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Half-life measurements of bare, mass-resolved isomers in a storage-cooler ring

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Abstract

Secondary beams from fragmentation of ^{58}Ni have been produced with energies of 200-220 A·MeV, separated with the fragment separator FRS and injected into the storage-cooler ring ESR for half-life measurements. The relative momentum spread $\Delta p/p \approx 1 \cdot 10^{-6}$ (FWHM) which we achieved allowed us to resolve the ground and isomeric states of cooled ^{52}Mn and ^{53}Fe nuclei in the measured mass spectra. The circulating beams were fully ionized which rendered possible, for the first time, to measure *pure* β^+ branches for ^{52g}Fe and ^{53g}Fe and the sum of pure β^+ and γ branches in the decay of the isomers ^{52m}Mn and ^{53m}Fe .

The β -decay probability of a nucleus is significantly affected by its ionic charge state. Striking examples are orbital electron capture (EC) which depends simply on the density of all bound electrons at the site of the nucleus, or the recently observed bound-state β^- decay of ^{163}Dy [1]. The number of bound electrons has implications for nucleosynthesis that occurs in stellar plasmas at high temperatures where a high degree of ionization prevails. Until recently such phenomena have been studied in highly ionized atoms including hydrogen-like ones [2, 3, 4], but not in bare ions. Bare radioactive nuclei can be generated by projectile fragmentation at relativistic energies, separated in flight and subsequently injected into a storage-cooler ring, where the specific decay properties can be studied over many hours.

In this Letter we report on half-life studies of fully stripped ions from fragmentation of ^{58}Ni beams. In several cases, β^+ decay was disentangled from the competing EC branch. Furthermore, by virtue of the experimental storage ring ESR [5] as a precision mass spectrometer, the ground- and isomeric states of

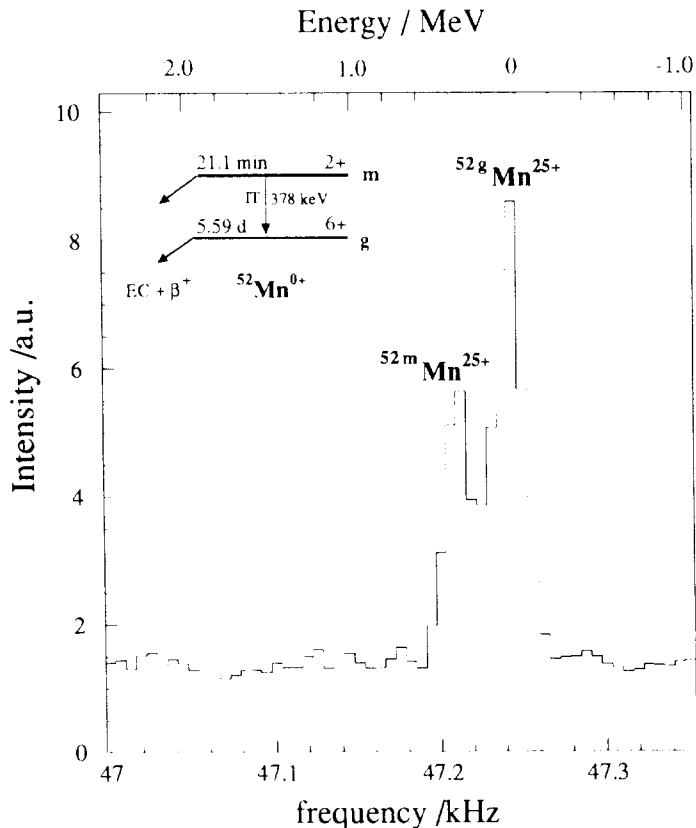


Figure 1: Schottky spectrum of fragments stored and cooled in the ESR, recorded at the 21th harmonic of the revolution frequency and averaged over 40 seconds. The frequency scale denotes the difference to an admixed external frequency of 33.15 MHz. The mass-resolved lines of the ground and isomeric states ($E^* = 378$ keV) of bare ^{52}Mn show a width of 21 Hz (FWHM), corresponding to a relative momentum spread of $\Delta p/p = 1.2 \cdot 10^{-6}$. The partial level scheme shown corresponds to neutral ^{52}Mn [9].

^{52}Mn and ^{53}Fe were mass-resolved and the corresponding half-lives determined separately.

Experiments with secondary nuclear beams at relativistic energies are performed at GSI with the heavy-ion synchrotron SIS [6], the fragment separator FRS [7], and the storage ring ESR. The most attractive tool in the ESR is an electron cooler that drastically increases the phase-space density of circulating beams offering unprecedented conditions for precision experiments.

In a first experiment with stored and cooled ^{20}Ne fragments we recently demonstrated the potential of cooled secondary beams to measure half-lives and masses of light nuclides [8]. With improved mass resolution we have now extended the mass region to $A \approx 50$ by studying bare fragments.

The fragments were produced by a 360-380 A·MeV ^{58}Ni beam in a 4 g/cm²

beryllium target placed at the entrance of the FRS (see Fig. 1 of Ref.[8]). The primary energy was chosen such that the magnetic rigidity of the nuclei injected into the ESR was always fixed at 4.5 Tm corresponding to 200-220 A·MeV.

The FRS was operated without profiled degraders in the intermediate focal plane which are normally used to provide monoisotopic secondary beams [7, 10]. In this experiment, a pure magnetic analysis of the fragment beam transmitting several nuclei with similar mass-to-charge ratios M/q is more advantageous: for mass measurements, nuclei with known masses contained in the fragment mixture can serve for calibration purposes; for half-life measurements, the method allows the half-lives of a series of nuclei to be measured simultaneously.

In the ESR electron cooling is applied to reduce the relative momentum spread of the stored fragment beam. With electron densities of typically $10^6/\text{cm}^3$ a relative momentum spread of $\Delta p/p \approx 10^{-6}$ can be achieved. Due to the cooling process, the mean velocities of all fragments are the same. The lengths of the closed orbit and, consequently, the revolution frequencies depend on the mass-to-charge ratio M/q , and the frequencies can be measured by Schottky noise spectroscopy.

The Schottky technique is based on current signals induced at pickup electrodes by the circulating ions [11]. The revolution frequency (in our experiment we used the 21th harmonic) was mixed with an external frequency to shift the frequency band to the range of 0-100 kHz before being processed by fast Fourier transformation. Examples of Schottky spectra are presented in Fig. 1 for the case of ^{52}Mn and in Fig. 2 for fragments with $A/Z = 2$.

For cooled beams, the Schottky spectra exhibit well-resolved peaks corresponding to the different M/q values of the fragments. The separation of two neighbouring peaks in the frequency spectra, Δf_{12} , is related to the (M/q) -difference by

$$\frac{\Delta f_{12}}{f_1} = -\frac{1}{\gamma_t^2} \cdot \frac{\Delta(M/q)_{12}}{(M/q)_1} \quad , \quad (1)$$

where γ_t is an ion-optical parameter characterizing the operation mode of the ESR. In our case we used an ionoptical setting with $\gamma_t = 2.6$. The width Δf_1 of a single peak in the frequency spectrum is determined by the velocity spread Δv_1 of the corresponding ion via

$$\frac{\Delta f_1}{f_1} = \left(1 - \frac{\gamma^2}{\gamma_t^2}\right) \cdot \frac{\Delta v_1}{v_1} \quad , \quad (2)$$

where γ is the Lorentz factor. The masses of two nuclei can be resolved if $\Delta f_{12} > \Delta f_1$.

Since our first experiment [8] the mass resolution has been improved by a factor of 6 to a value of $\Delta M/M = 4 \cdot 10^{-6}$ so that ^{52}Mn and ^{53}Fe ground and isomeric states can now be resolved. Fig. 1 demonstrates that this is possible even for $^{52\text{m.g}}\text{Mn}$, where the isomeric state is only 378 keV higher than the ground state. From the areas of the doublets in the Schottky spectra, the production ratios in the fragmentation reaction for the isomeric and the ground state, $\sigma^{\text{m}}/\sigma^{\text{g}}$, can

be derived (see below). For $^{52m,8}\text{Mn}$ (where the isomer has a low spin of $I = 2$ compared to $I = 6$ for the ground state) the result is $\sigma^m/\sigma^g = 0.62(3)$. For $^{53m,8}\text{Fe}$ (where the isomer is a high-spin state with $I = 19/2$ and the ground state has a spin of $I = 7/2$) we obtain a much smaller value of $\sigma^m/\sigma^g = 0.106(5)$. This indicates a preference for low-spin states to be populated in high-energy fragmentation.

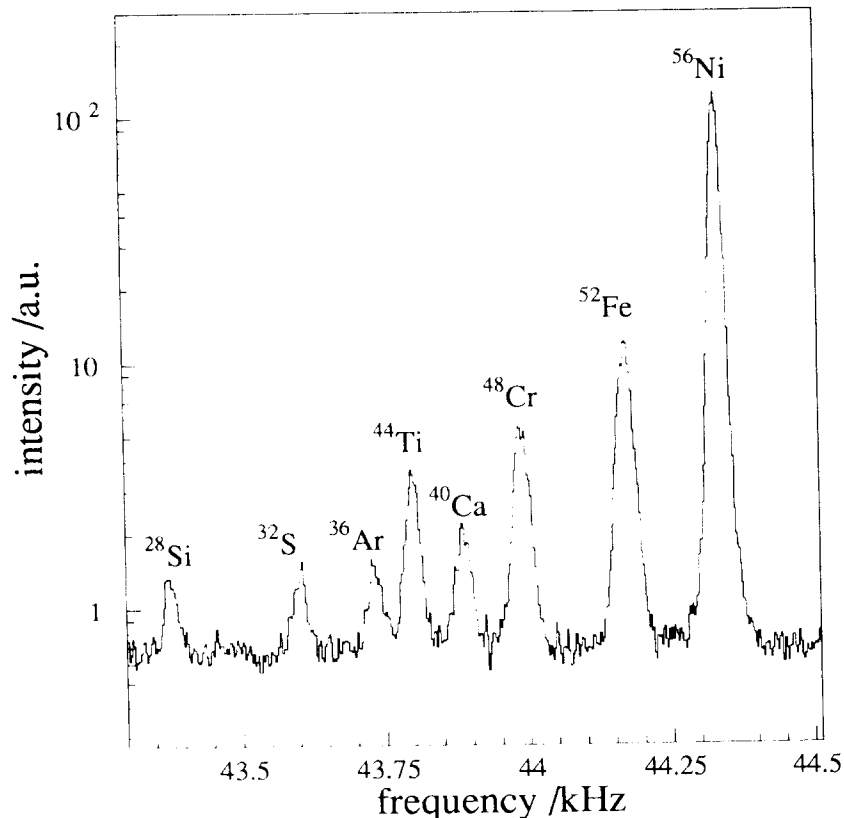


Figure 2: Frequency spectrum of bare $A/Z = 2$ fragments, averaged over 1 minute and recorded about 15 minutes after injection when the shorter-lived odd Z nuclides had decayed. The resolution is similar to that of the spectrum shown in Fig. 1.

In Fig. 2 we show a frequency spectrum of bare fragments with $A/Z = 2$. This spectrum was recorded 15 min after injection, so that short lived nuclei have already disappeared. Typically 10^3 to 10^4 fragments were stored in the ring. Taking the nuclides ^{44}Ti and ^{56}Ni as references, i.e. assigning to the corresponding peaks absolute masses from the compilation of Audi and Wapstra [12], the masses of ^{40}Ca , ^{48}Cr , and ^{52}Fe can be determined. The resulting values agree within an error of 20 keV with those given in Ref. [12] (quoted errors ≤ 10 keV). In this analysis the total energy of all bound electrons was taken into account. Since the separation in this $A/Z = 2$ cut directly reflects the

nuclear binding energy, effects such as the inversion of ^{40}Ca and ^{44}Ti due to shell closure are visible. The mass calibration yielded for the mass difference between the ground and isomeric states of $^{52}\text{Mn}^{25+}$ (Fig. 1) a value of 363 ± 20 keV, in good agreement with the literature value of 378 keV [9].

By using *fully ionized* fragments it is possible, for the first time, to study the β^+ decay alone. Note that under these conditions also internal conversion (IC) of isomeric transitions is also impossible. To detect β^+ decay in the ESR, one makes use of the fact that the decay alters the ionic charge state q by one unit. Therefore, the daughter ions leave the closed orbit and may hit a detector placed at a well defined position in the ring aperture. This method was used, e.g., to study bound-state β^- decay of ^{163}Dy [1].

In this experiment, however, we recorded the areas of the Schottky frequency spectra of the *mother* nuclei as a function of time. As the areas are proportional to the particle numbers, the half-lives of the corresponding ions can be determined and Lorentz-transformed from the laboratory frame to the ion rest frame. For each of the separate runs we have deduced the Lorentz factor γ of the circulating fragments from the terminal voltage of the cooler with a relative error of $\Delta\gamma/\gamma < 10^{-3}$.

During storage and cooling the intensity of radioactive nuclei in the ESR does not only change because of nuclear decay but also because of atomic interactions with the cooler electrons and with the atoms of the residual gas. Since the *bare* ions $^{36}\text{Ar}^{18+}$, $^{40}\text{Ca}^{20+}$, $^{44}\text{Ti}^{22+}$, $^{48}\text{Cr}^{24+}$, and $^{56}\text{Ni}^{26+}$ in Fig. 2 are essentially stable with respect to nuclear decay, i.e. $\lambda_{\text{nuc}} \approx 0$, their observed decay constants $\lambda_{\text{obs}} = \lambda^* + \lambda_{\text{nuc}}$ render directly the corresponding beam-loss constants λ^* due to atomic interactions. We assume that the measured values of λ_{obs} shown in Fig. 3 have a Z -dependence of $\lambda^* \propto Z^x$. From a fit to the data of Fig. 2 we obtain $x = 2.02$ if we exclude the data point for $^{52}\text{Fe}^{26+}$. This value agrees fairly well with the quadratic Z -dependence expected if radiative electron capture (REC) in the cooler is the dominant beam-loss process [13]. For $^{52}\text{Fe}^{26+}$, the fitted curve of Fig. 3 yields an interpolated value of $\lambda^* = 0.058(5)\text{h}^{-1}$. This value is subtracted from $\lambda_{\text{obs}}(^{52}\text{Fe}^{26+})$ to yield the β^+ decay constant in the laboratory system, $\lambda_{\text{nuc}}(^{52}\text{Fe}^{26+}) = 0.045(5)\text{h}^{-1}$. The β^+ decay half-life for $^{52}\text{Fe}^{26+}$ in its rest frame, $T_{1/2}^{\text{exp}} = (12.5_{-1.2}^{+1.5})$ h, is obtained by Lorentz transformation using $\gamma = 1.235(1)$.

Table 1 compares the experimental half-lives to those of the neutral atoms for ^{52}Mn (Fig. 1), ^{52}Fe , and $^{53\text{m,g}}\text{Fe}$ (Fig. 4). The table includes furthermore the expected half-lives of the bare nuclei derived from available spectroscopic informations. This calculated half-life of a bare nucleus in its rest frame is given by:

$$T_{1/2}^{\text{calc}}(\text{bare}) = T_{1/2} \cdot \left(\sum (I_{\beta}/\epsilon_f) + \sum I_{\gamma} \right)^{-1} \quad (3)$$

$T_{1/2}$ is the total half-life of the neutral atom, $\sum I_{\beta}$ and $\sum I_{\gamma}$ are the summed intensities per decay of all β^+ and γ branches, and $\epsilon_f = f^+(\text{neutral})/f^+(\text{bare})$ is the ratio of the β^+ -decay Fermi function for neutral and bare atoms, respectively [15].

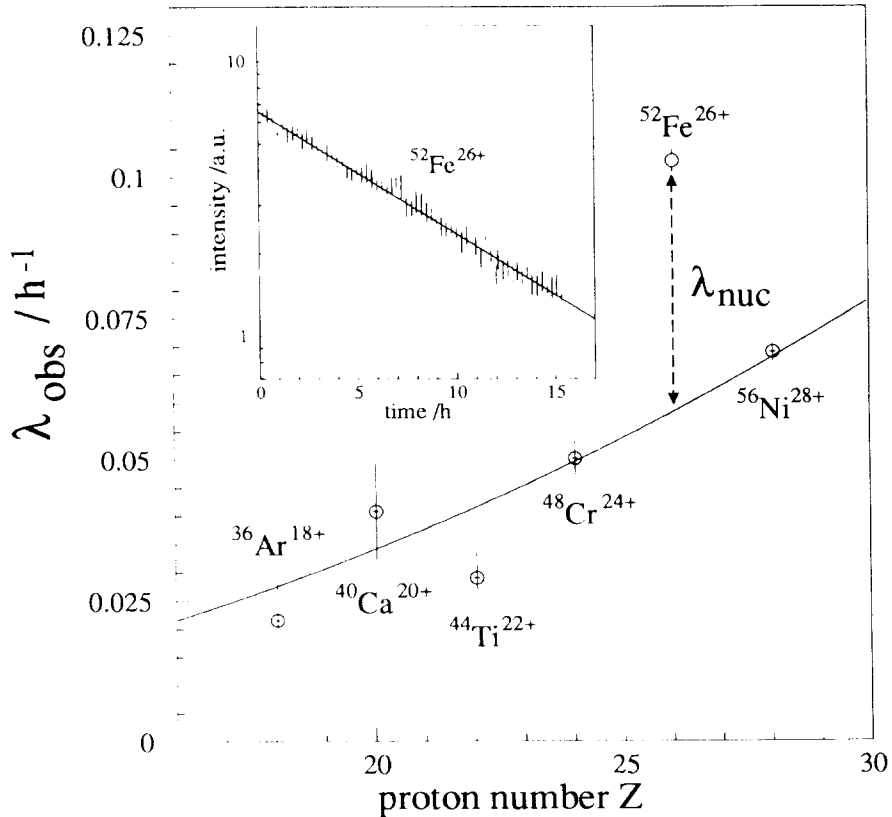


Figure 3: Observed total decay constants λ_{obs} of stored bare ions from the $A/Z = 2$ cut (Fig. 2). The full line is a fit of a $\lambda^* \propto Z^x$ dependence for atomic beam losses to all data, except $^{52}\text{Fe}^{26+}$, which is the only isotope in the series where the nuclear decay constant λ_{nuc} is comparable to λ^* ($\lambda_{\text{obs}} = \lambda^* + \lambda_{\text{nuc}}$). The insert shows the decay curve of $^{52}\text{Fe}^{26+}$, observed over more than 15 hours.

The decay curves for $^{53\text{m,g}}\text{Fe}$ are shown in Fig. 4. Whereas the decay of $^{53\text{m}}\text{Fe}$ shows only a single component, the decay of $^{53\text{g}}\text{Fe}$ also contains feeding by the isomeric transition. Therefore the half-life of $^{53\text{g}}\text{Fe}$ was obtained in a separate run after the isomer had decayed. The results were then used to calculate the isomeric production ratio $\sigma^{\text{m}}/\sigma^{\text{g}} = 0.106(5)$ mentioned above, taking into account a 100 % isomeric γ transition. With $\gamma = 1.227(1)$ the experimental half-lives $T_{1/2}^{\text{exp}}(\text{bare})$ given in rows 3 and 4 of Table 1 were obtained. In view of the short β^+ decay half lives, the losses due to REC in the cooler were neglected. The time dependence of the $^{52\text{m,g}}\text{Mn}$ Schottky peak areas was fitted by single exponentials, neglecting the weak ($\leq 2\%$) isomeric γ transition. The decay curve of $^{52\text{g}}\text{Mn}^{25+}$ with a neutral-atom half-life of 5.59 d is solely determined by REC in the cooler and therefore allows us to determine λ^* for this run. The resulting value together with $\gamma = 1.219(1)$ is then used to deduce for $^{52\text{m}}\text{Mn}^{25+}$ a half-life of $T_{1/2}^{\text{exp}}(\text{bare}) = (22.7 \pm 3.0)$ min.

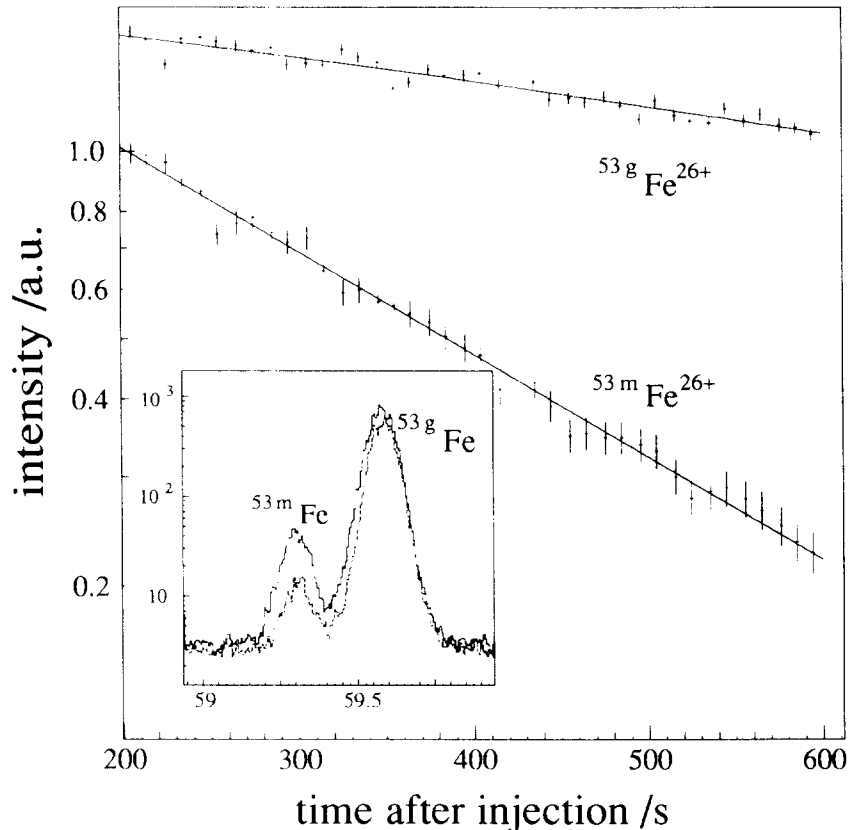


Figure 4: Schottky peak areas of $^{53m,g}\text{Fe}$ as a function of the time after injection. The insert shows the Schottky spectra after 200 s and after 600 s. The mass resolution (FWHM = 760 keV) is sufficient to resolve the $7/2^-$ ground state and the $19/2^-$ isomeric state at 3040 keV excitation energy.

For the cases of bare ^{52m}Mn and $^{53m,g}\text{Fe}$ the predicted half-lives $T_{1/2}^{calc}(\text{bare})$ from Eq. (3) are less than 4 % longer than the values for neutral atoms, because the EC and IC branchings and the screening corrections are small. Within the uncertainties they agree with the experimental values $T_{1/2}^{exp}(\text{bare})$ listed in Table 1.

This is not the case, however, for bare $^{52}\text{Fe}^{26+}$. For this isotope we have measured a half-life $T_{1/2}^{exp}(\text{bare}) = (12.5^{+1.5}_{-1.2})$ h. From this we deduce according to Eq. (3) a β^+ branching of $I_\beta = 0.67(8)$ and an EC branching of $I_{EC} = 0.33(8)$ for the *neutral* atom, using the appropriate values for the half-life of the neutral atom ($T_{1/2} = 8.275(8)$ h), for the Fermi correction factor ($\varepsilon_f = 1.015$) and for the γ -branch ($I_\gamma = 0$). This result differs significantly from the corresponding values quoted in the literature [9] ($I_\beta = 0.56(2)$, $I_{EC} = 0.44(2)$). The latter numbers were *calculated*, however, from the adjusted Q_{EC} value of 2372(12) keV [12], by assuming that the ^{52}Fe ground state decays by 100 % to the 1^+ level of ^{52}Mn at 546 keV. For such a simple case, there exists a

unique correspondence between the Q_{EC} value and the β^+/EC -ratio [15]. If we assume that the transition to the 1^+ level is the only decay channel, we get from our measured β^+ branching a value of $Q_{\text{EC}} = (2500_{-90}^{+100})$ keV, clearly in disagreement with Ref. [12]. A possible explanation for the discrepancy would be that the published level scheme [9] is incomplete, i.e. that additional β^+/EC decay channels must exist.

We have shown that the combination of the fragment separator FRS and the storage-cooler ring ESR provides a powerful tool for precise mass spectroscopy and lifetime measurements of exotic nuclei. Even more, it allows to mass-resolve ground states and isomers and to obtain valuable informations about specific decay branches.

For experiments of the type presented here, where the decreasing intensity of the stored mother nuclei is observed, beam losses due to atomic processes restrict the range of accessible half-lives to less than some hours. On the other hand, the need to first cool the unstable ions imposes a lower limit of about $T_{1/2} = 30$ s on nuclear half-lives that can be measured. It will be a challenge in future to observe directly in the Schottky spectra the EC channel. That is feasible if the Q_{EC} value is not too small ($Q_{\text{EC}} \geq 1$ MeV for $A \approx 100$). In such cases mother and daughter atoms appear as separate peaks in the frequency spectrum, and the pure EC life-time can be derived directly from the growth of the daughter peak area. Moreover, life-times up to several years become accessible, provided that 10^6 to 10^7 mother nuclei can be stored in the ring. On the other hand shorter half-lives limited only by the flight times of the fragments can be measured when a monoisotopic beam from the FRS is stored in the ESR: in this case cooling is not essential to identify the stored isotope. The authors would like to thank the GSI accelerator staff for providing excellent ^{58}Ni beams. The continuous efforts of the ESR and FRS engineers were indispensable for the success of the program presented in this Letter.

Table 1: Literature values [9, 14] of neutral-atom total half-lives, $T_{1/2}^{\text{exp}}(\text{neutral})$, half-lives of the corresponding bare ions, $T_{1/2}^{\text{calc}}(\text{bare})$, calculated according to Eq. (3), in comparison with experimental results from this work for bare ions, $T_{1/2}^{\text{exp}}(\text{bare})$.

Nucleus	$T_{1/2}^{\text{exp}}(\text{neutral})$	$T_{1/2}^{\text{calc}}(\text{bare})$	$T_{1/2}^{\text{exp}}(\text{bare})$
$^{52\text{m}}\text{Mn}$	21.1(2) min	21.5(6) min	22.7(3.0) min
^{52}Fe	8.275(8) h	15.1(5) h	12.5($^{+1.5}_{-1.2}$) h
$^{53\text{g}}\text{Fe}$	8.51(2) min	8.73(8) min	8.5(3) min
$^{53\text{m}}\text{Fe}$	2.58(4) min	2.58(4) min	2.48(5) min

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