LETTERS

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Ramsey-comb spectroscopy with intense ultrashort laser pulses

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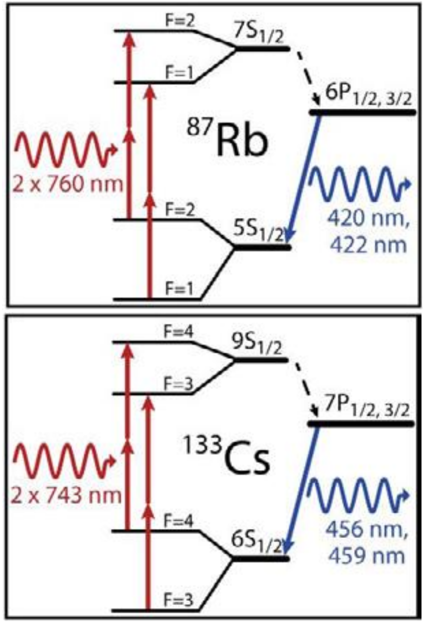
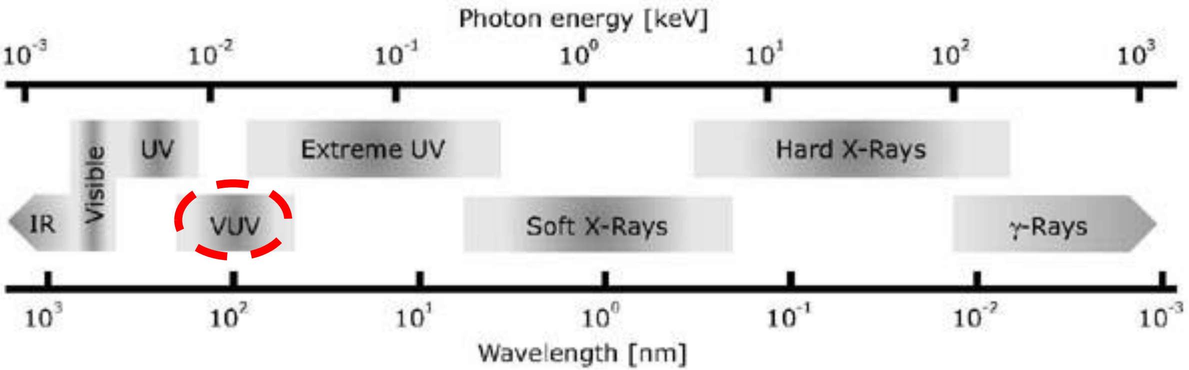
Abstract – much more precise measurement of atomic transition (Rb, Cs)

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[1,2]. 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[3,4]. Therefore full repetition rate laser amplifiers[5,6] and enhancement resonators[7,8] have been employed to produce up to microjoule-level pulse energies[9]. 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 - precision spectroscopy at VUV region



http://www.biox.kth.se/research/softxraysources/sxrproject.htm

One of the outstanding challenges in laser spectroscopy is the precise measurement of transitions in the vacuum ultraviolet (VUV: λ~10-200nm) 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 - A combination of two Nobel ideas

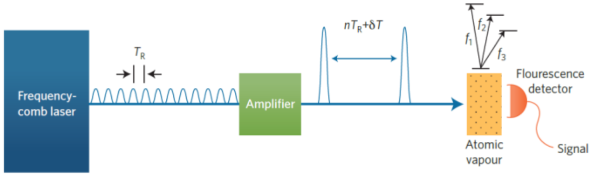
N. Ramsey Jr.

Ramsey spectroscopy
(Nobel Prize 1989)

J. Hall

T. Hänsch

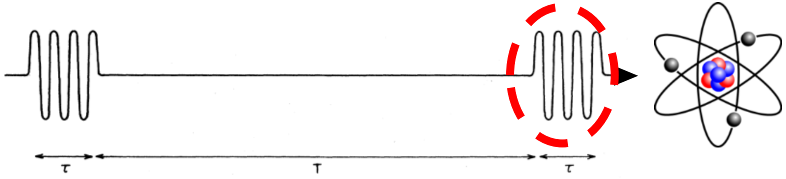
Optical frequency comb
(Nobel Prize 2005)



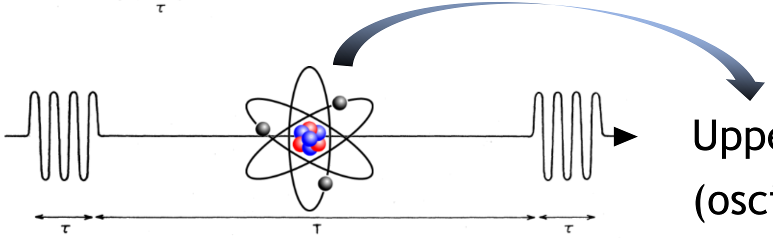
Ramsey-comb spectroscopy

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 in atomic physics

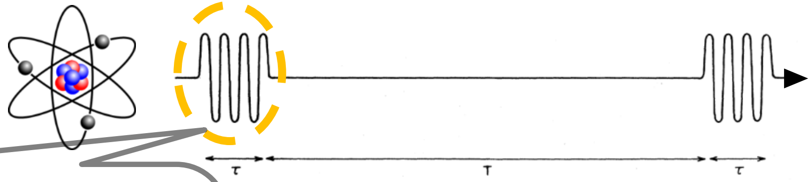


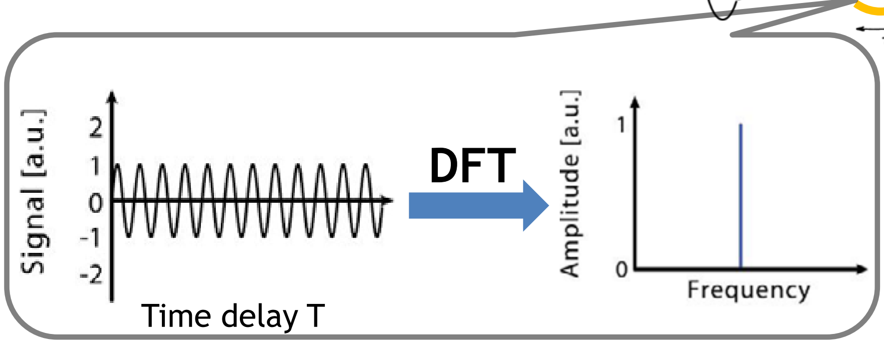
First pulse excites the atom.



Upper state pop. ~ $\boldsymbol{\cos(2\pi f_i t + \Delta\varphi)}$

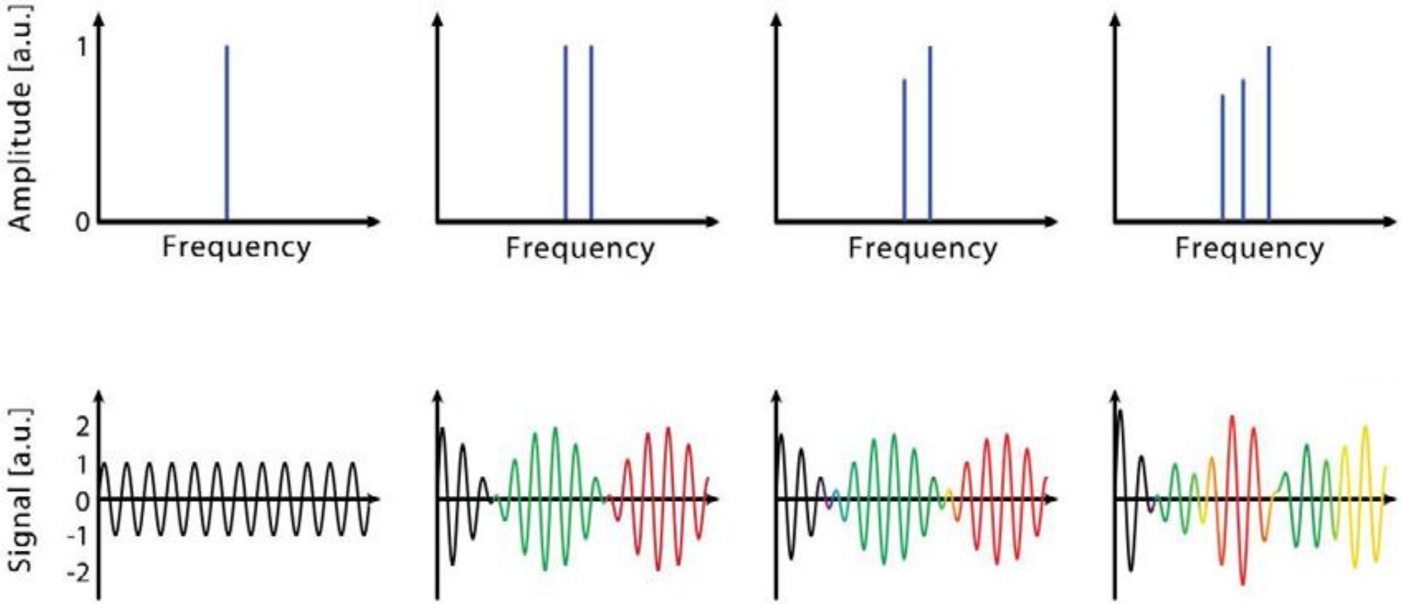
(oscillates with frequency $\omega_0 = \frac{W_i - W_f}{\hbar}$)





Second pulse detects the energy state of atom.

Ramsey spectroscopy – what if transition mode is not unique?

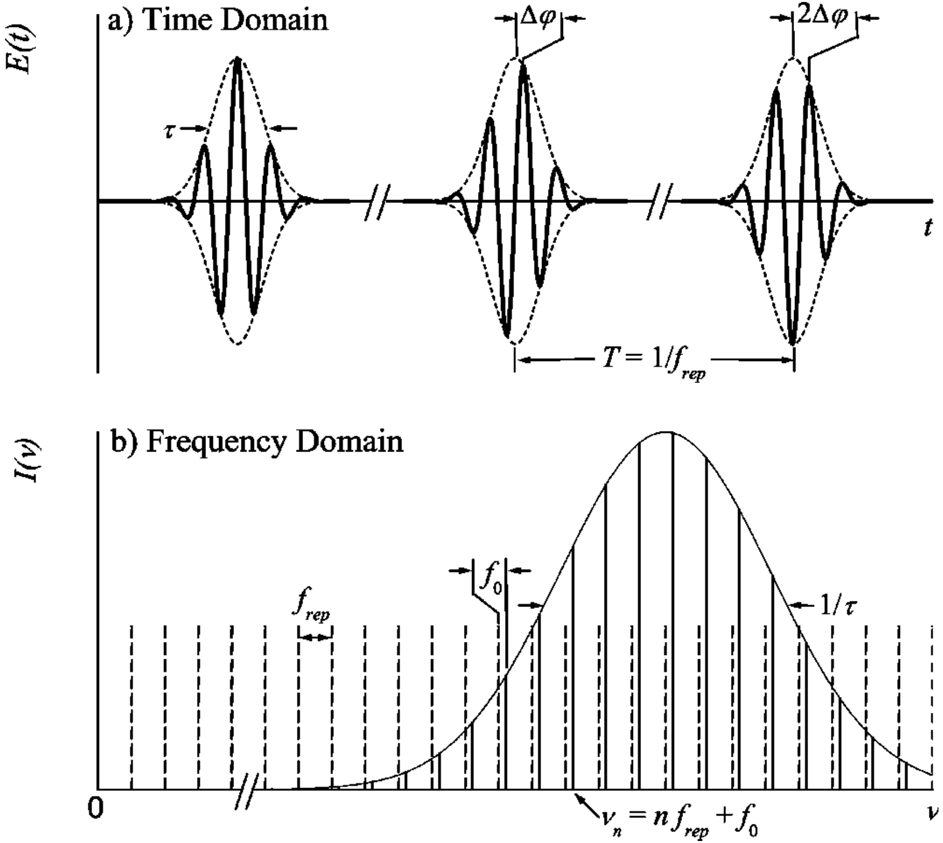


$$S = A_i \cos(2\pi f_i t + \Delta\varphi) \quad \Rightarrow \quad S = \sum_i A_i \cos(2\pi f_i t + \Delta\varphi)$$

Excitation signal is much more complex if more than one transition modes are involved.

-> Hard to measure precise amplitude A_i and frequency ω_i simultaneously ($\Delta E \cdot \Delta t \geq \frac{h}{2}$)

Optical frequency comb

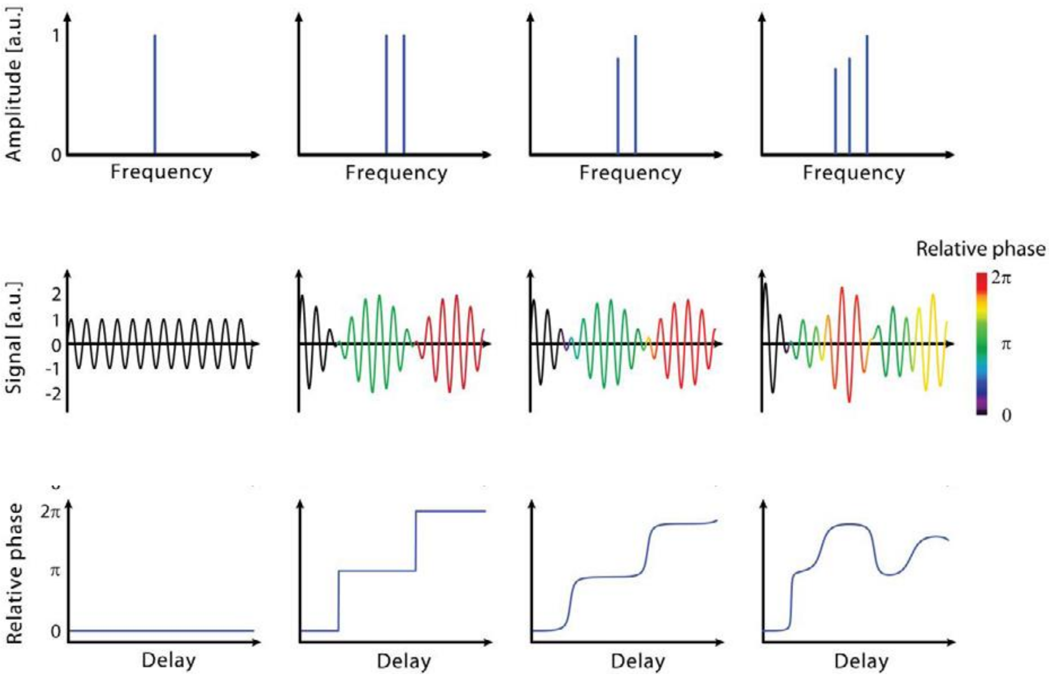


S. Cundiff & J. Ye, Rev. Mod. Phys., 75, 1. (2003)

A **frequency comb** is a light source whose spectrum consists of a series of discrete, equally spaced elements.

In frequency comb, phase relation between different modes is precisely defined.

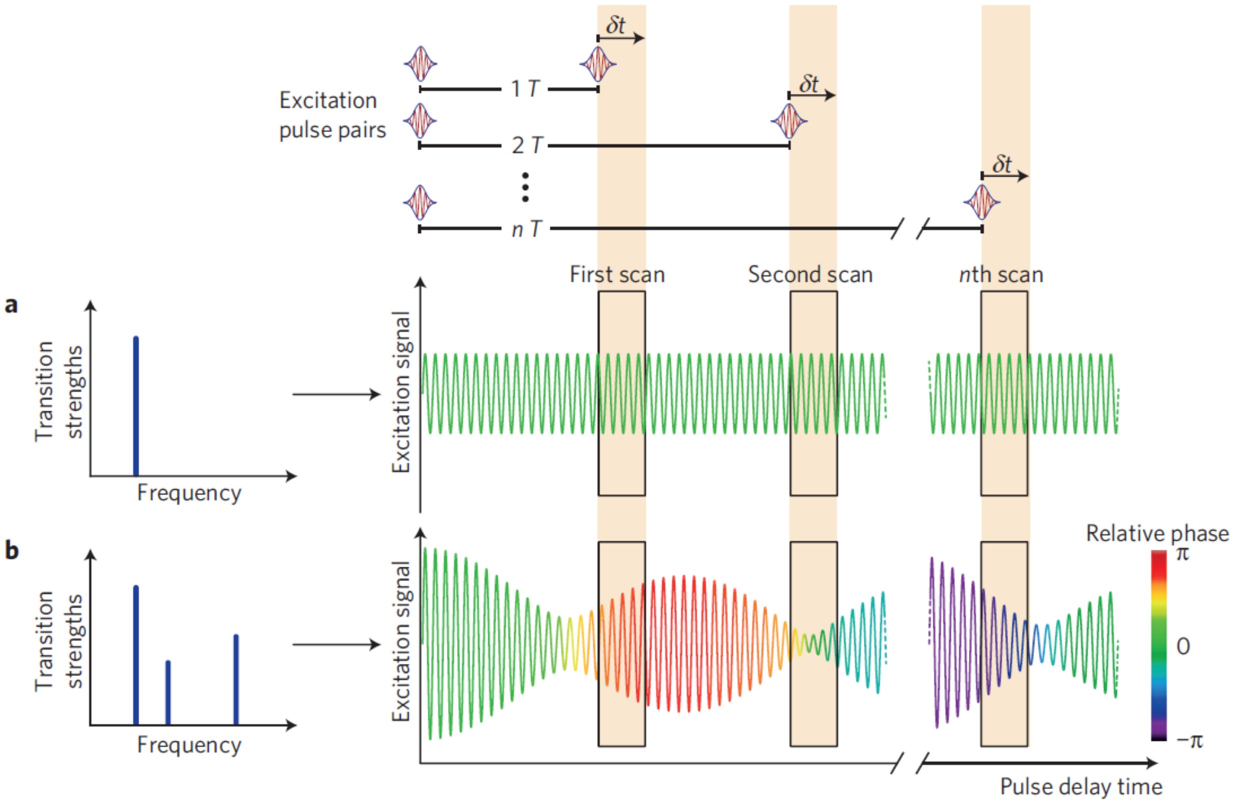
Ramsey spectroscopy + Optical frequency comb = ?



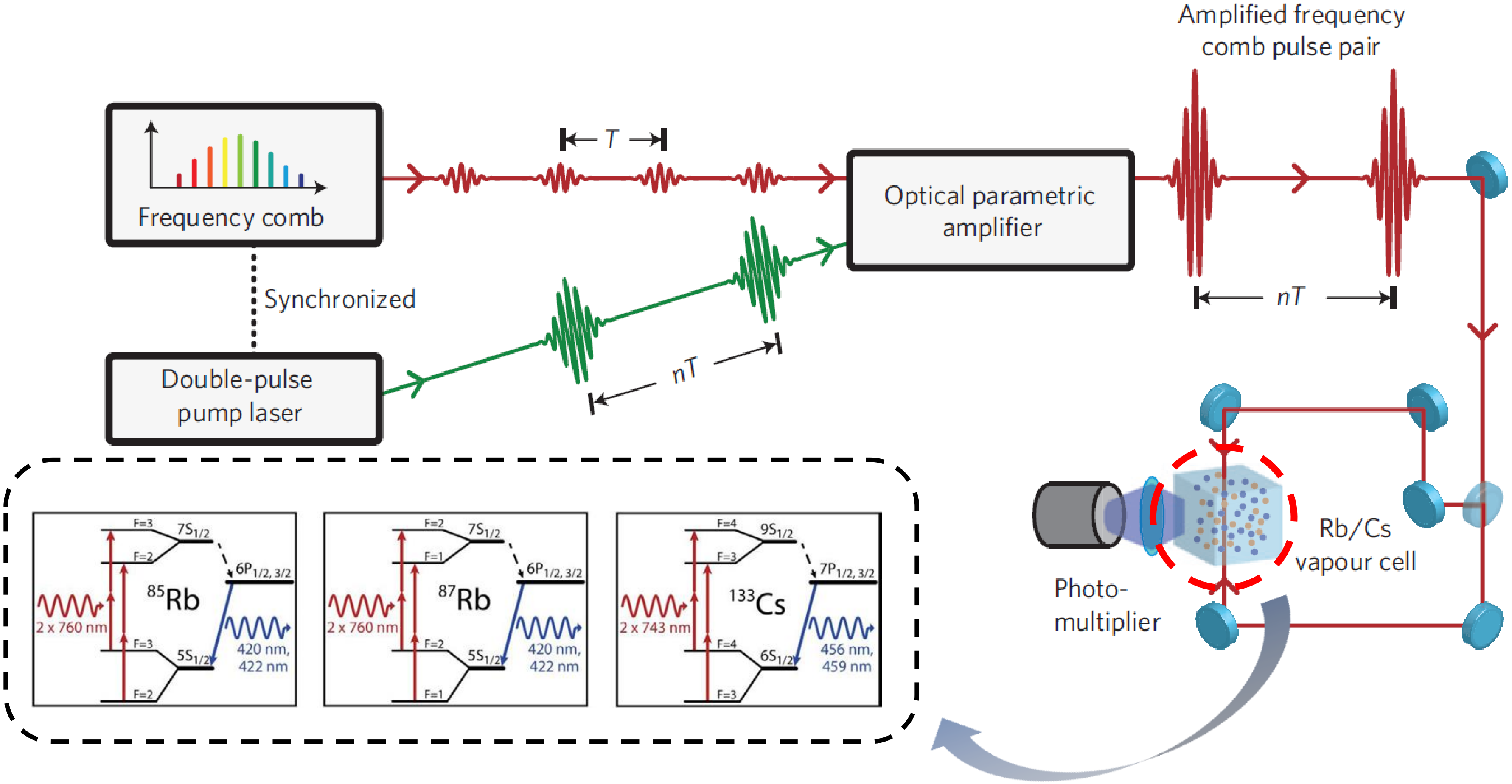
Since full spectral parameters are encoded in phase evolution, one can calculate the spectrum from the phase information, which is experimentally more robust than amplitude.

$$\Phi_{fit}(\Delta t; A_1, A_2, \ldots; f_1, f_2, \ldots; \Delta\phi) = \arg\left\{\sum_i A_i \exp(-i2\pi f_i \Delta t)\right\} + \Delta\phi - 2\pi f_0 \Delta t$$

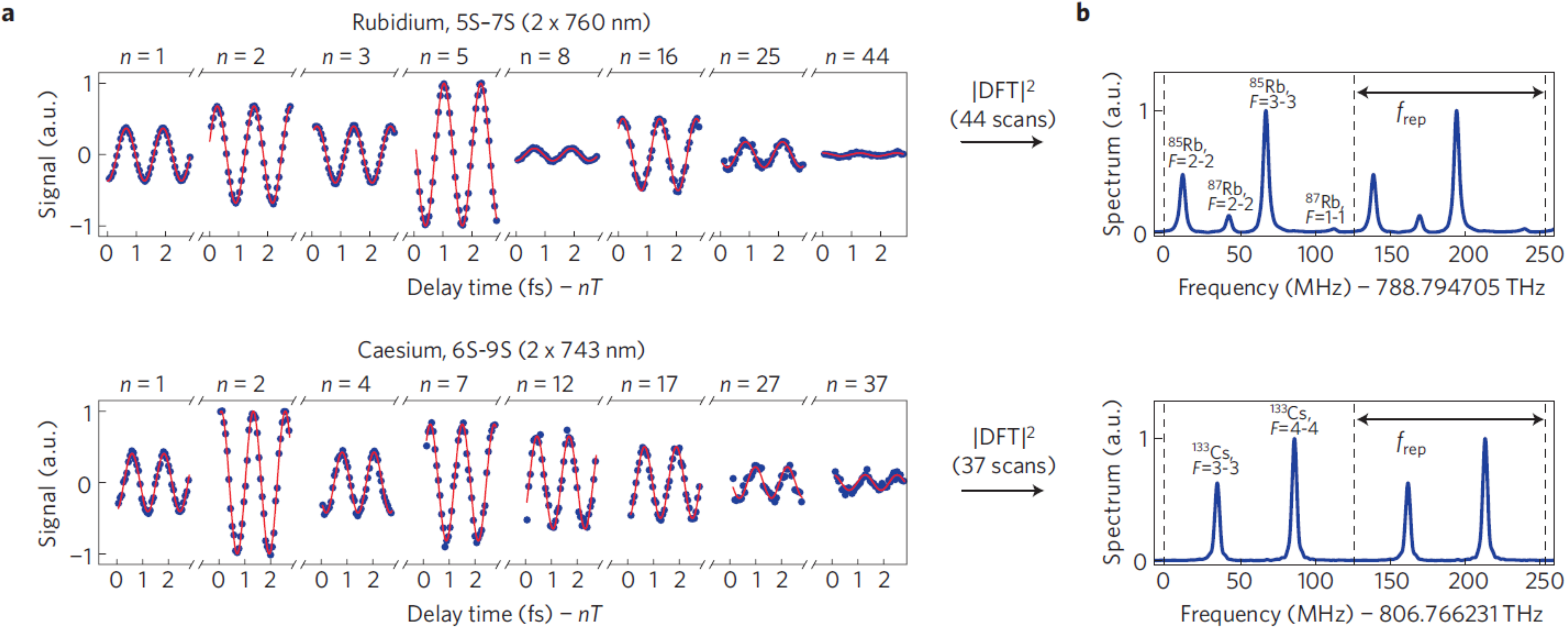
Ramsey-comb spectroscopy : principle



Ramsey-comb spectroscopy : setup



Ramsey-comb spectroscopy : results



Ramsey-comb spectroscopy : results

Frequency description		**Final result [kHz]**
^{85}Rb, $5S_{1/2}$ - $7S_{1/2}$	F=2 - F=2	$788{,}798{,}565{,}751(6)_{stat}(4)_{sys}$
	F=3 - F=3	$788{,}795{,}814{,}071(5)_{stat}(4)_{sys}$
	Centre of gravity	$788{,}796{,}960{,}604(4)_{stat}(4)_{sys}$
	Hyperfine A_{7S}	$94{,}684(2)_{stat}(1)_{sys}$
^{87}Rb, $5S_{1/2}$ - $7S_{1/2}$	F=1 - F=1	$788{,}800{,}964{,}104(9)_{stat}(4)_{sys}$
	F=2 - F=2	$788{,}794{,}768{,}945(7)_{stat}(4)_{sys}$
	Centre of gravity	$788{,}797{,}092{,}129(6)_{stat}(4)_{sys}$
	Hyperfine A_{7S}	$319{,}762(6)_{stat}(1)_{sys}$
	Isotope shift ^{87}Rb - ^{85}Rb	$131{,}525(7)_{stat}(3)_{sys}$
^{133}Cs, $6S_{1/2}$ - $9S_{1/2}$	F=3 - F=3	$806{,}766{,}286{,}786(8)_{stat}(4)_{sys}$
	F=4 - F=4	$806{,}757{,}534{,}152(7)_{stat}(4)_{sys}$
	Centre of gravity	$806{,}761{,}363{,}429(5)_{stat}(4)_{sys}$
	Hyperfine A_{9S}	$109{,}999(3)_{stat}(0)_{sys}$

Conclusion

- Atomic transitions in vacuum ultraviolet (VUV: λ~10-200nm) region were detected by new spectroscopic method combining Ramsey spectroscopy and frequency comb technique, namely Ramsey-comb spectroscopy.
- Much precise transition frequency of Rubidum and Caesium were experimentally determined.