Single-resonance optical pumping spectroscopy and application in dressed-state measurement with atomic vapor cell at room temperature

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Abstract: By monitoring the transmission of probe laser beam (also served as coupling laser beam) which is locked to a cycling hyperfine transition of cesium D_2 line, while pumping laser is scanned across cesium D_1 or D_2 lines, the single-resonance optical pumping (SROP) spectra are obtained with atomic vapor cell. The SROP spectra indicate the variation of the zero-velocity atoms population of one hyperfine fold of ground state, which is optically pumped into another hyperfine fold of ground state by pumping laser. With the virtue of Doppler-free linewidth, high signal-to-noise ratio (SNR), flat background and elimination of crossover resonance lines (CRLs), the SROP spectra with atomic vapor cell around room temperature can be employed to measure dressed-state splitting of ground state, which is normally detected with laser-cooled atomic sample only, even if the dressed-state splitting is much smaller than the Doppler-broaden linewidth at room temperature.

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OCIS codes: (300.6320) Spectroscopy, high-resolution; (020.2930) Hyperfine structure; (020.6580) Stark effect

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1. Introduction

High-resolution laser spectroscopy has important application in atomic and molecular physics, quantum optics, quantum metrology and so on. It is a very important tool for laser frequency stabilization [1, 2] and precision measurement of atomic hyperfine structure [3] and relevant important physical constants [4]. Normally, in atomic or molecular vapor cell, the spectral resolution will be obscured by the Doppler effect. Consequently, several kinds of the Doppler-free laser spectroscopy have been developed [1, 5–8]. One kind of the Doppler-free velocity-selective optical-pumping spectroscopy (VSOP) techniques can be used to eliminate crossover resonance lines (CRLs) which often appear on the saturated absorption spectroscopy (SAS) in atomic vapor [9]. In Ref [10], the structure of VSOP for the running multilevel atoms interacted with two co-propagated beams was studied with an atomic vapor cell. Additionally, once the two-photon Doppler-free condition is fulfilled by arranging different propagating configurations of two beams, the atomic coherence can reduce line-width of the spectrum, for example, co-propagation configuration for V-type three-level system while the very reverse for ladder-type [11,12]. However, if ones only pay attention to optical pumping and do not care for the atomic coherence, the direction of two overlapped beams could not be attention-getting. Based on the cascade two-photon resonance and optical pumping effect caused by two counter-propagating beams which interact with the ladder-type atomic system, another new type of the Doppler-free VSOP techniques, double-resonance optical pumping (DROP), was developed recently [1, 2]. It is much interested that the DROP reveals the physics of the intermediate excited state by detecting the population variation in ground state. So far, DROP has been applied in laser frequency stabilization [1, 2] and the precision measurement of Rydberg atomic level [13].

The dressed state picture is very helpful in quantum interpretation of some phenomena such as destructive interference in electromagnetically-induced transparency [14], double-dark resonances [15] and so on. Consequently, the precision measurement of the dressed state turns to be very important and has been studied widely [16, 17]. Although the above-mentioned DROP technique could be used to detect the dressed state of the excited atoms, unfortunately, it is not suitable for the dressed ground-state atoms with room-temperature atomic vapor cell, unless the dressed splitting is larger than the Doppler-broaden linewidth (about several hundred MHz around room temperature).

In our experiment, we lock the coupling laser to a cycling transition between the ground state and excited state. While the weak pumping beam is co-propagating with the coupling beam and resonant with the transition between ground state and another excited state, the zero-velocity atoms' population of one hyperfine fold of ground state will be partially pumped into another hyperfine fold. As a result, this changed population of hyperfine atomic ground state yields an obvious peak of the coupling-transmission, which we called the single-resonance optical pumping (SROP) spectrum by analogy with DROP. It is also similar to that kind of the VSOP in Ref. 9. However, in SROP only the swept pumping beam optically pumps the atoms to another hyperfine fold of ground state and redistributes the population, instead both of two lasers in Ref.9. The coupling laser is locked to a cycling transition for the purpose of probe the population variation of the ground-state atoms in a particular velocity. The SROP can be used to stabilize laser frequency [18] and to measure hyperfine splitting

accurately [9]. More importantly, it is turned to be possible the dressed-state splitting of ground state by the strong coupling laser can be measured by utilizing SROP 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 linewidth at room temperature.

In this paper, after the introduction of SROP, we analyzed some relevant experimental results. Then we measured the dressed ground state splitting with room-temperature cesium vapor cell by using SROP on cesium D_1 (894nm) and D_2 (852nm) lines. Interestingly, the experimental results of the dressed splitting at nonzero frequency detuning of the coupling laser were presented, which are different from the case of the dressed atoms confined in a MOT.

2. SROP spectroscopy

As shown in Fig. 1, cesium $6S_{1/2}$ F = 4 and $6P_{3/2}$ F" = 5 is labeled as the state |1> and |2>, respectively. The weak coupling laser (C in Fig. 1, $\Omega c < \Gamma_{21}$, $\Gamma_{21} = 2\pi X5.22$ MHz, is the spontaneous decay rate of cesium D_2 line) is locked to |1> - |2> cycling transition with $\Delta c \sim 0$ (Δc is the coupling laser's frequency detuning to |1> - |2>), by which the population variation of the atoms (in state |1>) moving perpendicular to the direction of the coupling beam is probed. In this way, the population redistribution of the ground state $6S_{1/2}$ F = 4 caused by the coupling beam is furthest avoided, but the energy level shift due to ac-Stark effect caused by it is also reduced. Meanwhile, the weak pumping beam P_1 (P_2) is scanned across transitions $6S_{1/2}$ F = 4 – $6P_{1/2}$ F' = 3, 4 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 atoms in the same velocity group (i.e. v ~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 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.

When the coupling beam is strong ($\Omega c \gg \Gamma_{21}$), both of the bare state |1> and |2> will split into two dressed states, as shown in Fig. 1. According to the dressed-atom approach [19], two dressed states of the ground state |1> can be depicted as follows:

$$\left|1\left(N-1\right)\right\rangle = \sin\theta\left|6S_{1/2}F = 4, N\right\rangle + \cos\theta\left|6P_{3/2}F'' = 5, N-1\right\rangle \quad (1)$$

$$\left|2\left(N-1\right)\right\rangle = \cos\theta\left|6S_{1/2}F = 4, N\right\rangle - \sin\theta\left|6P_{3/2}F'' = 5, N-1\right\rangle \quad (2)$$

here $2\theta = \arctan(-\Omega_C/\Delta_C), 0 \leq 2\theta < \pi$. When the detuning of the coupling laser $\Delta c \sim 0$, the dressed states |1(N-1)> and |2(N-1)> with the splitting space $\Omega = \Omega c$ deduced from the equation $\Omega = \sqrt{\Delta_C^2 + \Omega_C^2}$ will symmetrically distribute at the both sides of hyperfine fold $6S_{1/2}$ F = 4 in ground state according to the Eqs. (1) and (2). The dress-state splitting will also be demonstrated in SROP spectra.
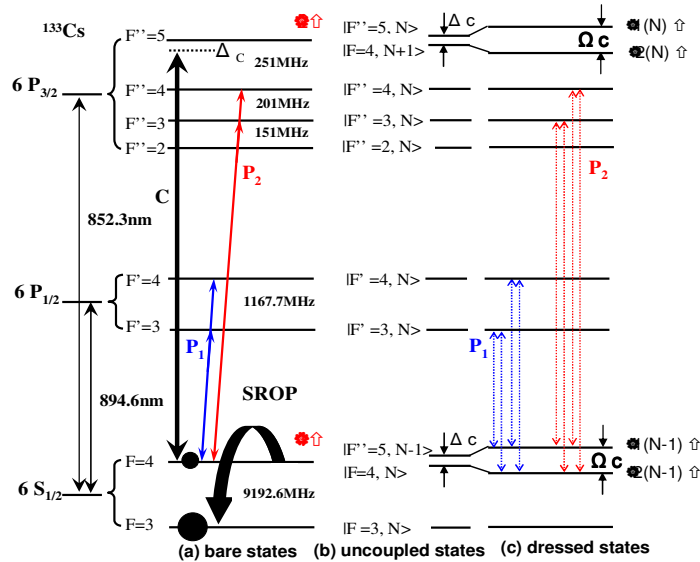
Fig. 1. (a) Relevant hyperfine levels of cesium bare states. (b) Uncoupled states. (c) Dressed states. Coupling laser (C) is locked to |1> - |2> cycling transition with frequency detuning of $\Delta c \sim 0$ and Rabi frequency of Ωc by utilizing modulation-free polarization spectroscopic technique. Pumping laser P_1 is scanned across transitions F = 4 – F' = 3, 4 (or F = 3 – F' = 3, 4) in D_1 line. Pumping laser P_2 is scanned across F = 4 – F'' = 3, 4 transitions in D_2 line.

3. Experimental setup

The schematic diagram of the experiment setup is depicted in Fig. 2. The coupling beam and pumping beam co-propagate through a cesium vapor cell (ϕ20mmX40mm) at room temperature.
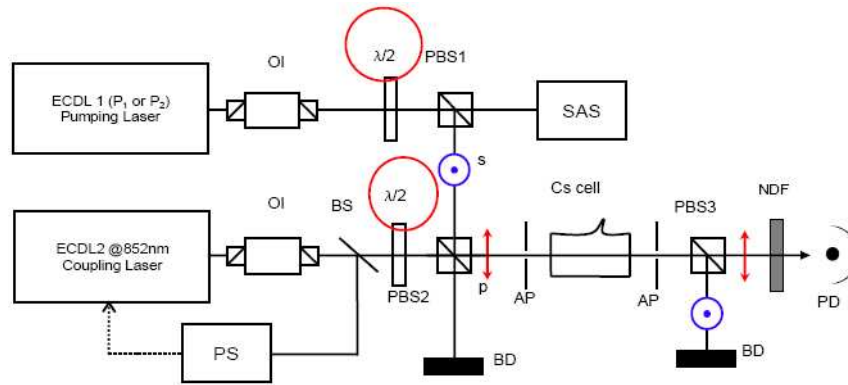

Fig. 2. Schematic diagram of experiment setup. The solid lines are for the optical path, and the dot line for the electronic connection. The key to figure: ECDL: external-cavity diode laser; OI: optical isolator; SAS: saturated-absorption-spectroscopy device; PS: polarization-spectroscopy device; BS: beam-splitting plate; PBS: wide-band polarization-beam-splitting cube; λ/2: half-wave plate; AP: aperture; NDF: neutral density filter; PD: photodiode; BD: laser beam dump; s: s polarization; p: p polarization.

After the output beam of pumping laser (grating external-cavity diode laser: ECDL, P_1@894nm or P_2@852nm) passing through the combination of a half wave-plate and a broad-band (800-920nm) polarization-beam-splitting cube PBS_1, it is separated into two parts whose power could be adjusted. One part is set to SAS device for frequency calibration. Another part is overlapped via broad-band PBS_2 with the co-propagated coupling beam through the

cesium cell. The cell is slightly deflective to avoid the diffuse reflection. The diameters of the pumping and coupling beams are set ~2mm by apertures (APs).

The coupling beam (C) is provided by an 852-nm ECDL and is locked to cesium $6S_{1/2}$ F = 4 – $6P_{3/2}$ F'' = 5 cycling transition utilizing the modulation-free polarization spectroscopy technique. We can also regulate the coupling beam's frequency detuning Δc by an AOM (Acousto-Optical Modulator) and change its optical intensity. The transmitted coupling beam is separated from pumping beam via broad-band PBS3 and collected by a photo-detector (PD, NewFocus-Model 2001) whose output (SROP signal) is observed and registered by a digital oscilloscope.

4. Experimental results and analysis

4.1 SROP for cesium D_1 line when Δc ~0

When the pumping laser is scanned across cesium $6S_{1/2}$ F = 4 (or 3) – $6P_{1/2}$ F' = 3, 4 hyperfine transitions in D_1 line, it leads to variation of transmission intensity of the coupling beam, and thus the Doppler-free SROP peaks (or dips) appear on the position where single resonance happen. As indicated in trace (b) in Fig. 3, the two enhanced transmission peaks at left hand correspond to $6S_{1/2}$ F = 4 – $6P_{1/2}$ F' = 3, 4 transitions, while the other two enhanced absorption dips at right-hand correspond to $6S_{1/2}$ F = 3 – $6P_{1/2}$ F' = 3, 4 transitions. SAS spectra are showed in trace (a) in Fig. 3 for frequency calibration.
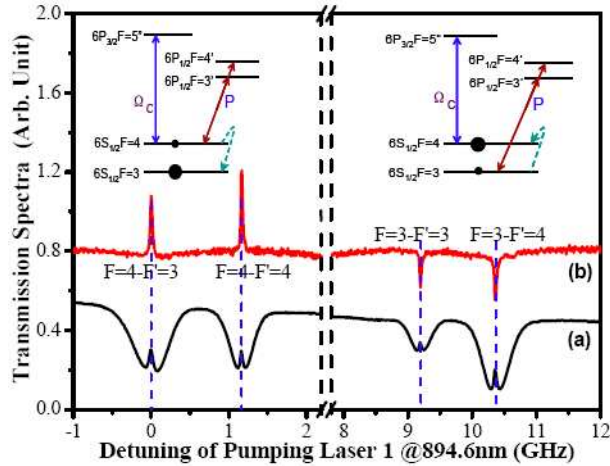

Fig. 3. Trace (b) shows SROP spectra corresponding to F = 4 –F' = 3, 4 (and F = 3 –F' = 3, 4) transitions in cesium D_1 line. The two peaks on left hand present enhanced transmission while the other two peaks on right hand for enhanced absorption. Insets give two different pictures for single-resonance optical pumping, in which the black spots indicate the atomic population. The relevant SAS shown in trace (a) is for frequency calibration.

We fix optical power of the pumping laser P_1 at 9.3 μW (Ωp ~0.3 Γ_{31}) and coupling laser's frequency detuning Δc ~0. High SNR Doppler-free SROP spectra is shown as trace (2) in Fig. 4(a). The linewidth of the SROP spectrum is mainly due to power broadening of the coupling laser. When the atoms in ground state $6S_{1/2}$ F = 4 are dressed by the coupling beam, the population of atoms with four different non-zero-velocity groups decreases due to optical pumping effect, once $\omega_P = \omega'_{43} \pm \Omega_C / 2$ and $\omega_P = \omega'_{44} \pm \Omega_C / 2$ (ω_p is angular frequency of the swept pumping laser, ω'_{43} is the angular frequency corresponding to cesium $6S_{1/2}$ F = 4 – $6P_{1/2}$ F' = 3 transition while ω'_{44} corresponds to cesium $6S_{1/2}$ F = 4 – $6P_{1/2}$ F' = 4 transition). Thus the enhanced transmission peaks are symmetrically distributed at two sides of $6S_{1/2}$ F = 4 state as shown in trace (3) in Fig. 4(a). In addition, the linewidth and frequency fluctuation of two lasers will contribute to SNR and linewidth of SROP spectra.
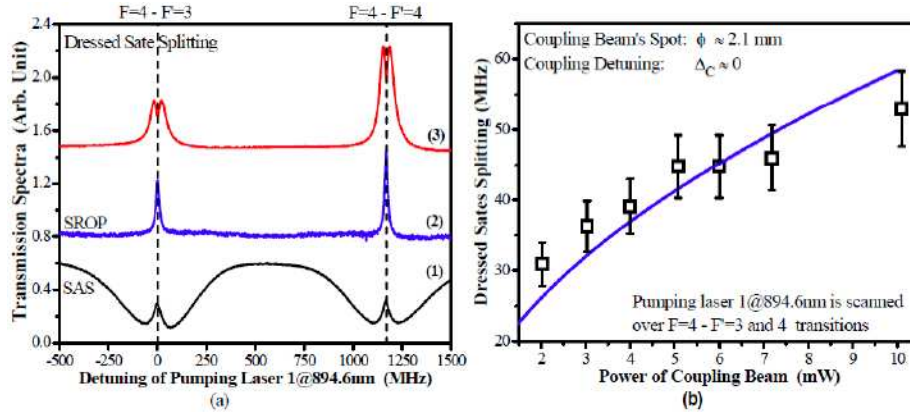
Fig. 4. **(a)** Trace (2) and (3) show the SROP spectra corresponding to cesium F = 4 –F' = 3, 4 transitions in D_1 line with different Ωc while the trace (1) shows the relevant SAS for frequency calibration. The pumping beam's power is ~9.3μW@894.6nm (Ωp ~0.3 Γ_{31}, Γ_{31} = 2πx4.56 MHz is the spontaneous decay rate of cesium D_1 line) for both of trace (2) and (3). The coupling beam's power is ~74μW@852.3nm (Ωc ~Γ_{21}) for trace (2) while ~3mW@852.3nm (Ωc ~6.3 Γ_{21}) for trace (3). The up-down shift of traces (2) and (3) 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 (Δc ~0). The hollow squares are experimental data with $\pm 2.5\%$ error bar. The solid line represents for theoretical curve.

As shown in Fig. 4(b), the splitting between the two dressed states (see trace (3) in Fig. 4(a)) vary with optical power of the coupling beam. Obviously here the dressed-state splitting for |1> ground state is much smaller than the Doppler-broaden linewidth (~500 MHz) at room temperature, and normally it is difficult to observe with atomic vapor cell because of the Doppler broadening. But it can be done with laser cooled atomic sample via simple absorption spectroscopy. Now it is clearly observed by using SROP technique.

The hollow squares in Fig. 4(b) are experimental data. The solid line is the fitting based on $\Omega = \Gamma_{21}\sqrt{I/2I_S}$ with no free parameter (I is intensity of the coupling beam, Is is the saturation intensity). The error bars are for $\pm$ 2.5% errors due to the nonlinearity of piezo-electrical transducer (PZT) in ECDL and the error of searching the peak values in trace (3) in Fig. 4(a). In future we will reduce the errors of searching peaks by utilizing derivation of SROP spectrum.

4.2 SROP for cesium D_1 line when $\Delta c \neq 0$

The SROP with Δ_C~0 introduced above is also suitable for the situation $\Delta c \neq 0$, except that the peak position of probe beam will move together with Δc at the given Ωc away from the SAS. This is also similar to the dressed atoms. More interestingly, when the locked coupling laser is strong, because of the Doppler shift, it will be resonant with another group of atoms whose velocity is Δ_C/k_C (where $k_C = 2\pi/\lambda_C$ and λ_C is the wavelength of the coupling laser) along the direction of probe beam and dress them. By utilizing the SROP technique, only these atoms with a particular velocity $\Delta c/k_c$ is selectively probed by the locked coupling beam when the pumping laser is swept. As a result, the double peaks of the dressed atoms appear on SROP at Δc distant from the Lamb dip of SAS. This can be proved experimentally as shown in Fig. 5. The offset between the center of the dressed splitting in SROP and the Lamb dip in SAS changes linearly along with Δc. Inserts are the experimental pictures of SROP and SAS obtained in different Δc at the given Ωc. Additionally, we find that the altitude of the SROP will decrease along with the increased Δc due to the Maxwell-Boltzmann velocity distribution of atoms.
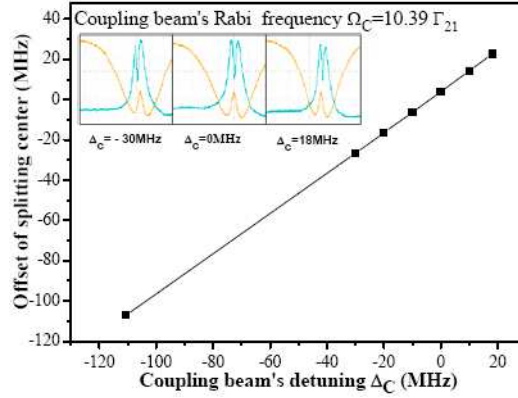
Fig. 5. The offset between the center of the dressed splitting in SROP and the Lamb dip in SAS change linearly along with Δc. Solid rectangles are the experimental data. Inserts are the experimental pictures of SROP and SAS obtained in different Δc at given Ωc.
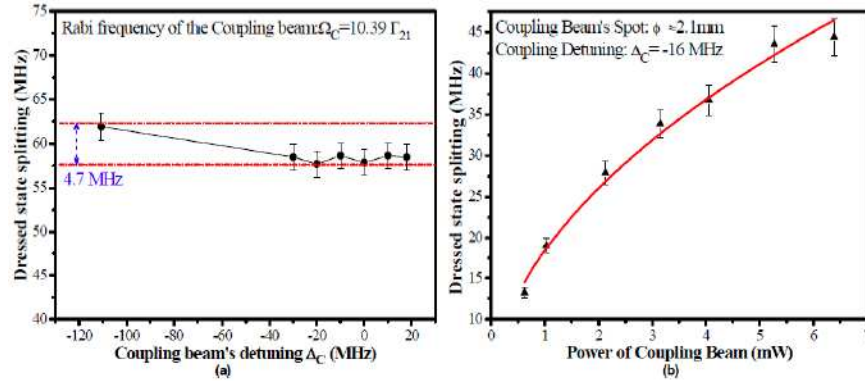

Fig. 6. **(a)** Dressed state splitting around a fixed value Ωc nearly unchanged in different Δc. Solid circles with $\pm 2.5\%$ error bar are experimental data. About 8.7% difference between them (4.7MHz) at the range of ~140MHz maybe caused mainly by the error of the power and the frequency drift of the coupling laser. **(b)** Dressed state splitting of the ground state $6S_{1/2}$ F = 4 is function of the coupling beam's power (Δc = −16 MHz). The solid triangles are experimental data with ± 2.5% error bar. The solid line represents for theoretical curve according to $\Omega = \Gamma_{21}\sqrt{I/2I_S}$.

The dressed state splitting is mainly controlled by Ωc and is independent on Δc. As the Doppler shift compensates the frequency detuning of the coupling laser, the atoms with a certain velocity group are exactly resonated with the coupling laser and thus are selected to contribute to the dressed state splitting. As shown in Fig. 6(a), the splitting is nearly unchanged at the given Ωc. This is very different from the case of the dressed atoms confined in a MOT. The atoms in MOT will be not exactly resonated with the coupling laser with a frequency detuning Δc.

If we fix the frequency detuning of the coupling laser, the splitting of the dressed ground state $6S_{1/2}$ F = 4 of the atoms with velocity Δc/kc is in good agreement with the equation $\Omega = \Gamma_{21}\sqrt{I/2I_S}$ as the solid line shown in Fig. 6(b).

4.3 SROP for cesium D_2 line

Similarly, we use P_2 as the pumping laser for cesium D_2 line. SROP spectra are observed as shown in trace (2) in Fig. 7(a) when P_2 is scanned across cesium $6S_{1/2}$ F = 4 - $6P_{3/2}$ F" = 3, 4

transitions. Here we should note that SROP spectra can eliminate CRLs, but this point is not shown in Fig. 4(a) in the case of cesium D_1 line. Because there are no CRLs even in SAS spectra in cesium D_1 line (see trace (1) in Fig. 4(a)) due to that the hyperfine splitting of $6P_{1/2}$ state (1167.7 MHz) is much larger than the Doppler-broaden linewidth at room temperature (~500 MHz). However, for cesium D_2 line, the situation is different. CRLs appear in SAS spectra (trace (1) in Fig. 7(a)), because the hyperfine splitting of $6P_{3/2}$ state (150 ~250 MHz) is much smaller than the Doppler-broaden linewidth at room temperature. In trace (2) of Fig. 7(a), the CRLs are eliminated due to feature of the zero-velocity selection in SROP.

The SROP spectra shown in trace (2) in Fig. 7(a) are for the case of weak coupling beam (~74μW@852.3nm, Ωc ~Γ_{21}). We find that linewidth and magnitude of SROP signals decrease when optical intensity of two beams is reduced. The dressed-state splitting peaks of $6S_{1/2}$ F = 4 ground state around Δc ~0 appear as shown in trace (3) in Fig. 7(a). When the coupling laser is getting stronger, the dressed-state splitting enlarges as well according to $\Omega = \Gamma_{21}\sqrt{I/2I_S}$ (see Fig. 7(b)).
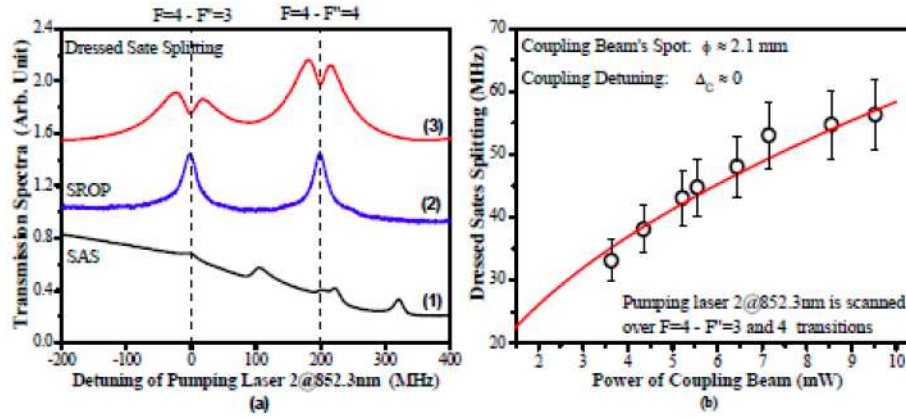

Fig. 7. **(a)** Trace (2) and (3) show the SROP spectra corresponding to cesium F = 4 – F" = 3, 4 transitions in D_2 line with different Ωc while the trace (1) shows the relevant SAS for frequency calibration. The pumping beam's power is ~10μW@852.3nm (Ωp ~0.4 Γ_{21}) for trace (2) and (3). The coupling beam's power is ~74μW@852.3nm (Ωc ~Γ_{21}) for trace (2) while ~4.3mW @852.3nm (Ωc ~7.5 Γ_{21}) for trace (3). The up-down shift of traces (2) and (3) 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 (Δc ~0). The circles are experimental data with ± 2.5% of error bar. The solid line represents for theoretical curve.

Both experimental results obtained from two independent experiments for cesium D_1 and D_2 lines fit well to the same theoretical model with no free parameters, as indicated in Fig. 4(b), Fig. 6(b) and Fig. 7(b). Consequently, it is confirmed that SROP spectrum could be used for making research on the dressed-atoms in ground state with room-temperature atomic vapor cell.

5. Conclusion

SROP spectra for cesium D_1 and D_2 lines have been demonstrated experimentally with room-temperature cesium vapor cell. As the coupling laser is locked to a cycling transition, only the optical pumping effect of the swept pump beam for ground state needs to be considered. The locked coupling laser serves as a selective probe. We find that the linewidth of SROP spectra mainly depends on the intensity of the coupling and pumping beams, and it will be broadened strikingly along with the increased coupling beam's intensity. Meanwhile, the altitude of SROP spectra is affected by the intensity of the pumping beam. When the intensity of both beams is weak enough, we could not observe the other peaks (too small) in SROP spectrum except those formed by the atoms which are resonant with the coupling laser. With features of

flat-background, narrow linewidth, high SNR, and elimination of CRLs, the SROP can be used to measure the atomic hyperfine splitting just as the work in Ref. 9.

The SROP can also be employed to measure the dressed-state splitting of ground state with room-temperature atomic vapor cell. This has been validated experimentally in both cases of cesium D_1 and D_2 lines. In SROP, the coupling beam can selectively probe and dress the atoms in a particular velocity belong to the Maxwell-Boltzmann velocity distribution. The peak position of probe beam will move together with Δc at the given Ωc away from the SAS. This is also similar to the dressed atoms. As a result, the splitting of the dressed ground state can be measured in the both case of $\Delta c \sim 0$ and $\Delta c \neq 0$ utilizing the SROP. In future, more efforts need to be done to reduce the errors of the measurement, and to study the atomic coherence that may be exist in SROP as well. We hope that the linewidth of the SROP will be reduced with the help of the atomic coherence.

Acknowledgments