Optics Communications 474 (2020) 126102

Contents lists available at ScienceDirect

Optics Communications

journal homepage: www.elsevier.com/locate/optcom

Double resonance optical pumping-polarization spectroscopy of an excited state transition

Baodong Yang [a,b,*], Yi Liu [a], Junmin Wang [b]

[a] College of Physics and Electronic Engineering, Shanxi University, Taiyuan 030006, China
[b] State Key Laboratory of Quantum Optics and Quantum Optics Devices and Institute of Opto-Electronics, Shanxi University, Taiyuan 030006, China

ARTICLE INFO

Keywords:
Polarization spectroscopy
Atomic coherence effect
Excited state spectroscopy
Frequency stabilization

ABSTRACT

We present an investigation of double resonance optical pumping-polarization spectroscopy (DROP-PS) of an excited state transition based on the cesium $6S_{1/2}$- $6P_{3/2}$- $8S_{1/2}$ ladder-type atomic system in a room-temperature vapor cell. A linearly polarized probe beam populates the atoms from ground state $6S_{1/2}$ to intermediate state $6P_{3/2}$, and then DROP-PS signal is obtained by detecting the anisotropy of atomic medium on the $6P_{3/2}$ state induced by a circularly polarized coupling beam operated on the $6P_{3/2}$ - $8S_{1/2}$ transition. This DROP-PS signal with dispersive shape is suitable for locking a laser to an excited state transition.

1. Introduction

It is an eternal pursuit for obtaining the high-resolution spectroscopy and developing its applications [1,2]. Over the years, the spectroscopy for transition between a ground state and an excited state have been paid more attention to. Recently, the excited state spectroscopy is of growing interest for many research fields such as precision measurement of energy structure of atoms [3,4], Rydberg states [5], frequency stabilization specially in optical communication [6], optical filter [7], frequency up/down-conversion [8–11], multiphoton laser cooling [12,13], nonlinear optics [14,15], and so on. The atoms are usually populated on the ground state according to Maxwell–Boltzmann velocity distribution in a room-temperature atomic vapor cell, so researchers often employ a laser to pump atoms into an intermediate excited state, and then another laser as probe light is scanned over the transition between an intermediate state and a higher excited state to obtain an excited spectrum, and this technique is known as optical–optical double resonance (OODR) [16,17]. In 2004, Moon et al. reported a double resonance optical pumping (DROP) technique for the observation of spectrum of an excited state transition [18], and its key idea is to detect the change of population on one of hyperfine sublevels of the ground state due to a two-photon optical pumping process based on a five-level atomic system, and the experiments have already proved that DROP technique can greatly improve the signal-to-noise ratio of spectra for some transitions between the excited states in comparison with OODR, especially for a atomic system with a large spontaneous emission rate, in which the intermediate state is not easily populated [19,20]. When applying DROP or OODR spectra into laser frequency stabilization, an extra frequency modulation signal is often needed to generate a dispersion-like error signal [18]. Another method of the two-photon dichroic atomic vapor laser lock can realize a modulation-free laser stabilization, but it requires an extra axial magnetic field [15]. Fortunately, in 2012, Carr et al. extended the use of popular polarization spectroscopy [21] to excited state transitions, and reported a two-color polarization spectroscopy (TCPS) in experiment [22], and subsequently Noh presented a theoretical calculation of TCPS for the cesium $6S_{1/2}$-$6P_{3/2}$-$7S_{1/2}$transition [23]: a circularly polarized pump beam populates atoms from ground state into intermediate state, and simultaneously induces the optical anisotropy in atomic medium, and then which is detected by a counter-propagating linearly polarized probe beam scanned across the upper transition of a three-level atomic system, and a TCPS signal with the dispersive shaped feature is obtained for laser stabilization [24–26]. Kulatunga et al. also experimentally investigated the dependency of the TCPS on the frequency detuning of pump laser in the ^{87}Rb atoms $5S_{1/2}$-$5P_{3/2}$-$5D_{5/2}$ system [24].

In this work, we demonstrated polarization spectroscopy of an excited state transition with high signal-to-noise ratio and completely flat spectral background as well as DROP due to the frequency-stabilized probe laser [18–20], and we call it as double resonance optical pumping-polarization spectroscopy (DROP-PS). Furthermore, different from the previous TCPS, where the two laser beams are often counter-propagating through the atomic medium, we realize the DROP-PS in the case of counter-propagating (CTP) and co-propagating (CP) beams experimental configurations, and have proved that the linewidth of DROP-PS for CTP configuration is obviously narrower that in the case of CP configuration due to the atomic coherence effect in a ladder-type atomic system.

* Corresponding author at: College of Physics and Electronic Engineering, Shanxi University, Taiyuan 030006, China.
E-mail address: ybd@sxu.edu.cn (B. Yang).

https://doi.org/10.1016/j.optcom.2020.126102
Received 18 March 2020; Received in revised form 20 May 2020; Accepted 23 May 2020
Available online 26 May 2020

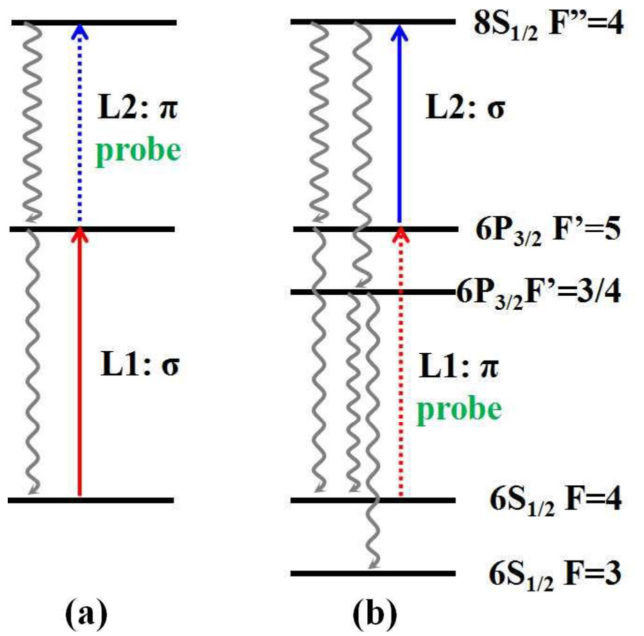

Fig. 1. Schemes of Cs $6S_{1/2}$-$6P_{3/2}$-$8S_{1/2}$ ladder-type atomic system: (a) Two-color polarization spectroscopy (TCPS) based on a three-level atomic system; (b) Double resonance optical pumping-polarization spectroscopy (DROP-PS) based on a five-level atomic system.

2. Principle and experimental setup

The related hyperfine levels of Cs atoms are shown in Fig. 1, the natural linewidth of excited states $6P_{3/2}$ and $8S_{1/2}$ are Γ_1 = 5.2 MHz and Γ_2 = 2.2 MHz, respectively. Different from the previous two-photon TCPS, where the probe is on the excited-state transition as shown in Fig. 1(a), the probe is on the ground-state transition in DROP-PS as shown in Fig. 1(b): A linearly polarized 852.3 nm laser (L1) as probe light is stabilized to the $6S_{1/2}$ F = 4 - $6P_{3/2}$ F′ = 5 transition by saturated absorption spectroscopy (SAS), and its frequency fluctuation is ~0.9 MHz. The probe laser populates some atoms on ground state $6S_{1/2}$ F=4 into intermediate state $6P_{3/2}$ F′ = 5. A circularly polarized 794.6 nm laser (L2) as coupling light is scanned across the $6P_{3/2}$ F′ = 5 - $8S_{1/2}$ F″ = 4 transition, and further excites atoms into the excited state $8S_{1/2}$ F″ = 4, and simultaneously induces an anisotropy in the atomic medium. Some atoms on the state $8S_{1/2}$ F″ = 4 can be optically pumped into another ground state $6S_{1/2}$ F = 3 through other intermediate states such as $6P_{3/2}$ F′ = 3/4 (or $7P_{3/2}$, $7P_{1/2}$ not shown in Fig. 1(b)), which leads to a change of the population in ground state $6S_{1/2}$ F = 4 due to the DROP effect. This optical anisotropy in the medium results in a change of the polarization of 852.3 nm probe beam, and then is detected using a balanced detector to subtract the signals of two orthogonal linear components of probe beam, which are resolved by the combination of a quarter wave plate (QWP) and a polarizing beam splitter (PBS) cube (See Fig. 2). By rotating the QWP, the angle of the plane of polarization of the probe beam with respect to the PBS is set to ~45 degree for a biggest DROP-PS signal. The probe and coupling beams have $1/e^2$diameters of ~1.6 mm, and they are overlapped and separated by dielectric filters (DFs) in a 5 cm long Cs vapor at room-temperature. To reduce the influence of earth's magnetic field, the Cs cell is wrapped with five-layer μ-metal sheets. When the two coherent laser fields interact with atoms in a ladder-type atomic system, there usually exists atomic coherence effect such as electromagnetically induced transparency (EIT). In order to study the influence of atomic coherence effect on DROP-PS, the probe and coupling beams counter-propagate or co-propagate through the Cs cell as shown in Fig. 2.

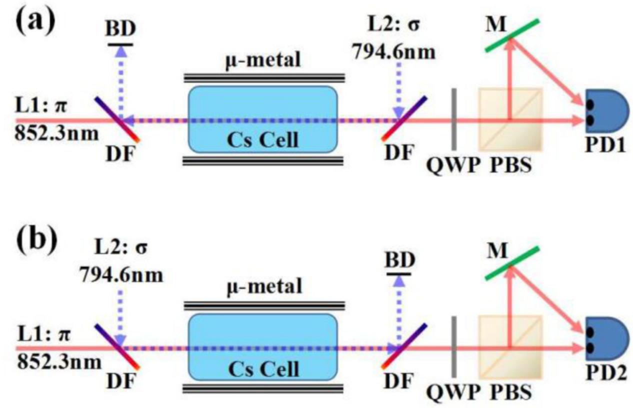

Fig. 2. Experimental setups for observation of a DROP-PS signal: (a) Counter-propagating (CTP) configuration between the probe and coupling laser beams; (b) Co-propagating (CP) configuration. (DF: dielectric filter, BD: beam dump, Cs cell: cesium vapor cell, QWP: quarter wave plate, PBS: polarizing beam splitter, M: mirror, PD: Si photodiode).

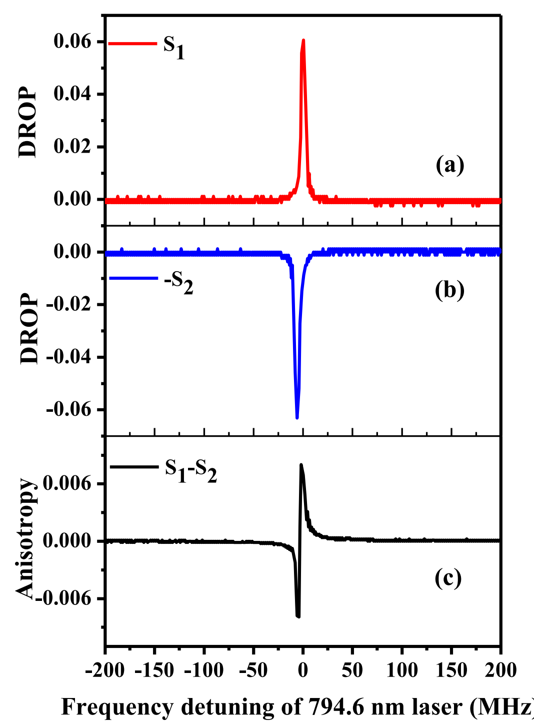

Fig. 3. A typical DROP-PS signal S_1-S_2 with a dispersive shaped profile between the excited states $6P_{3/2}$ F′ = 5 - $8S_{1/2}$ F″ = 4 transition in the CTP configuration. Individual signals S_1 and S_2 are DROP spectra recorded at the two photodiodes of PD1, respectively.

3. Experimental results and discussion

Fig. 3(c) is a typical DROP-PS signal (S_1-S_2) corresponding to the excited states $6P_{3/2}$ F′ = 5 - $8S_{1/2}$ F″ = 4 transition when the frequency of 852.3 nm probe laser is locked to the $6S_{1/2}$ F = 4 - $6P_{3/2}$ F′ = 5 transition and 794.6 nm coupling laser is scanned around the $6P_{3/2}$ - $8S_{1/2}$ transition. Here, the power of coupling and probe beams are ~200 μW and ~100 μW, respectively. They counter-propagate through Cs cell. The signals S_1 and S_2 are two individual DROP spectra recorded at the two photodiodes of balanced detector PD1, as shown in Fig. 3(a) and (b).

Fig. 4 shows the development of DROP-PS as a function of coupling power in the CTP (a) and CP (b) configurations when the probe power

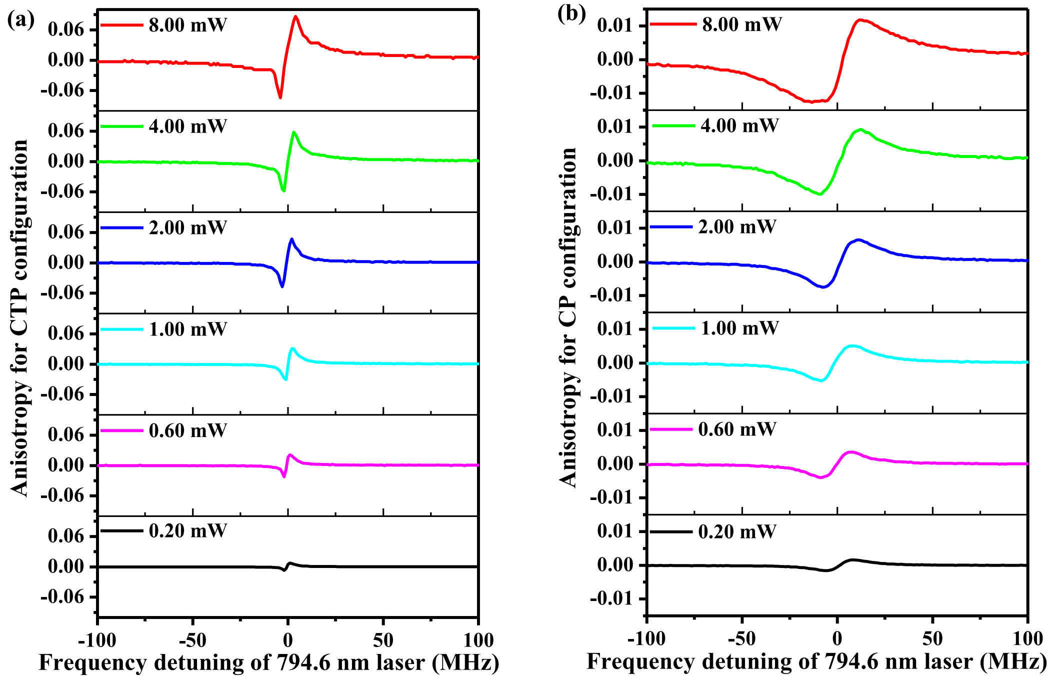

Fig. 4. Evolution of the DROP-PS signals with increasing coupling laser power (a) in the CTP configuration and (b) in the CP configuration.

is set to ~100 μW. With increasing the coupling power, the magnitude of DROP-PS increases and then is close to saturation. The linewidth of DROP-PS also increases with power broadening (The linewidth is defined as frequency difference between the dip and peak of DROP-PS lineshape). It is worth mentioning that there is an obvious differences in linewidth of DROP-PS between the CTP and CP experimental configurations: when the coupling power is increased from ~0.05 mW to ~10.00 mW, the linewidth of DROP-PS is slowly changed from ~3.0 MHz to ~8.0 MHz for the CTP configuration, while that is clearly broadened from ~12.4 MHz to ~27.5 MHz for the CP configuration as shown in Fig. 5. This significant difference is attributed to the atomic coherence between the ground and upper states driven by the two coherent laser fields via a common intermediate state, resulting in an EIT effect, which suppresses the linewidth of DROP-PS only for the CTP configuration in a ladder-type atomic system [27–30]. In theory, the susceptibility under the weak probe laser approximation in a ladder-type EIT atomic system is $\chi = \chi' + i\chi''$, the real part χ' and imaginary part χ'' are related to the dispersion and absorption of the atomic medium, respectively [27].

$$\chi(v)dv = \frac{4i\hbar g_{21}^2/\varepsilon_0}{\gamma_{21} - i\Delta_1 - i\frac{\omega_1}{c}v + \frac{\Omega_2^2/4}{\gamma_{21}-i(\Delta_1+\Delta_2)-i(\omega_1\pm\omega_2)v/c}} \cdot \frac{N}{u\sqrt{\pi}} e^{-v^2/u^2} dv \quad (1)$$

where ω_{21} is the frequency of the Cs $6S_{1/2}$ F = 4 - $6P_{3/2}$ F′ = 5 transition, ω_1 the frequency of probe laser, and so the corresponding frequency detuning $\Delta_1 = \omega_1 - \omega_{21}$. Similarly, ω_{32} the frequency of the $6P_{3/2}$ F′ = 5 - $8S_{1/2}$ F″ = 4 transition, ω_2 the frequency of coupling laser, and its detuning $\Delta_2 = \omega_2 - \omega_{32}$. g_{21} is the dipole moment matrix element for the $6S_{1/2}$ F = 4 - $6P_{3/2}$ F′ = 5 transition, and Ω_2 the Rabi frequency of coupling laser. If collisional dephasing and laser linewidths are negligible, the decay rates are defined by $\gamma_{ij} = (\Gamma_i + \Gamma_j)/2$, where $\Gamma_{i(j)}$ is the natural linewidth of level *i(j)*. *N* is the number density of Cs atoms, *v* the speed of Cs atoms, *c* the speed of light, *u* the most probable velocity. In formula (1), the term $-i(\omega_1 - \omega_2)v/c$ is corresponding to the case of CTP configuration, which is a small term especially for the case of $\omega_1 \approx \omega_2$, so it is a Doppler-free configuration, and a narrow linewidth EIT signal can be obtained due to atomic coherence effect; on the contrary, the term $-i(\omega_1 + \omega_2)v/c$ is corresponding to the CP configuration, EIT signal is not easily observed. The above discussions have been verified by recording the signals of one of the photodiodes

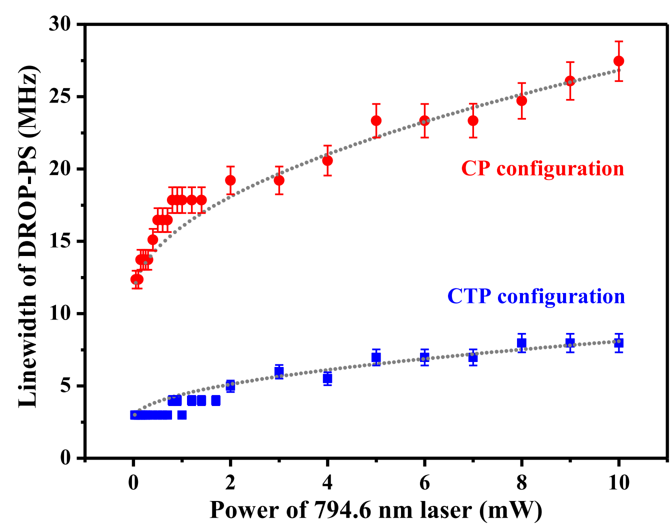

Fig. 5. Measured linewidth of the DROP-PS signals for varying coupling laser power in the CTP and CP configurations. The data points are fitted with the function ~square root of coupling power indicated by the gray-dotted lines.

of balanced detectors PD1 and PD2 in experiment as shown in Fig. 6: when the probe laser is scanned over the whole $6S_{1/2}$ - $6P_{3/2}$ transition, and the coupling laser is resonant on the $6P_{3/2}$ F′ = 5 - $8S_{1/2}$ F″ = 4 hyperfine transition, the PD1 shows a popular EIT signal with narrow linewidth due to atomic coherence effect for the CTP configuration; and PD2 also shows a transparent signal with broader linewidth for the CP configuration due to the DROP effect, because the DROP is a two-photon optical pumping process usually accompanied with spontaneous emission. The SAS is as a frequency reference in Fig. 6.

In addition, with the help of EIT coherence effect, we observe a minimum linewidth of DROP-PS with ~3.0 MHz in the CTP configuration as indicated in Fig. 5. In the weak probe limit, the minimum linewidth of spectroscopy of an excited state transition in a ladder-type atomic system driven by two coherent laser fields for the CTP configuration: $\Gamma_2 + \Gamma_1(k_2 - k_1)/k_1 = 2.6$ MHz (k_1, k_2 are the wave vector of 852.3 nm

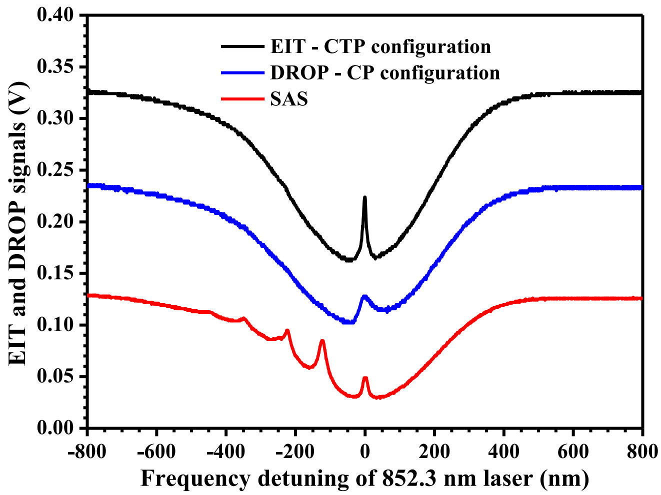

Fig. 6. The probe shows an EIT signal for the CTP configuration, and shows a DROP signal for the CP configuration when the probe laser is scanned over the whole $6S_{1/2}$-$6P_{3/2}$ transition, and the coupling laser is resonant on the $6P_{3/2}$ F′ = 5 - $8S_{1/2}$ F″ = 4 hyperfine transition. The saturated absorption spectroscopy (SAS) is as a frequency reference.

probe and 794.6 nm coupling lasers.), which is only slightly dependent on power broadening of the intermediate state [29,30]. It can be seen that experiment result is close to the value of theoretical prediction, and is also smaller than the natural linewidth $\Gamma_1 = 5.2$ MHz of intermediate state $6P_{3/2}$. However, the minimum linewidth of spectroscopy of an excited state transition for the CP configuration: $\Gamma_2 + \Gamma_1(k_2 + k_1)/k_1 =$ 12.2 MHz, which is agree with experimental minimum linewidth of DROP-PS of ~12.4 MHz as shown in Fig. 5, too [29,30].

4. Conclusions

We demonstrate a DROP-PS technique of an excited state transition based on Cs $6S_{1/2}$-$6P_{3/2}$-$8S_{1/2}$ atomic system in a room-temperature vapor cell. The evolution of DROP-PS lineshapes are measured as a function of coupling power in the CP and CTP experimental configurations. The experimental results have revealed that the linewidth of DROP-PS for the CTP configuration is obviously narrower than that for the CP configuration due to the ladder-type EIT atomic coherence effect. The narrow linewidth DROP-PS with dispersive shape and completely flat spectral background feature is very beneficial for stabilizing the laser frequency to an excited state transition line, which can be widely used in the study of optical filter, Rydberg gases, laser cooling/trapping, and many other experiments on the basis of a ladder-type atomic system.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgment

This work has been supported by the National Natural Science Foundation of China (Nos. 61975102, and 11774210), and the National Key Research and Development Program of China (No. 2017YFA0304502), and the Scientific and Technological Innovation Programs of Higher Education Institutions in Shanxi (No. 2019L0101).

Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.optcom.2020.126102.

References

[1] P.Y. Chang, S.N. Zhang, H.S. Shang, J.B. Chen, Appl. Phys. B 125 (2019) 196.
[2] H.S. Shang, T.Y. Zhang, J.X. Miao, T.T. Shi, D. Pan, X.W. Zhao, Q. Wei, L. Yang, J.B. Chen, Opt. Express 28 (2020) 6868.
[3] W.K. Lee, H.S. Moon, H.S. Suh, Opt. Lett. 32 (2007) 2810.
[4] J. Wang, H.F. Liu, G. Yang, B.D. Yang, J.M. Wang, Phys. Rev. A 90 (2014) 052505.
[5] A.K. Mohapatra, T.R. Jackson, C.S. Adams, Phys. Rev. Lett. 98 (2007) 113003.
[6] S.C. Bell, D.M. Heywood, J.D. White, J.D. Close, R.E. Scholten, Appl. Phys. Lett. 90 (2007) 171120.
[7] P.Y. Chang, T.T. Shi, S.N. Zhang, H.S. Shang, D. Pan, J.B. Chen, Chin. Opt. Lett. 15 (2017) 121401.
[8] M. Lam, S.B. Pal, T. Vogt, C. Gross, M. Kiffner, W. Li, Opt. Lett. 44 (2019) 2931.
[9] E. Becerra, R.T. Willis, S.L. Rolston, L.A. Orozco, Phys. Rev. A 78 (2008) 013834.
[10] J.T. Schultz, S. Abend, D. Döring, J.E. Debs, P.A. Altin, J.D. White, N.P. Robins, J.D. Close, Opt. Lett. 34 (2009) 2321.
[11] Y. Sebbag, Y. Barash, U. Levy, Opt. Lett. 44 (2019) 971.
[12] S.J. Wu, T. Plisson, R.C. Brown, W.D. Phillips, J.V. Porto, Phys. Rev. Lett. 103 (2009) 173003.
[13] B.D. Yang, J. Wang, J.M. Wang, Chin. Opt. Lett. 14 (2016) 040201.
[14] M. Song, T.H. Yoon, Phys. Rev. A 83 (2011) 033814.
[15] F.E. Becerra, R.T. Willis, S.L. Rolston, L.A. Orozco, J. Opt. Soc. Amer. B 26 (2009) 1315.
[16] R. Boucher, M. Breton, N. Cyr, C. Julien, M. Tetu, IEEE Photon. Technol. Lett. 4 (1992) 327.
[17] H. Sasada, IEEE Photon. Technol. Lett. 4 (1992) 1307.
[18] H.S. Moon, W.K. Lee, L. Lee, J.B. Kim, Appl. Phys. Lett. 85 (2004) 3965.
[19] B.D. Yang, Q.B. Liang, J. He, T.C. Zhang, J.M. Wang, Phys. Rev. A 81 (2010) 043803.
[20] H.S. Moon, L. Lee, J.B. Kim, Opt. Express 16 (2008) 12163.
[21] C.P. Pearman, C.S. Adams, S.G. Cox, P.F. Griffin, D.A. Smith, I.G. Hughes, J. Phys. B: At. Mol. Opt. Phys. 35 (2002) 5141.
[22] C. Carr, C.S. Adams, K.J. Weatherill, Opt. Lett. 37 (2012) 118.
[23] H.R. Noh, Opt. Express 20 (2012) 21784.
[24] P. Kulatunga, H.C. Busch, L.R. Andrews, C.I. Sukenik, Opt. Commun. 285 (2012) 2851.
[25] B.D. Yang, J. Wang, H.F. Liu, J. He, J.M. Wang, Opt. Commun. 319 (2014) 174.
[26] B.D. Yang, J.F. Zhang, J.M. Wang, Chin. Opt. Lett. 17 (2019) 093001.
[27] J. Gea-Banacloche, Y.Q. Li, S.Z. Jin, M. Xiao, Phys. Rev. A 51 (1995) 576.
[28] B.D. Yang, J. Gao, T.C. Zhang, J.M. Wang, Phys. Rev. A 83 (2011) 013818.
[29] M. Tanasittikosol, C. Carr, C.S. Adams, K.J. Weatherill, Phys. Rev. A 85 (2012) 033830.
[30] S.M. Iftiquar, G.R. Karve, V. Natarajan, Phys. Rev. A 77 (2008) 063807.