Measurements of quadrupole-interaction-resolved NMR on oriented $^{185}$Pt, $^{189}$Pt, and $^{191}$Pt in hcp-Co

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We report on measurements of quadrupole-interaction-resolved nuclear magnetic resonance on oriented nuclei (QI-NMR-ON) of $^{9/2}$+$^{185}$Pt ($I_{1/2}$=1.2 h), $3/2$+$^{189}$Pt ($I_{1/2}$=10.9 h), and $3/2$+$^{191}$Pt ($I_{1/2}$=2.9 d) in hcp-Co. Ratios of quadrupole moments were determined to be $Q(^{185}$Pt)/$Q(^{191}$Pt)=$-4.46(13)$ and $Q(^{189}$Pt)/$Q(^{191}$Pt)=$+1.235(8)$. [S0556-2813(98)01805-6]

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I. INTRODUCTION

The $^{77}$Ir, $^{79}$Pt, $^{79}$Au, and $^{80}$Hg nuclei lie in the transition region between the well deformed rare earth nuclei and the spherical nuclei near $^{208}$Pb. Depending on the neutron number, the nuclear shapes may vary between prolate deformation, oblate deformation, triaxial deformation and $\gamma$ softness. Most data on the ground-state properties are known from laser spectroscopy (LS) measurements [1]. For Hg, an oblate-prolate shape transition occurs between $^{187}$Hg and $^{185}$Hg [2]. The light even Hg isotopes, as well as the high-spin isomer $^{185m}$Hg, and those with $A \approx 186$ have a small oblate deformation, whereas the light odd isotopes $^{181}$Hg, $^{183}$Hg, and $^{185}$Hg are strongly deformed and prolate [3]. For Au, a similar behavior has been found: The isotopes with $A \approx 187$ have a small oblate deformation, whereas the isotopes with $A \approx 186$ are strongly deformed with a prolate shape [4–9]. For $^{184}$Au, shape coexistence was reported recently: The $I=5$ ground state is slightly more strongly deformed than the $I=2$ isomeric state [9]. A similar shape coexistence is also known for $^{186}$Ir: Here the $I=5$ ground state is also more strongly deformed than the $I=2$ isomeric state [10]. In this context, it is interesting whether a (sharp) nuclear shape transition exists also for Pt. The first direct evidence came from measurements of electrical quadrupole moments of neutron deficient radioactive Pt isotopes [11–13]: The quadrupole moments of $^{191}$Pt, $^{187}$Pt, and $^{185}$Pt — all isotopes have $I^u=3/2^+$ — are negative pointing to a predominantly oblate deformation, whereas the quadrupole moment of $^{185}$Pt ($I^u=9/2^+$) is large and positive, proving the existence of a large prolate deformation of $^{185}$Pt. Theoretical calculations yielded a smooth transition in the shape of odd-A Pt nuclei from a slightly deformed (nearly oblate) $^{195}$Pt via triaxial $^{193-187}$Pt to a strongly deformed (nearly prolate) $^{177}$Pt [13]. With the long-term aim of a better understanding of this transition, more accurate values for the quadrupole moments would be desirable. Therefore we performed measurements of quadrupole-interaction-resolved nuclear magnetic resonance on oriented nuclei (QI-NMR-ON) on $^{185}$Pt, $^{189}$Pt, and $^{191}$Pt. The results of these measurements are presented here.

II. QUADRUPOLE-INTERACTION RESOLVED NMR-ON WITH hcp-Co

In hcp-Co, in addition to the magnetic hyperfine field $B_{HF}$, an (axial symmetric) electric field gradient (EFG) $eQ$ exists, with the principal (z)-axis given by the c-axis of the crystal. The direction of spontaneous magnetization is also given by the crystal c-axis. For zero external magnetic field or if an external magnetic field $B_{ext}$ is applied parallel to the c-axis — this is the case for all measurements described in this paper — the magnetic and electric interactions are colinear. For spin I, the resonance signal is split into a set of 2I+1 equidistant subresonances. The center of the subresonance corresponding to rf transitions between state $|m\rangle$ and $|m+1\rangle$ is, on the condition that the $m=I$ state lies lowest in energy, given by

$$v_{m-m+1} = v_{mag} - \Delta v_Q(m+1/2),$$

$$v_{mag} = v_M + |g|\mu_B B_{ext} (1 + K) \text{ sgn}(B_{HF})/h,$$

$$v_M = |g|\mu_B B_{HF}/h,$$

$$v_Q = e^2 q Q/h,$$

$$\Delta v_Q = 3 v_Q/[2I(2I-1)],$$

where $v_M$ and $v_Q$ are the magnetic hyperfine splitting frequency and the quadrupole interaction frequency, $g$ and $eQ$ are the nuclear g-factor and the spectroscopic quadrupole moment, $B_{HF}$ and $eQ$ are the magnetic hyperfine field and the EFG, $B_{ext}$ and $K$ are the external magnetic field and the resonance shift parameter, sgn($B_{HF}$) is the sign of $B_{HF}$ with respect to $B_{ext}$, and $\Delta v_Q$ is the quadrupole subresonance separation. If $\Delta v_Q$ is larger than the inhomogeneous linewidth, this is fulfilled for our cases, a quadrupole-
subresonance-resolved spectrum can be measured and $\Delta \nu_Q$ is determined directly from the resonance centers of the subresonances.

The subresonance between the energetically lowest sublevels is denoted as $\nu_1$ resonance, the next $\nu_2$, etc. In low-temperature QI-NMR-ON experiments, the $\nu_1$ resonance has normally the largest amplitude and hence can be measured with the highest precision. The offset to the magnetic hyperfine splitting is given by

$$\nu_M - \nu_1 = (I - 1/2) \Delta \nu_Q.$$  \hspace{1cm} (6)

For $I = 3/2$ ($^{191}$Pt and $^{189}$Pt), the NMR-ON spectrum consists of 3 subresonances which are located at

$$\nu_1 = \nu_M - \nu_Q/2,$$

$$\nu_2 = \nu_M,$$

$$\nu_3 = \nu_M + \nu_Q/2.$$ \hspace{1cm} (7)

The $\nu_2$ resonance is difficult to observe because of its small amplitude. This is due to the fact that only transitions between the $m = -1/2$ and $+1/2$ sublevels are involved. Since the $\gamma$ anisotropy depends only on the sum of the sublevel population probabilities of the $-1/2$ and $+1/2$ substates, this is not accompanied by a direct change of the $\gamma$ anisotropy, and the indirect change of the $\gamma$ anisotropy via the rearrangement of the population probabilities of the $m = -3/2$ and $+3/2$ sublevels is small. The $\nu_3$ resonance is also difficult to observe because of the small sublevel population probability of the energetically higher sublevels.

For $I = 9/2$ ($^{189}$Pt), the complete NMR-ON spectrum consists of 9 subresonances. In this case the lowest subresonance ($\nu_1$) has the largest subresonance amplitude, too.

In cases, for which only the lowest subresonance can be observed, the quadrupole interaction can nevertheless be determined if the magnetic hyperfine interaction is known:

$$\nu_0 = 4I/3 (\nu_M - \nu_1).$$ \hspace{1cm} (8)

This is the case for $^{185}$Pt and $^{189}$Pt, for which the magnetic hyperfine splittings with respect to $^{191}$Pt are known experimentally from NMR-ON measurements on $^{189}$Pt, $^{191}$Pt, and $^{191}$Pt in Fe [14–16].

In the NMR-ON methods the resonant absorption of an applied radio frequency (rf) field is detected via nuclear radiation [17]. The angular distribution $W(\theta)$ and the anisotropy $A(\theta)$ of $\gamma$-rays emitted in the decay of oriented nuclei at the temperature $T$ is given by

$$A(\theta) = W(\theta) - 1 = \sum_{k \text{ even}} A_k B_k(a_m) P_k(\cos \theta) Q_k,$$ \hspace{1cm} (9)

where all parameters have their conventional meaning [18]. In the present work the ratio of count rates at $0^\circ$ and $90^\circ$

$$\epsilon = W(0^\circ)/W(90^\circ) - 1$$ \hspace{1cm} (10)

is analyzed, which is independent of the decreasing count rate due to the radioactive decay.

III. EXPERIMENTAL DETAILS

The samples were prepared with two different preparation techniques, mass-separator-implantation at ISOLDE/CERN and recoil-implantation at the cyclotron in Karlsruhe. The hcp-Co single crystal disks used for the implantations were prepared from a hcp-Co single crystal commercially available. Disks with a diameter of $\sim 10$ mm and a thickness of $\sim 0.5$ mm were spark cut from the crystal, the $c$-axis being oriented parallel to the plane of the disk. It should be mentioned that, especially for mass-separator implantation, the surface quality plays an essential role. (The implantation energy at ISOLDE/CERN is 60 keV.) Therefore the further treatment of the single-crystal disks consisted of many steps, which are described in detail in Ref. [19]: (i) Mechanical polishing with 15-µm, 9-µm, and 3-µm diamond emery paper; (ii) mechanical polishing with 1-µm, 0.5-µm, and 0.25-µm diamond paste. After each mechanical polishing step the crystal was cleaned ultrasonically. (iii) Electropolishing ($\sim 30$ min) in H$_2$PO$_4$ (85%) with an abrasion rate of $\sim 10$ µm/h. The final thickness of the disks used for mass-separator implantation and recoil implantation were $\sim 0.3$ mm and $\sim 7$ µm, respectively.

Mass-separator-implanted samples of $^{191}$PtCo$_{\text{(hcp)}}$, $^{189}$PtCo$_{\text{(hcp)}}$, and $^{185}$PtCo$_{\text{(hcp)}}$ were prepared at ISOLDE/CERN by implanting the radioactive precursor isotopes $^{191}$Hg, $^{189}$Hg, and $^{185}$Hg. Recoil-implanted samples of $^{189,191}$PtCo$_{\text{(hcp)}}$ were prepared at the cyclotron in Karlsruhe. A sandwich target consisting of a Cu foil (thickness 3 mg/cm$^2$) onto which Os (natural abundance) had been evaporated with a thickness of 1.5 mg/cm$^2$, followed by a hcp-Co single crystal (thickness 7 µm), was irradiated for 6 hours with 55-MeV $\alpha$-particles (beam current 2 µA). $^{189}$Pt and $^{191}$Pt are produced via the nuclear reaction $^{190,192}$Os($\alpha$,5$n$)$^{189,191}$Pt and recoil-implanted into hcp-Co.

In both cases, without any further heat treatment, the samples were soldered with GaIn to the Cu sample holder, loaded into a $^3$He–$^4$He-dilution refrigerator with a top-loading facility, and cooled to temperatures near 10 mK. The radio frequency was applied using a rf synthesizer. The experiments on the mass-separator implanted samples of $^{189}$Pt and $^{189}$Pt were performed at the NICOLE cryostat at CERN; the experiments on the recoil-implanted samples of $^{189,191}$Pt and the mass-separator implanted sample of $^{191}$Pt were performed at the cryostat in Munich.

IV. MEASUREMENTS AND RESULTS

Figure 1 shows the QI-NMR-ON spectrum of the mass-separator-implanted $^{191}$PtCo$_{\text{(hcp)}}$ sample measured for $B_{\text{ext}} = 6$ kG. The three quadrupole subresonances are perfectly resolved. By taking into account the results of additional measurements, the final results are

$$\nu_M = 235.18(6) \text{ MHz},$$

$$\nu_Q = +27.10(4) \text{ MHz}.$$
nique for the Co single crystals. This can be seen from the fact that the inhomogeneous line width in the present QI-
NMR-ON spectrum is by a factor of \(\frac{20}{20}\) smaller than that of Ref. [20].

For the mass-separator-implanted \(^{189}\text{Pt}^{191}\text{Co}(hcp)\) sample only the \(n_1\) resonance could be measured, because of difficulties with the rf equipment of NICOLE. It is shown in Fig. 2. The result is \(n_1 = 187.389(9)\) MHz. The magnetic hyper-
fine interaction of \(^{189}\text{Pt}^{191}\text{Co}(hcp)\) can be predicted as the ratio of \(g\) factors for \(^{189}\text{Pt}\) and \(^{191}\text{Pt}\) can be derived from NMR-ON measurements of \(^{189}\text{Pt}\) and \(^{191}\text{Pt}\) in Fe. Eder et al. [14] and Ohya et al. [15,16] determined the ratio \(\nu_M^{(189}\text{Pt})/\nu_M^{(191}\text{Pt})\) to be 0.8680(3) and 0.8679(2). Thus we get \(\nu_M^{(189}\text{Pt}^{191}\text{Co}(hcp)) = 204.11(9)\) MHz. According to Eq. (8) we get \(\nu_Q^{(189}\text{Pt}^{191}\text{Co}(hcp)) = +33.44(20)\) MHz. Thus the ratio of quadrupole splittings and hence the ratio of quadrupole moments of \(^{189}\text{Pt}\) and \(^{191}\text{Pt}\) is deduced to be \(Q^{(189}\text{Pt})/Q^{(191}\text{Pt}) = +1.234(8)\). This is very different from \(Q^{(189}\text{Pt})/Q^{(191}\text{Pt}) = +1.05(7)\) quoted by Duong et al. as result from LS measurements [11].

Because of this large discrepancy, another experiment was performed, in which recoil-implanted \(^{189}\text{Pt}\) and \(^{191}\text{Pt}\) were measured in the same sample. In this experiment the \(n_2\) resonance of \(^{189}\text{Pt}\) could be measured in addition to \(n_1\). It is shown in Fig. 3. The results are \(n_1(B_{\text{ext}}=6 \text{ kG}) = 186.18(11)\) MHz and \(n_2(B_{\text{ext}}=6 \text{ kG}) = 203.13(43)\) MHz. The extrapolation to zero external magnetic field yields \(n_1(B_{\text{ext}}=0 \text{ kG}) = 187.50(11)\) MHz and \(n_2(B_{\text{ext}}=0 \text{ kG}) = 204.45(43)\) MHz. Within the experimental error, \(n_1\) is in perfect agreement with the measurement on the mass-separator-implanted sample, and \(n_M\) is in perfect agreement with the value calculated by taking into account the ratio of \(g\) factors determined from NMR-ON of \(^{189,191}\text{Pt}\). Thus, we get the independent additional result of \(\nu_Q^{(189}\text{Pt}^{191}\text{Co}(hcp)) = +33.90(89)\) MHz. The average value of both measurements is

\[
\nu_Q^{(189}\text{Pt}^{191}\text{Co}(hcp)) = +33.46(20)\ \text{MHz}
\]

Thus, the final value for the ratio of quadrupole moments of \(^{189}\text{Pt}\) and \(^{191}\text{Pt}\) is deduced to be

\[
Q^{(189}\text{Pt})/Q^{(191}\text{Pt}) = +1.235(8)\]

It should also be stated that for the recoil-implanted sample the quadrupole splitting for \(^{191}\text{Pt}^{191}\text{Co}(hcp)\) was measured to be \(+26.88(40)\) MHz, in perfect agreement with the more precise result for the mass-separator implanted sample quoted before.

The experiment on \(^{185}\text{Pt}\) was difficult because of the short half-life of 1.2 h. Here only the \(n_1\) resonance was searched, for which the largest resonance effect is expected. For both the 461-and 641-keV transitions, a significant resonance signal was observed as shown in Fig. 4. The result from both

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FIG. 1. QI-NMR-ON spectrum of the 538 keV transition of mass-separator-implanted \(^{191}\text{Pt}^{191}\text{Co}(hcp)\) measured for \(B_{\text{ext}}=6.0 \text{ kG}\).

FIG. 2. \(n_1\) resonance of the 545 keV transition of mass-separator-implanted \(^{191}\text{Pt}^{191}\text{Co}(hcp)\) measured for \(B_{\text{ext}}=0.5 \text{ kG}\).

FIG. 3. \(n_1\) and \(n_2\) resonance of the 545 keV transition of recoil-implanted \(^{189}\text{Pt}^{191}\text{Co}(hcp)\) measured for \(B_{\text{ext}}=6.0 \text{ kG}\).

FIG. 4. \(n_1\) resonance (sum of 461 and 641 keV transition) of mass-separator-implanted \(^{189}\text{Pt}^{191}\text{Co}(hcp)\) measured for \(B_{\text{ext}}=0.5 \text{ kG}\).
transitions is $\nu_1 = 140.86(28) \text{ MHz}$. Here again, the quadrupole splitting is obtained by taking into account the magnetic hyperfine-splitting frequency of $^{185}\text{Pt}^{\frac{3}{2}}\text{Fe}$, which is derived as follows: The NICOLE Collaboration observed NMR-ON on $^{185}\text{Pt}^{\frac{3}{2}}\text{Fe}$, with a resonance center of $\bar{\nu} = 164.9(2) \text{ MHz}$ (extrapolated to $B_{\text{ext}} = 0 \text{ kG}$) [12,21]. Here the quadrupole splitting is unresolved because of the high spin $I = \frac{3}{2}$; it causes, however, an offset of the resonance center with respect to the pure magnetic hyperfine splitting. This offset is estimated to be $0.5(5) \text{ MHz}$ by taking into account the known quadrupole splitting of $^{191}\text{Pt}^{\frac{3}{2}}\text{Fe}$ [22] and the ratio of quadrupole moments $Q(^{185}\text{Pt})/Q(^{191}\text{Pt})$ as determined in this work. Taking now $\nu_{\text{ext}}(^{185}\text{Pt}^{\frac{3}{2}}\text{Fe}) = 164.4(7) \text{ MHz}$, we calculate $\nu_{\text{m}}(^{185}\text{Pt}^{\frac{3}{2}}\text{Fe}^{\text{hbp}}) = 120.7(5) \text{ MHz}$. According to Eq. (8) we get

$$\nu_{\text{m}}(^{185}\text{Pt}^{\frac{3}{2}}\text{Fe}^{\text{hbp}}) = -120.8(3.5) \text{ MHz},$$

and hence

$$Q(^{185}\text{Pt})/Q(^{191}\text{Pt}) = -4.46(13).$$

Here we want to mention that an uncertainty exists in the assignment of the observed resonance to $\nu_1$. Because of the short half-life of $^{185}\text{Pt}$ and the limited beam time we could not perform an additional dedicated experiment on that subject. If the observed resonance would have to be assigned to $\nu_2$, the ratio of quadrupole moments would be $Q(^{185}\text{Pt})/Q(^{191}\text{Pt}) = 5.94(17)$. 

### V. DISCUSSION

Our ratios of spectroscopic quadrupole moments $Q_s/Q_s(^{191}\text{Pt})$ are listed in column 3 of Table I, together with experimental results from nuclear orientation (column 4), laser spectroscopy of Duong et al. (column 5), nuclear orientation of Hilberath et al. (column 6), and theoretical calculations within a particle-triaxial rotor model (column 7). The change of deformation parameters between $^{191}\text{Pt}$ and $^{189}\text{Pt}$ can be obtained from the ratio $Q_s(^{191}\text{Pt})/Q_s(^{189}\text{Pt})$. The LS result of Hilberath et al. (Ref. [13]) and the NO result of Eder et al. (Ref. [12]) do not allow one to draw any conclusion because of the large experimental uncertainties. The LS result of Duong et al. (Ref. [11]) is more accurate, $1.05(7)$, but an increase of the quadrupole moment can also not be deduced unambiguously as $1.0$ is also included within their experimental uncertainty. These LS data are also consistent with a moderately small increase in the quadrupole moment. Theoretical calculations yield $1.04$–$1.08$ for this ratio [13], which was considered to describe the experimental result well. Our experiments have shown, however, that this ratio is $+1.235(8)$, i.e., that the change of deformation parameters, mainly $\gamma$, between $^{191}\text{Pt}$ and $^{189}\text{Pt}$ is much stronger than expected theoretically. The fact that the theoretical calculations of Ref. [13] describe the known experimental magnetic moments well cannot be interpreted that the nuclear deformation is described properly. This is due to the fact that, for the case of not too large deformations, the influence of Coriolis mixing on the magnetic moment cannot be calculated with sufficient accuracy. Recent measurements of magnetic moments of the first $2^+$ excited states in even Pt isotopes showed that these are remarkably constant between $A = 184$ and $198$ despite the undoubted shape change in this region [23].

The value of Duong et al. [11] for $Q_s(^{189}\text{Pt})/Q_s(^{191}\text{Pt})$ was too small is probably due to an incorrect decomposition of the magnetic and quadrupole contributions in the LS spectrum. This is supported by the fact that the magnetic moment of $^{180}\text{Pt}$ given by Duong et al. [11] is also at variance with the magnetic moments derived from NMR-ON [14] and LS of Ref. [13] (see Table II).

### TABLE I. Experimental and theoretical ratios of spectroscopic quadrupole moments $Q_s/Q_s(^{191}\text{Pt})$.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$I$</th>
<th>$Q_s/Q_s(^{191}\text{Pt})$</th>
<th>NO</th>
<th>LS</th>
<th>LS</th>
<th>Theory</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{185}\text{Pt}$</td>
<td>$\frac{3}{2}^+$</td>
<td>$-4.46(13)$</td>
<td>$-5.2(1.5)$</td>
<td>$-4.4(6)$</td>
<td>$-4.5$</td>
<td>$-5.3$</td>
</tr>
<tr>
<td>$^{187}\text{Pt}$</td>
<td>$\frac{3}{2}^-$</td>
<td>$+1.4(4)$</td>
<td>$+1.15(8)$</td>
<td>$1.3(2)$</td>
<td>$+1.17$</td>
<td>$+1.31$</td>
</tr>
<tr>
<td>$^{189}\text{Pt}$</td>
<td>$\frac{3}{2}^-$</td>
<td>$+1.235(8)$</td>
<td>$+1.3(3)$</td>
<td>$1.05(7)$</td>
<td>$+1.04$</td>
<td>$+1.08$</td>
</tr>
</tbody>
</table>

aQuadrupole-interaction-resolved NMR-ON: this work.
bNuclear orientation: Refs. [11,14,22].
dLaser spectroscopy: Ref. [13].

### TABLE II. Magnetic moments of Pt isotopes.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$I$</th>
<th>NMR-ON</th>
<th>$\mu_\text{m}$</th>
<th>LS</th>
<th>LS</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{185}\text{Pt}$</td>
<td>$\frac{3}{2}^+$</td>
<td>$0.774(14)$</td>
<td>$-0.83(1)$</td>
<td>$^{12,14,22}$</td>
<td></td>
</tr>
<tr>
<td>$^{187}\text{Pt}$</td>
<td>$\frac{3}{2}^-$</td>
<td>$0.408(8)$</td>
<td>$-0.397(5)$</td>
<td>$-0.427(20)$</td>
<td></td>
</tr>
<tr>
<td>$^{189}\text{Pt}$</td>
<td>$\frac{3}{2}^-$</td>
<td>$0.434(9)$</td>
<td>$0.427(9)$</td>
<td>$-0.421(5)$</td>
<td>$-0.440(8)$</td>
</tr>
<tr>
<td>$^{191}\text{Pt}$</td>
<td>$\frac{3}{2}^-$</td>
<td>$0.500(10)$</td>
<td>$0.492(10)$</td>
<td>$-0.501(5)$</td>
<td>$-0.494(8)$</td>
</tr>
</tbody>
</table>

aNMR on oriented nuclei: Refs. [12,14,22].
bNMR on oriented nuclei: Ref. [15].
dLaser spectroscopy: Ref. [13].
For $^{185}$Pt, there is a discrepancy for the experimental magnetic moments (see Table II). The ratios of quadrupole moments (Table I), however, are consistent. Here the theoretical ratio $Q_x^{(185)}Pt/Q_x^{(191)}Pt = -4.5 \ldots -5.3$ is in agreement with our experimental ratio of $-4.46(13)$.

In summary, we have measured precise ratios of quadrupole moments of Pt isotopes. Our results can serve for a better theoretical understanding of the change of nuclear shapes in the Ir-Pt-Au-Hg transition region.

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