

# Doppler-free spectroscopy of rubidium atoms driven by a control laser

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A scheme of Doppler-free spectroscopy is experimentally demonstrated with a co-propagating control laser locking to an atomic hyperfine transition, and the differential transmission of the probe and the reference laser is detected. Crossover resonances are eliminated by selecting the class of atoms with zero velocity in the direction of beam propagation. In addition, the sub-Doppler spectrum experiences optical gain compared to the conventional saturated-absorption spectrum as a result of optical pumping.

**Keywords** Doppler-free spectroscopy, crossover resonances, optical pumping

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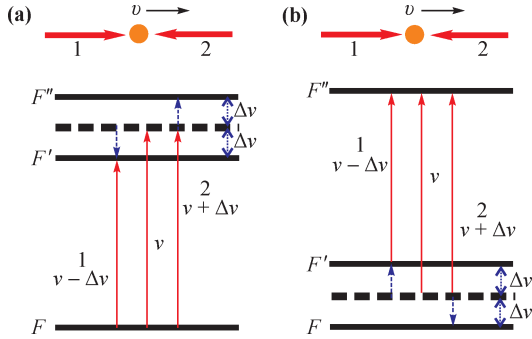
## 1 Introduction

The normal atomic absorption spectra are Doppler broadened due to random movements of atoms. Saturation absorption spectroscopy (SAS) [1–4] and polarization spectroscopy (PS) [5–8] are commonly used Doppler-free spectroscopic techniques to resolve the hyperfine structure of atomic spectra, and have proved useful tools in areas of laser frequency stabilization [8, 9] and the frequency reference application [10]. The SAS contains two counter-propagating beams splitted from one laser: One strong pump beam saturates atoms and the other much weaker probe beam is used to obtain the transmitted spectrum. The PS uses the similar setup but with a circularly polarized pump beam to induce the birefringence in the medium and a different detection scheme. In both of the two Doppler-free techniques, when the pump and probe beams are absorbed by atoms in the same velocity group simultaneously, several sub-Doppler absorption and dispersion features with the width approaching the atomic nature linewidth appear in the Doppler absorption profile. These sub-Doppler features

can be classified into two types: The first one happens when the pump and probe beams interact with atoms of zero velocity or velocity perpendicular to the beam propagation direction, corresponding to the real transitions of atoms; the second type occurs when the laser is tuned exactly halfway between two hyperfine levels and interacts with the atoms moving with a certain longitudinal velocity, which is called crossover resonances. Crossover resonances appear in the multiple unresolved hyperfine transitions that have a common level (Fig. 1), and provide a way of laser frequency stabilization with a frequency detuning from the exact transition, which is favorable in atomic laser cooling and trapping experiments. However, the presence of the crossover resonances makes the real resonances of atomic spectra less resolvable, especially in the case that adjacent transitions are very close to each other. It is noted that the amplitude of crossover resonances are usually larger than that of real transitions in SAS and PS, and these spurious resonances render the identification of closely spaced atomic structures difficult.

There are several methods to eliminate crossover resonances in multilevel structures of atoms. It is suggested

to use co-propagating probe and pump laser beams to avoid the appearance of crossover resonances [11]. The pump laser is locked to a hyperfine transition, and its transmission is monitored while the frequency of the probe laser is scanning over the range of the Doppler line width. When the probe comes into resonance with the same or other hyperfine transitions for atoms with zero longitudinal velocity, the pump laser transmission spectrum shows a Doppler-free dip due to the absorption reduction caused by the probe laser. In addition, elimination of crossover resonances can also be achieved with a nanocell [12, 13].



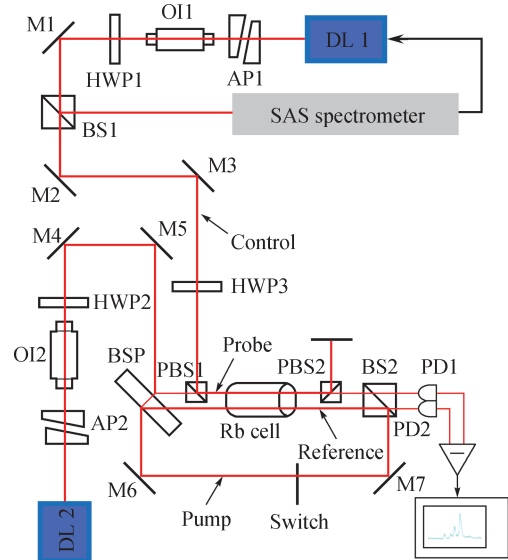
**Fig. 1** Schematic diagram of crossover resonances. The probe and pump beams drive two different hyperfine transitions. 1 and 2 represent the probe (pump) and the pump (probe) beam, respectively.  $\Delta\nu$  is the Doppler shift of lasers for atoms with longitudinal velocity of  $v$ . (a) Excitations sharing a common upper level; (b) Excitations sharing a common lower level.

In this work we demonstrate a new Doppler-free spectroscopy scheme in the control of laser driven multilevel atoms. Three co-propagating laser beams are used. The frequency of the control laser is stabilized to an atomic hyperfine transition, while the probe and reference lasers are scanning frequency over a range of several hundreds MHz. Unlike the scheme mentioned above [11], the differential transmission of the probe and reference laser beams instead of the transmission of the control laser, is monitored. The key idea is based on the velocity-selective optical pumping. Since the control laser is fixed on a hyperfine transition, only the atoms with zero velocity or velocity perpendicular to the laser propagation direction could absorb the control laser, and this contributes to the variation of the differential transmission of the probe and reference beams, resulting in the formation of sub-Doppler peaks on the flat background of transmission spectra. The presented scheme has the main characteristics as follows: i) the Doppler background is eliminated; ii) crossover resonances disappear; iii) real atomic resonances are optically amplified through optical pumping.

## 2 Experimental setup

The experimental setup is shown in Fig. 2. Two external cavity diode lasers (Toptica DL100) with the linewidth

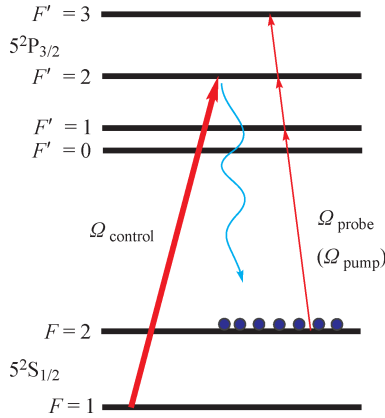
of  $\sim 1$  MHz are used. One laser beam from DL 2 is split into three parts. Two parallel weak beams act as the probe and the reference laser respectively, and the other much stronger counter-propagating beam is used as the pump laser. The output of DL 1 serves as a control laser. All the three beams co-propagate through a 10-cm-long room-temperature natural rubidium vapor cell with magnetic shielding. The probe and control lasers have orthogonal polarizations. They are combined with a polarizing beamsplitter cube (PBS 1), and then the control laser is separated by another polarizing beamsplitter cube (PBS 2) when it has passed through the Rb vapor cell. The profiles of the probe and control beams are reshaped by anamorphic prism pairs to obtain beam diameters of 1 mm and 3 mm respectively. The two beams are fully overlapped in the Rb vapor cell in order to achieve better resolution. Switching of the pump beam is controlled mechanically to provide reference spectra. Neutral density filters are used to vary powers of the two parallel beams independently. Transmissions of the probe and reference beams are detected by two photodiodes and the differential signal output is recorded by an oscilloscope.



**Fig. 2** Experimental setup of the Doppler-free spectroscopy. The frequency of the control laser is stabilized to a hyperfine transition, the probe and reference lasers scan around the  $D_2$  absorption line of Rb atoms. The presence of the pump laser is controlled by a switch. BS, beamsplitter cube; PBS, polarizing beamsplitter cube; BSP, beamsplitter plate; HWP, half-wave plate; PD, photodiode; M, mirror; OI, optical isolator; AP, anamorphic prism.

As shown in Fig. 3, in  $^{87}\text{Rb}$   $D_2$  line transition, the control laser drives the  $5^2S_{1/2} F = 1 \rightarrow 5^2P_{3/2} F' = 2$  transition and is actively locked to it using the SAS frequency modulation method. Most of the atoms are optically pumped to the  $5^2S_{1/2} F = 2$  state in the presence of the control laser. The probe laser has a frequency scanning range of several hundreds MHz around the  $5^2S_{1/2} F = 2 \rightarrow 5^2P_{3/2}$  transition. The powers of the probe and reference beams are adjusted to be ap-

proximately equal, so that the differential transmission between the two beams should be zero in the absence of the control laser, through which the Doppler broadened background is eliminated.

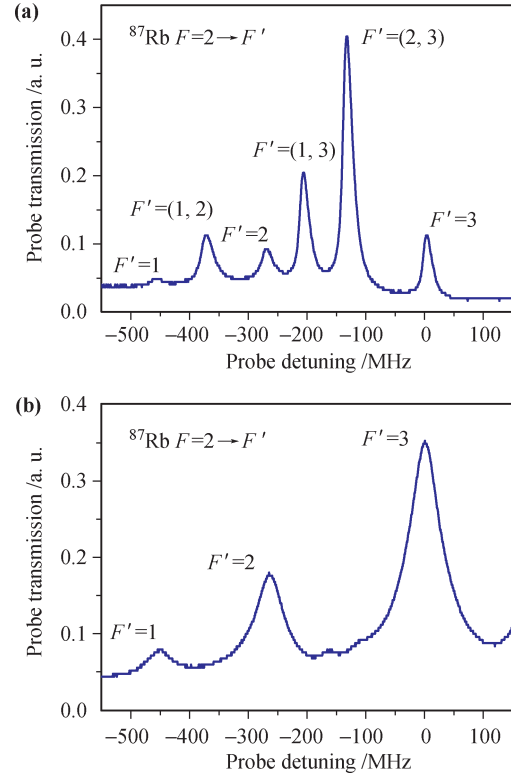


**Fig. 3** The relevant energy levels of D<sub>2</sub> line transition of <sup>87</sup>Rb atoms.

### 3 Results and analyses

In Fig. 4 (a), the conventional saturated-absorption spectrum is presented when the control laser is blocked. The individual hyperfine transitions are well-resolved including real atomic transitions and crossover resonances, and the amplitude of crossover resonances are much stronger than that of real transitions. As the control laser is resonant with the  $5^2S_{1/2} F = 1 \rightarrow 5^2P_{3/2} F' = 2$  transition, only the atoms with zero velocity in the beam propagation direction can absorb the control laser and are optically pumped to the  $5^2P_{3/2} F' = 2$  state. Then, the spontaneous emission from the  $5^2P_{3/2} F' = 2$  state to the  $5^2S_{1/2} F = 2$  state causes the increase of the population of the zero-velocity atoms in the  $5^2S_{1/2} F = 2$  state, resulting in the enhancement of the probe absorption and further the change of differential transmission of the probe and reference beams. While the probe laser is scanning around the  $5^2S_{1/2} F = 2 \rightarrow 5^2P_{3/2} F' = 1, 2, 3$  transition, a re-pumping process from the state  $5^2S_{1/2} F = 2$  to  $5^2S_{1/2} F = 1$  also occurs. In the case of the probe frequency is near to the  $5^2S_{1/2} F = 2 \rightarrow 5^2P_{3/2} F' = 2$  transition, and the control and the probe laser transitions form a three-level  $\Lambda$  system. If the control laser intensity is high enough, the atomic medium would become transparent for the probe in the vicinity of two-photon resonance as a result of the coherence induced by the control laser, which is known as electromagnetic induced transparency (EIT) [14, 15]. In our scheme, the control laser power is 200  $\mu$ W while the probe laser power is 20  $\mu$ W, therefore the optical pumping effect rather than the quantum coherence dominates. As shown in Fig. 4 (a), the  $5^2S_{1/2} F = 2 \rightarrow 5^2P_{3/2} F' = 3$  transition has the most prominent increments because it is a closed tran-

sition and re-pumping from state  $5^2S_{1/2} F = 2$  to state  $5^2S_{1/2} F = 1$  is forbidden. The line width of sub-Doppler features are slightly broadened due to the power broadening, the frequency stability of the control laser and the imperfect overlap of the co-propagating probe and control beams.

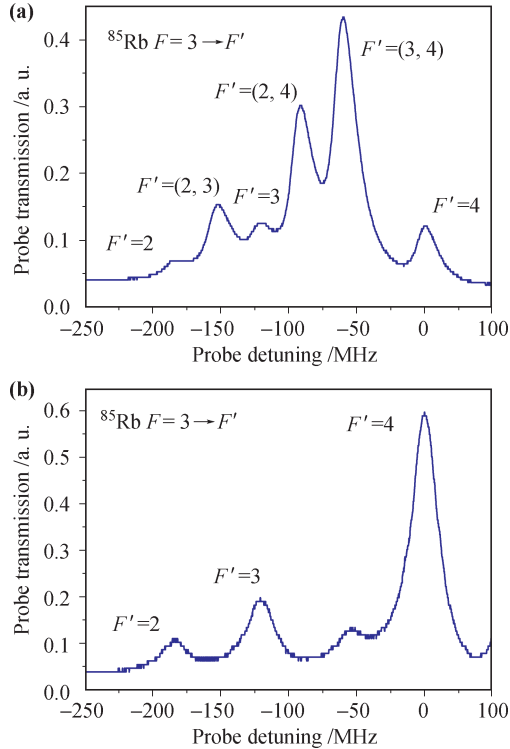


**Fig. 4** (a) Saturated-absorption spectrum of  $5^2S_{1/2} F = 2 \rightarrow 5^2P_{3/2} F'$  transition of <sup>87</sup>Rb D<sub>2</sub> line when the control laser is blocked; (b) The obtained Doppler-free spectrum in the presence of the control laser, and the pump laser is blocked. The control laser is locked to the  $5^2S_{1/2} F = 1 \rightarrow 5^2P_{3/2} F' = 2$  transition.

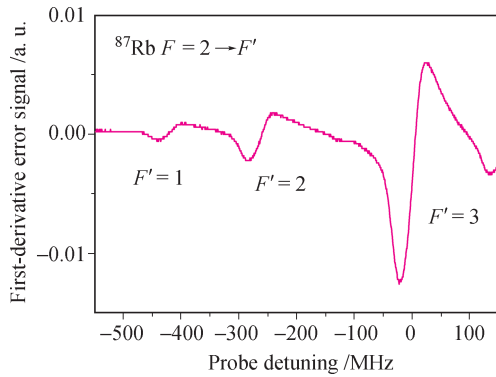
Figure 5 shows the Doppler-free spectrum in <sup>85</sup>Rb D<sub>2</sub> line transition with the same experimental setup. For <sup>85</sup>Rb, the control laser is locked to the  $5^2S_{1/2} F = 2 \rightarrow 5^2P_{3/2} F' = 3$  transition, and the probe scans around the  $5^2S_{1/2} F = 3 \rightarrow 5^2P_{3/2} F' = 2, 3, 4$  transitions. As shown in Fig. 5 (b), three sub-Doppler peaks with optical gain are clearly identified. It can also be seen that the closed transition of  $5^2S_{1/2} F = 3 \rightarrow 5^2P_{3/2} F' = 4$  has stronger amplitude than other open transitions.

In the following we obtain the first-derivative error signal corresponding to the Doppler-free profile presented in Fig. 4 (b) by using frequency modulation techniques. As shown in Fig. 6, three dispersion features are clearly resolved and can easily be matched to the corresponding hyperfine transitions. The intensity of the derivative signal is also enhanced due to optical pumping by the control laser. This provides an important advantage in high resolution spectroscopy and is also beneficial for the laser frequency locking for atomic multilevel structures. For other Doppler-free spectroscopy techniques such as

PS, the peaks of closely spaced transitions usually merge, making it difficult to match the dispersion features and the actual transitions exactly [6].



**Fig. 5** (a) Saturated-absorption spectrum of the  $5^2S_{1/2} F = 3 \rightarrow 5^2P_{3/2}$  transition of  $^{85}\text{Rb}$  D<sub>2</sub> line when the control laser is blocked; (b) The Doppler-free spectrum obtained with the new scheme. The control laser is locked to the  $5^2S_{1/2} F = 2 \rightarrow 5^2P_{3/2} F' = 3$  transition.



**Fig. 6** The derivative lineshape of the  $5^2S_{1/2} F = 2 \rightarrow 5^2P_{3/2}$  transition of  $^{87}\text{Rb}$  D<sub>2</sub> line obtained with the new scheme.

## 4 Conclusions

In summary, we have demonstrated a Doppler-free spectroscopy scheme by using the co-propagating probe and control lasers. The Doppler broadened background is eliminated by subtracting the probe transmission with that of a reference beam propagating through the same atomic vapor. The control laser locked to a hyperfine transition causes velocity selective optical pumping, and only the atoms with zero longitudinal velocity contribute

to the enhancement of the probe absorption. We detect the differential transmission of the probe and reference beams and obtain sub-Doppler peaks on a flat background. The presented Doppler-free scheme has features of eliminating crossover resonances and offering well-resolved atomic hyperfine structures. Meanwhile the signal experiences optical gain as a result of optical pumping. The scheme is meaningful for the high-resolution spectroscopy of closely spaced hyperfine levels (e.g. Zeeman spectra), for which the existence of crossover resonances complicates the identification of the relevant spectral features. Combined with the frequency modulation techniques [16], the presented scheme can be applied to laser frequency stabilization as well, making it much easier to match exactly the dispersion features to the atomic hyperfine transitions in comparison with the PS technique.

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