Drastic Increase of the Nuclear Deformation in Bromine between $A = 79$ and 77

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The electric quadrupole interaction frequencies eQV_{zz}/h of ^{77}Br ($I^\pi = 3/2^-$; $T_{1/2} = 57$ h) and ^{82}Br ($I^\pi = 5^-$; $T_{1/2} = 35$ h) in hcp Co were measured with modulated adiabatic fast passage on oriented nuclei. The electric quadrupole moment of ^{77}Br is deduced to be $Q = +0.530(22)$ b, which, on the condition of strong coupling, implies a nuclear deformation of $\beta_2 = +0.354(15)$. This manifests a drastic increase of the nuclear deformation between ^{79}Br ($\beta_2 \sim +0.22$) and ^{77}Br. [S0031-9007(98)06361-3]

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The study of nuclear properties in long isotopic and isotonic chains has been the subject of many investigations, both experimental and theoretical [1], the main source of information on nuclear ground state properties being optical spectroscopy [2]. Such investigations revealed that the nuclear shape may change drastically between two nuclei which differ only by one or two nucleons. It was first observed with $_{80}$Hg, for which a drastic change in the mean square charge radius was found between ^{187}Hg and ^{185}Hg [3]. This phenomenon was explained by nearly degenerate nuclear states of different shape and is now usually denoted as "shape coexistence." As N changes for a given Z or vice versa strong competition can occur between the different forces which separately drive the nucleus towards prolate, oblate, triaxial, and spherical shapes. For $_{80}$Hg, $_{79}$Au, and $_{78}$Pt—these elements are just below the $Z = 82$ shell closure—, the neutron-rich nuclei are weakly deformed with oblate shapes, whereas the neutron-deficient isotopes are strongly deformed with prolate shapes [4–6]. For Hg and Au the shape transition is sharp [4,7], whereas it is smooth for Pt [6]. Meanwhile it is generally adopted that the phenomenon of shape coexistence exists in many regions of the nuclear chart [1]. One of these is the region just below the $Z = 40$ shell closure, which has been studied extensively meanwhile [8]. In the isotopic chains of $_{38}$Sr, $_{37}$Rb, and $_{36}$Kr the nuclear shape changes from nearly spherical around $N = 50$ (^{88}Sr$_{50}$, ^{87}Rb$_{50}$, ^{86}Kr$_{50}$) to strong prolate deformation ($\beta_2 \sim 0.4$) at $N \sim 40$ and $N \sim 60$ [9–13]. For the light isotopes of Sr, Rb, and Kr the deformation develops gradually from $N \sim 46$ to $N \sim 40$. For several even-even nuclei β_2 can be determined also from lifetime measurements available in the literature [14,15]. In this context the nuclear shapes of $_{35}$Br would be interesting. For Br, however, laser spectroscopy (LS) measurements have not yet been reported in the literature. Thus direct information on the neutron-number dependence of the nuclear deformation in the bromine chain—as normally obtained from LS measurements of mean square radii and nuclear electric quadrupole moments—is not available. Concerning the ground-state properties of neutron-deficient Br isotopes, only the magnetic moments are known experimentally from nuclear orientation (NO) [16] and nuclear magnetic resonance on oriented nuclei (NMR-ON) measurements [17,18]. From these measurements Griffiths *et al.* drew the conclusion that their results pointed to a *gradual* increase in nuclear ground-state deformation from $\beta_2 = 0.20$, $\gamma = 0$ in ^{81}Br to $\beta_2 = 0.33$, $\gamma = 0$ in ^{75}Br [16]. Such a gradual increase of the deformation with decreasing N was actually in agreement with the change of the deformation of $_{36}$Kr, $_{37}$Rb, and $_{38}$Sr. For bromine, however, the overall deformation may be influenced considerably by the single-particle properties of the valence proton. If this valence proton occupies a strongly downsloping Nilsson orbit, it may drive the nucleus towards a large deformation. Such a situation is actually realized: The odd-A Br nuclei $^{75-87}$Br all have ground state spin $3/2^-$. Within the Nilsson scheme, the 35th proton is expected to occupy the $I^\pi[Nn_z\Lambda] = 3/2^-[301]$ orbital for quadrupole deformations up to $\beta_2 < 0.22$ and the $3/2^-[312]$ orbital at larger deformations in the range $0.30 < \beta_2 < 0.40$ [16]. The magnetic moments are considerably different for these two single-particle states, and from the fact that the experimental magnetic moment changes from $\mu = +2.106\,400(4)\mu_N$ for ^{79}Br to $\mu = (+)0.9738(5)\mu_N$ for ^{77}Br it was concluded that the deformation increases moderately from $\beta_2 \sim 0.22$ to ~ 0.28–0.30 between ^{79}Br and ^{77}Br. It is obvious that the large change of the magnetic moment must be due to the fact that different Nilsson orbitals are involved, and that it depends on the deformation which orbital is occupied. On the other hand, for $0.30 < \beta_2 < 0.40$, the dependence of the magnetic moment on the deformation is relatively weak. In addition, magnetic moments depend strongly on configuration mixing. This means that it is very difficult to predict the magnetic moment for intermediate deformations, $0.25 < \beta_2 < 0.30$. Thus, the uncertainty in the configuration prevents the exact determination of the nuclear deformation via the magnetic moment.

The electric quadrupole moment, however, is much more sensitive on the nuclear deformation. The quadrupole moment of ^{79}Br is known to be $Q = +0.331(4)$ b [19,20], which is consistent with a deformation of $\beta_2 \sim 0.22$. Thus the shape change between $A = 79$ and 77 can be determined via a measurement of the quadrupole moment of ^{77}Br. Here we report measurements of the quadrupole interaction of ^{77}Br and ^{82}Br in hcp Co. As measurement technique we chose modulated adiabatic passage on oriented nuclei (MAPON) [21,22]. Taking into account the free-atom electric quadrupole interaction constants B for ^{79}Br [19] and ^{82}Br [23] which are known with high precision from atomic-beam magnetic-resonance measurements, we get a precise value for the ratio of the quadrupole moments of $3/2^-$ ^{77}Br and ^{79}Br, which allows us to deduce the increase of deformation between ^{79}Br and ^{77}Br with high precision.

In hcp Co, in addition to the magnetic hyperfine field, an (axially symmetric) electric field gradient (EFG) exists, with the principal (z) axis given by the c axis of the single crystal. For NMR on impurity atoms with nuclear spin I, there exists a set of $2I$ equidistant subresonances. These are separated by

$$\Delta\nu_Q(\theta) = 3\nu_Q P_2(\cos\theta)/[2I(2I - 1)], \tag{1}$$

$$\nu_Q = eQV_{zz}/h, \tag{2}$$

where ν_Q is the quadrupole interaction frequency, eQ and V_{zz} are the spectroscopic quadrupole moment and the principal component of the EFG tensor, and θ is the angle between the direction of the hyperfine field and the single-crystal c axis. The $P_2(\cos\theta)$ dependence of the subresonance separation is exact only for $|\nu_Q| \ll \nu_M = |g\mu_n B_{HF}/h|$, where ν_M is the magnetic hyperfine interaction frequency, g is the nuclear g factor, and B_{HF} is the magnetic hyperfine field. If this condition is not fulfilled the subresonance frequencies have to be calculated by diagonalization of the Hamiltonian as outlined in Ref. [24].

If $\Delta\nu_Q$ is larger than the inhomogeneous line width Γ, a quadrupole-subresonance-resolved spectrum can be measured and $\Delta\nu_Q$ is determined directly from the resonance centers of the subresonances. If $\Delta\nu_Q$ is smaller than the inhomogeneous line width the quadrupole interaction can be determined with the MAPON technique. Its principle is described in detail in Refs. [21,22]. It is a variant of adiabatic fast passage (AFP) in which the rf frequency is slowly and continuously swept through the resonance conditions. The response of the nuclear spin system is detected via the anisotropy of the γ radiation emitted in the decay of low-temperature-oriented radioactive nuclei. With AFP on a quadrupole split system, cyclic permutation of the sublevel population probabilities a_m is obtained. A MAPON pulse consists of a sequence of *two* AFP pulses which are separated in frequency by $\Delta\nu$. The influence of the MAPON pulse on the time evolution of the sublevel populations is different for $\Delta\nu > \Delta\nu_Q$ and $\Delta\nu < \Delta\nu_Q$: (i) For $\Delta\nu > \Delta\nu_Q$ the MAPON pulse causes a distinctive reorganization of sublevel populations in which a net population transfer of the first *two* accessed substate populations is cyclically permuted through to the final two substates. (ii) For $\Delta\nu < \Delta\nu_Q$ the successive double transits through each subresonance yield a null result. The different sublevel populations a_m in these two cases cause different γ anisotropies, and $\Delta\nu_Q$ is obtained from the transition between the respective γ anisotropy values. In reality, the quadrupole interaction is inhomogeneously broadened which can be described by a Gaussian distribution of the quadrupole subresonance frequencies. The MAPON spectrum (γ anisotropy as function of $\Delta\nu$) is then given by the integral of this distribution. In the following the half value of the MAPON spectrum is denoted as $\Delta\nu_Q^{(\mathrm{MAP})}$.

Experimentally, two different geometries are used mainly for MAPON measurements with a hcp Co host: In the so-called "0° geometry" the single-crystal c axis is mounted parallel to the direction of B_{ext}. Here the best possible accuracy is normally obtained for the determination of ν_Q. However, 0°-geometry MAPON measurements may be prevented by too small an enhancement factor for the radio frequency (rf) field. The transition rate is proportional to the square of the effective rf field at the nuclear site $B_1^{(\mathrm{eff})}$, which is enhanced by the hyperfine interaction. The enhancement factor η is defined as

$$B_1^{(\mathrm{eff})} = \eta B_1^{(\mathrm{app})}, \tag{3}$$

where $B_1^{(\mathrm{app})}$ is the amplitude of the applied rf field.

In the "90° geometry" the single-crystal is mounted with the c axis perpendicular to the direction of B_{ext}. Here the enhancement factor $\eta^{\perp}$ has a resonancelike behavior around $B_A^{\perp} = 13.5$ kG [25],

$$\eta^{\perp} = 1 + \frac{B_{\mathrm{HF}}}{B_{\mathrm{ext}} - B_A^{\perp}}. \tag{4}$$

For the determination of ν_Q from 90°-geometry measurements of $\Delta\nu_Q^{(\mathrm{MAP})}$ several (small) corrections have to be applied:

$$\nu_Q = -2\,\frac{2I(2I - 1)}{3}\,[\Delta\nu_Q^{(\mathrm{MAP})} + \Delta\nu_Q^{(\alpha)} + \Delta\nu_Q^{(Q/\mathrm{M})} + \Delta\nu_Q^{(\mathrm{sw})} + \Delta\nu_Q^{(\mathrm{rf})}]. \tag{5}$$

(i) $\Delta\nu_Q^{(\alpha)}$ is due to the unavoidable misalignment of the single-crystal c axis with respect to the direction of B_{ext}; this angle is typically $\alpha = 89.5(5)^\circ$ instead of the exact 90°. (ii) $\Delta\nu_Q^{(Q/\text{M})}$ is due to the fact that the principal axes systems of the electric quadrupole interaction and the magnetic interaction are not collinear. (iii) $\Delta\nu_Q^{(\text{sw})}$ is due to the finite sweep time of the MAPON pulse with respect to the nuclear spin lattice relaxation time. (iv) $\Delta\nu_Q^{(\text{rf})}$ is due to rf power broadening. All of these corrections can be determined by model calculations.

It should be realized that the external magnetic field B_{ext} for the 90°-geometry MAPON measurement has to be chosen as a compromise between two conflicting conditions: (i) To get a large MAPON signal, B_{ext} near 13.5 kG is required [see Eq. (4)]. (ii) A small angular uncertainty and, hence, a small uncertainty in the respective correction $\Delta\nu_Q^{(\alpha)}$ requires $B_{\text{ext}} \gg 13.5$ kG. Therefore, the 90°-geometry MAPON measurements as described here were performed for $B_{\text{ext}} = 16$ and 17 kG.

A sample of ^{77}Br ($I^\pi = 3/2^-$; $T_{1/2} = 57.0$ h) in hcp-Co was prepared by mass separator implantation at ISOLDE/CERN. ^{77}Rb was implanted with $E = 60$ keV into a hcp-Co single crystal with high surface quality (total dose $\sim 5 \times 10^{13}$ ions/cm^2). The following decay chain is relevant: ^{77}Rb ($T_{1/2} = 3.9$ min) $\rightarrow$ ^{77}Kr ($T_{1/2} = 1.2$ h) $\rightarrow$ ^{77}Br. After the irradiation the sample was mounted on the coldfinger of a ^{3}He-^{4}He-dilution refrigerator, first with $c \parallel B_{\text{ext}}$ (0° geometry), and cooled to temperatures $\sim$10 mK. The γ rays emitted in the decay of ^{77}Br were detected with 4 Ge detectors placed at 0°, 180°, and 90°, 270° with respect to the single crystal c axis. The γ anisotropy $\epsilon = W(0^\circ)/W(90^\circ) - 1$ [here $W(\vartheta)$ is the angular distribution of γ rays emitted in the decay of oriented nuclei [26]] of all γ transitions with sufficient intensity was analyzed. The NMR-ON resonance for $B_{\text{ext}} = 0$ (in 0° geometry) was found at 295.64(9) MHz. Subsequent MAPON measurements, however, showed no significant MAPON signal. This was attributed to too small rf power at the nuclear sites. Therefore all further measurements were performed in 90° geometry. The MAPON spectrum measured for $B_{\text{ext}} = 16.0$ kG is shown in Fig. 1. The result of the *least-squares* fit (solid line in Fig. 1) is $\Delta\nu_Q^{(\text{MAP})} = -1.535(57)$ MHz. The corrections introduced before are $\Delta\nu_Q^{(\alpha)} = -25(25)$ kHz, $\Delta\nu_Q^{(Q/\text{M})} = -3(3)$ kHz, $\Delta\nu_Q^{(\text{sw})} = -3(3)$ kHz, and $\Delta\nu_Q^{(\text{rf})} = -2(2)$ kHz. Thus we obtain according to Eq. (5) as a final result

$$\nu_Q(^{77}\text{Br}\underline{\text{Co}}^{(\text{hcp})}) = +6.28(24)\ \text{MHz}.$$

Two samples of ^{82}Br ($I^\pi = 5^-$; $T_{1/2} = 35.3$ h) in hcp Co were prepared by implantation at the mass separator in Bonn. For the first sample total dose and energy were 3×10^{14} ions/cm^2 and $E = 120$ keV. The 90°-geometry MAPON results for $B_{\text{ext}} = 15$ kG were $\Delta\nu_Q^{(\text{MAP})} = -142(9)$ kHz and $\Gamma_{\Delta\nu_Q} = 106(30)$ kHz for the half width. For the second sample the total implantation dose was much lower, 6×10^{12} ions/cm^2, and the energy was $E = 140$ keV. A MAPON spectrum measured for $B_{\text{ext}} = 17$ kG is shown in Fig. 2. The width of the MAPON signal was now significantly smaller. The results are $\Delta\nu_Q^{(\text{MAP})} = -145.1(9)$ kHz; $\Gamma_{\Delta\nu_Q} = 17(3)$ kHz. Applying corrections as described before of $\Delta\nu_Q^{(\alpha)} = -1.5(1.5)$ kHz, $\Delta\nu_Q^{(Q/\text{M})} = -0.5(3)$ kHz, $\Delta\nu_Q^{(\text{sw})} = -0.3(3)$ kHz, $\Delta\nu_Q^{(\text{rf})} = -0.4(6)$ kHz, the final result is

$$\nu_Q(^{82}\text{Br}\underline{\text{Co}}^{(\text{hcp})}) = +8.87(11)\ \text{MHz}.$$

Our results are compiled in Table I, together with free-atom electric-quadrupole interaction constants B, and spectroscopic quadrupole moments of bromine isotopes. The Sternheimer correction has been applied as given in Ref. [20]. The change in deformation of the $3/2^-$ ground states of the odd bromine isotopes can be inferred from the ratio $Q(^A\text{Br})/Q(^{A+2}\text{Br})$. For ^{79,81}Br this ratio is 1.20, which reflects the smooth increase of the deformation as also realized for the neighboring elements Sr, Rb, and Kr. For ^{77,79}Br, however, this ratio is 1.60(7), indicating a drastic increase of the deformation between ^{79}Br and ^{77}Br. With the Sternheimer correction as given in Ref. [20] the

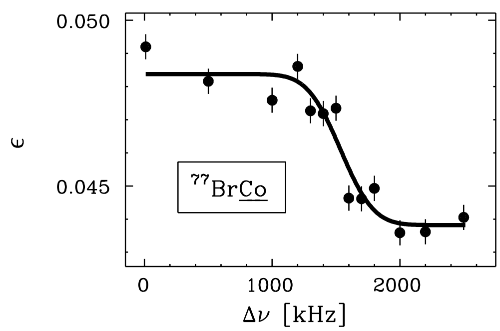

FIG. 1. MAPON spectrum of ^{77}Br in hcp Co. The solid line is the result of a *least-squares* fit, taking into account a Gaussian distribution for the quadrupole subresonance resonance separation. The MAPON signal is given by the integral of this distribution, i.e., an error function. It has its half value at $\Delta\nu_Q^{(\text{MAP})}$ and width $\Gamma_{\Delta\nu_Q}$.

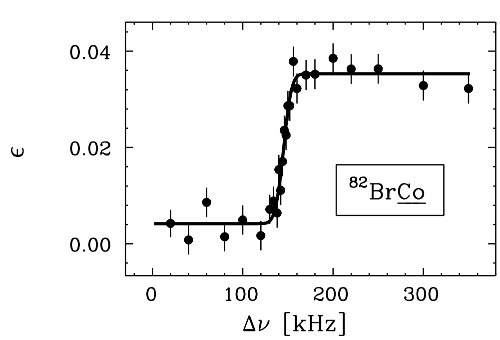

FIG. 2. MAPON spectrum of ^{82}Br in hcp Co. The different sign of the MAPON signal in comparison to ^{77}Br is due to the fact that the γ anisotropy of ^{77}Br is positive, whereas it is negative for ^{82}Br.

TABLE I. Quadrupole interaction frequencies ν_Q in hcp Co, free-atom electric-quadrupole interaction constants B, and spectroscopic quadrupole moments of bromine isotopes.

Isotope	I^π	E [keV]	ν_Q [MHz]	B [MHz]	Q [b]
^{77}Br	$3/2^-$	0	$+6.28(24)^{a}$		$+0.530(22)^{a}$
^{79}Br	$3/2^-$	0		$-384.878(8)^{b}$	$+0.331(4)^{b,c}$
^{81}Br	$3/2^-$	0		$-321.516(8)^{b}$	$+0.276(4)^{b,c}$
^{76}Br	1^-	0		$-314.329(22)^{d}$	$+0.270(4)^{d,c}$
^{80}Br	1^+	0		$-227.62(10)^{e}$	$+0.196(3)^{e,c}$
	2^-	37			$\pm0.173(6)^{c}$
	5^-	86		$-874.9(2)^{e}$	$+0.751(10)^{e,c}$
^{82}Br	5^-	0	$+8.87(11)^{a}$	$-870.7(9)^{f}$	$+0.748(10)^{f,c}$

[a]This work. [b]Ref. [19]. [c]Ref. [20]. [d]Ref. [27]. [e]Ref. [28]. [f]Ref. [23].

quadrupole moment of ^{77}Br is deduced to be

$$Q(^{77}\mathrm{Br}) = +0.530(22)\ \mathrm{b}\,.$$

The deformation parameter β_2 can be calculated using the rotational-model relationship

$$\beta_2 = \frac{\sqrt{5\pi}}{3ZR_\circ^2}\,\frac{(I+1)(2I+3)}{3K^2 - I(I+1)}\,Q\,, \tag{6}$$

which holds in the case of axially symmetric and strongly deformed nuclei. Taking $I = K = 3/2$ and $R_\circ = 1.25A^{1/3}$ fm, the deformation parameter for ^{77}Br is found to be $\beta_2 = +0.354(15)$, i.e., much larger than expected from the extrapolation of the smooth increase between ^{81}Br ($\beta_2 \sim +0.19$) and ^{79}Br ($\beta_2 \sim +0.22$). (In the case of a triaxial deformation and/or K mixing the actual β_2 would be even larger.) Our β_2 is considerably larger than the values deduced from the magnetic-moment measurements and the theoretical calculations. Thus the deformation-driving effect of the downsloping $\pi 3/2^-[312]$ orbital has been strongly underestimated until now.

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