Determination of the proton mass from a measurement of the cyclotron frequencies of D^+ and H_2^+ in a Penning trap

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We determine the cyclotron frequency ratio of H_2^+ and D^+, applying the two-pulse Ramsey-excitation technique in the Penning-trap mass spectrometer SMILETRAP. The final result, based on probing more than 100 000 ions, is a frequency ratio of 0.999 231 659 33(17). Using a value of the D^+ mass recently measured by the Seattle group, we obtain so far the most precise experimental H_2^+ mass value of 2.015 101 497 16(34) u. From this value a proton mass value of 1.007 276 466 95(18) u (0.18 ppb relative uncertainty) could be derived, in good agreement with the value of 1.007 276 466 89(14) u published by Van Dyck *et al.* [R. S. Van Dyck, Jr., D. L. Farnham, S. L. Zafonte, and P. B. Schwinberg, in *Trapped Charged Particles and Fundamental Physics*, edited by D. E. Dubin and D. Schneider, AIP Conf. Proc. No. 457 (AIP, Woodbury, NY, 1999)].

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I. INTRODUCTION

The masses of the very lightest nuclei are fundamental constants, needed in different fields of physics with an increasing demand of accuracy. One of the most fundamental mass values is the proton mass and the value is reported in the NIST CODATA database to a precision of 1 part in 10×10^9 [1]. However, so far it is only Van Dyck *et al.* who has measured it with this precision, reporting an uncertainty of 0.14 ppb [2]. Other measurements of the proton mass, carried out, e.g., by the MIT group [3] and by our group [4], have at least 3 times larger uncertainties. The Seattle trap uses a nondestructive technique where a single ion is observed for a long time. Because the axial frequency slightly depends on the cyclotron energy, due to a nonzero quadratic magnetic-field term and a nonzero hexadecapole electrostatic-potential term, the ion cyclotron frequency can be measured through the image current induced in the trap endcap electrodes by the axial motion of the ion [5]. Our Penning trap mass spectrometer (SMILETRAP) uses instead the so-called destructive time-of-flight (TOF) technique [6] where thousands of ions are used sequentially and the cyclotron frequency is determined via the ion TOF to a detector outside the trap. Therefore, an at least as precise measurement by this, different, method would give additional insight into possible systematic errors and would add to the reliability of the value.

In our previous measurements the H_2^+ molecular ion has frequently been used as a mass reference. This is mainly because it has a well-known mass, a q/A close to that of the highly-charged ions investigated and it is easily created from the rest gas H_2 in the first, cylindrical, trap of our setup, called the preparation trap (pretrap). A small uncertainty, 0.11 ppb, of the H_2^+ mass is assumed, thanks to its close relation to the proton mass. In the Appendix we show how the masses of H_2^+ and the proton are connected on a sub-0.1-ppb accuracy level. Using the value of the proton mass from the NIST CODATA database, an H_2^+ mass of 2.015 101 496 80(21) can be derived. So far, there is no direct high-precision measurement of the H_2^+ mass. Previously, Brunner *et al.* [7] measured the cyclotron frequency ratio of H_2^+ and $^4He^+$ in a Penning trap using the TOF technique and single-pulse excitation. The measurement was done at a time when the published values of the helium masses were strongly questioned as shown by Fritioff *et al.* [8]. Now, when a precise mass value of the ^{4}He isotope of 4.002 603 354 15(6) u is available [9], it is more justified to use the results of Brunner *et al.* to calculate the H_2^+ mass, though it was not the original objective. This calculation yields a H_2^+ mass of 2.015 101 496 9(32).

In 2006 Van Dyck *et al.* published an improved value of the mass of deuterium with an uncertainty of 0.07 ppb [9]. This is, however, only a preliminary result and in private communication Van Dyck has kindly given us a final value of the D^+ mass of 2.013 553 212 73(4) u, a relative uncertainty of 0.02 ppb [10]. Thus, D^+ is a suitable mass reference ion in a q/A doublet measurement of the H_2^+ mass.

Together with the fact that recent works [11,12] have shown that a precision increase by a factor of about 3 can be reached by applying two-pulse Ramsey excitation, this encouraged us to attempt a determination of the proton mass at close to 0.1 ppb precision, measuring the cyclotron frequency ratio of H_2^+ and D^+ ions.

II. MASS DETERMINATIONS IN SMILETRAP

The procedure of mass measurements in our Penning-trap mass spectrometer SMILETRAP is described in detail in [13]. Here we therefore only give a brief summary. The setup consists of a plasma ion source device (SMILIS), a 90° analyzer magnet, two Penning traps, and a Microchannel plate-(MCP-) based ion detector. The H_2^+ ions are produced in the cylindrical preparation trap (pretrap) by bombarding the rest gas with 3.4-keV electrons, while the D^+ ions are delivered from SMILIS. Before the D^+ ions enter the pretrap, a q/A ion selection is done in the analyzer magnet. On average, a

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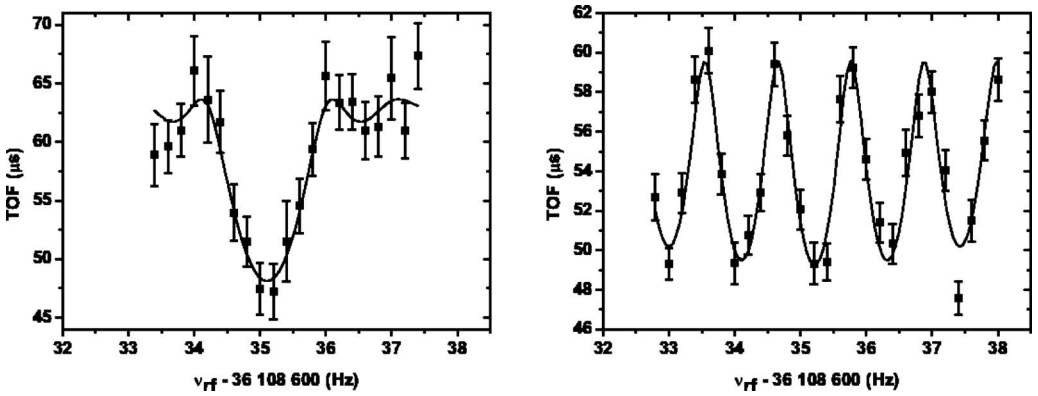

FIG. 1. The one-pulse TOF resonance of D^+ ions (left) and the corresponding two-pulse resonance (right). Specifically note that all fringes of the two-pulse resonance have almost the same intensity as opposed to the weak sidebands of the one-pulse resonance.

few thousand D^+ ions are able to enter the pretrap. Here the ions are retarded from the transportation energy of 3.4 q keV to the ground potential in a time of about 30 ms, then accelerated to −1 keV and transported to the hyperbolical precision trap. Before entering this trap, the ions are retarded to the ground potential. Only about 30 ions are able to reach the precision trap, mostly due to poor phase-space properties of the beam from SMILIS. In addition, an entrance aperture with a diameter of 1 mm prevents ions with too large radial energies from entering the trap. The hottest ions are then abolished in an evaporation process where the trap potential is lowered from 5 V to 0.1 V and then quickly restored. About 0–7 ions are left in the trap after this procedure and subjected to an rf-quadrupole excitation, typically during 1 s. It is possible in the offline analysis to single out data with different numbers of detected ions in the trap and, if necessary, to correct for any ion number effect. However, the ion detection efficiency in the present detector of about 0.50(10) [14] has to be taken into account.

The excitation frequency ν_{rf} is scanned in steps around the ion cyclotron frequency ν_c, which is given by $\nu_c = qeB/(2\pi m)$, where m is the mass of the ion and B the magnetic field strength (in our trap approximately 4.7 T). If the applied excitation frequency matches the ion cyclotron frequency, $\nu_{rf}=\nu_c$, a conversion of the magnetron motion into cyclotron motion will occur, increasing the ion radial energy [15]. If, on the other hand, the applied frequency differs from the cyclotron frequency, the gain in energy will be less than in the resonance case or even zero.

After excitation, the ions are extracted from the trap and enter the inhomogeneous part of the magnetic field. In this area the magnetic dipole moment of the ion motion couples to the gradient of the magnetic field, resulting in a conversion of the radial energy into axial energy [6]. By plotting the average TOF of the ions to a detector as a function of the applied frequency, the ion cyclotron frequency displays as a well-pronounced minima with weak sidebands (Fig. 1). By fitting the theoretical curve to this result, the center frequency, corresponding to the cyclotron frequency, can be obtained.

In the one-pulse excitation described so far, the rf-field is applied continuously during 1 s. The resulting resonance curve is made up of 21 frequencies separated by 0.2 Hz (Fig. 1). In the measurements reported in this paper, one-pulse excitation has only been used to roughly locate the resonance frequency, while the actual data were recorded using the Ramsey-excitation technique [11,12,16].

In the Ramsey case the rf-field pulse is divided into two or more pulses with a waiting time in between. Here we have used a two-pulse excitation with two 0.1-s excitation pulses separated by a 0.8-s waiting time. In order to better cover the Ramsey fringes (Fig. 1), the number of frequency steps was increased to 27 and sometimes even 31, keeping the separation between adjacent frequencies at 0.2 Hz. The benefit of using two-pulse Ramsey excitation is a more precise determination of the center frequency. This is a result of a narrower linewidth and a higher intensity of the Ramsey fringes compared to the weak sidebands of the one-pulse resonance (Fig. 1) [11,12]. One complete frequency measurement, covering the resonance region, is called a *scan*. A certain number of scans, grouped together in the evaluation for the resonance fit, are named a *bunch* of scans. Usually a bunch consists of about 80–120 scans.

In order to obtain the mass of the investigated ion, its cyclotron frequency ν_i is compared to the cyclotron frequency ν_{ref} of a reference ion of well-known mass, in this case D^+. If the change in the B field between the measurements of the two cyclotron resonances can be neglected, the ratio between the two cyclotron frequencies gives the mass of the ion under investigation:

$$R = \frac{\nu_i}{\nu_{ref}} = \frac{q_i m_{ref}}{q_{ref} m_i}. \tag{1}$$

In this measurement singly charged ions were used and therefore

$$m_i = \frac{\nu_{ref}}{\nu_i} m_{ref} = \frac{m_{ref}}{R}. \tag{2}$$

In general, to get the mass M of the neutral atom one has to add the mass of the missing electrons, qm_e, and their binding energies E_B:

$$M = m + qm_e - E_B/c^2. \tag{3}$$

However, in this work this is not necessary since the objective is to determine the mass of the H_2^+ ion.

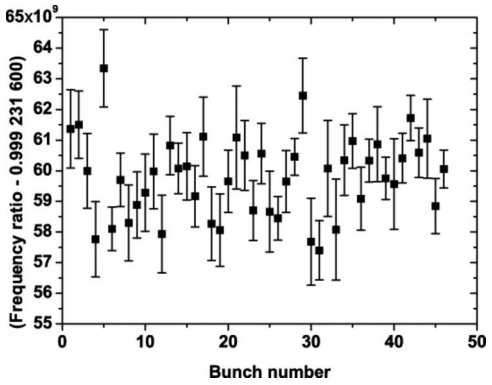

FIG. 2. The uncorrected cyclotron frequency ratios of H_2^+ and D^+ ions for subsequent bunches. The data include one to two detected ions and result in a weighted average of 0.999 231 659 83(18).

III. DATA ANALYSIS

A. Uncorrected data

Altogether our two-pulse Ramsey-excitation measurement resulted in more than 5300 scans of each ion species, grouped together in 46 bunches, collected with some interruptions over a period of three weeks. Each bunch of scans yields a value of the cyclotron frequency and an estimate of the uncertainty which, together with the corresponding frequency for the other ion measured in parallel, yields a frequency ratio. Figure 2 shows the frequency ratios, as obtained with subsequent bunches, using data with one to two detected ions. The weighted average with the weighted standard error of the mean becomes 0.999 231 659 83(18), corresponding to a relative uncertainty of 0.18 ppb.

B. Possible systematic effects

In previous measurements [13] we concluded that the most serious systematic effects originate from the following sources: (a) uncertainties in the electron mass and electron binding energies, (b) q/A asymmetry of the two ion species, (c) uncertainty in the reference mass, (d) variations in the magnetic field, (e) relativistic mass increase, and (f) ion-ion interactions.

It can be noticed immediately that the first two effects are not relevant in this measurement since our objective is to determine the ion mass rather than the atomic mass and since we are using perfect q/A ion doublets. With an ambition to reach a precision close to 0.1 ppb, also the uncertainty of 0.02 ppb in the D^+ mass can be neglected in the evaluation of the H_2^+ and proton masses. The remaining three effects are discussed below.

1. Variations in the magnetic field

The natural decay of the superconducting magnet is $\ll 10^{-9}$ per hour [13]. During this experiment, the measurement was switched between H_2^+ and D^+ every five scans, resulting in a measurement cycle of about 5 min. This corresponds to a change in the field by $\ll 0.08$ ppb, and the resulting systematic uncertainty of $\ll 0.04$ ppb can be neglected. To reduce the B-field variation caused by the fluctuations of ambient parameters such as atmospheric pressure and room temperature, an active regulation system was developed [13].

There is also an oscillation of the field originating from the temperature stabilization system itself with a period of about 100 min. However, since the lengths of the bunches used in the evaluation are in the same order as the temperature-induced oscillation period, this will result in a broadening of the resonance width, which limits the statistical uncertainty, rather than introducing any systematic uncertainty [17].

2. Relativistic mass increase

During the excitation, the ions gain energy and as a result the resonance frequency shifts due to the relativistic mass increase. This shift of the frequency is rather small, and it becomes even smaller when looking at the frequency ratio [13]. Thanks to previous calibration measurements [18], the relativistic shift of the frequency ratios can be corrected for by comparing the TOF of ions in and off resonance. In this experiment, the relativistic effect of each bunch was calculated individually and the ratios were corrected accordingly. As a conservative estimate, the full correction was used as the uncertainty of the correction of each ratio. The average relativistic correction in this measurement was −0.12 ppb. This is a small correction compared to the uncertainties of the individual ratios, which are of the order of 1 ppb. The result is a shift of the weighted average frequency ratio from 0.999 231 659 83 to 0.999 231 659 71 and an increase in the relative uncertainty from 0.1787 ppb to 0.1794 ppb.

3. Ion-ion interaction

It is known that having more than one ion in the trap could cause a shift of the ion cyclotron frequency [19,20], and since the number of trapped ions is a random variable this effect has to be addressed. One way to handle it would be to discard any data with more than one ion in the trap. This is unfortunately not possible with the present system because of the reduced detection efficiency of the TOF detector of 0.5 for singly charged light ions [14]. It also has the drawback that a lot of data would be wasted. In earlier measurements an upper limit of this correction was estimated and treated as a systematic uncertainty. With the present requirement of precision, it is necessary to instead do a number dependence correction. For this purpose each bunch in this experiment was analyzed for different, well-defined numbers of detected ions.

The measured ratio $R(n_H', n_D')$ depends on the number of detected ions as

$$R(n_H', n_D') = \frac{\nu_H(0) + k_H n_H'}{\nu_D(0) + k_D n_D'}, \tag{4}$$

where $\nu_x(0)$ and k_x are constants and n_x' are the average number of detected D^+ and H_2^+ ions, respectively. With the current detector efficiency of 0.50(10), the value corresponding to having only one ion in the trap is $R = R(0.5, 0.5)$. By ap-

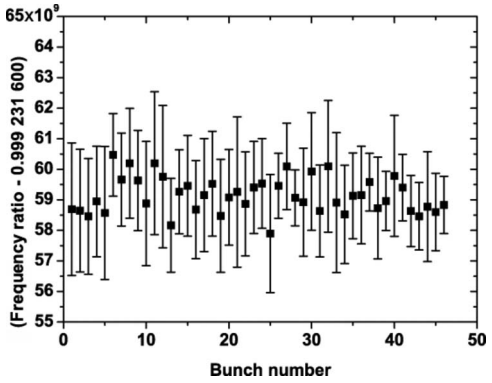

FIG. 3. Corrected cyclotron frequency ratios of H_2^+ and D^+ ions for subsequent bunches accepting one to two detected ions. The correction was done by extrapolation of the values of Fig. 2 to 0.5 detected ion.

plying the variable transformation $n_x' \Rightarrow n_x = n_x' - 0.5$, we obtain the equation

$$R(n_H, n_D) = \frac{\nu_H(0.5) + k_H n_H}{\nu_D(0.5) + k_D n_D} = \frac{\nu_H(0.5)}{\nu_D(0.5)} \frac{\left(1 + \frac{k_H}{\nu_H(0.5)} n_H\right)}{\left(1 + \frac{k_D}{\nu_D(0.5)} n_D\right)}. \tag{5}$$

The ratio $\nu_H(0.5)/\nu_D(0.5)$ is exactly the wanted R, and since k_D and n_D are of the order of 1 and $\nu_D(0.5)$ is of the order of 36 MHz, the denominator can be Taylor expanded, yielding (to the second order)

$$R(n_H, n_D) = R(1 + \epsilon n_H)[1 - \delta n_D + (\delta n_D)^2], \tag{6}$$

where ϵ and δ are small compared to unity. By making 46 simultaneous three-dimensional fits, one for each bunch, with ϵ_i and δ_i as free parameters and R as a common parameter of all fits, a value of $R = 0.999\,231\,659\,33(17)$ is obtained.

Worth noticing is how the uncertainty of the frequency ratio actually decreases from the correction. This can be understood by comparing Fig. 2 to Fig. 3 where the 46 ratios again are presented in sequential order. The values of Fig. 3 were obtained by extrapolating the values of Fig. 2 to 0.5 detected ion using Eq. (6) and the values of the coefficients obtained from the fits. It is evident that the spread of the ratios has decreased drastically, resulting in a more precise determination of the average ratio. The large error bars of the individual points are because these are extrapolated values and also because they originate from the one- to two-detected-ion data only, while the value of R from the least-squares fit incorporates all data up to four detected ions.

IV. RESULTS

The value of the frequency ratio from the three-dimensional fit becomes 0.999 231 659 33(17), yielding, together with the Seattle value of the D^+ mass, an H_2^+ mass of 2.015 101 497 16(34) u, a deviation by 0.18 ppb from the value calculated from the tabulated proton mass [1]. This result is presented in Fig. 4 and compared to the values derived from the tabulated proton mass [1] and the up-to-now best direct determination of the H_2^+ mass by Brunner *et al.* [7].

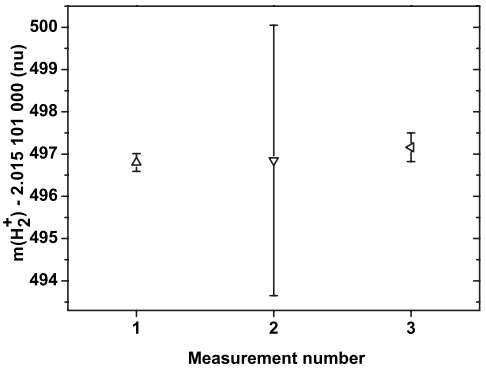

FIG. 4. Values of the H_2^+ mass. From left to right: (1) (△) the value calculated from the proton mass from the most recent NIST CODATA database [1], (2) (▽) the experimental value by Brunner *et al.* [7], and (3) (◁) the value derived in this experiment.

From our measured mass of H_2^+ a proton mass of 1.007 276 466 95(18) u can be derived (see the Appendix). This is in good agreement with both the value from the most recent NIST CODATA database of 1.007 276 466 77(10) u [1] and the most precise measurement by Van Dyck *et al.* [2] of 1.007 276 466 89(14) u (Fig. 5 and Table I).

V. CONCLUSIONS AND OUTLOOK

The SMILETRAP Penning-trap mass spectrometer was originally designed for mass measurements using highly charged heavy ions. This measurement has proven that SMILETRAP, with the implementation of the Ramsey-excitation technique and the improved handling of the ion-ion interaction effect, can compete with other mass spectrometers, reaching a precision close to 0.1 ppb, even for the lightest elements.

The main uncertainties in this experiment come from the temperature-induced magnetic-field oscillation which causes

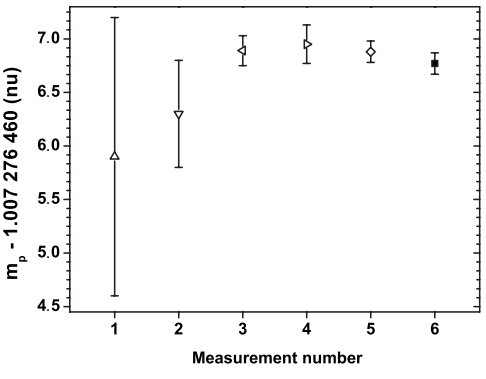

FIG. 5. Values of the proton mass. From left to right: (1) (△) the value by Borgenstrand [18], (2) (▽) the value by DiFilippo *et al.* [3], (3) (◁) the value by Van Dyck *et al.* [2], (4) (▷) the value from this experiment, (5) (◇) the weighted average of all Penning-trap measurements, and (6) (■) the proton mass from the most recent NIST CODATA database [1].

TABLE I. The mass of the proton from this experiment compared with other high-precision measurements and the value from the NIST CODATA database.

Reference		Proton mass	Comment
1	Borgenstrand [18]	1.007 276 465 90(130)	From measurements of the H_2^+ mass
2	DiFilippo *et al.* [3]	1.007 276 466 30(50)	
3	Van Dyck *et al.* [2]	1.007 276 466 89(14)	
4	This experiment	1.007 276 466 95(18)	From a measurement of the H_2^+ mass
5	Weighted average	1.007 276 466 88(10)	All Penning-trap measurements
6	NIST [1]	1.007 276 466 77(10)	Tabulated value

a broadening of the resonance and sometimes even renders data useless. Also the low detection efficiency of the TOF detector for light, singly charged ions is a weak point in the ion-ion interaction correction. For the new SMILETRAP II experiment which is currently being set up, a much better temperature stabilization system is being designed and a new detector with a detection efficiency close to 1 is being implemented. A more stable magnetic field will, furthermore, make it possible to increase the excitation time which will push the limit of the precision even further. SMILETRAP II will be located in the new electron-beam ion-trap source (S-EBIT) laboratory that is being built at the AlbaNova research center in Stockholm. S-EBIT will make it possible to fully exploit the precision increase with charge state.

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APPENDIX: CALCULATION OF THE PROTON MASS FROM THE H_2^+ MASS

The mass of the proton can be calculated from the mass of the H_2^+ ion in the following way:

$$m_p = \frac{1}{2}[m(H_2^+) - m_e - E_I(H_2) + 2E_I(H) + E_B(H_2)], \tag{A1}$$

where m_e=0.000 548 579 909 43(23) u [1] is the electron mass, $E_I(H)$=13.598 433 7(12) eV [1] the hydrogen ionization energy, $E_I(H_2)$=15.425 77(3) eV [21] the ionization energy of H_2, and $E_B(H_2)$=4.478 10(6) eV+ΔE [21] the molecular binding energy.

The conversion between electron volts and atomic mass units is done by the factor 1 eV=1.073 544 188(27) nu [1]. The uncertainties in these atomic and molecular constants are all smaller than 0.001 nu and thus negligible on a 0.1-nu level.

The term ΔE is the vibrational energy above the ground-state energy of the H_2^+ molecule and the energy for a certain vibrational quantum number n can be calculated from the semiempirical formula [21]

$$E(n) = k_1\left(n+\frac{1}{2}\right) + k_2\left(n+\frac{1}{2}\right)^2, \quad k_1 = 0.2848 \text{ eV},$$

$$k_2 = -0.007\,687 \text{ eV}. \tag{A2}$$

Because of the simple rotational symmetry of the H_2^+ molecule, the uncertainty of the energy predicted by this formula is entirely negligible at the uncertainty level of this paper.

ΔE depends on which vibrational quantum level the ion occupies. In our case we have to average, because we are using several thousand ions, excited into different vibrational states. Thus, what we measure is the average mass of the H_2^+ ion. Assuming that the H_2 molecule, before ionization and at room temperature, is in its vibrational ground state, we calculate the energy above ground energy, $\Delta E(n)=E(n)-E(0)$, of each state and fold it with the population probabilities to get the average energy.

The population probabilities of the vibrational levels of H_2^+ have been both measured [22,23] and calculated [23], and they all agree within two percentage points (Fig. 6). Moreover, the result by Weijun *et al.* [22] has been measured using about 100 000 ions [24], corresponding to a relative statistical uncertainty of 2% or less of all measured population probabilities. Using the calculated values from [23] to-

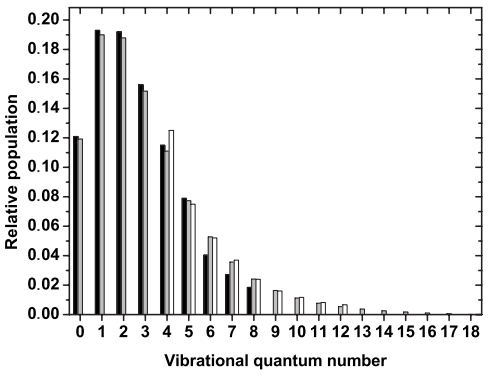

FIG. 6. Population probabilities of the first 19 H_2^+ vibrational levels as calculated by [23] (gray) and measured by [23] (white) and [22] (black).

gether with Eq. (A2), an average ΔE of 0.742 eV with a standard deviation of 0.548 eV can be calculated. If one instead calculates the average energy using the values by Weijun *et al.* [22], adding the theoretical values for levels above $n=8$, one gets 0.730 eV with a standard deviation of 0.547 eV. Normally this would allow a calculation of the standard error of the mean (the standard deviation divided by the square root of the total number of measurements), but in this case it is a bit more problematic since there are no data for the higher quantum states. Nevertheless, using the number 100 000 as an estimate of the statistics, a standard error of the mean of less than 0.002 eV is reached.

It was, furthermore, concluded that the population probability is independent of the electron energy in the keV energy domain and that the vibrational levels are stable. The latter is true since the H_2^+ molecule has no electric dipole moment and collisional depopulation is highly unlikely at our excitation time of about 1 s at a pressure of less then 10^{-11} mbar. Our averaging gives a value of 0.74 eV (in good agreement with the value presented in Ref. [13]). Considering the small uncertainties in the population distribution, we feel confident in claiming an uncertainty in ΔE of 10%. This uncertainty corresponds to 0.075 nu and has to be included in the mass analysis on the 0.1-nu level.

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