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Deformation and mixing of coexisting shapes in neutron-deficient polonium isotopes

Coulomb-excitation experiments are performed with postaccelerated beams of neutron-deficient $^{196,198,200,202}$Po isotopes at the REX-ISOLDE facility. A set of matrix elements, coupling the low-lying states in these isotopes, is extracted. In the two heaviest isotopes, $^{200,202}$Po, the transitional and diagonal matrix elements of the $2^+_1$ state are determined. In $^{196,198}$Po multistep Coulomb excitation is observed, populating the $4^+_2$, $0^+_1$, and $2^+_2$ states. The experimental results are compared to the results from the measurement of mean-square charge radii in polonium isotopes, confirming the onset of deformation from $^{196}$Po onwards. Three model descriptions are used to compare to the data. Calculations with the beyond-mean-field model, the interacting boson model, and the general Bohr Hamiltonian model show partial agreement with the experimental data. Finally, calculations with a phenomenological two-level mixing model hint at the mixing of a spherical structure with a weakly deformed rotational structure.

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I. INTRODUCTION

Nuclear shape coexistence is the remarkable phenomenon in which states at similar excitation energies exhibit different intrinsic deformations. By now it is established to appear throughout the whole nuclear landscape, in light, medium, and heavy nuclei [1]. A substantial number of data have been gathered in the neutron-deficient lead region, providing clear evidence for the coexistence of shapes in these nuclei from an experimental as well as a theoretical point of view.
Experimentally, shape coexistence is well established in mercury isotopes ($Z = 80$) around neutron midshell, e.g., the large odd-even staggering and large isomer shift in the measured charge radii [2]. Despite the relatively constant behavior of the $2^+_1$ energy and of the reduced transition probabilities $B(E2; 2^+_1 \rightarrow 0^+_1)$, a recent Coulomb-excitation study of the neutron-deficient, even-even $^{182-188}$Hg isotopes led to the interpretation of mixing between two different structures that coexist at a low excitation energy [3]. Mixing between a weakly deformed oblate-like band and a more deformed prolate-like band is proposed to gain importance when going towards neutron midshell nuclei. This mixing between two configurations is also predicted in recent theoretical efforts studying neutron-deficient mercury isotopes in the framework of the interacting boson model (IBM) with configuration mixing [4].

The $^{186}$Pb nucleus ($Z = 82$) is a unique case of shape coexistence since three $0^+$ states with different deformations have been observed within an energy span of 700 keV [5]. Also, many other lead isotopes display signs of shape coexistence [6]. However, the ground states of the neutron-deficient lead isotopes are found to stay essentially spherical while different shapes appear at low excitation energies [7,8].

In the polonium isotopes, above $Z = 82$, low-lying intruder states have also been identified. Early theoretical studies concluded that the ground state of the heavier $^{194-210}$Po isotopes remains spherical, with the first (oblate-like) deformed ground state appearing in $^{192}$Po [9]. A prolate deformation in the ground state was suggested for the lightest polonium isotopes with mass $A \leqslant 190$. These findings were supported by a series of experimental studies of the polonium isotopes employing a range of techniques that include $\alpha$-, $\beta$-, and in-beam $\gamma$-decay studies (e.g., see Refs. [6] and [10]). The intrusion of the deformed state, becoming the ground state, is an unexpected result as in the even-even mercury isotopes, which “mirror” the polonium isotopes with respect to $Z = 82$, the intruding $0^+$ deformed state never becomes the ground-state structure.

Recent results from the measurement of changes in mean-square charge radii $\delta(r^2)$ in a wide range of polonium isotopes point to an onset of deviation from sphericity around $^{199}$Po [8,11], which is significantly earlier, when going towards a lighter mass, than previously suggested (e.g., in [6]). Comparison of the mean-square charge radii of the polonium isotopes with their isotones below $Z = 82$, as shown in Fig. 1, suggests that the deviation from sphericity of the ground state sets in earlier above $Z = 82$ [8]. Extending the results towards the more neutron-deficient radon ($Z = 86$) and radium ($Z = 88$) isotopes could confirm this hypothesis [12]. The platinum isotopes with $Z = 78$ show a similar early, but less pronounced, onset of deviation from sphericity as the polonium isotopes [13,14].

The band structure of the neutron-deficient even-even polonium isotopes has been studied extensively. The relevant results of these studies are summarized in the energy systematics of $^{190-210}$Po shown in Fig. 1. Lifetime measurements of $^{194,196}$Po [15,16] and inelastic scattering studies of $^{210}$Po [17] provided information on reduced transition probabilities. The level structure of the polonium isotopes was interpreted as an anharmonic vibrator in, e.g., [18]. Although vibrational characteristics can be identified in the level systematics of the polonium isotopes, the observation of the downsloping trend of the $0^+_1$ states in $^{196-202}$Po is hard to fit into the vibrational picture. Recent literature and theoretical efforts have provided more evidence that points toward the importance of intruder structures [9,16,19].
Studies within the IBM point out that the energy systematics in platinum isotopes conceal the presence of two different structures, which are reproduced with the inclusion of configuration mixing [20]. Also, in the polonium isotopes, an increasing admixture of deformed configurations in the ground and isomeric states is proposed based on in-beam, α-decay, and lifetime studies [15,21,22]. Recent beyond-mean-field (BMF) studies of polonium isotopes result in potential-energy surfaces that are soft for heavier polonium isotopes (A > 198), pointing toward the possibility of triaxial structures [19].

Theoretical descriptions, such as phenomenological shape-mixing calculations [23–26], contemporary symmetry-guided models [4], and BMF approaches [19], can reproduce the elements, which are good fingerprints for shape coexistence [27]. It provides access to transitional and diagonal matrix nuclear quadrupole deformation in a model-independent way between coexisting states is missing for most of the isotopes in the lead region. However, more subtle experimental information was determined using scattered particles on the DSSSD during laser-ON/OFF runs. The purity is defined as the fraction of polonium isotopes in the beam and covered and

<table>
<thead>
<tr>
<th>A</th>
<th>$T_{1/2}$ (s)</th>
<th>$I_{Po,av}$ (pps)</th>
<th>Purity (%)</th>
<th>Target</th>
<th>$\theta_{CM}$ (deg)</th>
<th>$T_{exp}$ (min)</th>
<th>Year</th>
</tr>
</thead>
<tbody>
<tr>
<td>196</td>
<td>5.8(2)</td>
<td>2.3(2) × 10^4</td>
<td>59.51(7)</td>
<td>$^{196}$Po</td>
<td>66–128</td>
<td>1687</td>
<td>2012</td>
</tr>
<tr>
<td>198</td>
<td>106(2)</td>
<td>4.6(7) × 10^4</td>
<td>95.97(19)</td>
<td>$^{196}$Mo</td>
<td>66–128</td>
<td>1235</td>
<td>2012</td>
</tr>
<tr>
<td>200</td>
<td>691(5)</td>
<td>2.54(17) × 10^3</td>
<td>97.90(4)</td>
<td>$^{197}$Pd</td>
<td>77–136</td>
<td>2424</td>
<td>2009</td>
</tr>
<tr>
<td>202</td>
<td>26.8(2) × 10^2</td>
<td>6.67(7) × 10^4</td>
<td>98.3(2)</td>
<td>$^{198}$Pd</td>
<td>66–128</td>
<td>196</td>
<td>2012</td>
</tr>
<tr>
<td>202</td>
<td>26.8(2) × 10^2</td>
<td>4.69(1) × 10^4</td>
<td>98.1(2)</td>
<td>$^{198}$Mo</td>
<td>66–128</td>
<td>170</td>
<td>2012</td>
</tr>
</tbody>
</table>
projectile-target combination was well below the “safe value” ensuring a purely electromagnetic interaction between the colliding nuclei. States up to $4^+_1$ and $2^+_2$ were populated. The choice of the respective target for each isotope (see Table I) was made considering the $\gamma$-ray energies de-exciting the $2^+_1$ states in the beam and target, to avoid an overlap, and the excitation probability of the target nucleus. The scattered particles are detected with a double-sided-silicon-strip detector (DSSSD), which is also mounted in the target chamber and is divided into 48 secular strips, coupled pairwise and read out by 24 ADC channels, and 16 annular strips to ensure position sensitivity [35]. The distance between target and DSSSD was 32.5 mm during the experiment in 2009 and 26.5 mm in 2012, yielding an angular coverage of $15.5^\circ < \theta_{\text{LAB}} < 51.6^\circ$ and $18.8^\circ < \theta_{\text{LAB}} < 57.1^\circ$, respectively. The $\gamma$ rays are detected with the Miniball Ge-detector array that surrounds the target chamber in close geometry. The Miniball detector array consists of eight cluster detectors, of which only seven were operational during both experimental campaigns. Each cluster contains three individually encapsulated hyperpure germanium crystals, which are in turn divided by segmentation of the outer electrode into six segments and a central electrode. The high granularity of the Miniball detectors assures position sensitivity for the $\gamma$-ray detection as well. A combination of $^{152}$Eu and $^{133}$Ba calibration sources was used to calibrate the energy and to determine the absolute detection efficiency of Miniball over the entire relevant energy range. Caution was paid to the low-energy range so as to ensure a good description of the absolute photon-detection efficiency in the polonium $\gamma$-ray region [33]. More specifically, relative efficiency curves were normalized to absolute efficiencies using $\gamma\gamma$ coincidences [34].

B. Data taking at Miniball

The specific timing properties of REX-ISOLDE beams have an implication for the method of data taking at Miniball. As the beam delivered to the REX linear accelerator is bunched, the radio-frequency cavities are not continuously operational. Triggered by the EBIS signal, the linac is switched on during an active window with a length of 800 $\mu$s and 1 ms for the 2009 and 2012 experiments, respectively. During the full 800 $\mu$s/1 ms window, the Miniball data system acquires all the information in the $\gamma$-ray and particle detectors. This window is called the “beam-on” window. To ensure correct identification of all possible sources of background, during an equally long “beam-off” window the data acquisition system is turned on, 4–10 ms after the beam-off window, when no beam is coming from the linear accelerator.

Coincidences between a particle and a $\gamma$ ray ("p-$\gamma$ coincidences") are essential to select the interesting events (Coulomb-excitation events) among the background radiation. The $\gamma$-ray background originates from the room background, decay radiation from the radioactive beam, and x rays from the accelerator, while the particle background is essentially due to the elastic scattering process. Therefore a specific coincidence scheme is developed for the data system (see Fig. 16 in [34]). An 800-ns-wide coincidence gate is defined about each $\gamma$ ray that is detected in the Miniball array. Particles detected within this window are considered to be coincident with the $\gamma$ ray and treated as p-$\gamma$ events. In the case of high beam intensities at the Miniball secondary target, particles that do not fall inside the 800-ns coincidence gate can be downscaled. This means that all the coincident particles are registered, but only 1 in $2^N$ particles with a $\gamma$ ray outside of the coincidence gate is accepted (with $N$ the downsampling factor), thus reducing the dead time of the particle-detection electronics due to elastically scattered particles. This downsampling method was applied for all polonium isotopes (with downsampling factor $N = 4$) except for $^{208}$Po, where the beam intensity was significantly lower (see Table I). However, during the 2012 experimental campaign the p-$\gamma$ coincidence gate was not set correctly for two of the four quadrants of the DSSSD. This gave rise to downsampling of the p-$\gamma$ coincidences instead of the particles without coincident $\gamma$ rays for half of the data. The consequences of this incorrect downsampling procedure are discussed in more detail in Sec. III A 2.

III. OFF-LINE DATA ANALYSIS

A. Selection of events

1. Selection based on kinematic properties

The Miniball detection setup registers a large number of data on scattered particles and decay radiation. As Coulomb-excitation events are hidden in this background, identifying these events of interest is a crucial step in the data analysis. The detected particle energy in the DSSSD as a function of the scattering angle in the laboratory frame of reference $\theta$ for $^{200}$Po on $^{104}$Pd is shown in Fig. 2. It shows a typical inverse kinematics scattering pattern. The recoiling target atoms (hereafter called “recoils”) are scattered throughout the whole detection range of the DSSSD, while the heavier beam particles are detected only at smaller scattering angles in the laboratory frame of reference. It is thus possible to make a distinction between a beam particle and a recoil, based on the energy-versus-angle kinematics. Detected particles in the

![FIG. 2. (Color online) Particle energy versus scattering angle in the laboratory frame of reference $\theta_{\text{LAB}}$ for $^{200}$Po on $^{104}$Pd. The color scale on the vertical axis represents the intensity in each bin. Only particles that are coincident with at least one $\gamma$ ray are shown. Gates chosen to select the $^{104}$Pd recoil are shown in black. The two innermost strips are not taken into account, as it is not possible to distinguish between the beam and the recoil particles in this angular range.](054301-4)
FIG. 3. (Color online) Time difference between particle and γ ray, gated on the $2_+^+ \rightarrow 0_+^+$ γ-ray transitions (blue lines) and gated on polonium x rays (red lines). The prompt and random coincidence windows used are shown in black. (a) Sum of $p$-γ timing information for all quadrants (2009 data of $^{200}$Po on $^{104}$Pd). (b) $p$-γ timing information for quadrants 1 and 2 where the downscaling was not properly set (2012 data of $^{198}$Po on $^{94}$Mo).

DSSSD related to the scattering of beam on target are selected by “following” the recoils through the range of the DSSSD. For each case, specific E-versus-θ gates were adopted to select the recoils scattered in the particle detector and to avoid including noise into the analysis. As an example, the gates that were used for $^{200}$Po on $^{104}$Pd can be seen in Fig. 2. The two innermost strips of the particle detector were excluded from the analysis because in this region of the detector the beam and recoil particles are not separable. The range of center-of-mass scattering angles covered by applying this method is listed for each reaction in Table I.

2. Selection based on timing properties

Figure 3 shows the time difference between a particle and a γ ray detected during the Coulomb-excitation experiments on $^{200}$Po on $^{104}$Pd [Fig. 3(a)] and $^{198}$Po on $^{94}$Mo [Fig. 3(b)]. The different structure of the data on $^{198}$Po can be explained by a problem with the downscaling in 2012. A difference in time behavior is observed between the γ rays following Coulomb excitation (de-exciting the $2_+^+$ state in the polonium isotope) and the low-energy polonium x rays and is due to the energy dependence of the time response of the Ge detectors. The prompt $p$-γ coincidence window is defined broadly enough to include low-energy x rays. Random $p$-γ coincidences are selected with a second time window. In the normal case of the 2009 data on $^{200}$Po, the random window is chosen within the region where the events are not downscaled. This allows us to scale the prompt and random events using the difference in length of the two respective windows. Data with the wrongly downscaled events were treated in a slightly different way. As the prompt $p$-γ events fall inside the downscaled region in this case, the random window is also selected among the downscaled events.

The purification power of the event selection based on kinematics and timing is highlighted in Fig. 4, where all detected γ rays in the $^{200}$Po experiment are shown in Fig. 4(a). The γ rays following Coulomb excitation are not visible yet in this γ-ray energy spectrum. By selecting the prompt $p$-γ coincidences that satisfy the kinematic gates and subsequently subtracting the random coincidences from it, a clean γ-ray energy spectrum, associated with events following Coulomb excitation, is obtained [Fig. 4(c)]. As the γ rays of interest are emitted in flight, the angular information on the detected particle and γ ray can be used to perform a Doppler correction of the detected γ-ray energy. Finally, a γγ-coincidence window of 350 ns is defined to check for coincidences between the emitted γ rays.

B. Polonium x rays

In addition to the γ rays following the Coulomb excitation of target and projectile, the background-subtracted γ-ray energy spectra show, for all isotopes studied, two peaks, around 78 and 90 keV. These energies correspond to polonium $K_α$ and $K_β$ x rays, respectively. Origins of these polonium x

FIG. 4. (Color online) The γ-ray energy spectra shown here display the 2009 data of $^{200}$Po on the $^{104}$Pd target. (a) All γ rays that were detected. (b) Prompt γ rays (black line) and random γ rays [gray (red) line] that fulfill the kinematic conditions. (c) Subtracted prompt-minus-random γ spectrum.
The experimental data and theoretical predictions for the Coulomb excitation of the $^{204}$Po isotope are shown in Fig. 5. The ratio of the number of observed x rays to the number of (rescaled) expected x rays is plotted for all studied polonium isotopes. For $^{202}$Po two points are drawn: the circle represents the data on $^{194}$Pd; the square, the data on $^{206}$Po data. The solid horizontal black line represents the scaling factor with the associated uncertainty (dashed horizontal black lines) deduced from the $^{202,206}$Po data.

Using the efficiency-corrected intensity balance between the 871-eV and the 204-eV $x$ rays and the Coulomb-excitation cross section for $^{94}$Mo and $^{95}$Mo by a $^{198}$Po projectile, the $^{94}$Mo component in the target was determined to be $F_{94Mo} = 95(2)\%$. As the absolute Coulomb-excitation cross sections in the polonium isotopes are determined by normalization to the known Coulomb-excitation cross section probabilities of the low-lying excited states in $^{94}$Mo (Fig. 6) one $\gamma$-ray transition related to target excitation at 871 keV is expected. However, the background-subtracted $\gamma$-ray energy spectrum for $^{196}$Po on $^{94}$Mo in Fig. 7 shows a second transition around 200 keV. This $\gamma$ ray can be associated with Coulomb excitation of the $3/2^+$ state at 204 keV in the $^{95}$Mo impurity in the target as the FWHM of the peak decreases when a Doppler correction for the target recoil trajectory is applied to the $\gamma$-ray energies. The isotopic impurity of the target was independently observed in the analysis of other Coulomb-excitation experiments that used the same target.

![Figure 5](image1.png)

**FIG. 5.** (Color online) The ratio of the number of observed x rays to the number of (rescaled) expected x rays is plotted for all studied polonium isotopes. For $^{202}$Po two points are drawn: the circle represents the data on $^{194}$Pd; the square, the data on $^{206}$Po data. The solid horizontal black line represents the scaling factor with the associated uncertainty (dashed horizontal black lines) deduced from the $^{202,206}$Po data.

![Figure 6](image2.png)

**FIG. 6.** Level energies (in keV) and reduced transition probabilities for the low-lying excited states in $^{94}$Mo (Fig. 6) one $\gamma$-ray transition related to target excitation at 871 keV is expected. However, the background-subtracted $\gamma$-ray energy spectrum for $^{196}$Po on $^{94}$Mo in Fig. 7 shows a second transition around 200 keV. This $\gamma$ ray can be associated with Coulomb excitation of the $3/2^+$ state at 204 keV in the $^{95}$Mo impurity in the target as the FWHM of the peak decreases when a Doppler correction for the target recoil trajectory is applied to the $\gamma$-ray energies. The isotopic impurity of the target was independently observed in the analysis of other Coulomb-excitation experiments that used the same target.

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and mass 95) was used as a first-order estimate [44].

The excitation cross sections of the target (calculated with mass 94) was observed in 198Po in the 4\(^+\) state at 871 keV. In order to be sensitive to the second-order effect following the Coulomb excitation of the polonium isotope, the background-subtracted and Doppler-corrected spectrum for 198Po on 94Mo (black line) and Doppler corrected for the target [gray (red) line].

D. Experimental data analysis

This section describes the data analysis for the four isotopes studied in this work. For each isotope, the background-subtracted and Doppler-corrected \(\gamma\)-ray energy spectrum, following the Coulomb excitation of the polonium isotope, is shown. In order to be sensitive to the second-order effect of the diagonal matrix element of the 2\(^+\)+ state, the data are divided into a number of angular ranges. The adopted number of subdivisions per isotope depends on the statistics that were obtained in both the projectile and the target yields. The total statistics that were acquired, together with the deduced Coulomb-excitation cross section \(\sigma_{CE}\), are listed in Table III for all isotopes. The cross section was calculated using the integrated beam current, which was determined using the known cross section for Coulomb excitation of the target nucleus, taking into account the beam purity (see Table I), the target purity, and the Miniball detection efficiencies at the respective transition energies [33].

In the two heaviest isotopes studied, only the 2\(^+\)+ state was populated. The \(\gamma\)-ray energy spectra are shown in Figs. 8 and 9 for 202Po and 200Po, respectively. As in 196,198Po, multistep Coulomb excitation was observed; additional details related to the data analysis of these two isotopes are provided below.

I. Data obtained for 198Po

The background-subtracted \(\gamma\)-ray spectrum of 198Po on 94Mo is shown in Fig. 10(a). While in the 94Mo target, only the 2\(^+\)+ state was populated, multiple-step Coulomb excitation was observed in 198Po in the 4\(^+\), 0\(^+\)+, and 2\(^+\)+ states (see level scheme in Fig. 15). A clearer view of the 4\(^+\)+ \(\rightarrow\) 2\(^+\)+ transition results from gating on the 2\(^+\)+ \(\rightarrow\) 0\(^+\)+ \(\gamma\) ray at 605 keV [Fig. 10(b)]. There is only a weak indication of the transitions depopulating the 0\(^+\)+ and 2\(^+\)+ states, which is reflected in the size and relative error of the extracted intensities. The resulting intensities for all the observed transitions, together with the statistics in the 2\(^+\)+ \(\rightarrow\) 0\(^+\)+ transitions in projectile and target, are listed in Table III. The statistics on the 2\(^+\)+ \(\rightarrow\) 0\(^+\)+ transitions in the projectile and target nuclei allowed us to divide the data into five angular ranges.

D. Experimental data analysis

This section describes the data analysis for the four isotopes studied in this work. For each isotope, the background-subtracted and Doppler-corrected \(\gamma\)-ray energy spectrum, following the Coulomb excitation of the polonium isotope, is shown. In order to be sensitive to the second-order effect of the diagonal matrix element of the 2\(^+\)+ state, the data are divided into a number of angular ranges. The adopted number of subdivisions per isotope depends on the statistics that were obtained in both the projectile and the target yields. The total statistics that were acquired, together with the deduced Coulomb-excitation cross section \(\sigma_{CE}\), are listed in Table III for all isotopes. The cross section was calculated using the integrated beam current, which was determined using the known cross section for Coulomb excitation of the target nucleus, taking into account the beam purity (see Table I), the target purity, and the Miniball detection efficiencies at the respective transition energies [33].

In the two heaviest isotopes studied, only the 2\(^+\)+ state was populated. The \(\gamma\)-ray energy spectra are shown in Figs. 8 and 9 for 202Po and 200Po, respectively. As in 196,198Po, multistep Coulomb excitation was observed; additional details related to the data analysis of these two isotopes are provided below.

D. Experimental data analysis

This section describes the data analysis for the four isotopes studied in this work. For each isotope, the background-subtracted and Doppler-corrected \(\gamma\)-ray energy spectrum, following the Coulomb excitation of the polonium isotope, is shown. In order to be sensitive to the second-order effect of the diagonal matrix element of the 2\(^+\)+ state, the data are divided into a number of angular ranges. The adopted number of subdivisions per isotope depends on the statistics that were obtained in both the projectile and the target yields. The total statistics that were acquired, together with the deduced Coulomb-excitation cross section \(\sigma_{CE}\), are listed in Table III for all isotopes. The cross section was calculated using the integrated beam current, which was determined using the known cross section for Coulomb excitation of the target nucleus, taking into account the beam purity (see Table I), the target purity, and the Miniball detection efficiencies at the respective transition energies [33].

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TABLE III. Statistics obtained in the Coulomb-excitation experiments of 196–202Po on the 104Pd and 94Mo targets. \(N_{\gamma}\) represents the number of detected \(\gamma\) rays at the Miniball setup, and \(\sigma_{CE}\) is the deduced cross section for Coulomb excitation.

<table>
<thead>
<tr>
<th>Nucleus</th>
<th>Transition</th>
<th>(N_{\gamma})</th>
<th>(\sigma_{CE}) (b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>202Po</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>3.8(3) \times 10^2</td>
<td>0.45(6)</td>
</tr>
<tr>
<td>204Pd</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>1.04(4) \times 10^3</td>
<td>0.03(11)</td>
</tr>
<tr>
<td>204Pd</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>1.04(4) \times 10^3</td>
<td>0.03(14)</td>
</tr>
<tr>
<td>200Po</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>2.2(2) \times 10^2</td>
<td>0.39(8)</td>
</tr>
<tr>
<td>94Mo</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>75(13)</td>
<td></td>
</tr>
<tr>
<td>200Po</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>1.93(18) \times 10^4</td>
<td>0.48(3)</td>
</tr>
<tr>
<td>104Pd</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>4.37(3) \times 10^3</td>
<td></td>
</tr>
<tr>
<td>198Po</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>4.60(8) \times 10^3</td>
<td>1.00(16)</td>
</tr>
<tr>
<td>202Po</td>
<td>0(^+)+ (\rightarrow) 2(^+)+</td>
<td>171(39)</td>
<td>0.03(4)</td>
</tr>
<tr>
<td>200Po</td>
<td>0(^+)+ (\rightarrow) 2(^+)+</td>
<td>78(58)</td>
<td></td>
</tr>
<tr>
<td>202Po</td>
<td>2(^+)+ (\rightarrow) 2(^+)+</td>
<td>34(40)</td>
<td>0.010(12)</td>
</tr>
<tr>
<td>94Mo</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>5.3(3) \times 10^2</td>
<td></td>
</tr>
<tr>
<td>104Pd</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>6.05(9) \times 10^3</td>
<td>1.67(19)</td>
</tr>
<tr>
<td>198Po</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>373(41)</td>
<td>0.108(17)</td>
</tr>
<tr>
<td>198Po</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>79(12)</td>
<td>0.052(14)</td>
</tr>
<tr>
<td>104Pd</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>85(35)</td>
<td>0.06(3)</td>
</tr>
<tr>
<td>104Pd</td>
<td>2(^+)+ (\rightarrow) 0(^+)+</td>
<td>5.17(8) \times 10^3</td>
<td></td>
</tr>
</tbody>
</table>
FIG. 8. (Color online) Background-subtracted and Doppler-corrected γ-ray energy spectrum following the Coulomb excitation of 202Po, induced by the 202Po beam impinging (a) on the 104Pd and (b) on the 94Mo target. The gray (red) spectrum is Doppler corrected for the target; the black spectrum is Doppler corrected for the projectile. Observed transitions are highlighted.

The particle-gated γγ-energy spectrum also contains polonium x rays, which can be attributed to the conversion of observed coincident γ rays and to the E0 component of the $2^+_1 \rightarrow 2^+_1$ transition. The $4^+_1 \rightarrow 2^+_1$ transition, which is observed both in the “singles” particle-gated γ-ray energy spectrum and in the particle-gated γγ spectrum, is used to link the intensity in the γγ spectrum to the singles intensity. A scaling factor $S$ is defined as

$$S = \frac{I_{4^+_1 \rightarrow 2^+_1, p\gamma}}{I_{4^+_1 \rightarrow 2^+_1, p\gamma\gamma}}.$$  \hspace{1cm} (2) \hspace{1cm}

FIG. 9. (Color online) Background-subtracted and Doppler-corrected γ-ray energy spectrum following the Coulomb excitation of 200Po, induced by the 200Po beam impinging on the 104Pd target. The γ-ray energies are Doppler corrected for 200Po; the target Doppler correction is shown in Fig. 7. Observed transitions are highlighted. (b) Energy of γ rays coincident with the $2^+_1 \rightarrow 0^+_1$ γ ray at 605 keV in 198Po. The gated spectrum is background subtracted and Doppler corrected for 198Po. Observed transitions in 198Po are highlighted.

FIG. 10. (a) Background-subtracted and Doppler-corrected γ-ray energy spectrum following the Coulomb excitation of 198Po, induced by the 198Po beam on the 94Mo target. The γ-ray energies are Doppler corrected for 198Po; the target Doppler correction is shown in Fig. 7. Observed transitions are highlighted. (b) Energy of γ rays coincident with the $2^+_1 \rightarrow 0^+_1$ γ ray at 605 keV in 198Po. The gated spectrum is background subtracted and Doppler corrected for 198Po. Observed transitions in 198Po are highlighted.

Finally, following the method described in [46], 1σ upper limits were determined for the additional unobserved transitions. The upper limits, listed in Table IV, are taken into account in the Coulomb-excitation analysis described in Sec. IV B 1.

which is equal to 10(4) in this case. This factor is mainly determined by the γ-ray detection efficiency of the $2^+_1 \rightarrow 0^+_1$ transition and includes the difference in sorting procedure in the construction of the particle-gated γ spectrum and the particle-gated γγ spectrum [33]. The intensity of the polonium x rays in the γγ spectrum is then corrected for conversion of other γ-ray transitions in the γγ spectrum (assuming $\delta = 1.8(5)$ [45]) and for the (small) number of “atomic” x rays present in the γγ spectrum. Of this, a total of 80(130) x rays are associated with the E0 component of the $2^+_1 \rightarrow 2^+_1$ transition and 150(310) x rays are attributed to the E0 transition between the excited $0^+$ state and the ground state. Details of this calculation can be found in [33]. The large uncertainties of these numbers are due to the indirect method of determining these intensities.
TABLE IV. 1σ upper limits for unobserved transitions in the Coulomb excitation of 196Po on 94Mo. Values for the 1σ upper limits (UPL) are determined using the method described in [46] and are not efficiency corrected. The uncertainty of the upper limit represents the 1σ uncertainty of the value. F is the efficiency-corrected ratio of the 1σ upper limit to the intensity of the 21→01 transition.

<table>
<thead>
<tr>
<th>Transition</th>
<th>Energy (keV)</th>
<th>UPL</th>
<th>F</th>
</tr>
</thead>
<tbody>
<tr>
<td>61→41</td>
<td>559</td>
<td>44(30)</td>
<td>0.92%</td>
</tr>
<tr>
<td>22→02</td>
<td>223</td>
<td>31(50)</td>
<td>0.40%</td>
</tr>
<tr>
<td>22→01</td>
<td>1039</td>
<td>17(15)</td>
<td>0.48%</td>
</tr>
</tbody>
</table>

2. Data obtained for 196Po

The Coulomb excitation of 196Po was studied on a 104Pd target. The background-subtracted γ-ray energy spectrum in Fig. 11(a) shows that multistep Coulomb excitation was observed. The γ rays de-exciting the 41→21 and 22→02 states are certainly visible next to some lines that cannot be placed in the level scheme of 196Po (Fig. 15). The comparison of the Coulomb-excitation spectra, which were acquired during the laser-ON and laser-OFF periods of the laser-ON/OFF data in Fig. 11(a), shows that the unknown transitions originate from de-excitation of populated levels in the isobaric contaminant 196Tl.

The beam purity, time-integrated over all the laser-ON/OFF runs, was determined to be 59.51(7)%. The same method as in [32] was applied to extract the beam purity during the runs where the lasers were on continuously. In this approach, the intensity of the γ rays associated with Coulomb excitation of polonium (21→01 at 463 keV) and thallium (1→2 at 253 keV) were taken into account, together with an extrapolation factor from the laser-ON/OFF runs, yielding a total purity of 46(6)%). The larger relative error bar is due to the smaller statistics in the Coulomb-excitation transitions compared to the scattered particles on the DSSD. The target de-excitation γ-ray yields were extracted in a separate analysis for ON/OFF runs and ON runs, employing the respective correction factors for the beam purity.

The projection of the γγ-energy matrix with a gate on the 463-keV 21→01 transition is shown in Fig. 11(b). Of the 15(9) detected coincident Kα x rays, 4(2) are associated with E2/M1 conversion. The unknown E2/M1 mixing ratio of the 21→21 transition was taken into account by applying the same method as in the case of 198Po. The remaining 11(9) Kα x rays translate, using Eq. (2) to get to a scaling factor of S = 5.3(9), into 76(66) x rays that can be related to the E0 component of the 21→21 transition. The number of x rays originating from the 01→01 transition is compatible with 01→01 intensity is compatible with 01→01 given an upper limit of 140 counts. The detection of E00→01 transitions thus cannot be excluded.

Table III summarizes the intensities of the observed transitions in the Coulomb excitation of 196Po on 104Pd. Seven angular ranges were defined for the 21→01 transitions in target and projectile and for the 41→21 transition in 196Po to gain sensitivity to second-order effects. The deduced cross section for Coulomb excitation, σCE, is extracted based on the known cross section for Coulomb excitation of the target and taking into account the beam purity and the Miniball detection efficiencies at the respective transition energies. Finally, following the same method as in 198Po, 1σ upper limits were determined for the unobserved transitions (see Table V).

IV. GOSIA ANALYSIS

The unknown matrix elements coupling low-lying states in polonium isotopes are extracted using the coupled-channels Coulomb-excitation analysis code GOSIA [47,48]. Two approaches are used, depending on the number of states that are populated in the experiment. In the case where only the 21 state is populated, GOSIA2 is used (see Sec. IV A). When multistep
TABLE V. 1σ upper limits for unobserved transitions in the Coulomb excitation of 198Po on 104Pd. The values for the 1σ upper limits (ULPs) are determined using the method described in [46] and are not efficiency corrected. The uncertainty of the UPL represents the 1σ uncertainty of the value. $F$ is the efficiency-corrected ratio of the 1σ UPL to the intensity of the $2^+_1 \rightarrow 0^+_1$ transition.

<table>
<thead>
<tr>
<th>Transition</th>
<th>Energy (keV)</th>
<th>UPL</th>
<th>$F$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$6^+_1 \rightarrow 4^+_1$</td>
<td>499</td>
<td>9(20)</td>
<td>0.2%</td>
</tr>
<tr>
<td>$0^+_1 \rightarrow 2^+_1$</td>
<td>95</td>
<td>61(70)</td>
<td>0.6%</td>
</tr>
<tr>
<td>$2^+_2 \rightarrow 0^+_2$</td>
<td>301</td>
<td>48(60)</td>
<td>0.6%</td>
</tr>
<tr>
<td>$4^+_2 \rightarrow 4^+_1$</td>
<td>497</td>
<td>21(20)</td>
<td>0.4%</td>
</tr>
<tr>
<td>$4^+_2 \rightarrow 2^+_2$</td>
<td>529</td>
<td>528(50)</td>
<td>9.5%</td>
</tr>
<tr>
<td>$4^+_2 \rightarrow 2^+_1$</td>
<td>925</td>
<td>8(2)</td>
<td>0.2%</td>
</tr>
</tbody>
</table>

Coulomb excitation is observed, a combined approach using GOSIA and GOSIA2 is employed, as explained in Sec. IV B.

A. Exclusive population of the $2^+_1$ state

This section deals with the cases of 200,202Po in which only the $2^+_1$ state is populated. The Coulomb-excitation cross section is affected by the matrix element coupling the ground state to the populated $2^+_1$ state (01$^+_1$$\langle E2\rangle$$|2^+_1\rangle$) and, to second order, by the diagonal matrix element of the $2^+_1$ state (2$^+_1$$\langle E2\rangle$$|2^+_1\rangle$). The sensitivity to the second-order effect is determined by the obtained statistics, i.e., the number of subdivisions adopted.

Measuring the intensity of the incoming beam is difficult in a radioactive ion-beam experiment with a low beam energy, as the intensity is very low and can fluctuate. The beam can be contaminated as well. Another normalization method is thus needed. The Coulomb-excitation cross section of the projectile is normalized to the target-excitation cross section, which is calculated using the known matrix elements of the target. Table VI lists the matrix elements coupling the relevant states in the 104Pd and 94Mo targets used.

GOSIA2 is a special version of the GOSIA code that simultaneously minimizes the $\chi^2$ function for the projectile and target, thus resulting in a set of normalization constants and projectile matrix elements that best reproduce the experimental $\gamma$-ray yields. A drawback of the current version of GOSIA2 is that a proper correlated-error determination is not implemented. As only two parameters are determined, the correlated uncertainties are extracted by constructing a two-dimensional $\chi^2$ surface and projecting the 1σ contour of the total $\chi^2$ surface on the respective axis [44]. In this case the 1σ contour is defined as the points at which $\chi^2_{\text{min}} \leq \chi^2 \leq \chi^2_{\text{min}} + 1$.

I. 202Po

Coulomb excitation of 202Po was studied using two targets, 94Mo and 104Pd. Most of the statistics, especially on target excitation, were collected on the 104Pd target. The 4$^+_1 \rightarrow 2^+_1$ transition in 202Po was not observed above the level of 13% (5%) relative to the 202Po 2$^+_1 \rightarrow 0^+_1$ $\gamma$-ray in the 104Pd (94Mo) experiment. The higher upper limit for the 104Pd target is due to the overlap of the 4$^+_1 \rightarrow 2^+_1$ $\gamma$-ray energy with the target de-excitation transition energy. In both cases the assumption is made that only the 2$^+_1$ state is populated. Figure 12 shows the total $\chi^2$ surface constructed, in which $\chi^2$ is defined as

$$\chi^2 = \chi^2_{\text{Total,94Mo}} + \chi^2_{\text{Total,104Pd}},$$

where

$$\chi^2_{\text{Total}} = N_{P}^\text{data} \chi^2_{\text{P,GOSIA}} + N_{P}^\text{data} \chi^2_{\text{T,GOSIA}}.$$

Here, $N_{P}^\text{data}$ is the number of data points for the projectile [3(2) for the experiment on 104Pd (94Mo)] and $N_{P}^\text{data}$ represents the number of data points for the target [5(4) for 104Pd (94Mo)]. The number of data points for the target includes the known matrix elements provided to GOSIA (with their error bars). $\chi^2_{\text{P,GOSIA}}$ and $\chi^2_{\text{T,GOSIA}}$ are the reduced $\chi^2$ values given as output by the GOSIA code.

The correlated uncertainties of the transitional and diagonal matrix element can be deduced from the 1σ contour as shown in Fig. 12. The resulting matrix elements with their corresponding error bars are listed in Table VII. The value for the transitional matrix element, assuming that (2$^+_1$$\langle E2\rangle$$|2^+_1\rangle$) = 0 eb, i.e., with no influence of second-order effects, is also given. The error bar extracted in this way represents the quality of TABLE VI. Matrix elements coupling the relevant states in 104Pd and 94Mo. These matrix elements were used to determine the cross section for Coulomb excitation of the target.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Matrix element</th>
<th>Value (eb)</th>
<th>Ref. No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>104Pd</td>
<td>$0^+_1$$\langle E2\rangle$$</td>
<td>2^+_1\rangle$</td>
<td>0.73(2)</td>
</tr>
<tr>
<td></td>
<td>$0^+_1$$\langle E2\rangle$$</td>
<td>2^+_1\rangle$</td>
<td>0.134(7)</td>
</tr>
<tr>
<td></td>
<td>$2^+_2$$\langle E2\rangle$$</td>
<td>2^+_1\rangle$</td>
<td>-0.61(15)</td>
</tr>
<tr>
<td></td>
<td>$2^+_2$$\langle E2\rangle$$</td>
<td>4^+_1\rangle$</td>
<td>1.16(3)</td>
</tr>
<tr>
<td></td>
<td>$2^+_2$$\langle E2\rangle$$</td>
<td>0^+_2\rangle$</td>
<td>0.20(1)</td>
</tr>
<tr>
<td></td>
<td>$2^+_2$$\langle E2\rangle$$</td>
<td>2^+_1\rangle$</td>
<td>0.57(3)</td>
</tr>
<tr>
<td>94Mo</td>
<td>$0^+_1$$\langle E2\rangle$$</td>
<td>2^+_1\rangle$</td>
<td>0.451(4)</td>
</tr>
<tr>
<td></td>
<td>$2^+_2$$\langle E2\rangle$$</td>
<td>2^+_1\rangle$</td>
<td>0.17(11)</td>
</tr>
<tr>
<td></td>
<td>$2^+_2$$\langle E2\rangle$$</td>
<td>4^+_1\rangle$</td>
<td>0.78(6)</td>
</tr>
</tbody>
</table>

FIG. 12. (Color online) $\chi^2$ surface of the transitional and diagonal matrix element of the $2^+_1$ state in 202Po. The $\chi^2$ is the sum of the $\chi^2$ resulting from the experiment on 104Pd and the $\chi^2$ extracted from the 94Mo experiment. Projection of the 1σ contour (dashed lines) gives the correlated uncertainties of the two parameters extracted (see Table VII).
of the data in a simplified way and reflects the statistical error of the measured (projectile and target) $\gamma$-ray yields, the uncertainty of the $\gamma$-ray detection efficiency and of the beam and target purity, and the error bar on the matrix elements of the target.

2. 209Po

Coulomb excitation of 200Po was studied only with the 104Pd target. The $4^+_1 \rightarrow 2^+_1$ and $0^+_2 \rightarrow 2^+_1$ $\gamma$ rays were not observed above the level of 0.9% and 0.7% relative to the 200Po $2^+_1 \rightarrow 0^+_1$ $\gamma$ ray, respectively, so an exclusive population of the $2^+_1$ state was assumed. Figure 13 shows the resulting $\chi^2$ surface constructed applying the $\chi^2$ definition given in Eq. (4) with $N_{\text{data}}^p = 14$ and $N_{\text{data}}^t = 16$. A significantly higher sensitivity to the second-order effect of the diagonal matrix element results from the large statistics acquired, which allowed us to divide the data into 14 angular ranges. The resulting matrix elements with their corresponding error bars are listed in Table VII. An independent $\chi^2$-surface analysis, with 6 angular ranges instead of 14, yielded consistent results with a slightly larger error bar for the $\langle 2^+_1 | | E2 | 2^+_1 \rangle$ diagonal matrix element. Also, a value for the transitional matrix element, under the assumption that $\langle 2^+_1 | | E2 | 2^+_1 \rangle = 0$ eb, i.e., with no influence of second-order effects, is given.

B. Population of several low-lying excited states

In the case of multistep Coulomb excitation to states above the $2^+_1$ state, a combined approach between the standard version of the GOSIA code and GOSIA2 is implemented. The strategy combines the ability to simultaneously minimize the target and projectile $\chi^2$ in GOSIA2 and the correlated-error determination of GOSIA. The GOSIA2 $\chi^2$-surface analysis is provided as a first approximation in which the influence of higher-order excitations is not considered. The first-order solution for $\langle 0^+_1 | | E2 | 2^+_1 \rangle$ is used as absolute normalization in the second step, in which GOSIA is used to include couplings to higher-lying excited states. All populated states, observed $\gamma$-ray yields, and relevant spectroscopic data are included, and additionally, a number of “buffer” states are added to avoid artificial population buildup on top of the highest observed state. Including an E0 decay path in GOSIA has to be done in an indirect way by simulating the electron decay via a M1 transition [55]. Thus, nonexistent, additional $1^+_1$ states are included in the level scheme of the polonium isotope that take care of the E0 decay paths of the $0^+_2$ and $2^+_1$ states. As M1 excitation is orders of magnitudes weaker than E2 excitation, the $0^+_2$ and $2^+_1$ states are not populated via the $1^+_1$ states. In GOSIA the target de-excitation yields are used to determine relative normalization constants, which are related to the incoming beam intensity and the particle detection efficiency and link the different experimental subdivisions of the data to each other. The solution that results from the GOSIA $\chi^2$ minimization is then fed again to GOSIA2 to check the stability of the solution for $\langle 0^+_1 | | E2 | 2^+_1 \rangle$. In this step the couplings between states above the $2^+_1$ state are fixed and only $\langle 0^+_1 | | E2 | 2^+_1 \rangle$ and $\langle 2^+_1 | | E2 | 2^+_1 \rangle$ are free parameters of the GOSIA2 fit. Iterations between GOSIA and GOSIA2 are performed until a consistent solution is reached [44].

I. 198Po

The 94Mo target was used to study the Coulomb excitation of 198Po. Multistep Coulomb excitation up to the $4^+_1$, $2^+_2$, and $0^+_2$ states was observed. The first approximation with GOSIA2 yields a minimum at $\chi^2 = 3.9$ for $\langle 0^+_1 | | E2 | 2^+_1 \rangle = 1.14(12)$ eb and $\langle 2^+_1 | | E2 | 2^+_1 \rangle = 3.6(17)$ eb. The first-order solution for $\langle 0^+_1 | | E2 | 2^+_1 \rangle$ is then used as an additional data point in the GOSIA analysis, together with the known and relevant spectroscopic information on 198Po, which is listed in Table VIII. The E2/M1 mixing ratio determined in 202Po is assumed to stay constant for the neighboring polonium isotopes, which is an approximation. However, as the Coulomb-excitation data are insensitive to the M1 component of the mixed E2/M1 transitions, this does not influence the extracted matrix elements.
TABLE VIII. Spectroscopic information on $^{196,198}$Po included in the GOSIA analysis. The $E2/M1$ mixing ratio determined in $^{200}$Po is assumed to stay constant for the neighboring polonium isotopes. The $I_{2+}^1 → I_{2+}^2 /I_{2+}^0$ branching ratio is the $\gamma$-ray branching ratio and does not include $E0$ components.

<table>
<thead>
<tr>
<th>Observable</th>
<th>Value</th>
<th>Ref. No.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\tau_{2+}^1$</td>
<td>11.7(15) ps</td>
<td>[16]</td>
</tr>
<tr>
<td>$\tau_{2+}^2$</td>
<td>7.8(11) ps</td>
<td>[16]</td>
</tr>
<tr>
<td>$\tau_{2+}^0$</td>
<td>2.9(12) ps</td>
<td>[16]</td>
</tr>
<tr>
<td>$I_{2+}^2 → I_{2+}^1 /I_{2+}^0$</td>
<td>0.64(3)</td>
<td>[18]</td>
</tr>
<tr>
<td>$\delta(E2/M1)$</td>
<td>1.8(5)</td>
<td>[45]</td>
</tr>
</tbody>
</table>

Next to the populated states, the $6^+_1$ and $4^+_2$ states were included as buffer states. The $E0$-decay transitions of the $0^+_1$ and $2^+_2$ states were simulated via $M1$ transitions through two $1^+$ states included in the level scheme at 300 and at 700 keV. A $\chi^2$ minimization is performed resulting in four sets of matrix elements that reproduce the experimental data on a comparable level. The four solutions represent four different relative sign combinations for the matrix elements. Solutions 1 and 2, listed in Table IX, represent two solutions where $\langle 0^+_1 | E2 | 2^+_2 \rangle$ is positive. The solutions where $\langle 0^+_1 | E2 | 2^+_2 \rangle$ is negative (solutions 3 and 4) are not listed in Table IX, as they are not considered to be physical solutions. The relative signs of the matrix elements affect the Coulomb-excitation cross section in an important way. Every possible excitation path contributes to the cross section for multistep Coulomb excitation to a certain excited state. As the excitation amplitude for a given path is proportional to the product of the matrix elements involved, the relative signs of these matrix elements play a crucial role. The signs of the products of matrix elements were varied by carefully adopting various initial values, and all possible sign combinations were investigated [33,44].

In solutions 3 and 4 (not listed in Table IX), the population of the $2^+_2$ state is significantly lower than in solutions 1 and 2. In order to reproduce the experimental yields, the diagonal matrix element of the $2^+_2$ state has to be increased in these solutions to unphysically large values of $>4$ eb, far beyond the rotational limit. Because of these large values for the diagonal matrix element, solutions 3 and 4 are disregarded. The sign of the loop $(2^+_1 | E2 | 0^+_2) \cdot (0^+_2 | E2 | 2^+_2) \cdot (2^+_2 | E2 | 2^+_1)$ is the only difference between solution 1 and solution 2, which are listed in Table IX (positive for solution 1, negative for solution 2). Changing the sign of this loop does not change the population of any of the excited states significantly. However, the matrix element $(0^+_1 | E2 | 2^+_2)$ reaches the lower limit 0 in solution 2, hinting at the fact that a better solution would be obtained with a negative sign for this matrix element. When the sign of the matrix element between the $0^+_1$ and the $2^+_2$ state is changed, the first solution is reproduced exactly in magnitude, but with a negative value for $(2^+_1 | E2 | 0^+_2)$ and $(0^+_2 | E2 | 2^+_2)$. However, the positive sign for the $(2^+_1 | E2 | 0^+_2) \cdot (0^+_2 | E2 | 2^+_2) \cdot (2^+_2 | E2 | 2^+_1)$ loop is not changed. This is an argument for putting solution 1 forward as the correct sign combination.

The matrix elements of solution 1 are fixed in a new $\chi^2$ analysis in GOSIA2 where only $(0^+_1 | E2 | 2^+_2)$ and $(2^+_2 | E2 | 2^+_1)$ are allowed to vary. The resulting $1\sigma$ contour is shown in Fig. 14, yielding a result for both matrix elements which is consistent with the GOSIA minimum $(0^+_1 | E2 | 2^+_2) = 1.14(14)$ eb, $(2^+_2 | E2 | 2^+_1) = 2.41(21)$ eb.

2. $^{196}$Po

The Coulomb excitation of the lightest polonium isotope studied in this work, $^{196}$Po, was examined on a $^{104}$Pd target. The Coulomb excitation of $^{198}$Po on $^{90}$Mo shows the $1\sigma$ contour constructed by letting $\chi^2$ increase to $\chi^2_{\min} + 1 = 4.9$. Projection of the $1\sigma$ contour gives the correlated uncertainties of the two parameters that are extracted.

FIG. 14. (Color online) Result of the $\chi^2$-surface analysis for the Coulomb excitation of $^{198}$Po on $^{90}$Mo showing the $1\sigma$ contour constructed by letting $\chi^2$ increase to $\chi^2_{\min} + 1 = 4.9$. Projection of the $1\sigma$ contour gives the correlated uncertainties of the two parameters that are extracted.
target. Multistep Coulomb excitation to the \(4^+_1\), \(2^+_2\), and \(0^+_3\) states was observed. The relevant spectroscopic information on \(^{196}\text{Po}\) included in the GOSIA analysis is listed in Table VIII. The first approximation with GOSIA2, with all the relevant spectroscopic information included, yields a minimum at \(x_{\text{min}}^2 = 13.1\) for \((0^+_1||E2||2^+_1) = 1.36(5)\) eb and \((2^+_1||E2||2^+_1) = 0.1(5)\) eb. The first-order solution for \((0^+_1||E2||2^+_1)\) is used as an additional data point in the GOSIA analysis, together with the relevant spectroscopic information, which is listed in Table VIII. The \(E0\) transitions of the \(0^+_1\) and \(2^+_1\) states were simulated via \(M1\) transitions through two \(1^+\) states included in the level scheme at 300 and 650 keV. A \(\chi^2\) minimization, checking also the sensitivity of the signs of the loops of matrix elements, leads to two sets of matrix elements that reproduce the experimental data on a comparable level (see Table IX). A lack of experimental information on the coupling between the \(0^+_1\) state and the \(2^+_1\) and \(2^+_2\) states renders it impossible to extract information on the sign and magnitude of \((2^+_1||E2||0^+_1)\) and \((0^+_1||E2||2^+_2)\). However, to make sure that the correlations to these couplings are taken into account, the matrix elements were included in the GOSIA analysis, as well as the buffer states \(6^+_1\) and \(4^+_2\). It is clear from Table IX that the sign of the loop \((0^+_1||E2||2^+_1)\cdot(2^+_1||E2||2^+_2)\cdot(2^+_2||E2||0^+_1)\) influences only the value of the diagonal matrix element of the \(2^+_1\) state significantly. There is no model-independent way to distinguish between these two solutions with the present set of data.

### V. DISCUSSION

Mixing between coexisting structures has a large influence on the matrix elements and depends strongly on the proximity of energy levels of the same spin. Figure 15 shows systematically both the level energies and the transitional quadrupole moments \(|Q_i|\) for \(^{196–202}\text{Po}\).

The experimentally obtained results are compared using three theoretical approaches: the BMF method, the generalized Bohr Hamiltonian (GBH), and the IBM. The first two methods are based on the introduction of a mean field determined by the HFB method and the same SLy4 energy density functional. In the BMF method, the mean-field wave functions are first projected on the angular momentum and particle number and then mixed with respect to the axial quadrupole moment. Spectra and transition probabilities are calculated in the laboratory frame of reference and compared directly to the experimental data [19]. In the GBH method, the mass parameters of a Bohr Hamiltonian are derived thanks to a cranking approximation to the adiabatic time-dependent Hartree-Fock method and are rescaled to take into account the fact that time-odd contributions to the mass parameters are neglected. One of the benefits of this method is that it leads to calculations much less heavy than the BMF method and permits the treatment of triaxial quadrupole deformations [59,60]. Note that in both methods, the only parameters are those of the energy density functional and no specific adjustments are performed in their applications to the neutron-deficient isotopes around lead. The IBM is a very convenient method to put into evidence the group properties of nuclear spectra and to classify them using group theoretical methods.

However, it contains eight parameters per isotope in the form used here, which are adjusted for each isotope thanks to known experimental data. The measured energies for the yrast band up to \(I = 8^+\) states \(0^+_1, 2^+_1, 2^+_2, 2^+_3, 3^+_4, 4^+_4, 5^+_5, 6^+_6\), and \(7^+_7\), and the measured \(B(E2)\) values between the above states are used to fix the parameters of the Hamiltonian through a least-squares fit. The purpose of the IBM is therefore to analyze data but it is less suited to perform predictions for unknown nuclei [4].

The BMF approach overestimates the level energies in the four polonium isotopes studied here, as noted by Yao et al. [19]. The level energies in the neighboring mercury, lead, and radon isotopes are also too widely spaced in the BMF calculations. The results obtained using the GBH approach are significantly better, pointing out the importance of triaxial quadrupole deformations, although the renormalization of the GBH mass parameters does not allow a firm conclusion. The transition probabilities between the ground state and the \(2^+_1\) state are reproduced quite well for \(^{200,202}\text{Po}\), suggesting the correct description of the underlying structures. For mass \(A < 200\), these transition probabilities are underestimated. Further, significant differences can be noted in the transition probabilities related to the \(0^+_1\) versus \(2^+_2\) state resulting from the three theoretical descriptions. The triaxial quadrupole degree of freedom included in the GBH approach does not significantly affect the transition probabilities.

Figure 16 shows a comparison of extracted deformation parameters obtained from the measured charge radii \(\langle r^2 \rangle\), on the one hand, and from the sum of squared matrix elements \(\sum_i |(0^+_1||E2||2^+_1)|^2\), on the other. As the parameters extracted from these two approaches are not identical, separate notation is used. A deformation parameter, called \(\tilde{\beta}_{20}\), was estimated from the charge radii using the expression

\[
\langle r^2 \rangle_A \approx \langle r^2 \rangle_{A}^{\text{mean}} \left( 1 + \frac{5}{4\pi} \tilde{\beta}_{20}^2 \right),
\]

where \(\langle r^2 \rangle_{A}^{\text{mean}}\) is the mean-square charge radius of a spherical nucleus with the same volume, which was evaluated with the droplet model with a revised parametrization [61]. From the extracted \(E2\) matrix elements, a deformation parameter, \(\beta_2\), can be deduced, through the quadrupole invariant \(Q^2\), using the expression

\[
\sum_i |(0^+_1||E2||2^+_1)|^2 = \left( \frac{3}{4\pi} Z e R_0^2 \right) \beta_2^2,
\]

where a uniform charge distribution is assumed [62]. The sum of squared matrix elements \(\sum_i |(0^+_1||E2||2^+_1)|^2\) was evaluated over the \(2^+_1\) states populated for each case, i.e., only the \(2^+_1\) state in \(^{200,202}\text{Po}\) and the \(2^+_1\) and \(2^+_2\) states in \(^{196,198}\text{Po}\). In \(^{194}\text{Po}\), only the \(B(E2)\) value of the \(2^+_1\) state is known from the lifetime measurement [15]. The onset of deviation from sphericity around \(N = 112\) \((A = 196)\), observed in the laser spectroscopy studies (see also Fig. 1), is confirmed by the measured transition probabilities. An overall good agreement between the deformation parameter extracted from the charge radii and the squared matrix elements is observed within the error bars.
FIG. 15. Experimental levels of the low-lying structures in $^{196,198,200,202}$Po. Level energies (in keV) are taken from Nuclear Data Sheets. Transitional $|Q_t|$ values (in eb) are based on the experimentally determined matrix elements. The width of the arrows represents the relative size of the transitional quadrupole moments $|Q_t|$. Experimental level energies and $|Q_t|$ values are compared to the same information, predicted by the BMF [19], IBM [58], and GBH [59] models.
FIG. 16. (Color online) Deformation parameters of the ground state, extracted from the charge radii $\delta(r^2)$ (triangles) [61] and sum of squared matrix elements according to Eq. (6) (squares). For odd isotopes the deformation parameter for the $3/2^-$ ground state is shown. The data point for $^{194}$Po is deduced from the lifetime measurement [15].

The experimentally determined transitional quadrupole moments $|Q_t|$ connecting the $2^+_1$ and $2^+_2$ states to the ground state are displayed and compared to the predictions from the BMF, IBM, and GBH calculations in Fig. 17. The same trend of increasing deformation with decreasing mass is observed from the experimental $|Q_t|$ values. The BMF $|Q_t(2^+_1 \rightarrow 0^+_1)|$ values start to deviate from the experimental values at mass $A = 198$ as noted earlier. The inclusion of the new data points deduced in this work shows that the three theoretical approaches reproduce the experimental values quite well.

The reproduction of the experimental $|Q_t(2^+_1 \rightarrow 0^+_1)|$ values by the IBM follows directly from the fit performed to the measured $B(E2)$ values to fix the parameters of the IBM Hamiltonian. Nevertheless, the experimental trend in $|Q_t(2^+_2 \rightarrow 0^+_1)|$ is predicted well by the IBM, as well as the GBH and BMF models. The GBH model slightly overestimates the collectivity in the $2^+_1 \rightarrow 0^+_1$ transition for $^{196-200}$Po.

The deformation of the $2^+_1$ state can be understood in a Coulomb-excitation experiment through the measurement of the spectroscopic quadrupole moment $Q_s$. The observed trend of increasing deformation in the $2^+_1$ state when going down in mass number, shown in Fig. 18, is predicted by the three model descriptions.

A phenomenological two-state mixing model was used to calculate the $E2$ matrix elements between low-lying states in the neutron-deficient $^{192-198}$Hg isotopes [3,26] to test the assumption that the excited states in the mercury isotopes can be described by a spin-independent interaction between two rotational structures. A common set of matrix elements within the unperturbed bands (transitional as well as diagonal $E2$ matrix elements) for the four studied mercury isotopes was found to reproduce most of the experimental results. A similar approach has been used for the polonium isotopes studied in this work. However, in the polonium isotopes a rotational structure was assumed to mix with a more spherical structure. A fit with the variable moment of inertia model [63] of the yrast $4^+, 6^+, 8^+$, and $12^+$ levels in $^{196}$Po was used to determine the unperturbed energies of the $0^+$ and $2^+$ rotational states. The $10^+$ state was not included because of an ambiguity. In this procedure, no mixing was assumed for states with spin $I \geq 4$. Using the unperturbed $0^+$ and $2^+$ rotational energies from the variable moment of inertia fit, information on the size of the spin-independent mixing matrix element was extracted [33]. The mixing amplitudes, listed in Table X, were determined by combining the spin-independent mixing matrix element $V = 200$ keV with the mixed experimental level energies.

The experimental $E2$ matrix elements can then be expressed in terms of pure intraband matrix elements and a set of mixing amplitudes. No interband transitions between
TABLE X. Square of wave-function mixing amplitudes of the “normal” (vibrational) configuration, at spin 0+ ($\sigma^0_{E2}$) and spin 2+ ($\sigma^2_{E2}$). Details on the method applied to extract these values are provided in [33].

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$\sigma^0_{E2}$</th>
<th>$\sigma^2_{E2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{194}$Po</td>
<td>12%</td>
<td>29%</td>
</tr>
<tr>
<td>$^{196}$Po</td>
<td>85%</td>
<td>50%</td>
</tr>
<tr>
<td>$^{198}$Po</td>
<td>94%</td>
<td>69%</td>
</tr>
<tr>
<td>$^{200}$Po</td>
<td>97%</td>
<td>92%</td>
</tr>
<tr>
<td>$^{202}$Po</td>
<td>99%</td>
<td>88%</td>
</tr>
</tbody>
</table>

The unperturbed structures were allowed. A set of unperturbed matrix elements was fitted to optimally reproduce the experimental results, yielding $\langle 0_{1}^+ | E2 | 2_{1}^+ \rangle = 1.1$ eb, $\langle 0_{1}^+ | E2 | 2_{0}^+ \rangle = 1.5$ eb, $\langle 2_{1}^+ | E2 | 2_{0}^+ \rangle = -0.4$ eb, and $\langle 2_{0}^+ | E2 | 2_{0}^+ \rangle = 1.8$ eb. Here, $I$ represents the spherical structure, and $II$ the deformed one. In the fitting procedure the unperturbed diagonal matrix elements were not allowed to cross the rotational limit compared to the intraband transitional matrix element ($\langle |E2|2_{0}^+| \rangle < 1.19 \times (0^+_+||E2||2^+_+)$ [64]).

A comparison of the measured and calculated values of the $E2$ matrix elements is shown in Fig. 19. The best fit was found with solution 2 in $^{196}$Po (see Table IX), where the diagonal matrix element is positive and $\langle 0_{1}^+ | E2 | 2_{1}^+ \rangle$ is negative. Most of the experimental results are reproduced within the 1σ uncertainty. The total $\chi^2$ for this fit is equal to 102, while the total $\chi^2$ for the best fit to solution 1 is equal to 189. The extracted unperturbed $E2$ matrix elements describing the rotational structure in the polonium isotopes are comparable to those extracted in the two-state mixing approach in the mercury isotopes for the weakly deformed oblate structure, where the extracted unperturbed transitional and diagonal $E2$ matrix elements of the weakly deformed structure are 1.2 and 1.8 eb, respectively [3]. This supports the interpretation that a weakly deformed, oblate structure is intruding in the low-lying energy levels of the neutron-deficient polonium isotopes. The characteristics of this weakly deformed oblate structure seem to be related to those of the oblate structure in the mercury isotopes, which mirror the polonium isotopes with respect to $Z = 82$.

VI. SUMMARY AND CONCLUSIONS

A set of matrix elements coupling the low-lying states in the even-even neutron-deficient $^{196–202}$Po isotopes was extracted in two Coulomb-excitation campaigns, which were performed at the REX-ISOLDE facility at CERN. In the two heaviest isotopes, $^{200,202}$Po, the transitional and diagonal matrix elements of the $2_{1}^+$ state were determined. In $^{196,198}$Po multistep Coulomb excitation was observed to populate the $4_{1}^+$, $0_{2}^+$, and $2_{2}^+$ states. The relatively large uncertainty of the matrix elements related to the $0_{2}^+$ and $2_{2}^+$ states is due to the indirect observation of the $E0$ transitions between the $0_{2}^+$ and $0_{1}^+$ states and the $2_{2}^+$ and $2_{1}^+$ states through characteristic polonium x rays. For future experiments the electron spectrometer SPEDE will provide a direct way of detecting $E0$ transitions [65].

The experimental results were compared to the results from the measurement of mean-square charge radii in the polonium isotopes, confirming the onset of deformation from $^{196}$Po onwards. Three model descriptions were used to compare to the data. Calculations with the BMF model, the IBM, and the GBH model show partial agreement with the experimental data. The comparison between the BMF model and the GBH results does not permit a firm conclusion regarding the effect of triaxial quadrupole deformations. Finally, calculations with a phenomenological two-level mixing model hint at the spin-independent mixing of a more spherical structure with a weakly deformed oblate structure. Overall the comparison to theory would benefit from an increase in the experimental sensitivity. This increased sensitivity could be reached in Coulomb-excitation experiments with higher beam energies at HIE-ISOLDE [66,67].

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