Dual-frequency Doppler-free spectroscopy for compact atomic physics experiments

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ABSTRACT

Vapour-cell spectroscopy is widely used for the frequency stabilisation of diode lasers relative to specific atomic transitions — a technique essential in cold atom and ion trapping experiments. Two laser beams, tuned to different frequencies, can be overlapped on the same spatial path as an aid to compactness; this method also enhances the resulting spectroscopic signal via optical pumping effects, yielding an increase in the sensitivity of spectroscopically-generated laser stabilisation signals. Doppler-free locking features become visible over a frequency range several hundred MHz wider than for standard saturated absorption spectroscopy. Herein we present the measured Doppler-free spectroscopy signals from an atomic vapour cell as a function of both laser frequencies, showing experimental data that covers the full, 2D parameter space associated with dual-frequency spectroscopy. We consider how dual-frequency spectroscopy could be used for enhanced frequency-stabilisation of one laser, or alternatively to frequency-stabilise two lasers simultaneously, and analyse the likely performance of such stabilisation methods based on our experimental results. We discuss the underlying physical mechanism of the technique and show that a simple rate-equation model successfully predicts the key qualitative features of our results.

1 Introduction

Experiments employing cold, trapped atoms or ions underpin a wide range of fundamental research¹–³ and technological applications⁴–⁸. Such experiments require lasers that are accurately frequency-stabilised relative to specific atomic transitions, a function typically performed via feedback based on spectroscopy of a thermal atomic vapour. In addition many important direct applications rely on atom-light interactions in atomic vapour spectroscopy — for example the use of thermal atomic magnetometers in medical imaging⁹, ¹⁰ or the use of atomic spectroscopy to search for dark matter¹¹. Improvement of the signal strength and the frequency sensitivity of atomic vapour spectroscopic signals is therefore beneficial across a wide range of applications and research areas.

Frequency-stabilisation of diode lasers for cooling and trapping experiments is achieved via a feedback servo that controls the laser diode current and/or the alignment of a grating used for external optical feedback. The signal used to control this feedback is usually generated from spectroscopic measurements on an atomic vapour cell. The most common technique for this is based on saturated absorption spectroscopy¹², ¹³, combined with modulation of the laser current and phase-sensitive detection of the spectroscopic signal¹⁴.

It has been shown that the inclusion of two laser frequencies, overlapped on a single spatial path in orthogonal polarisation states, can increase the strength and sharpness of Doppler-free resonance features in saturated absorption spectroscopy¹⁵–¹⁸. This previous work on dual-frequency spectroscopy has considered a one-dimensional parameter space in which a single laser’s frequency is stabilised with high precision, usually to an ultra-narrow resonance feature produced via coherent atom-optical effects. The two frequency components previously considered have been produced via fixed-frequency modulation of light from a single laser, thus inherently restricting the dimensionality of the parameter space considered. Herein, we take a different approach and consider the full, two-dimensional parameter space obtained by using two independent laser sources whose frequencies can be freely varied over the relevant region. This serves a different experimental aim to precision stabilisation of a single, modulated laser and is likely to be particularly relevant where cost and compactness are important, as high-frequency optical modulation typically requires expensive, power-hungry hardware (such as electro-optic modulators) and can result in the need for additional beam-processing after modulation¹⁹, ²⁰. We find that, even in the absence of more complex effects that exploit atomic coherences, simple optical pumping effects create a lattice of Doppler-free resonance features in 2D frequency space; the size and spread of these features is considerably greater than that of those produced by conventional saturated absorption spectroscopy, potentially allowing lasers to be frequency-stabilised with improved accuracy and flexibility. We consider how the signals generated by this form of spectroscopy could be used to frequency-stabilise two independent lasers.
simultaneously and evaluate the likely performance of such stabilisation based on our experimental results.

Our results are of particular relevance to the burgeoning field of portable quantum technologies\textsuperscript{21,22}. Here, the reductions in size and weight offered by allowing multiple beams to share one spatial pathway can be important\textsuperscript{23}. Furthermore, miniaturised devices will increase the desirability of using small vapour cells with correspondingly reduced optical depths\textsuperscript{24,25}, thus making enhancement of the strength and sensitivity of the locking signal yet more important. The ambition to operate these systems outside the laboratory, with potential exposure to increased environmental noise, also adds to the desirability of increasing signal strength.

Below we give a brief outline of the physical basis of the approach and present our experimental results. We then discuss the implications of these results for two-laser stabilisation techniques in more detail and develop a theoretical model that explains many of the observed features.

2 Basic principle and experimental results

A diagram of our experimental setup is shown in figure 1(a). This differs from a standard saturated absorption spectroscopy apparatus\textsuperscript{13} in that two beams from different lasers are combined at a polarising beam splitter and co-propagate on the same spatial path through a 75 mm long vapour cell in orthogonal linear polarisation states, in an arrangement similar to that described in previous work on dual-frequency spectroscopy\textsuperscript{18}. Both beams have a diameter of $\sim 1.25 \text{ mm}$ with frequencies tuned close to resonance with the caesium D2 line: the ‘repumper’ laser such that it is resonant with transitions from the $F = 3$ hyperfine state of the lower manifold to the upper manifold, and the ‘cooler’ laser with transitions from the $F = 4$ state of the lower manifold, matching the standard naming convention in the field — see figure 1(b). Note that the hyperfine states in the upper manifold are spread over $\sim 600 \text{ MHz}$ of frequency space, while those in the lower manifold are separated by more than 9 GHz. For this reason, the separation of states in the lower manifold can be approximated as large compared to the Doppler shifts experienced by thermal vapour atoms, while the separation of the upper manifold states cannot. The beams are overlapped on a polarising beamsplitter and pass through the spectroscopy setup together. After interacting with the cell they are separated again with another polarising beamsplitter and their intensity is individually recorded on the two photodiodes shown in figure 1(a).

One mechanism by which this arrangement allows enhancement of the spectroscopic response is via optical pumping. It has previously been shown that optical pumping effects cannot be neglected in vapour cell spectroscopy\textsuperscript{26–28}; indeed optical pumping timescales (usually on the order of tens or hundreds of ns) are generally very much shorter than the typical time taken for thermal atoms to traverse the probing laser beam in such systems (usually on the order of tens of microseconds), and the lifetime of optically-induced atomic state disparities can be much longer still\textsuperscript{29}. When only one laser is used, significant attenuation of the spectroscopic signal therefore occurs via optical pumping of the vapour atoms into a state that is not accessible via transitions driven by the laser; this is exactly the same effect that necessitates the use of a ‘repumper’ laser beam for magneto-optical trapping\textsuperscript{30}. Simultaneous use of lasers operating on the cooler and repumper transitions prevents this effect and thus enhances signal strength. A more detailed analysis of this effect is presented in §5.

![Figure 1](image_url)

**Figure 1.** (a): Experimental setup used for dual-beam spectroscopy. For clarity, the spatially-overlapped laser beams are illustrated as side-by-side in the figure. (b): Level structure for $^{133}$Cs, indicating the role of the ‘cooler’ and ‘repumper’ lasers.

Our experimental results consist of the output signals from each of the photodiodes shown in figure 1(a), as a function of the frequencies of both the cooler and repumper lasers, and are shown in figure 2. These were obtained by synchronously scanning
both laser frequencies across the relevant frequency range. This was done by adding a linear ramp to the current supplied to each laser diode, and by simultaneously ramping the voltage supplied to piezoelectric transducers that control the alignment of diffraction gratings used for external cavity feedback. The result is laser frequencies described by the equations

\[ F_1 = A + Bt \quad \text{and} \quad F_2 = C + Dt, \]

where \( t \) is the time since the start of the ramp, \( F_1 \) and \( F_2 \) are the frequencies of the cooler and repumper lasers, respectively, and \( A-D \) are constant coefficients. The resulting equation describing \( F_2 \) as a function of \( F_1 \) is

\[ F_2 = \frac{D}{B} F_1 + \left( C - \frac{DA}{B} \right). \]

This equation clearly describes a diagonal line in the parameter-plane displayed in figure 2, with gradient \( D/B \) and offset \( C-(DA/B) \). By adjusting either of the static frequency offsets, \( A \) and \( C \), it is then possible to collect data along multiple such lines and build up a full, 2D dataset as displayed. An absolute frequency reference was provided in each case by simultaneously directing light from each of the lasers to a standard saturated absorption spectroscopy apparatus. The diameter of each beam was 1.25 mm and the powers used were 0.14 mW for the cooler laser and 0.23 mW for the repumper laser. One consequence of collecting the data in this way is that the boundaries of the region within which we collected experimental data are not aligned to the axes of the graph, hence the white triangles in the corners of the plots, which represent regions where data was not collected.

![Figure 2. Spectroscopic signals from the cooler laser photodiode (a) and repumper laser photodiode (b) as a function of the frequencies of both lasers. See figure 1 and main text for details. Frequencies for the cooler and repumper lasers are given relative to the F=4→F′=5 and F=3→F′=4 transitions respectively.](image)

In a separate experiment, 0.37 mW of cooler light, at a fixed frequency of -365 MHz (relative to the F=4→F′=5 transition), was directed into the same apparatus alongside 0.25 mW of repumper light, while the frequency of the repumper laser was scanned. The resulting absorption of the repumper light was determined by recording the output of “Photodiode Repumper” from figure 1(a) and is shown in figure 3(a). For comparison, the figure also shows the result when the light from the cooler laser was blocked, which then corresponds to a standard saturated absorption spectroscopy signal. It is evident from this figure that the use of dual-frequency spectroscopy substantially increases both the overall absorption of the light by the atomic vapour and the size and spread of Doppler-free resonance features. Figure 3(b) shows the corresponding “normalised error signal,” which we define as the fractional change in light transmission per unit frequency, given by \( T^{-1} (dT/dF_r) \), where \( T \) is the fraction of the repumper light that is transmitted through the atomic vapour and \( F_r \) is the frequency of the repumper laser. Analysis of the normalised error signal provides the most appropriate figure of merit for the efficacy of the technique when applied to laser stabilisation, as this describes the ratio of the response to changes in laser frequency to the overall optical signal. Once again, it is clear that the presence of the cooler light in the vapour increases the size and spread of spectroscopic features appropriate for use in laser frequency stabilisation.

3 Discussion

The results in figures 2 and 3 show that the presence of the additional cooler/repumper frequency light enhances the absorption signal of the repumper/cooler light by the atomic vapour, as expected. This already suggests that it may enable more accurate
Figure 3. (a): Spectroscopy signal from “Photodiode Repumper” (see figure 1(a)) while light from the cooler laser, tuned 365 MHz below the $F=4 \rightarrow F'=5$ transition, is also present in the cell (blue line). For reference, a standard saturated absorption spectroscopy signal, generated by blocking the beam from the cooler laser, is also shown (orange line). It can be seen that the addition of light tuned to the cooler transition substantially increases the size and spread of the Doppler-free features. (b): Normalised error signal (see text) corresponding to the spectroscopic signals shown in panel (a). The increased size and spread of features appropriate for laser frequency stabilisation is clearly visible. In both panels, repumper laser frequency is given relative to the $F=3 \rightarrow F'=4$ transition.

laser stabilisation by providing a feedback signal with improved signal to noise ratio (SNR). However, fig. 2 also demonstrates that sharp response features are created in a diagonal grid pattern over a frequency range of about 700 MHz — considerably broader than 450 MHz (for the cooler laser) or 350 MHz (for the repumper laser) over which conventional saturated absorption spectroscopy produces Doppler-free resonance features.

An intuitive explanation for the features in fig. 2 is as follows. Consider for example the absorption of the cooler beam. When the repumper light is on resonance with transitions in the same atoms within the thermal velocity distribution as the light from the cooler beam, it enhances the absorption of the cooler light. For co-propagating beams the simultaneous resonance condition requires that the two laser frequencies are separated by a fixed amount, thus creating a set of sharp features, similar to those seen in saturated absorption spectroscopy, that map out diagonal lines of positive slope within the frequency planes shown in figure 2. For counter-propagating beams, the same effect is present but the sign of the slope is reversed. Since both co-propagating and counter-propagating beams are present in the vapour cell, the dual-frequency optical pumping effects produce diagonal line features with both positive and negative slope, creating the observed grid patterns. A more detailed analysis is given in section 5.

4 Application to laser frequency stabilisation

We consider two ways in which dual-frequency spectroscopy is likely to be of use for laser frequency stabilisation: the first is that light from a second laser, already stabilised using independent equipment, could be injected into a spectroscopy apparatus for the purposes of enhancing the spectroscopic signal. The second involves two lasers being frequency-stabilised simultaneously using the same apparatus. Note that we do not consider the possibility of frequency-stabilising a single laser from which multiple frequency components are produced via electro-optic or laser current modulation, as this is relevant to a different range of applications and has been considered in detail in previous work.\(^{15–18}\)

An appropriate figure of merit to consider in each case is the factor by which the frequency deviations of a laser stabilised via dual-frequency spectroscopy could be reduced compared to those of a laser stabilised using conventional spectroscopic techniques, under the assumption that the feedback hardware works perfectly and stability is limited entirely by the quality of the spectroscopic signal used to control the feedback. We shall refer to this as the “signal limited stability enhancement” (SLSE).

In order to conduct a quantitative analysis of the benefits of dual-frequency spectroscopy for laser stabilisation, and to determine numerical values for the SLSE, we must consider a specific laser stabilisation scheme. We examine more closely the case of saturated absorption spectroscopy combined with laser current modulation and phase-sensitive detection of the corresponding spectroscopic signal.\(^{14}\) This is one of the most commonly used approaches to spectroscopic laser stabilisation and therefore seems a suitable choice, but it is worth noting that many other schemes for spectroscopic laser stabilisation exist — for example dichroic atomic vapour laser locking\(^{31,32}\) and polarisation spectroscopy\(^{33,34}\) — and that similar qualitative trends
4.1 Spectroscopy with injected light from an independently stabilised laser

In this case the SLSE is determined by two factors: the sensitivity of the spectroscopic feedback signal to changes in the frequency of the target laser about its ‘lock point’ (the frequency at which it is stabilised), and the scale of unwanted shifts in the lock point of the target laser that will result from changes in the frequency of the injected light. Since there is no reason for these two sources of instability to be correlated they can be added in quadrature when determining the likely magnitude of the laser’s frequency deviations from its desired value.

If we assume that the injected laser is stabilised using standard saturated absorption spectroscopy we can label the likely scale of its frequency deviations from its setpoint as $\delta F_0$. Locking based on saturated absorption spectroscopy and current modulation generates a response signal $S$ approximately proportional to the rate of change of the saturated absorption signal voltage with laser frequency:

$$S \approx C \frac{dV}{dF},$$

where $C$ is a system-specific proportionality constant, $F$ is the laser frequency and $V$ is the output voltage of the photodiode following the spectroscopy apparatus, normalised against its maximum value when the laser light is far off resonance and there is negligible absorption in the atomic vapour. The parameter of interest — the sensitivity of the response signal to changes in laser frequency — is therefore given by

$$\frac{dS}{dF} \approx C \frac{d^2V}{dF^2}. \quad (4)$$

To compare the efficacy of dual-laser spectroscopy to standard saturated absorption spectroscopy, we therefore compare the maximum values of $\frac{d^2V}{dF^2}$ measured with and without the second beam present in the vapour cell, defining the ratio between them as $R$. This ratio reaches values $>3$ for our experimental data. However, we must also take into account the effects of deviations in the injected light’s frequency, giving

$$\delta F \approx \sqrt{(\delta F_0 R)^2 + (\delta F_0 Q)^2}, \quad (5)$$

where $Q$ is defined by

$$Q = \frac{\frac{d^2V}{dF \, dF_{\text{injected}}}}{\frac{d^2V}{dF^2}}, \quad (6)$$

and we have assumed that the two different contributions to laser frequency variation — deviation from the lock point and lock-point mobility arising from variations of the injected laser frequency — are independent and can be added in quadrature. The derivatives of photodiode voltage with respect to $F$ in (6) are assumed to be evaluated at the chosen locking point and in the presence of the injected beam. We denote our figure of merit, the SLSE, by $E = \delta F_0/R$. Figure 4(a) shows a colour plot of the experimentally derived values of the SLSE for the repumper laser with cooler light injection, as a function of both laser frequencies. Note that the experimentally derived SLSE value requires numerical estimation of local gradient functions, and that the exact method by which these are calculated can have some influence on the result — see supplementary materials for more details. It can be seen that there are numerous regions in the plane where $E > 1$, with the maximum value of $E$ equal to 2.26. This demonstrates the clear advantages of the technique in terms of laser stability.

4.2 Simultaneous stabilisation of two lasers

For simultaneous stabilisation of two lasers composite feedback signals must be generated that depend (at least to first order about the desired lock point) only on the frequency of one laser. We consider two lasers with frequencies $F_1$ and $F_2$ generating corresponding photodiode outputs $V_1$ and $V_2$. We assume that the currents of the two lasers are modulated at different frequencies to avoid direct cross-talk.

Assuming once again that all derivatives are evaluated at the chosen locking point, the gradient of the spectroscopic signal after demodulation, $S_1$, is given by

$$\frac{dS_1}{dF_1} = C_1 \frac{d^2V_1}{dF_1^2}, \quad (7)$$
and
\[
\frac{dS_1}{dF_2} = C_1 \frac{d^2V_1}{dF_1^2} \frac{dF_2}{dV_1},
\]
with corresponding expressions for the gradient of $S_2$. We can therefore define a composite parameter $\chi_1$ with no first-order dependence on $F_2$:
\[
\chi_1 = S_1 - \nu_1 S_2,
\]
where
\[
\nu_1 = \frac{C_1}{C_2} \frac{d^2V_1}{dF_1^2} \left( \frac{d^2V_2}{dF_2^2} \right)^{-1}.
\]
Corresponding expressions obviously exist for $\chi_2$, following the same derivation. Thus it is possible to generate a feedback signal for each laser that is independent of the frequency of the other laser. This makes it possible to stabilise both lasers simultaneously based on the generated feedback signals.

We now determine the resulting laser frequency stability and compare it to that achieved with conventional saturated absorption spectroscopy. In this case the SLSE for laser 1 is given by
\[
E_1 = \frac{d\chi_1}{dF_1} \left| \frac{dS_{\text{con}}}{dF_{\text{con}}} \sqrt{1 + \nu_1^2} \right|,
\]
where $dS_{\text{con}}/dF_{\text{con}}$ is the gradient of the feedback signal about the lock point in an equivalent conventional saturated absorption spectroscopy apparatus. The factor of $\sqrt{1 + \nu_1^2}$ normalises against the amplification of the feedback signal that has been performed via post-processing, and consequently therefore amplifies the experimental noise as well as the desired signal and cannot produce an improvement in frequency stability. We thus find that
\[
E_1 = \left( \frac{dS_1}{dF_1} - \nu_1 \frac{dS_2}{dF_1} \right) \left| \frac{dS_{\text{con}}}{dF_{\text{con}}} \sqrt{1 + \nu_1^2} \right|,
\]
and the equivalent expression for $E_2$. For effective stabilisation of both lasers simultaneously, a conservative figure of merit is the minimum value of the SLSE for either laser (M-SLSE). This was calculated for a subset of the experimental data shown in figure 2 over which optical pumping effects are significant, i.e. where both lasers are close to resonance and dual-frequency spectroscopy differs significantly from conventional Doppler-free spectroscopy. The results are shown in figure 4(b), in which the repumper laser frequency is set 315 MHz below the F'=3 to F''=4 transition and the cooler laser frequency is varied. It can be seen that there are multiple locations in frequency space where the M-SLSE exceeds 1, with the maximum value of the M-SLSE that occurs in the displayed data being equal to 1.32. This shows that, even when stabilising two lasers using the same apparatus, and when considering only the laser for which the result is least favourable, dual-frequency spectroscopy still offers better performance than conventional Doppler-free spectroscopy.

4.2.1 Simultaneous stabilisation of two lasers with strongly asymmetric frequency tolerances
A surprisingly common experimental scenario is that one laser must be frequency-stabilised with high accuracy while another need not be stabilised so carefully. Consider for example the repumper and cooler lasers typically used for magneto-optical trapping of alkali metals: the efficacy of the trap is strongly sensitive to the frequency of the cooler laser but, provided that excess power is available, the frequency of the repumper laser can be varied quite widely with minimal effect on the performance of the trap. In such a scenario, performance considerably greater than that shown in figure 4(b) can be obtained, as a location in 2D frequency space can be chosen that optimises the stability of the target laser rather than optimising the ‘minimum stability’ across both lasers. Under such circumstances, our results already show values $>2$ for the SLSE of the target laser, with superior performance likely possible via fine-tuning of the experimental parameters.

In a related scenario, dual-frequency spectroscopy might prove useful where current modulation of a specific laser is undesirable, but it is also preferable to avoid the inclusion of the additional hardware typically required for frequency stabilisation schemes that do not employ current modulation — for example, the magnetic elements and polarisation optics required for dichroic atomic vapour laser locking. In such situations, dual-frequency spectroscopy has the potential to generate an appropriate stabilisation signal by using current modulation of the second laser only, leaving the first laser current unmodulated. This could be important if the first laser is to be used to stimulate or probe coherent quantum effects, for which laser current modulation typically proves deleterious. We do not examine this possibility in detail here, but it presents a promising avenue for future investigation.
Signal-limited stability enhancement (SLSE — see text for full definition) resulting from laser stabilisation based on dual-frequency spectroscopy, compared to the maximum achievable using conventional saturated absorption spectroscopy. (a) Shows the SLSE for the repumper laser resulting from the injection of independently stabilised cooler light, as derived by applying equation (5) to our experimental data. (b) Relates to simultaneous stabilisation of two lasers using the same apparatus; plotted is the minimum SLSE for either laser (M-SLSE), as a function of cooler laser frequency. The repumper laser frequency is set to -315 MHz and in each case the SLSE is determined by applying equation (12) to our data. It can be seen that multiple locations exist at which M-SLSE > 1, i.e. at which laser stabilisation based on dual-frequency spectroscopy can offer improved performance for both lasers simultaneously. Frequencies for the cooler and repumper lasers are given relative to the F=4 to F′=5 and F=3 to F′=4 transitions respectively.

5 Theoretical model

We develop a simple theoretical model that correctly predicts the key features of our results and elucidates the underlying physical mechanisms. It is not intended to accurately reproduce our experimental results on a quantitative basis; doing so would require a far more complex model that is beyond the scope of this work — see Supplementary Material for a discussion of the complexities involved in producing such a model.

We approximate the transient behaviour of the atoms as they traverse the beam in the following simplified way: we assume that some fraction of the atoms, $N_X$, have recently entered the laser beam and have not yet been optically pumped. These are assumed to be in an equal mixture of the two hyperfine states of the 6S$_{1/2}$ level. The remaining atoms that are interacting with the beam are assumed to be in a state of dynamic equilibrium. This is likely to be a reasonable approximation, as very few cycles of optical pumping are required to redistribute the atomic population.

In order to determine the influence of these ‘equilibrium state’ atoms on the spectroscopic signals, we adopt a similar approach to that presented in previous work on atom trapping using optical pumping effects$^{35}$, developing a rate-equation based model in which we consider the six-level system shown in figure 1(b). For convenience, we label the F=3 and F=4 hyperfine states of the 6S$_{1/2}$ level as ‘A’ and ‘B’ respectively, while the F′=2 through F′=4 hyperfine states of the 6P$_{3/2}$ level will be denoted by ‘C’ to ‘F’ respectively. For now we consider only a single atom with fixed laser detunings — the thermal distribution of atomic velocities and corresponding Doppler shifts will be factored in later. We define a set of rate coefficients, $\tau_{ij}$ and $\Gamma_{ij}$, such that the stimulated and spontaneous transition rates between, for example, states E and B are given by $\tau_{EB}$ and $\Gamma_{EB}$ respectively. The spontaneous decay rates for the relevant transitions are already known accurately — see for example$^{36}$. To determine the rate coefficients for stimulated transitions, we equate the steady-state results for the upper state population produced by our rate equation model to those produced by solving the full optical Bloch equations for a two level system. For a transition with spontaneous decay rate $\Gamma$, illumination of detuning $\delta$ and intensity $I$, with Rabi frequency $\Omega$, we obtain

$$\frac{\Omega^2}{4} \frac{\delta^2 + \Omega^2/2 + \Gamma^2/4}{\delta^2 + \Omega^2/2 + \Gamma^2/4} = \frac{\tau I}{2\tau I + \Gamma}.$$  

(13)
Therefore, labeling the dipole matrix element $\langle E | x | B \rangle$ between two levels as $X_{EB}$, we find that

$$
\tau_{EB} = \frac{\Omega_{EB}^2 \Gamma_{EB}}{4\epsilon_{0} (\delta_{EB}^2 + \Gamma_{EB}^2/2)} = \frac{e^2 |X_{EB}|^2 \Gamma_{EB}}{2\hbar^2 c \epsilon_{0} (\delta_{EB}^2 + \Gamma_{EB}^2/2)}
$$

(14)

and similar results for the other transitions. Hence, using each state’s label to denote the population fraction in that state, the rate equations governing the system are:

$$
\frac{dF}{dt} = (B - F) \tau_{FB} I_{FB} - FT_{FB},
$$

(15)

$$
\frac{dE}{dt} = (A - E) \tau_{EA} I_{EA} + (B - E) \tau_{EB} I_{EB} - E \Gamma_{EB} - E \Gamma_{EA},
$$

(16)

$$
\frac{dD}{dt} = (B - D) \tau_{DB} I_{DB} + (A - D) \tau_{DA} I_{DA} - D \Gamma_{DB} - D \Gamma_{DA},
$$

(17)

$$
\frac{dC}{dt} = (A - C) \tau_{CA} I_{CA} - C \Gamma_{CA},
$$

(18)

$$
\frac{dB}{dt} = (F - B) \tau_{FB} I_{FB} + (E - B) \tau_{EB} I_{EB} + (D - B) \tau_{DB} I_{DB} + FT_{FB} + E \Gamma_{EB} + D \Gamma_{DB},
$$

(19)

and

$$
\frac{dA}{dt} = (E - A) \tau_{EA} I_{EA} + (D - A) \tau_{DA} I_{DA} + (C - A) \tau_{CA} I_{CA} + E \Gamma_{EA} + D \Gamma_{DA} + C \Gamma_{CA},
$$

(20)

Setting all these time derivatives equal to zero and the total fractional population across all states equal to 1, we solve these equations to find the fractional steady-state populations in each state. The full derivation is given in the supplementary material. Here we simply quote the result, but we first introduce some notation to simplify the resulting expressions:

$$
\gamma_{ij} = \tau_{ij} I_{ij} + \Gamma_{ij},
$$

(21)

$$
k_{ij} = \frac{\tau_{ij} I_{ij}}{\sum_k \gamma_{ik}},
$$

(22)

$$
\zeta_{i} = \frac{1}{\sum_j \tau_{ji} I_{ji}},
$$

(23)

and the composite coefficient

$$
C_{1} = \frac{(k_{EA} \gamma_{EB} + k_{DA} \gamma_{DB}) \zeta_{B}}{1 - (k_{FB} \gamma_{FB} + k_{EB} \gamma_{EB} + k_{DB} \gamma_{DB}) \zeta_{B}},
$$

(24)

where the summations are carried out over all dipole-allowed transitions. Note that repeated indices do not imply summation here — summations are used only where explicitly stated. We now find that in the steady state

$$
A = \left[ 1 + \sum_i k_{iA} + C_{1} \left( 1 + \sum_j k_{jB} \right) \right]^{-1},
$$

(25)
with the remaining fractional populations given by

\[ B = C_1 A , \]  

(26)

and for the upper manifold

\[ i = k_{iA} A + k_{iB} B . \]  

(27)

Having obtained the steady state populations we can now determine the rate of photon loss per equilibrium-state atom, \( L_{eq} \), from a given beam as

\[ L_{eq} = \sum (j - i) \tau_{ij} I_{ij} , \]  

(28)

where the sum is taken over all combinations of upper manifold states \( i \) and lower manifold states \( j \) between which dipole-allowed transitions can be stimulated by the chosen beam (ignoring the negligible stimulation of cooler/repumper transitions by repumper/cooler lasers respectively). The un-pumped atoms can be taken into consideration at this stage as well, yielding

\[ L = (1 - F_N) \sum (j - i) \tau_{ij} I_{ij} + F_N \sum \frac{1}{2} \tau_{ij} I_{ij} . \]  

(29)

Equation (29) can be used to determine the rate of attenuation of a laser beam by stationary atoms subject to known illumination conditions. In order to accurately model a thermal atomic vapour, the velocity distribution of the atoms and corresponding Doppler shift in each beam’s effective detuning must be taken into account. This can be done by integrating equation (29) over the atomic velocity distribution, where the dependence of \( L \) on atomic velocity comes in via the dependence of the values of \( \delta_{ij} \) in equation (14) on atomic velocity (due to the Doppler shift) and the corresponding variation in the values of \( \tau_{ij} \). We define a new variable, \( L_{thermal} \), as the average loss rate of photons from the beam per atom, given the atoms’ thermal velocity distribution. In the case of a thermal atomic vapour at temperature \( T \), considering only the first-order Doppler shift, this is given by

\[ L_{thermal} = \frac{1}{N} \int_{-\infty}^{\infty} L(\nu) \exp (-mv^2/2k_B T) dv , \]  

(30)

where \( m \) is the mass of the atoms, \( k_B \) is the Boltzmann constant and the integration variable \( \nu \) corresponds to the atomic velocity component along the axis of the laser beams. \( N \) is the normalisation constant for the 1D Boltzmann distribution. Furthermore, we must consider that there are counter-propagating beams within the vapour cell. These can be taken into account simply by summing the contributions of the different beams to the stimulated transition rates, such that in the equations above \( \tau_{ij} I_{ij} \) becomes \( \tau_{ij} I_{ij}(\text{beam1}) + \tau_{ij} I_{ij}(\text{beam2}) \). Note that for counter-propagating beams the sign of the Doppler shift on the value of \( \delta_{ij} \) in equation (14) will be opposite for the two beams. With this substitution made equation (30) can then be applied individually to each laser beam present.

Equation (30) can now be used to deduce the mean optical absorption cross-section per atom as a function of both laser frequencies. Figure 5 shows the results of this. The same key features can be observed as in our experimental results (shown in figure 2). The presence of each beam increases the strength of the absorption of the other beam and creates sharp locking features, similar to those seen in saturated absorption spectroscopy, in a diagonal ‘criss-cross’ pattern across the whole Doppler valley.

This simple theory explains the qualitative behaviour of the system and can be used as a guide to the trends likely to be seen as experimental conditions are varied. However, a range of further effects must be accounted for in order to provide an accurate, quantitative description of the system, as laid out in more detail in the Supplementary Material. This is the subject of ongoing work.

6 Conclusions

Our results show that the use of spatially overlapping beams tuned to different atomic transitions can allow optical pumping effects to be exploited to enhance signal strength in atomic vapour spectroscopy. The technique also allows the two beams to
Figure 5. Predicted relative total optical cross section (cumulative over outward and return passes) per atom with respect to the cooler (a) and repumper (b) beams, as a function of the frequencies of both lasers. Frequencies for the cooler and repumper lasers are given relative to the F=4→F′=5 and F=3→F′=4 transitions respectively. The key features of the technique — the enhancement of the absorption strength from one laser when the other laser is close to resonance, and the ‘criss-cross’ pattern of sharp interaction features — are both clearly predicted by the simple rate equation model. The beam properties used in the simulation match those used in the experiment, except that it is assumed that the intensity of the return beam is always one half of that of the outward beam, when in reality this ratio would vary slightly according to the level of absorption in the atomic vapour. FN is set to 0.3. Note that an absolute scale is intentionally not provided for the absorption cross section, as this simple model is only expected to capture the qualitative trends seen in the system and not to provide accurate, quantitative information.

share the same optical path, thus reducing total system size and component usage, as well as allowing laser stabilisation at a greater range of frequencies than is possible via conventional spectroscopic techniques. In our experimental arrangement, which generates a feedback signal for laser stabilisation to the D2 line of Caesium via laser current modulation and demodulation of the corresponding spectroscopic signal at the same frequency, we found that the inclusion of a second, spatially overlapped beam could increase the frequency sensitivity of the resulting feedback signal by factors in excess of 3, corresponding to a reduction in signal-limited frequency deviation for lasers stabilised using the technique by a factor \( \sim 2 \).

The technique is likely to be of use wherever space, weight and optical components are at a premium as well as in situations where signal strength and frequency sensitivity are important. This will range from portable devices where reductions in size and weight are important and environmental noise sources make an increased signal strength desirable, through to precision lab-based experiments where the accuracy of laser frequency stabilisation is paramount and limited by the sensitivity or signal-to-noise ratio of the spectroscopic feedback signal.

The technique may prove particularly useful for spectroscopy of atomic Lithium. Firstly, Lithium reacts chemically with most standard glasses, meaning that vapour cells for Lithium are typically much larger and/or more costly than those for other alkali metals and so the benefit of using the same vapour cell for spectroscopy on two different lasers is strongly enhanced. Secondly, the absence of a ‘cycling transition’ in atomic Lithium, brought about by the small energy separation of the hyperfine states of the upper manifold of the D2 line, suggests that the benefits of avoiding optical pumping to dark states may be further enhanced for Lithium spectroscopy.

Future extensions of this technique could include demodulation of the signal from one photodiode at two different frequencies — corresponding to the two laser current modulation frequencies. This would allow measurement of, for example, \( dV_1/dF_1 \) and \( dV_1/dF_2 \) from a single photodiode; in the situation considered above \( dV_1/dF_1 \) is obtained by demodulation of the signal from one photodiode and \( dV_2/dF_2 \) from the other. In principle this could allow for both lasers to be stabilised using only one photodiode, which could be useful in systems where size and weight are paramount, or alternatively the extra information obtained by using two photodiodes, each demodulated at two different frequencies and thus generating four distinct feedback signals, could allow for yet more accurate stabilisation of both lasers.

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**Author contributions statement**

N.C. conceived the experiments, which were carried out by N.C., D.J. and S.M.. L.H. was responsible for funding acquisition and oversaw the work and provided advice and guidance. N.C. constructed the theoretical model and analysed the different approaches to laser stabilisation based on dual-laser spectroscopy. D.B. assisted with data analysis and presentation. N.C. wrote the paper, with assistance from all authors. All authors reviewed the manuscript.

**Additional information**

The authors declare no competing interests. All data necessary to support the conclusions of this article are given in the article. All further data related to this work are available from the authors upon request.
Supplementary materials for: Dual-frequency Doppler-free spectroscopy for compact atomic physics experiments

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ABSTRACT

Herein we give the full derivation of the solutions of our rate-equation model of the pumped atomic vapour system. We also provide full details of our method of numerical estimation of second derivatives — the method is not novel or of inherent interest, but the exact details of the estimation method used can affect the final results displayed in figure 4 of the main article, and it is therefore not possible to exactly reproduce our results without access to full details of the method. Finally, we briefly review some of the complications that would be involved in constructing a full theoretical model of the system, capable of accurately reproducing experimental results. Though such a model would be complex and unlikely to reveal new fundamental physics, it does have significant practical applications and is intended to be the subject of future research. We also present the data from figure 3(a) of the main article on a logarithmic scale, which illustrates how the technique is likely to be even more effective in miniaturised devices that contain only small vapour cells, with a correspondingly lower optical depth.

1 Solution to rate equations

In this derivation we will employ the terms defined in equations (11)-(12) and (19)-(22) of the main article. Setting all time derivatives to zero, as is the case in the steady state, equations (13)-(18) of the main text can then be re-arranged to give

\[ F = \frac{B\tau_FB_I_{FB}}{\gamma_FB} = Bk_{FB}, \]  
(1)

\[ E = \frac{A\tau_EA_I_{EA} + B\tau_EB_I_{EB}}{\gamma_EA + \gamma_EB} = Ak_{EA} + Bk_{EB}, \]  
(2)

\[ D = \frac{A\tau_DA_I_{DA} + B\tau_DB_I_{DB}}{\gamma_DA + \gamma_DB} = Ak_{DA} + Bk_{DB}, \]  
(3)

\[ C = \frac{A\tau_CA_I_{CA}}{\gamma_CA} = Bk_{CA}, \]  
(4)

\[ B = \frac{D\gamma_DB + E\gamma_EB + F\gamma_FB}{\tau_DB_I_{DB} + \tau_EBI_{EB} + \tau_FB_I_{FB}} = \frac{D\gamma_DB + E\gamma_EB + F\gamma_FB}{\zeta_B}, \]  
(5)

and

\[ A = \frac{C\gamma_CA + D\gamma_DA + E\gamma_EA}{\tau_CA_I_{CA} + \tau_DA_I_{DA} + \tau_EAI_{EA}} = \frac{C\gamma_CA + D\gamma_DA + E\gamma_EA}{\zeta_A}. \]  
(6)

Substitution of eqns (1) to (3) into (5) then yields

\[ B = \frac{\gamma_DB(Ak_{DA} + Bk_{DB}) + \gamma_EB(Ak_{EA} + Bk_{EB}) + \gamma_FBk_{FB}}{\zeta_B}. \]  
(7)
Collecting terms in $A$ and $B$ and dividing through by the coefficient of $B$, one finds that

$$B = C_1A.$$ (8)

We can now express all other state populations in terms of $A$. As a final constraint, we impose the condition that the sum over all state population fractions must be equal to 1. Expressing all state populations in terms of $A$ and setting their sum equal to 1 yields equation (23) of the main article. Combining this with equations (1) to (4) and (8) above directly gives all of the steady state population fractions.

2 Numerical estimation of second derivatives

Before gradient estimation is performed, linear interpolation is used to obtain, from our irregularly-spaced raw data points, data corresponding to the fractional absorption of each laser beam at each point on a regular, 2500 × 2500 point grid, running between the minimum and maximum frequencies of each laser. Once this regularised data is obtained, each first derivative is estimated using a symmetric, linear estimation based on the values at grid points ten spaces in either direction of the point in question, such that, for example

$$\frac{dS_1}{dx} \approx \frac{S_1(x+10,y) - S_1(x-10,y)}{20},$$ (9)

where $x$ and $y$ correspond to the indices of the grid points, such that $dS_1/dF_{\text{cooler}}$ etc. are ultimately found by multiplying the corresponding $dS_1/dx$ values by the frequency spacing between points on the corresponding axis. Equivalent expressions clearly exist for the three remaining first derivatives.

The calculated first derivatives are saved as arrays. This enables the corresponding second derivatives to be calculated, from the first derivative estimates, using exactly the method shown above.

This is a simplistic and un-optimised method of numerical derivative estimation, and more advanced techniques are certainly available. However, it was intentionally chosen in order to more accurately simulate the kind of low-complexity calculation that is likely to be performed in real time by experimental hardware, and to therefore better reflect a realistic implementation of our technique in an actual technological application. The use of more advanced gradient estimation techniques, potentially with a lower susceptibility to experimental noise, would likely improve upon the results given here but might overestimate the performance of the technique in realistic experimental implementations.

3 Complications of a full, theoretical model

In addition to requiring a quantum master-equation approach to achieve high accuracy under all conditions\(^1\), such a model would have to include a full analysis of transient, non-equilibrium effects in atoms that traverse the (spatially non-uniform) laser beam, integrated over all possible traversal speeds and trajectories in the thermal atomic vapour. It would also need to allow for the fact that the approximations typically made when analysing saturated absorption spectroscopy — that the ‘probe’ beam is weak and the ‘pump’ beam is strong\(^1\) — are not valid in this system. It would have to correctly handle the attenuation of the laser light as it passes through the atomic vapour, including the fact that the returning beam’s intensity will depend on the attenuation of the outward beam, which in turn depends on the return beam’s properties. The lifetime of the hyperfine states of the $6S_{1/2}$ manifold against collisional redistribution can not be neglected, even for atoms that leave the beam and might return to it later, necessitating an in-depth analysis of both interatomic collisions and collisions with the walls of the cell. All of the above effects can have non-negligible consequences for our results, and the authors do not believe the omission of any to be a good approximation in realistic systems of this kind. In short, the complexity of a model capable of providing accurate quantitative agreement with our results is very high, and since such a model is not necessary to support our conclusions or elucidate the underlying principles we leave it for future work.

4 Application within small vapour cells in miniaturised devices

Figure 3(a) of the main article was taken using a 75 mm long vapour cell. This size of cell is typical of existing, lab-based experiments but is not plausible in the next generation of miniaturised technologies for use outside the lab, or for spectroscopic devices that require a high spatial resolution. A smaller cell necessarily leads, for a given vapour pressure, to a lower 2D (projected) atom density and thus to a lower optical depth for laser beams passing through the cell. In addition, the experiment for which we give results was performed with Cs atoms, which have the highest vapour pressure of any commonly-used alkali metal species at 298 K.

As a result of the high total optical absorption in our experiment, our results — in particular figure 3(a) — do not make the degree of enhancement of the optical absorption via the use of dual-frequency spectroscopy immediately obvious. In order to
see how large an enhancement of the spectroscopic signal is likely to be possible when using a lower total optical depth, it is necessary to view the results in logarithmic space, thus illustrating the strength of the response per unit of atomic density. Supplementary figure 1 (below) shows the same results as given in figure 3(a) of the main article, but plotted on a logarithmic scale. Here, the degree of benefit offered by this technique in portable devices with small vapour cells, or in any other situation with reduced total optical depth, is clearly evident.

**Supplementary figure 1.** Natural logarithm of the spectroscopy signal from “Photodiode Repumper” (see figure 1(a) of main article) while light from the cooler laser, tuned 365 MHz below the $F = 4 \rightarrow F' = 5$ transition, is also present in the cell (blue line). For reference, a standard saturated absorption spectroscopy signal from the same apparatus (taken by blocking the light from the cooler laser) is also shown (orange line). Repumper laser frequency is given relative to the $F=3\rightarrow F'=4$ transition.

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