NUCLEAR CHEMISTRY PROGRESS REPORT  
Contract No. E(11-1)-2184

During the past fifteen months we have continued work in all three of our main areas of interest: (I) investigations of the chemical effects accompanying mu meson capture in atoms and molecules; (II) quadrupole interaction in metal and semimetal systems using perturbed gamma ray angular correlation; and (III) nuclear structure research using nuclear reaction spectroscopy.

(I) Investigations of the Chemical Effects Accompanying mu Meson Capture in Atoms and Molecules

The meson research has been exclusively carried out at the stopping muon channel of the Los Alamos Scientific Laboratory LAMPF Accelerator; the research there (experiment 60) has been a collaborative effort with members of the Los Alamos staff led by Dr. Jere Knight who is the spokesman for the experiment. The first paper of this collaboration has now been published in Physical Review A (COO-2184-22). Another paper in this area has been accepted for publication by the same journal. Since the "Great Shutdown" at LAMPF, experiment 60 has resumed in the Spring of 1976. Several striking new results from this work were communicated to the LAMPF Program Advisory Committee when they met in August of this year. Fig. 1 shows a striking variation in the intensity of the K series of muonic X-rays observed for metals as a function of atomic-number.

The research work at LAMPF involves travel by those of us participating in experiments at Los Alamos. During the last fifteen months, Dr. Gerhart Schmidt has made three trips of several weeks each and Robert Naumann also made four trips; the last consisting of a two month stay at Los Alamos during July and August. During this period, Robert Naumann defended LAMPF proposal 288 (attachment 1). Studies of Mu-minus Capture Probabilities for Solid State Solutions; the successor to experiment 60. The LAMPF Program Advisory Committee allocated
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150 hours for this experiment during the next experimental cycle probably beginning in February 1976 (attachment 2).

(II) Quadrupole Interaction in Metal and Semimetal Systems using Perturbed Gamma Ray Angular Correlation

The perturbed gamma angular correlation studies are the doctoral thesis research topic of Mr. Steven Lis, a graduate student in the Department of Chemistry.

As often occurs, the last year of research has been the most fruitful. Mr. Lis' study of intermetallic compounds has resulted in four reports (C00-2184-26, 27, 28, 29) which are in process of publication and two or three further articles will be prepared for publication. Mr. Lis has now ceased his experimental work and is devoting himself to the preparation of his doctoral thesis. During the summer months a quite novel experimental result appears to have been recognized: The quadrupole field gradient environment of an implanted atom located in a hexagonal rare earth metallic host changes from an axially asymmetric field to an axially symmetric field with increasing temperature. This has been observed for a cadmium atom (metallic radius 1.46Å) in five rare earth metals: Gadolinium, Terbium, Dysprosium, Holmium, Erbium (respective metallic radii 1.79, 1.77, 1.77, 1.76, 1.75Å). We tentatively attribute these results to a libration of the smaller cadmium atom in a cavity formed by the larger rare earth host atoms as the temperature is raised.

During the past year we have been attracted by a newly developing experimental technique for studying samples under high pressures of up to one million atmospheres - the diamond anvil press. It is our ultimate hope to inject small samples of metallic hosts (Ca.1 microgram) with carrier free amounts of

1 Physics Today 29, No 9, 44 (1976).
2.8 day indium 111 using our isotope separator. The sample would then be compressed using the diamond anvil pressure and the variation of field gradient of a substitutional cadmium atom in the metallic host measured as a function of pressure.

A diamond anvil press was supplied to us by Professor William Bassett of the University of Rochester Geology Department in March 1976. Since then Dr. Gerhart Schmidt of our group has been gaining experience in the delicate techniques involved in applying pressures and carrying out X-ray diffraction studies of the very small samples involved.

(III) Nuclear Structure Research using Nuclear Reaction Spectroscopy

The experimental nuclear structure research has been carried out at the Q3D spectrometer at the Princeton cyclotron and the Los Alamos tandem accelerator. At Los Alamos we have been collaborating with Dr. Ed Flynn of P I Division and Professor J. Kraushaar of the University of Colorado Physics Department.

This experimental program has involved \((p, t)\) and \((t, p)\) reaction spectroscopy. The interest in this research derives from the ability of these two reactions (which strip or add a pair of neutrons) to selectively populate collective states. Our interest has been a comparison of the stripping reaction observed for an odd proton nucleus with ground state spin \(1/2\) with the analogous results for an immediately adjacent even-even nucleus. The extent to which the stripping strengths and stripping strength ratios can be related for the even and odd targets tests the core coupling nuclear model in which the odd proton is considered as a spectator nucleon weakly coupled to an even-even core structure. The latter gives rise to the collective properties and participates in the two nucleon transfer reactions. Cases where the odd proton has spin \(1/2^+\) are particularly convenient since core coupling here results in
doublet pairs with spins \( (L \pm \frac{1}{2}) \) or singlets with spin \( \frac{1}{2} \) related to the collective states with spin \( L \) or 0 in the even-even nucleus.

During the last year we have extended these studies to the case of spin \( \frac{1}{2} \) yttrium 89 in a study of the \( ^{89}_{\text{Y}}(p,t)^{87}_{\text{Y}} \) which has just been published in the Physical Review (C00-2184-24). For this target with a 50 neutron closed shell the pattern of states observed is more complicated than in our comparison of the \((p,t)\) reactions in rhodium, palladium, silver. Here the first excited \( 2^+ \) state in the even-even strontium 86 lies considerably higher at 1.08 MeV; in yttrium 87 the expected \( 3/2^- \) and \( 5/2^- \) core coupled states admix with shell model proton states \( p_{3/2} \) and \( f_{5/2} \) with the same spin and parity. Thus in \( ^{87}_{\text{Y}} \) four states are observed near 1 MeV, two each with spin parity \( 3/2^- \) and \( 5/2^- \). The relative strengths observed in the \((p,t)\) reaction reflect the collective and single particle parentage of these states.

At the Los Alamos tandem the following \((t,p)\) spectroscopic studies have been carried out which complement the earlier Princeton \((p,t)\) studies:

\[ ^{103}_{\text{Rh}}(t,p)^{105}_{\text{Rh}}; ^{107}_{\text{Ag}}(t,p)^{109}_{\text{Ag}}; ^{109}_{\text{Ag}}(t,p)^{111}_{\text{Ag}}; ^{106}_{\text{Pd}}(t,p)^{108}_{\text{Pd}}; ^{108}_{\text{Pd}}(t,p)^{110}_{\text{Pd}}; ^{110}_{\text{Pd}}(t,p)^{112}_{\text{Pd}}. \]

The results of this investigation are in process of publication in Physical Review C (C00-2184-32). It is probably fair to characterize these investigations as providing the best examples of extended core coupling behavior so far observed.

The above \((p,t)\) and \((t,p)\) studies will constitute the doctoral thesis of Mr. Ivan Oelrich, a graduate student in the Department of Chemistry. During this summer Mr. Oelrich has been a summer student with the Radiochemistry Division of the Lawrence Livermore Laboratory where he has been working with staff members there on further charged particle nuclear spectroscopy research. Mr. Oelrich will remain at the Livermore Laboratory through the fall to undertake theoretical calculations with Dr. Gordon Struble related to his thesis research.
He will then return to Princeton late this year to prepare and defend his doctoral thesis.

At Los Alamos together with Dr. Ed Flynn we have also been investigating the spectroscopy of lead 207 using the $^{205}\text{Pb}(t,p)^{207}\text{Pb}$ reaction. This study using the Los Alamos Q3D spectrometer and tritons accelerated at the Van de Graaf accelerator employs targets of 30 million year lead 205 prepared and isotope separated at the Oak Ridge National Laboratory. Preliminary examination of the data indicates we have identified the monopole pairing vibrational state in lead 207 coupled to the $f_{5/2}$ neutron hole state of lead 205. This data is being presently analyzed.

Mr. David Allred, a graduate student enrolled in the joint Physics and Physical Chemistry program offered by the Physics and Chemistry Departments at Princeton University has almost completed the writing of his doctoral thesis. In the late fall he will join Oak Ridge National Laboratory on an N.S.F. post-doctoral fellowship in the area of energy research. Mr. Allred's thesis topic is an investigation of rotational-vibrational coupling. Our objective is to clarify some fundamental questions underlying the Variable Moment of Inertial Nuclear Model. This model seeks to simply parameterize the ground state "yrast" collective sequence for a broad range of nuclei and has been advocated by physicists at the Brookhaven National Laboratory.

Mr. Allred has been considering the quantization of a single particle moving in central force potentials having the form $V = kr^s$ where $s$ varies between 2 and 23. He has found a convenient method for deriving quite accurate eigenvalues for the problem using third order JWKB quantization conditions. His model calculations suggest that zero point vibrational effects are important and deserve consideration in semi empirical treatments of nuclei following VMI-type prescriptions.
Finally, Mr. Fred Girshick, a new graduate student in the Chemistry Department joined the nuclear chemistry group in April 1976 to undertake his doctoral research with us. At this time we learned that radioactivity was being produced in pure hydrogen-1 plasma experiments in components of the Princeton Large Torus Tokomak fusion reactor. The radioactivity occurred in a tungsten limiter used to skim heavy atom impurities from the plasma.

Using the Germanium Lithium spectrometer in our laboratory we have been able to definitively identify $^{182}$Ta, $^{181}$W and $^{185}$W products produced by $(ee',p)$; $(ee',n)$; and/or $(\gamma,p)$ or $(\gamma,n)$ reactions initiated in the diverter by energetic runaway electrons accelerated during the discharge. The gamma ray spectrum is shown in fig. 2. A copy of an article reporting these findings is being published together with staff members of the Plasma Physics Laboratory (C00-2184-35). The following page contains our publications for the last contract period.
PUBLICATION LIST

COO-2184-20 Evidence for Core-Coupled States in $^{87}$Y from a $^{87}$Y(p,t)$^{87}$Y and $^{88}$Sr(p,t)$^{86}$Sr Comparison, I. C. Oelrich, K. Krien, R. M. DelVecchio, and R. A. Naumann, Phys. Rev. C14, 563 (1976)


COO-2184-26 Temperature Dependence Study of the Quadrupole Interaction of $^{111}$Cd in Tellurium, S. A. Lis and R. A. Naumann, (Submitted to Hyperfine Interactions)

COO-2184-27 Temperature Dependence Study of the Quadrupole Interaction of $^{111}$Cd in Antimony, S. A. Lis and R. A. Naumann, (Submitted to Hyperfine Interactions)

COO-2184-28 TDPAC Studies of $^{111}$Cd in InBi and In$_2$Bi, S. A. Lis and R. A. Naumann (Submitted to Journal of Physics F: Metal Physics)

COO-2184-29 TDPAC Study of $^{111}$Cd in Ag, Cd, Ag, Al, and Ag, Zn, S. A. Lis and R. A. Naumann, (Submitted to Journal of Physics F: Metal Physics)


| C00-2184-35 | Photo-Neutron Production in the PLT Tokamak, J. D. Strachan, E. B. Meservey, W. Stodiek, R. A. Naumann, and F. Girshick, (to be submitted to Nuclear Fusion Journal) |
**Present Research Staff**

<table>
<thead>
<tr>
<th>Name</th>
<th>Position</th>
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</thead>
<tbody>
<tr>
<td>Professor R. A. Naumann</td>
<td>Principal Investigator</td>
</tr>
<tr>
<td>Dr. Gerhart Schmidt</td>
<td>Research Associate</td>
</tr>
<tr>
<td>Mr. Steven Lis</td>
<td>Completing graduate work early 1977</td>
</tr>
<tr>
<td>Mr. Ivan Oelrich</td>
<td>Completing graduate work early 1977</td>
</tr>
<tr>
<td>Mr. David Allred</td>
<td>Completing graduate work late 1976</td>
</tr>
<tr>
<td>Mr. Fred Girshick</td>
<td>Second Year Graduate Student</td>
</tr>
<tr>
<td>Mr. Fred Loeser</td>
<td>Research Staff</td>
</tr>
</tbody>
</table>
Fig. 1

μ− LYMAN X-RAY INTENSITY PATTERNS FOR PURE ELEMENTS (METALS)

Kα 2p-1s

Kα 2p-1s

Kβ=3p-1s

Kγ=4p-1s

LAMPF EXP. 60

LBL 184" CYCLOTRON

∑∞

N=8

2p-1s

TARGET Z
GAMMA RAY SPECTRUM FROM RADIOACTIVITY IN THE
PRINCETON LARGE TORUS LIMITER

COUNTS / CHANNEL

CHANNEL NUMBER
LAMPF EXPERIMENT PROPOSAL

Title: Studies of Mu-minus Capture Probabilities for Solid State Solutions

Spokesman: R. A. Naumann, Princeton University

Participants: J. D. Knight, L.A.S.L.
L. Mausner, L.A.S.L.
R. A. Naumann, Princeton University
C. Orth, L.A.S.L.
M. Schillaci, L.A.S.L.
G. Schmidt, Princeton University

Summary: Muon capture X-ray spectra will be recorded for five single phase systems of variable composition containing two or three elements. Relative muon capture probabilities will be determined from the summed elemental K X-ray intensities observed for alloy and ionic crystal samples.

The objective is to quantitatively test:
(1) the linearity of muon capture ratio with composition.
(2) the invariance of the individual elemental X-ray spectra to composition.

The experiment is intended to enhance knowledge of negative muon capture - cascade processes in atomic systems. The results relate directly to the use of muons for elemental analysis.

Beam Area: Area A, Stopping Muon Channel

List of Targets: (atomic/molecular percent shown)

Aluminum/Copper: 100% Cu, 95% Cu, 85% Cu, 33% Cu, 100% Al
Silver/Zinc: 100% Ag, 95% Ag, 85% Ag, 25% Ag, 100% Zn
Sodium Bromide/Sodium Iodide: 100% NaBr, 95% NaBr, 50% NaBr, 5% NaBr, 100% NaI
Potassium Bromide/Sodium Bromide: 100% KBr, 95% KBr, 50% KBr, 5% KBr
Potassium Iodide/Sodium Iodide: 100% KI, 95% KI, 50% KI, 5% KI

Running Time Requested: 200 hours at minimum proton beam intensity 20 microamperes or greater. (23 targets at 8 hours apiece + 16 hours beam tuning/contingency)

Scheduling Requirements: Would prefer to receive the beam time in approximately three 72 hour periods, each period spaced by approximately one week. Early summer (July) 1977 would be most convenient period.
For binary chemical compounds of form $Z'Z_m$, the relative muon capture probability is estimated by the Fermi Teller $Z$-law (1) as $\frac{W(Z)}{W(Z')} = \frac{mZ}{kZ'}$, where $Z$ and $Z'$ are the atomic numbers of the constituent elements. Rough verification of this rule has been claimed for inert gas mixtures, (2) binary metallic alloys (3) and for ionic salts (3). If such a variation, which neglects details of the valence electron structure of $Z$ and $Z'$, were indeed found to be valid, one of two conditions governing the capture of a negative meson by an atom would be indicated. Either (1) the meson passes through the entire capture energy range while it remains essentially localized within the region of one atom or (2) the majority of the meson capture events take place at sufficiently high meson energies that the detailed chemical nature of the stopping medium (valence electron properties) is unimportant, and the capture process is statistical.

A measurement of the relative muon capture ratio for one binary chemical compound, one alloy, or one mixture of fixed composition cannot provide an adequate test of the predicted composition linearity of the capture ratio. For such a test, single phases of variable composition are required. In a two component system e.g. a single phase alloy composed of metals A and B, the two limiting situations where component B or component A is very dilute, are of especial interest. Here the slowing of the muon is principally determined by the major component (here A or B respectively). High statistical accuracy measurement of the total intensity of the mesic X-rays of the two components and the relative intensity patterns will display any differences between the A dominated or B dominated stopping processes.

To show that variation in capture probability is occasioned by chemical bonding we cite from the extensive data of Zinov (3) per atom capture ratios for the antimony oxides $\text{Sb}_2\text{O}_3$ and $\text{Sb}_2\text{O}_5$:

<table>
<thead>
<tr>
<th></th>
<th>$\text{Sb}_2\text{O}_3$</th>
<th>$\text{Sb}_2\text{O}_5$</th>
</tr>
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<tbody>
<tr>
<td>$A$</td>
<td>$3.48 \pm 0.35$</td>
<td>$1.73 \pm 0.09$</td>
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</table>

Z law prediction $= \frac{51}{8} = 6.4$

As an example illustrating that the mesic X-ray intensity pattern varies for a given element in different chemical states, the results of the Chicago group (4)
may be given for the titanium muonic K X-ray intensities accompanying muon stops in titanium metal or titanium dioxide.

Table II

<table>
<thead>
<tr>
<th></th>
<th>$K_{\alpha}$</th>
<th>$K_{\beta}$</th>
<th>$K_{\gamma}$</th>
<th>$K_{\delta}$</th>
<th>$K_{\epsilon}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ti</td>
<td>1</td>
<td>0.102(3)</td>
<td>0.031(4)</td>
<td>0.036(3)</td>
<td>0.041(3)</td>
</tr>
<tr>
<td>TiO₂</td>
<td>1</td>
<td>0.093(6)</td>
<td>0.027(5)</td>
<td>0.025(5)</td>
<td>0.028(5)</td>
</tr>
</tbody>
</table>

The large effects on the cascade process caused by variations of muon angular momentum have recently been strikingly demonstrated by the experiments of the Daniel group\(^5\) and the subsequent theoretical analysis\(^6\). Table III shows the intensity patterns for the fluorine muonic K X-rays for muons stopping in pure gaseous sulfur hexafluoride and in hydrogen gas with sulfur hexafluoride as a minor component.

Table III

<table>
<thead>
<tr>
<th></th>
<th>$K_{\alpha}$</th>
<th>$K_{\beta}$</th>
<th>$K_{\gamma}$</th>
<th>$K_{\delta}$</th>
<th>$K_{\epsilon}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>SF₆</td>
<td>1</td>
<td>0.18(3)</td>
<td>0.15(3)</td>
<td>0.080(15)</td>
<td>0.050(11)</td>
</tr>
<tr>
<td>SF₆/H₂ = 2 x 10⁻⁴</td>
<td>1</td>
<td>0.51(8)</td>
<td>0.75(9)</td>
<td>0.46(7)</td>
<td>0.19(4)</td>
</tr>
</tbody>
</table>

As the data and theory verified, Coulomb transfer of the muon from the ground state of the proton-muon atomic system with low angular momentum is signalled by a strong enhancement of the higher members of the K X-ray series.

The evidence that the chemical environment does modify the capture processes of slowed negative mesons challenges the concept of an invariant per atom capture ratio for two elements and motivates this proposal. We note the technique of studying muon capture by dilute components in solutions has been previously
introduced in experiments at Los Alamos\(^7\) on aqueous systems. Here, however, the unique processes occurring in hydrogen containing systems require special interpretation.\(^8\)

We turn now to a discussion of some features of the systems chosen for study. The two component alloy systems each have single phases, stable at room temperature, which persist over a range of compositions\(^9\). In both alloys the two metallic atoms have comparable atomic radius \((r_{\text{Al}} = 1.25 \text{ Å}, r_{\text{Cu}} = 1.17 \text{ Å} \text{ and } r_{\text{Ag}} = 1.33 \text{ Å} \text{ and } r_{\text{Zn}} = 1.25 \text{ Å})\). In each case an atom of lower ionization potential is paired with another of higher ionization potential (I.P. \(^{13}\text{Al} = 5.98 \text{ ev}, ^{29}\text{Cu} = 7.72 \text{ ev} \text{ and } ^{47}\text{Ag} = 7.57 \text{ ev}, ^{30}\text{Zn} = 9.39 \text{ ev})\). The relation of atomic number and ionization potential however are reversed among the two pairs. Both metallic alloy systems possess mobile conduction electrons, charging after-effects due to auger ionization effects should be minimized.

The three pairs of ionic salts form continuous solid solutions which are stable at room temperature\(^10\). The radii of the ions involved vary markedly as do the ionization potentials.

\[
\begin{array}{c}
r_{\text{Na}^+} = 0.95 \text{ Å}, r_{\text{K}^+} = 1.33 \text{ Å}, r_{\text{Br}^-} = 1.95 \text{ Å}, r_{\text{I}^-} = 2.16 \text{ Å} \\
\text{I.P.} \quad \text{Na}^+ = 47.3 \text{ ev}, \text{K}^+ = 31.8 \text{ ev}, \text{Br}^- = 3.54 \text{ ev} \\
\quad \text{I}^- = 3.24 \text{ ev}
\end{array}
\]

On the basis of both size and ionization potential one may surmise that the anions would dominate the energy loss processes for muons of low energy. The comparison of the muon capture ratios sodium to potassium for the systems NaBr/KBr and NaI/KI will directly reveal any effect of varying the negative ion on relative capture. If there is no effect of the counter ion, the same sodium to potassium capture ratio will be observed for the two systems at the same atomic concentrations.

From previous experience at the LAMPF stopping muon channel, 8 hours of observation time per sample will afford spectra with sufficient statistical precision to enable capture ratios to be compared with a precision of ± 2%.
References

(4) Kessler, Anderson, Dixit, Evans, McKee, Hargrove, Barton, Hincks, McAndrew,
    (1976).
(8) LAMFF experiment 60 unpublished results.
(10) Phase Diagrams for Ceramists, The American Ceramic Society (1964) Supplement
August 25, 1976

Professor Robert A. Naumann  
Princeton University  
Department of Physics  
Joseph Henry Laboratories  
Jadwin Hall, P.O. Box 708  
Princeton, NJ 08540

Dear Professor Naumann:

On behalf of the LAMPF Program Advisory Committee, may I thank you for the time and effort devoted to the preparation and presentation of proposal No. 288, entitled, "Studies of Mu-minus Capture Probabilities for Solid State Solutions," by Professor R. A. Naumann, Spokesman. The summary and the discussion it engendered was most helpful to those who must propose priorities for the extensive LAMPF research program and recommend the allocation of resources.

I am pleased to inform you that 150 hours have been granted for carrying out your research program. The scheduling of your work will take place in reasonable sequence after #60 is completed.

You will be contacted by Don Hagerman, Chief of Operations, on the scheduling of your experiment. Where necessary, an engineering review will be held. All safety matters should be handled through Tom Putnam and Lew Agnew should be contacted for experimental floor support.

Best wishes on the success of your research program.

Sincerely yours,

Louis Rosen  
Director, LAMPF

LR/rg

cc: PAC