Mon. Not. R. Astron. Soc. **373,** L41–L44 (2006) doi:10.1111/j.1745-3933.2006.00239.x

Isotopically resolved calibration of the 285-nm Mg I resonance line for comparison with quasar absorptions

E. J. Salumbides,★ S. Hannemann, K. S. E. Eikema and W. Ubachs★
'Laser Centre, Vrije Universiteit, De Boelelaan 1081, 1081 HV Amsterdam, the Netherlands

Accepted 2006 August 29. Received 2006 August 15; in original form 2006 July 15

ABSTRACT

We have performed high-precision spectroscopy on the $3s^2\ ^1S \rightarrow 3s3p\ ^1P$ first resonance line of Mg I at 285 nm. This measurement is motivated by the recent studies comparing observed quasar absorption lines with laboratory measurements in the search for a possible variation of the fine structure constant α. The Mg transition is excited using the up-converted radiation of an injection-seeded Ti : sapphire pulsed laser system, while the frequency of the continuous wave seed light is measured using a femtosecond frequency comb laser. The line positions of the isotopes are fully resolved and have accuracies better than 8×10^{-9}. Using terrestrial abundances for the Mg isotopes $^{24}Mg : {}^{25}Mg : {}^{26}Mg$ of 78.99 : 10.00 : 11.01, we determine the composite line wavenumber to be 35 051.2808(2) cm^{-1}, which can be compared to the astronomical observations where the isotopic shifts are unresolved.

Key words: atomic data – methods: laboratory – techniques: spectroscopic – quasars: absorption lines – ultraviolet: general.

1 INTRODUCTION

There is a resurgent interest in the possibility of a variation of fundamental constants, like the fine structure constant $\alpha = e^2/4\pi\epsilon_0\hbar c$ (Webb et al. 1999, 2001) and the proton-to-electron mass ratio $\mu = M_p/m_e$ (Reinhold et al. 2006). The possible variation of such constants has far-reaching implications for modern physical theories – see for example Uzan (2003) for a recent review. The magnitude of such variations and their effect on the wavelengths of spectral lines of atoms, ions and molecules are expected to be minute, so that it is necessary to employ extremely precise spectroscopic techniques to probe for such effects. One approach is to measure very narrow transitions in atoms or ions over time intervals of months to years at accuracies of $\sim 10^{-15}$ yr^{-1} (Fischer et al. 2004). Another approach is to compare spectra taken over extremely large time intervals, comparing spectra in cold absorbing interstellar clouds at high redshift in the line of sight of quasars (providing look-back times of ∼10 billion years) and present-day measurements in the laboratory. The advantage of the latter approach is that the absolute differences are greater, thus the demands on the spectral accuracy are less stringent compared with those of the first approach.

The most accurate method for probing possible changes in α, based on a comparison of quasar absorption data and laboratory spectra, is the many-multiplet (MM) method (Dzuba, Flambaum & Webb 1999). Performing the MM analysis on data from quasar absorption systems that are observed with the Keck High Resolution Echelle Spectrometer telescope in the northern hemisphere have indicated that α was smaller at earlier epochs, with the most recent result of the variation at $\Delta\alpha/\alpha = (-0.57 \pm 0.10) \times 10^{-5}$ (Webb et al. 2003). However, when other groups (Quast, Reimers & Levshakov 2004; Srianand et al. 2004) applied the same method to data gathered from the Very Large Telescope (VLT) Ultraviolet and Visual Echelle Spectrograph in the southern hemisphere, they found a null result. Murphy et al. (2001, 2003) extensively studied possible sources of systematic errors that could explain this discrepancy; however, none could be found to date. The reliability of the results obtained from the MM method can be improved by maximizing the number of transitions that are included in the analysis, as well as improving the laboratory frequency calibrations (at zero redshift) so that those can be considered exact for the purpose of the comparisons. Berengut et al. (2006) have provided a list of spectral lines that require better laboratory calibrations for inclusion in the MM method.

The Mg I $3s^2\ ^1S \rightarrow 3s3p\ ^1P$ transition at 285 nm is one of several lines of atomic or ionic magnesium that are used in the MM method. Beverini et al. (1990) have previously calibrated this line using continuous wave (cw) excitation of an atomic beam, resolving all the naturally occurring isotopes. Pickering, Thorne & Webb (1998), and more recently Aldenius, Johansson & Murphy (2006), performed calibrations using Fourier transform spectroscopy (FTS) without resolving the isotopic substructure due to Doppler broadening. Isotope shifts have been determined by Hallstadius (1979) and Le Boiteux et al. (1988), as well as Beverini et al. (1990).

The ^{25}Mg isotope has a nuclear spin of 5/2, and, from the hyperfine structure constants obtained by Kluge & Sauter (1974), the total splitting between the three hyperfine components can be calculated to be less than 50 MHz. However, the hyperfine structure cannot be

★E-mail: ejsalum@nat.vu.nl (EJS); wimu@nat.vu.nl (WU)

resolved since the lifetime of the 3s3p ^{1}P state is 1.99 ± 0.08 ns (Lurio 1964), corresponding to a natural linewidth of about 80 MHz.

Following our recent measurements of the Mg I $3s^2\ ^1S \rightarrow 3s4p\ ^1P$ transition near 202 nm (Hannemann et al. 2006), we present here our frequency calibration of the Mg I $3s^2\ ^1S \rightarrow 3s3p\ ^1P$ transition with the isotopes fully resolved. Using a similar system consisting of a Ti : sapphire-based pulsed laser system, we performed laser-induced fluorescence (LIF) spectroscopy on a Mg atomic beam and obtained frequency calibrations with a stabilized femtosecond frequency comb laser.

2 EXPERIMENTAL METHOD

A pulsed laser source is used because of the relative ease and efficiency in the up-conversion process towards the deep ultraviolet (UV) region. We employed an almost identical system to that of the Mg I $3s^2\ ^1S \rightarrow 3s4p\ ^1P$ measurements except that the third harmonic of the fundamental is used instead of the fourth harmonic. Here, we present a concise description of the experimental setup and procedures. For an extensive and detailed discussion of the experiment as well as the resulting uncertainty evaluations, we refer to our previous publication (Hannemann et al. 2006).

The laser system consists of a 10-Hz repetition rate Ti : sapphire pulsed oscillator, which is injection-seeded with radiation from a tunable cw Ti : sapphire laser. The oscillator output pulse is centred around the cw-seed frequency at ∼856 nm. The required 285-nm UV excitation light is produced after two frequency up-conversion stages in Beta Barium Borate crystals, involving second-harmonic generation and a subsequent frequency-mixing process to produce the third harmonic.

We excite an effusive Mg atomic beam that is skimmed to reduce the transversal velocity spread, resulting in a residual Doppler width of about 15 MHz for the 285-nm transition. The laser-induced fluorescence, which is at the same wavelength as the excitation, is collected with a photomultiplier tube. In order to minimize the Doppler shift due to an imperfect alignment of the UV excitation and Mg atomic beams, an interferometric technique is employed. A Sagnac interferometer is set up by splitting the UV beam and then recombining the paths, thus creating exactly counter-propagating beams that cross the atomic beam. Successive measurements for the each of the counter-propagating beams are performed separately and, by taking the mean of the spectral line positions obtained from either of the beam paths, the resulting Doppler error is reduced to <300 kHz.

Frequency calibration is performed on the cw-seed radiation using a stabilized femtosecond frequency comb (Holzwarth et al. 2000; Jones et al. 2000). The frequency comb is referenced to a rubidium-clock standard and approaches a stability of the order of 10^{-12} over a 10-s averaging time (Witte et al. 2004). However, there can be a shift between the centre frequency of the pulse and that of the cw-seed radiation, caused by frequency excursions during the temporal evolution of the pulse due to the frequency chirp in the Ti : sapphire gain medium and by dynamical mode-pulling effects in the pulse oscillator cavity. In order to characterize these effects, we employ procedures similar to those used by White et al. (2003, 2004) to assess chirp phenomena in an injection-seeded and pulsed optical parametric oscillator. Using this method, we measure and correct for the frequency offset between the cw-seed radiation and the amplified pulse generated for every laser shot.

3 RESULTS AND DISCUSSION

A recording of the ^{24}Mg line with a Lorentz fit superimposed is displayed in Fig. 1. A recorded resonance consists of about 2500 data points, with each point representing a 0.1-s measurement. The observed linewidth is about 120 MHz, which is close to the expected width taking into account the natural linewidth of 80 MHz, the laser linewidth of about 35 MHz and the residual Doppler width of 15 MHz. The signal-to-noise ratio (SNR) is low because we reduce the intensity of the UV excitation in the measurements to the lowest possible value in order to avoid power shifts. We typically use peak intensities of less than 100 W cm^{-2} (pulse length ∼20 ns), and we do not detect any power shifts at higher laser peak intensities (∼1 kW cm^{-2}). An isotope-resolved overview recording of the Mg I $3s^2\ ^1S \rightarrow 3s3p\ ^1P$ transition is shown as an inset in Fig. 1.

We list the absolute line positions that we obtained for the different isotopes in Table 1. An uncertainty budget for the calibration is presented in Table 2 and we refer to Hannemann et al. (2006) for a more detailed discussion of the uncertainty estimates. The lower accuracy that we obtain for the present measurements compared with that of the 202-nm line can be attributed to the larger natural linewidth, the worse SNR since resonance fluorescence is detected, and the more severe frequency chirp in the Ti : sapphire pulsed oscillator at the fundamental wavelength of ∼856 nm. The first two factors increase the uncertainty in the determination of the line centre (errors iii and iv), while the last factor increases the error in the correction of the cw–pulse frequency offset (error vi).

Our value for the ^{24}Mg line position is in good agreement with that obtained by Beverini et al. (1990) of 35 051.272(5)(4) cm^{-1}, where the first number in parentheses refers to the experimental uncertainty and the second number refers to the confidence in the reference standard used for calibration. The isotope shifts are also listed in Table 1, along with some previous determinations (Hallstadius 1979; Le Boiteux et al. 1988; Beverini et al. 1990). It is satisfying to see that the values obtained from different measurement techniques are consistent with each other. In addition, the results of the *ab initio* calculations of Berengut, Flambaum & Kozlov (2005) for the isotope shifts are in good agreement with the experimental results. The relative uncertainty for the ^{24}Mg line position is $<5 \times 10^{-9}$, while it is $<8 \times 10^{-9}$ for the less abundant isotopes. The present values constitute a more than an order of magnitude improvement

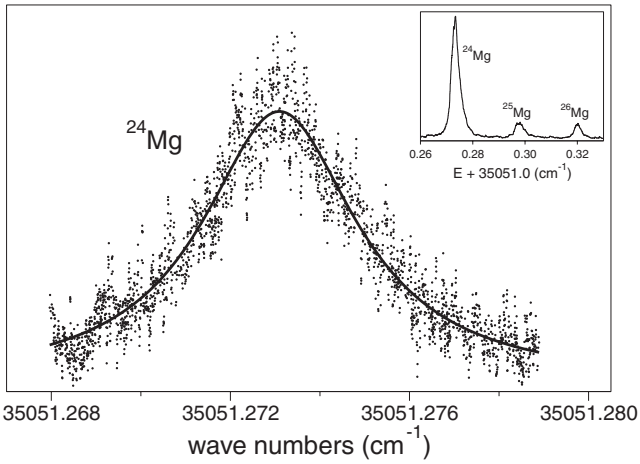

Figure 1. A recording of the ^{24}Mg resonance with the fitted Lorentzian profile. Each data point represents a 0.1-s measurement for every laser shot. Inset: an overview spectrum of the Mg $3s^2\ ^1S \rightarrow 3s3p\ ^1P$ transition with the ^{24}Mg, ^{25}Mg and ^{26}Mg isotopes resolved.

Table 1. The line positions of the Mg isotopes for the 3 s^2 ^{1}S → 3 s 3 p ^{1}P transition. The resulting isotope shifts are compared with previous determinations by Beverini et al. (1990), Hallstadius (1979) and Le Boiteux et al. (1988), as well as the theoretical calculations from Berengut et al. (2005). The uncertainties are shown in parentheses.

Isotope	Absolute position (cm^{-1})	Relative shift from ^{24}Mg (MHz) This experiment	Beverini[a]	Hallstadius	Le Boiteux	Berengut[b]
^{24}Mg	35 051.27311(17)	–	–	–	–	–
^{25}Mg	35 051.29784(25)	744.1(7.5)	743.8(3.0)	728 (12)	720(27)	740(30)
^{26}Mg	35 051.32015(25)	1413.8(7.5)	1415.3(5.0)	1412(21)	1391(30)	1420(30)

[a]Beverini et al. (1990) give a ^{24}Mg position of 35 051.272 cm^{-1}, where an experimental uncertainty of 0.005 cm^{-1} and a calibration confidence of 0.004 cm^{-1} are specified. [b]Theoretical results.

Table 2. Uncertainty budget for the absolute calibration at the deep-UV energy scale. The total uncertainty is obtained by taking the quadrature sum of the errors i–iv and linear summation of the result with the errors v–vi.

	Error	MHz
i	first-order Doppler	0.3
ii	frequency comb	<0.1
iii	fit error ^{24}Mg	2.0
iv	fit error ^{25}Mg, ^{26}Mg	4.5
v	AC-Stark	0.5
vi	cw–pulse offset	2.5
	total ^{24}Mg	5.0
	total ^{25}Mg, ^{26}Mg	7.5

on the absolute frequencies of the isotope-resolved transition. The relative isotope shift determinations of Beverini et al. (1990) are slightly more accurate than our values since they employed cw-laser excitation.

We use the weighted sum of the isotopic line positions using the terrestrial isotopic abundances to obtain the centre of gravity of the transition in order to compare our measurements with the composite line obtained by Aldenius et al. (2006), by Pickering et al. (1998) and by Risberg (1965), as shown in Table 3. [Rosman & Taylor (1998) provide values for the terrestrial Mg isotopic abundances of 78.99(4) per cent for ^{24}Mg, 10.00 (1) per cent for ^{25}Mg and 11.01(3) per cent for ^{26}Mg.] The centre of gravity that we obtain is in agreement with the value obtained by Aldenius et al. (2006) and that of Risberg (1965). It has been pointed out by Aldenius et al. (2006) that there is a systematic offset of -0.003 cm^{-1} between their results and those of Pickering et al. (1998) due to the use of new and improved calibration values for the (Ar II) reference standard in the more recent FTS measurements. If the results of Pickering et al. (1998) are recalibrated with respect to the improved reference standard, then the values are consistent.

Table 3. Comparison of the transition centre of gravity with previous determinations where the isotopes were not resolved. We use the terrestrial isotopic abundance ratio ^{24}Mg : ^{25}Mg : ^{26}Mg of 78.99 : 10.00 : 11.01, to weight the isotopic line centres for comparison with the composite line positions given by Aldenius et al. (2006), Pickering et al. (1998) and Risberg (1965). The values are given in cm^{-1}.

	Composite line
This experiment	35 051.2808(2)
Aldenius et al.	35 051.280(2)
Pickering et al.	35 051.277(1)
Risberg	35 051.26(4)

The isotopic structure is not resolved in the quasar absorption spectra with a typical linewidth of 0.78 cm^{-1} for this transition (corresponding to a velocity spread of 6.6 km s^{-1}). In general, the lineshapes and linewidths should be taken into account in synthesizing a composite line for comparison with the absorption spectra. However, for the large linewidths in the quasar absorption data, the difference from our centre-of-gravity value can be neglected. Hence the centre-of-gravity value that we provide here could be used directly in the comparison with quasar absorption spectra.

There is an implicit assumption in the MM method that the terrestrial isotopic abundances also hold in the absorption systems studied. The possibility that a variation in isotopic abundance might mimic an α-variation has been discussed in a number of papers (Murphy et al. 2001; Ashenfelter, Mathews & Olive 2004; Kozlov et al. 2004; Fenner, Murphy & Gibson 2005). Systematic shifts attributable to variations in isotopic abundance are more pronounced for the light species such as Mg, because the isotope shifts are larger relative to the expected spectral line shift induced by a changing α. For the Mg I 3s^2 ^{1}S → 3s4p ^{1}P transition, the isotopic shifts are more than twice that of the 3s^2 ^{1}S → 3s3p ^{1}P transition; however, the sensitivity coefficients to the variation of α of both transitions are almost equal (Kozlov et al. 2004). It might be possible to evaluate or correct for any systematic effect related to the isotopic abundance by comparing the spectral shifts of both transitions. The highly accurate spectroscopic data produced in our laser experiments may assist in resolving these issues.

4 CONCLUSION

We have reinvestigated the Mg I 3s^2 ^{1}S → 3s3p ^{1}P transition and obtained the line positions of the ^{24}Mg, ^{25}Mg and ^{26}Mg isotopes with unprecedented accuracies of better than 8×10^{-9}. This transition is seen in quasar absorption spectra and is used in the many-multiplet method to probe for a possible variation in the fine structure constant α. Using the terrestrial isotopic abundance ratio ^{24}Mg : ^{25}Mg : ^{26}Mg of 78.99 : 10.00 : 11.01, we evaluate the centre of gravity of the transition to be 35 051.2808(2) cm^{-1}. This value for the composite line including all three isotopes is in agreement with the values obtained by Aldenius et al. (2006) and the older measurements of Risberg (1965); it also agrees with that of Pickering et al. (1998) if the corrections for the FTS absolute calibrations are taken into account. Our more accurate absolute frequency determination of the ^{24}Mg line is in agreement with the previous measurements of Beverini

et al. (1990). The isotope shifts that we obtain are also consistent with the previous determinations of Hallstadius (1979), Le Boiteux et al. (1988) and Beverini et al. (1990). Furthermore, the experimental results agree very well with the recent *ab initio* calculations of the isotope shifts by Berengut et al. (2005). The transition centre of gravity that we provide can be used in the comparison with the quasar absorption spectra, where the isotopic structure is unresolved. In addition, our isotopically resolved frequency determinations of the $3s^2\ ^1S \rightarrow 3s3p\ ^1P$ and $3s^2\ ^1S \rightarrow 3s4p\ ^1P$ (Hannemann et al. 2006) transitions in Mg I might be helpful in resolving the issue of isotopic abundance evolution in the MM analysis which mimics an α-variation.

ACKNOWLEDGMENTS

We thank the Netherlands Foundation for Fundamental Research of Matter (FOM) for financial support.

REFERENCES

Aldenius M., Johansson S., Murphy M. T., 2006, MNRAS, 370, 444
Ashenfelter T., Mathews G. J., Olive K. A., 2004, Phys. Rev. Lett., 92, 041102
Berengut J. C., Flambaum V. V., Kozlov M. G., 2005, Phys. Rev. A, 72, 044501
Berengut J. C., Dzuba V. A., Flambaum V. V., Marchenko M. V., Webb J. K., 2006, Class. Quant. Grav. 21, 4771
Beverini N., Maccioni E., Pereira D., Strumia F., Vissani G., Wang Y.-Z., 1990, Opt. Commun., 77, 299
Dzuba V. A., Flambaum V. V., Webb J. K., 1999, Phys. Rev. Lett., 82, 888
Fenner Y., Murphy M. T., Gibson B. K., 2005, MNRAS, 358, 468
Fischer M. et al., 2004, Phys. Rev. Lett., 92, 230802
Hallstadius L., 1979, Z. Phys. A, 291, 203
Hannemann S., Salumbides E. J., Witte S., Zinkstok R. Th., van Duijn E.-J., Eikema K. S. E., Ubachs W., 2006, Phys. Rev. A, 74, 012505
Holzwarth R., Udem Th., Hänsch T. W., Knight J. C., Wadswoth W. J., Russel P. St. J., 2000, Phys. Rev. Lett., 85, 2264
Jones D. J., Diddams S. A., Ranka J. K., Stentz A., Windeler R. S., Hall J. L., Cundiff S. T., 2000, Sci, 288, 635
Kluge H.-J., Sauter H., 1974, Z. Phys., 270, 295
Kozlov M. G., Korol V. A., Berengut J. C., Dzuba V. A., Flambaum V. V., 2004, Phys. Rev. Lett., 70, 062108
Le Boiteux S., Klein A., Rios Leite J. R., Ducloy M., 1988, J. Phys. France, 49, 885
Lurio A., 1964, Phys. Rev., 136, A376
Murphy M. T., Webb J. K., Flambaum V. V., Churchill C. W., Prochaska J. X., 2001, MNRAS, 327, 1223
Murphy M. T., Webb J. K., Flambaum V. V., Curran S. J., 2003, Ap&SS, 283, 577
Pickering J., Thorne A. P., Webb J. K., 1998, MNRAS, 300, 131
Quast R., Reimers D., Levshakov S. A., 2004, A&A, 415, L7
Reinhold E., Buning R., Hollenstein U., Ivanchik A., Petitjean P., Ubachs W., 2006, Phys. Rev. Lett., 96, 151101
Risberg G., 1965, Ark. Fys., 28, 381
Rosman K. J. R., Taylor P. D., 1998, Pure Appl. Chem., 70, 217
Srianand R., Chand H., Petitjean P., Aracil B., 2004, Phys. Rev. Lett., 92, 121302
Uzan J.-P., 2003, Rev. Mod. Phys., 75, 403
Webb J. K., Flambaum V. V., Churchill C. W., Barrow J. D., 1999, Phys. Rev. Lett., 82, 884
Webb J. K., Murphy M. T., Flambaum V. V., Dzuba V. A., Barrow J. D., Churchill C. W., Prochaska J. X., Wolfe A. M., 2001, Phys. Rev. Lett., 87, 091301
Webb J. K., Murphy M. T., Flambaum V. V., Curran S. J., 2003, Ap&SS, 283, 565
White R. T., He Y., Orr B. J., Kono M., Baldwin K. G. H., 2003, Opt. Lett., 28, 1248
White R. T., He Y., Orr B. J., Kono M., Baldwin K. G. H., 2004, J. Opt. Soc. Am. B, 21, 1577
Witte S., Zinkstok R. T., Hogervorst W., Eikema K. S. E., 2004, Appl. Phys. B, 78, 5

This paper has been typeset from a TEX/LATEX file prepared by the author.