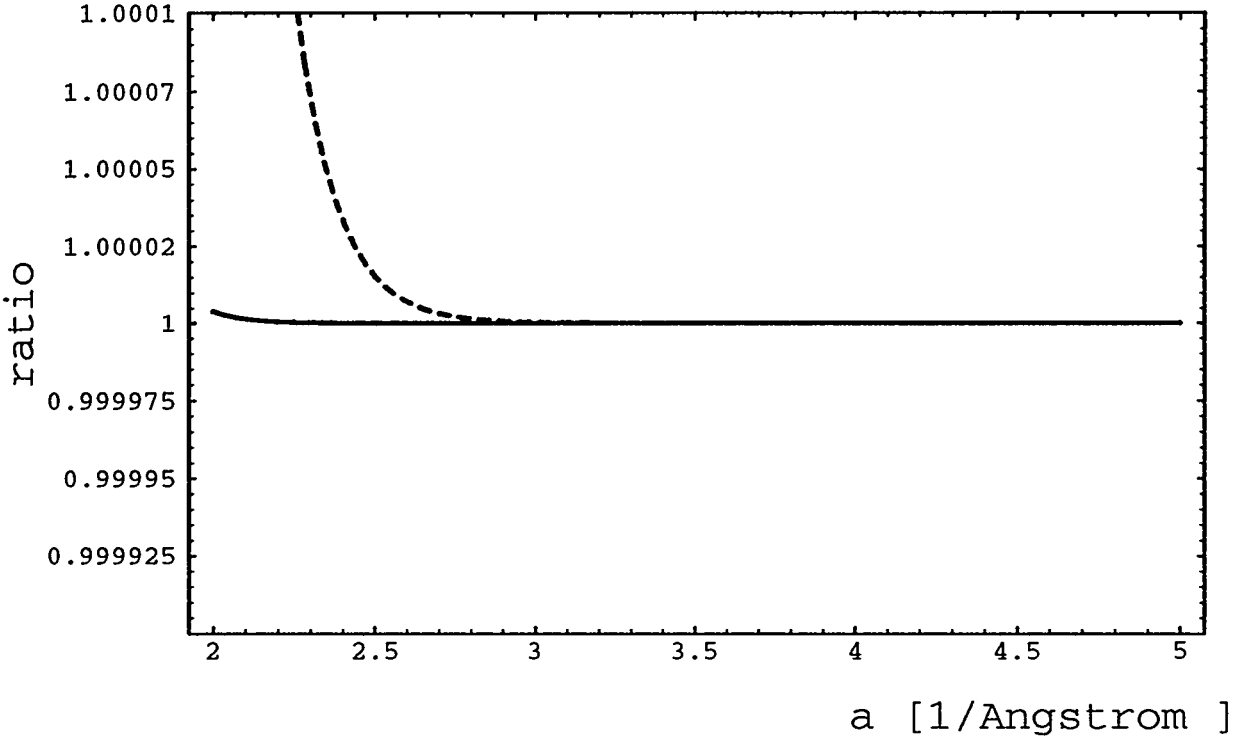
Figure 4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5
a [1/Angstrom]

PUBLICATIONS

(June 1, 1993 - July 31, 1994)

Journal articles:

1. "β^+ decay and cosmic-ray half-lives of ^{143}Pm and ^{144}Pm," M. M. Hindi, A. E. Champagne, M. T. F. da Cruz, R.-M. Larimer, K. T. Lesko, E. B. Norman, and B. Sur, Phys. Rev. C (in press, 1994).

2. "Neutrino Recoil Induced Desorption," L. Zhu, R. Avci, G. J. Lapeyre, M. M. Hindi, R. L. Kozub, and S. J. Robinson, J. Vac. Sci. Technol. A **12**, 2037-2044 (1994).

3. "Search for 17-keV neutrinos in the internal bremsstrahlung spectrum of ^{125}I," M.M. Hindi, R. L. Kozub, and S. J. Robinson, Phys. Rev. C **49**, 3289-3296 (1994).

4. "Search for the β^+ decay of ^{54}Mn," M. T. F. da Cruz, Y. Chan, A. García, M. M. Hindi, G. Kenchian, R.-M. Larimer, K. T. Lesko, E. B. Norman, R. G. Stokstad, F. E. Wietfeldt, and I. Žlimen, Phys. Rev. C **48**, 3110-3112 (1994).

5. "Thick-target yields of iodine isotopes from proton interactions in Te, and the double-β decays of ^{128,130}Te," M. T. F. da Cruz, D. W. Bardayan, Y. Chan, A. García, M. M. Hindi, R.-M. Larimer, K. T. Lesko, E. B. Norman, Della F. Rossi, R. G. Stokstad, F. E. Wietfeldt, and I. Žlimen, Phys. Rev. C **48**, 3106-3109 (1994).

Conference Contributions

1. "β^+ decay and cosmic-ray half-lives of ^{143}Pm and ^{144}Pm," M. M. Hindi, Bhaskar Sur, A. E. Champagne, M. T. F. da Cruz, R.-M. Larimer, K. T. Lesko, and Eric B. Norman, Proc. 23rd Int. Cosmic Ray Conf., Calgary, Canada, Vol. 1, 527-530 (1993).

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3. "β^+ decay and cosmic-ray half-life of ^{54}Mn," M. T. F. da Cruz, Y. Chan, A. García, M. M. Hindi, G. Kenchian, R.-M. Larimer, K. T. Lesko, E. B. Norman, R. G. Stokstad, F. E. Wietfeldt, and I. Žlimen, Proc. 23rd Int. Cosmic Ray Conf., Calgary, Canada, Vol. 2, 293-296 (1993).

4. "Thick-target yields of iodine isotopes from proton interactions in Te, and the double-β decays of ^{128,130}Te," M. T. F. da Cruz, D. W. Bardayan, Y. Chan, A. García, M. M. Hindi, R.-M. Larimer, K. T. Lesko, E. B. Norman, Della F. Rossi, R. G. Stokstad, F. E. Wietfeldt, and I. Žlimen, Proc. 23rd Int. Cosmic Ray Conf., Calgary, Canada, Vol. 3, 850-853 (1993).

5. "Neutrino Recoil Induced Desorption," L. Zhu, M. Hindi, R. Avci, G. J. Lapeyre, R. Kozub, 40th National AVS Symposium and Topical Conferences, Orlando, Florida (November, 1993).

Abstracts

1. "Neutrino Recoil Desorption and Internal Relaxation of ^{37}Cl Following Electron Capture Decay of ^{37}Ar," R. Avci, L. Zhu, G. J. Lapeyre, M. M. Hindi, R. L. Kozub, and S. J. Robinson, Bull. Am. Phys. Soc. **39**, 780 (1994).

2. "Measurement of the X-Ray-Internal Bremsstrahlung Coincidence Spectrum from the Decay of ^{55}Fe," I. Žlimen, Y. D. Chan, M. T. F. da Cruz, A. García, R.-M. Larimer, K. T. Lesko, E. B. Norman, R. G. Stokstad, F. E. Wietfeldt, and M. M. Hindi, Bull. Am. Phys. Soc. **38**, 1804 (1993).

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4. "Production Cross Sections from the Bombardment of Natural Mo with 1.85-GeV Protons," D. W. Bardayan, M. M. Hindi, M. T. F. da Cruz, Y. D. Chan, A. García, R.-M. Larimer, K. T. Lesko, E. B. Norman, D. F. Rossi, R. G. Stokstad, F. E. Wietfeldt, I. Žlimen, and A. F. Barghouty, *Ibid.* p. 1830.

5. "Search for 17-keV Neutrinos in the Internal Bremsstrahlung Spectrum of ^{125}I," R.L. Kozub, M. M. Hindi, J. G. Parker, and S. J. Robinson, *Ibid.* p. 1854.

6. "Search for Massive Neutrinos in the Recoil Spectrum of ^{37}Cl following the Electron Capture Decay of ^{37}Ar," M. M. Hindi, D. W. Bardayan, R. L. Kozub, S. J. Robinson, R. Avci, Lin Zhu, and G.J. Lapeyre, *Ibid.* p. 1855.

7. "Further Studies of a ^{14}C-Doped Germanium Detector," F. E. Wietfeldt, Y. D. Chan, M. T. F. da Cruz, A. García, E. E. Haller, W. L. Hansen, R.-M. Larimer, K. T. Lesko, P. N. Luke, E. B. Norman, R. G. Stokstad, I. Žlimen, B. Sur, and M. M. Hindi, *Ibid.* p. 1855.

TTU PERSONNEL
(June 1, 1993 - July 31, 1994)

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APPENDIX

Reprints & preprint removed and cycled separately.