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Improved frequency measurement and isotope shift of the $4d^95s^2\ ^2D_{5/2} \rightarrow 4d^{10}6p\ ^2P_{3/2}$ transition in silver by laser heterodyne spectroscopy

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Abstract. The frequency of the center of gravity of the transition $4d^95s^2\ ^2\mathrm{D}_{5/2} \rightarrow 4d^{10}6p\ ^2\mathrm{P}_{3/2}$ in Ag I has been determined using laser heterodyne spectroscopy of a collimated thermal metastable atomic beam. We find the value $\nu = 547\,376\,425(12)$ MHz, five times more accurate than our earlier published result. For the isotope shift, we obtain $\nu(^{109}\mathrm{Ag})-\nu(^{107}\mathrm{Ag})=-978.1(0.5)$ MHz, a ten-fold improvement over our previous measurement.

PACS. 32.30.Jc Visible and ultraviolet spectra

1 Introduction

1.1 Motivation

The silver atom is an interesting candidate for an optical frequency standard based on narrow two-photon transitions from the $4d^{10}5s\ ^2\mathrm{S}_{1/2}$ ground state (Fig. 1). Excitation of the $4d^95s^2\ ^2\mathrm{D}_{3/2}$ state, whose natural width is about 4 kHz, requires two photons near 576 nm. Detection of this transition *via* cascade fluorescence should be straightforward. The narrower $4d^95s^2\ ^2\mathrm{D}_{5/2}$ level (natural width less than 1 Hz) can be excited using two photons near 661 nm and was proposed as a frequency standard many years ago [1]. This transition is harder to detect because the upper level is metastable. To the best of our knowledge, two groups are currently working in this field. The group of Walther at the Max Planck Institut für Quantenoptik, Garching, has cooled and trapped a sample of silver atoms using the $5s$–$5p$ transition at 328 nm [2–4]. In our own laboratory, we employ a thermal atomic beam with a first aim of obtaining an experimental linewidth of a few kilohertz for the $4d^{10}5s\ ^2\mathrm{S}_{1/2} \rightarrow 4d^95s^2\ ^2\mathrm{D}_{5/2}$ resonance using near collinear excitation.

1.2 Previous measurement at 547.7 nm

We have achieved detection of metastable silver atoms present in an atomic beam using laser induced fluorescence. In these experiments, a small fraction of atoms effusing from a tantalum oven was excited to the metastable $^2\mathrm{D}_{5/2}$ level by electron bombardment. A dye laser was tuned to a hyperfine component of the $4d^95s^2\ ^2\mathrm{D}_{5/2} \rightarrow 4d^{10}6p\ ^2\mathrm{P}_{3/2}$ transition near 547.7 nm to transfer atoms from the metastable level to the short-lived $6p\ ^2\mathrm{P}_{3/2}$ level. Atoms in this state were detected *via* the observation of the $4d^{10}6p\ ^2\mathrm{P}_{3/2} \rightarrow 4d^{10}5s\ ^2\mathrm{S}_{1/2}$ transition at 206 nm. In a recent publication, we have described a measurement of the frequency, isotope shift and hyperfine structure of the 547.7 nm line which arose from this work [5]. Our aim was to check the results of measurements by other groups [6,7] since, from a knowledge of the frequencies of the 547.7 nm and 206 nm transitions, one can deduce the energy of $4d^95s^2\ ^2\mathrm{D}_{5/2}$ level [7,8]. First-order Doppler broadening

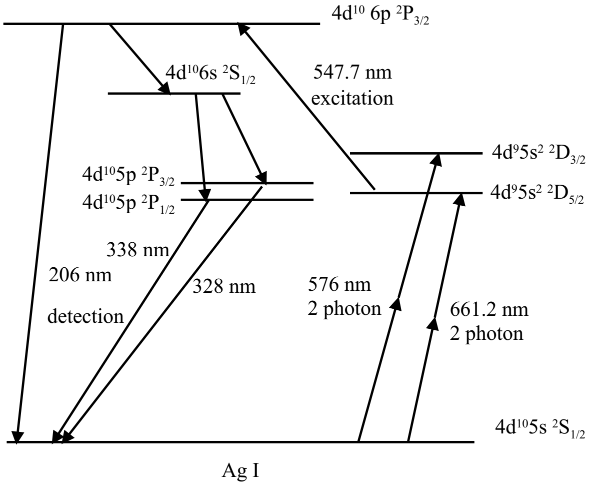

Fig. 1. Partial energy level diagram for Ag I showing the transitions of interest.

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was reduced to about 10 MHz using laser excitation perpendicular to a collimated atomic beam. Local calibration of the frequency axis involved the use of a Fabry Perot cavity (free spectral range ≃150 MHz) and the accurately known hyperfine splittings of the $4d^95s^2\ ^2\mathrm{D}_{5/2}$ levels in the two stable isotopes of this element ^{107}Ag (51.84%) and ^{109}Ag (48.16%) [9]. The frequency of each hyperfine component of the silver transition was linked to that of the hyperfine component a_1 of the resonance $R(22)27-1$ of the I_2 molecule.

2 Current experiment

In the remainder of this article, we describe a measurement of the frequency and isotope shift of the $4d^95s^2\ ^2\mathrm{D}_{5/2} \rightarrow 4d^{10}6p\ ^2\mathrm{P}_{3/2}$ transition using laser heterodyne spectroscopy. Our aim was to verify the result of our previous experiment while at the same time improving the accuracy. It appeared reasonable to reduce the uncertainty in the isotope shift by the use of the heterodyne calibration technique instead of marker fringes. A more accurate frequency measurement was possible thanks to the availability of improved values for iodine calibration lines [10]. The motivation for such an improvement is to know more accurately the frequency necessary to excite the two-photon transition described above. Although the current limiting factors are the frequency and isotope shift of the $5s$–$6p$ line at 206 nm, these quantities could be determined to within a few megahertz by high-resolution laser spectroscopy of an atomic beam. Indeed, in collaboration with the group of Biraben at Laboratoire Kastler Brossel, Paris, we have already observed the Doppler-broadened absorption resonance in a hollow cathode discharge.

2.1 Apparatus

The apparatus is shown in Figure 2. Here, as in our previous work [5], one laser excites the silver transition in the atomic beam but, in addition, the frequency of a second dye laser is servo-locked to the above-mentioned iodine resonance using saturated absorption spectroscopy. The beat signal between the two lasers provides direct calibration of the frequency axis without the need to know the hyperfine splitting of the metastable levels of either isotope. Furthermore, for a given scan time, more data points can be acquired for each atomic resonance since there is no need to generate marker fringes from an interferometer.

2.2 Detection scheme

As far as detection of the silver transition is concerned, collection of the fluorescence at 206 nm was employed to begin with, since this gave a low background signal (< 10 s^{-1}). Figure 3 shows a scan over the transition between the hyperfine levels $4d^95s^2\ ^2\mathrm{D}_{5/2}$ ($F = 3$) and $4d^{10}6p\ ^2\mathrm{P}_{3/2}$ ($F' = 3$) in ^{107}Ag. Subsequently, however, and following Dinger *et al.* [6], we detected the fluorescence at 328 and 338 nm resulting from the cascade decay of the $6p\ ^2\mathrm{P}_{3/2}$ level. Using an appropriate photomultiplier tube (Hamamatsu R6095) and interference filter (Oriel 340FS25), we obtained an eight times greater signal as a result of higher quantum efficiency (0.25), more favourable branching ratio (0.6) and better filter transmission. In both cases, a spherical mirror below the interaction region was used to increase the light collection efficiency. On the other hand, a much larger background arose (2000 s^{-1}) from light emitted by the filament of the electron gun. Nevertheless, the overall signal-to-noise ratio (≃60 for the largest peak) was superior to that for 206 nm detection (≃15).

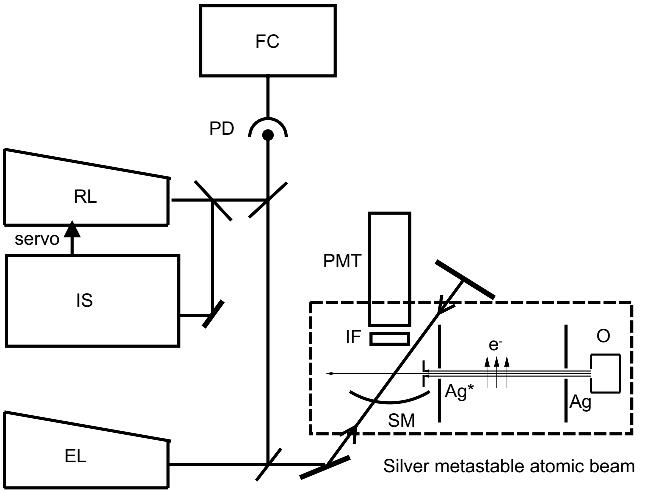

Fig. 2. Experimental set-up. EL: excitation laser, RL: reference laser, IS: iodine spectrometer, PD: photodiode, FC: frequency counter, O: oven, IF: interference filter, PMT: photomultiplier tube, SM: spherical mirror.

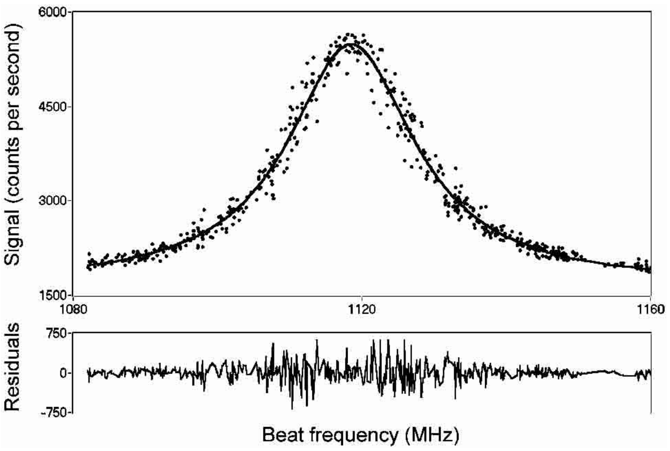

Fig. 3. Scan over the component $F = 3 \rightarrow F' = 2$ for the isotope ^{107}Ag. The vertical axis indicates the number of counts per second and the horizontal axis the beat frequency between the two lasers. The dots represent the data points and the solid line is a fitted Voigt profile.

Table 1. Values for the isotope shift $\delta\nu^{107,109}$ of the 547.7 nm transition in Ag I obtained by various authors.

Author	Isotope shift $\delta\nu^{107,109}$ (MHz)
This work	−978.1(0.5)
Guérandel *et al.* [5]	−982.6(5.4)
Dinger *et al.* [6]	−976(8)
Rasmussen [13]	−960(15)

2.3 Isotope shift

The laser frequency was scanned over a given hyperfine component of the 547.7 nm transition in both isotopes ^{107}Ag and ^{109}Ag, without the trajectory of the laser beam being modified. The centers of the resonances were found by least-squares fitting of a Voigt profile to the data. To calculate the isotope shift, we fixed the values of the hyperfine splittings in both the $4d^95s^2\ ^2\mathrm{D}_{5/2}$ [9] and upper $4d^{10}6p\ ^2\mathrm{P}_{3/2}$ [11] levels, since these have been determined accurately by other groups. For the hyperfine splitting of the upper level in ^{109}Ag, we used the value of Bengtsson *et al.* [11] together with the ratio of the ground state hyperfine splittings measured by Dahmen and Penselin [12] and neglected the hyperfine anomaly. The value $(A(6p\ ^2\mathrm{P}_{3/2}) = -10.4(3)$ MHz) is very close to that measured recently by our own group (−10.2(6) MHz) [5]. Although the choice of value makes a negligible difference to the calculated isotope shift, we prefer to use data independent of our own. The difference between corresponding hyperfine components of the 547.7 nm transition in each isotope then yields an updated value for the isotope shift. The sample standard deviation σ for $\Delta\nu$ is 150 kHz. As discussed in our previous article [5], the isotope shift measurement is insensitive to many systematic effects. The use of heterodyne calibration eliminates uncertainties associated with non-uniformities of the laser frequency scan. However, given the relatively small size of the data set (7 values), we prefer to ascribe a conservative uncertainty of 500 kHz ($\simeq 3\sigma$) to these measurements. This still represents an order of magnitude improvement over our previous work. A comparison with other experiments is given in Table 1. The value $\Delta\nu = -978.1(5)$ MHz lies within one standard deviation of the number we obtained using marker fringe calibration (−982.6(5.4) MHz). It is close to the value −976(8) MHz due to Dinger *et al.* [6] obtained by collinear laser spectroscopy of a fast beam and compares with the much earlier value of Rasmussen −960(15) MHz who observed the fluorescence from a cooled discharge lamp [13].

2.4 Frequency measurement

To measure the frequency of the hyperfine components of the silver transition we proceeded as described above. In our previous article, we used the frequency of the fine structure component of the molecular iodine line given by the Aimé Cotton atlas [14]. From the known hyperfine constants of this molecule one can deduce the frequency of the hyperfine component a_1 [15]. For the measurements presented here, however, we have taken advantage of recent more accurate data for the iodine molecule, namely a computer program (named IodineSpec) available from TuiOptics which provides the frequencies of the hyperfine components of interest directly [10]. In our case, the frequency of the component a_1 lies 31 MHz above the value given by the Aimé Cotton atlas. To minimise the first order Doppler shift which arises when the angle between the laser and atomic beams is different from 90°, the laser beam was retro-reflected so as to generate two resonances with equal but opposite frequency displacements. Again, the use of the heterodyne technique enabled more data to be acquired rapidly than with marker fringe calibration. A spectrum is shown in Figure 4 for the component $4d^95s^2\ ^2\mathrm{D}_{5/2}\ (F = 3) \rightarrow 4d^{10}6p\ ^2\mathrm{P}_{3/2}(F' = 2)$. For this measurement we detected fluorescence at 206 nm. The beat frequency 1065(1) MHz, obtained from an average of 3 measurements lies 4(1) MHz below our previous value obtained using marker fringe calibration [5]. Using our values for the isotope shift and those of hyperfine splittings measured by other authors [9,11,12], we calculated the frequencies of the centroids of both isotopes as well as that of a naturally abundant sample. Systematic effects, discussed in detail in [5], are negligible compared with the 12 MHz uncertainty in the frequency calibration of the iodine reference line. The results, presented in Table 2, are in agreement with the work of other authors [6–8]. Compared with our previous measurement, we find better accordance with the values of Larkins and Hannaford [7] and Pickering [8], both of whom used an independent calibration method. If one compares our values with those of Dinger *et al.* [6], one should increase the latter's frequencies by 31 MHz to account for the modification of the frequency of the molecular iodine calibration lines.

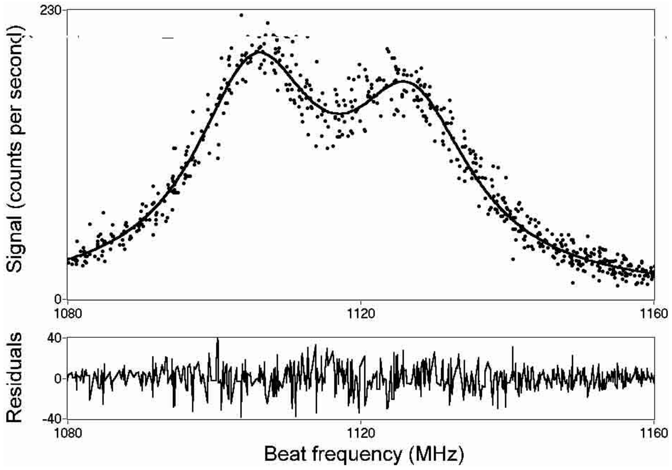

Fig. 4. Scan over the component $F = 3 \rightarrow F' = 2$ for the isotope ^{107}Ag with the laser beam close to normal incidence with the atomic beam and retro-reflected. The vertical axis indicates the number of counts per second and the horizontal axis the beat frequency between the two lasers. The dots represent the data points and the solid line is a fitted Voigt profile.

Table 2. Frequencies (in megahertz) of the hyperfine components of the transition $4d^9 5s^2\ ^2\mathrm{D}_{5/2}(F) \rightarrow 4d^{10} 6p\ ^2\mathrm{P}_{3/2}(F')$ in Ag I.

Isotope	F	F'	Frequency $-547\,370\,000$ MHz		
			this work	other authors	reference
^{109}Ag	2	2	5 656(12)		
^{109}Ag	2	1	5 676(12)		
^{109}Ag	3	2	6 091(12)		
^{109}Ag	centroid		5 918(12)	5 830(60)	[6]
^{107}Ag	2	2	6 668(12)		
^{107}Ag	2	1	6 686(12)		
^{107}Ag	3	2	7 047(12)		
^{107}Ag	centroid		6 896(12)	6 880(50)	[6]
^{109}Ag, ^{107}Ag	centroid of both		6 425(12)	6 449(60)	[7]
				6 449(45)	[8]

3 Conclusion

We have performed new measurements of the isotope shift and frequency of the $4d^9 5s^2\ ^2\mathrm{D}_{5/2} \rightarrow 4d^{10} 6p\ ^2\mathrm{P}_{3/2}$ transition in atomic silver using an improved frequency calibration procedure. Our results are in good agreement with those of our earlier experiment. The isotope shift is ten times more accurate than our previous one and represents, as far as we are aware, the most precise value currently available. Our measurement of the transition frequency provides a value five times more accurate than the result of our previous work. This improvement is due mainly to the use of an updated value for the frequency of the molecular iodine reference line.

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