Nonlinear Optics (WiSe 2018/19)

Lecture 19: December 21, 2018

Chapter 10: Interactions of light and matter

10.8 Extreme nonlinear optical response of two-level systems

Chapter 13: Strong-field physics in solids

13.4 Carrier-wave Rabi flopping

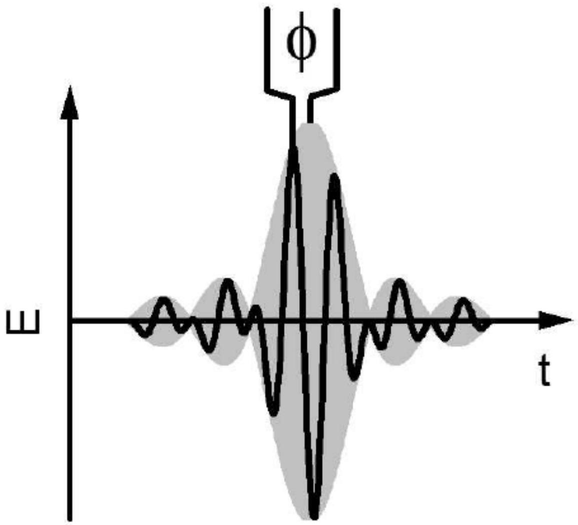
13.5 THG in disguise of SHG

10.8 Extreme nonlinear optical response of two-level systems

extreme nonlinear optics: $E(t)$ not $I(t) \propto |\tilde{E}(t)|^2$ matters

RWA and SVEA cannot be used

observables depend on CEP ϕ

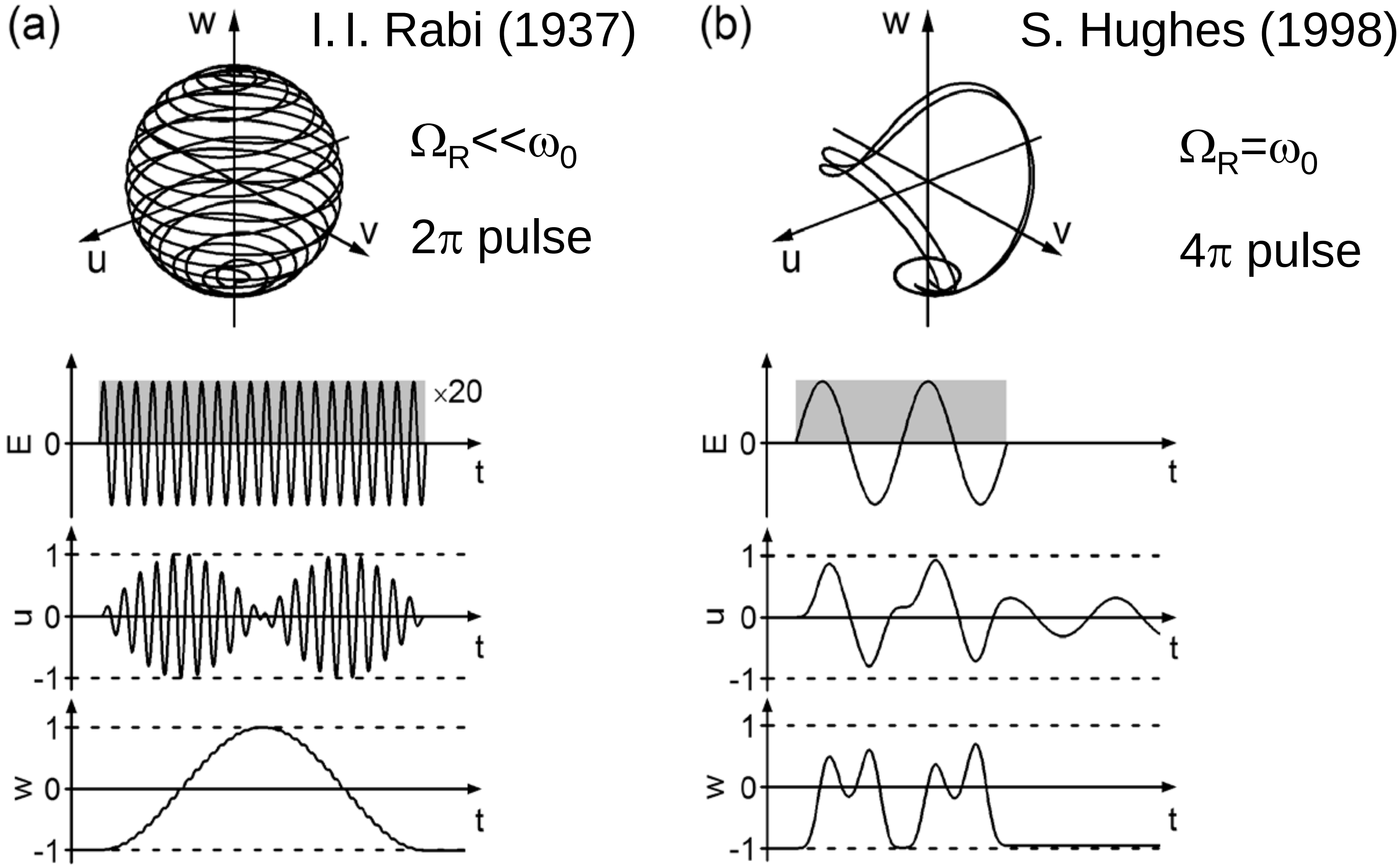


numerically **solve Bloch equations exactly**
(i.e., *without* employing RWA) driven by $E(t)$

M. Wegener, *Extreme Nonlinear Optics*, Springer, Berlin (2005)

Carrier-wave Rabi flopping

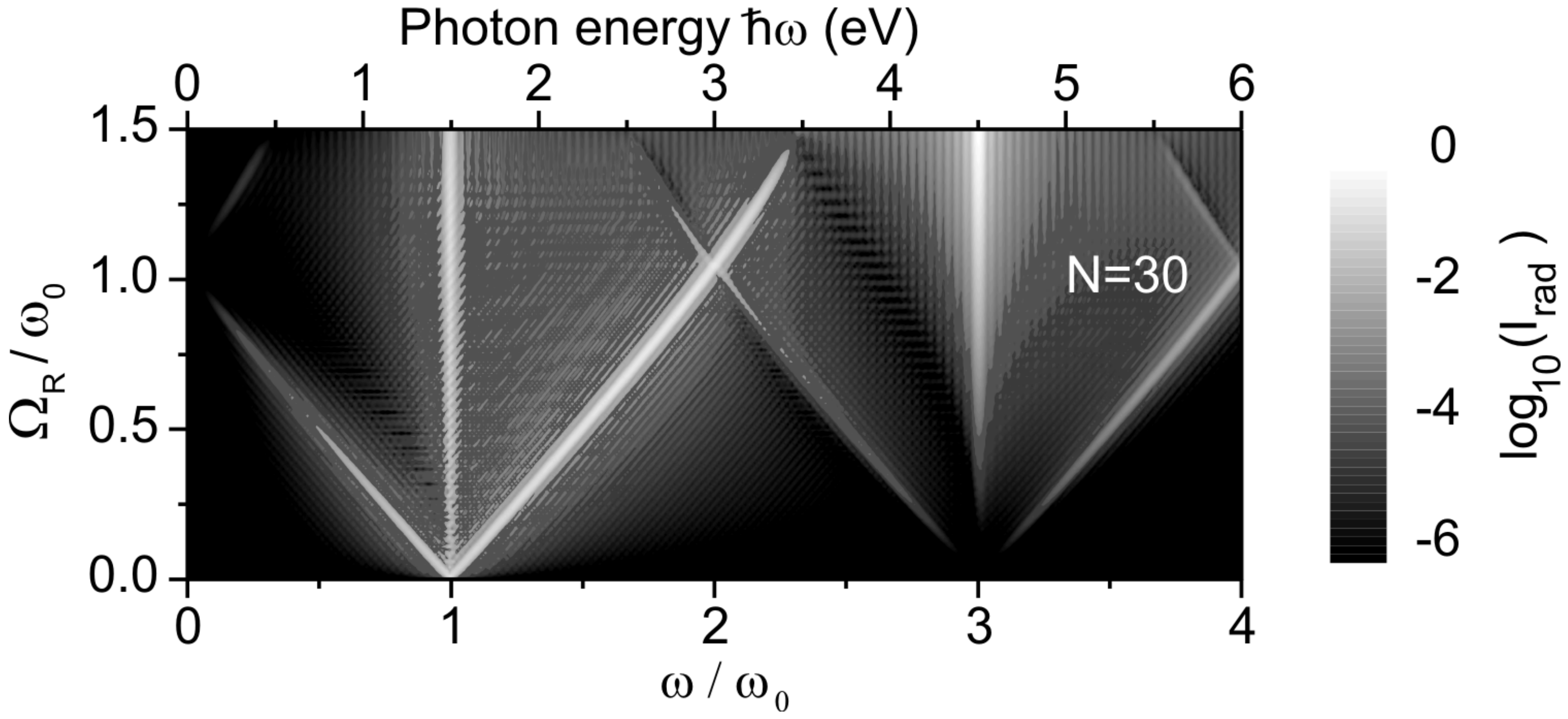
Bloch vector $(u,v,w)=(2\mathrm{Re}(p_{vc}),2\mathrm{Im}(p_{vc}),f_c-f_v)$ Rabi frequency $\Omega_R(t)\propto E(t)$



Carrier-wave Mollow triplets

B. R. Mollow (1969)

30 cycle long box-shaped pulses



Mollow sidebands at $(2n+1)\omega_0 \pm \Omega_R$

Within the dipole approximation, but without employing the RWA and without transverse or longitudinal damping, the Bloch equations of a two-level system with transition frequency Ω for the Bloch vector $(u, v, w)^{\mathrm{T}}$ can be written in matrix form as

$$\begin{pmatrix} \dot{u} \\ \dot{v} \\ \dot{w} \end{pmatrix} = \begin{pmatrix} 0 & +\Omega & 0 \\ -\Omega & 0 & -2\Omega_{\mathrm{R}}(t) \\ 0 & +2\Omega_{\mathrm{R}}(t) & 0 \end{pmatrix} \begin{pmatrix} u \\ v \\ w \end{pmatrix} . \tag{10.101}$$

The dots denote the derivative with respect to time t. Here, we have introduced the (instantaneous) Rabi frequency $\Omega_{\mathrm{R}}(t)$ via the (instantaneous) Rabi energy

$$\hbar\Omega_{\mathrm{R}}(t) = dE(t) \tag{10.102}$$

with dipole matrix element d and the laser electric field defined as

$$E(t) = \tilde{E}(t)\cos(\omega_0 t + \phi). \tag{10.103}$$

Note that the Rabi frequency itself oscillates with the carrier frequency of light and periodically changes sign. We shall call the peak of the Rabi frequency Ω_{R} [rather than $\Omega_{\mathrm{R}}(t)$] with $\hbar\Omega_{\mathrm{R}} = d\tilde{E}_0$, where $\tilde{E}_0$ is the peak of the electric-field envelope.

T. Tritschler *et al.*, PRA **68**, 033404 (2003)

The Bloch vector $(u, v, w)^{\mathrm{T}}$ thus allows an intuitive geometric representation of the state of the two-level system which was introduced by R. P. Feynman *et al.* [9]. The complex amplitude of the superposition state is encoded in the real and the imaginary part of the transition amplitude, i.e., in the components u and v of the Bloch vector. The component w is again the inversion of the two-level system, i.e., it is equal to -1 if all electrons are in the ground state, and it is +1 for complete inversion. The light intensity radiated by the two-level system is proportional to the square modulus of the second temporal derivative of the macroscopic polarization, hence proportional to $|\omega^2 u(\omega)|^2$ in the Fourier domain, where ω is the spectrometer frequency. For vanishing relaxation, the length of the Bloch vector is conserved and equal to one, i.e.,

$$\sqrt{u(t)^2 + v(t)^2 + w(t)^2} = 1\,. \tag{10.104}$$

Hence, all the physics can be represented as rotations of the Bloch vector on a sphere with radius unity, the so-called Bloch sphere. For vanishing electric field, the Bloch vector rotates in the uv-plane with a frequency given by the optical transition frequency Ω, for very large fields one gets a rotation in the vw-plane with frequency $\Omega_{\mathrm{R}}(t)$. This oscillation is the Rabi oscillation. If, for example, during the action of the electric field pulse, the Bloch vector performs one complete rotation in the vw-plane, the pulse area $\Theta = \frac{d}{\hbar}\int_{-\infty}^{+\infty} \mathrm{d}t\, \tilde{E}(t)$ is equal to 2π. There is, however, no simple analytical expression for Θ. For finite Ω and Ω_{R}, the dynamics of the Bloch vector is a combination of both rotations, one in the uv-plane and one in the vw-plane.

Most importantly the optical Bloch equations (10.101) are invariant under space inversion [8]: Space inversion means that we have to replace $\vec{r} \to -\vec{r}$. Thus, the dipole matrix element transforms as $d \to -d$, the electric field as $E(t) \to -E(t)$, and the Rabi frequency as $\Omega_{\mathrm{R}}(t) \to +\Omega_{\mathrm{R}}(t)$ according to Eq. (10.102). As a result, the optical Bloch equations (10.101) are invariant under space inversion and the solution for the Bloch vector $(u(t), v(t), w(t))^{\mathrm{T}}$ is also unchanged. Finally, the macroscopic optical polarization, which is given by $P(t) = n_{\mathrm{TLS}} d u(t)$ with the density of two-level systems n_{TLS}, transforms according to $P(t) \to -P(t)$. Consequently, in an expansion of the polarization in terms of powers of the electric field up to infinite order, strictly no even harmonic orders occur – even for arbitrarily large electric fields [8]. In the literature, one can find several papers reporting on symmetry breaking of two-level systems driven by strong laser fields, that is supposedly leading to second-harmonic generation. This claim is physically wrong, as proven by the invariance under space inversion! A more careful analysis reveals that although light can indeed be emitted at the spectral position of even harmonics, the corresponding carrier frequency and phases allow to clearly identify them belonging to odd-order harmonics, as we will below.

From this model, a complete overview of the rich behavior as a function of the four involved frequencies can be obtained [7, 8]: Carrier frequency of light ω_0, transition frequency Ω, Rabi frequency Ω_R, and spectrometer frequency ω. Thereby it is natural to scale all frequencies to ω_0, in which case the dependence of the radiated intensity on the three dimensionless parameters Ω/ω_0, Ω_R/ω_0, and ω/ω_0 has to be studied. In all calculations, we start from the ground state of the two-level system, i.e., from Bloch vector $(0, 0, -1)^T$.

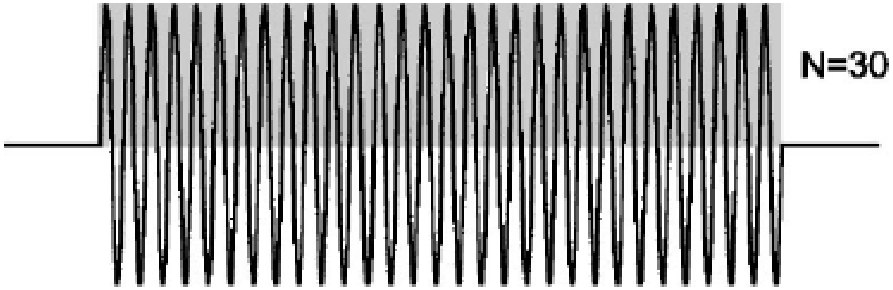


Figure 10.5: Box-shaped optical pulses $E(t)$: The integer number of cycles in the pulse is called N. The gray area indicates the electric-field envelope $\tilde{E}(t)$. [8]

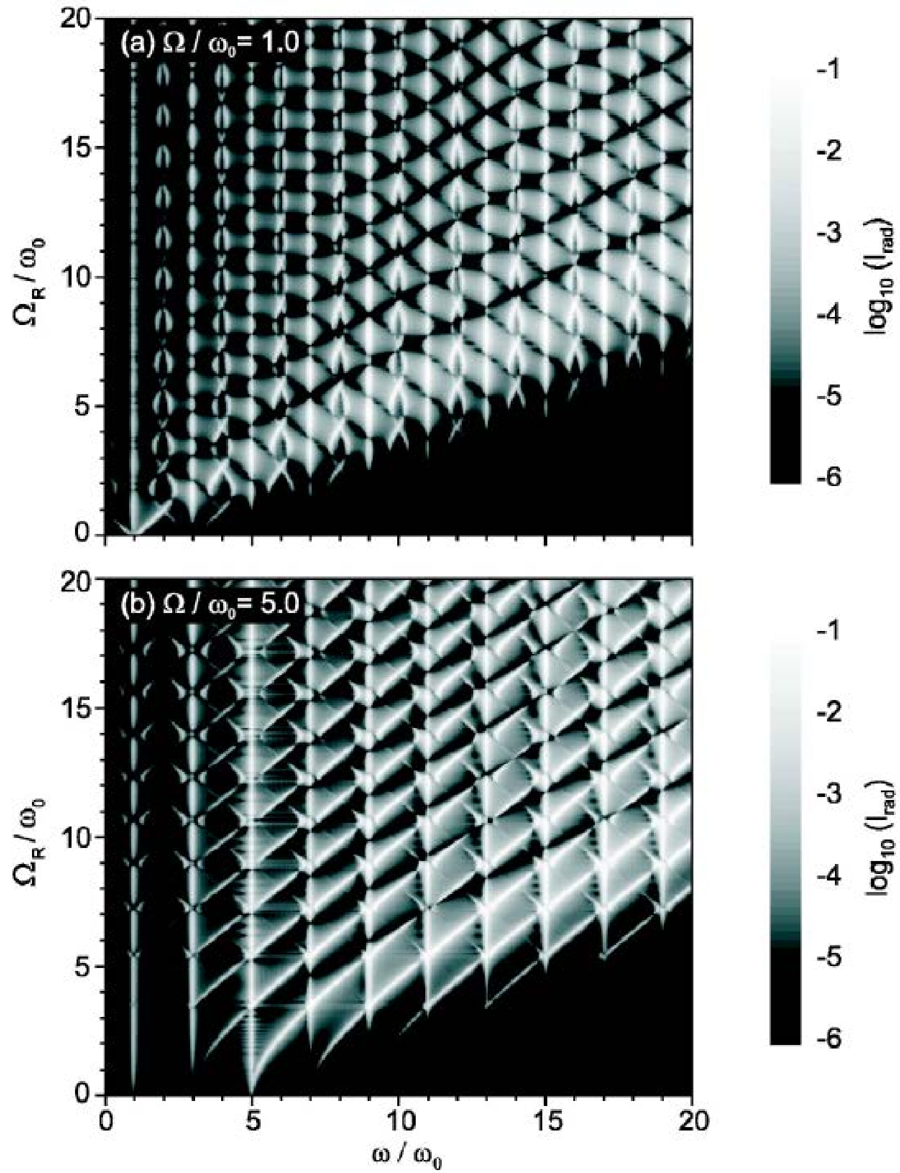


Figure 10.6: Gray-scale images of the radiated intensity spectra $I_{\mathrm{rad}}(\omega) \propto |\omega^2 u(\omega)|^2$ (normalized and on a logarithmic scale) from exact numerical solutions of the two-level system Bloch equations (10.101). The peak Rabi frequency Ω_{R} of the exciting $N = 30$ cycles long box-shaped optical pulses is plotted along the vertical axis. The transition frequency ω is parameter. (a) $\Omega/\omega_0 = 1$ and (b) $\Omega/\omega_0 = 5$. ω_0 is the carrier frequency of the laser pulses. [8]

resonant excitation

conventional Rabi flopping

carrier-wave Rabi flopping

carrier-wave Mollow triplets around odd harmonics

off-resonant excitation

T. Tritschler *et al.*,
PRA **68**, 033404 (2003)

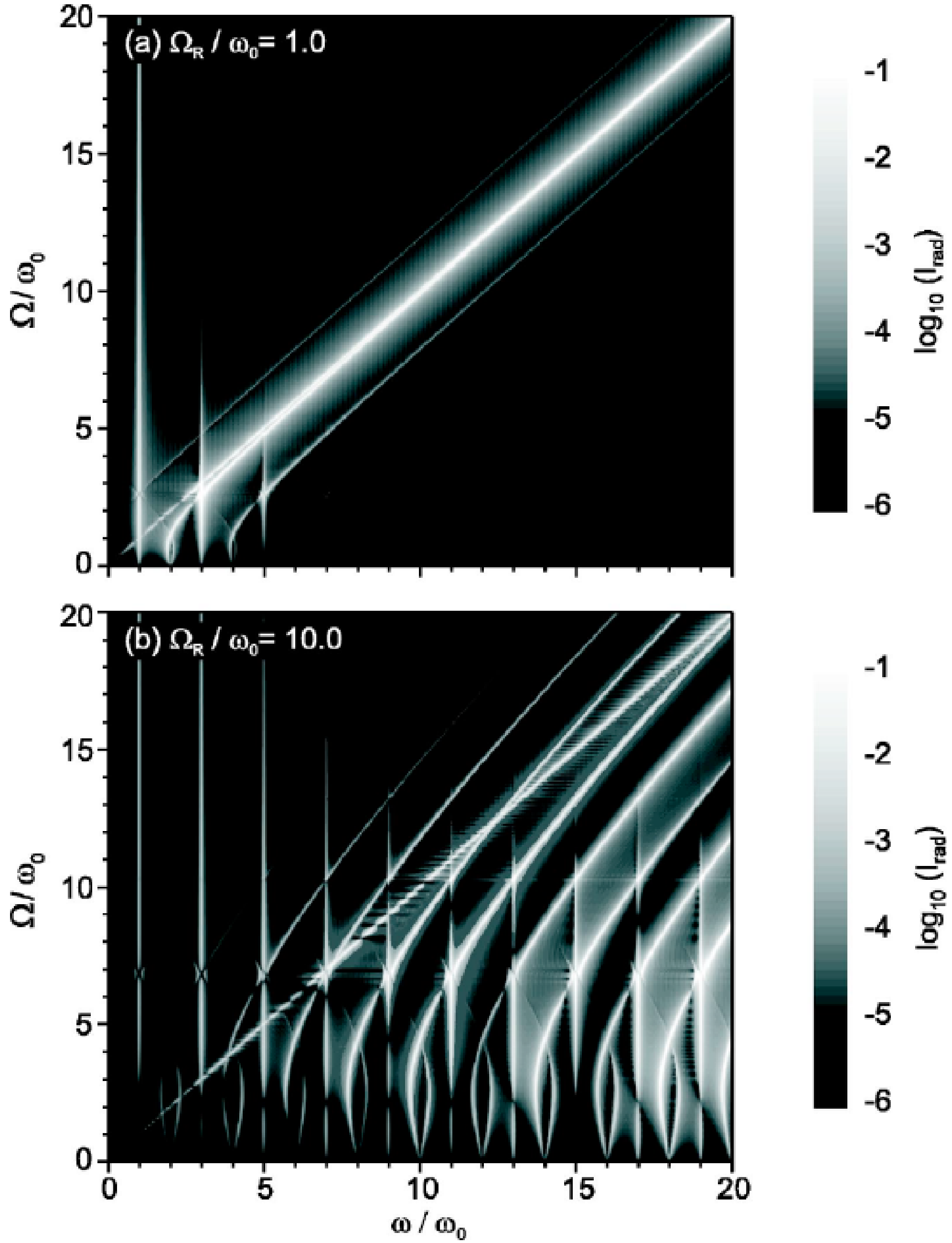


Figure 10.7: Same as Fig. 10.6, but versus transition frequency Ω for two fixed values of the peak Rabi frequency Ω_R. (a) $\Omega_R/\omega_0 = 1$ and (a) $\Omega_R/\omega_0 = 10$. [8]

On the diagonal, where $\omega = \Omega$, very large resonant enhancement effects

large contributions can occur at spectral positions of even harmonics

but no even harmonics (inversion symmetry)

THG in disguise of SHG

for SHG it would be carrier wave $\mathbf{2\omega_0}$
CEP $\mathbf{2\phi}$

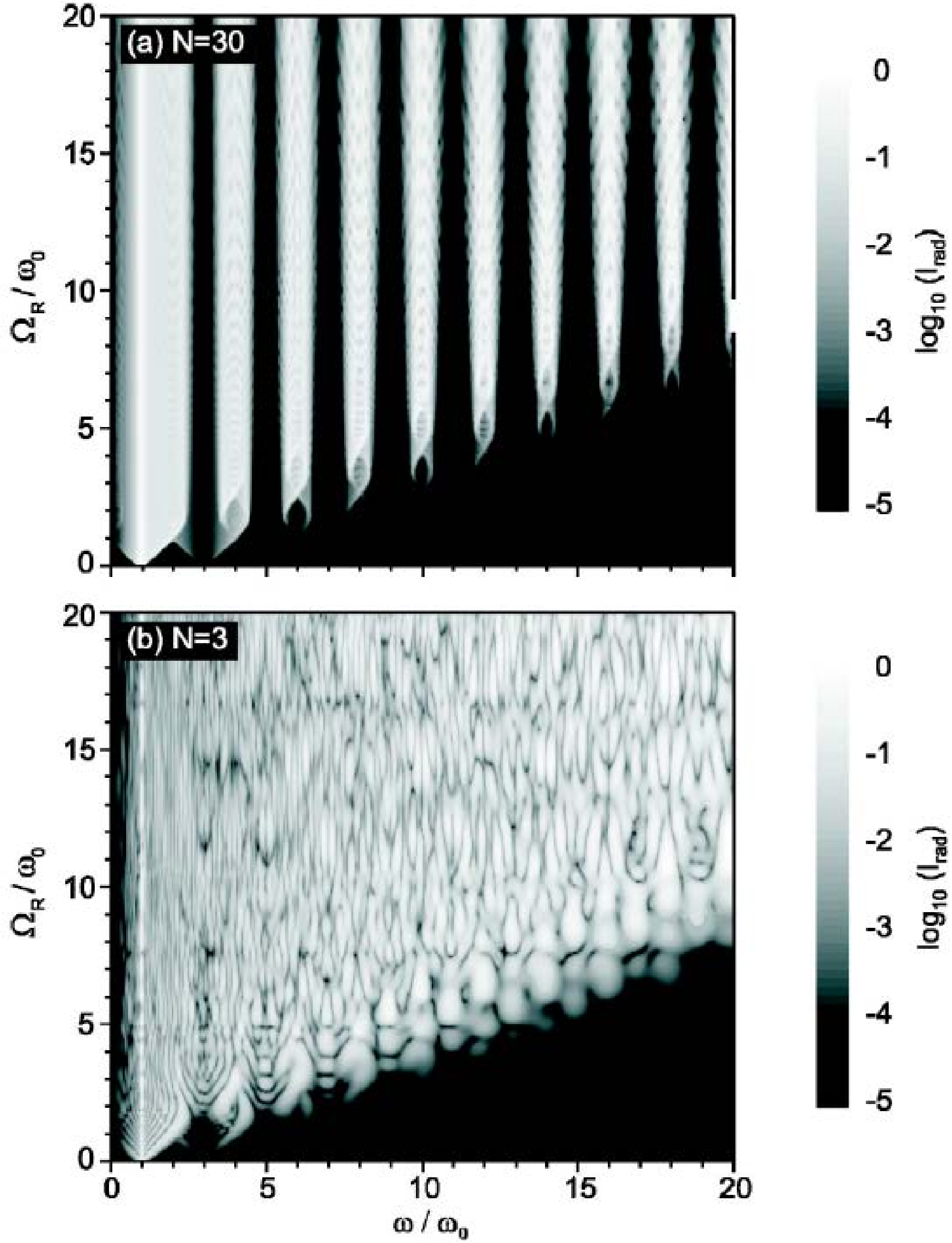


Figure 10.8: Same as Fig. 10.6(a), i.e., $\Omega/\omega_0 = 1$, but for Gaussian optical pulses with CEP $\phi = 0$ and with a FWHM of (a) $N = 30$ and (b) $N = 3$ optical cycles. [8]

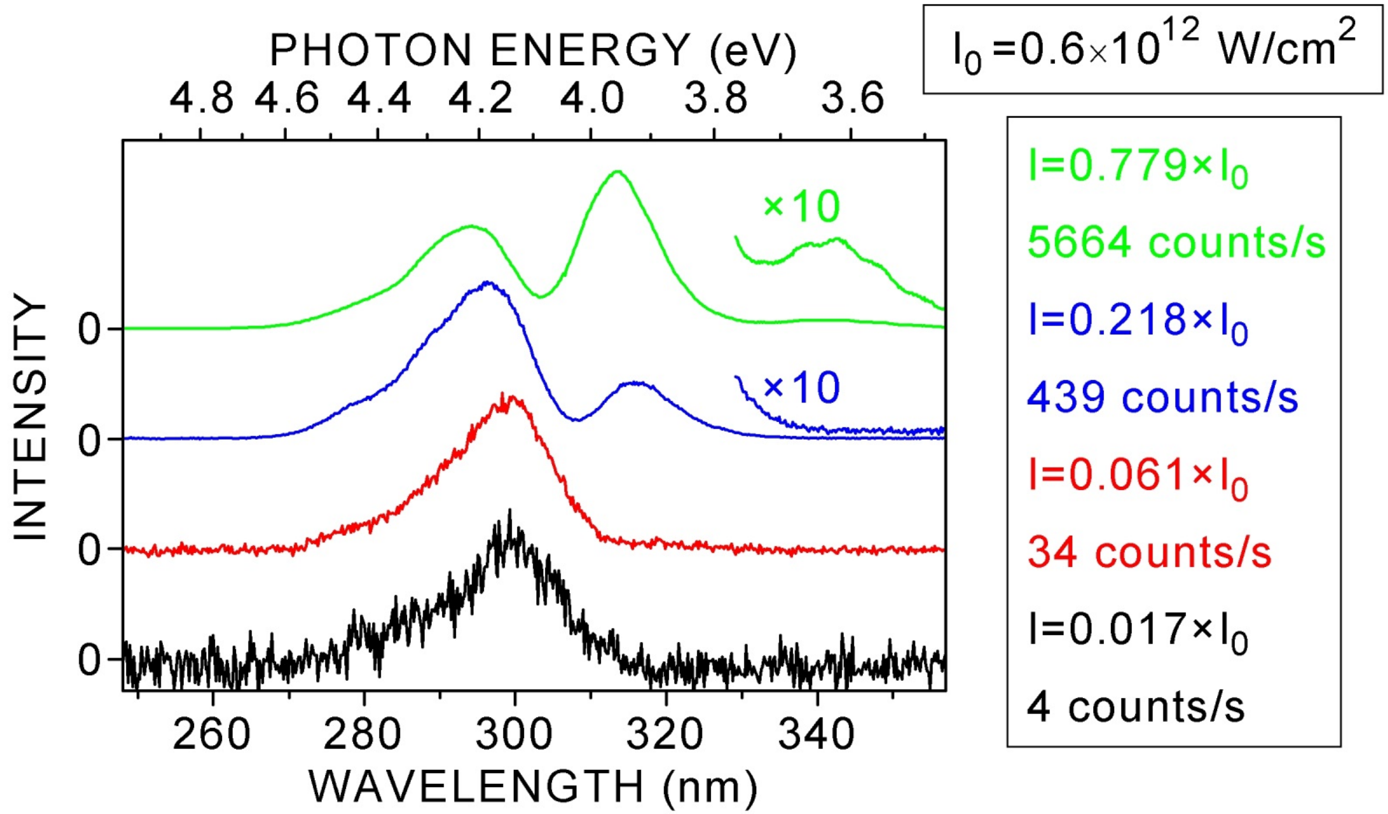
For Gaussian pulses, electric field envelope not constant in time

effectively averaging over vertical axis

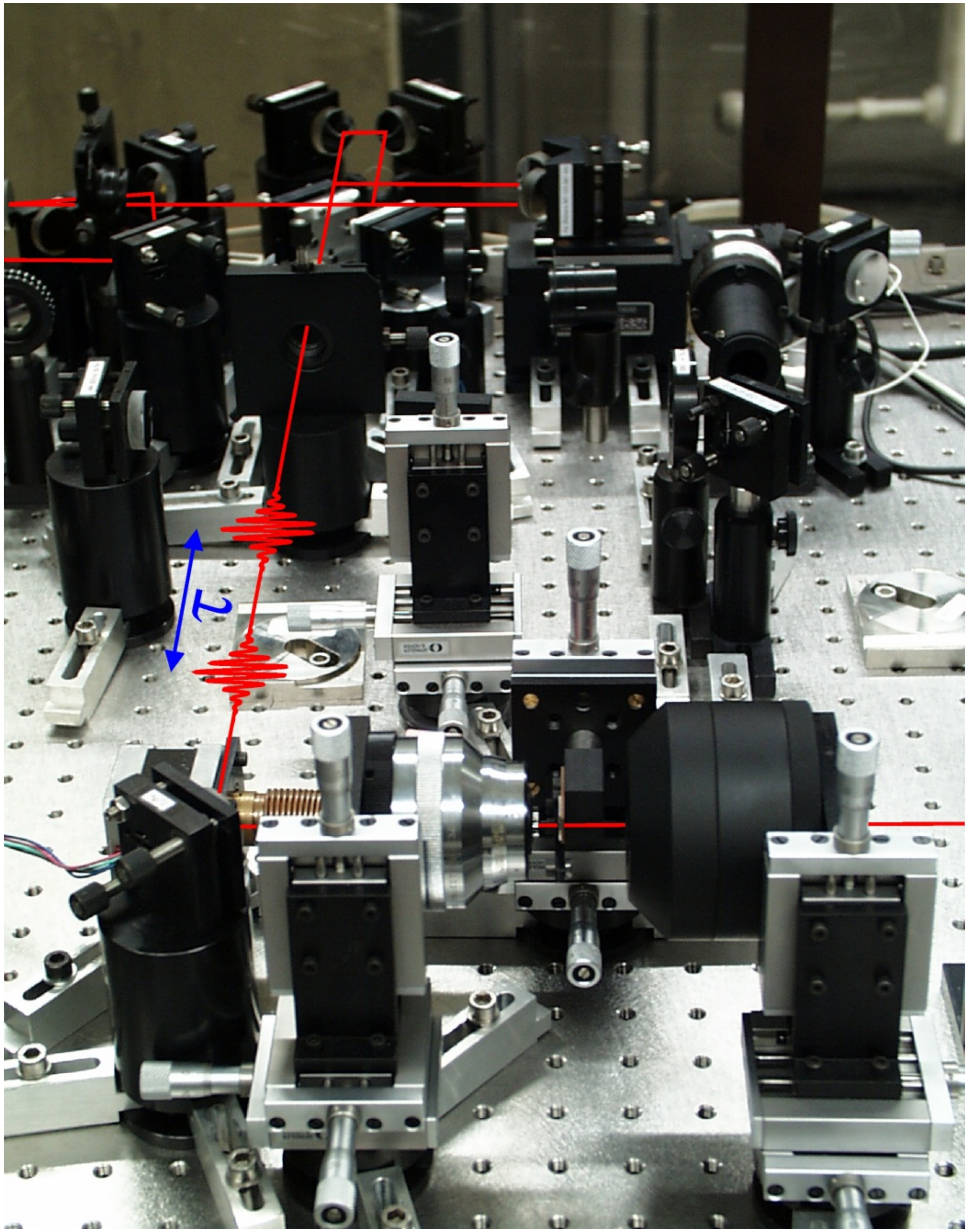
“messy” spectra

Experiment

GaAs / $Al_{0.3}Ga_{0.7}As$ double heterostructure (W. Stolz)



O. D. Mücke *et al.*, PRL **87**, 057401 (2001)
Q. T. Vu *et al.*, PRL **92**, 217403 (2004)



~5fs Ti:sapphire laser pulses

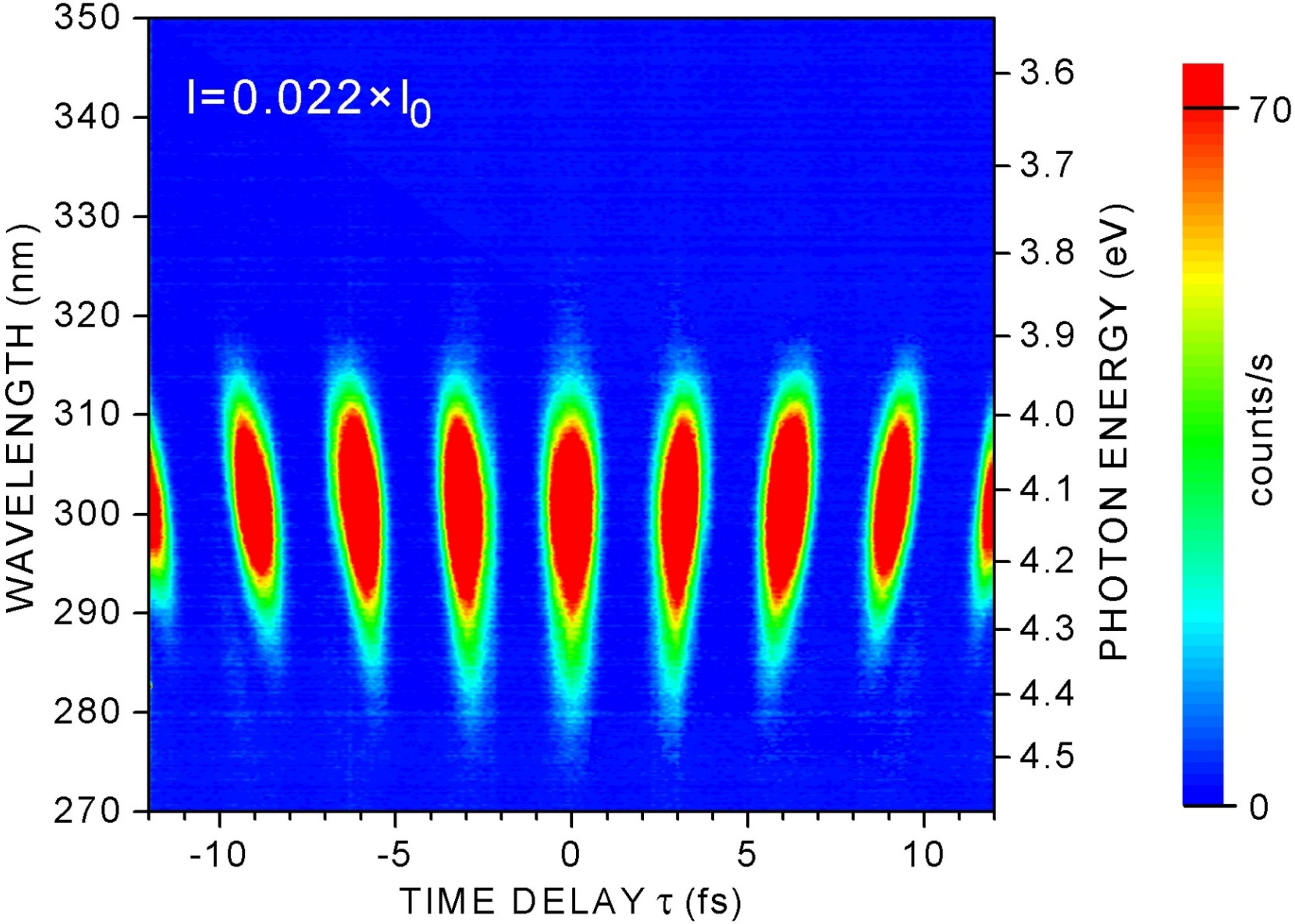
balanced Michelson interferometer is actively stabilized by a Pancharatnam screw [M. U. Wehner *et al.*, Opt. Lett. **22**, 1455 (1997)]

remaining fluctuations in time delay τ are <50 as

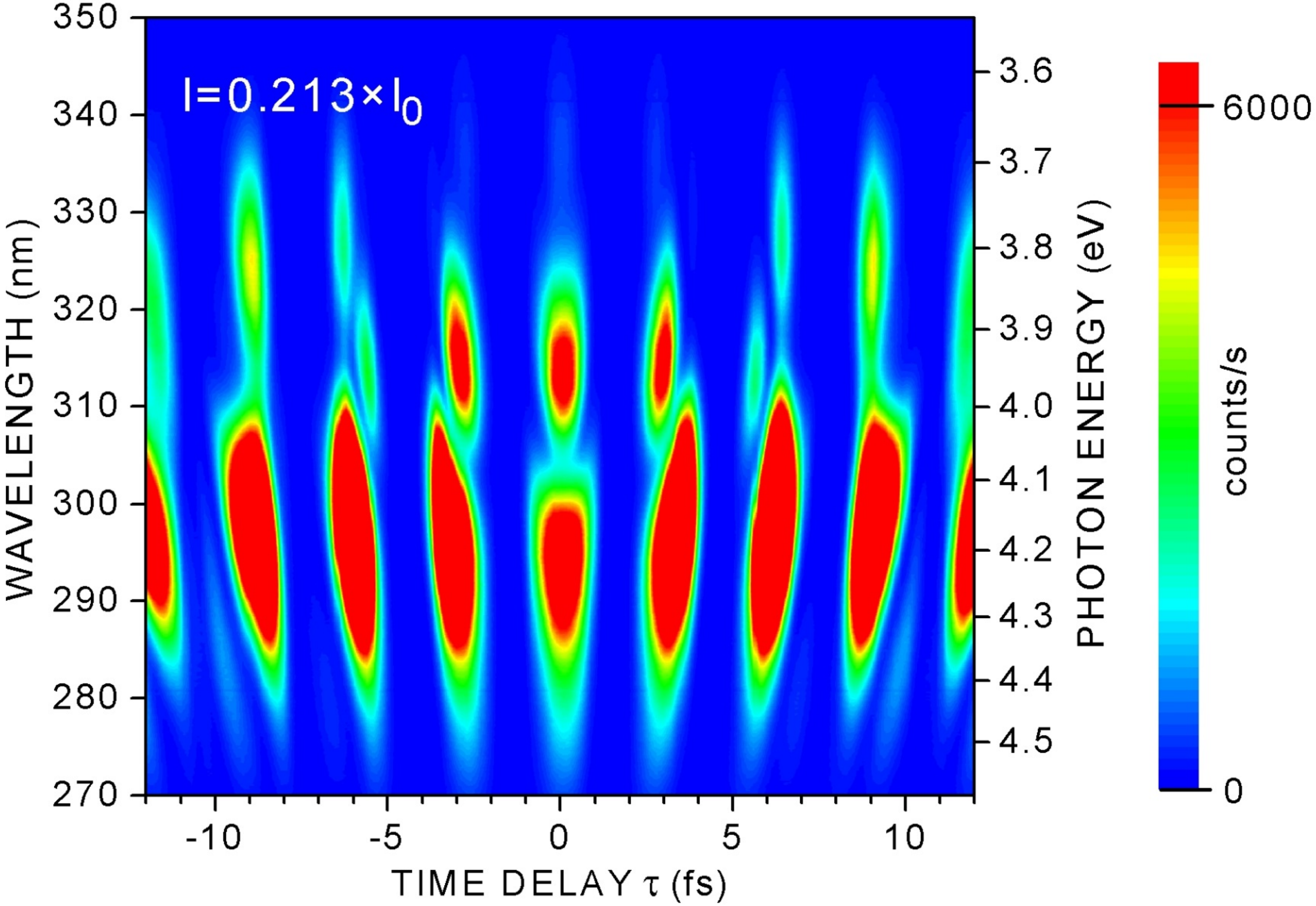
two reflective microscope objectives with NA=0.5 $\rightarrow$ 1 micron focus radius

Karlsruhe Institute of Technology
Wegener group, Applied Physics

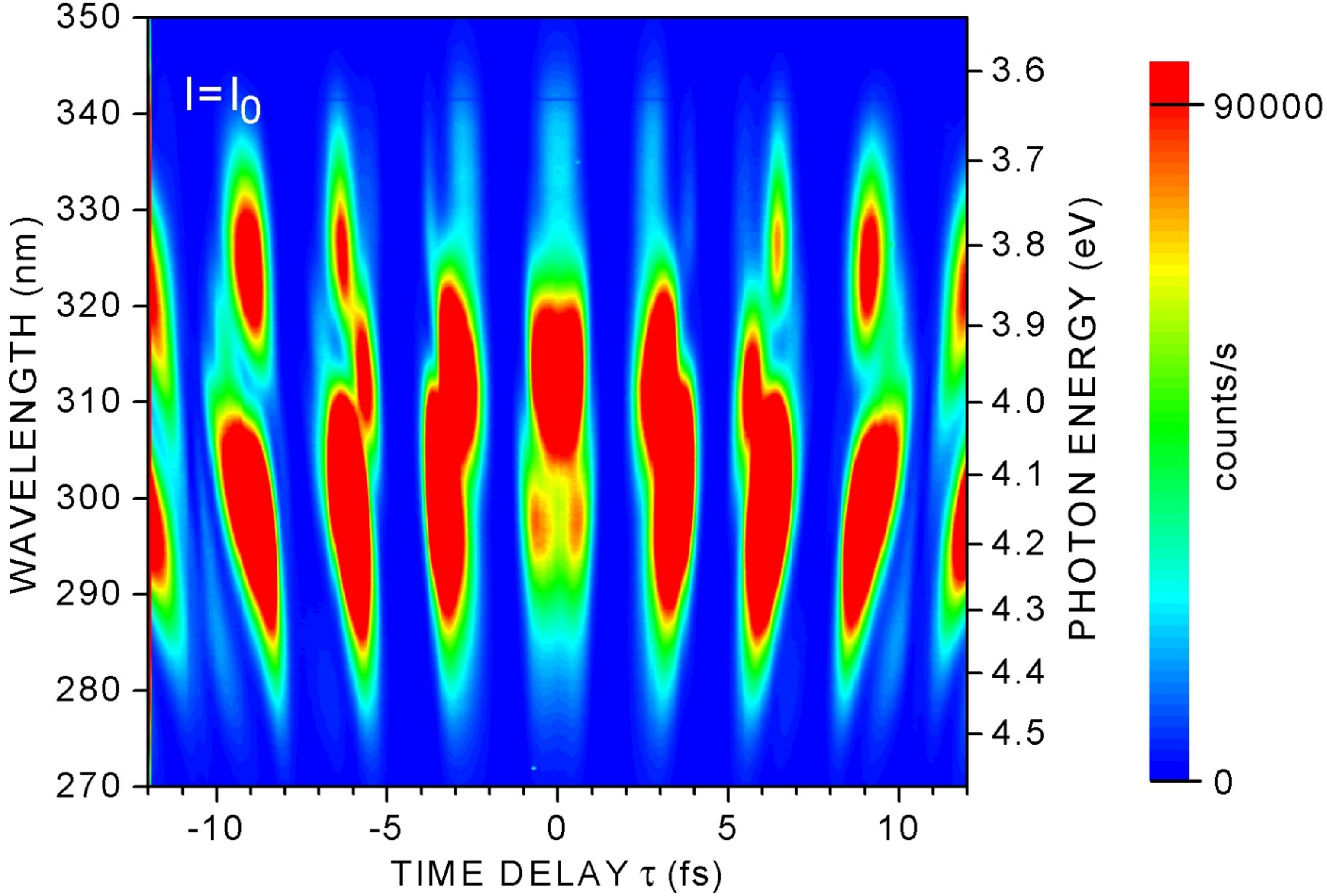
Interferometric measurements



Interferometric measurements



Interferometric measurements



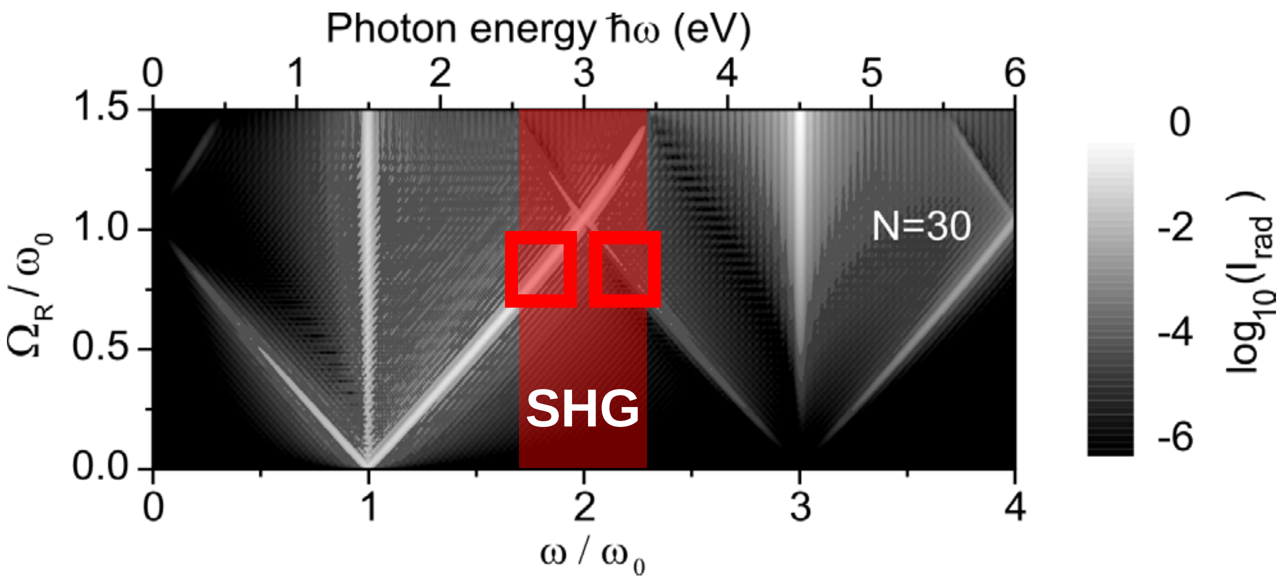
CEP dependence

fundamental / third-harmonic Mollow triplet: 1ϕ / 3ϕ

+

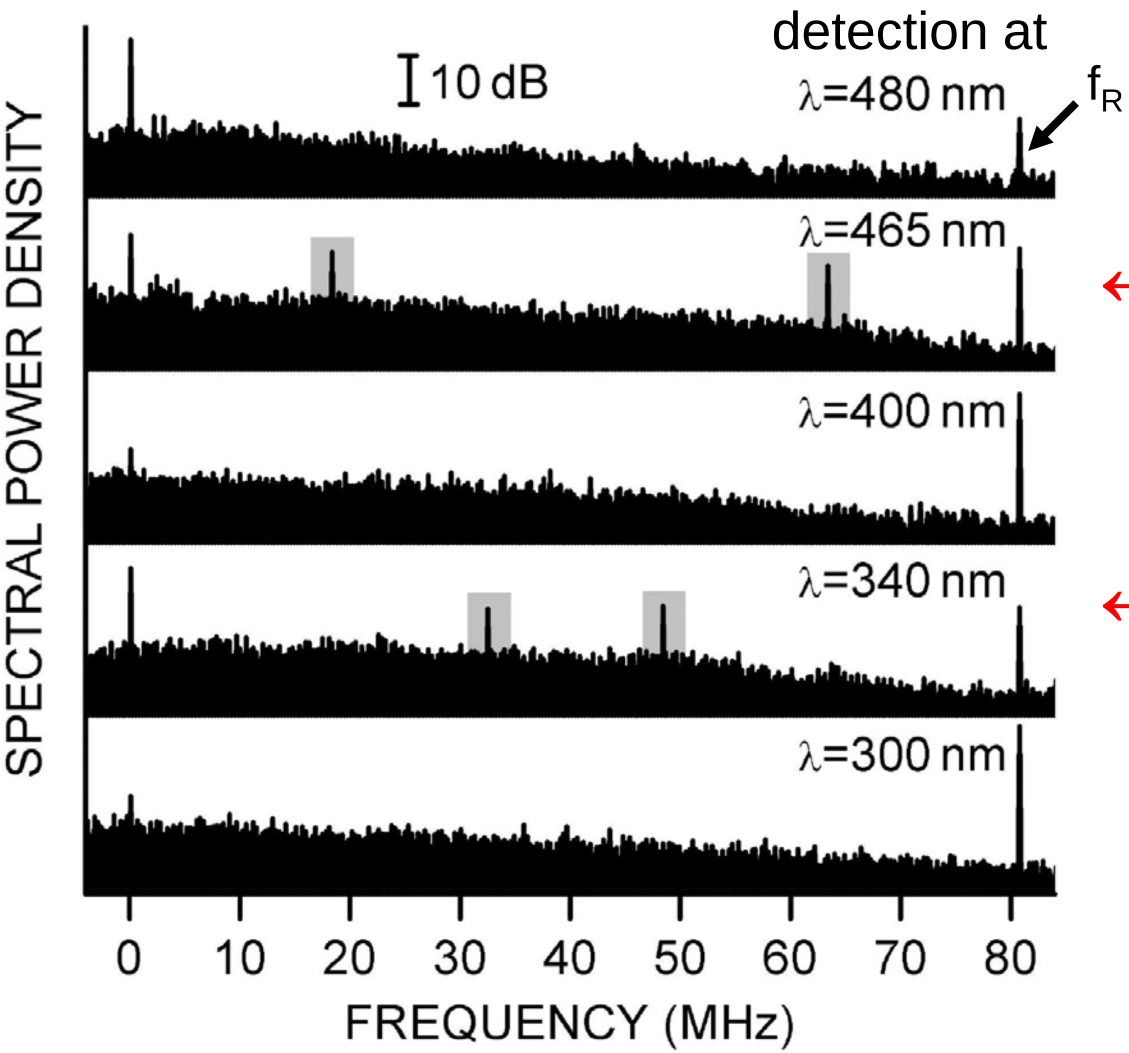
surface second-harmonic generation: 2ϕ

CE phase dependence: 1ϕ



O.D. Mücke *et al.*, PRL **89**, 127401 (2002)

Measuring the CEO frequency with GaAs



← f_{CEO} (SHG+fundamental)

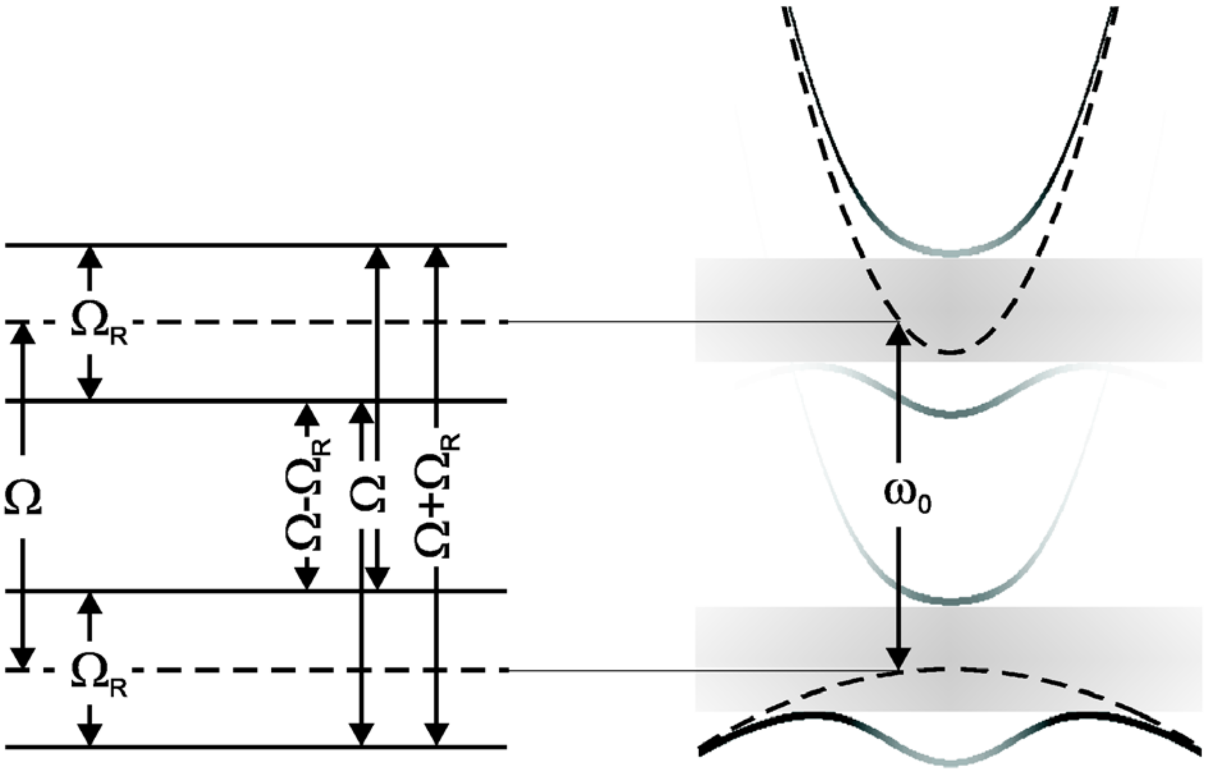
← f_{CEO} (SHG+THG)

O. D. Mücke *et al.*,
Opt. Lett. **29**, 2160 (2004)

Light-induced gaps in semiconductors

two-level system
Mollow triplet
B. R. Mollow (1969)

two-band semiconductor
light-induced gaps
V. F. Elesin (1971)



conduction band
+
1-photon sideband

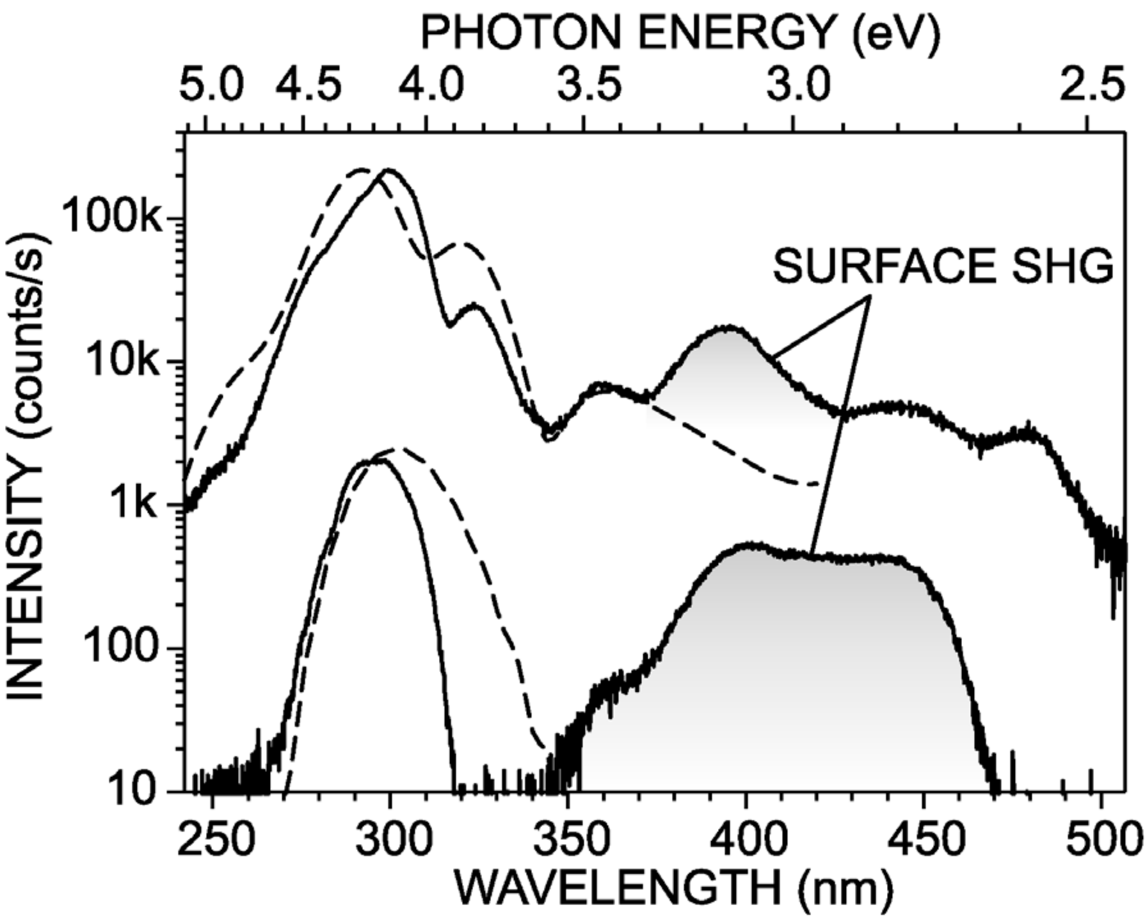
avoided crossing

light-induced gaps
triplet in third harmonic

Rabi splitting>damping

Q. T. Vu *et al.*, PRL **92**, 217403 (2004)

Light-induced gaps in semiconductors



Q. T. Vu *et al.*, PRL **92**, 217403 (2004)

theory (dashed curves):

- semiconductor Bloch equations
- full tight-binding bands
- density- and energy-dependent dephasing and relaxation
- no RWA

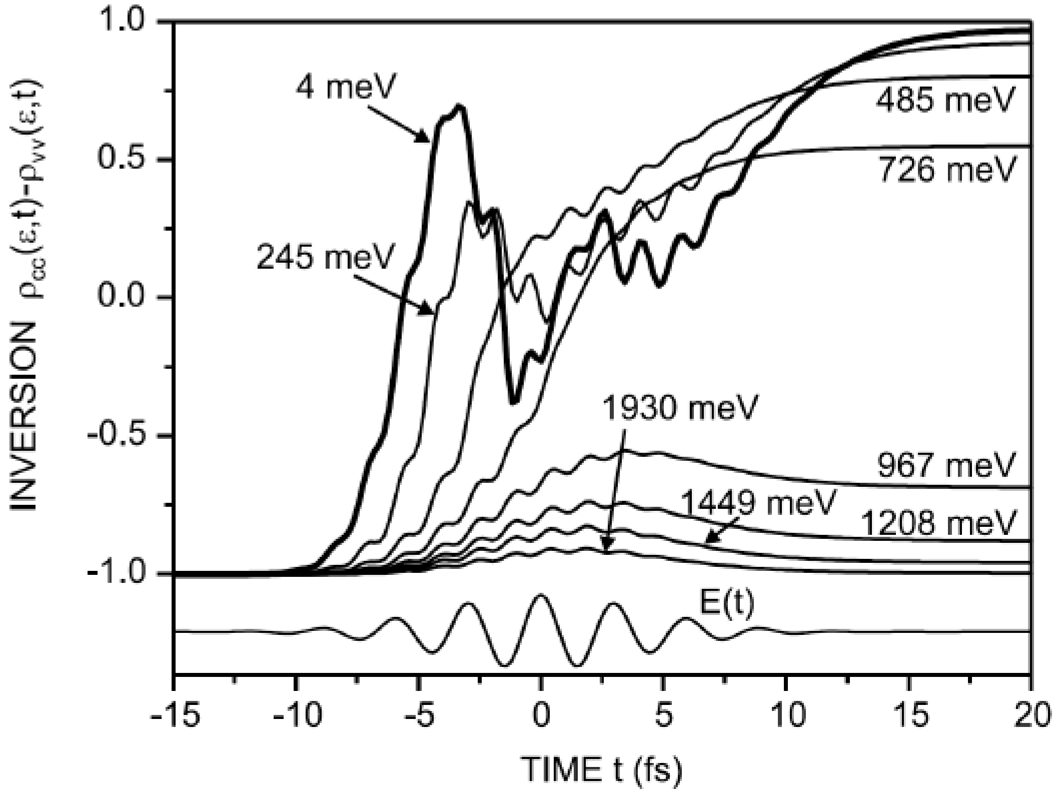
experiment (solid curves):

- 100nm thin GaAs film

high excitation
(upper curves)

low excitation
(lower curves)

Light-induced gaps in semiconductors

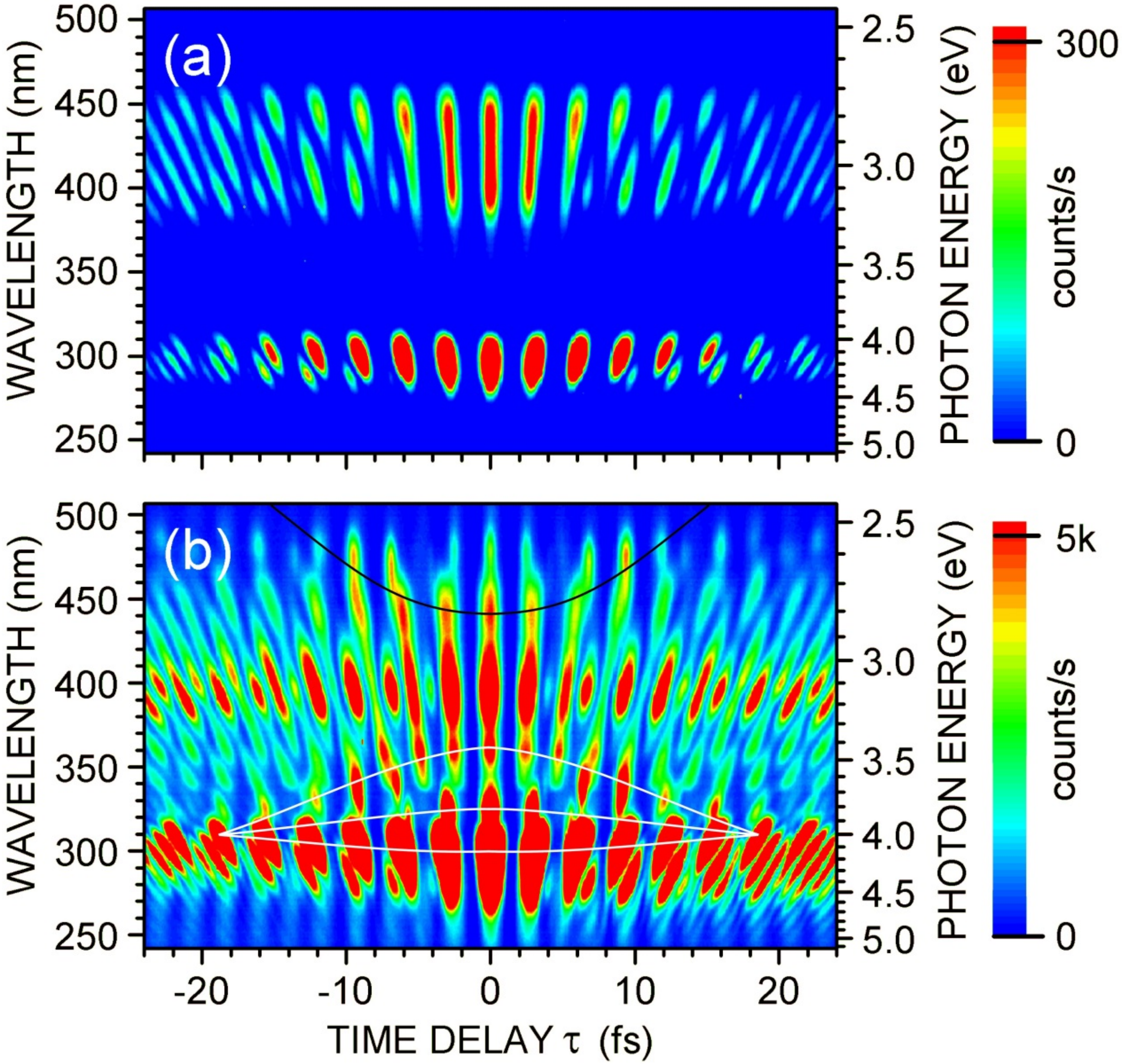


effect of carrier-density- and energy-dependent **dephasing** and **relaxation**

Figure 13.12: Computed inversion for various excess energies above the band gap versus time t for a peak electric field of 1.65×10^9 V/m. For $t = 20$ fs, the carrier density equals 1.1×10^{20} cm^3. The lower trace shows the laser field $E(t)$. [52]

Q. T. Vu *et al.*, PRL **92**, 217403 (2004)

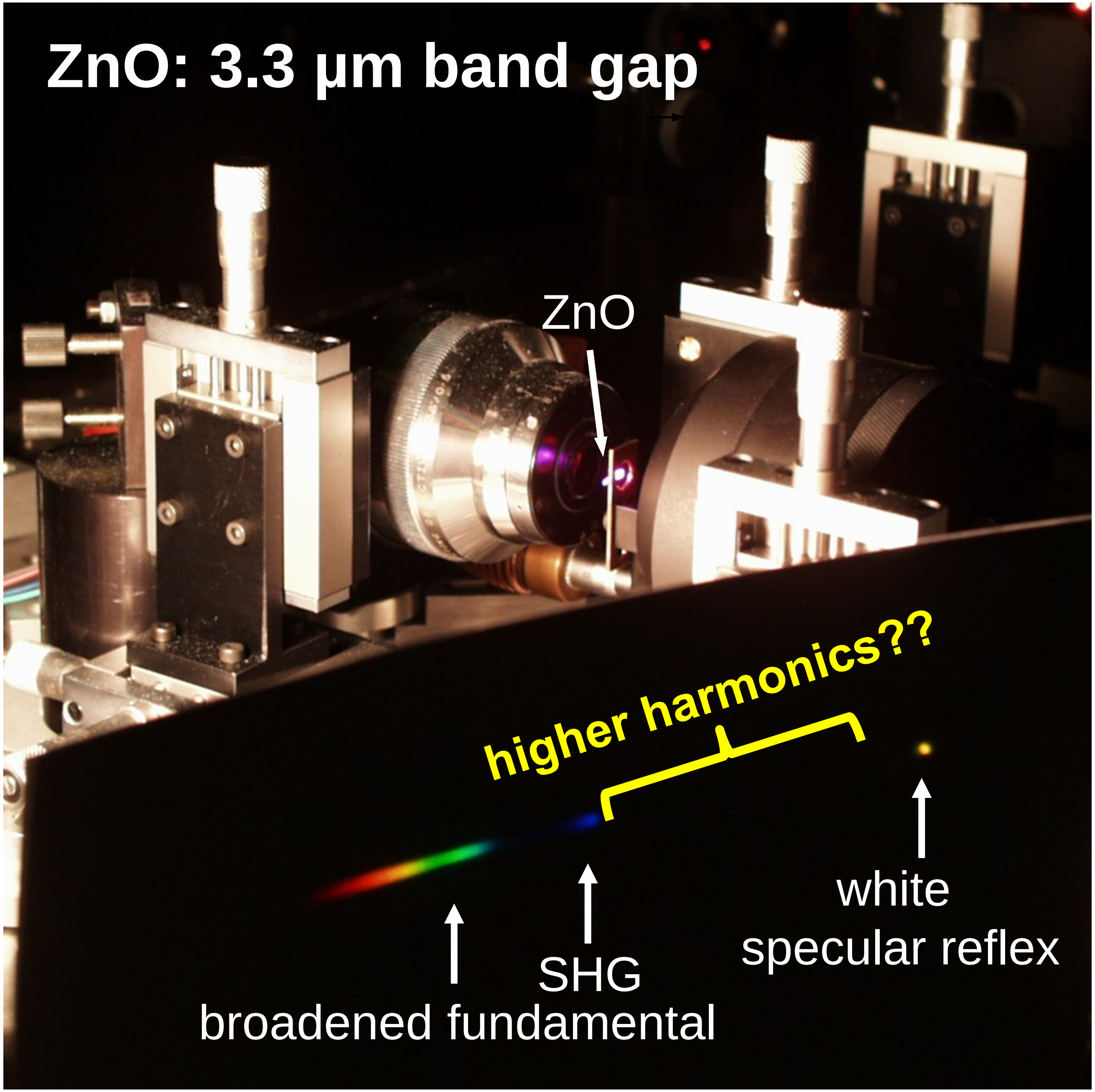
Light-induced gaps in semiconductors



low excitation

high excitation

O. D. Mücke *et al.*, OL **29**, 2160 (2004)



5-fs 800-nm pulses from Ti:sapphire oscillator

peak electric field

$E_0 = 6\mathrm{V/nm}$

Bloch energy

$\hbar\Omega_B = 3.0\mathrm{eV}$

Bloch period

$T_B = 1.4\mathrm{fs}$

optical period (800nm)

$T_L = 2.8\mathrm{fs}$

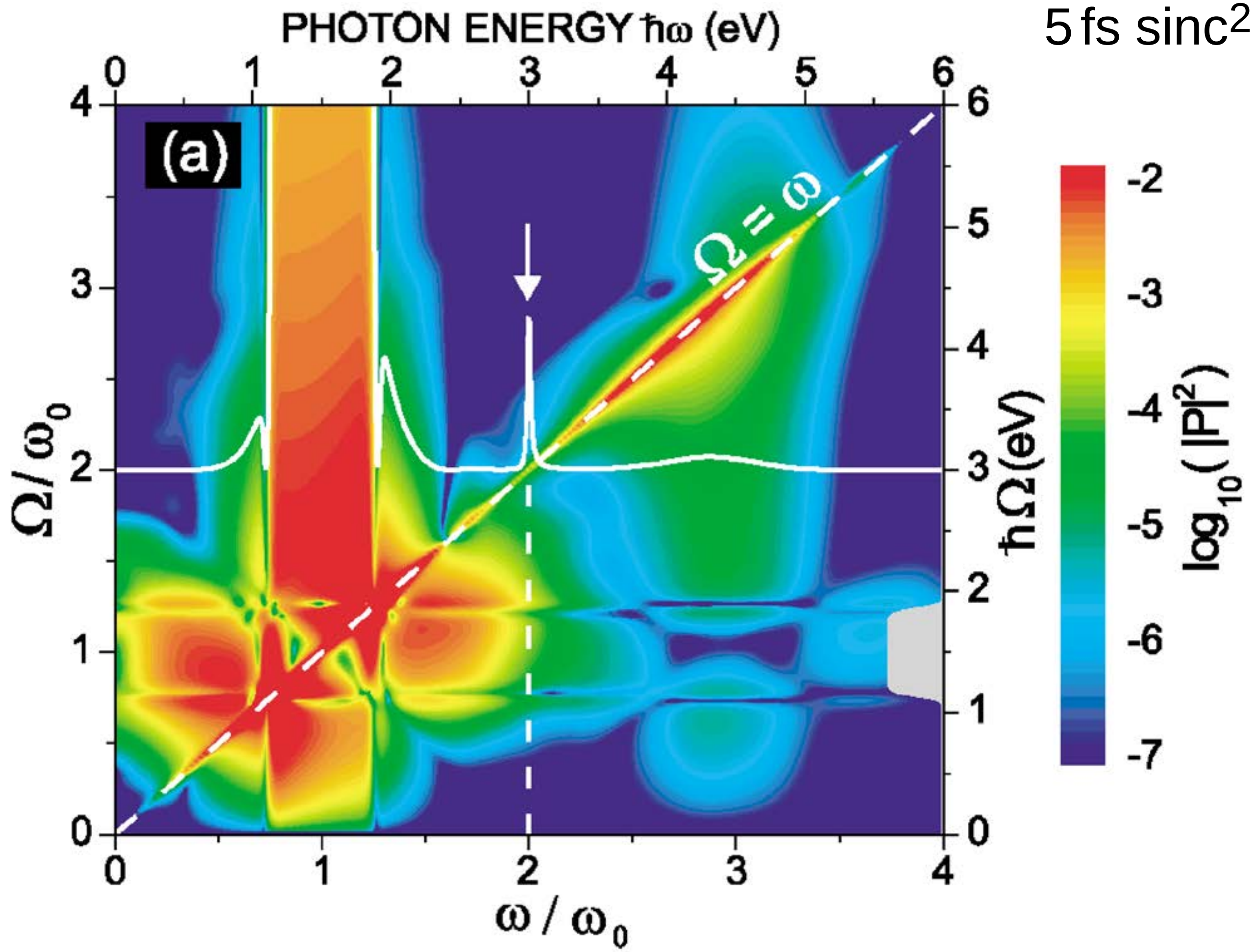
Bloch oscillations??

O. D. Mücke *et al.*,
Opt. Lett. **27**, 2127 (2002)

T. Tritschler *et al.*,
PRL **90**, 217404 (2003)

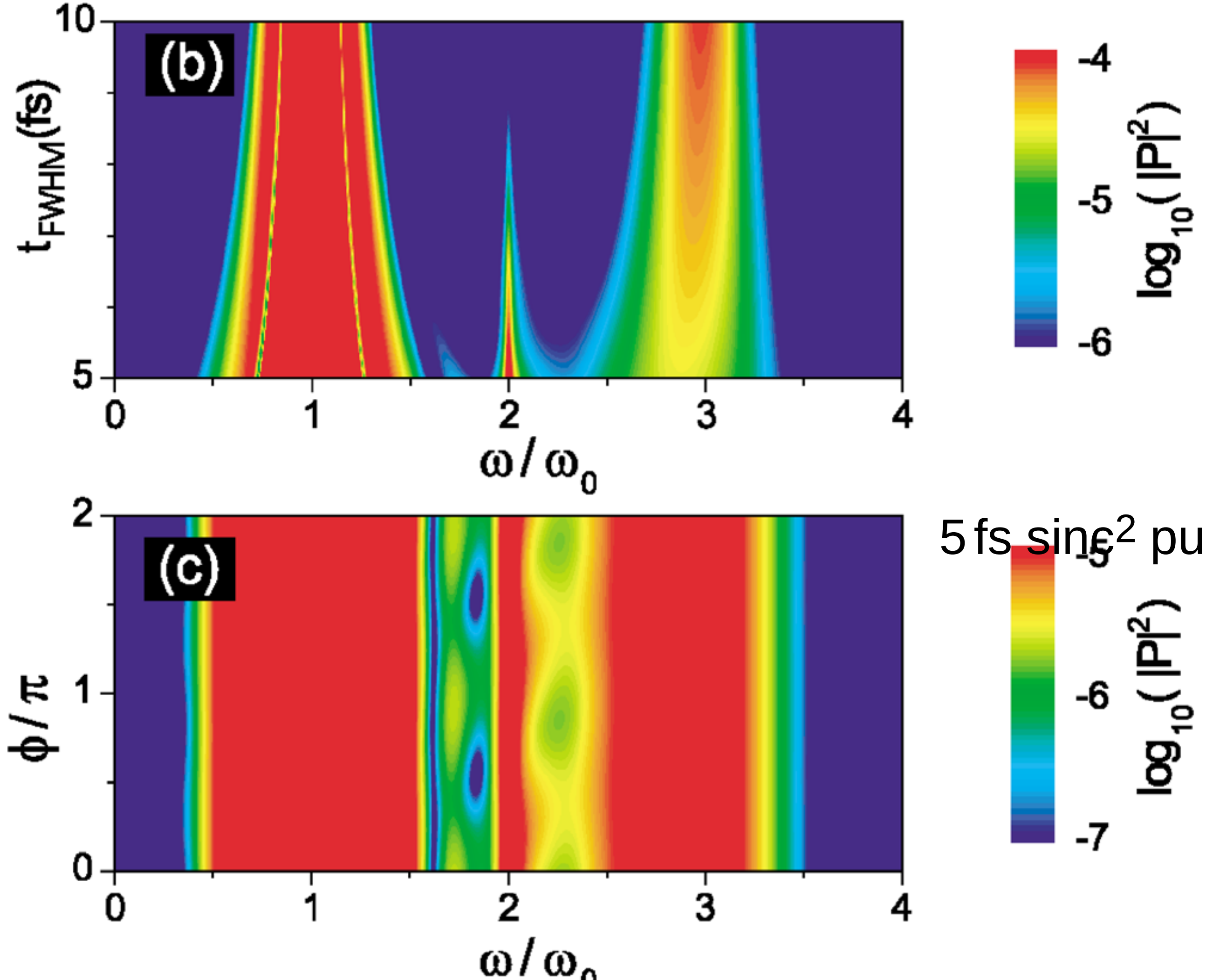
THG in disguise of SHG

5 fs sinc^2 pulses



T. Tritschler *et al.*, PRL **90**, 217404 (2003)
T. Tritschler *et al.*, PRA **68**, 033404 (2003)

THG in disguise of SHG



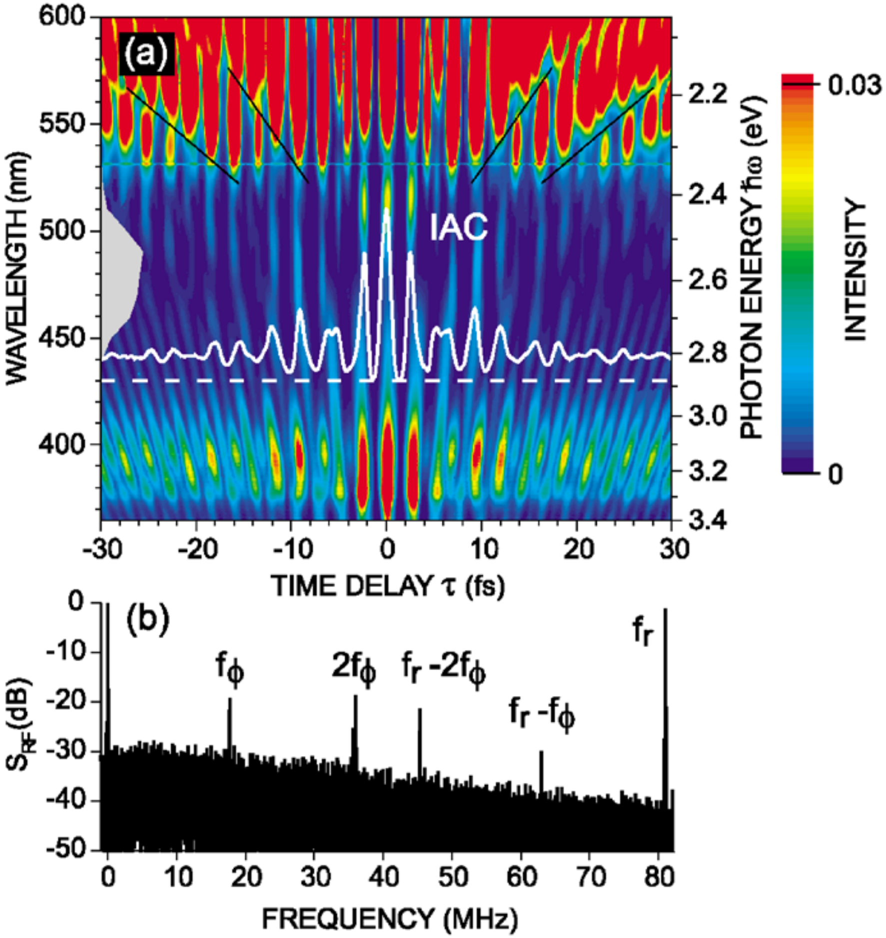
5 fs sinc^2 pulses

T. Tritschler *et al.*, PRL **90**, 217404 (2003)

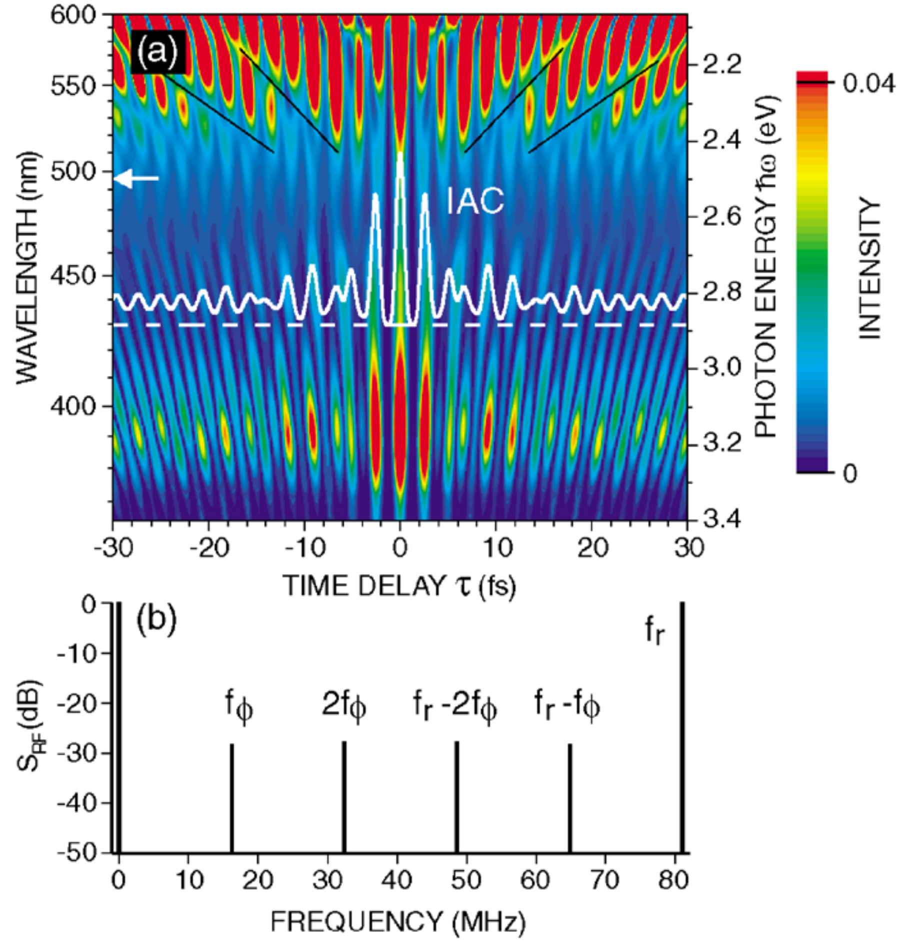
T. Tritschler *et al.*, PRA **68**, 033404 (2003)

THG in disguise of SHG

experiment



theory



Merry Xmas !!!