High resolution spectroscopy of Rb in magnetic field by far-detuning electromagnetically induced transparency

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We have presented a high resolution spectroscopy of Rb in magnetic field by far-detuning electromagnetically induced transparency (EIT). The EIT spectrum in the Ξ-type configuration is usually companied by a double resonance optical pumping (DROP) due to the strong optical coupling between the two upper states, leading to the spectral lines seriously deformed and widely broadened for complex relaxation processes in DROP. Here we demonstrate a high resolution spectroscopy by far-detuning EIT for ^{87}Rb $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$ in magnetic fields. The method of far-detuning eliminates the relaxation in DROP to the most extent and decreases the spectral linewidth from more than 20 MHz down to its natural linewidth limit (6 MHz). The deformation of the spectral lines also disappears and the observed spectra are well in accordance with the theoretical calculation. Our work shows that far-detuning EIT is a reliable high resolution spectroscopic method when the relaxation in DROP cannot be neglected, especially for the case of transition to low excited states.

Keywords: Zeeman, high resolution spectroscopy, electromagnetically induced transparency, off-resonance

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1. Introduction

The eigenstate energies of atoms and molecules are determined with high precision based on the high-resolution spectroscopic measurements of optical transitions in the frequency domain.[1–3] It stimulates the development of high resolution spectroscopy, for example, saturation absorption spectroscopy (SAS),[4,5] double resonance optical pumping (DROP),[6–9] electromagnetically induced transparency (EIT),[10–14] and optical–optical double resonance (OODR).[15–20]

As a high resolution spectroscopic method, EIT can make a light beam which is completely absorbed by an atomic medium suddenly go through within a transparency window just by shining in another certain light beam.[10–14] This feature is raised by a coherent optical process in EIT. A typical Ξ-type EIT is shown in Fig. 1(a), where the weak probe beam λ_p on resonance with the $|1\rangle \rightarrow |2\rangle$ transition would be strongly absorbed. When a strong "coupling beam" λ_c resonant with the $|2\rangle \rightarrow |3\rangle$ transition is added, however, absorption of the probe beam can be greatly reduced.[21] If the coupling and probe beams are counterpropagating and have similar frequencies, the total Doppler width of the two-photon process is also greatly reduced. The simulated spectra are shown in Fig. 1(b) for different frequency detunings, where the Doppler effect has also be included.[21,22] The spectral line intensities are all normalized as the detunings will decrease the line intensities greatly.

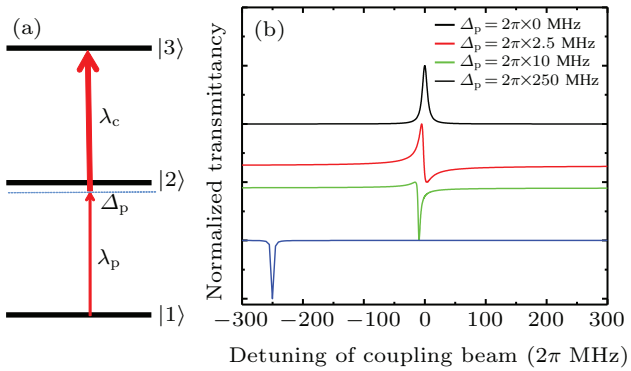


Fig. 1. A typical Ξ-type EIT configuration with cascading excitation (a) and its spectral simulation at different frequency detunings of the coupling beam (b). The spectral intensities are normalized for comparison. In the simulation, $\Omega_p = 2\pi \times 5$ MHz, $\Omega_c = 2\pi \times 5$ MHz, $\gamma_3 = 2\pi \times 3$ MHz, and $\gamma_2 = 2\pi \times 6$ MHz at room temperature $T = 24$ °C.

However, the high resolution of EIT spectroscopy is often obscured by the complex relaxation processes due to the strong optical coupling between states $|2\rangle$ and $|3\rangle$, which evolves into another spectral terminology, DROP.[6–9,23] It is shown in Fig. 2(a), demonstrated on the two photon transitions ^{87}Rb $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$. DROP and EIT methods have different physical mechanisms. DROP is the optical pumping phenomena in atomic system, which are based on the interaction of atoms with two optical fields that are resonant with two transitions that share a common state.[1,24] Unlike DROP arising

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due to population transfer between two dipole forbidden state levels,[6–9] EIT originates from destructive quantum interference mechanism, which is manifest as a decrease or absence of absorption of probe laser due to the quantum coherence effect when the probe laser is resonant with the atomic transition in three-level system.[10–14] These two processes usually occur at the same time for the spectral recording and we have to take both into consideration to account for the experimental observation. This makes it very difficult to investigate the spectrum in an energy range as the spectral line intensities are uncomparable since the DROP effect might be different at different energy or transition frequency with serious state-dependent spectral line broadening.[25,26] This DROP effect is usually neglected in the transition to high Rydberg states since the optical coupling to these states are very weak for small transition electric dipole moments[27–30] or not so serious,[31] which will be discussed later.

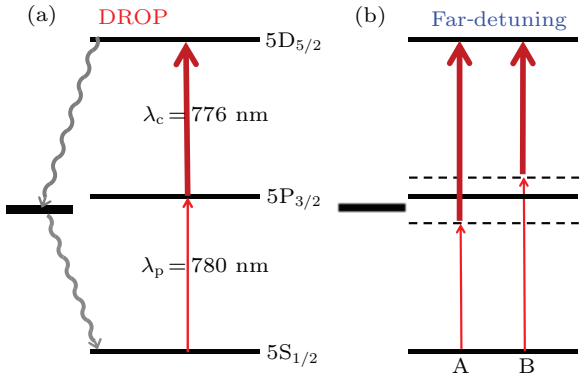


Fig. 2. The level diagram for the optical process in DROP at resonance, demonstrating for the field-free Ξ-type spectrum of ^{87}Rb for transition $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$ (a), and a far-detuning EIT scheme to avoid the pumping effect in DROP (b). The relaxation channel shown in the left of panel (a) plays an important role for DROP. In the latter case, the coupling and probe lasers are off-resonant to the intermediate states, either for blue-detuning (A) or red-detuning (B) of the coupling beams.

In this letter, we adopt a technique of far-frequency detuning shown in Figs. 1(b) and 2(b) to record the high resolution EIT spectrum of ^{87}Rb for $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$ in magnetic fields, which could eliminate the pumping effect completely and present a high quality of experimental spectrum sample that can be comparable with theory strictly.

2. Experimental setup

The EIT experimental setup with coupling beam ($\lambda_c \sim$ 776 nm) and probe beam ($\lambda_p \sim 780$ nm) counter-propagating through a thin Rb vapor cell is shown in Fig. 3. The two lasers are irradiated by two external cavity diode lasers (Moglabs) with linewidth less than 1 MHz. The cylindrical shaped Rb vapor cell (about 1 cm in length and 1.5 cm in diameter) is filled with both isotopes of Rb in their natural abundances ^{85}Rb (72%) and ^{87}Rb (28%). The small size of cell is to remove the effect of the inhomogeneity of the magnetic field. The longitudinal magnetic field is created by a pair of neodymium–iron–boron permanent magnets (M1–M2). The magnets should be separated far enough to keep the magnetic field homogeneous further through the cell.[31,32] A polarizing beam splitter (PBS) is used to combine the coupling and probe beams in space along the magnetic field. The linear polarization of both beams can be taken as the combination of σ^+ and σ^- since the quantum z axis is defined along the magnetic field. Their polarizations are mutually perpendicular. The probe beam (coupling beam) has a waist of about 3 mm (2 mm) and power of 30 μW (15 mW) in our measurement, corresponding to Rabi frequency $2\pi \times 0.56$ MHz and $2\pi \times 4.79$ MHz, respectively. The probe light is detected by a photo detector (PD). The cell is heated to a temperature of ~ 67 °C to enhance the spectral signal to noise ratio (SNR) while the DROP spectrum recorded at lower temperature ~ 55 °C. A spectrum can be recorded by scanning the coupling 776-nm laser.

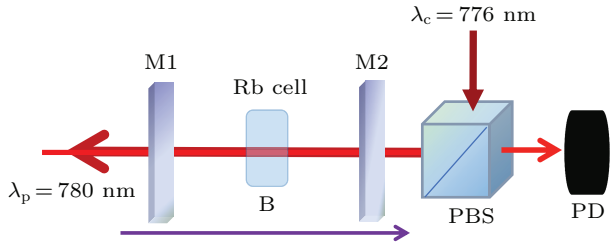


Fig. 3. EIT experimental setup with coupling beam 776 nm and probe beam 780 nm counter-propagating through a thin Rb vapor cell. Both of them are linearly polarized. The magnetic field is provided by a pair of magnets (M1–M2) and the probe light is detected by the photo detector (PD). Pay attention that the magnets should be separated far enough to keep the magnetic field homogeneous.

3. Results and discussion

We record a spectrum for the transition $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$ with different laser frequency detunings. It is presented in Fig. 4 where an SAS spectrum is also given as a frequency reference. For evaluating the quality of the spectrum, a magnetic field of 10.5 Gs (1 Gs = 10^{-4} T) is applied to see its Zeeman splitting and the spectral resolution. Higher magnetic fields require the pair of magnets matches closer but the inhomogeneity comes out. All spectral line intensities are normalized to the one at zero-detuning at point C in Fig. 4(a), corresponding to the transition from the ground state $5S_{1/2}(F = 2)$ to the intermediate state $5P_{3/2}(F = 3)$. We can see that the spectra at C seem to be composed of some narrow lines populated on a broaden background (Fig. 4(b)).[33,34] The narrow lines have different linewidth. The lines at right side are narrower than the left ones. Even we blue- or red-detune the frequency to points D and B, corresponding to $\Delta_p \sim \pm 250$ MHz, there seems no great change for the spectral patterns although the spectral line intensities are decreased half. It indicates that the pumping effect due to the strong coupling between the up-

per two states still predominates with the relaxation processes unable to be ignored.[23,33,34]

When we increase the detuning further, up to $\Delta_p \sim \pm 700$ MHz, as shown at points E and A, the broaden background disappear and the narrow spectral lines come out clearly. However, the spectral lines are weakened more than 50 times compared with that at zero-detuning at point C. Moreover, we can see that the red-detuning (at point A) could give a richer spectral structure than the blue-detuning (at point E). It could be due to the fact that the latter encounters an invasion of resonance from the ground state $F_g = 3$ of ^{85}Rb. While the spectrum at point A is completely pure without pollution from neighboring states since they are all far away in frequency domain. It can also be seen that the spectrum at E still keeps some irregular background compared with that at point A.

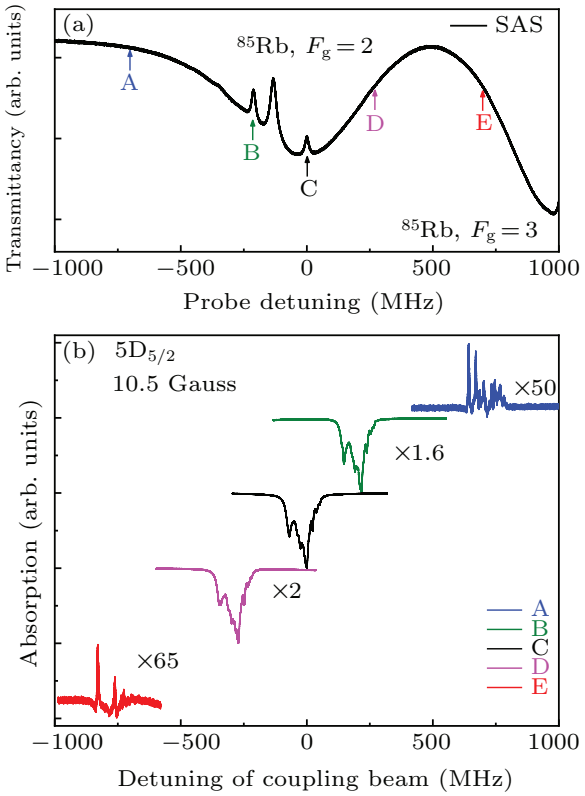


Fig. 4. The experimental observation of EIT spectrum of ^{87}Rb in magnetic fields for transition $5S_{1/2} \rightarrow 5P_{3/2} \rightarrow 5D_{5/2}$ with different laser frequency detunings (b) with an SAS spectrum as reference for the frequency-detuning magnitude (a). A far frequency detuning removes the broaden DROP background away and enhances the spectral resolution greatly although the spectral line intensities weakened as well. Taking spectrum C as reference, the spectra A, B, D, and E have been magnified with intensity scales indicated. The DROP effects are very strong in the spectra within frequency detuning < 500 MHz as shown for B, C, and D in panel (b) but rather weak for A and E at couple beam detuning 750 MHz and −750 MHz. However, obviously, spectrum E is intruded by ^{85}Rb, $F_g = 3$ and only the one of A is immune to the DROP effect and the perturbation from other states.

The spectra recorded at the zero-detuning (point C in Fig. 4(a)) and at far-detuning (point A in Fig. 4(a)) can be magnified for further comparison. It is shown in Fig. 5. The strong pumping laser beam leads to a strong optical coupling between the two upper states, $5P_{3/2}$ and $5D_{5/2}$. In this case, much population to the $5D_{5/2}$ occurs and the relaxation processes in DROP from it will mix with the EIT process.[23,33] As these two different processes happen in the same frequency domain, resulting in an overlap on spectrum in which DROP has a wide spectral broadening. It is shown in Fig. 5(a). While for the spectrum shown in Fig. 5(b), as the DROP effect has been removed completely, it displays a clear spectral structure with narrow linewidth. We have a spectral linewidth (6 MHz) of natural linewidth, much narrower than that ~ 20 MHz in DROP as shown in Fig. 5(a) and those measurements in other previous works.[6,35] We can expect that this spectrum can be well compared with the theory based on EIT in high qualities.

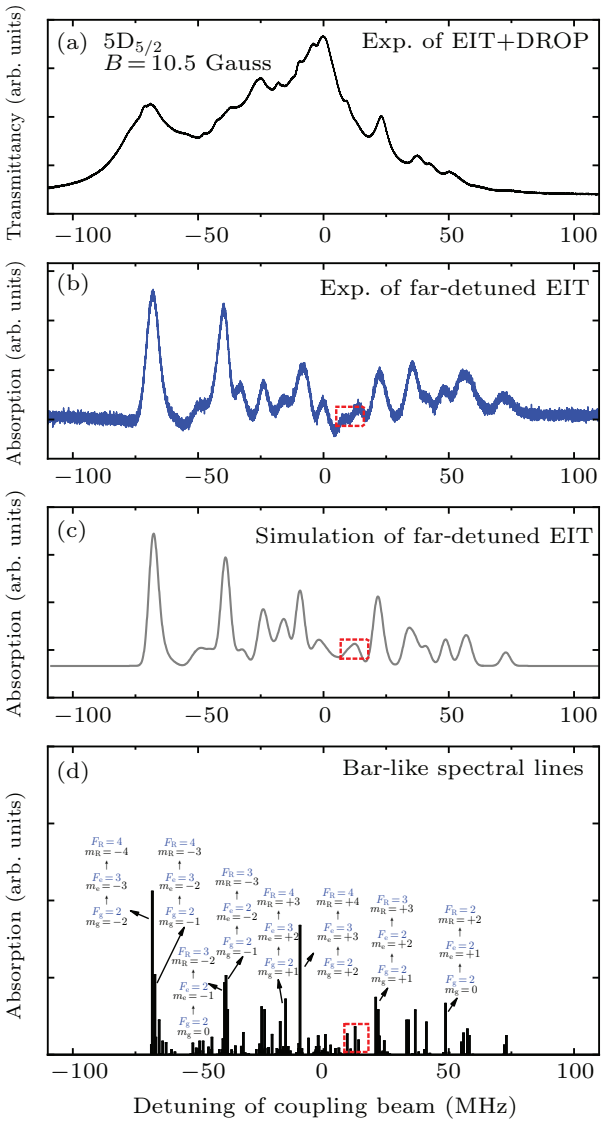


Fig. 5. Spectral comparison of DROP and far-detuning methods. The DROP spectrum (a) and far-detuning spectrum (b) are taken from Fig. 4(b) and magnified here. The simulation in panel (c) recovers the experimental observation very well except the part in dotted frame, which is convinced by a bar-like spectrum (d). A spectral broadening skill in simulation obscures this fine structure. The quantum state assignment for the transitions with considerable intensities are presented in panel (d).

The two optical transition simulation[30] is presented in

Fig. 5(c) where we can see that it has recovered the experimental observation in Fig. 5(b) excellently. This high consistence in spectral structure convinces the reliability of the far frequency detuning method in revealing the high resolution energy level splitting again. A small flaw can be found at frequency detuning at $\Delta_c \sim 12$ MHz where the spectral observation gives a splitting structure but the simulation does not (see the spectrum in the red dotted frame). We present a bar-like spectrum in Fig. 5(d) to have a look at its spectral components and find that there remain separated spectral lines indeed. As a natural linewidth broadening is used in the simulation, the structure has been obscured with a small resolution decreasing. The spectral assignment is also given in Fig. 5(d) to trace its quantum states concerned in the predominant transition. Obviously, DROP loses this ability of high resolution due to the spectral complexity of pumping effect.[8,9,34,36–41]

This comparison indicates that the far frequency detuning method is a nice spectroscopic means to reveal the fine structure in spectrum. It is due to the fact that the spectral method of DROP usually concern extra optical pumping channel, which rapidly reduces the population of the excited states.[23] The extra optical pumping can dump the atoms in the excited states down to another ground state through the intermediate states if it is permissible for the optical selection rule, resulting in the DROP signal showing a spectrum similar to EIT.[6,9,23] Moreover, if the upper excited state has extra decay channel allowed by the optical selection rule, the EIT can not work within a cycling transition any more. The open channel leads to a destructive defect for the EIT spectrum with spectral signal weakened or even reversed, called electromagnetically induced absorption (EIA).[39,42–45] These two optical processes can occur simultaneously and their contribution for the total spectral intensity depends on the applied laser intensity and the selected states. The spectral inhomogeneity on states makes it difficult to quantitatively compare the observation with the theoretical calculation although DROP method is a good candidate for laser frequency locking, where a certain individual spectral line with SNR is needed.[35,46]

As shown in the left panel of Fig. 2(a), the relaxation channel plays an important role for the DROP effect. This process is also dependent on which upper state you excite to. For example, the DROP effect is much stronger for transition to $5D_{5/2}$ through $5P_{3/2}$[33] than that to $5D_{3/2}$ through $5P_{3/2}$.[34] The vigorous optical pumping effect in the former case is also partly due to the fact that their two-photon transition probability is stronger. Obviously, the DROP effect will drop quickly when excited to high Rydberg states[27–30] since the transition electric dipole moment to these sates gets much smaller than to low excited states such as $5D_{3/2,5/2}$. The remained DROP will not destroy the main spectral pattern.[47]

4. Conclusion

In summary, we have recorded and successfully explained Ξ-type far-detuning EIT spectroscopy of ^{87}Rb of state $5D_{5/2}$ in magnetic fields and all the spectra are well explained by theory concerning two photon-transitions with Zeeman-splitting considered. Our work shows that far-detuning EIT is a reliable high resolution spectroscopic method when the relaxation in DROP cannot be neglected.

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References

[1] Demtröder W 2003 *Laser spectroscopy* (Berlin: Springer-Verlag)
[2] Zhang X B and Ye J 2016 *Natl. Sci. Rev.* **3** 189
[3] Bourzeix S, de Beauvoir B, Nez F, Plimmer M D, de Tomasi F, Julien L, Biraben F and Stacey D N 1996 *Phys. Rev. Lett.* **76** 384
[4] Pappas P G, Burns M M, Hinshelwood D D, Feld M S and Murnick D E 1980 *Phys. Rev. A* **21** 1955
[5] Brandenberger J R 1989 *Phys. Rev. A* **39** 64
[6] Yang B D, Gao J, Liang Q B, Wang J, Zhang T C and Wang J M 2011 *Chin. Phys. B* **20** 044202
[7] Zhao J, Zhu X, Zhang L, Feng Z, Li C and Jia S 2000 *Opt. Express* **17** 15821
[8] Moon H S, Lee L and Kim J B 2008 *Opt. Express* **16** 12163
[9] Moon H S and Noh H R 2012 *J. Opt. Soc. Am. B* **29** 1557
[10] Hamid R, Çetintaş M and Çelik M 2003 *Opt. Commun.* **224** 247
[11] Hamid R, Çetintaş M and Çelik M 2004 *Phys. Rev. A* **70** 025805
[12] Harris S E 1997 *Phys. Today* **50** 36
[13] Lezama A, Barreiro S and Akulshin A M 1999 *Phys. Rev. A* **59** 4732
[14] Boller K J, Imamoglu A and Harris S E 1991 *Phys. Rev. Lett.* **66** 2593
[15] Stwalley W C and Wang H 1999 *J. Mol. Spectrosc.* **195** 194
[16] Tao C, Richmond C A, Mukarakate C, Kable S H, Bacskay G B, Brown E C, Dawes R, Lolur P and Reid S A 2012 *J. Chem. Phys.* **137** 104307
[17] Sunahori F X, Nagarajan R and Clouthier D J 2015 *J. Chem. Phys.* **143** 224308
[18] Ip P C F, Bernath P F and Field R W 1981 *J. Mol. Spectrosc.* **89** 53
[19] Nishiyama A, Yoshida S, Nakajima Y, Sasada H, Nakagawa K, Onae A and Minoshima K 2016 *Opt. Express* **24** 25894
[20] Ishiwata T, Tokunaga A, Shinzawa T and Tanaka I 1984 *J. Mol. Spectrosc.* **108** 79
[21] Gea-Banacloche J, Li Y, Jin S and Xiao M 1995 *Phys. Rev. A* **51** 576
[22] Mondal S, Ghosh A, Islam K, Bhattacharyya D and Bandyopadhyay A 2018 *Chin. Phys. B* **27** 094204
[23] Noh H R and Moon H S 2012 *Phys. Rev. A* **85** 1
[24] Ray A, Sabir Ali M and Chakrabarti A 2013 *Euro. Phys. J. D* **67** 78
[25] Fulton D J, Moseley R R, Shepherd S, Sinclair B D and Dunn M H 1995 *Opt. Commun.* **116** 231
[26] Bhushan S, Chauhan V S, M D and Easwaran R K 2019 *Phys. Lett. A* **383** 125885
[27] Bao S, Yang W, Zhang H, Zhang L, Zhao J and Jia S 2015 *J. Phys. Soc. Jpn.* **84** 104301
[28] Bao S, Zhang H, Zhou J, Zhang L, Zhao J, Xiao L and Jia S 2016 *Phys. Rev. A* **94** 043822
[29] Naber J, Spreeuw R, Tauschinsky A and van Linden van den Heuvell B 2017 *SciPost Phys.* **2** 015
[30] Cheng H, Wang H M, Zhang S S, Xin P P, Luo J and Liu H P 2017 *Opt. Express* **25** 33575

[31] Zhang L, Bao S, Zhang H, Raithel G, Zhao J, Xiao L and Jia S 2018 *Opt. Express* **26** 29931
[32] Wang H M, Cheng H, Zhang S S, Xin P P, Xu Z S and Liu H P 2018 *Chin. Phys. B* **27** 094205
[33] Cheng H, Wang H M, Zhang S S, Xin P P, Luo J and Liu H P 2017 *Chin. Phys. B* **26** 074204
[34] Moon H S and Noh H R 2011 *Phys. Rev. A* **84** 033821
[35] Moon H S and Noh H R 2011 *J. Phys. B* **44** 055004
[36] Yuan J, Dong S, Wang L, Xiao L and Jia S 2020 *Laser Phys.* **30** 025201
[37] Moon H S, Lee L and Kim J B 2007 *J. Opt. Soc. Am. B* **24** 2157
[38] Moon H S, Lee W K, Lee L and Kim J B 2004 *Appl. Phys. Lett.* **85** 3965
[39] Failache H, Valente P, Ban G, Lorent V and Lezama A 2003 *Phys. Rev. A* **67** 043810
[40] Das B C, Bhattacharyya D, Das A, Chakrabarti S and De S 2016 *J. Chem. Phys.* **145** 224312
[41] Kim K, Kwon M, Park H D, Moon H S, Rawat H S, An K and Kim J B 2001 *J. Phys. B* **34** 4801
[42] Kim S, Moon H, Kim K and Kim J 2003 *Phys. Rev. A* **68** 063813
[43] Brazhnikov D V, Taichenachev A V and Yudin V I 2011 *Euro. Phys. J. D* **63** 315
[44] Dey S, Mitra S, Ghosh P N and Ray B 2015 *Optik* **126** 2711
[45] Rehman H U, Mohsin M Q, Noh H R and Kim J T 2016 *Opt. Commun.* **381** 127
[46] Yu H, Kim J D, Jung T Y and Kim J B 2012 *J. Korean Phys. Soc.* **61** 1227
[47] Wang H M, Xu Z S, Ma S C, Cai M H, You S H and Liu H P 2019 *Opt. Lett.* **44** 5816