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Direct mass measurements of short-lived A = 2Z - 1 nuclides ⁶³Ge, ⁶⁵As, ⁶⁷Se and ⁷¹Kr and their impact on nucleosynthesis in the rp process

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Mass excesses of short-lived A = 2Z - 1 nuclei ⁶³Ge, ⁶⁵As, ⁶⁷Se, and ⁷¹Kr have been directly measured for the first time to be -46921(37), -46937(85), -46580(67), and -46320(141) keV, respectively. The deduced proton separation energy of -90(85) keV for ⁶⁵As shows that this nucleus is only slightly proton-unbound. Xray burst model calculations with the new mass excess of ⁶⁵As suggest that majority of the reaction flow passes through ⁶⁴Ge via proton capture, indicating that ⁶⁴Ge is not a significant *rp*-process waiting point.

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Type I X-ray bursts occur on the surface of neutron stars accreting matter from a companion star in a stellar binary system. The accreted matter often contains hydrogen and helium, which accumulate on the neutron star surface for hours or days before igniting and burning explosively for 10 to 100 seconds causing a bright X-ray burst. The nuclear reaction sequence powering the bursts is the rapid proton capture process (*rp*-process) [1], a sequence of proton captures and β^+ decays near the proton drip line ending, in some extreme bursts, in the Sn region. Reliable nuclear physics input, including precise mass values, is needed for the nuclides along the *rp*-process path to compare models with observations of the light curve profiles and to extract quantitative information about the stellar environments [2].

It has been shown that long-lived rp-process waiting points beyond ⁵⁶Ni can qualitatively explain the extended tails that are observed in the light curves of many X-ray bursts [3, 4]. A waiting point is a nucleus near the proton drip line where a captured proton is unbound or only weakly bound and can easily be removed by photodisintegration. As a result, proton capture is effectively suppressed and the rp-process has to proceed via the slow β^+ decay. This delays the nuclear burning and leads to the extended burst tail. Three major waiting points that dominate the energy generation during the burst tail have been identified: ⁶⁴Ge, ⁶⁸Se, and ⁷²Kr [5]. Their estimated long lifetimes provide the explanation for the observed burst tails. As a significant amount of hydrogen is required for the rp-process to pass through the A=64–72 mass region, this has enabled astronomers to interpret long bursts as hydrogenrich, and short bursts as hydrogen-poor. This is important as the amount of hydrogen affects the opacity of the atmosphere and therefore distance estimates and attempts to extract properties of the underlying neutron star from burst observations.

For a quantitative analysis of burst light curves in terms of hydrogen contents and other system parameters, the effective lifetimes of the suggested waiting points need to be known. They determine the overall processing time scale, energy generation, and the composition of the burst ashes. As all β^+ half-lives are known, the most critical parameter becomes the proton separation energy (S_p) of the nuclei produced by proton capture on the waiting point nucleus, which can exponentially impact the effective lifetime of the waiting point [5]. $S_p(^{69}\text{Br})$ and $S_p(^{73}\text{Rb})$ are known to be negative based on experiments demonstrating that both ⁶⁹Br and ⁷³Rb are fast proton emitters [6, 7]. This makes it likely that 68 Se and 72 Kr are waiting points even though 2p-captures can reduce their effective lifetime [5]. On the other hand, for ⁶⁴Ge, which is the most important potential waiting point [8] because it is encountered first, only a somewhat model dependent lower limit of $S_n(^{65}As) > -250$ keV had been derived from the observation of the β decay of ⁶⁵As [9]. A precise measurement of $S_p(^{65}\text{As})$ is thus needed to determine to which degree ^{64}Ge is a waiting point, and if it is a waiting point at all. We address this long-standing problem here by measuring accurately the mass of ⁶⁵As ($T_{1/2}$ =128 ms), which, together with previous measurements of the ⁶⁴Ge mass [10], enables us to derive for

the first time the experimental $S_p(^{65}As)$ value.

Recently, Penning trap mass measurements have reached some nuclides along the *rp*-process path [10–15]. However, the more exotic ⁶⁵As was out of reach so far. Attempts have been made to determine S_p (⁶⁵As) by computing the Coulomb displacement energy (CDE) between ⁶⁵Ge and ⁶⁵As [16]. However, this theoretical approach needs to be verified experimentally, especially in this mass region where rapid shape changes and shape coexistence might affect nuclear masses.

The experiment has been performed at the HIRFL-CSR (Cooler Storage Ring at the Heavy Ion Research Facility in Lanzhou) [17]. A test experiment was conducted prior to the main run described here yielding a preliminary mass excess $ME(^{65}As) = -47428(\pm 530) \text{ keV}$ [18]. In this Letter we report on the accurate measurement of the masses of four A = 2Z - 1 nuclei 63 Ge, ^{65}As , 67 Se, and 71 Kr in a new experiment.

A primary ⁷⁸Kr¹⁹⁺ beam of 4 MeV/u was provided by a sector focusing cyclotron. The beam passed through a carbon stripper-foil placed inside the beam-transport line, and the ⁷⁸Kr²⁸⁺ ions were selected and injected into the main coolerstorage ring (CSRm). The ⁷⁸Kr ions were accumulated in CSRm and then accelerated to an energy of 483.4 MeV/u. A beam intensity of up to $\sim 10^8$ particles per spill has been achieved. The high-energy ⁷⁸Kr beam was fast extracted from CSRm and focused upon a ~ 15 mm thick beryllium production target placed at the entrance of the in-flight fragment separator (RIBLL2). The exotic nuclei were produced by projectile fragmentation of ⁷⁸Kr and emerged from the target predominantly as bare ions. After in-flight separation of the fragments in RIBLL2, the cocktail beam of exotic nuclei was injected into the experimental storage ring (CSRe). RIBLL2 and CSRe were set to a magnetic rigidity of $B\rho = 5.9493$ Tm to optimize the transmission of the A = 2Z - 1 nuclides. Other nuclides within the acceptance of the RIBLL2-CSRe system were also transmitted and stored in CSRe. After each injection, about 20 ions were simultaneously stored in CSRe.

The revolution times, T, of various stored ions are related (in first order) to their mass-to-charge ratios, m/q, via [19]

$$\frac{\Delta T}{T} = \frac{1}{\gamma_t^2} \frac{\Delta(m/q)}{(m/q)} - \left(1 - \frac{\gamma^2}{\gamma_t^2}\right) \frac{\Delta v}{v},\tag{1}$$

where γ is the relativistic Lorentz factor and γ_l denotes the transition point of the storage ring. In order to determine m/q, the revolution times of the ions have to be measured and the term containing their velocity spreads, $\Delta v/v$, needs to be minimized. For this purpose, the CSRe was tuned into an isochronous ion-optical mode [19] and the energy of the primary beam was chosen such that $\gamma \approx \gamma_l = 1.395$ for the ions of interest. Thus, the revolution times reflect directly m/q ratios of the stored ions, independent of their $\Delta v/v$.

The revolution times were measured using a timing detector [20] equipped with a 19 μ g/cm² thin carbon foil of 40 mm in diameter. Each stored ion passed through the timing detector at every revolution in CSRe. Secondary electrons released from the foil due to the passing ions were guided isochronously by perpendicularly arranged electrostatic and

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FIG. 1: The revolution time spectrum zoomed in on a subset of the A = 2Z - 1 nuclides. The insert shows the well-resolved ground and isomeric ($T_{1/2} = 247$ ms, $E^*=3174.3(1.0)$ keV [21]) states of ⁵³Co. Our result $E^*({}^{53}Co^m) = 3202(36)$ keV is in good agreement with the literature value.

magnetic fields to a set of micro-channel plates. The time resolution of the detector was about 50 ps, and the detection efficiency varied from 20% to 70% depending on the ion type and the number of the ions stored. The signals from the detector were sampled with a digital oscilloscope Tektronix DPO 71254 at a sampling rate of 50 GHz. The recording time was 200 μ s for each injection corresponding to ~320 revolutions.

In this experiment, 309, 49, 104, and 28 particles were accumulated for 63 Ge, 65 As, 67 Se, and 71 Kr, respectively. The revolution time of each particle and its identification were determined in a way similar to that described in Ref. [19]. Fig. 1 presents the histogram of the extracted revolution times for a part of the A = 2Z - 1 nuclei. The nuclides whose masses were determined in this work are indicated by bold symbols.

The resolving power of CSRe mass spectrometry is mainly determined by instabilities of the magnetic fields. These instabilities cause small shifts in the revolution time of the entire spectrum for different injections. In order to reduce the effect of these shifts, we measured the revolution time of each particle relative to the revolution time of a selected reference nuclide present in the same injection. As not all nuclides were present in each injection, we used seven reference peaks (¹⁹Ne, ²¹Na, ²³Mg, ²⁵Al, ²⁷Si, ²⁹P, and ³¹S) thus creating seven independent spectra which contain 69% of all particles. The resolving power of the shift-corrected spectra is 170'000.

All the A = 2Z - 1 nuclides with experimentally known masses [21, 22] from ³³Cl through ⁶¹Ga (except for ⁴³Ti since it has a low-lying isomeric state) were used to fit m/q versus T employing a second order polynomial. The ME values of ⁶³Ge, ⁶⁵As, ⁶⁷Se, and ⁷¹Kr were obtained and presented in Table I by extrapolating the fit function to the corresponding T. The seven sub-spectra and an integral spectrum have been analyzed separately and provided consistent results. In order to estimate possible systematic errors, the literature ME values

TABLE I: ME_{CSRe} and S_p values obtained in this work. ME_{CDE} values are derived from mirror nuclei (⁶³Ga [22], ⁶⁵Ge [10], ⁶⁷As [10] and ⁷¹Br [15]) using CDE [16]. ME_{AME03} values are taken from AME03 [22]. Values estimated in AME03 are marked with [#].

Atom	ME _{CSRe} [keV]	S_p [keV]	ME _{CDE} [keV]	ME _{AME03} [keV]
⁶³ Ge	-46921(37)	2210(46)	-46945(100)	$-46910(200)^{\#}$
⁶⁵ As	-46937(85)	-90(85)	-46776(100)	$-46980(300)^{\#}$
⁶⁷ Se	-46580(67)	1851(74)	-46548(100)	$-46490(200)^{\#}$
⁷¹ Kr	-46320(141)	2184(142)	-46025(100)	-46920(650)

have been re-determined in the analysis using different number of reference nuclides and different extrapolation ranges. For instance, the re-determined mass values for ⁵⁵Ni, ⁵⁷Cu, ⁵⁹Zn, and ⁶¹Ga (the extrapolation range is about the same as used to fit the new masses) are consistent with literature values [21, 22] within 1 σ . Mainly due to the small extrapolation range (for ⁷¹Kr it is just ~0.76 ns), the errors for the new masses are dominated by statistical errors. Details of the data analysis will be described in [23].

 S_p values for ⁶³Ge, ⁶⁵As, ⁶⁷Se, and ⁷¹Kr were deduced using our new masses and the previously known masses of ⁶²Ga [22], ⁶⁴Ge [10], ⁶⁶As [10], and ⁷⁰Br [15]. We emphasize that $S_p(^{65}As) = -90(85)$ keV. This confirms experimentally for the first time that ⁶⁵As is unbound against proton emission at a 68.3% confidence level. Our $S_p(^{65}As)$ agrees with the limit of $S_p > -250$ keV obtained in Ref. [9] based on the observed β -decay and theoretical estimates of the proton penetrability.

The new experimental ME values are compared in Table I with those calculated by the CDE method [16] and those from the latest Atomic Mass Evaluation (AME03) [22]. Good agreement between CSRe and CDE is found for ⁶³Ge, ⁶⁵As, and ⁶⁷Se, which confirms the predictive power of the CDE method in this mass region and shows that at least for these three nuclei the method is reliable. In the case of ⁷¹Kr, the CDE value differs by more than one standard deviation from our CSRe result. The recently measured ME(^{71}Br) = -56502.4(5.4) keV [15], combined with $Q_{\beta}(^{71}\text{Kr}) = -10140(320)$ keV [24], yields ME(⁷¹Kr) = -46360(320) keV, which agrees nicely with our CSRe result of ME(⁷¹Kr) = -46320(141) keV. The disagreement of the CDE value with ours for ⁷¹Kr may hint at a structure change along the N = Z line at N = Z = 35. Nuclei with N = Z < 35are nearly spherical while the heavier ones are strongly deformed. Hasegawa et al. [25] have suggested that there is a sudden shape change near the ground state at N = Z = 35, which may be regarded as a (spherical to deformed) phase transition. Furthermore, Kr isotopes with $N \approx Z$ are known to exhibit the shape-coexistent phenomenon [26, 27]; for example, deformed prolate and oblate shapes compete near the ground state with comparable energies. This suggests that deformation effects and shape-coexistence are the factors that future CDE calculations may need to consider.

The experimental $S_p(^{65}As)$ allowed us to determine the degree to which ^{64}Ge is an *rp*-process waiting point. Fig. 2



FIG. 2: Regions in the temperature and density plane where 64 Ge is a waiting point (WP) with an effective lifetime longer than 50% of the β -decay lifetime. Shaded areas are deduced assuming a 1 σ variation of the AME03 mass for 65 As (grey) and the 65 As mass from this work (black). Results are given for a proton mass fraction of 0.7.

shows regions in the temperature-density plane where proton captures reduce the effective lifetime of ⁶⁴Ge to less than 50% of the β -decay lifetime resulting in a less effective waiting point. With our new mass value the temperatures and densities needed to bypass the ⁶⁴Ge waiting point are rather well defined. In particular, it is clear that a fairly high density in excess of 2×10^4 g/cm³ will be needed. For densities below 2×10^5 g/cm³ the required temperature range is rather narrow around 1.3 GK. For lower temperatures, ⁶⁴Ge can only be bypassed at higher densities. To illustrate the importance of the mass accuracy, we also show the region obtained with a 300 keV uncertainty (AME03). Though it is bigger than the uncertainty quoted for CDE calculations of the ⁶⁵As mass, we stress that the theoretical uncertainties were estimated from the performance of the model in other mass regions.

We have implemented our new results in a one-zone X-ray burst model [4] to explore the impact on modeled light curves. Fig. 3 shows the calculated X-ray luminosity as a function of time. Varying our $S_p(^{65}As) = -90$ keV within 2σ provides essentially identical light curves demonstrating that our accuracy is sufficient to eliminate the effect of the ^{65}As mass uncertainty in X-ray burst calculations. 89-90% of the reaction flow passes through ^{64}Ge via proton capture indicating that ^{64}Ge is not a significant rp-process waiting point. In contrast, using the estimated upper 2σ limit for ^{65}As from AME03 ($S_p(^{65}As)\approx -650$ keV), leads to a reduction of the proton capture flow through ^{64}Ge to 54% with a significant effect on the light curves (see Fig. 3).

Fig. 4 shows the composition of the burst ashes. This is relevant for model calculations of the crusts of accreting neutron stars that exhibit X-ray bursts [28]. The sensitivity of the composition to the ⁶⁵As mass uncertainty had been pointed out before [8]. A 1 σ variation of the remaining 85 keV uncertainty of S_p (⁶⁵As) has overall a very small effect, except for a factor of three uncertainty in the A = 64 abundance. We emphasize that the A = 64 abundance is about an order of mag-



FIG. 3: (color online) Calculated X-ray luminosity in a one zone X-ray burst model varying S_p ⁽⁶⁵As) within 2σ for masses from AME03 [22] (dashed black lines) and from this work (solid red lines).



FIG. 4: (color online) The calculated abundance distribution of the X-ray burst ashes using $S_p(^{65}As)$ from AME03 (black filled circles) and from this work (red open squares). Error bars indicate uncertainties induced by the 1σ uncertainty of $S_p(^{65}As)$. Except for A = 64, the error bars of our new results are within the symbol size.

nitude lower than the most abundant constituents of the burst ashes. If the AME03 values are used, the uncertainty of the A = 64 abundance is much larger and includes the possibility of A = 64 being one of the main components of the ashes. It is clear that our new data have reduced the uncertainties significantly. For the data from the AME03, the changes in the calculated abundances (see Fig. 4) are entirely due to the lower bound of S_p that causes ⁶⁴Ge to become a significant waiting point. Interestingly this delays energy generation, resulting in high temperature conditions for a longer time and enhancement of the production of the heaviest isotopes. This is the opposite of what one might naively expect for a stronger bottleneck at the proton-unbound ⁶⁵As.

In summary, the masses of the short-lived A = 2Z - 1 nuclides ⁶³Ge, ⁶⁵As, ⁶⁷Se, and ⁷¹Kr have been directly measured for the first time using a new implementation of the storage ring mass spectrometry. While for ⁶³Ge, ⁶⁵As and ⁶⁷Se our data confirm theoretical predictions based on CDE

calculations, a 1.7σ discrepancy to theory for ⁷¹Kr may suggest the necessity of an experimental validation of such calculations for nuclei along the N = Z line. The measurement of the ⁶⁵As mass provides the first experimental $S_p(^{65}As)$ value. This establishes reliably the astrophysical conditions under which 64 Ge is an *rp*-process waiting point. For the particularly hydrogen rich X-ray bursts explored in our X-ray burst model, ⁶⁴Ge is now with certainty not a major waiting point in the rp-process in X-ray bursts. This increases the importance of possible subsequent waiting points such as ⁶⁸Se and ⁷²Kr, for which an experimental determination of $S_p(^{69}\text{Br})$ and S_p ⁽⁷³Rb) would be desirable, though existing limits suggest these nuclei as waiting points. Our measurement has removed a major uncertainty in X-ray burst models and paved the way for a quantitative analysis of observed X-ray burst tails. The current results may also be of importance in the v_p process [29, 30] in neutrino driven winds in core collapse supernovae, though more work needs to be done to explore the sensitivity of this site to masses around ⁶⁴Ge, ⁶⁸Se and ⁷²Kr.

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