Letters

Giant modification of atomic transition probabilities induced by a magnetic field: forbidden transitions become predominant

Armen Sargsyan¹, Ara Tonoyan¹, Grant Hakhumyan¹, Aram Papoyan¹, Emilio Mariotti² and David Sarkisyan¹

¹ Institute for Physical Research, NAS of Armenia, Ashtarak-2, 0203, Armenia
² CNISM–Department of Physical, Earth and Environmental Sciences, University of Siena, Via Roma 56, 53100 Siena, Italy

E-mail: papoyan@ipr.sci.am

Received 25 October 2013, revised 6 March 2014
Accepted for publication 7 March 2014 Published 4 April 2014

Abstract
The magnetic field-induced giant modification of probabilities for seven components of $6S_{1/2}, F_g=3 \rightarrow 6P_{3/2}, F_e=5$ transition of the Cs D₂ line, forbidden by selection rules, is observed experimentally for the first time. For the case of excitation with circularly polarized laser radiation, the probability of a $F_g=3, m_F=-3 \rightarrow F_e=5, m_F=-2$ transition becomes the largest of 25 transitions of the $F_g=3 \rightarrow F_e=2,3,4,5$ group in a wide-range magnetic field of 200–3200 G. Moreover, the modification is the largest among D₂ lines of alkali metals. A half-wave-thick cell (the length along the beam propagation axis $L=426$ nm) filled with Cs has been used in order to achieve sub-Doppler resolution, which allows the large number of atomic transitions that appear in the absorption spectrum to be separated when an external magnetic field is applied. For $B > 3000$ G the group of seven transitions $F_g=3 \rightarrow F_e=5$ is completely resolved and is located at the high frequency level of $F_g=3 \rightarrow F_e=2,3,4$ transitions. The applied theoretical model describes very well the experimental curves.

Keywords: hyperfine structure, optical nanocells, sub-Doppler spectroscopy, transition probability, selection rules, Zeeman effect, Paschen–Back effect

(Some figures may appear in colour only in the online journal)

1. Introduction

Alkali atoms are widely used in atomic physics due to the simplicity of their electronic structure and their strong atomic transitions from the ground state with wavelengths in visible and near-infrared, where diode lasers with good parameters are available. Cesium atoms are widely used in laser cooling experiments, information storage, spectroscopy, magnetometry, laser frequency stabilization, etc [1–3]. That is why any knowledge of the behavior of Cs atomic transitions, particularly in an external magnetic field, is of high importance. It is well known that in a quite moderate magnetic field $B$ the splitting of atomic energy levels to Zeeman sublevels deviates from linear behavior and the atomic transition probabilities undergo significant changes [4–6]. The simplest and most straightforward technique to study such a modification of atomic transitions (whose frequency distance belongs to the optical domain) is laser spectroscopy of atoms contained in an atomic vapor cell. For $B$ up to $\approx 1000$ G, the split Zeeman transitions remain overlapped because of Doppler broadening, and sub-Doppler techniques have to be implemented in order to spectrally...
As was demonstrated earlier, strong narrowing in the absorption spectrum can be attained with the use of an atomic vapor cell with a half-wavelength thickness \( L = \lambda/2 \), where \( \lambda \) is the resonant wavelength of laser radiation, \( L = 426 \text{ nm} \) for the case of the Cs D\(_2\) line [8–11]. Particularly, the absorption linewidth for the Cs D\(_2\) line reduces to \( \pm 100 \text{ MHz} \) full-width at half-maximum (FWHM), as opposed to \( \pm 400 \text{ MHz} \) in an ordinary cell. Moreover, the absorption lines for \( L = \lambda/2 \) exhibit a Voigt profile (a convolution of Lorentzian and Gaussian profiles) with a sharp (nearly Gaussian) peak, which allows separation of closely spaced individual transitions and the study of their transition probabilities in an external magnetic field. In addition, the \( \lambda/2 \)-method is tolerant to a 10% deviation of thickness (weak influence on the absorption linewidth). These benefits make it convenient to use the \( \lambda/2 \)-method for studies of closely spaced individual atomic transition components in a magnetic field.

In this letter we present, for the first time, the results of experimental and theoretical studies showing a giant transition probability modification for the Cs D\(_2\) line 6\(S_{1/2}\)–6\(P_{3/2}\) transition induced by a magnetic field. The \( \lambda/2 \)-method is tolerant to a 10% deviation of thickness (weak influence on the absorption linewidth). These benefits make it convenient to use the \( \lambda/2 \)-method for studies of closely spaced individual atomic transition components in a magnetic field.

2. Experimental details

Nanometric thin cells (NTCs), filled with Cs, have been used in our experiment because of their peculiar properties, which allow (a) the direct obtainment of sub-Doppler spectra without application of nonlinear techniques, and therefore (b) the resolution of even more complicated spectra. This has already been demonstrated in many papers over the last few years since the introduction of NTCs to laser spectroscopy [8].

The general design of NTCs is similar to that described in [12]. A compact oven was used to set the required temperature regime.

The temperature was set to 100\(^\circ\)C, which corresponds to the number density of isotopically pure \(^{133}\)Cs atoms \( N \approx 10^{13} \text{ cm}\(^{-3}\) \) sufficient to obtain >0.02% peak absorption for all of the studied individual transitions. Adjustment of the needed vapor column thickness without variation of thermal conditions was attained by smooth vertical translation of the cell+oven assembly.

A schematic diagram of the optical part of the experimental setup is shown in figure 1. A circularly polarized laser radiation beam (\( \lambda = 852 \text{ nm} \), \( P_1 = 5\text{ mW}, \Delta \nu_1 = 1 \text{ MHz} \)) resonant with the Cs D\(_2\) line was focused (\( \phi = 0.5 \text{ mm} \)) onto a Cs NTC with a vapor column of thickness \( L = \lambda/2 \) at normal incidence angle. The 8 mm thick assembly of the oven, with the main NTC inside, was placed between two permanent ring magnets (whose axis was directed along the laser radiation propagation direction \( k \)) with a gradually adjustable spacing, which provided a controllable longitudinal B-field (for details, see [6]). The extremely small thickness of NTCs is once more advantageous for the application of a very strong magnetic field, using permanent magnets which are otherwise unusable because of strong inhomogeneity: in NTCs, the variation of the B-field inside the cell is several orders less than the applied B value [13]. To record transmission and fluorescence spectra, the laser radiation was linearly scanned within up to a 15 GHz spectral region covering the studied group of transitions. The nonlinearity of the scanned frequency (<1% throughout the spectral range) was monitored by simultaneously recorded transmission spectra of a Fabry–Pérot etalon (not shown). About 30% of the pump power was branched to an auxiliary Cs NTC with thickness \( L = \lambda/2 \), providing a reference absorption spectrum for \( B = 0 \). All the spectra were detected by photodiodes with amplifiers followed by a four channel digital storage oscilloscope Tektronix TDS 2014B.

The diagram of relevant transitions between Zeeman sublevels of ground (\( F_g = 3 \)) and excited (\( F_e = 4, 5 \)) states for the D\(_2\) line of \(^{133}\)Cs (nuclear spin \( I = 7/2 \)) with \( \sigma^+ \) (left-hand) laser excitation. Transition labels are given in circles.

The temperature was set to 100\(^\circ\)C, which corresponds to the number density of isotopically pure \(^{133}\)Cs atoms \( N \approx 10^{13} \text{ cm}\(^{-3}\) \) sufficient to obtain >0.02% peak absorption for all of the studied individual transitions. Adjustment of the needed vapor column thickness without variation of thermal conditions was attained by smooth vertical translation of the cell+oven assembly.

A schematic diagram of the optical part of the experimental setup is shown in figure 1. A circularly polarized laser radiation beam (\( \lambda = 852 \text{ nm} \), \( P_1 = 5\text{ mW}, \Delta \nu_1 = 1 \text{ MHz} \)) resonant with the Cs D\(_2\) line was focused (\( \phi = 0.5 \text{ mm} \)) onto a Cs NTC with a vapor column of thickness \( L = \lambda/2 \) at normal incidence angle. The 8 mm thick assembly of the oven, with the main NTC inside, was placed between two permanent ring magnets (whose axis was directed along the laser radiation propagation direction \( k \)) with a gradually adjustable spacing, which provided a controllable longitudinal B-field (for details, see [6]). The extremely small thickness of NTCs is once more advantageous for the application of a very strong magnetic field, using permanent magnets which are otherwise unusable because of strong inhomogeneity: in NTCs, the variation of the B-field inside the cell is several orders less than the applied B value [13]. To record transmission and fluorescence spectra, the laser radiation was linearly scanned within up to a 15 GHz spectral region covering the studied group of transitions. The nonlinearity of the scanned frequency (<1% throughout the spectral range) was monitored by simultaneously recorded transmission spectra of a Fabry–Pérot etalon (not shown). About 30% of the pump power was branched to an auxiliary Cs NTC with thickness \( L = \lambda/2 \), providing a reference absorption spectrum for \( B = 0 \). All the spectra were detected by photodiodes with amplifiers followed by a four channel digital storage oscilloscope Tektronix TDS 2014B.
components appear with \(\pm 100\) MHz linewidth, thus being completely frequency resolved except for transitions 5, 6 and 7', resolved partially, and 6' and 7, which are fully overlapped. As can be seen from the inset, the amplitudes of 5, 6 and 7', which are fully overlapped.

The absorption spectrum for \(B = 3450\) G and otherwise invarian
tleum density, etc), it is expedient to present also the dependence of the frequency shifts and probabilities presented below have been performed using the formulas (1)–(7) given in [11].

The dependence of frequency shifts of transitions 1'–7' and 1–7 in a magnetic field, relative to the position of the \(F_g=3\rightarrow F_e=4\) transition at \(B = 0\) for the case of \(\sigma^+\) laser excitation, is shown in figure 6. Good agreement of theory and experiment is observed throughout the whole explored range of the \(B\)-field (up to 3500 G).

The calculated dependence of 1'–7' and 1–7 transition probabilities (absorption amplitudes) in a magnetic field, for the case of \(\sigma^+\) laser excitation, are shown in figure 7.

Since the absolute value of the absorption amplitude \(A\) depends on parameters of the experiment (laser intensity, atomic density, etc), it is expedient to present also the \(B\)-field dependence of the ratio of absorption amplitudes \(A_i\) of 1'–7' transitions to absorption amplitude \(A_7\) of the transition 7. The latter is the strongest in the \(F_g=3\rightarrow F_e=4\) group; moreover, the absorption amplitude \(A_7\) is nearly constant in a wide range 250 G \(< B < 4000\) G (see figure 7), which makes it convenient to use as a reference. The theoretical ratio \(A_i/A_7\) versus \(B\)-field is plotted in figure 8, together with experimental results (the ratio is easily measurable). The dashed line marks the unity ratio. As can be seen in figure 8, \(A_7/A_{1'} > 1\) holds in a wide range 200 G \(< B < 3200\) G, and the maximum value of the ratio is 2.3.

Thus, the transition 7', as well as 6' and 5', which are forbidden for \(B = 0\), undergo giant modification under the...
A Sargsyan et al

Zeeman transitions of the Cs D2 line. Fg = 3 maximum probability value for B = 60 G is over 30 times smaller than the B value achieved at induced by magnetic field. However, its maximum probability the Cs atom occurs for > B0. All the individual Zeeman transitions 1–7 and 1–7′ are completely resolved. Two shifted components of Fg = 4 → Fc = 5 transition appear at the low frequency region.

Figure 5. Absorption spectrum of Cs NTC with L = λ/2 for B = 3450 G and σ+ laser excitation. For transition labels, see figure 2. The bottom-left curve is the absorption spectrum of the reference NTC showing positions of Fg = 3 → Fc = 2,3,4 transitions for B = 0. All the individual Zeeman transitions 1–7 and 1–7′ are completely resolved. Two shifted components of Fg = 4 → Fc = 5 transition appear at the low frequency region.

Figure 6. Magnetic field dependence of frequency shift for atomic transition labeled 1–7 and 1–7 relative to the position of Fg = 3 → Fc = 4 transition at B = 0. Black squares: experimental results (inaccuracy ≈ 2%); solid curves: calculated dependence.

influence of a magnetic field, becoming predominant over the initially allowed transitions 1–7. It is worth noting that the maximum value of transition probability for a 7′ transition reaches 76% of the probability for a Fg = 4, mF = +4 → Fc = 5, mF = +5 transition, which is the strongest among all the 54 Zeeman transitions of the Cs D2 line.

We want also to stress that another group of transitions, forbidden at B = 0, Fg = 4 → Fc = 2 (five transition components for the case of σ+ excitation), also exhibits modification induced by magnetic field. However, its maximum probability value achieved at B = 60 G is over 30 times smaller than the maximum probability value for Fg = 3 → Fc = 5.

The decoupling of the total angular momentum J and the nuclear momentum I (hyperfine Paschen–Back regime) for the Cs atom occurs for B ≫ B0 = A_{HFS}/μB ≈ 1700 G, where

A_{HFS} = h × 2.3 GHz is the magnetic dipole hyperfine constant for 6S_{1/2}, and μB is the Bohr magneton. In this case the splitting of transitions is described by the projections mJ and mI [13–18]. At B > 6000 G, 16 transitions are observable in the absorption spectrum: by eight starting from the ground states 6S_{1/2}, mJ = −1/2 and 6S_{1/2}, mJ = +1/2. For B > 3000 G and σ+ excitation, the group of Fg = 3 → Fc = 5 transitions is always located at the high-frequency side of Fg = 3 → Fc = 2,3,4 transitions, with line intensities monotonically reducing and completely vanishing at B > 9000 G.

It is interesting to compare the maximum probability of a magnetic field-induced Cs Fg = 3 → Fc = 5 transition, normalized to the strongest D2 transition (the calculated value is 0.76), with the corresponding values for D2 line transitions, forbidden at B = 0, for other alkali metal atoms of practical interest. Our theoretical calculations show that for Rb transitions 85Rb 5S_{1/2}, Fg = 2 → 5P_{3/2}, Fc = 4 and 87Rb 5S_{1/2}, Fg = 1 → 5P_{3/2}, Fc = 3, the maximum value is smaller than in the case of Cs 1.10 and 1.36 times, correspondingly. Also for 3S_{1/2}, Fg = 1 → 3P_{3/2}, Fc = 3 transitions of Na and 4S_{3/2}, Fg = 1 → 4P_{3/2}, Fc = 3 of K the maximum values are smaller.
than in the case of Cs (1.31 and 1.33 times, correspondingly). Thus, the modification of the probabilities for cesium $F_g=3 \rightarrow F_e=5$ transitions is the strongest among the widely used alkali atoms.

4. Conclusion and outlook

Giant modification of probabilities of Cs D2 line transitions 6S$_{1/2}$, $F_g=3 \rightarrow$ 6P$_{3/2}$, $F_e=5$ induced by magnetic field B has been studied both experimentally and theoretically. It has been revealed for the first time that the absorption intensity for $F_g=3$, $m_F=-3 \rightarrow F_e=5$, $m_F=-2$ transition becomes the largest among all the 25 Zeeman transitions of the $F_g=3 \rightarrow F_e=2,3,4,5$ manifold for the case of $\sigma^+$ excitation in a wide range of magnetic field 200–3200 G. It is demonstrated that utilization of a half-wavelength-thick cell filled with Cs, favorable for strong reduction of Doppler broadening of absorption lines, allows the quantitative study of frequency positions and the modification of individual transition probabilities. As a result of a special interest, for $B > 3000$ G all the seven transitions of the $F_g=3 \rightarrow F_e=5$ group are completely separated. Calculated theoretical curves for transition frequency shifts and modification of probabilities induced by a magnetic field for all the transitions under study show very good coincidence with the experimental results.

The largest probabilities of ‘forbidden’ transitions labeled 7’, 6’ and 5’ in a wide range of magnetic field (see figure 8) make them attractive for formation of sub-natural electromagnetically induced transparency resonances in new frequency regions [3, 19] (for the realization of this experiment, the coupling laser frequency must be in resonance with $F_g=4 \rightarrow F_e=5$, which is easy to realize). It should be noted that the $h/2$-M method can be implemented successfully to study forbidden transitions of D2 lines of other alkalis, such as Rb, K and Na.

Acknowledgments

The authors are grateful to A Sarkisyan for his valuable participation in the development and fabrication of the NTC. This work has received partial funding from the EU Seventh Framework Programme (FP7/2007–2013) under Grant Agreement no 295264-COSMA. AS, GH and DS acknowledge financial support from the State Committee Science, MES of Armenia (Research Project No 13-1C029 and 13-1C089).

References

[1] Budker D, Kimball D F and DeMille D P 2008 Atomic Physics, Exploration Through Problems and Solutions 2nd edn (Oxford: Oxford University Press)
[2] Auzinsh M, Budker D and Rochester S M 2010 Optically Polarized Atoms: Understanding Light-Atom Interactions (Oxford: Oxford University Press)
[3] Kargapoltsvev S V, Kitching J, Hollberg L, Taichenachev A V, Velichansky V L and Yudin V I 2004 Laser Phys. Lett. 1 495
[4] Tremblay P, Michaud A, Levesque M, Thériault S, Breton M, Beaubien J and Cyr N 1990 Phys. Rev. A 42 2766
[5] Alexandrov E B, Châka M P and Khvostenko G I 1993 Interference of Atomic States (Berlin: Springer)
[6] Sargsyan A, Hakhumyan G, Papoyan A, Sarkisyan D, Atvars A and Auzinsh M 2008 Appl. Phys. Lett. 93 021119
[7] Sargsyan A, Sarkisyan D, Papoyan A, Pashayan-Leroy Y, Moroshkin P, Weis A, Khanbekyan A, Mariotti E and Moi L 2008 Laser Phys. 18 749
[8] Dutier G, Yarovitski A, Saltiel S, Papoyan A, Sarkisyan D, Bloch D and Ducloy M 2003 Europhys. Lett. 63 35
[9] Andreeva C, Cartaleva S, Petrov L, Saltiel S M, Sarkisyan D, Varzhapetyan T, Bloch D and Ducloy M 2007 Phys. Rev. A 76 013837
[10] Gazayazn E A, Papoyan A V, Sarkisyan D and Weis A 2007 Laser Phys. Lett. 4 801
[11] Hakhumyan G, Leroy C, Mirzoyan R, Pashayan-Leroy Y and Sarkisyan D 2012 Eur. Phys. J. D 66 119
[12] Keaveney J, Sargsyan A, Krohn U, Sarkisyan D, Hughes I G and Adams Ch S 2012 Phys. Rev. Lett. 108 173601
[13] Sargsyan A, Hakhumyan G, Leroy C, Pashayan-Leroy Y, Papoyan A and Sarkisyan D 2012 Opt. Lett. 37 1379
[14] Olsen B A, Patton B, Jau Y Y and Happer W 2011 Phys. Rev. A 84 063410
[15] Weller L, Kleinbach K S, Zentile M A, Knappe S, Adams Ch S and Hughes I G 2012 Opt. Lett. 37 3405
[16] Sargsyan A, Mirzoyan R, Papoyan A and Sarkisyan D 2012 Opt. Lett. 37 4871
[17] Weller L, Kleinbach K S, Zentile M A, Knappe S, Adams Ch S and Hughes I G 2012 J. Phys. B: At. Mol. Opt. Phys. 45 215005
[18] Sargsyan A, Mirzoyan R and Sarkisyan D 2012 JETP Lett. 96 303
[19] Fleischhauer M, Imamoglu A and Marangos J P 2005 Rev. Mod. Phys. 77 633