Study of CPO resonances on the intercombination line in $^{173}$Yb

Pushpander Kumar$^1$, Alok K Singh$^{1,3}$, Vineet Bharti$^1$, Vasant Natarajan$^1$ and Kanhaiya Pandey$^2$

$^1$ Department of Physics, Indian Institute of Science, Bangalore-560012, India
$^2$ Department of Physics, Indian Institute of Technology, Guwahati 781039, India

E-mail: vasant@physics.iisc.ernet.in

Received 15 June 2017, revised 11 December 2017
Accepted for publication 19 December 2017
Published 8 January 2018

Abstract

We study coherent population oscillations in an odd isotope of the two-electron atom Yb. The experiments are done using magnetic sublevels of the $F_\text{g} \rightarrow F_\text{e} = 5/2 \rightarrow 3/2$ hyperfine transition in $^{173}$Yb of the $^3S_0 \rightarrow ^3P_1$ intercombination line. The experiments are done both with and without an applied magnetic field. In the absence of an applied field, the complicated sublevel structure along with the saturated fluorescence effect causes the linewidth to be larger than the 190 kHz natural linewidth of the transition. In the presence of a field (of magnitude $330$ mG), a well-defined quantization axis is present which results in the formation of two M-type systems. The total fluorescence is then limited by spin coherence among the ground sublevels. In addition, the pump beam gets detuned from resonance which results in a reduced scattering rate from the $^3P_1$ state. Both of these effects result in a reduction of the linewidth to a subnatural value of about 100 kHz.

Keywords: coherent population oscillations, coherent control, two-electron atom

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

1. Introduction

Coherent population oscillation (CPO) is a phenomenon in which two levels are coupled by two coherent electromagnetic fields of different amplitudes and frequencies [1]. This results in the population difference between the two levels to oscillate periodically at the beat frequency of the two fields. The linewidth of the resulting resonance is limited by the population relaxation rate, and can be subnatural for optical transitions. In recent times, it has received much interest from the quantum optics community [2–4], because narrow CPO resonances can be used for applications in light storage [5, 6] and optical memories [7]. It has other applications similar to the related phenomenon of coherent population trapping (CPT) [8], but is more advantageous because of the following reasons.

(i) There is no third level involved, so it can be observed in a larger class of materials including gases, liquids, and room temperature solids.

(ii) It does not require a two-photon resonance, therefore laser jitter—both frequency and amplitude—becomes irrelevant.

(iii) It offers spectral features that are broad, which implies that the medium will respond to very short optical pulses (of picosecond duration or less), whereas the shortest pulses that can be manipulated by CPT-based schemes are of nanosecond duration.

We have recently reported the first experimental observation of CPT and CPO in the two-electron atom Yb [9], a theoretical model for which was presented in [10]. The study was novel because it was an observation in a V-type system, a system which does not have such resonances in a one-electron atom. We used the intercombination line ($^3S_0 \rightarrow ^3P_1$ transition) at 556 nm and the even isotope $^{174}$Yb. We used a small magnetic field (of order 0.5 G) to shift the CPT resonances away from line center, and retain only the CPO resonances. The linewidth of the central peak was then about 400 kHz, which was a factor of two higher than the natural linewidth of the upper $^3P_1$ state. This was because the peak was due to multiple CPO resonances, each of which had a resonant pump field.

3 Present address: Laboratoire Aimé Cotton, CNRS, Univ. Paris-Sud, ENS Cachan, Bât. 505, F-91405 Orsay, France.
Here, we report a study of CPO resonances in the odd isotope $^{173}\text{Yb}$. The main difference from our work on the even isotope is that there is a single peak with and without a magnetic field. This is because there are only CPO resonances and no CPT resonances. CPT resonances will move in the presence of a magnetic field, because the field will cause the pump beam to become detuned and the resonance will appear at the location where the probe beam has the same detuning. For CPO resonances, such detuning does not cause any shift from line center because it only deals with the two levels being coupled, but will cause a reduction in decoherence due to spontaneous emission from the excited state. The reduced scattering rate will result in a reduction in the linewidth of the CPO peak. The detuning also causes a decrease in the fluorescence signal, and an overall decrease in the signal-to-noise ratio. This explanation is borne out by a detailed density-matrix calculation of the levels involved.

We demonstrate this using the $F_g = 5/2 \rightarrow F_e = 3/2$ hyperfine transition $^{173}\text{Yb}$, but the same trend of linewidth reduction is observed for the other two hyperfine transitions in this isotope. $^{173}\text{Yb}$ was preferred over the other odd isotope $^{171}\text{Yb}$—which has only one transition—for the following reason. It gives us the ability to study transitions where the number of sublevels in the excited state is of three different kinds: smaller ($5/2 \rightarrow 3/2$), equal ($5/2 \rightarrow 5/2$), and larger ($5/2 \rightarrow 7/2$).

This ability to study transitions with different number of sublevels is one of the main differences from the earlier work on CPO resonances done with metastable He in [2]. In addition, those experiments were done with a vapor cell inside a magnetic shield. By contrast, our experiments are conducted with an atomic beam, because a vapor cell gets coated with Yb and becomes opaque due to the high sublimation temperature of Yb (about 500 °C). Therefore, it is practically impossible to make a vapor cell of Yb; hence all our previous Yb work has been done using fluorescence detection from an atomic beam [11, 12]. The theoretical model given in [2] is not applicable to our case mainly because of the large number of levels involved.

2. Experimental details

The experimental setup shown in figure 1 is the same as in our previous work in [9], and is reviewed here for completeness. Briefly, the $^1S_0 \rightarrow ^3P_1$ transition at 556 nm is generated by doubling the output of a fiber laser with linewidth of 70 kHz. The doubling is done in a commercial external cavity doubler using potassium niobate as the nonlinear crystal. As seen in the figure, part of the output beam from the doubler is used to lock the laser to the correct hyperfine transition. To achieve this, the laser beam is sent perpendicular to the atomic beam, and the resulting fluorescence is collected using a photomultiplier tube (PMT). The frequency dithered for generating the error signal is obtained by frequency modulating the acousto-optic modulator (AOM) in the path of the laser beam.

The remaining part of the laser beam is used for the experiment. It is divided into two parts—one as a pump beam and the other as a probe beam—with orthogonal linear polarizations using a polarizing beam splitter (PBS). The pump beam is fixed in frequency, and is resonant with the transition used for locking the laser. This is achieved by using a second AOM with an identical shift (of +100 MHz) as the one used for laser locking. The probe beam is scanned around this transition by double passing a third AOM at 50 ± 2 MHz. The double passing ensures directional stability as the AOM is scanned, and the variable shift of ±2 MHz allows the probe beam to be scanned by ±4 MHz around the resonance. The two beams are combined on another PBS, and then transported to the experimental chamber using a polarization maintaining fiber. The use of the fiber ensures perfect overlap of the two beams as the probe beam is scanned. The mode coming out of the fiber is perfectly Gaussian. The power in each beam is 40 µW and its 1/e² diameter is 5 mm, which gives a maximum Rabi frequency of 1.2 Γ at beam center. The output of the fiber is sent perpendicular to the atomic beam, and the resulting fluorescence signal is detected using a second identical PMT.

The atomic beam used both for laser locking and the experiment is contained in an ultra high vacuum (UHV) chamber. The beam is generated by resistively heating a quartz ampule containing an ingot of unenriched Yb to a temperature of about 400 °C. The vacuum chamber is maintained at a pressure below $10^{-8}$ torr using a 201 s⁻¹ ion pump.

The magnetic field required for the experiment is generated by winding three pairs of Helmholtz coils in three orthogonal directions on the outside of the UHV chamber. The value of the $B$ field was measured with a three-axis fluxgate magnetometer. The first set of experiments was done with nominally zero field. However, the presence of stray fields in the lab, coupled with the fact that no magnetic shield was used, meant that there was always a small non-zero field. The second set of experiments was done with a finite longitudinal field, which was generated by increasing the current in the pair of coils that is coaxial with the laser beam. The value of 330 mG is the same as the one used in our even
Transitions between sublevels coupled by the pump and probe fields are shown in figure 2. The first thing to notice is that there is a single peak, both with and without an applied $B$ field. The linewidth of the peak without a field—but with a resonant pump beam—is 270 kHz. This is slightly larger than the natural linewidth of the intercombination line of 190 kHz (determined by the lifetime of the $^3P_1$ state), and arises both because of multiple CPO resonances contributing to the peak and a saturated fluorescence effect. The saturated fluorescence effect is a two-level process in which there is a dip in the fluorescence at zero detuning. This has been previously observed in the intercombination line in Sr [13]. In the presence of a $B$ field of 330 mG, the linewidth reduces to a subnatural value of 100 kHz. This is primarily because of reduced spontaneous emission from the upper state—the $B$ field causes the pump beam to be detuned—as shown in our work on CPT resonances [14]. While this is the main effect, other mechanisms limiting the linewidth include the ground-state decoherence rate, power broadening, and transit-time broadening. The relative contributions of these factors is considered in detail in the ‘Theoretical analysis’ section.

The same behavior, i.e. a linewidth reduction in the presence of a $B$ field, is seen for the other two hyperfine transitions—$5/2 \rightarrow 5/2$ and $5/2 \rightarrow 7/2$. The results of linewidth measurements for all three cases are shown in figure 3. As seen, the linewidth of the resonance for the no-field case shows a steady increase from the one for the $5/2 \rightarrow 3/2$ transition. This is because the number of sublevels increases steadily, which results in more and more decay channels contributing to the peak.

4. Theoretical analysis

The complete sublevel structure for the $5/2 \rightarrow 3/2$ transition in the presence of a $B$ field is shown in figure 4. The presence of the field defines a quantization axis, which is independent of the direction of light polarization. Therefore, even though the light beams are linearly polarized, we do not have to consider transitions coupled by $\pi$ polarization (selection rule $\Delta m = 0$). Furthermore, the role of $\pi$ polarization has been discussed in detail in our earlier work on the even isotope [9]. It is also reasonable to consider only transitions in the presence of a $B$ field because, as mentioned in the experimental details section, it is practically impossible to nullify stray fields completely.

Each excited state sublevels shifts by an amount proportional to $\Delta B$, which is 139 kHz for our experimental field of 330 mG. The pump and probe beams are linearly polarized. This means that they decompose into equal amounts of right circular ($\sigma^+$) and left circular ($\sigma^-$) polarizations. $\sigma^+$ couples transitions with the selection rule $\Delta m = +1$ and $\sigma^-$ couples with selection rule $\Delta m = -1$.

Transitions between sublevels coupled by the pump and probe beams depicted in figure 4 show that the system separates into two M-type subsystems. This shows that no
CPT-like resonances are formed, because CPT requires a simple three level \( \Lambda \)-type system.

In order to see this theoretically, we do a density-matrix analysis of this system. To enable this we depict the two types of transition in the figure with different colors. The first one (shown in blue) involves the ground sublevels \( m_g = -5/2, -1/2, \) and \(+3/2\). The second one (shown in red) involves sublevels \( m_g = -3/2, +1/2, \) and \(+5/2\). For ease of writing the Hamiltonian, we define the levels in the first system from \([1]\) to \([5]\). The Rabi frequency of the probe beam driving the \([|i\rangle \rightarrow |j\rangle\) transition is \( \Omega_{ij}^p \) while its detuning is \( \delta_{ij}^p \). The corresponding parameters for the pump beam are \( \Omega_{ij}^p \) and \( \delta_{ij}^p \). The Rabi frequencies of the pump and probe beams are weighted by the respective Clebsch–Gordan coefficients. In the rotating wave approximation, the Hamiltonian is

\[
H = 0|1\rangle\langle 1| + \hbar \Omega_{12}^p e^{-i\delta_{12}^p t}|1\rangle\langle 2| + \hbar \Omega_{23}^p e^{-i\delta_{23}^p t}|2\rangle\langle 3| + \hbar \Omega_{34}^p e^{-i\delta_{34}^p t}|3\rangle\langle 4| + \hbar \Omega_{45}^p e^{-i\delta_{45}^p t}|4\rangle\langle 5| + \hbar \Omega_{51}^p e^{-i\delta_{51}^p t}|5\rangle\langle 1|
\]

As seen from the figure, the probe detunings are such that

\[
\delta_{12}^p = \delta_{23}^p = 0 \quad \text{and} \quad \delta_{34}^p = \delta_{45}^p = 0
\]

since there is no splitting of the ground state. The equation of motion of the density matrix \( \rho \) is given by

\[
\dot{\rho} = -i\hbar [H, \rho] - \frac{1}{2} \{\Gamma, \rho\},
\]

where \( \Gamma \) is the decay matrix. Its diagonal terms give the total decay rate (radiative and non-radiative) of the respective population terms. The off-diagonal terms represent the decoherence rate between states \([i]\) and \([j]\), such that

\[
\Gamma_{ij} = \frac{\Gamma_{ij}^a + \Gamma_{ij}^b}{2}.
\]

\( \Gamma_{ij} \) between two ground states is the spin relaxation rate, and is taken to be \( 2\pi \times 10 \text{ kHz} \).

The solution of equation (3) will be time dependent because the Hamiltonian is time dependent. Hence the solution can be written in the form

\[
\rho_{ij} = \rho_{ij}^0 + \rho_{ij}^{+1}e^{i\delta_{ij}^p t} + \rho_{ij}^{+2}e^{2i\delta_{ij}^p t} + \ldots
\]

\( \rho^0 \)—the DC component of \( \rho \) —is found by taking the Fourier transform of the numerical solution of \( \rho \), which includes all orders in the expansion.

The fluorescence signal obtained in the experiment is proportional to the population in the upper levels, i.e. \( \rho_{22} + \rho_{44} \). From the time dependence of this quantity shown in figure 5, we see that it oscillates around a mean value after a few lifetimes of the excited state. Thus, the system reaches ‘steady state’ after this time.

The other M-type system is analyzed in a similar manner. For clarity, it is denoted with prime symbols in figure 4. The ‘steady state’ populations in the upper levels for the two M-type systems, i.e. \( \rho_{22}^f + \rho_{44}^f \), are shown in figure 6. The main thing to notice is that there is only one peak at line center, exactly as seen in the experimental spectrum. This is different from the spectrum for the even isotope, shown in [9], where four shifted peaks corresponding to CPT resonances were seen in the presence of a B field. The linewidth of the peak is smaller than the natural linewidth because detuning will cause reduced decoherence from the upper state. But this comes at the price of reduced fluorescence signal, something which is also observed experimentally.

We have considered other factors limiting the linewidth. Ground state decoherence rate plays a negligible role. Transient broadening [15] contributes to less than 20%. Power broadening [16] contributes to less than 50%. Therefore, reduced scattering from the excited state remains the dominant factor.

The theoretical model presented above reproduces the fact that the experimentally observed linewidth is subnatural. But it does not give the exact linewidth observed, probably because it assumes a constant Rabi frequency across the laser beam while in reality it is a Gaussian distribution.

5. Conclusions

In summary, we have studied CPO resonances in the odd isotope \(^{173}\text{Yb}\). The magnetic sublevel structure of the
The intercombination line used in this study is such that the peak at line center does not split into multiple peaks in the presence of a $B$ field, which is quite different from the even isotope used in our previous work [9]. The effect of a $B$ field is to cause the pump beam to be detuned from resonance, which results in a reduced scattering rate from the excited state of the transition. The effect of this on the CPO resonance peak is to reduce its linewidth. The linewidth reduces to a subnatural value of 100 kHz with a magnetic field of 330 mG. Such features could lead to new mechanisms for sub-Doppler cooling in divalent atoms compared to the more commonly used monovalent alkali-metal atoms [17].

Acknowledgments

This work was supported by the Department of Science and Technology, India, PK and AKS acknowledge financial support from the Council of Scientific and Industrial Research, India; and VB from a D S Kothari post-doctoral fellowship of the University Grants Commission, India. The authors thank S Raghuvveer for help with manuscript preparation.

ORCID iDs

Vineet Bharti @ https://orcid.org/0000-0001-7927-3025
Vasant Natarajan @ https://orcid.org/0000-0002-1456-665X

References

[1] Bigelow M S, Lepeshkin N N and Boyd R W 2003 Phys. Rev. Lett. 90 113903
[2] Lauprître T, Kumar S, Berger P, Faoro R, Ghosh R, Bretenaker F and Goldfarb F 2012 Phys. Rev. A 85 051805
[3] Kapale K T, Lam W K and Agarwal G S 2013 The Rochester Conf. on Coherence and Quantum Optics and the Quantum Information and Measurement Meeting (Optical Society of America) p M6.35
[4] Maynard M-A, Bretenaker F and Goldfarb F 2014 Phys. Rev. A 90 061801
[5] de Almeida A J F, Sales J, Maynard M-A, Lauprître T, Bretenaker F, Felinto D, Goldfarb F and Tabosa J W R 2014 Phys. Rev. A 90 043803
[6] Neveu P, Maynard M-A, Bouchez R, Lugani J, Ghosh R, Bretenaker F, Goldfarb F and Brion E 2017 Phys. Rev. Lett. 118 073605
[7] Eilam A, Azuri I, Sharypov A V, Wilson-Gordon A D and Friedmann H 2010 Opt. Lett. 35 772
[8] Arimondo E 1996 Prog. Opt. 35 257
[9] Singh A K and Natarajan V 2015 New J. Phys. 17 033044
[10] Vafafard A, Mahmoudi M and Agarwal G S 2016 Phys. Rev. A 93 033848
[11] Pandey K, Singh A K, Kumar P V K, Suryanarayana M V and Natarajan V 2009 Phys. Rev. A 80 022518
[12] Singh A K, Angom D and Natarajan V 2013 Phys. Rev. A 87 012512
[13] Yang T, Pandey K, Pramod M S, Leroux F, Kwong C C, Hajiyev E, Chia Z Y, Fang B and Wilkowski D 2015 Eur. Phys. J. D 69 226
[14] Khan S, Kumar M P, Bharti V and Natarajan V 2017 Eur. Phys. J. D 71 38
[15] Thomas J E and Quivors W W 1980 Phys. Rev. A 22 2115
[16] Mallick N S, Dey T N and Pandey K 2017 J. Phys. B: At. Mol. Opt. Phys. 50 195502
[17] McFerran J J 2016 J. Opt. Soc. Am. B 33 1278