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# Improvement of the spectra signal-to-noise ratio of caesium $6P_{3/2}-8S_{1/2}$ transition and its application in laser frequency stabilization

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## Abstract

The spectra of the  $^{133}\text{Cs}$   $6P_{3/2}-8S_{1/2}$  transition have been demonstrated by using both the double resonance optical pumping (DROP) and the optical–optical double resonance (OODR) techniques. Compared with the OODR technique, the DROP spectra show a much better signal-to-noise ratio and narrow spectral linewidth. When the frequency of a 795 nm external-cavity diode laser is stabilized to the DROP spectrum of  $^{133}\text{Cs}$   $6P_{3/2} F' = 5-8S_{1/2} F'' = 4$  transition, the lowest square root of the Allan variance of  $4.1 \times 10^{-12}$  at an averaging time of 100 s was obtained.

## 1. Introduction

Laser frequency stabilization is very important for various studies from fundamental science to practical applications in all kinds of fields, such as high-resolution molecular and atomic spectroscopy, laser cooling and trapping of atoms, optical fibre communication, and so on [1–3]. In these applications, an absolute frequency reference (a transition line of atoms or molecules) is required for frequency stabilization of a laser diode [4]. The saturated absorption spectroscopy (SAS) technique is a standard technique used to generate such spectral profiles [5]. However, people pay less attention to the spectrum between the excited states compared with SAS which focuses on the spectrum between a ground state and an excited state. Moreover, it is difficult to obtain the spectrum between excited states due to the low thermal equilibrium population in the excited states [6–10].

Based on the virtue of the Doppler-free spectrum between the excited states, Sasada stabilized the frequency of a diode laser using the optical–optical double resonance (OODR) spectrum and measured the wavelengths of the  $^{87}\text{Rb}$   $5P_{1/2}-6S_{1/2}$  and  $5P_{3/2}-4D_{3/2}$ ,  $4D_{5/2}$  transitions [11].

Boucher *et al* [12] and Breton *et al* [13] also stabilized a distributed feedback (DFB) diode laser to the  $^{87}\text{Rb}$   $5P_{1/2}-6S_{1/2}$  transition using the OODR spectrum. Pandey *et al* [14] stabilized the frequency of a diode laser to the  $^{87}\text{Rb}$   $5P_{3/2}-7S_{1/2}$  transition using the blue fluorescence spectrum of  $^{87}\text{Rb}$   $6P_{1/2}-5S_{1/2}$ , which is due to the strong decay channel from  $^{87}\text{Rb}$   $7S_{1/2}-6P_{1/2}-5S_{1/2}$ . Although the OODR technique is a sophisticated tool for obtaining the spectra between excited states, its signal-to-noise ratio (SNR) is not high enough for a large spontaneous emission rate system because the intermediate state is not easily populated [11]. For the frequency stabilization of a laser diode, the spectral linewidth  $\Delta\nu$  and the SNR are very important factors. Recently, the SNR of the spectrum between excited states was improved through use of the double resonance optical pumping (DROP) method, which is based on the interaction of an atom with two optical fields that are resonant to two transitions that share a common state, and the key idea of DROP is to detect the population of the ground state instead of the excited state. Actually DROP is a ‘two-photon optical pumping’ process, in which an atom is populated to a higher excited state from a hyperfine ground state via an intermediate state by two photons and then spontaneously decays to another hyperfine ground

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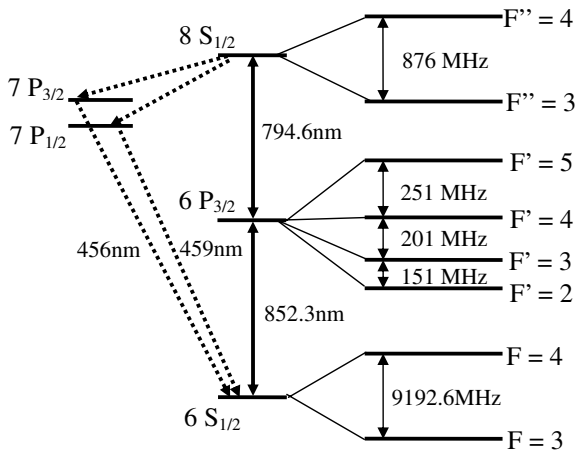


Figure 1. Schematic diagram of relevant states of caesium atoms.

state through other decay channels. Compared with DROP, hyperfine optical pumping is a ‘one-photon optical pumping’ process, in which an atom is populated to an excited state from a ground state by one photon and then spontaneously decays back to another hyperfine ground state. Moon *et al* [15] observed DROP of <sup>87</sup>Rb atoms and stabilized the frequency of a laser diode using DROP. Lee *et al* [16] measured the absolute energy level and the hyperfine structure of the <sup>87</sup>Rb 4D<sub>5/2</sub> state through DROP.

In this paper, we have presented the hyperfine spectrum of the <sup>133</sup>Cs 6P<sub>3/2</sub>–8S<sub>1/2</sub> transition using the OODR technique and then applied the DROP technique to the <sup>133</sup>Cs 6S<sub>1/2</sub>–6P<sub>3/2</sub>–8S<sub>1/2</sub> transitions and investigated the linewidth and magnitude of the DROP spectrum as a function of the laser power. Finally, we acquired the DROP spectrum with a narrow linewidth and a high SNR in comparison with the OODR spectrum and stabilized the frequency of a 795 nm external-cavity diode laser (ECDL) using this spectrum.

## 2. Principle and experimental setup

Figure 1 shows the schematic diagram of relevant hyperfine states of <sup>133</sup>Cs atoms. The center wavelength for the 6S<sub>1/2</sub>–6P<sub>3/2</sub> fine transition is 852.3 nm, and for the 6P<sub>3/2</sub>–8S<sub>1/2</sub> fine transition it is 794.6 nm. The hyperfine splitting between F'' = 3 and F'' = 4 states in 8S<sub>1/2</sub> is 876 MHz. The natural linewidth of the 6P<sub>3/2</sub>F' = 5–6S<sub>1/2</sub>F = 4 hyperfine transition is 5.22 MHz, and the natural linewidth of the hyperfine transitions between the 8S<sub>1/2</sub> and 6P<sub>3/2</sub> states is approximately 2.18 MHz.

Figure 2 shows the experimental scheme for the DROP and the OODR in the 6S<sub>1/2</sub>–6P<sub>3/2</sub>–8S<sub>1/2</sub> channel. In the experiment, two laser beams are counter-propagating through a Cs vapour cell (length of 5 cm, diameter of 2 cm), a slight angle (~0.02 rad) between the two light beams is set to permit separation of the beams before detection. The two lasers are commercial ECDLs (Toptica DL-100) with a typical linewidth of ~500 kHz. The ECDL1@852 nm is locked to one of the caesium 6S<sub>1/2</sub>F = 4–6P<sub>3/2</sub>F' = 3, 4, 5 hyperfine transitions by the SAS scheme, in which the error signal is

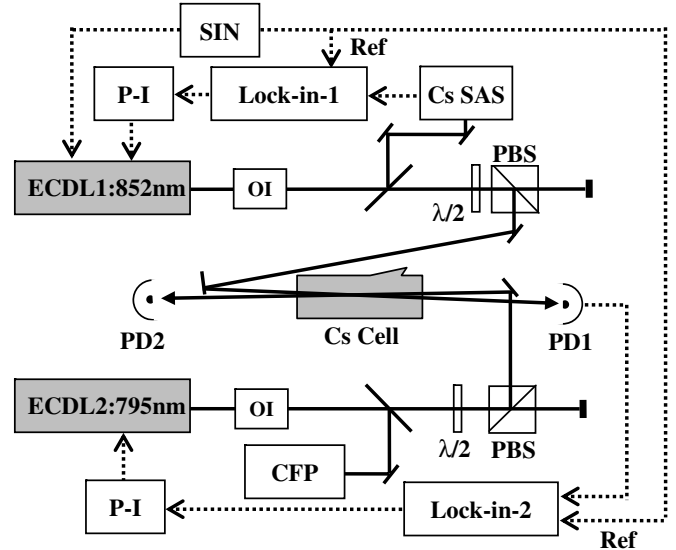
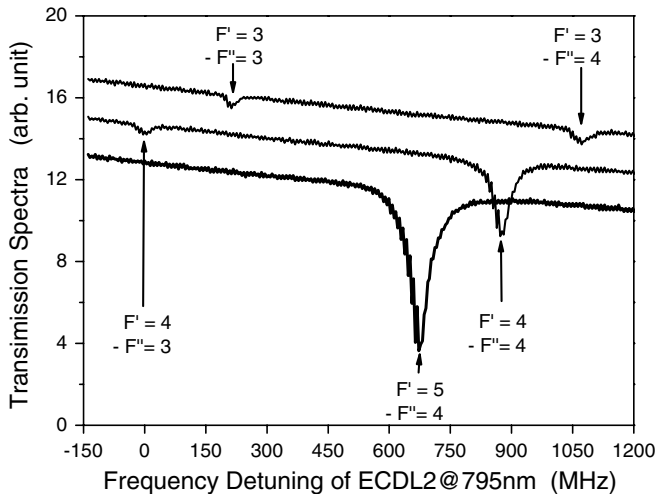


Figure 2. Experimental setup for the observation of the OODR and the DROP spectra and for the frequency stabilization of ECDL2@795 nm by using the DROP. Keys to figure: ECDLs: external-cavity diode lasers; SIN: sine-wave signal generator; Lock-in: lock-in amplifier; OI: optical isolator; CFP: confocal 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.

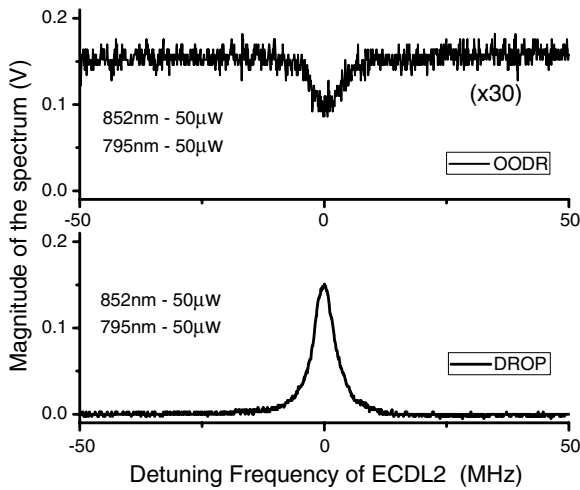
generated from a lock-in amplifier using 14 kHz sine wave modulation, and the ECDL2@795 nm is scanned over the whole range of excited states in the 6P<sub>3/2</sub>–8S<sub>1/2</sub> transition. Essentially, the DROP signal from photodiode 1 (PD1) is the transmission of ECDL1@852 nm through the Cs vapour cell, and the OODR signal from photodiode 2 (PD2) is the absorption of ECDL2@795 nm between the 6P<sub>3/2</sub> and 8S<sub>1/2</sub> transitions. The spectral linewidth of DROP is measured using a confocal Fabry–Perot (CFP) cavity with a calibrated free spectra range of 503 MHz, while the power of the two lasers is varied by a half-wave plate and a polarizing beam splitter cube (PBS). Simultaneously, we record the magnitude of DROP and stabilize the frequency of ECDL2@795 nm using it.

## 3. The experimental results and discussions

<sup>133</sup>Cs atoms are usually in the 6S<sub>1/2</sub> ground states, so it is difficult to get the spectra of the 6P<sub>3/2</sub>–8S<sub>1/2</sub> transition. When the ECDL1@852 nm is locked to the 6S<sub>1/2</sub>F = 4–6P<sub>3/2</sub>F' = 3 (F' = 4, F' = 5) transitions, some atoms can be populated to the relevant hyperfine states, respectively. When ECDL2@795 nm is scanned over the range 6P<sub>3/2</sub>–8S<sub>1/2</sub> transition, the relevant OODR spectra detected by PD2 can be obtained as shown in figure 3. The typical powers of the two lasers are of a few milliwatts in the experiment. The base lines are not flat because of residual amplitude modulation when ECDL2@795 nm is scanned by a triangle-wave voltage. The spectra are consistent with the energy level diagram in figure 1. Using the calibrated CFP cavity, the hyperfine splitting between the F'' = 3 and the F'' = 4 states in 8S<sub>1/2</sub> can be measured and it is 876 MHz.

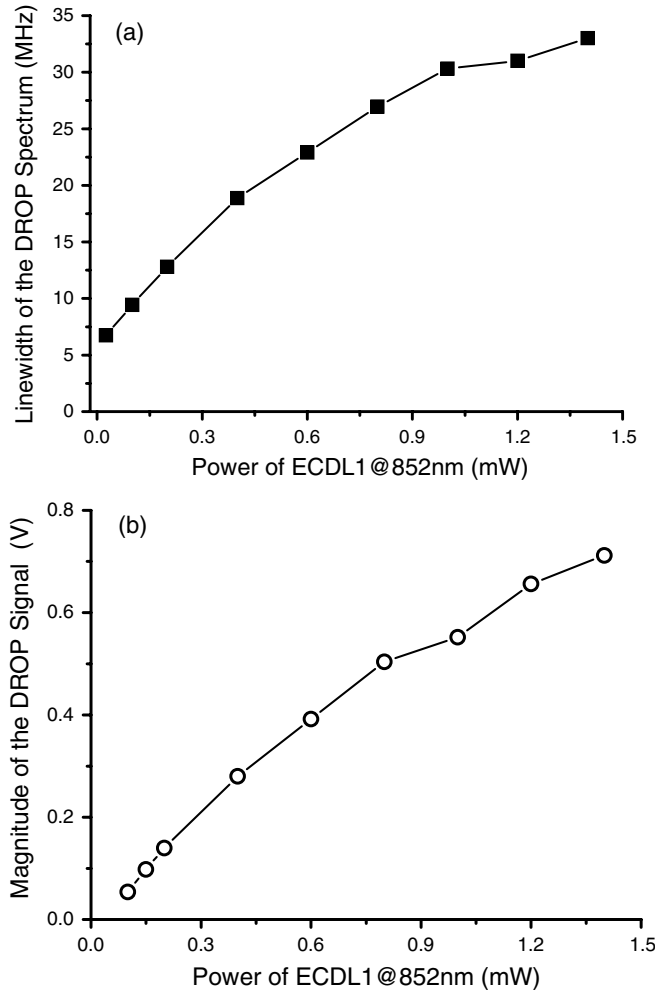


**Figure 3.** The OODR spectrum of the  $^{133}\text{Cs}$   $6P_{3/2}-8S_{1/2}$  transition. The measured hyperfine splitting between  $F'' = 3$  and  $F'' = 4$  states in  $8S_{1/2}$  is 876 MHz.



**Figure 4.** Comparison of the OODR spectrum and the DROP spectrum of the  $^{133}\text{Cs}$   $6P_{3/2} F' = 5-8S_{1/2} F'' = 4$  transition. Obviously the SNR of DROP is much better than that of OODR.

Figure 4 shows a typical DROP spectrum of the  $6P_{3/2} F' = 5-8S_{1/2} F'' = 4$  transition of the  $^{133}\text{Cs}$  atoms with PD1. This can be easily understood from the energy level diagram in figure 1. When ECDL1@852 nm is locked to the  $6S_{1/2} F = 4-6P_{3/2} F' = 5$  transition, some atoms are populated in the  $6P_{3/2} F' = 5$  state. And then when the frequency of ECDL2@795 nm is resonant with the  $6P_{3/2} F' = 5-8S_{1/2} F'' = 4$  transition, the atoms are excited to the  $8S_{1/2} F'' = 4$  excited states. Many atoms in this state spontaneously transfer to the  $6S_{1/2} F = 3$  state through the  $6P_{3/2} (F' = 3, F' = 4)$  intermediate states and other channels such as  $8S_{1/2}-7P_{3/2}-6S_{1/2}$  in which we observe strong blue fluorescence from the  $7P_{3/2}-6S_{1/2}$  transition which may be used to stabilize the frequency of ECDL2@795 nm to the  $6P_{3/2} F' = 5-8S_{1/2} F'' = 4$  transition similarly to the scheme in [14]. So the population of the  $6S_{1/2} F = 4$  state can be depleted; we obtain the DROP spectrum corresponding to the  $6P_{3/2} F' = 5-8S_{1/2} F'' = 4$  transition between the excited states by detecting the transmission of ECDL1@852 nm.



**Figure 5.** The linewidth (a) and the magnitude (b) of the DROP signal versus the power of ECDL1@852 nm.

Figure 4 also shows the comparison of DROP and OODR of the  $6P_{3/2} F' = 5-8S_{1/2} F'' = 4$  transition, where the power of ECDL1@852 nm is  $50 \mu\text{W}$  with the same power of ECDL2@795 nm. From this figure, we can clearly see that the SNR of DROP is much higher than that of OODR under the same conditions. The SNR of DROP is high due to the rapid decrease in the population of  $6S_{1/2} F = 4$  by optical pumping. However, the SNR of the OODR spectrum is very low because the spontaneous emission of the atoms excited in the  $8S_{1/2} F'' = 4$  state decreases the population in the intermediate state.

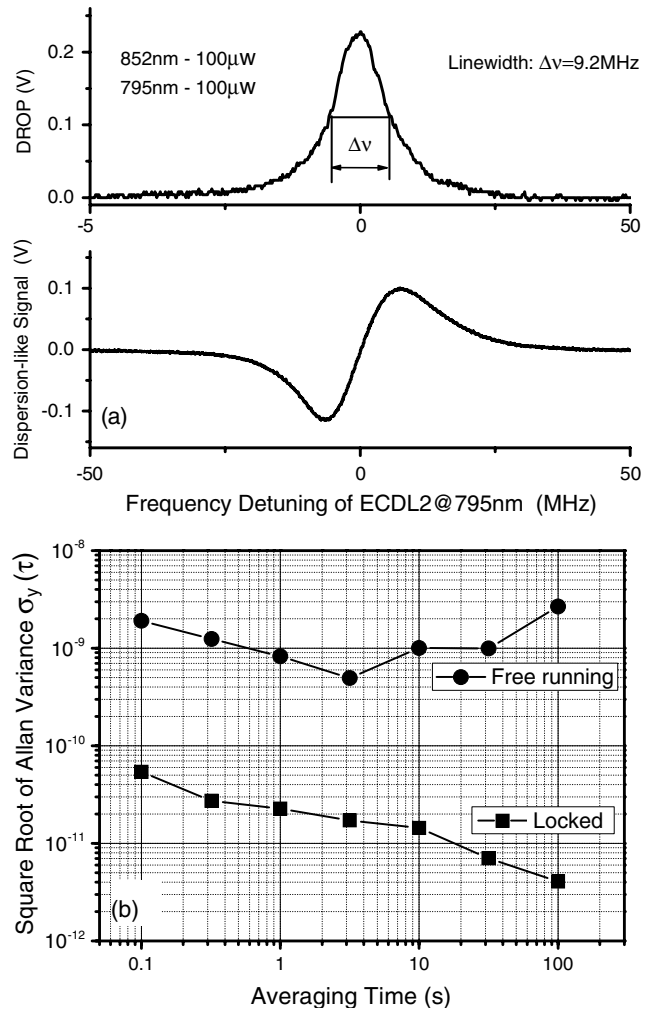
In order to study the effect of the power of ECDL1@852 nm on the DROP spectrum, we measured the spectral linewidth of the DROP spectrum which is related to the natural linewidth, the laser linewidth and the laser power. When the power of ECDL1@852 nm varied from 25 to  $1400 \mu\text{W}$ , and the power of ECDL2@795 nm was fixed at  $25 \mu\text{W}$ , we got results as shown in figure 5(a). We can see that the linewidth of the DROP spectra broadens nonlinearly with the increase in power of ECDL1@852 nm. The reasons for the broadening linewidth are complicated; there are two main factors: one is the power broadening effect and the other is the electromagnetically induced transparency effect, which can

be found in [17]. Meanwhile, we also recorded the magnitude of DROP as shown in figure 5(b), and it depends mainly on the optical pumping rate. While increasing the power of ECDL1@852 nm, the transfer rate from the  $6S_{1/2}F = 4$  to the  $6S_{1/2}F = 3$  ground state also increases, but it is limited by the power of ECDL2@795 nm and the spontaneous emission rates from  $8S_{1/2}-6P_{3/2}(6P_{1/2}, 7P_{1/2}, 7P_{3/2})-6S_{1/2}$  channels, so that the magnitude of DROP nonlinearly increases. Similarly, a result just like figure 5(b) has been obtained when changing the power of ECDL1@795 nm from 25 to 1400  $\mu\text{W}$  and fixing the power of ECDL2@852 nm to 25  $\mu\text{W}$ .

It is known that a spectrum with a narrow linewidth  $\Delta\nu$  and a high SNR is important to stabilize the laser frequency; the variance for the laser frequency stabilization  $\sigma_\nu(\tau) \propto \Delta\nu/\text{SNR}$ . But from figure 5, the  $\Delta\nu$  and SNR are inconsistent. Making a tradeoff between the two factors, the powers of both ECDL1@852 nm and ECDL2@795 nm are set to  $\sim 100 \mu\text{W}$ , and the linewidth of 9.2 MHz for the DROP spectrum of  $6P_{3/2}F' = 5-8S_{1/2}F'' = 4$  transition is obtained for frequency stabilization. At first, the frequency of ECDL1@852 nm was stabilized to the  $^{133}\text{Cs } 6S_{1/2}F = 4-6P_{3/2}F' = 5$  transition through SAS with frequency modulation. Then when ECDL2@795 nm was scanned over the  $6P_{3/2}-8S_{1/2}$  transition, the modulation of ECDL1@852 nm was transferred to the DROP spectrum. So we could get a dispersion-like signal, as shown in figure 6(a), with the DROP spectrum and the modulation from ECDL1@852 nm using a lock-in amplifier. Finally, this error signal was fed back to the piezo attached to the grating of ECDL2@795 nm, and we stabilized the frequency of ECDL2@795 nm without a direct modulation to ECDL2@795 nm. Compared with the case of free running, the frequency stability of the ECDL2@795 nm after locking was remarkably improved as shown in figure 6(b). The lowest square root of the Allan variance was  $\sigma_y(\tau) = 4.1 \times 10^{-12}$  with an averaging time of  $\tau = 100$  s.

#### 4. Conclusion

In conclusion, we demonstrated the hyperfine structure of the  $^{133}\text{Cs } 6P_{3/2}-8S_{1/2}$  transition using OODR technology and obtained a kind of Doppler-free spectrum between the  $6P_{3/2}F' = 5-8S_{1/2}F'' = 4$  excited states by DROP technology, which has a narrow linewidth and a high SNR compared with the OODR spectrum. Then we investigated the linewidth and the magnitude of the DROP spectrum as a function of the ECDL1@852 nm power. Finally, we stabilized the frequency of ECDL2@795 nm to the  $6P_{3/2}F' = 5-8S_{1/2}F'' = 4$  transition without direct modulation to ECDL2@795 nm, and the lowest square root of the Allan variance was  $4.1 \times 10^{-12}$  at an averaging time of 100 s. In our experiment, when ECDL1@852 nm and ECDL2@795 nm are locked to the  $6S_{1/2}F = 4-6P_{3/2}F' = 5$  transition and the  $6P_{3/2}F' = 5-8S_{1/2}F'' = 4$  transition, respectively, we could observe blue fluorescence due to the  $7P_{1/2}(7P_{3/2})-6S_{1/2}$  transitions, which is another way that can be used to stabilize the frequency of ECDL2@795 nm to the  $6P_{3/2}F' = 5-8S_{1/2}F'' = 4$  transition.



**Figure 6.** (a) The DROP spectrum of Cs  $6P_{3/2}F' = 5-8S_{1/2}F'' = 4$  transition and the corresponding dispersion-like frequency discriminating signal without direct modulation to ECDL2@795 nm. (b) The square root of the Allan variance for the free running case and after locking of ECDL2@795 nm.

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