# Nuclear Excitation by Electron Capture (NEEC) Measurements using the ESR Electron Cooler as a Target for Free Electrons

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Abstract: We propose to investigate the resonant process of nuclear excitation by electron capture (NEEC) under clean experimental conditions at the ESR storage ring. A recent measurement using  $\gamma$ -spectroscopy in a beam-solid-target arrangement was found to deviate by 9 orders of magnitude from a theoretical study that tried to model the experiment. Earlier attempts by other groups and plans to study NEEC were unsuccessful or were never realized.

At GSI we will use an approach that is unambiguous by design: He-like  $^{238}$ U<sup>90+</sup> will be stored and phase-space cooled by means of stochastic cooling. The ESR electron cooler serves as a target of free electrons. By changing the relative energy between cooler electrons and stored ions the nucleus will be resonantly excited with simultaneous capture of a free electron into the atomic excited states  $1s^2 2p_{1/2}$  or  $1s^2 2p_{3/2}$ . These states decay by emission of x-rays to the atomic ground state  $1s^2 2s$  vastly faster than the nucleus. The subsequent decay of the nucleus predominantly proceeds via internal conversion (IC) and is time-delayed in such a way that the reaction products can be counted using position-sensitive particle detectors in and behind the ESR dipole magnet located immediately down-beam behind the cooler. Normalization on the number of stored ions and the electron density of the cooler will deliver the NEEC cross section on an independent absolute scale.

Requested Shifts: 39 (13 days)

Requested beam facility: SIS18, SIS cooler, ESR, ESR cooler, stochastic cooling

Requested beam:  $^{238}$ U, min.  $2 \cdot 10^9$  ions in SIS18

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#### **GOALS**

The main goal of this proposal is the first unambiguous experimental demonstration of the process of nuclear excitation by electron capture (NEEC). Stochastically cooled He-like <sup>238</sup>U<sup>90+</sup> ions will be employed at the electron cooler of the heavy ion storage ring ESR. The experimental procedure is similar to the one that is routinely used to study atomic resonant reactions within the framework of the so-called electron-ion collision spectroscopy [1–7]. Stored and phase-space cooled ions in well-defined quantum states collide with the electrons of the storage ring's electron cooler or a dedicated free-electron target. Using this technique, in atomic physics, the process of dielectronic recombination (DR) is routinely studied with high precision and on an independent absolute scale, thus allowing for a clear assessment and in-depth comparison between experiment and theory (cf. [1–9] and references therein). In the two-step process of DR the initial "dielectronic" capture (DC) of an electron is time-mirrored to the Auger effect, and can thus be seen as the atomic physics sibling of NEEC. It is proposed to extend this well-established method of collision spectroscopy to the study of the NEEC process.

The NEEC measurements at the ESR will be the first clear-cut experimental assessment of the NEEC process. NEEC is the time-mirrored process of internal conversion (IC). Despite the simplicity of this fundamental process, an unambiguous experimental assessment is still pending. The theoretical examination [10] of a recent ion-beam-solid-target  $\gamma$ -spectroscopy measurement [11] revealed a 9-orders-of-magnitude discrepancy with knwon facts thus rendering doubts about the assignment of the experimental observations to the process of NEEC. The concept of the previous experiment in [11] is very involved with many different processes being entangled, finally, raising more questions than answers [10, 12]. In the fall of 2019 a full workshop was devoted to shed light on this discrepancy, to summarize the present status of NEEC experiments and theory as well as to discuss potential applications [12].

By its definition, in NEEC a free electron is resonantly captured into an atomic-shell vacancy under excitation of the nucleus. Thus, for a clean and unambiguous signature of the process the following conditions shall be met: (1) a free-electron target to exclude competing nuclear excitation processes such as Coulomb excitation, (2) well defined atomic and nuclear quantum states, to exclude bulk, many-particle, solid state, or NEET-like (nuclear excitation by electron transition) effects, (3) single collision conditions, (4) observation of the resonance condition, and (5) a quantitative determination of the resonance strength, i.e., the collision cross section. The latter allows for a robust comparison with theory. Our proposed experiment at the ESR is designed to meet all of the above criteria.

In our approach we will deploy a particular NEEC reaction path, termed nuclear resonant electron scattering with x-ray emission (NRESX) [13, 14] on an intense beam of stochastically cooled Helike  $^{238}$ U<sup>90+</sup> ions. In NRESX in the initial NEEC step, i.e., the inverse IC, the free electron is captured into an excited atomic state, in our case into  $1s^2 2p_{1/2}$  or  $1s^2 2p_{3/2}$  levels, thus forming a Li-like excited atomic configuration while the nucleus is excited from its  $0^+$  ground state to the first excited state (2<sup>+</sup>) at 44916 eV [15]. This intermediate state predominantly decays in a sequence of a fast  $2p_j \rightarrow 2s$  x-ray emission and subsequent time-delayed conversion of the  $|2^+; 2s\rangle$  state. This sequence produces a unique signature that allows one to discriminate the NRESX process against the 4 to 5 orders of magnitude stronger non-resonant electron capture channel of radiative recombination (RR) which is a dominant background channel for NEEC followed by photon emission. By tuning the electron energy of the cooler electrons with respect to the ion energy that is fixed by stochastic cooling, changes in the relative electron-ion collision energy are applied in order to scan the NEEC resonance shape.

# INTRODUCTION, MOTIVATION, SCIENTIFIC CONTEXT

The process of nuclear excitation by electron capture (NEEC) is the time-mirrored process of internal conversion (IC). In NEEC a free electron that possesses a matching kinetic energy is resonantly captured into a vacancy of the atomic shell under excitation of the nucleus. The required kinetic energy of the electron, i.e., the resonance energy is given by  $E_{\rm Res} = E_{\gamma} - E_{\rm B}$ where  $E_{\gamma}$  is the excitation energy of the nuclear transition, and  $E_{\rm B}$  the binding energy of the captured electron. The inverse process, IC is much better known to atomic and nuclear physicists. In IC, the de-excitation of the nucleus ejects an electron with a well-defined kinetic energy. In turn, this kinetic energy corresponds to the  $E_{\rm Res}$  of NEEC. As is well known, IC takes place in, but is not restricted to, neutral atoms [16]. In contrast, due to the need for the atomic vacancies, into which the free electron will be captured, NEEC is a reaction mechanisms that requires highly ionized matter. NEEC is expected to play an important role in hot laser-generated or stellar plasmas and/or nucleosynthesis scenarios [17–22]. In addition, NEEC is considered an effective mechanism to deliberately pump nuclear isomeric states or to trigger their decay [7, 23]. The latter feature also attracted some attention by large companies such as Google [24] which foster related basis research [25]. The goal is to utilize NEEC's potential to store and to release a substantial amount of energy on demand, and hence to provide a means to mitigate the storage problem of typical sources of renewable energy.

Although NEEC is a very elemental process at the interface of atomic and nuclear physics, the prerequisite for suitable atomic vacancies has hampered the experimental investigation of NEEC. Similar to IC (both processes are linked by detailed balance), large overlap of the atomic and nuclear wave functions, a large nuclear charge Z and a small  $E_{\rm kin}$  of the active electron are preferred for a sizeable resonance strength. Thus, suitable candidates for experiments possess similar nuclear and atomic transition energies, i.e.,  $E_{\rm B}$  should be of the same order as  $E_{\gamma}$ . Highly charged heavy ions with inner-shell vacancies are closest to meeting these criteria. The observation in plasmas is difficult due to the necessity to produce the high charge states and due to the lack of a very unique signature to discriminate NEEC and its decay products against competing background processes, e.g. photons from non-resonant recombination (RR), from resonant atomic recombination process such as dielectronic recombination (DR) and/or other atomic and nuclear reactions. The same is true for many beam-based scenarios in particular if Coulomb excitation of the nucleus may play a role. Until recently, several unsuccessful attempts by other groups as well as several merely proposed experiments have been reported in the literature or on conferences and workshops, e.g. [12, 26–28]. One promising experimental approach that is extensively discussed is to perform superelastic collisions, i.e., to start with a long-lived nuclear isomeric state whose decay is triggered by using NEEC via a doorway state slightly above the isomeric state [7, 23, 29, 30].

In a recent publication, the first experimental evidence of NEEC was claimed in a measurement that applies this "superelastic" approach [11]. In the beam–solid-state-target experiment  $\gamma$ -spectroscopy was applied. A specially designed combination of a transmutation target to produce  $^{93m}$ Mo isomers and a second target for subsequent immediate "gentle" stopping [31] was set-up to trigger the decay of the isomer via a NEEC transition to a state just 4.85 keV above the isomeric state. The collision energy and the (calculated) charge state distribution were chosen to ensure NEEC conditions (and not NEET), and the target materials were selected in order to minimize contributions by Coulomb excitation. NEEC was inferred from coincidences of a feeding photon (2.475 MeV) that populates the isomeric state and a cascade of three depletion photons (268 keV, 685 keV, 1478 keV) of the isomeric transition in  $^{93}$ Mo [11]. With this set-up the production and triggering of the isomer could be clearly proven. A theoretical examination of the experiment [10], however, revealed an about 9-

orders-of-magnitude discrepancy between experiment and theory and thus seriously questions the assignment of the observations to the NEEC process. This discrepancy is even more surprising since NEEC being the time-mirrored process of IC can be considered well understood from the theoretical point-of-view [13, 14, 32]. In addition, an order-of-magnitude estimate can be easily performed using detailed balance and tabulated conversion coefficients supporting the theoretical result. On the other hand, the experimental conditions are ambiguous in several aspects, e.g. the role of Coulomb excitation or the charge state distribution, to name just two aspects. Coulomb excitation is estimated by the authors of [11] to be of no significance, yet it is 4 to 5 orders of magnitude stronger than the calculated NEEC probability. Likewise, the calculated charge state distribution excludes an open K-shell. An open K-shell would allow for nuclear excitations accompanied by bound-bound transitions, so-called nuclear excitation by electron transition (NEET). NEET has already been observed and experimentally studied in some detail previously, cf. [33–36]. Yet, the coincidence condition on the photon cascade 2475 keV + 268 keV + 685 keV + 1478 keV artificially selects ions that have an excess energy close to 5 MeV, by far enough to ionize a K-shell electron with high probability, and in clear contradiction to the claimed charge state distribution. Thus, one can only speculate about the discrepancy between experiment and theory or whether the observed triggering of the nuclear level can be uniquely attributed to the NEEC process at all. In the fall of 2019 a 3-day workshop was conducted that was dedicated to the NEEC process, its applications and to potential methods for a clear and unambiguous experimental assessment [12].

Instead of trying to exclude alternative mechanisms that compete with the sought-after effect of NEEC, in our approach we propose to perform an experiment at the ESR storage ring which already by design allows for an unambiguous assignment of the experimental outcome to the process of NEEC. The list of experimental prerequisites that have to be met for a clear-cut NEEC study has been already noted in the previous section.

### EXPERIMENTAL DESIGN AND MEHTODS

## Theory background and experimental procedure

NEEC and IC are linked by detailed balance, most notably, if the inverse conversion proceeds into the ground state of the atomic shell. Thus, for a given combination of initial and final states, the resonance strength for NEEC is determined by the  $\gamma$ -transition rate  $A_{\gamma}$  and the partial conversion coefficient for the corresponding atomic level.

For our experiment we have chosen  $^{238}\mathrm{U}^{90+}$  which initially has two electrons ("He-like") in the K-shell and an empty L-shell. The even-even-nucleus ( $I^{\pi}=0^{+}$  ground state) has a first excited  $2^{+}$  state at an excitation energy of  $E_{\gamma}=44.916$  keV [15]. This state has  $A_{\gamma}(44.916\mathrm{keV};2^{+})=5.40\times10^{6}~\mathrm{s}^{-1}$  and a total conversion coefficient  $\alpha_{\mathrm{tot}}=609$  for neutral 238-uranium, corresponding to a total transition rate  $A_{\gamma+\mathrm{IC}}(44.916\mathrm{keV};2^{+})=(\alpha_{\mathrm{tot}}+1)\cdot A_{\gamma}=3.3\times10^{9}~\mathrm{s}^{-1}$ .

For He-like <sup>238</sup>U<sup>90+</sup> NEEC can proceed via capture of the initially free electron into the L-shell forming a Li-like <sup>238</sup>U<sup>89+</sup> with an excited nucleus. The partial conversion coefficients per electron for the Li-like ion, the conversion rates and the atomic intra-L-shell transition rates are [14]:

state	$lpha_{ m IC}$	$A_{\rm IC} [{\rm s}^{-1}]$	$A(2p_j \to 2s_{1/2}) \text{ [s}^{-1}]$
$2s_{1/2}$	4	$2.16 \times 10^{7}$	
$2p_{1/2}$	129	$7.00 \times 10^{8}$	$1.7 \times 10^{10}$
$2p_{3/2}$	67	$3.62 \times 10^{8}$	$8.5 \times 10^{13}$

Table I. Conversion coefficients, conversion rates and  $\Delta n = 0$  atomic transition rates of L-shell electrons for the first excited nuclear state (44.916 keV;  $2^+$ ) of  $^{238}\mathrm{U}^{89+}$ . Due to the large binding energy of the K-shell electron of > 130 keV K-conversion is energetically not possible.

It is apparent from these numbers that atomic transition rates are at least about a factor of 25 faster than the nuclear decays. This has several consequences: (a) NEEC into  $2p_j$ -states is followed with large probability by atomic intra-shell  $2p_j \to 2s$  x-rays. (b) Since  $\alpha_{2p_j} \gg \alpha_{2s}$  the lifetime of the nucleus is substantially prolonged. (c) Creation and decay of the excited nuclear state are separated in time, and consequently for the fast moving ion beam in the ESR also separated in space. (d) The population of the  $2p_j$ -states via NEEC proceeds with a much higher rate than the IC-decay of the 2s state. The latter evasion of regular detailed balance due to the rapid  $2p_j \to 2s$  x-ray yields an orders-of-magnitude higher resonance strength  $S_{\rm NEEC} = \int \sigma_{\rm NEEC} {\rm d} E_e$  for NEEC via the investigated channel compared to the standard NEEC process. Thus the intermediate x-ray facilitates the proposed NEEC experiment as described previously in [13]. The intermediate nuclear  $2^+$  state can decay by emission of a  $\gamma$ -photon or by IC. For the first pathway the acronym NEECX is commonly used:

$$^{238}\mathrm{U}^{90+}(1s^2;0^+) + e^- \rightarrow ^{238}\mathrm{U}^{89+}(1s^22p_j;2^+)$$
 
$$\rightarrow ^{238}\mathrm{U}^{89+}(1s^22s;2^+) + \text{x-ray}$$
 
$$\rightarrow ^{238}\mathrm{U}^{89+}(1s^22s;0^+) + \gamma \qquad (\text{NEECX}),$$

and for the latter NRESX (nuclear resonant electron scattering):

$$^{238}\mathrm{U}^{90+}(1s^{2};0^{+}) + e^{-} \rightarrow ^{238}\mathrm{U}^{89+}(1s^{2}2p_{j};2^{+})$$

$$\rightarrow ^{238}\mathrm{U}^{89+}(1s^{2}2s;2^{+}) + \text{x-ray}$$

$$\rightarrow ^{238}\mathrm{U}^{90+}(1s^{2}2s;0^{+}) + e_{\mathrm{IC}}^{-} \qquad (\text{NRESX}).$$

For the total pathway of NRESX the net charge state does not change: In the initial NEEC-step the ion captures an electron  $q+\to (q-1)+$  which due to the delayed IC leads back to the He-like charge state. The utilization of the NRESX pathway enables the detection of NEEC with a large detection efficiency by means of particle counters.

One of the genuine experiments that has been pursued since the advent of heavy ion storage rings is the technique of electron-ion collision spectroscopy [1–9]. Atomic recombination experiments, and especially dielectronic recombination measurements at storage ring coolers, are typically being performed by collecting all recombined product ions while the relative energy between electrons and ions is varied. The the electron energy of the cooler is ramped up and down in a swift and controlled way, thus introducing precise relative electron-ion collision energies. The recombination reaction products having changed their charge state during the capture process in the cooler can be easily separated from the circulating primary beam in the next dipole magnet behind the cooler and counted by means of particle detectors. These measurements have a large range of applications comprising atomic physics, nuclear physics, fundamental physics, astro- and plasma physics, cf. [1–9, 37] and references therein. At first sight, a similar procedure for NEEC under the conditions

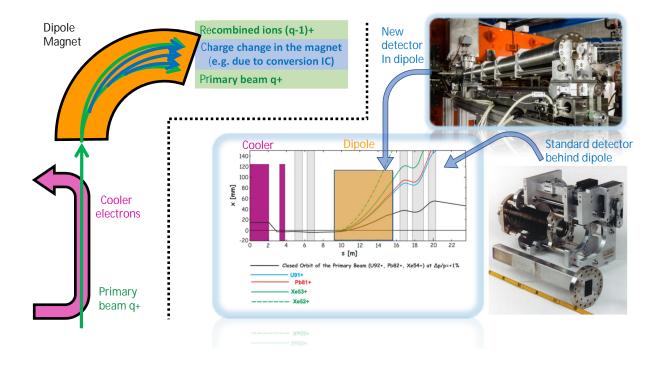
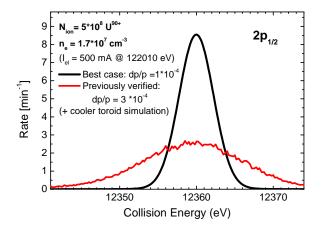


Figure 1. Experimental scheme for the proposed NRESX experiment at the ESR electron cooler. The ion beam is permanently stochastically cooled, and the cooler is used as a target for free electrons. Ions that resonantly capture an electron with simultaneous excitation of the nucleus as well as those which undergo non-resonant radiative recombination (RR) change their charge state from  $q+\to (q-1)+$  and will be separated from the primary beam in the next dipole magnet. In contrast to the prompt RR process, a large fraction ( $\approx 15\%$ ) of the excited nuclei undergoes IC, i.e. a change from (q-1)+ back to q+ **inside** the dipole magnet. On a position sensitive detector, these events will lead to signals **between** the ones from the primary beam and the ones from the recombined ions. These counts are unequivocal signatures from the NRESX (NEEC) process. The insert in the bottom middle shows the trajectories for a stochastically cooled primary beam and the recombined ions for several ions/charge states. The stochastic cooling requires an orbit that has a momentum deviation of up to +1% w.r.t. the nominal orbit. The positions of the particle detector pockets are at about 15 m and 21.6 m behind the center of the cooler and are indicated with blue arrows.

at the ESR appears promising. The total NEEC rate at the maximum of a resonance in  $^{238}$ U<sup>90+</sup> is of the order of 1–10 min<sup>-1</sup> which is a reasonable count rate (cf. Fig. 2). However, in this "brute force" approach this signal sits on a strong background signal of about 1-2 kHz that stems from radiative recombination (RR), i.e., the non-resonant capture of an electron under emission of a photon (time-inverse photo-effect). Observing NRESX instead of NEECX directly these prompt RR reaction products can be fully separated and suppressed (Fig. 1). For the proposed experiment, an intense, stochastically cooled  $^{238}$ U<sup>90+</sup> ion beam is stored in the ESR storage ring with an ion energy of 400 MeV/u ( $\beta_{\rm ion} = 0.715c$ ). The ESR electron cooler is freed from cooling tasks and is now used solely as an energy-tunable free-electron target that introduces the required relative electron-ion collision energies. It is worth noting that the application of this measurement scheme, i.e., the combination of a stochastically cooled ion beam and associated electron-ion collisions in the cooler has been successfully used by our group for precision high-resolution dielectronic recombination studies at the ESR, cf. [4]. For the proposed NEEC experiment with observation



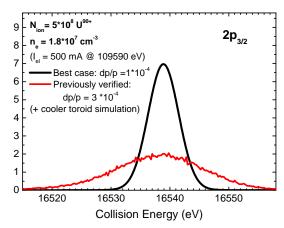


Figure 2. Total NRESX reaction rate associated with nuclear excitations  $0^+ \to 2^+$  and capture of the free electron into  $1s^2 \, 2p_{1/2}$  (left) and  $1s^2 \, 2p_{3/2}$  configurations (right) assuming a beam intensity of  $5 \times 10^8$  stored  $^{238}\mathrm{U}^{90+}$  ions. The black lines indicate an ion momentum spread of  $\delta p/p = 1 \cdot 10^{-4}$  which might be achieved in a best case scenario using notch-filter cooling [38]. In comparison, the red lines indicate a simulation with  $\delta p/p = 3 \cdot 10^{-4}$  which is a verified number that was achieved in previous experiments with stochastically cooled ion beams at the ESR. The latter simulation (red line) additionally takes into account the toroid sections of the electron cooler. Roughly 10 % of these products can be detected with our setup, see text. Expected beam intensities are  $2-5\cdot 10^8$  stored ions.

of the NRESX channel, a large fraction ( $\sim 15\%$ ) of the conversion processes is delayed and takes place *inside* the next dipole magnet behind the cooler (Fig. 1). Conversion in the dipole leads to orbits in the dipole that are located in *between* the ones of the  $^{238}\mathrm{U}^{90+}$  primary beam and the ones of the recombined ions  $^{238}\mathrm{U}^{89+}$ . Thus, using a position-sensitive particle detector the desired reaction products from NEEC/NRESX are separated from primary beam and recombined ions. The idea is sketched and supplemented by ion optical calculations in Fig. 1.

Using the relevant numbers for the proposed set-up at the ESR one obtains for the nuclear lifetime of the  $I^{\pi}=2^{+}$  state in Li-like uranium with the atomic configuration  $1s^{2} 2s_{1/2}$ , i.e., after the almost immediate  $2p_{j} \rightarrow 2s$  x-ray emission in the cooler:

$$\tau(^{238}{\rm U}^{89+}(I^*=2^+))=1/(\alpha_{2s_{1/2}}+1)\cdot A_{\gamma}^{-1}=37$$
ns

or  $T_{1/2}=25.7$  ns. At  $\beta_{\rm ion}=0.715c$  the relativistic Lorentz factor of the ion is  $\gamma_{\rm ion}=1.43$  leading to a half life in the laboratory system of  $T_{1/2}({\rm lab})=36.7$  ns which corresponds to a flight path of  $\sim 7.9$  m for one half life. The distance from the middle of the electron cooler to the entrance of the dipole is  $\sim 9.3$  m. There,  $\sim 44\%$  of the NEEC nuclei are still left in the  $2^+$ -state. Within the distance of  $\sim 6.5$  m that the ions travel in the dipole about  $\alpha_{2s_{1/2}}/(\alpha_{2s_{1/2}}+1)\cdot 45\%$  out of these ions undergo IC in the dipole. Thus, in total a number of  $\sim 16\%$  of the initial NEECX rate will lead to ions with trajectories between the two charge states 90+ and 89+ of primary beam and recombined ion beam, respectively. Since detection close to the primary beam is not possible due to the rim of the detector pocket and due to stray particle background from the intense primary beam we expect that 8-10% of the NEEC rate can actually be detected.

This approach to study IC of highly charged ions using a high-resolution ion spectrometer resembles the detection scheme used in single-pass experiments in Argonne by Phillips *et al.* [16, 39] or at GANIL by Attalah *et al.* [40, 41]. Table II gives the calculated resonance energies and resonance strengths for NRESX of  $^{238}U^{90+}$  [14, 42] and Fig. 2 provides the according simulations of the NRESX process in the ESR cooler assuming an ion beam momentum spread of  $\delta p_{\rm ion}/p_{\rm ion} = 1 \times 10^{-4}$  and  $\delta p_{\rm ion}/p_{\rm ion} = 3 \times 10^{-4}$ . For the latter case a full simulation of the relative electron-ion velocity

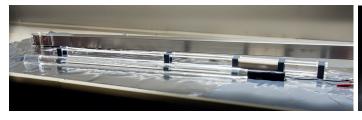




Figure 3. Newly developed detector pocket for the dipole magnet behind the electron cooler (left) and one-dimension position sensitive plastic scintillators. Left: scintillation detector with Bicron BC-418 / NE Pilot U head, thickness 5mm. BC-418 has a very short absorption length that leads to a continuous strong dependence of the pulse height on the particle impact position. Right: staircase-shaped plastic scintillator with discrete (10 steps) pulse height response to the impact position. Both detectors were successfully tested in 2015/2016.

distribution in the cooler was performed including the influence of the toroid bending sections. For the both simulations a cooler electron current of 500 mA and  $5 \times 10^8$  stored ions in the ESR were assumed, yet, the expected beam intensities in the ESR might be lower with roughly  $2-5 \times 10^8$  ions.

#### Count rate, detectors and background

Summarizing these numbers, for typical experimental conditions we expect a count rate in the maximum of the resonance of  $\sim 10-20$  counts per hour. With this count rate the proposed experiment is already feasible since background processes are essentially absent and/or well controlled (see initial discussion on RR suppression and estimates below). Still, control and reduction of background count rates are of primary importance. Realistic improvements that could be expected until the beam times in 2021/22 that would enhance this count rate by a factor of up to 20 are: (1) an increase of the ESR primary beam intensity to  $1 \times 10^9$  ions, (2) a higher electron density (×2), and (3) from improvements of the stochastic cooling technique using notch-filter cooling  $(\delta p_{\rm ion}/p_{\rm ion} = 3 \times 10^{-4} \to 1 \times 10^{-4})$  [38].

For low-noise position-sensitive detection in 2015/2016 one-dimensional position sensitive plastic scintillation counters were developed and successfully tested (Fig. 3). At the respective high ion energies of 400 MeV/u they possess a very narrow pulse height distribution. Using either a scintillation material with very *short* absorption length or a scintillator with variable (here: discrete) thickness the impact position can be derived to a level down to 1-2 mm for the continuous type and to the size of the staircase structures for the discrete type. The advantage of the latter type is that the position determination is more robust than for the continuous type. Both detectors showed a negligible dark count rate.

state	$E_{\mathrm{Res}}[eV]$	$S_{\text{NRESX}} = \int \sigma_{\text{NRESX}} dE_e \text{ [beV]}$
$2p_{1/2}$	12360.1	1.049
$2p_{3/2}$	16538.9	0.797

Table II. Theoretical resonance energies and resonance strengths for NRESX in  $^{238}$ U<sup>90+</sup> [14]. In supplement to the calculations in [14], QED energies for the atomic part [42] are used that slightly shift the resonances to lower energies. The uncertainty of the nuclear excitation energy of  $\sim$ 15 eV [43] exceeds the one from the atomic part by more than one order of magnitude.

The method how to suppress the non-resonant RR has been discussed in detail in the previous sections. Essentially all RR processes are prompt and appear in the cooler. It is noted, that at the CM collision energies well above 10 keV where the NEEC resonances occur the relative population of potentially long-lived high-n high-angular momentum Rydberg states arising from atomic RR and DR processes that can leave the cooler (and will be autoionized or field-ionized in the dipole) is suppressed by more than ten orders of magnitude [44] compared to the relative population at cooling conditions and is thus completely negligible.

For capture in the residual gas and for a typical ring vacuum of  $5 \times 10^{-11}$  mbar that possesses a H<sub>2</sub>-dominated residual gas composition, for  $10^9$  ions we expect a total capture rate of  $7 \times 10^{-6}$  s<sup>-1</sup>. This corresponds to less than one count per day, combined for both, non-radiative capture (NRC) and radiative electron capture (REC). The rate applies for the whole ring, but only the cooler section would contribute to the background. It is also noted, that the K-shell is filled. In absence of electron cooling,—the beam is continuously stochastically cooled—, the main loss mechanism (but not background) in the residual gas is ionization. Having said that, for the experiment very good vacuum conditions are required. The operation of the internal gas-jet target with heavy gases (Z > 2), and in particular difficult-to-pump heavy noble gases such as xenon, must be avoided in the weeks preceding the experimental run, since according capture rates scale with large powers of Z. In former experiments at the ESR it was anyhow good practice for collision experiments at the electron cooler to avoid heavy gases at the jet target prior to the experiment runs.

At present we consider single ions that are randomly lost from the primary beam and that fly with but that are not confined to the circulating ion beam in the storage ring as the main background process. Such single-ion "beam halos" might be created in particular during the kicking action of stochastic cooling but also due to intra-beam scattering. These background processes can be visualized and investigated using Schottky noise spectroscopy by means of the New continuous Schottky Time CAPture data acquisition (NTCAP) [45, 46]. According to our experience, using scrapers and unused detector pockets of the ESR to block most of the unoccupied acceptance of the ring such stray particles can be largely suppressed. In order to optimize the according settings we foresee at least three shifts at the beginning of the experimental run.

In addition, the NTACP system will be used to monitor the ion beam properties during the whole run. The non-destructive system has an enormous dynamic range from primary beam intensities of  $> 10^9$  ions down to single-particle sensitivity [45, 46]. The NTCAP system will also be used for precise ion beam energy determination and its constancy during the run (see next section).

## JUSTIFICATION OF BEAMTIME REQUEST

#### Finding the resonance and energy determination

Although the count rates for NRESX are very encouraging, the most difficult task is to find the resonances. On one hand, the theoretical position of the resonance is not known to better than about 15 eV  $(1\sigma)$ , mainly due to the uncertainty of the nuclear excitation energy of  $E_{\gamma}=44916$  eV. The energy value itself is confirmed in several experiments [15] but in most cases no error bars are given. Only reference [43] dating back to 1972 provides an estimates of 15 eV uncertainty.

On the other hand, the a priori experimental value for the relative collision energy has an uncertainty of the same order. This comprises the energies of the two contributing collision partners, the electrons and the ions. The voltage of the electron cooler can be determined on an absolute scale to a precision of better than  $10^{-5}$  [47, 48] using a high-voltage divider [49] provided by the

Physikalisch-Technische Bundesanstalt, Braunschweig. For electron cooling conditions by definition average electron and ion velocities are the same with a corresponding well defined revolution frequency of the ions in the ring. With the NTCAP system the constancy of the frequency and its width, e.g., the energy variation of the ion beam can be monitored down to relative changes of  $\Delta E_i/E_i \sim 10^{-7}$ . The measured voltages need to be corrected for the space charge potential of the electron beam which can be done using Schottky spectroscopy. The determination of the space charge is well established and is described e.g. in [46–48]. In our case, the procedure is time-consuming and needs to be performed for three different relevant cooler voltages of about 219.42 kV (reference for the ion energy), for 122 kV (electron energy around the  $2p_{1/2}$ -resonance) and for 109.6 kV ( $2p_{3/2}$ -peak). The energy calibration has to be carefully performed directly in advance of the NEEC measurement to obtain good initial energy values for electron and ion beam and, hence, to be able to find the resonance. The ion beam energy can be monitored during the whole beam time using the NTCAP system. The electron energy calibration has to be repeated regularly or in case of a failure of the storage ring or the cooler.

## Scope of the experiment and requested beamtime

For the initial set-up of the ESR including stochastic cooling we estimate 2 working days (6 shifts). Several initial preparations and calibrations are required for the experiment. This comprises energy calibration of electron and ion beam as described in the previous section, position calibration of the detectors with an electron-cooled ion beam and measures to minimize the background count rate of the detectors. Since this requires new settings for the ring, a deceleration cycle and a check that the new orbit corresponds to the initial orbit at stochastic cooling energy, we estimate at least 3 shifts for this work. Position calibration of the detectors and careful background optimization requires further 3 shifts each, respectively. For the production run we foresee one week (21 shifts). Due to the initial uncertainties in the theoretical and the experimental energies we need to scan at least an electron-ion energy range of 100 eV with a step width of 2 eV to not miss the resonance. Thus, for one energy step we have about 3 h of measurement time not including down-times of the accelerator. At a count rate of 10/h in the maximum of the resonance (estimated using previously established values for the beam intensities and ion beam momentum spread) this translate to roughly 30 counts in the maximum or about 100 counts integrated over the resonance.

Our beamtime request is summarized in the following table:

Task	Shifts
Set-up of ESR, Cooler and stochastic cooling	6
Energy calibration of ion beam, electron cooler and stochastic cooling	3
Position calibration of particle detectors	3
Background minimization (see text)	3
Production run / data taking	21
End-of-run calibration of ion beam, electron cooler and stochastic cooling	3
Total (Set-up + Production run)	39 (13 days)

#### 3 Publications of Spokesperson

- 1. Y. Litvinov and F. Bosch, Beta-decay of highly-charged ions, Rep. Prog. Phys. **74** (2011) 016301.
- 2. F. Bosch, Y. Litvinov and T. Stöhlker, Nuclear physics with unstable ions at storage rings, Prog. Part. Nucl. Phys. **73** (2013) 84.
- 3. M. Steck and Y. Litvinov, Heavy-Ion Storage Rings and Their Use in Precision Experiments with Highly Charged Ions, Prog. Part. Nucl. Phys. (in press) (2020), arXiv:2003.05201 [nuclex]
- [1] R. Schuch, Nucl. Instr. and Meth. A **532**, 56 (2004).
- [2] R. Schuch and S. Böhm, J. Phys.: Conf. Ser. 88, 012002 (2007).
- [3] A. Müller, Adv. At. Mol. Opt. Phys. 55, 293 (2008).
- [4] D. Bernhardt, C. Brandau, Z. Harman, et al., Phys. Rev. A 83, 020701 (2011).
- [5] C. Brandau and C. Kozhuharov, in Atomic Processes in Basic and Applied Physics, Springer Series on Atomic, Optical, and Plasma Physics, edited by V. Shevelko and H. Tawara (Springer, Berlin, Heidelberg, 2012) pp. 283–306.
- [6] C. Brandau, C. Kozhuharov, M. Lestinsky, et al., Phys. Scr. T166, 014022 (2015).
- [7] M. Lestinsky, V. Andrianov, B. Aurand, et al., Eur. Phys. J. Spec. Top. 225, 797 (2016).
- [8] S. Schippers, Nucl. Instr. and Meth. B 267, 192 (2009).
- [9] S. Schippers, Nucl. Instrum. and Meth. B **350**, 61 (2015).
- [10] Y. Wu, C. H. Keitel, and A. Pálffy, Phys. Rev. Lett. 122, 212501 (2019).
- [11] C. J. Chiara, J. J. Carroll, M. P. Carpenter, et al., Nature 554, 216 (2018).
- [12] "Workshop: Time domain control of atomic shell for nuclear excitation, Lerici, Italy, 7-9 Oct 2019," https://www.epfl.ch/labs/lumes/neec-workshop-oct-2019/ (2019).
- [13] A. Pálffy, Z. Harman, C. Kozhuharov, et al., Phys. Lett. B 661, 330 (2008).
- [14] A. Pálffy and Z. Harman, Phys. Rev. A 77, 042704 (2008).
- [15] E. Browne and J. K. Tuli, Nuclear Data Sheets 127, 191 (2015).
- [16] W. R. Phillips, I. Ahmad, D. W. Banes, et al., Phys. Rev. Lett. 62, 1025 (1989).
- [17] V. I. Goldanskii and V. A. Namiot, Phys. Lett. B 62, 393 (1976).
- [18] N. Cue, J.-C. Poizat, and J. Remillieux, Europhys. Lett. 8, 19 (1989).
- [19] M. R. Harston and J. F. Chemin, Phys. Rev. C 59, 2462 (1999).
- [20] G. Gosselin and P. Morel, Phys. Rev. C 70, 064603 (2004).
- [21] P. Morel, J. Daugas, G. Gosselin, V. Méot, and D. Gogny, Nucl. Phys. A 746, 608 (2004).
- [22] C. J. Cerjan, L. Bernstein, L. B. Hopkins, et al., J. Phys. G: Nucl. Part. Phys. 45, 033003 (2018).
- [23] A. Pálffy, J. Evers, and C. H. Keitel, Phys. Rev. Lett. 99, 172502 (2007).
- [24] Ross Koningstein, Google Inc, Why Is Google Interested in NEEC?, Workshop: Time domain control of atomic shell for nuclear excitation, Lerici, Italy, 7-9 Oct 2019 (2019).
- [25] "Google funds EPFL research on nuclear phenomena," https://actu.epfl.ch/news/google-funds-epfl-research-on-nuclear-phenomena/ (2019).
- [26] F. Gobet, C. Plaisir, F. Hannachi, et al., Nucl. Instr. and Meth. A 653, 80 (2011).
- [27] "ECT\* Workshop: Atomic Effects in Nuclear Excitation and Decay, ECT\*, Trento, Italy, 15-19 June 2009," (2009).
- [28] "EMMI-JINA Workshop: Nuclear Physics Processes in Dynamic High Energy Density Plasmas, GSI-Darmstadt, Germany & ND London Center, UK," (Oct. 13 - 17, 2012).
- [29] S. A. Karamian and J. J. Carroll, Phys. Atom. Nuclei 75, 1362 (2012).
- [30] J. Rzadkiewicz, M. Polasik, K. Słabkowska, et al., Phys. Rev. C 99, 044309 (2019).

- [31] M. Polasik, K. Słabkowska, J. J. Carroll, et al., Phys. Rev. C 95, 034312 (2017).
- [32] A. Pálffy, W. Scheid, and Z. Harman, Phys. Rev. A 73 (2006), 10.1103/PhysRevA.73.012715.
- [33] S. Kishimoto, Y. Yoda, M. Seto, et al., Phys. Rev. Lett. 85, 1831 (2000).
- [34] S. Kishimoto, Y. Yoda, Y. Kobayashi, et al., Nucl. Phys. A 748, 3 (2005).
- [35] S. Kishimoto, Y. Yoda, Y. Kobayashi, et al., Phys. Rev. C 74, 031301 (2006).
- [36] X. Cai, W. Xu, W. Luo, et al., Nucl. Phys. A 874, 1 (2012).
- [37] D. W. Savin, N. S. Brickhouse, J. J. Cowan, et al., Rep. Prog. Phys. 75, 036901 (2012).
- [38] W. Maier, C. Dimopoulou, R. Hettrich, et al., in COOL2013 (Mürren, Switzerland, 2013) pp. 142–145.
- [39] W. R. Phillips, J. Copnell, D. W. Banes, et al., Phys. Rev. A 47, 3682 (1993).
- [40] F. Attallah, M. Aiche, J. F. Chemin, et al., Phys. Rev. Lett. 75, 1715 (1995).
- [41] F. Attallah, M. Aiche, J. F. Chemin, et al., Phys. Rev. C 55, 1665 (1997).
- [42] Y. Kozhedub, "Priv. Comm," (2009).
- [43] M. Schmorak, C. E. Bemis, M. J. Zender, N. B. Gove, and P. F. Dittner, Nucl. Phys. A 178, 410 (1972).
- [44] C. Brandau, Messungen Zur Photorekombination Hochgeladener Lithiumähnlicher Ionen, PhD thesis, Justus-Liebig-Universität, Gießen (2000).
- [45] C. Trageser, C. Brandau, C. Kozhuharov, et al., Phys. Scr. **T166**, 014062 (2015).
- [46] C. Trageser, Aufbau Einer Datenaufnahme Zur Integration von Schottky-Signalen in Atomphysikexperimenten an Speicherringen, PhD thesis, Justus-Liebig-Universität, Gießen (2018).
- [47] J. Ullmann, Z. Andelkovic, C. Brandau, et al., Nat Commun 8, 1 (2017).
- [48] J. Ullmann, Laserspektroskopie an hochgeladenen Bismutionen zum Test der Quantenelektrodynamik, PhD thesis, Friedrich-Schiller-Universität, Jena (2017).
- [49] J. Hällström, A. Bergman, S. Dedeoğlu, et al., IEEE Trans. Instr. Meas. 63, 2264 (2014).