

Nonlinear Dual-Comb Spectroscopy with Two-Photon Excitation

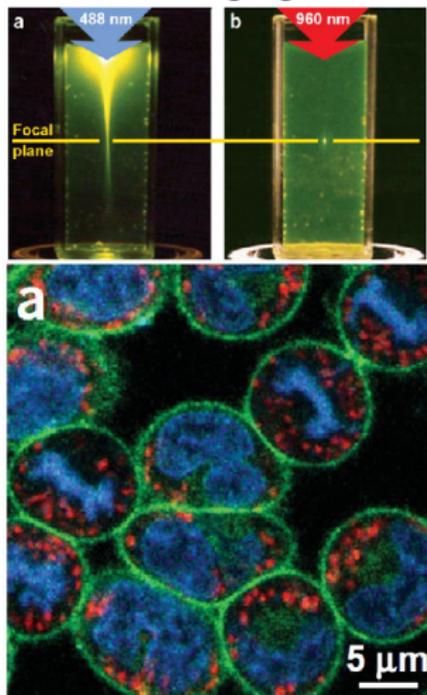
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Theodor W. Hänsch, Nathalie Picqué

Max Planck Institute of Quantum Optics
Garching, Germany

June 17, 2013

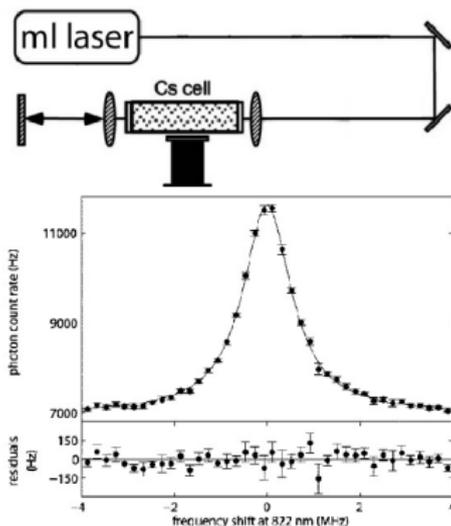
Applications of two-photon spectroscopy

Fluorescence imaging



Zipfel et al., Nature Biotech. 21, 1369–1377 (2003)

Precision spectroscopy

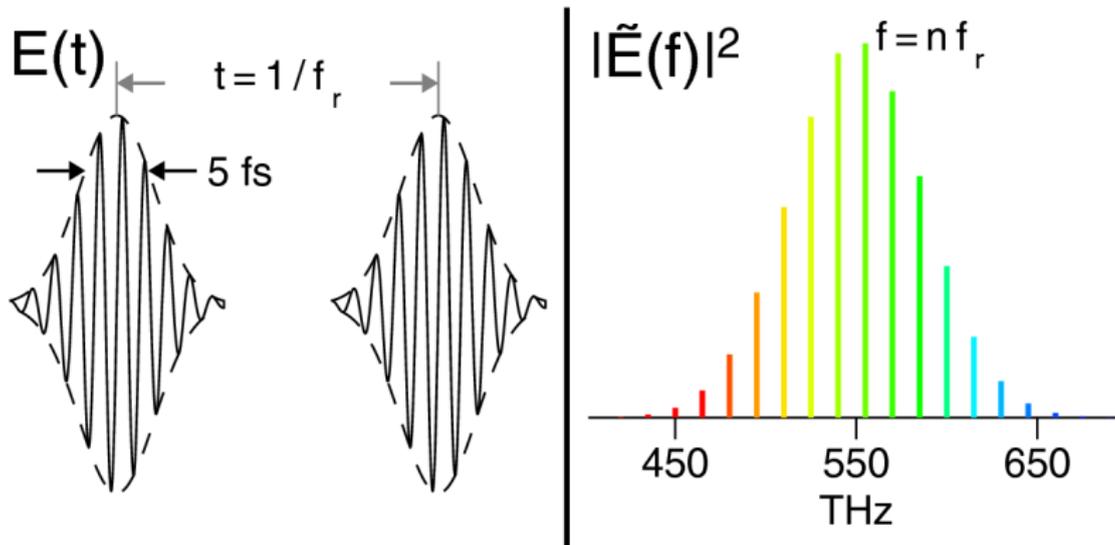


$$\text{Cs } 8s F' = 3 \leftarrow 6s F'' = 3$$

$$\nu = 364\,507\,238\,417(15) \text{ kHz}$$

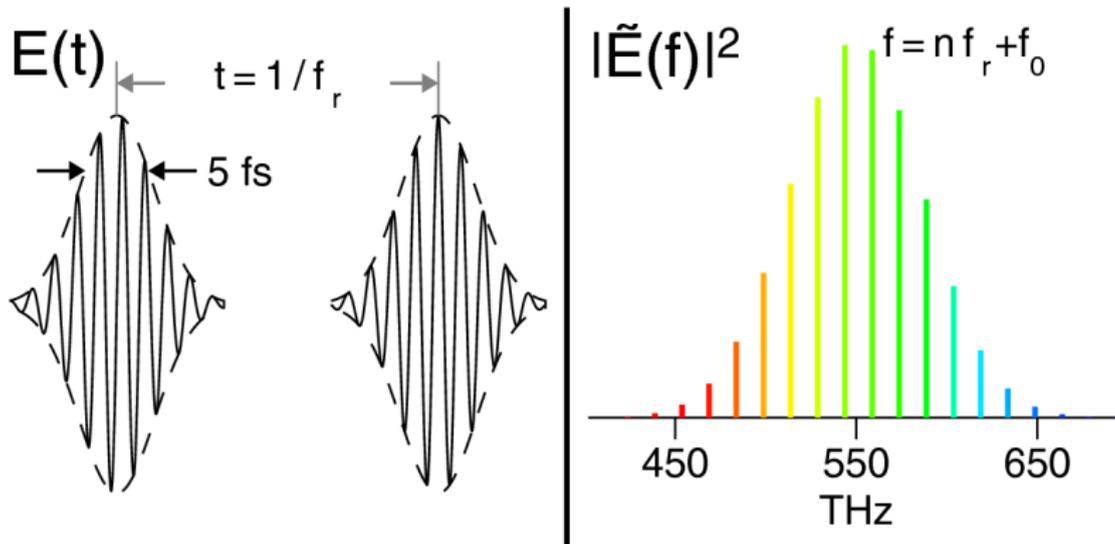
Adapted from Fendel et al., Opt. Lett. 702, 701–703 (2007)

Quick review of frequency combs



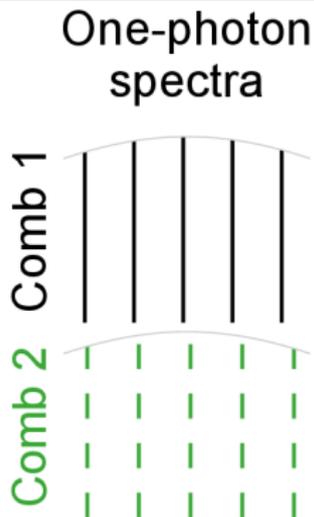
- ▶ Long train of identical pulses \rightarrow many evenly spaced CW lasers

Quick review of frequency combs



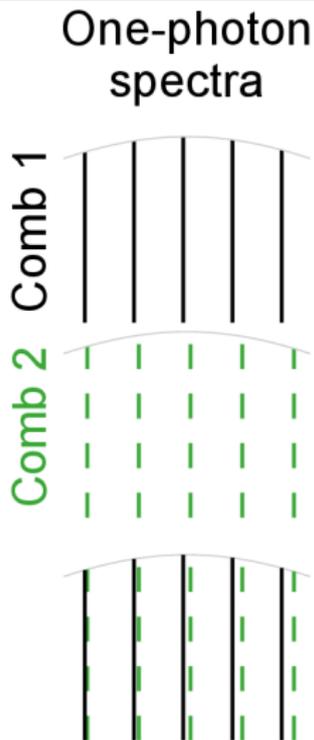
- ▶ Long train of identical pulses \rightarrow many evenly spaced CW lasers
- ▶ Pulse-to-pulse phase offset \rightarrow all comb lines shifted by equal amount

Principle — One-photon dual-comb spectroscopy



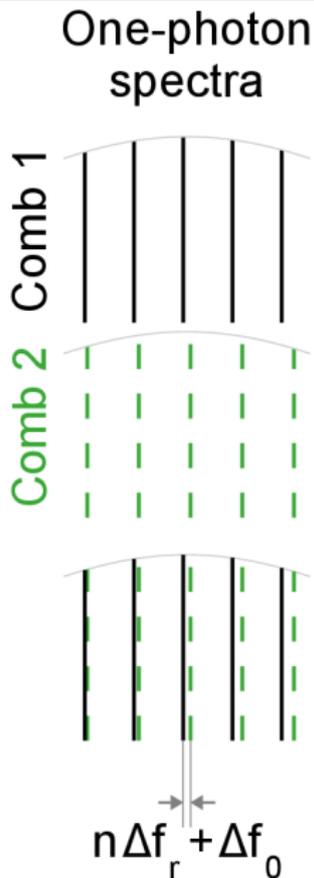
- ▶ Two combs with different repetition rates combined

Principle — One-photon dual-comb spectroscopy



- ▶ Two combs with different repetition rates combined

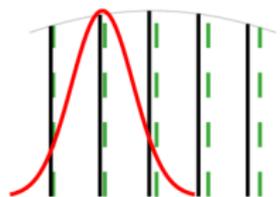
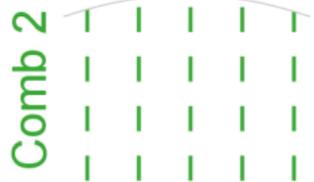
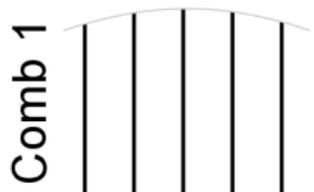
Principle — One-photon dual-comb spectroscopy



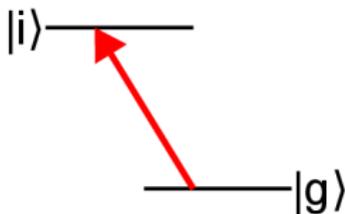
- ▶ Two combs with different repetition rates combined
- ▶ Unique RF beat notes produced at each optical frequency

Principle — One-photon dual-comb spectroscopy

One-photon spectra

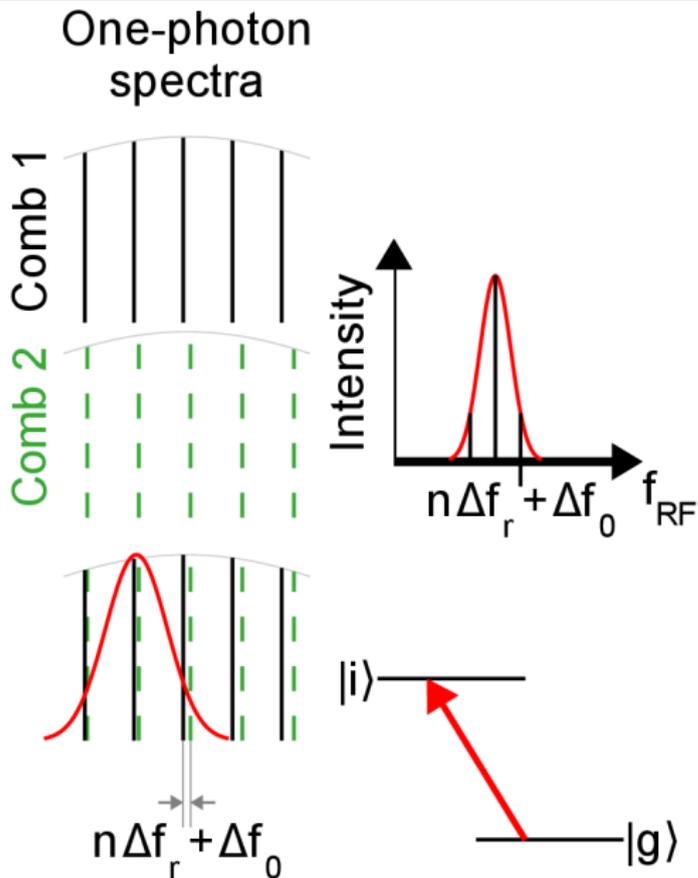


$$n\Delta f_r + \Delta f_0$$



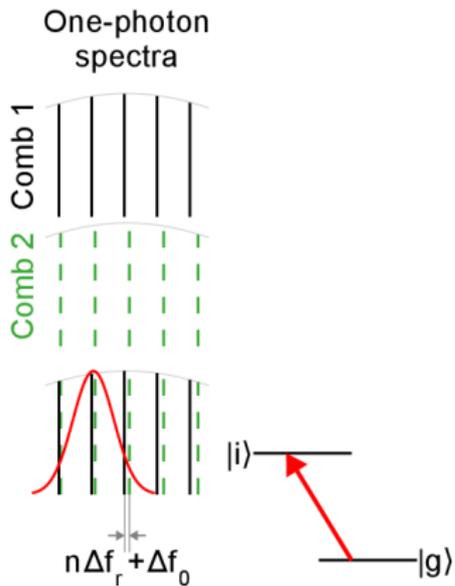
- ▶ Two combs with different repetition rates combined
- ▶ Unique RF beat notes produced at each optical frequency
- ▶ Molecular transitions modulated with RF beat note

Principle — One-photon dual-comb spectroscopy

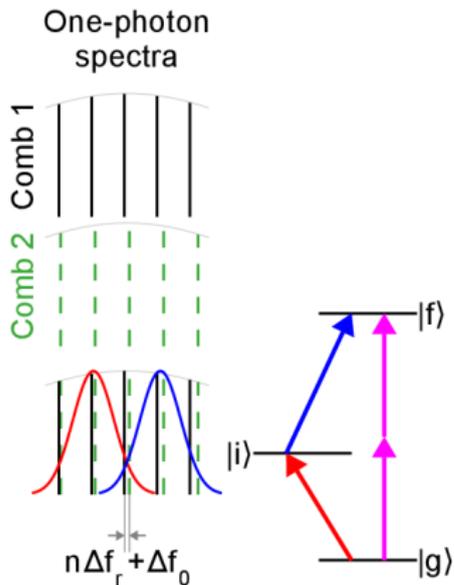


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Principle — Two-photon dual-comb spectroscopy

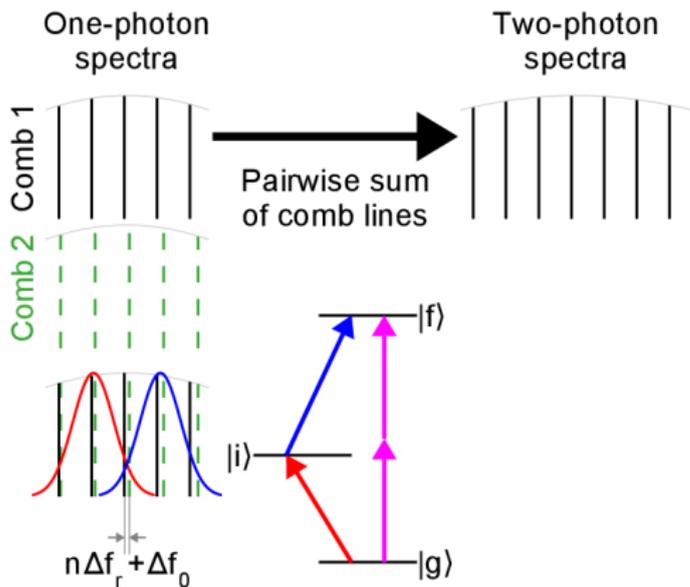


Principle — Two-photon dual-comb spectroscopy



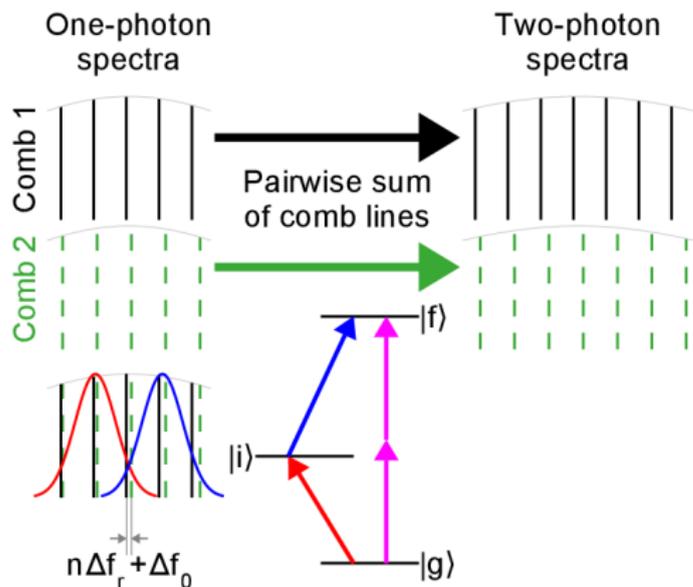
- ▶ Two-photon transitions excited directly or through intermediate state

Principle — Two-photon dual-comb spectroscopy



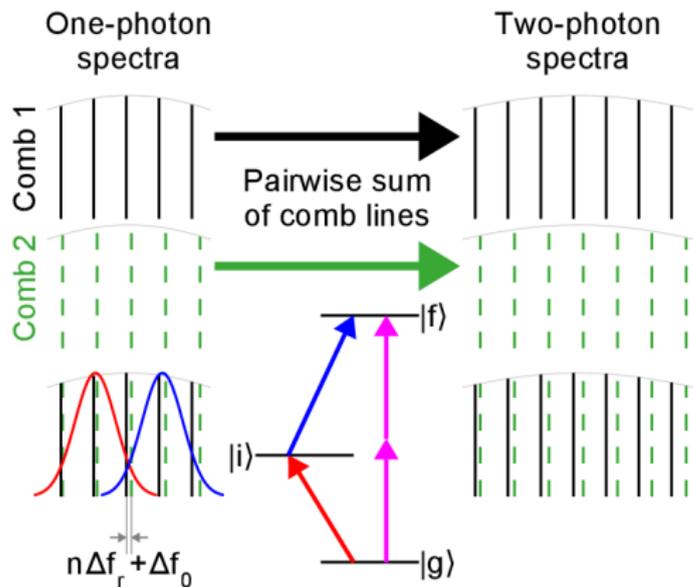
- ▶ Two-photon transitions excited directly or through intermediate state
- ▶ Direct two-photon excitation through pairwise sum of all comb lines

Principle — Two-photon dual-comb spectroscopy



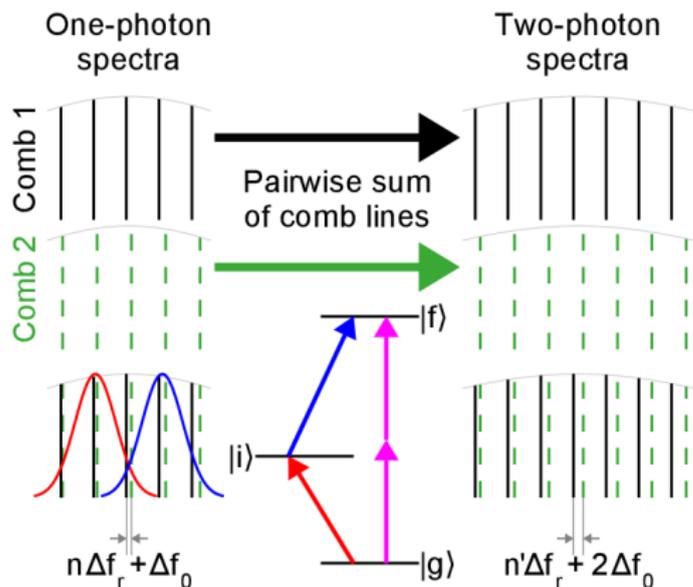
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Principle — Two-photon dual-comb spectroscopy



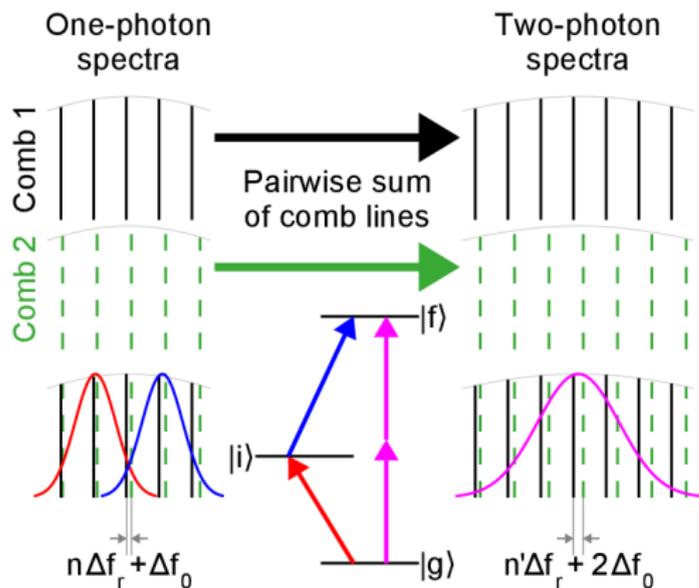
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Principle — Two-photon dual-comb spectroscopy



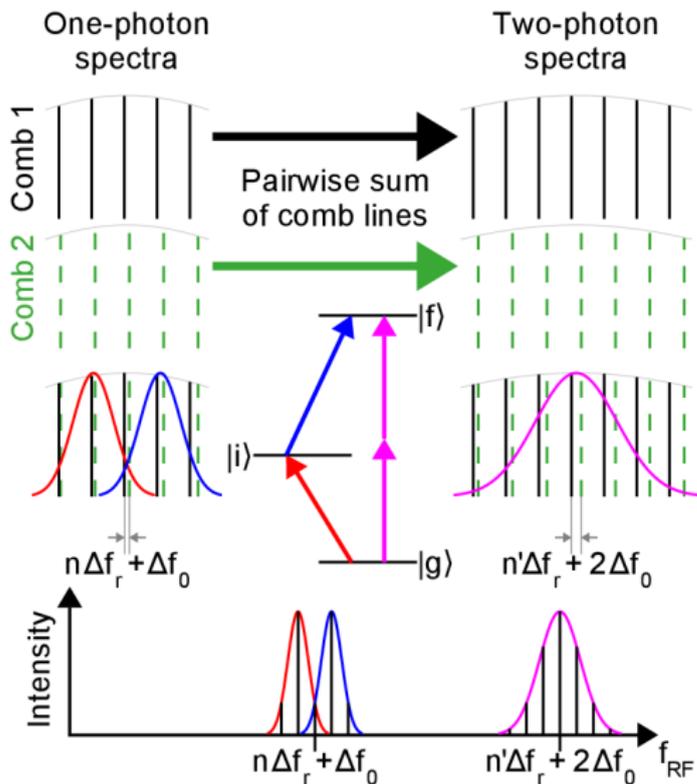
- ▶ Two-photon transitions excited directly or through intermediate state
- ▶ Direct two-photon excitation through pairwise sum of all comb lines
- ▶ RF beatnote frequency doubles for two photon excitation

Principle — Two-photon dual-comb spectroscopy



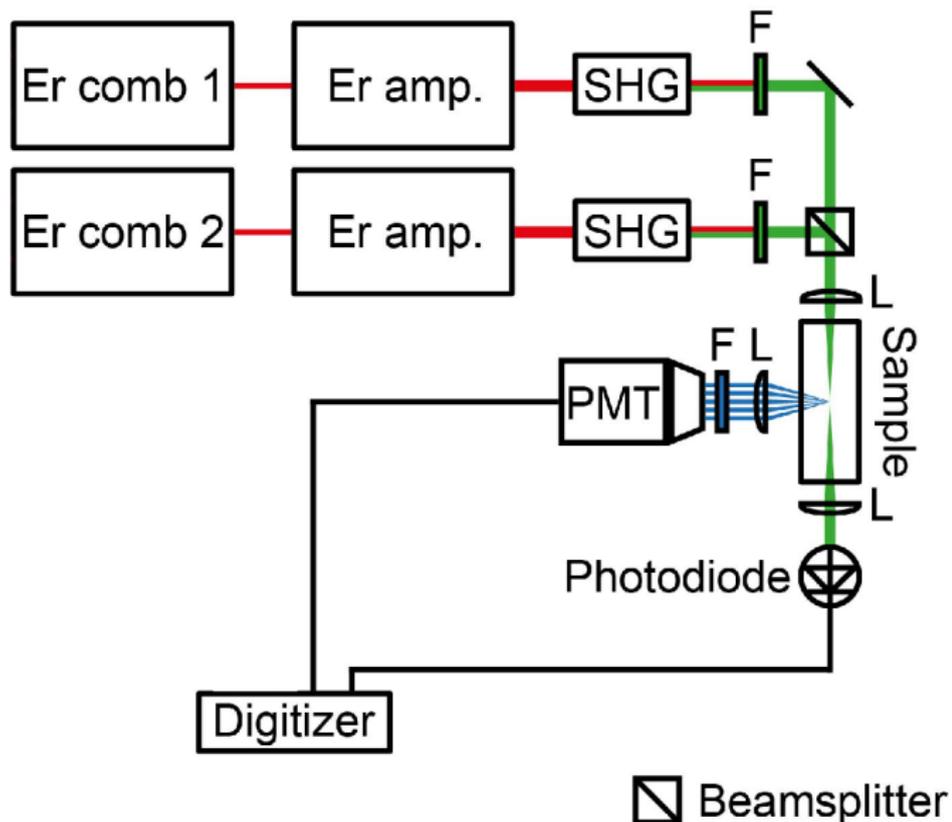
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Principle — Two-photon dual-comb spectroscopy



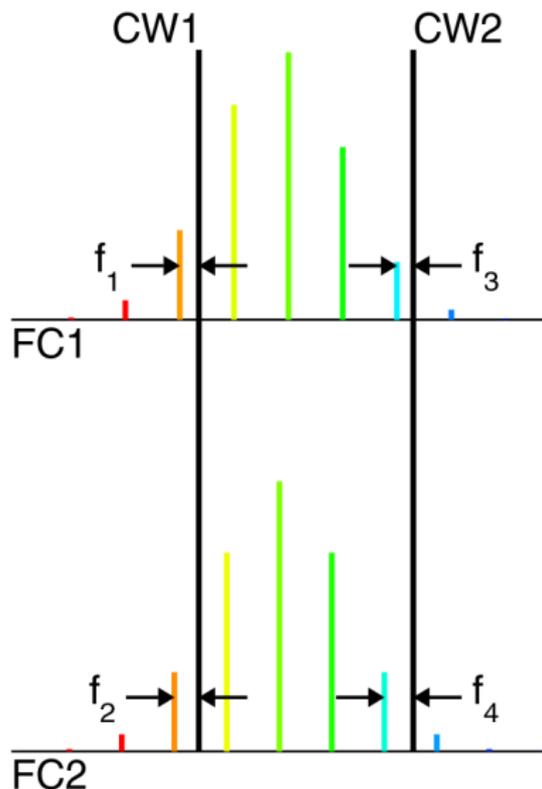
- ▶ Two-photon transitions excited directly or through intermediate state
- ▶ Direct two-photon excitation through pairwise sum of all comb lines
- ▶ RF beatnote frequency doubles for two photon excitation
- ▶ All three transitions observed in RF spectrum

Experimental setup

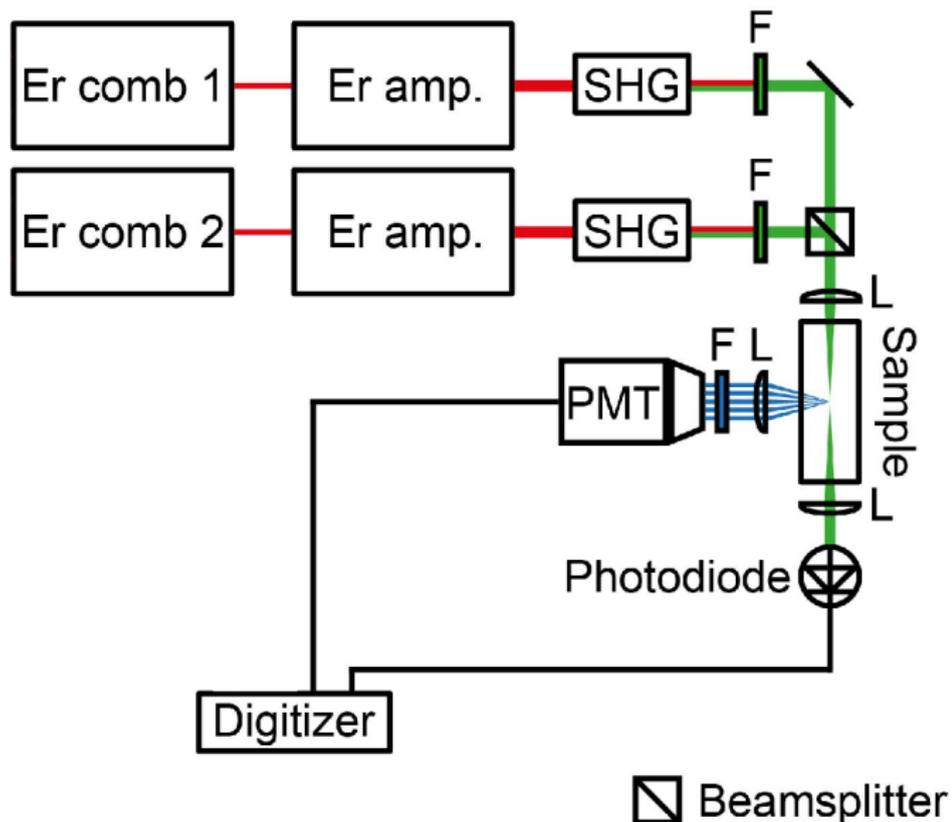


Two-laser adaptive sampling scheme

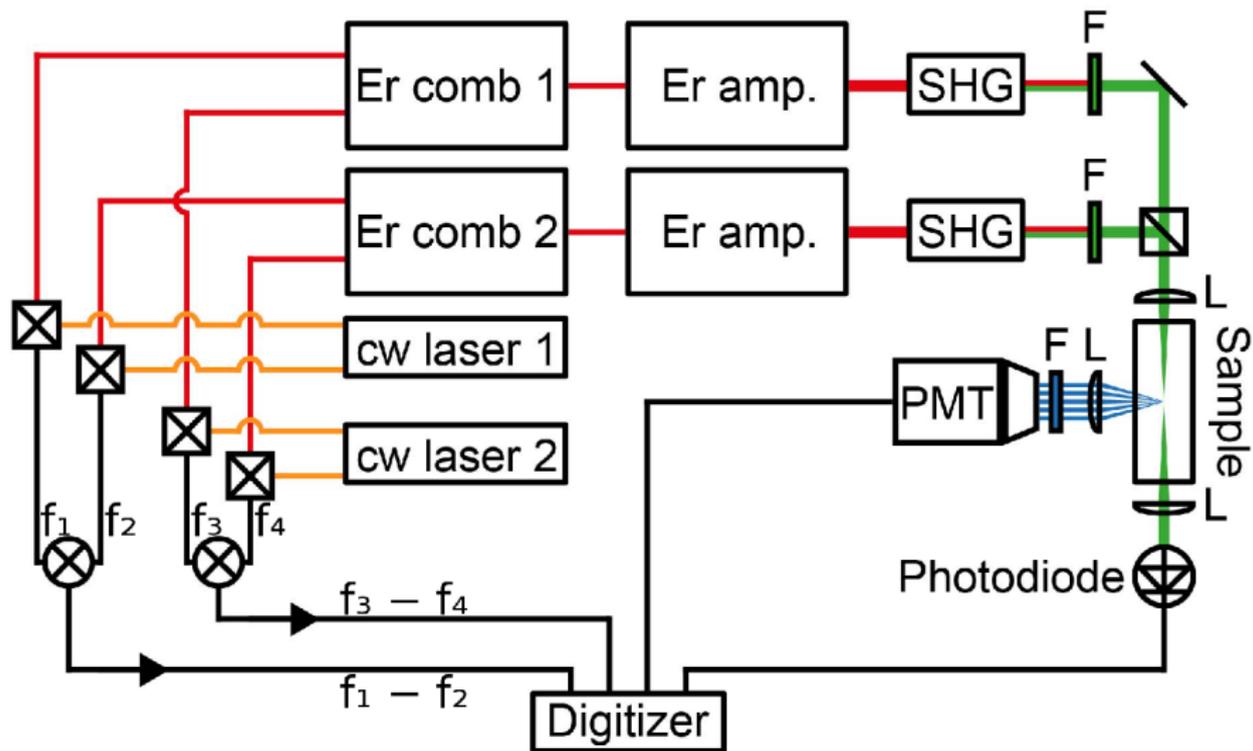
- ▶ $f_{\text{RF}} = n \Delta f_r + k \Delta f_0$ distorted by fluctuations in f_r and f_0
- ▶ Correct by measuring 4 beatnotes with 2 CW lasers
- ▶ Δf_0 correction: adjust phase by $(a + k)(f_1 - f_2) - a(f_3 - f_4)$
 - ▶ a - arbitrary real number
 - ▶ k - net photons absorbed
- ▶ Δf_r correction: scale sampling rate by $(f_1 - f_2) - (f_3 - f_4)$
- ▶ Corrections can be done in hardware (arXiv:1201.4177v2) or software (Deschênes et al. Opt. Express 18, p. 23358–23370 (2010))



Experimental setup

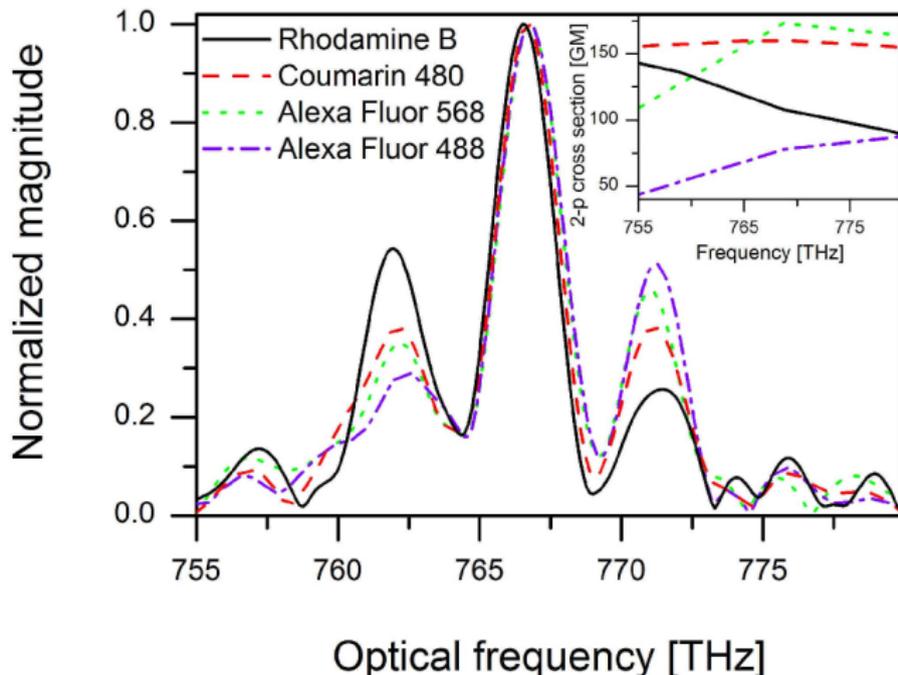


Experimental setup



⊠ Optical beat detection ⊗ Frequency mixer ◻ Beamsplitter

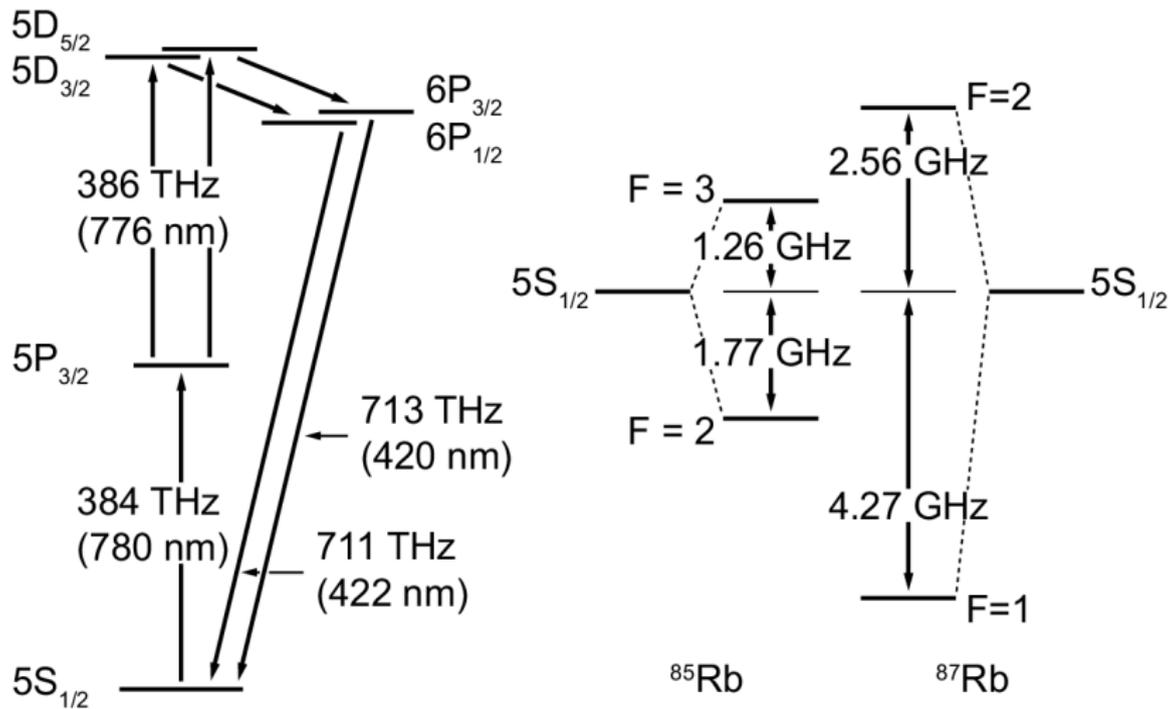
Dye spectrum



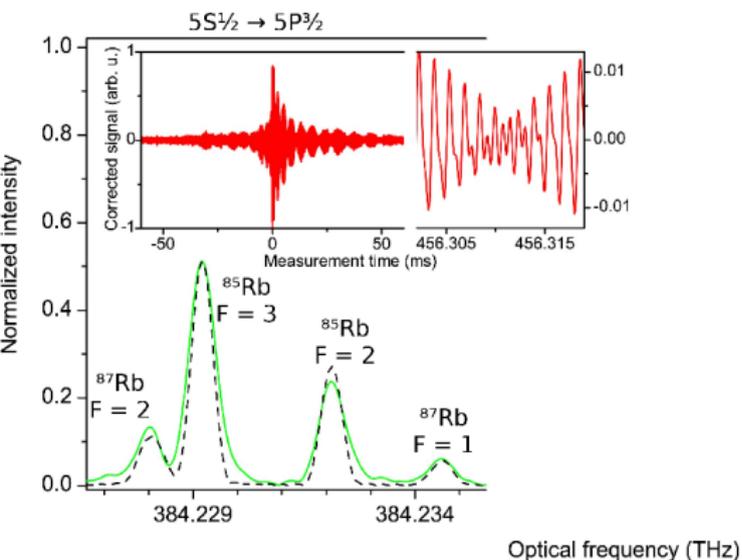
Resolution: 2 THz, Acquisition time: 500 μ s

- ▶ Spectrum distinguishes between dye samples
- ▶ Fast acquisition time

Rubidium level structure

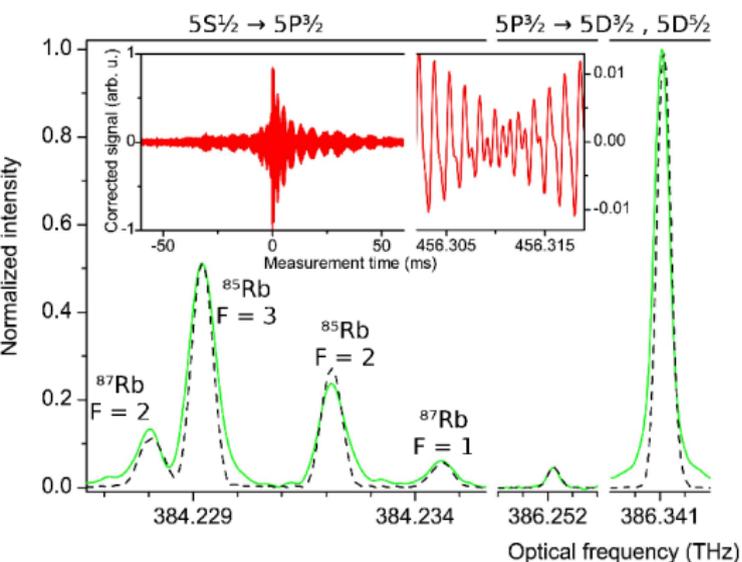


Rubidium spectrum - Doppler limited



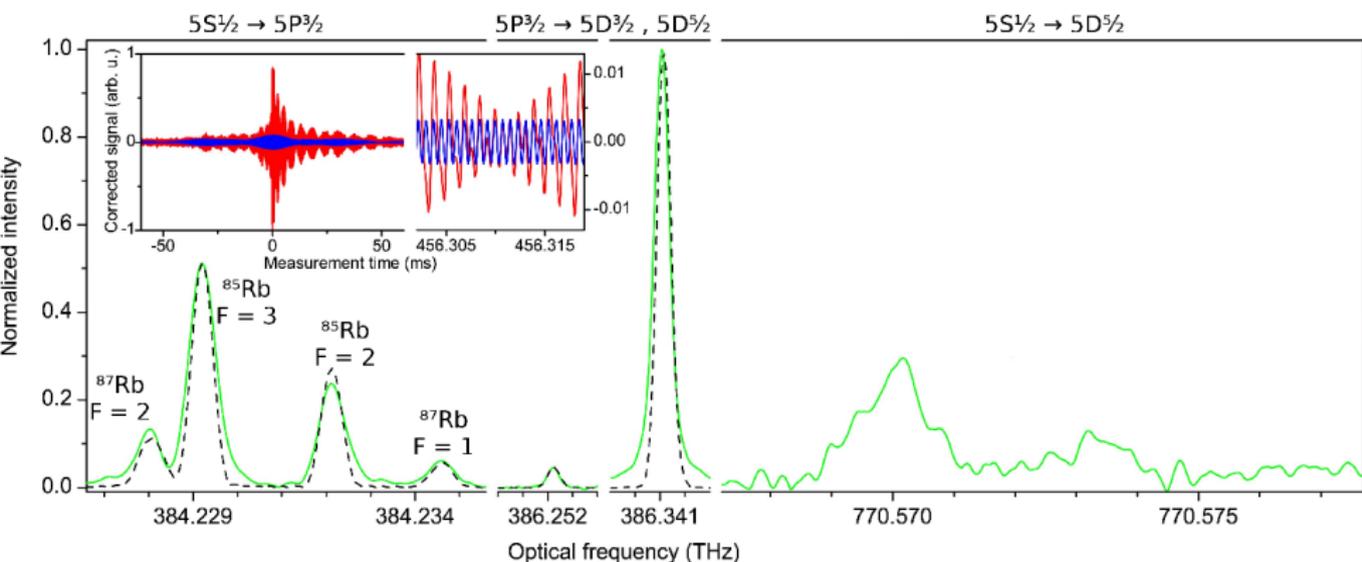
Resolution: 300 MHz, Acquisition time: 18 s

Rubidium spectrum - Doppler limited



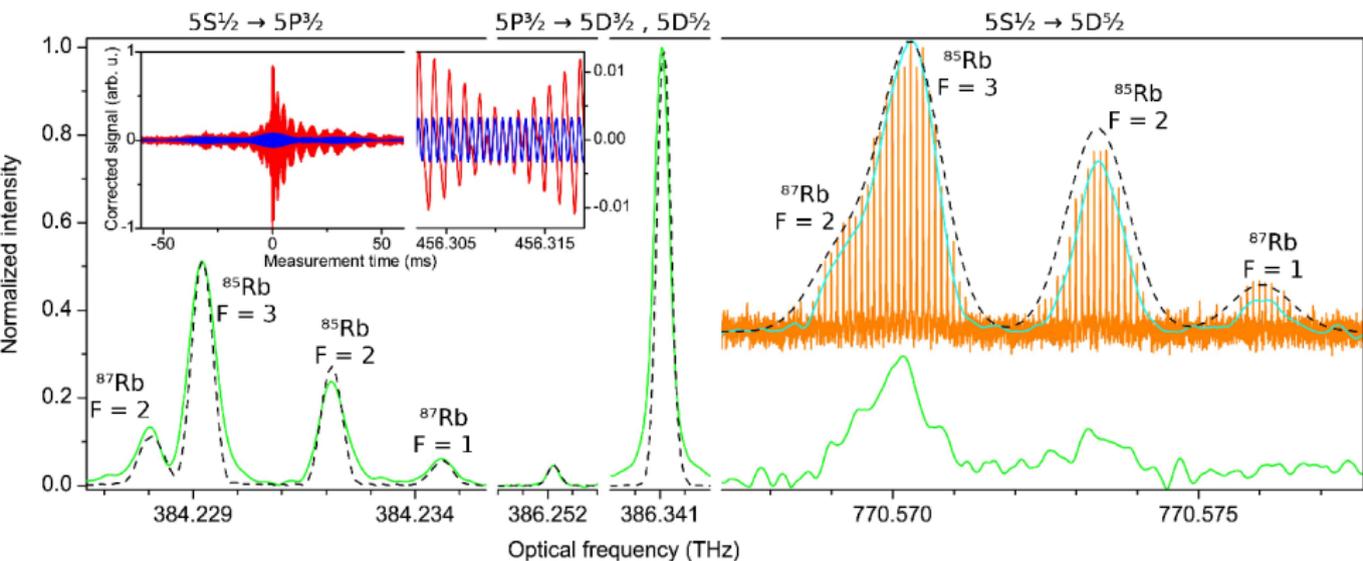
Resolution: 300 MHz, Acquisition time: 18 s

Rubidium spectrum - Doppler limited



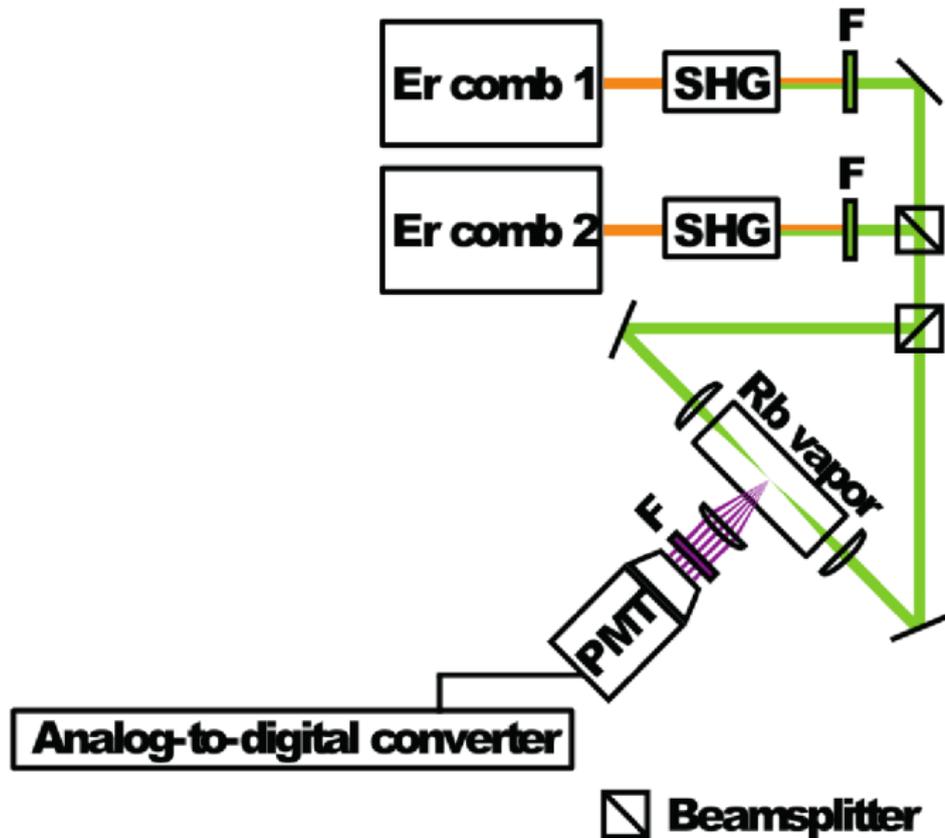
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Rubidium spectrum - Doppler limited

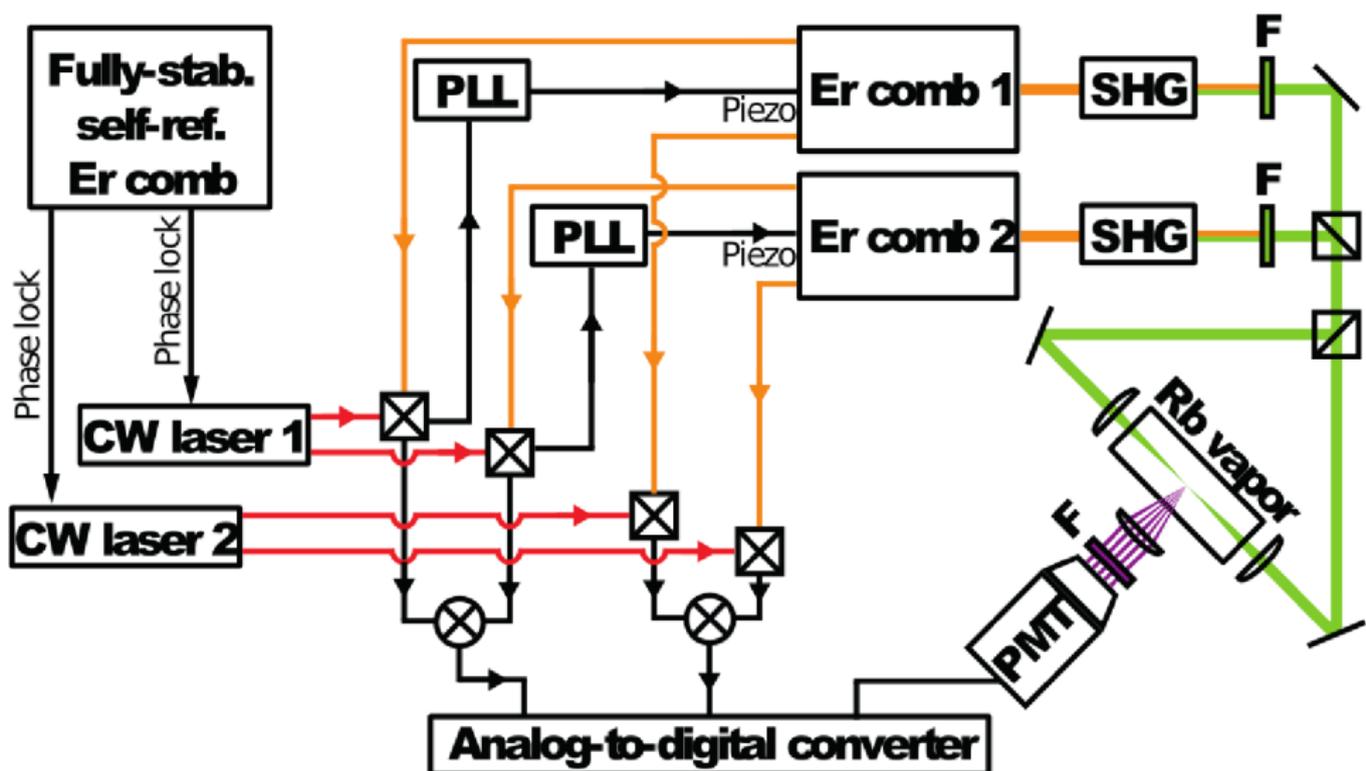


Resolution: 300 MHz, Acquisition time: 18 s

Experimental setup — Doppler-free spectroscopy

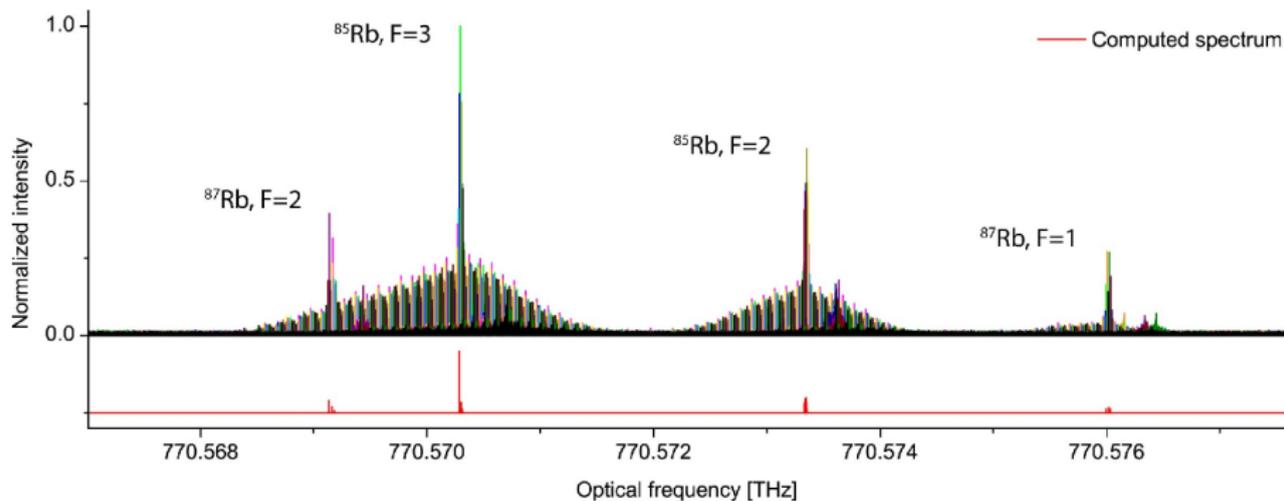


Experimental setup — Doppler-free spectroscopy



⊠ Optical beat detection ⊗ Frequency mixer ◻ Beamsplitter

Rubidium spectrum — Doppler-free



Resolution: 8 MHz, Acquisition time: 52 s

- Interleaving necessary to achieve sub-100 MHz resolution

- ▶ Two-photon excitations useful for fluorescence microscopy and precision spectroscopy
- ▶ Fast measurement times of dual-comb spectroscopy enable spectrally-resolved fluorescence microscopy
- ▶ Dual-comb technique enables precision Doppler-free broadband spectroscopy on complex systems

Acknowledgments



- ▶ Arthur Hipke

Principle Investigators

- ▶ Nathalie Picqué
- ▶ Theodor W. Hänsch