

## Dipole Moment of $\text{PH}^+$ and the Atomic Masses of $^{28}\text{Si}$ , $^{31}\text{P}$ by Comparing Cyclotron Frequencies of Two Ions Simultaneously Trapped in a Penning Trap

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(Received 2 August 2007; published 5 March 2008)

By trapping pairs of ions in a Penning trap, alternating each ion between large and small cyclotron orbits, we have measured the cyclotron frequency ratios  $^{12}\text{C}_2\text{H}_4^+ / ^{28}\text{Si}^+$ ,  $^{13}\text{C}_2\text{H}_2^+ / ^{28}\text{Si}^+$ ,  $^{28}\text{SiH}_3^+ / ^{31}\text{P}^+$ , and  $^{16}\text{O}_2^+ / ^{31}\text{PH}^+$ , all to <30 ppt precision. The  $^{16}\text{O}_2^+ / ^{31}\text{PH}^+$  data exhibit a bimodal distribution due to the polarizability of the  $\Lambda$ -doubling components of the  $\text{PH}^+$  ground state, from which we obtain the electric dipole moment of  $^{31}\text{PH}^+$ ,  $0.331(8) e a_0$ . Combined with other atomic mass measurements we also obtain improved values for  $m(^{28}\text{Si})$ ,  $27.976\,926\,535\,0(6)$  u, and  $m(^{31}\text{P})$ ,  $30.973\,761\,998\,9(9)$  u.

DOI: [10.1103/PhysRevLett.100.093002](https://doi.org/10.1103/PhysRevLett.100.093002)

PACS numbers: 32.10.Bi, 06.20.Jr, 07.75.+h, 33.15.Kr

The most precise measurements of atomic masses are obtained from ratios of cyclotron frequencies ( $f_c = qB/2\pi m$ ) of single ions, trapped in an ultrahigh vacuum in a cryogenic Penning trap [1]. An important limitation in the precision of such comparisons is the time variation of the magnetic field  $B$  between the measurements of  $f_c$  for each ion species. This has been addressed by careful stabilization of the magnetic field [2], or alternatively, by simultaneous measurement of the cyclotron frequencies of a pair of ions in a coupled magnetron orbit [3]. Otherwise, ions of the two species to be compared must be alternately introduced into the trap and their cyclotron frequencies measured, alternating between them as frequently as possible, e.g., see refs. [4,5]. As has been demonstrated in a 90 ppt comparison of the masses of  $\text{H}^-$  and the antiproton [6], it is also possible to simultaneously trap single ions of the two species in the same trap, but with one ion at the center of the trap—where its cyclotron frequency is measured—while the other ion is “parked” in a large-radius cyclotron orbit. The two ions are then interchanged by reducing the cyclotron radius of the outer ion while exciting that of the inner ion.

In this Letter we present a development of this alternating-cyclotron-orbit method to the measurement of cyclotron frequency ratios of pairs of heavy-ions [7]—for which perturbations due to ion-ion coulomb interaction are relatively larger—obtaining uncertainties below 30 ppt. We have applied this technique to measurements of the cyclotron frequency ratio  $^{16}\text{O}_2^+ / ^{31}\text{PH}^+$ —which show a bimodal distribution, with a relative separation of 1 ppb, due to the different polarizabilities of the  $\text{PH}^+$  ion in the two  $\Lambda$ -doubling components of its  $X^2\Pi_{1/2}$ ,  $J = 1/2$  ground state [8,9]. As first demonstrated for  $\text{CO}^+$  [10], the electric polarizability of an ion, and the motional electric field it experiences in its cyclotron motion, lead to a fractional shift in cyclotron frequency given approximately by  $\Delta f_c / f_c = -\alpha_{XX}(i)B^2/m$ , where  $\alpha_{XX}(i)$  is the polarizability in state  $|i\rangle$  for an electric field perpendicular

to the magnetic field. For a polar diatomic molecular ion in a cryogenic Penning trap, which has equilibrated with blackbody radiation into the lowest rotational levels of the ground electronic and vibrational state, the polarizability is dominated by an “orientation contribution” [8], which is proportional to the square of the molecule’s body-frame electric dipole moment. The constant of proportionality can be calculated using an effective Hamiltonian [11] with molecular parameters obtained from molecular spectra.

Our measurement of the cyclotron frequency shift between the two, opposite parity  $\Lambda$ -doubling components of  $\text{PH}^+$  is the second clear observation of polarizability shifts significantly affecting the cyclotron frequency of molecular ions in a Penning trap, and verifies the model introduced in Ref. [10]—but for a molecule with quite different electronic structure ( $^2\Pi_{1/2}$  vs  $^2\Sigma_{1/2}$ ). Together with the results of effective Hamiltonian calculations such as in Ref. [8], it can also be used to obtain a value for the electric dipole moment of  $\text{PH}^+$ , which can be compared with that obtained from molecular structure calculations. In contrast to neutral molecules where dipole moments can be obtained directly from Stark shifts, previous methods for measuring dipole moments of diatomic molecular ions do not match the precision claimed by current theory ( $\sim 0.01 e a_0$ ), or even, as in the case of  $\text{ArH}^+$ , differentiate between (earlier) conflicting theory (see Ref. [8] and references therein). Our result for  $\text{PH}^+$ , and the previous result for  $\text{CO}^+$ , provide the only accurate tests of dipole moment calculations of diatomic molecular ions.

The cyclotron frequency ratio (hereafter just referred to as the “ratio”) of  $^{16}\text{O}_2^+ / ^{31}\text{PH}^+$ , after corrections for the polarizability, can be used to obtain a precise mass of  $^{31}\text{P}$ . However, we observed that the mass of  $^{31}\text{P}$  obtained by this route was inconsistent, at the level of 0.1 ppb, with that obtained from measurements we made of the ratio  $^{31}\text{P}^+ / ^{28}\text{SiH}_3^+$ , combined with the mass of  $^{28}\text{Si}$  given in the atomic mass evaluation (AME) [12]. We therefore

decided to remeasure the mass of  $^{28}\text{Si}$ . We did this by measuring the ratios  $^{12}\text{C}_2\text{H}_4^+ / ^{28}\text{Si}^+$  and  $^{13}\text{C}_2\text{H}_2^+ / ^{28}\text{Si}^+$ , making use of previous precision measurements of the proton and  $^{16}\text{O}$  relative to  $^{12}\text{C}$  [12,13], and the simultaneous comparisons  $^{13}\text{C}_2\text{H}_2^+ / ^{14}\text{N}_2^+$  [3] and  $^{12}\text{C}^{16}\text{O}^+ / ^{14}\text{N}_2^+$  [10]. Combined with the value obtained from  $^{31}\text{P}^+ / ^{28}\text{SiH}_3^+$ , which provided a useful consistency check, we obtain a new atomic mass for  $^{28}\text{Si}$ , a factor of 3 more precise than the value in [12]. The atomic mass of  $^{28}\text{Si}$  is important since, combined with the mass of  $^{28}\text{Si}$ , in kg, and the density and lattice spacing of near-perfect silicon crystals, it enables Avogadro's number to be determined, possibly allowing the S. I. artifact kilogram to be replaced by a standard based on atomic mass. However at present the precision of the atomic mass measurement exceeds that of the current macroscopic measurements on silicon crystals by 3 orders of magnitude [14,15].

*Method.*—Details of our method and apparatus can be found in Refs. [7,16]. In brief, we obtain  $f_c$  of the inner ion by measuring its three normal mode frequencies,  $f_{ct}$ ,  $f_z$ , and  $f_m$ , corresponding to the trap-cyclotron, axial, and magnetron motions, respectively, and using the invariance theorem  $f_c^2 = f_{ct}^2 + f_z^2 + f_m^2$  [1]. We employ the “pulse-and-phase” technique [17], in which  $f_{ct}$  is obtained from the accumulated phase of a pulse-excited cyclotron motion, with an inner-ion radius  $\rho_{ci} \leq 150 \mu\text{m}$ . Only the axial motion is directly detected and damped—through image currents induced in a cryogenically-cooled (at 4.2 K) high- $Q$  inductor coupled to a dc SQUID. The radial motions are detected and damped by coupling to the axial motion using tilted quadrupolar rf drives. A single measurement of  $f_c$  with statistical precision of  $\sim 3 \times 10^{-10}$ , with contributions from variation in the magnetic field of  $\sim 10^{-9}/\text{h}$ , and uncertainty in the determination of cyclotron phase and  $f_z$  due to detector noise and voltage fluctuations, takes about 5 min. The ions are then interchanged by applying a series of cyclotron-axial coupling pulses to reduce the cyclotron radius of the outer ion, and a single cyclotron drive pulse to the inner ion. Starting with an outer-ion radius  $\rho_{ck}$  of 1.5 mm the time required for interchange is also about 5 min.

*Systematic shifts to the cyclotron frequency ratios.*—The coulomb interaction between the ions does perturb their normal mode frequencies relative to those in an ideal Penning trap. As discussed in [7], the main perturbations can be divided into static effects, equivalent to a perturbation of the trap electrostatic field by a “ring of charge” obtained by averaging over the outer-ion's motion, and dynamical effects, that are inversely proportional to the differences between the ions' mode frequencies. For the ion pairs considered here, the largest dynamical effect, a repulsion between the ions' axial frequencies that scales with ion separation as  $\rho_{ck}^{-6}$ , shifts the measured ratios  $< 10^{-11}$  for  $\rho_{ck} = 1.5 \text{ mm}$ . Regarding the static perturbations, we note that in the limit of small amplitudes for the

inner ion, the perturbation to the electrostatic potential is automatically allowed for by use of the invariance theorem, provided we measure  $f_{ct}$  and  $f_z$  together [18]. The remaining shifts to  $f_c$  can be considered as equivalent to a modification of the existing trap field imperfection parameters  $C_4$ ,  $C_6$ , etc. [1,7]. This leads to fractional shifts to the cyclotron frequency at the  $10^{-10}$  level. But, by obtaining cyclotron frequency ratios for “mass doublets”, we ensure that  $\rho_{ci}$ , the resulting axial amplitude  $a_{zi}$ , and also  $\rho_{ck}$  are, respectively, very similar for the two different ions, so these shifts nearly cancel in the ratio. The use of mass doublets also ensures that systematic effects due to “pushing” of the ion's axial frequency by interaction with the detector resonance, image charges in the trap electrodes, and possible differences in the ions' equilibrium positions [16], are all  $< 10^{-11}$ .

However, in our implementation of the pulse-and-phase technique there are small imbalances in  $\rho_{ci}$ ,  $a_{zi}$ , and  $\rho_{ck}$ , due to small detunings between the drives and respective motions [16]. We estimate these imbalances, and making use of the model of Ref. [7] and values for the trap field imperfections  $C_4$  (which we null to  $< 10^{-5}$ ),  $C_6 = 1.1(1) \times 10^{-3}$ , and the main relative magnetic imperfection  $B_2/B_0 = -5(3) \times 10^{-9} \text{ mm}^{-2}$ , we obtain small systematic corrections which we apply to our ratio data.

To verify our understanding of systematic effects due to ion-ion interaction we performed several tests, which we briefly summarize here. First, we measured the ratio  $^{12}\text{C}_2\text{H}_4^+ / ^{28}\text{Si}^+$  using deliberately imbalanced values of  $\rho_{ck}$  (1.0–1.7 mm) for the two ions to produce observable shifts in the measured ratio. These data were well described by the model specified by Eqs. (5) and (6) of Ref. [7]. Second, we verified that the respective  $\rho_{ck}$ 's were balanced, to within 2%, by measuring the shifts in  $f_z$  of the outer ion due to the large  $\rho_{ck}$  combined with  $B_2$  and  $C_6$ . Third, we obtained ratio data for  $^{12}\text{C}_2\text{H}_4^+ / ^{28}\text{Si}^+$  and  $^{13}\text{C}_2\text{H}_2^+ / ^{28}\text{Si}^+$  for a range of balanced  $\rho_{ck}$ , from 0.8 to 2.0 mm; see Fig. 1. We saw no systematic variation for the ratio over this range, for which ion-ion effects would be expected to vary by 2 orders of magnitude. (In this figure the gray bands indicate the maximum expected systematic shifts, corresponding to a detuning of the rf drives for one ion of 0.5 Hz and no detuning for the other.) Fourth, as an independent check, we measured the ratio  $^{13}\text{C}_2\text{H}_2^+ / ^{14}\text{N}_2^+$ , obtaining 0.999 421 460 858(29), in agreement with the ultrahigh-precision MIT result 0.999 421 460 888 (7) [3]. Finally, by looking for axial excitation from magnetron-axial coupling pulses applied to the inner ion, before and after the cyclotron frequency measurement, we verified that neither ion acquired significant magnetron motion.

*Determination of the dipole moment of  $\text{PH}^+$  and the masses of  $^{28}\text{Si}$ ,  $^{31}\text{P}$ .*—Our final cyclotron frequency ratios, obtained from data with  $\rho_{ck} \geq 1.5 \text{ mm}$ , are summarized in Table I. Each ratio is the weighted average of the results of more than 10 runs, each run lasting 10 to 14 h and with

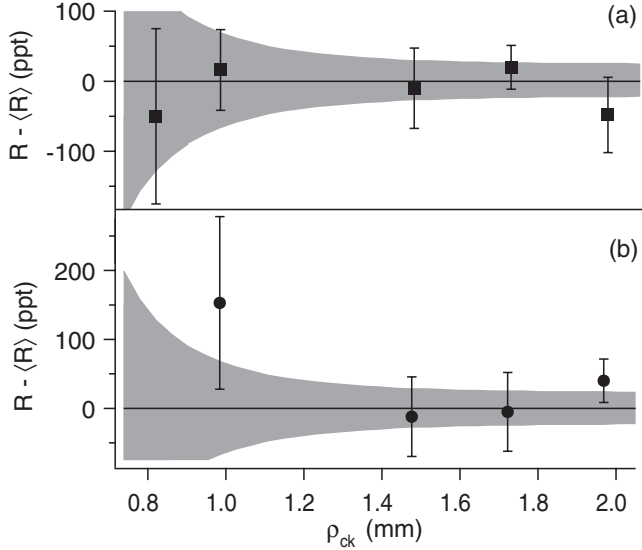


FIG. 1. Cyclotron frequency ratio measurements of (a)  $^{12}\text{C}_2\text{H}_4^+ / ^{28}\text{Si}^+$ , and (b)  $^{13}\text{C}_2\text{H}_2^+ / ^{28}\text{Si}^+$  for a range of parking radii,  $\rho_{ck}$ . Systematic corrections (but not uncertainties) for small imbalances in the amplitudes of the motion have been applied to the data. The solid line shows the final average ratio (see Table I); for gray bands, see text.

typically 50 alternating cyclotron frequency measurements. Estimated systematic corrections for ion-ion interaction, existing trap field imperfections, and for shifts to  $f_z$  due to interaction with the detector are also shown.

For  $^{16}\text{O}_2^+ / ^{31}\text{PH}^+$  there were 18 runs involving 8 different  $\text{PH}^+$  ions, with between 1 and 4 runs for each. Here we applied systematic corrections to each run and took the weighted average for all runs involving the same  $\text{PH}^+$  ion. As shown in Fig. 2 these data show a clear bimodal distribution which we interpret as due to the different polarizabilities of the  $\text{PH}^+$  ion in the opposite parity  $\Lambda$ -doubling components of the  $X^2\Pi_{1/2}$ ,  $J = 1/2$  ground state, which are separated by approximately 7 GHz [8,9]. We did not observe any change in the ratio for a given  $\text{PH}^+$  over several days, consistent with only the  $J = 1/2$  level of the ground electronic and vibrational state having significant occupation probability at 4.2 K, and an estimated mean transition time between the  $\Lambda$ -doubling states, due

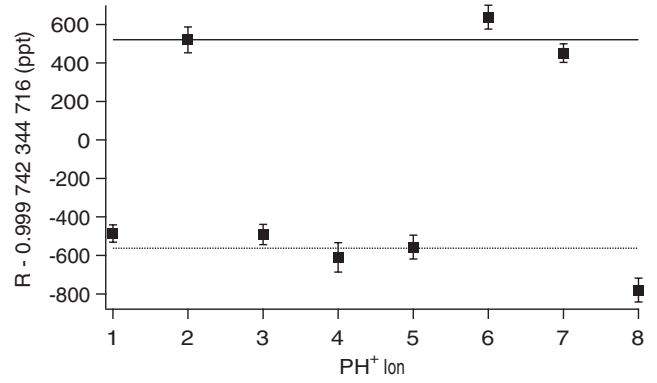


FIG. 2. Cyclotron frequency ratio measurements of  $^{16}\text{O}_2^+ / ^{31}\text{PH}^+$  showing shifts due to the polarizability of  $\text{PH}^+$ . Each point is the ratio measurement for a given  $\text{PH}^+$  ion. The solid (broken) line is the weighted average of the positively (negatively) shifted ratio, corresponding to the  $\Lambda^-$  ( $\Lambda^+$ ) state.

to thermal radiation, of  $\sim 10^3$  days. We treated the two regions separated by  $\sim 1 \times 10^{-9}$  individually and obtained the weighted average for each. From the difference,  $\Delta R = 1077(40)$  ppt, and the effective Hamiltonian calculations of Ref. [8] we obtain the dipole moment of  $\text{PH}^+$ . We find  $\mu = 0.331(8)ea_0$ , in good agreement with  $0.342(8)ea_0$  calculated using coupled-cluster molecular structure calculations, also given in Ref. [8].

Since the polarizabilities of the two  $\Lambda$ -doubling states are nearly equal and opposite, we can average the two measurements to cancel the polarizability shift—except for the small correction  $\Delta_{\text{pol}}$  obtained from the results of Ref. [8]—and obtain the ratio  $^{16}\text{O}_2^+ / ^{31}\text{PH}^+$  shown in Table I. [There is a small dependence of the polarizability of  $\text{PH}^+$  on the hyperfine state which is unresolvable in Fig. 2. We have estimated the resulting additional uncertainty in the ratio (error of  $\Delta_{\text{pol}}$  in Table I).]

The inverse mass ratios given in Table I can be converted into equations expressing the mass differences between neutral, isolated atoms. To do this we account for the mass of the missing electrons and the ionization and chemical binding energies [20,21]. The resulting mass difference equations, to be used in global mass evaluations, are shown in Table II.

TABLE I. Inverse mass ratios and systematic corrections for each ion pair.  $N$  is the number of runs.  $\Delta_{i-i}$ ,  $\Delta_{\text{trap}}$ ,  $\Delta_{f_z}$  and  $\Delta_{\text{pol}}$  are the systematic correction, with estimated uncertainty in parentheses, due to ion-ion interaction, trap field imperfections, shifts in  $f_z$  due to ion-detector interaction and differential voltage drift [19], and polarizability shifts (for  $\text{PH}^+$ ), respectively.  $\sigma_{\text{syst}}$  is the total systematic error and  $\sigma_{\text{stat}}$  is the corresponding statistical error (all in ppt).  $\langle R \rangle$  is the average inverse mass ratio after applying systematic corrections, with statistical and systematic uncertainties combined in quadrature.

Ion pair	$N$	$\Delta_{i-i}$	$\Delta_{\text{trap}}$	$\Delta_{f_z}$	$\Delta_{\text{pol}}$	$\sigma_{\text{syst}}$	$\sigma_{\text{stat}}$	$\langle R \rangle$
$^{12}\text{C}_2\text{H}_4^+ / ^{28}\text{Si}^+$	11	10(7)	12(9)	1(7)	0(0)	13	25	0.998 060 217 144(28)
$^{13}\text{C}_2\text{H}_2^+ / ^{28}\text{Si}^+$	15	0(7)	2(5)	-2(7)	0(0)	11	23	0.998 378 649 225(25)
$^{28}\text{SiH}_3^+ / ^{31}\text{P}^+$	11	-10(10)	-4(5)	0(12)	0(0)	16	24	0.999 140 653 321(29)
$^{16}\text{O}_2^+ / ^{31}\text{PH}^+$	18	-1(9)	-3(8)	-1(4)	-10(15)	20	18	0.999 742 344 690(27)

TABLE II. Mass difference equations corresponding to the ratios given in Table I. The statistical, systematic and total errors are shown in parentheses.

Mass Difference	Result (u)
$2(^{12}\text{C}) + 4(\text{H}) - (^{28}\text{Si})$	0.054 373 593 60 (69)(38)(79)
$2(^{13}\text{C}) + 2(\text{H}) - (^{28}\text{Si})$	0.045 433 199 86 (64)(30)(71)
$(^{31}\text{P}) - 3(\text{H}) - (^{28}\text{Si})$	-0.026 639 633 24 (74)(49)(89)
$2(^{16}\text{O}) - (^{31}\text{P}) - (\text{H})$	0.008 242 208 19 (58)(64)(86)

Here however, we choose to use these mass difference equations, the MIT results  $(^{13}\text{C}) + (\text{H}) - (^{14}\text{N}) = 0.008\,105\,862\,88(10)\text{ u}$  [3,22] and  $2(^{14}\text{N}) - (^{16}\text{O}) - (^{12}\text{C}) = 0.011\,233\,389\,32(42)\text{ u}$  [10], and the AME values [12] for  $m(^1\text{H}) = 1.007\,825\,032\,07(10)\text{ u}$  and  $m(^{16}\text{O}) = 15.994\,914\,619\,56(16)\text{ u}$ , to obtain three values for  $m(^{28}\text{Si})$  and one for  $m(^{31}\text{P})$ . These results are compared to previous measurements in Table III.

Our three values for  $m(^{28}\text{Si})$  are in good agreement. A weighted average, in which we propagate the errors in the reference masses, and, conservatively, we treat the systematic errors from the similar ratios  $^{12}\text{C}_2\text{H}_4^+ / ^{28}\text{Si}^+$  and  $^{13}\text{C}_2\text{H}_2^+ / ^{28}\text{Si}^+$  as completely correlated, gives  $m(^{28}\text{Si}) = 27.976\,926\,534\,96(62)\text{ u}$ . This differs from the value given in Ref. [12] by 2.5(2.0) nu, and is a factor of 3 more precise. Our mass of  $^{31}\text{P}$  is a factor of 200 more precise than that given in Ref. [12].

We have developed a mass comparison technique in which two ions are simultaneously confined in a Penning trap and alternated between large and small cyclotron orbits, allowing mass comparisons to be made at  $2 \times 10^{-11}$  fractional precision. We have used this technique to measure the electric dipole moment of the molecular ion  $\text{PH}^+$ , providing a useful test of molecular theory. Our work further demonstrates that electric polarizability must be

TABLE III. Masses for  $^{28}\text{Si}$  and  $^{31}\text{P}$  obtained from the different ratios compared with previous values.

Atom	Ion pair/Ref.	Atomic Mass (u)
$^{28}\text{Si}$	$^{12}\text{C}_2\text{H}_4^+ / ^{28}\text{Si}^+$	27.976 926 534 68 (88)
	$^{13}\text{C}_2\text{H}_2^+ / ^{28}\text{Si}^+$	27.976 926 534 78 (86)
	$^{28}\text{SiH}_3^+ / ^{31}\text{P}^+$	27.976 926 535 89 (134)
	Ref. [5]	27.976 926 533 90 (980)
	Refs. [4,12]	27.976 926 532 46 (194)
$^{31}\text{P}$	$^{16}\text{O}_2^+ / ^{31}\text{PH}^+$	30.973 761 998 86 (92)
	Ref. [7]	30.973 761 999 70 (610)
	Ref. [12]	30.973 761 63 (20)

taken into account of in precision atomic mass comparisons using molecular ions.

We thank D. E. Pritchard, S. Rainville, J. K. Thompson, and J. M. Brown for assistance and encouragement, and acknowledge support from NSF-PHY0354741 and the NIST PMG Program.

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