Beyond Antihydrogen:
Testing CPT with the Antihydrogen Molecular Ion

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Abstract

In response to CERN-SPSC-2019-028, SPSC-G-035 we outline the potential to test the CPT theorem in the baryon sector, with unprecedented precision, by radio-frequency, microwave and infrared spectroscopy on the antihydrogen molecular ion $\bar{H}_2^-$ ($\bar{p}\bar{p}e^+$). Due to the higher precision with which spectroscopy can be carried out on trapped ions versus weakly or un-trapped neutral atoms, and the intrinsic high sensitivity of $\bar{H}_2^-/H_2^+$ vibrational frequencies to $\bar{p}/p$ mass, such measurements have a sensitivity to proton/antiproton CPT violation surpassing that of measurements on $\bar{H}/H$ by more than four orders-of-magnitude. The spectroscopy requires only a *single* $\bar{H}_2^-$, which can be trapped indefinitely in a Penning or radio-frequency ion trap. $\bar{H}_2^-$ might be produced using the $\bar{H}^+ + \bar{p} \rightarrow \bar{H}_2^- + e^+$ reaction, with $\bar{H}^+$ produced using technology being developed by GBAR, although other options exist. Further work is required to ascertain the practicality of $\bar{H}_2^-$ production. If the results are positive, and assuming the necessary collaboration can be formed and funding obtained, we project a tentative timeline of proposal submission by 2022, equipment installation and connection to ELENA during LS3, and operation, requiring $\sim$1000 hours of antiproton beam time, after 2026.

I.   MOTIVATION

A direct way to test the CPT theorem [1, 2] is to compare the properties of a particle and its antiparticle. Recently there has been major progress in measurements of the mass and magnetic moment of the antiproton [3–5], and of the $1s − 2s$ transition and $1s$ Hyperfine splitting (HFS) [6, 7] of antihydrogen. For the bare antiproton the mass has been measured at a fractional uncertainty of 7×10^{-11}[3], and the magnetic moment at 1.5×10^{-9}[4]. For antihydrogen the current precision is 2×10^{-12} for the $1s − 2s$ transition [6], and 4×10^{-4} for the $1s$ HFS transition [7]. With $\bar{H}$ the goals are to reach the precision that has been achieved with H, namely 4×10^{-15} for the $1s − 2s$ transition [8], and 2.7×10^{-9} for the $1s$ hyperfine splitting (HFS) [9]. This will be difficult since both measurements on H employed high-flux atomic beams.

We propose that the same physics can be tested, *at four orders-of-magnitude higher precision*, by comparing the frequencies of vibrational and hyperfine transitions of the diatomic antihydrogen ion $\bar{H}_2^-$ with those of H_2^+ [10]. These measurements require only a single $\bar{H}_2^-$ ion, which, once produced, can be trapped and manipulated almost indefinitely [11]. In the case of vibrational transitions probed by infra-red lasers, the ion can be localized to a fraction of a wavelength of the probe light (Lamb-Dicke regime), resulting in complete suppression of the first-order Doppler shift. Atomic clock precision of better than 10^{-16} is then in reach, as has been analyzed in detail for H_2^+ [12, 13]. (For comparison, the Al^+ optical clock is already at the sub-10^{-18} level [14]). But further, it should be noted that $1s−2s$ $\bar{H}/H$ spectroscopy has sensitivity to the difference $m(e^+)/m(\bar{p}) − m(e^-)/m(p)$ mainly through the reduced mass correction. So, a 4×10^{-15} comparison of the $1s − 2s$ transition compares $m(e^+)/m(\bar{p})$ and $m(e^-)/m(p)$ at only the 7×10^{-12} level. By contrast, because the $\bar{H}_2^-/H_2^+$ vibrational frequencies scale as $\sim [m(e)/m(p)]^{1/2}$, a 10^{-16} measurement compares $m(e^+)/m(\bar{p})$ and $m(e^-)/m(p)$ at $\sim2\times10^{-16}$. Hence, combining the $\sim10^2$ experimental advantage with the $\sim10^3$ intrinsic advantage, $\bar{H}_2^-/H_2^+$ vibrational spectroscopy has a $>10^4$ advantage in sensitivity to a difference between the antiproton and proton mass, relative to the best-envisaged measurements of $1s − 2s$ in $\bar{H}/H$, or to direct antiproton/proton mass comparisons [3]. Further, using "correlation spectroscopy" [15] in which an $\bar{H}_2^-$ and H_2^+ are probed with the same laser, it is conceivable that a transition frequency *difference* could be measured at the sub-10^{-18} level. A fractional sensitivity matching that of measurements of the $\bar{K}^0$-K^0 mass difference [16] would then be in reach.

Radio-frequency measurement of the hyperfine structure of $\bar{H}_2^-/H_2^+$ probes the interactions between the e^+/e^- and $\bar{p}/p$ magnetic moments. It hence gives the same physics as measurement of $\bar{H}/H$ HFS [7, 9]. However, the tight localization and long interaction times in an ion trap may permit measurements of HFS with precision exceeding 10^{-14}. (A sub-10^{-13} measurement of the HFS of Be^+ in a Penning trap was made as long ago as 1985 [18]). Hence, for hyperfine spectroscopy also, there is a potential 4-order-of-magnitude advantage compared to the best envisaged measurements on $\bar{H}/H$ [9].

II. SPECTROSCOPIC MEASUREMENT SCHEMES

Only the ground electronic state of $\bar{H}_2^-/H_2^+$ is practically accessible for experiments. It supports 20 vibrational and over 400 bound rotational states. Because of the molecular symmetry, single-photon electric-dipole transitions between these levels are forbidden. Excited ro-vibrational levels decay by electric quadrupole transitions, resulting in excited level lifetimes of ~1 week. Correspondingly, the "natural" widths of ro-vibrational transitions are sub-μHz. While these extremely small widths are advantageous for ultra-precise spectroscopy (the ^{171}Yb single-ion clock uses an even narrower transition [19]), the lack of a strong laser-addressable transition has delayed progress on precision spectroscopy on H_2^+. A current project at LKB-Paris aimed at obtaining fundamental constants [20] uses photo-dissociation for detection [21]. Here we propose methods that use non-destructive detection on a single ion, that can be trapped, indefinitely, in the extreme vacuum of a cryogenically pumped ion trap.

A. Using the continuous Stern-Gerlach technique in a Penning trap

As detailed in [10], the ro-vibrational state and magnetic sub-state of a single $\bar{H}_2^-/H_2^+$ can be identified, non-destructively, via the continuous-Stern-Gerlach (CSG) effect [22]. This is the same detection technique as has been used in measurements of the electron g-factor of hydrogen-like ions, yielding the most precise value for the electron's atomic mass, by a collaboration of MPIK, Mainz and GSI [23]; and of the $\bar{p}$ magnetic moment by the BASE collaboration [4]. In these measurements a change in the spin direction of an electron [23], or anti-proton [4], was sensed by a change in the frequency of the ion's axial oscillation, which was monitored by detecting image currents. Applied to $\bar{H}_2^-/H_2^+$ the situation is closer to that of a hydrogen-like ion, in that it is the spin-flip of the single e^+/e^- that is detected. Because of the 650-times larger magnetic moment of the e^+/e^- compared to the $\bar{p}/p$, the e^+/e^- spin-flip transitions are much easier to detect than those of $\bar{p}/p$.

For the proposed $\bar{H}_2^-/H_2^+$ spectroscopy there is an important extension to the CSG technique: use is made of the fact that the microwave frequency that induces positron/electron spin-flips (around 140 GHz in a 5 tesla Penning trap), depends, in a resolvable and calculable way, on the ro-vibrational and hyperfine state. So, by measuring the microwave frequency at which a positron/electron spin-flip occurs (requiring relatively modest, $\sim 10^{-6} - 10^{-7}$ precision), the initial state can be determined. A try is then made at inducing the transition to be precisely measured (either Zeeman-hyperfine or vibrational) by applying radiation near the estimated transition frequency. Then, whether the transition has occurred, is probed by again measuring the positron/electron spin-flip transition frequency. The procedure is then repeated to build up a resonance curve of transition probability versus applied frequency. In essence, "electron-spin-resonance spectroscopy", with CSG detection, is used to identify the initial state and then signal the transition of interest.

Stark Quenching: Although this detection procedure can be applied to any of thousands of possible Zeeman-hyperfine or vibrational transitions, the transitions with the least sensitivity

to systematic uncertainties are between levels with the lowest vibrational and rotational quantum numbers. While it would take several weeks for an $\bar{H}_2^-/H_2^+$ formed in an excited ro-vibrational state to de-excite by spontaneous $E2$ decays to the vibrational groundstate, and many years for it to de-excite rotationally, this process can be accelerated by inducing electric-dipole transitions via the process of "Stark-quenching". By placing the $\bar{H}_2^-/H_2^+$ in a $\sim$20 mm radius cyclotron orbit in a 10-tesla Penning trap, the motional electric field it experiences speeds up the ro-vibrational decay. The $\bar{H}_2^-/H_2^+$ can then be prepared in one of the lowest 3 rotational levels of the ground vibrational level in $\sim$1 week [24].

B. Quantum-logic spectroscopy in an RF trap

"Quantum-logic spectroscopy" (QLS) is the scheme used to read-out the transition frequency in the sub-10^{-18} Al^+ optical clock [14]. Like H_2^+, Al^+ has no strong transition suitable for laser cooling or detection. Instead, a single laser-coolable ion, $e.g.$ Be^+, co-trapped in the same RF-trap, is used to sympathetically cool, and then monitor (via a coupled motional state) the clock transition in the Al^+. This scheme can be extended to transitions in molecular ions [25], with proposed extension to H_2^+, and then to $\bar{H}_2^-$, using a cryogenic, double-well RF-trap. QLS is expected to enable higher precision due to a faster read-out time and avoidance of high magnetic field. However, we expect that production and initial state preparation of H_2^+ will still use Penning traps.

C. Correlation spectroscopy

Unlike an atomic clock where one is concerned with the reproducibility and transfer of the absolute transition frequency, in order to test CPT one only needs to detect a *difference* between $\bar{H}_2^-$ and H_2^+. By performing simultaneous Ramsey spectroscopy on the two ions, trapped in close proximity in the same trap, and detecting correlation in the transition probability, a signal can be obtained in which sources of phase noise common to both ions cancel [15]. The frequency difference can then be probed with phase-evolution times of several seconds, much longer than the coherence time of the laser. This has recently been demonstrated in the comparison of two Al^+ clocks [26]. This may provide a path to a measurement at the sub-10^{-18} level, thereby exceeding the fractional precision of the $\bar{K}^0$-K^0 mass difference, often cited as the most precise test of CPT [16].

D. Portable Penning traps for transport of $\bar{H}_2^-$

Portable Penning traps for antiprotons are being developed both by the PUMA collaboration [27] and by the recently funded STEP project headed by C. Smorra at the University of Mainz. Further development would enable a single $\bar{H}_2^-$ to be transported from the AD hall to the less noisy environment of a precision measurements laboratory.

III. PRODUCTION OF $\bar{H}_2^-$

The proposed measurements require a single trapped $\bar{H}_2^-$ which could be manipulated and measured on for many weeks or months. Of the four two-body production reactions: (1) $\bar{H} + \bar{p} \to \bar{H}_2^- + \gamma$, (2) $\bar{H}(1s) + \bar{H}(n \geq 3) \to \bar{H}_2^- + e^+$, (3) $\bar{H}(2s) + \bar{H}(2s) \to \bar{H}_2^- + e^+$, (4) $\bar{H}^+ + \bar{p} \to \bar{H}_2^- + e^+$, reaction (1) has an extremely small cross-section, while reactions (2) and (3) have been considered in the context of laser-excited, cold, trapped antihydrogen by Zammit $et\ al.$ [28], who conclude that reaction (3) is superior, see below. However, assuming the availability of the atomic antihydrogen ion $\bar{H}^+$, whose synthesis is a goal of the GBAR collaboration [29], reaction (4) may provide an elegant, all-ion route to $\bar{H}_2^-$ production.

A. Production of $\bar{H}_2^-$ from $\bar{H}^+ + \bar{p} \to \bar{H}_2^- + e^+$ in a Penning trap

In this scheme, using methods already developed for $\bar{H}$ production [30–33], $\sim 10^5$ antiprotons, cooled to below 100 K, would be trapped in the central trap of a 5-tesla nested Penning trap, producing a cloud with a radius of 1 mm, with integrated density along the axis of $\sim 3 \times 10^6$ cm^{-2}. A single $\bar{H}^+$ is then injected into one of the side-traps for positive ions. The $\bar{H}^+$ sees the trap containing the antiproton cloud as a central potential hill. Using single-ion image-current techniques [3, 23, 34], the $\bar{H}^+$ in the side-trap is monitored, cooled in its axial and cyclotron modes to a few meV, and radially centered to a few microns. The axial energy of the $\bar{H}^+$ is then increased, using a chirped resonant electric field, until it is within a few meV of the potential hill. The $\bar{H}^+$ then oscillates back and forth through the antiproton cloud, or reverses direction within the antiproton cloud and oscillates in a single side-trap, until it reacts with an antiproton. A slow ramp is applied to the side-trap potentials, to compensate for the loss of axial energy of the $\bar{H}^+$ due to Coulomb collisions with the antiprotons. The continued presence of the $\bar{H}^+$ is monitored periodically by deepening the side-traps so they become sufficiently harmonic that the $\bar{H}^+$ can be again detected using image current techniques. The harmonic side-traps are also used to recenter and reset the axial energy of the $\bar{H}^+$ as necessary.

The $\bar{H}_2^-$ production reaction (4) [35] is in fact a 0.5% side branch of the mutual neutralization reaction which forms $\bar{H} + \bar{H}$ [36], which has a cross-section (inversely proportional to energy) of 1.2×10^{-11} $(E_{cm}/\text{meV})^{-1}$ cm^{-2} . Hence, assuming an average center-of-mass energy of 10 meV, the total probability for either reaction is $\sim 10^{-6}$ per pass, requiring a "mixing" time of ~ 10 s at $10\,\mu$s per pass. When the $\bar{H}^+$ is no longer detected, the antiprotons will be selectively removed from the reaction trap and the presence of an $\bar{H}_2^-$ searched for using image current techniques. The process is then repeated ~ 200 times till an $\bar{H}_2^-$ is detected. Provided the time required for the entire process, that results in the $\bar{H}^+$ undergoing either reaction, is less than the average time between the availability of successive $\bar{H}^+$, the $\bar{H}_2^-$ production rate will be limited by the $\bar{H}^+$ rate. Hence, conservatively, assuming the availability of $\bar{H}^+$ at a rate of $\sim 10^{-3}$ s^{-1} [29, 37], it follows that the $\bar{H}_2^-$ production rate will be $\sim 5 \times 10^{-6}$ s^{-1}, $i.e.$, one every few days. Given that a single $\bar{H}_2^-$ can then be used for months, a manufacturing time of several days is not unreasonable.

$\bar{H}^+$ *loss through the formation of Coulomb complexes:* The above assumes no $\bar{H}^+$ loss besides reaction with an antiproton. A potentially limiting loss mechanism is the formation of a weakly bound $\bar{p}\bar{H}^+$ "Coulomb complex" (or "Heavy Rydberg atom") through the three-body collision $\bar{p}+\bar{p}+\bar{H}^+$. This is the analogue of the $e^++e^++\bar{p}$ reaction that is understood to be the main route to antihydrogen formation via Rydberg antihydrogen [38]. Being neutral, such $\bar{p}\bar{H}^+$ are untrapped. If they are not disassociated by the trapping fields, they will cause the $\bar{H}^+$ to be lost from the trap. Estimates based on scaling the rates used to simulate Rydberg antihydrogen formation [39], indicate that at antiproton densities of $\sim10^6$ cm^{-3} and temperatures of ~100 K, the formation rate of $\bar{p}\bar{H}^+$ complexes that survive the trapping fields is less than that of the mutual neutralization reaction. However, detailed simulations are necessary to investigate this and other potential $\bar{H}^+$ loss mechanisms.

B. Production of $\bar{H}_2^-$ from $\bar{H}(2s) + \bar{H}(2s) \rightarrow \bar{H}_2^- + e^+$

Zammit *et al.* [28] have considered the production of $\bar{H}_2^-$ from collisions of two antihydrogen atoms in the $2s$ metastable state (mean lifetime 0.12 s), reaction (3), in the context of the ALPHA antihydrogen experiment. In one scenario, laser cooling is used to produce an $\bar{H}$ density of 10^3 cm^{-3} at 1 mK, followed by efficient laser excitation to the $2s$ state. They estimate an $\bar{H}_2^-$ production rate of 10^{-2} s^{-1}, but with difficult-to-estimate competing losses with a rate ~100 times greater. The viability of this route should become clearer after further developments in the production and laser cooling of antihydrogen.

IV. ONGOING AND COMPLETED ACTIVITIES TOWARD $\bar{H}_2^-$ SPECTROSCOPY

A. Demonstration of Zeeman, Zeeman-hyperfine and vibrational spectroscopy on H_2^+ in a Penning trap using the ALPHATRAP apparatus at MPIK

The ALPHATRAP Penning trap [40] was designed by S. Sturm and colleagues at MPIK to perform precision measurements of g-factors and other microwave and laser spectroscopy on externally-created highly-charged ions, with state detection via the continuous Stern-Gerlach technique. It is based on experience gained from several successful high-precision g-factor measurements on lower-charge ions, including one that produced the most precise measurement of the electron's atomic mass [23]. It is planned to use this system with H_2^+ to demonstrate the proposed Penning-trap based state-identification and spectroscopic technique. ALPHATRAP's capabilities are almost ideal for the proposed $\bar{H}_2^-/H_2^+$ experiments, and it serves as a model for the spectroscopy part of a future apparatus. In particular, thanks to a cryogenic valve, it allows for the injection of an externally produced ion while preserving the extreme vacuum necessary to store it for many months.

B. Calculation of H_2^+ Zeeman and Zeeman-hyperfine transition frequencies

The Penning trap detection technique requires knowledge of a large number of Zeeman transition frequencies in H_2^+ in a $\sim$5 tesla magnetic field, at the level of 10^{-6} to 10^{-7}. The necessary calculations are being performed at LKB by J.-P. Karr.

C. Calculation of H_2^+ Stark-quenching rates

Stark-quenching rates for ro-vibrational transitions have already been calculated by Karr [24]. These show that Stark-quenching using a large cyclotron orbit in a 10 tesla Penning trap will be an effective means for producing $\bar{H}_2^-/H_2^+$ in its lowest vibrational and rotational states.

D. Simulation of production of $\bar{H}_2^-$ from $\bar{H}^+ + \bar{p} \to \bar{H}_2^- + e^+$ in a nested Penning trap

$\bar{H}_2^-$ production from single $\bar{H}^+$ and a trapped cloud of antiprotons in a realistic nested Penning trap will be simulated [41], with a focus on loss mechanisms for the $\bar{H}^+$. Guided by the results, a cryogenic, extreme high-vacuum Penning trap will then be developed which will test the process by producing an H_2^+ from H^- ions and protons.

V. USE OF CERN RESOURCES

A. Outlook and timeline

Given a single $\bar{H}_2^-$ trapped in an ALPHATRAP-like apparatus, we have confidence that CPT can be tested with a sensitivity exceeding that obtainable using bare antiprotons or antihydrogen. Hence, the proposal's success hinges on the ability to make $\bar{H}_2^-$. This will become clearer after completion of the above simulations and test experiments to synthesize H_2^+ from H^-, and after further progress in the production of $\bar{H}^+$ by the GBAR experiment; or, by development of the alternative route to $\bar{H}_2^-$ using metastable antihydrogen. If these indicate a clear path to $\bar{H}_2^-$ production, and the necessary collaboration can be formed, a full proposal to CERN might be submitted by 2022. Assuming the $\bar{H}^+$ route, installation of the major components, namely, the $\bar{H}_2^-$ production apparatus (antiproton and $\bar{H}^+$ catching traps, mixing traps, and Stark-quenching solenoid), and the spectroscopy apparatus, with beamline connections to ELENA and GBAR, could then occur during 2024-2025, that is during LS3, with first experiments in 2026 or 2027.

B. Infrastucture and beamtime requirements

Assuming the above timeline, we tentatively envisage, beginning during LS3, requirements of 50 m^2 of floor space (for the $\bar{H}_2^-$ production apparatus, ALPHATRAP-like spectroscopy apparatus, laser hut, electronics racks, work space, extra space for future RF trap), 20 kW at 230 Vac, and cryogens of 100 liters/week LHe, 200 liters/week LN2. Assuming their collaboration, the space would be adjacent to GBAR, with beamline connection to GBAR for $\bar{H}^+$ as well as to ELENA for antiprotons. The requirements for antiprotons from ELENA will be determined by the $\bar{H}_2^-$ production process, and especially by the antiproton requirements to produce a few hundred $\bar{H}^+$. Although this is still to be determined, it is conceivable that all necessary commissioning, first $\bar{H}_2^-$ production, and a beyond-antihydrogen test of CPT could take place with 1000 hours of ELENA beamtime.

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