

Observation by two-photon laser spectroscopy of the $4d^{10}5s^2S_{1/2} \rightarrow 4d^95s^2D_{5/2}$ clock transition in atomic silver

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We report the observation of the very narrow $4d^{10}5s^2S_{1/2} \rightarrow 4d^95s^2D_{5/2}$ transition in atomic silver. The frequencies of the hyperfine components in ^{107}Ag and ^{109}Ag have been measured using Doppler-free two-photon laser spectroscopy of a thermal beam and heterodyne calibration with respect to the a_1 component of the $62P(4-5)$ line in molecular iodine near 661 nm. For the center of gravity of a mixture of natural abundance, we deduce the value 906 641 295.77(19) MHz. For the isotope shift, we obtain $\nu(^{109}\text{Ag}) - \nu(^{107}\text{Ag}) = 564.15(37)$ MHz, from which we deduce the frequency and isotope shift of the $4d^{10}5s^2S_{1/2} \rightarrow 4d^{10}6p^2P_{3/2}$ transition at 206 nm.

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I. INTRODUCTION

The silver atom is an interesting candidate for an optical frequency standard based on a narrow two-photon transition from the $4d^{10}5s^2S_{1/2}$ ground state (see Fig. 1). The metastable $4d^95s^2D_{5/2}$ 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 [1,2]. More recently, the group of Walther at the Max Planck Institut für Quantenoptik, Garching, has cooled and trapped a sample of silver atoms using the $5s^2S_{1/2} \rightarrow 5p^2P_{3/2}$ transition at 328 nm [3,4]. In our own laboratory, we have employed a thermal atomic beam where we excited the two-photon transition, never observed before by laser spectroscopy, as a prelude to laser cooling work.

II. CURRENT EXPERIMENT

In previous papers, we presented an optical technique to detect silver atoms in the metastable level [5,6]. We later reported two-photon excitation of the $4d^95s^2D_{3/2}$ state, whose natural width is about 4 kHz, using two photons near 576 nm [7]. The detection of this transition via cascade fluorescence was straightforward. In the present paper, we describe the optical two-photon excitation of the very narrow $4d^{10}5s^2S_{1/2} \rightarrow 4d^95s^2D_{5/2}$ transition. We present measurements of the frequency, isotope shift, and hyperfine coupling constants of this transition.

III. EXPERIMENTAL SETUP

The apparatus is shown in Fig. 2. Silver atoms in a thermal beam undergo two laser excitations. The first excitation

corresponds to the two-photon transition at 661 nm that brings atoms into the $^2D_{5/2}$ state. Detection of silver atoms in this level requires a second laser excitation that takes atoms from the metastable state to the short-lived ($\tau=80$ ns) $6p^2P_{3/2}$ level. We detect cascade fluorescence at 328 and 338 nm as a signature of the population of the metastable state.

A. Atomic beam

The beam of silver atoms is produced by heating up to 1200 °C a tubular tantalum oven filled with natural silver powder. It is very similar to the one described in Ref. [8]. Atoms effusing from the oven are collimated by two 10 mm holes. Note that natural silver is a mixture of two stable isotopes of nearly equal abundances: ^{107}Ag :51.84% and ^{109}Ag :48.16%. Both isotopes have the same nuclear spin 1/2.

B. Excitation laser

The laser used to excite the silver two photon transition is a coherent CR 699-21 ring dye laser (DCM dye) with a Radiant Dye Laser Accessories high pressure dye circulator system, pumped by a 6 W Ar^+ laser at 514.5 nm. A cw single-mode output power of 500 mW is typically obtained. The dye laser frequency jitter is reduced by the use of an intracavity electro-optic actuator and a fast electronic servo. A microwave sideband method [9] is used to lock the output frequency of the laser to the center of an isolated Fabry-Perot cavity (finesse 600 at 661 nm). We estimated the residual frequency jitter to be less than 10 kHz over a 1 s integration time [7].

C. Reference laser

The frequency of the excitation laser is obtained by analyzing the beat note between the excitation and the reference

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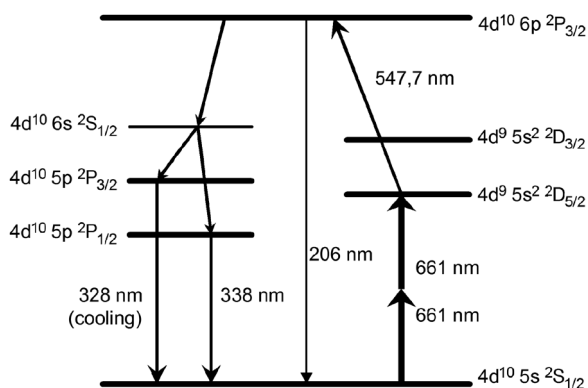


FIG. 1. Energy levels of Ag I.

lasers. One milliwatt of each laser beam is overlapped on a fast photodetector (Hamamatsu G4176) injecting an rf amplifier (Miteq, gain 40 dB). The frequency of the beat signal is measured by a counter (EIP 625A). The reference laser is a diode laser in a standard ECL (Littrow) configuration, sent to a molecular iodine spectrometer. We lock the frequency of the ECL to that of the a_1 component of the $62P(4-5)$ line in $^{127}\text{I}_2$. A typical spectrum of the component, obtained by saturated absorption technique, is shown in Fig. 3.

D. Power build-up cavity and magnetic shielding

To increase the two-photon transition probability as well as to ensure correct overlap of counterpropagating laser beams, a Fabry-Perot cavity (finesse 300 at 661 nm) is placed in the vacuum chamber around the two-photon interaction region. This cavity is formed by two mirrors of radii of curvature 100 mm, separated by 145 mm. The length of the cavity is servo-locked to maintain resonance with the frequency of the excitation laser using a 17 kHz dither and phase sensitive detection. With an injected power of 200 mW, we could obtain intracavity powers of over 12 W,

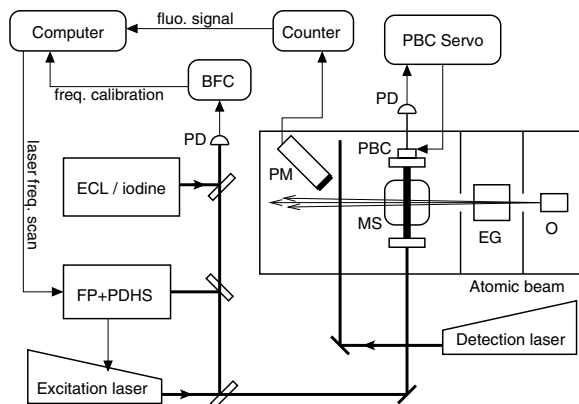


FIG. 2. Experimental setup. EL, excitation laser; FP+PDHS, Fabry-Perot cavity and Pound-Drever-Hall stabilization; ECL/iodine, extended cavity laser diode stabilized on iodine line; PD, photodiode; BFC, beat frequency counter; DL, detection laser; O, oven; EG, electron gun; MS, magnetic shielding; PBC, power build-up cavity; PM, photomultiplier; A+C, amplifier and counter.

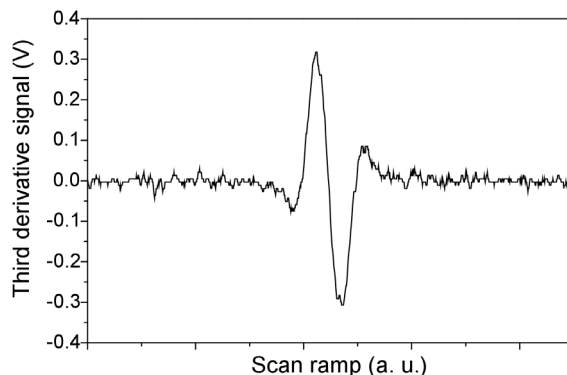


FIG. 3. Third-derivative spectrum of the a_1 component of the $62P(4-5)$ line in molecular iodine, obtained by saturation spectroscopy. The reference laser was locked to this component.

with a minimum beam radius of $97 \mu\text{m}$ located in the middle of the cavity.

In the study of another two-photon transition in silver [7], the first-order Zeeman shift had caused a visible splitting of hyperfine components into subcomponents, and even overlap of two different hyperfine components. This overlapping contributed significantly to the uncertainty in the determination of center frequencies for each component. To avoid this, we have now shielded the two-photon interaction region by a double-wall μ -metal enclosure, that reduces the magnetic field in the two-photon excitation region from $\|\mathbf{B}\| = 360$ milligauss (without shielding) to less than 5 milligauss.

E. Detection laser and electron gun

The detection laser is a commercial coherent CR 699-21 dye laser emitting at 547.7 nm, using rhodamine 560 dye, pumped by a 5 W Ar^+ laser at 514.5 nm. The purpose of this laser is to excite the atoms from the metastable level to the short-lived $6p \ ^2P_{3/2}$ level. The interaction region between atoms and the detection laser is located 30 cm downstream from the two-photon excitation region. Nevertheless, considering the long lifetime of atoms in the metastable state and the high speed of atoms (over 600 m s^{-1}), a negligible fraction of metastable atoms decays between the two laser excitations. The strong single photon $4d^9 5s^2 \ ^2D_{5/2} \rightarrow 4d^{10} 6p \ ^2P_{3/2}$ transition is easily saturated ($I_{\text{sat}} \approx 2.4 \text{ mW/cm}^2$) by the 100 mW beam (diameter 20 mm) incident on the silver atoms. Combined with a large Doppler broadening due to high divergence of the detection beam, the experimental linewidth of each hyperfine component of the detection transition was about 30 MHz. As the detection dye laser stabilized with its commercial system exhibits a 2 MHz linewidth and small frequency drift during the acquisition time, no further frequency stabilization was required for this laser.

The electron gun is fully described in Ref. [8]. It is used to populate the metastable level by electron bombardment, allowing a correct adjustment of the frequency of the detection laser (cf. Sec. IV B).

TABLE I. Hyperfine coupling constants of the $4d^{10}5s^2S_{1/2}$ and $4d^95s^2D_{5/2}$ levels in Ag.

Level	Isotope	This work (MHz)	Others (MHz)
$^2S_{1/2}$	107	-1 712.53(60)	-1 712.512 111 (18) [13]
	109	-1 976.99(42)	-1 976.932 075 (17) [13]
$^2D_{5/2}$	107	-126.28(20)	-126.281 8 (1) [14]
	109	-145.11(16)	-145.158 4 (5) [14]

F. Detection scheme

The detection setup is similar to that described in Ref. [7] and is located above the interaction region of the atoms and the detection laser. We detect fluorescence at 328 and 338 nm resulting from the cascade decay from the $6p^2P_{3/2}$ level via the $6s^2S_{1/2}$, $5p^2P_{3/2}$, and $5p^2P_{1/2}$ levels, using a photomultiplier tube in pulse counting mode. The active area of this tube is protected by two color glass filters (UG11, thickness 3 mm) centered on 330 nm. A spherical mirror ($R=-100$ mm, diameter 90 mm) was placed 3 cm below the interaction region to increase the light collection efficiency. We estimate the total detection efficiency of this setup to be about 0.0015. The pulses from the photomultiplier tube are sent via a high-speed amplifier (Stanford Research Systems SR445, gain 25) to a counter (Stanford Research Systems SR400). Unfiltered light from the oven is the main cause of the observed $20\,000\text{ s}^{-1}$ background signal.

IV. EXPERIMENTAL PROCEDURE

A. Previous knowledge of frequencies

The very narrow level in silver has been studied since the early 1940s. Measured values for the energy of the $5s^2^2D_{5/2}$ level measured can be found in Refs. [10,11] with an uncertainty of several gigahertz. In 1994, Larkins and Hannaford [12] measured the frequency of both 547.7 nm and 206.1 nm lines to deduce that of the 661.3 nm transition, using a hollow cathode lamp and a monochromator. Using known val-

ues for the magnetic hyperfine interaction constants (see Table I) of the fundamental [13] and the metastable [14] states, and estimating the isotope shift of the transition, they calculated the expected frequencies of the hyperfine components with a 240 MHz uncertainty (see Table II). In our group, we have followed the same approach. In previous experiments [5,6], we had studied the 547.7 nm line by laser spectroscopy of our atomic beam. The frequency of the center of gravity and the isotope shift of this line were measured and found to be $\nu_{547.7}=547\,376\,425(12)$ MHz and $\delta\nu_{547.7}^{\text{iso}}=-978.1(0.5)$ MHz. Later, we collaborated with the group of Biraben in Laboratoire Kastler-Brossel to measure the frequency of the 206.1 nm line in a silver hollow cathode lamp, using their twice frequency-doubled Ti:sapphire laser and an optogalvanic detection technique. A profile of the line was observed with a linewidth of 1 GHz corresponding to hyperfine components broadened and merged by Doppler and collision effects. By estimating the isotope shift, we could determine the frequency of the center of gravity of this profile, $\nu_{206.1}=1\,454\,017\,694(45)$ MHz with uncertainty dominated by that of the isotope shift. We could then derive an expected hyperfine spectrum with an uncertainty of 45 MHz for the frequency of the components (see Table II). Moreover, frequencies of the 547.7 nm and 206.1 nm lines were measured [15] for us by Pickering and Zilio by Fourier transform spectroscopy in a hollow cathode lamp (see Appendix B).

The expected spectrum of the 661.3 nm line included three components for each isotope of natural silver. The relative intensities of the components can be calculated simply from the expression of electric quadrupole transitions in I - J coupling [12,16], which yields intensities ratios of 2, 3, and 7 for the $F=1 \rightarrow F'=2$, $F=0 \rightarrow F'=2$ and $F=1 \rightarrow F'=3$ components, respectively.

B. Data acquisition and analysis

Prior to any two-photon excitation, it was necessary to adjust the frequency of the detection laser to match that of a suitable detection hyperfine component. We turned on the electron gun to populate the metastable state. By manually

TABLE II. Isotope shift and frequencies of the $4d^{10}5s^2S_{1/2} \rightarrow 4d^95s^2D_{5/2}$.

Isotope	Component	Rel. int.	This work (MHz)	Expected values (MHz) [17]
107	1 \rightarrow 3	7	906 641 294.36 (43)	906 641 265 (24)
	1 \rightarrow 2	2	906 641 673.19 (44)	906 641 644 (24)
	0 \rightarrow 2	3	906 639 960.66 (42)	906 639 931 (24)
	Centroid ν_{107}		906 641 024.07 (28)	
109	1 \rightarrow 3	7	906 641 901.08 (38)	906 641 877 (24)
	1 \rightarrow 2	2	906 642 336.42 (30)	906 642 312 (24)
	0 \rightarrow 2	3	906 640 359.42 (30)	906 640 335 (24)
	Centroid ν_{109}		906 641 588.22 (24)	
Centroid for natural silver			906 641 295.77 (19)	906 640 925 (210) [12] 906 641 075 (120) [15]
Isotope shift $\nu_{109}-\nu_{107}$			564.15 (37)	550 (60) [12]

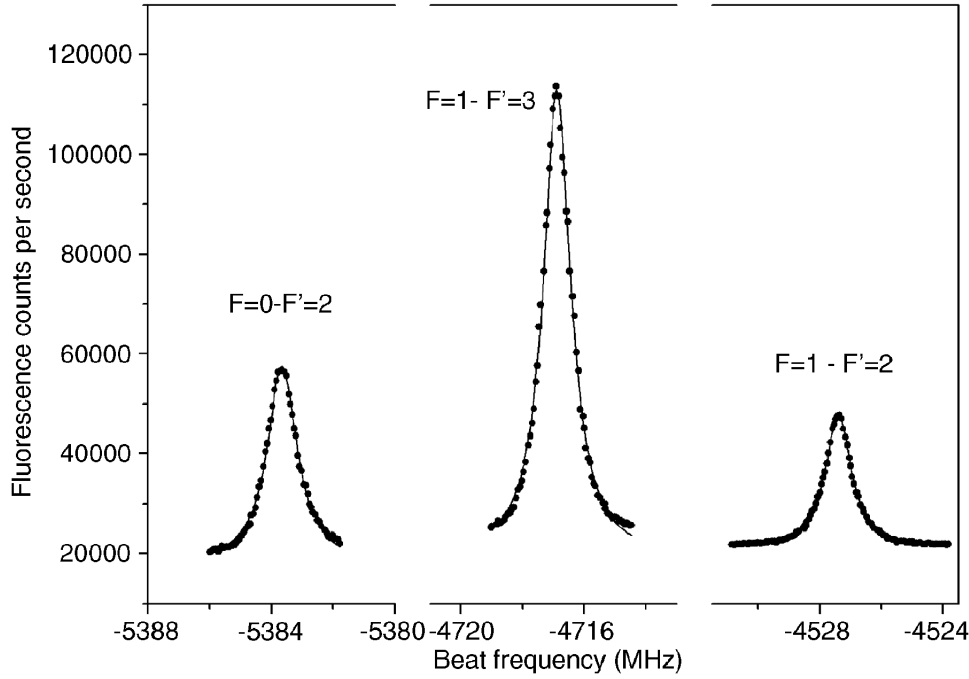


FIG. 4. The three hyperfine components at 661.3 nm in ^{107}Ag . Relative intensities roughly match their expected values of 3, 7, and 2, respectively. The linewidth of the components is about 1 MHz. Dots, experimental data. Line, fit by a Lorentzian curve.

scanning the detection laser and monitoring the fluorescence signal, we could observe the components of the 547.7 nm line and position the frequency of the laser at the top of one of them. Then the electron gun was switched off. The detection line has three allowed hyperfine components for each isotope, of relative intensities ratios of 1, 9, and 14 for the $F=2 \rightarrow F'=2$, $F=2 \rightarrow F'=1$, and $F=3 \rightarrow F'=2$ components, respectively. Naturally, for a given component of the two-photon transition, we used the strongest component of the detection line, namely $(F=2 \rightarrow F'=1)_{547.7}$ to detect $(F=0 \rightarrow F'=2$ and $F=1 \rightarrow F'=2)_{661.3}$ and $(F=3 \rightarrow F'=2)_{547.7}$ to detect $(F=1 \rightarrow F'=3)_{661.3}$.

To study a given component, we scan the excitation laser by steps of about 50 kHz over a few megahertz. We record the fluorescence count, the beat frequency between excitation and reference lasers and the power transmitted by the build-up cavity. The typical integration time for the fluorescence and frequency counters is 1 second.

Hyperfine components of the 661 nm line were found close to their expected frequencies [17] (about 15 MHz above), with amplitudes of several 10 thousands counts per second and an excellent SNR. The observed relative amplitude of components is in good agreement with I - J coupling theory. Figure 4 shows the three hyperfine components at 661.3 nm of ^{107}Ag . We had estimated [8] the excitation probability to be

$$\Gamma_{F=1 \rightarrow F'=3} = 6.3 \times 10^{-8} \frac{P^2}{S^2} \frac{1}{\Gamma_e} s^{-1}, \quad (1)$$

where P and S are, respectively, the power and the section of the incident beams. Based on this estimation and taking into account the detection efficiency of our experimental setup, we expected about 50 000 counts per second for the strongest component, with a large uncertainty due to the flux of

silver atoms in the two-photon interaction region. We obtained up to 100 000 counts per second for this component. The experimental linewidth of components is about 1 MHz which corresponds to the expected transit-time broadening for an optical beam of waist of 90 μm and a most probable atomic speed of $V_{\text{RMS}}=660 \text{ m s}^{-1}$. We have calculated [18] that the theoretical profile for components excited by a Gaussian laser beam in an atomic beam (ignoring the weak Gaussian pedestal due to absorption of copropagating photons) is the convolution of two Lorentzian curves deformed by light shift, and is indeed very close to a single Lorentzian profile. Thus, to determine the center frequency of each component, we fitted experimental data by a Lorentzian curve. Other attempts to fit by more complex functions gave no significant difference (less than 5 kHz) for the center frequency value. For each hyperfine component, five spectra were acquired and analyzed.

V. SYSTEMATIC EFFECTS

In the following section, we discuss the various perturbations affecting our measurements. Most systematic effects for the $4d^{10}5s^2S_{1/2} \rightarrow 4d^95s^2D_{5/2}$ transition have been studied in [8]. An error budget is given in Table III.

A. Reference laser frequency and stability

The reference laser has been calibrated at the International Bureau of Weights and Measures (BIPM) with a frequency comb based on a femtosecond laser whose repetition rate is stabilized to a hydrogen maser [19]. As the modulation depth of diode current and iodine temperature showed critical influence on the ECL frequency, the reference laser was run using the same settings for silver measurements and for calibration at BIPM: diode current modulation at 15.7 kHz, frequency modulation depth of 6 MHz, diode temperature of

TABLE III. Sources of uncertainty in the determination of frequencies of hyperfine components of the $4d^{10}5s^2S_{1/2} \rightarrow 4d^95s^2D_{5/2}$ transition in atomic silver (uncertainties given at twice the laser frequency).

Source	Contribution to absolute frequency (kHz)
Frequency of I_2 reference line $62P(4-5)$	<1
Reference laser lock point	30
Determination of centroid	<5
First-order Zeeman shift (residual circular polarization)	<1
Second-order Zeeman shift	1
d.c. Stark shift	<1
Second-order Doppler shift	<1
Light shift correction at 661.3 nm (intensity 4.5 kW cm^{-2})	<440

25 °C, cold point of the 20 cm long glass iodine cell maintained at 13 °C. The frequency of the ECL locked to a_1 has been measured to be $\nu_{a_1} = 453\,325\,363\,771(1)$ kHz. However, the transportation between the two laboratories may have perturbed slightly the reference laser, we enlarged the uncertainty of the frequency of the reference laser to 15 kHz.

B. Zeeman effects

In the weak field limit, the Zeeman shift of a hyperfine level $|F, m_F\rangle$ can be calculated by the usual expression:

$$\Delta E = g_F \mu_B B m_F. \quad (2)$$

Using the Landé factors of the $^2S_{1/2}$ and $^2D_{5/2}$ levels, we calculate that the maximum shift occurs for the $F=1 \rightarrow F=2$ component and is equal to 560 kHz G^{-1} , i.e., a 280 kHz G^{-1} shift of the frequency of a photon at 661 nm. Since we use linearly polarized light, the splitting should be symmetric and contribute only to the broadening of the line. We cannot exclude a degree of residual circular polarization in the build-up cavity that could induce a shift of components, but the μ -metal shielding of the two-photon interaction region decreases the magnetic field down to less than 5 mG making this shift thus negligible.

The second-order Zeeman effect for this transition has been fully calculated and is also negligible (see Table IV).

TABLE IV. Second-order Zeeman shift of the 661 nm two-photon components with a linearly polarized light.

$F_g \rightarrow F_e$	$\Delta \nu_B^{107}$ (kHz/G ²)	$\Delta \nu_B^{109}$ (kHz/G ²)
0 → 2	0.7	0.6
1 → 2	0.7	0.6
1 → 3	-3	-2.6

C. Light shift

The high intensity of the excitation beam allowed spectra with excellent signal-to-noise ratio, but also induced a significant light shift of the hyperfine components. Because of technical issues concerning the excitation dye laser, we could not achieve a complete experimental study of the light shift. So we choose to calculate the effect and correct the measured frequencies with the calculated value. For each acquired spectrum, the center frequency of the line was corrected with respect to the measured power in the build-up cavity, also taking into account the Gaussian distribution of energy in the beam (see Appendix A for details). We associated a 100% uncertainty with this correction, mainly due to assumptions in the calculation of the light shift coefficient c_{ls} .

D. Other negligible effects

The first-order Doppler shift is absent because the silver atoms are excited by two counterpropagating beams at the beams waist. The second-order Doppler shift in our thermal beam (most probable velocity 660 m s^{-1}) is less than 1 kHz and can be neglected. The dc Stark shift is also negligible.

VI. RESULTS AND DISCUSSION

Each hyperfine component in each isotope at 661.3 nm has been measured and its center frequency has been corrected for light shift. From the separation between components, we calculated the hyperfine splittings of the ground and excited states and hence the magnetic hyperfine constants A . The difference of the centers of gravity for each isotope gave the isotope shift. Finally, to compare our results with previously presented values, we calculated the center of gravity of the spectrum of a naturally abundant sample containing 51.84% ^{107}Ag and 48.16% ^{109}Ag . The results are shown in Tables II and I. Our values of the hyperfine frequencies lie about 15 MHz (at the laser frequency) above our expected values, probably due to the uncertainty on the value of the isotope shift of the $4d^{10}5s^2S_{1/2} \rightarrow 4d^{10}6p^2P_{3/2}$ transition at 206.1 nm. A critical test is provided by comparison with the hyperfine constants of the ground and excited states. Our values are fully compatible with those of the ground state [13] and metastable level [14], both obtained by radio-frequency measurements. Our value of isotope shift of the two-photon transition is the first direct determination of this quantity, and is in good agreement with previously estimated values. The uncertainty of our frequency measurements is dominated by the light shift. By contrast, as regards the optical frequency of the transition, there is no close agreement with other measured values deduced from Fourier transform spectroscopy of hollow cathode discharge emission lines. The value of the center frequency for natural silver measured by Pickering and Zilio lies 220 MHz away from ours. The value deduced by Larkins and Hannaford from measurements of the frequencies at 206.1 nm and 547.7 nm using a grating spectrometer lies even further away, though their estimation of the isotope shift at 206.1 nm was noticeably good. The amplitudes of the hyperfine components are com-

TABLE V. Frequencies and isotope shift of the $4d^{10}5s^2S_{1/2} \rightarrow 4d^{10}6p^2P_{3/2}$ at 206 nm.

Isotope	$F \rightarrow F'$	Relative intensity	Frequency (MHz)	Others (MHz)	
				Ref. [12]	Ref. [17]
107	0 \rightarrow 1	2	1 454 016 647 (12)		
	1 \rightarrow 1	1	1 454 018 359 (12)		
	1 \rightarrow 2	5	1 454 018 341 (12)		
	Centroïd 107		1 454 017 920 (12)		
109	0 \rightarrow 1	2	1 454 016 035 (12)		
	1 \rightarrow 1	1	1 454 018 012 (12)		
	1 \rightarrow 2	5	1 454 017 992 (12)		
	Centroïd 109		1 454 017 505 (12)		
Centroïd for natural silver			1 454 017 720 (12)	1 454 017 375 (240)	1 454 017 694 (45)
Isotope shift $\nu_{109} - \nu_{107}$			-414.45(62)	-410(50)	

patible with our calculation of the transition rate [8] using Cowan codes.

VII. CONCLUSION

We have performed measurements of frequency, isotope shift, and hyperfine structure of the $4d^{10}5s^2S_{1/2} \rightarrow 4d^95s^2D_{5/2}$ transition in atomic silver. As far as we know, this is the first observation and measurement of this very narrow two-photon transition. Our measurement of the frequencies of hyperfine components is a three order of magnitude improvement over previous values. Our values of hyperfine constants are compatible with values more precisely measured by radio-frequency spectroscopy. Finally, we have performed the first measurement of the isotope shift of the transition. The excellent signal-to-noise ratio obtained using thermal atoms opens the way to higher resolution studies on cold atoms.

ACKNOWLEDGMENTS

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APPENDIX A: LIGHT SHIFT CORRECTION

We have corrected the measured frequencies using a simple model of light shift in a Gaussian energy distribution. The frequency of a transition between the g and e levels is modified by light shift as follows:

$$\omega_{eg}(I) = \omega_{eg}(0) + c_{ls}I,$$

where $\omega_{eg}(0)$ is the transition frequency at zero power and I the incident intensity. For the two-photon transition in silver, the light shift coefficient c_{ls} has been recently calculated [20] using two multiconfigurational self-consistent-field methods, $c_{ls}/2\pi = -1.42(0.48) \times 10^{-3} \text{ Hz m}^2 \text{ W}^{-1}$. If the original line shape of this transition is a Lorentzian curve of effective width Γ_e , the light shift induced by a beam of uniform inten-

sity I_0 will modify the transition rate Γ_g expression as follows:

$$\Gamma_g \propto I_0^2 \frac{\Gamma_e}{[2\omega - \omega_{eg}(0) - c_{ls}I_0]^2 + \left(\frac{\Gamma_e}{2}\right)^2}.$$

In the case of a Gaussian TEM₀₀ beam of power P and waist w_0 , the intensity distribution varies as

$$I(r, z) = I_0 \left(\frac{w_0}{w(z)}\right)^2 e^{-[2r^2(z)/w^2(z)]} \quad (\text{A1})$$

$$\approx I_0 e^{-(2r^2/w^2)}, \quad \text{where } I_0 = \frac{2P}{\pi w_0^2}. \quad (\text{A2})$$

If the dimensions of the interaction region are small compared to the Rayleigh length of the beam, then the z dependence of the intensity can be ignored. With $\Omega = 2\omega - \omega_{eg}(0)$ and ω the frequency of a single photon at 661.3 nm, the transition rate in the Gaussian beam is

$$\Gamma_g \propto \int_0^\infty \frac{\Gamma_e 2\pi r dr}{(\Omega - c_{ls}I_0 e^{-2r^2/w_0^2})^2 + \left(\frac{\Gamma_e}{2}\right)^2} (I_0 e^{-2r^2/w_0^2})^2, \quad (\text{A3})$$

which becomes

$$\Gamma_g = 4K\Omega \left[\arctan\left(\frac{2\Omega}{\Gamma_e}\right) - \arctan\left(\frac{\Omega - c_{ls}I_0}{\frac{\Gamma_e}{2}}\right) \right] - K\Gamma_e \ln \left(\frac{\Omega^2 + \left(\frac{\Gamma_e}{2}\right)^2}{(\Omega - c_{ls}I_0)^2 + \left(\frac{\Gamma_e}{2}\right)^2} \right), \quad (\text{A4})$$

$$\text{where } K = \left(\frac{4\pi a_0^2}{m_e c^2 \alpha} \right)^2 |\gamma|^2 \frac{\pi w_0^2}{4c_{ls}^2}. \quad (\text{A5})$$

From Eq. (A4), one can see that light shift has a multiple influence on the experimental line: shift to red, asymmetry and broadening. If the shift is small with respect to the original effective width Γ_e , the deformation is negligible. In our case, $\Gamma_e/2\pi$ is about 1.1 MHz and the shift is about 145 kHz for $P=12$ W and $w_0=97$ μm . The deformation is small (center of gravity and frequency at the top of the line are distant by less than 1% of the linewidth) and the broadening is about 5%. Given the power at 661.3 nm in the build-up cavity, we could simulate the light shift and deformation and correct the

center frequency of each spectrum at 661.3 nm. Typical corrections (for one photon at 661.3 nm) are 150–220 kHz with a very conservative 100% uncertainty.

APPENDIX B: FREQUENCIES OF THE 206 nm LINE

The frequencies of the $4d^{10}5s^2S_{1/2} \rightarrow 4d^{10}6p^2P_{3/2}$ transition at 206.1 nm can be easily deduced from the frequencies of the two-photon transition at 661.3 nm and the frequencies of the $4d^95s^5^2D_{5/2} \rightarrow 4d^{10}6p^2P_{3/2}$ transition at 547.7 nm. They are summarized in Table V. It is noticeable that the estimated value of isotope shift $\Delta\nu_{\text{iso}}^{206,1} = -410(50)$ MHz of Ref. [12] is in very good agreement with that deduced from our measurements.

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