

## LETTERS

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# Ramsey-comb spectroscopy with intense ultrashort laser pulses

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# Outline

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- Introduction
- Ramsey spectroscopy
- Optical frequency comb spectroscopy
- Ramsey-comb spectroscopy
- Experimental demonstration of Ramsey-comb spectroscopy
- Conclusion



# Introduction

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Optical frequency combs based on mode-locked lasers have revolutionized the field of metrology and precision spectroscopy by providing precisely calibrated optical frequencies and coherent pulse trains. Amplification of the pulsed output from these lasers is very desirable, as nonlinear processes can then be used to cover a much wider range of transitions and wavelengths for ultra-high precision, direct frequency comb spectroscopy. Therefore full repetition rate laser amplifiers and enhancement resonators have been employed to produce up to microjoule-level pulse energies. **Here we present a spectroscopic method to obtain frequency comb accuracy and resolution by using only two frequency comb pulses amplified to the millijoule pulse energy level**, orders of magnitude more energetic than what has previously been possible. The new properties of this approach, such as cancellation of optical light-shift effects, are demonstrated on weak two-photon transitions in atomic rubidium and caesium, thereby improving the frequency accuracy by up to thirty times.



# Introduction

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One of the outstanding challenges in laser spectroscopy is the precise measurement of transitions in the vacuum ultraviolet (VUV:  $\lambda \sim 10\text{-}200\text{nm}$ ) region.

(e.g. atomic & molecular energy structure of small elements such as hydrogen, helium and antihydrogen, antiprotonic helium)

However, there is **no widely tunable narrow-linewidth lasers below about 180nm** that would be suitable for precision spectroscopy



# Introduction



N. Ramsey Jr.

**Ramsey spectroscopy**  
(*Nobel Prize 1989*)

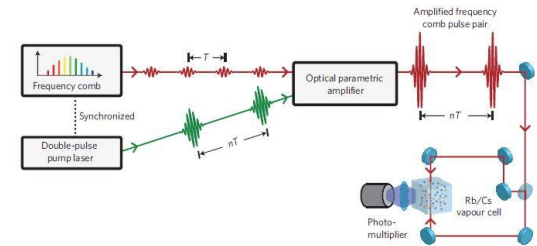
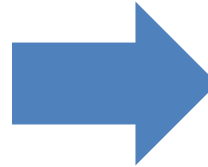


J. Hall



T. Hänsch

**Optical frequency comb**  
(*Nobel Prize 2005*)



**Ramsey-comb spectroscopy**

Basically, **Ramsey-comb spectroscopy** combines two spectroscopic methods to solve this problem, both of them are part of the Nobel Prize in Physics.



# Ramsey Spectroscopy

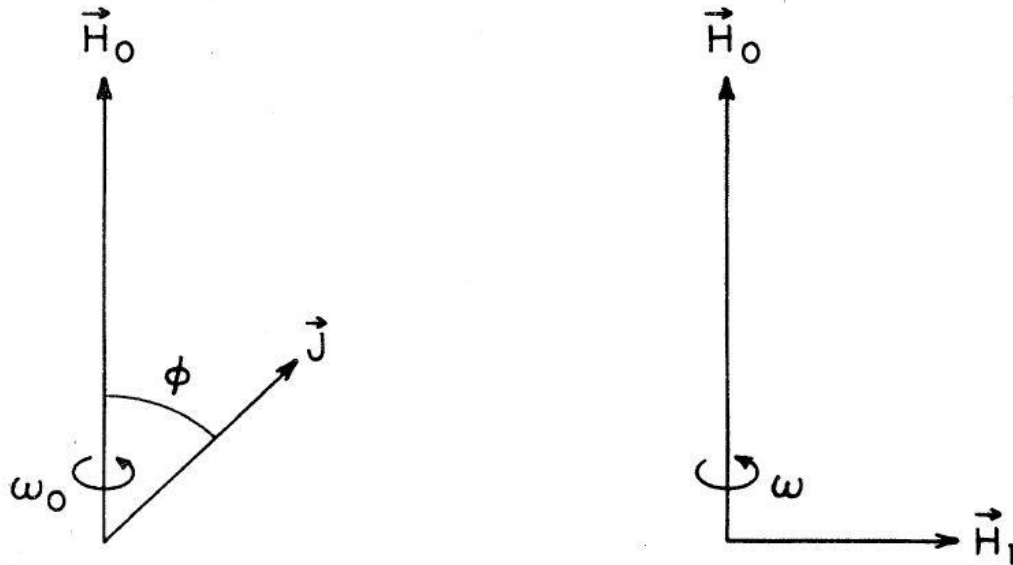


FIG. 3. Precession of the nuclear angular momentum  $\mathbf{J}$  (left) and the rotating magnetic field  $\mathbf{H}_1$  (right) in the Rabi method.

N. Ramsey, *Rev. Mod. Phys.* 62, 3 (1990)

If we consider the nucleus with angular momentum  $J$ , its Larmor frequency:

$$\omega_0 = 2\pi\nu_0 = \frac{\mu H_0}{\hbar I}$$

can be detected by measuring the oscillator frequency  $\omega$  at which there is maximum reorientation of the angular momentum and hence a maximum change in beam intensity.



# Ramsey Spectroscopy

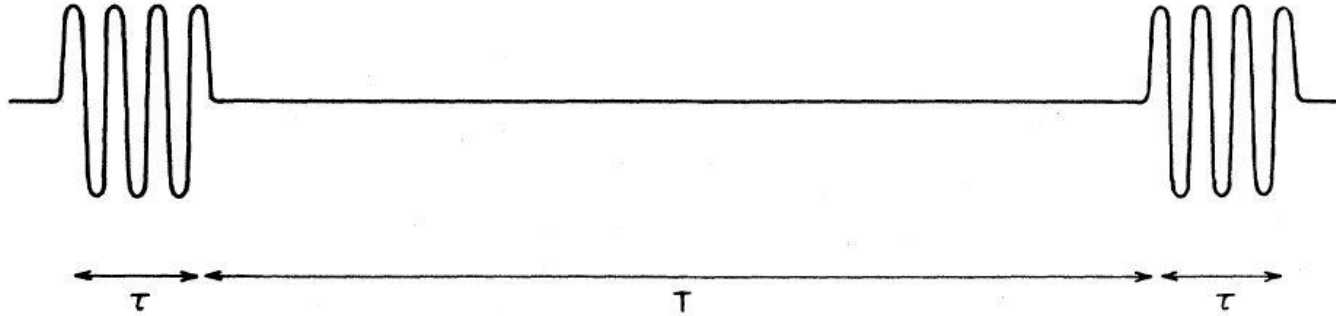


FIG. 4. Two separated oscillatory fields, each acting for a time  $\tau$ , with zero-amplitude oscillating field acting for time  $T$ . Phase coherency is preserved between the two oscillatory fields, so it is as if the oscillation continued, but with zero amplitude for time  $T$ .

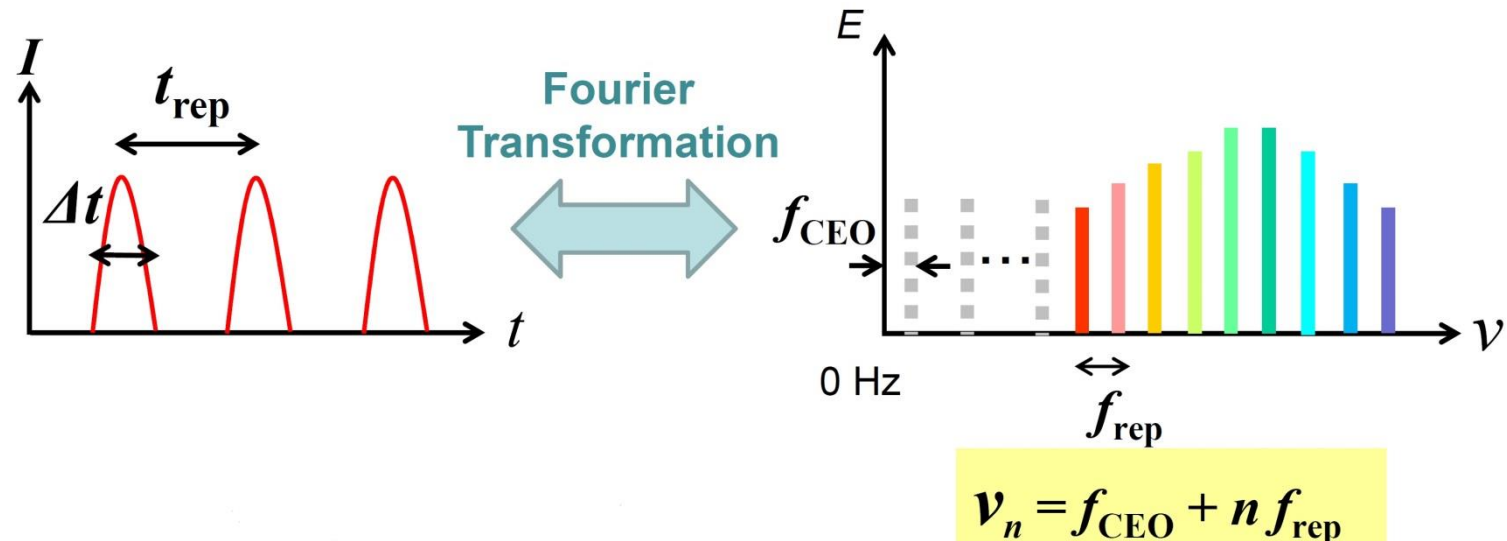
N. Ramsey, *Rev. Mod. Phys.* 62, 3 (1990)

Practically, Ramsey spectroscopy uses two separated oscillating fields and can be applied to any quantum-mechanical system. In this way, atomic resonance

frequency  $\omega_0 = \frac{W_i - W_f}{\hbar}$  can be determined.



# Optical Frequency Comb Spectroscopy

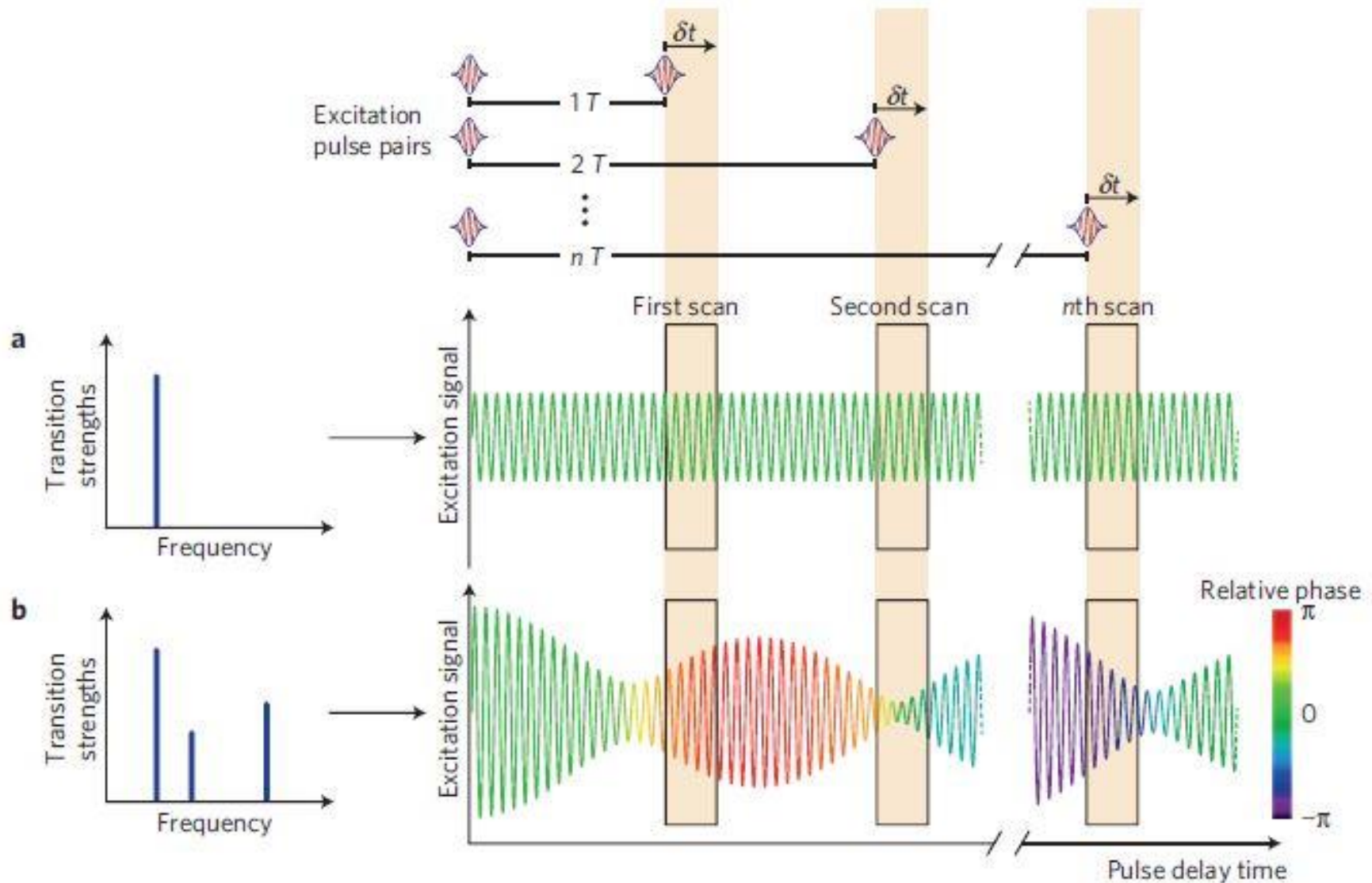


<http://www.phys.keio.ac.jp/guidance/labs/sasada/research/fibercomb-en.html>

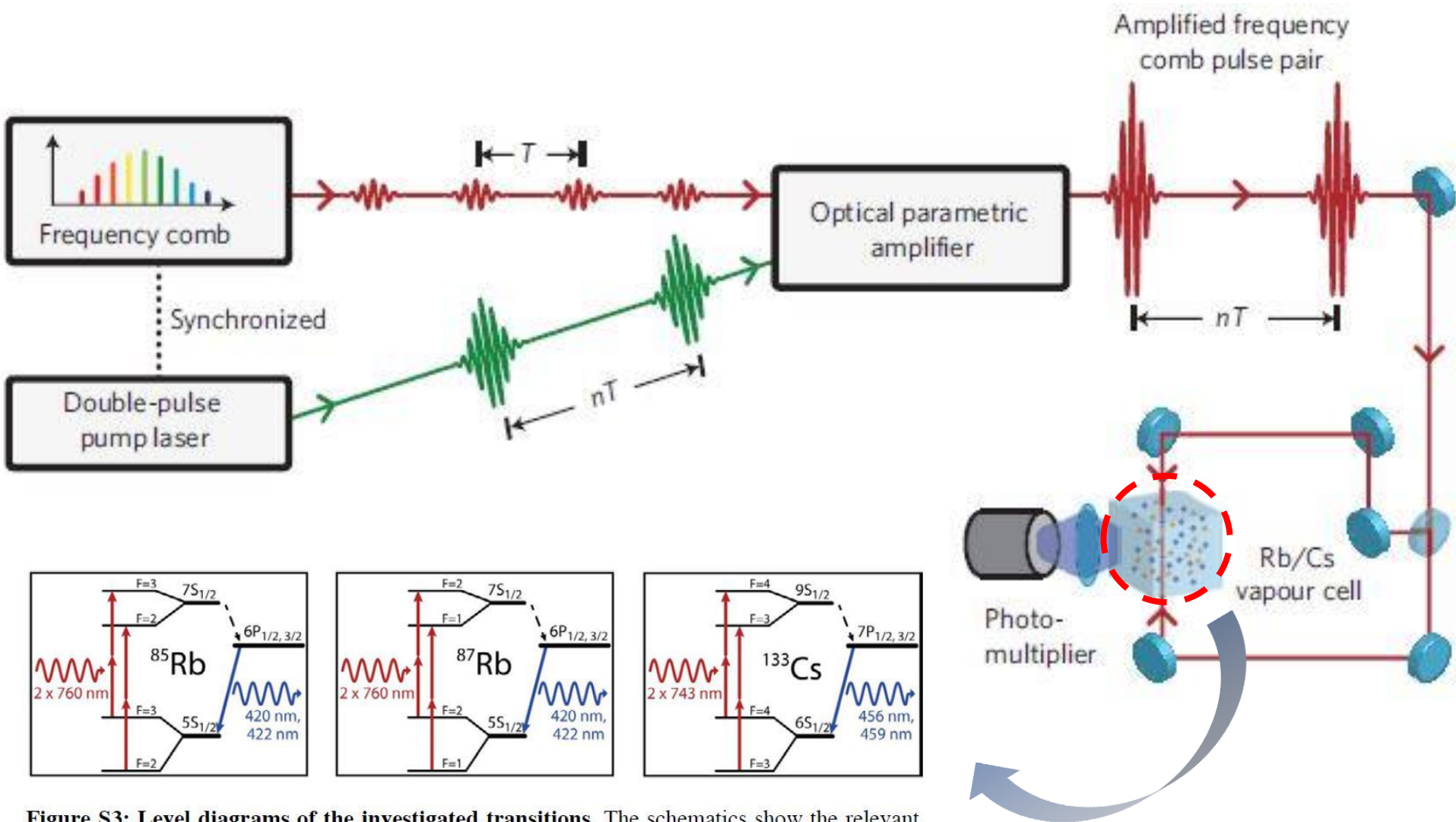
Optical frequency comb technique was employed to improve the precision.



# Ramsey-comb spectroscopy



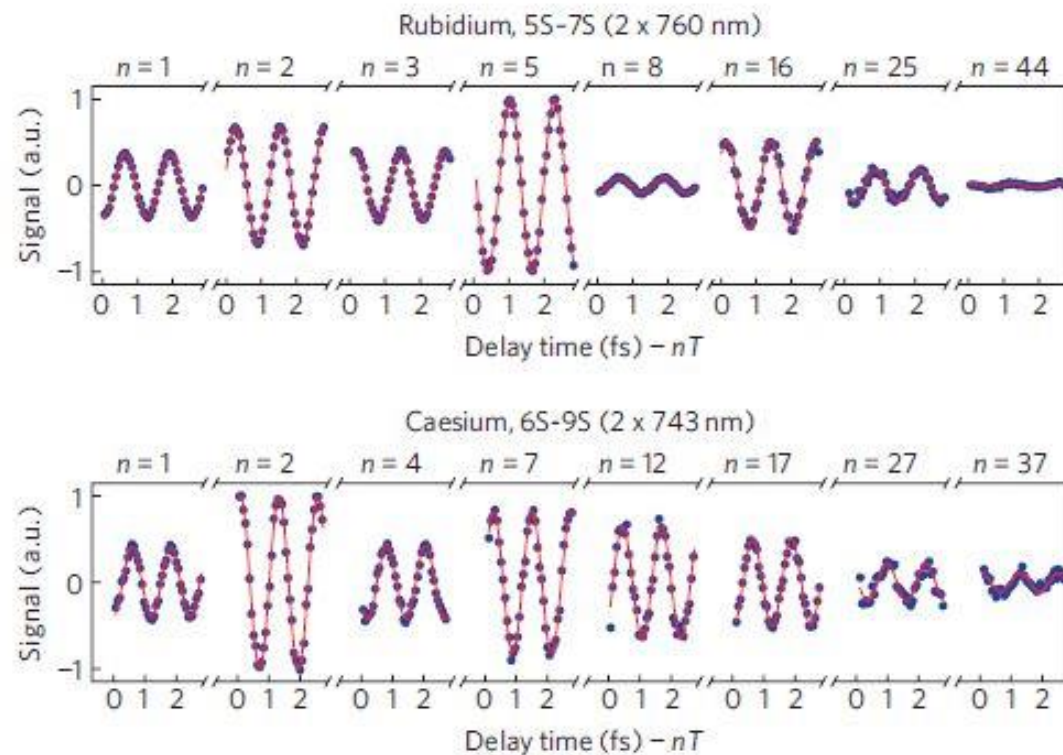
# Ramsey-comb spectroscopy: setup



**Figure S3: Level diagrams of the investigated transitions.** The schematics show the relevant levels for the two rubidium isotopes ( $^{85}\text{Rb}$  and  $^{87}\text{Rb}$ ) and caesium (only one isotope,  $^{133}\text{Cs}$ ). Indicated are the excitation paths in red and the fluorescence light used for detection in blue.

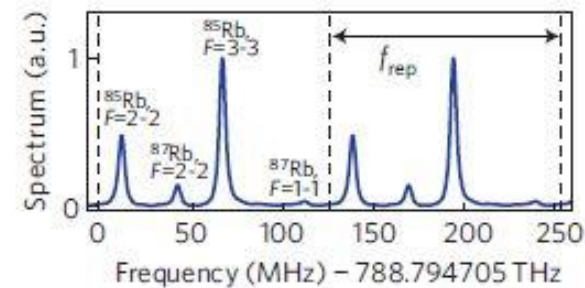
# Experimental result

a

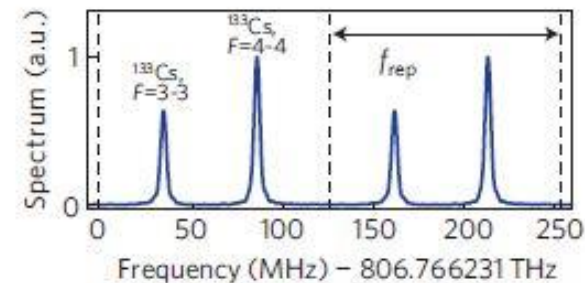


$|DFT|^2$   
(44 scans)

b



$|DFT|^2$   
(37 scans)



# Conclusion

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- Atomic transitions in vacuum ultraviolet (VUV:  $\lambda \sim 10\text{-}200\text{nm}$ ) region were detected by new spectroscopic method combining Ramsey spectroscopy and frequency comb technique, namely Ramsey-comb spectroscopy.
- Transition frequencies of  $^{85}\text{Rb}$ ,  $^{87}\text{Rb}$ ,  $^{133}\text{Cs}$  were studied with Ramsey-comb spectroscopy.

