Double-resonance *g*-factor measurements by quantum jump spectroscopy

W. Quint, B. Nikoobakht*, U. D. Jentschura*

Gesellschaft für Schwerionenforschung (GSI), 64291 Darmstadt, Germany

* Max-Planck-Institut für Kernphysik, 69029 Heidelberg, Germany and Institut für Theoretische Physik, Universität Heidelberg, 69120 Heidelberg, Germany

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With the advent of high-precision frequency combs that can bridge large frequency intervals, new possibilities have opened up for the laser spectroscopy of atomic transitions. Here, we show that laser spectroscopic techniques can also be used to determine the ground-state g-factor of a bound electron: Our proposal is based on a double-resonance experiment, where the spin state of a ground-state electron is constantly being read out by laser excitation to the atomic L shell, while the spin flip transitions are being induced simultaneously by a resonant microwave field, leading to a detection of the quantum jumps between the ground-state Zeeman sublevels. The magnetic moments of electrons in light hydrogen-like ions could thus be measured with advanced laser technology. Corresponding theoretical predictions are also presented.

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Recently, there has been a dramatic progress in the precision laser spectroscopy of atomic transitions, with uncertainties on the order of 10^{-14} for two-photon transitions in hydrogen [1] and even 10^{-17} for ultraviolet (UV) electric-quadrupole transitions in the mercury ion [2] (statistical effects led to a limitation on the order of 10^{-16} for the evaluation of the latter measurement). By contrast, microwave measurements of the bound-electron g-factor in hydrogen-like ions [3-5] have been restricted to a comparatively low level of accuracy, namely in the range of 10^{-10} , where the most accurate values have been obtained for bound electrons in hydrogen-like carbon ¹²C⁵⁺ and oxygen ¹⁶O⁷⁺ (for an introductory reviews on bound-electron g-factors and various related experimental as well as theoretical techniques, see [6-8]). It is tempting to ask if the accuracy gap between the two categories of measurements might leave room for improvement of the g-factor determination, as both measurements investigate the properties of bound electrons. More specifically, the question arises if the additional "channels" provided by laser excitation among the discrete states of the bound system, and the additional possibilities for the laser cooling of ions (following the experimental methods described in Refs. [9, 10]), can be used as auxiliary devices to improve the accuracy of the g-factor determination via quantum jump spectroscopy. We also note that double-resonance techniques for stored ions have already been shown to open up attractive experimental possibilities with re-

spect to hyperfine transitions as well as electronic and nuclear g-factors [11-13].

The bound-electron (Landé) g_j -factor for an electron bound in an ion with a spinless nucleus is the proportionality constant relating the Zeeman energy ΔE in the magnetic field B (directed along the z axis) and the Larmor precession frequency $\omega_{\rm L}$ to the magnetic spin projection $m_j = -\frac{1}{2}, \frac{1}{2}$ onto that same z axis. In natural units ($\hbar = c = \epsilon_0 = 1$), we have

$$\Delta E = m_i \, \omega_{\rm L} = m_i \, g_i \, \mu_{\rm B} \, B, \tag{1}$$

where $\mu_{\rm B}=-e/2m_e$ is the Bohr magneton, expressed in terms of the electron charge e and the electron mass m_e . Deviations from the Dirac–Breit [14] prediction $g_j(1S)=2\left(1+2\sqrt{1-(Z\alpha)^2}\right)/3$ are due to quantum electrodynamic (QED), nuclear and other effects.

The purpose of this note is to answer the following question: "Is it possible to apply ultra-high precision atomic laser spectroscopy to bound-electron g-factor measurements?". Our answer will be affirmative.

In contrast to the continuous Stern–Gerlach effect [15], and complementing a recent proposal for a high-precision measurement of the g-factor in a highly charged ion [16], the current proposal is based on a Penning trap and will be studied here in conjunction with the hydrogen-like helium ion ${}^{4}\text{He}^{+}$, which seems to be well suited for an experimental realization in the near future. The magnetic field strength B in the Penning trap can be calibrated via a measurement of the cyclotron

frequency ω_c of the trapped ion, and the Landé g-factor for the bound electron is determined by the relation

$$g_j = 2 \left(Z - 1 \right) \frac{m_e}{m_{\rm ion}} \frac{\omega_{\rm L}}{\omega_c}, \tag{2}$$

where the electron-ion mass ratio $m_e/m_{\rm ion}$ is an external input parameter and Z is the nuclear charge number.

In a Penning trap, a single ${}^{4}\text{He}^{+}$ ion is confined by a strong homogeneous magnetic field B in the plane perpendicular to the magnetic field lines and by a harmonic electrostatic potential in the direction parallel to the field lines [17]. The three eigenmotions of a stored ion are the trap-modified cyclotron motion (frequency ω_{+}), the axial motion (frequency ω_{z}), and the magnetron motion (frequency ω_{-}). The free-space cyclotron frequency $\omega_{c} = qB/m_{\text{ion}}$ of an ion with charge q can be determined from the three eigenfrequencies by [18]

$$\omega_c^2 = \omega_+^2 + \omega_z^2 + \omega_-^2. \tag{3}$$

Experimentally, the eigenfrequencies of the stored ion can be measured by non-destructive detection of the image currents which are induced in the trap electrodes by the ion motion. Measurements on the level of $\delta\omega_c/\omega_c=$ $=7\cdot10^{-12}$ are achieved [19-21] by careful anharmonicity compensation of the electrostatic trapping potential, optimizing the homogeneity and temporal stability of the magnetic field close to the Penning trap's center, and cooling the motional amplitudes of the single trapped ion to low temperatures. Further optimization of experimental techniques should make it possible to reach an accuracy of (better than) $\delta\omega_c/\omega_c=10^{-12}$. In our proposed g-factor measurement, advantage could be taken of the fact that two frequencies of the same particle are measured simultaneously (cyclotron vs. spin-flip), whereas in a mass measurement, the cyclotron frequencies of two different particles have to be determined.

In the following, we concentrate on the ${}^4\mathrm{He}^+$ system, where the total angular momentum is equal to the total electron angular momentum J. In the presence of the magnetic field B in the Penning trap, the Zeeman splitting of the electronic ground state $1S_{1/2}$ of the ${}^4\mathrm{He}^+$ ion is given by Eq. (1). Correspondingly, the excited state $2P_{3/2}$ is split into four Zeeman sublevels $\Delta E = m_j \, g_j (2P_{3/2}) \, \mu_{\mathrm{B}} \, B$, with $m_j = \pm \frac{1}{2}, \pm \frac{3}{2}$. The Landé g-factor can be obtained easily according to a modified Dirac equation which forms a basis for bound-state analysis [22]

$$g_j(2P_{3/2}) = \frac{4}{3} + \frac{\alpha}{3\pi} - \frac{2}{15}(Z\alpha)^2,$$
 (4)

where we take into account the leading QED and relativistic contributions and use Z = 2 for the relativis-

tic term of order $(Z\alpha)^2$. Suppose now that one single ${}^4\mathrm{He^+}$ ion in the Penning trap is prepared in the Zeeman sublevel $m_j=+\frac{1}{2}$ of the electronic ground state $1S_{1/2}$. Narrow-band UV electromagnetic radiation with σ^+ polarization and angular frequency $\omega_{\mathrm{UV}}=2\pi\times 9.87\cdot 10^{15}\,\mathrm{Hz}$ drives the Lyman- α transition $1S_{1/2}$ $(m_j=+\frac{1}{2})$ $\Leftrightarrow 2P_{3/2}$ $(m_j=+\frac{3}{2})$, see Fig.1. This is a closed cycle because decay by emission of a

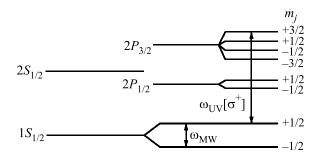


Fig.1. Laser-microwave double-resonance excitation scheme using circularly polarized UV light for excitation of the $1S_{1/2} \Leftrightarrow 2P_{3/2}$ transition and a microwave field for driving the spinflip transition $1S_{1/2}$ $(m_j=+\frac{1}{2}) \Leftrightarrow 1S_{1/2}$ $(m_j=-\frac{1}{2})$

fluorescence photon is only possible to the initial state $1S_{1/2}$ ($m_j=+\frac{1}{2}$) (if one ignores one-photon ionization into the continuum). Due to the short lifetime of the upper state $\tau(2P_{3/2})\approx 99.7\,\mathrm{ps}$, the fluorescence intensity of $[2\,\tau(2p_{3/2})]^{-1}\approx 5.01\cdot 10^9\,\mathrm{photons/s}$ under saturation conditions makes it possible to detect a single trapped ion with high sensitivity [23]. The Rabi frequency of the UV transition is given by $\Omega_{\mathrm{Rabi}}=1.308\cdot 10^7\,\mathrm{Hz}\,\sqrt{I_{\mathrm{UV}}},$ where I_{UV} is measured in units of W/cm².

Building a continuous-wave (cw) laser that operates at the Lyman- α transition of ${}^{4}\text{He}^{+}$, with a wavelength of 30.37 nm, is certainly not a trivial task. However, a cw laser operating at the corresponding Lyman- α transition for atomic hydrogen, with $\lambda = 121.56 \, \text{nm}$, has already been demonstrated [24]. A possible pathway is higher-harmonic generation which has recently been described in Ref. [25] and leads to a pulsed UV excitation with a high repetition rate and a potentially discontinuous probing of the Zeeman ground-state sublevel. Note that a discontinuous probing of the ground-state sublevels does not inhibit the quantum jump detection scheme as outlined below. Groundwork for a detailed analysis of the dynamics of a pulsed excitation scheme in a very much analogous atomic system has recently been laid in Ref. [26]; in principle, one only has to ensure that the light intensity of the Lyman- α source during a single laser pulse is sufficient to discern the presence or absence of fluorescence.

During excitation of $_{
m the}$ transition $1S_{1/2}$ $\left(m_{j}=+\frac{1}{2}\right)$ \Leftrightarrow $2P_{3/2}$ $\left(m_{j}=+\frac{3}{2}\right)$ and detection of the corresponding fluorescence photons, a microwave field with frequency $\omega_{\rm MW}$ in resonance with the spinflip transition $1S_{1/2}$ $(m_j = +\frac{1}{2}) \Leftrightarrow 1S_{1/2}$ $(m_j = -\frac{1}{2})$ in the electronic ground state is irradiated on the single trapped ⁴He⁺ ion (Fig.1). Successful excitation of the spinflip transition results in an instantaneous stop of the fluorescence intensity, because the lower Zeeman level $1S_{1/2}$ $(m_j = -\frac{1}{2})$ is not excited by the narrow-band Lyman- α radiation. A quantum jump is thus directly observed with essentially 100% detection efficiency [27]. A second spinflip $1S_{1/2} (m_j = -\frac{1}{2}) \rightarrow 1S_{1/2} (m_j = +\frac{1}{2})$ restores the fluorescence intensity. A plot of the quantum jump rate versus excitation microwave frequency at $\omega_{\rm MW} \approx \omega_{\rm L}$ yields the resonance spectrum of the Larmor precession frequency. The cyclotron frequency ω_c , which also enters Eq. (2), is measured simultaneously by nondestructive electronic detection of the image currents induced in the trap electrodes [5].

Table 1

Nuclear charge radius	$1S \Leftrightarrow 2P_{3/2}$ frequency
$\langle r^2 angle^{1/2} = 1.673(1) { m fm}$	9 868 722 559.240(237) MHz
$\langle r^2 angle^{1/2} = 1.680(5) { m fm}$	9 868 722 558.650(477) MHz

Absolute transition frequencies of ⁴He⁺ relevant to the excitation scheme given in Fig.1. The Zeeman splitting is excluded

While the UV laser light drives the transition $1S_{1/2}$ $(m_j=+\frac{1}{2}) \Leftrightarrow 2P_{3/2}$ $(m_j=+\frac{3}{2})$, the absorption of an additional photon can take place, resulting in ionization through the channel $2P_{3/2}$ $(m_j=+\frac{3}{2}) \Rightarrow \varepsilon D_{5/2}$ $(m_j=+\frac{5}{2})$, where εD are electronic continuum states (see Fig.2). For this process, we obtain an

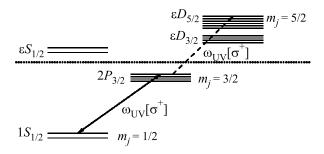


Fig.2. Schematic representation of the excitation scheme including ionization channels. The dotted line represents the ionization continuum threshold, and εL_j are electronic continuum states

ionization cross section of $1.631 \cdot 10^{-23} \,\mathrm{m}^2$. This leads to an ionization rate of $\gamma_{\rm i} = 2.495 \cdot 10^{-2} \, {
m s}^{-1} \times I_{\rm UV},$ where $I_{\rm UV}$ is the laser intensity measured in units of W/cm², corresponding to a depletion of the $2P_{3/2}$ $(m_i = +\frac{3}{2})$ state with a time-dependent exponential $\exp(-\gamma_i t)$. Here, we use the notational conventions of Ref. [26]. In principle, since the ionization rate is proportional to the laser intensity, whereas the Rabi frequency is only proportional to its square root, it might be preferable to work at reduced laser intensities in order to increase the lifetime of the hydrogen-like charge state of the ion. However, at an incident typical laser intensity of 100 W/cm², the lifetime of ⁴He⁺ is 0.401 s against ionization, and this has to be compared to a Rabi frequency of $1.308 \cdot 10^8$ Hz. The ${}^4\text{He}^+$ ion has about 10^8 Rabi cycles before it is ionized, and so the ionization channel does not limit the feasibility of the measurement at all.

Finally, we take notice of the ac Stark shift of the 1S-2P transition due to non-resonant levels, which is $0.0968 \,\mathrm{Hz} \times I_{\mathrm{UV}}$, with I_{UV} given in W/cm². The ac Stark shift of the UV transition affects the two groundstate Zeeman levels slightly differently, but the relative shift of the spinflip transition frequency between them is a fourth-order effect and is suppressed with respect to the ac Stark shift by a factor of $\omega_{\rm L}/\omega_{\rm UV}$ < 10^{-4} and thus negligible on the level 10^{-12} in units of the microwave frequency, at a typical laser intensity of 100 W/cm². Also, experimental procedures for previous q-factor measurements [4, 5] have included an extrapolation to zero intensity of the microwave fields, and the same can be done with the driving UV laser field in the proposed measurement scheme. Alternatively, one can perform the excitation of the Lyman- α and the spinflip transitions in a time sequence, thus eliminating any systematic uncertainties of the g-factor determination related to the intensity of the UV laser light.

In order to lay a theoretical ground for the evaluation of the $^4\mathrm{He}^+$ measurement, we present theoretical predictions for the transition frequencies and for the g-factor. According to the recent compilations [31-33], the ground state $^4\mathrm{He}^+$ Lamb shift values are $\mathcal{L}(1S)=107692.522(228)$ MHz for the "old" value [34] of the nuclear charge radius $\langle r^2 \rangle^{1/2}=1.673(1)$ fm and $\mathcal{L}(1S)=107693.112(472)$ MHz for the "new" value of the charge radius [35], which is $\langle r^2 \rangle^{1/2}=1.680(5)$ fm. (The uncertainty estimate for the "old" value has given rise to discussions, see Ref. [31].) For the $2P_{3/2}$ states, the Lamb shift is independent of the current uncertainty in the nuclear radius on the level of one kHz and reads $\mathcal{L}(2P_{3/2})=201.168$ MHz (see Tables 3 and 4 of Ref. [33]). Using a proper definition of the Lamb shift

Table 2

g -factor contribution ($^4\mathrm{He^+}$)	
$\alpha^{-1} = 137.035999070(98)$	$\alpha^{-1} = 137.03599911(46)$
Dirac eigenvalue	
1.999 857 988 825 2(2)	$1.999\ 857\ 988\ 825\ 3(9)$
Finite nuclear size	
0.000 000 000 002 3	0.000 000 000 002 3
One-loop QED	$(Z\alpha)^0$
0.002 322 819 466 0(17)	$\begin{array}{c} 0.002 \ 322 \ 819 \ 465 \ 4(76) \\ (Z\alpha)^2 \end{array}$
0.000 000 082 462 2	$0.000\ 000\ 082\ 462\ 2 \ (Zlpha)^4$
0.000 000 001 976 7	0.000 000 001 976 7
	h.o.,SE
$0.000\ 000\ 000\ 035\ 1(2)$	$0.000\ 000\ 000\ 035\ 1(2)$
	h.o.,VP-EL
0.000 000 000 002 0	0.000 000 000 002 0
	h.o.,VP-ML
0.000 000 000 000 2	0.000 000 000 000 2
$\geq ext{two-loop QED}$	$(Zlpha)^0$
-0.000 003 515 096 9(3)	$ \begin{array}{c ccccccccccccccccccccccccccccccccccc$
-0.000 000 000 124 8	$-0.000\ 000\ 000\ 124\ 8$ $(Zlpha)^4$
0.000 000 000 002 4(1)	0.000 000 000 002 4(1)
Recoil	m/M
0.000 000 029 198 5	0.000 000 029 198 5
Radiative recoil	$(m/M)^2$
-0.000 000 000 025 3	-0.000 000 000 025 3
Hadronic/weak interaction	
0.000 000 000 003 4 Total	0.000 000 000 003 4
2.002 177 406 727 1(17)	2.002 177 406 726 5(77)

Individual contributions to the 1S bound-electron g-factor for $^4\mathrm{He}^+.$ In the labeling of the corrections, we follow the conventions of Ref. [28]. The abbreviations used are as follows: "h.o." stands for a higher order contribution, "SE" for a self energy correction, "VP-EL" for the electric-loop vacuum-polarization correction, and "VP-ML" for the magnetic-loop vacuum-polarization correction. The value of $\alpha^{-1}=137.035~999~070(98)$ is the currently most accurate value from Ref. [29], whereas the value of $\alpha^{-1}=137.035~999~11(46)$ is the 2002 CODATA recommended value [30]

as given, e.g., in Eq. (10) of Ref. [33], we then obtain the transition frequencies as given in Table 1.

The ground-state g_j -factor can be described naturally in an intertwined expansion in the QED loop expansion parameter α and the electron-nucleus interaction strength $Z\alpha$ [28]. We follow the conventions of Ref. [28] and take into account all corrections that are relevant at the 10^{-12} level of accuracy (see Table 2).

The entry for the " $(Z\alpha)^0$ one-loop QED" is just the Schwinger term $\alpha/2\pi$ and it carries the largest theoretical uncertainty, because of the uncertainty in the fine-structure constant α itself [36, 37].

In this note, we attempt to formulate a proposal by which ultra-accurate q-factor measurements in hydrogen-like systems with low nuclear charge number might be accessible to laser spectroscopic techniques. Within the next decade, it is realistic to assume that the necessary requirements for experiments will be provided that fully profit from both the electric coupling of the electron (via optical electric-dipole allowed Lyman- α transitions) and from the magnetic coupling of the electron (via spin-flip transitions among the Zeeman sublevels of the ground state, see Fig.1). The accuracy of the measurement of the free-electron g-factor has recently been increased to a level of $7.6 \cdot 10^{-13}$ [38, 29]. Within our proposed setup, an accuracy on the level of $10^{-12} \dots 10^{-13}$ seems to be entirely realistic for boundelectron g-factors in hydrogen-like ions with a low nuclear charge number. It might be very beneficial if the extremely impressive, ultra-precise new measurement of the free-electron g-factor [29] could be supplemented by a potentially equally accurate measurement of the bound-electron g-factor in the near future, as an alternative determination of the fine-structure constant is urgently needed in conjunction with an improved determination of the electron mass.

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- 36. This observation illustrates that if two g_j -factors could be measured to sufficient accuracy in the low-Z region, then the fine-structure constant could be inferred in addition to the electron mass. Namely, we would have two equations of type (2) and two unknowns: the fine-structure constant and the electron mass. See also [37].
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