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SROP and DROP Spectra with Alkali Atomic Vapor Cell and Applications

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ABSTRACT

Two schemes of Doppler-free high-resolution velocity-selective optical-pumping atomic spectroscopy, named single-resonance optical pumping (SROP) and double-resonance optical pumping (DROP), are performed and characterized with room-temperature cesium vapor cells. Due to velocity-selective optical pumping from one hyperfine fold of ground state to another via one-photon excitation in SROP or cascade two-photon excitation in DROP and decay processes thereafter, the atomic population variation of one hyperfine fold of ground state is indicated by SROP and DROP spectra by using of the transmission of the probe laser which is usually frequency locked to a cycling hyperfine transition. As a result, SROP and DROP spectra often have flat background and higher signal-to-noise ratio. Therefore, SROP and DROP spectra are very useful for measurement of the dressed-state splitting of ground state with an alkali atomic vapor cell, precise measurement of hyperfine splitting of alkali atomic excited states, frequency references for laser frequency stabilization, two-color MOT, and so on.

Keywords: single-resonance optical pumping (SROP); double-resonance optical pumping (DROP); dressed state splitting; two-color cesium magneto-optical trap; frequency stabilization

1. INTRODUCTION

High-resolution laser spectroscopy has important applications in atomic and molecular physics, quantum optics, and quantum metrology, such as laser frequency stabilization [1, 2], precision measurement of atomic hyperfine structure and determination of relevant important physical constants [3]. Here, two schemes of the Doppler-free velocity-selective optical-pumping spectroscopy, named single-resonance optical pumping (SROP) and double-resonance optical pumping (DROP), are performed and characterized with room-temperature cesium vapor cells.

In SROP and DROP schemes, the probe laser is usually frequency locked to a cycling hyperfine transition between a ground state and an excited state with an adjustable detuning. The swept pumping laser beam is co-propagating or counter-propagating with the probe beam and the "zero-velocity" atoms' population of one hyperfine fold of ground state will be partially pumped into another once the pumping beam is resonant with the transition between that ground state and another excited state in SROP, or between that intermediate excited state and other higher excited state in DROP. As a result, this vibration of atom population in hyperfine ground state yields peaks or dips in probe beam's transmission signal which reveals the atomic hyperfine energy level structure. The SROP and DROP spectra often have flat background and higher signal-to-noise ratio (SNR), therefore, can be used to stabilize laser frequency [2, 4] and to measure hyperfine splitting accurately [5]. Our group stabilize the 795 nm external-cavity diode laser (ECDL) to the DROP spectrum of cesium $6P_{3/2}$ F'= 5 - $8S_{1/2}$ F''= 4 transition without the direct frequency modulation [2]. And this

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method is also used to realize two-color cesium magneto-optical trap (MOT) [6, 12] and to improve frequency stability of 1.5 μm diode laser by DROP spectrum of rubidium $5P_{3/2}$ - $4D_{3/2}$ transitions [4].

Importantly, the locked probe laser in SROP can be strong enough to dress the atoms, and this allows us to directly detect the dressed-state splitting of ground state with room-temperature atomic vapor cell, instead of the laser-cooled atomic sample that usually can only be done with, even if the dressed-state splitting is much smaller than the Doppler-broaden line-width [7]. Additionally, once the two-photon Doppler-free condition is fulfilled by arranging different propagating configurations of two beams, atomic coherence can reduce the spectrum line-width [8, 9]. We also demonstrate experimentally that electromagnetically induced transparency (EIT) exist in DROP spectrum whose line-width is reduced by EIT [9], and further investigate EIT in a multilevel ladder-type cesium atomic system [10].

2. PRINCIPLE OF SROP AND DROP SPECTROSCOPY OF CESIUM ATOMS

As shown in Fig. 1(A), the probe/coupling laser C is locked to |1⟩ - |2⟩ cycling hyperfine transition with frequency detuning Δ_C (usually $\Delta_C \sim 0$ MHz). Meanwhile, the weak pumping beam P_1 (P_2) is scanned across cesium $6S_{1/2}$ F = 4 – $6P_{1/2}$ F' = 3, 4 transitions in D_1 line ($6S_{1/2}$ F = 4 – $6P_{3/2}$ F'' = 3, 4 transitions in D_2 line).When it is resonant with the same velocity group atoms (i.e. ν ~0), the population of state |1⟩ will be transferred to another hyperfine fold ($6S_{1/2}$ F = 3) of ground state due to optical pumping effect of single-resonance pumping laser. Because the decreased population weaken the absorption of the probe/coupling beam at $\Delta_C \sim 0$, then the Doppler-free SROP with high SNR and narrow linewidth would appear on the flat background of transmission signal. Similarly, this kind of the transmission signal (called DROP) can be obtained when both of the lasers are double resonant with the atoms which are two-photon optical pumped from ground state to the highest excited state then decay spontaneously, see Fig. 1(B).

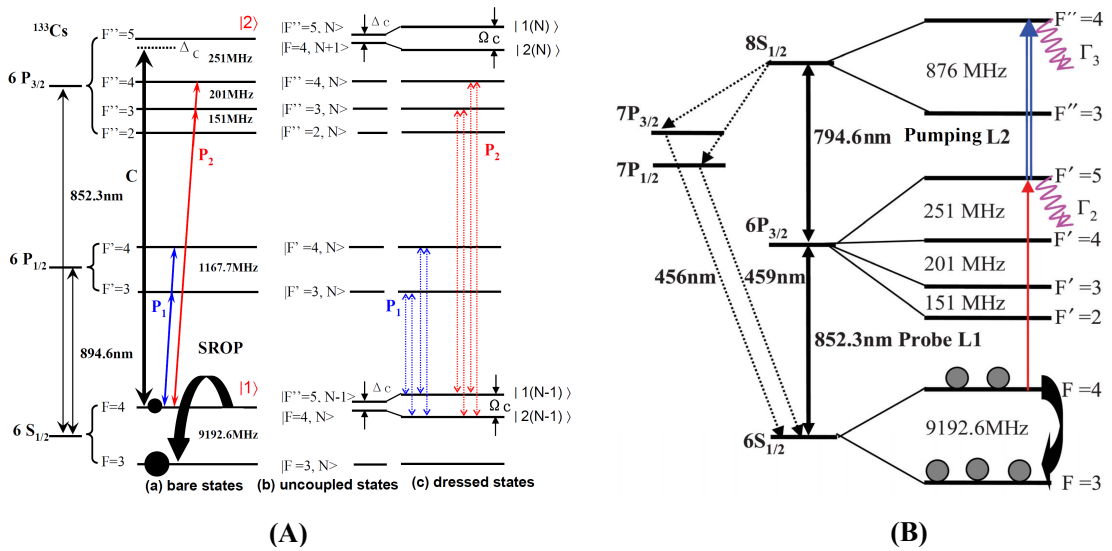


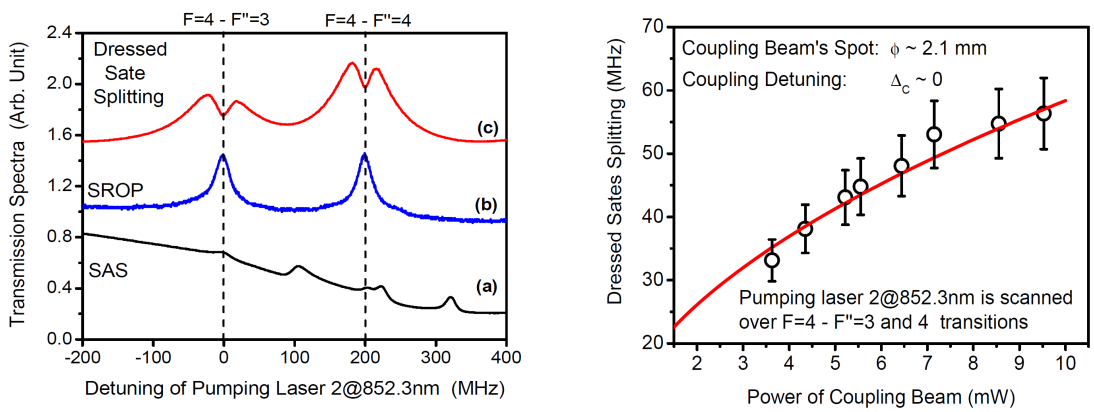
Fig. 1 (Color online) **(A)** Relevant hyperfine levels of cesium for SROP. (a) Bare states. (b) Uncoupled states. (c) Dressed states. Coupling/Probe laser (C) is locked to |1⟩ - |2⟩ cycling hyperfine transition with frequency detuning of Δc and Rabi frequency of Ωc. Pumping laser P_1 is scanned across F=4 – F'=3, 4 (or F=3 – F'=3, 4) transitions in D_1 line. Pumping laser P_2 is scanned across F=4 – F''=3, 4 (or F=3 – F''=3, 4) transitions in D_2 line. **(B)** Relevant hyperfine levels of cesium for DROP. Probe laser is locked to F=4 – F'=5 cycling hyperfine transition. Pumping laser is scanned across F'=5 – F''=4 excited state transition.

It should be noted that just because the probe/coupling is locked to a cycling hyperfine transition, not only the population redistribution of the ground state $6S_{1/2}$ F = 4 is farthest avoided, but also the probe laser can be strong enough to dress the atoms. In addition, it is also made the probe/coupling laser is capable to selectively detect the atoms in ground state with the relative velocity of Δ_C/k_C (where $k_C = 2\pi/\lambda_C$ and λ_C is the wavelength of the probe/coupling laser). Consequently, it is available for us to measure the dressed-state splitting of ground state by utilizing SROP with room-temperature atomic vapor cell and to investigate the off-resonant DROP spectra and its application in two-color cesium MOT [6, 12].

3. EXPERIMENTAL RESULTS AND ANALYSIS

3.1 SROP with cesium atoms and measurement of the dressed-state splitting of ground state

The experiment setup can be found in Fig. 2 in ref. [7]. Due to the probe/coupling is locked to a cycling transition, when the coupling beam is strong ($\Omega_C >> \Gamma_{21}$, $\Gamma_{21}=2\pi$ x 5.22 MHz is the spontaneous decay rate of cesium F'=5 – F=4 transition), both of the bare state$|1\rangle$ and $|2\rangle$ will split into two dressed states, as shown in (c) of Fig. 1(A). According to the dressed-atom approach [11], when the detuning of the probe/coupling laser $\Delta_C \sim 0$, two dressed states of the ground state $|1\rangle$ will symmetrically distribute at the both sides of $6S_{1/2}$ F = 4 hyperfine fold, as shown in curve (c) of Fig. 2 (A). The frequency interval of the dressed state splitting can be seen clearly in curve (c) of Fig. 2 (A). It changed with the optical power of the probe/coupling beam, which is in good agreement with the equation $\Omega = \Gamma_{21}\,[I/(2I_s)]$ with no free parameter (I is the coupling beam's intensity, I_s is the saturation intensity), as shown in Fig. 2(B).



(A) **(B)**

Fig. 2 (Color online) **(A)** Curves (b) and (c) show the SROP spectra corresponding to cesium F=4 - F''=3, 4 transitions with different Ωc, while the curve (a) shows the saturated absorption spectra (SAS) for calibration. The pumping beam's power is ~ 10 μW@852.3nm ($\Omega p \sim 0.4\ \Gamma_{21}$) for the curves (b) and (c). The coupling beam's power is ~ 74 μW@852.3nm ($\Omega c \sim \Gamma_{21}$) for the curve (b) and ~ 4.3 mW @852.3nm ($\Omega c \sim 7.5\ \Gamma_{21}$) for the curve (c). The up-down shift of the curves (b) and (c) does not mean that transmission baseline changes and just for convenient comparison. **(B)** The splitting of dressed ground state $6S_{1/2}$ F=4 is function of the coupling beam's power ($\Delta c \sim 0$). The circles are experimental data with ±2.5% of error bar. The solid line represents for theoretical predictions.

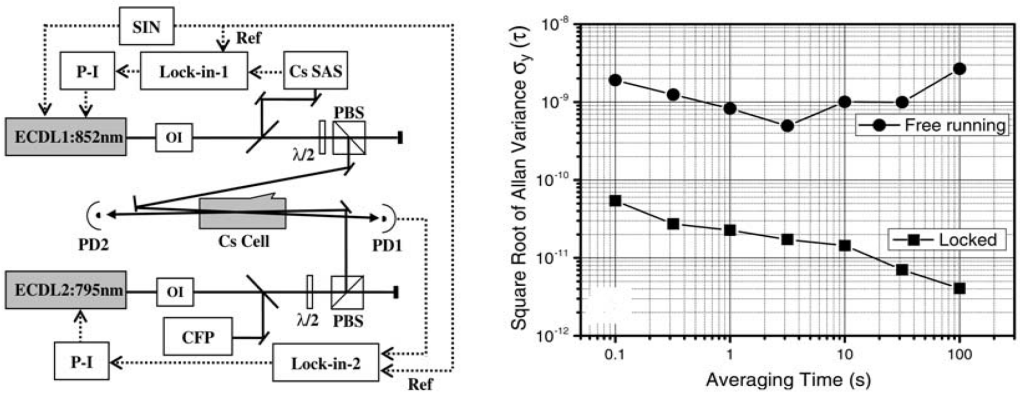
If the probe/coupling laser is weak enough, the normal SROP with flat background and higher SNR can be obtained as shown in the curve (b) of Fig. 2(A). Additionally, the crossover resonance lines which appeared on the saturated absorption spectra (SAS) in the curve (a) of Fig. 2(A), disappear in SROP spectra due to the feature of velocity-selection. As a result, the spectral resolution can be improved, which is benefit for the application of the precision measurement of atomic hyperfine structure.

Utilizing SROP scheme one can also measure the structure of atomic hyperfine excited state [7] and realize the frequency stabilization of the pumping laser without direct modulation as we have done with DROP [2].

3.2 DROP for cesium atoms and applications

3.2.1 Laser frequency stabilization to excited state transition and two-color cesium MOT

In some physical fields, such as high-resolution molecular and atomic spectroscopy, laser cooling and trapping of atoms, and optical fiber telecommunication, laser frequency stabilization is very important. An absolute frequency reference (a transition line of atoms or molecules) is normally required for laser frequency stabilization. Doppler-free spectrum of optical-optical double resonance (OODR) has been used widely before. However, OODR normally has quite weak signal [1] and bad SNR, these limit application in frequency stabilization and other fields.



(a) (b)

Fig. 3 (a) Experimental setup for OODR and DROP as well as laser frequency stabilization of the ECDL@795nm by DROP. SIN: sine-wave signal generator; Lock-in :lock-in amplifier; OI: optical isolator; CFP: confocal Fabry-Perot cavity; PD: photodiode; P-I: proportion and integration amplifier; λ/2: half-wave plate; PBS: polarizing beam splitter cube; SAS: saturated absorption spectroscopy; Ref: reference channel of lock-in amplifier. **(b)** Square root of the Allan variance for the case of the ECDL2@795nm free running and after being locked.

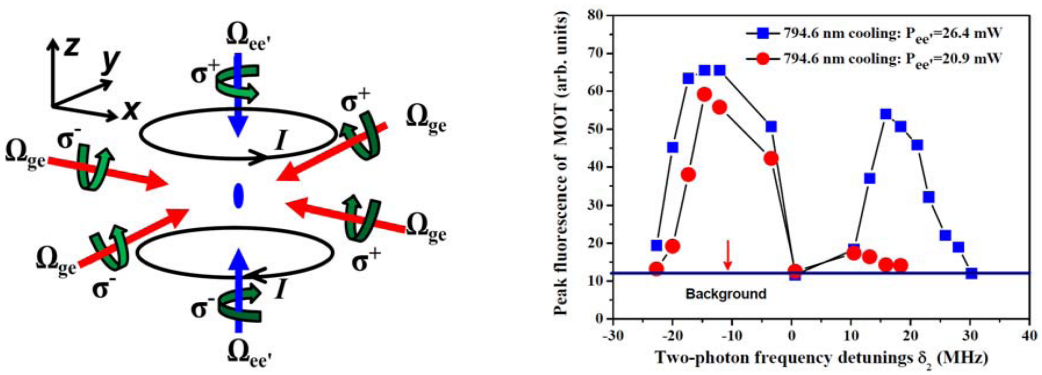
Based on the key idea of two-photon optical pumping and the population variation detection of the ground state, instead of the excited state, DROP has a flat background and higher SNR compared to OODR [2]. It is known that a spectrum with a narrow line-width $\Delta\nu$ and a high SNR is important for laser frequency stabilization ($\sigma_\nu(\tau) \propto \Delta\nu/\mathrm{SNR}$).

Fig. 3(a) shows the experimental scheme for the observation of OODR and DROP spectra of cesium atoms, and also for the frequency stabilization of the pumping laser (ECDL2@795 nm) by using of DROP. The detailed

experimental process can be found in Ref. [2]. In experiment, the probe laser (ECDL1@852 nm) is locked to cesium 6 $S_{1/2}$ F = 4 – 6 $P_{3/2}$ F' = 5 cycling hyperfine transition by SAS scheme, in which the error signal is generated by a lock-in amplifier with 14 kHz sine-wave frequency modulation, while the pumping laser (ECDL2@795 nm) is scanned across $6P_{3/2}$–$8S_{1/2}$ transition. In fact, this sine-wave frequency modulation for probe laser will be transfer automatically to the DROP spectra due to the two-photon optical pumping effect. As a result, a dispersion-like signal without the direct modulation of the pumping laser's frequency can be obtained to stabilize the pumping laser. Compared with the free running case, the frequency stability of the pumping laser after being locked is remarkably improved, as shown in Fig. 3(b). The lowest square root of the Allan variance is $\sigma_y(\tau) \sim 4.1 \times 10^{-12}$ with an averaged time $\tau = 100$ s.

3.2.2 Two-color cesium MOT

Additionally, as the probe laser of DROP can be locked to a cycling transition with certain frequency detuning, when it is detuned from the lower transition, the pumping laser is oppositely detuned from the upper transition to meet the two-photon resonance. This provides us an effective method to offset lock the pumping laser utilizing the same technique introduced above by employing DROP spectra under off-resonance condition. The two-photon detuning can be controlled conveniently by changing the probe laser's detuning. We have applied this offset locking scheme into frequency control system of a new-type cesium MOT configuration – two-color MOT [6, 12]. It allows completely background-free detection of laser-induced- fluorescence (LIF) photons of trapped cold atoms in a MOT, especially in a single atoms MOT [13].



(a) (b)

Fig. 4 (Color online) **(a)** Two-color cesium MOT configuration. **(b)** Peak fluorescence of cold atoms trapped in the two-color cesium MOT vs the two-photon frequency detuning δ_2. The solid lines are only for guiding eyes.

The two-color cesium MOT configuration is shown in Fig. 4(a), which is similar to that in Ref. [12]. It employs the radiation force came from |e⟩ (cesium $6P_{3/2}$ F'=5) - |e'⟩ (cesium $8S_{1/2}$ F''=4) transition along one axis of MOT (here ***z*** axis, see Fig. 4(a)), and that from |g⟩ (cesium $6S_{1/2}$ F=4) - |e⟩ (cesium $6P_{3/2}$ F'=5) transition along other two axis (here ***x*** and ***y*** axis). It is clearly different from the traditional standard MOT in which the atoms are cooled and trapped by the radiation force of |g⟩ - |e⟩ transition along all three axis.

We plot the peak fluorescence of cold cesium cloud vs the two-photon detuning δ_2 in Fig. 4(b). It clearly indicates

that two-color cesium MOT can efficiently trap atoms in both cases of red and blue two-photon detuning. One point should be addressed here that it is quite different to the standard MOT in which the Doppler and sub-Doppler cooling mechanisms require red detuning. When the 794.6 nm cooling/trapping laser beam along z axis is ~ 20.9 mW, the two-color cesium MOT still can cool and trap atoms at up to ~ + 15 MHz of two-photon blue detuning. Further, when the 794.6 nm cooling/trapping laser beam along z axis is increased to a higher level (for example, ~ 26.4 mW), the two-photon blue detuning remarkably extends up to ~ + 30 MHz. The highest fluorescence intensity at δ_2 ~ -14.6 MHz was found in Fig. 4(b). In this case ~ 5 x 10^6 cesium atoms are trapped in our two-color cesium MOT and typical effective temperature of cold cloud is 70 ~ 100 μK. The result is very different to conventional 852.3 nm cesium MOT but similar to ref. [12].

With the red two-photon detuning, the cooling/trapping mechanism of the two-color MOT can be understood using two-photon Doppler cooling picture which is similar to the standard MOT. With the blue two-photon detuning, the cooling/trapping mechanism is now not very clear and should be investigated deeply, and it probably can be explained using two-color polarization gradient cooling mechanism. This two-color MOT may be used in the background-free detection of LIF photons of trapped few atoms even single atom in MOT [13]. And in principle it also probably can be extended to directly trapping Rydberg cold atoms when the upper laser couples the intermediate excited state to Rydberg state. The detailed investigation and results will publish in another paper [6].

3.2.3 Atomic coherence in DROP with ladder-type cesium system

EIT in a ladder-type atomic system has been studied widely and it can be understood by the physical picture of dark state or the quantum interference between the different atomic internal one-photon path and two-photon path [14]. Recently, DROP spectrum based on velocity-selective population detection and two-photon optical pumping in a ladder-type atomic system has been developed and been used in many kinds of fields. The main difference in experiment between typical EIT and DROP in a ladder-type atomic system is, the probe laser which couples the lower transition is swept and the coupling laser which couples the upper transition is locked in typical EIT, but the probe laser is locked and the coupling laser is swept in DROP. According to the equation (1) (refer to Ref. [9]), if the wavelength of two lasers in DROP scheme are matched (i.e., $\omega_{probe} \sim \omega_{pumping}$), and they are overlapped and counter-propagated with each other, the two-photon Doppler-free condition will be fulfilled and ladder-type EIT caused by the atomic coherence will exist no matter whether the two lasers are locked or swept.

$$\chi(\upsilon)d\upsilon = \frac{(4i\hbar g_{21}^2/\varepsilon_0)N(\upsilon)d\upsilon}{\gamma_{21} - i\Delta_1 - i\frac{\omega_{probe}}{c}\upsilon + \frac{\Omega_C/4}{\gamma_{31} - i(\Delta_1 + \Delta_2) - i(\omega_{probe} \pm \omega_{pumping})\upsilon/c}} \quad (1)$$

Although we can obtain the DROP spectra in both co-propagation (CP) and counter-propagation (CTP) configurations of two beams, EIT will happened only in the case of the CTP configuration. This has been demonstrated experimentally as shown in Fig. 5. Compared with DROP for CP configuration, DROP spectrum for CTP configuration has a little bit higher magnitude and much narrower line-width due to EIT. The similar phenomena in Λ- and V-type atomic systems have been studied as well [15, 16].

Furthermore, we can adjust the EIT effect mixed in DROP by optimize the intensity of the two laser beams. This provides us a method to obtain a new type of EIT without Doppler background. In particular experiment case, DROP

spectrum for the CTP configuration clearly shows two components: the narrow part due to EIT and the broad part due to DROP [9]. But the DROP spectrum with two different components is never seen in the CP configuration.

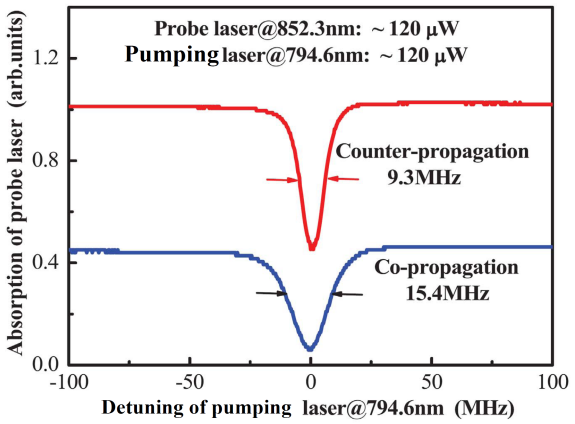


Fig. 5 (Color online) DROP spectrum of cesium $6P_{2/3}$ F' = 5 - $8S_{1/2}$ F" = 4 hyperfine transition under the same intensity of the pumping and probe laser beams for CTP and CP configurations.

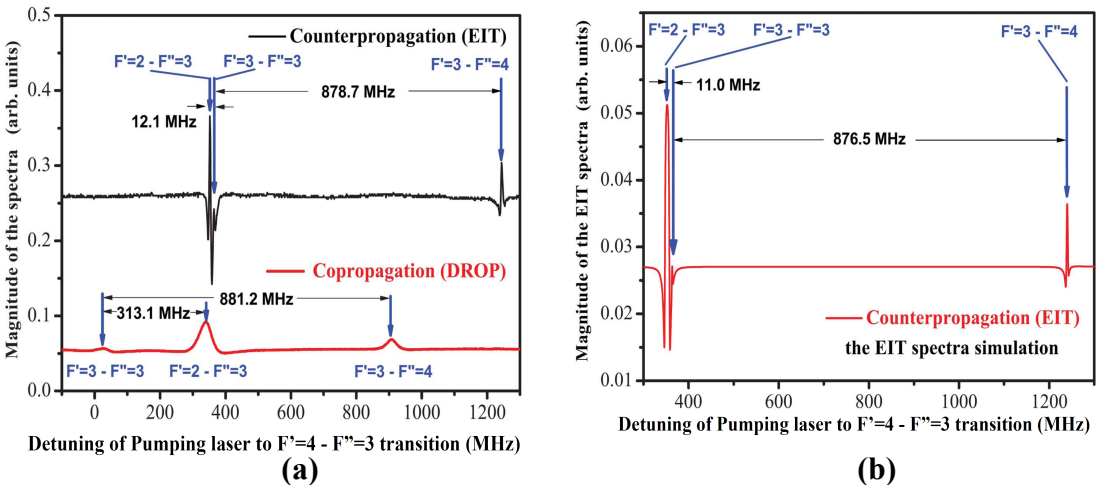


Fig. 6 (Color online) **(a)** EIT spectra for CTP configuration without Doppler background and DROP spectra for CP configuration. The probe laser is locked to the $6S_{1/2}$ F = 3 - $6P_{3/2}$ F' = 2 transition, while the pumping laser is scanned over the $6P_{3/2}$ - $8S_{1/2}$ transition. **(b)** Simulation results for EIT spectra for CTP configuration in (a) based on a multilevel ladder-type model.

We also studied the new type of EIT in a multilevel ladder-type system. It is known that two-photon Doppler-free condition will be fulfilled even if the locked laser is detuned to one of the atomic transition in ladder-type system. Take cesium D_2 line for example; there are four hyperfine levels. When the system $6S_{1/2}$ F = 3 - $6P_{3/2}$ F' = 2 - $8S_{1/2}$ F" = 3 is used for the resonant EIT, in which the probe laser is locked to $6S_{1/2}$ F = 3 - $6P_{3/2}$ F' = 2 transition while the upper laser is scanned across the $6P_{3/2}$ - $8S_{1/2}$ transition, some atoms with particular none-zero velocity will also be populated on the

neighborhood hyperfine levels, such as $6P_{3/2}$ F' = 3 in our experiment (F' = 3 is far from $6P_{3/2}$ F' = 2 are negligible due to little contribution to the spectra). As a result, the $6S_{1/2}$ F = 3 - $6P_{3/2}$ F'=3 - $8S_{1/2}$ F" =3(4) system will be −151.2 MHz detuning EIT signal with respect to the $6S_{1/2}$ F = 3 - $6P_{3/2}$ F' = 2 transition. Meanwhile, these atoms with none-zero velocity feel different detuning relative to two lasers, which also leads to the different structure of the DROP for CP configuration and multilevel EIT for CTP configuration. As shown in Fig. 6(a) and (b), from which we can clearly see that the experimental results and numerical simulation are in good agreement. In addition, the measured frequency intervals are insensitive to frequency detuning of the probe laser beam due to the feature of velocity-selective detection. Compared with DROP spectra, the new type EIT spectra for the CTP configuration has a higher SNR and a narrower line-width. Thus it can be applied to improve measurement's precision of the hyperfine splitting and the hyperfine structure constant.

4. CONCLUSION

We have introduced two schemes of the Doppler-free velocity-selective optical-pumping spectroscopy, named SROP and DROP, which have a flat background and high SNR. Although they are suitable for the different atomic systems, V-type for SROP and ladder-type for DROP, both of them are based on the velocity-selective detection and optical pumping. SROP and DROP spectra can provide us a good absolute frequency standard which can be used to improve the measurement's precision of atomic hyperfine splitting. With the help of SROP, we realize the measurement of the dressed-state splitting of ground state with room-temperature cesium vapor cell, and find that the Doppler shift will compensate the frequency detuning of the locked laser and the frequency intervals of the dressed-state splitting will not change with the detuning. This is also suitable for DROP (EIT) spectroscopy under off-resonant condition. We have also developed a frequency stabilization method by the way of modulation transfer using DROP. It has been applied in 1.5 μm DFB diode laser frequency stabilization by DROP spectrum of rubidium $5S_{1/2}$ - $5P_{3/2}$ - $4D_{3/2}$ transitions. We applied DROP for cesium $6S_{1/2}$ - $6P_{3/2}$ - $8S_{1/2}$ transitions in two-color cesium MOT in ultimate purpose of suppressing the background of a single atom MOT. We have also investigated EIT and DROP in ladder-type atomic system with the different configurations (CP and CTP) of two laser beams in atomic vapor cell.

ACKNOWLEDGMENT

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