

Measurement of the Spectroscopic Quadrupole Moment of ^{186}Au : Experimental Verification of the Large Prolate Deformation of the ^{186}Au Ground State

B. Hinfurtner, E. Hagn, and E. Zech

Physik Department, Technische Universität München, D-8046 Garching, Federal Republic of Germany

R. Eder

PPE Division, CERN, CH-1211 Geneva 23, Switzerland

NICOLE Collaboration and ISOLDE Collaboration

CERN, CH-1211 Geneva 23, Switzerland

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We report the first on-line measurements of quadrupole-interaction-resolved nuclear magnetic resonance on oriented nuclei. The 10-min ^{186}Au activity was obtained as daughter after cold implantation of mass-separated ^{186}Hg into a hcp Co single crystal at the NICOLE facility at ISOLDE-3 (CERN). The quadrupole interaction of $^{186}\text{AuCo}(\text{hcp})$ was fully resolved, and the spectroscopic quadrupole moment of ^{186}Au was determined to be $Q = +3.12(20)$ b. This implies $\beta_2 = +0.246(16)$, proving the large prolate deformation of the ^{186}Au ground state.

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From optical isotope-shift measurements of very light gold isotopes with laser spectroscopy, a drastic change of the nuclear charge radius was observed recently between ^{187}Au and ^{186}Au , which has been interpreted as an onset of strong prolate deformation of $\beta_2 \approx 0.25$ in ^{186}Au and ^{185}Au [1]. Although the interpretation of the large $\delta\langle r^2 \rangle$ has been very convincing, it should be kept in mind that neither the sign nor the *absolute* value of the deformation can be inferred from those experiments. Thus, independent information would be valuable, e.g., from a direct measurement of the spectroscopic ground-state quadrupole moment. Recently, Le Blanc *et al.* reported a laser-spectroscopy measurement of the ground-state quadrupole moment of ^{186}Au , $Q = +2.69(8)$ b, from which the deformation parameter $\beta_2 = 0.21$ can be calculated within the rotational model, which is significantly smaller than the optical isotope-shift result [2]. In addition, they reported $Q(^{191}\text{Au}) = -1.1(1)$ b. However, (i) it cannot be anticipated that the Sternheimer effect has been taken into account correctly, and (ii) the negative quadrupole moment of ^{191}Au would be very hard to understand.

Thus, in addition to the well established laser technique, an independent technique would be desirable, with which the magnitude *and* sign of electric quadrupole moments of nuclei far off stability can be determined unambiguously. Here we report the first successful application of such a new technique: quadrupole-interaction-resolved NMR on oriented nuclei (QI-NMR-ON) after on-line cold implantation into a hcp Co single crystal. The basis for this new technique had been the observation that the quadrupole substructure of ^{198}Au ($I^\pi = 2^-$) and ^{199}Au ($I^\pi = \frac{3}{2}^+$) in hcp Co could be well resolved [3]. In this pilot experiment, the Au isotopes had been implanted with the rather high implantation voltage of 350 kV available at the mass separator at Konstanz. Thus, before being able to proceed to on-line experiments, two to-

pics had to be solved. (i) It had to be shown that it is possible to improve the surface quality of the hcp Co single crystals enough that the quadrupole substructure can be resolved despite the relatively small implantation voltage of 60 kV available at the on-line mass separator ISOLDE-3 at CERN. (ii) It had to be shown that the quadrupole splitting can also be detected after cold implantation.

Both problems have been solved. (i) By a sophisticated combination of different polishing steps the surface quality of hcp Co single crystals could be improved enough that no deterioration of the NMR-ON linewidths at 60-kV implantation voltage (in comparison to 350 kV) was observed [4]. (ii) After cold implantation of ^{198}Au in hcp Co exactly the same quadrupole splitting was obtained as already known from warm implantation.

For the case of a combined magnetic dipole plus electric quadrupole interaction, both being collinear, the sub-level energies E_m of a nuclear state with spin I are given by

$$E_m = -h\nu_M m + h\nu_Q [3m^2 - I(I+1)]/4I(2I-1). \quad (1)$$

Here ν_M and ν_Q are the magnetic and electric hyperfine-splitting frequencies defined as

$$\nu_M = |g\mu_N B_{\text{hf}}/h|, \quad \nu_Q = e^2 q Q/h, \quad (2)$$

where g and eQ are the nuclear g factor and the spectroscopic quadrupole moment, and B_{hf} and eq are the magnetic hyperfine field and the electric-field gradient (EFG), respectively. The condition for nuclear magnetic resonance is fulfilled for a set of $2I$ subresonances: Assuming that the $m=I$ state lies lowest in energy, the center of the subresonance corresponding to rf transitions between states $|m\rangle$ and $|m+1\rangle$ is given by

$$\nu_{m \rightarrow m+1} = \nu_M - \Delta\nu_Q(m + \frac{1}{2}), \quad (3)$$

where Δv_Q is the subresonance separation defined as

$$\Delta v_Q = 3v_Q/2I(2I-1). \quad (4)$$

The subresonance between the energetically lowest sub-levels is denoted as the v_1 resonance, the next as v_2 , etc. The v_1 resonance has the largest amplitude and can hence be measured with the highest precision. The offset to the magnetic hyperfine splitting is given by

$$v_M - v_1 = (I - \frac{1}{2})\Delta v_Q. \quad (5)$$

In the NMR-ON method, the resonance absorption is detected via the change of the angular distribution of the emitted radiation. The angular distribution $W(\theta)$ and the anisotropy $A(\theta)$ of γ rays emitted in the decay of oriented nuclei at the temperature T are given by

$$A(\theta) = W(\theta) - 1 \\ = \sum_{k=2,4} A_k B_k(v_M, v_Q, T) P_k(\cos\theta) Q_k. \quad (6)$$

The parameters A_k are products of the normally used angular correlation coefficients U_k and F_k which depend on the spins and the multiplicities of the decay cascade. The $P_k(\cos\theta)$ are Legendre polynomials, θ being the angle between the quantization axis (here the c axis of the hcp Co single crystal) and the direction of observation, and Q_k are solid-angle-correction coefficients. The B_k describe the degree of orientation; they depend on v_M , v_Q , and T . For hcp Co, $v_M \gg |v_Q|$, the degree of orientation is mainly fixed by the ratio $h v_M/k_B T$ and is nearly independent of v_Q .

Samples were prepared from a hcp Co single crystal available commercially. Disks with a diameter of ≈ 10 mm and a thickness of ≈ 0.2 mm were spark cut from the crystal, the c axis being oriented parallel to the plane of the disk. The further treatment consisted of many steps: (i) mechanical polishing with 15-, 9-, and 3- μ m diamond emery paper; (ii) mechanical polishing with 1-,

0.5-, and 0.25- μ m diamond paste, after each mechanical polishing step the crystal was cleaned ultrasonically and polished chemically; (iii) electropolishing (~ 30 min) in H_3PO_4 (85%) with an abrasion rate of $\approx 1 \mu\text{m/h}$.

The pilot cold-implantation experiment with ^{198}Au in hcp Co was performed in the following way: A 100- $\mu\text{g}/\text{cm}^2$ Au layer was evaporated onto a 3- mg/cm^2 Nb foil. Four NbAu samples ($2 \times 4 \text{ mm}^2$) were irradiated for 6 days with thermal neutrons ($\phi = 8 \times 10^{13} \text{ n/cm}^2\text{s}$) in the Munich research reactor FRM. The activated samples were mounted in an empty target holder of the mass separator ISOLDE-3 at CERN, and ^{198}Au was cold implanted into a hcp Co single crystal kept at temperatures below 100 mK in the NICOLE ^3He - ^4He dilution refrigerator. The NMR-ON spectrum is shown in Fig. 1. The linewidth is $\Gamma = 1.5(1)$ MHz, i.e., a factor ≈ 3 larger than obtained with 350-kV warm implantation in a single crystal with a comparable surface quality [5]. However, despite the larger linewidth, the quadrupole splitting [$v_Q = -15.25(20)$ MHz] is in perfect agreement with the warm-implantation data [$-14.92(20)$ MHz [3], $-15.12(5)$ MHz [5], and $-15.4(5)$ MHz [6]], proving the applicability of the method for on-line measurements.

The main experiment on ^{186}Au was performed as follows: ^{186}Hg ($T_{1/2} = 1.4$ min), obtained from a $\text{Pb}(p, xpyn)$ spallation reaction using a liquid Pb target, was implanted continuously at a rate of 2×10^6 atoms/s into a hcp Co single crystal kept at temperatures around 10 mK in the NICOLE ^3He - ^4He dilution refrigerator. ^{186}Au ($T_{1/2} = 10$ min) is produced *in situ* by the decay of ^{186}Hg . As the spin-lattice relaxation time is much shorter than the half-life, ^{186}Au becomes oriented to a large degree before it decays to levels in ^{186}Pt . Thus, the strong γ rays with $E = 192$ and 299 keV are emitted anisotropically and can thus be used as a detector for NMR. Figure 2 shows a NMR-ON spectrum of the

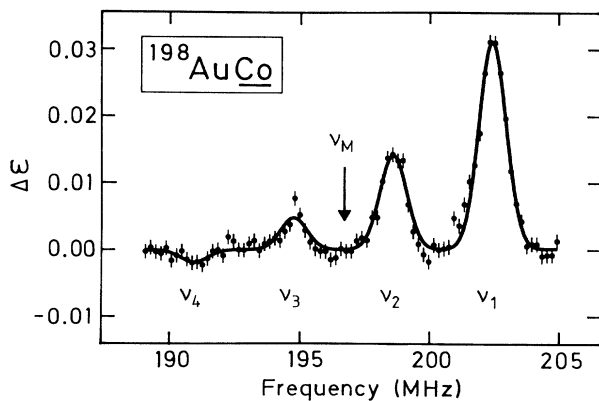


FIG. 1. NMR-ON resonances of the 412-keV transition of ^{198}Au in hcp Co measured with frequency steps 0.2 MHz, modulation bandwidth ± 0.25 MHz, and total counting time 10 h. [$\varepsilon = W(0^\circ)/W(90^\circ) - 1$].

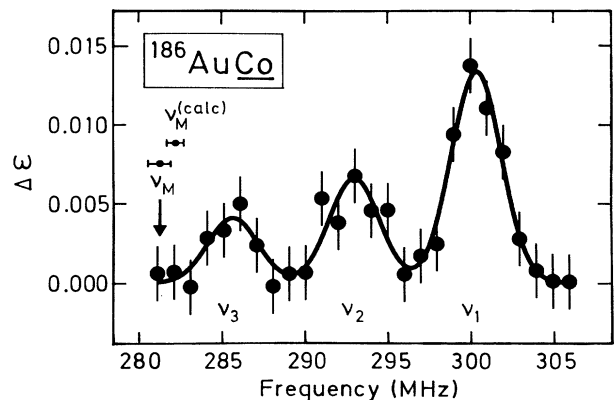
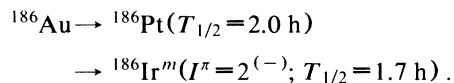


FIG. 2. NMR-ON resonances of the 192-keV transition of ^{186}Au in hcp Co measured with frequency steps 1 MHz, modulation bandwidth ± 1 MHz, and total counting time 6 h. The magnetic hyperfine-splitting frequency v_M is marked: top, calculated as described in text; bottom, experimental.

192-keV transition. The lowest three subresonances are well resolved, which is sufficient for the determination of ν_Q and ν_M . The results are

$$\nu_Q = -74.1(3.1) \text{ MHz}, \quad \nu_M = 281.6(7) \text{ MHz}.$$

The following facts deserve to be mentioned. (i) The absolute values for the resonance amplitudes are rather small. The reduction with respect to the expectation according to the experimental γ anisotropy is estimated to be 0.35(15). The relative amplitudes of the three observed subresonances $\nu_{1,2,3}$ follow well the decreasing trend expected according to a Boltzmann distribution of the sublevel populations. (ii) The magnetic hyperfine-splitting frequency, which according to Eq. (5) is separated (for $I=3$) from the lowest subresonance by $\nu_M - \nu_1 = \frac{5}{2} \Delta\nu_Q$, is in good agreement with the expectation: Taking into account the hyperfine-splitting frequencies of ^{198}Au and ^{186}Au in Fe, $\nu_M(^{198}\text{AuFe}) = 259.5(1)$ MHz [7], $\nu_M(^{186}\text{AuFe}) = 372.0(6)$ MHz [8], and $\nu_M(^{198}\text{AuCo}) = 196.86(4)$ MHz [5], we calculate $\nu_M(^{186}\text{AuCo}) = 282.2(5)$ MHz. This proves that the three equidistant resonances originate from a quadrupole splitting and that the spin assignment $I=3$ is correct. (iii) During the on-line measurement, $^{186}\text{Ir}^m$ was accumulated via the decay



After the on-line measurement, the NMR-ON resonance of $^{186}\text{Ir}^m\text{Co}$ was investigated. The result for the ν_1 resonance is $\nu_1 = 277.57(9)$ MHz, in perfect agreement with the respective result after warm implantation, $\nu_1 = 277.66(2)$ MHz [9]. (Because of $I=2$ and an unfavorable A_4/A_2 ratio only the ν_1 resonance has a reasonable amplitude for $^{186}\text{Ir}^m\text{Co}$.) However, the resonance amplitude of the $^{186}\text{Ir}^m\text{Co}$ ν_1 resonance after cold implantation was reduced to 0.25(8) of the amplitude observed after warm implantation with the same experimental conditions. Taking this observation into account we can understand the small absolute amplitudes for $^{186}\text{AuCo}$: Either Hg is substituted with a smaller fraction to the regular lattice sites in Co—with the cold implantation of ^{198}Au no significant decrease of the resonance amplitudes had been observed—or, if Hg is initially implanted with a large fraction onto regular lattice sites, the binding of Hg onto these sites is so weak that the high-energy β decay may induce a site change. From the perfect agreement of the ν_1 resonance center for $^{186}\text{Ir}^m$ —despite the small amplitude—we conclude that only the nuclei on regular lattice sites contribute to the observed resonance signals and that the observed quadrupole-splitting frequency can be ascribed to the (small fraction of) nuclei on regular lattice sites.

The presently known quadrupole-splitting frequencies of Au isotopes in hcp Co are listed in Table I. For the

TABLE I. Quadrupole moments of Au isotopes and quadrupole-splitting frequencies in hcp Co.

Isotope	I^π	ν_Q (MHz)	Q (b)	Reference
^{186}Au	3^-	-74.1(3.1)	+3.12(20)	a
^{191}Au	$\frac{3}{2}^+$		+0.716(21)	[10]
^{193}Au	$\frac{3}{2}^+$		+0.664(20)	[10]
^{195}Au	$\frac{3}{2}^+$	-14.35(2)	+0.604(22) ^b	[9]
^{197}Au	$\frac{3}{2}^+$		+0.547(16)	[11]
^{198}Au	2^-	-15.12(5)	+0.637(24)	[3,5],a
^{199}Au	$\frac{3}{2}^+$	-12.06(10)	+0.508(20)	[3]

^aThis work.

^bLinear interpolation between $^{191,193,197}\text{Au}$; see text.

derivation of absolute values for the quadrupole moments the electric-field gradient of Au in hcp Co has to be known. It is obtained in the following way: The hyperfine constants $B(^2D_{3/2})$, which are proportional to the spectroscopic quadrupole moments, for $I^\pi = \frac{3}{2}^+$ $^{191,193,197}\text{Au}$ are experimentally known to be -1192.4(6.2), -1106.5(4.6), and -911.21(72), respectively [10], indicating a nearly perfect linear dependence on A . With the quadrupole moment $Q(^{197}\text{Au}) = +0.547(16)$ b [11], the quadrupole moments of $^{191,193}\text{Au}$ are obtained [10] as listed in Table I, and the quadrupole moment of ^{195}Au is predicted to be $Q(^{195}\text{Au}) = +0.604(22)$ b. With the quadrupole splitting $\nu_Q(^{195}\text{AuCo}) = -14.35(2)$ MHz [9], we find $eq(\text{AuCo}) = -0.982(36) \times 10^{17}$ V/cm². The quadrupole moment of ^{186}Au is then deduced to be

$$Q(^{186}\text{Au}) = +3.12(20) \text{ b},$$

which is significantly larger than the laser-spectroscopy result [2].

The configuration of ^{186}Au is believed to be $\{\pi \frac{3}{2}^-\} [532] \otimes \nu \frac{9}{2}^+ [624] \}_{3^-}$, i.e., $K=I$ [12]. The deformation parameter β_2 can be calculated using the rotational-model relationship

$$\beta_2 = \frac{\sqrt{5}\pi}{3ZR_0^2} \frac{(I+1)(2I+3)}{3K^2 - I(I+1)} Q. \quad (7)$$

Taking $I=K=3$ and $R_0 = 1.25A^{1/3}$ fm, the deformation parameter for ^{186}Au is found to be $\beta_2 = +0.246(16)$, the absolute value being in perfect agreement with $|\beta_2| = 0.246$ [13], and $|\beta_2| = 0.252$ [14], deduced from laser-spectroscopy isotope-shift measurements.

The EFG as derived here differs slightly from $eq(\text{AuCo}) = -0.90(4) \times 10^{17}$ V/cm² derived via the chain $\nu_Q(^{197}\text{AuFe})$, $\nu_Q(^{199}\text{AuFe})$, and $\nu_Q(^{199}\text{AuCo})$ [3]. In this derivation there is one step which has an inherent uncertainty: the ratio of the quadrupole splittings of ^{197}Au and ^{199}Au in Fe, as these have been measured with different techniques, namely, NMR for stable ^{197}Au and NMR-ON for radioactive ^{199}Au . Although there is no evidence in the literature that these techniques have

different response functions for nuclei on nonequivalent lattice sites, as resulting, e.g., from different spin-lattice relaxation times, such an effect cannot be excluded *a priori*. It should be noted that the quadrupole moment of ^{199}Au as derived with $eq(\text{AuCo}) = -0.982(36) \times 10^{17}$ V/cm² (see Table I) follows also the linearly decreasing trend of Q vs A , supporting the derivation of the EFG as presented here.

In conclusion, we have shown the following. (i) QI-NMR-ON is a new method which allows the on-line measurement of electric quadrupole moments of radioactive nuclei far off stability. (ii) The resonance amplitudes after direct cold implantation of Au are larger than the amplitudes obtained if a Hg precursor is implanted. This indicates either a large degree of nonsubstitutional implantation of Hg or, as supported by other experiments [9], a β -decay-induced lattice-site change. (iii) The spectroscopic quadrupole moment of ^{186}Au determined in this work proves the large prolate deformation of ^{186}Au .

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