Resolving closely spaced levels for Doppler mismatched double resonance

Elijah Ogaro Nyakang’o and Kanhaiya Pandey

Department of Physics, Indian Institute of Technology Guwahati, Guwahati, Assam 781039, India

(Dated: January 21, 2021)

In this paper, we present experimental techniques to resolve the closely spaced hyperfine levels of a weak transition by eliminating the residual/partial two-photon Doppler broadening and cross-over resonances in a wavelength mismatched double resonance spectroscopy. The elimination of the partial Doppler broadening is based on velocity induced population oscillation (VIPO) and velocity selective saturation (VSS) effect followed by the subtraction of the broad background of the two-photon spectrum. Since the VIPO and VSS effect are the phenomena for near zero velocity group atoms, the subtraction gives rise to Doppler-free peaks and the closely spaced hyperfine levels of the 6P3/2 state in Rb are well resolved. The double resonance experiment is conducted on 5S1/2 → 5P3/2 strong transition (at 780 nm) and 5S1/2 → 6P3/2 weak transition (at 420 nm) at room temperature.

PACS numbers:

I. INTRODUCTION

Saturated absorption spectroscopy [1] is a commonly used technique in the field of laser spectroscopy to overcome the Doppler broadening effect by canceling it in the counter-propagation configuration of the probe and pump lasers. However, the drawback of this technique is the formation of spurious (or cross-over) resonance peaks within the spectrum peaks, which swamps the real resonance peaks if the levels are closely spaced within the Doppler profile.

Further, the cancellation of the Doppler effect for two-photon (or multi-photon) processes such as electromagnetically induced transparency (EIT) [2, 3] requires appropriate lasers propagation direction. However, this cancellation is only possible if the wavelength of the lasers is approximately the same [4] otherwise suffers through partial Doppler broadening due to wavelengths mismatch of the transitions involved [7–10]. Recently this mismatch has been recovered using velocity dependent light shift for detuned control laser and with an extra dressing laser [11, 12].

In this work, we eliminate both of these problems, i.e. (i) the cross-over peaks formed within the spectrum peaks and (ii) wavelength mismatched partial Doppler broadening, for double resonance at 780 nm and 420 nm of a V-type system to resolve closely spaced hyperfine broadening, for double resonance at 780 nm and 420 nm peaks and (ii) wavelength mismatched partial Doppler broadening due to wavelengths mismatch of the transitions involved [11, 12].

The method used to overcome the above mentioned two problems are velocity induced population oscillation and velocity selective saturation (VSS) effects. In the atomic frame for the moving atoms, the two counter-propagating lasers with the same polarization and driving the same transition, will be beating due to opposite Doppler shift. The beating of the two lasers causes a temporal modulation of population difference between the levels driven by the lasers and the phenomenon is called population oscillation [23–29]. Since the two beating fields have same frequency, the induced population oscillation is dependent on the velocity of the atom and hence the name velocity induced population oscillation (VIPO) [30]. The VIPO effect occurs only for a narrow range of beat frequencies (i.e. near zero velocity range) because of the inherent population inertia i.e. the slow response of electric dipoles to incident fields. The range of beat frequencies is determined by the inverse of population relaxation times of the upper levels [29, 31]. Similarly, VSS effect is also for near zero velocity group atom and hence the effect of partial Doppler broadening and cross-over peaks is removed for multi-photon resonance.

This paper is organized in the following way. In section II we describe the relevant energy levels with the transitions of the various configurations and the experimental setup. In section III we describe the density matrix formalism for the various systems considered and the numerically simulated absorption profile of the probe. In section IV we present the experimental results on resolving the closely spaced hyperfine levels of the 6P3/2 state in 85Rb and 87Rb. Finally in section V we give the conclusion on this work.

---

*Electronic address: eogaro@gmail.com
†Electronic address: kanhaiyapandey@iitg.ac.in
II. ENERGY LEVELS AND EXPERIMENTAL SET UP

The relevant energy levels and transitions is illustrated in Fig. 1 and 2 for the V-type system and optical pumping system respectively. The propagation direction of the probe and the pump (or control) lasers at 780 nm (IR) and 420 nm (blue) transitions in various configurations is shown below the energy level scheme.

The probe and the counter-propagating pump lasers at 780 nm are locked to resonance on $5S_{1/2}(F = 3) \leftrightarrow 5P_{3/2}(F = 4)$ transition. The lifetime, $\tau_1$ of the state, $5P_{3/2}(F = 4)$ is 26.25 ns [33]. The absorption of the probe is monitored as the 420 nm pump laser scans across the $6P_{3/2}$ hyperfine levels on $5S_{1/2}(F = 3) \leftrightarrow 6P_{3/2}$ weak transition for a V-type system or on $5S_{1/2}(F = 2) \leftrightarrow 6P_{3/2}$ weak transition in the case of optical pumping system. The lifetime, $\tau_2$ of $6P_{3/2}$ is 120.7 ns [35].

The 780 nm laser beam is generated from the throrlab laser diode L785H1 which is a home-assembled extended cavity diode laser (ECDL) with typical linewidth of 500 kHz. This laser is locked to resonance on $5S_{1/2}(F = 3) \leftrightarrow 5P_{3/2}(F = 4)$ transition shown in Figs. 1 and 2 using saturated absorption spectroscopy (SAS) set-up. The error signal for locking the laser is generated by frequency modulation using the current of ECDL at 50 kHz. The recorded experimental spectra is frequency scaled using the resolved peaks location of the green trace (in each of the configurations) for the hyperfine splitting values given in reference [14].

The 420 nm laser beam is generated from a commercially available ECDL from TOPTICA of model no. DL PRO HP with a typical linewidth of $<200$ kHz and output power of 70 mW. A portion of the beam is fed to Fabry-Perot Interferometer for monitoring the single-mode operation of the blue laser. The beam diameter of the 780 nm probe and pump beams is $2\times3$ mm and that of 420 nm pump beams is $3\times4$ mm. The power of the probe beam used in the experiment is $42 \mu W$ (or peak intensity, $I = 1.78 \text{ mW/cm}^2$).

The detailed experimental set up is shown in Fig. 3. In order to extract the narrow linewidth, the probe laser beam is divided into two beams with same polarization and power and propagated in the Rb cell with a spatial separation of about 5 cm. The blue beam is also divided into two beams with the same polarization as the IR beams and co-propagates with the two probes as shown in experimental set-up of Fig. 3. The IR pump beam which counter-propagates with one of the probe beam, has the same polarization as the probe beam since having the same polarization is key for the interference/beating of the two fields. The interference/beating of the fields requires the polarization of the two fields to be identical and this aspect has been verified experimentally by rotating the polarization of one of the fields. When the polarization of the two fields are orthogonal, the VIPO dip disappear. There is a retro-mirror for reflecting the blue beam (which is overlapping with the IR pump beam) to generate counter-propagating blue beams in the cell when shutter 2 is open. It is very important to keep the angle between the beams as minimum as possible (i.e. near zero angle) and also use a magnetic shield to minimize broadening of the spectrum.

There are three shutters which are used to generate various conditions and configurations in the experiment. The configuration represented by Fig. 1 or 2a is generated with all the shutters closed. The configuration represented by Fig. 1 or 2b is generated with shutter 1 open and shutter 2 closed. The configuration represented by Fig. 1 or 2c is generated with shutter 1 and shutter 2 open. Opening the shutter 3 removes the broad background of the transparency and EA peaks. The broad background is removed by the subtraction of the absorption/transparency spectra of the two probes using two
III. THEORETICAL MODEL

We have conducted the experiments in the six configurations shown in Fig. 1 and 2, three of them are for the V-type open system (Fig. 1a, 1b and 1c); and the other three are for optical pumping system (Fig. 2a, 2b and 2c). The V-type open system is further sub-categorized into: (i) V-type open system shown in 1a, (ii) VIPO at IR transition for V-type open system shown in Fig. 1b and (iii) VIPO at IR and VSS at blue transition for V-type open system shown in Fig. 1c. We discuss the theory for all these configurations one by one.

A. Transparency for V-type open system

1. V-type open system

This corresponds to the energy level and the configuration shown in Fig. 1a and is achieved by closing all the shutters of the experimental setup in Fig. 3. This system is very well known and has been extensively studied [8, 30]. This V-type of system is open as the population from 6P\(_{3/2}\) decays to the other ground state hyperfine level, 5S\(_{1/2}\)(F = 2) and can not be recycled. In the presence of the blue pump laser, c\(_2\), there is transparency of the IR probe laser due to two effects, one is coherence effect i.e. EIT in a V-type atomic system [37, 38] and the other is optical pumping effect [39–41]. The Hamiltonian of the system, the equations of motion and the analytical expression for the absorption of the probe, \(\rho_{12}\), are given in Eq. A1, A2 and A3 respectively.

The mixing rate, \(\Pi_p\), for the hyperfine ground states (appearing in the equations of motion) is due to thermal collisions and the time of flight of atoms across the laser beam [3, 42]. The contribution due to time of flight is defined as \(\tilde{v}\) where, \(\tilde{v}\) is the thermal velocity of the atoms in the atomic medium and \(d\) is the diameter of the laser beam. The numerically simulated absorption spectrum of the IR probe laser locked to resonance on 5S\(_{1/2}\)(F = 3) ↔ 5P\(_{3/2}\)(F = 4) cycling transition vs detuning of the blue pump laser is plotted in Fig. 4 (see the blue trace). The Lorentzian fitting to this curve gives a linewidth of 16 MHz, while the linewidth is 11 MHz if the pump laser wavelength is taken to be 780 nm instead of 420 nm (see the table I). This broadening by 1.5 times is due to residual or partial Doppler broadening caused by wavelength mismatch between the probe and the pump laser.

2. VIPO at IR transition for V-type open system

This configuration corresponds to the energy scheme given in Fig. 1b and the experimental set-up when shutter 1 is open and shutter 2 is closed. This is theoretically modeled by considering the Hamiltonian \(H\) under electric-dipole and rotating-wave approximation and in the interaction picture as follows,

\[
H = \hbar \left\{ (\Omega_{c1} + \Omega_pe^{i\delta_1 t})|1\rangle\langle 2| + \Omega_{c2}|1\rangle\langle 3| - \Delta_{c1}|2\rangle\langle 2| - \Delta_{c2}|3\rangle\langle 3| + h.c. \right\}
\]  

where, 5S\(_{1/2}\)(F = 3) = |1\>, 5P\(_{3/2}\)(F = 4) = |2\>, 6P\(_{3/2}\)(F = 2) = |3\>, \(\delta_1 = (\omega_p - k_1v) - (\omega_{c1} + k_1v) = -2k_1v\) is the frequency difference between IR probe and pump beams in the atomic frame (since \(\omega_p = \omega_{c1}\)), \(k_1 = 2\pi/\lambda_1\) is the wave-vector of the IR laser and \(\lambda_1\) is the wavelength, \(v\) is the velocity of the atom in the direction of the probe, \(\Delta_{c1} = \omega_{c1} - (\omega_p - \omega_1) + k_1v\) is the detuning of the IR control laser, \(\Delta_{c2} = \omega_{c2} - (\omega_3 - \omega_1) - k_2v\) is

![Figure 3: (Color online). The experimental setup for resolving the closely spaced hyperfine levels of the 6P\(_{3/2}\) state in Rb atom using the VIPO and VSS effects.](image)
the detuning of the blue laser, \( k_2 = 2\pi/\lambda_2 \) is the wave-vector of the blue laser and \( \lambda_2 \) is the wavelength. The Rabi frequency for the fields is \( \Omega_i = -d_{ij}E_i/h \) where, \( d_{ij} = \langle i|\hat{d}|j\rangle \) is the dipole matrix element, \( \hat{d} \) is the atomic dipole operator and subscript \( L = p, c1, c2 \) represent the fields (i.e. \( p \) is the probe and \( c1 \) is the pump of the 780 nm laser and \( c2 \) is the pump of the 420 nm laser).

The atom-field interaction is described by writing the Liouville-von Neumann equation for the density matrix,

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

where, \( \rho \) is the atomic density operator, \( \Gamma \) is the relaxation operator defined as \( \langle i|\Gamma|j\rangle = \gamma_i\delta_{ij} \) (\( \delta_{ij} = 1 \) if \( i = j \) and 0 if \( i \neq j \) and \( \gamma_i \) is the decay rate of state \( |i\rangle \). The temporal behavior of the element of density matrix governed by Eq. 2 is velocity dependent due to the Doppler effect and oscillates at the harmonics of the beat frequency \( \Delta_1 = -2k_1v \). The oscillation is due to the beating of the two fields addressing the same transition \( 5S_{1/2}(F = 3) \leftrightarrow 5P_{3/2}(F = 4) \) in Fig. 1c. The equations of motion of the density matrix elements is given in Eq. [A4] and is obtained using Eq. [1] and 2. The harmonically oscillating density matrix elements at beat frequency can be written in the Floquet expansion \( \rho^{(n)}_{ij} \) in the following form

\[
\rho_{ij}(t) = \sum_{n=-\infty}^{\infty} \rho^{(n)}_{ij}(t)e^{i\delta_1 t}
\]

where, \( \rho^{(n)}_{ij}(t) \) are \( n \)th harmonic amplitudes of the density matrix elements. The imaginary part of the zeroth harmonic, \( \rho^{(0)}_{ij} \) corresponds to the IR pump absorption, while the imaginary part of the first harmonic, \( \rho^{(+1)}_{ij} \) is for IR probe absorption in first order and all the others are for wave-mixing \([16]\). In the steady state condition \( \rho^{(n)}_{ij} = 0 \) for all \( n, i \) and \( j \), the absorption of the probe laser \( \rho^{(+1)}_{ij} \) is obtained by substituting the truncated series of the Floquet expansion given in Eq. 3 up to first order into Eq. [A4]. The coefficients of the same power in \( n\delta_1 \) are then compared which yields a set of steady state equations of motion in the Floquet expansion. The \( \rho^{(+1)}_{12} \) element of the density matrix is expressed as follows,

\[
\rho^{(+1)}_{12} = \frac{\Omega_p}{2(\gamma_{12} + i\delta_1)}(\rho_{11}^{(0)} - \rho_{22}^{(0)}) + \frac{i\Omega_{c1}}{2(\gamma_{12} + i\delta_1)}(\rho_{11}^{(+1)} - \rho_{22}^{(+1)}) - \frac{i\Omega_{c2}}{2(\gamma_{12} + i\delta_1)}\rho_{32}^{(+1)}
\]

where, \( \gamma_{12} = i\Delta_p + \gamma_{dec}, \Delta_p = \Delta_{c1} = 0, \gamma_{ij} = \frac{1}{2}(\Gamma_i + \Gamma_j) \) and \( \Gamma_i \) is the decay rate of the \( i \)th level. The quantity \( (\rho_{11}^{(0)} - \rho_{22}^{(0)}) \) in term I is the population inversion created by the pump lasers at IR and blue transition. The quantity \( (\rho_{11}^{(+1)} - \rho_{22}^{(+1)}) \) in term II is the population oscillation difference and its contribution is significant for the velocity group atoms in the range of \( v | \sim \Gamma_p/k_1 \) and forms a dip inside the transparency window. The density matrix element \( \rho_{32}^{(+1)} \) in term III is the coherence oscillation which further modifies the lineshape of the dip inside the transparency window. The role of individual terms for the probe absorption is shown in Fig. A2.

The absorption of the probe laser is obtained by thermal averaging of Eq. 4 at room temperature as follows,

\[
\overline{\rho_{12}} = \frac{1}{\nu} \int \rho_{12} e^{-\frac{\nu t}{k_B T/m}} d\nu,
\]

where, \( \nu = \sqrt{k_BT/m} \) is the atomic mass and \( T (= 300 \text{ K}) \) is the temperature. The lineshape of the probe absorption after thermal averaging is shown in Fig. 4 (see the red trace marked by circles). The linewidth of the dip inside the transparency window is around 7 MHz which is less than the linewidth for a V system if the pump laser had wavelength at 780 nm instead of 420 nm. The linewidth of the dip is determined by fitting with a Gaussian line-profile (which fits better than a Lorentzian line-profile). The FWHM of a Gaussian fit (i.e. \( \sigma = \frac{\sqrt{2\ln 2}}{\pi} \)), is \( 2\sqrt{2\ln 2}\sigma \) where \( \Lambda, x_0 \) and \( \sigma \) are the fitting parameters and \( x \) is the frequency detuning.

3. VIVO at IR and VSS at blue transition for V-type open system

The energy scheme for this configuration is given in Fig. 1 where the probe and IR pump are similarly locked to resonance on \( 3S_{1/2}(F = 3) \leftrightarrow 5P_{3/2}(F = 4) \) cycling.
transition. The blue pump scans across the hyperfine levels of the $6P_{3/2}$ at the weak transition $5S_{1/2}(F = 3) \leftrightarrow 6P_{3/2}$ and is retro-reflected by mirror M to generate the two counter-propagating beams inside the Rb vapor cell. The VIPO on $5S_{1/2}(F = 3) \leftrightarrow 5P_{3/2}(F = 4)$ transition will induce a dip on the transparency peak as previously explained in the Sec. 3. This dip is further enhanced by the VSS effect of the two counter-propagating blue pump laser beams.

The VSS effect can be understood in the following simple way. We consider population dynamics between the two states, $|1\rangle$ ($5S_{1/2}, F=2$) and $|3\rangle$ ($6P_{3/2}$) due to two counter-propagating blue pump laser beams only in the absence of the IR laser. For simplicity, consider three velocity group of atoms, $\pm v$, 0 and $-v$. For detuned case of the blue pump laser, $(\Delta k_2)$ both the non-zero velocity group of atom $\pm v = \Delta c_2/k_2$ will be in resonance with either of the two counter-propagating blue pump laser and hence the number of atoms in the excited state will be twice. For zero detuning case, the near-zero ($< \Gamma_3/k_2$) velocity group of atom will be in resonance with both the blue pump laser and hence intensity seen by this group of atoms will be twice. However, the excited state population will be less than twice due to saturation effect, thus inducing a dip on the absorption spectrum of the probe beam with the scan of the blue pump laser. The linewidth of this dip is in the range of $\Gamma_3$. This qualitative picture is also presented in Table I. Mathematically, the population transfer due to blue pump lasers will be given by the following equation \[23\].

$$\rho_{33} = \frac{1}{2} \left\{ \frac{S_{\Delta -2 -k_2 v} + S_{\Delta -2 +k_2 v}}{1 + S_{\Delta -2 -k_2 v} + S_{\Delta c 2 +k_2 v}} \right\}$$

with,

$$S_{\Delta c 2 +k_2 v} = \frac{S_0}{1 + \frac{4(\Delta c_2 +k_2 v)^2}{\Gamma_3^2}}, \quad S_{\Delta -2 -k_2 v} = \frac{S_0}{1 + \frac{4(\Delta -2 -k_2 v)^2}{\Gamma_3^2}}$$

where $S_0(= 2\Omega^2/\Gamma_3)$ is the saturation intensity of the blue transition for the stationary atoms. In the presence of the IR pump laser i.e. when shutter 1 and 2 are open, the VSS effect will induce a dip on both the transparency spectra of both the IR pump and the probe.

The detailed formalism for the VIPO at IR and VSS at blue transitions is as follows. For the given velocity $v$ there is a beating for the two-counter-propagating blue pump lasers in the atomic frame with a beat frequency ($\delta_2 = -2k_2 v$). The Hamiltonian $H$ of a V-type system shown in Fig. 3 under electric-dipole and rotating-wave approximation and in the interaction picture is as follows,

$$H = \frac{\hbar}{2} \left\{ \frac{(\Omega_{c 1} + \Omega_{c 2} e^{i\delta_2 t})|1\rangle\langle 2| + (\Omega_{c 2} + \Omega_{c 2} e^{i\delta_2 t})|1\rangle\langle 3|}{2} - \Delta c_2 |2\rangle - \Delta c_2 |3\rangle \right\} + \text{h.c.}$$

The equations of motion of the density matrix elements is given in Eq. [A3] which is obtained using Eq. 2 and [3]. The coefficients of the harmonically oscillating density matrix elements have two different time dependencies, which is also the case for the Hamiltonian in Eq. [3]. The Floquet expansion for the density matrix elements in such a case is modified and written as follows,

$$\rho_{ij}(t) = \sum_{m=-\infty}^{\infty} \sum_{n=-\infty}^{\infty} \rho_{ij}^{(n,m)}(t) e^{i(n\delta_1 + m\delta_2) t}$$

where, $n$ is the $n^{th}$ harmonic component due the beating of the IR laser beams and $m$ is the $m^{th}$ harmonic component due the beating of the blue pump laser beams.

The imaginary part of $\rho_{12}^{(0,0)}$ corresponds to the IR pump absorption, while the imaginary part of $\rho_{12}^{(+1,0)}$ is for IR probe absorption and all the others are for wave-mixing. In the steady state condition (i.e. $\dot{\rho}_{ij}^{(n,m)} = 0$ for all $n, m, i$ and $j$), $\rho_{12}^{(+1,0)}$ is obtained by substituting the truncated series of the Floquet expansion given in Eq. [4] up to first-order into Eq. [A3]. The coefficients of the same power in $(n\delta_1, m\delta_2)$ are similarly compared and yields a set of steady state equations of motion in the Floquet expansion. The $\rho_{12}^{(+1,0)}$ element of the density matrix is expressed as follows,

$$\rho_{12}^{(+1,0)} = \frac{1}{2(\gamma_{12} + i\delta_1)} \left\{ \frac{i\Omega_{c 1}(\rho_{11}^{(0,0)} - \rho_{22}^{(0,0)})}{2(\gamma_{12} + i\delta_1)} + \frac{i\Omega_{c 1}(\rho_{11}^{(-1,1)} - \rho_{22}^{(-1,1)})}{2(\gamma_{12} + i\delta_1)} \right\}$$

In Eq. 8 the quantity $(\rho_{11}^{(+1,0)} - \rho_{22}^{(+1,0)})$ in term I is the population inversion induced by the IR pump and the saturation of the counter-propagating blue pumps and $(\rho_{11}^{(+1,0)} - \rho_{22}^{(+1,0)})$ in term II is the population oscillation induced by the beating of the IR probe and

### Table I: Comparison of numerically calculated linewidth for various configuration

| System and configuration | Linewidth (MHz) |
|--------------------------|-----------------|
| Configuration as shown in Fig. 1a | 16 |
| Configuration as shown in Fig. 1b but considering pump wavelength 780 nm instead of 420 nm | 11 |
| Configuration as shown in Fig. 2b | 7±1 |
| Configuration as shown in Fig. 2c & 2d but considering pump wavelength 780 nm instead of 420 nm | 6±1 |
| Configuration as shown in Fig. 2b | 17 |
| Configuration as shown in Fig. 2c & 2d but considering pump wavelength 780 nm instead of 420 nm | 12±1 |
| Configuration as shown in Fig. 2c & 2d | 9±1 |
| Configuration as shown in Fig. 2b | 6±1 |
pump laser beams and the saturation of the counterpropagating blue pumps. The density matrix elements, $\rho^{(+1,0)}_{32}$ and $\rho^{(+1,-1)}_{32}$ in term III, are the oscillating coherence terms due to the beating of the fields on IR and blue transitions. The thermal averaged probe absorption, $\frac{1}{2\pi\Delta v} \int \rho_{12}(+1,0) e^{-(\pi^2 t)} dv$ is calculated numerically and is plotted in Fig. 4 (see the green trace marked with dots). The linewidth of the induced dip is around 6 MHz.

B. Enhanced absorption for optical pumping system

1. Optical pumping system

This system corresponds to the energy level and the configuration shown in Fig. 2a and is achieved when all the shutters in the experimental setup of Fig. 3 are closed. Again, the probe laser is locked to resonance on $5S_{1/2}(F = 3) \leftrightarrow 5P_{3/2}(F = 4)$ transition. The absorption of the probe is monitored as the co-propagating blue pump laser scans across the $6P_{3/2}$ hyperfine levels on $5S_{1/2}(F = 2) \leftrightarrow 6P_{3/2}$ transition instead of $5S_{1/2}(F = 3) \leftrightarrow 6P_{3/2}$ transition. The absorption of the probe is increased by optical pumping of population to the upper ground hyperfine level $5S_{1/2}(F = 3)$ via $5S_{1/2}(F = 2) \rightarrow 6P_{3/2}(F = 1, 2, 3)$ excitation and various decay channels (i.e. direct, $6P_{3/2}(F = 2, 3) \rightarrow 5S_{1/2}(F = 3)$ and indirect decay channels [47-48] such as $6P_{3/2}(F = 1) \rightarrow 6S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5S_{1/2}(F = 3)$). Therefore, optical pumping gives rise to enhanced absorption (EA) Doppler-free peaks of the $6P_{3/2}$ hyperfine levels. The numerically simulated absorption spectrum considering only one hyperfine level is plotted in Fig. 5 (see the blue trace). Note that the Hamiltonian of the system, equations of motion and the analytical expression for the absorption of the probe, $\rho_{12}$, are given in Eq. (11) and (12) respectively. The Lorentzian fitting to this curve gives a linewidth of 17 MHz, while it is 11 MHz if we consider the pump laser wavelength to be 780 nm instead of 420 nm (see the table). This broadening by 1.5 times is again due to residual or partial Doppler broadening caused by wavelength mismatch between the probe and the pump laser.

2. VIPO at IR transition for optical pumping system

This corresponds to the energy level and the configuration shown in Fig. 2a and is achieved when shutter 2 is closed in the experimental setup of Fig. 3. This is theoretically modeled by considering the Hamiltonian $H$ of the optical pumping system shown in Fig. 2b under electric-dipole and rotating-wave approximation and in the interaction picture as follows,

$$H = \frac{\hbar}{2} \{ (\Omega_{c1} + \Omega_pe^{i\delta t})|1\rangle\langle 2| + \Omega_{c2} e^{i\delta t} |4\rangle\langle 3| - \Delta_{c1}|2\rangle\langle 2| - \Delta_{c2}|3\rangle\langle 3| + h.c. \} \tag{9}$$

where, $5S_{1/2}(F = 3) = |1\rangle$, $5P_{3/2}(F = 4) = |2\rangle$, $6P_{3/2}(F = 1) = |3\rangle$ and $5S_{1/2}(F = 2) = |4\rangle$. The Hamiltonian is time dependent and the equations of motion of the density matrix elements is given in Eq. (B4). The equations of motion are solved in steady state after the Floquet expansion given in Eq. (3) and the imaginary part of the density matrix element $\rho^{(+1)}_{12}$ gives the absorption of the probe as follows,

$$\rho^{(+1)}_{12} = \frac{i\Omega_p}{2(\gamma_{12} + i\delta_1)} (\rho^{(0)}_{11} - \rho^{(0)}_{22}) + \frac{i\Omega_{c1}}{2(\gamma_{12} + i\delta_1)} (\rho^{(+1)}_{11} - \rho^{(+1)}_{22})$$ \tag{10}

The Eq. (10) is similar to the Eq. (4) except the coherence term. The first term, I in Eq. (10) is due to population inversion created by the pump laser at IR and blue transition and gives only the EA line-shape. The second term is due to VIPO at IR transition and gives a dip inside the EA spectrum as shown Fig. 5 (see the red trace marked by circles). The linewidth of the dip is 9 MHz using Gaussian line profile fit. The contribution of each of the terms I and II is given in Eq. (B2).

3. VIPO at IR and VSS at blue transition for optical pumping system

The energy levels for this configuration is given in Fig. 2a. The probe and the IR pump lasers are again locked to resonance on $5S_{1/2}(F = 3) \leftrightarrow 5P_{3/2}(F = 4)$ cycling transition. The blue pump laser is scanning across the hyperfine levels of $6P_{3/2}$ at the weak transition, $5S_{1/2}(F = 2) \leftrightarrow 6P_{3/2}$ and is retro-reflected to generate the two counter-propagating beams inside the Rb vapor cell.

The Hamiltonian $H$ of the optical pumping system shown in Fig. 2b under electric-dipole and rotating-wave approximation and in the interaction picture is given as follows,

$$H = \frac{\hbar}{2} \{ (\Omega_{c1} + \Omega_pe^{i\delta t})|1\rangle\langle 2| + (\Omega_{c2} e^{i\delta t} + \Omega_{c2} e^{i\delta t}) |4\rangle\langle 3| - \Delta_{c1}|2\rangle\langle 2| - \Delta_{c2}|3\rangle\langle 3| + h.c. \} \tag{11}$$

The probe absorption is similarly obtained in the steady state condition using the equations of motion given in Eq. (B5) and the Floquet expansion given in Eq. (7). The imaginary part of the density matrix element $\rho^{(+1,0)}_{12}$ in
the Floquet expansion gives the probe absorption and is expressed as follows,

\[
\rho_{12}^{(+1,0)} = \frac{\Omega_c}{(\gamma_2 + i\delta_1)^2} + \frac{i\Omega_c(\rho_{11}^{(+1,0)} - \rho_{22}^{(+1,0)})}{2(\gamma_2 + i\delta_1)^2}
\]

(12)

In Eq. 12, the quantity \((\rho_{11}^{(0,0)} - \rho_{22}^{(0,0)})\) in term I is the population inversion induced by the 780 nm and 420 nm pump lasers. The quantity \((\rho_{11}^{(+1,0)} - \rho_{22}^{(+1,0)})\) in term II is the population oscillation induced by the beating of the 780 nm laser beams and saturation effect induced by the counter-propagating 420 nm pump beams. The thermal averaged absorption in this configuration is shown in Fig. 5 (see the green trace marked with dots). The linewidth of the induced dip on the EA peak is about 6 MHz.

IV. EXPERIMENTAL RESULTS

A. Resolving 6P_{3/2} hyperfine levels in ^85Rb

1. V-type open system

The transparency spectrum of the energy scheme in Fig. 4 is shown by the red dashed trace of Fig. 6. This spectrum is obtained when all the three shutters in the experimental set-up of Fig. 3 are closed. The three peaks of the 6P_{3/2}(F = 2, 3, 4) hyperfine levels are merged forming a broad transparency spectrum due to the residual Doppler broadening effect. When shutter 1 is open, dips corresponding to three hyperfine levels are induced inside the broad transparency peaks caused by VIPO at IR transition (see the blue trace marked with dots in Fig. 6a). However, the dips appear very small due to the broad transparency background. The effect is removed when shutter 3 is open to subtract the broad transparency profile and the spectrum of the resolved hyperfine levels is shown by the green trace of Fig. 6 (see Fig. 6). The line width of the resolved peaks are as follows: F = 4 is 13.3 MHz, F = 3 is 14.1 MHz and F = 2 is 12.1 MHz. The power of the pump beams labeled c1, c2 and c3 used for optimal signal-to-noise ratio of the spectrum are 276.2 \mu W (or peak intensity I = 11.7 mW/cm^2), 5.02 mW (or peak intensity I = 106.5 mW/cm^2) and 3.64 mW (or peak intensity I = 77.2 mW/cm^2) respectively.

Further line narrowing of the resolved peaks is achieved using the configuration shown in Fig. 1c (i.e. VIPO at IR and VSS at blue transition). The energy configuration scheme in Fig. 1c (i.e. VIPO at IR and VSS at blue transition) is implemented in the experimental set-up given in Fig. 3 when shutter 1 and shutter 2 are open. Lower power of IR pump beam is used in this
configuration since the induced dips by VIPO at IR are enhanced by VSS effect at blue transition. The transparency spectrum of this configuration is shown by the blue trace marked with dots in Fig. 6(b). The broad transparency background is removed when shutter 3 is open and the well resolved peaks of the 6P$_3/2$ ($F = 2, 3, 4$) hyperfine levels is shown by the green trace of Fig. 6(b). The linewidth of the resolved peaks are as follows: $F = 4$ is 10.8 MHz, $F = 3$ is 9.1 MHz and $F = 2$ is 11.4 MHz. The power of the pump beams labeled c1, c2 and c3 used for optimal signal-to-noise ratio of the spectrum are 176.4 µW (or peak intensity $I=7.5$ mW/cm$^2$), 6.01 mW (or peak intensity $I=127.5$ mW/cm$^2$) and 8.62 mW (or peak intensity $I=182.9$ mW/cm$^2$) respectively.

In the final result of the resolved peaks (see the green trace of Fig. 6(b)), there are small peaks between the main peaks of $F = 3$ and $F = 4$ and between $F = 2$ and $F = 3$. These are not cross-over peaks (or real peaks), but the residue due to incomplete removal of the broad transparency background in the overlapped regions. The effect also occur for the optical pumping system when the broad absorption background is removed (see the green trace of Fig. 7(b) the small peak between $F = 2$ and $F = 3$).

2. Optical pumping system

The EA spectrum of the optical pumping system is shown by the red dashed trace in Fig. 7. This spectrum is obtained when all the three shutters in the experimental set-up of Fig. 2 are closed. The absorption peaks corresponding to the 6P$_3/2$ ($F = 1, 2, 3$) hyperfine levels are completely merged. The levels 6P$_3/2$ ($F = 2, 3$) are detected by the probe via both the direct decay and indirect decay channels while level 6P$_3/2$ ($F = 1$) is detected via the indirect decay channels to 5S$_1/2$($F = 3$) only. When shutter 1 is open, dips corresponding to the hyperfine levels are induced inside the broad EA peaks due to VIPO at IR transition (see the blue trace marked with dots in Fig. 7). The dips appear small due to broad EA background caused by the residual Doppler broadening effect. The broad EA background is removed when shutter 3 is open and the dips corresponding to the hyperfine levels 6P$_3/2$ ($F = 1, 2$) are still not resolved while the 6P$_3/2$ ($F = 3$) peak is resolved (see the green trace of Fig. 7(b)). The linewidth of the resolved peak is $F = 3$ is 13.9 MHz. The power of the pump beams labeled c1, c2 and c3 used for optimal signal-to-noise ratio of the spectrum are 806.2 µW (or peak intensity $I=34.2$ mW/cm$^2$), 5.01 mW (or peak intensity $I=106.3$ mW/cm$^2$) and 1.84 mW (or peak intensity $I=39.0$ mW/cm$^2$) respectively.

The peaks corresponding to the 6P$_3/2$ ($F = 1, 2, 3$) hyperfine levels, can be completely resolved using the configuration shown in Fig. 2 i.e. VIPO at IR and VSS at blue transition. This configuration is implemented when shutter 1 and shutter 2 are open in the experimental set-up of Fig. 3. The broad EA spectrum is removed when shutter 3 is open and the green trace of Fig. 7(b) shows well resolved peaks of the 6P$_3/2$ ($F = 1, 2, 3$) hyperfine levels. Note, the frequency scaling of the spectra in Fig. 7(b) is assigned using the peak locations of $F = 2$ and $F = 3$ after the complete resolution of all the three peaks of 6P$_3/2$ ($F = 1, 2, 3$) hyperfine levels. The linewidth of the resolved peaks are as follows: $F = 3$ is 9.8 MHz, $F = 2$ is 10.1 MHz and $F = 1$ is 7.2 MHz. The power of the pump beams labeled c1, c2 and c3 used for optimal signal-to-noise ratio of the spectrum are 276.3 µW (or peak intensity $I=34.2$ mW/cm$^2$), 5.01 mW (or peak intensity $I=106.3$ mW/cm$^2$) and 1.84 mW (or peak intensity $I=39.0$ mW/cm$^2$) respectively.

Besides the main peaks due to near zero-velocity group atoms in Fig. 7(b), the extra peaks (or cross-over peaks) formed outside the main spectrum are caused by atoms moving with velocities of 94 ms$^{-1}$ and 143 ms$^{-1}$ respectively. Atoms moving with velocities of 94 ms$^{-1}$ and 143ms$^{-1}$ along the propagation direction of the IR probe, will see the probe laser to be on resonance with the 5S$_1/2$($F = 3$) → 5P$_3/2$($F = 3$) and 5S$_1/2$($F = 3$) → 5P$_3/2$($F = 3$) transitions.
Figure 8: (Color online). The transparency spectrum of the 6P\textsubscript{3/2} hyperfine levels in \textsuperscript{87}Rb recorded for similar configurations of \textsuperscript{85}Rb shown in Fig. 1. The red dashed trace is for the V-type open system (Fig. 1a), the blue trace marked with dots is for V-type open system with VIPO effect at IR and VSS effect at blue transition (Fig. 1c) and the green trace is the final result after removing the broad transparency background and it is magnified 3 times for visibility purpose.

5P\textsubscript{3/2}(F = 2) transitions respectively. The corresponding extra peaks location will be at 224 MHz and 342 MHz from the main peaks. In Fig. 7b, the counter-propagating blue laser beams will form extra peaks on both the left and right side of the main peaks. Ideally the extra peak on the right side of the green spectrum should vanish, but it is still visible due to incomplete subtraction.

B. Resolving 6P\textsubscript{3/2} hyperfine levels in \textsuperscript{87}Rb

1. V-type open system

The 6P\textsubscript{3/2} hyperfine levels of \textsuperscript{87}Rb were also resolved using similar configurations shown in Fig. 1 and 2. The results of VIPO at IR plus VSS at blue transition configuration both in the case of a V-type open system and optical pumping system are reported here. In this configuration, the probe and the counter-propagating pump lasers at 780 nm are locked to resonance on 5S\textsubscript{1/2}(F = 2) \leftrightarrow 5P\textsubscript{3/2}(F = 3) transition. The 420 nm pump laser scans across the 6P\textsubscript{3/2} hyperfine levels on 5S\textsubscript{1/2}(F = 2) \leftrightarrow 6P\textsubscript{3/2} weak transition in the case of a V-type system and on 5S\textsubscript{1/2}(F = 1) \leftrightarrow 6P\textsubscript{3/2} weak transition in the case of optical pumping system.

The transparency spectrum of the configuration given in Fig. 1a for \textsuperscript{87}Rb, is shown by the red dashed trace of Fig. 8 when all the three shutters in the experimental set-up of Fig. 3 are closed. The peaks of the 6P\textsubscript{3/2}(F = 2, 3) hyperfine levels are well resolved but the peaks of 6P\textsubscript{3/2}(F = 1, 2) are partially resolved due to the residual Doppler broadening effect. When shutters 1 and 2 are open, the dips induced by VIPO at IR and VSS at blue transition inside the broad transparency peaks corresponds to the three hyperfine levels of the 6P\textsubscript{3/2}(F = 1, 2, 3) state (see the blue trace marked with dots in Fig. 8). The residual Doppler broadening effect is removed when shutter 3 is open and the spectrum of the resolved hyperfine levels is shown by the green trace of Fig. 8. The linewidth of the resolved peaks are as follows: F = 3 is 14.4 MHz, F = 2 is 15.7 MHz and F = 1 is 15.8 MHz. The power of the pump beams labeled c1, c2 and c3 used for optimal signal-to-noise ratio of the spectrum are 302.6 \mu W (or peak intensity I=12.8 mW/cm\textsuperscript{2}), 4.82 mW (or peak intensity I=102.3 mW/cm\textsuperscript{2}) and 13.2 mW (or peak intensity I=280.1 mW/cm\textsuperscript{2}) respectively.

2. Optical pumping system

The EA spectrum of the optical pumping system is shown by the red dashed trace in Fig. 9 when all the three shutters in the experimental set-up of Fig. 3 are closed. The absorption peaks corresponds to the 6P\textsubscript{3/2}(F = 0, 1, 2) hyperfine levels in \textsuperscript{87}Rb. The peaks for 6P\textsubscript{3/2}(F = 0, 1) are completely merged while the peaks for 6P\textsubscript{3/2}(F = 1, 2) are partially merged. The levels 6P\textsubscript{3/2}(F = 1, 2) are detected by the probe via both the direct decay and indirect decay channels \textsuperscript{35} while level 6P\textsubscript{3/2}(F = 0) is detected via the indirect decay channels to 5S\textsubscript{1/2}(F = 2) only. When shutters 1 and 2 are open, dips corresponding to the hyperfine levels are induced inside the broad EA peaks due to VIPO at IR and VSS at blue transition (see the blue trace marked with dots in Fig. 9). The dips appear small due to broad EA background caused by the residual Doppler broadening effect. The broad EA background is removed when shutter 3 is open and the dips corresponding to the hyperfine levels 6P\textsubscript{3/2}(F = 0, 1, 2) are resolved (see the green trace of Fig. 9). The linewidth of the resolved peaks are as follows: F = 2 is 16.4 MHz, F = 1 is 13.1 MHz and F = 0 is 12.3 MHz. The power of the pump beams labeled c1, c2 and c3 used for optimal signal-to-noise ratio of the spectrum are 823.6 \mu W (or peak intensity I=35.0 mW/cm\textsuperscript{2}), 4.82 mW (or peak intensity I=102.3 mW/cm\textsuperscript{2}) and 15.3 mW (or peak intensity I=325.5 mW/cm\textsuperscript{2}) respectively.

3. Power broadening effect

The contribution of the IR pump power broadening effect to the final result (i.e. the resolved spectrum of the 6P\textsubscript{3/2} state), is illustrated in Fig. 10. The configuration used here is given in Fig. 2b (i.e. VIPO at IR transition) for the case of \textsuperscript{87}Rb. The power of the blue pump laser beams is fixed (i.e. c2 is 4.26 mW and C3 is 3.27 mW) as the the power of IR pump is changed. At 1 mW of the IR pump, all the three peaks corresponding to the 6P\textsubscript{3/2}(F = 0, 1, 2) hyperfine levels are well resolved (see the red trace of Fig. 10b). However, as the IR pump power is increased to 5 mW, the peaks corresponding
Figure 9: (Color online). The EA spectrum of the 6P3/2 hyperfine levels in 87Rb recorded for similar configurations of 85Rb shown in Fig. 2. The red dashed trace is for the optical pumping system (Fig. 2a), the blue trace marked with dots is for the optical pumping system with VIPO effect at IR and VSS effect at blue transition (Fig. 2c) and the green trace is the final result after removing the broad absorption background and it is magnified 3 times for visibility purpose.

to the 6P3/2(F = 0, 1) are completely merged as shown by the cyan trace marked by circles in Fig. 10(b). High intensity of the IR pump broadens the VIPO dips and limits the resolution of the closely spaced hyperfine levels of F = 0 and F = 1 which are 23.739 MHz apart [14]. The frequency scaling of the spectra in Fig. 10 is assigned using the resolved peak locations of F = 1 and F = 2 of the red trace. The variation of the linewidth of the resolved peak corresponding to F = 2 with the IR pump power is shown in Fig. 10(b).

V. CONCLUSIONS

In conclusion we have presented a detailed experimental technique to eliminate the residual (or partial) Doppler broadening in a Doppler mismatched double resonance spectroscopy for a transparency spectrum (or enhanced absorption spectrum). The residual two-photon Doppler broadening is removed using the VIPO at IR transition, VSS at blue transition and the combination of the two effects followed by the subtraction of the broad transparency background or EA background. The technique has been used to resolve the closely spaced hyperfine levels of weak transitions for a Doppler mismatched double resonance at 780 nm and 420 nm in Rb at room temperature.

Acknowledgement

E.O.N. would like to acknowledge Indian Council for Cultural Relations (ICCR) for the PhD scholarship. K.P. would like to acknowledge the funding from SERB of grant No. ECR/2017/000781.

Appendices

Appendix A: Transparency for V-type open system

1. V-type open system

The Hamiltonian $H$ of a V-type open system shown in Fig. 1, under electric-dipole and rotating-wave approximation and in the interaction picture is given as follows,

$$H = \frac{\hbar}{2} \left( \Omega_p |1\rangle \langle 2| + \Omega_{c2} |1\rangle \langle 3| - \Delta_p |2\rangle \langle 2| - \Delta_{c2} |3\rangle \langle 3| + h.c. \right)$$

(A1)

where the levels are 5S1/2(F = 3) = |1\rangle, 5P3/2(F = 4) = |2\rangle, 6P3/2(F = 2) = |3\rangle and 5S1/2(F = 2) = |4\rangle. The equations of motion of the density matrix are obtained
using Eq. 2 and A1 and are given as follows,

\[ \dot{\rho}_{12} = \frac{i\Omega_p}{2}(\rho_{11} - \rho_{22}) - \frac{i\Omega_{c2}}{2}\rho_{32} - \gamma_{12}\rho_{12} \]

\[ \dot{\rho}_{13} = \frac{i\Omega_p}{2}(\rho_{11} - \rho_{33}) - \gamma_{13}\rho_{13} - \frac{i\Omega_{p}}{2}\rho_{23} \]

\[ \dot{\rho}_{14} = -\frac{i\Omega_{c2}}{2}\rho_{34} - \gamma_{14}\rho_{14} - \frac{i\Omega_p}{2}\rho_{24} \]

\[ \dot{\rho}_{22} = \frac{i\Omega_p}{2}\rho_{21} - \frac{i\Omega_{p}}{2}\rho_{12} - \Gamma_2\rho_{22} \]

\[ \dot{\rho}_{23} = -\frac{i\Omega_{p}}{2}\rho_{23} + \frac{i\Omega_{p}}{2}\rho_{13} - \gamma_{23}\rho_{23} \]

\[ \dot{\rho}_{24} = -\frac{i\Omega_{c2}}{2}\rho_{34} - \gamma_{24}\rho_{24} \]

\[ \dot{\rho}_{33} = -\frac{i\Omega_{p}}{2}\rho_{33} + \frac{i\Omega_p}{2}\rho_{13} - \gamma_{34}\rho_{33} \]

\[ \dot{\rho}_{34} = -\frac{i\Omega_2}{2}\rho_{34} - \gamma_{34}\rho_{34} \]

\[ \dot{\rho}_{44} = \Gamma_{34}\rho_{33} + \Pi_2(\rho_{11} - \rho_{44}) \]

where, \( \gamma_{12} = i\Delta_{c1} + \gamma^{dec}_{12} \), \( \gamma_{13} = i\Delta_{c2} + \gamma^{dec}_{13} \), \( \gamma_{14} = \gamma^{dec}_{14} \), \( \gamma_{23} = i(\Delta_{c2} - \Delta_{p}) + \gamma^{dec}_{23} \), \( \gamma_{24} = -i\Delta_{p} + \gamma^{dec}_{24} \), \( \gamma_{34} = -i\Delta_{c2} + \gamma^{dec}_{34} \). The steady state solution of the equations of motion is obtained by substitution of Eq. 1 into Eq. 2.

\[ \rho_{12} = \frac{i\Pi_2\Omega_p(\Gamma_3 + \Pi_2)^2 + 4\Delta_p^2)}{(\Omega_p^2(\Gamma_3 + \Pi_2)(\Gamma_{34} + 3\Pi_2) + 2\Gamma_3\Pi_2((\Gamma_3 + \Pi_2)^2 + 4\Delta_p^2))} \]

The following set of equations of motion are obtained by numerical simulation of the full density matrix given in Eq. 2.

\[ \dot{\rho}_{12} = \frac{i}{2}(\Omega_{c1} + \Omega_{c1}e^{i\delta_1}) (\rho_{11} - \rho_{22}) - \frac{i\Omega_{c2}}{2}\rho_{32} - \gamma_{12}\rho_{12} \]

\[ \dot{\rho}_{13} = \frac{i\Omega_p}{2}(\rho_{11} - \rho_{33}) - \gamma_{13}\rho_{13} - \frac{i}{2}(\Omega_{c1} + \Omega_{c1}e^{i\delta_1})\rho_{23} \]

\[ \dot{\rho}_{14} = -\frac{i\Omega_2}{2}\rho_{34} - \gamma_{14}\rho_{14} - \frac{i}{2}(\Omega_{c1} + \Omega_{c1}e^{i\delta_1})\rho_{24} \]

\[ \dot{\rho}_{22} = \frac{i}{2}(\Omega_{c1} + \Omega_{c1}e^{i\delta_1})\rho_{21} - \frac{i\Omega_{c2}}{2}\rho_{21} - \gamma_{23}\rho_{23} \]

\[ \dot{\rho}_{23} = -\frac{i}{2}(\Omega_{c1} + \Omega_{c1}e^{i\delta_1})\rho_{13} + \frac{i\Omega_{c2}}{2}\rho_{21} - \gamma_{24}\rho_{24} \]

\[ \dot{\rho}_{24} = \frac{i}{2}(\Omega_{c1} + \Omega_{c1}e^{i\delta_1})\rho_{14} - \gamma_{34}\rho_{34} \]

\[ \dot{\rho}_{33} = -\frac{i\Omega_p}{2}\rho_{33} + \frac{i\Omega_{c2}}{2}\rho_{31} - \Gamma_3\rho_{33} \]

\[ \dot{\rho}_{34} = -\frac{i\Omega_p}{2}\rho_{34} - \gamma_{34}\rho_{34} \]

\[ \dot{\rho}_{44} = \Gamma_{34}\rho_{33} + \Pi_2(\rho_{11} - \rho_{44}) \]

The solution of a V-type open system given in Eq. A3 is graphically represented in Fig. A.1 and is well matched with the numerical simulation of the full density matrix given in Eq. A2.

2. VIPO at IR transition for V-type open system

The Hamiltonian of the VIPO at IR transition for a V-type open system shown in Fig. 1, is given in Eq. 1 and the individual contribution of the terms I, II and III is illustrated in Fig. A2.
Appendix B: Enhanced absorption for optical pumping system

1. Optical pumping system

The Hamiltonian $H$ of the optical pumping system consider in Fig. 2h under electric-dipole and rotating-wave approximation and in the interaction picture is given as follows,

$$H = \frac{\hbar}{2}\left\{\Omega_p |1\rangle\langle 2| + \Omega_c |2\rangle\langle 3| - \Delta_p |2\rangle\langle 2| - \Delta_c |3\rangle\langle 3| + h.c.\right\}$$  \hfill (B1)

The equations of motion of the density matrix is obtained from Eq. 2 and B1 and set of equations are given as follows,

$$\dot{\rho}_{12} = i\frac{\Omega_p}{2}(\rho_{11} - \rho_{22}) - \gamma_{12}^{dec}\rho_{12}$$  \hfill (B2)
$$\dot{\rho}_{13} = -i\frac{\Omega_p}{2}\rho_{23} + \frac{i\Omega_c}{2}\rho_{14} - \gamma_{13}\rho_{13}$$
$$\dot{\rho}_{14} = -i\frac{\Omega_p}{2}\rho_{24} + \frac{i\Omega_c}{2}\rho_{13} - \gamma_{14}\rho_{14}$$
$$\dot{\rho}_{22} = -i\frac{\Omega_p}{2}\rho_{12} + \frac{i\Omega_c}{2}\rho_{21} - \Gamma_2\rho_{22}$$
$$\dot{\rho}_{23} = -i\frac{\Omega_p}{2}\rho_{13} - \gamma_2\rho_{23} + \frac{i\Omega_c}{2}\rho_{24}$$
$$\dot{\rho}_{24} = -i\frac{\Omega_p}{2}\rho_{14} - \gamma_2\rho_{24} + \frac{i\Omega_c}{2}\rho_{23}$$
$$\dot{\rho}_{33} = -i\frac{\Omega_c}{2}\rho_{43} + \frac{i\Omega_c}{2}\rho_{34} - \Gamma_3\rho_{33}$$
$$\dot{\rho}_{34} = -i\frac{\Omega_c}{2}\rho_{43} - \gamma_3\rho_{34}$$
$$\dot{\rho}_{44} = -i\frac{\Omega_c}{2}\rho_{34} + \frac{i\Omega_c}{2}\rho_{43} + \Gamma_3\rho_{33} + \Pi_g(\rho_{11} - \rho_{44})$$

where, $\gamma_{12} = i\Delta_p + \gamma_{dec}^p$, $\gamma_{13} = \gamma_{13}^{dec}$, $\gamma_{14} = \gamma_{14}^{dec}$, $\gamma_{23} = -i\Delta_p + \gamma_{23}^{dec}$, $\gamma_{24} = i(\Delta_c - \Delta_p) + \gamma_{24}^{dec}$, $\gamma_{34} = i\Delta_c + \gamma_{34}^{dec}$. The steady state solution of Eq. B2 in the weak probe approximation which gives enhanced absorption spectrum of the probe is expressed as follows,

$$\rho_{12} = \frac{i\Omega_p(\Omega_{c2}^2(\Gamma_3 + \Pi_g)(\Gamma_{31} + \Pi_g) + \Gamma_3\Pi_g(\Gamma_3 + \Pi_g)^2 + 4\Delta_c^2))}{(\Gamma_2 + \Pi_g + 2i\Delta_p)(\Omega_{c2}^2(\Gamma_3 + \Pi_g)(\Gamma_{31} + 3\Pi_g) + 2\Gamma_3\Pi_g(\Gamma_3 + \Pi_g)^2 + 4\Delta_c^2))}$$  \hfill (B3)

The solution of optical pumping system given in Eq. B3 is graphically represented in Fig. B1 and is well matched with the numerical simulation of the full density matrix given in Eq. B2.
2. VIPO at IR transition for optical pumping system

The Hamiltonian of the VIPO at IR transition for the optical pumping system consider in Fig. 2b is given in Eq. \[1\]

The equations of motion of density matrix elements is obtained from Eq. 2 and 9 which gives the following set of equations.

\[
\begin{align*}
\dot{\rho}_{12} &= \frac{i}{2}(\Omega c + \Omega_p e^{i\delta t})\rho_{11} - \gamma_{12} \rho_{12} \\
\dot{\rho}_{13} &= -\frac{i}{2}(\Omega c + \Omega_p e^{i\delta t})\rho_{23} + \frac{i\Omega_p^*}{2}\rho_{14} - \gamma_{13} \rho_{13} \\
\dot{\rho}_{14} &= -\frac{i}{2}(\Omega c + \Omega_p e^{i\delta t})\rho_{24} + \frac{i\Omega_p^*}{2}\rho_{13} - \gamma_{14} \rho_{14} \\
\dot{\rho}_{22} &= -\frac{i}{2}(\Omega c^* + \Omega_p^* e^{-i\delta t})\rho_{12} + \frac{i}{2}(\Omega c + \Omega_p e^{i\delta t})\rho_{21} - \Gamma_2 \rho_{22} \\
\dot{\rho}_{23} &= -\frac{i}{2}(\Omega c^* + \Omega_p^* e^{-i\delta t})\rho_{13} + \gamma_{23} \rho_{23} + \frac{i\Omega_p^*}{2}\rho_{24} \\
\dot{\rho}_{24} &= -\frac{i}{2}(\Omega c^* + \Omega_p^* e^{-i\delta t})\rho_{14} - \gamma_{24} \rho_{24} + \frac{i\Omega_p^*}{2}\rho_{23} \\
\dot{\rho}_{33} &= -\frac{i\Omega_p}{2}\rho_{33} + \frac{i\Omega_p^*}{2}\rho_{34} - \Gamma_3 \rho_{33} \\
\dot{\rho}_{34} &= -\frac{i\Omega_p}{2}(\rho_{33} - \rho_{44}) - \gamma_{34} \rho_{34} \\
\dot{\rho}_{44} &= -\frac{i\Omega_p}{2}\rho_{43} + \frac{i\Omega_p^*}{2}\rho_{43} + \Gamma_3 \rho_{33} + \Pi_p(\rho_{11} - \rho_{44})
\end{align*}
\]

The steady state solution of the equations of motion given in Eq. [B4] is given in Eq. [10] and the solution of the various density matrix components \(\rho_{11}, \rho_{22}, \rho_{11}^{(+1)}\) and \(\rho_{22}^{(+1)}\) is given as follows. The individual contribution of the terms I and II given in Eq. [10] is also illustrated in Fig. B.2.

3. VIPO at IR and VSS at blue transition for optical pumping system

The Hamiltonian of the VIPO at IR transition and VSS at blue transition for the optical pumping system consider in Fig. 2b is given in Eq. [11]

The equations of motion of density matrix is obtained from Eq. 2 and 11 as follows.

\[
\begin{align*}
\dot{\rho}_{12} &= \frac{i}{2}(\Omega c + \Omega_p e^{i\delta t})\rho_{11} - \gamma_{12} \rho_{12} - \gamma_{12c} \rho_{12} \\
\dot{\rho}_{13} &= -\frac{i}{2}(\Omega c + \Omega_p e^{i\delta t})\rho_{23} + \frac{i\Omega_p^*}{2}(1 + e^{-i\delta t})\rho_{14} - \gamma_{13} \rho_{13} \\
\dot{\rho}_{14} &= -\frac{i}{2}(\Omega c + \Omega_p e^{i\delta t})\rho_{24} + \frac{i\Omega_p^*}{2}(1 + e^{-i\delta t})\rho_{13} - \gamma_{14} \rho_{14} \\
\dot{\rho}_{22} &= -\frac{i}{2}(\Omega c^* + \Omega_p^* e^{-i\delta t})\rho_{12} + \frac{i}{2}(\Omega c + \Omega_p e^{i\delta t})\rho_{21} - \Gamma_2 \rho_{22} \\
\dot{\rho}_{23} &= -\frac{i}{2}(\Omega c^* + \Omega_p^* e^{-i\delta t})\rho_{13} + \gamma_{23} \rho_{23} + \frac{i\Omega_p^*}{2}\rho_{24} \\
\dot{\rho}_{24} &= -\frac{i}{2}(\Omega c^* + \Omega_p^* e^{-i\delta t})\rho_{14} - \gamma_{24} \rho_{24} + \frac{i\Omega_p^*}{2}\rho_{23} \\
\dot{\rho}_{33} &= \frac{i\Omega_p}{2}\rho_{33} + \frac{i\Omega_p^*}{2}\rho_{34} - \Gamma_3 \rho_{33} \\
\dot{\rho}_{34} &= \frac{i\Omega_p}{2}(\rho_{33} - \rho_{44}) - \gamma_{34} \rho_{34} \\
\dot{\rho}_{44} &= \frac{i\Omega_p}{2}\rho_{43} + \frac{i\Omega_p^*}{2}\rho_{43} + \Gamma_3 \rho_{33} + \Pi_p(\rho_{11} - \rho_{44})
\end{align*}
\]

\[1\] K.-B. Im, H.-Y. Jung, C.-H. Oh, S.-H. Song, P.-S. Kim, and H.-S. Lee, Phys. Rev. A 63, 034501 (2001), URL https://link.aps.org/doi/10.1103/PhysRevA.63.034501

\[2\] K.-J. Boller, A. Imamoğlu, and S. E. Harris, Phys. Rev. Lett. 66, 2593 (1991), URL https://link.aps.org/doi/10.1103/PhysRevLett.66.2593

\[3\] Y.-q. Li and M. Xiao, Phys. Rev. A 51, 4959 (1995),
[39] M. S. Feld, M. M. Burns, T. U. Kühl, P. G. Pappas, and D. E. Murnick, Opt. Lett. 5, 79 (1980), URL http://ol.osa.org/abstract.cfm?URI=ol-5-2-79

[40] D. A. Smith and I. G. Hughes, American Journal of Physics 72, 631 (2004), https://doi.org/10.1119/1.1652039, URL https://doi.org/10.1119/1.1652039

[41] H. R. Noh, European Journal of Physics 30, 1181 (2009), URL http://stacks.iop.org/0143-0807/30/i=5/a=025

[42] V. Sautenkov, H. Li, Y. Rostovtsev, G. Welch, J. Davis, F. Narducci, and M. Scully, Journal of Modern Optics 56, 975 (2009), https://doi.org/10.1080/09500340902836275, URL https://link.aps.org/doi/10.1080/09500340902836275

[43] J. H. Shirley, Phys. Rev. 138, B979 (1965), URL https://link.aps.org/doi/10.1103/PhysRev.138.B979

[44] Z. Ficek and S. Swain, Quantum interference and coherence: theory and experiments, vol. 100 (Springer Science & Business Media, 2005).

[45] U. D. Giovannini and H. Hübener, Journal of Physics: Materials 3, 012001 (2019), URL https://doi.org/10.1088%2f2053-4461%2fabe3a0

[46] R. W. Boyd, M. G. Raymer, P. Narum, and D. J. Harter, Phys. Rev. A 24, 411 (1981), URL https://link.aps.org/doi/10.1103/PhysRevA.24.411

[47] C. Y. She and J. R. Yu, Appl. Opt. 34, 1063 (1995), URL http://ao.osa.org/abstract.cfm?URI=ao-34-6-1063

[48] H.-R. Noh and H. S. Moon, Phys. Rev. A 85, 033817 (2012), URL https://link.aps.org/doi/10.1103/PhysRevA.85.033817

[49] D. J. Fulton, S. Shepherd, R. R. Moseley, B. D. Sinclair, and M. H. Dunn, Phys. Rev. A 52, 2302 (1995), URL https://link.aps.org/doi/10.1103/PhysRevA.52.2302

[50] V. B. Tiwari, S. Singh, H. S. Rawat, M. P. Singh, and S. C. Mehdendale, Journal of Physics B: Atomic, Molecular and Optical Physics 43, 095503 (2010), URL https://doi.org/10.1088%2f0953-4075%2f43%2f9%2f095503