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PROPOSAL TO THE ISOLDE COMMITTEE

MEASUREMENTS OF ELECTRIC QUADRUPOLE MOMENTS OF
NEUTRON-DEFICIENT Au, Pt, AND Ir NUCLEI WITH NMR-ON IN
hcp-Co

CERN¹ - Munich² - NICOLE Collaboration

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Summary:

We propose on-line measurements with the method of quadrupole-interaction-tuned NMR on oriented nuclei ("QIT-NMR-ON") developed recently in Munich. During the last two years we have shown with experiments at ISOLDE/NICOLE [1]:

- 1) After cold-implantation of ^{198}Au the quadrupole structure was fully resolved, the results being in perfect agreement with the warm-implantation results.
- 2) The first on-line experiment was successful: The quadrupole moment of ^{186}Au was determined; our result is larger than that known from Laser-spectroscopy.
- 3) The quadrupole splittings of $^{185,189,191}\text{Pt}$ could be resolved well. In comparison to published Laser-spectroscopy data, i) the values from QI-NMR-ON are more precise and ii) there is a clear discrepancy for ^{189}Pt . The quadrupole splittings of $^{184,186,186m,188}\text{Ir}$ could also be resolved well, yielding highly precise electric quadrupole moments.
- 4) The dependence of the hyperfine field and the electric field gradient on the angle between the magnetization and the single-crystal c-axis was investigated for Lu, Ir, Pt and Au. For Iridium a strong anomaly was found.

We propose to continue these experiments with the following main aims:

- 1) On-line measurements of nuclear magnetic moments and electric quadrupole moments of isotopes far off stability in the region Os-Ir-Pt-Au. Especially, the quadrupole moments are very interesting in this region as they are related directly to the deformation. In addition, with QIT-NMR-ON spins can also be determined unambiguously via resonance-frequency measurements.
- 2) For the understanding of the magnetism in Co: Study of the anisotropy of hyperfine fields in hcp-Co.
- 3) Study of the electric field gradients in hcp-Co, especially for the understanding of the Ir-anomaly.
- 4) Study of the strong, resonance-like magnetization-dependence of the spin-lattice relaxation matrix element. This feature could have an important application for pulsed-beam nuclear orientation studies far off stability, as it is possible to "tune" the spin-lattice relaxation time.

We ask for 20 shifts Hg (Au) for the on-line experiments.

The investigations of items 2) - 4) can be performed parasitically with (short) off-line implantations (typical 10 min ... 1 h). For these experiments a total beam-time corresponding to 2 - 3 shifts would be required.

All our experiments can be performed with the GPS.

1. Introduction

From Laser-spectroscopy measurements it is known that the nuclear charge radius changes drastically between ^{187}Au and ^{186}Au and that the isotope shift data indicate an onset of strong deformation of $\beta \sim 0.25$ in ^{186}Au and ^{185}Au [2]. This can be understood qualitatively as the result of the $\pi h_{9/2}$ intruder configuration being the ground state proton configuration in both ^{185}Au and ^{186}Au [2], with some analogy to ^{184}Ir , ^{185}Ir and ^{186}Ir , for which the $\pi h_{9/2}$ intruder configuration is known to be the ground-state proton configuration, too [3,4]. The consequence of this proton configuration is the dominance of the $K = 1/2$ component in the ground-state wave function in the odd-A isotopes. For the even-A isotopes strong Coriolis mixing occurs if the proton is in the $1/2^- [541]$ state. (In the case of $I = 5$ ^{184}Ir , the main components in the ground state wave function have $K = 4$ and 5, with a possible $K = 3$ admixture, in the case of $I = 5$ ^{186}Ir , the main components are $K = 1$ and 0 [3,4].) For well-deformed nuclei, in the frame of the rotational model, the K -quantum number enters into the relation between the intrinsic quadrupole moment Q_0 and the spectroscopic quadrupole moment Q . Thus, a measurement of Q yields, together with Q_0 , the (effective) K -quantum number and hence information on the components in the ground state wave functions, or, if there is no doubt on the ground state wave function, the ground state deformation β_2 .

In the last two years we have established the new QIT-NMR-ON technique which allows the determination of magnetic moments and electric quadrupole moments with resonance precision. We have applied this method successfully, both off-line and on-line: i) With off-line measurements, the quadrupole splittings of $^{184,186,186m,188}\text{Ir}$, $^{185,189,191}\text{Pt}$, $^{195,198,199}\text{Au}$ and $^{169,171,172,173,177,177m}\text{Lu}$ were determined with high precision. The samples for these experiments had been prepared to a large extent parasitically at ISOLDE (typical implantation time 10 min ... 1 h).

ii) With an on-line measurement on ^{186}Au after implantation of ^{186}Hg the interesting quadrupole moment of ^{186}Au could be determined [5], the absolute value being larger than the literature value obtained with direct Laser spectroscopy [6].

Thus we propose to continue these experiments on the more neutron deficient Au, Pt and Ir isotopes. We would start with $^{184,184m}\text{Au}$, for which, in addition to the determination of the magnetic moments and electric quadrupole moments, unique spin assignments are expected to be possible with QIT-NMR-ON. Then we would proceed successively to the more neutron-deficient A -chains.

2. Essential features of quadrupole-interaction-tuned NMR-ON in hcp-Co

For nuclear-orientation measurements of quadrupole moments with resonance precision, a non-cubic ferromagnetic matrix is necessary, in which a (large) electric field gradient (EFG) is present in addition to a large magnetic hyperfine field. With a series of experiments we have shown that hcp-Co fulfills this condition in an ideal way, with the prospects of a very general applicability:

1) With mass-separator implantations of ^{198}Au and ^{199}Au (at Konstanz) we have demonstrated that hcp-Co is well suited as matrix for nuclear magnetic resonance on oriented nuclei (NMR-ON) and that the quadrupole interaction could be resolved well for both isotopes [7,8].

2) With a $^{191}\text{PtCo}(\text{hcp})$ sample prepared at ISOLDE by implanting ^{191}Hg in a hcp-Co single crystal, NMR-ON measurements were performed for the "standard" 0° -geometry (magnetization M parallel to the c -axis) and, for the first time, the 90° -geometry (M perpendicular to the c -axis). (In order to achieve $M \perp c$, an external magnetic field $B_0 > 13.2$ kG is necessary.) The quadrupole interaction was fully resolved for both cases, $M \parallel c$ and $M \perp c$. The quadrupole interaction for $M \perp c$ was $-1/2$ compared to the case $M \parallel c$, as expected according to the $P_2(\cos\theta)$ dependence (Fig. 1).

Completely unexpected, it was found that the resonance amplitudes for $M \perp c$ were drastically enhanced compared to the case $M \parallel c$. This has been explained as being due to the enhancement factor F for the rf-field which was shown to be

$$F^{\parallel} = \frac{B_{HF}}{B_0 + B_A} \quad \text{for } B_0 \parallel c; \quad F^{\perp} = \frac{B_{HF}}{|B_0 - B_A|} \quad \text{for } B_0 \perp c, \quad (1)$$

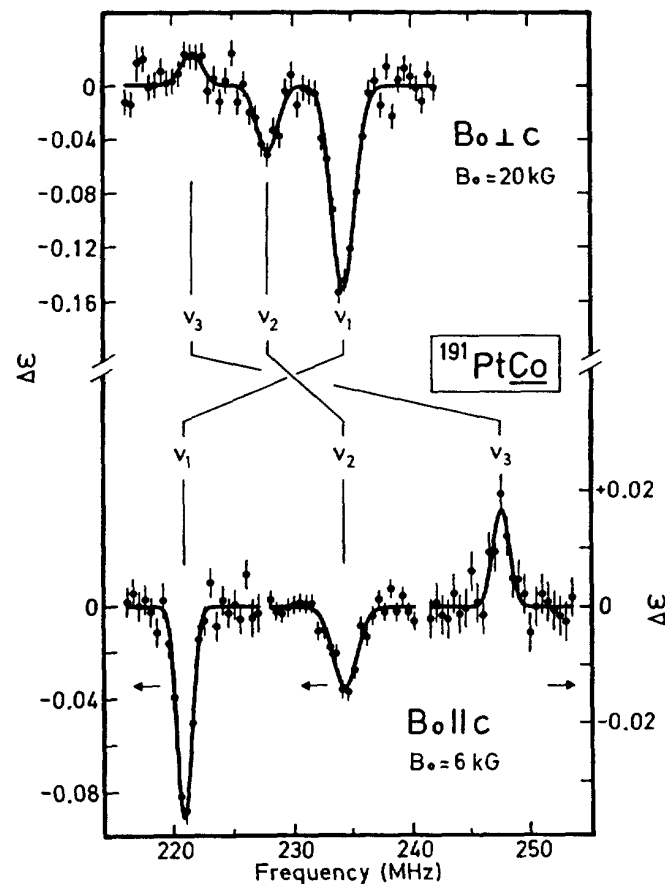


Fig. 1: NMR-ON resonance structure of $^{191}\text{PtCo}(\text{hcp})$ in "0°-geometry" and "90°-geometry". (Taken from Ref. 9.)

where B_A is an anisotropy field, defined as the magnetic field which is necessary for complete orientation of the magnetization in the case $B_0 \perp c$ -axis [9].

This observation has important consequences: For the 90° -geometry the enhancement factor for the rf-field is comparable in magnitude to low-field enhancement factors obtained with Fe as host lattice. As the spin-lattice relaxation matrix elements for hcp-Co are by a factor of ≈ 2 smaller than those for Fe and the resonance frequencies in hcp-Co are typically $\approx 30\%$ smaller than the corresponding resonance frequencies in Fe, the efficiency for resonance detection is expected to be considerably larger for hcp-Co than for Fe. With the results for the 90° -geometry, the relevant frequency region for the 0° -geometry can be predicted with good accuracy. Then, in a narrow frequency region, 0° -geometry measurements can be performed despite of the smaller resonance amplitudes, allowing then the most precise determination of the quadrupole interaction.

3) For $^{198}\text{AuCo}(\text{hcp})$ prepared with the mass-separator at Konstanz the full dependence of

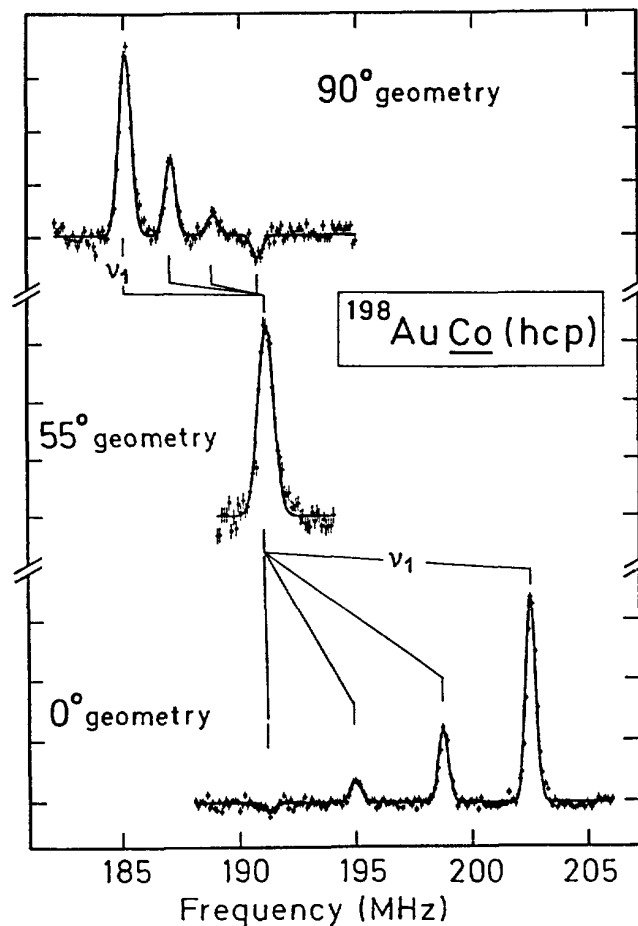


Fig. 2: Dependence of the quadrupole splitting of $^{198}\text{AuCo}(\text{hcp})$ on the angle between the magnetization and the c -axis. For $\theta = 55^\circ$ the 4 quadrupole subresonances (according to $I = 2$) coincide as the effective quadrupole interaction vanishes. (Taken from Ref. 10.)

the effective quadrupole splitting on the angle between the magnetization and the c-axis was measured recently. We have shown that a controlled "tuning" of the effective EFG is possible: By choosing the proper value for the external magnetic field B_0 , any value of the angle θ between the magnetization and the c-axis can be adjusted. Especially, it is easily possible to achieve $\theta = 55^\circ$, for which the effective quadrupole interaction vanishes. This feature will be extremely useful for the detection of new resonances of high-spin states, as all sub-resonances coincide in the 55° -geometry, improving the detection efficiency for the resonance search (Fig. 2).

4) From systematic studies of the line widths and resonance effects on $^{198}\text{AuCo}(\text{hcp})$ we recognized that the surface preparation procedure of the single crystals is the most crucial

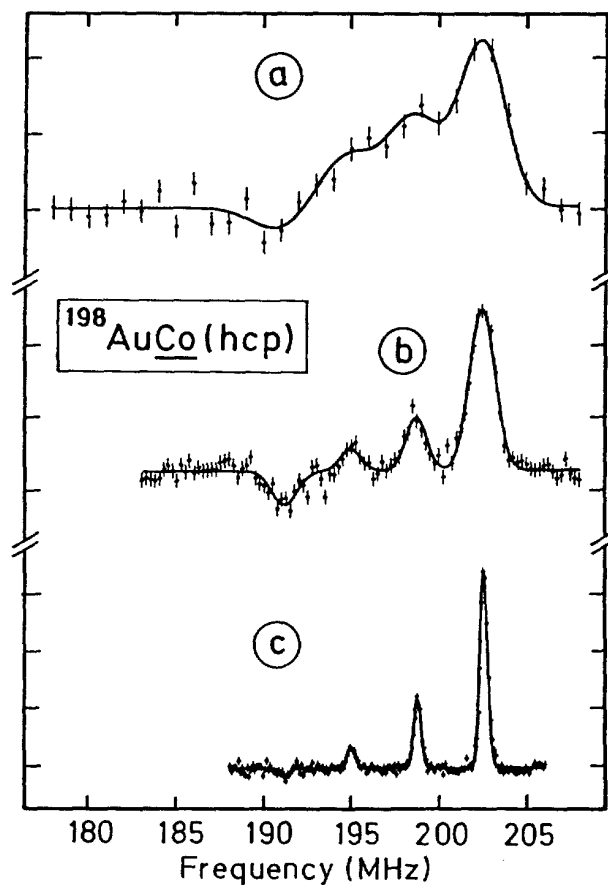


Fig. 3: Illustration of the extreme importance of the surface treatment of the single crystals.

- a) First evidence for the quadrupole splitting as observed in 1985. The line width is $\Gamma = 2.5$ MHz [7].
- b) Considerable improvement by a refined electropolishing procedure. $\Gamma = 1.5$ MHz [8].
- c) Further improvement by a sophisticated procedure of mechanical polishing, electropolishing and chemical polishing as observed 1990. $\Gamma = 0.5$ MHz [10].

point. Therefore much time was devoted to develop a technique with which high-quality surfaces can be prepared reproducibly (Fig. 3).

5) There were several unresolved problems with the ground state quadrupole moments of the neutron-deficient Ir isotopes. After it had been shown with $^{188}\text{IrCo}(\text{hcp})$ that the quadrupole substructure can be resolved well, further QI-NMR-ON measurements were performed for ^{184}Ir , ^{186}Ir and ^{186m}Ir [11]. Here, completely unexpected, a strong deviation of the effective quadrupole interaction from the $P_2(\cos\theta)$ behaviour was observed, which had initially prevented the interpretation of the measured spectra, but which was finally established in the measurements on all Ir-isotopes. The total EFG could be separated into two contributions with different angular distribution: The lattice gradient, which follows the $P_2(\cos\theta)$ dependence, and an EFG which is collinear with the electronic magnetization. This EFG could arise from a huge unquenched orbital momentum or from a hitherto not observed mechanism such as an angular dependence of the Sternheimer effect. A detailed measurement of the complete angular distribution, which is planned for the future, will be very interesting. For $^{186}\text{IrCo}(\text{hcp})$ in 0° -geometry, the lowest four subresonances have been resolved well (Fig. 4). The line widths are ~ 0.2 MHz at ~ 600 MHz, i.e. $\Gamma/\bar{\nu} \sim 3 \times 10^{-4}$. This is the highest relative resolution ever observed with NMR-ON in a metal matrix,

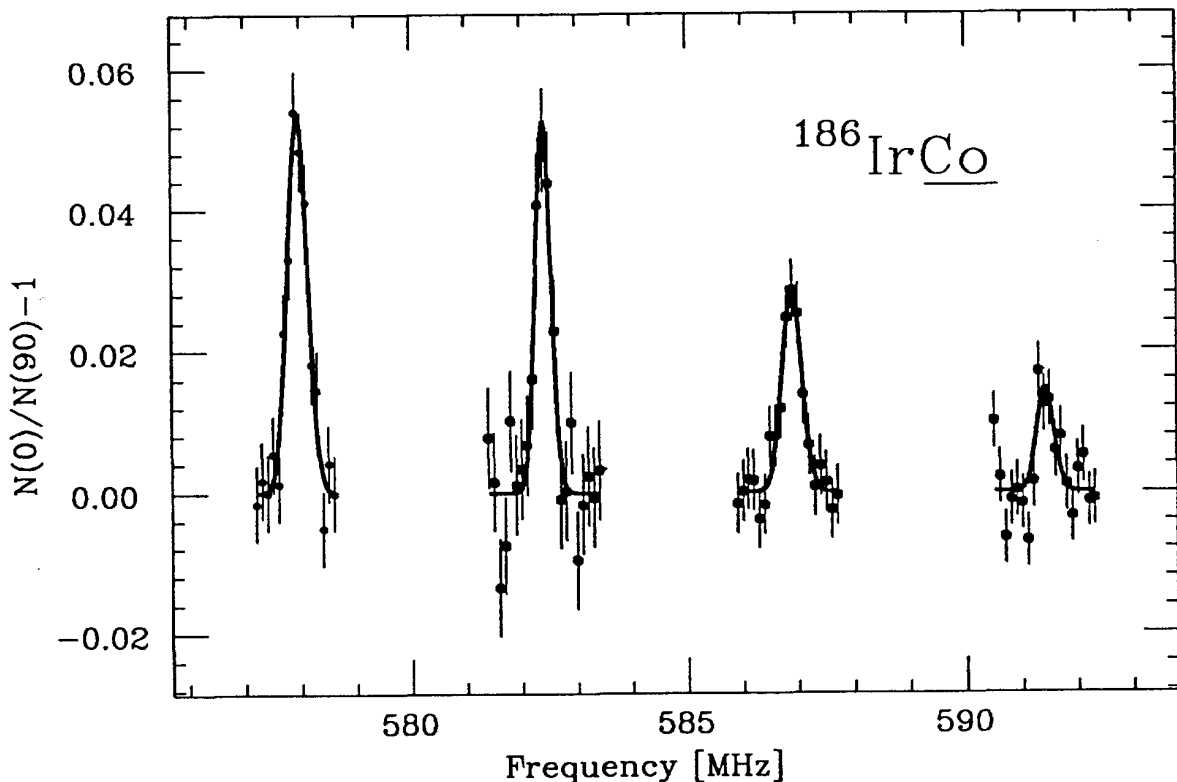


Fig. 4: NMR-ON resonances of $I = 5$ ^{186}Ir in 0° -geometry.
(Taken from Ref. 11.)

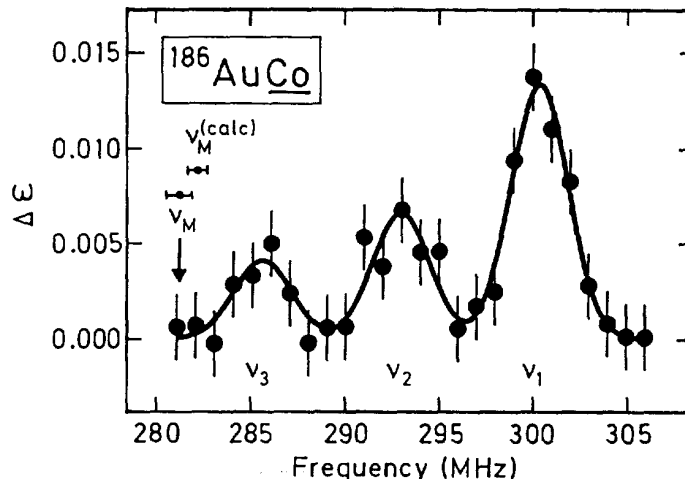


Fig. 5: NMR-ON resonance of $^{186}\text{AuCo}(\text{hcp})$ after on-line implantation of ^{186}Hg at NICOLE/ISOLDE-3. (Taken from Ref. 5.)

although the implantation voltage was only 60 kV and the samples were *not* annealed after the implantation. For ^{184}Ir the combination of 0° and 90° QI-NMR-ON measurements allowed a unique determination of the ground state spin via the resonance-offset of the ν_1 -resonance:

$$I = (\nu_M - \nu_1) / \Delta\nu_Q + 1/2 \quad (2)$$

This new method for the spin-determination is completely free of any assumptions and decay parameters and should be well applicable to nuclei far off stability.

6) In the first on-line experiment ^{186}Hg was continuously implanted into a hcp-Co single crystal kept at ~ 10 mK in the NICOLE ^3He - ^4He -dilution-refrigerator. The quadrupole splitting was fully resolved (Fig. 5) and the quadrupole moment was determined to be $Q = +3.12(20)$ b [5], which is larger than the result known from direct Laser-spectroscopy measurements [6].

3. Proposed experiments

We ask for 20 shifts, either Hg- or Au-beam. Because of the excellent warm-implantation behaviour of Au in hcp-Co we would prefer a Au-beam, if available. We think, however, that the proposed experiments can also be performed with the Hg-beam.

We would start with $^{184,184m}\text{Au}$ for which moderately good estimates of the frequency regions for the resonance search can be made taking into account the results of Laser-spectroscopy measurements [12] and nuclear orientation measurements of the NICOLE collaboration [13]. We are confident that the spins of ^{184}Au and ^{184m}Au can be determined uniquely and the configurations can be determined via the magnetic moments and electric quadrupole moments.

We would continue with ^{183m}Pt and ^{183}Ir for which also moderately good estimates of the frequency regions for the resonance search exists from NO measurements [13], and

then proceed successively to the more neutron-deficient A -chains.

Here we want to point out that, although high-precision NO data on the $A = 180 - 184$ chains exist, the determination of magnetic moments from these data contain severe uncertainties because of effects such as β -decay-induced lattice site changes which have been neglected up to now. For isotopes not too far away from stability such effects did not play an essential role as the recoil energy due to the EC- and/or β^+ -decays was generally below the displacement thresholds. For nuclei farer away from stability the Q_{EC} and Q_{β^+} values become so large, however, that the recoil energy may exceed the displacement thresholds, which has the consequence that, after a precursor-implantation, the nuclei undergo partly an EC- or β^+ -decay-induced lattice site change, and the assumption of a unique hyperfine interaction is no longer fulfilled. The existence of this effect has been proven recently by a double-resonance NMR-ON experiment on the decay of ^{90}Nb in Fe [11,16]. This means that, among the different low-temperature nuclear-orientation techniques, *only* resonance techniques yield precise *and* reliable results for the nuclear moments.

The on-line experiments can be started after the reinstallation of the NICOLE cryostat at the new beam-line.

Before the reinstallation of the NICOLE cryostat, we would like to perform off-line QIT-NMR-ON experiments with parasitical implantations (typical implantation time 10 min ... 1 h) into hcp-Co and transportation of the samples to Munich. (Here we would like to mention that several open problems in the literature concerning magnetic moments and magnetic hyperfine fields were resolved recently with such off-line implantations [14–16].) We would like to perform studies on:

- i) The anisotropy of the magnetic hyperfine fields and the electric field gradients in hcp-Co; an accurate measurement of the angular dependence of the resonance structure of Ir in hcp-Co could yield to an understanding of the Iridium anomaly.
- ii) The resonance-like magnetization-dependence of the spin-lattice relaxation matrix element. This feature could have an important application for pulsed-beam nuclear orientation studies far off stability, as it is possible to “tune” the spin-lattice relaxation time and thus to optimize the experimental conditions which is not possible with Fe as host matrix as done up to now. For these off-line experiments we would ask for parasitical beam-time. The typical implantation times are 10 min - 1 h, which, depending of the half-life of the respective isotope, then allow measurement times between 1 day and several weeks. The total beam-time for these off-line implantations would correspond to 2 - 3 shifts.

All our experiments can be performed with the GPS.

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