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Nuclear Chemistry Progress Report.

Our program of research has been concerned with three main areas: The first involves nuclear structure and reactions. The second is concerned with nuclear hyperfine interaction in metals and chemical compounds. The third area involves experimental and theoretical investigations of the chemical effects accompanying Mu meson capture in chemical compounds.

Because of the delays in the schedule for Mu meson beams at the LAMPF accelerator at Los Alamos, and the limited residence period permitted Mr. Len Mausner in the Graduate School of Princeton University, we have shifted a portion of the Mu meson capture program to the 184 inch cyclotron at the Lawrence Berkeley Laboratory. 200 hours of beam time were allocated to our experiment by the Scheduling Committee of the 184 inch cyclotron. We have already obtained the first results in a collaboration with Doctor Selig Kaplan and Joyce Monard of the Lawrence Berkeley Laboratory.

We have continued our program in nuclear hyperfine spectroscopy, a very fruitful collaboration with scientists at the Bell Telephone laboratories. We believe we have observed several striking and novel effects in our investigations of the hyperfine interaction of radioactive atoms implanted into solid metals. Mr. Lis, one^{of} our second year graduate students, will be extending this type of research to insulators, both extended solids and solids where discrete molecules exist.

The program at the isotope separator beside the implantation technique is being used to investigate short-lived radioactive nuclei in an on-line mode. A new high temperature source has been built and tested; the solid state ion source has been modified to minimize the handling time needed in changing radioactive sources. Further investigations of liquid-ion sources are being carried out.

During the Spring of 1974 the Q3D heavy particle magnetic spectrograph became operational at the Princeton University sector-focus cyclotron. This instrument has one of the highest resolution-transmission capabilities available anywhere. This spectrograph in combination with the cyclotron which provides beams of sharply defined high energy particles affords a unique spectroscopic facility. We have commenced using the spectrograph to continue our studies of the odd A rhodium and silver isotopes by the (p,t) reaction. The high resolution spectra taken with the spectrograph has revealed a doublet structure which was not resolved using semiconductor particle detectors. Because of the high resolution and transmission figures of merit, this heavy particle spectrograph should also function as an alpha spectrograph of superior resolution. We are commencing to assess this capability using natural radioactive sources. We intend to follow these initial studies with investigation of rare earth and trans lead alpha emitting isotopes prepared at the Princeton cyclotron and deposited with the isotope separator.

In the year ahead the mu meson channel at the Los Alamos Meson Facility will be providing the highest fluxes of stopping mu mesons so far available. We intend to continue our collaboration with staff members of the C.N.C. 11 Division of Los Alamos to investigate the initial capture mechanisms for negative mu mesons using these unique beams. At Princeton we will continue our program in nuclear structure using both radioactive source spectroscopy and the heavy particle reaction method. At Princeton we will also continue our productive collaboration with the Bell Telephone Laboratories investigating nuclear hyperfine interactions in solids.

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Study of Short-Lived Isomers at the Los Alamos Tandem Accelerator, R. A. Naumann
(with V. R. Casella and J. D. Knight, CNC 11 division Los Alamos Scientific
Laboratory)

A program of studying nuclear isomers in the 0.1 to 10 second range has been initiated at the Los Alamos tandem accelerators. To accomplish this it has been necessary to have a chopped beam available so that the short-lived nuclear states can be observed between beam bursts. This has been accomplished in two ways. For two stage operation a pulser has been installed at the ion source of the tandem van de Graaf accelerator. This pulsergates the ion source and provides an optical signal when the ion source is on. The optical signal is transmitted to ground potential through a fibre-optic light link to a photodiode. The fibre optic link withstands the 150 kilovolt negative potential of the ion source and the photodiode provides a timing signal at the end of the beam burst.

For three stage operation, it is not practical to transmit a signal from the negative terminal of the first van der Graaf which is at -5 to -7 Megavolts with respect to ground. Instead, magnetic deflection of the fully accelerated beam is employed and the magnet switching circuit provides the timing signal at the end of the beam burst to the experimental area.

The timing signals at the end of the beam burst turn on a computer which records ten successive 1024 channel spectra from a Ge(Li) gamma

spectrometer which views the reaction chamber and target where the isomers are produced.

For the initial work E3 isomers in Yttrium 93 and Silver 101 have been investigated. The Yttrium 93 isomer investigation exploits the availability of the unique triton beam at Los Alamos and our study has resulted in the discovery of the 0.82 second isomeric state. The abstract of the paper given at the April 1973 meeting of the Washington American Physical Society follows:

Observation Of The E-3 Isomer Of ^{93}Y

Using the reaction $^{94}\text{Zr}(t,\alpha)$ with 12-to 16-MeV tritons, we have measured the decay of the E3 isomer of ^{93}Y . The activity was produced by short bursts of beam on an isotopically enriched ^{94}Zr target; the decay γ -rays were measured with a Ge(Li) detector - PHA system operated by a computer arrangement set up to turn on the γ -spectrometer at the end of each beam burst, record up to 10 successive 1024-channel spectra at intervals as short as 0.2 sec, and sum up corresponding spectra for a long series of bursts. The $^{93\text{m}}\text{Y}$ half life was found to be 0.82 ± 0.04 sec. Decay of the isomer is accompanied by γ -rays of 169 and 590 keV, in agreement with other nuclear spectroscopic data. The radiative lifetime of the isomeric state is 2.7 Weisskopf units, consistent with a shell-model picture (based on published $^{94}\text{Zr}(d, ^3\text{He})$ data) of a $g_{9/2}$ state decaying to a $J^\pi = 3/2^-$ state containing about 25% $p_{3/2}^{-1}$.

The discovery of the silver 101 isomer continues our investigations of the low lying isomers of the odd A silver isotopes. We have irradiated an enriched palladium 102 target with 24 MeV protons producing the isomeric

state by the $^{102}\text{Pd}(p,2n)^{101\text{m}}\text{Ag}$ reaction. Figure I shows three of the successive gamma spectra accumulated. The decay of the 176 and 98 keV gamma transitions is evident. Figure II shows the 3.07 ± 0.09 sec half-life derived from these data.

Earlier work at the ISOLDE project at CERN had sought this isomer using electron capture of mass separated 1.2 minute cadmium 101 sources. This investigation revealed the 176 and 98 keV transitions we have observed and included an E3 multipolarity determination for the 176 keV transition but failed to find the isomeric half life.

After correction for internal conversion a radiative lifetime of 6.55 seconds is found. It is of interest to compare the reduced radiative lifetimes for the 6 isomers now known in the odd silver isotopes involving $7/2^+$ and $1/2^-$ states. This is shown in the table below after multiplying by the factor $A^2 E^7 S$ to correct for size, energy, and nuclear spin statistical factors:

Table I

Reduced Radiative Lifetimes Odd A Silver Isomers

A	E^7 (MeV ⁷)	S	$T_{1/2}$ rad (secs)	$A^2 E^7 S T_{1/2}$
101	5.13×10^{-6}	4	6.55	13.7×10^{-1}
103	9.52×10^{-7}	4	24.3	9.81×10^{-1}
105	7.01×10^{-12}	1	9.37×10^6	7.25×10^{-1}
107	6.51×10^{-8}	1	9.48×10^2	7.06×10^{-1}
109	3.99×10^{-8}	1	1.13×10^3	5.36×10^{-1}
111	4.90×10^{-9}	1	8.81×10^3	5.21×10^{-1}

The trend towards decreasing reduced lifetime with increasing mass number is evident. We believe this trend is most simply explained by postulating an increasing admixture of the $g_{7/2}$ shell model state into the $7/2^+$ state as the nuclear deformation increases with increasing neutron number.

1 24MEV PROTONS ON 102PD RUN 10 3/1/74 98KEV

$T_{1/2}$
3.071 ± 0.09 SEC

03/04/74 21.59.59

Study of Nuclear Quadrupole Interaction in Metals using Perturbed Gamma-Ray Angular Correlation, E. Ansaldi and R. A. Naumann (with R. Raghavan, P. Raghavan and E. Kaufman, Bell Telephone Laboratories)

During the past year we have continued to investigate the electric field gradients acting on the nuclei of atoms implanted into metals of non-cubic structure. This work uses perturbed gamma ray angular correlation as an indicator of the electric field gradient. The perturbation depends upon the product of this gradient and the nuclear quadrupole moment upon which the field acts.

Our work has primarily involved the 247 keV first excited state of cadmium 111 which has a lifetime of 84 nanoseconds, a nuclear spin of $5/2$ and a sizeable quadrupole moment (approximately 0.77 barns). The electric field gradients in solids (and molecules) can act on the 247 keV state during its lifetime and the perturbation revealed using the time dependence of the perturbed angular correlation. The effect appears as the periodic modulation of the decay curve derived from the time to amplitude converter of a fast gamma-gamma coincidence circuit. The frequencies appearing are directly related to the nuclear quadrupole hyperfine energy differences.

The 247 keV level of interest may be populated in three ways: beta minus decay of 7.5 day silver 111, decay of the 49 minute isomeric state of cadmium 111 or electron capture of 2.8 day indium 111. The silver 111 has been prepared by neutron irradiation of natural palladium or palladium enriched in the 110 mass number isotope. The cadmium 111 isomer and the indium 111 have been prepared by bombardment using alpha particles from the Princeton cyclotron. Atoms of all three parents have been implanted using the Princeton isotope separator with post acceleration at the collector to implant the radioactive atoms at 90 keV energy.

Some results of our work are:

- (1) The temperature dependence of the nuclear quadrupole interaction of cadmium atoms in zinc has been measured by implanting indium 111 in polycrystalline metallic zinc. The temperature ranged from 4°K to 400°K . Previous theoretical treatments using a point positive ion lattice have suggested that the quadrupole interaction should increase with temperature due to the elongation of the unit cell with temperature. Instead we observe a decrease with temperature using a source which has been annealed after implantation. The cadmium nuclei under these conditions occupy substitutional sites in the crystal lattice. Our data indicate that the conduction electrons make a larger contribution of opposite sign to the electric field gradient than that expected from the lattice.
- (2) Usually the temperature dependence of the electric field gradient acting at the nuclei of atoms implanted into metals depends only upon the host lattice and is independent of the nature of the atom implanted. The temperature dependence of the nuclear quadrupole interaction of cadmium and tantalum nuclei located at substitutional sites in titanium have been measured over the temperature range $4^{\circ} - 400^{\circ}\text{K}$. A striking difference has been found; the relative cadmium quadrupole interaction decreases far more rapidly with temperature than that for tantalum. This is the first time that a significant dependence of the temperature variation of the electric field gradient on the nature of the implanted atom has been observed.
- (3) It is expected that the nuclear quadrupole interaction frequencies observed by perturbed angular correlation for the same metallic host crystal and the same nuclear excited state should be independent of the radioactive process by which the given nuclear excited state is formed. This expectation

rests on two assumptions (a) the electronic relaxation processes in metals are rapid and (b) that the radioactive parent atoms occupy uniform and identical sites in the host crystal. The observation of well defined periodic precession curves checks that the implanted atoms occupy uniform sites.

We have implanted cadmium 111 m and indium 111 into metallic beryllium. As expected, within experimental error, identical quadrupole frequencies of 52.5 MHz and 54.6 MHz respectively are found.

For the case of silver 111 in the same metallic beryllium host a frequency about 30% of the former value is found. (16.0 and 17.1 MHz). Preliminary results of channeling experiments carried out at the Rutgers tandem accelerator on the implanted beryllium crystals indicate that the implanted cadmium and indium atoms occupy one type of site in crystalline beryllium while the implanted silver atoms occupy a different second site. The sharp periodic modulations observed in the perturbed angular correlation measurements indicate these sites, whenever populated, contains the majority of the implanted atoms.

The following three abstracts concerning our work on nuclear hyperfine interactions have been submitted and reported at the Conference on Hyperfine Interactions, Institute of Physics, University of Uppsala, Sweden June 10-15, 1974.

SIGN OF THE QUADRUPOLE INTERACTION AT ^{111}Cd IN NON-CUBIC METALS BY β - γ PERTURBED ANGULAR CORRELATIONS.

by E. N. Kaufmann, P. Raghavan^{*} and R. S. Raghavan
Bell Laboratories, Murray Hill, N. J.
and E. J. Ansaldo and R. A. Naumann
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Knowledge of the sign of the nuclear electric quadrupole interaction (QI) is vital to the understanding of electric field gradients in solids. This is especially true for metallic systems in which the sign may be largely determined by the conduction electrons. A significant advance in the methods available to measure the sign of the QI has been the recent introduction of the β - γ perturbed angular correlation (PAC) technique,¹ which simplifies the measurement considerably. An important extension of this technique was the demonstration² that it is applicable to the 247 keV level of ^{111}Cd , which has been utilized to measure the largest number of QI constants by PAC experiments. This opens the way to the determination of the sign of the QI in most of these cases.

In this work we report the sign of the QI at ^{111}Cd embedded in several non-cubic metals. The unique first-forbidden (UFF) β -decay ($\Delta I=2$, yes) of ^{111}Ag (7-day) populates the β - γ cascade

$$\frac{1}{2}^- \xrightarrow{\beta^-} \frac{5}{2}^+ \xrightarrow{\gamma(E2)} \frac{1}{2}^+$$

through the 247 keV ($T_{1/2} = 84$ ns) level of ^{111}Cd . As a result of parity non-conservation in β -decay, the emission of the first radiation leaves the 247 keV level polarized. Because only one nuclear matrix element contributes to a UFF transition, the degree and sign of the nuclear polarization is uniquely determined by the β^- emission direction. The quadrupole precession of the nucleus in a single crystal has the special property of converting the polarization to an alignment, the sign of which is determined by the sign of the QI. This alignment can then be observed as an anisotropy in the emission of the succeeding γ -ray relative to the β -particle direction and the crystalline symmetry ($\bar{C}$) axis. The sign of the anisotropy leads directly the sign of the QI coupling constant, e^2qQ .

When the β -active nuclei are embedded in only one face of the host crystal, the most convenient detection geometry is that shown in Fig. 1a. The β -particles are observed normal to the $\bar{C}$ -axis which is in the plane of the crystal and the γ -rays are detected alternately at the positions denoted by $\phi = \pm \pi/2$ in the figure. The theory shows that the asymmetry ratio

$$A(t) = 2[W(-\pi/2) - W(\pi/2)] / [W(-\pi/2) + W(\pi/2)],$$

which can be computed from the observed β - γ delayed coincidence spectra, is given by $A(t) = a \sin 2\omega_0 t + b \sin 3\omega_0 t$ for the cascade in question, where a and b are positive and

$\omega_0 = \frac{3\pi}{10} (e^2qQ/h)$. Thus the sense of measured $A(t)$ spectra directly reveals the signs of e^2qQ .

^{111}Ag sources were produced in the reaction $^{110}\text{Pd}(n, \gamma)^{111}\text{Pd} \xrightarrow{\beta^-} ^{111}\text{Ag}$. In the first application of this technique,² chemically separated ^{111}Ag was diffused into a Cd single crystal and the $A(t)$ spectrum is reproduced in Fig. 1b. For subsequent experiments, in order to insure that all of the activity was actually in the host lattice while still sufficiently close to the crystal surface to minimize β scattering and absorption, the ^{111}Ag was implanted at 90 keV energy using an isotope separator. The resulting spectra for Sn, Ti and Be host crystals are shown in Figs. 2a, b, c. It can be seen from the sign of the initial excursion of $A(t)$ near $t = 0$ that for the cases of Cd, Sn and Ti (Fig. 1b and Figs. 2a, b) that $e^2qQ > 0$ and that for Be (Fig. 2c) $e^2qQ < 0$. From nuclear systematics on generically related 5^+ levels in Cd isotopes it is believed that $Q > 0$; thus the sign of the field gradient, e^2q , is determined. Point ion lattice sum calculations for these metals indicated that e^2q (lattice only) is positive for Ti and Be and negative for Cd and Sn. The effect of the electronic contribution to e^2q , therefore, overwhelms and reverses the sign of the lattice field in Cd (as has been theoretically predicted³) and in Sn, whereas not in Ti. Be must be omitted from this consideration since, unlike in the other three hosts, a very interesting anomaly between this measurement and previous γ - γ PAC measurements in Be has been observed which is the subject of another contribution to this Conference.⁴

^{*}Resident Visitor from Rutgers University, New Brunswick, N. J.; supported in part by the N.S.F.

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Fig. 1a Experimental geometry showing the emission directions of the β and γ radiations relative to the single crystal symmetry (c) axis.

Fig. 1b $A(t)$ spectrum from Ref. 2 for the case of ^{111}Ag diffused into a single crystal of hexagonal close packed Cd.

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Fig. 2a $A(t)$ spectrum for ^{111}Ag implanted into a single crystal of tetragonal β (white) Sn.

Fig. 2b $A(t)$ spectrum for ^{111}Ag implanted into a single crystal of hexagonal close packed Ti.

Fig. 2c $A(t)$ spectrum for ^{111}Ag implanted into a single crystal of hexagonal close packed Be.

OBSERVATION OF LOCAL TEMPERATURE DEPENDENCE IN THE IMPURITY ELECTRIC QUADRUPOLE INTERACTION.

by R. S. Raghavan, E. N. Kaufmann and P. Raghavan*
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and K. Krien** and R. A. Neumann
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Measurements of the nuclear electric quadrupole interaction (QI) at impurities in non-cubic metals have been reported by several groups recently. It is hoped that such information will shed light on the electronic structure of the metal as well as on the specific impurity-host interaction. To this end, the temperature dependence of the QI has been measured in several cases¹ where it was found that for a given host the magnitudes of the QI varied from one impurity to another but the relative variation of the QI with temperature was independent of the specific impurity. Such observations are consistent with the notion that the temperature variation arises mainly from the thermal expansion of the host lattice and from thermal redistribution of host conduction electrons among states of different spatial symmetry and is therefore a property only of the host characteristics.

We report here the first observation of a markedly different temperature dependence at two impurities in the same host. Using the time-differential perturbed angular correlation technique on the well-known 247 keV level of ¹¹¹Cd and 482 keV level of ¹⁸¹Ta, the QI was measured as a function of temperature for these impurities in a Ti metal host. The parent activities, ¹¹¹In (produced by ¹⁰⁹Ag (α, n) ¹¹¹In) and ¹⁸¹Hf (from ¹⁸⁰Hf (n, γ) ¹⁸¹Hf), were implanted at 90 and 70 keV energy, respectively, into a freshly cleaned Ti single crystal using the Princeton and Bonn isotope separators. Unique QI frequencies were found in each case and were assigned to impurities at regular lattice sites. This assignment is based on a previous channeling measurement on In implanted Ti² (for the Cd case) and on the agreement of the QI frequency observed with TiHf alloys³ (for the Ta case). The room temperature values for the QI frequency $\nu_Q = e^2qQ/h$ are 27.8 ± 0.3 MHz for ¹¹¹Cd and 345 ± 5 MHz for ¹⁸¹Ta. The temperature variations normalized to 4.2°K are shown in Fig. 1 (where some data from Ref. 3 are included).

The substantial difference between $\nu_Q(T)$ for Cd and Ta impurities must be attributed to distinctly local differences in the way each impurity interacts with the Ti host. Local lattice distortion as well as impurity charge screening mechanisms will probably play an important role in the explanation of this effect.

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Fig. 1. Values of ν_Q normalized to their respective values at 4.2°K plotted against temperature for the Cd probe (○), the Ta probe (□), and the data from Ref. 3 (x). (The points at 77°K have been slightly displaced horizontally for the sake of clarity.) The solid lines have been drawn to guide the eye.

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QUADRUPOLE INTERACTION OF ^{111}Cd IN Be FOLLOWING RADIOACTIVE ION-IMPLANTATION OF PARENT Ag, Cd AND In: A PUZZLE

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 R. S. Raghavan and E. N. Kaufmann, Bell Laboratories, Murray Hill, N. J.
 E. J. Ansaldo and R. A. Naumann, Princeton University, Princeton, N. J.

As part of a program¹ to determine the sign and magnitude of the quadrupole interaction (QI) coupling constant e^2qQ of the 247 keV level of ^{111}Cd after radioactive ion-implantation into a number of non-cubic metals, we have performed a series of experiments in Be metal. Normally one expects the QI frequency to be independent of the type of parent nucleus which populates the level. The present experiments show that e^2qQ for this level in Be differs by more than a factor of 3 depending on whether the implanted parent nuclide is ^{111}Ag on the one hand or ^{111}mCd and ^{111}In on the other. Such a result has not been encountered before and remains, at the present time, a puzzle.

The 247 keV level of ^{111}Cd can be populated by the β^- decay of ^{111}Ag (7d), the isomeric γ -decay of ^{111}mCd (49 min.), or the electron-capture (E.C.) decay of ^{111}In (2.8d), permitting β - γ (^{111}Ag) or γ - γ (^{111}Ag , ^{111}mCd , ^{111}In) perturbed angular correlation (PAC) experiments on the ^{111}Cd level. The activities mentioned, were produced by reactor irradiation of Pd or ^{109}Ag or ^{115}In bombardment of Pd or Ag foils in the Princeton University cyclotron and implanted with an energy of 90 keV using the Princeton University Isotope Separator facility into a single crystal or polycrystalline foil of Be metal. Experiments with ^{111}Ag sources utilized the $\frac{1}{2}^+ \rightarrow \frac{5}{2}^+ \rightarrow \frac{7}{2}^+$ cascade for β - γ PAC measurements² in the single crystal while the (93-247) keV cascade was employed for the γ - γ PAC experiments with mono- or polycrystalline sources. The (150-247) and the (173-247) keV cascades were utilized for γ - γ PAC measurements following ^{111}mCd and ^{111}In decays, respectively. All implants and PAC measurements were performed at room temperature; and sources were used without any further treatment after the ion-implantation. To make a clear comparison of the results with implants of the three parents, the same single crystal was used in all three cases, the residual activity being etched away after each experiment. The QI constants obtained are listed in Table I, and typical PAC spectra are displayed in Figs. 1 and 2. The main features of the results can be summarized as follows:

- The QI of the $\frac{5}{2}^+$ level in ^{111}Cd in Be metal, measured after ^{111}Ag implantation is more than a factor of 3 smaller than that observed after ^{111}mCd or ^{111}In implantation.
 - The PAC quadrupole precession spectra are very well-defined in each case, indicating that the great majority of atoms are at unique final sites, regardless of the implanted parent species.
 - The single crystal experiments show that the axis of symmetry of the QI is apparently along the c-axis of the crystal, regardless of the type of the parent species.
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- The fact that the same frequency is observed with ^{111}mCd (isomeric γ -decay) or with ^{111}In (E.C. decay) rules out the possibility that E.C. after effects play a role. The "anomalous" result, at present, appears to be that obtained with ^{111}Ag .
 - The dependence of QI coupling constant on the nature of the parent species is observed so far only in Be. Measurements with ^{111}Ag and ^{111}In in Ti, Sn, and Cd show the same QI frequency with either parent. The anomaly appears to be linked to the impurity-host pair Ag-Be only.

A satisfactory understanding of the principal result, viz. observation a) above, is lacking at this time. We are pursuing two main lines of investigation to solve this puzzle. (a) A difference in the nature of the final sites of Ag in Be as contrasted to Cd and In; and (b) vacancy-associated field-gradients preferentially produced at the Ag sites. Channeling experiments, to elucidate the lattice location and investigations on the annealing behavior of the QI observed with each parent species, are currently in progress.

*Supported in part by the N.S.F. Resident Visitor at Bell Labs.

- E. N. Kaufmann, P. Raghavan, R. S. Raghavan, E. J. Ansaldo and R. A. Naumann, Contribution to this Conference.
- P. Raghavan, R. S. Raghavan and E. N. Kaufmann, to be published.

Table I. Summary of QI frequencies of the 247 keV level of ^{111}Cd in Be.

Parent	Decay Mode	Crystalline form of Be	PAC method	e^2qQ/h (MHz)
^{111}Ag	β^-	single crystal	β - γ	-16.0(2)
^{111}Ag	β^-	single crystal	γ - γ	$\pm 17.1(7)$
^{111}mCd	γ	single crystal	γ - γ	$\pm 52.5(15)$
^{111}In	E.C.	single crystal	γ - γ	$\pm 54.6(5)$
^{111}In	E.C.	Polycrystal	γ - γ	$\pm 54.9(5)$

Fig. 1 Quadrupole precession curves for 5^+ level of ^{111}Cd in a Be single crystal following implantation of ^{111}Ag . Upper curve: β - γ PAC. Lower curve: γ - γ PAC with c-axis perpendicular to detector plane.

Fig. 2 Quadrupole precession curves for 5^+ level of ^{111}Cd in a Be single crystal (c-axis perpendicular to detector plane) by γ - γ PAC following ^{111}In implantation (upper curve) and $^{111\text{m}}\text{Cd}$ implantation (lower curve).

PROGRESS REPORT: Chemical Structure Effects in the
Capture of Negative Muons in Matter;

L. F. Mausner and R. A. Naumann
(with Jeri Knight, Los Alamos Scientific
Laboratory and Selig Kaplan, Joyce Monard,
Lawrence Berkeley Laboratory)

Experiments in the last decade have provided much evidence that the molecular structure of matter affects the process of the atomic capture of negative muons. In particular, a current model¹ discussed in the latest Annual Reviews of Nuclear Science predicts that in slowing down and stopping in a chemical compound a large fraction of the muons are initially captured into orbitals of the molecule as a whole rather than into the orbitals of individual atoms. In this model for these highly excited initial states the mesoatomic orbits are comparable to the size of the capturing molecules and exceed by hundreds of times the characteristic radius of the K mesoatomic orbital near the nucleus. The muon then de-excites by radiative and auger transitions. Further questions arise. For example, what is the mechanism of initial capture and what angular momentum distribution does it produce? In the subsequent de-excitation what process determines the ratio of muon captures on each atomic center? We are attempting a systematic study of these phenomena using isoelectronic sequences of compounds.

To this end, we are measuring muonic X-ray spectra at the 184 inch cyclotron of the Lawrence Berkeley Laboratory and the LAMPF accelerator in a carefully chosen series of targets with a high resolution Ge(Li) detector system, looking for correlations of X-ray intensity ratios with chemical structure. We have taken our initial experimental data at the 184" cyclotron of the Lawrence Berkeley Laboratory because of the delays in the LAMPF schedule. We have recently completed a series of bombardments on the sequence Ar(liquid), KCl and CaS. Since KCl and CaS are to a great extent ionic compounds, each atom in the sequence may be considered to be surrounded by an equal number of electrons. With this parameter held constant we may more easily investigate the effect of atomic charge on the muon captures. Preliminary analysis of the data shows that the positive ions, K and Ca, capture a larger fraction of the muons than the negative ions, larger even than the ratio of nuclear charges predicted by the Fermi-Teller Z Law. At first sight this seems unsurprising but actually is contrary to our initial expectations based on the molecular orbital model. We have also extracted the muon population distributions for s, p, and d states up to $n=7$. The distributions are heavily weighted toward the circular orbits, pointing to a similarly weighted non statistical initial angular momentum distribution.

There is also interest in the liquid argon target apart from its place in this series. In 1971 at CERN Backenstoss et al.² stopped negative muons in a high pressure gaseous Ar target. They observed the $K\alpha$ line ($2p \rightarrow 1s$ transition) but no other higher members of the Lyman series. This result was unexpected, unexplained, and in disagreement with some earlier NaI work by Budyashov.³ Also, it was unlike muonic X-ray spectra we obtained at the SREL cyclotron in 1970 for the isoelectronic Cl^- ion in LiCl. At Berkeley we were easily able to observe 7 members of the Lyman series in Ar. The attached plot of our data clearly shows this. If there is no error on the part of the CERN group these results imply a very large physical state influence on the muon capture mechanism. We are presently discussing the feasibility of using a solid Ar target.

Our next runs at Berkeley and Los Alamos are scheduled during the summer. Among proposed targets are pure Ca metal, pure S, and CaSO_4 , all of which will be compared with our CaS results. This will enable us to investigate the effect of formal valence charge on the X-ray intensity ratios. We will also study NaClO_4 , MgSO_4 and AlPO_4 , an isoelectronic, isostructural series.

Concurrent with this experimental work we have been investigating methods to calculate eigenfunctions and eigenvalues for excited states of the helium atom with one electron replaced by a muon. Instead of using unscreened hydrogenic wavefunctions as is usual we have developed a simple method that utilizes the Virial Theorem to help determine proper values of screening parameters for both the muon and the electron. It gives improved results over simple first order perturbation theory with only slightly more computational effort. We are now using these wavefunctions and energies to calculate radiative and auger transition rates from states where the electron and muon orbital radii are approximately equal ($Nu = 15$).

References:

1. L. I. Ponomarev; Annual Reviews of Nuclear Science 395 (1973)
2. G. Backenstoss, H. Daniel, K. Jentzsch, H. Koch, H. Povel, F. Schmeissner, K. Springer, R. Stearns; Phys. Letters 36 B, 422 (1971)
3. Yu. Budyashov, P. Ernolov, V. Zinov, A. Konin, A. Mutkhin; Sov. J. Nucl. Phys. 5, 529 (1967)

Liquid Argon Target
184" Cyclotron
April 9, 1974

Research Employing The Q3D Spectrometer (R. Del Vecchio, I. Oelrich, and R. A. Naumann)

The Q3D heavy particle spectrograph affords both high resolution ($\frac{\Delta p}{p} < 10^{-4}$) and high transmission, 13 milliradian. The apparatus has been completed during early 1974 and experimental programs using this instrument are underway. As noted in our 1972 progress report, we have been interested in the nuclear level structure of the odd A rhodium and silver isotopes ($Z=45, 47$). The ground state spin of these nuclei is $1/2^-$ reflecting the participation of the $p_{1/2}$ proton shell model state in the ground state particle configuration of this nucleus. Certain excited states in rhodium 101, silver 105 and silver 107 might then be expected to arise from coupling the intrinsic ground state ($1/2^-$) with the collective excited states of the core nuclei (ruthenium 100, palladium 104 and palladium 106). These core nuclei are known to have lowest lying near harmonic collective excited states, ie. the first excited state has spin 2^+ followed by a nearly degenerate triplet of states with spins 0^+ , 2^+ and 4^+ . Thus the core coupling model predicts, for example in rhodium 101, a doublet states with spins $3/2^-$ and $5/2^-$ near the 2^+ state of ruthenium 100 and a quintet of states with spins $1/2^-$, $3/2^-$, $5/2^-$, $7/2^-$ and $9/2^-$ arising near the triplet of states in the even core. In earlier work using the reaction $^{103}\text{Rh}(p,t)^{101}\text{Rh}$ at the University of Pittsburgh van de graaf we have resolved the doublet and some members of the quintet. Now using the Princeton Q3D spectrograph we have resolved the expected 5 members of the quintet expected in Rhodium 101. A system resolution of 10 keV at 30 MeV was required to distinguish the quintet members. At this resolution less energetic triton groups corresponding to higher excited states of rhodium 101 also revealed doublet structure.

We have separated 3.6 day radium 224 from 1.9 year thorium 228 by ion exchange. Figure A shows the alpha spectrum of the radium 224 and

daughter activities taken with a solid state alpha spectrometer. The Q3D spectrometer offers the possibility of taking high resolution alpha spectra. We have been exploring this possibility using the radium 224 sources. Figure B shows the high energy alpha group. The primary limitation of the instrument as an alpha spectrometer appears to be thickness of sources. We are investigating the preparation of essentially weightless sources.

Fig 8

2.78 MeV α peak
of ^{212}Po .
5 cm E γ detector
used in Q.D.D.D.
20 minute count

← Here detector
was moved
5 cm to see
tail of peak.

← F.W.H.M. = 24 keV

Target Development for Princeton On-Line Isotope Separator (E. Ansaldo, J. Lind, R. A. Naumann, and I. Oelrich)

Liquid Target Systems

The liquid target system described in last year's progress report was designed to release radioactive isotopes of inert gases. The short-lived isotope 1.5 second argon-35 formed in a volatile liquid target of dichloroethylene (Boiling Point 84°C) by the reaction $^{35}\text{A} (\text{p},\text{n}) ^{35}\text{Ar}$ was dispersed through the Princeton on-line mass separator in on-line experiments. This demonstrated that short-lived activities can be released from a liquid during bombardment and delivered to the separator ion source at sufficiently low pressure to permit its operation.

In attempts to prepare more neutron deficient and even shorter-lived argon isotopes, reactions such as $^{32}\text{S} (^4\text{He}, 2\text{n}) ^{34}\text{Ar}$ and $^{32}\text{S} (^3\text{He}, 2\text{n}) ^{33}\text{Ar}$ have been attempted. The volatile liquid dimethyl disulfide, $(\text{CH}_3)_2\text{S}_2$ Boiling Point 117°C , has been used. With this liquid target 0.8 second ^{34}Ar was observed but no 0.17 second ^{33}Ar was detected. Presumably the ^{33}Ar has such a small production yield that no evidence for its production was obtained even using a solid state detector to search for the known beta delayed protons. In addition to the malodorous property of dimethyl disulfide, a further disadvantage of this liquid target was radiation decomposition. Significant quantities of hydrogen were liberated from the organic target during bombardment; when stable argon was introduced into the ion source the stable mass spectrum showed an additional peak at mass 41, the molecular ion ArH^+ , in addition to the mass 40 peak ^{40}Ar . The intensities of these two mass peaks were comparable.

To overcome this disadvantage, carbon disulfide, CS_2 Boiling Point 46°C , has been used. Special ventilation equipment was installed near the target to

minimize build up of explosive mixtures of this vapor with air. The carbon disulfide target has yielded the highest levels of argon-34 observed to date, however again no argon-33 could be detected. The results of our experiences using volatile liquid targets in an on-line separator system is being written up for publication.

Solid Target Systems

The solid target system in use at the Princeton isotope separator consists of a resistance heated target tube connected to the rear plate of the ion source. A less volatile solid target is contained at the end of the target tube behind a thin foil window of tantalum or tungsten which admits the cyclotron beam. Radioactive isotopes of elements volatile at the target tube temperature pass to the ion source of the separator.

In our initial design, changing the radioactive target tubes after bombardment entailed significant radiation exposure to personnel. The tubes and ion source were threaded and considerable close handling was required during both connection and disconnection. The threaded connection method has been replaced by a friction fit. The end of both the target tube and the boron nitride back plate of the separator ion source have been tapered. Target tube change over can now be rapidly accomplished. In addition the target tubes have been equipped with gas inlets so that chemically reactive gases can be introduced near the target during bombardment.

Higher temperatures of the target tube have been realized by adding a second resistance winding to the lavite heating form which surrounds the target tube. The target tube can now be reliably operated at temperatures as high as 1100°C as measured by an optical pyrometer.

We have been attempting to develop selenium and tellurium targets for the on-line separator system. The first targets employed were copper and zinc

selenides and tellurides. These however proved unsatisfactory under the heated bombardment conditions since the compounds decomposed too rapidly. Aluminum selenide and aluminum telluride have proved to be more stable targets. From bombardment of a natural tellurium target with alpha particles we have observed a number of xenon isotopes including the short-lived isomers 57 second xenon 125m and 72 second xenon 127m.

Tin dioxide has been used as a target and has yielded tellurium activities in on-line tests. Similarly a heated on-line target using a mixture of palladium and graphite has yielded cadmium activities when bombarded with alpha particles.

Spallation of Copper by 3.9 GeV ^{14}N and 3.9 GeV Protons (J. B. Cumming, P. E. Haustein, R. W. Stoenner (Brookhaven National Laboratory) L. Mausner, R. A. Naumann (Princeton University))

Our analysis of the comparative reaction yields of products when copper is irradiated with 3.9 GeV nitrogen ions and protons is complete and has been accepted for publication in the August, 1974 Physical Review. The close similarity in the yield distribution are shown in the accompanying charge dispersion yield curves. The upper curve and points refer to the nitrogen ion induced reactions, the lower curve refers to the proton data. The similarity in relative yield is apparent.

Figure 7

Beta Decay of ^{60}Zn (J. Lind, E. Ansaldi, G. Garvey and R. A. Naumann)

The isotope separator has been used "on-line" to study the beta decay of 2.3 min ^{60}Zn . This isotope has the interesting property of having an energetically allowed decay from its 0^+ , $T=0$ ground state to the 0^+ , $T=2$ state of ^{60}Cu at 2.5 MeV. Such a $\Delta T=2$ beta decay is forbidden by isospin selection rules. The relevant levels in ^{60}Cu are shown in fig. 1. The main features of the ^{60}Zn beta decay have been measured by others.¹ The de-excitation of the 2-5 MeV level is known and the three gamma transitions are indicated along with their approximate relative intensities. We searched for evidence of the 1866 keV line from this decay.

The ^{60}Zn activity was produced via the $^{58}\text{Ni}(^3\text{He},n)^{60}\text{Zn}$ reaction. A target consisting of a mixture of very fine mesh natural nickel and graphite powders was bombarded with 44 MeV ^3He . The powder was held in a heated tube which was attached to the end of the separator's ion source. Volatile zinc activity rapidly diffused from the target powder into the separator. The activity was collected for two minute periods on a moving tape transport system and then moved in front of a 35cc Ge(Li) detector for two minutes. During the first minute of this counting period the spectrum was routed into one 2048 channel array and during the second minute it was routed into another. This procedure generated two spectra showing the time evolution of the activity. Sample spectra are shown in fig. 2. The only significant contamination is the 24 minute ^{60}Cu daughter.

From the decay of peaks in the gamma spectra we can identify several very small, previously unreported gamma transitions at 1260, 1841 and 1986 keV. We calculate branching ratios for these transitions of 6.9×10^{-4} , 1.2×10^{-3} , and 2.3×10^{-4} respectively. We propose that the 1270 keV gamma de-excites a known level of ^{60}Cu at 1930 keV by cascading through the 670 keV level; that the

1841 keV gamma de-excites a level at 2175 keV by cascading through the 334 keV level; and that the 1986 keV gamma is a ground state transition de-exciting a known level at 1986 keV. If these decay schemes are correct they correspond to log ft values of 6.0, 5.3 and 6.4 respectively. A log ft of 5.3 is unusually low for a Gamow Teller beta decay to a state with such higher excitation as 2175 keV and may warrant further study.

In searching for evidence of a transition to the 2.5 MeV T=2 state we were hampered by a number of factors. The energy of this state is only known to plus or minus 15 keV. The most prominent feature of the spectrum in this region is the 1862 keV transition in ^{60}Ni following the beta decay of the daughter ^{60}Cu . However the area under this peak scales with the area of other prominent peaks from the decay of ^{60}Cu by the known ratios of intensity. From the size of the other peaks we know what size this peak should be and can eliminate it. We find a limit to the beta decay branching ratio from ^{60}Zn to the T=2 state of ^{60}Cu of 7.9×10^{-5} . This corresponds to a log ft of 5.7. This limit is above the level required to test the theoretically expected isospin mixing. A more accurate knowledge of the energy of the T=2 state in ^{60}Cu as well as further suppression of gamma background due to the annihilation in flight of high energy positrons is desirable.

1. Dulfer et al. Z. Physik 251, 416 (1972)

Fig 1 ^{60}Zn decay scheme

60ZN GAMMA SPECTRUM FIRST ARRAY

60ZN GAMMA SPECTRUM SECOND ARRAY

Publications and Abstracts

- (1) Upper Limit of an Isotensor Beta Decay in ^{60}Zn , J. M. Lind, E. J. Ansaldi, G. T. Garvey and R. A. Naumann, Bull. Am. Phys. Soc. Ser II, 19, 90 (1974) paper JF 3
- (2) Observation of the E3 Isomer of ^{93}Y , V. R. Casella, J. D. Knight, and R. A. Naumann, Bull. Am. Phys. Soc. Ser II, 19, 473 (1974) paper DG 8
- (3) High Spin States in ^{155}Dy and ^{154}Dy from $(^{12}\text{C}, \text{xn}, \gamma)$ Studies, K. Krien, R. A. Naumann, J. O. Rasmussen, and I. Rezanka, Nucl. Phys. A209, 572 (1973).
- (4) The Princeton On-Line Separator, R. A. Naumann, Proceedings of the 8th International EMIS Conference, Chalmers and Gothenberg Universities, August, 1973.
- (5) Spallation of Cu by 3.9-geV ^{14}N ions and 3.9-geV Protons, J. B. Cummings, P. E. Haustern, R. W. Stoenner, L. Mausner and R. A. Naumann, In Press August 1973 Physical Review C.

U. S. Atomic Energy Commission

Proposed Budget

October 1, 1974 - September 30, 1975

SALARIES

Prof. R. A. Naumann,	(2 mths. Summer Full Time)	\$
Principal Investigator	(No charge Acad. Yr.)	
Research Associate	(12 mths. Full Time)	
Technician F. Loeser	(12 mths. Full Time)	
Graduate Students	(3) (2 mths. Summer Full Time)	
	(2) ($\frac{1}{2}$ Time Acad. Yr.)	

TOTAL SALARIES \$ 36,065

EMPLOYEE BENEFITS @ 22.5% of non-student salaries 6,090

Tuition @ \$2400/student/year 4,800

INDIRECT COSTS @ 85% of total salaries 30,655

TOTAL SALARY RELATED COSTS \$ 77,610

CAPITAL EQUIPMENT 5,000

MATERIALS, EXPERIMENTAL SUPPLIES AND SHOP SERVICES 4,990

COMPUTER USAGE 1,000

TRAVEL 3,000

PUBLICATIONS 400

TOTAL ONE (1) YEAR PROPOSED BUDGET \$ 92,000