Characterizing double-resonance optical-pumping spectra of cesium $6P_{3/2}$ - $8S_{1/2}$ excited-state transition and its application

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ABSTRACT

The spectra of cesium $6P_{3/2}$ - $8S_{1/2}$ excited-state transition have been obtained using double resonance optical-pumping (DROP) technique in a room-temperature vapor cell, and have shown a much better signal-to-noise ratio (SNR) compared with that using the traditional optical-optical double resonance (OODR) method. Furthermore, the line-width of DROP spectra is obviously narrowed by electromagnetically-induced transparency (EIT) effect in cesium $6S_{1/2}$ F=4 - $6P_{3/2}$ F'=5 - $8S_{1/2}$ F''=4 transitions. Finally, such DROP spectrum of $6P_{3/2}$ F'=5 - $8S_{1/2}$ F''=4 transition with a high SNR and a narrow line-width is applied into frequency stabilization of a 795 nm external-cavity diode laser, and the residual frequency fluctuation is ~ 600 kHz within 500 s.

Keywords: Double-resonance optical pumping (DROP), Optical-optical double resonance (OODR), Electromagnetically-induced transparency (EIT), Ladder-type atomic system, Laser frequency stabilization

1. INTRODUCTION

Compared with the spectra of the transition between a ground state and an excited state, the spectra of transitions between atomic excited states have a distinct virtue of natural Doppler-free in a room-temperature vapor cell due to velocity selection mechanism, are widely used in measurement of hyperfine splitting, determination of hyperfine structure constant, Rydberg atomic state detection, laser frequency stabilization and so on ¹⁻⁴. The transitions between atomic excited states can be approached traditionally by the optical-optical double-resonance (OODR) technique, which is based on the interaction of molecules or atoms with two optical fields resonantly tuning to two transitions that share a common state in a ladder-type atomic system ⁵. However, the signal-to-noise ratio (SNR) of OODR spectrum is poor especially in some atomic systems with large spontaneous emission rates. Recently double-resonance optical-pumping (DROP) technique, detecting the population variation of ground state instead of intermediate state, can be adopted to improve the SNR of the spectrum between atomic excited states ⁶.

In substance, DROP is a kind of optical pumping effect, which is often accompanied with spontaneous decay. In fact, in such atomic system based upon the interaction of atoms with two optical fields, there always exits quantum coherence such as EIT, which can fully suppress the line-width of DROP spectrum ⁷⁻⁸. In this paper, we made a comparison between DROP and OODR spectrum in cesium $6S_{1/2} - 6P_{3/2} - 8S_{1/2}$ ladder-type atomic system, and employed EIT effect to reduce the line-width of DROP spectrum, and applied it to frequency stabilization of a 795nm external-cavity diode laser system.

2. EXPERIMENTAL SETUP

Figure 1 shows the energy level diagram of the $6S_{1/2} - 6P_{3/2} - 8S_{1/2}$ transitions of ¹³³Cs atoms. The center wavelength of $6S_{1/2} - 6P_{3/2}$ transition is 852.3nm and its natural line-width is 5.22MHz. The $6P_{3/2} - 8S_{1/2}$ transition is 794.6nm and its natural line-width is 2.18MHz. The probe laser is locked to the $6S_{1/2}$ F=4 - $6P_{3/2}$ F'=5 cycling transition, which can populate the atoms around zero velocity in the direction of probe beam to the F'=5 intermediate state from the F=4

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ground state. The coupling laser is scanned over the $6P_{3/2}$ F'=5 to $8S_{1/2}$ F''=4 hyperfine transition, which can further populate the atoms to the F''=4 higher excited state from the F'=5 intermediate state. Here, the transmission signal of the coupling laser is recorded as function of its frequency detuning, which is called OODR spectrum. Some of the atoms on the F''=4 state spontaneously decay to the F'=4, F'=3 states and the $7P_{1/2}$, $7P_{3/2}$ states (not shown in Fig. 1), then decay to the F=3 ground state. Due to this two-photon excitation and spontaneous decay, the population of the F=4 ground state are optically pumped to the F=3 ground state, so the absorption of probe laser accordingly decreases forming DROP spectrum, which is the transmission signal of probe laser as the function of coupling laser' frequency detuning.



Fig.1 Relevant energy level of ¹³³Cs atoms



Fig. 2 Schematic diagram of experimental arrangement. The coupling laser @794.6 nm takes the path of beam 1 for counter-propagating (CTP) configuration, or it takes the path of beam 2 for co-propagating (CP) configuration. Keys to figure: ECDLs: external cavity diode lasers; SIN: sine-wave signal generator; Lock-in: lock-in amplifier; P-I: proportion and integration amplifier; Ref: reference channel of lock-in amplifier; SAS: saturated absorption spectroscopy; OI: optical isolator; CFP: confocal Fabry-Perot cavity; PD: photodiode; $\lambda/2$: half-wave plate; PBS: polarization beam splitter cube; DF, dichroic filter.

The schematic diagram of experimental setup is shown in Fig. 2. A commercial grating-feedback external-cavity diode laser (ECDL) at 794.6nm (Toptica DL-100L) with typical line-width of ~ 500 kHz (in 50 ms) is employed as the coupling laser with Rabi frequency Ωc and detuning Δ_2 . A home-made ECDL at 852.3nm with the roughly same line-width is used as the probe laser with Rabi frequency Ωp and detuning Δ_1 , which can be locked to the F=4 - F'=5

cycling transition by the conventional frequency modulation technique for a saturated absorption spectral profile. Optical power of the probe and coupling lasers can be adjusted by corresponding sets of module consisted of halfe-wave plate (HP) and polarization beam splitter (PBS) cube. The two laser beams are arranged to counter-propagate passing through a cesium vapor cell (~ ϕ 25 mm×50 mm) at room temperature when the coupling laser takes the path of beam1 after PBS2 as shown in Fig.2. They are also arranged to co-propagate in vapor cell when the coupling laser takes the path of beam2 by removing the photodiode 1(PD1) and blocking the beam1. Two dichroic filters are used to combine and separate two beams. The beams with a diameter $\phi p \sim 2.0$ mm for the probe beam and $\phi c \sim 2.2$ mm for the coupling beam are overlapped inside cesium vapor cell. The probe laser's transmission signal (DROP spectrum) is collected by a PD2 (New Focus, Model 2001) while the coupling's transmission signal (OODR spectrum) is obtained by a PD1. A cofocal Fabry-Perot cavity @795nm (not shown in Fig. 2) with finesse of ~100 and calibrated free spectra range (FSR) of 503 MHz is used for measurement of the line-width of the signals.

3. EXPERIMENTAL RESULTS AND DISCUSSIONS

3.1 Comparison between OODR spectrum and DROP spectrum

The OODR is the absorption spectrum, which can be accounted for the resonance absorption of ECDL2@794.6nm between the $6P_{3/2}$ intermediate state and the $8S_{1/2}$ excited state. However, the DROP is the transmission spectrum, which can be illustrated by the two-photon optical pumping from the F=4 ground state to another F=3 ground state companied by spontaneous decay. Fig. 3 shows the comparison of OODR and DROP in the F' = 5 – F'' = 4 transition of the ¹³³Cs atoms, where the power of ECDL1@852nm is 50µW with the same that of ECDL2@795nm. From Fig.3, we can obviously see that the SNR of the DROP is much higher than that of the OODR. The SNR of the DROP is high due to the rapid decrease of the population of the F = 4 ground state by two-photon optical pumping. However, that of the OODR is small because the spontaneous emission of the atoms excited in the F''=4 state lowers the population in the F'= 5 intermediate state ⁶. Another reason: the OODR is related to the population of the F'=5 intermediate state, however, in order to avoid power broadening of the spectrum, the low power of the ECDL1@852.3nm results in the low population of the F'=5 intermediate state, so the SNR of OODR spectrum is relatively low.



Fig. 3 Comparison between OODR spectrum and DROP spectrum in the F' = 5 - F'' = 4 transition of ¹³³Cs atoms

3.2 Comparison of DROP spectra between the CP and CTP configurations

Figure 4 is the comparison of DROP spectrum in the F' = 5 - F'' = 4 transition of ¹³³Cs atoms between the CP and CTP configurations, here the power of probe beam with 120µw is the same with that of coupling beam. The magnitude of DROP spectrum for CTP is bigger than that for CP, because more atoms will contribute to the DROP spectrum in that Doppler Effect can be in part eliminated in the vapor cell for CTP. On the other hand, the line-width of DROP spectrum with 9.3MHz for CTP is narrower than that with 15.4 MHz for CP.



Fig. 4 Comparison of DROP spectrum in the F' = 5 - F'' = 4 transition of ¹³³Cs atoms between the CP and CTP configurations

We find that the experimental setup for the DROP in the case of CTP is the same as that for the EIT in ladder-type Doppler broadened atomic system. The only difference is that the frequency of probe laser is scanning while the coupling laser is fixed resonantly with the transition in the normal EIT experiments. On the contrary, it is the frequency of coupling laser that is scanning while the probe laser is locked on resonance in the DROP experiments. We thus believe that the EIT effect makes the spectrum for CTP narrower than that for CP without the EIT. The explanations are as follows.

EIT is a kind of destruction interference atomic coherence effect, and is substantially interpreted that the atoms are prepared in a dark superposition state of the ground state and higher excited state for the ladder-type atomic system, so the system under the above two scanning modes in the CTP configuration should all exist EIT effect. Further, in EIT theory, there is a very popular formula: the complex susceptibility $\chi = \chi' + i\chi''$, the real part χ' and imaginary χ'' are related to the dispersion and absorption of the atomic medium, respectively ("+" for CP; "-" for CTP). Here N(v) is one-dimensional Maxwell-Boltzmann velocity distribution, and u is the most probable speed. k is the Boltzmann constant, T temperature, m the mass of a Cs atom. ^{9,10}

$$\chi(v)dv = \frac{4i\hbar g_{21}^2 / \varepsilon_0}{\gamma_{21} - i\Delta_1 - i\frac{\omega_p}{c}v + \frac{\Omega_c^2 / 4}{\gamma_{31} - i(\Delta_1 + \Delta_2) - i(\omega_p \pm \omega_c)v / c}} N(v)dv$$
(1)
$$N(v) = \frac{N}{u\sqrt{\pi}} e^{-v^2/u^2} dv$$
(2)

$$u = \sqrt{\frac{2kT}{m}} \tag{3}$$

In scanning mode of DROP for CTP configuration, formula (1) gives the new EIT profile which is similar with the line-shape of DROP as shown in Fig.4, so that EIT signal is often mixed with DROP signal and it is difficult to distinguish the difference of spectra between them. However, for CP configuration, EIT is almost submerged by Doppler effect according to formula (1) because the term $(\omega_P + \omega_C) v / c$ cannot be ignored in both scanning modes⁷. So according to the comparison of spectrum between CP and CTP configuration, we can clearly discover EIT effect from

DROP with the similar spectral profile, simultaneously, an explanation is given: the line-width of DROP spectrum for CTP is narrower than that for CP due to atomic coherence in EIT.

3.3 Application of DROP spectrum in laser frequency stabilization

The above experimental results and discussions will help us get a DROP spectrum with a narrow line-width Δv and a high SNR, which are desired for laser frequency stabilization according to the variance $\sigma_v(\tau)$ for laser frequency stabilization $\sigma_{\rm v}(\tau) \propto \Delta v/SNR$. Now, when the powers of both ECDL1@852 nm and ECDL2@795 nm are 120 uW, we obtain a DROP spectrum with line-width of 9.3 MHz at the F' = 5 - F'' = 4 transition for CTP configuration, and apply it to the frequency stabilization of the ECDL2@795nm diode laser. The detailed process is as following: First, the frequency of ECDL1@852 nm is stabilized to the F = 4 - F' = 5 transition using the SAS profile with frequency modulation. Second, when ECDL2@795 nm is scanning over the F' = 5 - F'' = 4 transition, the modulation of ECDL1@852 nm is transferred to the DROP spectrum. So we can get a dispersion-like signal (error signal) as shown in Fig. 5 by a lock-in amplifier with the DROP spectrum and the modulation reference signal from SIN generator as shown in Fig. 2. At last, the error signal is fed back to the piezo attached with the grating of ECDL2@795 nm for realizing frequency locking. Here, we stabilize the frequency of ECDL2@795 nm without the direct modulation to it. The frequency stability of ECDL2@795 nm after locked is 0.6 MHz as shown in Figure 6, which is remarkably improved compared with the case of free running with frequency fluctuation 14.6 MHz. This technique of frequency locking without modulation will be more significant in the field of laser frequency stabilization for optical communication frequency band. For example, we can also get a DROP spectrum based on rubidium atoms instead of cesium atoms using the same method.



Fig. 5 The DROP spectrum of F'=5-F''=4 transition (CTP configuration) and the corresponding dispersion-like frequency-discriminating signal without direct modulation of ECDL2@795 nm.



Fig. 6 The frequency fluctuation of ECDL2@795 nm for locking and free running cases.

4. CONCLUSIONS

We demonstrate Doppler-free spectrum between the $6P_{3/2} F' = 5 - 8S_{1/2} F'' = 4$ excited states of ¹³³Cs atoms by DROP technique, which has a higher SNR compared with that of OODR spectrum. Then we have investigated the ladder-type EIT and DROP in this atomic system. Results show that there are the EIT and DROP effects for CTP while there is only the DROP effect for CP for the moderate coupling laser (neglectable EIT for CP configuration), which are proved theoretically and experimentally. Furthermore, EIT effect makes the line-width of DROP spectrum for CTP configuration much narrower than that for CP configuration. Finally, we stabilize the frequency of a 795-nm ECDL to the F' = 5 - F'' = 4 transition using DROP spectrum without direct modulation of this 795-nm ECDL.

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