Doppler-free spectroscopy in driven three-level systems

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Abstract. We demonstrate two techniques for studying the features of three-level systems driven by two lasers (called control and probe), when the transitions are Doppler broadened as in room-temperature vapor. For Λ-type systems, the probe laser is split to produce a counter-propagating pump beam that saturates the transition for the zero-velocity atoms. Probe transmission then shows Doppler-free peaks which can even have sub-natural linewidth. For V-type systems, the transmission of the control beam is detected as the probe laser is scanned. The signal shows Doppler-free peaks when the probe laser is resonant with transitions for the zero-velocity group. Both techniques greatly simplify the study of three-level systems since theoretical predictions can be directly compared without complications from Doppler broadening and the presence of multiple hyperfine levels in the spectrum.

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1 Introduction

There have been several recent theoretical and experimental studies where control lasers have been used in three-level systems to modify the absorption properties of a weak probe laser [1–4]. For example, in electromagnetically induced transparency (EIT), an initially absorbing medium is made transparent to a probe beam when a strong control laser is turned on [5,6]. EIT techniques have several practical applications in probe amplification [7], lasing without inversion [2] and suppression of spontaneous emission [8–10]. Experimental observations of EIT have been facilitated by the advent of low-cost tunable diode lasers which can be used to access transitions in alkali atoms such as Rb and Cs. Alkali atoms have convenient energy levels with strong oscillator strengths which form almost ideal three-level systems. However, spectroscopy in room-temperature vapor of these atoms is often limited by Doppler broadening, which is typically a factor of 100 larger than the natural linewidth. Therefore, in many experiments, large laser intensities are needed to overcome this broadening and observe the predicted effects [4], since the scale for the Rabi frequency of the control laser is set by the width of the transition. Moreover, the Doppler-broadened spectrum contains several closely-spaced hyperfine levels, so that the system is not a simple three-level system but a multi-level system. In such situations, theoretical calculations based on the three-level model are only approximately valid.

In this paper, we demonstrate two techniques to overcome the Doppler broadening in room-temperature vapor and make the system a true three-level system. The first technique applies to Λ-type systems, where the control and probe lasers couple different ground levels to the same excited level. The second technique applies to V-type systems, where the two lasers couple the same ground level to different excited levels. We demonstrate these techniques in a room-temperature vapor of Rb atoms, with two diode lasers tuned to different hyperfine transitions in the D_2 line. We obtain Doppler-free spectra and observe predicted features such as sub-natural linewidths for the dressed states created by the control laser [11]. The separation and linewidth of the states depends on control-laser intensity as predicted from a simple model of a driven two-level system. In addition, the creation of resonances with sub-natural linewidth has applications in precision spectroscopy [12], better stabilization of lasers to atomic transitions, and attaining sub-Doppler temperatures in laser cooling of atoms and ions [13].

2 Experimental details

The experimental set up, shown schematically in Figure 1, consists of a probe beam and a control beam derived from two frequency-stabilized diode laser systems operating near the D_2 line in ^{87}Rb ($5S_{1/2} \leftrightarrow 5P_{3/2}$ transition at 780 nm). The linewidth of the lasers after stabilization is less than 1 MHz. The probe beam has $1/e^2$ diameter of 3 mm while the control beam is slightly larger with

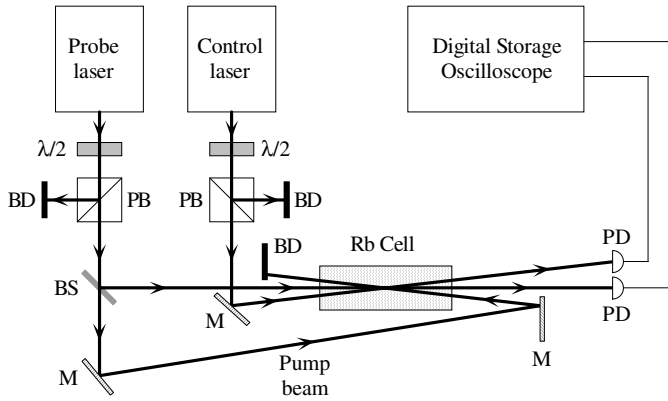[a] e-mail: `vasant@physics.iisc.ernet.in`

Fig. 1. Schematic of the experiment. The probe and control beams are derived from frequency-stabilized, tunable diode laser systems. The angle between the beams has been exaggerated for clarity; in reality it is close to 0, ensuring good overlap of the beams in the Rb cell.

Fig. 2. Three-level systems in Rb. Rb can be used to form either a Λ-system or a V-system, depending on which hyperfine levels are coupled by the two lasers. The control laser has detuning Δ_c and Rabi frequency Ω_R. The probe laser has detuning Δ.

diameter of 4 mm. The two beams are mixed in a beamsplitter and co-propagate through a room-temperature vapor cell containing Rb. The absorption through the cell is about 25%. The two beams have identical polarization and the angle between them is about 17 mrad. For some experiments, a counter-propagating pump beam is generated from the probe laser using a beamsplitter.

The D_2 line in ^{87}Rb has two hyperfine levels in the ground state ($F = 1, 2$) and four hyperfine levels in the excited state ($F' = 0, 1, 2, 3$). Thus, as shown in Figure 2, it can be used as either a Λ-system or a V-system, depending on which levels are coupled by the lasers. For the Λ-system, the same excited level ($F' = 2$) is coupled to two ground levels: to the $F = 1$ level by the control laser and to the $F = 2$ level by the probe laser. For the V-system, the same ground level ($F = 2$) is coupled to two excited levels: to the $F' = 3$ level by the control laser and the $F' = 1$ (or 2) level by the probe laser. The control laser has Rabi frequency of Ω_R and detuning from resonance of Δ_c. The weak probe laser has detuning Δ. The spontaneous decay rate from the excited levels is Γ, which is $2\pi \times 6.1$ MHz in Rb.

3 Λ-system

We first consider the Λ-system. The absorption of the weak probe is proportional to $\mathrm{Im}(\rho_{13})$, where ρ_{13} is the induced polarization on the $|1\rangle \leftrightarrow |3\rangle$ transition coupled by the probe laser. From the density-matrix equations, the steady-state value of ρ_{13} is given by [3]:

$$\rho_{13} = \frac{(\Omega_p/2)(\Delta - \Delta_c)}{|\Omega_R/2|^2 - i(\Gamma - i\Delta)(\Delta - \Delta_c)}, \qquad (1)$$

where Ω_p is the Rabi frequency of the (weak) probe beam. The pole structure of the above equation shows that probe absorption has two peaks at the following detunings:

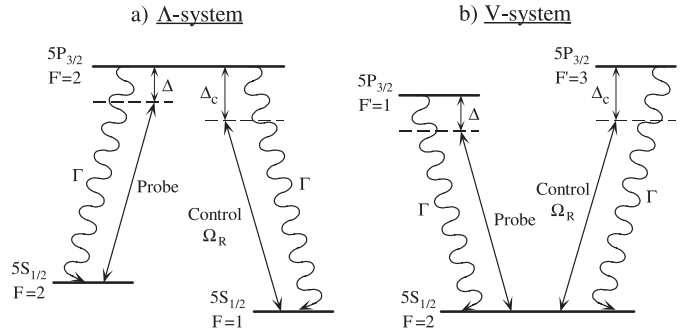$$\Delta_\pm = \frac{\Delta_c}{2} \pm \frac{1}{2}\sqrt{\Delta_c^2 + \Omega_R^2}, \qquad (2)$$

with the corresponding linewidths:

$$\Gamma_\pm = \frac{\Gamma}{2}\left(1 \mp \frac{\Delta_c}{\sqrt{\Delta_c^2 + \Omega_R^2}}\right). \qquad (3)$$

The two peaks are the well-known Autler-Townes doublet appearing at the location of the two dressed states created by the control laser [14]. Due to coherence between the dressed states, the peaks have asymmetric linewidths for non-zero detuning of the control laser, in such a way that the sum of the two linewidths is equal to the unperturbed linewidth, Γ.

The above analysis shows how three-level systems are useful in many applications. For example, probe absorption at line center ($\Delta = 0$) is strongly suppressed in the presence of a resonant control laser because the dressed states created by the control laser are shifted by the Rabi frequency. This is the basis for EIT experiments. Similarly, it is seen from equation (3) that the linewidth Γ_+ of the second dressed state can be much below the natural linewidth when the control-laser detuning is large.

However, observing such effects in room-temperature vapor is complicated by effects due to Doppler broadening. The above expressions are valid for a stationary atom; in room-temperature vapor they have to be averaged over the Maxwell-Boltzmann velocity distribution of the atoms taking into account the different frequencies seen by each atom in its velocity frame. The effect of this averaging is that the location of the peaks given in equation (2) does not change, but the linewidth of the peaks changes to [3]:

$$\Gamma_\pm = \frac{\Gamma + D}{2}\left(1 \mp \frac{\Delta_c}{\sqrt{\Delta_c^2 + \Omega_R^2}}\right), \qquad (4)$$

where D is the Doppler width, which is 560 MHz for room-temperature Rb atoms. Thus, in EIT experiments, the Rabi frequency of the control laser has to be very large to see significant reduction in absorption at line center. Another complication due to the appearance of the Doppler width is that the different excited-state hyperfine levels, spaced a few 100 MHz apart, all lie within the Doppler-broadened profile. Any comparison with the predictions of
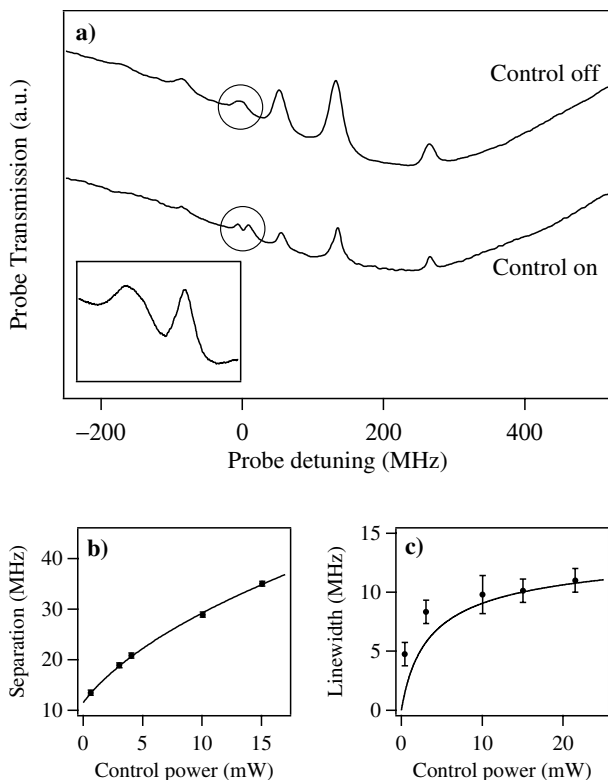
Fig. 3. Doppler-free spectroscopy in Λ-system. In (a), we show the transmission of the probe laser as it is scanned across all the $F = 2 \leftrightarrow F'$ transitions. The upper trace is with the control beam turned off and shows the usual saturated absorption peaks. The lower trace is with the control beam turned on, and shows the $F' = 2$ peak splitting into an Autler-Townes doublet. The control laser has detuning of -11.5 MHz. The inset is a close-up of the doublet showing the asymmetric linewidths of the two peaks. In (b) and (c), we plot the separation of the peaks and the linewidth of the smaller peak as a function of control-laser power. The solid lines are the expected variation from equations (2, 3), as explained in the text.

the above equations is difficult because the control laser couples to several hyperfine levels, but with different detunings. The simple model of a two-level system driven by the control laser is no longer valid.

We have solved this problem for the Λ-system in the following manner. A part of the probe laser is split off and sent through the cell so that it is counter-propagating with respect to the probe and control beams. The intensity of this pump beam is chosen to be about 5 times higher than the probe. In this configuration, the zero-velocity group of atoms preferentially absorbs from the pump and the probe gets transmitted. This is a standard technique used in Doppler-free saturated-absorption spectroscopy [15], which we have adapted to the three-level case. Note that the pump beam is still very weak compared to the control and any coherent driving from this beam can be neglected as in the case of the probe beam [16].

In Figure 3a, we show the probe spectrum as the laser is scanned across all the $F = 2 \leftrightarrow F'$ transitions. The control laser is locked to the $F = 1 \rightarrow F' = 2$ transition with a

detuning of $\Delta_c = -11.5$ MHz. With the control beam off, the spectrum shows the typical Doppler-broadened profile (with a linewidth of 560 MHz), and "Doppler-free" peaks at the location of the hyperfine transitions due to saturation by the pump beam. The words "Doppler-free" are set in quotes because the linewidth of the hyperfine peaks is usually larger than the natural linewidth. The primary cause for this is power broadening from the pump laser and a small misalignment angle between the counter-propagating beams. Indeed, we have studied the variation of the linewidth as a function of pump power. With near-perfect alignment of the beams and with a magnetic shield [17] around the cell, the linewidth extrapolated to zero power is only 6.5 MHz, close to the natural linewidth of 6.1 MHz. This shows that collisional broadening in the vapor cell is negligible. The effect of stray magnetic fields in the vicinity of the cell is to increase the linewidth by 15–20%, which we have verified by measuring the linewidth with and without the magnetic shield.

The near-perfect alignment of the pump-probe beams is possible only by mixing and separating the beams using polarizing-beamsplitter cubes. This requires the use of orthogonal polarization for the two beams. For the experiments reported here, we have not used this technique but rather used a small angle (of $\sim$17 mrad) between the beams. There are two reasons for this. The first is that we require the beams to have the same polarization. Otherwise, the different magnetic sublevels are driven differently, and the analysis becomes complicated. The second reason is that the separation of orthogonal-polarization beams using a polarizing beamsplitter is not perfect. Any leakage of one beam into the other makes the interpretation of the detected signal questionable. Thus, under typical conditions, the observed linewidth has three broadening mechanisms: power broadening due to the pump beam, which increases the linewidth from 6.1 MHz to about 20 MHz; misalignment by 17 mrad, which increases it further to 25 MHz, and residual magnetic fields, which increases it to 30 MHz. This accounts for the linewidth of the hyperfine peaks in Figure 3a, but note that this is still 20 times smaller than the Doppler width and is thus essentially Doppler free.

As seen in the bottom trace in Figure 3a, when the control laser is turned on, the $F' = 2$ peak splits into two as predicted by the earlier analysis of the three-level system. The two peaks are clearly resolved even though their separation is only of order 20 MHz. As shown in the inset, the peaks also have asymmetric linewidths. This is because of the non-zero detuning of the control laser ($\Delta_c = -11.5$ MHz), which results in unequal linewidths from equation (3).

We have thus achieved a situation where the Doppler broadening is virtually eliminated and the three-level assumption is valid. Therefore the predictions of equations (2, 3) can be applied without worrying about the presence of other hyperfine levels. To demonstrate this, we have studied the probe-absorption spectra as a function of control-laser power and detuning. As shown in Figure 3b, the separation of the peaks [18] increases with

power as expected from equation (2). We have plotted the separation as a function of control-laser power and not Rabi frequency since the power is what is measured in the laboratory. The relation between the power and the Rabi frequency depends on factors such as the intensity profile across the control beam, overlap between the beams, losses at the cell surfaces, absorption through the cell, and so on. It is difficult to experimentally determine these factors with any degree of certainty, therefore we have left this factor as an overall fit parameter in obtaining the solid line in Figure 3b. It is important to note that this fit parameter does not determine the trend in the data, and changes in its value would cause the solid line to move up or down without changing its shape. The best fit shown in the figure is obtained with a parameter that assumes that the control-laser power is spread uniformly over a circle of diameter 4.6 mm, which is reasonable given that the measured size of the Gaussian beam is 4 mm.

The linewidth of the smaller peak also follows the trend predicted by equation (3), as shown in Figure 3c. There are no additional fit parameters to obtain the solid line except that we have to increase the unperturbed linewidth in equation (3) from the natural linewidth of 6.1 MHz to 30 MHz. As explained earlier, this is the linewidth obtained for the hyperfine peak without the control laser. The two points at low power lie slightly above the curve, possibly because in these cases it is hard to determine the linewidth accurately since the lineshape is distorted by the underlying Doppler profile and peak pulling by the other dressed state. However, despite these effects and the somewhat large unperturbed linewidth, the linewidth of the smaller peak is below the natural linewidth for small control powers. We have recently demonstrated this as a technique to achieve sub-natural resolution in room temperature vapor [11]. We have also combined the narrow linewidth and the variation with detuning to make precise measurements of hyperfine intervals. We have used this technique to measure a hyperfine interval in Rb with an accuracy of 44 kHz [12].

4 V-system

We now turn to the V-system. The theoretical analysis proceeds along similar lines as for the Λ-system [7]. In room temperature vapor, the location of the two Autler-Townes peaks is the same as before (Eq. (2)). Similarly, the linewidths of the dressed states (given by Eq. (3)) are increased by the Doppler width after thermal averaging. In this case, we obtain Doppler-free spectra not by using a counter-propagating pump beam but by detecting the transmission of the control laser through the vapor cell. The control laser is locked to $F = 2 \leftrightarrow F' = 3$ transition while the probe laser is scanned across all the $F = 2 \leftrightarrow F'$ transitions. Since the control laser is locked, it is only the zero-velocity atoms that absorb from this laser. When the probe laser is also resonant with a transition starting from the same zero-velocity atoms, the absorption from the control laser is slightly reduced and the transmitted signal shows a narrow peak. Thus, as the probe laser is
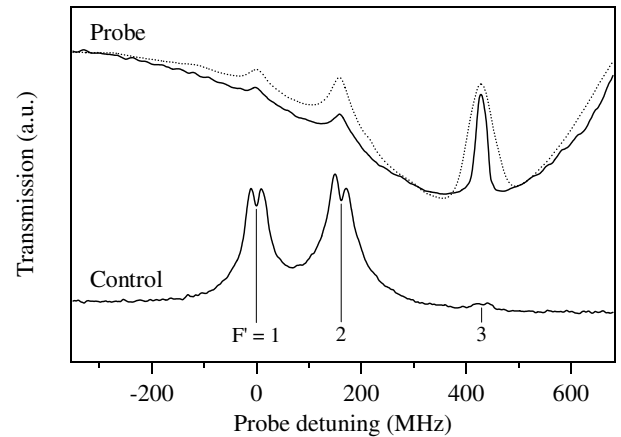

Fig. 4. Spectroscopy in V-system. The upper trace is the transmission of the probe laser as it is scanned across all the $F = 2 \leftrightarrow F'$ transitions. The spectrum shows the expected Doppler broadening. The peaks at $F' = 1$ and 2 are due to EIT effects from a resonant control laser. The transparency increases to nearly 100% as the control-laser power is increased from 0.8 mW (solid line) to 7 mW (dotted line). The peak at $F' = 3$ is due to saturation by the control laser. The lower trace is the transmission of the control laser. The Autler-Townes doublets near the $F' = 1$ and 2 levels are clearly resolved with Doppler-free linewidths.

scanned, Doppler-free peaks appear in the transmission of the control-laser at the exact locations where the probe is absorbed.

The resultant spectra are shown in Figure 4. The experimental parameters are similar to the case of the Λ-system, except that the beam sizes are reduced by a factor of 3, and there is no counter-propagating pump beam. Under these conditions, the probe spectrum shows the expected Doppler broadening with some increased transmission at line center for the $F' = 1$, 2 and 3 transitions. However, there is no apparent signature of the dressed states created by the control laser. By contrast, the control transmission clearly shows the Autler-Townes doublets near the $F' = 1$ and 2 levels. It is interesting to note the increased transmission of the probe laser at line centers. In the case of the $F' = 1$ and 2 levels, the increased transmission is precisely due to EIT because the location of the dressed states is shifted away from the line center. Indeed, at higher power (dotted line in figure), the transparency approaches 100%. In the case of the $F' = 3$ level, both probe and control lasers couple the same levels and the increased transmission of the probe is due to saturating effects from the much stronger control laser.

One of the disadvantages of the pump-probe technique used for the Λ-system is that the Doppler-free peaks appear on top of a broad Doppler profile, as seen in Figure 3a. However, in the V-system, our technique of measuring the control-laser transmission results in a constant background signal corresponding to absorption by the zero-velocity atoms. The signal changes only when the zero-velocity atoms start absorbing from the probe. Therefore, the Doppler-free peaks appear on a flat background.
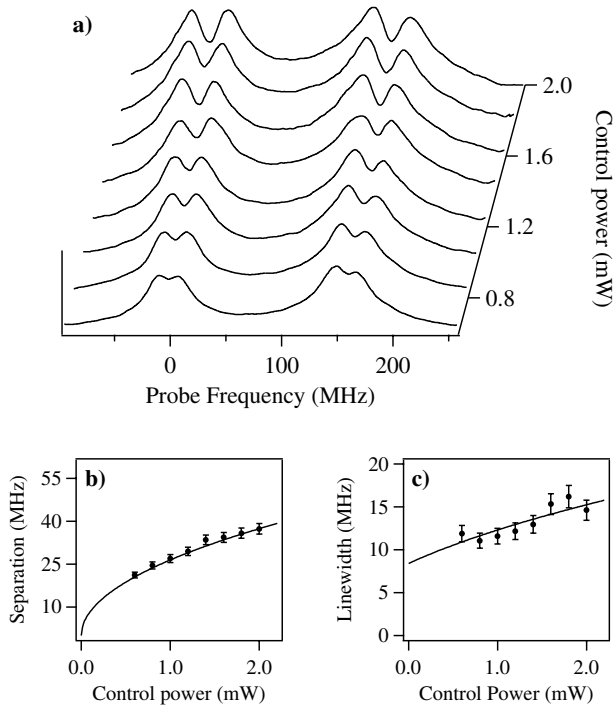
Fig. 5. In (a), we show a waterfall plot of the transmission of the control laser as a function of probe-laser detuning, for different values of control-laser power. The control laser has zero detuning. As the control power is increased, the signal near the $F' = 1$ and 2 levels shows the Autler-Townes doublets. In (b), we plot the separation of the peaks as a function of power. The solid line is the variation expected from equation (2) in the text. In (c), we show the linewidth of one of the peaks as a function of control power. The solid line is the increase due to power broadening, as explained in the text.

This is shown in Figure 5a, which is a waterfall plot of control transmission spectra for different values of control power. The $F' = 1$ and 2 peaks split into Autler-Townes doublets, with the separation of the doublets increasing with power. At very low powers (below 0.5 mW) the splitting is smaller than the unperturbed linewidth, and the doublet peaks are not well resolved. One other feature of the spectrum is that the doublet peaks in the $F' = 2$ level are not of equal height. This kind of asymmetry has also been observed in the case of Doppler-broadened doublets [4].

As in the case of the Λ-system, the Doppler-free spectra allow the predictions of the theoretical analysis to be applied with confidence. This is seen in Figure 5b, where we plot the separation of the peaks in the Autler-Townes doublet as a function of control-laser power. The solid line is the variation predicted by equation (2). As before, the relation between the power and the Rabi frequency is left as a fit parameter. With the best fit, the agreement with the observed separation is quite good, indicating that the three-level model is adequate to explain the data.

The analysis of the linewidth of the two peaks is slightly different from the case of the Λ-system. Since the control-laser detuning is zero, equation (3) predicts

that the linewidth should remain constant at $\Gamma/2$. From Figure 5c, we see that the measured linewidth is about 12 MHz (corresponding to a total linewidth of 24 MHz) and shows a gradual increase as the power is increased. If we assume an unperturbed linewidth of 7 MHz, then it increases to 14 MHz due to the misalignment and further to 17 MHz due to stray magnetic fields, which is still lower than the measured value. We attribute the remaining linewidth to power broadening by the control laser. In the case of the Λ-system, the control laser is detuned from resonance and its influence on the linewidth is negligible, therefore the only power broadening comes from the pump beam.

The power-broadened linewidth varies as $\Gamma\sqrt{1+s}$, where s is the saturation parameter. However, at the control-laser powers used in the experiment, the power-broadened linewidth is 40 MHz, which is much larger than the observed linewidth of 24 MHz. This is because the control laser is primarily driving the atoms coherently, and power broadening is a smaller effect. To account for this reduction, we have fitted the linewidth to $\Gamma\sqrt{1+ks}$, with k as a fit parameter. In Figure 5c, the solid line is obtained with a value of $k = 0.16$, or 16%. The curve fits the data reasonably well and explains why the linewidth increases with control power. However, it is important to note that the measured linewidth is still much smaller than the Doppler width, as in the case of the Λ-system.

5 Conclusions

We have thus demonstrated techniques to overcome Doppler broadening when studying the properties of three-level systems driven by two lasers. The Λ-system requires the use of a counter-propagating pump beam to saturate the transition for the zero-velocity atoms. In the V-system, the control laser transmission is detected as the probe laser is scanned. The transmitted signal shows Doppler-free peaks when the probe is absorbed by the zero-velocity atoms. In the Λ-system, probe absorption is to the two dressed states in the upper level created by the control laser. On the other hand, in the V-system, probe absorption is from the two dressed states created in the ground state. Probably because of this difference, each technique works only in its respective system and not in the other system. But the techniques allow the predictions of simple theoretical models to be applied directly since there are no complications arising from Doppler broadening or the presence of multiple levels in the Doppler-broadened spectrum. We expect these techniques to find widespread use in the study of driven three-level systems. We have already demonstrated the application of these ideas for high-resolution hyperfine measurements in Λ-systems [12]. In future, we plan to apply such ideas to precision spectroscopy in V-systems also.

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