Many-mode Floquet theoretical approach for probing high harmonic generation in intense frequency-comb laser fields

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Abstract

We extend the many-mode Floquet theorem (MMFT)[1)] for the investigation of high harmonic generation of a two-level system driven by intense frequency-comb laser fields. The frequency comb structure generated by a train of short laser pulses can be represented by a combination of the main frequency and the repetition frequency. The MMFT allows non-perturbative and accurate treatment of the interaction of a quantum system with the frequency comb laser fields. We observe that harmonic generation of the two-level system is dramatically enhanced by controlling the repetition frequency and the phase difference between pulses, due to simultaneous resonances.

Introduction

- A train of pulses generates the comb structure in the frequency domain due to quantum interference by the phase difference between pulses[2)].
- The time interval between pulses τ and the pulse-to-pulse carrier-envelope phase (CEP) shift $\Delta\phi$ stabilize comb frequencies[3)].
- Harmonic generation driven by comb laser is expected to have the comb structure[4)].

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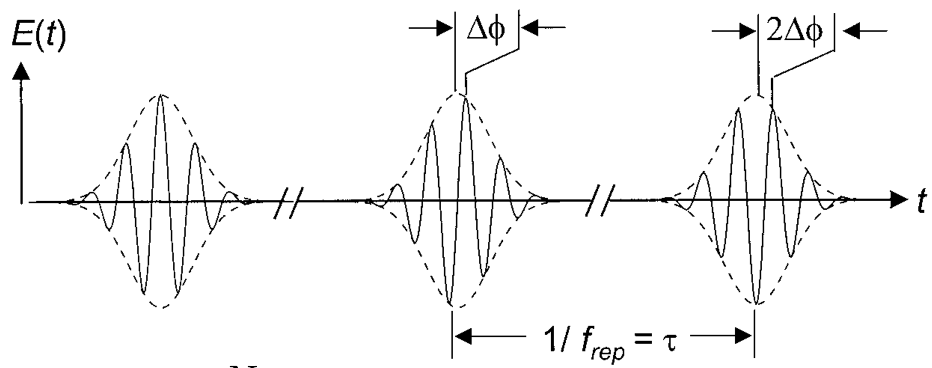
Computational Details

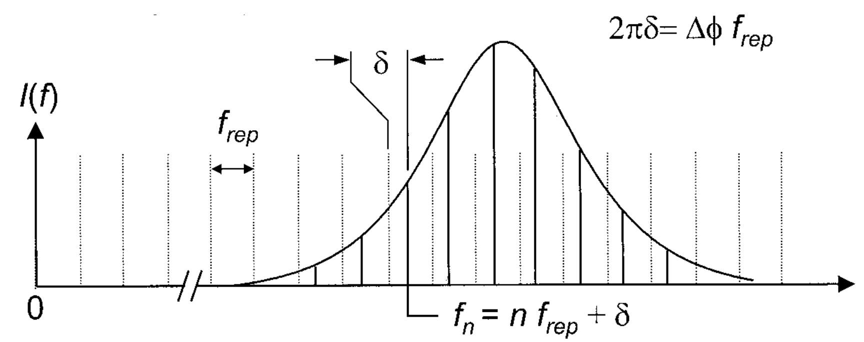
In the time domain*



$$E(t) = \sum_{n=1}^{N_p} f(t - n\tau) e^{i(\omega_c t - n\omega_c \tau + n\Delta\phi)}$$

$$f(t) = f_0 e^{-t^2/2\sigma^2}$$

In the frequency domain*



FT

$$\tilde{E}(\omega) = \tilde{f}(\omega - \omega_c) \sum_{n=1}^{N_p} e^{-in(\omega\tau - \Delta\phi)}$$

$$N_p \to \infty$$

$$\tilde{E}^\circ(\omega) = \tilde{f}(\omega - \omega_c)\omega_r \sum_{k=-\infty}^{\infty} \delta(\omega - \omega_k)$$

IFT

$$E^\circ(t) = \sum_{k=-N}^{N} E_k e^{i\omega_k t}$$

$$E_k = \frac{f_0 \sigma \omega_r}{\sqrt{2\pi}} e^{-\sigma^2(\omega_0 - \omega_c + k\omega_r)^2/2}$$

Frequency-comb: $\omega_k = \omega_0 + k\omega_r$

* Pictures taken from Cundiff *et al.*, *Rev. Sci. Instrum.* **72**, 3749 (2001)

Time-dependent Hamiltonian

$$H(\mathbf{r},t) = \hat{H}_0(\mathbf{r}) - \sum_{k=-N}^{N} \boldsymbol{\mu}(\mathbf{r}) \cdot \mathbf{E}_k \, \Re\left[e^{i\omega_k t}\right]$$

$$= \hat{H}_0(\mathbf{r}) - \tfrac{1}{2} \sum_{k=-N}^{N} \hat{z} E_k \left(e^{i(\omega_0 + k\omega_r)t} + e^{-i(\omega_0 + k\omega_r)t}\right)$$

Generalized Floquet basis state[5)]: $|\alpha n m\rangle = |\alpha\rangle \otimes |n\rangle \otimes |m\rangle$

$$H(\mathbf{r},t) = \sum_{n,m} H^{[n,m]}(\mathbf{r}) e^{-i(n\omega_0 + m\omega_r)t}$$

$$H^{[0,0]} = H_0$$

$$H^{[-1,0]} = H^{[+1,0]} = -\tfrac{1}{2}\hat{z}E_0$$

$$H^{[-1,-1]} = H^{[+1,+1]} = -\tfrac{1}{2}\hat{z}E_1, \quad H^{[-1,+1]} = H^{[+1,-1]} = -\tfrac{1}{2}\hat{z}E_{-1}$$

$$H^{[-1,-2]} = H^{[+1,+2]} = -\tfrac{1}{2}\hat{z}E_2, \quad H^{[-1,+2]} = H^{[+1,-2]} = -\tfrac{1}{2}\hat{z}E_{-2}$$

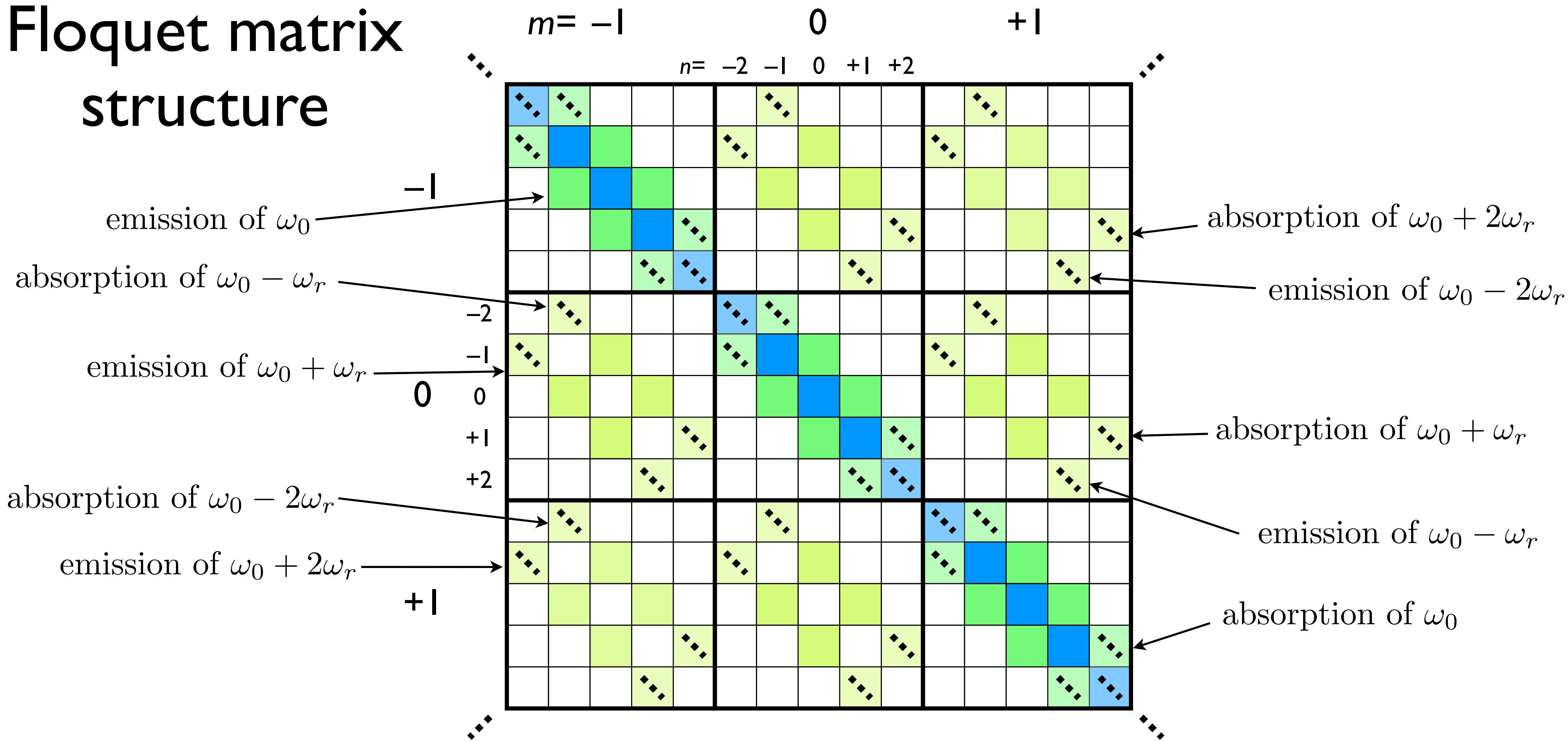
$$\vdots$$

$$H^{[-1,-N]} = H^{[+1,+N]} = -\tfrac{1}{2}\hat{z}E_N, \quad H^{[-1,+N]} = H^{[+1,-N]} = -\tfrac{1}{2}\hat{z}E_{-N}$$

From MMFT[1,5)], the time-dependent problem can be transformed into a time-independent infinite-dimensional matrix eigenvalue problem.

$$\sum_{\beta} \sum_{n'} \sum_{m'} \langle \alpha n m | H_F | \beta n' m' \rangle \langle \beta n' m' | \lambda \rangle = \lambda \langle \alpha n m | \lambda \rangle$$

Floquet matrix structure



$$\langle \alpha n m | H_F | \beta n' m' \rangle = H_{\alpha\beta}^{[n-n',m-m']} + (n\omega_0 + m\omega_r)\delta_{\alpha,\beta}\delta_{n,n'}\delta_{m,m'}$$

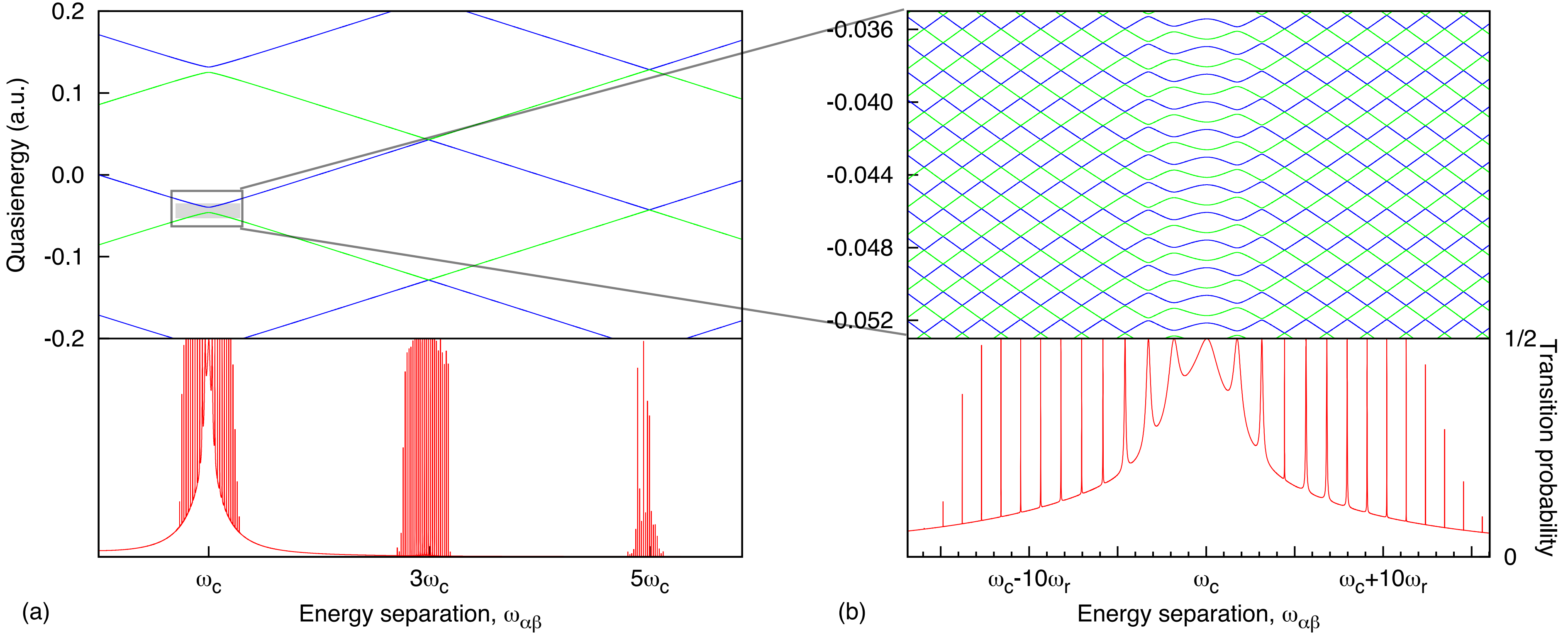
$$H_{\alpha\beta}^{[n-n',m-m']} = \langle \alpha | H^{[n-n',m-m']} | \beta \rangle$$

$$= \varepsilon_\alpha \delta_{\alpha,\beta}\delta_{n,n'}\delta_{m,m'} + \sum_{k=-N}^{N} V_{\alpha\beta}^{(k)} \left(\delta_{n+1,n'}\delta_{m+k,m'} + \delta_{n-1,n'}\delta_{m-k,m'}\right),$$

$$\varepsilon_\alpha = \langle \alpha | \hat{H}_0 | \alpha \rangle \quad \text{and} \quad V_{\alpha\beta}^{(k)} = -\tfrac{1}{2} E_k \langle \alpha | \hat{z} | \beta \rangle$$

Results

Plot of quasienergies as a function of $\omega_{\alpha\beta}$



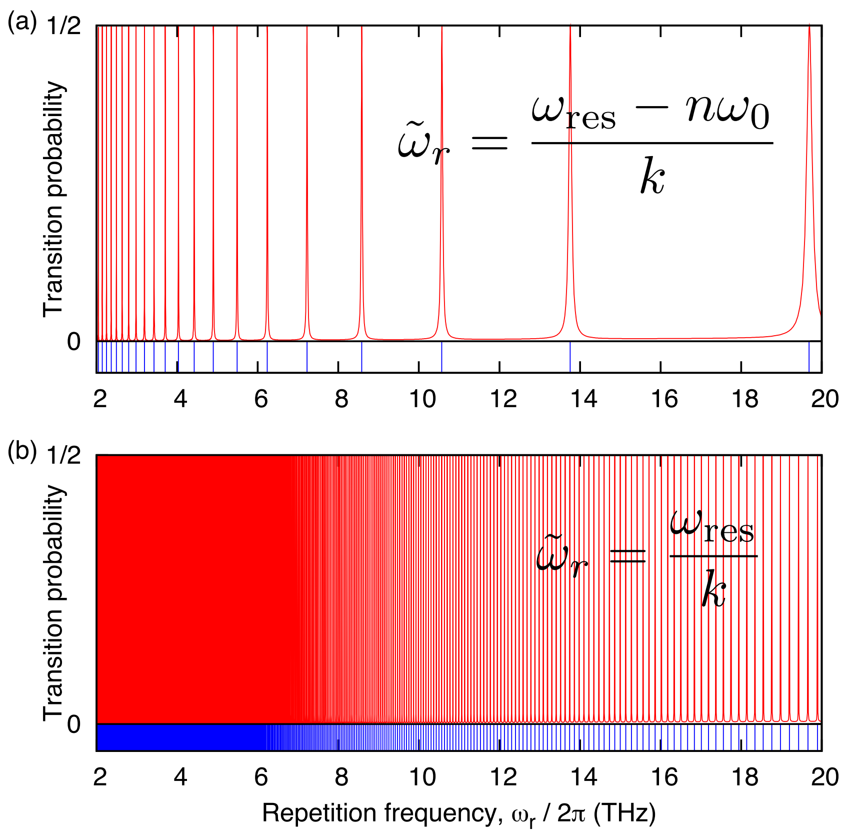
Comb laser: 2.5×10^{15} W/cm^2, 532 nm, f_{rep}=10 THz, 20 fs FWHM Gaussian pulses

Two-level system: $\omega_{\alpha\beta} = \varepsilon_\beta - \varepsilon_\alpha, \quad \langle\alpha|z|\beta\rangle = 0.1$ a.u.

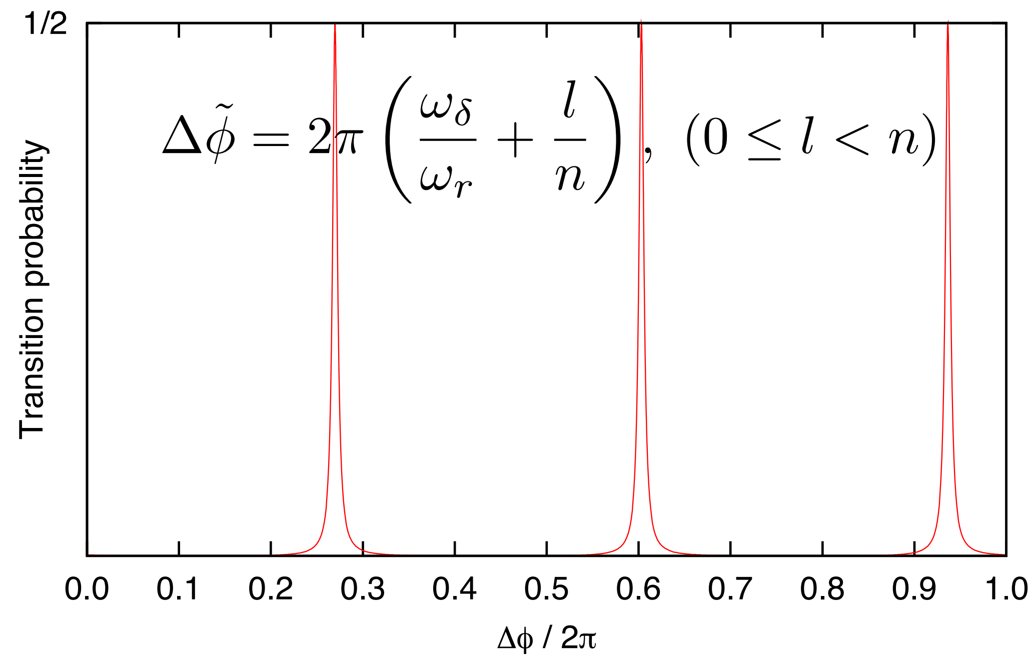
Quasienergy: $\lambda_{\gamma mn} = \lambda_\gamma + n\omega_0 + m\omega_r \quad (n, m: \text{ integer})$

3-photon dominant resonance cases:

Plot of transition probabilities as a function of ω_r



Plot of transition probabilities as a function of Δϕ

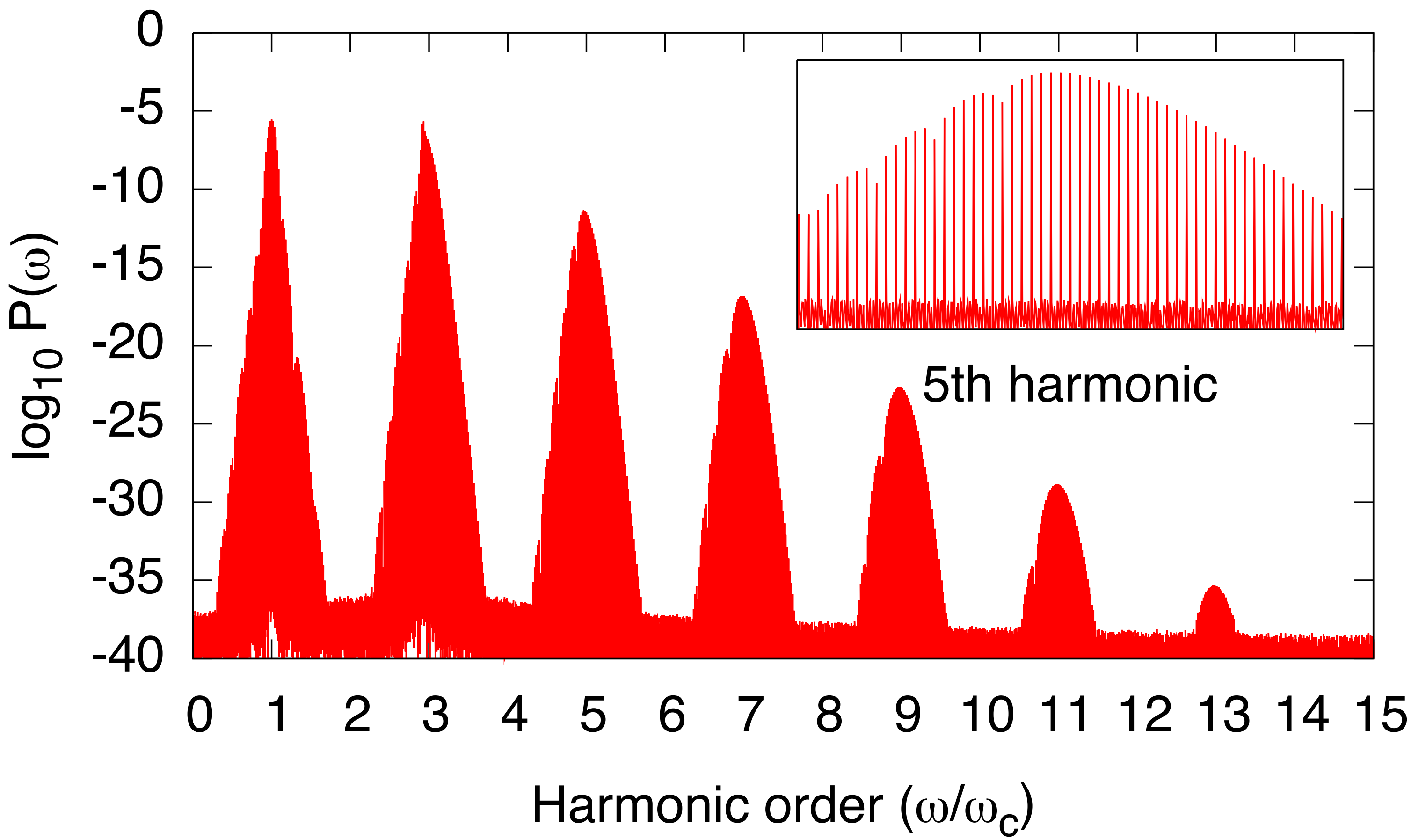


n-photon resonance condition

one-mode: $\omega_{\rm res} = n\omega$

comb: $\omega_{\rm res} \equiv n\omega \pmod{\omega_r}$

Power spectra driven by intense comb laser field



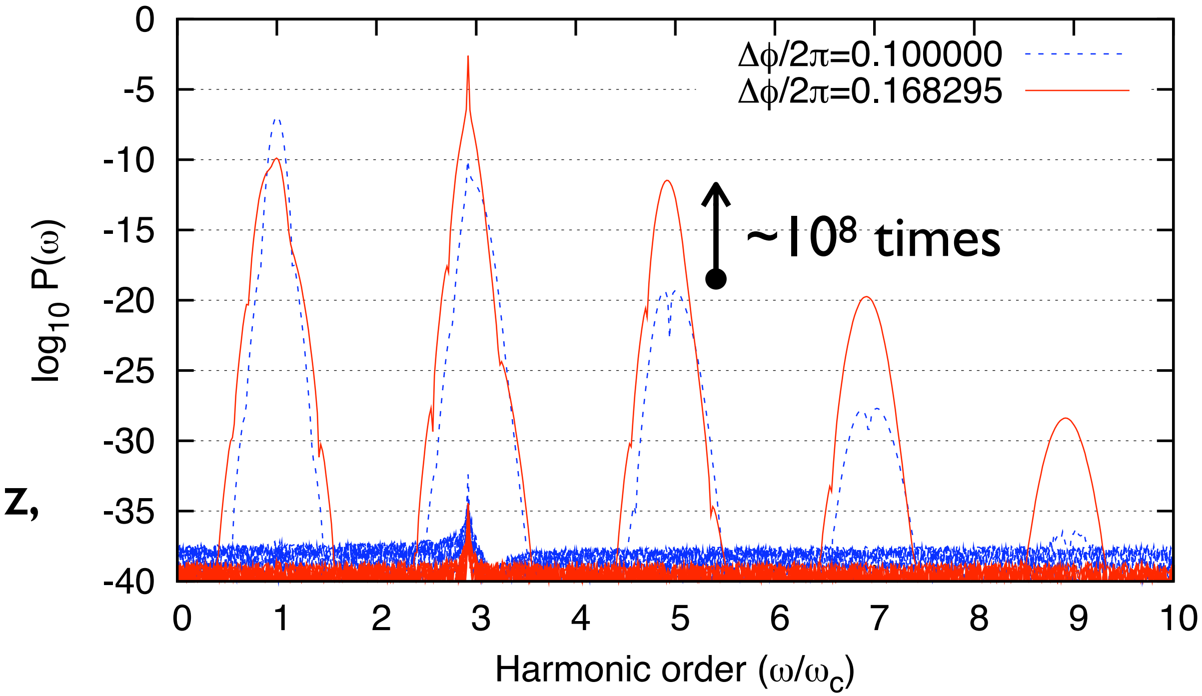
n-th harmonic of combs: $\{n\omega_0 + k\omega_r\}$ combs

repetition angular frequency: ω_r

offset angular frequency: $n\omega_0 \pmod{\omega_r}$

Enhancement of HHG by controlling Δϕ to tune resonances

1×10^{14} W/cm^2, 532 nm, f_{rep}=10 THz, 20 fs FWHM Gaussian pulses



1×10^{14} W/cm^2				1×10^{15} W/cm^2				2.5×10^{15} W/cm^2			
A		B		A		B		A		B	
q	$P(q\omega_c)$	q	$P(q\omega_c)$	q	$P(q\omega_c)$	q	$P(q\omega_c)$	q	$P(q\omega_c)$	q	$P(q\omega_c)$
2.92	9.15[−11]	2.92	2.50[−3]	2.92	7.07[−8]	2.92	2.41[−3]	2.92	1.33[−6]	2.92	2.01[−3]
5.00	4.53[−20]	4.91	3.42[−12]	5.00	6.62[−15]	4.93	3.39[−10]	5.00	1.28[−12]	4.94	2.07[−9]
7.02	1.99[−28]	6.92	1.83[−20]	7.00	3.10[−21]	6.93	1.82[−16]	7.00	4.13[−18]	6.95	6.82[−15]
9.03	5.58[−37]	8.92	4.07[−29]	9.01	6.00[−28]	8.92	3.99[−23]	9.01	5.30[−24]	8.95	9.40[−21]
				11.02	5.45[−35]	10.92	4.04[−30]	11.00	3.14[−30]	10.95	5.95[−27]
								12.98	1.05[−36]	12.95	1.96[−33]

A: no resonance, $\Delta\phi/2\pi = 0.1$

B: resonance, $\Delta\phi/2\pi = 0.168295$, 0.20634, and 0.269741 for 1×10^{14}, 1×10^{15}, and 2.5×10^{15} W/cm^2

Conclusion

- The frequency-comb structure can be expressed by the main frequency and the repetition frequency.
- Multiphoton resonances with the system and comb laser can be achieved by controlling the repetition frequency and the CEP shift.
- HHG driven by intense frequency-comb laser has the comb structure with the same repetition frequency and different offset for each harmonic.
- HHG shows immense enhancement by controlling the CEP shift due to simultaneous multiphoton resonance among comb frequencies.

References

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3. D. J. Jones, *et al.*, *Science* **288**, 635 (2000).
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