Method for Detection of Nuclear-Plasma Interactions in a 134Xe-Doped Exploding Pusher at the National Ignition Facility

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Angular momentum changes due to nuclear-plasma interactions on highly-excited nuclei in high energy density plasmas created at the National Ignition Facility can be measured through a change in isomer feeding following gamma emission. We propose an experiment to detect these effects in 133Xe in exploding pusher capsules.

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1. Introduction

Electron-mediated nuclear-plasma interactions (NPIs) such as Nuclear Excitation by Electron Capture (NEEC) or Transition (NEET) are expected to cause significant changes in reaction cross sections in High Energy Density Plasmas (HEDPs) such as nuclear weapons tests, National Ignition Facility (NIF) shots, and astrophysical settings. However, NPIs remain largely unobserved due to the extreme narrowness of nuclear transitions ($\Gamma \leq 1 \mu$eV). A schematic of the NEEC and NEET processes are shown in Fig. 1. In both cases, the atomic binding energies are modified in the plasma environment due to charge state and screening effects. For NEEC, an electron is resonantly captured into an atomic orbital where the binding energy plus the electron kinetic energy is equal to the first excited nuclear state (for excitations off the ground state). For the NEET process, atomic electron transition with a binding energy difference equal to the nuclear excitation energy. Then, the binding energy (for NEET) or the free electron energy plus the binding energy (for NEEC) is resonantly transferred to the nucleus via a virtual photon and the nucleus becomes excited.

HEDP environments are expected to have a significant effect on cosmogenic nucleosynthesis, the formation of heavy elements from pre-existing nucleons in astrophysical plasmas [1]. Nuclei in stellar plasmas reach a thermal population of low-lying excited nuclear states from photo-
pared to ground state nuclei is referred to as the stellar enhancement factor (SEF). In addition to this well-known but experimentally-unobserved effect of NPIs on low-lying nuclear states, NPIs may affect thermal neutron capture cross sections after the absorption of a neutron but before subsequent emission of a gamma ray. In such a case, small increases in angular momentum near the neutron separation energy due to NPIs dramatically increase the probability of neutron re-emission. The relevance of these processes to post-capture nuclei is subject to a competition between the short lifetimes (∼fs) of highly-excited states versus the large number of nuclear transitions available and high electron and photon fluxes. Modeling of these effects is unwieldy due to the large number of both atomic and nuclear transitions possible, necessitating experimental measurements.

Although there have been extensive theoretical studies on this subject, previous attempts to measure NEEC have been unsuccessful [2], including our own attempts [3] to detect the NEEC process by resonantly exciting the ground state of 181Ta and 187Os in plasmas generated at the Omega Facility at the Laboratory for Laser Energetics at the University of Rochester. The limited observations of the parallel bound-state process NEET are controversial and have been restricted to non-plasma environments [4–6]. We will overcome this challenge by inducing NPIs on highly-excited (~1–5 MeV) nuclear states produced by nuclear reactions prior to their decay by spontaneous gamma-ray emission. The large density of nuclear states (≥ 10^{-1}, 10^9 eV^{-1}) at these excitation energies increases the probability that the energy from the atomic transition will resonantly match an available nuclear transition. In addition, we will exploit a new experimental signature—the differential population of a high-spin isomer—to observe these effects.

2. The Double Isomer-to-Ground State (DIGS) Ratio: a Signature of NPIs

A major challenge in the search for NPIs is the observation of a clear signature of the effect in a highly-chaotic HEDP environment. The energy transfer is usually small (keV) and the subsequently emitted radiation is overwhelmed by the background radiation in the HEDP. We propose a new signature for NPIs on highly-excited nuclei: the differential population of an isomeric state versus the ground state of the de-excited nucleus.

While (n, γ) reactions are of primary importance to astrophysical and other HEDP environments, we may observe the same NPI effects following (n, 2n) reactions at the NIF, which initially populate states several MeV below the neutron separation energy prior to gamma emission. An experimentally-accessible candidate is 133Xe*, created in 134Xe(n, 2n) reactions, which has a long-lived 11/2- isomer and a 3/2+ ground state. The capability of NPIs to produce differential isomer population for reactions in versus out of a plasma is shown schematically in Fig. 2. The double isomer-to-ground-state (DIGS) ratio is defined as

\[ R_{D I G S} = \frac{N_{Xe-133m}^{\text{capsule}}}{N_{Xe-133g}^{\text{capsule}}} \frac{N_{Xe-133m}^{\text{TOAD}}}{N_{Xe-133g}^{\text{TOAD}}} \]  

where N is the number of each state populated, determined from its characteristic-energy gamma ray emissions measured in a high-purity germanium detector. A non-unity DIGS ratio for the 133mXe/133gXe fraction formed in an exploding pusher capsule plasma divided by same quantity for an externally-mounted, non-plasma “TOAD” (Target Option Activation Device) sample will show NPIs took place on highly-excited states in 133Xe.

3. Proposed Experiment

The National Ignition Facility (NIF) uses up to 2 MJ of laser energy to implode capsules of deuterium-tritium fusion fuel to pressures, densities, and temperatures similar to the core of the sun [7]. The combination of the large instantaneous neutron, photon and electron fluxes present in a NIF capsule is a unique environment for probing the interaction between highly-excited nuclear states with a HEDP. We propose an experiment at the NIF to observe NPIs on highly-excited 133mXe* using an existing platform and diagnostics. A standard indirect-drive exploding pusher platform [8] will be used, consisting of a gold hohlraum filled with low-density (0.03 mg/cc) helium gas surrounding a thin (120 - 175 μm) plastic (CH with a graded silicon dopant) shell/capsule filled with 10 atm DT gas. Because of the low compression and low capsule shell areal density, the exploding pusher platform, originally designed for diagnostic calibration, has also been used for controlled experiments [9] requiring evenly-heated and evenly-compressed plasmas and an unattenuated, isotropic, 14 MeV neutron source. The capsule gas
will be doped with 0.03 atm isotopically-pure (>99%) 134Xe. Indirect laser drive using 933 kJ of laser energy at a peak power of 325 TW will produce $>10^{14}$ DT fusion neutrons, inducing 134Xe$(n,2n)$ reactions in a $kT > 5$ keV plasma to produce 133Xe$^*$ in a highly-excited state a few MeV below the neutron separation energy.

Repeated NPIs in the high energy density plasma will transfer energy and angular momentum, altering the subsequent isomer population. The xenon originally loaded in the capsule will be collected in the existing RAGS (Radiochemical Analysis of Gas Samples) collectors, with measured collection efficiency greater than 50%, and transported to a low-background counting facility. As a control, 134Xe will also be loaded into one or more “TOAD” sealed containers located 50 cm from the implosion on up to three Diagnostic Instrument Manipulators (DIMs) irradiated with the same neutron spectrum. The thin shell and low compression of the exploding pusher ensures minimal difference between the original fusion neutron spectrum (overwhelmingly 14 MeV) in the plasma and that incident on the TOADs. The post-shot TOAD samples will be retrieved and also taken to a low-background facility for counting on the same detectors such that systematic uncertainties are the same between each sample and cancel on the TOADs. The post-shot TOAD samples will be doped with 0.03 atm isotopically-pure (similar to the NIF shot designated N130503), has shown this contributes no more than 10$^{-5}$ the number of reactions as in the capsule.

A previous NIF direct-drive exploding pusher shot (N120228-001) with 0.057 atm of a 52/44/4 mix of 124Xe, 126Xe and 128Xe showed the feasibility of irradiating, collecting, and counting xenon activation products [10]. The radioactive 123, 125, 127Xe products were collected using RAGS and counted $\approx$5 hours after the shot at the Building 151 nuclear counting facility at Lawrence Livermore National Laboratory.

The measured relative 126Xe$(n,2n)/124Xe(n,2n)$ ratio was 1.22 $\pm$ 0.05 ($\sigma_{\text{statistical}} = 4.1\%$). We anticipate better counting statistics in our experiment since the 133$m^+$Xe and 133$^f$Xe are significantly longer-lived than the 123, 125Xe radionuclides (2.00 and 17.1 hr respectively) resulting in fewer lost decays during retrieval and transport. Furthermore, the samples will be counted much closer to the detector (since only the ratio of the gamma-ray intensities and not their absolute magnitudes are important), and the yield will be higher. A statistical uncertainty of less than 1% is easily achievable based on these improvements.

The TOAD has an interior gas volume of 34.3 cm$^3$. For a NIF shot yield of 10$^{14}$ neutrons, the fluence at 50 cm from the implosion where the TOAD is located will be 3 $\times$ 10$^9$ n/cm$^2$, inducing 6 $\times$ 10$^6$ $(n,2n)$ reactions. This is more than sufficient to detect more than 10,000 $\gamma$ rays, achieving < 1% statistical uncertainty, in a high-purity germanium detector with $\sim$1% photopeak efficiency over a several-day measurement.

A potential background source of non-plasma xenon reaction products collected by RAGS is due to neutrons reacting with residual xenon in the capsule fill tube. An MCNP [11] simulation, conservatively assuming no attenuation, constant temperature and pressure, and a capsule compression to 200 $\mu$m (similar to the NIF shot designated N130503), has shown this contributes no more than 10$^{-5}$ of the number of reactions as in the capsule.

Another potential background source of 134Xe production is from thermal neutrons, which may induce 132Xe$(n,\gamma)$133Xe reactions on small impurities in the xenon gas. With an expected 0.3% 132Xe impurity in a 99% enriched 134Xe gas and a thermal neutron fraction of 3% of the 14 MeV fluence, the fraction of 133Xe produced from these $(n,\gamma)$ reactions only is 2.5 $\times$ 10$^{-5}$.

While the thermal cross section for 133Xe$(n,\gamma)$134Xe is quite high (190 b), no significant depletion of the 133Xe product is expected due to the low number density of nuclei produced ($\sim$10$^{-12}$ of the 132Xe impurity).

The Hauser-Feshbach nuclear reactions code, TALYS [12] was used to predict isomer and ground state population from this reaction. By default, TALYS uses the first ten known discrete levels in 131Xe, above which a level density model is used. This number was increased to 28 to include known, prominent, low-lying, isomer-feeding states (such as the twelfth level at 1.1695 MeV with $J = 13/2^-$). Changing this parameter, TALYS predicts cross sections of the 134Xe$(n,2n)$ reaction leading to the isomer and ground state of 133Xe to be, respectively, 0.75 b and 0.93 b, consistent with the limited experimental data available for this reaction [13, 14]. Thus these TALYS level parameters were later used (see next Section) to predict the cross sections at lower energies where measurements are unavailable. The ratio of this isomer cross section to the ground state cross section is 0.8. This near-equal population of each state provides ideal detection statistics and sensitivity to perturbation.

The baseline population 133$m^+$Xe/133$^f$Xe ratio from 14 MeV neutrons was measured using the DT generator located at iThemba LABS in South Africa. Results from that experiment are currently under analysis. Further quantification of background reactions and rate parameters could be obtained through thermal neutron capture experiments on 132Xe at a reactor or other thermal neutron source, and radiative strength function/level density measurements using the “Oslo method” [15] in inverse kinematics at iThemba using the $d$(132Xe,131Xe)$/p$ reaction.

4. MCNP Modeling of the Neutron Scatter Effect on the DIGS Ratio

A significant uncertainty in Eq. 1 can arise from neutrons scattering off DIMs and other chamber components such that the neutron spectra in and out of the plasma (capsule vs. TOAD) may not be precisely identical. This ef-
With the calculated cross sections including all 28 known TOAD are also shown in Fig. 4. Convoluting these spectra to lower energies (down to the \( \text{MeV} \)) does not affect the previous Section) shown in Fig. 4, it is apparent that neutron spectrum isomer-to-ground state ratio calculated with TALYS (see experimental section) in the capsule (red) and the TOAD (blue) above the 8.6 MeV threshold, along with the isomer-to-ground state \((m-g)\) cross section ratio (green, right axis) produced in \( ^{134}\text{Xe}(n, 2n) \) reactions predicted by TALYS for both the default number of discrete levels used (DL = 10) and increased values (DL = 20 and 28). Experimentally-measured ratios \([13, 14]\) are noted by datapoints. Statistical uncertainties in the neutron spectra are smaller than the datapoints.

Fig. 3 Geometry of NIF chamber and internal components modeled with MCNP to determine neutron downscatter in the TOAD xenon. The NIF chamber and target bay (left) surround the DIMs and cryo-TARPOS (right). Each DIM holds a TOAD (bottom left) and the capsule is contained within a hohlraum in the thermomechanical package (bottom right) at the end of the cryo-TARPOS.

The deployment of multiple TOADs of varying materials and thicknesses on up to three DIMs with different lines-of-sight in test shots will provide confidence in this modeling. Any changes in the DIGS ratio from one position to another, will indicate scattering effects rather than NPIs.

5. Predicted Results

Predicting the exact impact of NPIs on the relative populations of \( ^{133}\text{Xe} \) and \( ^{133}\text{Xe}^* \) is a significant challenge, due in part to the extremely large number of possible atomic transitions coupled to a similarly large number of nuclear transitions. The probability of an NPI is also extremely uncertain due to its dependence on the low-energy (keV) photon transition strength, characteristic of the Xe M-, L-, and K-shell atomic binding energies.

Top to bottom, respectively, Fig. 5 shows the initial excitation energy and angular momentum states populated immediately after \( ^{134}\text{Xe}(n, 2n)^{133}\text{Xe}^* \), the subsequent gamma decay path predicted (in the absence of NPIs), and the angular momentum distribution of the nucleus’ actual levels predicted by TALYS. Multiple NPIs will on average shift the angular momentum towards the actual available level distribution, overall lowering the angular momentum distribution as the nucleus decays by gamma-ray emission as indicated in the lower panel of Fig. 5. This lowering of angular momentum will increase the proportion of decays that populate the lower-spin ground state of \( ^{133}\text{Xe} \) versus the higher-spin \( ^{133}\text{Xe}^* \) isomer.

In order to determine the impact of NPIs on isomer population, we must consider both the transition rate of the NPIs, and how this additional decay channel will modify normal nuclear decay.
5.1 NPI rate

To determine the rate of NPIs on an excited state, we assume a local thermal equilibrium environment and use the principle of detailed balance, following the methodology of Gosselin and Morel [16] in which the rate of NEEC between an initial state \( i \) and a final state \( f \) is given by

\[
\lambda_d^{\text{NEEC}} = \frac{2J_f + 1}{2J_i + 1} \frac{T_{\text{ref}}^{\gamma}}{T_{\text{ref}}^{\gamma+}} f_{\text{FD}}(E_b)(1 - f_{\text{FD}}(E_i))
\]

\[
\times \left( \text{erf} \left( \frac{E_f}{E_b} \right) - \text{erf} \left( \frac{E_{b}}{E_f^\gamma} \right) \right) ,
\]

(2)

where \( J_i \) and \( J_f \) are the spins of states \( i \) and \( f \); \( \alpha(T_c) \) is the internal conversion coefficient for the \( i \rightarrow f \) transition; \( T_{\text{ref}}^{\gamma} \) is the radiative lifetime of the transition; \( f_{\text{FD}} \) is the Fermi-Dirac function; \( E_f \) and \( E_b \) are the free and bound electron energies, which are assumed to resonantly match the \( i \rightarrow f \) transition, and \( \epsilon \) is the dispersion of the electronic transition energy around the average atom value.

Rather than using the average atom model, however, we use the calculated energies of specific states and configurations from the atomic physics code FLYCHK [17], which replaces the error functions with a sum over all bound states \( b \). Furthermore, we assume that our initial state \( i \) is in the quasi-continuum, and so instead of a single state \( f \), we consider all possible final nuclear states, which we represent with the level density \( \rho(E_i + (E_f + |E_b|), J_f) \), and replace \( T_{\text{ref}}^{\gamma} \) with the average transition strength,

\[
\lambda_d^{\text{NEEC}} = \sum_{a,b} \frac{2J_f + 1}{2J_i + 1} \frac{\alpha(T_c)\ln(2)}{T_{\text{ref}}^{\gamma+}} \rho(E_i + (E_f + |E_b|), J_f)
\]

\[
\times f_{\text{FD}}(E_b)(1 - f_{\text{FD}}(E_i)).
\]

(3)

Now we substitute the expression for the photon strength function, \( S(E_i) \),

\[
S(E_i) = \frac{\hbar \rho(E_i + E_f, J_f)}{2 - \left\{ \frac{T_{\gamma}^{\gamma+}}{E_f^\gamma} \right\}} ,
\]

(4)

where \( E_f = E_i + |E_b| \), and finally, we integrate over all possible electron energies with a differential electron flux \( \frac{d\Phi(E_r)}{dE_r} \), yielding

\[
\lambda_d^{\text{NEEC}} = \int dE_r \frac{d\Phi(E_r)}{dE_r} \sum_{a,b} \frac{2J_f + 1}{2J_i + 1} \alpha(T_c)\ln(2) \frac{E_f^\gamma}{\hbar}
\]

\[
\times S(E_i)f_{\text{FD}}(E_b)(1 - f_{\text{FD}}(E_i)).
\]

(5)

Unfortunately, no measurements of the photon strength function, \( S(E_i) \), in the quasi-continuum have been made below \(~1\) MeV, which is well above the transition energies where \( \alpha(T_c) \) is substantial. To bound the possible NEEC rate, we consider two possible photon strength functions. The lower bound is based on the assumption that the photon strength function at low energies follows the generalized Lorentzian form seen at higher energies. With this photon strength function and a typical level density in the quasi-continuum, even in very hot, dense plasmas, the NEEC rate is on the order of 1/ns. To determine the upper bound, we assume that the photon strength function is given by the single-particle estimate, in which case the NEEC rate can be as high as \(~10\) fs\(^{-1}\).

5.2 Gamma-decay modification due to NPIs

To estimate the impact on high spin isomer production, we must include NPIs in a Hauser-Feshbach model. For some initial excited state \( i \), the total decay width is the sum of all possible partial decay widths. The branching ratio to a specific final state \( f \) is given by the ratio of the partial decay width to this total decay width.

To include the effects of NPIs, we assume a total NPI rate and convert it to a decay width to include among the existing decay channels. We use the reaction code TALYS to perform the standard Hauser-Feshbach calculation, and include the effects of NPIs manually. The initial population in \( E, J \) space for \(^{133}\)Xe following the \(^{134}\)Xe\((n,2n)\) reaction is calculated using TALYS, as well as the total decay width for normal reactions for each \( E, J \) bin. We then sequentially calculate the decay from the highest energy bin. For the assumed NPI rate, the NPI branching ratio for each state is calculated, and this ratio is applied to the population of the state to determine the population which undergoes NPIs. The remaining population is allowed to decay by a single gamma ray, and the new population is added to the initial \( E, J \) population matrix. Because the atomic transition energies are small compared to the typical bin size in the calculation, it is assumed that NPIs do not change the energy bin, and only change the spin. The spin change is allowed to be \(-1, 0, \) or \(+1\), with a rate proportional to the final state level density for each spin. This results in a new population in the maximum energy bin with a new spin distribution. The process is then repeated, with the proper fraction of the new population being allowed to undergo NPIs, while the rest are allowed to decay normally.
until the population of the highest energy bin falls below $10^{-8}$ mb/MeV. The process is then begun again with the next highest energy bin. This is repeated until the decay is complete, and then the isomer to ground state ratio is computed. Based on these results, an NPI rate of $\sim 0.4$ fs$^{-1}$ is required to reduce the isomer to ground state ratio by 5%. A rate of 1/fs will reduce this ratio by 10%. For the lower limit of the NPI rate, based on the generalized Lorentzian photon strength function, no visible impact will be seen in the DIGS ratio, but a 5 - 10% effect will be measurable for the single particle estimate of the PSF.

This analysis neglects the NEET process which may contribute significantly more to the NPI rate. Furthermore, this estimate is also sensitive to the low-energy portion ($\sim$keV) of the photon strength function characteristic of the Xenon M-, L-, and K-shell atomic binding energies. The low-energy portion of the strength function in similar-mass nuclei has exhibited an unexpected and unexplained dipole enhancement at low energies [18, 19], deviating dramatically from the Lorentzian models. Only the tail of this enhancement above 1 MeV has been measured, however. Depending on the keV nature of the enhancement, which is not included in the generalized Lorentzian used in TALYS calculations, reaction rates could increase by many orders of magnitude. Even if no change in the DIGS ratio is observed, limits can be set on the low-energy photon transition strength in the quasi-continuum, where no measurements are currently possible and theoretical estimates vary by many orders of magnitude.

6. Conclusions

The DIGS ratio is introduced as a signature for detecting angular momentum changes due to NPIs on highly-excited nuclei prior to gamma emission. We propose an experiment at the NIF to detect NPIs using the DIGS ratio in highly-excited $^{131}$Xe$^+\cdot$, produced in $^{134}$Xe($n,2n$) reactions, loaded into an indirect drive exploding pusher capsule. If the DIGS ratio does not deviate from unity, indicating NPI effects, limits can be set on the unknown low-energy photon transition strength in the quasi-continuum. Deviation from unity will indicate, for the first time, the alteration of a nuclear reaction due to a plasma environment.

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