# GSI Research Proposal

**G-22-00018**

Experiment title:
**Nuclear two-photon decay and bound-state pair conversion**

**Proposal type:** Standard **(ST)**

**Scientific College:** G-PAC

Co-Proposers of entire sub-collaboration: ILIMA

Abstract:
The aim of this proposal is to continue our studies of the rare nuclear two-photon decay in the decay of low-lying excited 0+ states. In the previous experiment (E143) we have successfully established the technique of Isochronous Schottky Mass Spectroscopy to study isomers in the ms range as a new technique to search for short-lived isomers. Candidates for the two-photon decay of the excited 0+ state in 72Ge indicate a much shorter lifetime than expected from previous experiments. In the new proposal we plan to study the isobars 98Mo and 98Zr, which each have two-particles outside the doubly semi-magic isotope 96Zr (Z=40,N=56) and therefore ideal candidates for shell-model calculations of the two-photon decay rate, which cannot easily access the previously studied 72Ge. In addition, we plan to study 194Pb, which has a first excited 0+ state at 931 keV and is also a candidate for bound-state electron-positron pair conversion, where the electron is captured in an atomic orbital.

**Proposer** (All correspondence concerning this proposal will be sent to the proposer)

Name and first name:
Dr. Korten, Wolfram, DE

Address:
CEA Paris Saclay
IRFU/DPhN
JARDINS DES MERISIERS
91191 Gif-sur-Yvette
France

Email:
w.korten@cea.fr

**Co-proposer:**

| | | |
|---|---|---|
| Dr. Akinci, Fatma Cagla, TR | U Istanbul | fcaglaozturk@gmail.com |
| Dr. Albers, Helena, DE | GSI Darmstadt | h.albers@gsi.de |
| Prof. Dr. Blaum, Klaus, DE | MPI HD | klaus.blaum@mpi-hd.mpg.de |
| Dr. Brandau, Carsten, DE | GSI Darmstadt | c.brandau@gsi.de |
| Dr Bruno, Carlo, IT | Univ Edinburgh | c.bruno@gsi.de |
| Dr. Chen, Rui Jiu, CN | IMP CAS | chenrj13@impcas.ac.cn |
| Chiara, Christopher, US | DEVCOM | christopher.j.chiara2.civ@army.mil |
| Dellmann, Sophia Florence, DE | JWGU Frankfurt/M. | s.f.dellmann@gsi.de |
| Dr. Dickel, Timo, DE | GSI Darmstadt | t.dickel@gsi.de |
| Dr. Dillmann, Iris, DE | TRIUMF | dillmann@triumf.ca |
| Dr. Forstner, Oliver, AT | FSU Jena | o.forstner@hi-jena.gsi.de |
| Freire Fernández, David, ES | MPI HD | d.freirefernandez@gsi.de |
| Prof. Dr. Geissel, Hans, DE | GSI Darmstadt | h.geissel@gsi.de |
| Dr. Gerl, Jürgen, DE | GSI Darmstadt | j.gerl@gsi.de |
| Dr. Glorius, Jan, DE | GSI Darmstadt | j.glorius@gsi.de |
| Dr. Gorska-Ott, Magdalena, PL | GSI Darmstadt | m.gorska@gsi.de |
| Dr Griffin, Christopher James, GB | TRIUMF | cgriffin@triumf.ca |
| Dr. Gumberidze, Alexandre, GE | GSI Darmstadt | a.gumberidze@gsi.de |
| Prof. Görgen, Andreas, DE | Univ. Oslo | andreas.gorgen@fys.uio.no |

| | | |
|---|---|---|
| Dr. Haettner, Emma, SE | GSI Darmstadt | e.haettner@gsi.de |
| Dr. Hagmann, Siegbert, DE | GSI Darmstadt | s.hagmann@gsi.de |
| Harayama, Sakumi, JP | RIKEN, JP | s.harayama.109@ms.saitama-u.ac.jp |
| Dr. Hillenbrand, Pierre-Michel, DE | JLU | p.m.hillenbrand@gsi.de |
| Dr. Hubbard, Nicolas James, GB | GSI Darmstadt | n.hubbard@gsi.de |
| Dr Jurado, Beatriz, FR | CENBG | jurado@cenbg.in2p3.fr |
| Dr. Kalaydjieva, Desislava, BG | IJCLab | dnkalaydjieva@gmail.com |
| Dr Kanika, Kanika, IN | IC London | k.kanika@gsi.de |
| Dr. Kozhuharov, Christophor, DE | GSI Darmstadt | c.kozhuharov@gsi.de |
| Mr Leckenby, Guy, AU | TRIUMF | gleckenby@triumf.ca |
| Apl. Prof. Litvinov, Yuri, RU | GSI Darmstadt | y.litvinov@gsi.de |
| Prof. Liu, Zhong, CN | IMP CAS | liuzhong@impcas.ac.cn |
| Dr. Lorentz, Bernd Alfred, DE | GSI Darmstadt | b.lorentz@gsi.de |
| Marsh, Jordan, GB | Univ Edinburgh | jordan.marsh@ed.ac.uk |
| Ozawa, Akira, JP | Tsukuba Ac | ozawa@tac.tsukuba.ac.jp |
| Prof. Pietralla, Norbert, DE | TU Darmstadt | pietralla@ikp.tu-darmstadt.de |
| Prof. Podolyak, Zsolt, GB | Surrey AC | z.podolyak@gsi.de |
| Prof. Reifarth, Rene, DE | JWGU Frankfurt/M. | reifarth@physik.uni-frankfurt.de |
| Dr. Sanjari, Mohammad Shahab, DE | GSI Darmstadt | s.sanjari@gsi.de |
| Prof. Dr. Scheidenberger, Christoph, DE | GSI Darmstadt | c.scheidenberger@gsi.de |
| Dr. Sidhu, Ragandeep Singh, IN | Univ Edinburgh | r.s.sidhu@gsi.de |
| Dr. Spillmann, Uwe, DE | GSI Darmstadt | u.spillmann@gsi.de |
| Dr. Steck, Markus, DE | GSI Darmstadt | m.steck@gsi.de |
| Prof. Stoehlker, Thomas, DE | HI Jena | t.stoehlker@hi-jena.gsi.de |
| Vesic, Jelena, SI | IJS Ljubljana | jelena.vesic@ijs.si |
| Prof. Walker, Philip, GB | Surrey AC | p.walker@surrey.ac.uk |
| Dr. Weick, Helmut, DE | GSI Darmstadt | h.weick@gsi.de |
| Dr. Werner, Volker, DE | TU Darmstadt | vw@ikp.tu-darmstadt.de |
| Dr. Wimmer, Kathrin, DE | GSI Darmstadt | k.wimmer@gsi.de |
| Dr. Yamaguchi, Takayuki, JP | Saitama Univ | yamaguti@ribf.riken.jp |
| Prof. Zhang, Yuhu, CN | IMP CAS | yhzhang@impcas.ac.cn |

| | |
|---|---|
| **FAIR Experiment Pillar:** NUSTAR | **Collaboration:** ILIMA |

**Declaration of peaceful purpose**

GSI pursues scientific work solely for peaceful purposes.
In this respect, only proposed research projects that serve and promote exclusively peaceful purposes can be accepted.

I confirm the peaceful purpose of the proposal ☒

**Experiment time allocated for**

| Target station | Allocated experiment time | Link scientist |
|---|---|---|
| 2.1-E: FRS-ESR (Main beam) |  | Yury Litvinov |
| 2.2-E: ESR (Main beam) | 15 Shifts | Yury Litvinov |

Next page: scientific case

### Nuclear two-photon decay and bound-state pair conversion (Continuation of E143)

*W. Korten (co-spokesperson)*, D. Kalaydjieva, A.M. Corsi, M. Vandebrouck et al., IRFU, CEA, Université Paris-Saclay, F-91191 Gif-sur-Yvette, France

*Yu.A. Litvinov (co-spokesperson),* C. Akinci, M.S. Sanjari, H. Weick, C. Brandau, S.A. Litvinov, M. Steck, H. Albers, T. Dickel, H. Geissel, J. Gerl, J. Glorius, M. Gorska , A. Gumberidze, E. Haettner, S. Hagmann, R. Heß, P.-M. Hillenbrand, K. Kanika, C. Kozhuharov, B. Lorentz, C. Scheidenberger, U. Spillmann, K. Wimmer, GSI Darmstadt, D-64291 Darmstadt, Germany

K. Blaum, D. Freire, MPI-Kernphysik, D-69117 Heidelberg, Germany

N. Pietralla, V. Werner, N. Hubbard, TU Darmstadt, 64294 Darmstadt, Germany

T. Yamaguchi, S. Harayama et al., Saitama University, Saitama, 338-8570, Japan

I. Dillmann, C. Griffin, G. Leckenby, TRIUMF, Vancouver BC V6T 2A3, Canada

B. Jurado et al., CNRS - IN2P3/IP2I Bordeaux, 33170 Gradignan, France

T. Stöhlker, O. Forstner, Helmholtz Institut Jena, 07743 Jena, Germany

A. Görgen, University of Oslo, 1048 Blindern, N-0316 Oslo, Norway

Ph. Walker, Zs. Podolyak, University of Surrey, Guildford, GU2 7XH, UK

F.G. Kondev, Argonne National Laboratory, Lemont, Illinois 60439, U.S.A.

J.J. Carroll, C.J. Chiara, US Army Research Laboratory, Adelphi, 20783, Maryland, U.S.A.

C. Bruno, J. Marsh, R. S. Sidhu, Univ. of Edinburgh, Edinburgh, EH9 3FD, UK

J. Vesic et al., Institut Jožef Stefan, 1000 Ljubljana, Slovenia

R. Chen, Z. Liu, Y.H. Zhang, Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, People's Republic of China

S. Dellmann, R. Reifarth, Goethe Universität, D-60438 Frankfurt, Germany

A. Ozawa, Tsukuba Univ., Japan

**and the ILIMA collaboration**

## Abstract

The aim of this proposal is to continue our studies of the rare nuclear two-photon ($2\gamma$) emission from the decay of low-lying excited $0^+$ states. In the previous "A"-rated experiment (E143) we have successfully established the technique of combined Isochronous plus Schottky Mass Spectroscopy to study isomers in the millisecond range as a new technique to search for low-lying $0^+$ isomers. We found candidates for the $2\gamma$ decay of the excited $0^+$ state in $^{72}$Ge indicating a much shorter half-life than extrapolated from previous experiments.

In the new proposal we plan to study the isobars $^{98}$Mo and $^{98}$Zr, which each have two-particles outside the doubly semi-magic isotope $^{96}$Zr (Z=40,N=56) and therefore ideal candidates for shell-model calculations of the two-photon decay rate. In addition, we plan to study $^{194}$Pb, which has a first excited $0^+$ state at 931 keV and is also a candidate for *bound-state electron-positron pair conversion*, where the emitted electron is captured in an atomic orbital. Measuring a possible $2\gamma$ decay branch is a prerequisit for a future search of this so far never observed decay mode. Finally, these experiments open the possibility to search yet unknown low-lying $0^+$ states in both mass regions.

The highly charged ions will be produced in-flight from $^{100}$Mo (for $^{98}$Mo and $^{98}$Zr) and from $^{209}$Bi (for $^{194}$Pb). The isotopes of interest can be produced in the direct beam line from SIS18 to the ESR and separated using the ESR. This approach was also used successfully for the previous experiment on $^{72}$Ge. In this way other NUSTAR experiments can be operated at the FRS in parallel to our experiment. The experiment is unique to GSI/FAIR and will employ a range of instrumentation and methodology developed within NUSTAR/ILIMA. Experience gained in the previous measurements, enabled us to significantly improve both experimental hardware and software as well as accelerator operation. Within the proposed experiment we aim at testing the ILIMA detectors and methods which will undoubtedly lead to their further developments.

We request 15 shifts with a $^{100}$Mo beam at 450 MeV/u to study $^{98}$Mo and $^{98}$Zr as well as 15 shifts with a $^{209}$Bi beam at 550 MeV/u to study $^{194}$Pb. This includes *3 shifts per isotope for setting-up and commissioning time of the ESR* in order to assure optimal operation conditions of the ESR in the isochronous mode.

## The nuclear two-photon decay

The nuclear two-photon decay, also called double-gamma ($2\gamma$) 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 hindered.

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 $2\gamma$ 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 electron-positron 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 2$\gamma$ decay proceeds through virtual excitation of intermediate, higher-lying states. The sum energy of the two $\gamma$-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 the 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 2$\gamma$ 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 2$\gamma$ decay width can be expressed as:

$$\Gamma_{\gamma\gamma} = \omega^7/105\pi\ [\alpha(E1)^2 + \chi^2(M1) + \omega^4\alpha(E2)^2/4752]$$

Here, $\omega$ is the energy difference of the initial and final state, while $\alpha$ denotes the (electric) transition polarizability and $\chi$ 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, which are small 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 photonuclear reactions, while the magnetic dipole susceptibility can be deduced from (e,e') measurements. The 2$\gamma$ 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 $\chi(M1)$.

## The bound-state electron-positron pair creation

Internal pair creation (IPC) is an electromagnetic decay process where the excitation energy of the nucleus allows it to create spontaneously an electron-positron pair. Because of the rest mass of the $e^+$-$e^-$ pair this decay mode is limited to excited states with energies above 1022 keV and its probability rises strongly with excitation energy [7]. The excess in excitation energy is shared between the $e^+$-$e^-$ pair. In $^{194}$Pb the first excited $0^+$ state is an isomer with a half-life of 1.1 ns located at an energy of 930.7 keV. Since it is below the pair-creation threshold it decays to the ground solely by internal conversion.

Similar to the other cases discussed in this proposal, fully-stripped ions will have no first-order electro-magnetic decay mode and may hence decay only by two-photon emission. If, however, the electron is captured into the empty atomic K-Shell an additional (atomic) binding energy of 101.3 keV [8] becomes available, which increases the total emission energy above the pair threshold. Therefore, the *bound-state electron-positron pair creation* becomes energetically possible [9]. $^{194}$Pb is in fact the only known nucleus where this decay mode is open, but not the normal electron-positron pair creation. Both decays channels, 2$\gamma$ and IPC, are of interest in the present study and will compete in $^{194}$Pb.

**Experimental background and referencing previous experiments**

Experimentally, many early attempts have been made to observe the 2$\gamma$ decay often with conflicting results [10]. 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 of the 2$\gamma$ decay 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 2$\gamma$ 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 4$\pi$ NaI(Tl) detection system, in order to identify the tiny 2$\gamma$ 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 [6,11] and $^{90}$Zr [6,11]. More recently, also the competitive 2$\gamma$ decay was observed in the decay of the $11/2^-$ isomer in $^{137}$Ba in experiments using the fast-timing method at TU Darmstadt [12].

The most surprising result obtained in the investigation of the nuclear two-photon decay from the $0^+ \rightarrow 0^+$ decay by the Heidelberg group is the fact that the 2M1 and 2E1 transitions are of equal strength. This has been explained [6,11] 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, respectively. Without a detailed knowledge of the nuclear structure effects it is therefore difficult to obtain a reliable estimate for the (partial) halflife of the 2$\gamma$ 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 $\sim$31 $s^{-1}$ $MeV^{-7}$, while the partial 2$\gamma$ decay widths vary by 4 orders of magnitude (see left part of Fig. 2). 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.

In our previous experiment (E143) we studied $^{72}$Ge, which is a mid-shell nucleus where the excited $0^+$ state is located at much lower excitation energy and interpreted as a shape isomer, i.e. located in secondary minimum of the potential energy surface. In this experiment we applied Schottky Mass Spectroscopy for the first time in the isochronous mode of the ESR. No cooling is needed in the isochronous mode thus enabling access to the shortest-lived nuclides. Furthermore we employed the new highly-sensitive non-destructive Schottky detector developed for ILIMA within the ERC grant "ASTRUm". This enabled us to measure nuclear isomers with half-lifes as short as several tens of ms. We also confirmed that excited states with energies as low as 100 keV can be separated from the ground state by tailoring the beam to the linear part of the isochronicity curve (see Fig. 1, left) [13]. We observed several hundred events for two-photon decay candidates of the $0^+$ isomer in $^{72}$Ge, which are separated from the ground state by the frequency expected for a state at 690 keV (see right part of Fig. 1).

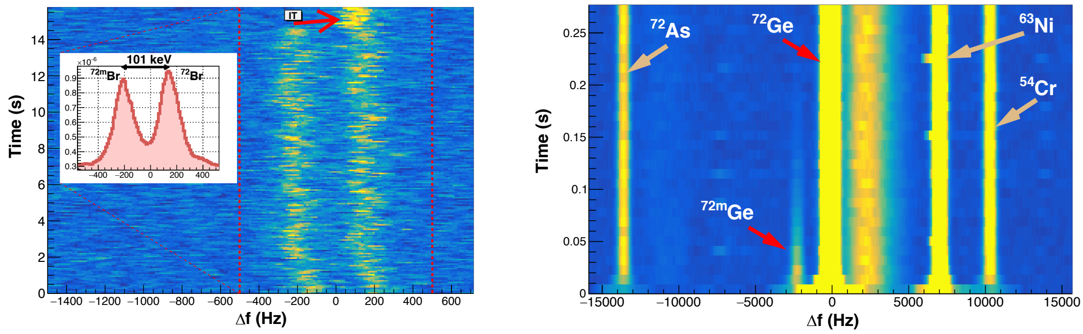

*Figure 1: Preliminary results from experiment E143: [left] Observation of the 101 keV isomer in $^{72}$Br: a single ion in the isomeric state decays at ~14s to the ground state, indicating a mass resolution of better than $10^{-6}$ for single ions in the novel combined isochronous plus Schottky mode of the ESR. [Right] First observation of the $0^+$ isomer in fully-stripped $^{72}$Ge ions: Overlay of a few hundred events, where the isomer is present.*

The preliminary analysis showed that the half-life of the isomer is much shorter than expected from the systematics (see Fig. 2). The detailed analysis is still in progress. We believe that the shorter half-life is related to a larger electric dipole polarizability in deformed $^{72}$Ge, since the cancellation effect previously observed in all (semi-) magic isotopes is probably not present. Therefore, it might not be too surprising that the (partial) half-life for the two-photon decay turned out to be much shorter than extrapolated from the “magic” nuclei studied earlier.

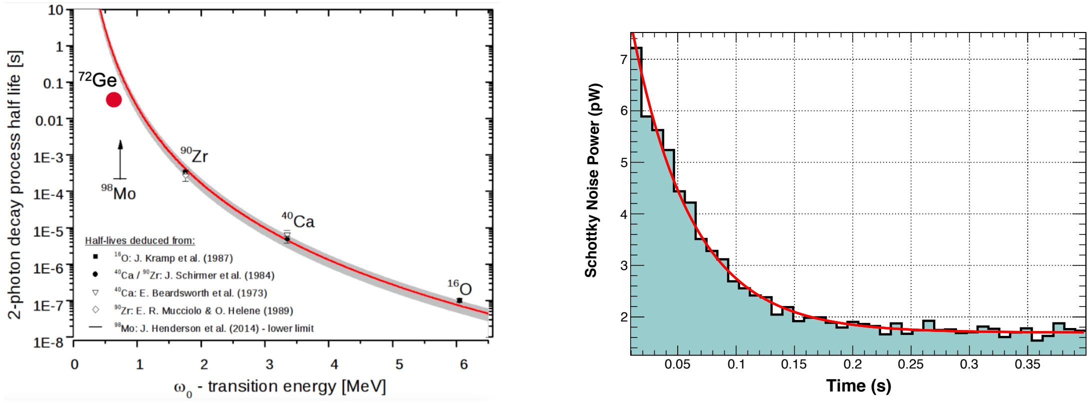

*Figure 2: [Left] Partial half-life for the two-photon decay previously observed in $^{16}$O, $^{40}$Ca and $^{90}$Zr extrapolated to lower energies for the proposed measurements and preliminary value for $^{72}$Ge$^m$ from E143 (reproduced from [14] which obtained the indicated lower limit for $^{98}$Mo). [Right] Power density of the signal from the $^{72}$Ge isomer (shown in Fig. 1) as function of time after injection indicating a preliminary half-life of 30-40 ms.*

In the new proposal we would like to extend our studies in two directions. First, we would like to study the isobars $^{98}$Mo and $^{98}$Zr, which have two-particles outside the “doubly semi-magic” isotope $^{96}$Zr (Z=40,N=56). They are therefore ideal candidates for shell-model calculations of the two-photon decay rate, which cannot easily access the previously studied isotope $^{72}$Ge. Other theoretical approaches such as (Q)RPA, which are capable to access $^{72}$Ge, do not allow to study the *transitional dipole polarizability*, where the initial and final state are not identical. Beyond mean-field calculation, e.g. using the HFB approach with different (Skyrme or Gogny) functionals, usually preserve parity as good quantum number, and do not allow to calculate negative-parity states other than collective octupole excitations. We are currently evaluating to what extent ab-initio calculations can access this quantity.

The second part of the experiment aims as at $^{194}$Pb, which is also a semi-magic nucleus, but with a large number of valence neutrons. In this case the measurement of the two-photon decay rate is a prerequisite to tackle the bound-state electron-positron pair creation. In view of the expected very long partial half-life for this even more exotic decay mode, the two-photon decay branch is actually a competing process, which we need to establish first.

## Experimental Technique and Expected Results

In this proposal we want to apply the recently pioneered technique of Isochronous plus Schottky Mass Spectroscopy (ISMS) at the ESR in order

i. *to establish the "exotic" nuclear two-photon decay mode in $^{98}$Mo, $^{98}$Zr and $^{194}$Pb,*
ii. *to search for new low-lying $0^+$ states in both mass regions*

Searching for the nuclear two-photon decay of low-lying excited $0^+$ states with ISMS exploits the unique capability of the GSI facility in producing fully stripped ions of exotic nuclei by in-flight fragmentation, which can be separated either by the GSI fragment separator (FRS) or in the direct connection (TE-)line from SIS18 to the ESR, and subsequently stored in the ESR. By using the isochronous mode of the ESR short-lived isotopes with half-lifes in the few tens of ms regime can be accessed. We also showed that by carefully preparing the beam in the ESR a mass resolution of better than $10^6$ (<100 keV for mass A~70 nuclei) can be achieved.

In the present proposal, the highly charged ions will be produced in-flight from primary beams of (i) $^{100}$Mo and (ii) $^{209}$Bi. The isotopes of interest will be produced by impinging on a 10 mm Be plate in the direct beam line from SIS18 to the ESR since their separation is possible using solely the ESR (see LISE++ calculations below). This approach has been successfully used for several storage ring experiments of high-intensity beams of artificially synthesized isotopes [15] and also in our previous experiment. Further developing and optimizing ISMS as well as the related instrumentation 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 half-life 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 half-life increases considerably [16]. Consequently, bare nuclei stored in the ESR are trapped in a long-lived isomeric state, which can only decay by $2\gamma$ emission to the ground state or by particle emission (alpha or beta decay) for unstable isotopes. The half-lifes of all isotopes under consideration is sufficiently long as to not compete with the sub-second half-life of the $2\gamma$ emission.

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

The ESR allows high-precision mass measurements using time-resolved 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) [18]. 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 use the newly developed highly-sensitive non-destructive resonant Schottky detectors [19]. Such cavity-based detectors enabled us to monitor in time steps of about 10-20 ms the frequencies and intensities of all secondary ions stored in the ESR. Combined with the New Time Capture Data Acquisition System (NTCAP) [20], which has been taken into operation in the E121 experiment in Spring 2020, we are able to observe with high time- and frequency resolution the entire acceptance of the ESR.

The use of *combined isochronous and Schottky mass spectrometry* was first demonstrated at GSI [21,22] and more recently employed at the CSRe in Lanzhou [23,24]. The mass resolving power of the IMS depends on the quality of the ion-optical setting and can be improved by tailoring the beam to the flat region of the isochronicity curve. In our previous experiment (E143) we achieved a mass resolving power of better than $10^6$, which was sufficient to separate the 101 keV isomer in $^{72}$Br from the ground state (see left side of Fig. 1). As in the experiment E143, the 2$\gamma$ decay of the isomer would be identified by time-resolved SMS [17,25], i.e., by observing the disappearance of the isomer peak in the SMS spectrum (see right side of Fig. 1) 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 [26].

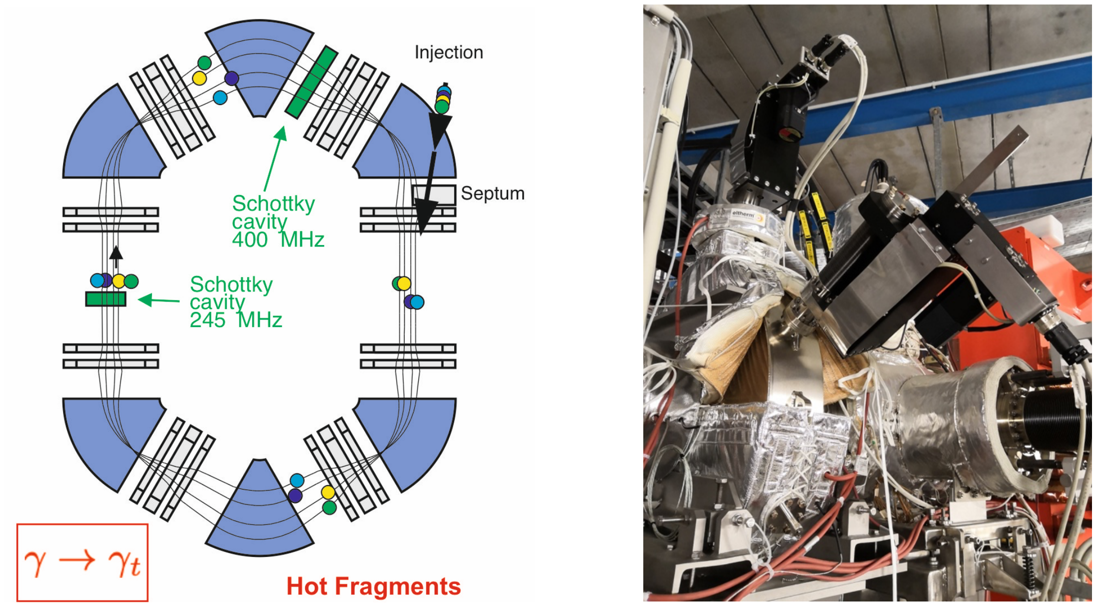

*Figure 3: Using combined Isochronous and Schottky Mass Spectrometry (ISMS) the ESR is tuned in isochronous mode where particles with* $\gamma = \gamma_t$ *have identical revolution frequencies, which is measured with high frequency Schottky detectors. Developed within the ERC grant “ASTRUm”, the latest edition working at 410 MHz is shown on the right hand part [27].*

As discussed above a reliable prediction for the partial 2$\gamma$ halflife of these $0^+$ isomers is difficult to obtain due to the uncertainties in the influence of nuclear matrix elements. Using as first-order estimate the constant value of the ratio $\Gamma_{\gamma\gamma}/\omega^7$~31 $s^{-1}MeV^{-7}$ (measured in the closed-shell nuclei [6,10]) gives halflifes of 300 and 100 ms for the 2$\gamma$ decay branch in $^{98}$Mo and $^{98}$Zr, respectively. In the case of $^{194}$Pb with a higher lying $0^+$ state the half-life could be as short as

60 ms, but as demonstrated in E143, using the newest sensitive 410 MHz Schottky detector [27], we are able to measure half-lifes as short as a very few tens of ms. Finally, the relativistic energies facilitate the detection of such short-lived isomers since their half-lifes are extended in the laboratory frame by a Lorentz factor of $\gamma\sim1.4$.

High gain component: Although not decisive for this experiment, we will employ the electron spectrometer available at the ESR to eventually measure the energies of the rarely emitted monochromatic positrons in the bound-state IPC decay of $^{194}$Pb. For this purpose, only the polarity of the spectrometer magnets will need to be changed which is a relatively simple and quick action. In this experiment we like to check the feasibility of such a measurement. However, if successful, the energy of positrons will directly be converted into the binding energy of the 1s orbital in even-even $^{194}$Pb nucleus and, dependent on the achieved precision, can potentially be used to benchmark the atomic physics calculations [8].

### Justification of Beamtime Request

The $^{98}$Mo and $^{98}$Zr nuclei can be produced rather abundantly in the fragmentation of a $^{100}$Mo beam (see Table 1). Mo beams have been used at GSI in the past. Previous fragmentation experiments have shown that low-lying excited $0^+$ states are populated with a probability of several percent. This was also confirmed in our previous experiment.

| Primary beam, intensity & energy | Beamline Target Degrader | Particle rate and energy | Bρ settings [Tm] | Total rate and other principal fragments |
|---|---|---|---|---|
| $^{100}$Mo<br>$10^9$ pps<br>450 MeV/u | FRS:<br>1.8 g/cm$^2$ Be<br>1 mm Al wedge | $^{98}$Mo:<br>**2.1 $10^5$ pps**<br>350 MeV/u | D1/D2:<br>7.0573<br>D3/D5:<br>6.9161 | Total: 2.2 $10^5$ pps<br>$^{96}$Nb: 11000 pps<br>$^{93}$Zr: 20 pps |
| | | $^{98}$Zr:<br>**590 pps**<br>382 MeV/u | D1/D2:<br>7.4549<br>D3/D5:<br>7.3219 | Total : 740 pps<br>$^{100}$Nb : 50 pps<br>$^{95}$Y : 60 pps |
| $^{100}$Mo<br>$10^9$ pps<br>450 MeV/u | TE-LINE:<br>1.8 g/cm$^2$ Be<br>No degrader | $^{98}$Mo:<br>**2.7 $10^5$ pps**<br>367 MeV/u | Beamline<br>7.0372 | Total: 3.2 $10^5$ pps<br>$^{96}$Nb: 12800 pps<br>$^{93}$Zr: 18000 pps |
| | | $^{98}$Zr:<br>**1050 pps**<br>371 MeV/u | Beamline<br>7.43763 | Total: 1810 pps<br>$^{95}$Y: 150 pps |

*Table 1: Rate estimate for the production of $^{98}$Mo and $^{98}$Zr from a primary $^{100}$Mo beam using the FRS (top) and the direct (TE) beam line from SIS18 (bottom).*

The production of radionuclides in this experiment can either be done in the FRS or in the transfer beamline between SIS18 and ESR, the TE-line. In Table 1 we show the results of LISE++ simulations for a $^{100}$Mo primary beam to produce $^{98}$Mo and $^{98}$Zr fragments. If employing the FRS, a higher purity can be reached even with a rather thin (1 mm) Al degrader in the FRS-S2. However, taking into account the huge load of the FRS, we propose to conduct the experiment employing the TE line. We will use the 10 mm Be plate (1.8 g/cm$^2$) available in the TE line as production target and utilize the ESR as an isotope separator in full analogy to our successful experiment E143. In this way other NUSTAR experiments at the FRS can be operated in parallel to our experiment. For the more exotic case of $^{98}$Zr the production rate via the TE line is expected to be a factor 2 higher[1]. 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. The details of the production and experimental settings either via the FRS or via the TE-line are summarized in Table 1.

| Primary beam, intensity & energy | Beamline Target & Degrader | **$^{194}$Pb rate** and energy | Bρ settings [Tm] | Total rate and other principal fragments |
|---|---|---|---|---|
| $^{209}$Bi<br>$10^9$ pps<br>550 MeV/u | FRS:<br>1.8 g/cm$^2$ Be<br>1 mg/cm$^2$ Nb<br>2 mm Al | **420 pps**<br>355 MeV/u | D1/D2: 7.5432<br>D3/D5: 7.0041 | **Total: 2900 pps**<br>$^{191}$Tl: 630 pps<br>$^{192}$Tl: 560 pps<br>$^{190}$Tl: 280 pps<br>$^{195}$Pb: 130 pps<br>$^{193}$Pb: 110 pps<br>$^{190}$Hg: 70 pps |
| $^{209}$Bi<br>$10^9$ pps<br>520 MeV/u | TE-line<br>1.8 g/cm$^2$ Be<br>1 mg/cm$^2$ Nb<br>no degrader | **2500 pps**<br>367 MeV/u | Beamline:<br>7.1377 | **Total: 82600 pps**<br>$^{192}$Tl: 3600 pps<br>$^{191}$Tl: 3100 pps<br>$^{189}$Hg: 5900 pps<br>$^{186}$Au: 4600 pps<br>$^{184}$Pt: 4800 pps<br>$^{181}$Ir: 3600 pps |

*Table 2: Rate estimate for the production of $^{194}$Pb from a primary $^{209}$Bi beam using the FRS (top) and the direct (TE) beam line from SIS18 (bottom).*

In Table 2 we show the results of LISE++ calculations for producing $^{194}$Pb using a $^{209}$Bi primary beam. For this second part of the experiment we also request the beam via the direct TE-line. However, if the FRS scheduling allows, we would prefer to utilize its purification capability since the total rate in the ESR will be quite large when using the TE line. This might lead to a rather complicated analysis of the high density of frequency lines, though there are no critical contaminants in the immediate vicinity of $^{194}$Pb frequency lines. In addition, the isotope of interest will present only ~3% of all stored nuclides. Using the FRS will increase this proportion to 20%, while still maintaining a reasonable rate for $^{194}$Pb of 420 pps.

In both parts of the experiment the estimated rates are high enough to *perform the pure data taking within ~3 shifts,* even taking into account that we might require to further reduce the Bρ acceptance inside the ESR in order to achieve the necessary mass resolution as shown in experiment E143. For the more challenging cases of $^{98}$Zr and $^{194}$Pb we expect to observe an isomer decay every few spills. With a repetition time of 10 seconds we should be able to select a few hundred events per shift. However, the experience from the previous experiment has

---

[1] The same result could of course also be obtained without a degrader in the FRS.

shown that the main part of the beamtime will go into the *preparation of the isochronous mode of the ESR in order to achieve the necessary mass resolution of <$10^{-5}$* to resolve the isomer from the ground state. Therefore, *we also request 2 days of parasitic preparation time with any (Z>6) direct SIS18 beam.*

In conclusion, we request **15 shifts with a $^{100}$Mo beam at 450 MeV/u** to study $^{98}$Mo and $^{98}$Zr and **15 shifts with a $^{209}$Bi beam at 550 MeV/u** to study $^{194}$Pb. This time includes for each isotope *3 shifts for setting-up and commissioning time of the ESR* in order to assure optimal operation conditions of the ESR in the isochronous mode. For the experiment the direct beam from SIS18 can be used so that the FRS could be used in parallel.

## 8. Three Major Publications of the Spokesperson(s)

K. Wimmer, W. Korten et al.,
Shape changes in the mirror nuclei $^{70}$Kr and $^{70}$Se.
Phys. Rev. Lett. 126, 072501 (2021)
https://doi.org/10.1103/PhysRevLett.126.072501

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

M. Steck and Yu.A. Litvinov,
*Heavy-ion storage rings and their use in precision experiments with highly charged ions*
Prog. Part. Nucl. Phys. 115, 103811 (2020)
*https://doi.org/10.1016/j.ppnp.2020.103811*

## *References*

[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] T. Kibédi, T.W. Burrows, M.B. Trzhaskovskaya, P.M. Davidson, C.W. Nestor Jr., Nucl. Instr. Meth. A 589, 202 (2008) and priv. comm.

[8] G. C. Rodrigues et al., At. Data Nucl. Data Tables 86 (2004) 117-233.

[9] F. Bosch et al., EPJ Web of Conf. 123, 04003 (2016)

[10] 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

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

[12] C. Walz et al., Nature 526, 406 (2015)
P.A. Söderström et al, Nature Communication 11, 3242 (2020)

[13] M. Hausmann et al., Nucl. Instr. Meth. A 446, 569 (2000)

[14] J. Henderson et al., Phys. Rev. C 89, 064307 (2014)

[15] C. Brandau, et al., Phys. Scr. 2013, 014050 (2013) and references therein

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

[17] Yu.A. Litvinov, Nucl. Phys. A 756, 3 (2005)
F. C. Ozturk, Phys. Lett. B 797, 134800 (2019)

[18] B. H. Sun et al., Nucl. Phys. A 812, 1 (2008)
B. Franzke, H. Geissel, and G. Münzenberg, Mass Spectr. Rev. 27, 428 (2008)

[19] F. Nolden et al., Nucl. Instr. Meth. A659, 69 (2011)
S. Sanjari et. al. Phys. Scr. 2013 014088 (2013)

[20] C. Trageser et al., Phys. Scripta T166, 014062 (2016)

[21] B. Sun et al., GSI Scientific Report 2011, 163, PHN-NUSTAR-FRS-21

[22] P. Walker et al., Int. J. Mass Spectrometry 349, 247 (2013)

[23] X.L. Tu et al, Phys. Rev. C 97, 014321 (2018)

[24] Y. H. Zhang et al., Phys. Scripta 91, 073002 (2015)

[25] P. Kienle et al., Phys. Lett. B 726, 638 (2013)

[26] Yu. A. Litvinov, Nucl. Phys. A 734, 473 (2004)

[27] S. Sanjari et al., Rev. Sci. Instr. 91, 083303 (2020)

# Technical requirements ESR

**G-22-00018-2.1-E**

| | | |
|---|---|---|
| Target station<br>**FRS-ESR** | Allocated experiment time: | Link scientist:<br>Yury Litvinov |

**Mode of operation:** **Main beam**

Comments, e.g. on n° of runs: We request two runs each for 15 shifts; one with 100Mo beam and ESR only, the other one with 209Bi beam and FRS-ESR.

N° of days for set-up and disassembling w/o beam (if > 2 days) [days]

**Ion Beam Specifications (*for parasitic mode please enter 'none' or '0' in the obligatory fields)*:**

Ion Species and Isotope (e.g. 197-Au) 209Bi

Enriched? ☒ Yes ☐ No

Charge State (e.g. 67)

Energy (e.g. 1250 MeV/u) 550 [MeV/u]

Intensity [particle nA, ions/s] e.g. 1e11 ions/s 1E9

Pulse Duration 50 [ns]

Duty Cycle (e.g. 5 Hz) [Hz]

On SIS18 ☐ slow extraction ☒ fast extraction

Extraction time needed? (e.g. 10 s) 10 [s]

Special requests on beam properties Isochronous mode of the ESR should have been established previously with parasitic beam from SIS18

**Additional information**

Use of ... ☒ an existing setup ☐ a new setup

Detector(s) used in experiment Schottky detectors and ESR in isochronous mode

# Technical requirements ESR

**G-22-00018-2.2-E**

| | | |
|---|---|---|
| Target station<br>**ESR** | Allocated experiment time:<br>**15 Shifts** | Link scientist:<br>Yury Litvinov |

**Mode of operation: Main beam**

Comments, e.g. on n° of runs: We request two runs each for 15 shifts; one with 100Mo beam and ESR only, the other one with 209Bi beam and FRS-ESR.

N° of days for set-up and disassembling w/o beam (if > 2 days) [days]

**Ion Beam Specifications (*for parasitic mode please enter 'none' or '0' in the obligatory fields)*:**

Ion Species and Isotope (e.g. 197-Au) 100Mo

Enriched? ☒ Yes ☐ No

Charge State (e.g. 67)

Energy (e.g. 1250 MeV/u) 450 [MeV/u]

Intensity [particle nA, ions/s] e.g. 1e11 ions/s 1E9

Pulse Duration 50 [ns]

Duty Cycle (e.g. 5 Hz) [Hz]

On SIS18 ☐ slow extraction ☒ fast extraction

Extraction time needed? (e.g. 10 s) [s]

Special requests on beam properties Isochronous mode of the ESR should have been established previously with parasitic beam from SIS18

**Additional information**

Use of ... ☒ an existing setup ☐ a new setup

Detector(s) used in experiment Schottky detectors and ESR in isochronous mode

# Safety Declaration

**G-22-00018**

## 1. General Safety

**Do you use combustable or hazardous gases within your experiment (e.g. gas target, gas detectors) ?**

☐ Yes ☒ No

If yes, what sort of gases?

Which quantities or flow rates?

(A flow scheme and description of the safety concepts have to be submitted to the Safety Engineers at GSI)

Upload safety concept

**Do you use any other dangerous (e.g. toxic, inflammable, biologically hazardous, etc.) materials / chemicals within your experiment?**

☐ Yes ☒ No

(Note: Only biological material of biological safety level 1 must be irradiated at GSI)

If yes, what sort of materials/chemicals?

Which quantities?

**Is your vacuum setup equipped with fragile parts like thin glass or foil windows, etc. (danger of implosion)?**

☐ Yes ☒ No

**Is it intended to move heavy parts for setting up your equipment or during the experiment?**

☐ Yes ☒ No

If yes, brief description of the equipment and working procedure:

## 2. Radiation Safety

**Do you use radioactive sources or materials onsite?**

☐ Yes ☒ No

If yes, which isotopes/type?

Which activities [Bq]?

**Do you use a target?**

☐ Yes ☒ No

If yes, position:

**Indicate thickness of target** [mm] or [g/cm²]**, and Interaction probability** [%] **with primary beam:**

Material: Car

**Do you use a secondary target/degrader?**

☒ Yes ☐ No

If yes, position: FRS and/or TE line (SIS18-ESR)

**Indicate thickness of target [mm] or [g/cm²]/ and Interaction probability [%] with primary/secondary beam:**

~2 g/cm2 Be or C, ~10% interaction probability

Material: Be and/or C

**Do you use a beam stop for primary/secondary beam?**

☒ Yes ☐ No

If yes, position: FRS

## 3. Electrical / Laser Safety

**Do you use electrical instruments that you bring on site?**

☐ Yes ☒ No

If yes, please describe devices above 1kV, self-made equipment etc.

**Do you use high-intensity radio frequency (rf) sources onsite?**

☐ Yes ☒ No

If yes, frequency region/power:

Brief description of the rf sources:

**Do you use lasers in your equipment?**

☐ Yes ☒ No

If yes, laser-type(s):

Max. power/energy:

Class:

Repetition rate:

## 4. Special Safety

**Is there any other special safety aspect to be considered in connection with your proposal?**

☐ Yes ☒ No

If yes, brief description:

# Host Lab Resources

**G-22-00018**

The timely knowledge on requirements of host lab resources by our users permits a solid in-house planning and allocation of respective resources. Please indicate here roughly, what you will need, and discuss details with the respective department later, if beamtime is granted. You might discuss your entries here with your link scientist before submission of your proposal.

**Target Laboratory**

Do you need targets from the department Target Laboratory?
☐ Yes ☒ No

If yes, please specify targets:

**Detector Laboratory**

Do you need support from the Detector Laboratory?
☐ Yes ☒ No

If yes, please specify:

**Experiment Electronics**

Do you need support from the Experiment Electronics department?
☐ Yes ☒ No

If yes, please specify:

**IT Department**

Do you need resources from the IT department?
☒ Yes ☐ No

Needed data storage: 100TB

Computing requirements: Data storage on analysis on Luster

Indicate further requirements here:

**Vacuum Systems**

Do you need support from the department Vacuum Systems?
☐ Yes ☒ No

If yes, please specify:

**Transport and Installation**

Do you need support from the department Transport & Installation for transporting or installing heavy equipment? (formerly "Großraummontage")
☐ Yes ☒ No

If yes, please specify:

**Mechanical Workshop**

Do you need resources from the department Mechanical Workshop?
☐ Yes ☒ No

If yes, please specify:

**Other Host Departments**

Do you need resources from other host departments?

☐ Yes ☒ No

If yes, please specify: