Optical pumping and electromagnetically induced transparency in a lithium vapour

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Abstract

We report the first study of electromagnetically induced transparency (EIT) on the D1 and D2 lines of 7Li in a vapour cell. The effect of different polarizations, background gas pressures and experimental designs on the dark resonance are examined. It is found that EIT is more prominent on the D1 line than on the D2 line and is present at the D2 transition under incident orthogonal linearly polarized fields and parallel circularly polarized fields, but absent for parallel linearly polarized fields and for orthogonal circularly polarized fields. It is shown that the contrast of the dark resonances can be affected by the presence of an inert background gas. An analysis of a further nine resonances observed at the D1 transition and three resonances at the D2 transition is presented.

Keywords: electromagnetically induced transparency, coherent population trapping, optical pumping, lambda systems, lithium

(Some figures in this article are in colour only in the electronic version)

1. Introduction

There has recently been increasing interest in the role that nonlinear optical phenomena play within atoms and molecules [1]. These can be clearly demonstrated when two light fields are incident on an atom or molecule with three internal levels where effects such as saturation and optical pumping of a particular velocity class by a strong ‘pump’ laser beam can be detected through the increased transmission or absorption of a weak ‘probe’ laser beam that interrogates the same velocity class of atoms. The presence of the two laser beams can also create quantum coherences within the atoms whereby at two-photon resonance an ‘uncoupled’ state exists such that the probability amplitudes for optical excitation from each of its components due to the two light fields exactly cancel each other. The population in this state is trapped, and so this phenomenon is called coherent population trapping, and where the transmission of the probe beam through the medium is observed to increase under these conditions, this phenomenon is called electromagnetically induced transparency (EIT). Applications of this process include lasing without inversion [2], the slowing down of light signals in dilute media by many orders of magnitude [3, 4], and the measurement of transition dipole moments (see [5] for such a measurement in the Li2 molecule).

The theory that describes saturation, optical pumping and coherent population trapping for three-level systems that can interact with a bichromatic light field for a long time is well understood. A typical three-level lambda system is shown in figure 1. We shall label the energy of the i-th bare state (i.e. the energy in the absence of the laser fields) by $E_i$ and take the energy separation between states $|i\rangle$ and $|j\rangle$ to be $E_i - E_j = \hbar \omega_{ij}$. The atoms are irradiated by two monochromatic laser fields (which are treated semiclassically) of amplitude $\varepsilon_1$ and $\varepsilon_2$ and angular frequency $\omega_1$ and $\omega_2$. We assume that the frequency of laser 1 is near resonant with the transition frequency $\omega_{12}$, that laser 2 is near resonant with $\omega_{23}$, and that the laser polarizations are such as to couple the lasers to their respective transitions. It is convenient to define the (small) detunings of the lasers from resonance as $\Delta_1 = \omega_1 - \omega_{12}$.
and $\Delta_2 = \omega_2 - \omega_{12}$. The two-photon resonance, or Raman resonance, condition then corresponds to $\Delta_1 = \Delta_2$.

In particular, this model is a useful one with which to describe the effect of a bichromatic light field on the D transitions of the alkali atoms. For these atoms the $^2S_{1/2}$ electronic ground state is split into two levels due to hyperfine interactions and a fine-structure component of the upper $^2P$ term is split into hyperfine components, one member of which plays the role of state $|2\rangle$ in this model. In reality, however, the actual levels within the alkali atoms are more complicated than figure 1 would indicate—each hyperfine level comprises several degenerate (in the absence of any external fields) $M_F$ states and, for the $^3P_{3/2}$ levels of both lithium and potassium, the small hyperfine splitting ($A = -3.06$ MHz for $^7$Li and $A = 6.06$ MHz for $^{39}$K, for example [6]) raises the interesting possibility of investigating the effect of very dense excited state hyperfine structure on the formation of trapping states at the D2 transitions. This structure can be expected to result in significant deviations from theoretical predictions based solely on an analysis of the simple system shown in figure 1. In this paper we report and compare the presence (and absence) of EIT at the D1 and D2 transitions of lithium atoms in a vapour cell for the first time, and the effect of different experimental conditions upon the spectral features.

Previous experimental studies on the effects of hyperfine structure on EIT have investigated the spectroscopy of cesium, rubidium and sodium in both $\Lambda$ and cascade configurations. Stähler et al [7] compared the visibility of the EIT peak at the D1 transitions in $^{85}$Rb ($A(\ ^2P_{3/2}) = 121$ MHz) and the D2 transitions ($A(\ ^2P_{1/2}) = 25$ MHz), and observed a significant increase in contrast, and decrease in resonance width, for the D1 system. Using a small magnetic field Nagel et al [8] have recorded the amplitude and FWHM of the coherence formed between the states $|F_s = 3, M_{Fs} = 0\rangle$ and $|F_s = 4, M_{Fs} = 0\rangle$ at the cesium D2 transition with circularly polarized light (i.e. with $|F_s = 3, M_{Fs} = 1\rangle$ or $|F_s = 4, M_{Fs} = 1\rangle$ playing the role of state $|2\rangle$ in this system). They observed that as the carrier frequency and its 9.2 GHz sideband were simultaneously scanned across the $^2P_{3/2}$ manifold and detuned away from either of the upper states of the $\Lambda$ system, the amplitude of the dark resonance decreased. This was consistent with transitions from one of the components of the uncoupled state to either the $|F_s = 2, M_{Fs} = 1\rangle$ or the $|F_s = 5, M_{Fs} = 1\rangle$ state—i.e. a ‘leaky’ $\Lambda$ system exhibiting one-photon losses. Renzoni et al [9] investigated the evolution of Zeeman coherences involved in different transitions within the sodium D1 line by using $\sigma^+ / \sigma^-$ incident polarization. The $|F_s = 1\rangle \rightarrow |F_s = 2\rangle$ transition displayed no dark resonance, which was attributed to the leaky nature of the single $\Lambda$ system present, and the dark resonances at the $|F_s = 1\rangle \rightarrow |F_s = 1\rangle$ and the $|F_s = 2\rangle \rightarrow |F_s = 2\rangle$ transitions demonstrated very similar amplitudes to each other. The dark resonance at the $|F_s = 2\rangle \rightarrow |F_s = 1\rangle$ transition had the greatest contrast; this was because this system was least ‘leaky’ (within five cycles of spontaneous decay from the $^2P_{1/2}$ state, only one of these would be to $|F_s = 1\rangle$, outside the $\Lambda$ system). Studies on the effect of hyperfine structure in EIT observed in cascade systems have included those of McGloin et al [10] and Badger et al [11]. In both these studies there was a significant dependence of the strength of the two-photon resonance peak upon the incident laser polarization at the 5s $\rightarrow$ 5p $\rightarrow$ 5d transitions in rubidium. In the former study, the ratios of the observed peak heights agreed with those calculated from transition dipole matrix calculations.

This work presents some results due to two incident laser beams on a vapour of lithium atoms at both the $^7$Li D1 and D2 transitions, in the first study on this species. Lithium is of general interest in atomic physics; in addition to being a system in which the effects of closely-spaced hyperfine states can be studied, it is the simplest of the alkalies, for which very accurate atomic structure calculations can be performed [12]. A theoretical analysis of the behaviour of dark resonances for atoms with upper-level structure in a high-pressure buffer gas has been developed by Tătărenachev et al [13], where the assumptions used correspond to buffer gas pressures significantly greater than used here.

This paper is presented as follows. In section 2 we describe the experiment in which various configurations of laser polarization and beam geometry were used, in section 3 we present the spectra obtained by detecting the transmission of either the probe laser, or the pump and the probe simultaneously, and in section 4 we present an analysis of the results and discuss their significance.

2. Experimental details

The lithium vapour cell consists of a stainless steel tube of 3/4 inch external diameter with glass windows at each end and a single core heating element (Thermoncoax) wrapped around the centre of the tube. The cable was wound in a bifilar arrangement with an equal number of windings in both directions to reduce any induced magnetic field. A thermocouple is held near the centre of the tube and the tube is rigidly held at each end by water-cooled aluminium plates. This ensures that lithium vapour does not reach and react with the glass windows, but necessarily means that the atomic vapour is not in thermodynamic equilibrium. The cell has two gas inlets/outlets; one is connected to a capsule pressure gauge (Edwards CG3) and the other to a pumping station consisting of a diffusion pump and a rotary backing pump. The cell can be pumped down to approximately $10^{-4}$ mbar and could be filled to a predetermined pressure with a buffer gas (argon, 99.998% purity, BOC).

Inside the cell lithium in its natural abundance (93% $^7$Li, 7% $^6$Li) is placed on a molybdenum boat. Although the melting point of lithium is 180 °C, in our experience there are no visible signs of melting at or above this temperature. We conclude that the sample is surrounded by a solid oxide or hydroxide layer which cracks upon heating and releases lithium vapour.
Atomic absorption profiles at the $^7\text{Li}$ D1 and D2 transitions (centred at 446 800 and 446 810 GHz, as measured on an Advantest Q8326 wavemeter) are visible when the temperature of the thermocouple reads between 400 and 500 °C. At these temperatures the atomic number density in the middle of the cell is expected to be $\approx 10^{12}$ cm$^{-3}$ [14]. Whilst the precise depth of each absorption profile is observed to be highly temperature dependent, the overall shape is approximately Gaussian with width 4 GHz. This is consistent with a ground state hyperfine splitting in $^7\text{Li}$ of 803.5 MHz [15] and a Doppler width of 3 GHz for each contributing transition. The D2 line of $^6\text{Li}$ (with ground state hyperfine splitting of 226 MHz [15]) occurs within the same spectral region as the D1 line of $^7\text{Li}$, and these transitions are expected to make a small contribution to the net absorption profile at 446 800 GHz.

The lasers used were single-mode external cavity diode lasers in the Littrow configuration [16] in which the zeroth-order beam from the grating constitutes the laser output. Limited frequency tuning of these lasers without mode hops ($\approx 1$ GHz) can be accomplished by changing the voltage across a piezoelectric stack within the diode mount, thereby changing the length of the external cavity. To ensure a single longitudinal mode remained in common with both the external and internal cavity as the laser is tuned over a broader range (up to 10 GHz), the current through the laser diode (Toshiba TOLD9225) was increased proportionally to the stack voltage using a simple amplifier circuit.

For these experiments two co-propagating laser beams were passed through the vapour cell in two different configurations: with the pump and probe laser beams at a small angle (3–4 mrad) to each other (the ‘crossed’ beam configuration) and also with the beams in a ‘collinear’ configuration (see figure 2). The diameter of each laser beam was 1–2 mm and the power in the pump beam was approximately 40 times greater than the probe beam (see below for precise values). In the crossed configuration the states of polarization of the pump and probe lasers could be set independently of each other and, due to the geometry of this configuration, the transmission of the probe beam could always be selectively recorded. In general, in this case there will be a small amount of mixing (less than 1%) of different polarization states in the resultant laser field in the interaction region associated with the addition of the non-collinear electric field vectors from each beam. However, to a very good level of approximation we use the same polarization labels as for the collinear case to describe the incident polarization in these experiments. For selective detection of the probe beam in the collinear configuration the polarizations of the pump and probe beams were oriented in a lin $\perp$ lin arrangement and a polarizing beamsplitter cube was used, as seen in figure 2. We also present spectra from both beams with parallel linear polarization, in which case the transmission of both beams was recorded.

For all but one of the spectra shown here, the pump laser was scanned across the frequency range of interest in a typical time of 0.5 s and the transmission of the probe beam was recorded. This had the advantage that the features observed in the spectra were on a flat background, instead of on a Doppler profile, and this enabled more accurate assessments of peak heights. For the case of the collinear beam configuration with parallel polarizations, the power of both beams was recorded, and the Doppler profile, of course, was then evident.

The linewidth of the diode lasers were initially determined by heterodyning them on a fast photodiode (EOT ET-2030A) and the beat signal observed with a spectrum analyser (Hameg...
5012-2) with 40 ms sweep time. The resulting spectra with spans of 50 MHz demonstrated a peak of $-3$ dB linewidth of less than 2 MHz, indicating that the linewidth of each laser was $\approx 1$ MHz, recorded in a 1 ms measurement time. A series of these spectra taken one minute apart resulted in a change in beat frequency of 4 MHz per minute, indicating that the frequency drift of each laser during a 0.5 s scan was 2–3 MHz.

The probe (unscanned) laser was placed near the centre of the Doppler-broadened absorption profile at the $^7$Li D1 line or D2 line before each scan. Part of the pump laser beam was sent to an interferometer with path length difference of approximately 3.3 m. This enabled the spectrum to be put on a linear frequency scale and provided an absolute calibration of the frequency difference was achieved by fitting a quadratic function to the interference fringes and setting the spacing between two assigned features in the spectrum to values obtained from the literature [6, 16, 17].

3. Results

3.1. Spectra at the $^7$Li D1 transition

Figure 3 shows the probe transmission spectrum in the collinear configuration with lin $\perp$ lin polarization, as the pump laser is scanned. The incident pump and probe powers are 4 mW and 110 $\mu$W respectively in a 1.5 mm diameter beam, and no buffer gas is present in the cell. For the allowed transitions within the D1 line, the pump intensity corresponds to a Rabi frequency of $173 \times 10^6$ rad s$^{-1}$ for the (strongest) $|F_e = 2, M_{F_e} = \pm 2\rangle \leftrightarrow |F_e = 1, M_{F_e} = \pm 1\rangle$ transitions with electric dipole moment [18] of $1.4 \times 10^{-29}$ C m, and $71 \times 10^6$ s$^{-1}$ for the weakest transitions with electric dipole moment a factor of $\sqrt{6}$ lower. The corresponding range of Rabi frequencies for the probe laser is from $28 \times 10^6$ to $11 \times 10^6$ rad s$^{-1}$. These can be compared with the width of each upper level due to its radiative lifetime, which is $37 \times 10^6$ rad s$^{-1}$.

The frequency axis has been calibrated by fitting a quadratic function to the interference fringes and setting the spacing between the peaks labelled $\Lambda P_1$ and $\Lambda P_2$ to 1607 MHz [6], which was consistent with the frequency difference determined from the coarse frequency calibration determined solely by counting interference fringes. The abscissa values in this figure are relative to the central peak; note that positive frequencies correspond to the pump being higher in frequency than the probe and vice versa. Below the spectrum are displayed the residuals from the quadratic fitting routine showing a slight systematic low-frequency sinusoidal difference between the fitted frequency and that corresponding to each fringe, which is probably attributable to the diode current and stack voltage being imperfectly scanned together. Nevertheless, the quadratic fit (with only two parameters and an arbitrary offset) is good to better than 5 MHz over the 1800 MHz scan range.

The spectrum shows 11 different resonances in $^7$Li that can be explained in terms of subsystems of its energy level structure. These results are similar to ones recently obtained by Wong et al [19] for resonances of the sodium D1 line and some of their notation will be adopted here. The central transmission peak labelled (T) is principally due to optical pumping and, to a lesser extent, saturation in a two-level subsystem when both lasers are resonant on the same transition. Due to Doppler shifting of the frequency of radiation seen by different velocity groups of atoms there are four possible subsystems involved, shown in figure 4. The two peaks on either side of the central peak labelled $V_1$ and $V_2$ result from optical pumping.

Figure 3. Probe transmission on the D1 line as a function of pump frequency in the collinear configuration and lin $\perp$ lin polarization. (a) Complete spectral range, (b) enlargement of low-frequency peaks, (c) enlargements of structure near zero frequency, (d) enlargements of high frequency structure.
and saturation in a three-level V subsystem. Here, the lasers are resonant with transitions from a common ground state to different excited states. These peaks differ in frequency from the central peak by the hyperfine splitting of the two excited states which is 91.8 MHz [6, 17]. For each of the peaks there are two possible subsystems involved due to Doppler shifting, one for each of the hyperfine ground states shown in figure 4.

The transmission peaks and dips labelled $\Delta P_1/\Delta D_1$ and $\Delta P_2/\Delta D_2$ are due to electromagnetically induced transparency and conventional optical pumping, respectively, in a three-level $\Lambda$ subsystem. For each peak and dip there are two possible contributing $\Lambda$-subsystems for the two different common excited states. The pump transfers population to the ground state from which the probe is exciting, resulting in increased absorption of the probe. However, at Raman resonance, where the frequency difference of the two fields equals the ground state hyperfine splitting of 803.5 MHz, the combined action of the pump and probe results in an eigenstate from which excitation to the upper state of the $\Lambda$ system is not possible (see section 4). The population of this uncoupled state can be further increased by spontaneous decay from the upper state, and under the conditions typically found in a vapour cell the height of this peak is indicative of both the initial population of this state and its increase through optical pumping.

The two outer pairs of shallow transmission dips, labelled $F_1$ to $F_2$ in figure 3, are due to optical pumping in a four-level subsystem as shown in figure 4. Note that in contrast to the other spectral features shown in the spectra we present, only one four-level subsystem contributes to each of these well-resolved dips, which makes them most amenable to quantitative analysis (see below). Each of these four features corresponds to the two laser beams being resonant with transitions from different ground states to different excited states. The dips are therefore separated from the EIT peaks by the splitting of the upper hyperfine states. The strong pump transfers population to the other ground state through spontaneous emission from the excited state resulting in increased absorption of the probe.

Finally, there are two small peaks to the left of $V_1$ and right of $V_2$, separated by 460 MHz in the spectrum. Within experimental uncertainty, this is twice the hyperfine splitting of the ground state of $^6\text{Li}$ (natural abundance 7%) and these are therefore due to transitions within the D2 line of this atom, which occurs within the same spectral region as the D1 transitions of $^7\text{Li}$.

Figure 5 shows two transmission spectra obtained using the crossed configuration with no buffer gas when the beams are vertically polarized, and when they are circularly polarized in the same sense. The incident pump and probe powers are 3.1 mW and 140 $\mu$W respectively in a 2 mm diameter beam. The top trace has been shifted vertically upwards by 1% for ease of comparison, but the traces are otherwise on the same scale. These spectra show the same resonances as figure 3 although the features are broader and less pronounced. The four resonances labelled $F_1$ to $F_4$ in figure 3 that are due to the single subsystems mentioned above have a characteristic intensity pattern for each of the two traces. We discuss this further in section 4.

Figure 6(a) shows spectra for parallel circularly polarized beams in the crossed configuration at different argon background gas pressures. Some of the traces are vertically scaled as depicted in the figure, and all have been shifted vertically for easy comparison. Therefore information about the absolute transmission for each trace is not provided, although the heights of each spectral feature may be reliably compared. The traces demonstrate a systematic change in both the optical pumping features and the EIT features as the pressure is monotonically varied. The capsule gauge used is accurate to $\approx 1$ mbar, and it can be concluded that the optimal pressure for observation of the EIT peaks lies between 0 and 1 mbar, whilst the optical pumping peaks are most visible at the lowest pressure reached. At a pressure of 1 mbar, and a temperature of 400 °C, the mean free path of gas atoms of diameter 1 Å is of the order of 2–3 mm, which is a typical distance of travel during which the atoms are irradiated by the laser beams. It may therefore be expected that collisions play a role here, and this is discussed in section 4.
of electromagnetically induced transparency peaks when the (F) dips of the D1 line and again we see the clear presence of the two dips at the centre peak is equivalent to the D1 (T) and (V) peaks, fieldscouple all the hyperfine excited states (complex) polarization vector of the medium, $P(\omega)$, to the oscillating (complex) electric field $E(t)$ through the relation $P(t) = i \hbar \chi(\omega) E(t)$, where $\chi(\omega)$ is the complex susceptibility. This relationship assumes that the (multilevel) atoms are in some sense at ‘steady state’ in the laser field, which is often a good approximation for atoms within a vapour cell when interrogated by a weak probe beam.

Figure 8 shows probe transmission spectra, in the crossed configuration for parallel circularly polarized beams, over a similar range of background gas pressure to those shown in figure 6. The top trace corresponds to the lowest pressure, and traces below to increased pressure. These spectra were taken without the capsule gauge in place and therefore the absolute pressures are not presented. Some of the traces are scaled for linearly polarized light. The EIT peaks are less distinct for linearly polarized light. The EIT peaks are more pronounced, whereas the optical pumping resonances diminish. Such spectra taken with fields polarized linearly in the same sense revealed no presence of EIT, even in the presence of argon background gas.

4. Analysis and discussion
4.1. Theoretical background to EIT
The coherent evolution of the three-level model system shown in figure 1 can be determined exactly (within the rotating wave approximation) either by solving the time-dependent Schrödinger equation for the state vector of the system when irradiated by two laser fields, or, equivalently, by solving the Liouville equation for the density operator [20]. The matrix elements of the density operator in a particular basis set then give the relevant populations (diagonal elements) and coherences (off-diagonal elements) of the system. In fact, the system shown in figure 1 does not include any decay, either out of the system (e.g. ionization, or fluorescence to other states) or within the system (e.g. spontaneous decay to states $|1\rangle$ and $|3\rangle$, or collisional decay between the states). The former process can be included within the state vector approach by adding a negative decay term to the equation of motion of the upper state, resulting in an imaginary term in the Hamiltonian matrix [21] and works because the subsystem remains in a pure quantum state [22]. In order to model decay within the system the density matrix approach is more appropriate as it permits the system to evolve from a pure state to a mixed state, through the phenomenological addition of decay terms to the equations of motion of the density matrix elements.

The absorption of a probe beam through a dielectric medium is then usually determined by relating the oscillating (complex) polarization vector of the medium, $P(t)$, to the oscillating (complex) electric field $E(t)$ through the relation $P(t) = \varepsilon_0 \chi(\omega) E(t)$, where $\chi(\omega)$ is the complex susceptibility. This relationship assumes that the (multilevel) atoms are in some sense at ‘steady state’ in the laser field, which is often a good approximation for atoms within a vapour cell when interrogated by a weak probe beam.

Figure 6. (a) Probe transmission in the crossed configuration for different argon gas pressures and parallel circular polarization at the D1 transition. Some of the traces are vertically scaled as shown in the figure, and all are shifted vertically for easy comparison (see text). (b) As in (a), but for parallel linear polarization.

For small amounts of background gas the EIT peaks emerge strongly and they remain prominent for increased argon pressure up to approximately 1 mbar. At buffer gas pressures above this they appear to decrease in contrast (there is very little transmission of any of the probe light at these pressures). Of particular interest is the reduced contrast of the dark resonance with very low buffer gas pressure, and its disappearance at pressures $>1.5$ mbar. This is discussed below. The optical pumping dips generally show opposite behaviour to that of the EIT feature, and disappear almost entirely for pressures of 1 mbar and above. Figure 6(b) shows similar spectra for linearly polarized light. The EIT peaks are less distinct than for circularly polarized light but all features show the same dependence as shown in figure 6(a) on background gas pressure.

3.2. Spectra at the $^7\text{Li} D2$ transition
Figure 7 shows the transmission spectra from both pump and probe beams in the collinear configuration where the beams have parallel polarization. As discussed above, the hyperfine splitting of the $^3P_{3/2}$ excited state is small and so the two laser fields couple all the hyperfine excited states ($F_x = 0, 1, 2, 3$) simultaneously. Therefore, we do not see the resonances previously labelled (F) and (V) resolved, as on the D1 line. The centre peak is equivalent to the D1 (T) and (V) peaks, the two dips at $\approx \pm 800$ MHz are equivalent to the AD and (F) dips of the D1 line and again we see the clear presence of electromagnetically induced transparency peaks when the fields differ in frequency by the ground state hyperfine splitting. However, the dark resonance disappears completely under parallel linear polarized fields which is not the case on the D1 line. Moreover, we have observed the EIT resonances to be present under orthogonal linear polarizations but absent under orthogonal circular polarizations. This behaviour will be discussed in section 4, below.

### Figure 8
Transmission spectra in the crossed configuration for parallel circularly polarized beams, over a similar range of background gas pressure to those shown in figure 6. The top trace corresponds to the lowest pressure, and traces below to increased pressure. These spectra were taken without the capsule gauge in place and therefore the absolute pressures are not presented. Some of the traces are scaled for linearly polarized light. The EIT peaks are less distinct for linearly polarized light. The EIT peaks are more pronounced, whereas the optical pumping resonances diminish. Such spectra taken with fields polarized linearly in the same sense revealed no presence of EIT, even in the presence of argon background gas.
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Figure 7. (a): Transmission of both pump and probe beams at the D2 line as a function of pump frequency in the collinear configuration, in parallel circularly polarized fields. (b) and (c): Enlargements of the low-frequency feature for parallel circularly and linearly polarized beams, respectively.

Figure 8. Probe transmission spectrum for different argon gas pressures under parallel circularly polarized fields. The top trace is with the cell evacuated (≈10⁻⁴ mbar) and the traces taken at increased pressure lie below this. The bottom three traces are scaled vertically by a factor of 5 and all have been shifted vertically for ease of comparison.

beam. In a dilute medium such as a gas, the absorption of this beam is then given by the imaginary part of the susceptibility, which is, in turn, proportional to the imaginary part of the coherence between the bare states of the transition coupled to the probe beam. If the probe laser corresponds to field $\varepsilon_3$, it is the reduction in the magnitude of $\text{Im}[\rho_{12}]$ that results in EIT, and this can be seen from several sources in the literature to occur when the Raman resonance condition is fulfilled; for recent reviews, see [23, 24].

In this experiment, the lasers are not locked to each other, and therefore the visibility of features that rely upon their mutual coherence can be expected to be diminished. If the incident electric fields in figure 1 are represented by cosines with phases of $\phi_1$ and $\phi_2$ respectively, it can be shown that the ‘dark’ state (i.e. the state uncoupled from the laser fields) takes the form [25]:

$$|\Phi_{NC}\rangle = e^{-iE_2t\hbar} \left[ \frac{\Omega_1}{\tilde{\Omega}} e^{i(\omega_1 t + \phi_1)} |1\rangle - \frac{\Omega_2}{\tilde{\Omega}} e^{i(\omega_2 t + \phi_2)} |3\rangle \right]$$

where $E_2$ is the energy of the bare state $|2\rangle$, and $\tilde{\Omega} = \sqrt{\Omega_1^2 + \Omega_2^2}$. The coupled state is orthogonal to this, and therefore if the relative phase between the two lasers changes sufficiently quickly, a dark state at one instant of time will rapidly oscillate between a dark and bright state. In our experiment it can be seen that the mutual coherence between the lasers over the timescale in which the EIT peaks are recorded is sufficient for the preservation of some atomic coherence. This may, however, be expected to contribute to the observed linewidth of the dark resonances.

One significant difference between the lithium D1 and D2 transitions is that the former couples ground state atoms to the $2\text{P}_1/2$ level with well-separated hyperfine states, whilst the latter involves the $2\text{P}_3/2$ level with different $|F_i\rangle$ states separated by 9 MHz or less, which is less than the Rabi frequency for the allowed one-photon transitions.

4.2. Resonances at the D1 transition

The width of the $\Delta P_1$ and $\Delta P_2$ peaks can be seen to be significantly less than that of the other features in the spectrum. In the collinear configuration the width of the EIT peaks should be determined principally by the coherence of the lasers, power broadening due to the pump beam, beam divergence and transit-time broadening. The residual Doppler broadening (associated with the differential Doppler shift due to the different wavelengths of pump and probe) is estimated to be 115.
approximate width, and is therefore negligible on this scale, as is the rate of decay of the ground state coherence. The EIT features in figure 3 have a full width at half maximum which is slightly less than 6 MHz, and is consistent with a spectral feature with sub-natural linewidth being broadened by small amounts of inhomogeneous and homogeneous broadening.

For the other resonances in figure 3 the energy spread of the excited states that participate in the optical pumping process also contributes to the width (states within the $^3P_m$ manifold have a radiative lifetime of 27 ns). The width of these other resonances ($\approx 20$ MHz) is consistent with the limiting width (displayed by the EIT peaks) broadened due to one-photon transitions involved in the optical pumping process.

The increased broadening of all resonances visible in figure 5 is principally due to the angle at which the beams cross. If two laser beams with similar laboratory frame frequencies cross at a small angle of $\theta$ radians, then the Doppler width due to a gas composed of atoms travelling in all directions at speed $v$ is given by

$$\Delta \nu = \frac{2\bar{\nu} \theta}{c},$$

(3)

where $\bar{\nu}$ is the mean frequency of the lasers in the laboratory frame. The average speed of lithium atoms at 700 K is about 1500 m s$^{-1}$, so for a crossing angle of 3–4 mrad the total broadening is $\approx 30$ MHz. The width of the central peak in figure 5 is indeed seen to be about 50 MHz, which is consistent with what we expect.

We attribute the decrease in contrast of the optical pumping features with increased buffer-gas pressure in figure 6 to velocity changing collisions between atoms of argon and lithium. Optical pumping relies on the laser fields interacting with the same velocity group of atoms for long enough for pumping to take place. Collisions with the background gas cause the lithium atoms to change their velocity so that they no longer see the laser fields at the same frequencies. Frequent collisions mean that the laser fields are constantly interacting with ‘fresh’ atoms which have not had time to be optically pumped.

There are a number of possible reasons for the sharpening of the EIT peaks when a small amount of background gas is released into the cell. One explanation is that frequent collisions between the argon and lithium atoms increase the time the lithium spends in the beam thus allowing more optical pumping into the non-coupled state. Pumping into the non-coupled state will take place as long as the condition of Raman resonance holds which, in contrast with single-photon resonance, is largely independent of the atoms’ velocity. It is well known that collisions causing decay between the ground states should have a negative effect on coherent population trapping but it is also well known that collisions with rare gases such as He, Ne and Ar which have no nuclear spin do not cause dephasing of the ground states [26]. Additionally, because the argon is free to move out of the hot centre of the vapour cell and cool down whereas the lithium vapour is confined to the centre, the argon will be colder than the lithium. Elastic collisions will therefore tend to decrease the average speed of the lithium atoms, increasing the time they spend in the beam. The above reasoning would suggest that the small $\Delta P_1$ and $\Delta P_2$ peaks observed with no buffer-gas present are mostly due to the initial projection of the ground states onto the non-coupled state and that optical pumping is needed to increase this population substantially. This may also contribute to the EIT being more pronounced for collinear beams as the interaction volume is much larger in the collinear beam setup.

Finally, we turn to an analysis of the relative heights of the features that are due to conventional optical pumping. These depend both on the transition dipole moments of the transitions involved and on the number of subsystems that contribute to the resonance. The feature height is indeed consistent with the number of subsystems with (T) being largest, then (V) and (AP/AD) and finally (F) being the smallest. The $F_1$–$F_4$ dips are the simplest to analyse quantitatively as each is only due to one four-level subsystem (encompassing in general 16 Zeeman sublevels). Under parallel linear polarization $F_1$ is larger than $F_2$ and $F_4$ is larger than $F_3$, whilst for parallel circularly polarized light $F_1$ is larger than $F_2$ and $F_3$ is larger than $F_4$.

An approximate model that can describe this behaviour is one that uses an Einstein rate equation approach [27] to describe the evolution of the atoms under the influence of the pump laser. The pump laser is assumed to excite the population within the Zeeman sublevels of a particular $|F_g\rangle$ state to sublevels of the relevant $|F_e\rangle$ state shown in figure 4 for the spectral features $F_{1-4}$. The forms of the coupled differential equations for the excited states, $|e\rangle$, and ground states, $|g\rangle$, are:

$$\frac{dN_e}{dt} = B_{ge}(N_e - N_g)\rho - AN_e$$

(4)

$$\frac{dN_g}{dt} = B_{ge}(N_e - N_g)\rho + \sum_i A_{iF}N_i,$$

(5)

where each ground state is coupled by the pump laser to the appropriate excited state by light of spectral energy density, $\rho$, and a particular excited state $|i\rangle$ radiatively decays with total rate $A_i$, but with partial decay rate of $A_{iF}$ to ground state $|g\rangle$.

The absorption of the probe laser at time $\tau$ after an atom enters the laser beams, $R_{abs}(t)$, is then assumed to be proportional to the sum of the population in each Zeeman sublevel times its transition dipole moment [18] to the excited state dictated by the polarization of the probe laser. We have calculated the relative values of $R_{abs}(t)$ for a pump beam spectral energy density of $5 \times 10^{-11}$ J m$^{-2}$, which is consistent with a pump beam power of 3 mW in a 2 mm diameter beam, and linewidth of 1 MHz. Figure 9 shows the calculated $R_{abs}(t)$ for each of the four ‘F’ resonances for the field polarizations shown in figure 6. It can be seen that at times corresponding to those that an average atom is in the laser field (1–2 $\mu$s), $F_1$ is larger than $F_2$ for both polarizations whereas the relative strengths of $F_3$ and $F_4$ reverse between polarizations. This is consistent with the behaviour of these resonances in figure 6, and indicates that a rate equation approach is able to qualitatively explain the effect of laser polarization on these subsystems. At spectral energy densities greater than that of saturation [27] the atomic dynamics is determined principally by the spontaneous decay rate of each transition, and so these plots are relatively insensitive to the precise value chosen for $\rho$.

4.3. EIT at the D2 transition

Electromagnetically induced transparency is observed to be less prominent on the D2 line than on the D1 line in $^7$Li. This
is in agreement with the results of Stühler et al [7] who find greater contrast of the dark resonance on the D1 line than the D2 line of rubidium. They attribute this to the closely spaced hyperfine excited states of the D2 line which are not part of the Λ system. Atoms can escape from the non-coupled state through one-photon excitation to states which are not part of the Λ system, which could be termed a 'leaky Λ system'.

As the excited states are closely spaced compared to the incident Rabi frequencies, at Raman resonance each laser field can excite from either the $|F_g = 1\rangle$ or $|F_g = 2\rangle$ sublevels to all upper-state levels permitted by the electric dipole selection rules. When the pump laser is 800 MHz red-detuned with respect to the probe, then the probe laser interacts with the $|F_g = 1\rangle \leftrightarrow |F_e = 0, 1, 2\rangle$ transitions and the pump laser mediates the $|F_g = 2\rangle \leftrightarrow |F_e = 1, 2, 3\rangle$ transitions. The opposite is true at the high-frequency Raman resonance in the spectra shown above. Figure 10 shows the allowed transitions between the Zeeman sublevels of the D2 line of $^7$Li for a variety of different incident polarizations.

For parallel linearly and parallel circularly polarized fields (figures 10(a) and (b)) there are four and five possible Λ systems, respectively (denoted by bold lines). The excited state population can spontaneously decay to several ground states with the possibility of being excited on a number of non-Λ transitions (denoted by light lines), which can reduce the population that is trapped. A close look at figure 10(a) shows the presence of a 'leaky' transition from $|F_g = 2\rangle$ but not from $|F_g = 1\rangle$. If the presence of leaky-Λ systems was the principal factor that determined the behaviour of the EIT resonances at the D2 transition, then when the frequency of the pump laser is scanned and the transmission of the probe laser is recorded, it would be expected that there would be a significant difference between the EIT features on either side of the spectrum, corresponding to whether the probe or the pump laser coupled with the ‘leak’ transition. This, however, we do not observe. Moreover, the presence or absence of a leaky-Λ system does not explain the lack of any EIT feature at the D2 transition under parallel linearly polarized fields, as the same situations exist for both these polarizations.

The most likely explanation for the presence of EIT at the D2 transition in $^7$Li under parallel circularly polarized fields, and its absence under parallel linearly polarized fields, can be inferred from the presence or absence of ‘double-Λ systems’ [28]. It is known that, in systems with closed interaction contours, unless the total interaction phase, Φ, is an integer number of π, then there is destruction of EIT [29].
This total phase is dependent upon the phase of the light fields and it can be seen from figure 10(a) that all lambda systems have a closed interaction contour, whilst there is a \( \Lambda \) system without a closed contour (albeit a 'leaky' one) in figure 10(b). As we are using lasers that are not phase-locked, it is highly unlikely that the required phase relation for incident parallel linearly polarized fields is met [30]. It should be noted that experiments using phase-locked lasers have demonstrated the presence of EIT in a system with closed interaction contours [31]. Figure 10(c) corresponds to orthogonal circularly polarized fields; here only closed contours are present, and there was no EIT observed. We observe EIT under orthogonal linearly polarized fields, and figures 10(d) and (e) denote the interaction systems that are present in this case (where one light beam is linearly polarized, and the other a superposition of left-hand and right-hand circular polarizations). When the linearly polarized light is at higher frequency (i.e. figure 10(d)) and when this is the lower frequency (i.e. figure 10(e)) there remains coherent coupling between several levels in the \( |F_g = 1\rangle\) and \( |F_g = 2\rangle\) levels with interaction contours that are not closed. These systems are more complicated extensions of the three-level \( \Lambda \) system shown in figure 1, and some of these have been previously studied (the four-level tripod system, for example, seen in figure 10(d), can exhibit EIT without the strict phase condition discussed above being satisfied [32]).

5. Conclusions

This experiment has provided the first evidence of electromagnetically induced transparency at the D1 and D2 transitions in \(^7\)Li atoms. The D1 line of lithium has revealed 11 resonances on a Doppler background. These are due to optical pumping and saturation in different four-level subsystems of the D1 line and electromagnetically induced transparency. It has been shown that the EIT peaks are strengthened by the introduction of argon buffer-gas into the lithium vapour cell. This may be due to argon atoms from the colder regions of the cell reducing the velocity of the lithium atoms in the interaction region, thus allowing more optical pumping into the non-coupled state to take place. Velocity changing collisions are, however, detrimental to the optical pumping which relies on one-photon resonance with individual transitions as this depends on the velocity of the atoms relative to the fields. This is not the case for optical pumping into the non-coupled state. The resonances in the collinear configuration are sharper than in the crossed configuration, although this latter case does allow separation of the probe and pump beams for all polarization conditions.

There are fewer resolved resonances present on the D2 line of lithium and the contrast of the dark resonance is much diminished, and only visible for parallel circularly polarized and orthogonally linearly polarized light. Consideration of the presence of leaky-\( \Lambda \) systems does not appear to provide an explanation for the observed behaviour. Instead the dependence of EIT on the polarization on the D2 line of \(^7\)Li can be explained through the presence, or absence, of systems with closed interaction contours. More theoretical and experimental work is required to fully understand the general problem of the absorption of bichromatic laser light by an optically dense, gaseous sample of atoms with many internal quantum states, but these results from lithium have provided some insight towards this goal.

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