# EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

# Shape-coexistence and shape-evolution studies for bismuth isotopes by insource laser spectroscopy and beta-delayed fission in <sup>188</sup>Bi.

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### Abstract

The Proposal aims at the two main goals: 1) the studies of shape-coexistence and shape-evolution phenomena in the long chain of bismuth isotopes (Z=83) by in-source laser spectroscopy measurements of isotopic shifts (IS) and hyperfine structures (hfs), and 2) beta-delayed fission ( $\beta$ DF) of two isomeric states in <sup>188</sup>Bi. Isomer-selective  $\beta$ DF-studies for <sup>188m1,188m2</sup>Bi isomers will enable us for the first time to investigate the spin-dependence of the  $\beta$ DF process and to check theoretical predictions of asymmetrical fission fragment mass-distribution in this region of nuclei. The measurements will be performed with the well-proven Windmill and MR-TOF MS/Penning Trap techniques.

Requested shifts: 29 shifts in one run on a UCx target, with RILIS.

## Section 1. Shape-coexistence and shape-evolution in the bismuth isotopes

### Subsection 1.1. Physics motivation and earlier experiments

The primary aim of this part of the experiment is to study, through the measurements of the changes in the nuclear charge radii and electromagnetic moments, the shape evolution in the bismuth isotopes (Z=83), which have one extra proton relative to the Z=82 shell closure. A comparison of charge radii and electromagnetic moments for the Bi and Tl (Z=81) chains will be one of the crucial outcomes of this study and will provide essential insight in the understanding of the origin of the low-lying 0+ states in the lead isotopes (see ref. [1]).

In the past several years, our collaboration has performed extensive successful laser and nuclearspectroscopic studies of shape evolution/coexistence in the lead region by using in-source laser spectroscopy with RILIS at ISOLDE. Long chains of Pb (Z=82) [2], Po (Z=84) [3], Tl (Z=81) [4], At (Z=85), Au (Z=79) [5] and Hg (Z=80) [6] isotopes were investigated, by employing the combination of particle detection techniques ( $\alpha/\beta/\gamma$ ) with the Windmill setup, and direct ion counting with the ISOLTRAP's multi-reflection time-of-flight mass spectrometer (MR-TOF MS). The bismuth isotopes are an important, albeit yet missing link in the charge radii systematics in this region, as they lie between the chains of predominantly spherical (in their ground states) lead isotopes [2] and polonium isotopes, which demonstrate a strong onset of deformation by approaching the neutron mid-shell at N=104 [3].

The emergence of shape coexistence (both experiment-wise and theory-wise) and underlying configurations in the lightest odd-A <sup>185-195</sup>Bi isotopes were discussed in detail in e.g. [7] (see also reference therein). Experimentally, all known odd-A bismuth isotopes have  $I^{\pi}=9/2 [\pi 1 h_{9/2}]$ ground state (with an exception of  $I^{\pi} = 1/2^+$  for <sup>185</sup>Bi [7]) and some of them have a coexisting  $1/2^+$ state (presumably oblate-deformed), due to proton excitation from the  $\pi 3s_{1/2}$  orbital, see, e.g. [7, 8]. However, the actual situation is expected to be much more complex, as can be illustrated by the example of a mid-shell isotope <sup>187</sup>Bi (N=104). As shown by potential-energy surface and particle-rotor calculations [7], the states in bismuth isotopes could be considered as originating from the coupling of a valence proton (e.g.  $\pi 1h_{9/2}$ ,  $\pi 3s_{1/2}$ , or  $\pi 1i_{13/2}$ ) to states in the lead core. In particular, by coupling a  $\pi 1h_{9/2}$  proton to three coexisting 0<sup>+</sup> states (spherical-oblate-prolate) in <sup>186</sup>Pb [9], one can expect three coexisting states in <sup>187</sup>Bi (spherical gs with I<sup> $\pi$ </sup>=9/2<sup>-</sup> and two excited deformed states, see Fig. 8 of [7]), while coupling the  $\pi 3s_{1/2}$  proton should result in coexistence of two closely-spaced (oblate-prolate)  $1/2^+$  states (Fig. 8c of [7]). One can expect that, for Bi isotopes with A<190 (N<107), the  $1/2^+$  states might become a mixture of oblate and prolate configurations, or a change of their character from predominantly oblate in the heavier isotopes to prolate in the lighter ones might even happen (see [7] for details). This could be revealed by different excited bands in these nuclei (see e.g. [10]) or by the charge radii / electromagnetic moments measurements, as considered in this Proposal. In the odd-odd Bi isotopes the coupling of the  $i_{13/2}$ ,  $p_{3/2}$  etc. neutrons to normal and intruder proton states may lead to the occurrence of isomers with different deformation [11].

A 'reciprocal' situation occurs in the chain of odd-A thallium isotopes, which can be considered as 'mirrors' for the Bi isotopes with respect to the Z=82 shell closure. Namely, for Tl the coexistence of the spherical  $1/2^+$  ground state ( $\pi 3s_{1/2}$ ) and of a weakly-oblate  $I^{\pi}=9/2^-$  [ $\pi 1h_{9/2}$ ] intruder state is well-documented in <sup>181-201</sup>Tl [1]. Therefore, the comparison of charge radii and electromagnetic moments for the Bi and Tl chains (both for the  $1/2^+$  and  $9/2^-$  states) will be one

of the crucial outcomes of this study.

In the past, Bi's electromagnetic moments and the changes of the charge radii,  $\delta < r^2 >$  were studied by laser spectroscopy at ISOLDE [12] with gas cell laser spectroscopy (for A=202-210, 212-213) and recently at IRIS@PNPI (Gatchina) (A=189-198, 211) [13], see Fig.1. The three most interesting finding are as follows:

1. A gradual onset of deformation in the 9/2 gs of the lightest isotopes (up to  $<\beta^2>^{1/2}\sim 0.2$  in <sup>189</sup>Bi, see Fig. 1). This effect is further stressed by comparing  $\delta < r^2 >$  values for the even-N Pb, Bi and Po isotopes, see Fig.2, which clearly show that the radii for the  $9/2^{-1}$  state start to deviate from the nearly-spherical trend of the lead isotopes from  $^{191}$ Bi (N=108) on, while the  $1/2^+$  gs of the Tl isotopes perfectly follow the Pb isotope trend (see Fig. 3). This onset of deformation in what was previously believed to be the spherical 9/2<sup>-</sup> ground state in odd-A Bi isotopes is a new phenomenon, and will be investigated in more detail in this work, by re-measuring a somewhat uncertain data for <sup>189</sup>Bi from IRIS [13] and measuring <sup>187</sup>Bi (N=104), for which this effect is expected to be maximized.



Fig. 1.  $\delta < r^2 >$  isotopic dependency for Bi nuclei [12,13].



Fig. 3. Comparison of  $\delta < r^2 >$  values for the even-N Pb [2] and for the  $(I^{\pi}=1/2^+)$  gs of the Tl isotopes.



Fig. 2. Comparison of relative  $\delta < r^2 >$  for the even-N Pb [2], Bi  $(I^{\pi}=9/2^{-})$  [13], Po [3] and odd-odd Bi  $(I^{\pi}=10^{-})$  isotopes.

2) At the same time the radii of odd-odd (even-A) bismuth isotopes follow the corresponding nearspherical trend of Pb isotopes until N=107 (<sup>190</sup>Bi) (see Fig. 2). This results in the strong odd-even staggering (see Fig. 1). This again stresses the importance of extending  $\delta < r^2 >$  measurements to at least <sup>187,188</sup>Bi

3) Large isomer shift for intruder Bi isomers  $(I^{\pi} = 1/2^{+})$  for <sup>193,195,197</sup>Bi (Fig. 1), which is the signature of shape coexistence in the corresponding nuclei. It is crucial to extend these measurements both for the heavier <sup>199,201,203</sup>Bi lighter <sup>189,191</sup>Bi isotopes. and Especially interesting is the behaviour of the isomer shift for <sup>189,191</sup>Bi where the well-known departure of the

energy of the  $1/2^+$  intruder states from the parabolic N-dependency (observed for adjacent

isotopic chains of Tl and Pb) occurs [1, 7].

On the neutron-rich side,  ${}^{214-218}$ Bi (N=131-135) need to be studied, as at these neutron numbers an inverse odd-even staggering in charge radii was found for  ${}_{85}$ At (IS534, unpublished),  ${}_{86}$ Rn,  ${}_{87}$ Fr and  ${}_{88}$ Ra isotopic chains (see [14] and references therein). This effect is believed to be connected with the possible stable octupole deformation in this region. Measurements for  ${}^{214-}$ <sup>218</sup>Bi will help to further delineate the region of this phenomenon.

### Subsection 1.2. Proposed laser spectroscopy with RILIS.

Three-step resonance ionization scheme for Bi atoms is well-known and was tested by RILIS in off-line [15] and on-line [16] experiments. In [16], the on-line measurements were carried out with a limited laser power P<sub>3</sub> available for the 3<sup>rd</sup> excitation step (copper vapor laser,  $\lambda$ =511 and 578 nm) and a linear dependence of the ionization efficiency  $\varepsilon$  on the laser power was demonstrated. Namely, with P<sub>3</sub>=1.5W,  $\varepsilon$ =1% was achieved on-line [16], to be compared to P<sub>3</sub>=6W and  $\varepsilon$ =6% in off-line tests [15]. With the presently-available Blaze Nd:YVO<sub>4</sub> laser for the 3<sup>rd</sup> step we can ensure P<sub>3</sub>=20W. Therefore, we can conservatively estimate that in the proposed experiment the Bi ionization efficiency should be not less than  $\varepsilon$ =10%, which was used for the yield calculations in Table 1.

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А	Ι	T <sub>1/2</sub> , s	Yield (expected, at 1.5		time,
			$\mu$ A and P <sub>3</sub> =20W) 1/s	measurement	shifts
218	(6-8)	33	220	WM/MR-TOF	0.5
217	(9/2)	98.5	>1.0E+03	MR-TOF	0.5
216m1	(6,7)	135	>2.0E+03	MR-TOF	0.5
216m2	(3)	396	>1.0E+03	MR-TOF	0.5
215g	(9/2)	456	2.1E+04	MR-TOF	- 1
215m	(29/2-25/2)	36.9	1.9E+03	MR-TOF	1
214	1	1190	>2.0E+04	MR-TOF	0.5
203m	1/2	0.305	2.8E+07	MR-TOF	0.5
201g	9/2	6180	1.1E+08	Penning trap (PT)	1.5
201m	1/2	3450	4.0E+07	PT	1.5
200g	7	2184	>1.0E+07	MR-TOF	
200m1	(2)	1860	>1.0E+07	MR-TOF	1
200m2	(10)	0.4	>1.0E+04	MR-TOF	
199g	9/2	1620	1.8E+08	PT	1.5
199m	(1/2)	1482	6.0E+06	PT	1.3
191g	(9/2)	12.4	4.50E+05	WM	0.5
191m	(1/2)	0.125	1.10E+03	WM	0.5
190m1	(3)	6.3	3.7E+03	WM	0.5
190m2	(10)	6.2	1.1E+04	WM	0.5
189g	(9/2)	0.658	2.1E+03	WM	0.5
189m	(1/2)	0.005	3	WM	1
188m1	(3)	0.06	60	WM	1
188m2	(10)	0.265	320	WM	1
187g	(9/2)	0.037	1.5	WM	1

Table 1. Estimated yields for Bi nuclei (see text for details). The simultaneous scans of both gs
and isomer(s) will be performed for isotopes with multiple states.

Table 1 shows that for all isotopes/isomers in question the yield is expected to be larger than 1

ion per second, which is quite accessible for in-source laser spectroscopy (cf. [2, 3]). The shortlived and predominantly  $\alpha$ -decaying isotopes <sup>187-191,218</sup>Bi will be studied with the Windmill setup, the other isotopes (longer-lived/beta-decaying) will be studied with the MR-TOF MS.

The excitation energies of the isomeric states in <sup>199,201</sup>Bi are slightly too low for separation using the MR-TOF technique, while their combined HFS spectrum is difficult to disentangle. Therefore we propose to study these cases using the ion counting behind the Penning Trap (PT), where we will benefit from the higher mass resolving power. These isotopes are also perfect test cases for a new Penning-trap phase-imaging detection technique [17], which is currently being developed for ISOLTRAP. This will be the first ever in-source RIS study combined with Penning Trap assisted ion counting. We thus request 1.5 shifts for each two cases.

It should be added that, as a by-product, a wealth of nuclear spectroscopic information  $(\alpha/\beta/\gamma)$  decays at the Windmill) will be obtained for the daughter Tl (after  $\alpha$ -decay) and Pb (after  $\beta$ -decay) without additional beam time requirements.

To summarize, we intend to study IS and hfs of  $^{187-191, 199-201, 203m, 214-218}$ Bi. Beam time request: 12 shifts + 1 shift for the reference measurements, **total 13 shifts**.

# Sect. 2. Beta-delayed fission (βDF) of isomerically-pure beams of <sup>188m1,m2</sup>Bi

About 6 years ago, our collaboration initiated  $\beta$ DF studies at ISOLDE. Our first experiments resulted in a discovery of unexpected asymmetric mass distribution of fission fragments (FFs) from <sup>178,180</sup>Hg, produced after the



Fig. 4. Calculated mass distributions (black) for even-N neutron-deficient isotopes (based on Möller et al. calculations [20]). The calculated yields are compared with experimental data, shown in red and blue insets for a selection of isotopes.

beta-decay of <sup>178,180</sup>Tl [18]. These data established a new region of asymmetric fission, in addition to the previously known 'classical' region of asymmetric fission in the heavy actinides. These studies generated a lot of interest from theory, aimed at clarifying the reason for the fission mass-asymmetry in the light lead region [19]. Fig. 4 shows the predicted fission fragments mass distributions calculated with the macroscopicmicroscopic approach by P. Möller et al. [20]. The confirmed calculations the asymmetric split mass of

 $^{178,180}$ Hg. Furthermore, calculations also reproduced the symmetric mass split around  $^{204}$ Rn and  $^{209}$ Ra (both shown in Fig. 4), measured at GSI via Coulex of relativistic radioactive beams. Therefore, one expects that a transitional region between the asymmetric mass split of  $^{178,180}$ Hg and symmetric split of nuclei around  $^{204}$ Rn/ $^{209}$ Ra should occur, somewhere in nuclei along the red sloped line in Fig. 4. Motivated by these predictions, in the follow-up experiments at ISOLDE our collaboration successfully studied  $\beta$ DF of  $^{194,196}$ At and  $^{200,202}$ Fr [20], in which, in



Fig. 5. HFS spectra for the two isomers in <sup>190</sup>Bi [13]. Vertical lines mark the frequency positions for the narrow-band 1st step laser with the pure low-spin (blue) or high-spin (red) isomer production.

contrast with the Moller's model, multimodal fission of the daughter nuclides <sup>194,196</sup>Po and <sup>202</sup>Rn was discovered (see insets in Fig. 4). It is clear that these nuclei are indeed lying in the transitional region, where both asymmetric and symmetric mass splits mix — see detailed discussion in [20], including the potential-energy surfaces (PES) analysis, which illustrates the vast difference in the 'classical' actinide region of fission and in this new region of asymmetric (or multimodal) fission in the lead region.

In the present proposal we aim to study the lowenergy fission of  $^{188}$ Pb via the  $\beta$ DF of two isomeric states in <sup>188</sup>Bi and more specifically — to investigate the difference in the  $\beta DF$  properties of these two isomers. As seen from Fig. 4, <sup>188</sup>Pb lies exactly on the red line, mentioned above, in the middle of the transitional region. Recently BDF was firmly established for <sup>188</sup>Bi at the experiments at the velocity filter SHIP [21] and corresponding ratio N<sub>ff</sub>/N<sub>alpha</sub> was measured (N<sub>ff</sub> is the number of fission fragments, Nalpha is the number of alpha-decays for the same nucleus during the same time). However, due to the presence of two isomers in <sup>188</sup>Bi with comparable production rate, rather similar half-lives and yet unknown β-branching ratios (which are needed for determination of the BDF probability), the assignment of fission events to a particular isomer was impossible at SHIP. In the proposed ISOLDE-RILIS experiment a clean isomer separation will be possible, due to expected prominent difference in the HFS structure for two isomers. The HFS for <sup>188</sup>Bi will be measured in the first part of the experiment, dedicated to the IS/HFS studies (see Section 1), but it is expected that it will be very similar to the already measured HFS for <sup>190,192</sup>Bi at IRIS [13]. This is because the spins, parities and configurations for <sup>188,190,192</sup>Bi are identical, based on nuclear spectroscopy data [11,22]: high-spin (10<sup>-</sup>) isomers <sup>188,190,192</sup>Bi<sup>m1</sup> ( $\pi$ 1h<sub>9/2</sub>×v1i<sub>13/2</sub>), low-spin (3<sup>+</sup>). isomers <sup>188,190,192</sup>Bi<sup>m2</sup> ( $\pi$ 1h<sub>9/2</sub>×v3p<sub>3/2</sub>). Fig. 5 shows the HFS spectra for two isomers in <sup>190</sup>Bi measured at IRIS. It is clearly seen that with the properly-chosen frequencies of the 1st-step narrow-band laser one obtains isomerically-pure beams for each isomer. Correspondingly, the isomer separation will be certainly possible for <sup>188</sup>Bi, thus we will be able to determine which of the two isomers (or both?) undergo  $\beta DF$ .

Thus, the goals of this part of the study are two-fold: a) FFs mass distribution measurements of different states in <sup>188</sup>Pb, populated via  $\beta$  decay of two isomers in <sup>188</sup>Bi, and b) how the respective βDF probabilities compare, which will probe the spin-dependence of fission (this is a totally open question in fission). Table 2 gives the expected number of fission fragments per hour and of coincident fission events per day. (Reliable FFs mass distribution may be obtained only from the analysis of the coincident ff events). Alpha detection efficiency and coincidence detection efficiency were set to 0.6 and 0.21 respectively [21].

	Y, 1/s	$\alpha$ count, 1/s	$N_{\rm ff}/N_{alpha}$ [19]	N <sub>ff</sub> , 1/h	coincidence events, 1/day
<sup>188m1</sup> Bi (I=3)	6.00E+01	3.5E+01	2.66E-05	3.4	30
<sup>188m2</sup> Bi (I=10)	3.20E+02	1.9E+02	4.00E-05	26	220

Table 2. Expected numbers of fission events for βDF of <sup>188m1,m2</sup>Bi

In this estimation we (arbitrarily) supposed that ff-events in SHIP-experiment [21] were distributed equally between isomers. It was shown in [18, 21] that to obtain a reliable fission fragment mass-distribution 70-100 coincident fission events is sufficient. Therefore,  $\beta$ DF measurements for <sup>188m2</sup>Bi (I=10) and <sup>188m1</sup>Bi (I=3) need 1 day (3 shifts) and 4 days (12 shifts), respectively (with the 1st step laser frequency at the maxima of corresponding hfs spectra).

We will also try to search for  $\beta DF$  in <sup>190</sup>Bi. Our recent semi-empirical approach, connecting partial  $\beta DF$  half-life with the  $(Q_{\beta}-B_f)$  difference [23], predicts  $N_{ff}=(1\div 200)$  1/h (at the yields from Table 1). We are planning to spend **1 shift** to check this prediction. In the favorable case, the FFs mass distribution for <sup>190</sup>Pb will be determined. Even the upper limit for  $\beta DF$  probability will be helpful for the models comparison and for probing isospin dependence of  $\beta DF$ .

In total, we ask 16 shifts for  $Bi-\beta DF$  studies.

# Summary of requested 29 shifts

13 shifts for Bi in-source laser spectroscopy with RILIS (see Table 1) and 16 shifts for Bi  $\beta$ DF studies (see Table 2).

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# Appendix

# **DESCRIPTION OF THE PROPOSED EXPERIMENT**

The experimental setup comprises a Windmill system with 2-4 Si detectors inside, and 1-2 Ge detectors outside. WM system was successfully used in the runs IS387, IS407, IS456, IS466, I-086 and IS534, therefore solid understanding of all possible hazards is available.

Part of the Choose an item.	Availability	Design and manufacturing
ISOLTRAP/MR-TOF MS and	Existing	$\boxtimes$ To be used without any modification
Windmill		

# HAZARDS GENERATED BY THE EXPERIMENT

Hazards named in the document relevant for the fixed ISOLTRAP + MR-TOF MS installation.

Additional hazards:

Hazards	Windmill	ISOLTRAP + MR TOF MS
	w mamm	ISOLI KAP + MK TOF MS
Thermodynamic and fluidic		1
Pressure	-	
Vacuum	Standard ISOLDE vacuum	
Temperature		
Heat transfer		
Thermal properties of		
materials		
Cryogenic fluid	LN2, 2 Bar, 1501	
Electrical and electromagneti	c	·
Electricity	Standard power supplies	
Static electricity		
Magnetic field		
Batteries		
Capacitors		
Ionizing radiation	•	·
Target material [C-foils]	The C foils, where the radioactive samples are implanted, are very fragile. Should they break upon opening the Windmill, the pieces are so light that they would become airborne. Great care must be taken when opening the system and removing them (slow pumping/venting protective equipment: facial mask).	
Beam particle type (e, p, ions,		
etc)		
Beam intensity		
Beam energy		
Cooling liquids		
Gases		
Calibration sources:	<u>   </u>	
Open source		
Sealed source		
• Isotope	<sup>239</sup> Pu, <sup>241</sup> Am, <sup>244</sup> Cm	
Activity	1 kBq each	
Use of activated material:		

Description		
• Dose rate on		
contact and in 10 cm distance		
• Isotope		
Activity		
Non-ionizing radiation		
Laser	Standard RILIS operation	
UV light	*	
Microwaves (300MHz-30		
GHz)		
Radiofrequency (1-300MHz)		
Chemical	·	
Toxic	Pb shielding, 30-40 bricks	
Harmful		
CMR (carcinogens, mutagens		
and substances toxic to		
reproduction)		
Corrosive		
Irritant		
Flammable		
Oxidizing		
Explosiveness		
Asphyxiant		
Dangerous for the		
environment		
Mechanical		
Physical impact or	The chamber is heavy and	
mechanical energy (moving	needs to be handled with care	
parts)	during installation/removing	
Mechanical properties (Sharp,		
rough, slippery)		
Vibration		
Vehicles and Means of		
Transport		
Frequency		
Intensity		
Confined spaces		
High workplaces		
Access to high workplaces		
Obstructions in passageways		
Manual handling		
Poor ergonomics		

## 0.1 Hazard identification

3.2 Average electrical power requirements (excluding fixed ISOLDE-installation mentioned above): Negligible