Absolute absorption on the potassium D lines: theory and experiment

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Abstract

We present a detailed study of the absolute Doppler-broadened absorption of a probe beam scanned across the potassium D lines in a thermal vapour. Spectra using a weak probe were measured on the 4S $\rightarrow$ 4P transition and compared to the theoretical model of the electric susceptibility detailed by Zentile et al (2015 Comput. Phys. Commun. 189 162–74) in the code named ElecSus. Comparisons were also made on the 4S $\rightarrow$ 5P transition with an adapted version of ElecSus. This is the first experimental test of ElecSus on an atom with a ground state hyperfine splitting smaller than that of the Doppler width. An excellent agreement was found between ElecSus and experimental measurements at a variety of temperatures with rms errors $\sim 10^{-3}$. We have also demonstrated the use of ElecSus as an atomic vapour thermometry tool, and present a possible new measurement technique of transition decay rates which we predict to have a precision of $\sim 3$ kHz.

Keywords: potassium vapour, ElecSus, D lines

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

1. Introduction

The study of thermal vapours is a rapidly expanding field of physics with many applications. These range from all optical delay lines [1], quantum atomic memories [2–5], clocks [6, 7] and compact magnetometry [8–10] to name but a few. The knowledge of the absorptive and dispersive properties of an atomic vapour has many benefits for existing and potential applications. This has been shown to facilitate the design of optical components, such as optical isolators (OIs) [11] and atomic Faraday filters [12–14], without the need for exhaustive trial and error. Furthermore, spectroscopy of alkali-metal elements, such as rubidium and caesium, is used for many laser-locking schemes [15–17]. Knowledge of the response of a thermal vapour to a weak resonant beam can aid the effective design of such systems.

The absolute Doppler-broadened absorption of a probe beam resonant on the D lines of rubidium has been extensively studied by Siddons et al [18]. They presented a theoretical framework which was utilized to correctly predict the absorption profile of a weak probe beam resonant on the D lines. Adapted from this theoretical framework, Zentile et al developed ‘ElecSus’ [19]. ElecSus is a program which calculates the electric susceptibility of a thermal vapour in the vicinity of the alkali-metal D lines which in turn allows one to calculate many optical measurables, such as the transmission of a weak probe beam, the dispersive properties of the medium [20] and the magnetic shifts of spectra [21]. The absolute Doppler-broadened absorption on the potassium D lines is yet to be studied extensively. The D lines of potassium are currently extensively used for gray molasses cooling [22–25] to create ultracold ensembles of atoms, which are used to simulate many-body problems in physics [26, 27]. It has also been shown that using the 4S $\rightarrow$ 5P D line transitions allows for greater cooling of the potassium atoms due to the reduced linewidth [28, 29]. Potassium is of particular interest due to the existence of both bosonic and fermionic species [30–33] which allows one to study interactions in Bose–Fermi mixtures [34].

Spectroscopy of potassium is different from that of rubidium due to the small spacing of its hyperfine energy levels. In particular, the ground state hyperfine splitting in potassium is less than the Doppler width and hence only one absorption profile is seen on each of the D lines [35, 36]. The
aim of this work is to establish the weak-probe regime for the 4S $\rightarrow$ 4P and 4S $\rightarrow$ 5P D lines in potassium in order to test the quality of ElecSus on an atom where the ground state hyperfine splitting is less than the Doppler width.

The paper is structured as follows. In section 2, we give a brief description of the weak-probe regime and the physics behind ElecSus. Section 3 provides details of the experimental apparatus and procedure. Section 4 details the atomic structure of the potassium D line transitions along with our experimental results. Section 5 outlines a possible new experiment technique to measure transition decay rates. We conclude our findings in section 6.

2. Theory

2.1. Weak-probe regime

The weak-probe regime of a resonant probe beam is defined as the incident intensity $I_0$ at which the probe beam intensity is sufficiently weak that the absorption coefficient is independent of incident intensity. This regime in rubidium is typically several orders of magnitude smaller than the saturation intensity $I_{sat}$ of the transition [18]. The saturation intensity of a two-level system is defined by

$$I_{sat} = \frac{\pi \hbar c \Gamma}{3 \lambda^2},$$

(1)

where $\hbar$ is Planck’s constant, $c$ is the speed of light, $\Gamma$ is the linewidth and $\lambda$ is the wavelength of the transition [37].

Despite their simple atomic structure, alkali-metal atoms cannot be described by a simple two-level model. In particular, the presence of multiple hyperfine levels in the ground state can significantly alter the transmission of resonant probe light. Consider a system with two ground states $\left| g_1 \right>$ and $\left| g_2 \right>$ and one excited state $\left| e \right>$ where the transitions $\left| g_1 \right>$ $\rightarrow$ $\left| e \right>$ and $\left| g_2 \right>$ $\rightarrow$ $\left| e \right>$ are allowed and $\left| g_1 \right>$ $\rightarrow$ $\left| g_2 \right>$ is forbidden. If the probe beam is resonant with the $\left| g_1 \right>$ $\rightarrow$ $\left| e \right>$ transition, atoms are excited to $\left| e \right>$ but they may spontaneously decay from $\left| e \right>$ $\rightarrow$ $\left| g_1 \right>$ or $\left| e \right>$ $\rightarrow$ $\left| g_2 \right>$. As the light is off-resonance with the $\left| g_2 \right>$ $\rightarrow$ $\left| e \right>$ transition, the atoms which have decayed into $\left| g_2 \right>$ remain there. This leads to a depletion in the ground state population of $\left| g_1 \right>$ and hence an increase in transmission of the probe beam. Redistributing the ground state populations using light is known as optical pumping [38]. This system therefore has non-trivial dependencies on the determination of the weak-probe regime due to optical pumping.

2.2. ElecSus

The frequency dependent complex electric susceptibility of a dielectric $\chi(\Delta)$, where $\Delta$ is the detuning from resonance, connects the macroscopic polarization to the applied electric field [39]. The real part $\chi_R(\Delta)$ describes the dispersive properties and the imaginary part $\chi_I(\Delta)$ describes the absorptive properties.

In order to calculate $\chi(\Delta)$ in the vicinity of the D line transitions, ElecSus first calculates the transition strengths of allowed electric-dipole transitions between all hyperfine sublevels. To account for Doppler broadening due to the line-of-sight thermal motion of the atoms, a lineshape of the resonance is applied to each transition. This lineshape is a convolution of the Lorentzian atomic lineshape, dependent on $\Gamma$, and the Gaussian velocity distribution which is a function of temperature. Finally, $\chi(\Delta)$ is calculated by summing over all transitions. From $\chi(\Delta)$, many optical observables may be predicted.

As $\chi_R(\Delta)$ describes the absorptive properties of the medium, the absorption coefficient $\alpha$ is given by $\alpha(\Delta) = k\chi_R(\Delta)$ [18] where $k$ is the wavevector of the incident radiation. The absorption coefficient is dependent on the linewidth of the transition and the atomic number density $N$ in the vapour which is exponential in the temperature, $T$. For a particular hyperfine transition, the absorption coefficient is given by [18]

$$\alpha(\Delta) = \frac{k \gamma^2 \delta^2 N}{2(2\Gamma + 1)} \frac{1}{\hbar^{2}e^{2}} \frac{1}{\delta^4(\Delta)},$$

(2)

where $C_\delta$ is the total transition strength, $\Gamma$ is the nuclear spin, $\hbar$ is the reduced Planck’s constant, $\delta$ is the permittivity, $\delta^4(\Delta)$ is the transition lineshape and $d$ is given by

$$d = \sqrt{\frac{3c_0 \hbar \Gamma \lambda^2}{8\pi^2}}.$$

(3)

where $\lambda$ is the wavelength of the transition. Therefore, using the Beer–Lambert law, one is able to predict the transmission of a weak probe beam resonant on the alkali-metal D lines. For rubidium and caesium a vapour cell of a few cm length at room temperature has sufficient number density to observe significant absorption features. However, due to the higher melting point of potassium [40], the vapour cell must be heated to ensure a large enough atomic vapour number density such that absorption features are observed.

3. Experimental details

The experimental configuration is shown in figure 1. A home-built external cavity diode laser (ECDL) in the Littrow configuration [41] was the light source. An Eagleyard EYP-RWE-0790-04000-0750-SOT01-0000 laser diode was used for the 4S $\rightarrow$ 4P transition and a Sanyo DL5146–101S laser diode was used for the 4S $\rightarrow$ 5P transition. The output beam from the laser passed through an OI before impinging on a 70:30 beamsplitter (BS). The stronger beam was directed into a scanning Fabry–Perot etalon, which was used for frequency calibration. A Toptica FPI-100-0750-γ scanning Fabry–Perot etalon with a free spectral range of 1.00 ± 0.01 GHz and a Thorlabs SA200-3B scanning Fabry–Perot etalon with a free spectral range of 1.50 ± 0.01 GHz were used for the 4S $\rightarrow$ 4P and 4S $\rightarrow$ 5P transitions respectively. The weaker beam was sent through a variable neutral density filter (ND), to ensure control of the probe beam intensity, before passing through a resistively heated, 10 cm, natural abundance potassium vapour cell (K Cell). The magnetic field over the cell was measured to be of the order of $10^{-4}$ G. Note that no attempt
was made to null the field. The probe beam was then focused onto a calibrated photodiode (PD) which was integrated into a low-pass filter circuit in order to remove high-frequency noise and allow for greater sensitivity. The minimum detection power was ~1 nW.

The wavelength of light from the ECDL was tuned to the D lines by adjusting the angle of the diffraction grating. The wavelength was measured using a HighFinesse WS-5 wavelength meter to ensure the correct transitions were being probed. The laser was then scanned by applying a triangular ramp voltage at 10 Hz to a piezo stack actuator controlling the grating position. This allowed the laser to be scanned approximately 8 GHz before a mode hop. This was sufficiently large to scan the whole Doppler-broadened transition. Note that current feed-forward was employed on the blue 4S → 5P transition in order to increase the scan range of the laser.

The voltage output from the calibrated PD was recorded on a digital oscilloscope in order to record the transmission spectrum. The ramp voltage and etalon transmission peaks were used to convert from the measurement time into frequency, as well as linearizing the scan. The spacing between the etalon peaks was plotted as a function of observed time. A polynomial was then fitted to a plot of the difference between observed and expected transmission peak times and subsequently used to linearize the scan.

The weak-probe regime was established by varying the intensity of the probe beam incident on the cell from $1 \times 10^{-3} I_{sat} \rightarrow 1 \times 10^{2} I_{sat}$ by inserting a variety of ND filters into the path of the probe beam and measuring the minimum transmission. This was taken to be the mean over several points at the minimum of the transmission spectrum. Using a CCD camera, the $1/e^2$ widths of the probe beam were measured by taking cross sectional images of the probe beam before the cell and fitting Gaussian profiles to the images. The incident power was measured using the PD in the wings of the transmission spectrum. Spectra in the weak-probe regime were recorded at a variety of temperatures. Theoretical spectra generated by ElecSus were fit to the experimental spectra. A linear fit was made to the wings of the spectrum in order to normalize the data to account for a weak variation in the laser power during the laser scan.

4. Results and discussion

4.1. 4S → 4P D lines

In order to have measurable absorption of a probe beam with a good signal-to-noise ratio, the potassium vapour cell was heated to a typical temperature of ~40 °C, as at this temperature there is sufficient atomic vapour number density to absorb a significant proportion of the incident beam. At a temperature of 40 °C, the Doppler width is ~0.8 GHz. This is much larger than the ground state hyperfine splitting of the potassium isotopes shown in figure 2. Consequently, one would expect to observe only a single Doppler-broadened transmission spectrum for each of the D lines with contributions from both ground states. This can be seen in plots (i) and (iii) of figure 2. The saturation intensity and decay rates for the $D_1$ and $D_2$ transitions are 1.71 mW cm$^{-2}$, 1.75 mW cm$^{-2}$, $\Gamma/2\pi = 5.956$ MHz and $\Gamma/2\pi = 6.035$ MHz respectively [42].

In order to test ElecSus, the weak-probe regime was experimentally determined. In figure 3, the main figure shows the minimum transmission of a probe beam as a function of incident intensity, normalized to the saturation intensity, of the $D_2$ transition. The red data points show the minimum transmission of a probe beam, with $1/e^2$ widths of $w_x = 0.341 \pm 0.001$ mm and $w_y = 0.351 \pm 0.002$ mm, through the vapour cell at a temperature of $T = 45 \pm 1$ °C. The temperature was measured by inserting a thermocouple between the vapour cell and the surrounding metal heater assembly. The results demonstrate the significance of optical pumping. At low intensities, the probe beam is weak enough
Figure 2. An energy level diagram of the 4S → 4P lines of 39K and 41K along with their natural abundance. Note that we have neglected 40K due to its natural abundance of 0.0117%. Data taken from [30, 43–47]. Plots (i) and (iii) show the calculated contributions to the total transmission spectrum of a probe beam resonant with the D1 and D2 transition passing through a 10 cm natural abundance potassium vapour cell at 40 °C respectively. The blue (dotted) and green (solid) spectra correspond to transitions from \( F' = 1 \rightarrow F'' \) and \( F' = 2 \rightarrow F'' \) for 39K respectively. The red (dashed) and cyan (dot-dashed) spectra correspond to transitions from \( F = 1 \rightarrow F' \) and \( F = 2 \rightarrow F' \) for 41K respectively. The thick black profile is the total transmission spectrum. Plots (ii) and (iv) show the transition strengths, weighted by the isotopic abundance, where the red and green lines correspond to 39K and 41K respectively. The linear detuning (\( \Delta/2\pi \)) is referenced to the weighted line-centre of the transition.

Figure 3. The main figure shows a plot of the minimum transmission as a function of probe intensity, normalized to the saturation intensity, for a natural abundance potassium cell with light resonant on the D2 transition. The data corresponds to \( T = 45 \pm 1 \) °C and probe beam widths \( w_p = 0.341 \pm 0.001 \) mm and \( w_s = 0.351 \pm 0.002 \) mm. The red dashed line is the theoretical value predicted by ElecSus. The inset is a plot of the transmission profile as a function of detuning from line centre, along with a theoretical fit (black dashed line). The four colours (from the bottom to the top trace) represent different probe intensities where \( I_{tot} \), \( I_{blue} \), \( I_{orange} \) and \( I_{pink} \) correspond to incident intensities of \( (8 \times 10^{-3} \), \( 5 \times 10^{-1} \), \( 4 \) and 100 \) \( I_{sat} \) respectively. The residuals for the weak-probe regime are shown below.

such that optical pumping effects are negligible and hence a change in probe beam intensity does not change the transmission. However, even at intensities \( \sim I_{sat} \) the scattering rate is sufficient to lead to a noticeable modification of the transmission. It is striking that at high intensities the amount of absorption is negligible compared to that of the weak-probe regime. From the transmission data, we experimentally established the weak-probe regime to be at an incident intensity of \( \sim 10^{-1} I/I_{sat} \). The larger error bars at the low incident intensities are a consequence of low light levels and hence a poor signal-to-noise ratio.

The red data points in the inset of figure 3 show a transmission spectrum in the weak-probe regime. The theory (black dashed line) is fitted to the data along with the transmission spectra for probe intensities higher than that of the weak probe to demonstrate the effect of optical pumping. The four transmission spectra colours represent the different probe intensities where \( I_{red} \), \( I_{blue} \), \( I_{orange} \) and \( I_{pink} \) correspond to incident intensities of \( (8 \times 10^{-3} \), \( 5 \times 10^{-1} \), \( 4 \) and 100 \) \( I_{sat} \) respectively. The residuals for the theoretical fit to the red data show limited structure and hence the quality of the theoretical model.

A weak probe on the D1 and D2 transitions was passed through the cell at a variety of temperatures as shown in figure 4, in order to further test the predictions of ElecSus. Plot (i) shows transmission spectra on the D1 transition at temperatures of 34.0 \( \pm 0.7 \) °C, 43.8 \( \pm 0.4 \) °C and 56.0 \( \pm 0.2 \) °C, as shown by the blue, green and red data points, with rms errors between theory and experiment of \( 5 \times 10^{-3} \), \( 5 \times 10^{-3} \) and \( 8 \times 10^{-3} \) respectively. Here the temperature of the vapour was measured by fitting ElecSus to a weak-probe transmission spectrum, using a Marquard–Levenberg algorithm to minimize the weighted squares of the residuals [48]. The errors on the temperatures were determined by calculating the error bars from \( \chi^2 \) statistic from the fits [48]. The fitted temperatures compare favourably with
the thermocouple measurements, being consistently ~3 °C higher due to the fact that the thermocouple was poorly contacted to the cell. Plot (ii) shows transmission spectra on the D2 transition at temperatures of 26.8 ± 0.7 °C, 43.3 ± 0.5 °C and 51.7 ± 0.4 °C with rms errors between theory and experiment of 5 × 10⁻³ and 1 × 10⁻² respectively for (i) and 2.8 ± 0.7 °C, 40.4 ± 0.5 °C and 63.7 ± 0.4 °C with rms errors between theory and experiment of 5 × 10⁻³, 2 × 10⁻² and 1 × 10⁻² respectively for (ii). The residuals for each temperature are shown below.

Figure 4. The main figure shows a plot of the transmission as a function of detuning for three different temperatures for a natural abundance potassium cell with a probe intensity in the weak-probe regime with light resonant on the D1 (i) and D2 (ii) transitions. The black dashed lines are the theoretical spectra predicted by ElecSus. The blue, green and red spectra (from top to bottom trace) correspond to temperatures of 34.0 ± 0.7 °C, 43.8 ± 0.4 °C and 56.0 ± 0.2 °C with rms errors between theory and experiment of 5 × 10⁻³, 5 × 10⁻³ and 8 × 10⁻³ respectively for (i) and 26.8 ± 0.7 °C, 43.3 ± 0.5 °C and 51.7 ± 0.4 °C with rms errors between theory and experiment of 5 × 10⁻³, 2 × 10⁻² and 1 × 10⁻² respectively for (ii). The residuals for each temperature are shown below.

4.2. 4S → 5P D lines

The atomic structure of ³⁹K and ⁴¹K on the 4S → 5P D lines is shown in figure 5. One should note that to the best of our knowledge, the isotope shift for the D2 transition has never been experimentally measured. Hence we have adopted the notation of δ to label the unknown isotope shift. The 4S → 5P transition is similar in structure to that of the 4S → 4P as an increase in principal quantum number does not change the angular components of the system. However, we note a different isotope shift and a smaller hyperfine splitting.

In rubidium for example, sub-Doppler spectroscopy is used to create many frequency markers to linearize the frequency scan as a consequence of a small scan range producing a limited number of etalon peaks. This therefore shows that the fitting procedure is sensitive to the scan linearization.

However, sub-Doppler spectroscopy of potassium does not lead to clear frequency markers due to the small hyperfine spacing and hence this method was not employed. However, our method of linearization appears to be sufficiently accurate, as shown by the quality of the other fits.

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5P transition which could prove 39K and 41K along with their natural abundance. Data taken from [28, 30, 46, 47, 52–54]. Plots (i) and (iii) show the calculated contributions to the total transmission spectrum of a probe beam resonant with the D1 and D2 transition passing through a 10 cm natural abundance potassium vapour cell at 100 °C respectively. The colours correspond to those in figure 2. Plots (ii) and (iv) show the transition strengths, weighted by the isotopic abundance, where the red and green lines correspond to 39K and 41K respectively. Detuning is reference to the weighted line-centre of the transition.

Figure 5. An energy level diagram of the 4S − 5P D transition of 39K and 41K along with their natural abundance. Data taken from [28, 30, 46, 47, 52–54]. Plots (i) and (iii) show the calculated contributions to the total transmission spectrum of a probe beam resonant with the D1 and D2 transition passing through a 10 cm natural abundance potassium vapour cell at 100 °C respectively. The colours correspond to those in figure 2. Plots (ii) and (iv) show the transition strengths, weighted by the isotopic abundance, where the red and green lines correspond to 39K and 41K respectively. Detuning is reference to the weighted line-centre of the transition.
waveguide EOM driver by an RF synthesizer, avoiding the nonlinearities associated with scanning the ECDL.

5. Future experiment

The plots in figure 6 show that ElecSus can be adapted to accurately model the $4S \rightarrow 5P$ transition. This leads one to consider an experiment where two parallel beams resonant with the $4S \rightarrow 4P$ and $4S \rightarrow 5P$ transitions propagate through the same vapour cell. The $4S \rightarrow 4P$ beam may be used to accurately determine the temperature of the atoms to a precision of $0.2^\circ C$. The other beam may be used for spectroscopy of the atoms. For example, the line centre transmission is dependent on the strength of the transition which is directly proportional to $\Gamma$. Therefore, having characterized the atoms with one probe beam, we would be able to fit to experimental data of the transmission of the other probe beam in order to measure the decay rate of the transition. This two beam method could also prove useful in experiments in which the temperature of an atomic vapour must be known accurately, such as experiments attempting to measure the Boltzmann constant from the Doppler-width of an absorption feature [55, 56].

By varying $\Gamma$ for the $4S \rightarrow 5P$ transition in simulations and assuming the best signal-to-noise we experimentally obtained in our experiment, we believe one should be able to determine $\Gamma$ to a precision of $\sim 3 \text{kHz}$ at a temperature of $100^\circ C$ which is comparable to that given in the literature which is of the order of $\text{kHz}$ [47]. If the improvements to experimental noise were implemented, then it should be possible to surpass the experimental error on $\Gamma$ given in the literature.

6. Concluding remarks

Transmission spectra using a weak probe were measured on the $4S \rightarrow 4P$ and $4S \rightarrow 5P$ transitions. Theoretical spectra generated by ElecSus, and its adapted version, were fit to the experimental spectra. This is the first experimental test of ElecSus on an atom with a ground state hyperfine splitting smaller than that of the Doppler width. An excellent agreement was found between ElecSus and experimental measurements at a variety of temperatures on both transitions with rms errors $\sim 10^{-3}$ for all three data sets for (i) and temperatures of $83.1 \pm 0.4^\circ C, 103.0 \pm 0.2^\circ C$ and $117.0 \pm 0.7^\circ C$ with rms errors between theory and experiment of $2 \times 10^{-3}$, $4 \times 10^{-3}$ and $1 \times 10^{-2}$ respectively for (ii). The residuals for each temperature are shown below. The inset shows the rms error between the red data and ElecSus as a function of the D2 isotope shift, $\delta$.

Figure 6. The main figure shows a plot of the transmission as a function of detuning for three different temperatures for a natural abundance potassium vapour cell with a probe intensity in the weak-probe regime with light resonant on the $D_1$ (i) and $D_2$ (ii) transitions. The black dashed lines are the theoretical spectra predicted by ElecSus. The blue, green and red spectra (from top to bottom trace) correspond to temperatures of $101.0 \pm 0.8^\circ C, 110.7 \pm 0.4^\circ C$ and $125.4 \pm 0.3^\circ C$ with rms errors between theory and experiment of $5 \times 10^{-3}$ for all three data sets for (i) and temperatures of $83.1 \pm 0.4^\circ C, 103.0 \pm 0.2^\circ C$ and $117.0 \pm 0.7^\circ C$ with rms errors between theory and experiment of $2 \times 10^{-3}$, $4 \times 10^{-3}$ and $1 \times 10^{-2}$ respectively for (ii). The residuals for each temperature are shown below. The inset shows the rms error between the red data and ElecSus as a function of the D2 isotope shift, $\delta$. 
the possibility of an experiment where two probe beams could be used to simultaneously interrogate an atomic vapour on different transitions. Using one beam for thermometry enhances the accuracy of the information which can be determined with the second beam, allowing accurate decay rate measurements for example.

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