Dynamics in direct two-photon transition by frequency combs

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Abstract – Based on the proposed theoretical model of a three-level system, the optical Bloch equations including the direct two-photon transition (DTPT) process using the optical frequency comb (OFC) were derived and the population distribution of particles in the upper states varying with the velocity of the atoms was obtained. Comparing to the resonance two-photon transition process, that population was increased by a factor of 1.4 without the Doppler shift, which is consistent with our previous experimental results. Simultaneously, the relationships between momentum transfers, and atomic velocity and pulse number were analyzed. When applied to a multi-level system it was found that the population of particles in the excited states increased by a few percentages. The novel approach of DTPT using OFC improved the utilization of comb teeth and atoms, increased the momentum transfer path, reduced the reachable Doppler temperature limit, and encouraged us to use OFC to cool multiple elements simultaneously through the DTPT process. By analyzing the Doppler temperature of $^{133}$Cs and $^{87}$Rb in one dimension, it was found that this process can lower a temperature below 100 mK and generate dipolar molecules $^{133}$Cs$^{87}$Rb via photoassociation, which provides us with a new tool to create dipolar molecules and to investigate their complex rovibrational spectra in ultra-cold chemistry.

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Introduction. – Interaction of atoms with continuous bichromatic laser can give rise to two-photon resonances based on the cascade three energy level system. This technology has been used for two-photon Doppler cooling of alkaline-earth metal [1], as well as metastable helium atoms [2], hydrogen atoms [3] and so on. This method is limited by the availability of cw lasers to a subset of atoms and molecules that have complex internal structure. With the development and wide applications of femtosecond optical frequency combs (OFCs) [4–6], scientific researches in fundamental physics measurements [7], ultrafast optics, strong optical field, ultraviolet and deep ultraviolet measurement [8] have been revolutionized and achieved great advances. The application of OFCs together with two-photon transitions (TPTs) generates a new subject named direct frequency comb spectroscopy [9]. In addition, high-precision spectroscopy measurement [10,11] and the optical frequency standard [12] can also be realized. As a new tool for manipulating atoms, Kielpinski proposed a scheme of using direct two-photon transition (DTPT) to laser cool atoms and molecules [13], and Jayich et al. observed the cooling phenomenon in experiments by using this technology [14]. However, some details of the DTPT process lack of transparency, especially the dynamics in DTPT process when using OFC as the laser source, necessitating further study.

In the previous reports of our group, we used Er-doped fiber OFCs to excite cold $^{87}$Rb 5S-5D DTPT [15], and obtained stronger spectral signal by enhancing resonant TPT process. This is due to the fact that there is a real energy level of 5P state between 5S and 5D states of the $^{87}$Rb atom. We also compared the DTPT process with the resonant TPT process and found that they made equal contribution to the two-photon fluorescence signal [16]. In our recent experiments only DTPT played a major role when Ti:sapphire mode-locked laser is used to interrogate 1S-2S transition of H atoms [17]. There are two reasons, 1) no intermediate real energy level therebetween, and 2) no commercially available deep ultraviolet laser to generate 243 nm or shorter wavelength to drive the transition. The OFC is now showing its unique usage in the DTPT process. After further analysis of the DTPT dynamic effects of the OFC, this article presents another potential application of the OFC to DTPT processes, that is, cooling heteronuclear atoms to achieve the conditions via photoassociation to produce dipolar molecules, and it can also be used to investigate the complex vibration-rotation spectra of dipolar molecules, hence playing an important role in precision manipulation in ultra-cold chemistry.

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absorbing photons with angular frequencies \( \omega_1 \) and \( \omega_2 \), Fig. 1, includes two TPTs processes, one of the two TPTs processes is the resonant TPTs process in which atoms go from ground state 1 to intermediate state 2 by absorbing photons with angular frequencies \( \omega_1 \), and then go from state 2 to final state 3 by absorbing photons with angular frequencies \( \omega_2 \). The other process is the DTPT process in which the atoms go from ground state 1 to excited state 3 directly by absorbing two photons with sum frequency equal to \( \omega_3 \). The population dynamics of these three states can be described by density matrix elements \( \rho_{ij} \) (i, j = 1, 2, 3), which can be calculated by solving the optical Bloch equation (OBEs) under the rotating-wave approximation given by

\[
\dot{\rho}_{33} = i(\Omega_{13}^* \sigma_{13} + \Omega_{23}^* \sigma_{23} - \text{c.c.}) - \Gamma_{33} \rho_{33},
\]

\[
\dot{\rho}_{22} = i(\Omega_{12}^* \sigma_{12} - \Omega_{23}^* \sigma_{23} - \text{c.c.}) + \Gamma_{33} \rho_{33} - \Gamma_{22} \rho_{22},
\]

\[
\dot{\sigma}_{23} = i[\delta_{23} \sigma_{23} + (\Omega_{12}^* \sigma_{13} - \text{c.c.}) + \Omega_{23}(\rho_{33} - \rho_{22})] - \Gamma_{23} \sigma_{23},
\]

\[
\dot{\sigma}_{12} = i[\delta_{12} \sigma_{12} + \Omega_{14}(2 \rho_{22} + \rho_{33} - 1) + \Omega_{13}^* \sigma_{13} - \Omega_{23}^* \sigma_{23}] - \Gamma_{12} \sigma_{12},
\]

\[
\dot{\sigma}_{13} = i[\delta_{13} \sigma_{13} + \Omega_{15}(2 \rho_{22} + \rho_{33} - 1) + \Omega_{13}^* \sigma_{13} - \Omega_{23}^* \sigma_{23}] - \Gamma_{13} \sigma_{13},
\]

where the frequency detuning is defined as \( \delta_{12} = \omega_{12} - \omega_1 \), \( \delta_{23} = \omega_{23} - \omega_2 \), \( \delta_{13} = \omega_{13} - \omega_3 \), \( \omega_1 \) and \( \omega_2 \) are single-photon frequencies that drive transitions from 1 to 2 and from 2 to 3, respectively, \( \omega_3 = \omega_1 + \omega_2 \) is the sum frequency of two photons that drive the transition from 1 to 3 directly. The frequency between two energy levels is defined as \( \omega_{ij} = (E_j - E_i)/\hbar \). \( \Gamma_{ij} \) is the spontaneous decay rate, \( \sigma_{12}, \sigma_{13}, \sigma_{12} \) are the transformed off-diagonal elements of the density matrix and are related with one another by

\[
\sigma_{12} = \rho_{12} e^{-i \omega_1 t}, \quad \sigma_{23} = \rho_{23} e^{-i \omega_2 t}, \quad \sigma_{13} = \rho_{13} e^{-i (\omega_1 + \omega_2) t}.
\]

We define the single-photon Rabi frequency as \( \Omega_{ij} = \mu_{ij} \epsilon / \hbar \) and the two-photon Rabi frequency [18] as

\[
\Omega_{ij} = \beta_{ij} I,
\]

where \( \mu_{ij} \) is the dipole moment of the corresponding transition, \( \epsilon = E(t) e^{-i \omega_1 t} \) is the slowly varying envelope of the laser pulse, \( I \) is the intensity of the laser beam, the coefficient [18] is

\[
\beta_{ij} = \frac{\epsilon^2}{\alpha_0 \hbar^2} \sum_r \left[ \frac{(f|x|\langle r|x|i \rangle)}{\omega_1 - \omega_{ir} - \frac{\hbar}{2}} + \frac{(f|x|\langle r|x|i \rangle)}{\omega_2 - \omega_{ir} - \frac{\hbar}{2}} \right]. \tag{3}
\]

The two-photon Rabi frequency depends on both the laser intensity and the laser frequency detuning. For a train of pulses travelling in the \( x \)-direction with intensity of 0.65 Wm\(^{-2}\) for each comb tooth and beam waist width at about 1.5 mm, the two-photon Rabi frequency for an alkaline metal atom varies with both two-photon detuning \( \Delta \) and single photon detuning \( \delta \) as shown in Fig. 2(a), in which \( \Delta = \omega_1 + \omega_2 - \omega_{ij} \), and \( \delta = \omega_1 - \omega_{ir} \), and \( i, r, f \) indicate initial ground state, intermediate state, final state, respectively. Although the corresponding peak values of different alkali metals are different, the Rabi frequency as shown in Fig. 2 can always be obtained by adjusting the light intensity. The peak value actually represents the value of the resonance TPT enhancement (i.e., \( \Delta = \Delta = 0 \)). For non-resonance of the TPT, we can set the Rabi frequency as the half-peak value in the calculation thereinafter, that is about 0.5 MHz as shown in Fig. 2(b). Although theoretically the Rabi frequency varies continuously with the single-photon detuning as shown in Fig. 2(b), for the OFC,
when the laser detuning frequency becomes larger, the probability of the TPT is almost negligible, which is consistent with our previous observations [16].

**Population distribution.** To clearly illustrate the effects in the DTPT process, we compare the results obtained from eqs. (1) by employing the method introduced by Felinto et al. [19]. Assuming that atoms are initially at the ground state and interact with a train of Gaussian-shaped pulses, the envelope function is \( E(t) = E_0 \exp(-t^2/(2T_p^2)) \). We set the pulse width as \( T_p = 50 \) fs and the pulse repetition period as 10 ns, the typical values of Rabi frequencies and decay rates for alkali atoms are as follows: \( \Omega_{12} = 0.7 \) MHz, \( \Omega_{23} = 0.2 \) MHz, \( \Gamma_{13} = 0.5 \) MHz, \( \Gamma_{33} = 4.2 \) MHz, \( \Gamma_{22} = 37.5 \) MHz, \( \Gamma_{12} = 18.7 \) MHz, \( \Gamma_{23} = 20.8 \) MHz, \( \Gamma_{13} = 2.1 \) MHz.

The population distribution of the upper state \( \rho_{33} \) depends on atomic velocity which is shown in fig. 3. For the Doppler frequency shift caused by the atomic motion, the population of particles at the upper state is related to the amount of frequency shift. Different velocities of the atoms correspond to different frequency detuning. The red solid line indicates that the population of particles at the upper state is related to the change of the velocity when DTPT is taken into consideration. The blue dashed line indicates the situation where DTPT does not occur. The peak of the figure shows the constructive interference. This is because the time interval between pulses is less than the life time of the upper state, which leads to the accumulation of the population of coherent particles.

For the zero speed atoms in fig. 3, where there is no Doppler frequency shift, the two-photon resonance can excite 28% of the atoms from the ground state to the upper state. In this situation, more comb pairs participate in the DTPT process, making approximately 1.4 times more of the population distributed at the upper level state. As previous research has shown, when considering the DTPT process the fluorescence signal intensity increased by a factor of 2 compared to that of resonance TPT process [20], and its amplitude increased by a factor of \( \sqrt{2} \approx 1.4 \), which is consistent with our calculation. By manipulating the two-photon process, our group also observed the fluorescence signal that has equal contribution from DTPT and from traditional TPT processes [16].

Taking the velocity of 155 m/s for Rb and Cs atoms as another example, this velocity is within the room temperature range. The Doppler shift produced by this velocity is 200 MHz for Rb and 180 MHz for Cs atoms. With (without) the DTPT process, an estimate of 5.1% (3.8%) of the population can be distributed at the upper state. In comparison with the resonance frequency when the speed is 0, if we set the repetition frequency of OFC to be 100 MHz, shifting 2 comb teeth can compensate the Doppler frequency shift generated by the speed of 155 m/s (for Rb), or can reduce the Doppler frequency shift to 20 MHz (for Cs). It is because of the compensation effect of the comb teeth that more atoms of different speeds gather at the upper state.

**Momentum transfer.** The momentum transferred on a three-level atom subjected to TPT process can be calculated by [21]

\[
\Delta P = \sum_i \hbar k_i \Delta \rho_{ii},
\]

where \( \hbar k_i \) is the momentum transfer by one pulse when atoms are excited from the ground state to the excited state. As shown in fig. 4, the momentum kick reaches the maximum at specific velocities, which compensate for Doppler shift caused by the atomic motion. The red and blue lines indicate the atom-pulse interaction with and without DTPT, it is clearly shown that the momentum transfer on atoms increases if we consider the DTPT process.

For more pulses interactions, we have compared our results with the single-photon transition (SPT). From fig. 5...
Fig. 5: Total momentum transfer varies with the number of pulses, the red line indicates the result with the DTPT process, while the blue line indicates the result without the DTPT process, and the purple line indicates the result of the SPT process.

we can see that the momentum transfer on atoms driven by SPT is slightly smaller than that driven by traditional TPT, and four times smaller than that driven by the DTPT process.

A train of pulses interact with atoms initially at its ground state. During the DTPT process, the momentum transferred to atoms tends to increase significantly at 90 pulses of interaction. With more pulses, a steady state is gradually attained, concomitant with an equilibrium state between absorption of photons and spontaneous radiation of atoms. The maximum change of momentum depends on the TPT Rabi frequency, which is related to optical intensity. Without considering the DTPT process, we compared the momentum transfer accumulation between TPT and SPT processes and found the momentum transfer produced by the resonance TPT process is slightly higher than that produced by the SPT process. The final momentum transfer depends on the number of the population in the excited state, which is inversely proportional to the spontaneous decay rate of the excited state. For the DTPT process, it is determined by the upper energy level, while the traditional resonance TPT and SPT are determined by the intermediate real energy level. The ratio of the spontaneous decay rate between the upper state and the real intermediate state is 1:9, which leads to the difference in the number of pulses required for momentum transfer in different processes. The increase in the number of pulses involved in momentum transfer also means that the atom absorbs more photons and transfers more momentum. The final momentum transfer depends on the light intensity and the final population of upper-level particles.

Multi-level DTPT. – Due to wavelength limitations of cw laser, the typical TPT process often happened between the ground state and one excited state resulting in a large population of particles staying in the ground state. The application of OFCs can allow multiple excited states to participate in the TPT process, so that more ground-state particles can absorb photons to obtain recoil momentum and multi-level simultaneous TPT is no longer restricted. In the situation where multiple TPT processes may occur at the same time, we consider the isotope $^{133}\text{Cs}$, whose fine structure is shown in fig. 6. Six levels with transition wavelengths close to each other are illustrated. The corresponding transition wavelengths from 761 nm to 921 nm are also labeled.

As discussed above, in the three-level system, the atom obtains greater momentum transfer after the DTPT process. The change in momentum will give the atom a force along the propagation direction of the photons. If the moving direction of the atom is opposite to the force, the atom can be decelerated, thereby reducing the temperature of the atom cluster. The pushing force on atoms during TPT process has been observed in previous studies, such as in the $^{87}\text{Rb}$ experiment [14]; in this report, it is extended to a six-energy-level system to further study its effect on atoms. The recoil momentum is proportional to the population of excited particles. Thus, if the population of pulses required for the excited state to reach the population balance is inversely proportional to the spontaneous decay rate of the excited state. For the DTPT process, it is determined by the upper energy level, while the traditional resonance TPT and SPT are determined by the intermediate real energy level. The ratio of the spontaneous decay rate between the upper state and the real intermediate state is 1:9, which leads to the difference in the number of pulses required for momentum transfer in different processes. The increase in the number of pulses involved in momentum transfer also means that the atom absorbs more photons and transfers more momentum. The final momentum transfer depends on the light intensity and the final population of upper-level particles.
excited particles increases under the interaction of DTPT process, the cooling effect will eventually be enhanced. Reference [14] shows that the Doppler limit of TPT cooling is proportional to the spontaneous decay rate of the upper state, and more DTPT processes mean that lower cooling limits are achieved. By solving the OBEs for the 6-level system, all the parameters are defined in the similar way as we do for the 3-level system, we obtain that the distribution of particle population varies with time taken as shown in fig. 7.

Here we consider the changes in the population of particles in the excited states after the DTPT process. It can be seen from fig. 7 that the population of the excited state including the intermediate state has increased from 1% to about 7%, and only the population on $8S_{1/2}$ state has increased from can be seen from fig. 7 that the population of the excited particles in the excited states after the DTPT process. It as shown in fig. 7.

Fig. 7: The changes of the population distribution in the DTPT process of $^{133}$Cs on the five excited states when compared to that in the DTPT-free process.

Fig. 8: Calculated transition probability of two alkaline metal atoms when scanning $f_{\text{rep}}$, while fixing $f_{\text{ceo}}$ of the mode-locked laser. The dashed line indicates the two-photon transition probability spectrum scanned at $-0.5f_{\text{rep}}$, which is the laser frequency detuned from the resonance transition frequency.

Simultaneously cooling $^{133}$Cs and $^{87}$Rb. – Nowadays, the spectrum output from one mode-locked Ti:sapphire laser can easily cover all above transitions, which ensures the DTPT process occurs throughout a multilevel atomic system. The optical frequency of a particular comb mode can be expressed as $f_n = f_{\text{ceo}} + nf_{\text{rep}}$, where $f_{\text{ceo}}$ is the initial frequency, $f_{\text{rep}}$ is the pulse repetition rate, and $n$ is the order of the comb which is about several millions for the transition in fig. 6 for cesium, and similar order for rubidium. For resonant TPTs, atoms are initially at ground state, there are two intermediate P states and three upper states that are highly likely to occur in multiple TPTs with fixed $f_{\text{ceo}}$ while scanning $f_{\text{rep}}$. The resonant TPTs and DTPTs in rubidium atoms have been experimentally demonstrated in our group [16].

We now consider that the optical comb acts on two different elements $^{87}$Rb and $^{133}$Cs at the same time. We kept $f_{\text{ceo}}$ fixed, and scanned $f_{\text{rep}}$ and one set of transition lines appeared every time when the range of about 30 Hz was scanned. As shown in fig. 8, it can be clearly seen that $^{87}$Rb has 16 fine peaks when the vertical axis was changed to logarithm, while only 14 of the 20 spectral lines of $^{133}$Cs can be clearly distinguished. This is because the parameters we chose were based on experiments using $^{87}$Rb, while the transition frequencies of Cs are very close to each other resulting in partial mergers.

Theoretically speaking, as long as the total frequency of the two photons is equal to the frequency between the upper and lower energy levels, a DTPT can occur. It can be seen from fig. 2 that the Rabi frequency reaches the largest value when each photon meets the resonance condition. This is the resonance enhancement of the intermediate energy level. When a two-photon process does not satisfy resonance conditions, the transition probability will decrease as the amount of photon frequency detuning from resonance increases. The dashed line in fig. 8 shows the two-photon transition probabilities scanned when the laser frequency is detuned $-0.5f_{\text{rep}}$ from the resonance transition frequency. When the detuning exceeds one $f_{\text{rep}}$,
Table 1: Predicted parameters of $^{133}$Cs and $^{87}$Rb for the one-photon and two-photon Doppler cooling.

| Element | Transitions | $\tau$ (ns) [22,23] | $T_{\text{Doppler}}$ ($\mu$K) |
|---------|-------------|---------------------|------------------|
| $^{133}$Cs | 6S$_1/2$-6P$_{1/2}$ | 35 | 109 |
|          | 6S$_1/2$-6P$_{3/2}$ | 31 | 123 |
|          | 6S$_1/2$-6D$_{3/2}$ | 58 | 49 |
|          | 6S$_1/2$-6D$_{5/2}$ | 62 | 46 |
|          | 6S$_1/2$-8S$_1/2$ | 104 | 28 |
|          | 5S$_1/2$-5P$_{1/2}$ | 27.7 | 138 |
|          | 5S$_1/2$-5P$_{3/2}$ | 26.2 | 146 |
|          | 5S$_1/2$-5D$_{3/2}$ | 241 | 12 |
|          | 5S$_1/2$-5D$_{5/2}$ | 241 | 12 |
|          | 5S$_1/2$-7S$_{1/2}$ | 91 | 31 |
| $^{87}$Rb | | | |

the DTPT processes can be ignored. The main contribution to the DTPT process comes from the comb teeth with the frequency near the resonance, even though that DTPT process makes the same contributions as the resonant TPT in those processes.

From the above discussion, it appears the DTPT process allows more atomic levels to participate in the TPT processes, thereby increasing the amount of momentum transfer. Laser cooling results from photons of a laser beam imparting momentum to the atoms.

As a new tool for manipulating atoms, the OFC can obtain a lower atomic Doppler cooling temperature through the DTPT process according to ref. [14]. The one-dimensional Doppler temperature is $3\hbar \Gamma / 2k_B$, while the single-photon Doppler temperature is $\hbar \Gamma / 2k_B$. $\Gamma$ is the decay rate of the upper state and inversely proportional to its lifetime $\tau$. The predicted parameters for the one-photon and two-photon Doppler cooling are shown in table 1. The Doppler temperature is $28 \mu$K for $^{133}$Cs atoms and $12 \mu$K for $^{87}$Rb atoms resulting from DTPT processes by OFC. Those temperatures are lower than the typical single-photon Doppler cooling limits, which are $109 \mu$K for $^{133}$Cs and $138 \mu$K for $^{87}$Rb, respectively. In previous studies using the two-photon Doppler cooling method for Mg atoms, the observed temperature relative to the single-photon Doppler temperature was reduced by more than a factor of 2.5 [24].

As a potential application, the simultaneous Doppler cooled $^{133}$Cs and $^{87}$Rb can be used to create ultra-cold dipolar molecules $^{87}$Rb$^{133}$Cs through photo-association at a temperature below $100 \mu$K [25] which is within the two-photon cooling limit but beyond the single-photon Doppler cooling limit as shown in table 1. At the same time, dipolar molecules have a much more complex rovibrational spectrum structure than atoms, and the DTPT process of the optical comb can also play an important role in precise spectrum measurement.

**Conclusion.** – In conclusion, we analyzed the dynamic effects that appear in the three-level atomic system resulting from direct application of the DTPT process using the OFC. By numerically solving the OBEs, the population of particles in the upper state presents a comb-like distribution with different speeds. Compared to the resonance TPT process, the population is increased by a factor of 1.4 without the Doppler shift in the DTPT process, which is consistent with our previous experimental results. This also leads to the momentum transfer obtained by the atoms. After application of the DTPT process, the total momentum transfer increased as the number of pulses increased. When applied to a multi-level system, it was found that the population of particles in the excited states changed from $-1\%$ to $7\%$. This novel approach improved the utilization of comb teeth and atoms, increased the momentum transfer path, reduced the reachable Doppler temperature limit, and promotes the use of OFC to cool multiple elements simultaneously through the DTPT process. By analyzing the Doppler temperature of $^{133}$Cs and $^{87}$Rb in one dimension, it was found that this process can reach a temperature below $100 \mu$K to generate the dipolar molecules $^{133}$Cs$^{87}$Rb via photoassociation, which provides a new tool for creating dipolar molecules and investigating their complex rovibrational spectra in ultra-cold chemistry.

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