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RESEARCH ARTICLE

Coherent effects in a three-photon CPT process of Ca^+

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Linewidth narrowing and other quantum coherent effects based on three-photon coherent population trapping (CPT) in Ca^+ ions are investigated. If the propagation directions of the three lasers obey the phase matching condition, the dark linewidth resulting from the CPT can be very narrow, and it can be controlled by adjusting the parameters of the lasers.

Keywords coherent population trapping (CPT), linewidth, coherent effects

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1 Introduction

Microwave atomic clocks have achieved fractional uncertainties below 1 part in 10^{15} after 50 years of development [1]. In the development of the frequency standard, some new technologies are used, one of which is coherent population trapping (CPT) method [2]. Besides high-precision clock [3], CPT can be used in quantum phase gate [4] and atomic transistors [5]. CPT phenomenon has been well investigated both in theory and in experiment [2, 3, 6]. Recently, two-photon CPT for an optical clock based on neutral ^{88}Sr [7] and three-photon coherent processes in four-level systems in both neutral alkaline earth atoms and ions have been studied [8, 9]. Some interesting effects have been investigated, such as the variable width of the clock transition which can be continuously adjusted from the MHz level to sub-mHz [8].

In the scheme described here, a three-photon CPT process is able to be involved in a Doppler-free configuration [10]. This process can occur in several ions (Ca^+, Sr^+, Ba^+) which at present are used or proposed for optical clock [11, 12]. For those ions listed above, the narrow dark line resulting from CPT corresponds to the quadrupole transition between the $n\mathrm{S}_{1/2}$ ground states and $n\mathrm{D}_{5/2}$ metastable states, whose electric-dipole forbidden line has been studied and probed directly in former experiments [11, 13–15]. The three-photon resonance is analogous to a two-photon CPT configuration where a trade-off between robustness and clock precision has to be made. By varying the intensities of the three optical beams, the dark linewidth can be adjusted.

2 Atomic structure and model analyzing

The atomic structure under consideration consists of four energy levels interacting with three light fields: a strong coupling field, a weak coupling field, and a probe field, as shown in Fig. 1. The three light fields connect the state $|1\rangle$ to the state $|4\rangle$ via two intermediate states, $|2\rangle$ and $|3\rangle$. For simplicity, we ignore all effects of polarization of the light fields. Ω_p, Ω_s, and Ω_w are Rabi frequencies of the probe field, the strong coupling field, and the weak coupling field associated to atomic transitions $|1\rangle \rightarrow |2\rangle$, $|2\rangle \rightarrow |3\rangle$, and $|3\rangle \rightarrow |4\rangle$, respectively. $\Delta_w = \omega_w - \omega_{21}$, $\Delta_s = \omega_s - \omega_{32}$, and $\Delta_p = \omega_p - \omega_{43}$ are the detunings between the field frequencies (ω_w, ω_s, and ω_p) and the atomic resonance frequencies (ω_{21}, ω_{32}, and ω_{43}) respectively.

The decay rate from state $|2\rangle$ and $|4\rangle$ to $|1\rangle$ is written as γ_{21} and γ_{41}, respectively. The total decay rate of state $|3\rangle$ is written as γ, while γ_{31}, γ_{32} and γ_{34} denote the decay from state $|3\rangle$ to $|1\rangle$, $|2\rangle$ and $|4\rangle$, respectively. It is evident that $\gamma = \gamma_{31} + \gamma_{32} + \gamma_{34}$.

Under the rotating wave approximation (RWA), the

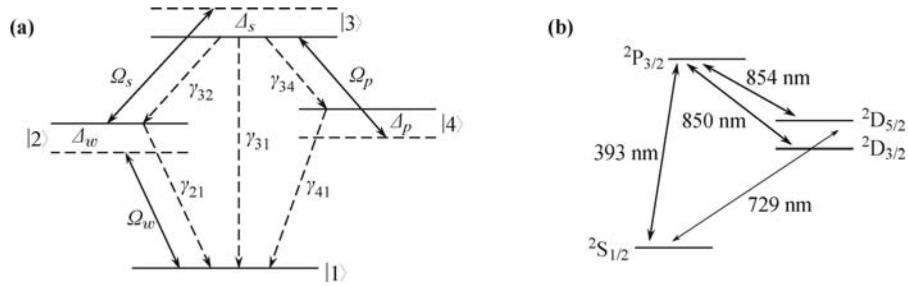

Fig. 1 **(a)** Energy level structure and optical couplings of the four-level system. **(b)** Specific case of the Ca^+ as an example of the scheme.

four-level atomic system coupled to the three light fields can be described by the following density matrix equation:

$$\frac{\mathrm{d}\rho(t)}{\mathrm{d}t} = -\frac{\mathrm{i}}{\hbar}[H,\rho] + L[\rho] \tag{1}$$

where $\rho(t)$ is the atomic density matrix. The sum of the diagonal elements satisfies the probability normalization, i.e., $\rho_{11} + \rho_{22} + \rho_{33} + \rho_{44} = 1$. And the matrix for the system Hamiltonian is defined by

$$H = \begin{pmatrix} 0 & -\frac{\Omega_w}{2} & 0 & 0 \\ -\frac{\Omega_w^*}{2} & -\Delta_w & -\frac{\Omega_s}{2} & 0 \\ 0 & -\frac{\Omega_s^*}{2} & -\Delta_s - \Delta_w & -\frac{\Omega_p}{2} \\ 0 & 0 & -\frac{\Omega_p^*}{2} & \Delta_p - \Delta_s - \Delta_w \end{pmatrix} \tag{2}$$

The Liouvillian matrix $L[\rho(t)]$, defined in Eq. (3), describes relaxation by spontaneous decay.

$$L[\rho] = \begin{pmatrix} \gamma_{31}\rho_{33} + \gamma_{41}\rho_{44} + \gamma_{21}\rho_{22} & -\frac{\gamma_{21}}{2}\rho_{12} & -\frac{\gamma}{2}\rho_{13} & -\frac{\gamma_{41}}{2}\rho_{14} \\ -\frac{\gamma_{21}}{2}\rho_{21} & \gamma_{32}\rho_{33} - \gamma_{21}\rho_{22} & -\frac{\gamma+\gamma_{21}}{2}\rho_{23} & -\frac{\gamma_{21}+\gamma_{41}}{2}\rho_{24} \\ -\frac{\gamma}{2}\rho_{31} & -\frac{\gamma+\gamma_{21}}{2}\rho_{32} & -\gamma\rho_{33} & -\frac{\gamma+\gamma_{41}}{2}\rho_{34} \\ -\frac{\gamma_{41}}{2}\rho_{41} & -\frac{\gamma_{21}+\gamma_{41}}{2}\rho_{42} & -\frac{\gamma+\gamma_{41}}{2}\rho_{43} & \gamma_{34}\rho_{33} - \gamma_{41}\rho_{44} \end{pmatrix} \tag{3}$$

In the following, we are interested in the three-photon transition process, where the quantum coherent effects arise from the transitions between $|1\rangle$ and $|4\rangle$ through two intermediate levels $|2\rangle$ and $|3\rangle$. The corresponding atomic levels in Ca^+ are $^2S_{1/2}$, $^2P_{3/2}$, $^2D_{3/2}$, and $^2D_{5/2}$. Terahertz signal can be derived if state $|1\rangle$, $|2\rangle$, $|3\rangle$ and $|4\rangle$ are set according to Champenois's proposal [9].

Here we choose Ca^+ optical clock system to demonstrate the effects. State $|1\rangle$, $|2\rangle$, $|3\rangle$ and $|4\rangle$ are set to be $^2S_{1/2}$, $^2D_{3/2}$, $^2P_{3/2}$, and $^2D_{5/2}$ states, respectively. The decays of the metastable states in alkaline earth ions can not be ignored, which is different from neutral Sr [7] and Yb [8] atomic systems. Since in $^{40}Ca^+$ system, the $^2D_{3/2}$ state and the $^2D_{5/2}$ state have almost the same lifetime, we use γ_1 to represent the decays of the two states, which is about 1 Hz [16]. And the total decay rate γ is about 140 MHz for $^{40}Ca^+$ [17, 18].

Since Eq. (1) is not easy to solve in general cases, we restrict the conditions to Ω_p, Ω_w, Δ_s, $\Delta_w \ll \gamma$ and $\Omega_s > \gamma$, and study the coherent effects. Considering the system in steady state ($\mathrm{d}\rho/\mathrm{d}t = 0$) and retaining the probe field and weak field only to the lowest order in Ω_p^2 and Ω_w^2, we obtain the dark linewidth (Full Width of Half Maximum) closed to the sharp absorption peak (which means that the three-photon detuning $\Delta = \Delta_p - \Delta_s - \Delta_w$ is very small, i.e. $\Delta \ll \gamma$) as

$$FWHM = \sqrt{\gamma_1^2 + \frac{\Omega_w^2}{\Omega_s^2}M} \tag{4}$$

with

$$M = \frac{2(\gamma-\gamma_{32})\Omega_s^2(\gamma\gamma_1^2\Omega_s^2 + \gamma_1\Omega_p^2\Omega_s^2 + \gamma\Omega_p^2\Omega_w^2) + \gamma^3\gamma_{34}\Omega_w^4 + \gamma^2\gamma_1(\gamma-\gamma_{32}+2\gamma_{34})\Omega_s^2\Omega_w^2}{(\gamma-\gamma_{32})\gamma_1\Omega_s^4 + \gamma\gamma_{34}\Omega_w^2\Omega_s^2}$$

According to Eq. (4), the dark linewidth can be tuned continuously by adjusting Ω_p, Ω_s and Ω_w. For instance, as Ω_s increases, the linewidth narrows down; and when Ω_w and Ω_p increases, the linewidth broadens up. Thus, the system shows rich convenience in controlling coherently the linewidth of the clock transition.

3 Numerical simulations

In order to figure out the roles of the coherent effects acting on the linewidth narrowing and broadening processes, the steady state solution to Eq. (1) is investigated. For simplicity, we are concerned with the population of the $|3\rangle$ state, that is, the upper-level state of the four-state system. The numerical results of $^{40}Ca^+$ system are shown in Fig. 2. In Fig. 2(a), with the strength of the strong coupling field increasing, the linewidth of the absorption peak narrows down. However, the behavior of the absorption peak acts differently from the variation of the probe field, that is, when the strength of the probe field increases, the linewidth of the upper-level absorption peak broadens up, which is clearly shown in Fig. 2(b). And the same case is with the trend of the absorption peak varying with the weak coupling field, as shown in Fig. 2(c). In a word, the system provides a lot of methods of controlling the behavior of the absorption peak. And according to the optical frequency standard, the best results must be a trade-off between a narrower linewidth and a lager signal.

To further investigate the quantum coherence generated in the CPT structure, we will look into the coherent term in the four-level atomic system. As known to all, the real part and the imaginary part of the coherent term stand for the optical dispersion and absorption, respectively. With a study on the properties of the coherent terms of the system, the variation law of the linewidth of the absorption peak will be revealed. In the system, the behaviors of one-photon transition term like ρ_{34}, two-photon resonant transition term ρ_{24} and ρ_{13} indicate the absorption enhancement or restraining of the corresponding driving fields, which is mostly the same as those of the three-level system like the Λ or V types of system. Here we are interested in the behavior of the most important three-photon transition term ρ_{14}.

The three-photon absorption corresponding to the situation with respect to Fig. 2(a) is shown in the following Fig. 3(a). It is evident that the three-photon absorption term determinate the absorption peak of the whole system. And the left two situations are shown in Figs. 3(b) and (c).

The dark linewidth can not be arbitrarily small as the two-photon CPT. The fundamental limit for the linewidth of the coherent dark line is defined by the lifetime of the metastable states, which can be clearly observed in Eq. (4). From the equation, we can easily obtain that the dark line is limited by the decay rate of the metastable states. (Since $M > 0$, it gets FWHM $> \gamma_1$.) And the dark line width is not affected by the detuning of the lasers also observed in Eq. (4), implying that the linewidth is insensitive to the fluctuations of the detuning.

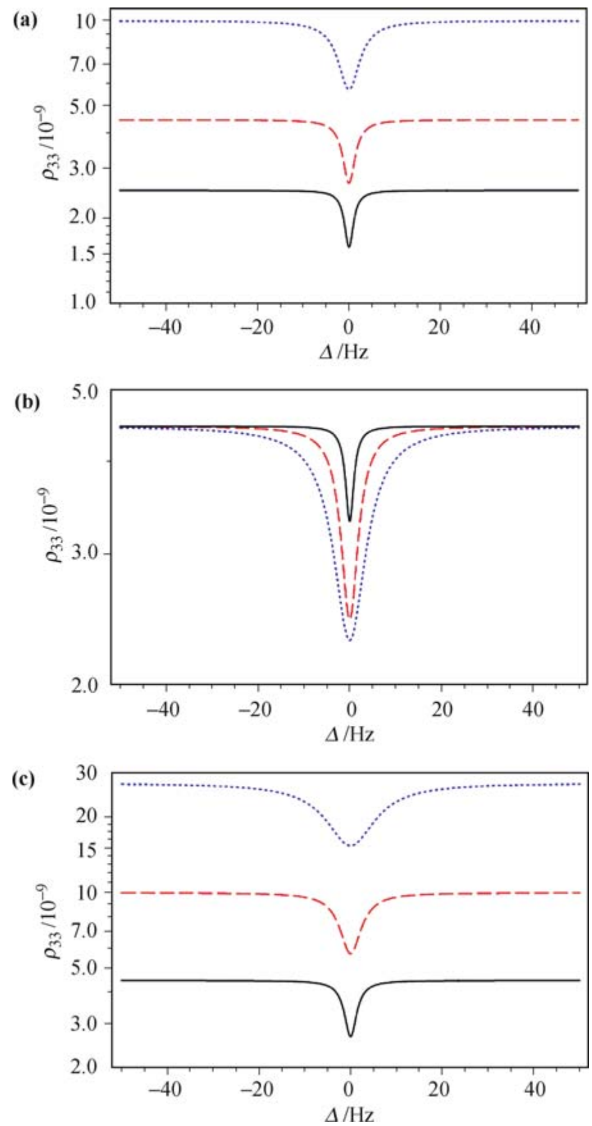

Fig. 2 **(a)** Upper-level population ρ_{33} (absorption peak) versus three-photon detuning Δ under different Ω_s. *Dotted line*: $\Omega_s = \gamma$; *dashed line*: $\Omega_s = 1.5\gamma$; *solid line*: $\Omega_s = 2\gamma$. Other parameters are: $\Omega_w = 10^{-4}\gamma$, $\Omega_p = 2\times10^{-4}\gamma$, $\Delta_s = -10^{-4}\gamma$, $\Delta_w = 3\times10^{-4}\gamma$. **(b)** Upper-level population ρ_{33} versus three-photon detuning Δ under different Ω_p. *Dotted line*: $\Omega_p = 6\times10^{-4}\gamma$; *dashed line*: $\Omega_p = 3\times10^{-4}\gamma$; *solid line*: $\Omega_p = 10^{-4}\gamma$. Other parameters are: $\Omega_s = 1.5\gamma$, $\Omega_w = 10^{-4}\gamma$, $\Delta_s = -10^{-4}\gamma$, $\Delta_w = 3\times10^{-4}\gamma$. **(c)** Upper-level population ρ_{33} versus three-photon detuning Δ under different Ω_w. *Dotted line*: $\Omega_w = 2.5\times10^{-4}\gamma$; *dashed line*: $\Omega_w = 1.5\times10^{-4}\gamma$; *solid line*: $\Omega_w = 10^{-4}\gamma$. Other parameters are: $\Omega_s = 1.5\gamma$, $\Omega_p = 2\times10^{-4}\gamma$, $\Delta_s = -10^{-4}\gamma$, $\Delta_w = 3\times10^{-4}\gamma$.

In the scheme of the three-photon resonance, the Doppler-free configuration is defined by the phase matching condition [10]:

$$\Delta k = \Delta k_s + \Delta k_w - \Delta k_p = 0$$

This can be done through accurate control of the angles of the laser beams.

4 Conclusion

In conclusion, we have investigated the dark line of a

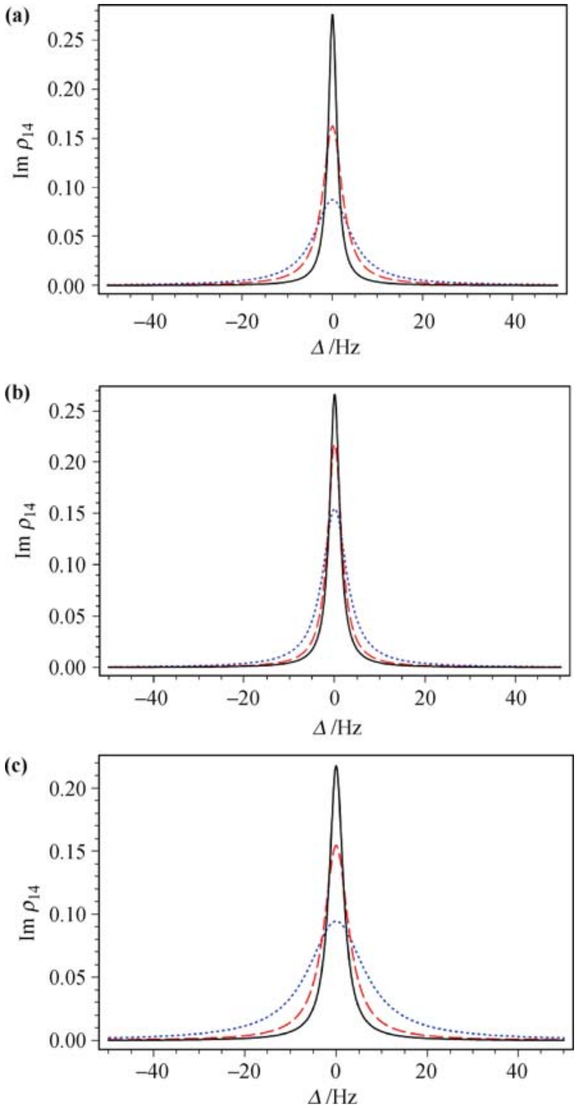

Fig. 3 **(a)** The three-photon transition term (Im ρ_{14}) varies with three-photon detuning with respect to Fig. 2(a). **(b)** The three-photon transition term (Im ρ_{14}) varies with three-photon detuning with respect to Fig. 2(b). **(c)** The three-photon transition term (Im ρ_{14}) varies with three-photon detuning with respect to Fig. 2(c).

four-level atomic system interacting with three laser fields. Our results show that the linewidth of the spectral line can be very narrow, and can be controlled by adjusting the parameters of the lasers, but still limited by the decay rate of the metastable states. And the relationship of the dark line and laser intensities has been studied.

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