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Precision measurement of cesium 6S–7S two-photon spectra with single trapped atoms

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A new method to precisely measure the hyperfine spectra of a weak atomic transition line with very low probe power is presented by combining the optical pumping and state detection techniques. By adopting such a method, we demonstrate an experiment to measure the cesium 6S–7S two-photon spectra with optically trapped single atoms. The power of the probe beam in our experiment is on the micro-watt level and the corresponding intensity is on the level of hundreds of watts per square centimeter. Both are much weaker than that of traditional fluorescence detection measurements. The corresponding hyperfine coupling constant A_{hps} of the 7S state is also determined, and is in good agreement with former measurements.

1. Introduction

The hyperfine spectra of weak dipole forbidden transitions are very difficult to observe directly in atomic physics. The two-photon process, in which the two photons separately couple an intermediate state to the ground and excited states with dipole permitting transitions, has been a commonly used method to measure these dipole forbidden hyperfine spectra with relatively low-power lasers.[1)] The retro-reflecting configuration to measure the two-photon spectra (TPS) also promises a Doppler-free spectrum line, and thus precision measurements of atomic structures, such as the Rydberg constant,[2)] hyperfine coupling constants,[3–6)] and state energy levels of unstable atomic isotopes,[7)] can be performed. For example, with the aid of an optical cavity to increase the probe beam power the technique has been used to study the famous hydrogen 1S–2S structure with increasing precision for decades.[2,8)] The S–S transitions of alkali atoms have no linear Zeeman effect and have been regarded as an optical frequency standard. Therefore, the S–S TPS of cesium and rubidium has been studied extensively.[6,9–14)] Up to now, the cesium 6S–8S, 9S[6,9,14)] and rubidium 5S–7S TPS[10)] have been reported and the absolute frequencies of cesium 6S–8S[12)] and rubidium 5S–7S[11)] two-photon transitions have been measured with unprecedented precision. Although one-photon spectroscopy of cesium 6S–7S performed using 540 nm laser has been studied for the testing of the standard model,[15–17)] the corresponding TPS have not been formally reported. In all the TPS measurements with atomic vapors, a high-power laser beam is often required to obtain an observable signal due to the weak transition strength. The power of the probe beam is usually on the level of tens to hundreds of milliwatts (mW) with a beam waist of tens of micrometers. To measure the spectra with a weaker-transition, higher-power probe beam is required.

In this paper, we present a method to precisely measure the hyperfine spectra of a weak transition in cold atoms by combining the optical pumping and atomic state detection techniques. The cold atoms provide a long interaction time with the probe beam. In principle, no matter how weak the the atomic transition and the probe beam power are, the atoms could be pumped to other states with sufficiently long interaction times. The state detection then reveals the result of the optical pumping process. We experimentally demonstrate this method by the cesium 6S–7S TPS measurement based on optically trapped single atoms. Benefitting from the long interaction time between atom and probe beam, the intensity of the probe beam in our experiment is much weaker than that of the traditional fluorescence detection TPS method. The corresponding hyperfine coupling constant A of the 7S state is also determined, and our result is in good agreement with the former experimental data obtained by one-photon spectroscopy[15)] and the optical–optical double resonance (OODR) method.[18,19)]

2. Theoretical model

In our method, we consider a three-level atom with two ground states $|1\rangle$ and $|2\rangle$ and one excited state $|e\rangle$ [see Fig. 1(a)]. The atom is initially prepared in state $|1\rangle$. A weak probe laser pulse couples state $|1\rangle$ and $|e\rangle$ with Rabi frequency Ω. Once the atom is pumped into the excited state $|e\rangle$, it will decay back to $|1\rangle$ and $|2\rangle$ with rates γ_1 and γ_2, respectively. Thus, the dynamics of the atomic populations on three energy levels obeys the Bloch equations:

$$\begin{aligned}
\dot{\rho}_{11} &= -\frac{i\Omega}{2}(\tilde{\rho}_{e1} - \tilde{\rho}_{1e}) + \gamma_1\rho_{ee},\\
\dot{\rho}_{ee} &= -\frac{i\Omega}{2}(\tilde{\rho}_{1e} - \tilde{\rho}_{e1}) - \Gamma\rho_{ee},\\
\dot{\rho}_{22} &= \gamma_2\rho_{ee},\\
\dot{\tilde{\rho}}_{1e} &= -(\gamma + i\Delta)\tilde{\rho}_{1e} - \frac{i\Omega}{2}(\rho_{ee} - \rho_{11}),
\end{aligned} \tag{1}$$

where $\tilde{\rho}_{1e} = \rho_{1e}\exp(-i\Delta t)$ with Δ the frequency detuning of probe beam, $\Gamma = \gamma_1 + \gamma_2$ the total decay rate from excited state, and γ the transverse decay rate. In our case, only radiative damping exists, and thus $\gamma = \Gamma/2$. Here, we consider a long probe pulse with time duration $t \gg 1/\Gamma$, and in this limit the Rabi oscillation between $|1\rangle$ and $|e\rangle$ can be reduced and the assumption $\dot{\tilde{\rho}}_{1e} = 0$ is physically reasonable. Having this, and by solving Eq. (1), we obtain the population in state $|2\rangle$ as:

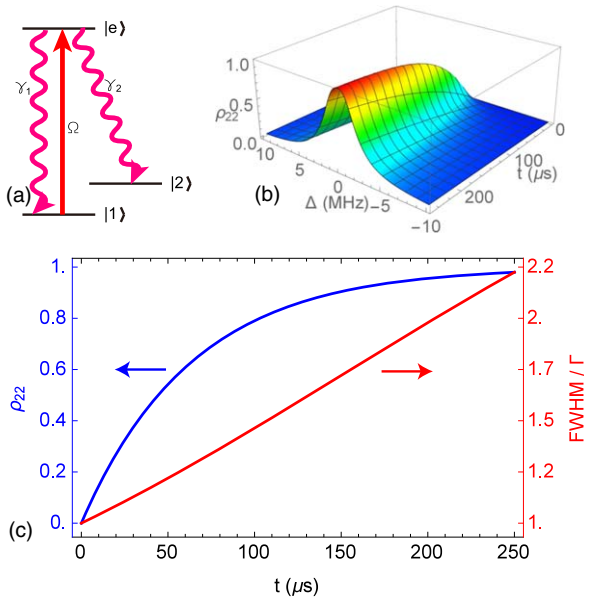

Fig. 1. (Color online) (a) Model of the optical pumping process with two ground states $|1\rangle$, $|2\rangle$ and excited state $|e\rangle$. A weak probe laser pulse couples states $|1\rangle$ and $|e\rangle$ with Rabi frequency Ω. The excited state $|e\rangle$ decays to $|1\rangle$ and to $|2\rangle$ with rates γ_1 and γ_2. (b) Dependence of population ρ_{22} on frequency detuning Δ of probe beam and probe pulse duration t according to Eq. (3). The population on state $|2\rangle$ gives the spectrum of atomic transition $|1\rangle \leftrightarrow |2\rangle$. (c) Population on $|2\rangle$ at resonance $\Delta = 0$ and ratio of the spectrum FWHM over natural linewidth Γ versus the probe pulse length t. In (b) and (c), the parameters $\beta = 0.5$, $\Omega = 0.25$ MHz, and $\Gamma = 2$ MHz are used.

$$\rho_{22} = 1 - \frac{\lambda_+}{\lambda_+ - \lambda_-} e^{-\lambda_- t} - \frac{\lambda_-}{\lambda_- - \lambda_+} e^{-\lambda_+ t}, \tag{2}$$

with $\lambda_\pm = \frac{\Gamma(s+1)}{2}[1 \pm \sqrt{1 - 2\beta s/(1+s)^2}]$, where $s = 2\Omega^2/(\Gamma^2 + 4\Delta^2)$ and $\beta = \gamma_2/\Gamma$. In the case of low pumping regime with $\Omega \ll \Gamma$ ($s \ll 1$) Eq. (2) can be approximated by:

$$\rho_{22} = 1 - \exp\left[-\frac{\Omega^2 \beta \Gamma t}{\Gamma^2 + 4\Delta^2}\right]. \tag{3}$$

Figure 1(b) shows the dependence of the population ρ_{22} on the frequency detuning Δ of the probe beam and probe pulse duration t. One can see that with a constant pulse duration the population on state $|2\rangle$ gives the spectrum of the transition $|1\rangle \leftrightarrow |e\rangle$ when the frequency of the probe beam scans across the resonance. When $\frac{\Omega^2\beta\Gamma t}{\Gamma^2+4\Delta^2} \ll 1$, Eq. (3) can be further simplified to $\rho_{22} = \frac{\Omega^2\beta\Gamma t}{\Gamma^2+4\Delta^2}$, which is exactly the natural Lorentzian line shape obtained by traditional fluorescence or absorption detection methods. By increasing the probe pulse length, more population is pumped to state $|2\rangle$; meanwhile, the full width at half-maximum (FWHM) of the spectrum becomes wider [see Fig. 1(c)]. This linewidth broadening can be considered the result of the saturation effect. When the pulse length is further increased, the population in $|2\rangle$ will be saturated obviously and a flattened top will appear on the spectrum. We also see here that no matter how weak the coupling between probe beam and atomic transition $|1\rangle \leftrightarrow |e\rangle$ is, the spectrum can still be obtained by measuring the population in $|2\rangle$ with an efficient long probe pulse. The population measurement of state $|2\rangle$ can be achieved by probing it through a self-recycling transition with very high fidelity and high signal-to-noise ratio. In other words, once the atom is pumped from $|1\rangle$ to $|2\rangle$, the pumping effect can be recorded with an efficiency of 1. In a real system, γ_1 is comparable to γ_2, and thus after scattering only several photons from the atom in state $|1\rangle$ can be pumped to $|2\rangle$. The effective pumping efficiency for one-photon is $\eta = 1/n$ with n the total scattered photon number. This efficiency is much higher than the detection efficiency of scattered photons in traditional fluorescence detection measurements, in which a large-aperture lens and photon-multiplying tube are usually used. Thus, the method provided here should be more sensitive and could be used to measure the spectrum of weak transitions with very low probe power. Of course, our method relies on the optical pumping effect between two ground states via excited states, and does not work for those systems with only one hyperfine ground state.

3. Experiment of cesium 6S–7S TPS measurement

Next, we demonstrate our method by measuring the cesium 6S–7S TPS with optically trapped single cesium atoms. The laser wavelength to drive the 6S–7S two-photon transition is 1079.01 nm. In our experiment, the atom is first loaded from a magneto-optical trap (MOT) into a micro-sized dipole trap formed by a strongly focused 1079 nm laser beam with linear polarization. The minimum beam waist of the trap is approximately 2 μm. The setups of the MOT, micro-sized optical trap, and loading process are the same as in our previous work.[20] The loading time of the optical trap from the atomic ensembles prepared by MOT is very short since that the optical trap overlaps with the MOT very well. As long as the optical trap is switched on the atom will be loaded into the trap immediately. With the aid of light assistant collision, the atom would lose two-by-two, and in the end there will be either one atom or no atom left in the trap.[21,22] The lifetime of the single atom in the trap is measured as about 30 s. The trap beam is locked to a reference cavity made from ultra-low expansion glass to ensure long-term frequency stability. The trap beam has a two-photon frequency detuning of approximately 10 GHz above the 6S–7S transition. Thus, it provides only a trapping effect for the cesium atoms. With 15 mW laser power we can achieve a trap depth of approximately -0.5 mK. The 1079 nm trap beam is also phase-modulated by a fiber-based electro-optical modulator (EOM) with a tunable modulation frequency from 1 to 10 GHz. The driving microwave for the EOM is from a frequency synthesizer locked to a rubidium frequency standard. The sideband with an order of -1 is used as the probe beam to excite the two-photon 6S–7S transition.

Although in our theoretical model the optical pumping beam couples the ground state $|1\rangle$ and excited state $|e\rangle$ via a one-photon process, the model still works for the two-photon excitation case since the excitation of the intermediate state can be adiabatically eliminated due to very large one-photon frequency detunings.[23] The coupling between the ground state 6S and excited state 7S via two-photon excitation can then be reduced as a two-level system interacting with an effective single beam, for which the effective Rabi frequency of the two-photon excitation process depends on the one-photon detuning δ_i through $\Omega = \sum_i \frac{\Omega 1_i \Omega 2_i}{\delta_i}$, where i represents the number of hyperfine states in the intermediate state $6P_{1/2}$ or $6P_{3/2}$, $\Omega 1_i$ is the one-photon Rabi frequency between 6S and 6P states, and $\Omega 2_i$ is for 6P $\leftrightarrow$ 7S transitions. The two

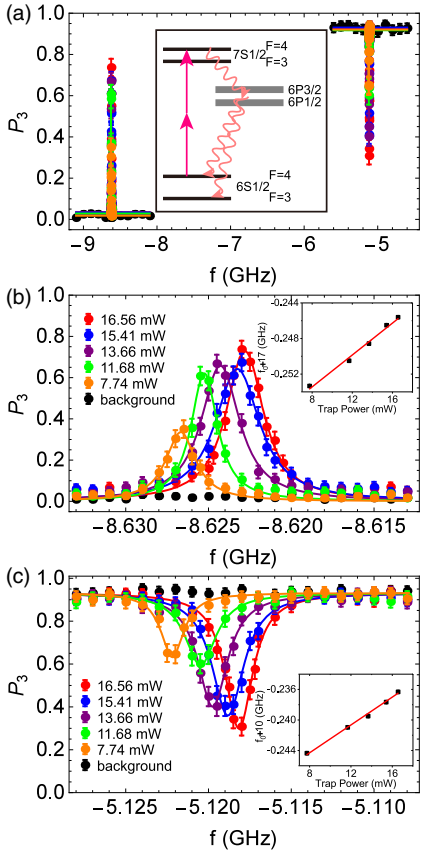

Fig. 2. (Color online) (a) Spectroscopy of the cesium 6S–7S TPS measured by the aforementioned optical pumping and state detection method. The inset shows the related state level configuration of 6S–7S TPS. (b) and (c) are the magnified spectra of $6S_{1/2}$ $F=3$–$7S_{1/2}$ $F=3$ and 6S1/2 $F=4$–$7S_{1/2}$ $F=4$ transitions on single atoms trapped with different trap powers. Probe powers are kept as 0.3% and 0.23% of the trapping power, respectively, in these two measurements. The solid data points are the backgrounds with no probe beam applied when the deepest trap are used. In (c), the atom is initially prepared in state of $6S_{1/2}$ $F=3$ and the atoms that have been pumped to $6S_{1/2}$ $F=4$ are pushed out of the trap. Thus, the background is approximately 1. Insets of these two plots show the dependence of spectrum line center as a function of trap power and the red lines are the linear fittings. In (b) and (c), each data point is obtained by using approximately 120 atom samples, and the error bars show the uncertainty of $\pm\sigma$, with σ being one standard deviation of statistics on the binomial distribution.

one-photon detunings for $6P_{1/2}$ and $6P_{3/2}$ are approximately -73.9 and -57.3 THz, respectively, which reduce the two-photon Rabi oscillation dramatically in our case. Owing to the selection rule of two-photon excitation, only transitions of $6S_{1/2}$ $F=3$–$7S_{1/2}$ $F=3$ and $6S_{1/2}$ $F=4$–$7S_{1/2}$ $F=4$ are permitted.[24,25] Once the atom is pumped to 7S $F=3$ state by the two-photon process, it will decay back to 6S $F=3$ and $F=4$ states via intermediate states $6P_{1/2}$ and $6P_{3/2}$ with probabilities of 3/8 and 5/8, respectively. However, when the atom is pumped to 7S $F=4$, it will decay back to 6S $F=3$ and $F=4$ states with probabilities of 7/24 and 17/24, respectively. Thus, it can be seen that the atom will be pumped to the other state from the $6S_{1/2}$ $F=3$ or $F=4$ states by only scattering several photons. The 6S–7S TPS can then be obtained by detecting the specific state, as mentioned above.

The typical energy levels of 6S–7S TPS measurements made using our method are shown in the inset of Fig. 2(a). The atom is initially prepared in $6S_{1/2}$ $F=3$ or $6S_{1/2}$ $F=4$ by optical pumping by MOT beams depending on which transition is probed. When the transition $6S_{1/2}$ $F=4$–$7S_{1/2}$ $F=4$ is probed, the atom is prepared in $6S_{1/2}$ $F=4$. Along with the probe sideband of the 1079 nm laser beam approaching the resonance frequency of the two-photon transition, the atom will be optically pumped to the $6S_{1/2}$ $F=3$ state via $7S_{1/2}$ $F=4$ with a certain probability. After the two-photon optical pumping process, atoms in the $6S_{1/2}$ $F=4$ state are pushed away from the trap by an 852 nm laser beam resonating to the transition of $6S_{1/2}$ $F=4$–$6P_{3/2}$ $F=5$. Finally, the MOT beams are illuminated on the trap again to see if the atom exists or not. If it is in the trap, it means that the atom is in the $6S_{1/2}$ $F=3$ state; otherwise, it is in the $6S_{1/2}$ $F=4$ state. This state detection has a fidelity of 0.97 in our experiment. The details of the state preparation and detection can be found in Ref. 20. The measured spectra are shown in Fig. 2(b). Here, we show a set of spectra for the $6S_{1/2}$ $F=4$–$7S_{1/2}$ $F=4$ line when the atom is loaded in traps with different depths. The duration of the probe pulse is fixed to 0.2 ms and the probe beam power is kept as 0.3% of the total 1079 nm laser beam. Thus, the minimum probe power is only 24 μW. When taking the 2 μm beam size into account, the minimum intensity is approximately 380 W cm^{-2}, which is much lower compared to that in traditional TPS measurements made with an atom vapor cell. If a longer probe pulse is used, the intensity can be even lower. Each spectrum curve is fitted by Eq. (3) with the replacement of $\Delta=2\omega-\omega_0$, where ω and ω_0 are the frequencies of the probe beam and resonance frequency, respectively, of the atomic transition. Since there is light shift on state $6S_{1/2}$ $F=4$ induced by the trap beam, the center of the transition line shifts along with trap power, which is shown in the inset of Fig. 2(b). Clearly, the shift of the center frequency is linear to the trap power. This is actually the ac Stark shift on the $6S_{1/2}$ $F=4$ state due to one-photon interaction with 6P states and thus is linear to the trap power. If a blue detuned trap[26] where the intensity is zero at the trap center is used the ac Stark shift would be eliminated. The center frequency at zero trap power is determined as $\nu_{44}=-17.260\,61\pm0.000\,72$ GHz from curve fitting. The decay rate of the 7S excited states is 1.8 MHz.[27] However, the measured linewidth in our work is greater than 2 MHz, which is mainly limited by the saturation-like effects when considerable part of the population is pumped to the other hyperfine state. This effect has been discussed with the theoretical model.

Figure 2(c) shows the spectra of $6S_{1/2}$ $F=3$–$7S_{1/2}$ $F=3$ when the 1079 nm trap laser is locked to same longitudinal mode of the reference cavity. In this measurement, the atom is initially prepared in state $6S_{1/2}$ $F=3$ and is then pumped to $6S_{1/2}$ $F=4$ by the two-photon process. The state detection process only detects the population on $6S_{1/2}$ $F=3$; thus, the spectra for this transition line are upside down relative to that of $6S_{1/2}$ $F=4$–$7S_{1/2}$ $F=4$. The fitting function is also modified accordingly. In these measurements the length of the probe pulse is also fixed to 0.2 ms and the probe beam power is kept as 0.23% of the overall 1079 nm laser beam. Thus, the minimum probe power is only 19 μW and the corresponding intensity is 300 W cm^{-2}. The inset of Fig. 2(c) shows the dependence of center frequency on the trapping power, and the linear fitting gives the center frequency in free space to be $\nu_{33}=-10.251\,60\pm0.000\,58$ GHz.

Figure 2(a) shows the overall spectra of 6S–7S transition. The frequency difference of the probe beam for the two

transitions in free space is determined as $\delta\nu = \nu_{33} - \nu_{44} = 7.009\,01 \pm 0.001\,30$ GHz. Taking into account the hyperfine splitting between the 6S $F = 3$ and $F = 4$ states $\delta_{\mathrm{hps,6S}} = 9.192\,631\,77$ GHz, we can obtain the hyperfine splitting between 7S $F = 3$ and $F = 4$ states as $\delta_{\mathrm{hps,7S}} = \delta_{\mathrm{hps,6S}} - \delta\nu = 2.183\,61 \pm 0.001\,28$ GHz. The hyperfine magnetic dipole coupling constant for the 7S state is then $A_{\mathrm{hps,7S}} = \delta_{\mathrm{hps,7S}}/4 = 545.90 \pm 0.32$ MHz, which is in good agreement with the former measurements obtained by the one-photon excitation[15] and OODR methods.[18,19]

4. Discussions and conclusions

In our measurement, we used trapped single atoms with the probe beam propagating along with the trap beam. The trapped single atom ensures that there is no pressure-induced or collisional shift of the transition frequency. The atom temperature in the trap is below 10 μK,[20] corresponding to an atom velocity slower than 0.035 m s^{-1}. Thus, the Doppler shift is estimated to be less than 40 kHz. The main uncertainty of the hyperfine coupling constant A_{hps} comes from the uncertainty of the ac Stark shift due to the statistical error of the data points. Each data point in the spectrum is obtained using approximately 120 atom samples, and the error bars show the uncertainty of $\pm\sigma$, with σ one standard deviation of statistics on the binomial distribution. In principle, if more atoms are used, the uncertainty will be further reduced.

The data-acquisition time for a single run of the measurement is approximately 3 s and 200 runs are used to effectively load approximately 120 single atoms. Therefore, the total time for one data point in Fig. 2 requires approximately 10 min. The time to obtain a single spectrum is then approximately 3.5 h. If more atoms are loaded into a larger trap or atom-lossless state detection[28,29] for single atoms is adopted, the data-acquisition time can be dramatically reduced. For example, if 10^4 atoms are loaded in to a larger trap, each data point could be obtained in only one experimental run. The time to obtain a single spectrum would be reduced to approximately 1 min. At the same time, due to the large number of atoms being used, the data uncertainty could be also suppressed.

Our method depends on optical pumping and atomic state detection, and it is assumed that the ground states should have no decay channels. However, the storage time of the trapped atom and lifetime of a ground state suffer from the collisions of background atoms, and they are no longer infinite. This will be the final limitation on the intensity of the probe beam. In our experiment, the typical storage time of the trapped single atom is about 30 s and the lifetime of the ground state hyperfine states is approximately 0.7 s. If the probe pulse duration is taken as 0.7 s, the probe intensity can be approximately 3500 times weaker than what we have adopted for a 0.2 ms pulse duration.

In conclusion, we have presented a method to precisely measure the hyperfine spectroscopy of a weak atomic transitions with low probe power by combining both the optical pumping and state detection techniques. We have finished the measurement of cesium 6S–7S TPS with optically trapped single atoms. The intensity of the probe beam is only on the μW level, and the pump intensity is much lower than that in traditional fluorescence-dependent methods. We also determined the hyperfine coupling constant A_{hps} of the 7S state. The result is in good agreement with that of the former measurements. Our method can also be used to probe the spectroscopy of other weak transitions, such as electric quadrupole transitions with a one-photon process, with relatively low probe power and even on the single atom level.

Acknowledgments

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