Inverted crossover resonance within one Zeeman manifold

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Received 9 February 2017, revised 8 July 2017
Accepted for publication 14 July 2017
Published 2 August 2017

Abstract

We carry out investigations of inverted crossover resonances (ICRs) in \( \pi \)-driven four-level systems where \( \Delta F \) can be zero. Through the use of sub-Doppler frequency modulation spectroscopy of the \((6\alpha^2)\) \( ^1S_0 - (6\alpha\rho)\) \( ^3P_1 \) transition in \(^{171}\)Yb the resonance becomes manifest. The centre frequency is inherently insensitive to first-order Zeeman shifts and equates to the two-level resonance frequency in the absence of a magnetic field. A rate equation model is used to help validate the nature of the resonance. Optical frequency measurements of the \( F' = 1/2 \) hyperfine line recorded over two months demonstrate a statistical uncertainty of \( 2 \times 10^{-11} \). The ICR found with the \( F' = 3/2 \) line is used for 556 nm laser frequency stabilisation, which is an alternative means when applied to magneto-optical trapping of \(^{171}\)Yb.

Keywords: laser spectroscopy, inverted crossover resonance, ytterbium, four-level rate equations

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

1. Introduction

Explorations in degenerate Fermi gases [1–3], quantum many body simulations with lattices [4–6], and atomic clocks [7–12] each make use of neutral ytterbium. A common approach to producing atomic cloud temperatures at a few tens of microkelvin is to employ two stages of laser cooling with 399 nm and 556 nm light, where the latter exploits the \((6\alpha^2)\) \( ^1S_0 - (6\alpha\rho)\) \( ^3P_1 \) intercombination line. The natural linewidth of which is 184 kHz and has a corresponding Doppler cooling limit of 4.4 \( \mu \)K. To generate frequency-stable 556 nm light a method often employed is stabilisation by locking to a high-\( Q \) optical cavity [13, 14]. Rather than take this approach, and despite the low scattering rate associated with the intercombination line, we employ sub-Doppler spectroscopy upon a thermal Yb beam that leads to a Zeeman slower and magneto-optical trap. In doing so, we have identified an inverted Lamb dip with the \( ^1S_0 F = 1/2 \) - \( ^3P_1 F' = 3/2 \) and \( ^1S_0 F = 1/2 \) - \( ^3P_1 F' = 1/2 \) hyperfine transitions in \(^{171}\)Yb. This was first noted recently in [15], where an emphasis was made in relation to its applicability to laser cooling of \(^{171}\)Yb. Here we provide a detailed description of the nature of the resonance, but also show the suitability of the \(^1S_0 F = 1/2 \) - \( ^3P_1 F' = 1/2 \) line as an effective frequency reference. For the two hyperfine lines the application of a dc magnetic field transforms a two-level scheme into a four-level scheme, with degeneracy only marginally lifted in the ground state. The atoms are excited through \( \pi \)-transitions, while the de-excitation process completes a simple optical pumping scheme. The enhanced absorption is as an inverted crossover resonance (ICR), where the principal lines involved in the crossover are transitions to symmetrically separated Zeeman substates. In the case of \(^{171}\)Yb \( F' = 1/2 \) this appears to be the first instance of an ICR where \( \Delta F = 0 \). While the ICR for \(^{171}\)Yb \( F' = 3/2 \) is successfully used to stabilise the frequency for laser cooling, the \(^{171}\)Yb \( F' = 1/2 \) line also acts as a simple and reliable frequency reference with statistical and systematic uncertainties in the 10 kHz range. In contrast to an ordinary crossover resonance where the signal is limited to approximately the summed strength of the principal lines, the ICR magnitude can be sizeably larger than this because the saturation behaviour of (closed) two-level systems no longer applies. We note that

To distinguish the two \(^{171}\)Yb \( ^1S_0 \) - \( ^3P_1 \) hyperfine lines we will state the upper \( F \) value as a subscript.
such resonances are rarely observed on atomic beams because the natural linewidth of the transition needs to be below a few megahertz (for a sufficiently collimated atomic beam). By carrying our similar measurements with $^{173}$Yb (I=5/2) and detecting extremely weak ICRs, we find that such ICRs are most apparent when $I = 1/2$. We note that saturation spectroscopy has recently been performed elsewhere with the Yb intercombination line, but in a configuration that may make observing the ICR rather difficult [16]. The four-level behaviour we observe is similar to the double-$N$ description given in the work of Scotto et al [17]. There they investigate Rb in a vapour cell at significantly higher $B$-field, and in our case the pump and probe signals are of approximately the same intensity.

In this paper we open with a description of the sub-Doppler spectroscopy on the Yb intercombination line and commence with an example of an ordinary crossover resonance, before showing results relating to, and describing, the ICR. We develop a four-level rate equation model that adequately describes the behaviour seen experimentally. Section 4 describes how the ICR arises from the Zeeman splitting of a $^{1}S_{0} - ^{3}P_{1}$ transition in an atomic beam of Yb. There is also a concave field plane of polarisation;

2. Sub-Doppler spectroscopy

The scheme for the sub-Doppler frequency modulation (FM) spectroscopy is illustrated in figure 1, where a retro-reflected 556 nm beam passes perpendicularly through the ytterbium atomic beam and the 556 nm fluorescence is detected by a photo-multiplier tube. A lens-mirror combination (cat’s eye) is used to produce the retro-reflection. The scheme is similar to that used for many saturated absorption spectroscopy schemes [18, 19], however, given that we observe enhanced absorption, rather than saturation, we refer to the scheme as sub-Doppler FM spectroscopy. FM at 33 kHz is applied to the light via an acousto-optic modulator allowing for $n$th harmonic detection with a lock-in amplifier. Unless otherwise mentioned $n = 3$. Helmholtz coils set the magnetic field in the vertical direction. The polarisation, $E$, of the 556 nm light is set either parallel to that of $B$ or perpendicularly. For the cases of enhanced absorption, $E$ is parallel to $B$, which is the case for most of the results presented here. The intensity profile of the 556 nm beam is elliptical with the major axis (4 mm in diameter) aligned with the atomic beam. The 556 nm light is produced via resonant frequency doubling of 1112 nm light, where a bow-tie type doubling cavity has at its prime focal point an 18 mm length periodically poled potassium titanyl phosphate crystal for the nonlinear conversion, and comprises mirrors with 75 nm radius of curvature. The 1112 nm light is sourced from a commercial fibre laser that injection-locks a ridge waveguide diode laser for amplification to $\sim 45$ mW [20].

An effusion cell heated above 400 °C produces a beam of Yb atoms (with natural isotopic abundance) that escape a reservoir through a trio of stainless steel tubes with a diameter of 1 mm and length 20 mm. Any accumulation of Yb within the tubes is likely to increase the aspect ratio and reduce the angular divergence of the atoms. No further collimation techniques are performed. The fluorescence from the atom–light interaction passes through a spatial filtering scheme to minimise stray light, along with an edge filter to block scattered 399 nm light from a Zeeman-slower beam. The fast-linewidth of the 556 nm light we presume to be $\sim 240$ kHz, based on the manufacturer’s linewidth specification for the 1112 nm fibre laser (thus, slightly larger than the natural linewidth of the $^{1}S_{0} - ^{3}P_{1}$ line). Frequency tuning of the 556 nm light is carried out via a piezo transducer in the 1112 nm laser. We calibrate the frequency tuning by use of a frequency comb that is referenced to a hydrogen maser. Further comments regarding the comb are made below when characterising the frequency instability of the light locked to the ICR.

Crossover resonances are commonly observed in saturated absorption spectroscopy. ICRs are less frequently observed, but their occurrence has been known for several decades [21–23]. The element that is unique here is that the ICR arises from the Zeeman splitting of a $\Delta F = 0$ transition. Crossover resonances are most easily observed in vapour cells where there is a full range of velocity components. Atomic beams, on the other hand, have a very restricted range of transverse velocity; hence the possibility of observing crossover resonances is very much reduced.

Crossover resonances preserving the saturated absorption can be produced with the bosonic ytterbium isotopes (even mass number) when the excitation occurs through $\sigma$-transitions. This is made possible when $E$ and $B$ are orthogonal to each other (i.e., with linear-$\sigma$ polarisation [24]. An example is shown in figure 2 taken for $^{174}$Yb and the $^{1}S_{0} - ^{3}P_{1}$ line. Here the nuclear spin $I = 0$ (for isotopic and hyperfine line frequency separations for the intercombination line in ytterbium see [15, 25–27]). Figure 2(a) is the third harmonic output of the lock-in amplifier and figure 2(b) is the corresponding absorption signal. The outer absorption, rather than saturation, we refer to the scheme as sub-Doppler FM spectroscopy. FM at 33 kHz is applied to the light via an acousto-optic modulator allowing for $n$th harmonic detection with a lock-in amplifier. Unless otherwise mentioned $n = 3$. Helmholtz coils set the magnetic field in the vertical direction. The polarisation, $E$, of the 556 nm light is set either parallel to that of $B$ or perpendicularly. For the cases of enhanced absorption, $E$ is parallel to $B$, which is the case for most of the results presented here. The intensity profile of the 556 nm beam is elliptical with the major axis (4 mm in diameter) aligned with the atomic beam. The 556 nm light is produced via resonant frequency doubling of 1112 nm light, where a bow-tie type doubling cavity has at its prime focal point an 18 mm length periodically poled potassium titanyl phosphate crystal for the nonlinear conversion, and comprises mirrors with 75 nm radius of curvature. The 1112 nm light is sourced from a commercial fibre laser that injection-locks a ridge waveguide diode laser for amplification to $\sim 45$ mW [20].

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dips are produced by the $J = 0, m_J = 0 \rightarrow J' = 1, m_J = \pm 1$ transitions, and the central dip is the crossover resonance. An explanation for the line spectrum is summarised in figure 3. As is commonly observed, the crossover resonance is stronger than the principal lines because there are two groups of atoms contributing to the signal, as opposed to one. This is portrayed in the upper left panel of figure 3. The principal lines are those generated by the atoms marked $P$ (those flowing perpendicularly to the 556 nm laser beams), and the crossover resonance is produced by the combined subsets of (1) and (2). Note, this image should not be taken too literally, because the subsets are in velocity space, while the image is drawn in a spatial sense. The transitions in this case are closed, having no alternative pathways. In the measurement of figure 2 the 556 nm combined beam intensity was $\sim 160 I_{sat}$ (where the saturation intensity $I_{sat}$ is 1.4 $\text{W m}^{-2}$), thus causing artificial broadening of the lines. Also affecting the resonance widths is the strength of the modulation applied to the 556 nm light. This is discussed below. The Zeeman line separation, here at 0.28 mT, is consistent with the first-order Zeeman shift of 21 MHz mT$^{-1}$ for the $m_J = \pm 1$ substates (which applies to all the bosonic isotopes).

3. ICR: measurement and modelling

The same experimental arrangement was applied to the $^{171}\text{Yb}$ atoms (with $I = 1/2$), except with $\vec{E}$ parallel to $\vec{B}$, ensuring that only $\pi$-excitations occur. A series of spectra are shown in figure 4 for the $^3S_{1/2} \rightarrow ^3P_{1/2}$ line with each trace recorded for a different bias magnetic field. The upper traces are experimental data and the lower traces are lineshape models. A similar sequence occurs when the direction of the bias is reversed (for the case of zero field, see [15]). One first notices the Zeeman components with $B$ at 0.24 mT and at slightly higher fields they are easily resolved. Note the opposing slopes of the main discriminator and the Zeeman components indicating that the crossover resonance is inverted. The mean width across the main discriminator for this set of measurements is $\sim 1.7$ MHz, which is dominated by intensity broadening, the intensity having been set to 90 $I_{sat}$, and the effect of strong FM. The applied modulation amplitude was 2 MHz. At zero $B$-field there is usually a weak saturated absorption signal arising because the four-level structure collapses into a two-level scheme. This is more readily observed at higher intensities. At low intensities ($\sim 50 I_{sat}$) the ICR may remain, but is very much suppressed. At $\sim 0.12$ mT the ICR reaches its maximum contrast and above which the sequence in figure 4 follows.

Before giving a fuller description, the resonances can be modelled in a basic way with a sum of Lorentzian lineshapes: two separate saturated lines and a central inverted line. The Lorentzian function has the form: $f_L(\delta, b) \propto (1 + (2(\delta - b)/\Delta)^2)^{-1}$, where $\delta$ is the frequency detuning, $\Delta$ is the full-width-half-maximum and $b = g_F \mu_B B/h$ we denote for the Zeeman shift of the magnetic sublevels, with $g_F$ the Landé $g$-factor and $\mu_B$ the Bohr magneton. Representing the signal from the lock-in amplifier with third harmonic detection we have,

\[ S_L(\delta) = \frac{d^3}{d\delta^3} [f_L(\delta, -b) + \mathcal{R} f_L(\delta, 0) - f_L(\delta, b)]. \]  

The ratio, $\mathcal{R}$, between the strength of the inverted line to the principal (Zeeman) lines is treated as a free parameter here. Such an assumption is not needed below in a more complete description of the four-level system. For both the experimental data and the summed Lorentzian model the third derivative is shown in figure 4. One sees a clear correspondence between the two sets of traces, with similar features appearing as the Zeeman components separate. For the $^{171}\text{Yb}_{F'=1/2}$ line here $b_F = 28$ MHz mT$^{-1}$, and $\mathcal{R} = 2.7$, notably larger than two.

A partial explanation for the ICR is seen in figure 5. A key aspect is the four-level structure and two branching routes for each upper Zeeman level (the ground state Zeeman splitting is 2.7 $\times 10^{-4}$ times weaker than that of the upper state splitting). The branching ratios from the upper states are readily calculated for the $^{171}\text{Yb}_{F'=1/2}$ and $^{171}\text{Yb}_{F'=3/2}$ systems [28], given that state-mixing with the $^3P_1$ state permits the $^3S_{1/2} \rightarrow ^3P_1$ transition to occur. Figure 5 shows the branching ratio for the $^3S_{1/2} \rightarrow ^3P_{1/2}$ hyperfine line. Two subsets of atoms combine to produce the ICR, each with the same absolute value of Doppler shift ($\omega_0$), but opposing sign. For the excitation phase the 556 nm light polarisation is parallel to the magnetic field, implying $\Delta \omega_B = 0$. When the optical frequency, $\omega$, lies midway between the upper state separation, $\omega_0$, then both subsets of atoms participate maximally in the absorption and cycling process ($\omega_B$ is the mean of $\omega_0$ and $\omega_H$). Atoms (1) with $+\nu_B$ Doppler shift interact with the $-\nu$ beam, exciting transition (B) such that $\omega_B = \omega_0 + \nu$. Alternatively, these atoms can interact with the $+\nu$ beam, exciting transition (A), where $\omega_A = \omega_0 - \nu$. Whichever decay channel occurs there is always the possibility for re-excitation, hence atoms are not lost to a dark state. The situation is reversed for atom subset (2).
The width of the central dispersive curve reduces to \(~800\) kHz when the 556 nm beam intensity is 30 \(I_{\text{sat}}\) (there are other factors influencing the width that are discussed below). For the most probable velocity of \(330 \text{ m s}^{-1}\), this corresponds to 1.4 mrad of separation between the two diverging sets of atoms responsible for the ICR.

The strength of the resonance exhibits symmetrical behaviour for opposing magnetic field directions, as shown in figure 6(a). The vertical axis represents the size of the discriminator from the output of the lock-in amplifier, again recorded with third harmonic detection. At zero \(B\)-field the Zeeman sublevels remain degenerate, thus acting like a two-level system and the usual saturated absorption behaviour occurs. Applying the \(B\)-field in either direction causes the Zeeman level splitting and two decay channels to occur for each excited Zeeman state. The situation is similar for the \(^{171}\text{Yb}\) \(^{1}S_{0} F=1/2 \rightarrow ^{3}P_{J=3/2}\) line (there is a frequency separation of 5937.78 MHz between the two \(^{171}\text{Yb}\) hyperfine lines); however, there is a reversal of branching ratios where the \(\sigma\) channel de-excitation occurs with 1/3 probability and the \(\pi\) channel de-excitation occurs with 2/3 probability. The maximum signal strength and contrast is greater for the \(F=1/2 \rightarrow 1/2\) line than the \(F=1/2 \rightarrow 3/2\) line. The reduction in signal strength at higher \(B\)-field takes place as the two Zeeman lines separate (also seen in figure 4). The Zeeman shift of 14 MHz mT\(^{-1}\) for \(F=1/2 \rightarrow 1/2\) is twice as strong as that for \(F=1/2 \rightarrow 3/2\); hence the lines become resolved at lower fields for the former and the main discriminator strength falls away more rapidly with \(B\)-field. The dashed lined curve fits away from saturation of figure 6(a) are derived from the simple summed Lorentzian model described above (and cannot reproduce saturation as it is void of any population dynamics—hence the disconnect near zero \(B\)). The abscissa scaling between the \(F'=1/2\) and \(F'=3/2\) data fits precisely the factor of two ratio of the Zeeman shifts. The peak-to-peak values versus magnetic field in figure 4 has a slightly different trend compared to the data in figure 6(a), with respect to the \(F'=1/2\) data. The main reason being that the intensity used for figure 6(a) was \(\sim 180 I_{\text{sat}}\), thus broadening the lines and influencing the progression seen in figure 4 (the FM amplitude was also twice as strong).

To encompass the full range of behaviour of the four-level system a rate equation model was developed to make comparisons with the experimental data. With a 4 mm laser beam width (in the direction of the flow of atoms) and a most probable velocity of \(330 \text{ m s}^{-1}\), the atomic transit time across the beam is estimated to be about 12 \(\mu\)s, which is...
large compared to the lifetime of the \(^3P_1\) level (0.86 \(\mu\)s). To a good approximation the atomic evolution should reach the steady-state, even at high saturation. In this case, the result of the laser–atom interaction can be estimated by the result of the quantum model.

A set of rate equations is solved describing population changes in a four-level system, where the fractional population of the four levels is \(n_j\), \(j \in \{1, 4\}\). The labelling of the states is shown in figure 7(a). In steady state, the rate equations for a \(\pi\) polarised light become,

\[
0 = \begin{cases} 
  n_1(W_{12} + \gamma_1) - n_4(W_{12} + a_{21}\Gamma) - n_2a_{41}\Gamma \\
  n_2(W_{12} + \Gamma + \gamma_1) - n_1W_{12} \\
  n_3(W_{34} + \gamma_3) - n_4(W_{34} + a_{43}\Gamma) - n_2a_{23}\Gamma \\
  n_4(W_{34} + \Gamma + \gamma_3) - n_3W_{34} 
\end{cases}
\]  

(2)

where \(\Gamma\) is the spontaneous rate of emission, \(W_{ij}\) is the pumping rate of the transition with levels \(i, j\), and \(a_{ij}\) are the relaxation branching coefficients and \(\gamma_{ij}\) is a relaxation term introduced to account for the finite interaction time as the atoms transit the laser beams. The ground states will have equal population before the interaction (denoted \(n_{10}\)) and without any loss of atoms the sum \(\sum_{i=1}^{4} n_i = 1\) holds. The solutions for \(n_i\) are given in equation (6) in the appendix.

The driving rates are found from,

\[
W_{12} = a_{21}\sigma_0 \delta \eta N_g \\
W_{34} = a_{43}\sigma_0 \delta \eta N_g
\]

\[\times \left\{ \begin{array}{c}
  \frac{\Gamma_2\Gamma/4}{\left[ \delta_{12} - kv + a_m \cos(x) \right]^2 + (\Gamma_2/2)^2} \\
  + \frac{\Gamma_3\Gamma/4}{\left[ \delta_{12} + kv + a_m \cos(x) \right]^2 + (\Gamma_2/2)^2} \\
\end{array} \right\}
\]

\[\times \left\{ \begin{array}{c}
  \frac{\Gamma_2\Gamma/4}{\left[ \delta_{34} - kv + a_m \cos(x) \right]^2 + (\Gamma_2/2)^2} \\
  + \frac{\Gamma_3\Gamma/4}{\left[ \delta_{34} + kv + a_m \cos(x) \right]^2 + (\Gamma_2/2)^2} \\
\end{array} \right\}
\]  

(3)
where $s_0$ is the on-resonance saturation parameter, $c_0 = 3\lambda^2/2\pi$ is the light-on scattering cross section (on-resonance), $k$ is the wave number and $\Gamma_2$ is the combined linewidth associated with spontaneous emission, transit time and laser frequency noise, $\beta = \Gamma + \gamma_f + \gamma_L$, with $\gamma_L$ the laser linewidth, $a_{\text{at}} \cos(x)$ is the frequency modulation term which will be defined more completely below. $N_{\beta} = I_{\text{at}}/h\nu_0$ represents the number of photons per unit area per second at the saturation intensity. The frequency detuning terms account for the Zeeman shift through,

$$\delta_0 = \delta + (m_1 g_s - m_j g_j) B \mu_0 / h,$$

where $\delta = \omega - \omega_0$ is the laser frequency detuning from the centre of resonance in null magnetic field, $\omega_0$; $g_s$, $g_j$ is the Landé factor of the level $i$, and $h$ is the reduced Planck constant. We note the ground state splitting is negligible in comparison to the upper state splitting. The laser frequency is sinusoidally modulated and the fluorescence signal is detected by means of third harmonic output of a lock-in amplifier. The modulation frequency, $\omega_\text{m}$, is fast in comparison to the mean laser frequency scanning rate, i.e. the detuning, $\delta$, is approximately constant during a modulation period, so that we can insert the modulation term in the steady-state equations. The $a_{\text{at}}$ term is the FM amplitude (in rad s$^{-1}$) and $x = \omega_0 t$ is the modulation term’s phase with $t$ the time. In this case, following from (29–31), the $q$-th Fourier-series coefficient of the modulated signal forms the signal yielded by the lock-in detection employing in-phase detection at the $q$-th harmonic of the modulation frequency. The combined signal is evaluated through,

$$S_q(\delta) = \frac{2\Gamma}{\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} [n_2(\nu, \delta, x) + n_4(\nu, \delta, x)] \cos(q\delta)f(\nu) d\nu dx,$$

where the rate equations are solved in a stepwise manner assuming a top-hat transverse velocity distribution $f(\nu)$ truncated at $-v_t$ and $v_t$. We choose $v_t = 10$ m s$^{-1}$, which is well beyond the Doppler components that compensate for the Zeeman shifts explored here. The equation represents an evaluation of the photon fluorescence rate produced by atoms in the two upper states. We note that equation (5) remains valid if the modulation amplitude is not large in comparison to the intensity broadened linewidth. Figure 7(b) provides an example of resonance width versus FM amplitude at $s_0 = 90$ and $B = 0.1$ mT for both measurement and model. The two agree reasonably well until a modulation amplitude of $\sim 2.5$ MHz is reached. The dashed line indicates a linear trend for the measured data.

The relaxation branching ratios of equation (2) can be summarised with $a_{21} = 1 - a_{23} = a_{43}$ and $a_{24} = a_{23}$. For the $S_0 F = 1/2 - 3P_1 F = 1/2$, $a_{23} = 2/3$ and for the $S_0 F = 3/2, a_{23} = 1/3$. These branching ratios may not be exact, since the intercombination line is a spin-flip transition (that is permitted through mixing with the $P_1$ state). Figure 6(b) shows the spectrum for the $S_0 F = 1/2 - 3P_1 F = 1/2$ line with third harmonic detection, $s_0 = 88, B = 0.15$ mT and a FM amplitude of 2 MHz. The solid (green) line is generated by the rate equation model for the same magnetic field, intensity and modulation amplitude. Only vertical scaling is applied to the computed result. The agreement between the experimental data and the model lends credence to its validity.

The comparison between model and data is also shown in figure 6(a). The solid lines are the computed signal strengths for the $S_0 F = 1/2 - 3P_1 F = 1/2$ and $S_0 F = 3/2 - 3P_1 F = 3/2$ lines. The input parameters match those of the experiment, where $s_0 = 176$ and the FM amplitude is 4 MHz. The model shows the same characteristic behaviour across the full magnetic field range: from saturation at and near zero field, the reverse peaks away from zero field. However, we have applied independent scaling between the data and model for the two hyperfine lines. The model creates different ICR strengths for the two hyperfine lines because of the different branching ratios, but the ratio is not in accord with that observed in the experiment. In the model the $F' = 3/2$ resonance is $\sim 15\%$ stronger than the $F' = 1/2$ ICR. This difference in ratio may be because the model does not take into account the spatial dependence of fluorescence. The quantity of fluorescence heading in the direction of the PMT differs depending on whether the released radiation is $\pi$ or $\sigma$ polarised [24]. For the two cases the ratio of $\pi$-to-$\sigma$ polarised light changes depending on the branching ratios. Another possible, but unlikely, factor influencing the behaviour of the $F' = 3/2$ line shape is the presence of the $171\text{Yb}^+ S_0 F = 5/2 - 3P_1 F = 3/2$ line, which lies only $\sim 2.6$ MHz away (the $171\text{Yb}$ line is approximately one sixth the strength of the $171\text{Yb}^+ F = 3/2$ line).

### 4. Frequency stabilisation

The ICR with the FM spectroscopy produces a dispersive signal that one can use to stabilise the 556 nm laser frequency. The correction signal is sent to the 1112 nm fibre laser through a piezo transducer with a servo bandwidth of approximately 20 Hz (deliberately set low to avoid adding technical noise at higher Fourier frequencies). We assess the frequency instability by counting the frequency of the beat generated by mixing 1112 nm light with the adjacent element.
of a frequency comb, whose mode spacing is referenced to a H-maser signal. Here we consider first harmonic detection in the FM spectroscopy as it confers approximately a factor of two improvement in the frequency instability compared with third harmonic detection (and is relevant for the laser cooling below). The frequency instability for an atomic flow rate of $N \sim 4 \times 10^{10}$ s$^{-1}$ through the beams is shown in figure 8(a). This result is for the $^{171}$Yb$_{F=3/2}$ hyperfine line. The left ordinate is the instability made fractional with respect to the carrier frequency of the 556 nm light. The instability reduces with a $\tau^{-1/2}$ dependence out to about 600 s. At 50 ms, which is the transfer duration for the second stage of the MOT, the instability is 45 kHz, and is well below the natural linewidth of the $^3P_1 - ^5S_0$ line (184 kHz). The atomic flow rate was estimated by recording the dc level of the PMT (registering 556 nm photons), accounting for the collection efficiency and applying the standard scattering rate equation (for zero frequency detuning). The uncertainty for $N$ is $\sim 50\%$. Based on particle flow rates through a cylindrical tube and accounting for atomic beam divergence in the vertical plane, $N$ should be an order of magnitude greater. The difference may be explained if Yb has accumulated to the sides of the tubes and reduced the aspect ratio.

Further improvements in frequency instability can be produced by increasing the atomic flow rate as seen in figure 8(b). A power law fit gives a dependence of $N^{-0.7}$. This rate of fall is likely a coincidental combination of noise terms proportional to $(N^{-2} + bN^{-1})^{1/2}$, with $b$ some constant. We do not observe a change in frequency instability with changes by an order of magnitude in optical intensity, suggesting that photon shot-noise is not a limiting factor, thus an unidentified noise source in the detection system contributes to the instability. The falling frequency instability is a useful trait for lasers employed in laser cooling. Moreover, by locking to the $^3S_0 - ^3P_1$ line one has immediate access to the frequency detuning (unlike in the case of locking to an optical cavity).

The symmetry of the ICR is again apparent when the line-centre frequency is measured as a function of magnetic field. The laser is locked to the centre of the dispersive curve and frequency counting of the IR beat is made for approximately 100 s at each $B$-field setting. On this occasion we consider the $^{171}$Yb$_{F=1/2}$ hyperfine line and third harmonic detection. Figure 9(a) shows the mean line-centre frequency, $\nu_{F=1/2}$, versus magnetic field. The frequency variations are constrained to within $\pm 5$ kHz across $\sim 1.5$ mT (or 15 G). In fractional terms this is $9 \times 10^{-12}$, demonstrating that the first order Zeeman shift is efficiently cancelled with this ICR (and the second order dependence is not apparent). Other systematic shifts have been investigated for the $^{171}$Yb$_{F=1/2}$ line. One can measure the frequency dependence on the separation between the beam and the optical axis of the cat’s eye, both in the direction of the atomic beam and in the vertical direction. In both cases there is a monotonic shift that is linear over small displacements, as seen in figure 9(b). In the horizontal plane the shift is 26 kHz mm$^{-1}$. Zero displacement is made evident by observing the signal-to-noise ratio of the signal, or equivalently the Allan deviation of the locked laser frequency. Alignment between the 556 nm beams and the optical axis can be determined to within $\pm 0.4$ mm, which equates to an uncertainty of $\pm 11$ kHz. Displacing the lens in the vertical direction creates a linear frequency shift over $\pm 0.3$ mm from the optical axis. A displacement greater than $\sim 0.2$ mm is relatively easily identified and the associated systematic shift is 13 kHz. We note that for larger ranges of displacement in the direction of the atoms the frequency variation can exhibit a quadratic dependence, the curvature of which may depend on the angle between the 556 nm beams and the optical axis of the cat’s eye. Shifts associated with the wavefront curvature were tested by changing the lens-mirror separation in the cat’s eye. When set to the focal length of the lens the shift is less than 3 kHz mm$^{-1}$.

The AC Stark shift was measured by varying the 556 nm beam intensity over a range of $s_0 \in \{10, 600\}$ where $s_0$ is the on-resonance saturation parameter ($s_0 = I/\hbar\omega_c$). The shift is almost indiscernible at 0.034 $\pm 0.019$ kHz W$^{-1}$ m$^2$. For the absolute frequency measurements made here the 556 nm intensity is $\sim 80$ W m$^{-2}$ (for the two beams), thus the associated correction required is smaller than the statistical variations between repeated measurements. The insensitivity to magnetic field and low sensitivity to other shifts prompted the recording of (semi-) regular measurements of $\nu_{F=1/2}$ over an extended period of time as a test of the repeatability. Measurements were taken over a period of two months and are shown in figure 9(c). The mean optical frequency from this series is 539 384 469 031(8)(23) kHz, where the standard deviation from the weighted mean is 8 kHz (which is $\sim 5\%$ of the natural linewidth). The reduced $\chi^2$ for the set is 2.1. Through the quadratic sum of uncertainties associated with systematic shifts (mentioned above) the total systematic uncertainty is 23 kHz. Also included is an uncertainty associated with servo error (more specifically, identifying the centre of the line)$^4$. We note this overall uncertainty is more than an order of magnitude smaller than the previous most accurate measurement of a Yb intercombination line frequency [32]. While not at all competitive with cold-atom atomic frequency standards, the $^{171}$Yb$_{F=1/2}$ ICR provides a simple and effective thermal beam frequency reference. The $^{171}$Yb$_{F=1/2}$ line has the advantage that it is free from neighbouring isotopic lines that can perturb the lineshape, unlike the $^{171}$Yb$_{F=3/2}$ line. Although calibration of the H-maser was not carried out during this measurement run it is

\[\text{Figure 8. (a) Frequency instability of the 556 nm light when locked to the }^{171}\text{Yb}_{F=1/2} \text{ inverted crossover resonance for an atomic flow rate of } \sim 4 \times 10^{10} \text{s}^{-1}. (b) Frequency instability at } \tau = 1 \text{ s and its dependence on atomic flow rate.}\]
is applied for all measurements to create the ICR. The offset frequency is the 556 nm beams with the optical axis of the cat’s eye lens position with use of the ICR. Zero corresponds to alignment of the 556 nm beams with the optical axis of the cat’s eye.

The stage cooling is performed with 399 nm light on the 1S0 – 1P1 line measurements. The mean deviation from previously published values of the clock transition frequency is less than 10 kHz. The method used for the clock line frequency measurements was reported in [32], though notably, we now perform all the measurements free of light shifts associated with the MOT.

In the case of the 171YbF=3/2 ICR, the frequency stabilised 556 nm light is used to cool atoms in a dual-colour magneto-optical trap. The experimental set-up is depicted in figure 10, where the sub-Doppler spectroscopy is carried out prior to the Yb atoms reaching a Zeeman slower. The first-stage cooling is performed with 399 nm light on the 1S0 – 1P1 transition, then second stage cooling is carried out with 556 nm light driving the intercombination line. Further details about the magneto-optical trap are described in [15]. In the FM scheme for the laser lock we use the first harmonic detection. We performed atomic cloud temperature measurements as a function of the 556 nm light frequency instability, where the temperature was estimated based on the expansion rate of the cloud and imaging onto a CCD. Examples of temperature measurements for 171Yb are shown in figure 11, where the sub-Doppler spectroscopy is carried out through sub-Doppler spectroscopy of the intercombination line. Free expansion of the atom-cloud is recorded with the CCD camera. The λ/4 refers to achromatic quarter wave plates suitable for 399 nm and 556 nm wavelengths. SPF (LPF), short (long) wavelength pass filters (505 nm). Other labels are defined in figure 1.

5. Conclusions

We have presented results in relation to the recently discovered crossover resonances in neutral ytterbium, notably the ICRs produced with the two hyperfine lines in 171Yb. They arise through sub-Doppler spectroscopy of the intercombination line (\(\Gamma/2\pi = 184\) kHz) when the atoms are formed into a thermal beam. This appears to be the first instance where the ICR occurs with \(\Delta F = 0\) and the principal lines belong to the same Zeeman manifold. The I = 1/2 isotope creates a circumstance where only two narrow velocity groups are involved in the generation of the resonance. The ICR is apparent when the separation of the principal lines ranges from ~1 MHz to tens of megahertz, which is unlike the case of ICRs in gas cells, where the separation is typically hundreds of MHz [33]. In such cases the crossover occurs between principal lines that involve different hyperfine states. There is a high degree of symmetry in the line profiles due to the nature of the phenomenon and this is confirmed by use of a four-level rate equation model. The
enhanced absorption is particularly striking in the case of the $^{171}\text{Yb}$ $^5S_0 F=1/2 - ^3P_1 F=1/2$ hyperfine line. By frequency locking to the centre of the ICR, one can generate a frequency reference that is independent of the Zeeman shift to first order. We observe fractional variations less than $9 \times 10^{-12}$ over a magnetic field range of $\pm 0.8$ mT. Hence, the ICR for $^{171}\text{Yb}$ $F=1/2$ gives a very good representation of line-centre at zero $B$-field. Absolute frequency measurements of the $^{171}\text{Yb} F=1/2$ line against a hydrogen maser were recorded over a two month period. The mean frequency is $539.384469031(8)(23)$ kHz. The combined $1\sigma$ uncertainty of 24 kHz in fractional terms is $4.5 \times 10^{-11}$. We have investigated relevant systematic shifts, the most significant being the overlap of the forward and return beams used in the sub-Doppler spectroscopy, and errors associated with identifying line-centre. The accuracy of the hydrogen maser was verified by carrying out $^5S_0 - ^3P_0$ line frequency measurements in $^{171}\text{Yb}$. We believe this to be the most accurate Yb intercombination line frequency measured to date. The reproducibility of the line frequencies points to a modestly accurate frequency reference that is simple and with the potential to be compact and robust [34]. We note that with the atomic flow rates estimated here, 1 g of Yb would last over one hundred years with continuous operation. The $^{171}\text{Yb} F=1/2$ frequency can be used to compute a value for the $^{171}\text{Yb} F=3/2$ frequency based on the hyperfine frequency separation of 5937 779(57) kHz [15]. The result is $539.390406810(62)$ kHz, which is consistent with that previously reported, but with reduced uncertainty [32].

For the $^{171}\text{Yb} F=3/2$ hyperfine line the ICR was used as a means of laser frequency stabilisation for laser cooling. By locking to the centre of the crossover resonance we show that temperatures of sub-20 μK can be produced in a dual-colour magneto-optical trap. By measuring the atom-cloud temperature at various levels of frequency instability, we find that the remaining instability of the 556 nm light is not responsible for the temperature limit; hence locking to the ICR is an effective means of laser stabilisation for laser cooling. Similar ICRs with an atomic beam are expected to manifest whenever the species has a nuclear spin of 1/2 and the electronic transition has a sufficiently narrow natural linewidth; other examples include the intercombination lines in $^{199}\text{Hg}$ ($\Gamma/2\pi = 1.3$ MHz), $^{111}\text{Cd}$ and $^{115}\text{Cd}$ ($\Gamma/2\pi = 65$ kHz).

**Acknowledgments**

This work was supported by the Australian Research Council (grant LE11101000054). JM was supported through an ARC Future Fellowship (FT110100392). We thank members of the ARC CoE Engineering Quantum Systems and OBEL for the use of diagnostic equipment, and Nils Nemitz, Frédéric du Burck and Stefan Weyers for valuable discussions.

**Appendix. Populations at steady-state**

The solutions to the steady-state four-level rate equation for the atomic populations are the following. The parameters are described in the main text.

\[
\begin{align*}
n_1 &= \gamma_1(W_{12} + \Gamma + \gamma_2)(n_0^0 a_1 W_{34} \Gamma \\
    &\quad + n_1^1 W_{34}(\Gamma(1 - a_{43}) + 2 \gamma_2) + \gamma_6(\Gamma + \gamma_6))/D \\
n_2 &= \gamma_2 W_{12}(n_0^0 a_1 W_{34} \Gamma \\
    &\quad + n_1^1 W_{34}(\Gamma(1 - a_{43}) + 2 \gamma_2) + \gamma_6(\Gamma + \gamma_6))/D \\
n_3 &= \gamma_3(W_{34} + \Gamma + \gamma_6)(n_0^0 a_{23} W_{12} \Gamma \\
    &\quad + n_1^0 W_{12}(\Gamma(1 - a_{23}) + 2 \gamma_3) + \gamma_6(\Gamma + \gamma_6))/D \\
n_4 &= \gamma_4 W_{34}(n_0^0 a_{23} W_{12} \Gamma \\
    &\quad + n_1^0 W_{12}(\Gamma(1 - a_{23}) + 2 \gamma_3) + \gamma_6(\Gamma + \gamma_6))/D \\
D &= \gamma_6(\Gamma + \gamma_6)[W_{34}(\Gamma(1 - a_{43}) + 2 \gamma_2) + \gamma_6(\Gamma + \gamma_6)] \\
    &\quad + W_{12}(\gamma_1(\Gamma + \gamma_1))(\Gamma(1 - a_{23}) + 2 \gamma_3) \\
    &\quad + W_{34}[\Gamma^{2}(1 - a_{23} - a_{43} + a_{23}a_{43} - a_{23}a_{43}) \\
    &\quad + 2 \gamma_3 \Gamma(2 - a_{23} - a_{43}) + 4 \gamma_3^2].
\end{align*}
\]

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**Figure 11.** (a) Atomic cloud expansion versus time (squared units) for (i) $^{172}\text{Yb}$, and (ii) $^{171}\text{Yb}$, after release from the dual-colour MOT. (b) A histogram of temperature measurements for $^{171}\text{Yb}$. (c) $^{171}\text{Yb}$ cloud temperature for a range of frequency instabilities at $\tau = 1$ s. The solid (green) line marks the Doppler cooling limit (for zero intensity).
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