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Nuclear physics and heavy element research at Lawrence Livermore National Laboratory

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Abstract. This paper highlights some of the current basic nuclear physics research at Lawrence Livermore National Laboratory (LLNL). The work at LLNL concentrates on investigating nuclei at the extremes. The Experimental Nuclear Physics Group performs research to improve our understanding of nuclei, nuclear reactions, nuclear decay processes and nuclear astrophysics; an expertise utilized for important laboratory national security programs and for world-class peer-reviewed basic research. The bibliography includes 42 references.

I. Introduction

It is fitting that we commemorate Dmitri Ivanovich Mendeleev on this occasion of the 175th anniversary of his birth and the 140th anniversary of his remarkable achievement of constructing the first periodic table of the chemical elements. In 1869, Mendeleev recognized the periodicity of the chemical properties of the known elements, constructed a tool to organize the elements based on that periodicity, and then used that tool to predict the existence of as yet undiscovered elements [see his presentation to the Russian Chemical Society called 'Dependence between the Properties of the Atomic Weights of the Elements (March 1869) and the textbook 'Principles of Chemistry' (1869 – 1871)]. The periodic table in a variety of different forms [1] is still used today to guide chemists and physicists studying the fundamental building blocks of our world. It is with great pleasure, and distinct honor, that we discuss the scientific efforts at LLNL in nuclear physics and heavy element research, and show how some of those efforts are shaped by understanding the periodicity of the chemical elements.

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The experimental nuclear physics effort at LLNL is centred on investigating nuclei at the extremes, in particular, extremes of spin, isospin, neutron richness, excitation energy, decay and detectability, mass and stability. Clearly, many of these areas are interrelated. This paper will discuss some examples of recent work in these extreme areas of nuclear physics. The work at LLNL is aimed to support the U.S. nuclear physics goals as indicated in the Nuclear Science Long Range Plan,[2] namely, to develop a comprehensive and unified description of nuclei, which requires nuclear data on exotic nuclei, to make use of existing and future facilities (such as the FRIB to be sited at Michigan State University) and to investigate neutrino properties and fundamental symmetries.

II. Main avenues of nuclear physics research at LLNL

1. Spin

We are involved in investigating shell structure effects when one increases the angular momentum of the nucleus — in other words, the evolution of shell structure with high spin. This work utilizes Coulomb-excitation reactions and transfer reactions at Argonne National Laboratory with the Gammasphere [3] high-purity Ge (HPGe) detector gamma-ray array and CHICO parallel plate avalanche counter particle detector.[4] A ^{242m}Am target was Coulomb excited using a 170.5 MeV ^{40}Ar beam at ATLAS and the resultant deexcitation gamma rays that were detected with Gammasphere are shown in Fig. 1. The level scheme constructed from coincident gamma-ray data is shown in Fig. 2. As can be observed from the level scheme and gamma-ray spectrum, the levels built on the $K = 5$ and $K = 6$ states are very strongly and nearly equally populated. Strong K-mixing results in nearly equal populations of the $K^{\pi} = 5^{-}$ and 6^{-} states. More details of the ^{242m}Am experiment can be found in Ref. 6.

Gammasphere and CHICO were also used for an experiment aimed at investigating high-spin states in Np isotopes. States in ^{235}Np were populated using the two-neutron transfer reaction ^{237}Np(^{116}Sn,^{118}Sn)^{235}Np. The ^{116}Sn beam energy of 801 MeV was chosen to be slightly above the Coulomb barrier and to maximize the one-neutron and two-neutron transfer reaction channels. As such, the rich data

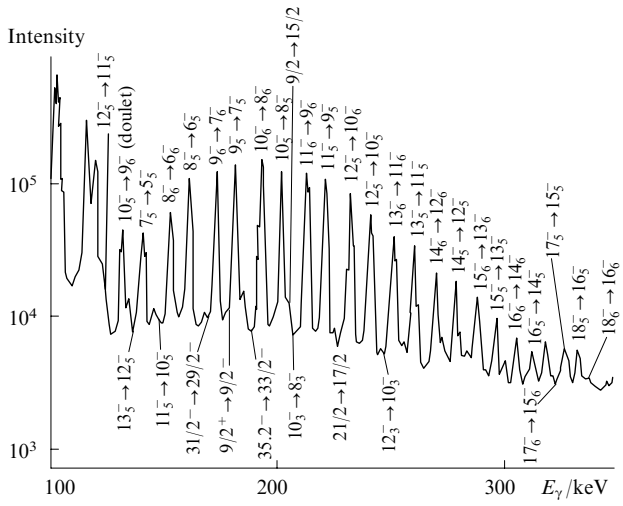


Figure 1. Gamma-ray energy spectrum with particle-gamma coincidence with known transitions labeled with the initial and final spin, parity and K values. States of half-integer spin belong to ^{241}Am target contaminants.

set also contains information on inelastic excitation of ^{237}Np and additional information on the ground state band from one neutron transfer to ^{236}Np. Both transfer reactions allow the study of rotational alignments in Np nuclei. The observed gamma-ray energy spectrum and partial level scheme for ^{235}Np are shown in Fig. 3. More details of the ^{237}Np experiment can be found in Ref. 7.

2. Isospin

We are involved in investigating shell-structure effects when one adds additional neutrons to the nucleus — in other words the evolution of shell structure with N/Z. This work utilized radioactive ion beams from TRIUMF ISAC-II (see Ref. 8) and the TIGRESS/BAMBINO detector setup[9] as shown in Fig. 4. The low-lying nuclear states in ^{29}Na were investigated using Coulomb excitation of a 70 MeV beam of ^{29}Na impinging on a ^{110}Pd target. The HPGe clover detectors of TIGRESS detected deexcitation gamma rays in coincidence with scattered particles which were detected in the segmented silicon detectors of BAMBINO. The observed gamma-ray spectrum is shown in Fig. 5. The reduced transition matrix element for the transition from the first excited state to the ground state in ^{29}Na was measured to be 0.237(21) eb [corresponding $B(E2) \sim 18(3)$ W.u.] and indicates a significant admixture of both sdand pf components in the wave function. More details about this experiment may be found in Ref. 10.

3. Astrophysics

We are involved in experiments to improve our understanding of stellar evolution and nucleosynthesis by performing critical cross section measurements using novel techniques. A principle observable in supernova remnants is the γ-ray decay of ^{44}Ti. The amount of ^{44}Ti produced in supernovae is an indicator of the internal dynamics of supernova evolution. The ^{40} Ca$(\alpha,\gamma)^{44}$Ti reaction rate has recently been measured by two groups whose reaction rates disagree with each other by factors of 3 to 5.[11,12] At LLNL, we have used the tandem accelerator at the Center for Accelerator Mass Spectrometry (CAMS) to measure the ^{40}Ca$(\alpha,\gamma)^{44}$Ti thick target yield in beam. One of the targets was analyzed as well with low background counting of the ^{44}Ti produced in this reaction. For the first time, we have determined the ^{40}Ca$(\alpha,\gamma)^{44}$Ti cross section in the 2–4 MeV region relevant to supernova physics in a self-consistent way. We find that the rec ent reported results overestimate the reaction rate by more than 35%. More details of this experiment may be found in Ref. 13.

In stellar evolution, we are studying nuclear branch point nuclei using the surrogate reaction technique. A nuclear branch point arises when the neutron capture rate

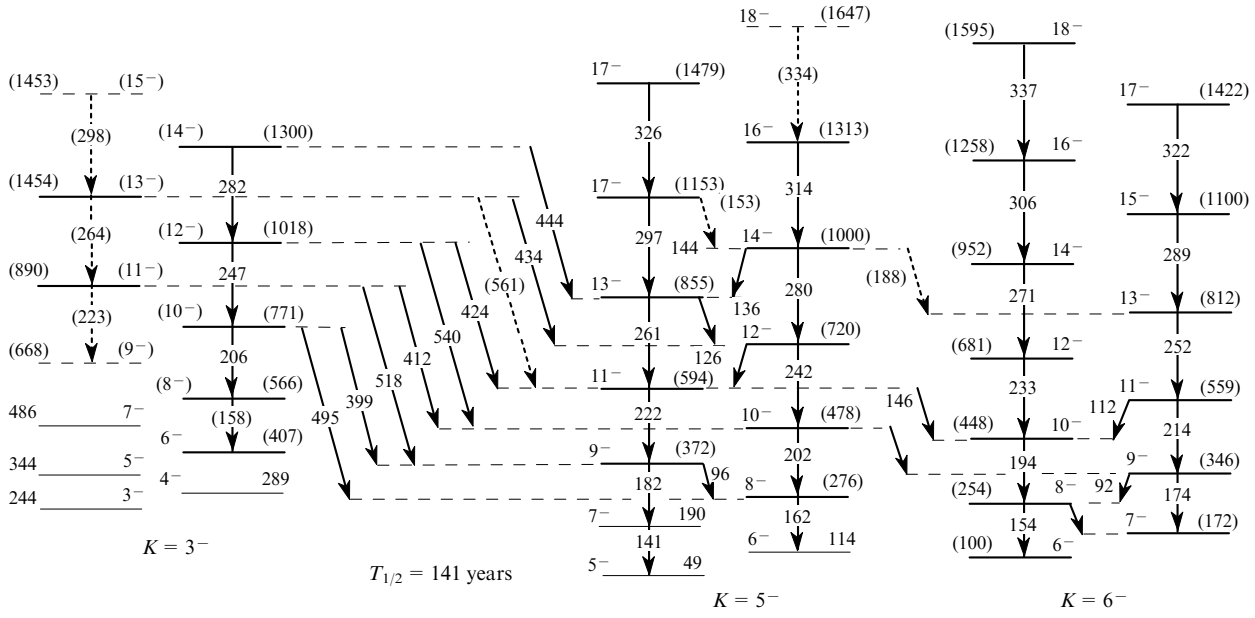


Figure 2. Partial level scheme for ^{242m}Am deduced from many gamma-ray coincidence spectra. Levels with bold lines are those observed in the Coulomb excitation experiment. Previously known states (thin lines) are from Ref. 5.

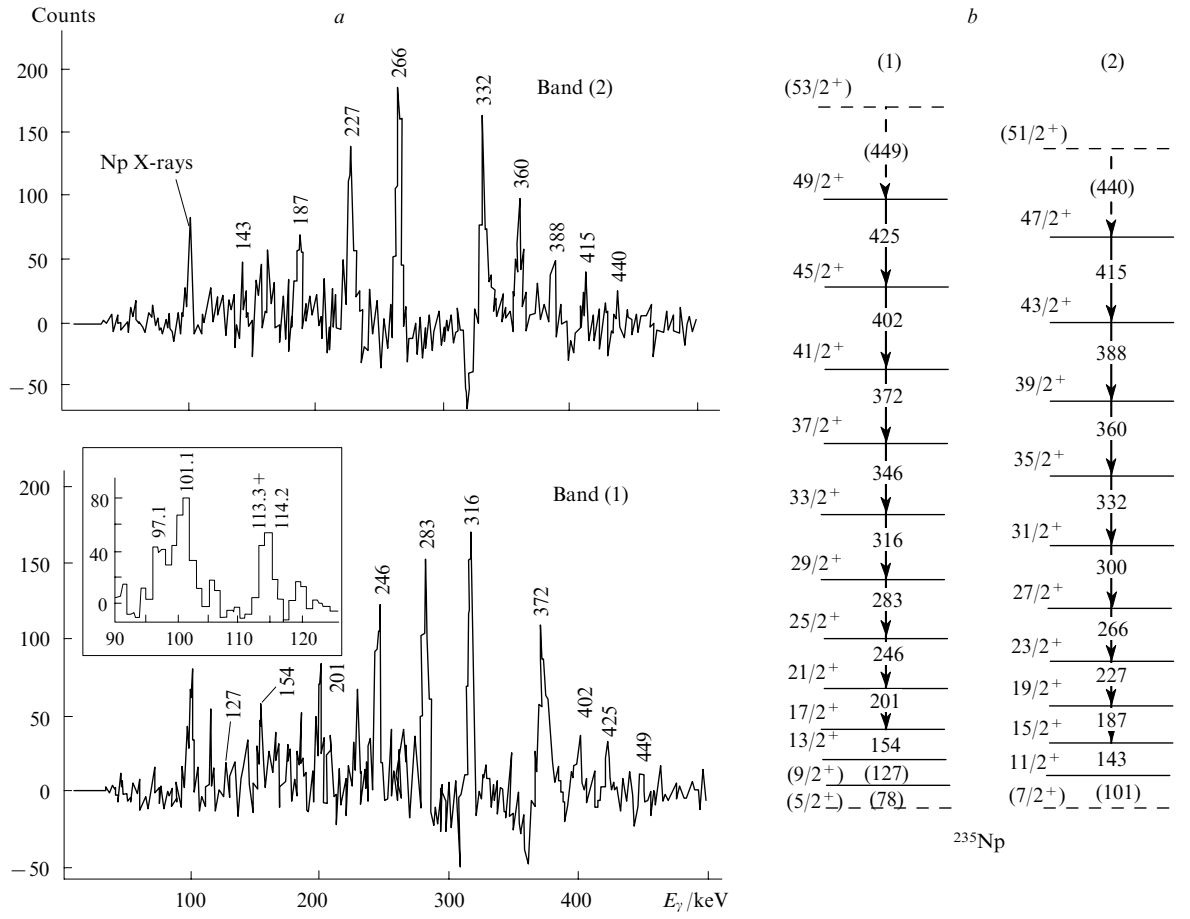


Figure 3. Gamma-ray energy spectrum gated on the two-neutron transfer reaction showing both signature partners in the ground band of ^{235}Np (*a*) and a proposed partial level scheme of ^{235}Np based on gamma-gamma coincidences (*b*).
On incut is part of spectrum on a large scale.

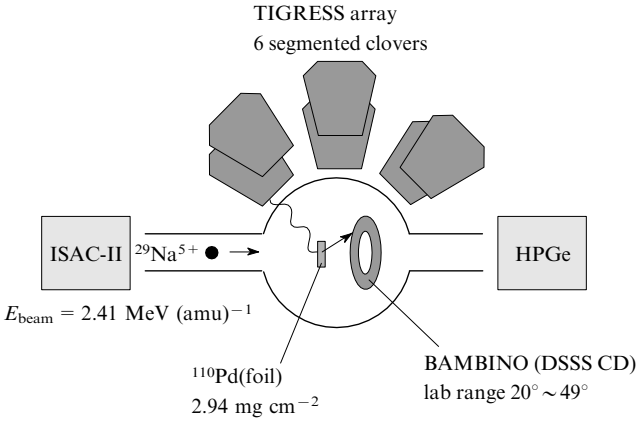


Figure 4. Schematic of the experimental setup for the ^{29}Na Coulomb excitation experiment.
Note that the beam intensity was on the order of 500 particles s^{-1}. DSSS CD is double sided silicon strip detector.

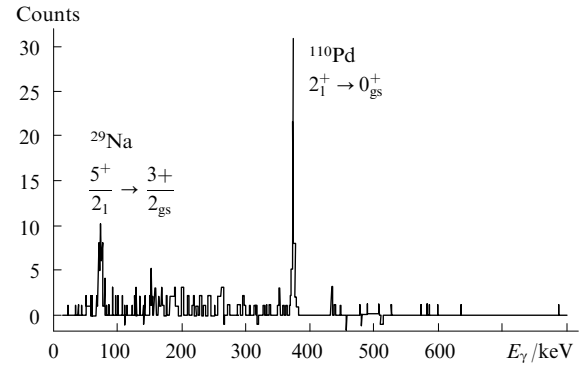


Figure 5. Particle-gamma-ray coincident, background subtracted gamma-ray energy spectrum following ~70 h beam on ^{110}Pd target showing the observed Coloumb excitation of the beam (^{29}Na) at 72 keV and target (^{110}Pd) at 374 keV; gs is ground state.

from the s-process competes with the β-decay rate of a given nucleus. In particular, we are measuring the ^{153}Gd(n,γ) branch point by performing the ^{154}Gd(p,p)^{154}Gd* surrogate reaction. A direct measurement is not possible as the ^{153}Gd half-life is 241.6 days and fabricating a target would result in a target activity of 3.5 Curies. We have benchmarked this approach using ^{156}Gd(p,p′) and ^{158}Gd(p,p′) as surrogates for the known ^{155}Gd(n,γ) and ^{157}Gd(n,γ) cross sections. For the first time, theory and experiment are being used to produce an absolute cross section in the (n,γ) energy

region below 1 MeV. More details about this experiment may be found in Ref. 14.

4. Neutron-richness

We are involved in the study of neutron-rich nuclei such as those produced in fission or at radioactive-ion beam facilities. Studies of prompt gamma-rays emitted from fission products in ^{252}Cf spontaneous fission has yielded a wealth of nuclear structure data,[15,16] data on the dynamics of fission,[17] and even information about ternary fission.[18] In-beam fission studies using $\alpha + ^{238}$U nuclear reactions provided a wealth of higher-spin data in the $A = 120$ region.[19] The determination of the excited states for ^{118,120}Pd has permitted considerable insight into the collective and non-collective behavior of Pd nuclei over a wide range of neutron numbers. Moreover, these data also reveal clear differences between collective and non-collective states as protons are removed from the Sn closed shell for $N = 72$ and $N = 74$ isotopes. New gamma-ray transitions in ^{120}Pd have been observed following alpha-particle induced fission of ^{238}U, building on the low-energy gamma rays previously identified in decay studies of ^{120}Rh. Transitions in ^{118}Pd were confirmed, reducing the discrepancy with prior work. A search for gamma-rays in ^{122}Pd was inconclusive. The systematics of neutron-rich isotopes of Pd indicates remarkable symmetry of the 2^+ levels in nuclei surrounding ^{114}Pd, which is somewhat maintained even to spins as high as 10^+. More details about this experiment may be found in Ref. 19.

The scientific interest in radioactive-ion beam facilities is prompted by understanding the r-process nucleosynthesis path and s-process nuclei in nuclear astrophysics. Recently, the next generation of radioactive ion beam facilities in the US, FRIB, was sited at Michigan State University, building on the projectile-fragmentation facility existing at the National Superconducting Cyclotron Facility. FRIB will provide beams of radioactive nuclei farther from stability than ever before, allowing exploration of nuclei residing in *terra incognita* between the valley of stability and the r-process pathway.

5. Excitation energy

We are involved in the study of nuclei at extreme excitation energy and in extreme environments. The recently completed National Ignition Facility (NIF) at LLNL is a 192-beam laser-driven inertial confinement fusion facility designed to produce ignition and energy gain by fusing deuterium and tritium nuclei together.[20] In addition to demonstrating a new source of energy, the facility will provide a laboratory for investigating plasma science, nuclear astrophysics and exotic nuclei in extreme environments. An indirect drive target is shown in Fig. 6. An igniting capsule produces 10^{19} 14 MeV neutrons in picoseconds, resulting in fluxes approaching those of supernovae, $\sim 10^{34}$ neutron cm^{-2} s^{-1}. Fluxes of lower energy neutrons are reduced but not insignificant. NIF will provide a unique environment for studying behaviour of nuclei in excited states. Reactions on excited states could provide insight into reactions on nuclei far from stability and investigate whether shell structure is quenched in these excited states.

There are several aspects of excited states in plasmas that we are exploring, namely, the effects of the population of excited states in a plasma on the network of reactions that proceed because of the neutrons, charged particles and photons in the burning plasma, and the mechanisms for

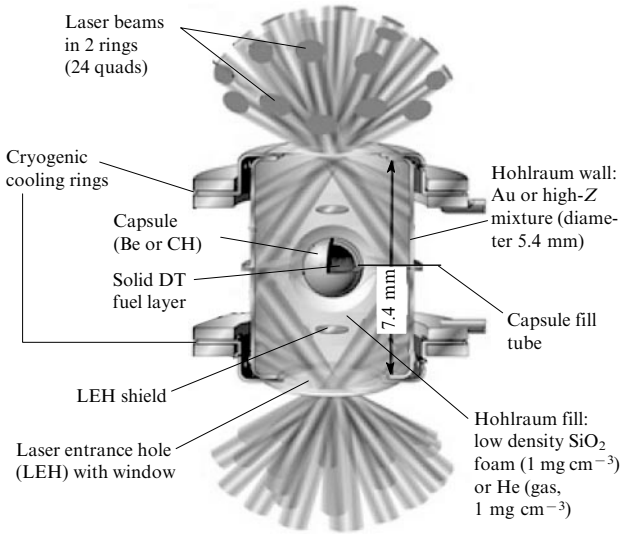


Figure 6. Schematic diagram of the point design NIF ignition capsule. Laser beams are directed into the ends of a cylindrical hohlraum, converted to X-rays, which then ablate the outer portion of the capsule shell. The remaining capsule shell and DT fuel is accelerated inward to a compression of approximately 30. The fuel is then at temperatures and pressures conducive to ignition and a hot spot in the very centre ignites the fuel.

population of excited states for nuclei residing in this plasma environment. An example of the first aspect is the study of reactions on ^{171}Tm nuclei doped in the ablators of NIF capsules in early NIF implosions. This offers the opportunity to study stellar (n,γ) processes in a near stellar-like high energy density environment created in a NIF implosion. Comparison of amounts of observed product nuclei with amounts produced in an accelerator experiment would elucidate differences between the vastly different environments. The s-process path near ^{171}Tm is shown in Fig. 7. Both ^{170}Tm and ^{171}Tm are s-process

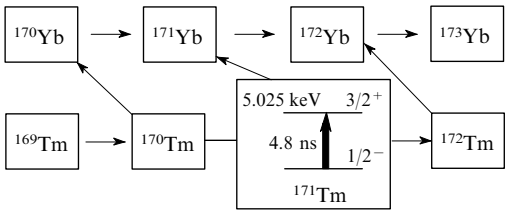


Figure 7. Section of chart of nuclides showing the *s*-process path near Tm.
Both ^{170}Tm and ^{171}Tm are s-process branching point nuclei. The low lying level scheme of ^{171}Tm is shown to highlight the presence of a 5 keV 'isomer' that could be populated *via* the NEEC mechanism discussed in the text.

branching points. Calculations have indicated that some (n,γ) reactions near s-process relevant nuclei are enhanced by 20%–40% (see Fig. 8)[21] due to excited states or 'isomers'. If we can obtain (n,γ) cross-sections in a stellar-like environment on these branch-point nuclei, they could then be used as 'thermometers' to determine interior stellar temperatures.

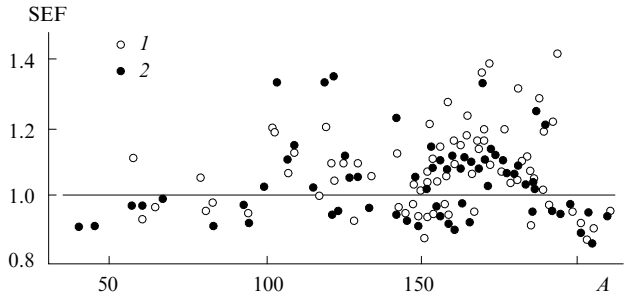


Figure 8. Figure from Ref. 17 showing the s-process enhancement factors (SEF) as a function of mass for a variety of nuclei.
Note in the $A = 170$ region that the enhancement factors are 1.2 – 1.4. (*1*) and (*2*) are data from various references.

The second aspect regarding the mechanism for population of the excited states in burning plasma involves exploration of inverse internal conversion processes known as nuclear excitation by electron capture (NEEC).[22] Again we use ^{171}Tm as an example. The high electron flux in the NIF plasma can populate the 5 keV 4.8 ns level in ^{171}Tm by this process as shown in Fig. 9. By varying the plasma conditions in NIF implosions, one can learn the population probability for these excited states in a stellar-like environment.

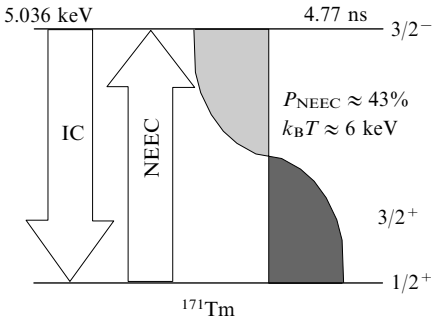


Figure 9. Low-lying level scheme of ^{171}Tm showing the internal conversion (IC) and nuclear excitation by electron capture (NEEC) processes connecting the 5-keV first-excited state to the ground state. Note the calculated probability for NEEC population of the first excited state at typical temperatures in NIF implosions of 43%.

Finally, as one increases the excitation energy of the nucleus near the neutron separation energy at the top of the nuclear potential well, the single particle levels are rearranged, which may change the influence of nuclear structure on reactions. Such a situation may provide insight into nuclear reactions with neutron-rich nuclei far from stability.

The performance of NIF capsules will be monitored with a variety of prompt diagnostics to measure neutron, X-ray and gamma-ray emission, and radiochemical diagnostics to measure mix, fuel and shell ρR and capsule asymmetries.[23] Radiochemical diagnostics are currently being developed based on doping NIF capsules with specific nuclides and then measuring the production of radionuclides following the implosion a nd all of the nuclear reactions occurring on the dopant. Current doping schemes favor insertion of the dopant into the inner part of the ablator just adjacent to the DT fuel (Fig. 10). The inner part of the ablator is not blown off during the compression phase of the implosion, so the dopant is ideally located at the interface between the shell and the fuel and very near the high flux region of the capsule. The amount of dopant put into the ablator is small enough to not perturb the implosion, on the order of $10^{14} - 10^{15}$ atoms. The dopant depends upon what the diagnostic is designed to measure and which nuclear reaction is being used. For example, to probe the amount of ablator that might be mixed into the fuel because of hydrodynamic instabilities during the implosion, a charged particle reaction might be used due to the shorter range of charged particles in the hot dense plasma. Because of the shorter range of charged particles, only when material is intimate with the burning fuel and hence the source o f charged particles, will copious reactions occur. Examples of such reactions include ^{127}I(d,2n)^{127}Xe, ^{79}Br(d,2n)^{79}Kr and ^{18}O(α,n)^{21}Ne, though since ^{21}Ne is a stable isotope, the latter may have background issues. These reactions produce noble gases which can be collected by pumping on the target chamber using the rapid automated gas sampling (RAGS) system designed for NIF (Fig. 11) following an implosion. Schemes for collection of solid samples are also under development.[24] Both sample collection methods will be important for not only diagnostics but also the science experiments highlighted above.

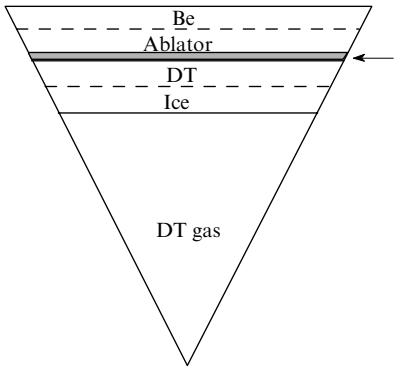


Figure 10. Schematic pie diagram of a NIF point design capsule indicating the location of the dopant in red and with the arrow.
Note that the ablator and DT ice regions of the capsule are not to scale and enlarged specifically to highlight dopant location.

6. Neutrino physics

We are involved in experiments to understand fundamental properties of the neutrino utilizing detection of rare decay processes in ultra-low background counting experiments. In order to answer questions of whether the neutrino mass scale has inverted or normal hierarchy, whether neutrinos are Majorana particles (*i.e.*, neutrinos are their own antiparticles, differing from antineutrinos only by helicity, see Fig. 12), and whether lepton number conservation is violated, searches for neutrinoless double-beta decay in ^{130}Te at the CUORICINO array of TeO_2 bolometers are continuing [25] at the Laboratori Nazionali del Gran Sasso in Italy. No evidence for this rare decay mode is observed in these experiments and a half-life limit $T_{1/2}(^{130}\text{Te}) \geqslant 3.0 \times 10^{24}$ years was set. In order to increase the sensitivity of the experiment, approximately 20 times more TeO_2 bolometers are being assembled into the CUORE array in Italy.

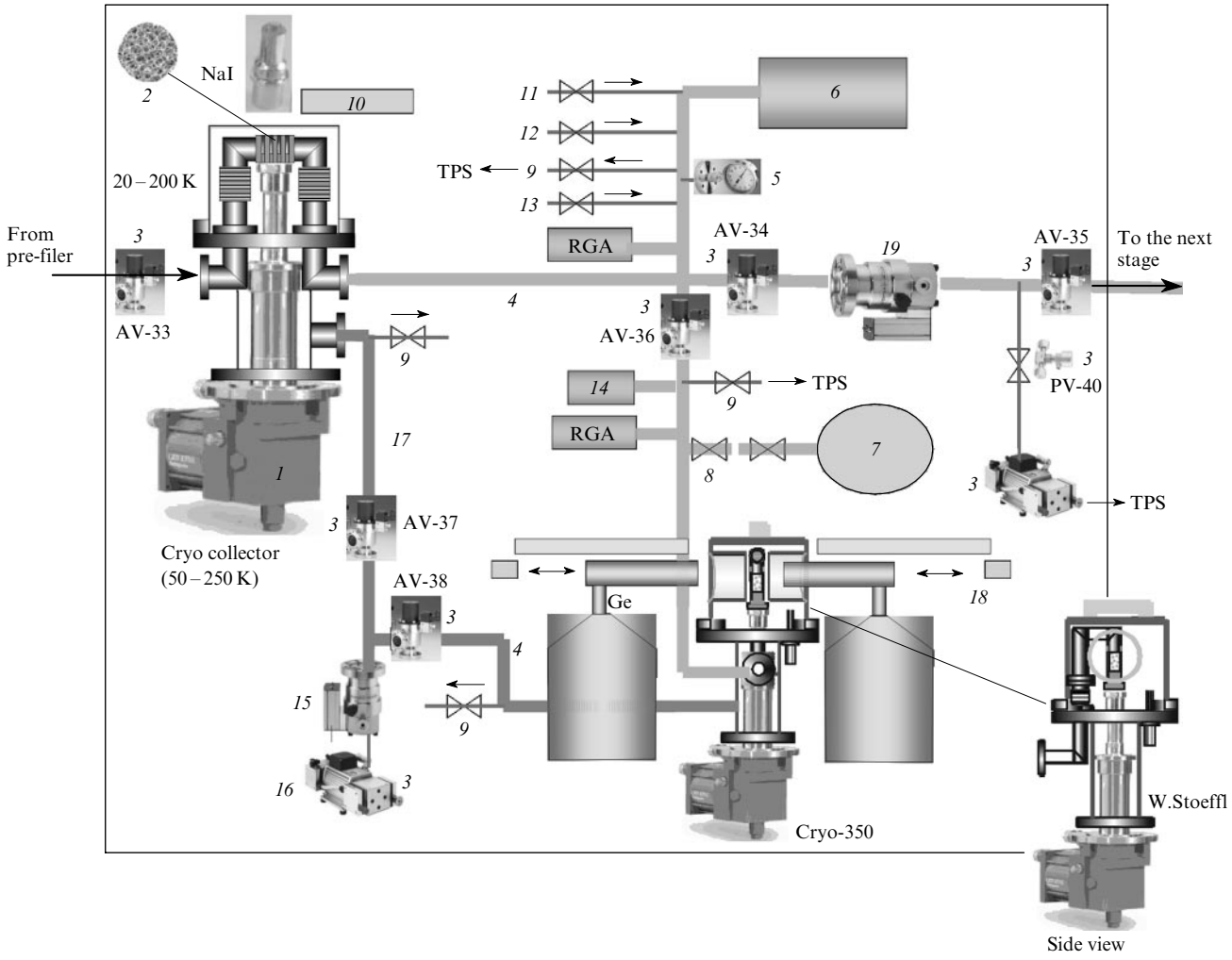


Figure 11. Schematic diagram of the Rapid Automated Gas Sampling (RAGS) system designed for NIF. The RAGS system relies on cryogenic separation and purification of noble gases and has 3 or 4 of these units each operating at temperatures necessary to collect different noble gases (shown is the Xe subsystem). A prefilter system (not shown) is used to remove unwanted water vapor and gases other than noble gases and resides between the target chamber and this unit. A helium gas puff can be injected into the system to maintain gas flow through RAGS.
Two-stage cryo head (*1*), copper foam filled tube (*2*), Xe traps (*3*), 1.5" tube (flex) (*4*), simple pressure gauge (*5*), additional equipment (*e.g.*, mass spectrometer) (*6*), removable trap (*7*), Nupro manual (*8*), relief (*9*), lead shield (*10*), He flush (*11*), N_2 flush (*12*), vent (*13*), cold cathode (*14*), turbo pump insulation vacuum (*15*), no TPS, it's clean (*16*), independent cryo insulation vacuum (*17*), overhead slide (*18*) and turbo pump (*19*). RGA is residual gas analyzer and TPS is tritium processing system.

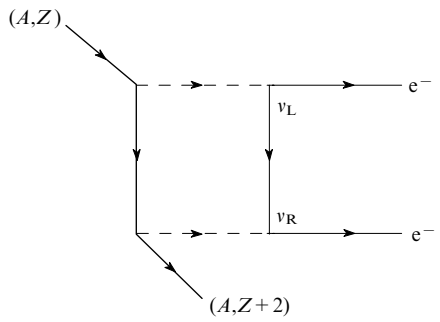


Figure 12. Feynman diagram showing the neutrinoless double-beta decay process.

7. Superheavy elements

One of the most productive collaborations over the last 25 years has been the Joint Institute for Nuclear Research (JINR, Dubna, Russia)/LLNL collaboration to investigate the nuclear and chemical properties of the heaviest elements. The Livermore Heavy Element group has a long and accomplished history of fundamental nuclear research, with spectroscopic, chemical and decay studies dating back to the 1950s. In the 1980s, two fission modes were discovered which competed in the spontaneous fission of several heavy actinide nuclides. This 'bimodal fission' decay challenged nuclear theory and resulted in fundamental changes in the way the fission barrier was modeled.[26,27] A collaboration between scientists at JINR and scientists at LLNL was established in 1989. In the early 1990s, this collaborative work resulted in the confirmation of a recently predicted region of nuclei that owe their extra stability to the nuclear shapes, the effect being strongest for nuclei near neutron number 162 and proton number 108.[28–30] In late 1998 and 1999, again in collaboration with our Russian colleagues, we performed experiments that resulted in the first observation of an 'island of stability' of superheavy ele-

ments.[31–33] Nuclei on this island, long predicted by theory to be centred around neutron number 184 and proton number 114, have been the subject of many experimental searches over the last 30 years. They owe their unusual stability to their proximity to nuclei with filled major nucleon shells, resulting in a spherical nuclear shape. This is the same effect that imparts the extra stability associated with nuclei in the lead region and doubly magic ^{208}Pb in particular. The collaborative work since 1998, described in a review article,[34] has resulted in the discovery of elements 113, 114, 115, 116 and 118, and over 30 new isotopes.

We continue to investigate the region of the chart of nuclides near the 'island of stability with our colleagues at JINR in Dubna. Recently, an experiment was performed to attempt to synthesize element 120 using the ^{58}Fe + ^{244}Pu reaction — an exciting attempt to continue hot-fusion reactions utilizing beams other than ^{48}Ca. The results of this particular experiment set a cross-section limit for the production of element 120 and are discussed in more detail in Ref. 35. We also recently performed a ^{48}Ca + ^{226}Ra experiment to produce Hs isotopes and several candidate decay chains were observed.[36]

Indeed, there has been exciting progress in the study of the properties of the heaviest elements, and several other experiments are noteworthy of mention. Nuclear spectroscopic studies of ^{254}No and ^{256}Rf isotopes have for the first time identified states built on the single-particle levels from above the $Z = 114$ shell gap in spherical nuclei.[37–39] Additionally, the Dubna/LLNL element 112 and 114 results have been reproduced by GSI,[40] PSI/Dubna[41] and LBNL[42] recently. Finally, a Dubna/LLNL/ORNL collaboration is working to perform a ^{48}Ca + ^{249}Bk experiment in the Fall of 2009 to attempt to produce element 117. The ^{249}Bk is produced in the ORNL High Flux Isotope Reactor (HFIR) and is chemically purified at ORNL. This experiment has severe time considerations because ^{249}Bk has a half-life of only 330 days.

III. Conclusion

Many of the research areas discussed in this paper are interconnected. All are focused on elucidating a better understanding of nuclei and nuclear reactions — an organization of our knowledge regarding the nucleus of a chemical element. Mendeleev's efforts to organize the chemical elements, and hence derive a basic understanding of nature, continue today with our work on the chemistry and physics of new heavy elements, with our work to develop a complete fundamental description of nuclei by studying exotic nuclei and nuclei in exotic environments and with our work to understand the basic properties of the neutrino.

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