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High pressure line shapes of the Rb D_1 and D_2 lines for ^{4}He and ^{3}He collisions

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ABSTRACT

Line shapes for the Rb D_1 ($5\,^2S_{1/2} \leftrightarrow 5\,^2P_{1/2}$) and D_2 ($5\,^2S_{1/2} \leftrightarrow 5\,^2P_{3/2}$) transitions with ^{4}He and ^{3}He collisions at pressures of 500–15,000 Torr and temperatures of 333–533 K have been experimentally observed and compared to predictions from the Anderson–Talman theory. The ground $X\,^2\Sigma^+_{1/2}$ and excited $A\,^2\Pi_{1/2}$, $A\,^2\Pi_{3/2}$, and $B\,^2\Sigma^+_{1/2}$ potential energy surfaces required for the line shape predictions have been calculated using a one-electron pseudo-potential technique. The observed collision induced shift rates for ^{4}He are dramatically higher for the D_1 line, 4.60 ± 0.12 MHz/Torr, than the D_2 line, 0.20 ± 0.14 MHz/Torr. The asymmetry is somewhat larger for the D_1 line and has the same sign as the shifting rate. The ^{3}He broadening rate for the D_2 line is 4% larger than the ^{4}He rate, and 14% higher for the D_1 line, reflecting the higher relative speed. The calculated broadening rates are systematically larger than the observed rates by 1.1–3.2 MHz/Torr and agree within 14%. The primary focus of the current work is to characterize the high pressure line shapes, focusing on the non-Lorentzian features far from line center. In the far wing, the cross-section decreases by more than 4 orders of magnitude, with a broad, secondary maximum in the D_2 line near 735 nm. The potentials do not require empirical modification to provide excellent quantitative agreement with the observations. The dipole moment variation and absorption Boltzmann factor is critical to obtaining strong agreement in the wings.

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1. Introduction

The diode-pumped alkali laser (DPAL) was proposed in 2001 as an alternative to high-power, diode-pumped, solid-state lasers [28,30]. The radiation from the unphased diode laser bars or stacks is absorbed on the $D_2\,^2S_{1/2} \rightarrow {}^2P_{3/2}$ transition and collisional energy transfer to the spin–orbit split $^2P_{1/2}$ state yields lasing on the $D_1\,^2P_{1/2} \rightarrow {}^2S_{1/2}$ transition in potassium, rubidium, or cesium vapor.

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A rubidium laser pumped by a 1.28 kW diode stack with a 0.35-nm bandwidth has achieved 145-W average power [52]. More recently, 1 kW Cs laser with closed loop transverse flow was demonstrated with 48% optical-to-optical efficiency [11]. The fine structure splitting in Cs is large, and hydrocarbon collision partners are generally required to prevent bottlenecking. The presence of hydrocarbons can lead to soot and alkali hydride formation [29]. In contrast, helium is sufficient to induce fine structure mixing in Rb with the rates required to support high power development [36]. Helium pressures of 10–20 atm are required to avoid bottlenecking on the fine structure mixing and to broaden the absorption line shapes sufficiently to accept modestly narrowed diode bar radiation. Characterizing the high pressure Rb–He line shapes is critical to: (1) design the pump diode spectral band, (2) design the optical resonator, (3) assess the effects of atmospheric transmission on high power propagation, and (4) evaluate the rates of ionization via far wing absorption. In this section we observe and compare with theory the high-pressure line shapes for the Rb D_1 and D_2 lines induced by collisions with ^{4}He and ^{3}He.

The Rb–He gas line shapes near resonance (in the core) have been investigated experimentally in considerable detail [45,44,51,42,34,40,23,47,6,25,49,19,27]. The broadening and shifting rates for the Rb D_1 line induced by collisions with ^{4}He using modern methods agree to be within better than 4% [45,44,51,42,34]. The agreement for the D_2 line is poorer, with a 9% variance for the broadening rate and 20% for the shift rate. The shift rate for the D_2 line is small due to the combined effects of two electronic surfaces. The temperature range where these rates have been determined is modest, 314–394 K, and span several different studies. Older measurements during the period 1940–1980 exhibit a 30% variance in broadening rates and disagree on the sign of the shift [40,23,47,6,25,49,19]. The corresponding rates for collisions with ^{3}He were all performed at high pressures, >1 atm, and vary by about 20% [44,27,32].

Several computational approaches have also been applied to compute the broadening and shift rates [26,46,9,33,8]. However, the results are sensitive to the long range portion of the interaction potentials. Indeed, the two ab inito potentials [35,10] used in our recent study of Cs line shapes [20] both require empirical modification to adequately describe the observed spectra. Furthermore, different line shape theories applied to the same interaction potentials do not agree on the sign of the shift for the Rb–He interaction [9].

In the current work we focus on the high pressure, non-Lorentzian behavior of the Rb–He D_1 and D_2 line shapes. For pressures exceeding 1000 Torr, a significant asymmetry has been observed in the core of the line, [44,42,34] as predicted by several theoretical calculations [12,13,17,24]. However, the magnitude of the asymmetry is not well predicted and further refinement of the interaction potential appears necessary [26]. A blue satellite is observed in the far wing of the Rb D_2 line and is most pronounced for the heavier rare gases [12,13,17,24,2]. A comparison of the theoretical predictions for the far wing spectra of Rb–He over a broad range of temperatures has recently been published [12]. In the recent work, potential surfaces were generated using SA-CASSCF-MRCI calculations. In this paper we report observations of the absorption spectrum in far wings of the Rb D_2 and D_1 lines perturbed by ^{4}He and ^{3}He at pressures as high as 15,000 Torr. We then employ the Anderson–Talman theory, [3,4] including the effects of dipole moment variation, [1] to predict the line shapes. The sensitivity of dipole moment variation on the wing line shapes is also evaluated. Our longer-term goal is to unify the Rb–He DPAL kinetics with potential surfaces that are sufficient to predict the temperature dependence of the fine structure mixing rates and collisional line shape parameters.

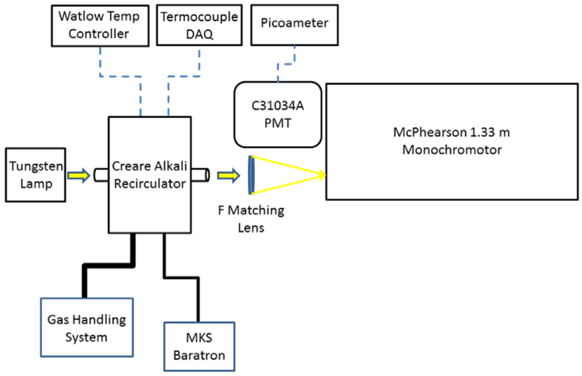

Fig. 1. Experimental setup for the high-pressure line shape study.

2. Experiment

Absorption spectra for rubidium vapor in the spectral range 600–875 nm were observed using a grating monochromator, as shown in Fig. 1. The broadband visible emission from an Ealing 100 W tungsten lamp, with Oriel 68831 300 W lamp power supply, was collimated with an $f=2.5$ cm lens to pass through a Rb sample maintained in a gas recirculation cell. The transmitted light was focused with another $f=2.5$ cm, 5 cm diameter lens onto the entrance slit of a McPherson model 209 $f=1.33$ m ($f/\#=9.4$) monochromator. With a 500 nm blaze, 1200 g/mm grating and slits widths of 20.8 µm for the entrance and 34.7 µm for the exit, the instrumental line shape exhibited a full width at half maximum spectral resolution of 0.05 nm (24.7 GHz). An Ultraviolet Products krypton pen lamp was positioned at 17 mm in front of the monochromator entrance slit to provide dynamic wavelength calibration. Wavelength calibration was stable to within 0.035 nm from over 6 months of data acquisition. Wavelength calibrations were performed dynamically, with the lamp and absorption spectra acquired simultaneously. During a single run, the accuracy of the calibration was limited to about 10% of the instrumental line shape. At 15,000 Torr, this corresponds to an uncertainty is shift and broadening rates of about 0.17 MHz/Torr.

An uncooled Burle C31034A photomultiplier tube biased at 1275 V exhibited a dark signal bias of 4–6 nA with a noise fluctuation of 0.04 nA, as monitored on a