Nonlinear Optics (WiSe 2019/20)

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Chapter 10: Interactions of light and matter

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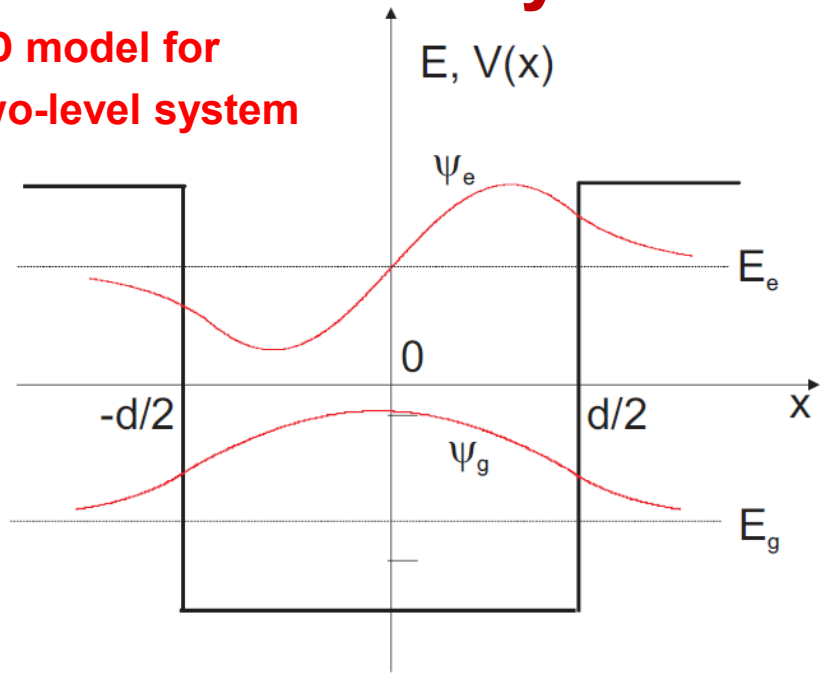
Chapter 10: Interactions of light and matter

10.1 Two-level systems

1D model for two-level system



$$\Psi(\vec{r},t) = c_g(t)\ \psi_g(\vec{r}) + c_e(t)\ \psi_e(\vec{r})$$

$$\begin{aligned} \mathbf{H}_A\ \psi_e(\vec{r}) &= E_e\ \psi_e(\vec{r}) \\ \mathbf{H}_A\ \psi_g(\vec{r}) &= E_g\ \psi_g(\vec{r}) \end{aligned}$$

time evolution from Schrödinger equation

$$\mathrm{j}\,\hbar\frac{\partial}{\partial t}\Psi\,(\vec{r},t) = \mathbf{H}_A\ \Psi\,(\vec{r},t)$$

$$\mathrm{j}\,\hbar\,(\dot{c}_g(t)\ \psi_g(\vec{r}) + \dot{c}_e(t)\ \psi_e(\vec{r})) = (E_g\ c_g(t)\ \psi_g(\vec{r}) + E_e\ c_e(t)\ \psi_e(\vec{r}))$$

by multiplication of this equation from the left with the complex conjugate ground-state or excited-state wavefunctions, integration over ***r***, and using the orthogonality relations for the energy eigenstates, we obtain two separate equations for the time dependence of the coefficients

$$\dot{c}_e = -\mathrm{j}\omega_e c_e, \text{ with } \omega_e = E_e/\hbar,$$
$$\dot{c}_g = -\mathrm{j}\omega_g c_g, \text{ with } \omega_g = E_g/\hbar.$$

This procedure is equivalent to projecting the Schrödinger equation onto the energy eigenstates.

time-dependent solution of the Schrödinger equation of the free atom

$$\Psi(\vec{r},t) = c_g(0)e^{-\mathrm{j}\omega_g t}\ \psi_g(\vec{r}) + c_e(0)e^{-\mathrm{j}\omega_e t}\ \psi_e(\vec{r})$$

How does the atomic dynamics change in the presence of an external electromagnetic (EM) field and environmental perturbations?

10.2 Atom-field interaction within the dipole approximation

induced dipole moment $\vec{d} = -e\vec{r}.$

Schrödinger equation for an atom in EM field $\mathbf{H}_{AF} = \mathbf{H}_A - \vec{d} \cdot \vec{E}(\vec{r}_A, t)$

new equations of motion contain matrix elements of dipole moment of atom

$$\vec{M}_{ee} = \int \psi_e^*(\vec{r})\, \vec{d}\, \psi_e(\vec{r})\, d\vec{r} = -e \int \psi_e^*(\vec{r})\, \vec{r}\, \psi_e(\vec{r}),$$

$$\vec{M}_{eg} = \int \psi_e^*(\vec{r})\, \vec{d}\, \psi_g(\vec{r})\, d\vec{r} = -e \int \psi_e^*(\vec{r})\, \vec{r}\, \psi_g(\vec{r}),$$

$$\vec{M}_{ge} = \int \psi_g^*(\vec{r})\, \vec{d}\, \psi_e(\vec{r})\, d\vec{r} = \vec{M}_{eg}^*,$$

$$\vec{M}_{gg} = \int \psi_g^*(\vec{r})\, \vec{d}\, \psi_g(\vec{r})\, d\vec{r} = -e \int \psi_g^*(\vec{r})\, \vec{r}\, \psi_g(\vec{r}).$$

0

0

symmetric/ antisymmetric atomic wavefunctions

new equations of motion for probability amplitudes

$$\dot{c}_e = -\mathrm{j}\omega_e c_e + \mathrm{j}c_g \frac{1}{\hbar}\left(\int \psi_e^*(\vec{r})\ \vec{d}\ \psi_g(\vec{r})\ d\vec{r}\right)\cdot\vec{E}(t), \tag{10.17}$$

$$\dot{c}_g = -\mathrm{j}\omega_g c_g + \mathrm{j}c_e \frac{1}{\hbar}\left(\int \psi_g^*(\vec{r})\ \vec{d}\ \psi_e(\vec{r})\ d\vec{r}\right)\cdot\vec{E}(t). \tag{10.18}$$

Separating the electric field into its polarization vector $\vec{e}$ and field strength $E(t)$

$$\vec{E}(t) = E(t)\ \vec{e}, \tag{10.19}$$

the Schrödinger equation becomes

$$\dot{c}_e = -\mathrm{j}\omega_e c_e + \mathrm{j}c_g \frac{\vec{M}_{eg}\cdot\vec{e}}{\hbar}E(t), \tag{10.20}$$

$$\dot{c}_g = -\mathrm{j}\omega_g c_g + \mathrm{j}c_e \frac{\vec{M}_{eg}^*\cdot\vec{e}}{\hbar}E(t). \tag{10.21}$$

The expectation value for the dipole moment of an atom in state (10.3) can also be expressed in terms of the dipole matrix elements

$$\begin{aligned}\left\langle\vec{d}\right\rangle &= |c_e|^2\vec{M}_{ee} + |c_g|^2\vec{M}_{gg} + c_e^*c_g\vec{M}_{eg} + c_g^*c_e\vec{M}_{ge}\\ &= c_e^*c_g\vec{M}_{eg} + c.c.,\end{aligned} \tag{10.22}$$

atom only has dipole moment if in superposition of energy eigenstates

monochromatic field

$$E(t) = \frac{1}{2}\left(\underline{E}_0 e^{\mathrm{j}\omega t} + \underline{E}_0^* e^{-\mathrm{j}\omega t}\right), \tag{10.23}$$

where $\underline{E}_0$ is the complex electric field amplitude. We expect strong interaction between the field and the atom if the atomic transition frequency between the states, $\omega_{eg} = \omega_e - \omega_g$, is close to the frequency of the driving field, i.e., $\omega_{eg} \approx \omega$. It is advantageous to transform to new probability amplitudes, that take some trivial oscillations already into account

$$C_e = c_e\, e^{\mathrm{j}\left(\frac{\omega_e+\omega_g+\omega}{2}t\right)} \tag{10.24}$$

$$C_g = c_g\, e^{\mathrm{j}\left(\frac{\omega_e+\omega_g-\omega}{2}t\right)}, \tag{10.25}$$

which leads to the new equations of motion

$$\dot{C}_e = \left[\mathrm{j}\left(\frac{\omega_e+\omega_g+\omega}{2}\right) - \mathrm{j}\omega_e\right] c_e e^{\mathrm{j}\left(\frac{\omega_e+\omega_g+\omega}{2}t\right)} + \mathrm{j}c_g \frac{\vec{M}_{eg}\cdot\vec{e}}{\hbar}\vec{E}(t)\, e^{\mathrm{j}\left(\frac{\omega_e+\omega_g+\omega}{2}t\right)},$$

$$\dot{C}_g = \left[\mathrm{j}\left(\frac{\omega_e+\omega_g-\omega}{2}\right) - \mathrm{j}\omega_g\right] c_g e^{\mathrm{j}\left(\frac{\omega_e+\omega_g-\omega}{2}t\right)} + \mathrm{j}c_e \frac{\vec{M}_{eg}^*\cdot\vec{e}}{\hbar}\vec{E}(t)\, e^{\mathrm{j}\left(\frac{\omega_e+\omega_g-\omega}{2}t\right)}.$$

Introducing the detuning between the atomic transition and the electric field frequencies

$$\Delta = \frac{\omega_{eg} - \omega}{2} \tag{10.26}$$

and the Rabi frequency

$$\Omega_r = \frac{\vec{M}_{eg}^* \cdot \vec{e}}{\hbar} \left(\underline{E}_0 + \underline{E}_0^* e^{-\mathrm{j}2\omega t} \right), \tag{10.27}$$

we obtain the following coupled-mode equations for the probability amplitudes

$$\frac{d}{dt} C_e = -\mathrm{j}\Delta C_e + \mathrm{j}\frac{\Omega_r^*}{2} C_g \tag{10.28}$$

$$\frac{d}{dt} C_g = +\mathrm{j}\Delta C_g + \mathrm{j}\frac{\Omega_r}{2} C_e. \tag{10.29}$$

If the Rabi frequency is small compared to the optical transition $|\Omega_r| \ll \omega_{eg} \approx \omega$, the so-called Rotating-Wave Approximation (RWA) [3] can be made, where we only keep the slowly varying components in the interaction, i.e.,

$$\Omega_r \approx \frac{\vec{M}_{eg}^* \cdot \vec{e}}{\hbar} \underline{E}_0 = const. \tag{10.30}$$

10.3 Rabi oscillations

resonant excitation

detuning Δ=0

$$\frac{d^2}{dt^2}C_e = -\frac{|\Omega_r|^2}{4}C_e \tag{10.31}$$

$$\frac{d^2}{dt^2}C_g = -\frac{|\Omega_r|^2}{4}C_g. \tag{10.32}$$

The solution to this set of equations are oscillations. If the atom is initially at time $t = 0$ in the ground state, i.e., $C_g(0) = 1$ and $C_e(0) = 0$, we arrive at

$$C_g(t) = \cos\left(\frac{|\Omega_r|}{2}t\right) \tag{10.33}$$

$$C_e(t) = -\mathrm{j}\sin\left(\frac{|\Omega_r|}{2}t\right). \tag{10.34}$$

Then, the probabilities for finding the atom in the ground or excited states are

$$|c_g(t)|^2 = \cos^2\left(\frac{|\Omega_r|}{2}t\right) \tag{10.35}$$

$$|c_e(t)|^2 = \sin^2\left(\frac{|\Omega_r|}{2}t\right), \tag{10.36}$$

as illustrated in Fig. 10.2. For the expectation value of the dipole operator under the assumption of a real dipole matrix element $\vec{M}_{eg} = \vec{M}^*_{eg}$, we obtain

$$\left\langle \vec{d} \right\rangle = \vec{M}_{eg}c_e c_g^* + c.c. \tag{10.37}$$

$$= -\vec{M}_{eg}\sin\left(|\Omega_r|\,t\right)\sin\left(\omega_{eg}t\right). \tag{10.38}$$

resonant excitation

detuning Δ=0

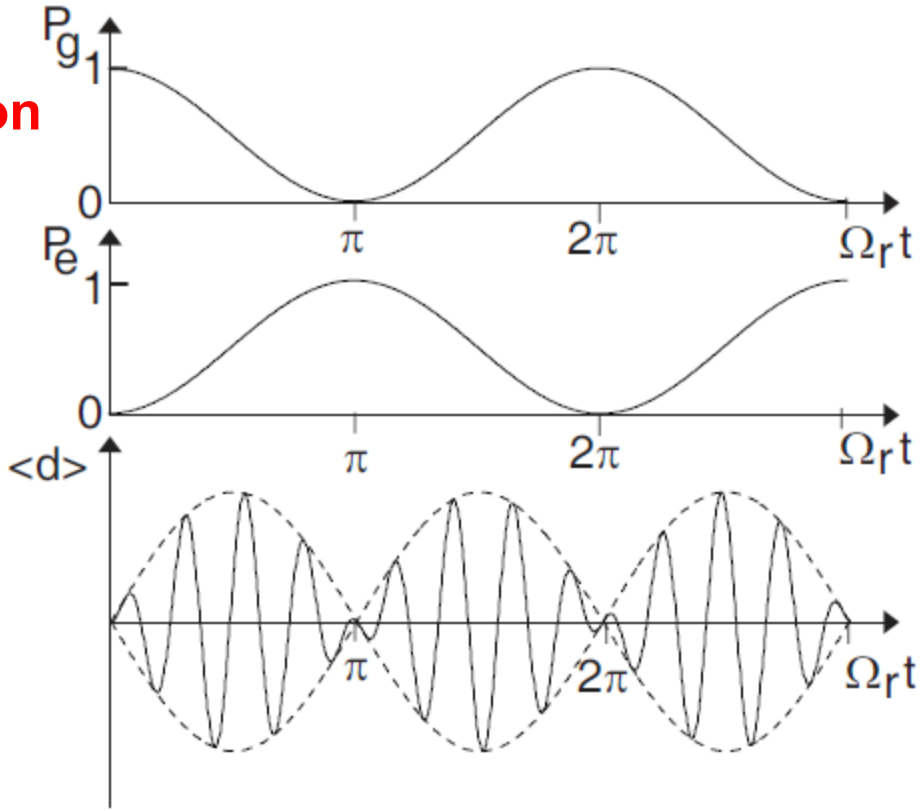


Figure 10.2: Evolution of occupation probabilities of ground and excited state and the average dipole moment of a two-level atom in resonant interaction with a coherent classical field.

population inversion *w* $w = P_e - P_g = |c_e|^2 - |c_g|^2$

Mollow sidebands $\omega_\pm = \omega_{eg} \pm \Omega_r$

loss of coherence in the atomic system due to additional interactions of the atom with its environment

dissipative processes can not easily be included in Schrödinger equation formalism

dissipative quantum systems:

open quantum systems = quantum system coupled to bath

Here: include **relaxation and dephasing** phenomenologically into equations of motion

From the equations of motion for the coefficients of the wave function, Eqs. (10.28) and (10.29), we derive equations of motion for the complex slowly varying dipole moment defined as

$$\underline{d} = c_e^* c_g e^{-\mathrm{j}\omega t} = C_e^* C_g. \tag{10.40}$$

By applying the product rule, we find

$$\begin{aligned}
\frac{d}{dt}\underline{d} &= \left(\frac{d}{dt}C_e^*\right) C_g + C_e^* \left(\frac{d}{dt}C_g\right) && (10.41)\\
&= \mathrm{j}\Delta C_e^* C_g - \mathrm{j}\frac{\Omega_r}{2} C_g^* C_g + \mathrm{j}\Delta C_e^* C_g + \mathrm{j}\frac{\Omega_r}{2} C_e^* C_e && (10.42)\\
&= \mathrm{j}\Delta\underline{d} + \mathrm{j}\frac{\Omega_r}{2}\cdot w && (10.43)
\end{aligned}$$

and

$$\begin{aligned}
\frac{d}{dt}w &= \left(\frac{d}{dt}C_e\right) C_e^* - \left(\frac{d}{dt}C_g\right) C_g^* + c.c. && (10.44)\\
&= \left(-\mathrm{j}\Delta C_e C_e^* + \mathrm{j}\frac{\Omega_r^*}{2} C_g C_e^* - \mathrm{j}\Delta C_g C_g^* - \mathrm{j}\frac{\Omega_r}{2} C_e C_g^*\right) + c.c. && (10.45)\\
&= +\mathrm{j}\Omega_r^*\underline{d} + c.c && (10.46)
\end{aligned}$$

For the monochromatic wave of Eq. (10.23), we find for the dynamics of a two-level system interacting with a coherent driving field with Rabi frequency Ω_r

$$\frac{d}{dt}\underline{d} = \mathrm{j}\Delta\underline{d} + \mathrm{j}\frac{\Omega_r}{2}\cdot w \tag{10.47}$$

$$\frac{d}{dt}w = +\mathrm{j}\Omega_r^*\underline{d} + c.c. \tag{10.48}$$

10.4 Energy and phase relaxation

difficult to completely isolate atom from its environment due to interaction with

- electric field from all the free-space modes of surrounding EM field
- phonons in solids

random interaction leads to a **thermalization and decoherence** of atom

Example:

To provide an example for the interaction of an atom with its environment in thermal equilibrium, we consider the interaction of a two-level system with the free-space EM field, that is in thermal equilibrium with temperature *T*

the electric field amplitude in the Bloch equations (10.47) and (10.48) is a random quantity and represents the field of the black-body radiation

$$\Omega_{bbr}(t) = \frac{\vec{M}_{eg}^{*} \cdot \vec{e}_{bb}}{\hbar} \underline{E}_{bb}(t), \qquad (10.49)$$

$\underline{E}_{bb}(t)$ is the random field and $\vec{e}_{bb}$ the random polarization

analysis yields

$$\frac{d}{dt}w = -\frac{1}{T_1}w(t). \qquad (10.58)$$

$$\text{with } \frac{1}{T_1} = \frac{\left|\vec{M}_{eg}\right|^2}{3\hbar^2}\frac{2\omega_{eg}^3\hbar}{\pi c^3\epsilon}\left(n_{th}(\omega_{eg})+\frac{1}{2}\right), \qquad (10.52)$$

$$\text{and } n_{th}(\omega_{eg}) = 1/(\exp(\hbar\omega_{eg}/kT)-1). \qquad (10.53)$$

random field fluctuations of the EM vacuum lead to an **exponential decay of population inversion**

This result can easily be interpreted: The first factor $\frac{\left|\vec{M}_{eg}\right|^2}{3\hbar}$ comes from the average of the projection of the dipole matrix element onto a unit vector, when averaged over every possible polarization direction. The second factor $\frac{4\omega_{eg}^3\hbar}{\pi c^3\epsilon}\left(n_{th}(\omega_{eg})+\frac{1}{2}\right)$ originates from expressing the power spectral density of the electric field amplitudes at the transition frequency $\omega \approx \omega_{eg}$ by the spectral energy density of the black-body radiation. However, we did not only include in the energy the part due to the thermal photon population of the mode, but also its ground-state energy $\hbar\omega_{eg}/2$. Thus even at temperature $T \to 0$, $\frac{1}{T_1}$ stays finite. The white noise property helps us to find an equation of motion

Similarly the analysis yields

$$\frac{d}{dt}\underline{d} = \mathrm{j}\Delta\underline{d} - \frac{1}{2T_1}\underline{d}. \qquad (10.59)$$

this equation describes now the average dipole moment in an ensemble of identical atoms

relaxation to steady-state inversion

$$\frac{d}{dt}w = -\frac{w - w_0}{T_1}. \qquad (10.60)$$

comparison with phenomenological discussion of how thermal equilibrium between thermal radiation and a two-level system is reached using

Einstein's A and B coefficients

$$\Gamma_e = \frac{1}{\tau_{sp}}(n_{th}+1)$$
$$\Gamma_a = \frac{1}{\tau_{sp}}n_{th},$$

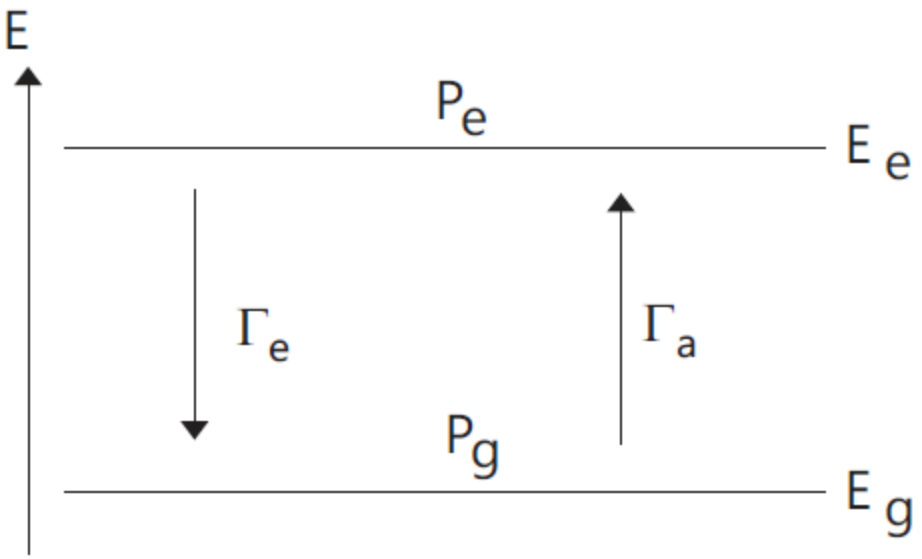


Figure 10.3: Two-level atom with transition rates due to induced and spontaneous emission and absorption.

$$\frac{dP_g}{dt} = -\frac{dP_e}{dt} = \frac{1}{\tau_{sp}} \left[(P_e - P_g)\, n_{th} + P_e \right] \tag{10.62}$$

or rewritten as

$$\frac{dP_g}{dt} = -\frac{dP_e}{dt} = \Gamma_e P_e - \Gamma_a P_g \,. \tag{10.63}$$

with abbreviations

$$\Gamma_e = \frac{1}{\tau_{sp}} (n_{th} + 1), \tag{10.64}$$

$$\Gamma_a = \frac{1}{\tau_{sp}} n_{th}, \tag{10.65}$$

see Fig. 10.3. For the inversion, we then obtain

$$\frac{d}{dt} w = \frac{d}{dt} P_e - \frac{d}{dt} P_g = \frac{-2}{\tau_{sp}} \left[(P_e - P_g)\, n_{th} + P_e \right] \tag{10.66}$$

$$= -\frac{2}{\tau_{sp}} \left[w n_{th} + \frac{w+1}{2} \right] = -\frac{2n_{th}+1}{\tau_{sp}} \left[w + \frac{1}{2n_{th}+1} \right]. \tag{10.67}$$

Note, here we used that $P_e + P_g = 1$ and thus $P_e = \frac{w+1}{2}$. Comparing coefficients between Eqs. (10.60) and (10.67), we find

$$\frac{1}{T_1} = \frac{2n_{th}+1}{\tau_{sp}} = \Gamma_e + \Gamma_a \tag{10.68}$$

$$w_0 = \frac{\Gamma_a - \Gamma_e}{\Gamma_a + \Gamma_e} = \frac{-1}{2n_{th}+1} = -\tanh\left(\frac{\hbar \omega_{eg}}{2kT} \right). \tag{10.69}$$

For zero temperature, the decay time T_1 approaches the spontaneous lifetime of the atom due to the zero-point fluctuations of the electromagnetic field

$$\frac{1}{T_1} = \frac{1}{\tau_{sp}} = \frac{\left|\vec{M}_{eg}\right|^2 \omega_{eg}^3}{3\pi\hbar c^3 \epsilon}.$$

This is an expression for the spontaneous lifetime of an atom in terms of the dipole matrix element and the density of modes in the electromagnetic field at the transition frequency ω_{eg}.

In summary, the equation for the dipole moment $\underline{d}$ and the inversion w due to its interaction with the environment can be written as

$$\dot{\underline{d}} = \left(\mathrm{j}\,(\omega_{eg} - \omega) - \frac{1}{T_2}\right)\underline{d} \tag{10.70}$$

$$\dot{w} = -\frac{w - w_0}{T_1}. \tag{10.71}$$

The time constant T_1 denotes the energy relaxation in the two-level system and T_2 the phase relaxation. T_2 is the correlation time between amplitudes c_e and c_g. The coherence between the excited and ground states described by the dipole moment is destroyed by the interaction of the two-level system with the environment.

T_1 energy relaxation time
T_2 dephasing time

in general need to be computed from the scattering processes involved, in solids, e.g., electron-electron, electron-phonon scattering

If the inversion deviates from its equilibrium value, w_0, it relaxes back into equilibrium with a time constant T_1. Eq. (10.69) shows that for all temperatures $T > 0$ the inversion is negative, i.e., the population of the lower level is higher than the upper level. Thus with incoherent thermal light, inversion in a two-level system cannot be achieved. Inversion can only be achieved by pumping with incoherent light, if there are more levels and subsequent relaxation processes into the upper laser level. Due to these relaxation processes, the rate Γ_a deviates from the equilibrium expression (10.65), and it has to be replaced by the pump rate Λ. If the pump rate Λ exceeds Γ_e, the inversion corresponding to Eq. (10.69) becomes positive

$$w_0 = \frac{\Lambda - \Gamma_e}{\Lambda + \Gamma_e}. \tag{10.74}$$

If we allow for artificial negative temperatures, we obtain with $T < 0$ for the ratio of relaxation rates

$$\frac{\Gamma_e}{\Gamma_a} = \frac{1 + n_{th}}{n_{th}} = e^{\frac{\hbar\omega_{eg}}{kT}} < 1. \tag{10.75}$$

Thus the pumping of the two-level system drives the system away from thermal equilibrium. Now, we have a correct description of an ensemble of atoms in thermal equilibrium with its environment, which is a much more realistic description of media especially of typical laser media.

10.5 Bloch equations

Thus, the total dynamics of the two-level system including the pumping and dephasing processes from Eqs.(10.70) and (10.71) is given by

$$\dot{\underline{d}} = -\left(\frac{1}{T_2} - \mathrm{j}\,(\omega_{eg} - \omega)\right)\underline{d} + \mathrm{j}\frac{\Omega_r}{2}\,w, \tag{10.76}$$

$$\dot{w} = -\frac{w - w_0}{T_1} + \mathrm{j}\Omega_r^*\,\underline{d} - \mathrm{j}\Omega_r\,\underline{d}^*. \tag{10.77}$$

These equations are called the Bloch equations (within the RWA). They describe the dynamics of a statistical ensemble of two-level atoms interacting with a classical electric field. Together with Maxwell's equations, where the polarization of the medium is related to the expectation value of the dipole moment of the atomic ensemble, these result in the Maxwell-Bloch equations.

10.6 Dielectric susceptibility and saturation

The Bloch equations are nonlinear. However, for moderate field strength $\underline{E}_0$, i.e., the magnitude of the Rabi frequency is much smaller than the optical frequency, $|\Omega_r| \ll \omega$, the inversion does not change much within an optical cycle of the field. We assume that the inversion w of the atom will only be slowly changing and it adjusts itself to a steady-state value w_s. For a constant field strength $\underline{E}_0$, Eqs. (10.76) and (10.77) reach the steady-state values

$$\underline{d}_s = \frac{\mathrm{j}}{2\hbar} \frac{\left(\vec{M}_{eg}^* \cdot \vec{e}\right) w_s}{1/T_2 + \mathrm{j}(\omega - \omega_{eg})} \underline{E}_0 \tag{10.78}$$

$$w_s = \frac{w_0}{1 + \frac{T_1}{\hbar^2} \frac{1/T_2 \, |\vec{M}_{eg}^* \cdot \vec{e}|^2}{(1/T_2)^2 + (\omega_{eg} - \omega)^2} |\underline{E}_0|^2}. \tag{10.79}$$

We introduce the normalized lineshape function, which is in this case a Lorentzian

$$L(\omega) = \frac{(1/T_2)^2}{(1/T_2)^2 + (\omega_{eg} - \omega)^2}, \tag{10.80}$$

and connect the square modulus of the field $|\underline{E}_0|^2$ to the intensity I of a propagating plane wave, according to $I = \frac{1}{2Z_F} |\underline{E}_0|^2$,

$$w_s = \frac{w_0}{1 + \frac{I}{I_s} L(\omega)}. \tag{10.81}$$

Thus the stationary inversion depends on the intensity of the incident light.

Therefore, w_0 is called the unsaturated inversion, w_s the saturated inversion and I_s, with

$$I_s = \left[\frac{2T_1 T_2 Z_F}{\hbar^2} |\vec{M}^*_{eg} \cdot \vec{e}|^2 \right]^{-1}, \tag{10.82}$$

is the saturation intensity. The expectation value of the dipole operator (10.22) is then given by

$$\left\langle \vec{d} \right\rangle = \vec{M}_{eg} \underline{d}\, e^{\mathrm{j}\omega t} + c.c. \tag{10.83}$$

Multiplication with the number of atoms per unit volume, N, relates the dipole moment of the atom to the macroscopic polarization $\vec{P}$. As the electric field, also the polarization can be written in terms of complex quantities

$$\vec{P}(t) = \frac{1}{2} \left(\underline{\vec{P}}_0 e^{\mathrm{j}\omega t} + \underline{\vec{P}}^*_0 e^{-\mathrm{j}\omega t} \right) \tag{10.84}$$

$$= N \vec{M}_{eg} \underline{d}_s\, e^{\mathrm{j}\omega t} + c.c. \tag{10.85}$$

or

$$\underline{\vec{P}}_0 = 2N \vec{M}_{eg} \underline{d}_s. \tag{10.86}$$

With the definition of the complex susceptibility

$$\underline{\vec{P}}_0 = \epsilon_0 \chi(\omega) \vec{e} \underline{E}_0 \tag{10.87}$$

and comparison with Eqs. (10.86) and (10.78), we obtain for the linear susceptibility of the medium

$$\chi(\omega) = \vec{M}_{eg} \vec{M}^+_{eg} \frac{\mathrm{j}N}{\hbar \epsilon_0} \frac{w_s}{1/T_2 + \mathrm{j}(\omega - \omega_{eg})}, \tag{10.88}$$

which is a tensor. In the following we assume that the direction of the atom is random, i.e., the alignment of the atomic dipole moment, $\vec{M}_{eg}$, and the electric field is random. Therefore, we have to average over the angle enclosed between the electric field of the wave and the atomic dipole moment, which results in

$$\overline{\begin{pmatrix} M_{egx}M_{egx}^* & M_{egx}M_{egy}^* & M_{egx}M_{egz}^* \\ M_{egy}M_{egx}^* & M_{egy}M_{egy}^* & M_{egy}M_{egz}^* \\ M_{egz}M_{egx}^* & M_{egz}M_{egy}^* & M_{egz}M_{egz}^* \end{pmatrix}} = \begin{pmatrix} \overline{M_{egx}^2} & 0 & 0 \\ 0 & \overline{M_{egy}^2} & 0 \\ 0 & 0 & \overline{M_{egz}^2} \end{pmatrix} = \frac{1}{3}|\vec{M}_{eg}|^2\,\mathbf{1}. \tag{10.89}$$

How to arrive at this average over the orientation is also discussed in Appendix A. Thus, for homogeneous and isotropic media the susceptibility tensor shrinks to a scalar

$$\chi(\omega) = \frac{1}{3}|\vec{M}_{eg}|^2 \frac{\mathrm{j}N}{\hbar\epsilon_0} \frac{w_s}{1/T_2 + \mathrm{j}(\omega - \omega_{eg})}. \tag{10.90}$$

Real and imaginary part of the susceptibility

$$\chi(\omega) = \chi'(\omega) + \mathrm{j}\chi''(\omega) \tag{10.91}$$

are then given by

$$\chi'(\omega) = -\frac{|\vec{M}_{eg}|^2 N w_s T_2^2(\omega_{eg} - \omega)}{3\hbar\epsilon_0} L(\omega), \tag{10.92}$$

$$\chi''(\omega) = \frac{|\vec{M}_{eg}|^2 N w_s T_2}{3\hbar\epsilon_0} L(\omega). \tag{10.93}$$

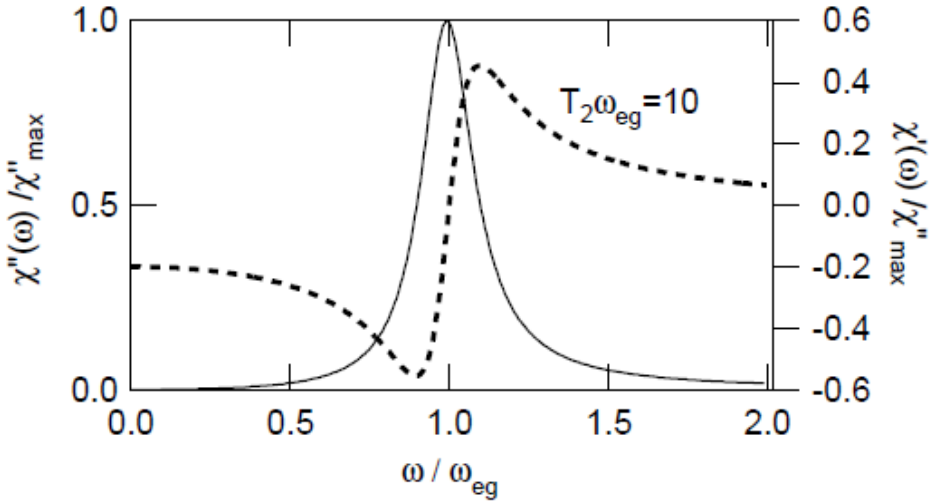


Figure 10.4: Real and imaginary part of the complex susceptibility for an inverted medium $w_s > 0$. The positive imaginary susceptibility indicates exponential growth of an electromagnetic wave propagating in the medium.

If the incident radiation is weak, i.e.,

$$\frac{I}{I_s} L(\omega) \ll 1 \tag{10.94}$$

we obtain $w_s \approx w_0$. For optical transitions there is no thermal excitation of the excited state and $w_0 = -1$. For an inverted system, $w_0 > 0$, the real and imaginary parts of the susceptibility are shown in Fig. 10.4.

The shape of the susceptibility computed quantum mechanically compares well with the classical susceptibility (2.22) derived from the harmonic oscillator model close to the transistion frequency ω_{eg} for a transition with reasonably high $Q = T_2\omega_{eg}$. Note, the quantum mechanical susceptibility is identical to the complex Lorentzian one encounters in the discussion of loss and gain. There is an appreciable deviation, however, far away from resonance. Far off resonance, the RWA should not be used.

The physical meaning of the real and imaginary part of the susceptibility is of course identical to our earlier discussion. The propagation constant k of a TEM-wave in such a medium is related to the susceptibility by

$$k = \omega\sqrt{\mu_0\epsilon_0(1+\chi(\omega))} \approx k_0\left(1+\frac{1}{2}\chi(\omega)\right), \quad \text{with} \quad k_0 = \omega\sqrt{\mu_0\epsilon_0} \tag{10.95}$$

for $|\chi| \ll 1$. Under this assumption we obtain

$$k = k_0(1+\frac{\chi'}{2}) + \mathrm{j}k_0\frac{\chi''}{2}. \tag{10.96}$$

The real part of the susceptibility contributes to the refractive index $n = 1+\chi'/2$. In the case of $\chi'' < 0$, the imaginary part leads to an exponential damping of the wave. For $\chi'' > 0$, amplification takes place. Amplification of the wave is possible for $w_0 > 0$, i.e., in an inverted medium.

The phase relaxation rate $1/T_2$ of the dipole moment determines the width of the absorption line or the bandwidth of the amplifier. The amplification can not occur forever, because the amplifier saturates, when the intensity reaches the saturation intensity. This is a strong deviation from the linear susceptibility we derived from the classical oscillator model. The reason for this saturation is twofold: First, the light can not extract more energy from the atoms than stored in them, i.e., energy conservation holds. Second, the induced dipole moment in a two-level atom is limited by the maximum value of the matrix element. In contrast, the induced dipole moment in a classical oscillator grows proportionally to the applied field without limits.

10.7 Rate equations and cross-sections

limit of fast dephasing, i.e., T_2 much shorter than dynamics we are interested in

the magnitude of the dipole moment relaxes instantaneously into the steady state and follows the slowly varying electric field envelope $E_0(t)$, which evolves on a much longer time scale. We obtain with the quasi-steady-state solution for the dipole moment (10.78), which may now have a slow time dependence due to the slowly varying field envelope $E_0(t)$, for the time-dependent inversion in the atomic system

$$\dot{w} = -\frac{w(t) - w_0}{T_1} - \frac{w(t)}{T_1 I_s} L(\omega) I(t), \tag{10.97}$$

where $I(t) = |E_0(t)|^2 / (2Z_F)$ is the intensity of the electromagnetic wave interacting with the two-level atom. In this limit, the Bloch equations are replaced by a simple rate equation for the population. We only take care of the counting of population differences due to spontaneous and stimulated emissions.

The interaction of an atom with light at a given transition with the stream of photons on resonance, i.e., $\omega = \omega_{eg}$ is often described by the mass action law. That is, the number of induced transitions from the excited to the ground state, is proportional to the product of the number of atoms in the excited state and the photon flux density $I_{ph} = I/\hbar\omega_{eg}$

$$\dot{w}|_{induced} = -\sigma w I_{ph} = -\frac{w}{T_1 I_s} I. \tag{10.98}$$

This defines an interaction cross-section σ that can be expressed in terms of the saturation intensity as

$$\sigma = \frac{\hbar\omega_{eg}}{T_1 I_s} \tag{10.99}$$

$$= \frac{2\omega_{eg} T_2 Z_F}{\hbar} |\vec{M}^*_{eg} \cdot \vec{e}|^2. \tag{10.100}$$

To summarize the findings of the discussions in this chapter so far, we found the most important spectroscopic quantities that characterize an atomic transition, which are the lifetime of the excited state or often called upper-state lifetime or longitudinal lifetime T_1, the phase relaxation time or transverse relaxation time T_2 which is the inverse half-width at half maximum (HWHM) of the line, and the interaction cross-section σ that only depends on the dipole matrix element and the linewidth of the transition.

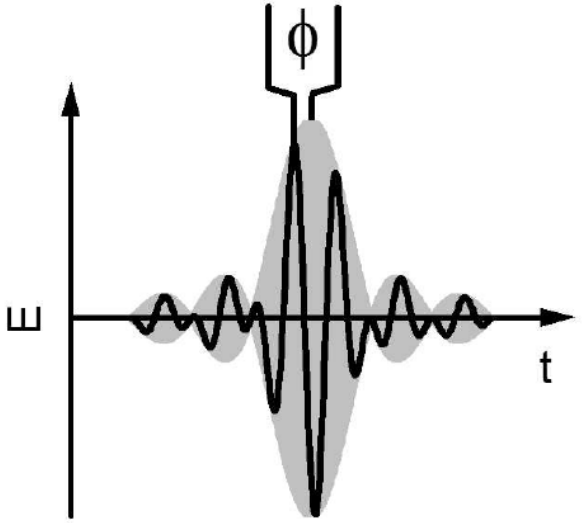
10.8 Extreme nonlinear optical response of two-level systems

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 $E(t)$

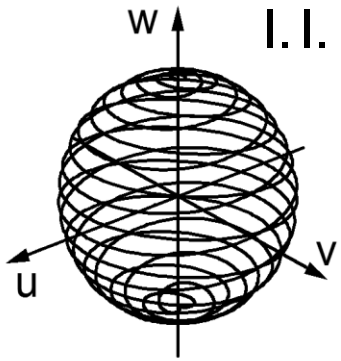


M. Wegener, *Extreme Nonlinear Optics*, Springer, Berlin (2005)

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 $\Omega_R(t)\propto E(t)$

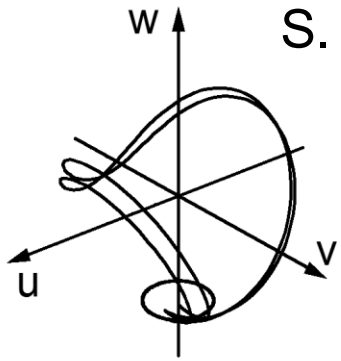
(a) I.I. Rabi (1937)



$\Omega_R \ll \omega_0$

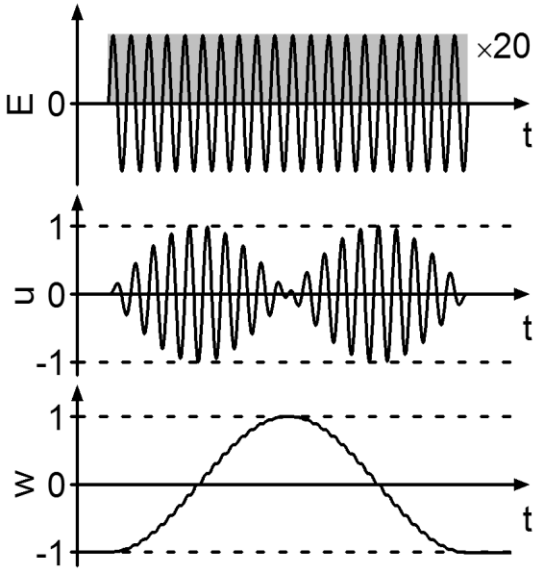
2π pulse

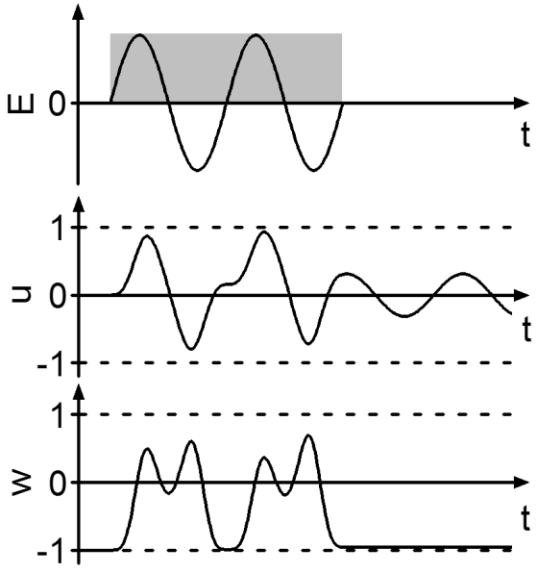
(b) S. Hughes (1998)



$\Omega_R = \omega_0$

4π pulse

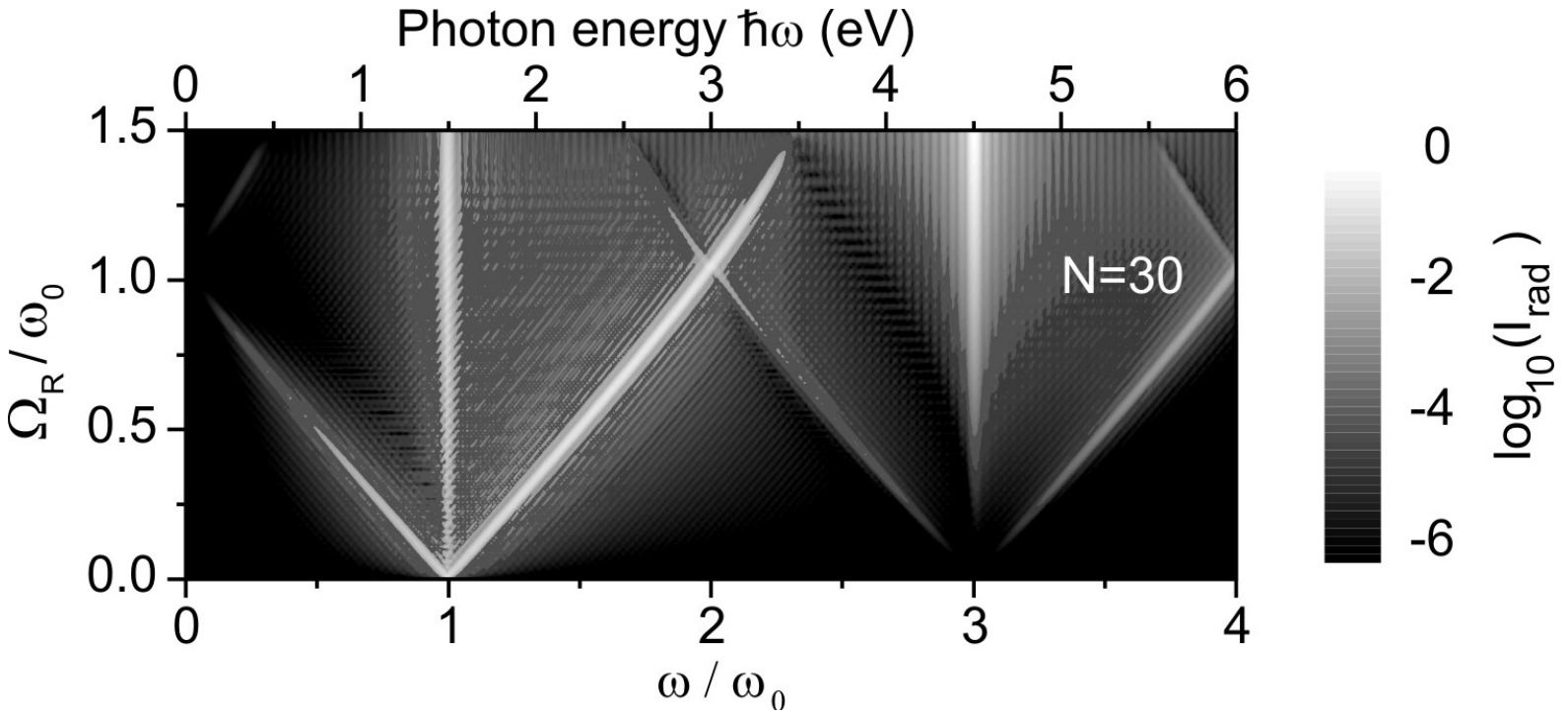




Carrier-wave Mollow triplets

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 $(0, 0, -1)^T$.

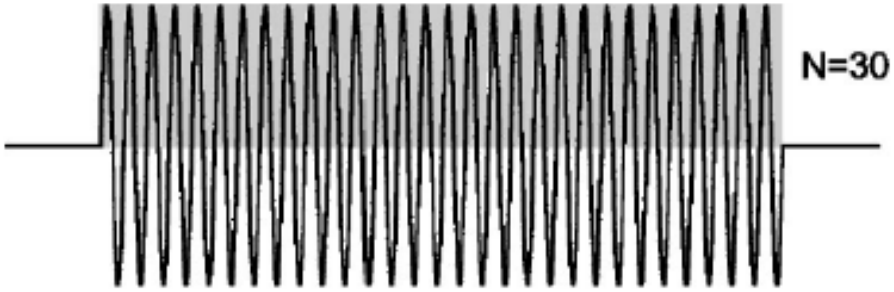


Figure 10.5: Box-shaped optical pulses $E(t)$: The integer number of cycles in the pulse is called N. The gray area indicates the electric-field envelope $\tilde{E}(t)$. [8]

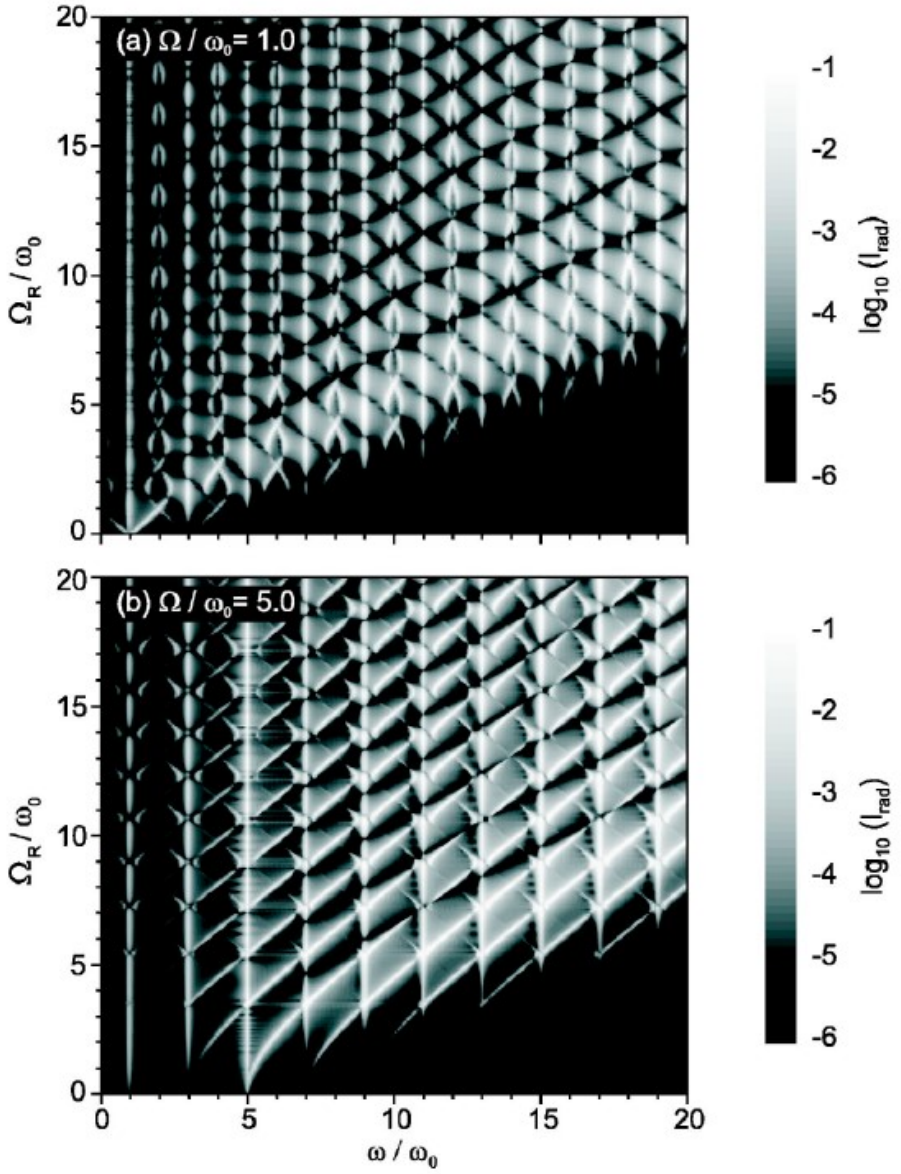


Figure 10.6: Gray-scale images of the radiated intensity spectra $I_{\rm rad}(\omega) \propto |\omega^2 u(\omega)|^2$ (normalized and on a logarithmic scale) from exact numerical solutions of the two-level system Bloch equations (10.101). The peak Rabi frequency $\Omega_{\rm R}$ of the exciting $N = 30$ 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)

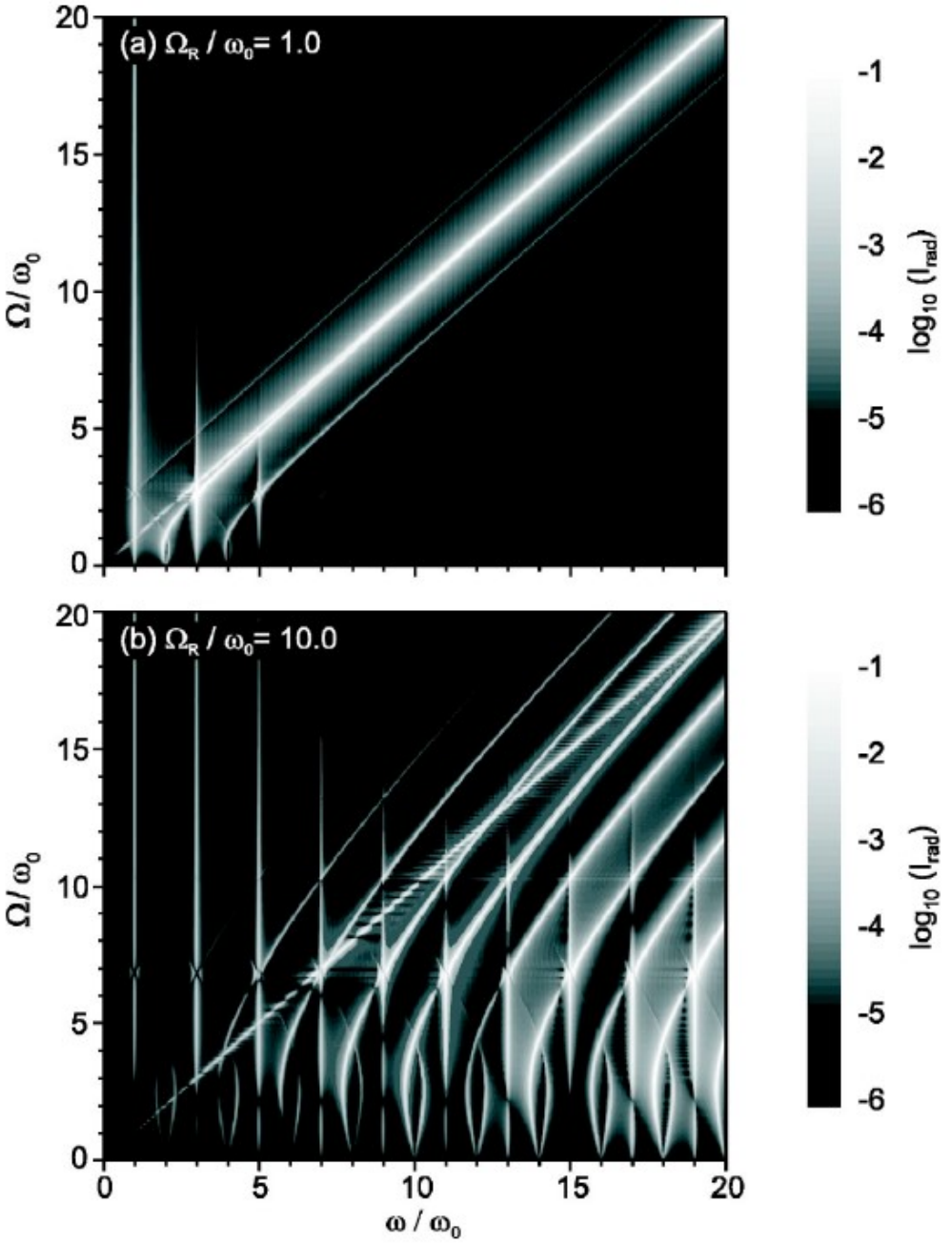


Figure 10.7: Same as Fig. 10.6, but versus transition frequency Ω for two fixed values of the peak Rabi frequency Ω_R. (a) $\Omega_R/\omega_0 = 1$ and (a) $\Omega_R/\omega_0 = 10$. [8]

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 $2\omega_0$ CEP 2ϕ

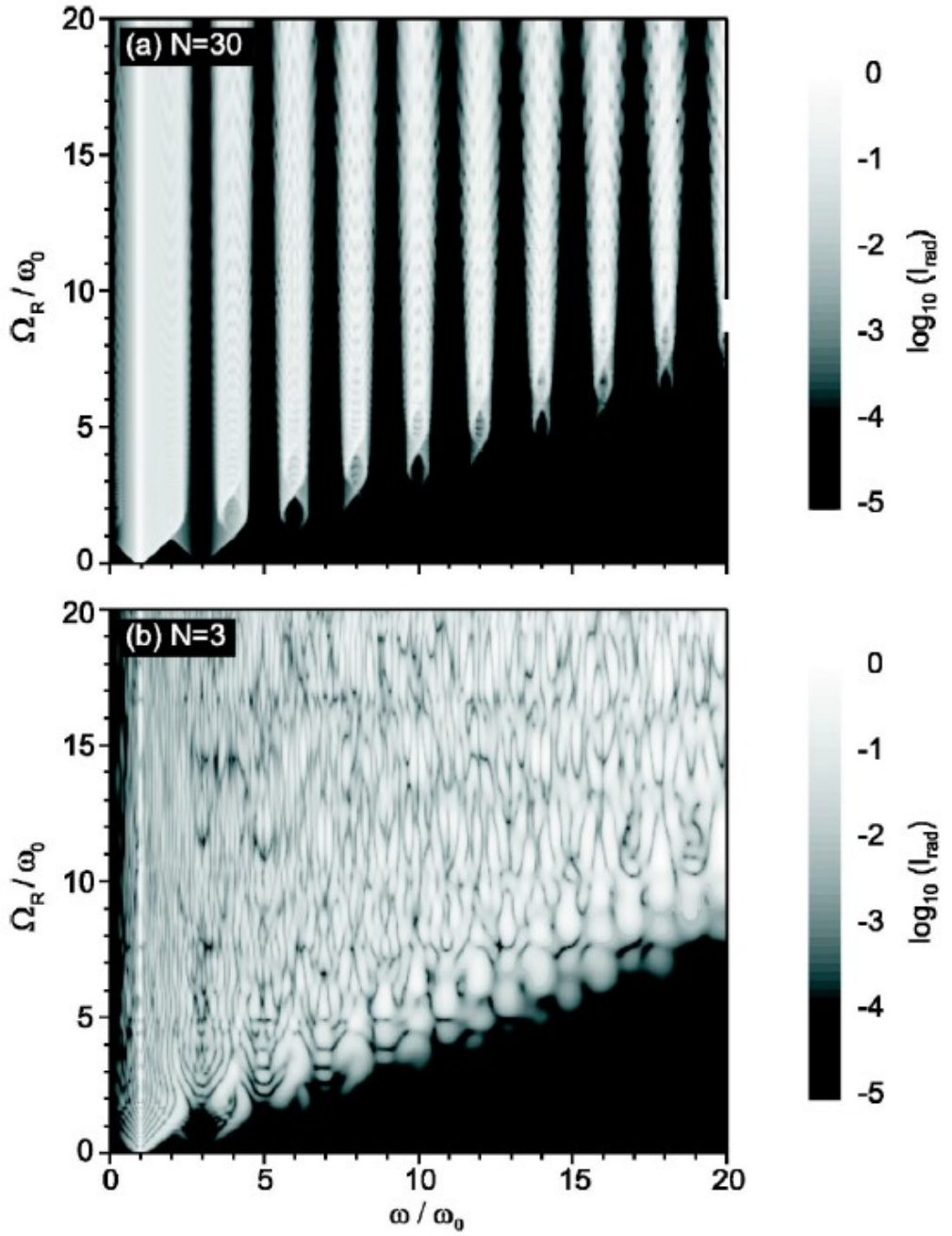


Figure 10.8: Same as Fig. 10.6(a), i.e., $\Omega/\omega_0 = 1$, but for Gaussian optical pulses with CEP $\phi = 0$ and with a FWHM of (a) $N = 30$ and (b) $N = 3$ optical cycles. [8]

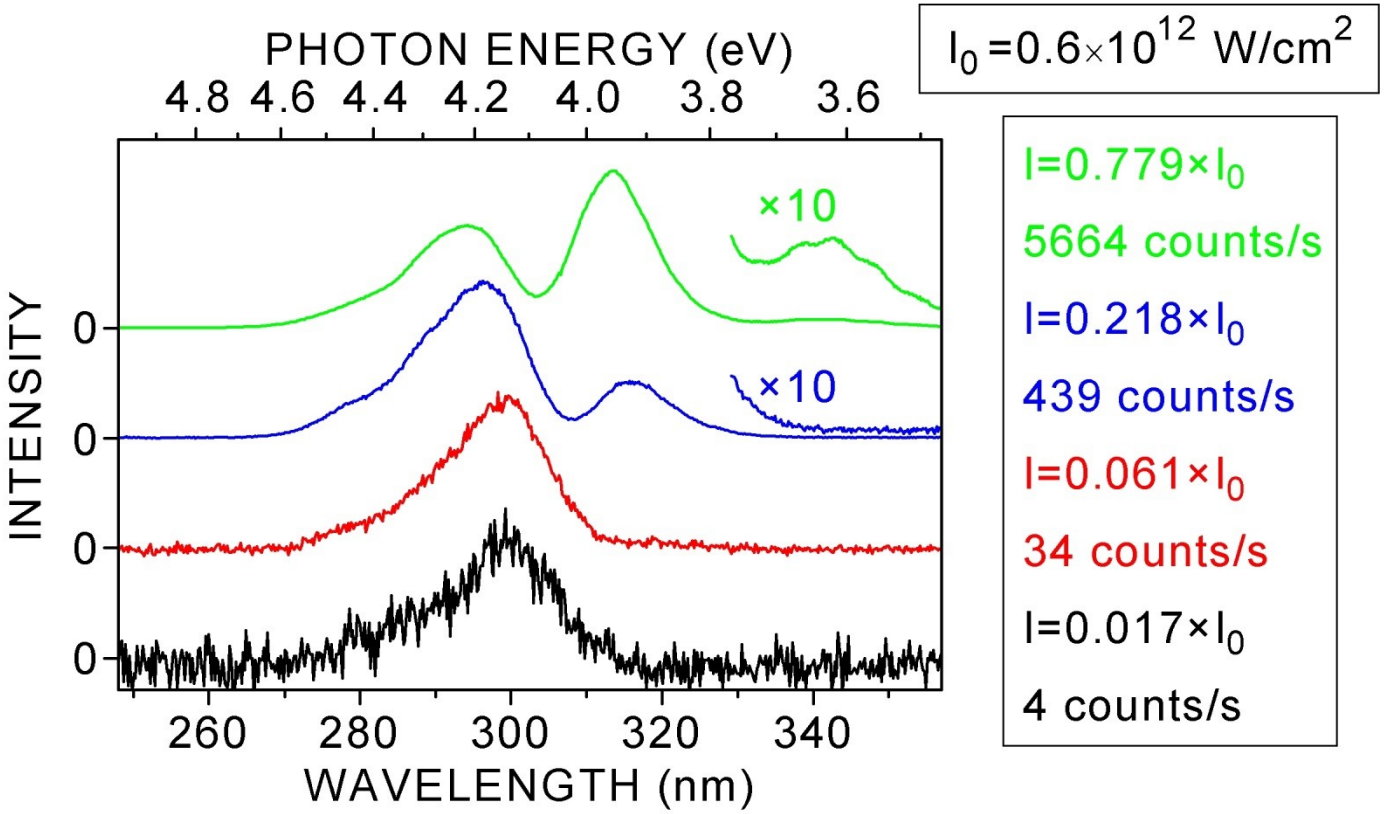
For Gaussian pulses, electric field envelope not constant in time

effectively averaging over vertical axis

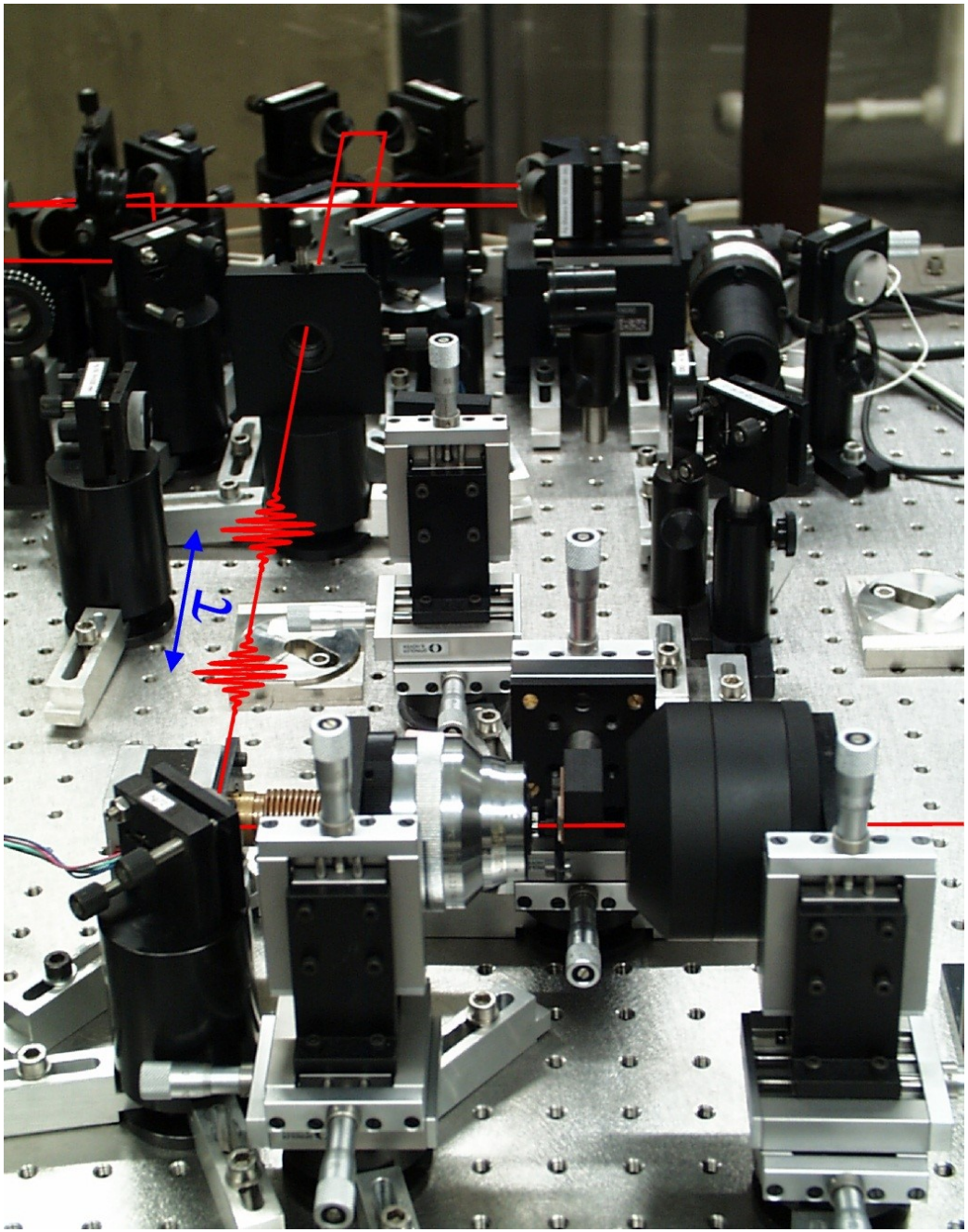
“messy” spectra

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)



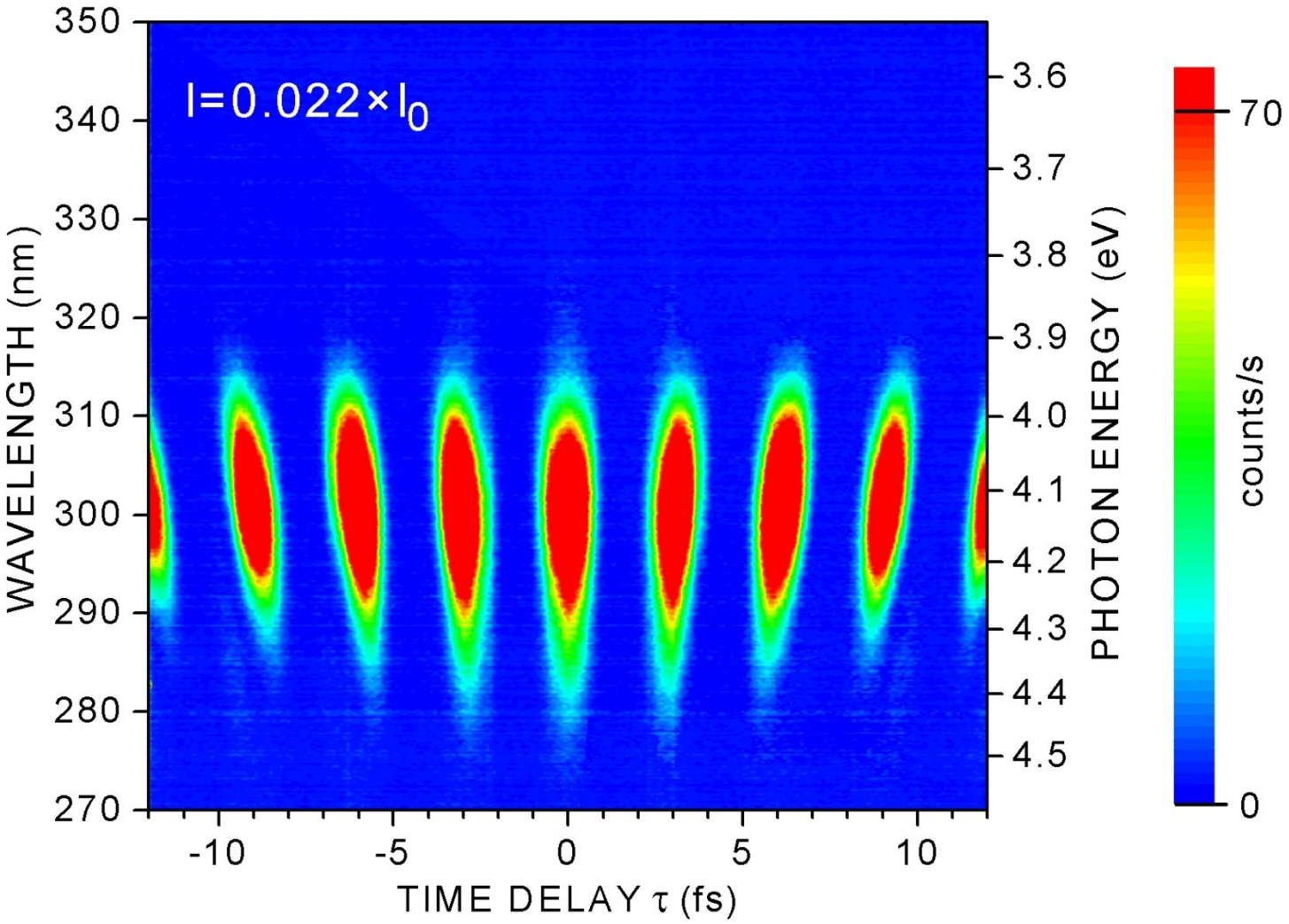
~5fs Ti:sapphire laser pulses

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

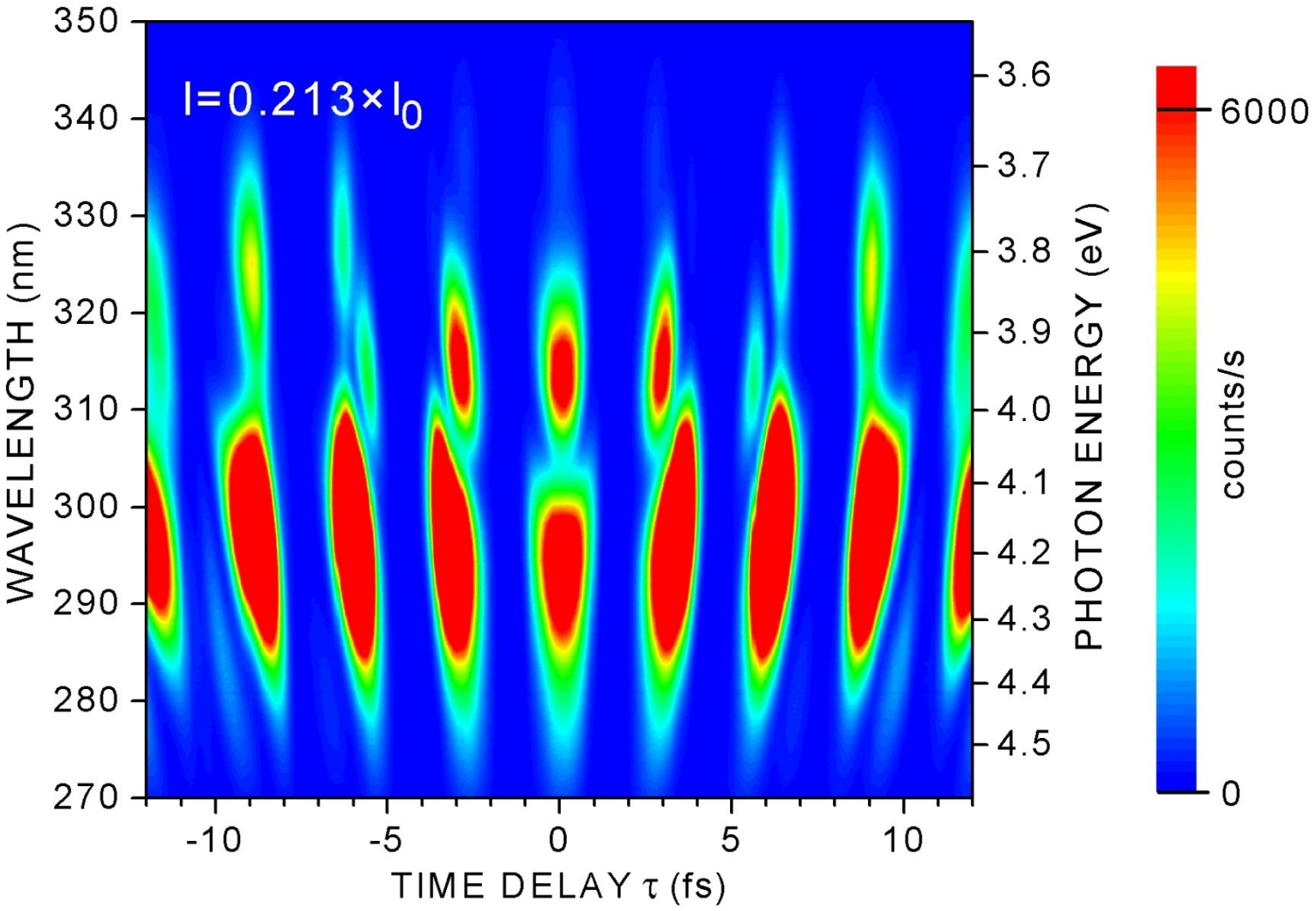
remaining fluctuations in time delay τ are <50 as

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

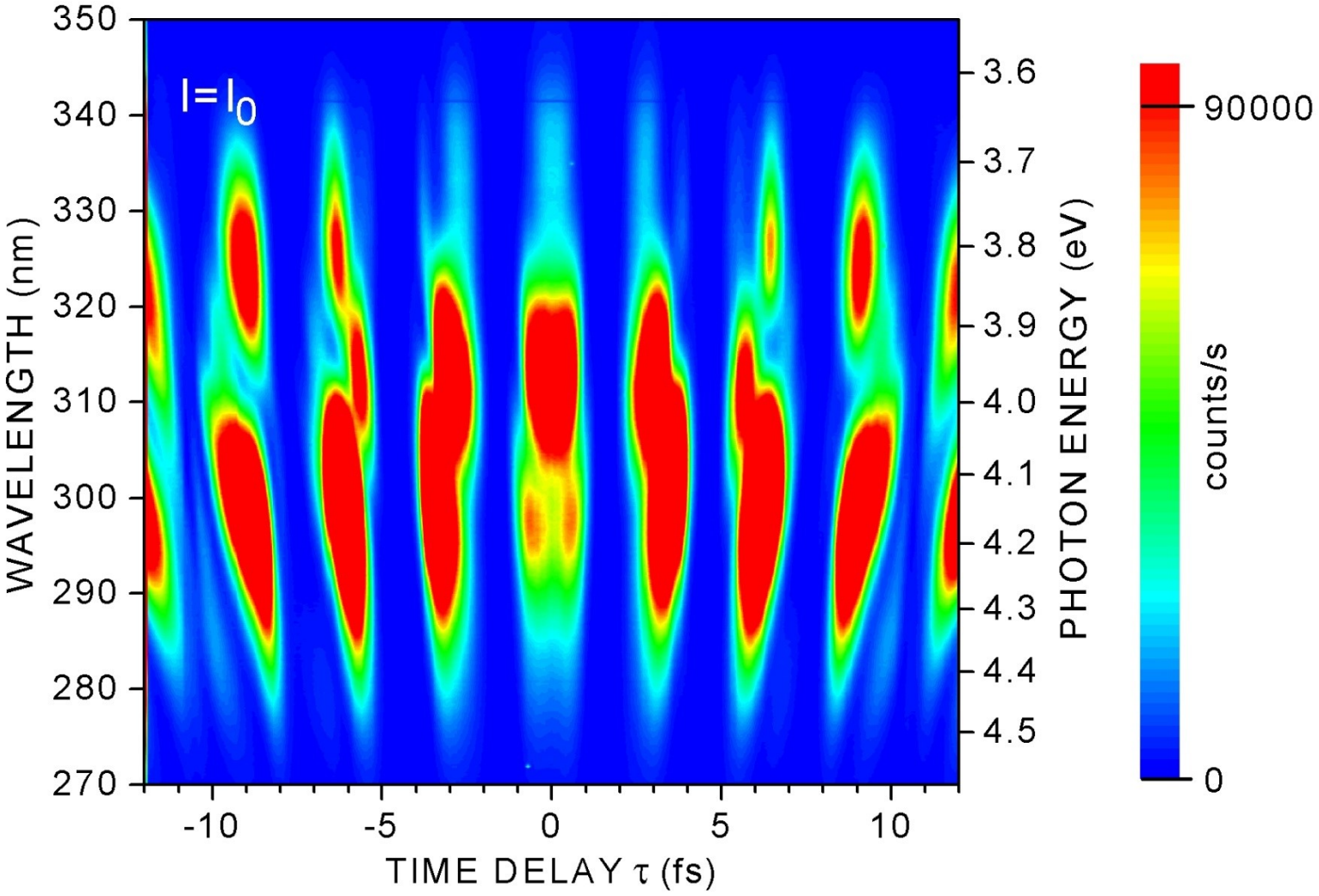
Interferometric measurements



Interferometric measurements



Interferometric measurements



CEP dependence

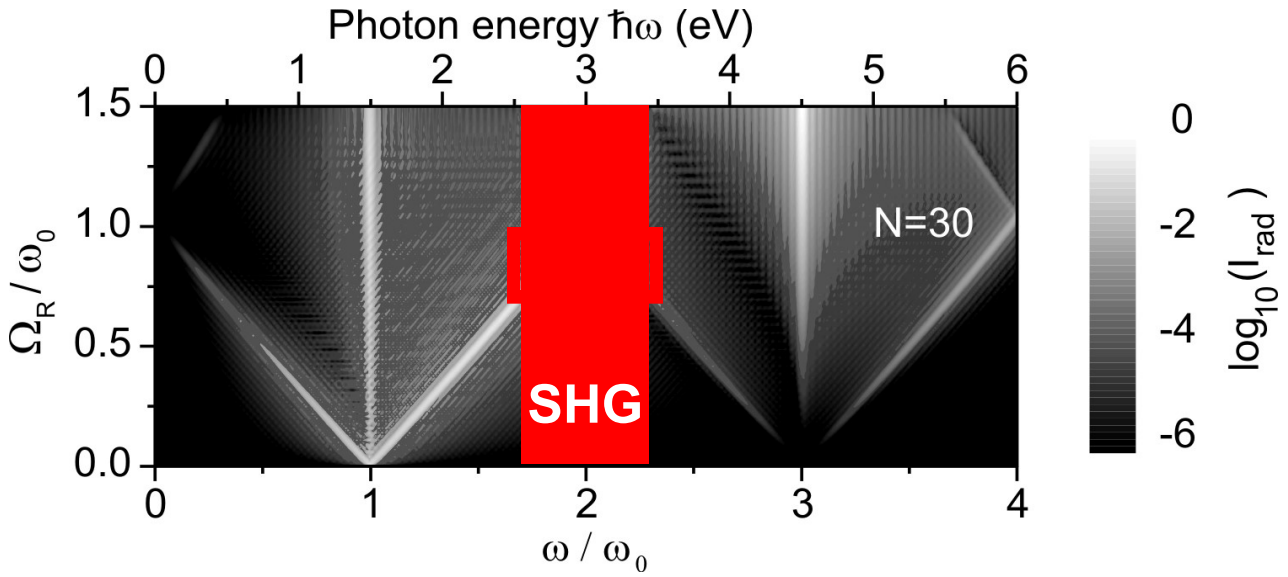
fundamental / third-harmonic Mollow triplet: 1ϕ / 3ϕ

+

surface second-harmonic generation: 2ϕ

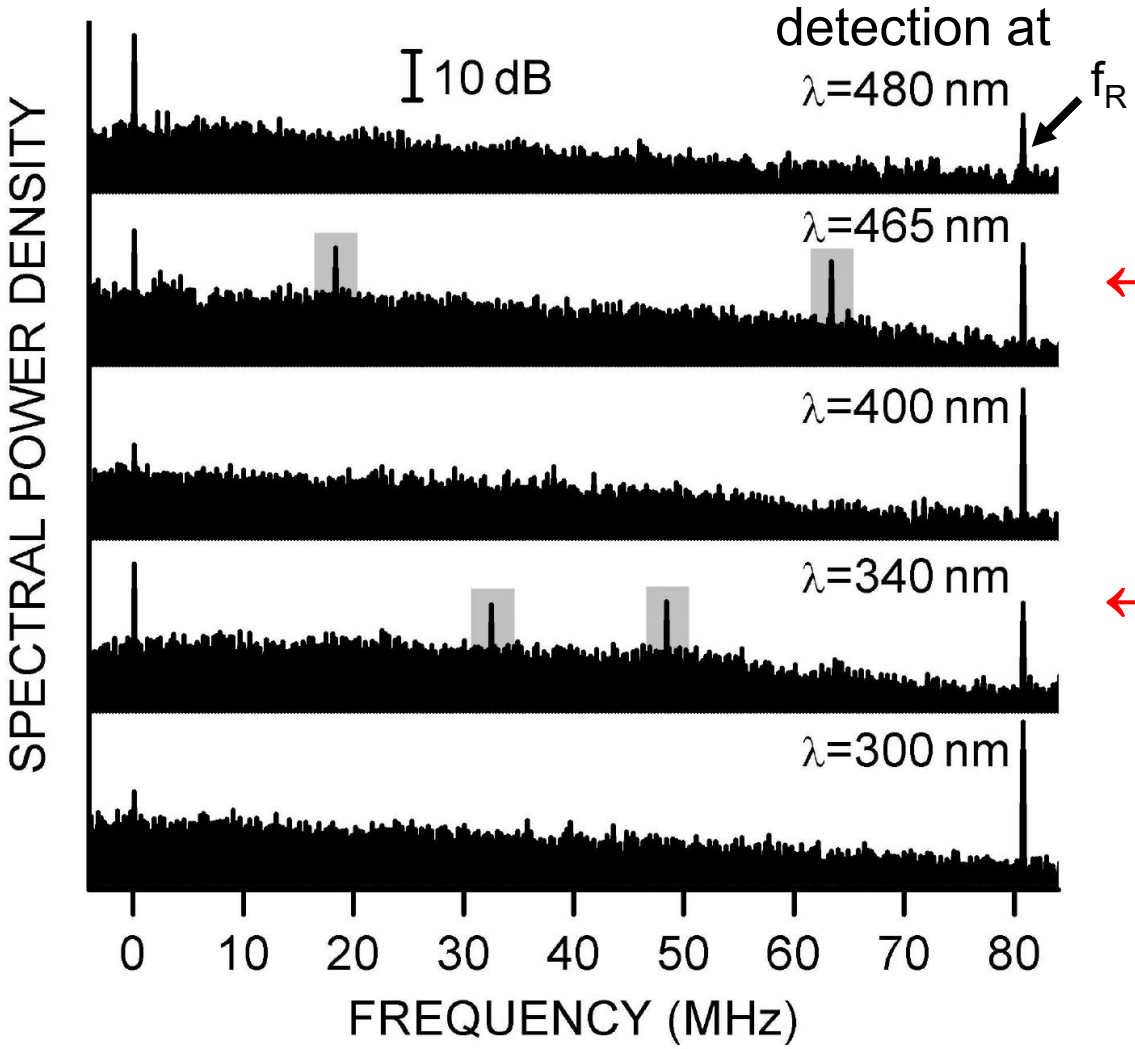
↓

CE phase dependence: 1ϕ



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

Measuring the CEO frequency with GaAs



← f_{CEO} (SHG+fundamental)

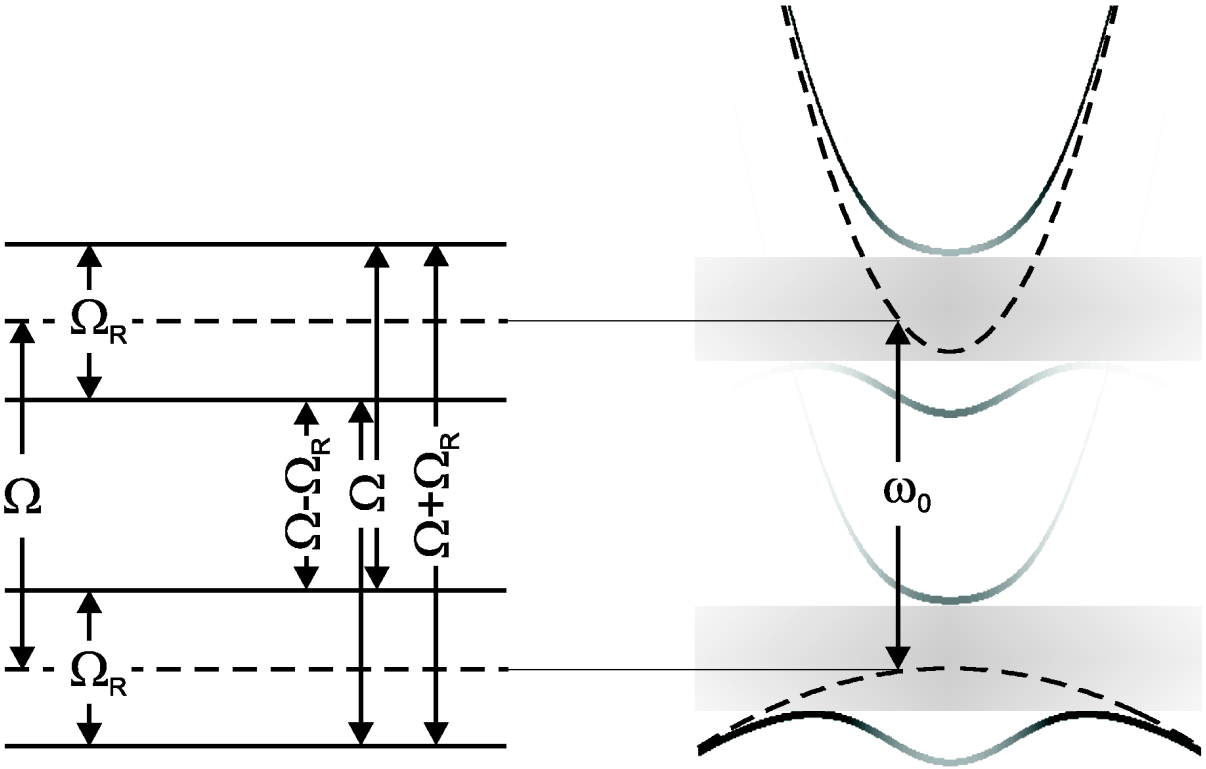
← f_{CEO} (SHG+THG)

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

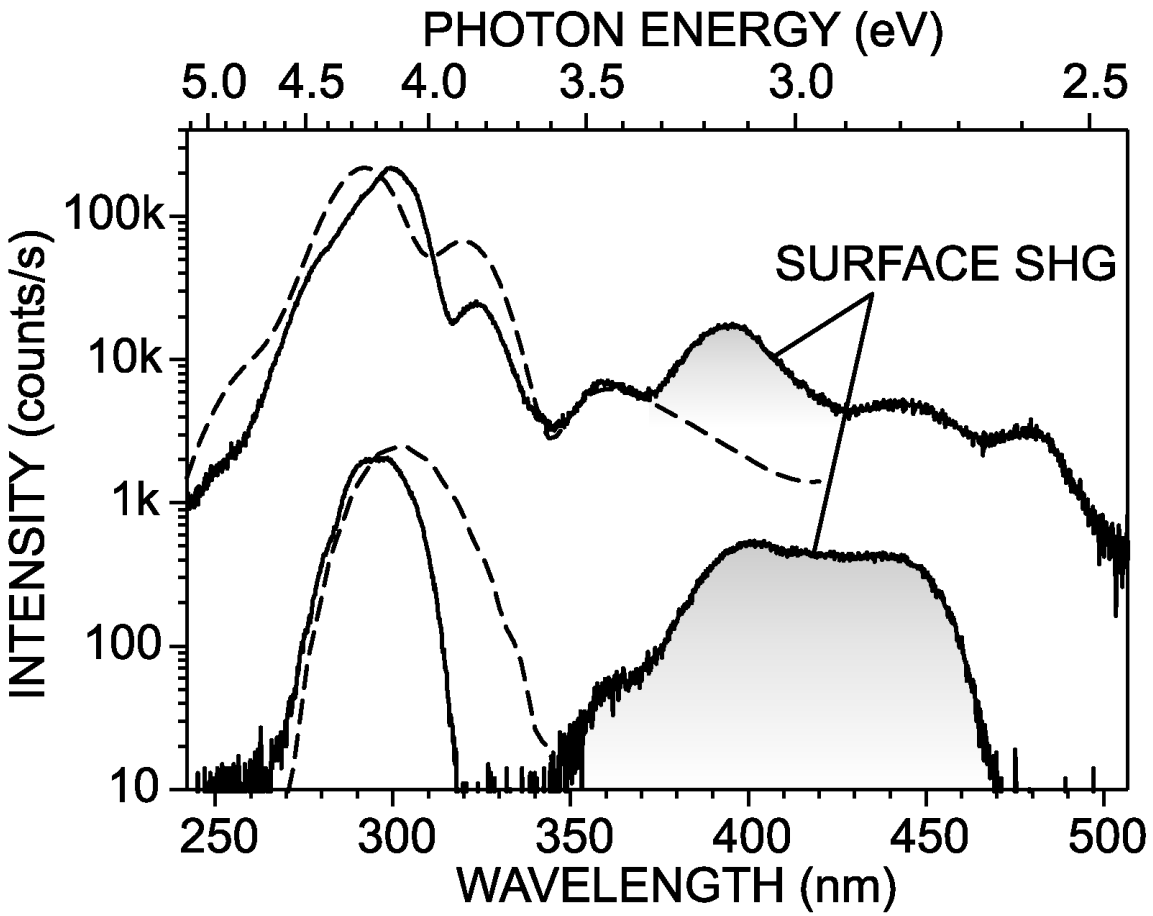
avoided crossing

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)

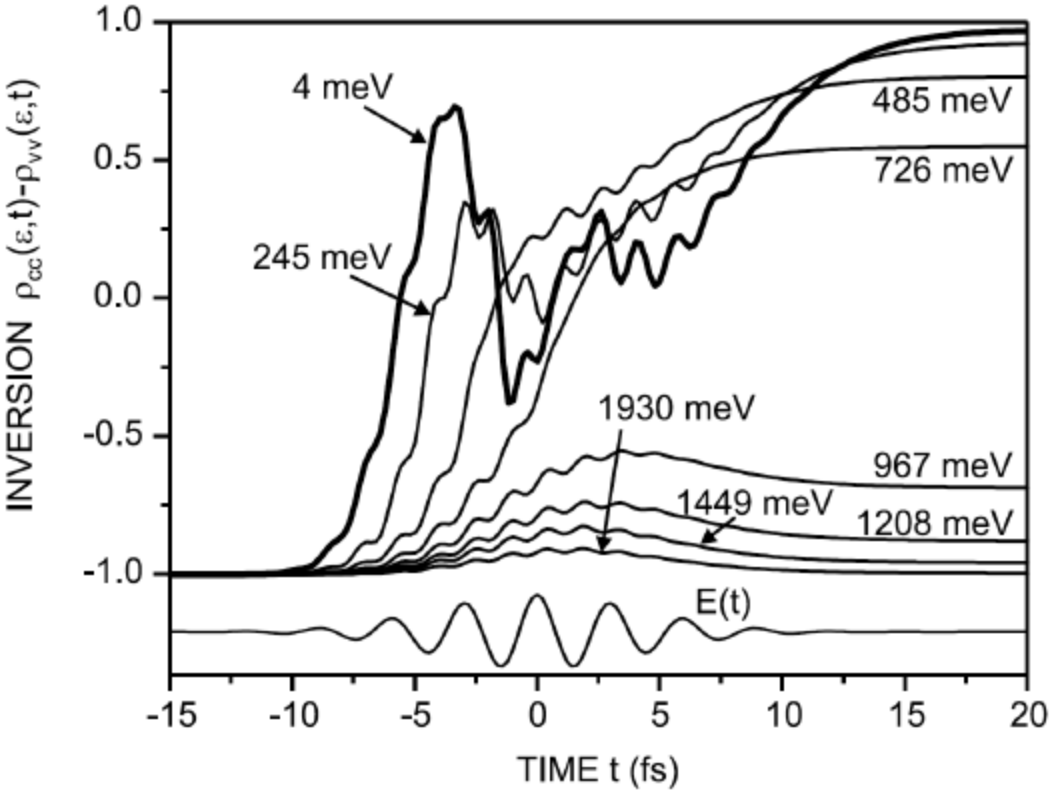
theory (dashed curves):
- semiconductor Bloch equations
- full tight-binding bands
- density- and energy-dependent dephasing and relaxation
- no RWA

experiment (solid curves):
- 100nm thin GaAs film

high excitation
(upper curves)

low excitation
(lower curves)

Light-induced gaps in semiconductors

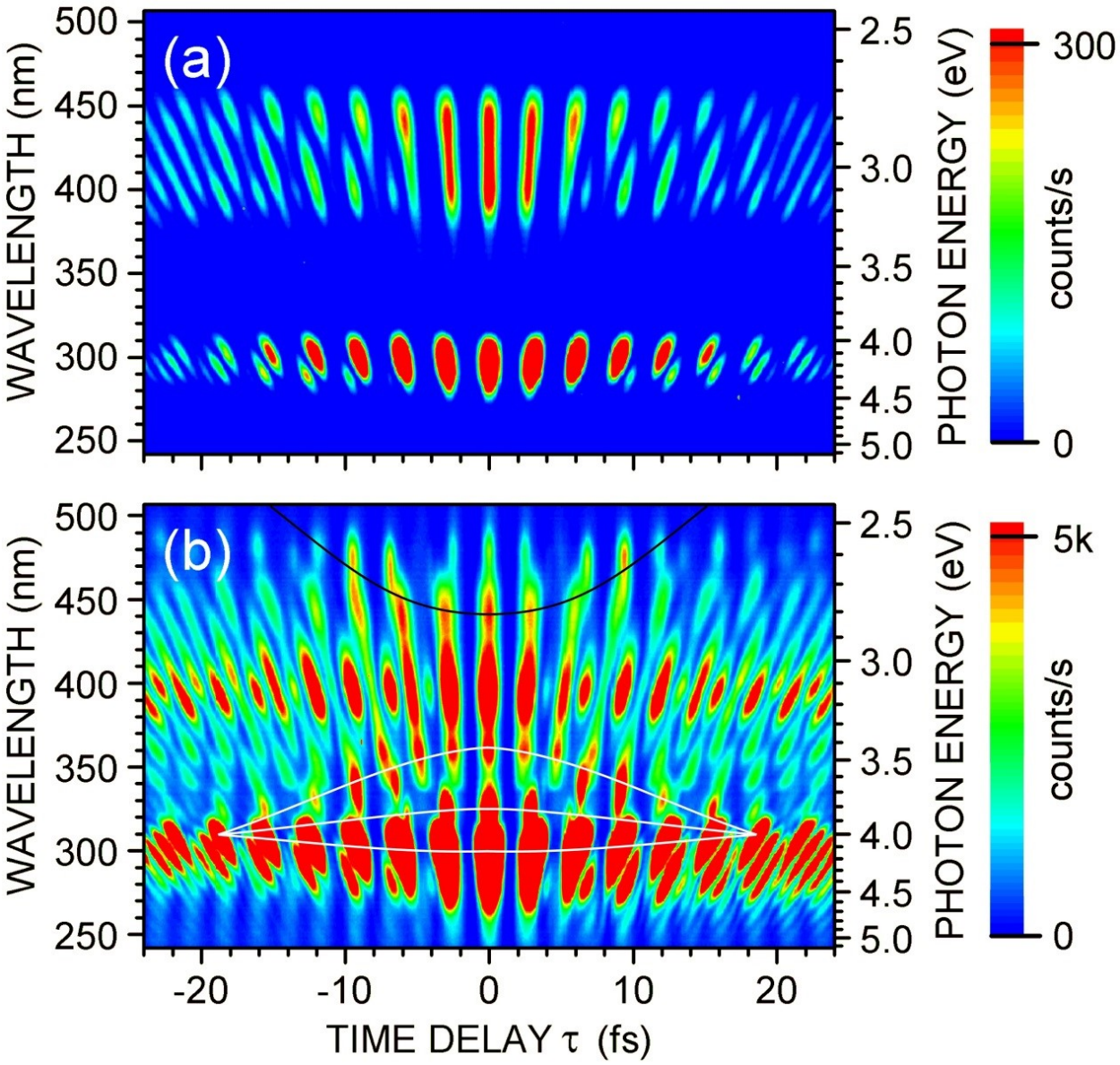


effect of carrier-density- and energy-dependent **dephasing** and **relaxation**

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)

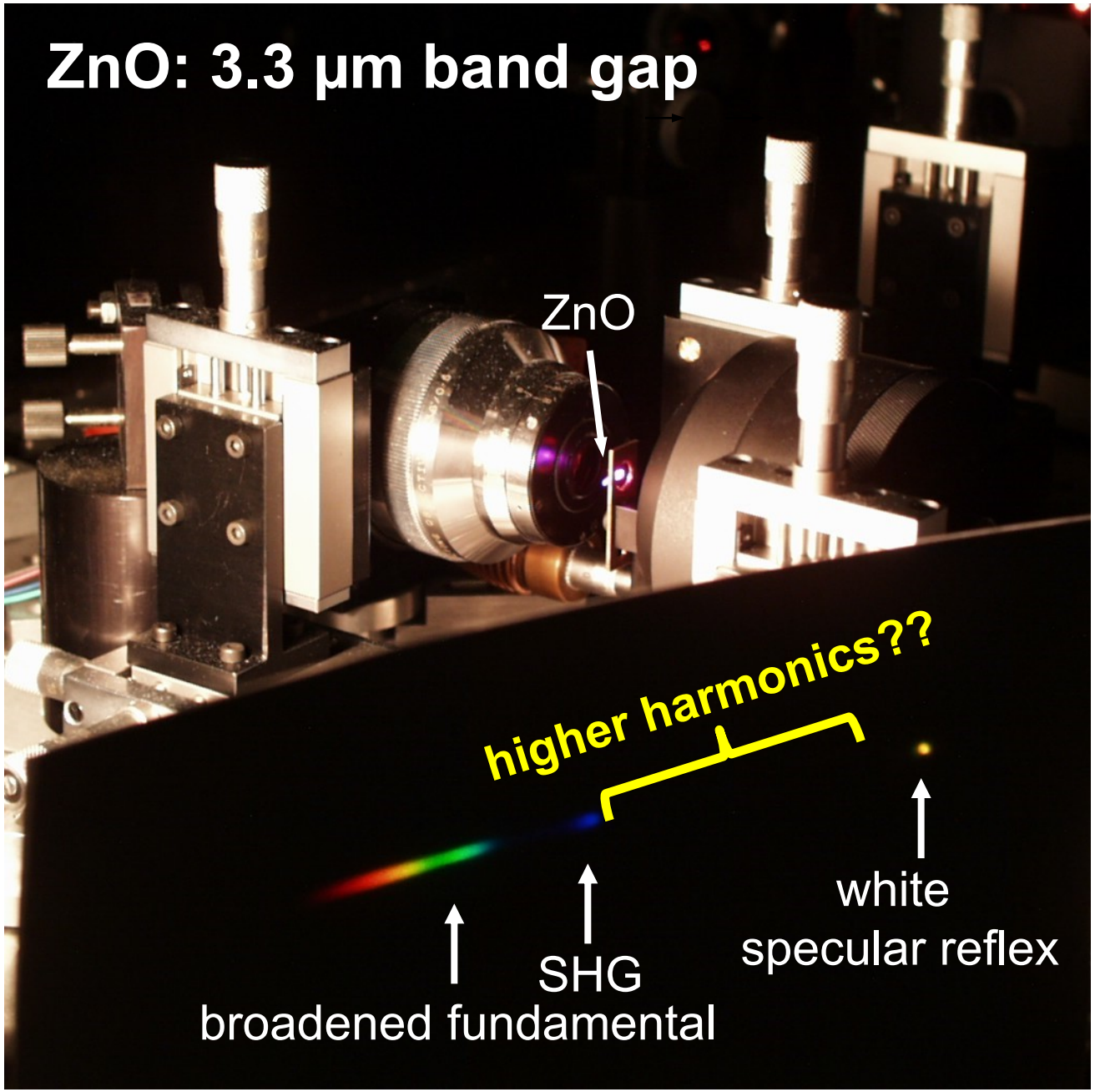
Light-induced gaps in semiconductors



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 = 6V/nm

Bloch energy

$\hbar$)eV

Bloch period

$T_B = 1.4\text{fs}$

optical period (800nm)

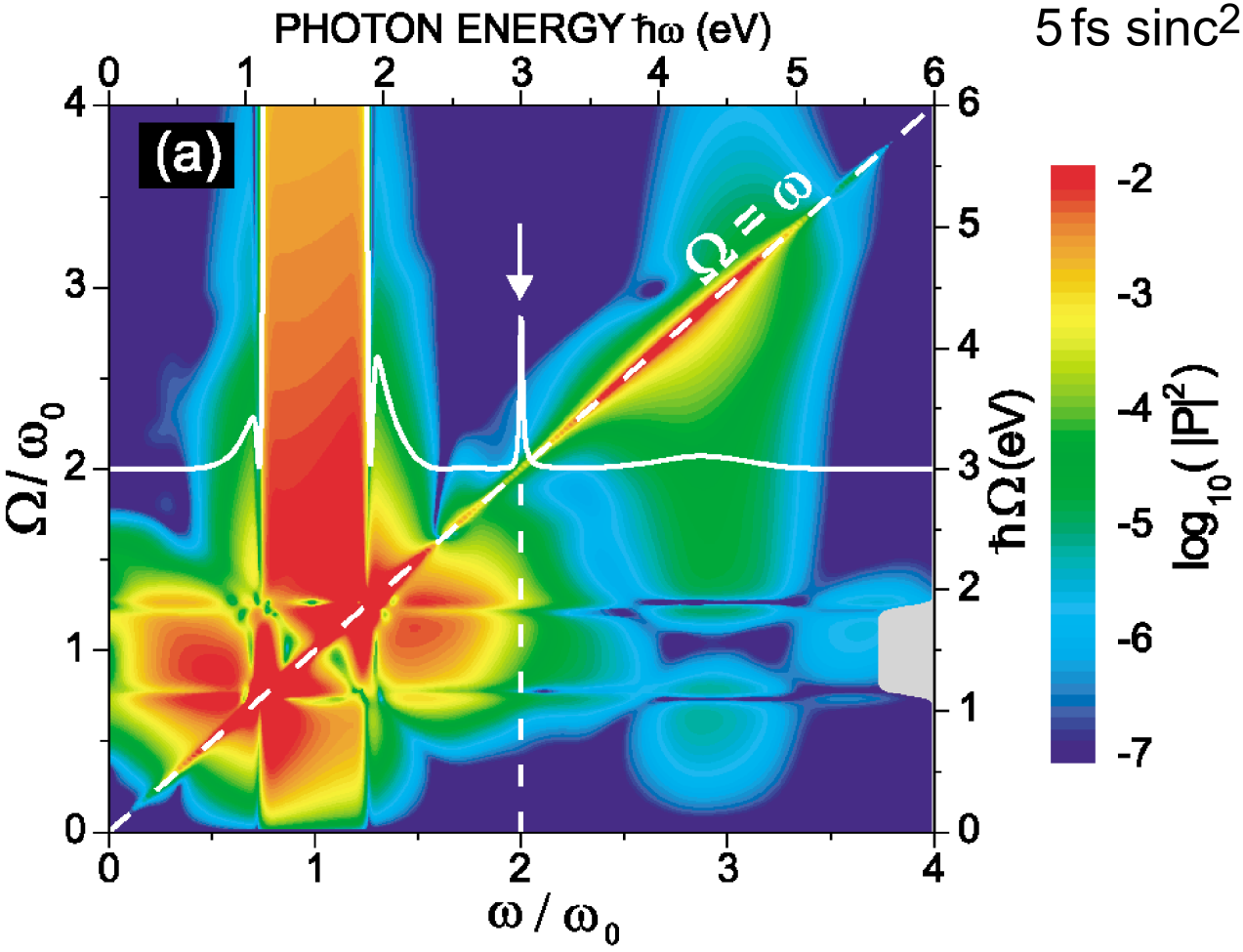
$T_L = 2.8\text{fs}$

Bloch oscillations??

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

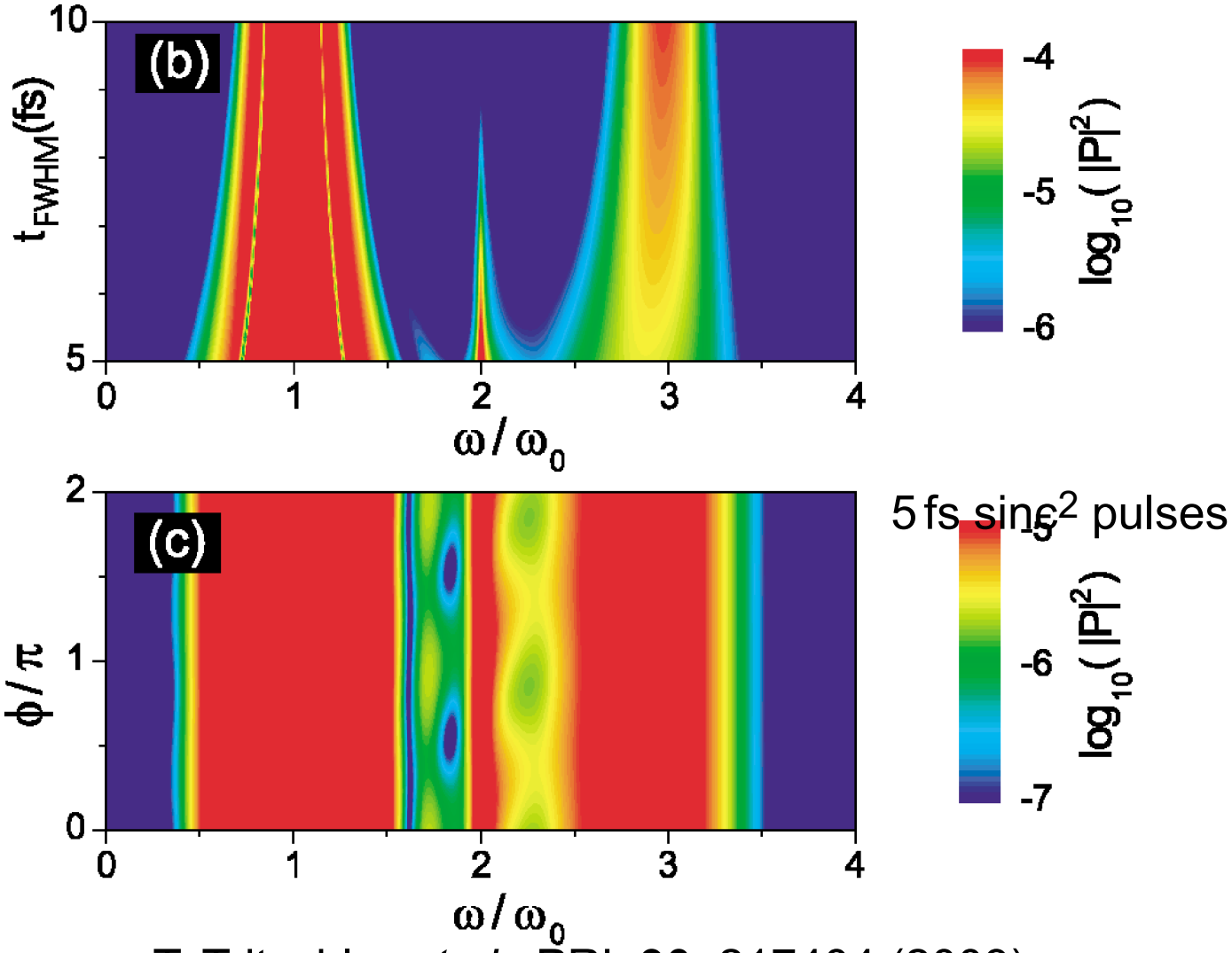
T. Tritschler *et al.*,
PRL **90**, 217404 (2003)

THG in disguise of SHG



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

THG in disguise of SHG

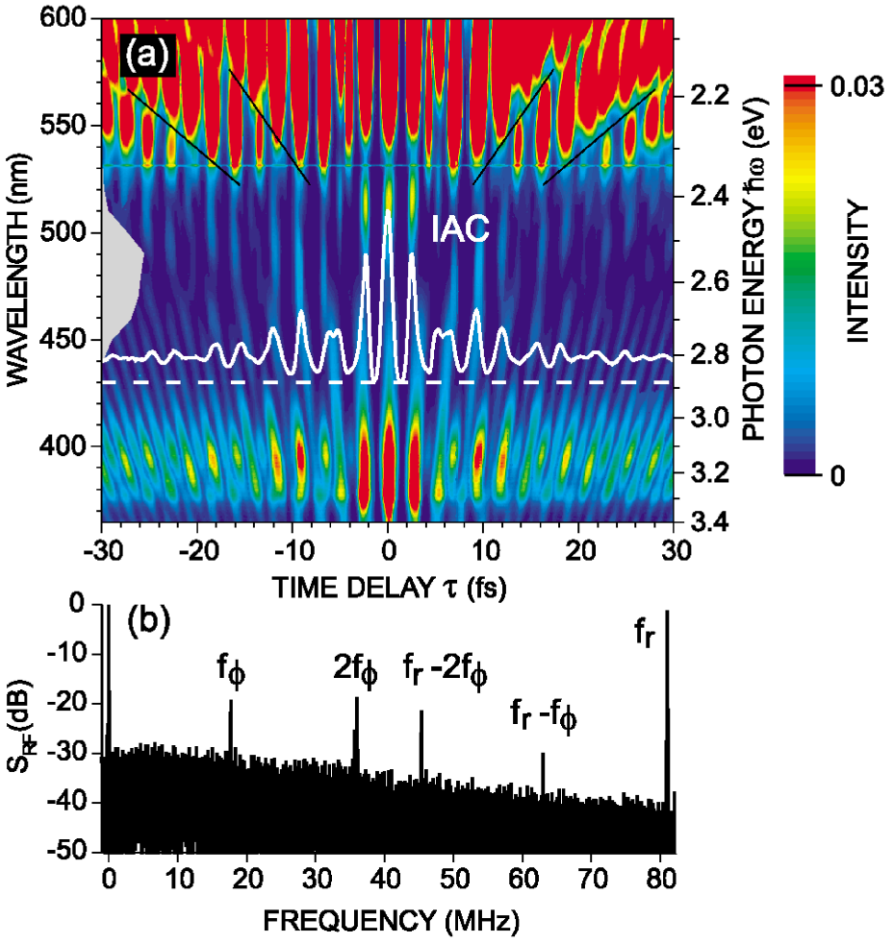


T. Tritschler *et al.*, PRL **90**, 217404 (2003)

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

THG in disguise of SHG

experiment



theory