

α particle angular distributions of $^{189,191,193}\text{Bi}$

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Angular distribution data for α particles emitted in the favored decay of on-line oriented neutron deficient isotopes $^{189,191,193}\text{Bi}$ near midshell ($N=104$) are presented. They give additional support for the recent finding that anisotropic α emission in favored decays from near-spherical nuclei is mainly determined by nuclear structure effects. [S0556-2821(98)03611-5]

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I. INTRODUCTION

The mechanisms which determine the α decay observables are still poorly understood despite much experimental and theoretical work on the subject. Indeed, the various theories developed to describe the total α decay rate and the angular distribution of α particles emitted by oriented nuclei make different predictions, but the experimental data available up to the mid-nineties did not allow a clear choice between these models. In an attempt to clarify this situation we have recently studied anisotropic α emission in favored decays of the near-spherical odd nuclei $^{199-211,217}\text{At}$ and $^{205,207,209}\text{Rn}$ [1]. Surprisingly large α anisotropies were found for these nuclei with the largest being observed for ^{211}At situated at the $N=126$ shell closure. It was shown that the experimental anisotropies cannot be reproduced in a satisfactory way by the extreme cluster model of Berggren [2-4] nor by the model dominated by deformed Coulomb barrier penetration as was recently developed by Delion *et al.* [5,6]. In the latter, the α particle formation amplitude at the nuclear surface is calculated in a large shell-model base with nucleon-nucleon pairing. The proton-neutron interaction is not explicitly considered but "effectively" taken into account by fitting the pairing gap for each isotope. The tunneling process of the α through the Coulomb barrier is treated semiclassically. For the At and Rn nuclei studied [1] the angular distribution coefficients calculated as a function of the deformation parameter β_2 are almost identical for each isotope [6]. Thus, in the model by Delion *et al.* the observed variation in anisotropy with changing neutron number can only be explained by a change in deformation. This, however, would lead to the unacceptable conclusion that for the light Rn and At nuclei nuclear deformation would *increase* when going *towards* the neutron shell closure, with for At a maximum being found at $N=126$. As a result it must be concluded that the anisotropy in the α emission of the nearly spherical At and Rn nuclei is not dominated by deformation

but rather by the nuclear structure of the decaying nucleus.

Our interpretation of the At and Rn α anisotropy data presented in [1] is based on spherical shell model calculations [7,8] using the formalism of Mang and Rasmussen [9] and taking into account BCS pairing [10]. The α particle formation amplitude at the nuclear surface was obtained from shell model wave functions near ^{208}Pb . In computing the tunneling probability, the quadrupole part of the Coulomb barrier is neglected as only nuclei with very small (or zero) deformations are treated. This implies that in this approach, the tunneling of the α particle through the Coulomb barrier does not cause any anisotropy in its angular distribution although the centrifugal barrier does damp the partial α waves with higher angular momentum L . It was found [1] that for odd- Z nuclei (e.g., At and Bi), besides the inclusion of BCS pairing, it is necessary to explicitly consider the p - n interaction between the valence neutron holes/particles and protons to explain the observed data. For odd Z nuclei near $N=126$ the major part of the $L=2$ partial α wave arises from the protons transferred to the α particle, which give a positive contribution to the anisotropy. Away from the $N=126$ closed shell, the quadrupole part of the p - n interaction polarizes the core, thus producing a mixed ground state containing 2^+ neutron components. These neutron excitations give a negative contribution to the $L=2$ amplitude in the α particle wave function. The magnitude of this contribution increases with increasing number of neutron holes (particles) below (above) the $N=126$ shell closure as the total proton-neutron interaction becomes stronger, thus causing a change in anisotropy. The qualitative trend of the observed α anisotropies for odd At isotopes agrees well with this model, i.e., a large positive anisotropy at the $N=126$ shell closure which decreases continuously with more neutrons being removed from the shell. Furthermore, a pure shell model calculation for the $N=126$ closed shell isotope ^{211}At turned out to be in excellent agreement with the experimental result [1]. A minimum in the anisotropy is expected at mid-shell ($N=104$) because the p - n quadrupole interaction is maximum at this point. In order to check the validity of these arguments, we have carried out anisotropy measurements for the

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avored $9/2^- \rightarrow 9/2^-$ α transitions of ^{189}Bi ($T_{1/2}=0.68(3)$ s [11]), ^{191}Bi ($T_{1/2}=12(1)$ s [11]) and ^{193}Bi ($T_{1/2}=67(3)$ s [11]) with $N=106$, 108 and 110, respectively. These nuclei are situated rather close to the proton drip line as ^{185m}Bi ($T_{1/2}=44(16)$ ms [12]) is a pure proton emitter.

II. EXPERIMENT AND ANALYSIS

The bismuth nuclei were produced with a 1 GeV proton beam on a ThC₂ target at the ISOLDE mass separator at CERN [13] and ionized in a plasma-discharge ion source. After mass separation they were oriented by implanting them (at 60 keV) at low temperature (down to 12.5 mK) into a magnetized, high-purity iron foil soldered onto the cold finger of the ^3He - ^4He dilution refrigerator of the NICOLE on-line nuclear orientation set-up [14].

The α spectra were measured with three Si P-I-N diodes mounted inside the 4 K radiation shield of the refrigerator, at angles $\theta=14^\circ$, 51° and 90° with respect to the orientation axis, and operated at a temperature of about 4 K. The energy resolution for 6 MeV α particles was 32 keV for the 14° detector and 20 keV for the other two. The resolution is affected by scattering of the α particles in the sample foil and is poorer for the first detector because of the rather small angle between the plane of the foil and the detector axis. The 90° detector was mounted off the vertical axis and the foil tilted 20° to the horizontal magnetic field axis. Conventional Ge detectors recorded the γ spectra at 0° , 90° and 180° with respect to the orientation axis. All measurements were carried out in a magnetic field of 0.2 T after the iron foil had been initially magnetized in 0.5 T.

For each α transition, the angular distribution function $W(\theta)$ was calculated from the ratio of the intensities $N(\theta)$ at low temperatures (i.e., $T < 100$ mK; ‘‘cold’’) to those at 1.4 K (where no orientation is present; ‘‘warm’’). This function can be written as [15,16]

$$W(\theta) = 1 + f \sum_{k \neq 0} A_k B_k Q_k P_k(\cos \theta). \quad (1)$$

Here the factor f represents the effective fraction of nuclei oriented by the hyperfine interaction, and it is assumed that the remainder $(1-f)$ is not oriented at all. The P_k are Legendre polynomials, the Q_k correct for the finite dimensions of source and detector and the B_k parameters describe the nuclear orientation. For α emission the directional distribution coefficients A_k can be written as [16]

$$A_k = \frac{\sum_{L,L'} a_L a_{L'} \cos(\sigma_L - \sigma_{L'}) F_k^\alpha(L, L', I_f, I_i)}{\sum_L a_L^2}, \quad (2)$$

where F_k^α are the F -coefficients modified for α decay [16], and a_L and σ_L are the amplitude and phase of the α wave with angular momentum L . From the a_L the mixing ratios $\delta_{0L} \equiv a_L/a_0$ are defined. Since parity is conserved in α decay only $L=0,2,4, \dots$ are involved in the favored decays investigated in the present work. In the data evaluation terms up to $L=4$ have been taken into account. In order to be

independent of the lifetime of the nuclei and of variations in the beam intensity, double ratios are constructed by combining the data of two detectors. The anisotropy is then defined as

$$R_{ij}(\theta_i, \theta_j) = \frac{[N(\theta_i)/N(\theta_j)]_{\text{cold}}}{[N(\theta_i)/N(\theta_j)]_{\text{warm}}} - 1. \quad (3)$$

In order to determine the L -mixing ratios from the anisotropy data for the favored α transitions the ‘‘fraction at good sites’’ f and temperature T have to be known. The $^{57}\text{CoFe}$ and $^{54}\text{MnNi}$ nuclear thermometers that were mounted on the cold finger could, in most cases, not be used because the γ spectra, mostly originating from isobaric contaminants in the ion beam, were so intense that the Ge detectors had to be positioned at a distance where the γ rays from the thermometer sources were too weak in intensity for a proper temperature evaluation. This situation could not be improved by reducing the beam intensity because the yield of ^{189}Bi was very low, i.e., only a few tens of ions per second.

The problem was solved by using the anisotropies observed for the $9/2^- \rightarrow 1/2^+$ pure $L=5$ α transition from the $^{191,193}\text{Bi}$ ground states to the $^{187,189}\text{Tl}$ ground states, respectively, for a simultaneous determination of magnetic splitting to temperature ratios $\mu B/k_B T$ (with μ the nuclear magnetic moment, B the total magnetic field experienced by the nuclei and k_B the Boltzmann constant) and the parameter f . For these transitions the A_k directional distribution coefficients are known. The parameter f is presumably the same for different Bi isotopes due to identical implantation conditions. Also, in the evaluation of temperature it was assumed that the $^{191,193}\text{Bi}$ isotopes experience the same hyperfine interaction. In columns 2–4 of Table I the experimental anisotropies of the $L=5$ α transitions as measured in different parts of the experiment are listed. For the fraction f the weighted average value $f=0.87(2)$ was obtained. This value is in agreement with $0.82 < f < 0.96$ which resulted from a fit of the anisotropy of the 847 keV pure $E2\gamma$ transition in the decay of ^{203}Bi , the magnetic moment of which is well known and which was also implanted into the same iron foil.

The derived magnetic splitting to temperature ratios $\mu B/k_B T$ are given in column 5 of Table I. Effective inverse temperatures $1/T^*$, shown in column 6, were calculated from these magnetic splitting to temperature ratios by using the precisely known hyperfine field $B_{\text{hyp}} = 119.0(13)$ Tesla for Bi in Fe [17,18] and an estimated value of $\mu = 3.9(2) \mu_N$ for the magnetic moments of the $\pi h_{9/2}$ ground states of $^{189,191,193}\text{Bi}$ since these have not been measured. This estimate is based on the experimentally determined moments of the corresponding state in the thallium isotopes $^{189,191,193}\text{Tl}$ [19].

For the first measurement with ^{193}Bi (line 1 in Table I) an effective on-line base temperature $T^* = 11.3(23)$ mK was calculated from the magnetic splitting to temperature ratio. In this measurement thermometry with γ rays from $^{54}\text{MnNi}$ was possible and this yielded $T = 13.3(5)$ mK. The good agreement between the values of T^* and T gives confidence in the correctness of the assumption made to derive T^* . Nevertheless, in order to make the analysis independent of this estimate, the data for the favored transitions were analyzed in terms of the magnetic splitting to temperature ratio, and the

TABLE I. Experimental anisotropies R_{ij} (three different detector ratios are given) for the 6639 keV and 6174 keV $9/2^- \rightarrow 1/2^+$ pure $L=5$ α transitions in the decay of ^{191}Bi and ^{193}Bi , respectively. Also listed is the magnetic splitting to temperature ratio $\mu B/k_B T$, which was calculated with $f=0.87(2)$, and the inverse effective temperature $1/T^*$ obtained from this when assuming $\mu=3.9(2)\mu_N$. The measurements are listed in chronological order.

Measurement	$R(14^\circ, 90^\circ)$	$R(51^\circ, 90^\circ)$	$R(14^\circ, 51^\circ)$	$\mu B/k_B T$	$1/T^* [\text{K}^{-1}]$
$^{193}\text{Bi}/1$		-0.763(11)		15(3)	88(18)
$^{193}\text{Bi}/2$		-0.334(26)		2.7(2)	15.9(15)
$^{191}\text{Bi}/1$	-0.931(13)	-0.721(15)	-0.753(45)	10.4(10)	63(8)
$^{191}\text{Bi}/2$	-0.499(40)	-0.252(36)	-0.330(30)	2.0(1)	11.8(8)
$^{193}\text{Bi}/3$	-0.938(21)	-0.740(90)	-0.760(13)	12.0(25)	70(15)
$^{191}\text{Bi}/3$	-0.946(14)	-0.739(13)	-0.793(54)	11.8(12)	69(8)

effective temperature T^* extracted from it was only used to present the data graphically. For ^{189}Bi the $9/2^- \rightarrow 1/2^+$ transition could not be observed. However, since the measurement with ^{189}Bi was carried out just before the third measurement on ^{191}Bi and ^{193}Bi , which both yielded $\mu B/k_B T \cong 12$ at base temperature (see Table I) we estimate $\mu B/k_B T = 12.0(25)$ for the measurement on ^{189}Bi as well.

The experimental anisotropies R_{ij} for the favored $9/2^- \rightarrow 9/2^-$ α transition are listed in Table II and shown graphically in Fig. 1. They were analyzed using the values for f and $\mu B/k_B T$ obtained from the analysis of the $9/2^- \rightarrow 1/2^+$ transition discussed above. Due to the short half-life of ^{189}Bi ($T_{1/2} = 0.68(3)$ s [11]) one expects incomplete relaxation for this isotope so that not all nuclei become fully oriented before decaying. The relaxation time for Bi in Fe has been measured by NMR/ON (nuclear magnetic resonance on oriented nuclei) on ^{206}Bi cold implanted in iron [18]. The anisotropy data for ^{206}Bi were fitted with a single exponential to give empirically an effective relaxation time $T'_1 = 0.84(3)$ s at $T = 12.6(10)$ mK in an external magnetic field of 0.08 T. Scaling this value to ^{189}Bi with the relation [20]

$$C_K^{189} \left[\frac{\mu^{189}}{I^{189}} \right]^2 = C_K^{206} \left[\frac{\mu^{206}}{I^{206}} \right]^2, \quad (4)$$

where C_K is the Korringa constant which characterizes the relaxation process, and taking into account the observed magnetic field dependence of the relaxation rate for impurities in iron [21], one finds $T'_1(^{189}\text{BiFe}, 0.2 \text{ T}) = 1.39(22)$ s. The magnetic interaction temperature for ^{189}Bi is $T_{\text{int}} = \mu B / I k_B \cong 38$ mK, but our measurements were made at $T \cong 14$ mK. Therefore the low temperature limit [22] is applicable in which case the effective relaxation time T'_1 is temperature independent. Using this effective relaxation time and assuming again an empirical single exponential behavior of anisotropy, an effective orientation parameter [23] B_2 could be calculated for ^{189}Bi and applied in the analysis of the anisotropy data. The derivation of the relaxation time and the subsequent correction for incomplete relaxation were thus performed in a consistent way so that systematic errors due to this correction should cancel to first order. Because we assume the same magnetic moment for $^{189,191,193}\text{Bi}$ and since the data for ^{191}Bi and ^{193}Bi were taken at the same temperature and in the same magnetic field as the ^{189}Bi data, $T'_1 = 1.39(22)$ s is valid for $^{191,193}\text{Bi}$ as well. Due to the half-life of ^{191}Bi of 12(1) s, the correction here turned out to be small, but non-negligible, so that it was employed for this as well. For ^{193}Bi [$T_{1/2} = 67(3)$ s] the correction has no effect. The calculated reduction factors for the B_2 orientation parameters and the resulting A_2 coefficients are listed in Table III. In the case of ^{189}Bi two approaches were used for the B_4

TABLE II. Experimental anisotropies R_{ij} (three different detector ratios are given) for the 6672 keV, 6311 keV, and 5899 keV $9/2^- \rightarrow 9/2^-$ favored α transitions in the decay of ^{189}Bi , ^{191}Bi , and ^{193}Bi , respectively. Also listed is the magnetic splitting to temperature ratio $\mu B/k_B T$ which was calculated from the measured anisotropies R_{ij} for the $9/2^- \rightarrow 1/2^+$ pure $L=5$ α transition in the decay of ^{191}Bi and ^{193}Bi , and the inverse effective temperature $1/T^*$ obtained from this when assuming $\mu=3.9(2)\mu_N$. The magnetic splitting to temperature ratio for ^{189}Bi is an estimate as is discussed in the text. The measurements are listed in chronological order.

Measurement	$\mu B/k_B T$	$1/T^* [\text{K}^{-1}]$	$R(14^\circ, 90^\circ)$	$R(51^\circ, 90^\circ)$	$R(14^\circ, 51^\circ)$
$^{193}\text{Bi}/1$	15(3)	88(18)		0.077(14)	
$^{193}\text{Bi}/2$	2.7(2)	15.9(15)		0.039(11)	
$^{191}\text{Bi}/1$	10.4(10)	63(8)	-0.107(11)	-0.0574(77)	-0.053(10)
$^{191}\text{Bi}/2$	2.0(1)	11.8(8)	-0.054(11)	-0.0168(80)	-0.038(10)
^{189}Bi	12.0(25)	70(15)	-0.173(45)	-0.119(30)	-0.060(50)
$^{193}\text{Bi}/3$	12.0(25)	70(15)	0.220(17)	0.090(12)	0.119(13)
$^{191}\text{Bi}/3$	11.8(12)	69(8)	-0.098(10)	-0.0485(73)	-0.0524(94)

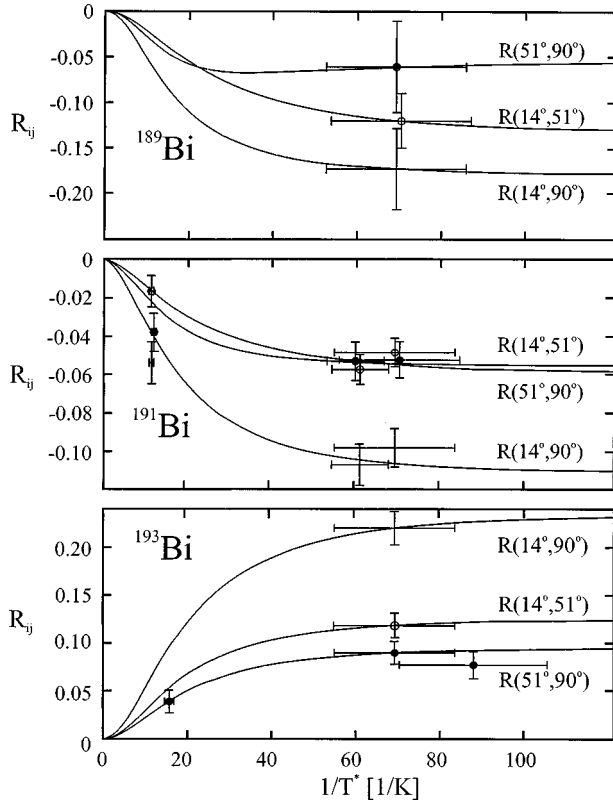


FIG. 1. Anisotropies R_{ij} versus inverse effective temperature $1/T^*$ for the 5899 keV, 6311 keV and 6672 keV $9/2^- \rightarrow 9/2^-$ favored α transitions in the decay of ^{193}Bi , ^{191}Bi , and ^{189}Bi , respectively.

orientation parameter. First we did not apply a correction for incomplete relaxation so that a lower limit was obtained for the A_4 coefficient. In a second approach (line 2 of Table III) we used the reduction factor k_2 as derived above to get the corresponding reduction factor for the B_4 orientation parameter from the tables in Ref. [20]. The resulting A_4 coefficients and mixing ratios are listed in Table III. For $^{191,193}\text{Bi}$ a correction of the B_4 parameters did not affect the results.

The resulting δ_{0L} ($L=2,4$) mixing ratios between the $L=0,2,4$ partial α waves for the favored $\pi h_{9/2} \rightarrow \pi h_{9/2}$ α decay of the odd $^{189,191,193}\text{Bi}$ isotopes are listed in Table III (see also Fig. 2). Note that the use of one or the other approach to take into account incomplete relaxation in the case of ^{189}Bi

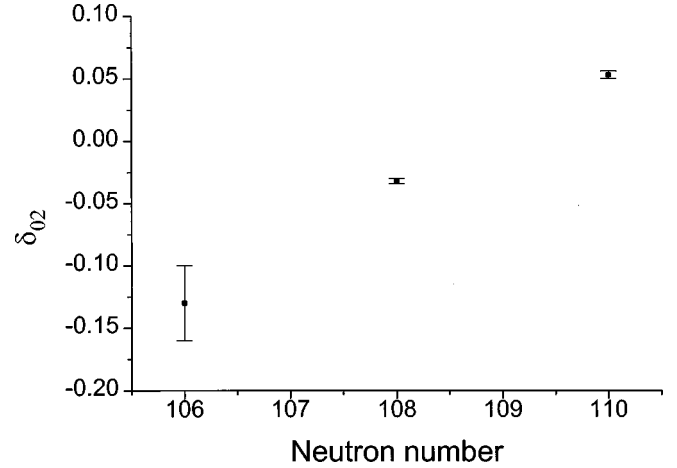


FIG. 2. Experimentally obtained δ_{02} mixing ratios versus neutron number N for the favored $9/2^- \rightarrow 9/2^-$ α transitions in the decay of $^{189,191,193}\text{Bi}$.

has no significant effect on the value of the mixing ratio δ_{02} . As can be seen in Fig. 2, the value of δ_{02} decreases more or less linearly with mass number A .

III. DISCUSSION

The quadrupole moments of $^{189,191,193}\text{Bi}$ have not been determined experimentally. The calculations of Möller *et al.* [24] however, predict the same, very small, deformation parameter $\beta = -0.052$ for all three isotopes. As a result, the model of Delion *et al.* [5,6], in which the α anisotropy is dominated by barrier penetration, would predict the same result for each of the three isotopes. This is in clear contradiction with the experimental observations as the data show a similar behavior to that previously observed for the odd isotopes $^{199-211}\text{At}$ [1,25], i.e., a decrease of δ_{02} with increasing number of neutron holes in the $N=126$ closed shell (see Fig. 2). As was the case for the At isotopes, the δ_{02} values for the Bi isotopes investigated here can be understood in terms of the shell model. Since Bi has only one proton outside the $Z=82$ closed shell the $L=2$ component in the α particle arising from the protons is mostly due to the 2^+ coupling of a proton hole pair in the $2d_{3/2}$ shell (of the Tl daughter nucleus) below $Z=82$. Because the quadrupole part of the p - n interaction polarizes the nuclear core, thus creating a

TABLE III. Values for the experimental directional distribution coefficients A_2 and A_4 , the mixing ratios δ_{02} and δ_{04} , and the corresponding intensities of the $L=2$ partial α waves for the $9/2^- \rightarrow 9/2^-$ favored α transition in the decay of ^{189}Bi , ^{191}Bi , and ^{193}Bi on the basis of the experimental anisotropies R_{ij} listed in Table II. In the second and third column the reduction factors k_2 and k_4 for the B_2 respectively B_4 orientation parameters are listed that were used to take into account the effects from incomplete relaxation when extracting A_2 and A_4 from the experimental data. The intensity of the $L=2$ wave is defined as $\delta_{02}^2 / (1 + \delta_{02}^2 + \delta_{04}^2)$.

Isotope	k_2	k_4	A_2	A_4	δ_{02}	δ_{04}	$L=2\%$
^{189}Bi	0.415(40)	1.000	-0.260(56)	$>0.073(39)$	-0.136(32)	0.036(23)	1.8(9)
	0.415(40)	0.287(41) ^a	-0.260(56)	0.26(14)	-0.130(30)	0.15(10)	1.6(8)
^{191}Bi	0.926(12)	1.000	-0.064(4)	$>0.016(7)$	-0.032(2)	0.10(4)	0.10(1)
^{193}Bi	0.986(2)	1.000	+0.107(7)	0.00(1)	0.053(3)	-0.001(6)	0.28(3)

^aFrom [20]; see text.

mixed ground state containing 2^+ neutron excitations, also an $L=2$ component arising from the neutrons is expected in the α decay. Since the $L=2$ component arising from the protons (neutrons) in the α particle gives rise to a positive (negative) contribution to the α anisotropy, and since the contribution of the protons to the $L=2$ partial α wave should not vary much for the three isotopes, one expects a decrease of δ_{02} with decreasing neutron number (and increasing neutron hole number), as is indeed observed. The variation of δ_{02} with neutron number for the three Bi isotopes discussed here is thus caused mainly by the variation of the strength of the 2^+ neutron excitations due to core polarization. This clearly indicates, at least for nuclei near the $Z=82$ and $N=126$ shell closures, the need for an explicit consideration of the proton-neutron interaction in the calculation of the angular distribution of α decay.

Since proton holes from the $d_{3/2}$ shell (case of $\text{Bi} \rightarrow \text{TI}$ decay) can only give rise to $L=0$ and $L=2$ partial waves, whereas protons from the $h_{9/2}$ shell (case of $\text{At} \rightarrow \text{Bi}$ decay) can give rise to $L=0, 2, 4, 6,$ and 8 waves, the positive $L=2$ contribution arising from the protons in the α particle is expected to be larger for Bi than for At α decay. This fact is also reflected in the so-called ‘‘coefficients of fractional parentage’’ which describe the coupling of the nucleons in a given shell [26]. For the neutrons no large difference between Bi and At is expected. Therefore, the zero crossing of the δ_{02} values (and thus of the anisotropy) is expected to occur further from the $N=126$ shell closure in the case of Bi, compared to At α decay. In fact, we observe that for At the zero crossing occurs between $N=118$ and $N=116$ [25], while for Bi it occurs between $N=110$ and $N=108$.

For a given proton number the strength of the quadrupole interaction $Q_{\pi\nu}$ in this region is approximately given by $Q_{\pi\nu} \propto N_\nu \sqrt{\Omega_\nu - N_\nu}$ [27] with N_ν being the number of neutron hole/valence pairs and Ω_ν the maximum number of neutron pairs in the shell, i.e., 22 in this case. It follows that, close to the $N=126$ closed shell (where N_ν is small compared to Ω_ν), $Q_{\pi\nu} \propto N_\nu$ to first order (as was observed for the odd isotopes $^{199-211}\text{At}$ [1,25]), while the increase of $Q_{\pi\nu}$ slows down towards mid-shell where it reaches a maximum before decreasing again. Unfortunately, due to the relatively large error bar on the δ_{02} value for ^{189}Bi (with $N=106$, i.e., only

two neutrons above mid-shell), compared to $^{191,193}\text{Bi}$, no definite conclusion as to whether this behavior is reflected by the experimental data (see Fig. 2) can be drawn.

In conclusion, the data presented here for $^{189,191,193}\text{Bi}$ constitute substantial support for the recent findings that the α anisotropy of favored transitions of near-spherical nuclei near the $Z=82$ and $N=126$ shell closures depends on the structure of the decaying nucleus, rather than on deformation changes in the mean field. Furthermore, our data stress the importance of the p - n interaction in the description of α decay, even for heavy nuclei close to the proton drip-line. Additional confirmation for these observations could be given by a measurement of the anisotropy for the favored $9/2^- \rightarrow 9/2^-$ α transition of the isotopes ^{187}Bi and ^{185}Bi , which are exactly on and just beyond the $N=104$ mid-shell respectively. Unfortunately, however, apart from the fact that both isotopes cannot yet be produced in sufficient abundance, incomplete relaxation will most probably seriously affect, or even prohibit, such measurements on ^{185}Bi and ^{187}Bi with the low temperature nuclear orientation method as the half-lives of these isotopes are in the millisecond region. Finally, it may be noted that our data do not give information on the importance of the p - n interaction in the case of large deformations. For these nuclei one would indeed expect that the tunneling process of the α particle through the deformed Coulomb barrier plays a much more important role in the determination of the angular distribution in α decay. Although this topic has not yet been clearly verified by quantitative experimental data, it is presently being investigated [28].

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