Collisional depolarization of Zeeman coherences in the $^{133}\text{Cs} \, 6p \, 2P_{3/2}$ level: Double-resonance two-photon polarization spectroscopy

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An experimental investigation of the Cs $6s^2S_{1/2} \rightarrow 6p^2P_{3/2} \rightarrow 10s^2S_{1/2}$ double-resonance two-photon polarization spectrum has been made. In the investigation, a linear polarization degree was measured to extract the alignment depolarization cross section in the cesium $6p \, 2P_{3/2}$ level due to collisions between ground-level argon and excited $6p \, 2P_{3/2}$ cesium atoms over the Zeeman coherences. The spectra show strong depolarization of the Cs $6p \, 2P_{3/2}$ level. The measured alignment depolarization cross section value of 186 (58) $\text{Å}^2$ is in good agreement with the experiment and theoretical predictions.

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I. INTRODUCTION

Experimental studies of depolarization in excited level alkali-metal atoms under the influence of collisions with rare-gas molecules can provide valuable information on the relaxation rates of the electronic moments and the state-multipole-dependent depolarization cross sections [1]. Collisions between alkali-metal and rare-gas atoms are important in numerous applications, particularly for remotely sensing the composition of planetary atmospheres, the interstellar medium, and fusion plasmas. The study of collisional dynamics is not new, but recent advances in techniques for cooling and trapping of atoms has enormously increased the interest in this area of atomic and molecular physics. For example, high-resolution molecular spectroscopy of colliding cold atoms has become a leading experimental technique to investigate the collisional dynamics and to accurately determine the potential curves of molecules [2,3].

Measurement of the depolarization cross sections of $2P_{3/2}$ alkali-metal atoms, up to now, was accomplished using incoherent light sources (alkali lamps) and magnetic fields to decouple the nuclear spin from the total angular momentum. In particular, Guiry et al. [4] has experimentally studied the alignment-dependent depolarization collisions between the $6p \, 2P_{3/2}$ Cs atoms with ground level argon atoms using linearly polarized Cs resonance radiation. In their work, after the alignment is optically produced in the excited level, the polarized Cs atoms become depolarized in subsequent collisions with Ar atoms using a Zeeman scanning technique. In the technique, an external magnetic field (0–10 kG) is applied in order to decouple the nuclear spin from the electronic angular momentum. There are no other experimental data available for comparison of the alignment-dependent depolarization cross section of the Cs $6p \, 2P_{3/2}$ level. Theoretical depolarization cross section $\sigma_2$ for the spin-coupled and decoupled cases was studied in Refs. [5,6]. Although there are large discrepancies in the value of $\sigma_2$, there is broad agreement between the theory and experiment within experimental error in both spin-coupled and decoupled cases.

Therefore, we focus on measurements of the depolarization cross section, for comparison, specifically the alignment produced in the $6p \, 2P_{3/2}$ Cs atoms.

While highly sophisticated narrow bandwidth lasers are available for high resolution spectroscopy to investigate collisional dynamics in excited level alkali, time resolved double-resonance two-photon polarization spectroscopy is still very attractive. It allows high-resolution spectroscopic investigations employing relatively simple pulsed broadband dye lasers. These pulsed dye lasers can be temporally overlapped using a pump-probe technique to create coherences in the Zeeman sublevels of the excited level alkali, and thus collisional dynamics between the alkali and buffer gas can be studied. Although the polarization spectroscopy using the two-photon pump-probe technique provides a wealth of information on the collisional dynamics, such as excited level hyperfine splittings and collisional depolarization cross sections, [7], there are no experimental observations of $\sigma_2$ of the $6p \, 2P_{3/2}$ Cs atoms using such a technique. We report on the alignment-dependent depolarization cross section of the $6p \, 2P_{3/2}$ Cs atoms due to collisions with the ground level Ar atoms based on the double-resonance two-photon polarization spectroscopy. In the experiment, a short-pulse pump-probe technique was employed to obtain a linear polarization spectrum as a function of the Ar buffer gas pressure. This method is a direct measure of the importance of the polarization to the alignment-dependent inelastic process in alkali-rare-gas collisions. The depolarization cross section is extracted from the linear polarization spectrum. Our measurements are unique in that they are sensitive only to the relative polarization directions of the pump and probe lasers. Our results agree, within stated errors, with the results of Ref. [4].

II. EXPERIMENTAL APPROACH

The schematic overview of the experimental apparatus is shown in Fig. 1. Two dye lasers pumped by a pulsed neodymium-doped yttrium aluminum garnet (Nd:YAG) laser are used for pumping and probing the atomic cesium. The output from the second harmonic generator of the Nd:YAG
laser produces light at 532 nm with an average power of 2 W at a pulse repetition rate of 20 Hz. The dye laser oscillators are constructed using a grazing incidence Littman-Metclaf cavity and operate in a single transverse mode. Dye laser 1 runs with a dye circulating system to maintain the power stable at about 1 mW with a bandwidth of approximately 0.9 GHz. The output power of dye laser 2 after the oscillator is increased by about a factor of 10 using a dye amplification system generates an average power of about 4 mW. The dye laser 1 has a bandwidth of approximately 300 MHz. The temporal pulse width of the dye lasers was measured to be ~6.5 ns using an ultraviolet photodetector with 0.2 ns rise time.

Dye laser 1 (pump) is on resonance at 852.1 nm (D2 line) to promote the atoms from the 6S 3/2 ground level to the 6P 3/2 level, and dye laser 2 (probe) is on resonance for the 6P 1/2 → 10S 1/2 transition at 603.4 nm. The frequency of the pump laser is fixed at the Cs D2 line while the frequency of the probe laser is tuned at ±11 cm⁻¹ in the vicinity of the 6P 1/2 → 10S 1/2 resonance transition.

The output of the lasers is strongly polarized by using Glan-Thompson prisms which have extinction ratios of better than 10⁻⁵. After purifying the polarization state of the lasers, a liquid crystal variable retarder (LCR), with an extinction ratio of 10⁻³, is used in the probe laser path. It should be noted that the LCR is a temperature sensitive device. Thus, electrical components of the LCR cavity are maintained by a temperature controller with an accuracy of ±0.01 °C. The LCR alternately switches the polarization direction of the probe laser parallel (along a laboratory z axis) and perpendicular (x axis) to that of the pump laser by applying a voltage to the retarder.

The pyrex cesium cells, 25.4 mm diameter by 50.8 mm length, with and without buffer gas at various pressures, were prepared using an oil-free vacuum system. [8]. The background pressure of the pure Cs cell is about 10⁻⁸ mbar. Our cells are placed inside a resistively heated nonmagnetic cylindrical aluminum oven wrapped with an aluminum oxide blanket to maintain a uniform temperature inside the oven. The cell temperature is stabilized to ±0.01 °C using a grounded thermocouple attached to the cell, and its temperature is varied by the heater as the thermocouple feeds back to the temperature controller. A bi-convex mirror is used behind the cell to collect more signal at the detection side of the window.

The signal was observed at the oven window by using a photomultiplier tube (PMT) at right angles to the propagation direction of the lasers which copropagate into the oven-cell system. A combination of interference and color glass filters were used in front of the PMT in order to remove background due to scattered and atomic resonance fluorescence. The signals were detected by measuring the cascade fluorescence from the 9p 3/2 → 6s 3/2 transition. The output from the PMT signal is then sent to a three-stage amplifier and gated boxcar averager with a gate width of 600 ns opened 1 ns after the lasers fire. That delay along with the gatewidth of the boxcar, the shielded cables and grounded optical table, eliminated electronic pickups and noise from the observed signal. The boxcar averager operates in a shot-to-shot mode and the signal detected within the gatewidth is digitized and stored while monitoring the size of the signal in real time. Our typical signal size is about 10³ photons for each laser pulse shot.

The ratio of the signals, detected when polarization states of the lasers are collinear (S‖) and perpendicular (S⊥) to each other, generates a linear polarization spectrum. A linear polarization degree can be expressed as

\[ P_L = \frac{S - S_\perp}{S + S_\perp}. \]  

The measured S‖ and S⊥ signals contain two-photon transition intensities \( I_1 \) and \( I_\perp \) as \( S_\parallel = I_1 I(9p - 6s) \) and \( S_\perp = I_\perp I(9p - 6s) \). Here, the \( I_1 = I_{1,1} J_{2,2} \) and \( I_\perp = I_{1,1} J_{2,1} \) intensities, spectrally integrated over the on resonant atomic line, are proportional to the total population in the final 10S 3/2 level, and \( I(9p - 6s) \) is the total intensity of the cascade fluorescence. The important transitions and their corresponding wavelengths [9] are indicated in Fig. 2.

It is important to point out that measuring a linear polarization degree has a significant advantage in reducing the experimental background. An absolute intensity ratio of the signals is sensitive only to the relative polarization directions of the lasers. Thus, any variations of the lasers intensities...
with experimental factors such as absorbing medium density, fluorescence background, sensitivity of the gated boxcar averager and amplifier, collectively do not affect the intensity ratio.

A typical experimental protocol is as follows: the frequency of the probe laser was measured by a wavemeter and the signals from several data runs were averaged at two different polarization states. The frequency of the probe laser was then detuned by 0.001 nm using a computer-controlled stepper motor installed on the tuning mirror mounting stage. For each detuning, the same procedure is repeated to determine the polarization dependence in the excitation spectrum at that detuning. The polarization switch of the lasers, the boxcar signal level, the control of the probe laser frequency scan interval, and the LCR are all managed by a computer controlled data acquisition system using LabVIEW programming. A typical polarization spectrum scan for the $6S_1/2 \rightarrow 6p^2P_{3/2} \rightarrow 10S_1/2$ transition is shown in Fig. 3.

III. RESULTS AND DISCUSSION

A. Systematic effects on the polarization spectrum

In this paper, our measurements were made using a short pulse pump-probe technique. One of the advantages of using this technique is that one can essentially eliminate the systematic effects related to on-resonance excitations. The main systematic effects that tend to reduce the measured polarization degree are radiation trapping and unobserved hyperfine structure. Therefore, two critical time scales play an important role: the radiative decay time and the shortest hyperfine precession time of the excited level.

First, due to the short pump-probe temporal overlap time ($T \approx 1$ ns) compared to the radiative lifetime of the excited level ($\tau \approx 33$ ns), approximately 3% of the excited atoms will have radiatively decayed during the pulse. Thus, radiation trapping has a negligible effect on the excited level. We have confirmed our results by varying the Cs cell temperature, as shown in Fig. 4.

Second, one could effectively eliminate the depolarization due to unobserved hyperfine structure if the temporal overlap time of the pulse lasers is made much smaller than the inverse of the highest hyperfine frequency component of the excited level. For the atoms with nonzero nuclear spin $I$, electronic angular momentum $J$ of the excited level couples with $I$ to form the total atomic angular momentum $F$. In our case, the nuclear spin $I=7/2$ of $^{133}$Cs [10] couples with $J=3/2$ and initiates the precession between $J$ and $I$ about $F$. This introduces an oscillating time dependence in the average tensor multipoles. In our experimental condition, that is the average axisymmetric alignment $\langle A_s(t) \rangle$ of the excited cesium atoms. Alignment, a second rank tensor, is a measure of the quadrupole moment of the excited level produced by excitation of an initially unpolarized ground state cesium atomic gas with the linearly polarized pump laser. The polarization direction of this laser defines the axis of symmetry of the $6p^2P_{3/2}$ electronic charge distribution. Thus, the time evolution of the electronic alignment under the influence of the hyperfine-structure interaction can be described as,

$$\langle A_s(t) \rangle = \langle A_0 \rangle g^{(2)}(t),$$

where $\langle A_0 \rangle$ is defined in terms of the angular momentum operators [11–14] as

$$\langle A_0 \rangle = \frac{(3J_z^2 - J^2)}{J'(J' + 1)} = \sum_{m'} |a(m')|^2 \frac{3m'^2 - J'(J' + 1)}{J'(J' + 1)}.$$

In Eq. (2), $g^{(2)}$ is the depolarization coefficient in the alignment, and can be represented in the form
$g^{(2)}(t) = \sum_{F,F'} \frac{(2F+1)(2F'+1)}{(2I+1)} W^2(k)\cos(w_{F,F'} t)$,

where $W(k) = W(JFJ'F'; I2)$ is the Racah coefficient and $w_{F,F'}$ is the frequency splitting between two hyperfine levels. The $g^{(2)}(t)$ coefficient decreases from its maximum value of 1 as $t$ becomes greater than zero and oscillates between the values of 1 and −1. This oscillation changes the average alignment in the excited level. However, by overlapping the pump and probe lasers for less than the hyperfine precession time ($T \leq 1/w_{FF}$) by delaying the arrival of the probe laser at the interaction region of the cell, one can freeze out the electronic alignment of the excited level. That is, the atoms promoted to the final state by the double-resonance two-photon excitation emit the photons before the hyperfine precessional motion can take place.

The temporal overlap time $T$ of the pulses can easily be made much shorter than the hyperfine precession time for selected excited levels of alkalies, specifically in Rb [15]. However, in the case of Cs, we have observed that the average value of the alignment over the pump-probe laser widths reduced the $g^{(2)}$ value from its maximum value of 1. This is the case even though we delayed the arrival time of the probe laser so that $T$ is approximately 1 ns while the inverse of the highest hyperfine frequency component in the excited level is 1.6 ns. Due to comparable time scales and unknown laser pulse shapes, the effective pulse length of the dye lasers may still be larger than 1 ns. Under this condition, the alignment is expected to be perturbed by the hyperfine interaction which in return decreases the polarization spectrum.

The linear polarization degree can be defined theoretically in terms of alignment for the right-angle observation [11] of the experiment as

$$P_L = \frac{3h^{(2)}(J,J')(A_0)g^{(2)}(t)}{4 + h^{(2)}(J,J')(A_0)g^{(2)}(t)},$$

where $h^{(2)}(J,J')$ is a ratio of Racah coefficients that is derived from the reduced matrix elements of the density matrices and depend on the angular momenta of the initial $J$ and final $J'$ levels. The value of $h^{(2)}(3/2,1/2)$ is $-5/4$ for the $2P_{3/2} - 2S_{1/2}$ excitation. In the absence of any systematic effects such as collisions, radiative trapping, and hyperfine structure, the alignment produced in the Cs $6p^2P_{3/2}$ level by a linearly polarized pump laser is $-4/5$, corresponding to a linear polarization degree of 60% according to Eq. (3). With $I$ coupled to $J$, the value of $g^{(2)}=0.219$ [16] would produce a linear polarization degree of 15.57%. We measure $P_L = 15.60(3)\%$ which shows an excellent agreement with Ref. [16].

We made an additional check of the polarization spectra and experimental technique by probing the 9$^2S_{1/2}\rightarrow 10^2S_{1/2}$ transition in Rb. With $I$ coupled to $J$, the value of $g^{(2)}=0.219$ [16] would produce a linear polarization degree of 15.57%. We measure $P_L = 15.60(3)\%$ which shows an excellent agreement with Ref. [16].

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B. Rate equation analysis of Zeeman coherences and the depolarization cross section

The variety of possible distributions of atoms in the Zeeman sublevels depends on the given experimental conditions such as optical pumping with a circularly or linearly polarized light source. In our studies, the selective excitation to the Zeeman sublevels of the $6p^2P_{3/2}$ level was done using a linearly polarized pulsed laser. The alignment of the excited electronic level was defined by this laser which selectively populates the $m_J=\pm 1/2$ Zeeman sublevels according to the selection rule $\Delta n=0$. In the presence of Ar atoms, collisions between the excited Cs atoms in the $m_J=\pm 1/2$ Zeeman sub-
levels and the ground level argon atoms mix the population in the Zeeman sublevels, and thus transfer the population between different magnetic sublevels. The population \(N_{±1/2} \) mixing among the \(m=±1/2, ±3/2\) magnetic sublevels is responsible for the reduction of the alignment produced in the excited level. This process is illustrated in Fig. 5. Note that all collisional rates can go both ways with equal values. The population variations among the Zeeman sublevels of the 6S-10S transition can be expressed by a simple theoretical model using the rate equations of Zeeman coherences \[12,20\]. These are written as follows:

\[
\dot{N}_{1/2} = - (\gamma + \Gamma_1 + \Gamma_2)N_{1/2} + (\Gamma_1 + \Gamma_2)N_{3/2} + \Gamma_p,
\]

\[
\dot{N}_{3/2} = - (\gamma + \Gamma_1 + \Gamma_2)N_{3/2} + (\Gamma_1 + \Gamma_2)N_{1/2},
\]

where \(\Gamma_p\) is the pump pulse rate. Here we have used \(N_{1/2} = N_{-1/2}\) and \(N_{3/2} = N_{-3/2}\) applicable to an electronically aligned level. The laser intensity profile was modeled using a square pulse shape. Other pulse shapes may be used and lead to the same polarization results \[7,21,22\]. The time dependent total population density and the alignment in the excited level can be written as follows:

\[
N(t) = 2\Gamma_pP_b,
\]

\[
\langle A_0(t) \rangle = - \frac{4}{5} \Gamma_pP_a
\]

where \(P_b = \frac{1}{\gamma}(1-e^{-\gamma t})\) and \(P_a = \frac{1}{2\gamma}(1-e^{-2\gamma t})\). Here, \(\gamma = \gamma + (2\Gamma_1 + 2\Gamma_2)\) is the rate at which the alignment is altered due to the collision between the cesium and argon atoms, \(\gamma\) is the radiative decay rate, and \((2\Gamma_1 + 2\Gamma_2)\) is the total collisional decay rate which is defined to be \(\Gamma\). Note that \(\Gamma_p\) in Eqs. (4) and (5) cancels out once the intensity ratio is taken, thus this term was not carried. The population densities in the Zeeman \(m=±1/2, ±3/2\) sublevels in terms of total population and alignment are

\[
N_{1/2}(t) = \frac{N(t)}{4} - \frac{10}{32}\langle A_0(t) \rangle s^{(2)},
\]

\[
N_{3/2}(t) = \frac{N(t)}{4} + \frac{10}{32}\langle A_0(t) \rangle s^{(2)}.
\]

These population densities can be rewritten in terms of \(P_a\) and \(P_b\) as follows:

\[
N_{1/2}(t) = (P_b + P_a)/2,
\]

\[
N_{3/2}(t) = (P_b - P_a)/2.
\]

Thus, the measured signals, integrated over the temporal overlap time of the pulses, can be written in terms of \(P_a\) and \(P_b\) as follows:

\[
S_1 = \frac{1}{6}\int_0^T P_b dt + \frac{1}{6}\int_0^T P_a dt,
\]

\[
S_\perp = \frac{1}{6}\int_0^T P_b dt - \frac{1}{12}\int_0^T P_a dt.
\]

The constants in front of the integrals are the Clebsch-Gordan coefficients and are obtained by using the Wigner-Eckart theorem \[14\]. Finally, after substituting Eqs. (8a) and (8b) into Eq. (1), the linear polarization degree can be expressed in terms of the depolarization cross section and the pressure of the buffer gas as

\[
P_L = \frac{S - S_\perp}{S + S_\perp} = \frac{3Z}{4 + Z},
\]

where

\[
Z = g^{(2)}\gamma\frac{1 - \frac{1}{\gamma}(1 - e^{-\gamma T})}{\gamma_a\left(1 - \frac{1}{\gamma_a}(1 - e^{-\gamma_a T})\right)}.
\]

In Eq. (10), \(\gamma_a\) is defined to be \(\gamma + \Gamma\), and

\[
\Gamma = \rho_{A_d}k_d = \rho_{A_d}\sigma_d\tilde{v}_{CSAr} = \frac{P}{kT}k_d,
\]

where \(k_d\) is the disalignment rate coefficient, \(P\) is the buffer gas pressure, \(kT\) is the thermal energy constant, and \(\sigma_d\) is the alignment-dependent depolarization cross section. Here, we assumed that \(k_d = \langle \sigma_d\tilde{v} \rangle\), and \(\langle \sigma_d\tilde{v} \rangle\) may be factored so that \(k_d = \sigma_d\langle \tilde{v} \rangle\). In Eq. (11), we denoted \(\langle \tilde{v} \rangle\) as \(\tilde{v}_{CSAr}\) which is the average velocities of the colliding Cs-Ar atoms over the Maxwell-Boltzmann distribution of relative velocities at the

\[
N_{3/2}(t) = (P_b - P_a)/2.
\]

Thus, the measured signals, integrated over the temporal overlap time of the pulses, can be written in terms of \(P_a\) and \(P_b\) as follows:

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We have experimentally studied the double-resonance two-photon linear polarization spectrum of the $6s^2P_{3/2} \rightarrow 6p^2P_{3/2} \rightarrow 10s^2P_{1/2}$ and $6s^2P_{1/2} \rightarrow 6p^2P_{3/2} \rightarrow 5d^2D_{5/2}$ transitions. The measurements of the hyperfine depolarization coefficient for these transitions are in excellent agreement with Ref. [16]. The measured linear polarization degrees were found to be independent of the power of the lasers, temperature, and variation in the intensity of the fluorescence signal. The effect of excited cesium atoms in subsequent collisions with ground level argon atoms is evident on the polarization spectrum of the $6s^2P_{1/2} \rightarrow 10s^2P_{1/2}$ transition. From the measurements, we obtained the alignment-dependent depolarization cross section in the cesium $6p^2P_{3/2}$ level. Our results are in good agreement with the theory [5] and Guiry et al. [4] within the error limits.

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[8] M. D. Havey (private communication).