

Unlocking Auger electron therapy via heavy-ion measurements

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Unlocking Auger electron therapy via heavy-ion measurements

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Within the radiotherapeutic arsenal, Auger Electron Therapy (AET) has attracted much interest recently as a method for targeted radiotherapy to cancerous cells with minimal impact on the surrounding healthy tissue (Cornelissen and Vallis 2010; Ku et al. 2019; Bolcaen et al. 2023). AET is a type of targeted radionuclide therapy that uses the Meitner-Auger effect, a stochastic physical process that competes with X-ray emission, in which low-energy electrons are emitted during atomic relaxation. These electrons, due to their low energy (eV – 100 keV level) and high linear energy transfer (LET), deposit their energy over an incredibly short range (on the order of nm), comparable to the size of the DNA within a biological cell (Cole 1969; Kassis 2010). A critical barrier to implementation remains however, in the determination of the dose deposited and its spatial disposition. This is largely dictated by the number and energy of electrons emitted in the decay process, but cannot be determined comprehensively by conventional electron-spectroscopy techniques. Overcoming this challenge is crucial for both patient safety and the efficacy of the targeted therapy. A state-of-the-art model, BrIccEmis (Lee et al. 2012; Lee et al. 2016), has been developed, and requires benchmarking against these medically relevant observables. Here, we present a novel experimental approach, in which laboratory measurements of ¹⁹⁷Au are compared to state-of-the-art atomic model calculations. Critically, the atomic processes in this method occur in a vacuum, avoiding interference with matter and thus making an ideal

laboratory for testing the model. The observed Auger electron multiplicity is compared with calculations using the BrIccEmis code, which well reproduce the experimental data. Thanks to the widely applicable nature of the method, we argue that this provides a route towards the complete resolution of the Auger-electron multiplicity challenge and a crucial step towards accurate dosimetry for Auger-electron therapy.

The Meitner-Auger effect is a quantum mechanical process that occurs when a vacancy is present in an atomic shell. An electron from a higher atomic shell drops down to fill the vacancy, creating a new vacancy as it does so, and the energy associated with this de-excitation is transferred to an outer-shell electron, which is then ejected from the atom. The effect is shown schematically in Fig. 1 along with other atomic emissions that follow nuclear decay by internal conversion. The effect also arises following nuclear decay by electron capture. As the vacancy propagates towards the outermost shell, the atom can emit many Auger electrons. Since the Meitner-Auger effect competes with X-ray emission, the number of Auger electrons emitted follows a statistical distribution, and, because it is a purely atomic process, the electrons released have relatively low energies (<10 keV). This makes Auger electrons uniquely well suited to targeted therapy for treating highly localised tumour regions, whereas nuclear emissions, such as beta or alpha decay, deposit energy over a much greater distance (on the order of mm), providing unnecessary dose to healthy tissue. Furthermore, the high probability for emission of multiple Auger electrons during a single atomic cascade increases the likelihood of inducing double-strand breaks in the DNA of cancerous cells. Despite renewed interest in the medical and scientific communities, the use of Auger electron emitters for radiotherapy is still unrealised, with only a few agents evaluated in clinical trials, and there are no AET-specific radiopharmaceuticals that have received regulatory approval for clinical use (Ku et al. 2019, Wulfmeier et al. 2023).

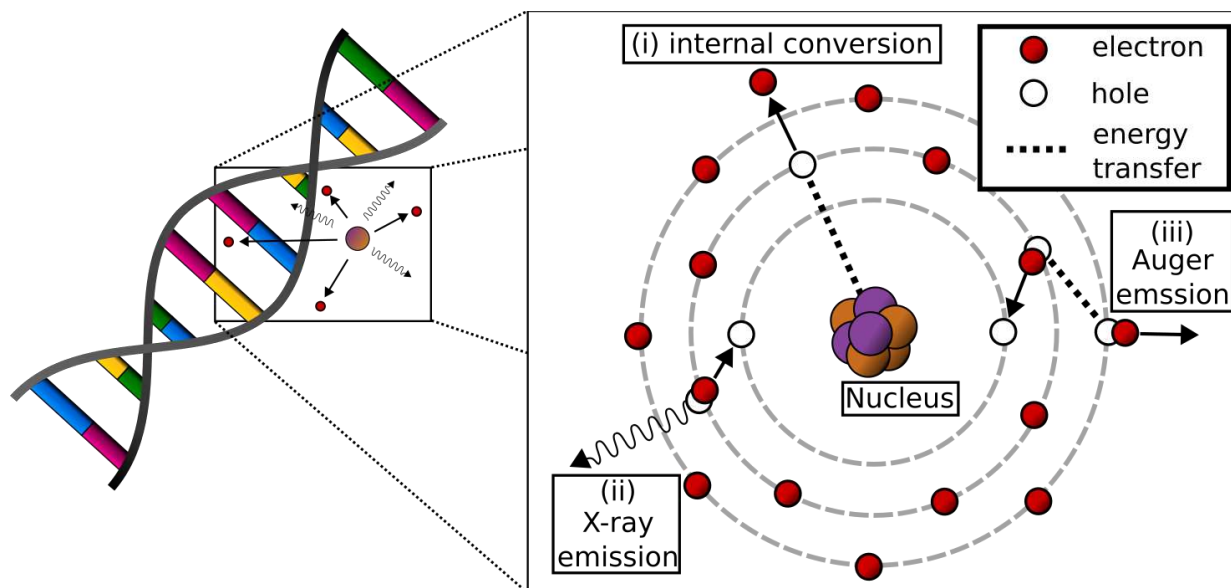


Figure 1. Schematic diagram showing atomic emissions resulting from the internal conversion of a nuclear transition. Auger-electron emission competes with characteristic X-ray emission, releasing low-energy electrons with ranges comparable to the size of DNA within a cell.

The Auger emission process results from the complex evolution of atomic vacancies as an ion relaxes into its lowest energy state, however, the dosimetric quality of an AET candidate radioisotope can be reduced to five terms (Bolcaen et al. 2023). Those terms include the physical half-life of the radioisotope, and three terms that relate to the cellular dosimetry, which arise from the efficacy of the radiopharmaceutical in providing dose to the biological nucleus of the cell. The fifth term, which the authors deem to have particular importance to assessing the dosimetric quality of a radionuclide, is simply the average number of Auger electrons emitted by a neutral atom in a single decay process (N_{AE}), however, this apparent simplicity is deceptive. Due to the low energy of some of the Auger electrons emitted in a typical cascade, many are experimentally unobservable in conventional experiments, preventing a simple counting measurement. In this work, we demonstrate a novel technique to determine the Auger multiplicity in a manner that is not limited by the low-electron energy. We present the charge reset method – a new technique for measuring average Auger electron emission multiplicities – and the results of its application to ^{197}Au , the nucleus populated in the β^+ /electron capture decay of ^{197}Hg , which has been identified as a promising candidate radioisotope for AET (Bolcaen et al. 2023). The results are compared to predictions from the theoretical code BrIccEmis (Lee et al. 2012).

Charge reset methods were originally developed (Ulfert et al. 1978) and refined (Barber et al. 2020; Heery et al. 2021) for excited-state lifetime measurements in fundamental nuclear physics. In its present application, for Auger-electron multiplicity measurements, an accelerated beam of ions impinges on a solid-foil target, scattering the target nuclei in a forward direction while some fraction of them are excited by the Coulomb interaction. The recoiling target ions then pass through a carbon charge-reset foil, serving to equilibrate the ionic charge about a mean value, defined largely by the velocity of the ions (Schiwietz and Grande 2001). Nuclei that undergo excited-state decay via electromagnetic processes prior to passing through the charge-reset foil have their charge states equilibrated subsequent to the decay and continue without any change in their charge state. The charge-reset foil therefore erases the previous electronic history of the ion, regardless of whether the decay proceeded via γ -ray emission or internal conversion. Nuclei that undergo excited-state decay via internal conversion after the charge-reset foil have their charge-state (Q) increased relative to the equilibrated value. This is due to the emission of an internal conversion electron ($\Delta Q=+1$) and the subsequent Auger cascade ($\Delta Q=+N_{AE}$). The charge-reset foil therefore converts the observed charge-state increase into a measure of the number of Auger electrons emitted during the selected decay. Thus, by selecting excited states with sufficiently long lifetimes so that the nucleus decays after the charge-reset foil, and with high internal conversion probability, one can probe the Auger-electron emission multiplicity through a simple measurement of the charge-state distribution. In practice, this is simplified through the selection of the excited state of interest by observing γ -rays populating the state. One thus requires a target of the material of interest, a charge-reset foil at an appropriate distance from the target, a mass-analysing recoil separator with a good ion mass-to-charge resolution to measure the charge-state distribution, and a γ -ray spectrometer to selectively identify the population of the state(s) of interest (see Methods Fig. 4).

The proof-of-principle experiment was performed at the ATLAS facility of Argonne National Laboratory, where a ^{40}Ar heavy-ion beam was accelerated and impinged on a $613\text{-}\mu\text{g}/\text{cm}^2$ thick

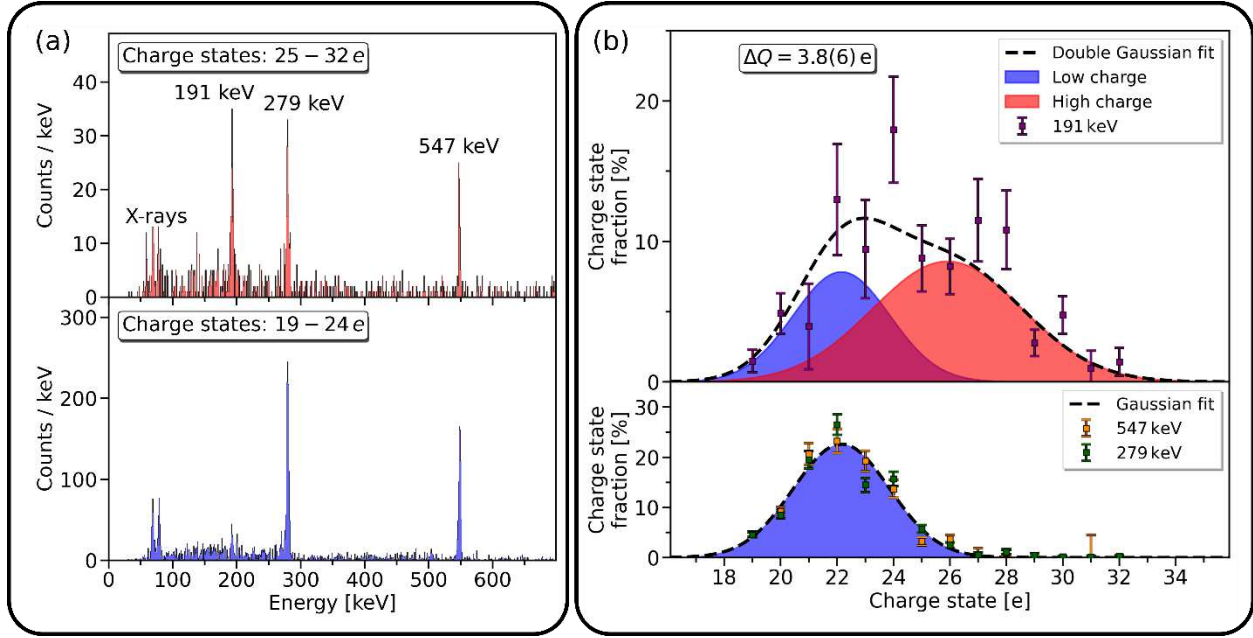


Figure 2. a) Gamma-ray spectrum coincident with ^{197}Au heavy ions for high-charge states (top) and low-charge states (bottom). The intensity of the 191-keV γ ray, relative to the 279- and 547-keV γ rays, is much stronger when in coincidence with the high-charge states. b) Charge-state distributions for the 191-keV transition to the 77-keV state of interest (top) and for transitions directly feeding the ground-state (bottom).

self-supporting ^{197}Au target. Two different beam energies were used to investigate different initial charge states of the recoiling ion. Following interaction with the ^{40}Ar beam, the ^{197}Au nuclei recoiled in the forward direction with velocities of about 9 mm/ns and 7 mm/ns respectively. The Coulomb interaction between the beam and target nuclei populated excited states of the ^{197}Au recoils. A $61 \mu\text{g}/\text{cm}^2$ carbon charge-reset foil was placed 5 mm downstream of the target, equivalent to about 0.5-0.7 ns of flight time for the scattered ^{197}Au recoils. After the charge-reset foil, the recoils were selected by their mass-to-charge ratio (M/Q) by the Fragment Mass Analyzer (FMA) (Davids and Larson 1989). The γ -ray energy tracking in-beam nuclear array (GRETINA) (Paschalis et al. 2013) surrounded the target and reset foil, and was used to detect prompt γ rays emitted near the target.

In the case of ^{197}Au (Huang and Zhou 2005), the excited state of interest lies at 77.3 keV, and decays to the ground state with a 1.91 ns half life. There is an 81% probability that the decay proceeds via internal conversion rather than γ -ray emission (Kibédi et al. 2008). The charge-state distribution following the de-excitation of the 77-keV level can be isolated through its population from above, via an excited state at 268.8 keV followed by the emission of a γ ray with energy 191.4 keV. This γ ray can clearly be seen in coincidence with high-charge state ions in the top panel of Fig. 2 (a), while it is almost absent in coincidence with low-charge state ions, shown in the bottom panel. This indicates that the decay of this state is associated with higher charge state recoils, the population of higher charge states arising from the emission of the internal-conversion electron plus the subsequent Auger-electron cascade. By extracting the intensity of this 191-keV γ ray as a function of M/Q , one can deduce the charge-state distribution associated with the decay of the 77-keV state, shown in the top panel of Fig. 2 (b). This is clearly distinct from charge-state

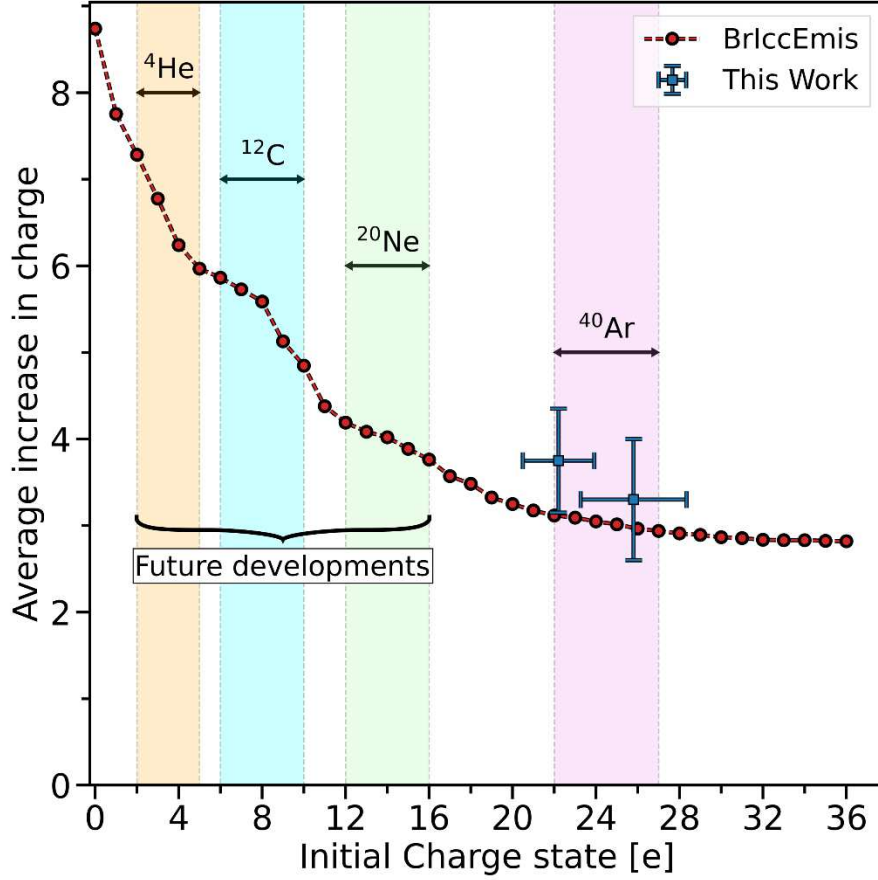


Figure 3. Average increase in charge for ^{197}Au plotted against the average initial charge state. Experimental data ("This Work") are shown, alongside calculated values from BrIccEmis. Vertical coloured bands indicate the approximate range of initial charge states that would be populated using the present method with different incoming beams.

distributions (shown in the bottom panel of Fig. 2 (b)) coincident with γ -ray decays directly to the ^{197}Au ground state, with no associated Auger cascade.

We have thus demonstrated the Auger multiplicity associated with the internal conversion of the 77-keV state in ^{197}Au . Due to the use of two different beam energies, we can investigate this multiplicity as a function of the initial charge state. For an initial low-charge distribution with mean $\bar{Q}_L=22.2(1)$ e, the average increase in charge was found to be $\Delta Q=3.8(6)$ e, including the emission of the conversion electron, whereas for an initial charge-state distribution with mean $\bar{Q}_L=25.8(2)$ e it was found to be $\Delta Q=3.3(7)$ e. To relate our observed results to those of a neutral atom - as relevant for AET - we make use of the BrIccEmis model, which allows the initial charge-state distribution to be accounted for. Having benchmarked BrIccEmis against the measured highly ionized systems, the model predicts an average charge increase of 8.7 electrons for a neutral ^{197}Au atom. Accounting for the internal-conversion electron, this leads to an average Auger emission multiplicity of 7.7 for a single atomic cascade. We highlight that the present method is an ideal laboratory for testing this model due to the measurement being performed in vacuum and thus excluding any interference from interactions with matter. A more realistic calculation would have

to assume multiple cascades emitted from ions present within a suitable medium (e.g. water). In such a case, electron filling from the medium into vacant outer-shell orbitals can significantly increase the Auger-electron multiplicity. Such calculations can be straightforwardly accounted for within the BrIccEmis framework. The calculations pushed the BrIccEmis model to the limit, beginning with an ion in a highly charged state and requiring more than fourteen million calculated atomic configurations. The results of the present study and those from BrIccEmis are compared in Fig. 3. Clearly, the scale for the Auger-emission multiplicity is well reproduced by the BrIccEmis calculations.

The present method can be readily deployed at laboratories around the globe, providing a route for dramatically improving Auger multiplicity data. One improvement would be to reach lower initial charge states, reducing the systematic uncertainty associated with extrapolation to neutral atoms. This can be achieved by reducing the energy of the incoming beam, as was done in this work, but also by using different mass beams to induce the backscatter. As the mass of the beam reduces, the velocity of the backscattered ions reduces, resulting in a lower equilibrated charge state. For a ^{12}C beam, for example, this would reduce the initial charge state of the ^{197}Au ions to less than 10+. The initial charge states that might be achieved in ^{197}Au with different choices of beam are shown in the filled bands of Fig. 3, clearly showing the potential of this method. We therefore see the technique as having the potential to completely resolve the question of Auger multiplicity within the next decade, eliminating it as a concern in the deployment of AET as a radiotherapy. Importantly, as an atomic process, the mass of the isotope alone is anticipated to have only a minor impact on the Auger-electron multiplicity for a single atomic cascade. Experimenters will thus be free to select the most favourable candidate isotope of the element of interest, further simplifying the method.

Here, we have presented a novel solution to the challenge of determining the emission multiplicities of Auger electrons for isotopes and elements relevant to Auger electron therapy. This method opens a new paradigm in the investigation of atomic structures with highly charged ions, and with the potential extension to radioactive species and heavy elements in which atomic spectroscopy is inherently challenging. We demonstrate this method in application to ^{197}Au , an isotope with medical relevance as the electron capture daughter of ^{197}Hg . We found average Auger multiplicities of $\Delta Q=3.8(6)$ e and $\Delta Q=3.3(7)$ e for initial average ^{197}Au charge states of $\widetilde{Q}_L=22.2(1)$ e and $\widetilde{Q}_L=25.8(2)$ e, respectively. We find that the state-of-the-art BrIccEmis model closely reproduces the observed results. The method presented is broadly applicable at ion-beam facilities across the world, requiring only the use of an M/Q-sensitive recoil separator and an array of HPGe detectors, both of which are commonplace at international heavy-ion beam facilities. We therefore propose that this new method represents a route towards a complete resolution of the Auger-electron multiplicity challenge.

Methods

Internal conversion and Auger emission

When an excited nuclear state decays, there is a probability that it will do so by the emission of an atomic electron, known as internal conversion. This process results in a vacancy in an inner atomic shell and therefore triggers an Auger cascade, providing the present method with a route to determine the Auger electron multiplicity. Crucial to extracting this information, however, is to isolate a single internal conversion process, ensuring that only a single Auger cascade occurs. To achieve this, the charge-reset method is used.

When an excited nuclear state decays via electromagnetic transitions, it might do so through multiple sequential processes, any of which could be an internal conversion. To isolate an individual internal conversion decay, we make use of the lifetimes of the nuclear state of interest. In the case of ^{197}Au , for example, the first excited state at 77 keV has a half-life of 1.9 ns, compared to half-lives of <20 ps for higher-lying states. Since the nucleus is in motion, one can equate this time to a distance. For example, in the space of 1.9 ns, the recoiling ^{197}Au ions will travel about 1.7 cm, whereas in 20 ps, the ions will travel less than 0.2 mm. If one can “reset” the charge of the ion at 1 mm or greater, one will be sensitive predominantly to charge-state increases from the decay of the 77 keV state of interest.

This is achieved with a thin carbon charge-reset foil located 5 mm from the target. As the ions pass through this foil, their charge states will alter through multiple electron stripping and pick-up interactions. This results in a charge-state distribution that is independent of the charge of the ions entering the reset foil.

From the above, one is able to select events that contain at most a single internal conversion process and thus, a single Auger cascade. One then measures the difference in charge-state distributions between states that have not undergone internal conversion (through selection of ions in their nuclear ground state) and those in which internal conversion (and thus Auger cascades) can occur post-reset. By doing this, one can access the average increase in charge and, thus, the Auger electron multiplicity. This is shown schematically in Fig. 4.

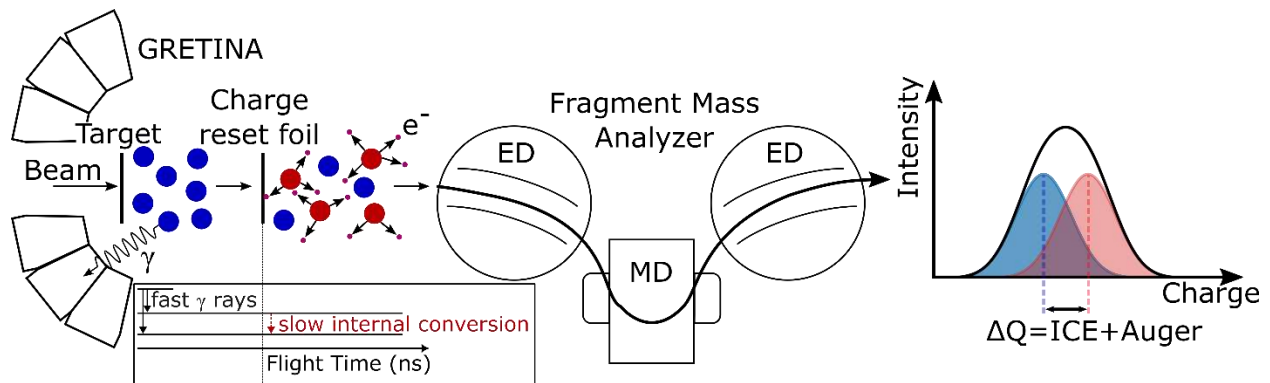


Figure 4. Experimental setup for the charge reset method. The ^{40}Ar beam produces forward-scattered target atoms. These ^{197}Au heavy ions recoil towards the mass analyzer. The combination of a γ -ray detector array, such as GRETINA, and the charge reset foil selects cases where internal conversion and Auger electrons are emitted.

To investigate how the Auger multiplicity changes for a different initial charge state, the experiment was split into two parts. In the first part, the ^{40}Ar beam particles were accelerated to an energy of 150 MeV, and in the second part, the ^{40}Ar beam was accelerated to an energy of 101 MeV. These two separate measurements are herein referred to as high-energy beam and low-energy beam, respectively. The high-energy beam produces ^{197}Au recoils with a higher velocity which, according to empirical formulas (Schiwietz and Grande 2001), leads to a higher average initial charge state.

Measuring ion charge states

The Fragment Mass Analyzer (FMA) device was used to measure the spectrum of ionic charge states of heavy-ion recoils of ^{197}Au . The FMA consists of focusing entrance and exit magnetic quadrupole doublets, two electric dipoles and a magnetic dipole magnet, which bend the path of the ion according to the Lorentz equation. The combination of electric and magnetic fields selects ions according to their energy-to-charge and momentum-to-charge ratios and disperses them at the focal plane of the separator by their mass-to-charge ratio. The FMA fields are set to select a reference particle with energy E_{ref} , mass M_{ref} and charge state Q_{ref} , which is transmitted to the centre of the focal plane of the separator, where it is detected. In this experiment, M_{ref} was set to 197 atomic mass units (u), and E_{ref} was set depending on the kinematics of ^{197}Au heavy ions that were scattered forward by the ^{40}Ar beam. When the ^{197}Au ions travelled through the separator, the charge state of the ion determined the horizontal distance from the centre of the focal plane where

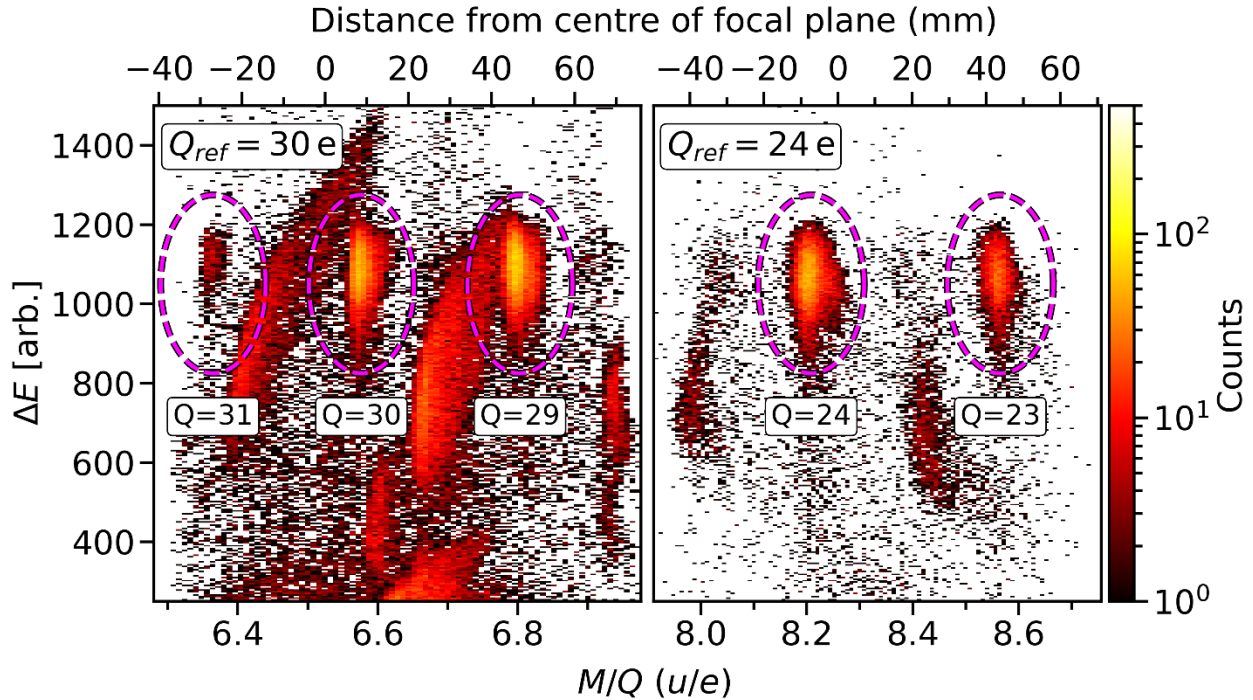


Figure 5. Typical FMA focal plane spectra showing the recoil energy deposited in the ionization chamber versus M/Q value determined by the position of the ion in the PPAC. The FMA electromagnetic fields were tuned to select ^{197}Au ions with reference charge states $Q_{\text{ref}}=24\text{ e}$ and $Q_{\text{ref}}=30\text{ e}$ respectively. The dashed magenta ellipses indicate the position ^{197}Au ions. Adjacent charge states of $Q=23\text{ e}$, $Q=29\text{ e}$, and $Q=31\text{ e}$ can also be seen, along with heavy-ion contaminants in the beam.

the ion was detected. A position-sensitive parallel-plate avalanche counter (PPAC) was used to measure the position of ^{197}Au ions at the focal plane of the FMA. An ionization chamber was placed behind the PPAC to measure the energy of the ions. The combination of the PPAC and the ionization chamber offered a highly selective method of identifying ^{197}Au recoils. Due to the finite length of the separator and size of the focal plane, a typical mass-to-charge spectrum from the FMA consists of only a portion of the charge-state distribution (~ 2 -5) charge states. To observe the entire ^{197}Au charge-state distribution, the total measurement time was divided into segments, where, for each segment, the FMA electromagnetic settings of the separator were tuned for a different Q_{ref} . Example spectra for ^{197}Au ions arriving at the FMA focal plane are shown in Fig. 5 for cases where the separator fields were set to select ^{197}Au ions with reference charge $Q_{\text{ref}}=30$ e and $Q_{\text{ref}}=24$ e. The separator was used to scan along the charge-state distribution, applying an appropriate normalisation value to account for changes in measurement time and beam intensity.

Normalisation

When measuring the ionic charge-state distribution, it is critical to be able to normalise each data point according to their integrated exposure. This can vary throughout an experiment due to fluctuations in beam intensities or stoppages due to instrument failures, among other things. The experimental data here are normalised according to three different metrics:

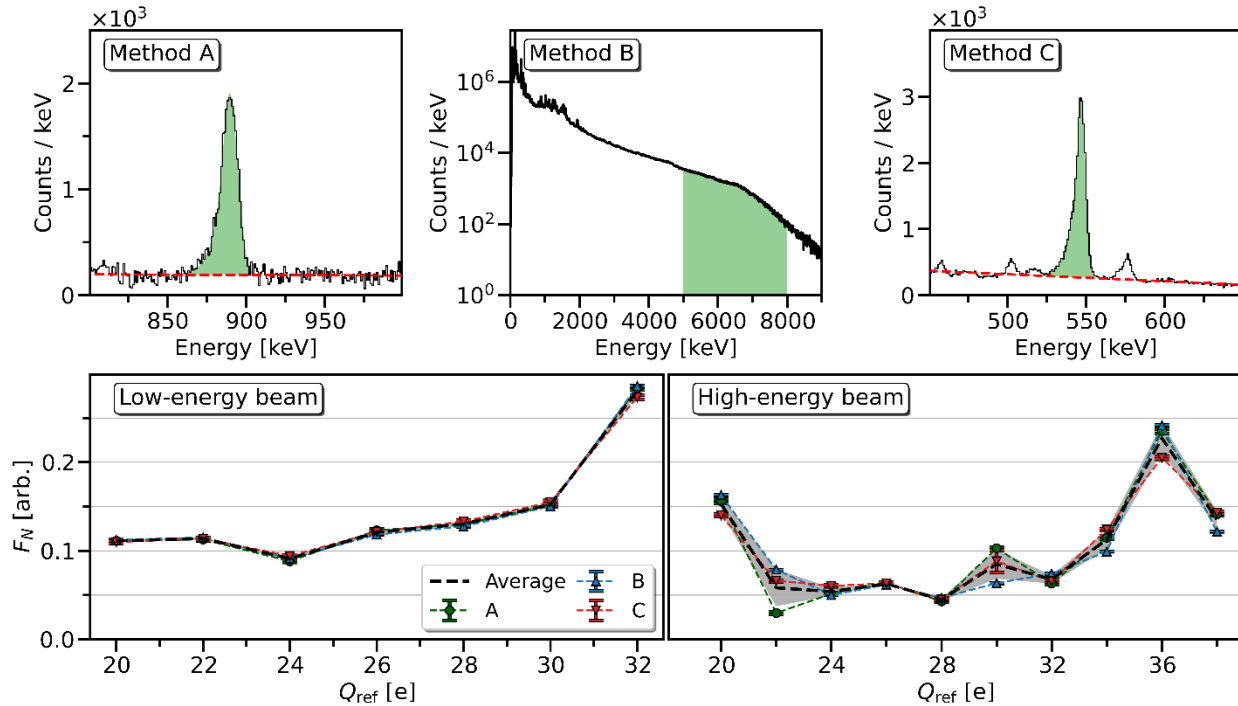


Figure 6. The top panels show the different methods, A, B and C, used to normalise the data points obtained in the charge-state distribution scans. The bottom panels show the normalisation values, F_N , obtained from these three methods, along with the average value (black dashed line) and standard deviation (grey band) that are used to produce the charge-state distribution spectra observed with FMA. For the high-energy beam measurement, there is a significant spread in F_N at $Q_{\text{ref}}=22$ e, 30 e, and 36 e which contributes to larger uncertainties in these data points. See text for details on the different methods used.

- A. Gamma-rays produced in nuclear reactions between the ^{40}Ar beam and the ^{12}C reset foil. Specifically, the 889-keV γ -ray emitted in the decay of the first-excited state of ^{46}Ti was used.
- B. The integrated yield of high-energy γ -rays between 5-8 MeV observed in GRETINA. These γ -rays are too high-energy to originate from terrestrial sources and must therefore be produced by beam-induced reactions.
- C. Coincidences between backscattered ^{40}Ar particles and ^{197}Au γ -rays observed in the data with no charge-state coincidence required. The backscattered ^{40}Ar particles were detected using a Micron silicon S3 detector (Micron, 2019) that was positioned upstream of the target. The ^{197}Au γ -ray energies were Doppler corrected based on the kinematics determined by the detected ^{40}Ar particles. Specifically, the 547-keV transition in ^{197}Au was used.

Examples of methods A, B and C are shown in the top panel of Fig. 6. The normalisation factor, F_N , that results from these three different strategies are shown in the lower panel of Fig. 6, for the low- and high-energy beams, as a function of Q_{ref} . The values of F_N that were used to normalise the data points in the charge-state distribution were calculated using the average of methods A, B and C. The uncertainties on the values of F_N were calculated by combining the statistical uncertainties from methods A, B and C with the standard deviation of the three methods in quadrature. This strategy accounted for systematic uncertainties arising from the choice of normalisation method.

Charge-state distribution measurements

Charge-state distribution data are shown for the low-energy beam Figs. 7(a) and 7(b), and the high-energy beam Figs. 7(c) and 7(d). The charge-state distribution of ^{197}Au recoils correlated with the 191-keV γ -ray transition that feeds the 77-keV state of interest are shown in Figs. 7(a) and 7(c). The equivalent charge-state distribution for ^{197}Au recoils that are correlated with the 279-keV or 547-keV γ -ray transitions that directly feed the ground state, and thus only contain a low-charge component are shown in Figs. 7(b) and 7(d). In Figs. 7(a) and 7(b) the corresponding paths in which excited states decay to the ground state are shown inset. For each charge state, the intensity of the γ -ray transition is divided by the relevant normalisation factor, F_N . This is then divided by the sum over the distribution so that each value represents the fractional percentage of ^{197}Au recoils in that charge state. The uncertainties are calculated by combining in quadrature the statistical uncertainty of the γ -ray transition intensity with the uncertainty on F_N .

A single-Gaussian lineshape is fit to the data in Figs. 7(b) and 7(d) to obtain the centroid ($\widetilde{Q}_L$) and width (σ_L) of the initial low-charge component. A double-Gaussian lineshape is fit to the data in Figs. 7(a) and 7(c), fixing the centroid and width of the low-charge component, to obtain the centroid ($\widetilde{Q}_H$) of the high-charge component. Systematic uncertainties on the parameterisation of the low-charge component are accounted for by repeating the fit for upper and lower bounds of $\widetilde{Q}_L$ and σ_L . A condition is placed so that the width (σ_H) of the high-charge component must be equal to or larger than σ_L . This is reasonable since the high-charge component arises from the combination of two distributions, i.e. the initial low-charge component and the internal conversion

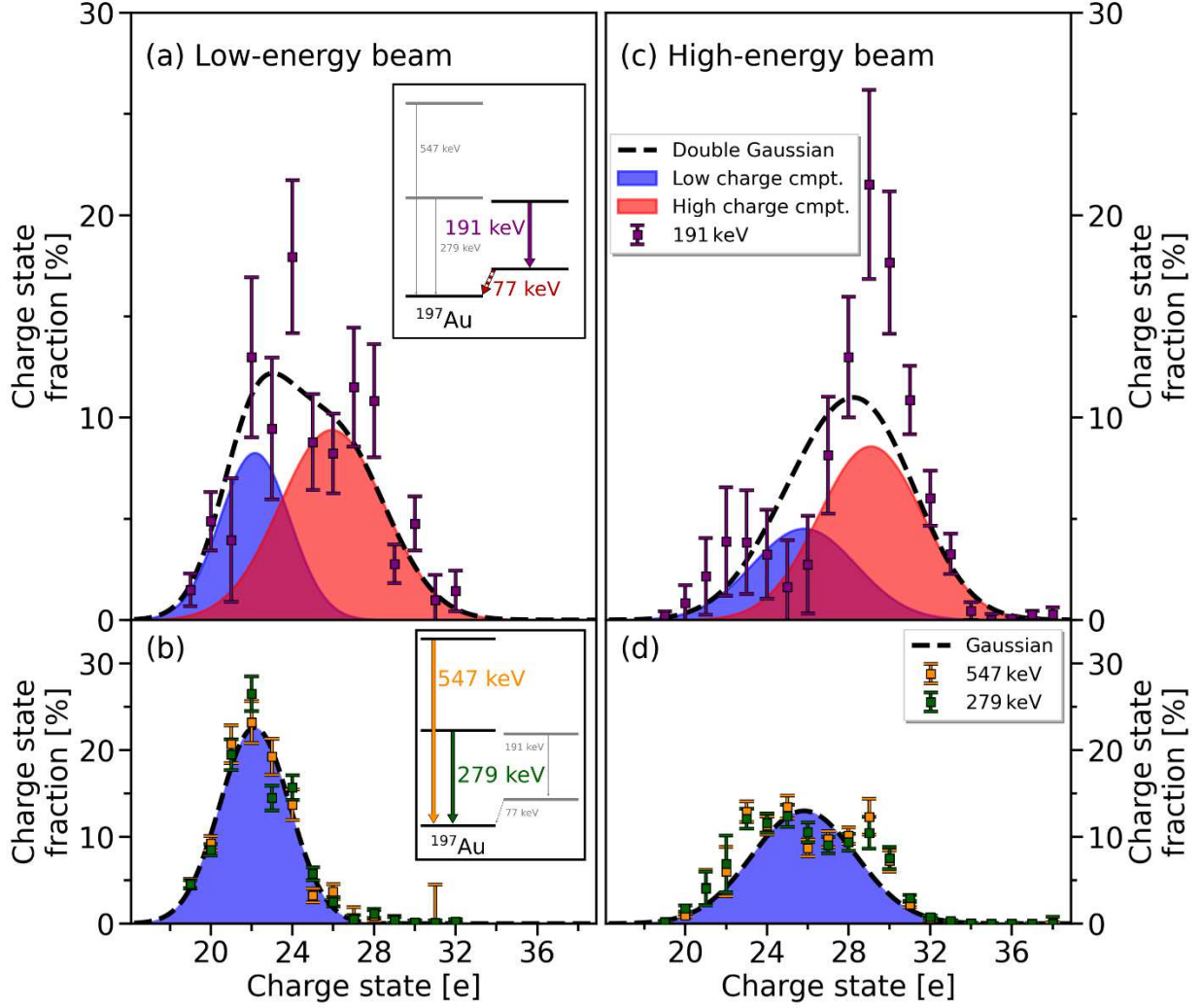


Figure 7. Charge-state distribution of ^{197}Au heavy-ions for (a) and (b) the low-energy beam, and (c) and (d) the high-energy beam. The distributions in (a) and (c) are coincident with the 191 keV γ ray that feeds the 77 keV state of interest. The distributions in (b) and (d) are coincident with the 279 keV or 547 keV γ -ray transitions that directly feed the ground state.

plus Auger-electron cascade. A further condition is placed so that the ratio of the amplitudes of the high-charge component to the low-charge components is,

$$\frac{A_H}{A_L} = \frac{P_{IC} \times P_{after}}{1 - P_{IC} \times P_{after}}, \quad (1)$$

where P_{IC} is the probability that the 77 keV state decays via internal conversion and P_{after} is the probability that the 77 keV state decays after the charge-reset foil, given by,

$$P_{after} = 2^{-\frac{t_f}{t_{1/2}}}. \quad (2)$$

Here, t_f is the flight time for ^{197}Au recoils to travel between the target and the charge-reset foil, and $t_{1/2}$ is the half-life of the 77-keV state. The value A_H in Eq. 1 accounts for ions that survive

beyond the charge-reset foil and subsequently decay by internal conversion, whereas A_L contains all other cases. The ratio $A_H/A_L = 1.9$ and 1.7 for the high-energy beam and low-energy beam, respectively. The increase in charge following internal conversion plus Auger-electron cascade is obtained via,

$$\Delta Q = \widetilde{Q}_H - \widetilde{Q}_L. \quad (3)$$

For low-energy beam measurement, values of $\widetilde{Q}_L=22.2(1)$ e, $\widetilde{Q}_H=26.0(5)$ e, are obtained. For the high-energy beam measurement, values of $\widetilde{Q}_L=25.8(2)$ e, $\widetilde{Q}_H=29.1(4)$ e, are obtained. The reduced chi-squared (χ_{red}) values obtained from fitting the charge-state distributions for heavy-ion recoils that are coincident with the 191 keV transition feeding the 77 keV state of interest (Fig. 7(a) and 7(c)) are $\chi_{\text{red}} = 1.5$ for the low-energy beam data and $\chi_{\text{red}} = 2.3$ for the high-energy beam data. Following the convention given by the Particle Data Group (Patrignani et al. 2016), we have inflated the uncertainties on ΔQ by $\sqrt{\chi_{\text{red}}}$. The resulting charge increase due to the internal conversion and following Auger cascade is therefore $\Delta Q=3.8(6)$ e for the low-energy beam measurement and $\Delta Q=3.3(7)$ e for the high-energy beam measurement.

BrIccEmis calculations

Nuclear and atomic decays of recoiling ^{197}Au ions have been simulated using the BrIccEmis code (Lee et al. 2012; Lee et al. 2016), originally developed for the calculation of the Auger electron and X-ray spectra following electron capture and internal conversion decays of isotopes with neutral atomic charges. For the present study, the model has been extended for ions.

The internal conversion process responsible for the creation of vacancies in the ^{197}Au recoils depends on the degree of atomic ionization. Theoretical internal conversion coefficients (ICC) and atomic binding energies have been calculated using the RAINE, relativistic Dirac-Fock model (Band et al. 2002, Kibédi et al. 2008). The model was successfully applied to study the internal conversion decay of the metastable nuclear state in ^{192}Os in a hydrogen-like charge state (Akber et al. 2015). Atomic transition rates were obtained from the EADL data base (Perkins et al. 1991) and various corrections have been applied as described in (Lee et al. 2012). Atomic transition energies also depend on the ionisation state of the atom. These were calculated with the RAINE code, which - due to the omission of the quantum electrodynamic (QED) and Breit magnetic electron interaction - overestimates the transition energies. Semi-empirical corrections (Tee et al. 2020) were applied, which reduces the differences below a few eV.

The actual calculations have been carried out for a single nuclear transition and initial charge state. The initial charge state of Au ions was defined by removing electrons starting from the last atomic shell of a neutral gold atom. The initial nuclear decay event was either the emission of a γ -ray or an internal-conversion electron. In the former case, no new atomic vacancy is created, and the calculation was terminated. Internal conversion decay will primarily produce atomic vacancies on the inner shell. In the case of the 77 keV transition, no K-shell conversion is allowed. The exact location of the initial vacancy was randomly selected from the occupied atomic shells of the ion, using the sub-shell ICC's as weighting factors. As the ions were moving in vacuum, vacancies reaching the outer shell will remain unfilled and the vacancy cascade will be terminated.

The internal conversion decay was followed by an atomic cascade of Auger electrons and X-rays, which is a multi-step, highly stochastic process. The next step of the atomic cascade was selected randomly from the pool of energetically possible atomic transitions using the transition rates as weights. This involves a full atomic configuration calculation of the atomic binding energies and the re-evaluation of the atomic transition rates at every step of the atomic cascade. To optimize the Monte Carlo simulations, the results of the atomic configurations are accumulated in the memory and saved on to disk. During X-ray emission, the number of atomic vacancies will be retained. On the other hand, Auger electron emission creates an extra atomic vacancy. The atomic vacancy propagation will proceed until no further transition is possible. It should be noted that, due to the vacuum environment in which the ion is travelling, a vacancy propagated to the outer atomic

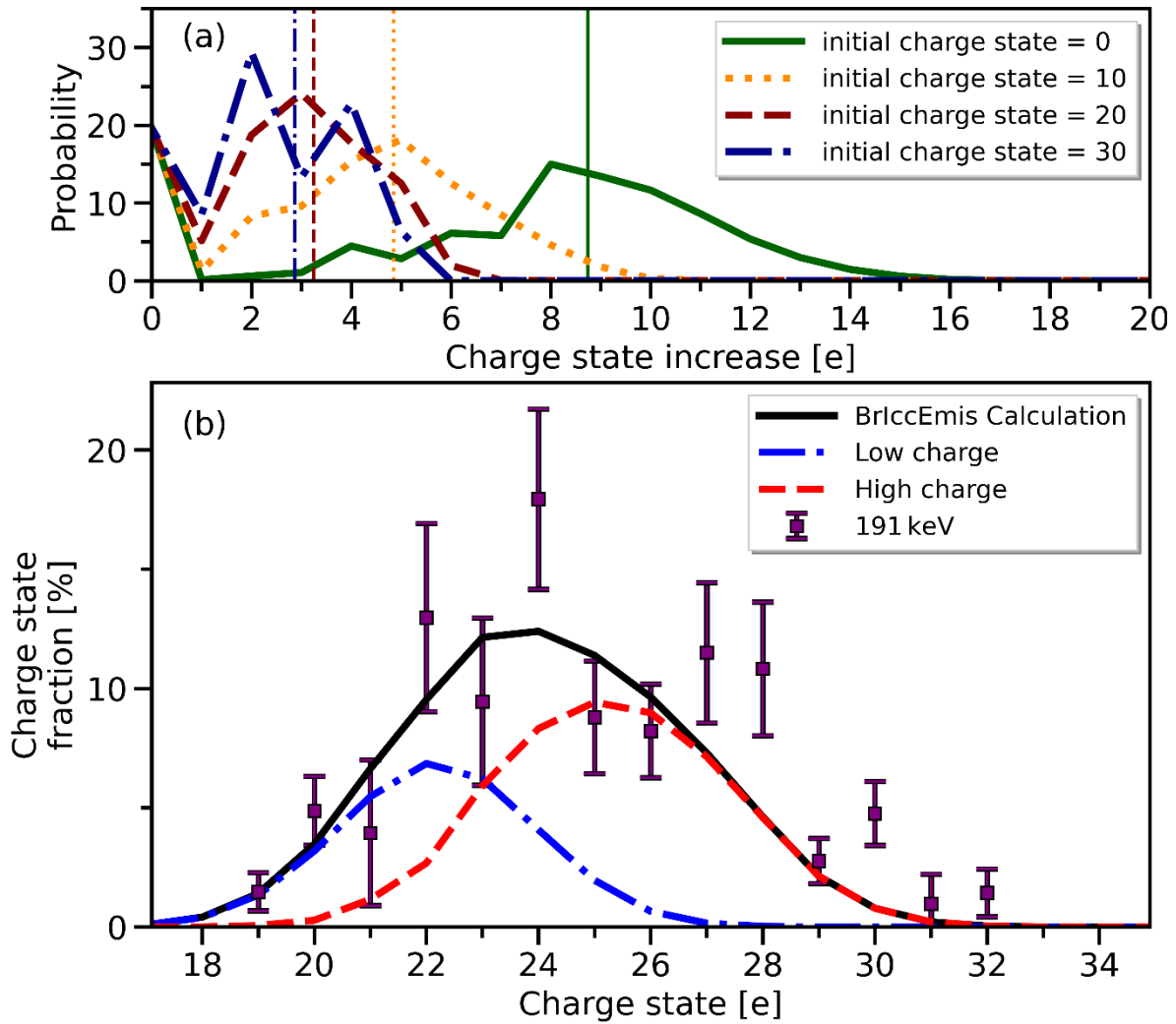


Figure 8. a) BrIccEmis calculations for the distribution of final charge states given an initial charge state of 0 e, 10 e, 20 e, and 30 e. Vertical lines give the average increase in ionic charge state when an internal conversion event occurs. b) The distribution of final charge states calculated using the BrIccEmis calculations is compared to the experimental charge-state distribution for the 191 keV transition to the 77 keV state of interest. The low- (corresponding to a charge increase of 0 e) and high-charge components of the BrIccEmis calculation are shown.

orbitals cannot be refilled from the environment. This is not the case for the radioactive decay of a medical isotope in a biological tissue, where a vacancy in the outer atomic shells can be readily filled and leads to an enhancement of the number of Auger electrons emitted during radioactive decay.

Every decay event, consisting of a γ ray or an internal conversion is written onto disk for offline processing. For internal conversion events only, the subsequent Auger- and X-ray emissions is also written to disk. The data collected from the simulation of 1 million nuclear decay events is used to produce energy spectra, transition energies and intensities for many atomic transitions, and a distribution of final charge states. The distribution of final charge states is shown in Fig. 8(a) for a selection of initial charge states, 0 e, 10 e, 20 e, and 30 e. A charge increase of 0 e corresponds to γ -ray emission.

To compare the BrIccEmis calculations with low-energy beam experimental data, the final distribution of charge states for each initial charge state are added together, with a weighting factor applied. The weighting factor is obtained from the experimental charge-state distribution shown in the lower panel of Fig. 2(b), where no internal conversion event occurs after the charge-reset foil. Fig. 8(b) shows the resulting charge-state distribution obtained by the BrIccEmis calculations compared to the experimental data for the high-energy beam measurement.

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Author contributions

J. Heery was the spokesperson for this experiment and was responsible for the conceptualization, proposal, and data analysis. J. Heery, J. Henderson, DGJ, and TK developed the interpretation of the results and prepared the manuscript. TK performed the BrIccEmis calculations. CMG, DS and MS were responsible for coordinating the apparatus used in the experiment. J. Heery, AE, CMG, VK, AK, NS, DS, MS and WR were responsible for preparing the experiment. JAMH, JH, DGJ, MC, CC, DD, JF, HJ, VK, FK, TL, GL, CMG, BSNS, EOS, WR, DS, MS and RSS performed the experimental data taking. All authors contributed to reviewing the manuscript.

Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.