## Anisotropic Alpha Emission from On-Line–Separated Isotopes

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A systematic on-line nuclear-orientation study of heavy isotopes by use of anisotropic  $\alpha$  emission is reported for the first time. The anisotropies recorded for <sup>199</sup>At, <sup>201</sup>At, and <sup>203</sup>At are remarkably pronounced and strongly varying. At lower neutron number the  $\alpha$  particles are more preferentially emitted perpendicularly to the nuclear-spin direction. This may be interpreted in terms of the high sensitivity of the  $\alpha$ -emission probability to changes in the nuclear shape.

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Over the last few years, it has been pointed out on various occasions that  $\alpha$ -particle angular distributions display a remarkable behavior.<sup>1-3</sup> This is usually interpreted as due to the strong dependence of  $\alpha$  emission on the penetrability of the Coulomb barrier and hence on the shape of the nucleus. No attempt has been made yet to exploit this phenomenon systematically for a study of relative shape changes in a series of isotopes. We report for the first time a study of this type by use of  $\alpha$  emission. Results on short-lived At and Po isotopes have been obtained by use of a combination of production by heavy-ion reactions, on-line separation, and on-line implantation, and orientation at low temperatures.<sup>4-8</sup>

The experiments are performed at temperatures in the millikelvin region. Since  $\alpha$  detection precludes the use of any thermal shield between source and detector, a first prerequisite for a successfuly study is the development of reliable  $\alpha$  detectors operating at very low temperatures (4 K). Previously, work on  $\alpha$  emission from some long-lived oriented nuclei has been reported.<sup>9-11</sup> At that time the source preparation as well as the quality of the detectors did not allow the resolved detection of various  $\alpha$  branches; also, the reproducibility of the data could not be guaranteed. No far-reaching conclusions could be drawn in those early studies, also in view of the limited range of potential candidates. Now the number of available  $\alpha$ emitters-if one works on line-has increased tremendously. Moreover, the implantation technique is another advantageous aspect for particle detection at our on-line nuclear-orientation setup.

The strong advances over the last decade in highpurity (HP) semiconductor materials have been used for systematic tests of particle detectors developed in cooperation with commercial suppliers. These tests confirm that the high purity of the starting material is very important for detector operation at 4 K. Good results were obtained with 2- and 3-mm planar HP Ge detectors and some types of Si surface barriers.

The technical work in overcoming the various cryogenic problems (among others, the construction of leak-tight feedthroughs at 4 K to have a short connection between detector and field-effect transistor) will be reported in a separate paper.<sup>12</sup> We are now able to detect particles at 4 K with an energy resolution and stability at least comparable to the ones achieved in "normal" operating conditions.

The first experiments with this technique were carried out on Po and At isotopes. These nuclei were produced at LISOL<sup>13</sup> in the fusion reaction of a 120-180-MeV <sup>20</sup>Ne beam (from the CYCLONE cyclotron) on a combined Re-Ir target in a forced electronbombardment-induced arc discharge ion source. We separated the masses 199 to 204 with <sup>199</sup>mPo ( $t_{1/2} = 4.2$  min), <sup>201</sup>mPo ( $t_{1/2} = 8.9$  min), <sup>203</sup>mPo ( $t_{1/2} = 1.2$  min), <sup>199</sup>At ( $t_{1/2} = 7.0$  s), <sup>201</sup>At ( $t_{1/2} = 1.5$  min), <sup>203</sup>At ( $t_{1/2} = 7.4$  min), and the odd-odd nuclei <sup>200,202,204</sup>At as most important activities. By continuous lowtemperature implantation (down to 11 mK) in a magnetized iron host foil, the nuclei were oriented. The isotopes decay by  $\beta^+$  emission and electron capture as well as by  $\alpha$  emission and, besides the  $\alpha$  and conversion-electron anisotropies,  $\gamma$ -anisotropy data have been accumulated, too. Nuclear information on At, Po, Bi, and some Pb nuclei is available from masses 195 to 204. Here we concentrate on the results obtained through the detection of  $\alpha$  particles.

In our specific experimental arrangement the influence of the external magnetic field on the emitted  $\alpha$ particles can be neglected and scattering effects are small (for a complete discussion we refer to Ref. 12). An accumulation of some  $\alpha$  spectra taken at different masses is shown in Fig. 1. Anisotropy data of some  $\alpha$ decays are presented in Figs. 2 and 3. Temperatures were recorded with  ${}^{57}\text{Co}Fe$  as a thermometer. It should also be remarked that at present  ${}^{199}\text{At}$  is the shortest-lived isotope ever oriented on line after direct implantation.

For a description of the angular distribution of the emitted radiation from oriented nuclei we refer to the work of Krane.<sup>14</sup> The radiation parameters in the  $\alpha$ -particle distribution formula contain the new nuclear information and can be written as

$$A_{k} = \frac{\sum_{L,L'} a_{L} a_{L'} \cos(\sigma_{L} - \sigma_{L'}) F_{k}^{\alpha}(L,L',I_{f},I_{i})}{\sum_{L} a_{L}^{2}}.$$



FIG. 1. An accumulation of  $\alpha$  spectra taken at different masses with particle detectors at liquid-helium temperature.

Here  $F_k^{\alpha}$  are Ferentz coefficients adjusted for  $\alpha$  decay, and  $a_L$  is the amplitude and  $\sigma_L$  the phase of the  $\alpha$ wave with angular momentum L. It has been pointed out before in several calculations<sup>11, 15-17</sup> that the phase shifts are small and hence  $\cos(\sigma_L - \sigma_{L'}) = \pm 1$  is a good approximation. Because of parity conservation only even-k terms are nonzero and only angular momenta  $L = 0, 2, \ldots$  contribute because the  $\alpha$  decays of the odd nuclei in this work are all favored transitions. A first analysis of the  $\alpha$ -particle distributions reveals the admixtures of  $L \neq 0$  in these decays to be very small, although they are responsible for the observed anisotropies. Therefore, we will consider only s and d waves: The hindrances increase with angular momentum. The foregoing enables us to express the radiation parameters as  $A_2 \approx 2\delta$  and  $A_4 \approx 0$ , where  $\delta = a_2/a_0$  with the phase factor incorporated in the parameter  $\delta$ . The error implied by this approximation is smaller than 3% for the  $\alpha$  decays under study. Thus the temperature-independent factor  $f\delta$  and the interaction  $\mu B$  are the only parameters left in these  $\alpha$ particle angular distribution functions, where f stands for the fraction of nuclei which experience the full hyperfine field.

To illustrate this the  $\alpha$  anisotropy of <sup>199</sup>mPo and  $\chi^2$  curve are shown in Fig. 2. The  $\nu \frac{13}{2}$  + isomer <sup>199</sup>mPo decays by a 6059-keV  $\alpha$  transition to the similar state in <sup>195</sup>Pb and the positive anisotropy in  $W(0^{\circ})/W(90^{\circ})$  implies preferential  $\alpha$  emission along the nuclear spin vector. The isotopes <sup>201</sup>mPo and <sup>203</sup>mPo have small  $\alpha$  branchings and results are much more difficult to extract. The change in anisotropies of the  $\alpha$  decays of <sup>199,201,203</sup>At is well pronounced, as can be seen in Fig. 3. As one goes further away from the N = 126 shell closure, the intensity of  $\alpha$  particles emitted perpendicularly to the nuclear spin direction increases strongly.



FIG. 2. The  $\alpha$  anisotropy of the 6059-keV transition of <sup>199</sup>mPo. Inset: The  $\chi^2$  surface as a function of the parameters  $\mu B$  and  $fA_2$  in the angular distribution, indicating that both can be extracted fairly accurately.

If we consider the formalism this means that the  $\delta$  value of these  $\alpha$  decays varies. For each of these three At nuclei, the following is found.

(i) We have similar  $\pi h_{9/2}$  to  $\pi h_{9/2} \alpha$  decays from <sup>A</sup>At to <sup>A-4</sup>Bi. The possibility of a ground-state spin of  $\frac{7}{2}$  in the neutron-deficient odd At nuclei has been suggested<sup>18</sup> but our  $\alpha$  anisotropy does not agree in sign and magnitude with a  $\frac{7}{2}$  to  $\frac{9}{2}$  transition.

(ii) The anisotropy curves (Fig. 3) show saturation at temperatures below 30 mK and in such cases the temperature-independent parameter can be determined very accurately. The  $f\delta$  values are tabulated in Table I. The large hyperfine field of At in Fe is responsible for the saturation effect.

(iii) The implantation characteristics are the same (no difference in f). These isotopes were directly produced and measured relatively under identical conditions. A deviation of the fraction f from 100% would



FIG. 3. Anisotropies as a function of 1/T of the  $\alpha$  emission in the decay of <sup>199,201,203</sup>At.

TABLE I. The  $f\delta$  values of the  $\alpha$  decays of odd At and Po nuclei. For <sup>199</sup>At we obtained a lower limit of  $|f\delta|$  from our fits because relaxation can become important at this short lifetime.

Isotope	t <sub>1/2</sub>	$E_{\alpha}$ (keV)	Transition	fδ
<sup>199</sup> At	7.0 s	6643	$\pi h_{9/12} \rightarrow \pi h_{9/12}$	-0.059(2)
<sup>201</sup> At	1.5 min	6344	<i>y</i> <sub>1</sub> <i>L y</i> <sub>1</sub> <i>L</i>	-0.023(1)
<sup>203</sup> At	7.4 min	6088		0.004(2)
<sup>199</sup> <sup>m</sup> Po	4.2 min	6059	$\nu i_{13/2}^{+} \rightarrow \nu i_{13/2}^{+}$	0.131(5)

even increase the three  $\delta$  values proportionally.

Hence only the variation of the mixing ratio  $\delta$  of the  $\alpha$  particles with L = 2 and L = 0 remains as main cause for the change in anisotropy when we go from <sup>203</sup>At to <sup>199</sup>At. In a qualitative way one can understand that a change in  $\delta = a_2/a_0$  might be correlated to an enhanced deformation. The picture wherein the penetration through the Coulomb barrier is considered has been described before in a simplified manner.<sup>19</sup> For instance, the larger the  $P_2$  deformation, the more  $L = 2 \alpha$  particles will be emitted over the isotropic L = 0 waves. This can also be seen if one considers for the L = 2 particles the tunneling through the additional centrifugal barrier: It is also dependent on the deformation.

Specific calculations of the  $\alpha$ -emission distribution in relation to deformation are being performed<sup>20</sup> and preliminary results point to nonunique solutions. More experimental information is certainly needed to solve this. Apart from the need to explain the observed  $\alpha$ -mixing ratios more quantitatively, it remains a problem why the  $\alpha$  anisotropy is apparently changing sign at <sup>203</sup>At. This may be an indication for a more elaborate approach where, besides the  $\alpha$  tunneling, also the (nonuniform) probability of  $\alpha$ -particle formation is taken into account, which is related to the observed  $\alpha$ -decay widths. We also want to stress that the deformation enters in the  $\alpha$  distributions by means of the  $\delta$ -mixing ratios (in general, every interference of angular momenta L' and L).

We calculated total potential energy (TPE) surfaces for <sup>195-205</sup>At and some Po nuclei with the method as described by Heyde *et al.*<sup>21</sup> The calculated TPE surfaces are rather flat with minima at the oblate as well as at the prolate side. However, if one adopts the idea that the nucleus has the shape corresponding to the deformation of the lowest minimum, then oblate shapes can be preferred for the three At isotopes under study (similar results have been found elsewhere<sup>22</sup>). Furthermore, the deformation increases as one goes further away from the shell closure at N = 126, though its variation is rather small ( $\epsilon_2$  varies from -0.05 to -0.09 as one goes from <sup>203</sup>At to <sup>199</sup>At).

As the TPE surfaces of the Po isotopes are rather

similar to those of Hg at the same neutron numbers, we expect the  $\nu \frac{13}{2}$  + states to be also slightly oblate. There is a small difference between Hg and Po because the proton shell corrections, which are larger for Hg than for Po, induce more pronounced oblate shapes for Hg than for Po.<sup>23,24</sup>

All these calculations agree with the sign and the variation of the  $\alpha$  anisotropies as observed in our measurements; e.g., the positive  $\alpha$  anisotropy of <sup>199</sup>mPo agrees with a small projection value  $\Omega$  of the unpaired neutron angular momentum and an oblate shape of the core.<sup>19</sup> These first data show that information on the shape of the nucleus can be obtained from anisotropic  $\alpha$  emission and at the same time the magnetic hyperfine interaction may be extracted as well. The relation of this information on shape changes to that obtained from other observables (quadrupole moments,  $\delta \langle r^2 \rangle$  values is not yet straightforward but allows interesting tests of various theoretical models.

In summary, we have measured  $\alpha$ -emission anisotropies of short-lived Po and At isotopes.  $\alpha$  anisotropies have been measured with separated beams of  $10^3$ ions/s which clearly shows the sensitivity of the technique and the feasibility in connection with on-line systems. This experiment provides the first systematic study of anisotropic  $\alpha$ -particle decay. Qualitatively, the observed increase in  $\alpha$  anisotropy of the  $1^{99, 201, 203}$ At nuclei can be interpreted as an indication of deformation change in going away from the neutron shell closure, although these nuclei lie in a quasispherical region. A comparison with calculations points to the fact that quite large variations in  $\alpha$  anisotropy correspond to small deformation changes.

More generally, the technical realization of particle detection of on-line-oriented isotopes opens new avenues for nuclear structure studies. Besides  $\alpha$ -decay studies of nuclear deformations, also the angular distribution of  $\beta$  decay, conversion electrons, and fragments of spontaneously fissioning nuclei can be investigated now in isotopic series down to very short life-times.

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