

High-Precision Mass Measurements of Hydrogen-Like $^{24}\text{Mg}^{11+}$ and $^{26}\text{Mg}^{11+}$ ions in a Penning Trap

On the need of precise mass values of hydrogen-like ions in g -factor measurements of bound electrons

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Abstract. For the determination of the bound-electron g factor in hydrogen-like heavy ions the mass of the ion is needed at a relative uncertainty of at least 1 ppb. With the SMILETRAP Penning trap mass spectrometer at the Manne Siegbahn Laboratory in Stockholm several mass measurements of ions with even-even nuclei at this level of precision have been performed so far, exploiting the fact that the mass precision increases linearly with the ion charge. Measurements of masses of the hydrogen-like ions of the two Mg-isotopes ^{24}Mg and ^{26}Mg are reported. The masses of the hydrogen-like ions are 23.979 011 054 (17) u and 25.976 562 347 (32) u, corresponding to the atomic masses 23.985 041 687 (17) u and 25.982 592 979 (32) u, respectively. The possibility to use these two isotopes for the first observation of an isotope effect in the bound-electron g factor in hydrogen-like heavy ions is discussed.

PACS. 07.75.+h Mass spectrometers and related techniques – 21.10.Dr Binding energies and masses – 31.30.Jv Relativistic and quantum electrodynamic effects in atoms and molecules – 32.10.Bi Atomic masses, mass spectra, abundances, and isotopes

1 Introduction

In this paper the importance of accurate atomic mass values for the determination of the bound-electron g factor in hydrogen-like heavy ions with even-even nuclei is emphasized. This fact will be evident by considering a few equations relevant for the experimental determination of these g factors.

The energy of an electron in a magnetic field B is given by $\mu_B B$. The energy difference when the electron has spin up or spin down relative to the direction of B in a Penning trap is given by

$$(\mu_B)\uparrow - (\mu_B)\downarrow = \frac{1}{2} [g \mu_b - (-g \mu_b)] B = g \mu_b B . \quad (1)$$

Here, $\mu_B = e\hbar/2m$ is the Bohr magneton and m is the electron mass.

Transitions between the two spin states are induced by a microwave field resonant with the Larmor precession frequency ω_L of the bound electron

$$\hbar\omega_L = g \frac{e\hbar}{2m} B . \quad (2)$$

The magnetic field B is calibrated by using the ion cyclotron frequency

$$\omega_c = 2\pi\nu_c = \frac{qe}{M} B , \quad (3)$$

where qe/M is the charge-to-mass ratio of the ion. Thus, the g factor of the bound electron can be expressed as

$$g = 2 \frac{\omega_L}{\omega_c} \cdot \frac{qe/M}{e/m} . \quad (4)$$

The charge-to-mass ratio (e/m) of the electron was determined in a Penning trap with a precision of 2.2×10^{-9} [1, 2] and recently by Beier *et al.* with a 3 times higher precision [3]. From equation 4 it is evident that when determining the value of g to an uncertainty in the low ppb range, the mass M of the hydrogen-like ion has to be known at an uncertainty ≤ 1 ppb.

Recently the g factor of the hydrogen-like $^{12}\text{C}^{5+}$ ion was measured by the group of Quint *et al.* [4] with a relative uncertainty of 2.5×10^{-9} . The same group is now determining the g factor of $^{16}\text{O}^{7+}$ and plan to measure the one of $^4\text{He}^{1+}$ and $^{24}\text{Mg}^{11+}$ [5]. In the first three cases the masses of the hydrogen-like ions are known with an uncertainty $\ll 1$ ppb; in $^{12}\text{C}^{5+}$ by the definition of the atomic mass unit u and in the cases of $^{16}\text{O}^{7+}$ [6] and $^4\text{He}^{1+}$ [7] by very accurate measurements of the Seattle group. For heavier hydrogen-like ions like $^{24}\text{Mg}^{11+}$ one has to rely upon new accurate mass measurements. It is here that the Penning trap mass spectrometer SMILETRAP [8] enters the picture. It is connected to the electron beam ion source CRYISIS for production of highly charged ions [9, 10]. In this work considerably improved atomic masses of ^{24}Mg and ^{26}Mg will be presented and their possible use for the observation of an isotope effect in the bound-electron g factor will be discussed.

2 Mass determination in SMILETRAP

The procedure of mass measurements in SMILETRAP has been described in detail previously [8]. Thus, only a shorter

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description is given here, sufficient for the measurements of the ^{24}Mg and ^{26}Mg masses.

2.1 Ion production and transportation of ions to the precision Penning trap

The electron beam ion source CRYISIS is connected to a high mass-resolution isotope separator that on-line can produce singly charged ions of any element to be injected into CRYISIS for charge breeding [9,10]. With the present performance highly charged hydrogen-like ions up to about $^{40}\text{Ca}^{19+}$ can be produced. Since the electron beam in CRYISIS can trap a certain maximum amount of charges, low abundances of some isotopes of interest can be compensated by a longer injection time of the singly charged ions. Tests have shown that it is possible to produce sufficient amounts of highly charged ions for mass determinations in SMILETRAP even for isotopes having an abundance as low as 0.1%.

For Mg the confinement time in CRYISIS was 1 s, the ion injection time 0.4 s, the electron energy 14.4 keV and the electron current 138 mA. A typical charge spectrum of ^{26}Mg ions obtained with the given parameters is shown in Fig. 1. A bunch of highly charged ions with a pulse width of 100 ms is transported to the SMILETRAP area, located at a distance of about 20 m, using conventional beam elements. The experimental setup of SMILETRAP is shown in Fig. 2. Before the ions under investigation enter the cylindrical retardation trap, here named pretrap, a charge selection is done in a double focusing magnet. In the pretrap the ions are retarded from the transportation

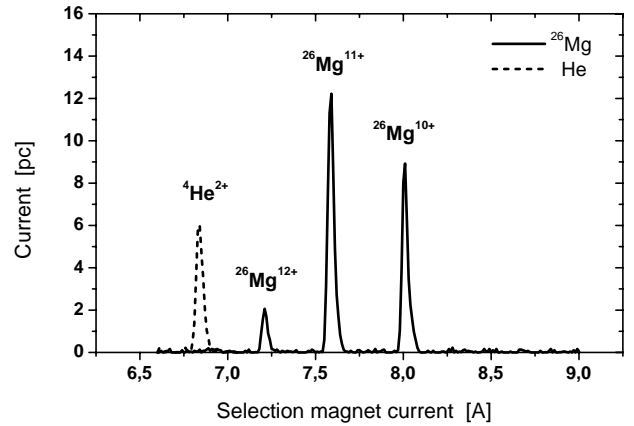


Fig. 1. Charge spectrum of $^{26}\text{Mg}^{q+}$ ions recorded in the focal plane of the double focusing magnet used for charge selection. The Mg-ions were cooled in CRYISIS by helium ions but in normal runs no detectable $^4\text{He}^{2+}$ ions were present. The dotted peak shows the presence of these ions at an extreme concentration of helium gas in CRYISIS.

energy of $3.4 \cdot q$ keV to ground within 30 ms, afterwards accelerated again to 1 keV and then finally transported to the hyperboloidal precision Penning trap. An aperture with a diameter of 1 mm prevents ions with too large initial radial energies to enter the precision trap. Before entering this trap the ions are again retarded and subject to an evaporation process by lowering the trap voltage from 5 to 0.1 V leaving only the coldest ions in the trap. On average not more than 1–2 ions are left in the trap after this procedure.

2.2 Mass determination of ^{24}Mg and ^{26}Mg

The mass measurement in a Penning trap is based on the determination of the cyclotron frequency given in Eq. 3. The magnetic field is in the case of SMILETRAP 4.7 T,

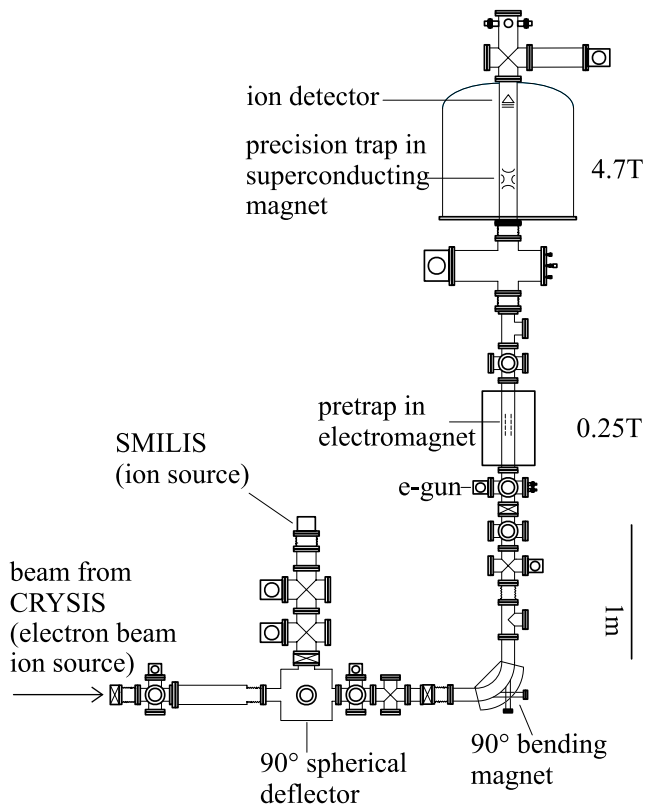


Fig. 2. The SMILETRAP Penning trap mass spectrometer connected to the ion source CRYISIS at the Manne Siegbahn Laboratory, Stockholm University. The overview sketch shows the 90° bending magnet used for charge selection, the first Penning trap in an electromagnet for ion retardation and bunched ejection of the ion under investigation, and the precision trap installed in a superconducting solenoid for the actual mass determination. For the time-of-flight detection of the cyclotron resonances the ion detector on top of the apparatus is used.

which is calibrated by measuring the well-known mass of a reference ion. Here this is H_2^+ , produced in the pretrap by bombarding the rest gas with 3.4 keV electrons. Since the mass resolving power $m/\Delta m = \nu/\Delta\nu$ increases linearly with the charge of the ion one gains a factor of 11 in precision by using Mg^{11+} ions as compared to singly charged ions.

The homogeneous magnetic field confines the ions in a plane perpendicular to B . In order to prevent the ions to disappear in the axial direction of B , two electrodes (end caps) are added as well as a segmented ring, used for quadrupole excitation at the truth cyclotron frequency ν_c . The end caps and also the ring have hyperboloidal shapes, which create an axial electrostatic quadrupole field. In this field the ions move in three independent modes; an axial motion with frequency ν_z , and the so-called magnetron and reduced cyclotron frequencies, ν_- and ν_+ , respectively. It can be shown [11] that:

$$\nu_c = \nu_- + \nu_+ . \quad (5)$$

By shining in an azimuthal quadrupole RF-field with the frequency ν_c into the precision trap, the two radial motions are coupled together [11] and excited. When the ions are released from the precision trap after an excitation time of 1 s, they are sent to a micro-channel-plate (MCP) detector located about 50 cm above the trap. In resonance, the radial energy of the ion is increased. In the inhomogeneous-magnetic-field region between the center of the trap and the MCP detector, the magnetic moment associated with this radial energy then leads to an increase in axial energy, which results in a pronounced time-of-flight minimum [12]. In Fig. 3 this can be seen for the case of $^{24}\text{Mg}^{11+}$. The cyclotron frequency ν_c is obtained by fitting a Gaussian to the center of the resonance.

Usually 21 frequency steps around the resonance frequency are used. This frequency scanning procedure takes about 25 s and was done in this investigation four times (usually twice) for each of the two ion species; the refer-

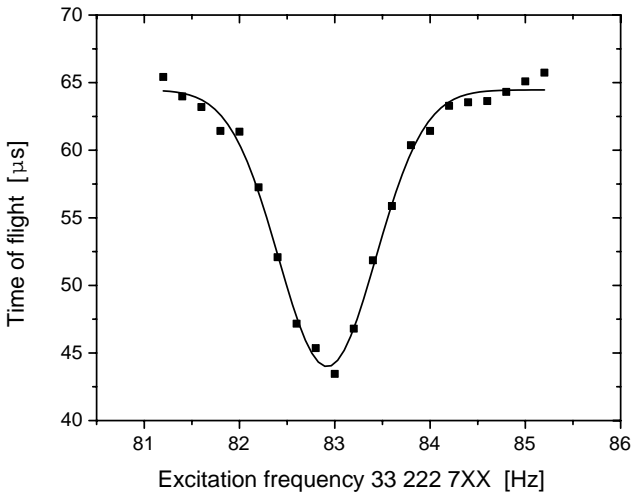


Fig. 3. The time-of-flight resonance for $^{24}\text{Mg}^{11+}$ after 7 hours data collection. The central part of the resonance is approximated with a Gaussian (solid line) in the data evaluation.

reference ion H_2^+ and the ion under investigation $^{24,26}\text{Mg}^{11+}$. The change of ion species takes only about 1 s. The cycle time is thus slightly less than 4 min and the relevant observable, the frequency ratio between the two ion species, is almost a direct observation. If the two frequency measurements are performed in similar ways, the systematic uncertainties in the frequency ratio cancel to a great extent. This is, in particular, the case for ions which have the same value of q/A . Therefore, one should compare the cyclotron frequency of the reference ion and the ion under investigation which, as much as possible, have the same values of q/A . The Mg^{11+} ions are close to this requirement.

Since the ion species are changed so rapidly the magnetic field does not change in a detectable way during the measurements of the cyclotron frequencies of H_2^+ and Mg^{11+} . Therefore, one can divide the frequencies given by

Eq. 3 and the final result is then expressed as the ratio of the cyclotron frequencies of two ions:

$$M(\text{Mg}^{11+}) = m_{\text{ref}}(\text{H}_2^+) \cdot \frac{\nu_c(\text{Mg}^{11+})}{\nu_{c,\text{ref}}(\text{H}_2^+)} \quad (6)$$

where the reference mass $m(\text{H}_2^+) = 2.015\,101\,487\,04\,(26)\,\text{u}$ is a mass obtained by multiplying the proton mass $m_p = 1.007\,276\,466\,89\,(13)\,\text{u}$ [13] by a constant $2.000\,544\,600\,49$ [8]. In order to get the mass M_0 of the neutral atom one has to correct for the mass $q \cdot m$ of the missing q electrons and their binding energies (E_B):

$$M_0 = M + q \cdot m - E_B/c^2. \quad (7)$$

For light ions such as Mg^{q+} , E_B can be calculated by summing the experimentally determined ionization energies [14] for different ion charges. In this way E_B is found to be $3488.395\,\text{eV}$ for $q = 11$. The electron mass m [1, 2] and E_B are known so accurately that they contribute to an uncertainty in the mass M_0 of the Mg-isotopes by much less than 0.1 ppb.

2.3 Systematic uncertainties

In addition to statistical uncertainties there are systematic uncertainties leading to frequency shifts. The main ones are discussed in detail in [8] and are due to

- relativistic mass increase,
- q/A -asymmetry of the two ion species,
- having more than one ion trapped,
- impurity ions coming from CRYISIS.

The relativistic corrections are usually less than 0.2 ppb and can be done to an uncertainty of about 0.1 ppb

either by using the time-of-flight values before and after excitation or by using a retardation technique [8]. ▀

The largest mass uncertainty originates from frequency shifts when the two ion species have different values of q/A . In [8] it is shown that the q/A -asymmetry for two ion species having $q/A = 0.5$ and 0.25 , respectively, introduces a maximum frequency shift of 1 ppb. This uncertainty is mainly due to limited statistics in this estimate. Thus, in the case of ^{24}Mg and ^{26}Mg with $q/A = 0.46$ and 0.42 , respectively, this effect should contribute to a maximum frequency shift of 0.16 and 0.32 ppb, respectively.

The ion number dependence can be investigated by studying the shift of the resonance frequency for different numbers of trapped ions [8], using a graph like the one shown in Fig. 4. In the evaluation of the mass values, only data from 1 and 2 trapped ions at the same time were taken into account.

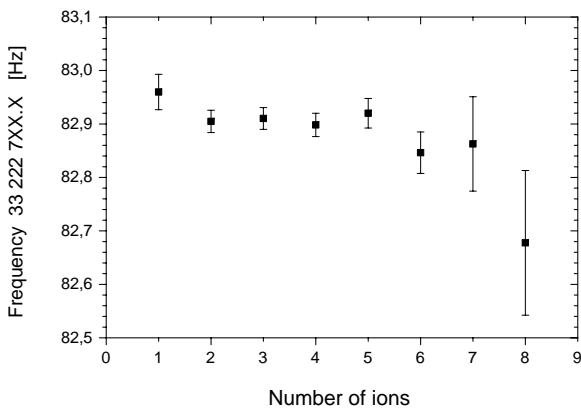


Fig. 4. The frequency shift of $^{24}\text{Mg}^{11+}$ ions as a function of the number of trapped ions. About 10% of the total data is included. In the off-line evaluation only data from 1 and 2 trapped ions were accepted.

The impurity ions are of two kinds; ions that arise from charge exchange of the highly charged ions with the rest gas and ions coming from CRYISIS having the same q/A as the ion of interest. At the pressure in SMILETRAP of about 5×10^{-12} mbar the charge exchange process can be neglected. The latter impurity ions are the worst ones but the amount can be checked by driving the dipole frequency of the highly charged ions and studying the time-of-flight spectra of all ions. The excited ions are then entirely resolved from the impurity ions and thus the relative impurity concentration can be obtained from which a limit for a frequency shift can be concluded [8]. Therefore, one has to be careful when investigating ions for which $q/A = 0.5$, because there is, in particular, a risk for having present contaminant ions like $^4\text{He}^{2+}$, $^{14}\text{N}^{7+}$, $^{16}\text{O}^{8+}$ originating from CRYISIS.

3 Results and possible improvements

In Table 1 the systematic uncertainty budget as well as the statistical uncertainty for the two hydrogen-like Mg^{11+} ions is given. The measurement time was four days for $^{24}\text{Mg}^{11+}$ and two days for $^{26}\text{Mg}^{11+}$. Therefore the difference in the statistical uncertainty.

In Table 2 the masses of the directly measured $11+$ ions as well as the atomic masses of the two Mg isotopes are listed. They were measured at a total uncertainty of 0.71 ppb ($^{24}\text{Mg}^{11+}$) and 1.23 ppb ($^{26}\text{Mg}^{11+}$), respectively. Thus, SMILETRAP was able to improve the precision by more than one order of magnitude in comparison to the accepted mass values [15]. Another factor of about 3 may

Table 1. Uncertainty budget for the masses of the hydrogen-like ions $^{24}\text{Mg}^{11+}$ and $^{26}\text{Mg}^{11+}$. The larger statistical uncertainty for ^{26}Mg is due to a lower amount of data.

Uncertainty	$^{24}\text{Mg}^{11+}$	$^{26}\text{Mg}^{11+}$
	[ppb]	[ppb]
Reference mass	0.18	0.18
Electron binding energies	0.10	0.10
Relativistic mass increase	0.23	0.23
Ion number dependence	0.25	0.25
q/A -asymmetry	0.16	0.32
Contaminant ions	0.10	0.10
Magnetic field drift	0.06	0.06
Total systematic uncertainty	0.45	0.52
Statistical uncertainty	0.55	1.12
Total uncertainty	0.71	1.23

Table 2. The masses of the neutral Mg isotopes as compared with previously accepted values and the masses of the hydrogen-like Mg ions.

Isotope	This work	Accepted value [15]
^{24}Mg	23.985 041 687 (17)	23.985 041 87 (17)
^{26}Mg	25.982 592 979 (32)	25.982 593 00 (26)
$^{24}\text{Mg}^{11+}$	23.979 011 054 (17)	
$^{26}\text{Mg}^{11+}$	25.976 562 347 (32)	

be gained for ions which are q/A doublets by extending the excitation time in the trap to 2 s and by applying Ramsey technique for the determination of the cyclotron frequency. For the latter, three excitation times interrupted with periods of no excitation will be used, a technique,

which was already tested in SMILETRAP [8] with H_2^+ and highly charged ions ($^{16}\text{O}^{7+}$ and $^{76}\text{Se}^{22+}$). However, this method has to be developed to a routine technique. The higher precision is likely to require a measuring time of about one week per isotope.

4 Possible observation of an isotope effect in g factor measurements

In the introduction the importance of accurate masses for the determination of the bound-electron g factor was emphasized. This is even more important when comparing the g factors of different ions of the same element.

An interesting question related to the sensitivity of the g -factor experiment by Häffner *et al.* [4] is whether there would be a measurable difference between the g factors of two isotopes of the same element, mainly due to the fact that in the heavier isotope the electron wave function is located slightly closer to the nucleus and thus feeling a somewhat stronger Coulomb field. A suitable isotope pair for a test could, for example, be the two investigated magnesium isotopes ^{24}Mg and ^{26}Mg .

Labelling the two ions with 1 and 2 having the masses $M(1)$ and $M(2)$ where $M(1) > M(2)$, the isotope effect in the g factor can be defined as

$$\Delta g = \frac{g(2) - g(1)}{g(1)} = \frac{g(2)}{g(1)} - 1, \quad (8)$$

or using Eq. 4:

$$\Delta g = \frac{\omega_L(2)}{\omega_c(2)} \cdot \frac{\omega_c(1)}{\omega_L(1)} \cdot \frac{M(1)}{M(2)} - 1. \quad (9)$$

Note that Δg is independent of the electron mass.

Table 3. Accurately measured masses of some selected isotope pairs suitable for measuring the isotope effect on the bound-electron g factor.

Atom	Abund. (%)	Mass (u)	Uncert. (ppb)
^{20}Ne	90.9	19.992 440 175 4	0.12 [16]
^{22}Ne	8.8	21.991 385 115	0.90 [8]
^{24}Mg	78.6	23.985 041 687	0.72 [this work]
^{26}Mg	11.3	25.982 592 979	1.23 [this work]
^{28}Si	92.3	27.976 926 532 4	0.07 [16]
^{30}Si	3.1	29.973 770 223	1.20 [15]
^{36}Ar	0.4	35.967 545 105	0.42 [8]
^{40}Ar	99.6	39.962 383 122	0.10 [16]

In the GSI/Mainz experiment it was demonstrated [4], that for hydrogen-like ions the ratio ω_L/ω_c can be determined at an uncertainty of 0.3 ppb in a Penning trap. Thus, the total uncertainty in the ratio of these two quantities in Eq. 9 is about 0.42 ppb. In order to exploit this potentially higher precision the masses of the two ion species should be known at least to the same precision. This is in principle possible by improving some of the mass values given in Table 3, where the most accurate masses for isotope pairs are listed.

The size of the isotope effect was estimated by the calculation of the g -factor correction term g_{recoil} due to the finite mass of the nucleus coming from relativistic calculations of strong-field QED [17]. This so called recoil contribution is the dominant ion mass dependent term

contributing to the g factor of the bound electron and reads

$$g_{\text{recoil}} = (Z\alpha)^2 \left[\left(\frac{m}{M} \right) - (1+Z) \left(\frac{m}{M} \right)^2 \right] + (Z\alpha)^2 \left(\frac{\alpha}{\pi} \right) \left[-\frac{1}{3} \left(\frac{m}{M} \right) + \frac{3-2Z}{6} \left(\frac{m}{M} \right)^2 \right]. \quad (10)$$

From this equation the isotope effect in the g factor for the two Mg isotopes results in the value $\Delta g = 6.7 \times 10^{-9}$, and can thus be observed, due to the improved mass values reported here. Similar estimates of the isotope effect were done for other pairs of even-even isotopes. It should be mentioned, that there are two additional isotope effects besides the recoil effect. They are due to the finite nucleus size and due to the nuclear polarization, i.e. the virtual excitation of nuclear states. Both effects are about an order of magnitude smaller than the recoil contribution [17].

As shown in Fig. 5 the isotope effect Δg in the bound-electron g factor for hydrogen-like ions is about $\Delta g \approx 7 \times 10^{-9}$ for the mass number difference $\Delta A = 2$. When ΔA is larger than 2, the isotope effect is larger. In the case of Sn the biggest isotope effect can be expected ($\Delta g \approx 3.2 \times 10^{-8}$). In Figure 6 the accepted mass values [15] of these atoms with even-even nuclei suitable for g -factor measurements of hydrogen-like ions are summarized. As can be seen, the masses of several light ions are already known with high precision, while most heavier ions have much larger mass uncertainties. However, they can be improved to the required precision of ≤ 1 ppb. At present the most promising isotope pairs for checking Eq. 9 are those ones given in Table 3.

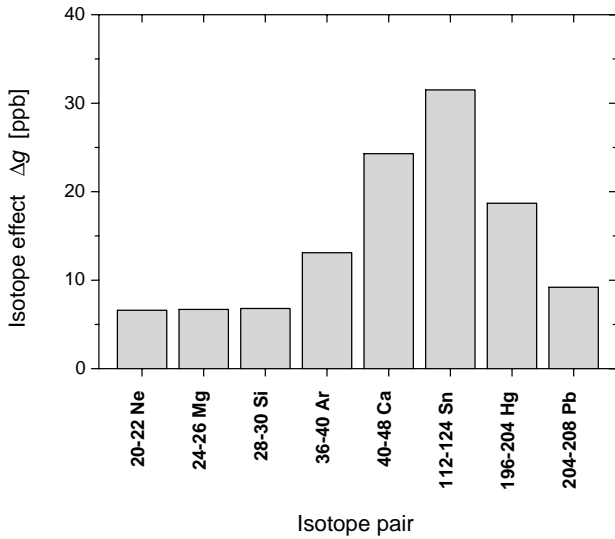


Fig. 5. The isotope effect Δg as calculated from Eq. 10 for certain isotope pairs ordered after increasing atomic number.

In the case of Sn the biggest isotope effect in the bound-electron g factor in hydrogen-like ions can be expected.

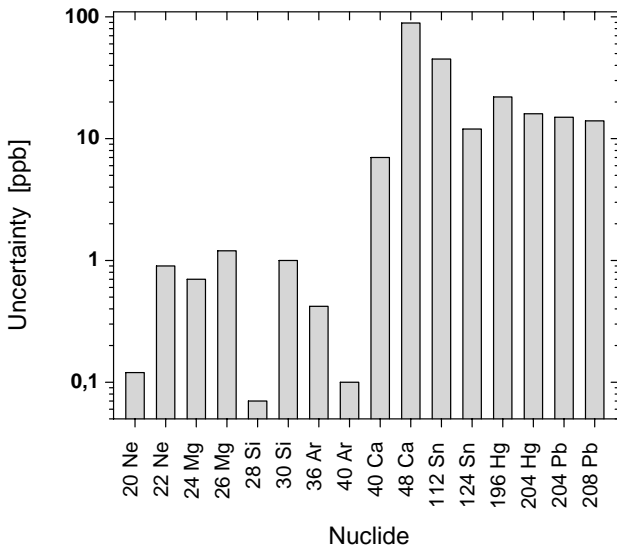


Fig. 6. Present mass uncertainties in some selected isotope pairs. The most precise mass values have been determined with Penning traps. All the accepted mass values with large uncertainties can in principle be improved to the precision needed.

It can be questioned why one should try to detect the isotope effect in such a small correction as the bound-state g factor. The isotope effect of g factors may admittedly not add to answering questions in fundamental physics. However, it should again be emphasized that the ratio of any two g factors is independent of the electron mass and can be determined with a precision of < 0.5 ppb if the ratio of the two ion masses is known to within 0.2 ppb. Furthermore, the reduced mass enters the different terms in the bound-electron g -factor expressions differently. A measurement with different isotopes can therefore be used to test the theoretical descriptions more detailed.

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