Search for the nuclear two-photon decay in swift fully-stripped heavy ions

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Abstract

The aim of this proposal is to search for the rare nuclear two-photon (2γ) emission in the decay of low-lying excited 0^+ states and to establish a new technique to search for low-lying 0^+ isomers. In a pilot experiment we plan to study the stable nucleus ^{72}Ge, i.e. the most easily accessible nucleus having a first excited 0^+ state below the pair creation threshold and to measure its partial life time for two-photon emission. In addition, we plan to search for a low-lying 0^+ state in ^{70}Se. The highly charged ions will be produced in-flight from a ^{78}Kr primary beam and separated using solely the ESR. This approach that bypasses the FRS has been successfully developed and used for several storage ring experiments of high-intensity beams of artificially synthesized isotopes and also isomers. The proposed experiment is unique for GSI/FAIR and will employ a range of instrumentation and methodology developed within NUSTAR/ILIMA.

We request a *total of 6 shifts of data taking* with a ^{78}Kr beam at 450 MeV/u to study both isotopes. In addition *6 shifts of set-up and commissioning time of the ESR is needed* in order to further develop the operation of the ESR in the isochronous mode. In both cases the direct beam from SIS18 can be used so that the FRS could be used in parallel.

The nuclear two-photon decay

The nuclear two-photon decay, also called two-gamma (2γ) decay, is a rare decay mode in atomic nuclei whereby a nucleus in an excited state emits two gamma rays simultaneously. The simultaneous emission of two photons as a second order quantum mechanical process was first treated for the case of atomic transitions by Göppert-Mayer [1] in 1931. First order processes usually dominate the decay by many orders of magnitude, but two-photon emission may become significant when first order processes are forbidden or strongly retarded.

Even-even nuclei with a first excited 0^+ state, such as ^{16}O, ^{40}Ca or ^{90}Zr are favourable cases to search for a 2γ decay branch, since the emission of a single gamma ray is strictly forbidden for $0^+ \rightarrow 0^+$ transitions by angular momentum conservation. The remaining first-order decay modes are the emission of atomic internal-conversion electrons (ICE) or internal positron-electron pair creation (IPC). For the IPC mode the excitation energy must exceed the rest mass of the pair of 1.022 MeV. The second-order 2γ decay proceeds through the virtual excitation of intermediate, higher-lying states. The sum energy of the two γ-rays must be equal to the transition energy, but the energy spectra of the individual gamma rays are continuous. Since the transition matrix elements are largest for low multipolarities, electric or magnetic dipole decays are predominant. The decays pass through a virtual excitation of intermediate 1^- or 1^+ states, which are usually located at (much) higher energy than the initial 0^+ state, i.e. in the Giant resonance region. Finally, it might be interesting to note that in a case where the ground and first excited state of a nucleus have spin zero but opposite parities the nuclear two-photon decay would be (amongst) the most probable decay modes leading to very long lived isomeric states [2].

The early theoretical treatment of the 2γ decay using second-order perturbation theory [3, 4] was completed by Friar et al. [5] and later generalised by considering not only dipole but also higher multipolarities by Kramp et al. [6]. The total 2γ decay rate can be expressed as:

$$\Gamma_{\gamma\gamma} = \omega^7/105\pi\,[\alpha(E1)^2 + \chi^2 + \omega^4\alpha(E2)^2/4752]$$

Here, ω is the energy difference of the initial and final state, while α denotes the (electric) transition polarizability and χ the (magnetic) transition susceptibility, which determine the probability for the emission of two E1 (or E2) or two M1 quanta, respectively.

The nuclear polarizabilities and susceptibilities describe the response of the nucleus to a perturbation by electromagnetic fields with frequencies small as compared to the characteristic nuclear transition frequencies. The *static* electric dipole polarizability of a nucleus in its ground state can be determined from the cross section measured in photo-nuclear reactions, while the magnetic dipole susceptibility can be deduced from (e,e') measurements. The 2γ decay on the other hand offers access to the *transition polarizabilities*, namely the electric dipole transition polarizability $\alpha(E1)$ and the magnetic dipole transition susceptibility χ.

Experimental Background and referencing previous experiments

Experimentally, many early attempts have been made to observe the 2γ decay often with conflicting results [7]. Most of the studies have concentrated on a few stable nuclei having a first excited 0^+ state at energies above 1.022 MeV (^{16}O, ^{40}Ca, ^{90}Zr). Due to the strong energy dependence a higher excitation energy increases the branching ratio, but the two-photon decay remains a very small decay branch ($\sim 10^{-4}$) competing with the dominant IPC (and ICE) modes. The positron created in the IPC mode will subsequently annihilate. In this process a pair of 511 keV gamma rays is created, but only if the annihilation takes place at rest. Otherwise the total energy, including the kinetic energy of the positron, is shared between the two gamma rays. Any experiment searching for the 2γ decay at energies well above 1.022 MeV must therefore discriminate against a continuous background originating from pair creation. The first conclusive experimental results were obtained about 30 years ago using the Heidelberg-Darmstadt Crystal Ball spectrometer, a highly selective 4π NaI(Tl) detection system, in order to identify the tiny 2γ decay branch. The two-photon decay probability has so far only been measured for the $0^+_2 \rightarrow 0^+_1$ transitions in ^{16}O [6], ^{40}Ca [6,8] and ^{90}Zr [6,8]. More recently, also the competitive 2γ decay was observed in the decay of the $11/2^-$ isomer in ^{137}Ba in an experiment using the fast-timing method at TU Darmstadt [9].

The most surprising result obtained in the detailed investigation of nuclear two-photon decay of the Heidelberg group is the fact that the 2M1 and 2E1 transitions are of equal strength. This has been explained [6,8] by a strong cancellation effect in the electric dipole transition polarizability, while the magnetic dipole transition susceptibility is of single particle strength. This cancellation effect is due to the structure of the 0^+ states, i.e. 0p-0h and np-nh states across closed shells. Without a detailed knowledge of the nuclear structure effects it is therefore difficult to obtain a reliable estimate for the (partial) halflife of the 2γ decay in other cases. It is however interesting to note that in all three cases where a reliable measurement has been performed (^{16}O, ^{40}Ca and ^{90}Zr) the ratio $\Gamma_{\gamma\gamma}/\omega^7$ has a rather constant value of 35 ± 4 s^{-1} MeV^{-7}, while the partial 2γ decay times vary by 4 orders of magnitude. This might indicate that the structure of the nuclei studied so far is indeed extremely similar, but it could also be due to a more general behaviour of the two-photon decay which is not yet understood.

Therefore it is very important to study other cases, especially for lower decay energies, in order to better understand the properties of this decay mode.

Ideally, the search for nuclear two-photon decays is performed in even-even nuclei having a first excited 0^+ state below the pair creation threshold. In this case the only first-order E0 decay mode to the ground state is the emission of internal conversion electrons. But only two stable nuclei are known to show such low-lying excited 0^+ states (^{72}Ge and ^{98}Mo). Further examples have been observed recently in several nuclei located far from the valley of stability and thus requiring the use of radioactive ion beams. Probably the most noted cases are ^{186}Pb [10] (studied at GSI) and ^{188}Pb [11]. In both cases the observation of two excited 0^+ states below 1 MeV has been interpreted as evidence for a unique triple shape coexistence of the nucleus. A similar coexistence between two different shapes is observed in nuclei around mass A~70 close to the N=Z line, where a single low-lying excited 0^+ state is observed, which becomes the first excited state in ^{72}Kr [12] and ^{72}Ge [13]. Searching for low-lying 0^+ states requires the observation of conversion electrons, which is experimentally more difficult than detecting gamma rays. It is therefore possible, or maybe even likely, that many more unstable nuclei with a first excited 0^+ state exist, but have so far escaped experimental observation (see Fig. 1). As an example, our observation of low-lying 0^+ states in ^{72,74}Kr [12] has ended more than 40 years of speculation about its existence. Similar 0^+ states are expected to exist in ^{68,70}Se, but remain yet to be observed. Besides their importance for a better understanding of the structure of N=Z nuclei around A~70, the 0^+ isomers play a major role in nuclear astrophysics as these nuclei are thought to be waiting points for the rp process.

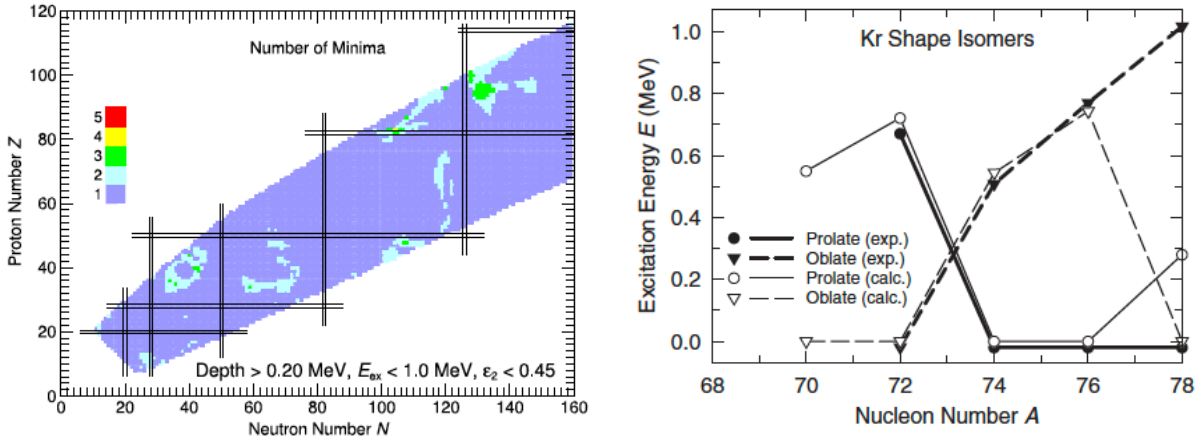

Fig. 1: (Left) Prediction of shape isomers from the Finite Range Liquid Drop Model [14], i.e. 0^+ states with a shape different from the ground state with an excitation energy below 1 MeV. (Right) Systematics of 0^+ states in Kr isotopes [12] and FRLDM predictions [14].

Experimental Technique and Expected Results

In this proposal we want to pioneer a novel experimental technique in order

i. *to search for low-lying 0^+ states and*
ii. *to establish the "exotic" nuclear two-photon decay mode* of these states.

The method takes advantage of the unique capability of the GSI facility in producing fully stripped ions of exotic nuclei by in-flight fragmentation, which can be separated either if needed by the GSI fragment separator (FRS) or in the straight connection line from SIS18 to the ESR, and subsequently stored and cooled in the experimental storage ring (ESR). In the

present proposal, the highly charged ions will be produced in-flight from a ^{78}Kr primary beam and separated using solely in the ESR. This approach that bypasses the FRS has been successfully developed and used for several storage ring experiments of high-intensity beams of artificially synthesized isotopes and also isomers [15]. If successful it will open up a wide range of studies not only with the current facility, but also for the Super-FRS and the CR storage ring at the new FAIR facility.

The basic idea of our experiment is to produce, select and store exotic nuclei in their excited 0^+ state. For neutral atoms the excited 0^+ state is a rather short-lived isomeric state with a lifetime of the order of a few tens to hundreds of nanoseconds. At relativistic energies available from SIS18, however, all ions are fully stripped of their atomic electrons and decay by ICE emission is hence not possible. If the state of interest is located below the pair creation threshold the IPC process is not possible either and the lifetime increases considerably [12]. Consequently, bare nuclei stored in the ESR are trapped in a long-lived isomeric state, which can only decay by 2γ emission to the ground state or by particle emission (alpha or beta decay) for unstable isotopes.

In the present proposal we aim for a pilot experiment to demonstrate the experimental technique in the most easily accessible nucleus having a first excited 0^+ state below the pair creation threshold, namely ^{72}Ge. However, if proven the method should be applicable to study the 2γ decay as well as to search for new low-lying 0^+ isomers. Furthermore, other exotic decays like, e.g., the bound-state electron-positron decay will be possible to address [16].

Experimental Design and Methods, Technical Requirements and Proposed Work-Plan

The ESR allows high-precision mass measurements using Schottky mass spectroscopy (SMS) [17]. However, in order to be able to measure these still rather short-lived isomers we will employ the isochronous ion optical setting of the ESR, routinely used in the past as basis for the Isochronous Mass Spectrometry (IMS). The IMS does not require the lengthy cooling of the ions and the revolution frequencies of the stored ions can be measured right after the injection, i.e. a few hundred nanoseconds after the radionuclides are produced. As a pioneering feature, instead of the time-of-flight detectors used in the past, we will use the newly developed highly-sensitive non-destructive resonant Schottky detectors [18]. Such cavity-based detectors enabled us to monitor in time steps of about 32 ms the frequencies and intensities of all secondary ions stored in the ESR. Combined with the new universal data acquisition system, which has been taken into operation in the E121 experiment in Spring 2020, the NTCAP (New Time Capture Data Acquisition System) [19], we will be able to observe with high time- and frequency resolution the entire acceptance of the ESR.

The use of *combined isochronous and Schottky mass spectrometry* has been demonstrated at GSI [20] and more recently at the CSR-e in Lanzhou [21]. The mass resolving power of the IMS depends on the quality of the ion-optical setting. In Lanzhou a mass resolving power of 2-4x10^5 is routinely obtained without cooling the beam, which is sufficient to separate states at an excitation energy of few hundred keV [22] as is the case of the ground and isomeric states of ^{72}Ge. At the ESR, a mass resolving power of 10^5 has been reported [23], which in principle would be sufficient to successfully conduct the proposed experiment. However, the experience gained in the last decade at the CSR-e shall allow us to improve this figure by a factor of 2 to 4. The operation of the ESR in the isochronous mode was not yet demonstrated

with the new control system. It is requested also by the experiment of Walker et al. and shall be commissioned in advance.

The 2γ decay of the isomer would be identified by time-resolved SMS [24], i.e., by observing the disappearance of the isomer peak in the SMS spectrum with a characteristic decay time. In cases where particle emission from the isomer is possible, we will identify it by the appearance of the corresponding daughter ions at the corresponding revolution frequency.

As discussed above a reliable prediction for the 2γ halflife of the 0^+ isomer in ^{72}Ge is difficult to obtain due to the uncertainties in the influence of nuclear matrix elements, but a first-order estimate can be made using the constant value of the ratio $\Gamma_{\gamma\gamma}/\omega^7 = 35\pm4$ $s^{-1}MeV^{-7}$ measured in previous experiments [6,8]. This gives a halflife of 400 ms for a 2γ decay at ~700 keV. Measuring such short lifetimes can only be achieved by using isochronous mass spectroscopy as discussed above. The already achieved time-resolution of the Schottky detectors of 32 ms suites well the expected lifetime. In addition the relativistic energies facilitate the detection of such isomers since their lifetimes are extended in the laboratory frame by Lorentz γ of about 1.4.

Though very unlikely, if the isomeric lifetime turns out to be much shorter than expected we will use a more sensitive, even higher-frequency detector, which has been implemented and commissioned in the ESR in 2020 for the FAIR Phase-0 program. This detector has even higher time resolution though at a cost of lower bandwidth. In any case the proposed experiment is unique for GSI/FAIR and will employ a range of instrumentation and methodology developed within NUSTAR/ILIMA.

Justification of Beamtime Request

The stable isotope ^{72}Ge nucleus can be produced very abundantly, and the isomeric state could in principle be populated by inelastic scattering at relativistic energies. However, a pure electro-magnetic interaction does not allow the direct excitation of a 0^+ state. Alternatively, few-nucleon removal reactions at relativistic energies are known to produce low-lying isomeric states with rather high cross sections. The 0^+ isomer in ^{72}Kr, for example, is populated with a probability of ~5% in the fragmentation of a primary ^{78}Kr beam at energies between ~70 MeV/u [12] and 350 MeV/u [25].

For this experiment a primary Kr beam of almost any isotope can be used. In the following we show the results using LISE++ calculations for a ^{78}Kr primary beam. If employing the FRS, a high purity can be reached even using only a rather thin (1mm) Al degrader in the FRS. However, taking into account the huge load of the FRS, we propose to use for this experiment a thick stripper target, i.e. a 1 mm (1.8 g/cm^2) Be plate in the transfer beamline between SIS18 and ESR, the TE-line, for production and the ESR utilized as an isotope separator. In [15] it was shown that the intense primary beam could be suppressed up to a few ions per spill, while storing 10^5 ions of the isotope of interest. Furthermore, with the NTCAP we will be able to monitor the fate of all species stored in the ESR. In this way other NUSTAR experiments at the FRS can be operated in parallel to our experiment. The details of the production and experimental settings either via the FRS (if available) or via the TE-line are summarised in table 1.

Primary beam, intensity & energy		Target/ Degrader	^{72}Ge rate and energy	Bρ settings [Tm]	Total rate and other principal fragments
^{78}Kr 10^8 pps 460 MeV/u	FRS:	2 g/cm^2 Be 1 mm Al	260 pps 372 MeV/u	D1/D2: 6.9447 D3/D5: 6.8442	Total: 480 pps ^{74}As: 50 pps ^{70}Ga: 30 pps
	TE-line:	2 g/cm^2 Be no degrader	500 pps 382 MeV/u	Beamline: 6.9447	Total : 5100 pps ^{74}As: 710 pps ^{65}Cu: 180 pps ^{63}Ni: 100 pps

Table 1: Rate estimate for the production of ^{72}Ge from a primary ^{78}Kr beam using the FRS (top) and the direct (TE) beam line (bottom). Similar rates can also be achieved with heavier Kr isotopes.

With both approaches the estimated rates are high enough to *perform the data taking within 3 shifts*. Assuming an isomer ratio of 4% we expect 10-20 isomer decays per injected spill, with a repetition time between 2-10 seconds depending on the actual lifetime. However, the main amount of beamtime will go into the preparation of the ESR in order to achieve the mass resolution of $<10^{-5}$ necessary to resolve the two peaks. We thus estimate a preparation time of 2 days of direct SIS18 beam. It should be noted that similar developments are planned for the experiment of Walker et al., albeit in a different mass region. Therefore, a commissioning during an engineering beamtime should precede these experiments.

In a second setting, we plan to search for a new 0^+ isomer in ^{70}Se, which has been searched for many years using gamma-ray and conversion-electron spectroscopy. The rate estimate is given in Table 2 and very similar to the ^{72}Ge case. Here again, we ask *for 3 shifts for data taking.*

Primary beam, intensity & energy		Target/ Degrader	^{70}Se rate and energy	Bρ settings [Tm]	Total rate and other principal fragments
^{78}Kr 10^8 pps 460 MeV/u	FRS:	2 g/cm^2 Be 1 mm Al	430 pps 364 Mev/u	D1/D2: 6.2950 D3/D5: 6.1832	Total: 550 pps ^{68}As: 55 pps
	TE-line	2 g/cm^2 Be no degrader	860 pps 376 Mev/u	Beamline: 6.2950	Total: 6900 pps ^{72}Br: 590 pps ^{68}As: 810 pps (^{66}Ge: 640 pps)

Table 2: Rate estimate for the production of ^{70}Se from a primary ^{78}Kr beam using the FRS (top) and the direct (TE) beam line (bottom).

We request a *total of 6 shifts of data taking* with a ^{78}Kr beam at 450 MeV/u to study both isotopes. In addition *6 shifts of set-up and commissioning time of the ESR is needed* in order to further develop the operation of the ESR in the isochronous mode. In both cases the direct beam from SIS18 can be used so that the FRS could be used in parallel. Future experiments devoted to the 2γ decay of the N=Z nucleus ^{72}Kr or to a search for new 0^+ isomers, e.g. in ^{68}Se,

would need to use of the FRS. These nuclei have multiple interests related to nuclear structure (pn-pairing, shape coexistence) and astrophysics (rp-process) and the observables in the 2γ decay might give some new interesting insights.

8. Three Major Publications of the Spokesperson, References.

E. Bouchez, I. Matea, W. Korten et al,
New Shape Isomer in the Self-Conjugate Nucleus ^{72}Kr
Phys. Rev. Lett. 90, 082502 (2003)
https://doi.org/10.1103/physrevlett.90.082502

A. Görgen and W. Korten
Coulomb excitation studies of shape coexistence in atomic nuclei
J. Phys. G: Nucl. Part. Phys. 43 (2016) 024002
https://doi.org/10.1088/0954-3899/43/2/024002

K. Wimmer, W. Korten, T. Arici, et al.,
Shape coexistence and isospin symmetry 1 in A = 70 nuclei: Spectroscopy of the $T_Z = -1$ nucleus ^{70}Kr
Phys. Lett. B. 785, 441 (2018)
https://doi.org/10.1016/j.physletb.2018.07.067

[1] M. Göppert-Mayer, Ann. Phys (Leipzig) 9, 273 (1931)

[2] R.G. Sachs, Phys. Rev. 57, 194 (1940)

[3] D.P. Grechukhin, Nucl. Phys. 47, 273 (1963)

[4] J. Eichler, Phys. Rev. A 9, 1762 (1974)

[5] J.L. Friar and M. Rosen, Ann. of Physics 87, 289 (1974) and
J.L. Friar Ann. of Physics 95, 170 (1975)

[6] J. Kramp et al., Nucl. Phys. A 474, 412 (1987)

[7] B.A. Watson et al., Phys. Rev. Lett. 35, 1333 (1975);
E. Beardsworth et al., Phys Rev. C 8, 216 (1973);
Y. Asano and C.S. Wu, Nucl. Phys. A 125, 557 (1973), and references therein

[8] J. Schirmer et al., Phys. Rev. Lett. 53, 1897 (1984)

[9] C. Walz et al., Nature 526, 406 (2015)

[10] A. Andreiev et al., Nature 405, 430 (2000)

[11] Y. Le Coz et al, Eur. Phys. J. direct A3, 1 (1999)

[12] E. Bouchez et al., Phys. Rev. Lett. 90, 082502 (2003)

[13] W.-T.Chou and M.M.King, Nucl. Data Sheets 73, 215 (1994)

[14] P. Möller at al., Phys. Rev. Lett. 103, 212501 (2009)

[15] C. Brandau, et al., Phys. Scr. 2013, 014050 (2013) and references therein
[16] F. Bosch et al., EPJ Web of Conf. 123, 04003 (2016)
[17] B. Schlitt et al. Hyperfine Interactions 99, 117-125 (1996)
[18] F. Nolden et al., Nucl. Instr. Meth. A659, 69 (2011)
[19] C. Trageser et al., Phys. Scripta T166, 014062 (2016)
[20] P. Walker et al., Int. J. Mass Spectrometry 349-350 (2013) 247-254
B. Sun et al., GSI Scientific Report 2011, 163, PHN-NUSTAR-FRS-21
[21] X.L. Tu et al, Phys. Rev. C 97, 014321 (2018)
[22] Y. H. Zhang et al., Phys. Scripta 91, 073002 (2015)
[23] B. Sun et al., Nucl. Phys. A811 (2008)
[24] F. Bosch, J. Phys. B 36, 585 (2003)
[25] K. Wimmer et al., Eur. Phys. J. A, in press