Progress Report Submitted to

U. S. DEPARTMENT OF ENERGY

by

MASTER

Princeton University

NUCLEAR CHEMISTRY PROJECT

Contract EY-76-S-02-2184

January 1, 1979 - December 31, 1979

December 21, 1979

DISCLAIMER

This book was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

REA

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

PROGRESS REPORT
TO THE
UNITED STATES DEPARTMENT OF ENERGY

Over the past year we have continued our research in three main areas of interest a) Coulomb capture of negative muons by atoms and molecules b) nuclear structure research c) use of the Princeton electromagnetic isotope separator to isolate radioactive isotopes for radioactive spectroscopy and optical hyperfine spectroscopy.

Our research effort has been very significantly enhanced by the arrival of Dr. Konrad Krien from the University of Bonn in West Germany who has joined our group as research physicist. Dr. Krien who has extensive experience in nuclear spectroscopy and nuclear hyperfine effects has proposed and initiated on line experiments at the Princeton cyclotron to determine the quadrupole moments of nuclear excited states.

During the past year the first on-line operation of the Princeton electron spectrometer was achieved and its capability for fast life time measurements demonstrated. The first operation of the heavy ion source was achieved and beams of 75 MeV $^{12}\text{C}^{+4}$ ions and 37 MeV $^6\text{Li}^{+2}$ ions demonstrated.

The analysis of LAMPF experiment 288 - the study muon capture by components in solid state solutions - was completed during a two month summer stay by Naumann at Los Alamos. The paper completed during the summer which reports this research has been accepted by Physical Review A for publication.

Data taking is complete for LAMPF experiment 374, the study of muon capture probabilities by alkali metal halides, group II metal oxides, sulfides and the compounds boron phosphide, aluminum nitride and aluminum phosphide. The data analysis for the alkali metal halides and the group II metal sulfides is now complete, but the muon capture data for the oxides and the three compounds is still in process of reduction.

Finally the capability of the isotope separator to provide pure mass separated samples of radioisotopes is being utilized for three different applications. We have studied the very weak electron capture branch of 19 sec krypton 81m to bromine 81, a naturally occurring isotope. This transition probability is of crucial importance for a possible solar neutrino detection method based on natural bromine. Lead 202, a 3×10^5 year neutron deficient specie, has been prepared and mass separated for optical hyperfine spectroscopic studies. The isotope shift which measures the second moment, $\langle r^2 \rangle$, of the nuclear charge indicates the more neutron deficient lead isotopes are becoming increasingly soft toward nuclear deformation. Finally the isotope separator is furnishing mass separated samples 1.67 second Neon 18. In a joint project with members of the cyclotron group, the positron decay from the 0^+ ground state of this isotope to a 0^- state in fluorine 18 is being searched for. The extent to which this pseudoscalar transition proceeds, ordinarily highly forbidden, is expected to be sensitive to mesonic exchange effects in the nucleus and hence is of great interest.

AN ON-LINE ORANGE SPECTROMETER FOR IN-BEAM CONVERSION ELECTRON STUDIES

(F. Girshick, K. Krien, R.A. Naumann)

The Orange conversion electron spectrometer has become operational on-line during the past year. A major problem addressed during this time is the elimination of background in the scintillation detection system of the spectrometer, when the cyclotron beam hits the target. This background is due to delta rays produced in the target as well as neutrons and gamma rays from both the target and the beam stop. In order to differentiate these effects time-of-flight techniques are necessary. However, the timing characteristics of the original electron detection system (two EMI 9536 B photomultiplier tubes viewing the same scintillator and operating in coincidence in order to reduce the multiplier noise) were found inadequate for such fast timing or to measure new lifetimes down into the nanosecond region in future studies.

Since the delta rays produced in the target during in-beam runs limit us, as expected, to electron energies above ~ 80 keV in most cases, we designed and installed a new detection system using only one photomultiplier tube with much improved time properties. The new system consists of a thin shell of Pilot B scintillator (also to reduce the detection efficiency for neutrons and γ rays) a short Lucite light pipe, a RCA 8575 phototube and an Ortec 256 phototube base. The improved timing capability was tested by measuring a time spectrum relative to the natural beam burst of the cyclotron. With a ^{197}Au target and exciting states in ^{196}Hg via a $(p, 2n)$ reaction we obtained a time resolution of $\text{FWHM} = 1.6$ nsec for the 496 keV K-conversion electrons. A similar test with the delta rays at much lower energies resulted in a prompt time curve of $\text{FWHM} = 3$ nsec. This increase in FWHM is due to a number

of effects at these lower energies, most importantly, the increase in spread of transit time of the conversion electrons through the spectrometer.

Using the improved timing capability for time-of-flight particle identification and also making use of the beam pulser of the cyclotron to detect delayed particles within up to ~ 400 nsec time ranges, we have identified background contributions due to γ rays and fast and slow neutrons from the beam dump as well as the target. The beam dump shielding has been improved; additional lead shielding greatly reduced the number of gamma rays reaching the detector. Paraffin surrounded by borax is to be used to first thermalize and then capture the neutrons. Another lead shield installed near the spectrometer chamber stops the gamma rays from neutron capture.

The background which originates in the target of the electron spectrometer is harder to eliminate. The large lead shield which originally surrounded the scintillator was removed to reduce the production of neutron capture gamma rays close to the detector. Instead a much smaller, but equally efficient conically-shaped "shadow shield" has been installed between target and the detector. These procedures have reduced the total background by a factor of approximately 30.

To further reduce the background effects, we have taken spectra for small electron-momentum regions of interest by setting tight windows on the pulse height spectrum from the photomultiplier tube thus suppressing events due to low energy scattered delta rays, photomultiplier noise and high energy gamma rays. As an example see Figure 1, where a comparison of the two modes of operation for the 253 keV K- and 203 L-conversion lines in ^{211}At (see below)

FIG. 1

is presented. The two spectra displayed have been acquired simultaneously and therefore directly show background reduction by about a factor of 10.

While we improve the performance of the spectrometer (e.g., obtain better momentum resolution by adding baffles) we have started our conversion electron spectroscopy research. In an effort to extend the systematics of the E3 transition probabilities between the $(g_{9/2})^3_{7/2^+}$ and $p_{1/2}$ -spherical shell model states in the odd-A silver isotopes known for $^{101}\text{Ag} \rightarrow ^{111}\text{Ag}$ to ^{99}Ag , we have first remeasured the respective lifetime in ^{101}Ag . The states in ^{101}Ag were excited by the $^{102}\text{Pd}(p,2n)^{101}\text{Ag}$ reaction. The $1/2^-$ isomeric state in ^{101}Ag decays by a 176 keV E3 transition to the $7/2^+$ state at 98 keV. A conversion electron spectrum showing the K + L lines of this transition is shown in Figure 2.

FIG. 2

Setting the spectrometer current so that the 176 keV L-conversion electrons are detected, we followed the decay of the conversion electron counting rate in the multiscaling mode, for a period of 50 seconds after the cyclotron beam is shut off. As a result we obtained the time-spectrum shown in Fig. 3. From this the half life of the $1/2^-$ isomer is deduced to be $T_{1/2}(1/2^- \text{ }^{101}\text{Ag}) = 3.45 \text{ sec}$ in agreement with previous^{1,2)} investigations.

An attempt to measure the corresponding isomeric half life in ^{99}Ag making use of the $^{102}\text{Pd}(p,4n)^{99}\text{Ag}$ reaction failed, because of the many other reaction channels ($p, p', 3n$, etc.) open at these very neutron deficient nuclei at much higher cross sections. In order to circumvent this problem we are attempting to populate the excited states in ^{99}Ag via the $^{102}\text{Pd}(^3\text{He}, p5n)^{99}\text{Ag}$ reaction.

FIG. 3

A second experiment involves the isotope ^{211}At . In ^{211}At at excitation energies between 1.12 MeV and 1.417 MeV a series of states are known with proposed spins between $11/2^-$ to $21/2^-$, two of which exhibit lifetimes of 50 nsec and 13 nsec, respectively. These states are due to the different couplings of three $h9/2^-$ protons outside the doubly magic ^{208}Pb . They therefore are considered to be rather pure spherical shell model configurations and afford a good opportunity to test specific predictions for such configurations (see section III c).

To observe the conversion electrons arising via decays from these states, we made use of the reaction $^{209}\text{Bi}(\alpha, 2n)^{211}\text{At}$. The target consisted of $\sim 175 \mu\text{g}/\text{cm}^2$ of Bi evaporated on approximately $2 \text{ mg}/\text{cm}^2$ Be. This type of target was also chosen to prove that such layered targets, which are

needed for our future quadrupole interaction studies are actually suitable for conversion electron studies. A conversion electron spectrum obtained without background suppression by setting an energy-window on the multiplier pulse height spectrum but selecting only events delayed relative to the cyclotron beam burst making use of the fast timing capability of the orange spectrometer is shown in Figure 4. Delayed K, L, and M-

FIG. 4

conversion electrons, deexciting those levels of interest in ^{211}At are observed. We also measured the lifetimes for several of the observed conversion lines making once again use of the cyclotron beam pulser system. As an example, a raw time spectrum obtained for the 96 keV L-conversion line in ^{211}At is shown in Figure 5.

References

- 1) S.I. Hayakawa, I.R. Hyman and F.K.P. Lee, Nucl. Phys. A296, 251 (1978).
- 2) V.R. Casella, F.D. Knight and R.A. Naumann, Nucl. Phys. A239, 83 (1975).

FIG. 5

ACCELERATION OF HEAVY ION BEAMS IN THE PRINCETON CYCLOTRON

During the past year heavy ion beams have been accelerated in the Princeton Cyclotron for the first time. The heavy ion source construction follows the design developed at the Michigan State University Cyclotron and employs a cold cathode discharge struck between tantalum electrodes. The 7 kilovolt-7 ampere fast current regulated arc power supply has been contributed by the Lawrence Livermore Laboratory as part of a continuing cooperative program of nuclear structure research.

The first heavy ion accelerated was $76 \text{ MeV } \text{C}^{+4}$. The source gas was carbon monoxide and a current of 0.4 microamperes was accelerated to the full radius of the cyclotron. To verify that carbon ions had indeed been accelerated, an irradiation of natural palladium foil ($Z = 46$) was carried out in the 0° bombardment facility. The production of tellurium radioisotopes ($Z = 52$) was confirmed by recording sequential gamma spectra after bombardment. Figure 1 shows a typical gamma spectrum and decay curves. Transitions from the decay of ^{115}Te , ^{116}Te , ^{117}Te decaying with the correct half lives confirm the acceleration of the carbon ions.

The second heavy ion accelerated was $^6\text{Li}^{+2}$. The heavy ion source was loaded with a lithium 6 fluoride pellet and the discharge maintained with neon gas. The lithium ions were formed by secondary bombardment with the neon ions and a 300 nanoampere beam of $37 \text{ MeV } ^6\text{Li}^{+2}$ ions was accelerated to the full radius of the cyclotron. In this case, a foil of monoisotopic niobium 93 ($Z = 41$) was bombarded and the production of ruthenium isotopes ($Z = 44$) was confirmed. Figure 2 shows a typical gamma spectrum recorded from the irradiated niobium. The observation of gamma radiations following the decay of ^{95}Ru (see insert of Fig. 2) confirms that the acceleration of lithium 6 predominantly causes the reaction:

The large excitation of the known $21/2 + {}^{93\text{m}}\text{Mo}$ isomer (also confirmed by its 6.9 hour decay) indicates the capability of lithium induced reactions to excite high spin isomers. The availability of these heavy ion beams widens the opportunities for nuclear structure studies at the Princeton Cyclotron both using the Q3D spectrograph for particle transfer studies and for in beam spectroscopic studies using the Orange electron spectrometer and Ge(Li) gamma ray spectrometers.

Of particular interest to us are isomer high spin states lying off the stability line. The possibility of imparting high recoil energies by means of these heavy ion reactions to these isomeric nuclei enables their implantation into solid substrates for subsequent measurement of their nuclear moments (see sec. I).

FIG. 1: Gamma spectrum obtained after bombardment of natural Pd with 76 MeV $^{12}\text{C}^{4+}$ -beam.
 INSERT: Decay curves for prominent lines in $^{115}, ^{116}, ^{117}\text{Te}$

FIG. 2: Gamma spectrum obtained after bombardment of ^{93}Nb with 37 MeV $^6\text{Li}^{2+}$ -beam.

INSERT: Decay curves for prominent lines in ^{95}Ru .

THE ELECTRON CAPTURE BRANCH OF 13 SECOND KRYPTON 81m; THE DETECTION OF SOLAR NEUTRINOS BY BROMINE 81

C.L. Bennett, M.M. Lowry, R.A. Naumann, F. Loeser, and W.H. Moore

The low number of solar neutrinos detected by Ray Davis using the $^{37}\text{Cl} \xrightarrow{(\nu, e^-)} ^{37}\text{Ar}$ reaction has prompted interest in investigating other nuclear reactions which may be driven by solar neutrinos. R. D. Scott¹⁾ in an article published in "Nature" during 1976 proposed to measure solar neutrinos by the amount of ^{81}Kr ($T_{1/2} = 200,000$ years) produced in natural bromine deposits by the reaction $^{81}\text{Br} \xrightarrow{(\nu, e^-)} ^{81}\text{Kr}$. The ^{81}Kr could be detected by isolating a Krypton sample from a natural bromine deposit and counting the single ions at mass 81. Such single ion counting has been demonstrated by the detection of carbon 14 at lower levels in natural carbon than can be achieved by direct counting of the beta radioactivity of the sample. For the selective single ion counting, a van de Graaff accelerator, magnetic deflection and particle identification counting is used as a highly selective mass spectrometer. This permits the traversal of a single ion from the source to the detector to be registered.

Bromine 81 (49% abundant) has a ground state spin parity, $3/2^-$. The solar neutrino reaction populates the second excited state of Krypton 81, an isomeric state with spin parity $1/2^-$ lying 190 kilovolts above the $7/2^+$ ground state of Krypton 81. The half life of this isomer is 13 seconds. The $1/2^-$ Krypton 81 decays predominantly by an E3 transition to the $7/2^+$ ground state. The neutrino threshold for the $^{81}\text{Br} \rightarrow ^{81m}\text{Kr}$ reaction is 0.512 MeV as compared to 0.814 MeV for the $^{37}\text{Cl} \rightarrow ^{37}\text{Ar}$ reaction. Unlike the chlorine reaction, which involves two $T = 1/2$ mirror

states, ^{81m}Br decay involves initial and final states for which the beta interaction matrix element cannot be predicted with confidence. Accordingly, this matrix element must be measured by experiment before serious consideration can be given to the Bromine 81 neutrino detection method.

A cooperative effort utilizing the isotope separator mounted on-line at the Princeton cyclotron was undertaken. A heated target of potassium bromide - graphite was bombarded with 22 MeV protons. This target attached to the isotope separator ion source provided microcurie isotopically separated sources of the 13 second isomer ^{81m}Kr at the collector of the isotope separator. These were deposited onto the moving tape receiver mounted after the isotope separator.

The electron capture decay of the isomer, $^{81m}\text{Kr} \rightarrow ^{81}\text{Br}$, involving the beta matrix element of interest are signalled by the emission of a Bromine K x ray from the Bromine atom formed with a vacancy by the electron capture process. A highly selective detection scheme for these 11.0 keV bromine K x rays is necessary since the Krypton 81m predominantly decays by the highly converted 190 keV E3 isomeric transition to the ground state of Krypton 81. This results in the emission of 12.6 keV krypton x rays with an intensity approximately ten thousand times greater than those of the bromine x rays.

The scheme used for the measurement is shown in Fig. 1. The moving tape collector transports consecutively collected Krypton 81m sources to a counting station where all x rays are recorded for a total of 36 seconds in four sequential periods of 9 seconds each by a Silicon-Lithium photon detector having a resolution of 250 keV. At this position, the moving tape also passes between two thin pilot B electron scintillation

detectors which provided simultaneously both coincident and anti-coincident events with the x ray detector. The 177 keV K conversion electrons signal the isomeric decay of Krypton 81m; the x ray spectrum recorded in anti-coincidence with these electrons thus attenuates the Krypton K x ray photons while not affecting the bromine K x ray photons. The x ray spectrum recorded in coincidence with the electron detectors corresponds to the simple Krypton x ray spectrum with Compton scattering from the 190 keV gamma ray strongly suppressed.

The detection of the bromine K x rays was accomplished by establishing very accurately the Krypton K x ray line shape from the coincidence spectrum. This line shape was subtracted from the anti-coincidence spectrum to reveal the bromine K x rays with correct energies. The decay of these lines was consistent with the expected 13 second half life.

During 24 hours of observation 890 ± 210 bromine K_{α} x rays and 4.7×10^6 krypton K_{α} x rays were detected at the mass 81 position. After correcting for the conversion coefficient for krypton and bromine, one obtains the branching ratio for K capture to all decays for krypton 81m of $(5.64 \pm 1.38) \times 10^{-5}$. The corresponding logft value is 4.88 ± 0.11 .

With this provisional logft value, the detection of solar neutrinos using natural bromine deposits appears quite possible. Assuming a logft 5, Kirsten¹⁾ has estimated that 1000 tons of sylvinites would yield 3.4×10^5 atoms of krypton 81 and 10^{19} atoms of natural krypton. With this ratio the krypton 81 can be detected using the accelerator mass spectrometer technique.

References

- 1) R.D. Scott, Nature 264, 730 (1976).
- 2) R. Kirsten, Proc. of Informal Conf. on Solar Neutrino Res., BNL 50879 (1978).

FIG. 1: Isotope Separation and Study of Krypton-81m

ISOTOPE SEPARATION AND OPTICAL HYPERFINE SPECTROSCOPY OF LEAD 200 AND LEAD 202

F. Loeser, R.A. Naumann, and Henry Stroke, Olav Redi, New York University

During 1978, a two week irradiation of natural thallium metal was carried out at the Brookhaven Linear Accelerator Isotope Production facility BLIP. The bombardment with a 100 microampere beam lasted for two weeks and produced several microgram quantities of 3×10^5 year Lead 202 by the $^{203}\text{Tl}(p, 2n)$ and $^{205}\text{Tl}(p, 4n)$ reactions. The target has now been chemically fractionated using anion exchange chromatography and the lead fraction containing the lead 202 isolated. This lead fraction has been mass separated using the Princeton isotope separator in order to provide enriched samples of lead 202 for investigation of the isotope shift of this nuclide.

The optical spectrum of the enriched sample of lead 202 was recorded photographically and photoelectrically using the grating spectrograph operated by Professor Henry Stroke's group at New York University.

Figure 1 compares the line spectrum photographically recorded from both the enriched lead 202 sample and a natural lead sample. The presence of the lead 202 fine structure line is definite. By comparing the optical shift measured for the isotopes lead 208, 206, 204, and 202, one finds an abnormally large decrease in the isotope shift between lead 204 and lead 202. This in turn indicates that the mean square radius $\langle r^2 \rangle$ for lead 202 has significantly increased. For this sequence of isotopes neutron pairs are being successively removed from the doubly magic and spherical closed shell nucleus lead 208. The rapid increase in $\langle r^2 \rangle$ for lead 202 is best interpreted in terms of an increasing softness of the lighter lead nuclei toward nuclear deformation resulting in larger zero point amplitudes.

This experimental research will be extended to 21.5 hour lead 200. This isotope will be produced by the $^{203}\text{Tl}(p,4n)$ reaction at the Princeton cyclotron. Chemical fractionation of the bombarded target will be immediately followed by mass separation and transportation of the enriched lead 200 sample to New York for recording the optical spectra.

In connection with these experiments an extensive effort has been made to reduce the background levels of natural lead contamination arising in the isotope separation step and collector foils. This program has significantly reduced the lead background making it possible to attempt the lead 200 experiment involving very small quantities of radioisotope where obscuring effects from natural lead are particularly significant.

FIG. 1: Hyperfine Optical Spectra for Lead Isotopes ($2833\text{\AA}$ line).

Research Staff

Professor R. A. Naumann

Dr. Konrad Krien

Mr. Fred Girshick

Mr. Fred Loeser

Principal Investigator

Research Physicist (Aug. 1979 -)

Fifth Year Graduate Student

Research Staff

Publication List

"Temperature Dependence Studies of the Electric Field Gradient at Cadmium Impurities in Heavy Rare Earth Metals", S. A. Lis, R. A. Naumann, and G. Schmidt, Hyperfine Interactions 5, 431 (1978).

"Temperature Dependence Studies of the EFG at ^{111}Cd Probes Located in Some Intermetallic Compounds", S. A. Lis, R. A. Naumann, and G. Schmidt, Hyperfine Interactions 5, 445 (1978).

"Multiplet Structure in ^{203}Pb ", R. A. Naumann, L. Dinescu, and D. Mueller, Phys. Rev. C18, 2506 (1978).

"Coulomb Capture of Negative Muons in Solid Solutions", R. A. Naumann, G. Schmidt, J.D. Knight, L.F. Mausner, C.J. Orth, and M. Schillaci, Bull. Am. Phys. Soc. 24, 26 (1979).

"Nuclear Spin of 3×10^7 -y ^{205}Pb ", F. Moscatelli, R.L. Wiggins, R.A. Naumann, O. Redi, and H.H. Stroke, Bull. Am. Phys. Soc. 24, 17 (1979).

"Constant Negative Muon Capture Ratios for Simple Ionic Crystals", H. Daniel and R.A. Naumann, Abstract for the Hawaii Meeting of the Amer. Chem. Soc., April 2, 1979. Abstract no. 71, Nuclear Chem. & Tech. Section.

"The Quadrupole Interaction of ^{100}Rh in Various Intermetallic Compounds of Palladium", R. Vianden, E.N. Kaufmann, G. Schmidt and R.A. Naumann, Hyperfine Interaction 7, 247 (1979).

"A Model Requiring Constant Negative Muon Coulomb Capture Ratios for Simple Ionic Crystals", R. A. Naumann and H. Daniel, Z. Physik A291, 33 (1979).

"Negative-muon Capture Ratios for Alloys and Alkali Halide Solid Solutions", G. Schmidt, J.D. Knight, L.F. Mausner, C.J. Orth, M.E. Schillaci, and R.A. Naumann, in press, Phys. Rev. A. (1979).

"Temperature Dependence of a Lattice Defect Induced Electric Field Gradient of ^{181}Ta in Pt", H.G. Muller, K. Krien, U. Putz, R. Feuschenbach, R. Trzcinski and K. Freitag, Z. Physik B32, 315 (1979).

Publication List (cont'd)

"The Magnetic Hyperfine Field and the Electric Field Gradient of ^{181}Ta Impurities in Gd Metal", K. Krien, M. Forker, F. Reuschenbach and R. Trzcinski, Hyperfine Interactions 7, 19 (1979).

"The Electric Field Gradients for Hg at Different Lattice Sites in Be", K. Krien, F. Reuschenbach, F.C. Soares, P. Herzog, H.-R. Falle, B. Perscheid, R. Trzcinski, K. Freitag and E.N. Kaufman, Hyperfine Interactions, in press.

"Isotope Shift Between 3×10^5 -y ^{202}Pb and ^{208}Pb ", R.A. Naumann, O. Redi, F. Moscatelli, R.L. Wiggins, and H.H. Stroke, Bull. Am. Phys. Soc. 24, 624 (1979).