Velocity-selective resonance dips in the probe absorption spectra of Rb D$_2$ transitions induced by a pump laser.

Keywords: Velocity-selective resonance, hyperfine spectra, Rubidium D-line, control laser, shifting of resonances.

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Abstract

We report experimental observation of velocity-selective resonances in the Doppler-broadened probe absorption spectra of $^{85}\text{Rb}$ and $^{87}\text{Rb}$ D$_2$ transitions in the presence of a strong copropagating pump laser locked to a frequency within the Doppler profile of the transition. The set of three dips having the separation of allowed hyperfine transitions can be moved along the Doppler profile by tuning the pump laser frequency indicating a resonance between the pump laser frequency and the velocity shifted probe laser frequency.
Manipulation of atomic response to a probe radiation field by using an intense pump laser has attracted considerable attention in recent years. Typical atomic systems considered for the purpose are three level atoms in the ladder or Λ configuration. The absorption profile of an atomic transition when the upper level is coupled coherently to a third level by a strong laser field exhibits reduction of absorption [1] leading to electromagnetically induced transparency (EIT). Since the pioneering work on quantum interference leading to zero absorption at the line centre by Harris and co-workers [2] and Agarwal and co-workers [3,4] a large number of theoretical and experimental observations on EIT were reported [5-9]. Most of the experimental work reported observation of EIT in a Λ type system in Rb. These work involved two lower hyperfine levels with a common upper hyperfine level in a D transition. The hyperfine split components of D₁ or D₂ transitions in alkali atoms are embedded in a Doppler broadened Gaussian background that makes it impossible to measure the splitting by using single resonance spectroscopy [10]. In order to recover the hyperfine components from the Doppler background standing wave Lamb dip spectroscopic method is used [10,11]. This method uses two counter propagating laser beams originating from the same source and crossing each other within the gas cell. If one of the beams is stronger than the other then it saturates the transition and creates a hole in the lower level Gaussian population distribution. At resonance the probe absorption shows a dip in the absorption spectra because of the population depletion of the lower level. If there are two closely spaced upper levels, in addition to the two dips at resonance frequencies Ω₁ and Ω₂ an additional "crossover" resonance is observed at a frequency intermediate between the two frequencies i.e. at \(\frac{(Ω₁+Ω₂)}{2}\) [12,13]. Such a resonance occurs when the frequency tuned probe absorption of one beam meets the hole created by the other beam. Hence, in such cases three resonance dips are observed all of which may have similar strength [13]. In the case of hyperfine spectroscopy the atomic energy levels commonly involved are a single ground state and three closely spaced upper level, because of the selection rule \(ΔF = 0, ± 1\). If the lower state hyperfine splitting is much larger the transitions involving different lower levels do not overlap and can be observed as separate transitions, commonly used in Λ type systems. For the three hyperfine transitions Lamb dip spectroscopy reveals three Lamb dips and three crossover resonance dips. The Lamb dips for \(^{85}\text{Rb}\) (F = 2 \(→\) F’ and F = 3 \(→\) F’) and for \(^{87}\text{Rb}\) (F = 1 \(→\) F’ and F = 2 \(→\) F’) have been studied in detail [14], mainly because of the importance in laser cooling and Bose-Einstein Condensation with Rb. Usually the hyperfine transition F=2 \(→\) F’=3 in \(^{87}\text{Rb}\) is frequency locked for using it as cooling transition. However, all the six components may not be fully resolved because of the close spacing of the six frequencies. Recent theoretical simulation [15] shows the difficulty of isolating the components in Lamb dip spectroscopy.

In the traditional methods of double resonance spectroscopy in a three level system with one common level copropagating pump and probe beams are used to observe induced emission power of the probe [16,17] in the case of homogeneously broadened microwave-microwave transitions.
The emission line shape and a splitting of the absorption peak may be observed depending on the power of the saturating beam [17]. In the case of inhomogeneously broadened transitions like $D_1$ and $D_2$ transitions in the alkali atoms similar copropagating beams from two laser sources may be used. Very recently such a pump-probe spectroscopic method using copropagating laser radiations has been reported to observe additional non-linear resonances of the Sodium $D_1$ line [18]. They used a four level system with two lower hyperfine levels and two upper hyperfine levels. The system is a combination of double Λ and two V-type transitions. In addition to the EIT of the Λ-type resonance they reported difference frequency crossing in the V-type configuration. We report a pump-probe experiment on Rb $D_2$ lines using copropagating laser beams where the pump beam (or the control laser) is locked at a certain frequency within the Doppler broadened background, the probe frequency is tuned and the probe power is monitored. At all the lock points of the pump beam we observe very clearly a set of three dips corresponding to the hyperfine components. The set of dips can be shifted by tuning the pump frequency thus exhibiting velocity selective resonances (VSR). Such a set of drifting VSR dips are reported for the first time.

The pump-probe experiment carried out in this work uses an atomic rubidium vapour cell having a length of 5 cm and window of diameter 2.5 cm (Fig. 1). The vapour cell is sealed with a pressure of one micro-Torr and there is no buffer gas. The pump frequency is produced by an external cavity diode laser (ECDL), TEC 100 from Sacher Lasertechnik. The weak probe frequency is obtained from another ECDL, Velocity 6312 from New Focus. The pump beam is split into two components using a 70:30 beam splitter. The transmitted beam is sent through another cell (cell 2) of the same configuration for saturation absorption spectroscopy (SAS) or Lamb dip measurements. The Lamb dip signal consisting of the hyperfine and crossover resonance transitions is fed to the input of the PID lock circuit for frequency locking purpose. The pump frequency is locked at different frequencies around the $^{85}\text{Rb } 5^2S_{1/2}(F = 3) \rightarrow 5^2P_{3/2}(F' = 2, 3, 4)$ transitions (Fig. 2) and their crossover resonance peaks appearing in the Doppler broadened background by using a home built PID lock circuit that has a long term frequency stability of 1 MHz. The reflected pump beam is sent through cell 1. The pump and probe fields in cell 1 are copropagating and colinear. A Coherent wavemeter is used to monitor the wavelengths of the pump and probe transitions. The typical pump and probe intensities used in the sample cell are 22 mW/cm$^2$ and 1.27 mW/cm$^2$ respectively. The powers are measured by a Melles Griot power meter. The lock point is varied using the offset of the lock circuit from the lower frequency to the higher frequency region. For each lock frequency of the pump, the probe scans a frequency range of nearly 1 GHz around the $^{85}\text{Rb } D_2 (F \rightarrow F')$ transitions. The pump beam is sent through an optical isolator to avoid feedback of laser power. Fig. 3a is the probe transmission in the absence of pump field and this may be used as the reference spectrum. Figs. 3b-3f show the probe transmission in presence of the pump field with different lock frequencies as shown. The set of three dips in the Doppler broadened background of probe absorption shift from the lower frequency to the higher frequency region.
region in harmony with the increasing lock frequency of the pump laser. The third dip has the highest intensity and it always coincides with the pump frequency. The separations between the dips match with the known hyperfine splittings ($\Omega_2 - \Omega_1 = 63.43$ and $\Omega_3 - \Omega_2 = 120.91$ MHz) of the excited state hyperfine levels $5^2P_{\frac{3}{2}}$ ($F' = 2, 3, 4$) of $^{85}\text{Rb}$ [14]. The relative strengths of the dips also change with shift of the lock point of the pump field. The set of three dips representing emission or reduction of absorption, and their shifts are reproducible with all pump powers larger than the probe power. By changing the pump power the peak height and width of the dips can be changed. The collision broadening is negligible at the pressure used. So the dips are power broadened Lorentzians.

The same experiment is repeated for the other $D_2$ transitions of $^{85}\text{Rb}$ and $^{87}\text{Rb}$. In all cases the hyperfine triplets moving with the pump laser frequency are observed. However they are much weaker than those in Fig. 3. Three well separated peaks in $^{87}\text{Rb}$ ($F = 2 \rightarrow F'$) are shown in Fig. 4. Since one of the dips is weak vertical arrows are used to mark them. The separation being very large, with the movement of the pump frequency, all the dips cannot be seen, as one of them may fall beyond the Gaussian curve (Fig. 4f). In fact this dip is very weak, but distinctly visible in other cases (Fig. 4b-4e). The observed separations of the triplet are $\Omega_2 - \Omega_1 = 157.09$ and $\Omega_3 - \Omega_2 = 267.17$ MHz. This agrees with the known hyperfine splitting values [14]. In all the spectra the intermediate dip is the strongest and it coincides with the pump frequency.

When the pump laser is operating at any frequency within the Doppler broadened envelope, it can cause non-resonant excitations for all the transitions $\Omega_1$, $\Omega_2$, and $\Omega_3$. Considering one-dimensional motion of atoms, this will happen for the velocities $v_i$ of atoms given by $\Omega_i = \omega_{pu}(1 - \frac{v_i}{c})$, $i=1,2,3$. These excitations will cause population holes for the three velocity groups $v_1$, $v_2$ and $v_3$. The atoms will meet the probe laser at shifted frequencies $\omega_{pr}(1 - \frac{v_i}{c})$ for these velocities. So the tuned probe laser absorption will exhibit dips at $\omega_{pr} = \omega_{pu}$, $\omega_{pr} - \omega_{pu} = \pm |(\Omega_3 - \Omega_1)|$, $\pm |(\Omega_3 - \Omega_2)|$, $\pm |(\Omega_2 - \Omega_1)|$. In principle there may be seven dips in the absorption curve.

However, the dips will not all be of the same magnitude as the population depletion for the three velocity groups will not be the same. The population depletion will depend on two factors - (i) the number of atoms with velocity $v_i$: $N_i$, which will be obtained from the Maxwell-Boltzmann velocity distribution and (ii) the fraction $f_i$ of the atoms excited to the higher state by the transition of frequency $\Omega_i$. $f_i$ will be determined by the transition probability of the particular case. It will also depend on the power of the pump laser but that is kept constant during the experiment. So if $n_i$ is the depth of holes for the transition $\Omega_i$, we may write $n_i = f_i N_i$. Simple numerical estimates show that for $\omega_{pu}$ in the Doppler broadened region, the velocities $v_i$ for the non-resonant excitations are such that the variations in $N_i$ are not very large if the hyperfine splitting is small. However, the relative transition probabilities for the three hyperfine transitions may be quite different. If one particular $f_i$ is much larger compared to the other two, then $n_i$, the
the $^{85}$Rb D$_2$ transitions, it is known that $\Omega_3$ has the strongest transition probability [19]; in that case $f_3$ is much greater than $f_1$ or $f_2$ and so the population depletion for the $v_3$ group will be the largest. One can then see that the strongest dips will occur whenever $\omega_{pr} = \omega_{pu}$ or $\omega_{pr} = \omega_{pu} - |(\Omega_3 - \Omega_2)|$ or $\omega_{pr} = \omega_{pu} - |(\Omega_3 - \Omega_1)|$ as long as $\omega_{pu}$ is within the Doppler-broadened region. The experimental observation agrees with this (Fig. 3). In principle, there could be four other peaks, but they are too weak to be observed by our experimental setup.

The data for the $^{87}$Rb transitions can also be explained by similar arguments. However, in this case the separations between the hyperfine transitions are much larger. Hence $N_i$ may have wide variations. In this case we have to calculate the complete absorption coefficient by considering the absorption cross section and the lower level population depletion caused by the non-resonant pump in order to determine the relative strength of the dips.

The experimental results obtained by us are similar to the difference frequency crossing proposed by Wong et al.[18]. But we do not observe symmetrically located dips on two sides of the pump laser frequency. Such dips will be very small and have negligible intensity. The two symmetrical dips reported for the single V-type system by Wong et al.[18] also had large difference in intensities. In our case of a double V-system (single lower level and three upper levels) considering one dimensional motion of atoms we predict seven dips. But the intensities of such dips differ considerably depending on the frequency difference $|\omega_{pu} - \Omega_i|$. The difference is prominent in Rb as it has a narrower Doppler width compared to Na.

Using copropagating laser radiation the probe absorption always reveals three strong dips corresponding to the hyperfine components and having the same separation as the hyperfine splitting. There should be no crossover resonance as it is observed in the case of Lamb dip spectroscopy. But it must be clear that the observed transmission peaks are not the hyperfine transitions. The positions may be variable points of the Doppler broadened curve depending solely on the pump laser frequency. It is found and explained that the set of transmission peaks, that "represent" the hyperfine components, tread along the Doppler profile and the pump laser actually pulls or drags the triplet peaks. Thus we can control and manipulate the positions and intensities of transmission peaks by shifting the position of the pump laser frequency. We can derive highly accurate values of hyperfine splitting free from the problem of any crossover resonance in a simple pump-probe experiment, though the absolute frequencies cannot be determined in this process. No effect of Autler-Townes splitting can be found in this experiment. The interesting feature of this experiment is that the triplet of 'hyperfine dips' appear at all pump frequencies within the Doppler background.

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**Figure Captions**

**Figure 1**: Schematic representation of the experimental arrangement for velocity-selective resonances; ECDL: external cavity diode laser, OI: optical isolator, BS: beam splitter, CBS: cubical beam splitter, PD: photo detector, M: mirror, L: convex lens, BD: beam dump.

**Figure 2**: Energy level diagram for the double V configuration used in this work. F=2,3 for \(^{85}\text{Rb}\) and F=1,2 for \(^{87}\text{Rb}\). \(\Omega_i\) are resonance frequencies for \(D_2\) transition.

**Figure 3**: The probe absorption profile of \(^{85}\text{Rb}\) transition, when the pump laser is locked to different points of the Lamb dip spectrum of \(^{85}\text{Rb}\) \(5^2S_{1/2} \rightarrow 5^2P_{3/2}(F=3) \rightarrow 5^2P_{3/2}(F'=2,3,4)\): (a) the pump is switched off, (b)-(f) pump is on, the upward pointing arrows represent the lock points.

**Figure 4**: The probe absorption profile of \(^{87}\text{Rb}\) transition, when the pump laser is locked to different points of the Lamb dip spectrum of \(^{87}\text{Rb}\) \(5^2S_{1/2} \rightarrow 5^2P_{3/2}(F=2) \rightarrow 5^2P_{3/2}(F'=1,2,3)\): (a) pump is off, (b)-(f) pump is on, the upward pointing arrows represent the pump lock points, while the downward pointing arrows represent the velocity selective resonances.
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