Coherence in semiconductor nanostructures
Part IV: Notions of nonlinear spectroscopy & a particular experimental realization

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Outline

Linear Response

Polarization $\propto$ EM field

- A solution of Maxwell's equations:

$$\varepsilon(\mathbf{r}, t) = \varepsilon e^{i(\mathbf{kr}-\omega t)} + c.c. \quad (1)$$

- Induced macroscopic polarization:

$$P(\mathbf{r}, t) = \chi(\mathbf{r}, t)\varepsilon(\mathbf{r}, t) \quad (2)$$

- $\chi(\mathbf{r}, t)$: first-order susceptibility $\Rightarrow$ response of the matter to an electromagnetic field.
- The dielectric function ϵ and the complex refractive index n are related to it by:

$$\epsilon(\mathbf{r}, t) = \epsilon_0(1 + \chi(\mathbf{r}, t)), \;\; n = \sqrt{1 + \chi(\mathbf{r}, t)} \quad (3)$$

Absorption, Reflectance

Optical Nonlinear Phenomena

Induced polarization P scales with the 2nd, 3rd, ...M-th power of the impinging field $\mathcal{E}$

$\chi^{(2)}$ process	$\chi^{(3)}$ process	Higher order process
sum frequency generation	four-wave mixing	n-wave mixing
difference frequency generation	coherent Raman scattering	mode locking
optical retrification	phase modulation/conjugation	generation of continuum
parametric scattering	optical solitons	n-photon absorption

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VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

$$P=\chi E\left(1+\frac{E}{E_1}+\frac{E^2}{E_2^{\,2}}+\cdots\right),$$

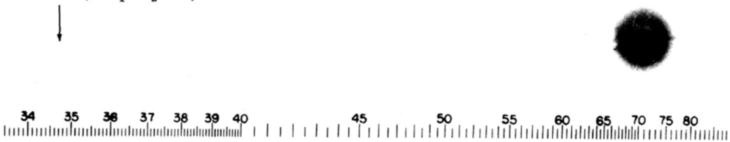

FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

Outline

Harmonic Generation in MoS_2

Multi-photon microscope: exciting $\hbar\omega$, detecting $2\hbar\omega$, $3\hbar\omega$, $5\hbar\omega$

A. Säynätjoki et al.Nature Communications 8, 893 (2017)

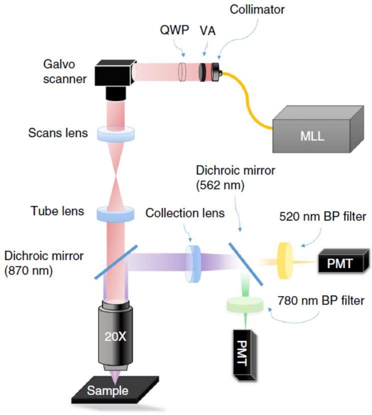

Fig. 2 Schematic diagram of multiphoton microscope. MLL, linearly

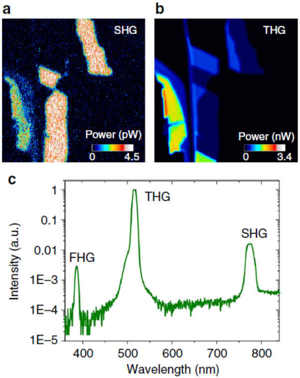

Fig. 3 Multiphoton images of MoS_2 flakes. **a** SHG and **b** THG map of the flakes in Fig. 1a. **c** Optical spectrum of the nonlinear signal from 1L-MoS_2 with a peak irradiance ~30 GW cm^{-2}

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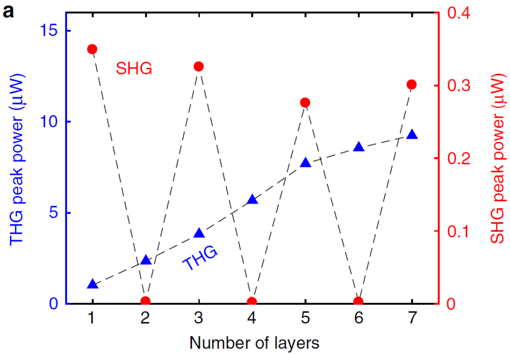

High-resolution imaging via multi-photon microscopy

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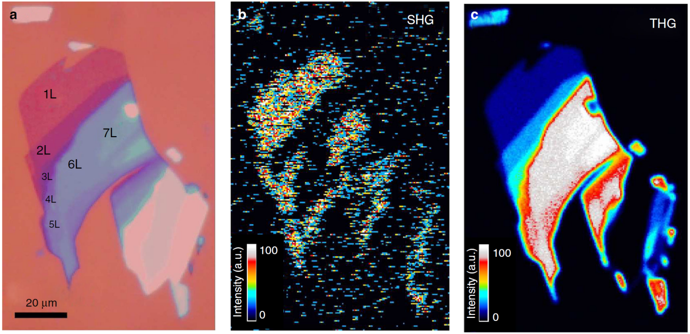


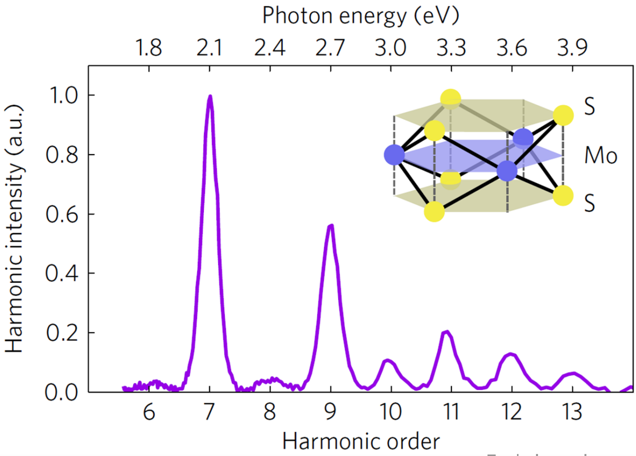
Fig. 6 Optical and multiphoton images of few-layer MoS_2 flake. **a** Optical micrograph, **b** SHG, and **c** THG images of flake with few-layer areas under 1560 nm excitation

High-resolution imaging via multi-photon microscopy

High-Harmonic Generation in MoS_2

Femto-second excitation: exciting $\hbar\omega = 0.3\,\text{eV}$, detecting up to $13\hbar\omega$

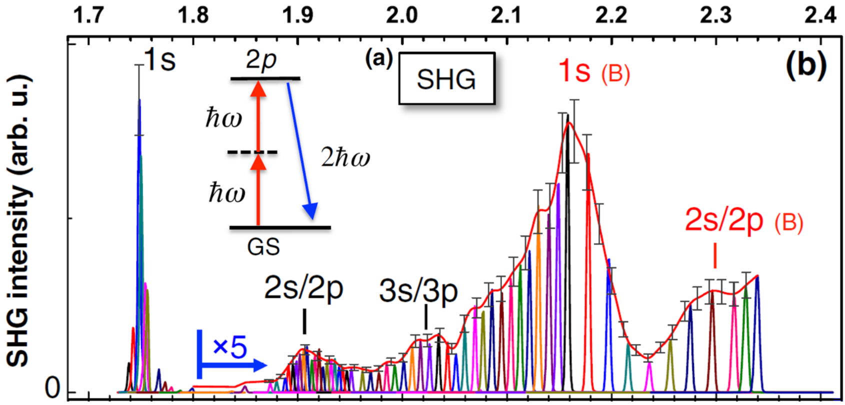
H. Liu et al.Nature Physics 13, 262 (2017)



Frequency-tuned Second Harmonic Generation

Level structure of excited states in WSe_2 monolayer

G. Wang et al. Phys. Rev. Lett. 114, 197403 (2015)



Toward the four-wave mixing

- In case of pair of fields: $\varepsilon(\mathbf{r}, t) = \varepsilon_1(\mathbf{r}_1, t) + \varepsilon_2(\mathbf{r}_2, t - \tau)$, one has:

$$P^{(\mathrm{N})} \propto \varepsilon_1^{a_1} \varepsilon_1^{*\,b_1} \varepsilon_2^{a_2} \varepsilon_2^{*\,b_2} \quad (4)$$

- For $\mathbf{N} = (0, 1, 2, 0)$, the resulting 3rd order polarization is:

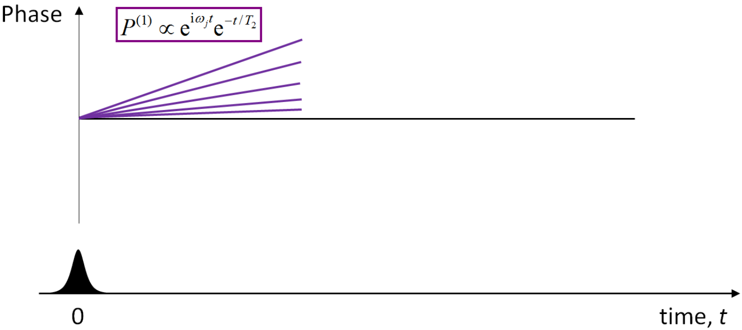
$$\mathrm{P}^{(3)} \propto R^{(3)}(\omega_3, t)\varepsilon_1^* \varepsilon_2^2 e^{-i(\mathbf{k_1}\mathbf{r}+\omega_1 t)} e^{2i(\mathbf{k_2}\mathbf{r}+\omega_2 t)} \propto \varepsilon_1^* \varepsilon_2^2 e^{+i[(2\mathbf{k_2}-\mathbf{k_1})\mathbf{r}+(2\omega_2-\omega_1)t]}, \ (5)$$

- Such $P^{(3)}$ propagates onto the $2\mathbf{k}_2 - \mathbf{k}_1$ direction and oscillates at $2\omega_2 - \omega_1$ frequency,
and is called degenerate **four-wave mixing** or **FWM**.

Outline

Why it is worth to look into FWM$\propto \mathcal{E}_1^* \mathcal{E}_2 \mathcal{E}_2$?

Because it offers an access to homogeneous dephasing time $\mathrm{T}_2 = 2\hbar/\gamma$
in the presence of spectral inhomogeneous broadening σ
via formation of a **photon echo**



Rephasing of all polarizations at $t = 2\tau \Rightarrow$

FWM is only sensitive to microscopic dephasing, independent of σ.
σ is inferred through the time-spread of the echo.

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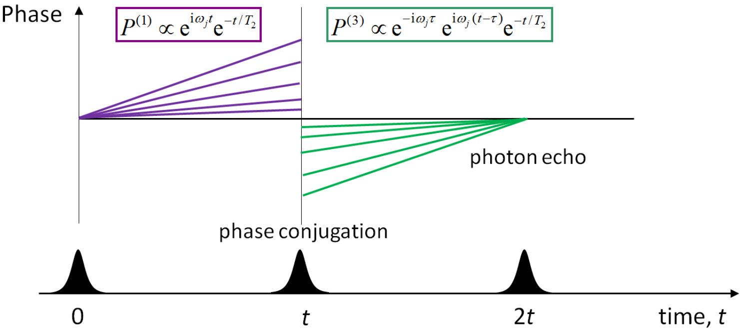

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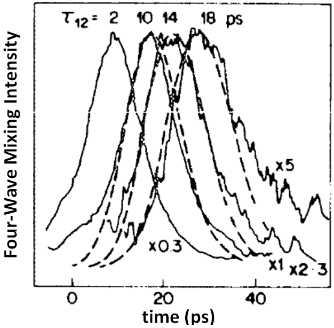
Photon echoes from two-dimensional excitons in GaAs-AlGaAs quantum wells

L. Schultheis[a] and M. D. Sturge J. Hegarty

Appl. Phys. Lett. **47** (9), 1 November 1985



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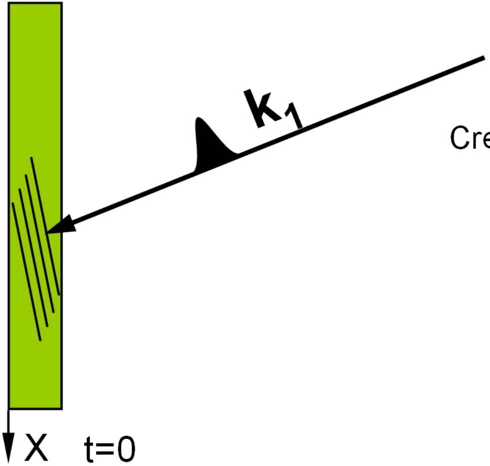
How to measure FWM ?

Issues: 1. spatial averaging, 2. fails for nano-objects like single excitons



Creation of the polarization along direction k_1

$\sim e^{i(k_1 x - \omega t)} e^{-t/T_2}$

T_2 – dephasing time

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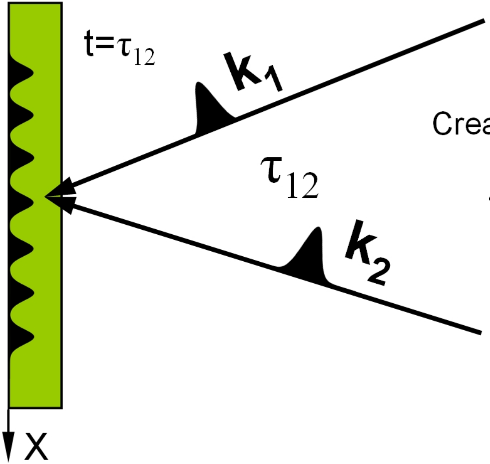

Creation of the density grating

$$\sim\cos[(k_2-k_1)x]\cdot e^{-(t-\tau_{12})/T_2}$$

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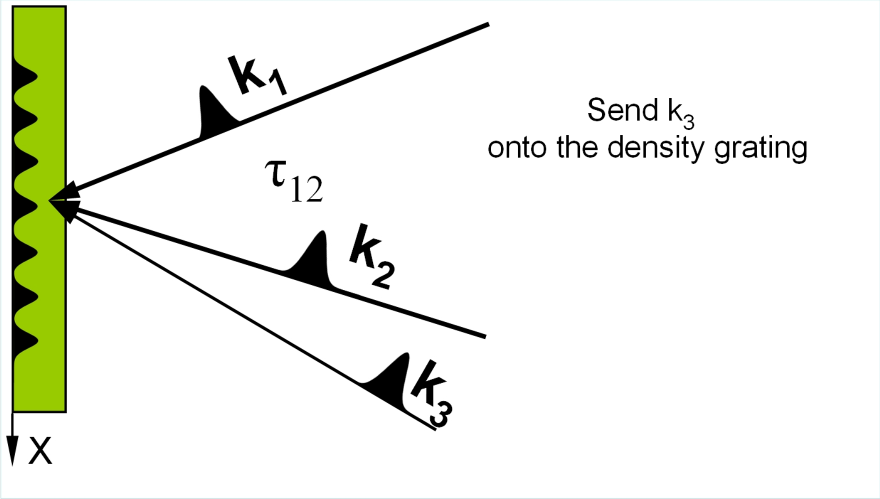

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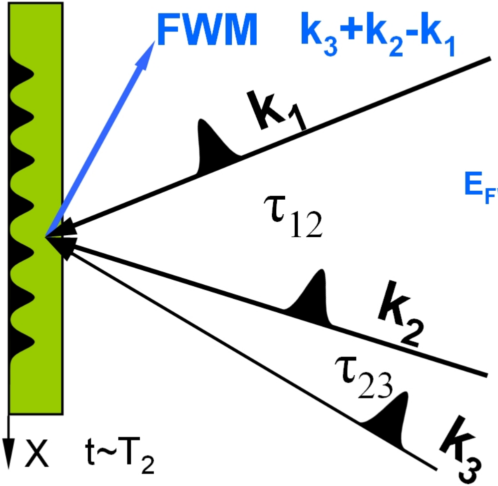

Diffration of k_3
on the density grating
E_{FWM} **in direction $k_3+k_2-k_1$**

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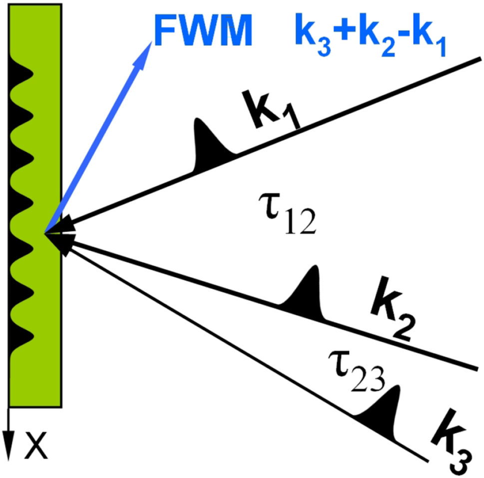

diffraction of $\mathbf{k_3}$ by the density grating created by $\mathbf{k_1}$ and $\mathbf{k_2}$

FWM$(\tau_{12}) \rightarrow$ coherence dynamics, dephasing time

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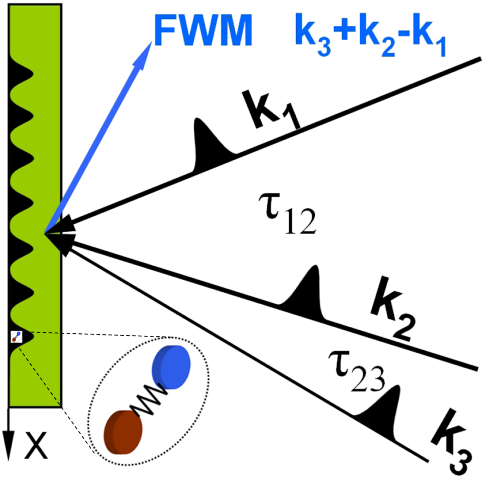

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? How to extract FWM of a localized state ?

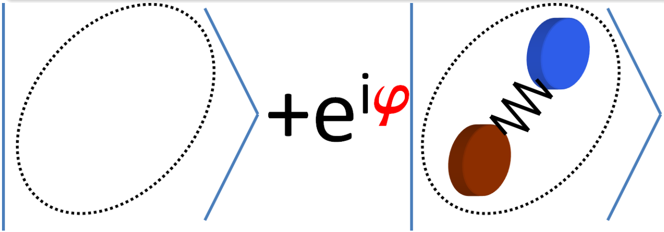
Microscopy required $\Rightarrow$ colinear arrangement of $\mathcal{E}_{1,2,3}$ $\Rightarrow$

Optical Heterodyning

$$\mathrm{E}_{\mathrm{FWM}} \propto \exp[\imath(\mathbf{k}_{\mathbf{FWM}}\mathbf{x} - \omega_{\mathrm{FWM}}\mathrm{t})]$$

distinct by directions - $\mathbf{k} \Longrightarrow \omega$ - distinct in frequency

spatial homogeneity - $\mathbf{x} \Longrightarrow \mathrm{t}$ - temporal invariance



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FWM retrieval by spectral interference of the reference field with the heterodyne beat at the FWM frequency

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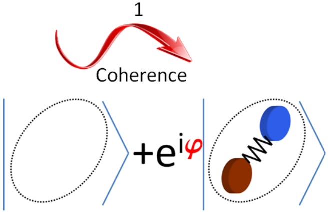

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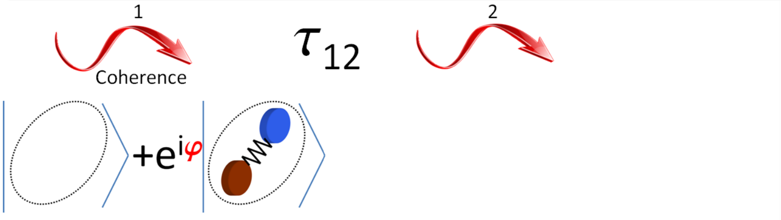

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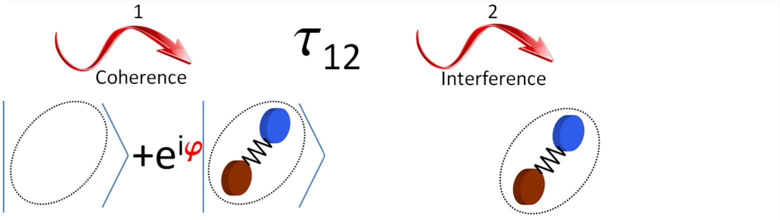

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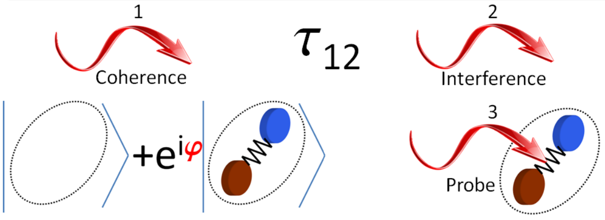

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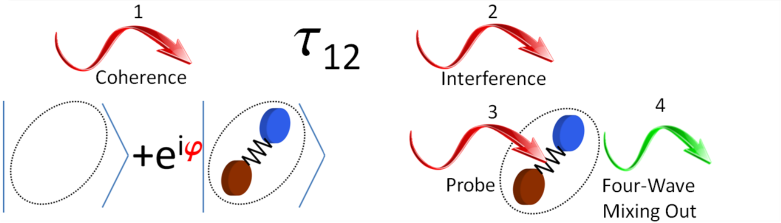

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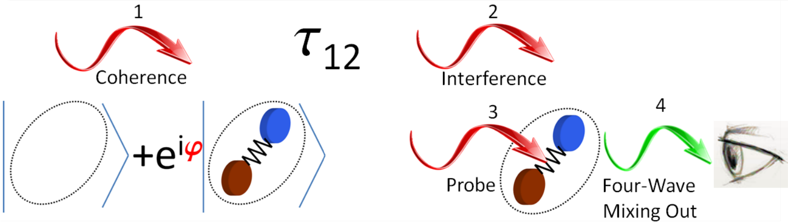

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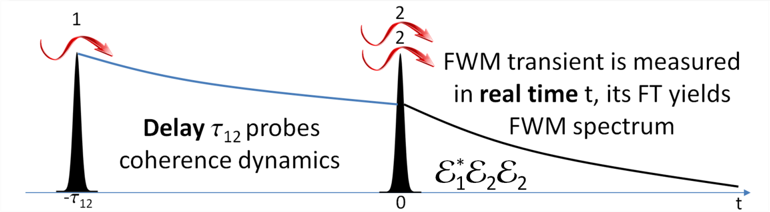

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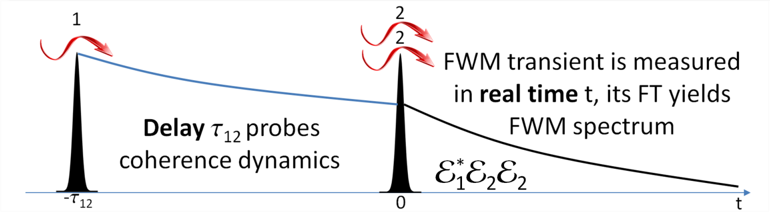

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Outline

Heterodyne FWM in 3 equations

- Pulse train: N delta pulses centered at ω with the repetition rate τ^{-1}:

$$\mathcal{E}(t) = A(t)e^{-i\omega t} + A(t-\tau)e^{-i\omega t} + A(t-2\tau)e^{-i\omega t} + \ldots = \sum_{n=0}^{N} A(t-n\tau)e^{-i\omega t}$$

- One needs a proper phase shifter at frequency θ acting on a pulse train:

$$\mathcal{E}(t) = A(t)e^{-i\omega t} + A(t-\tau)e^{-i(\omega t+\theta\tau)} + \ldots = e^{-i\omega t}\sum_{n=0}^{N} e^{-in\theta\tau}A(t-n\tau)$$

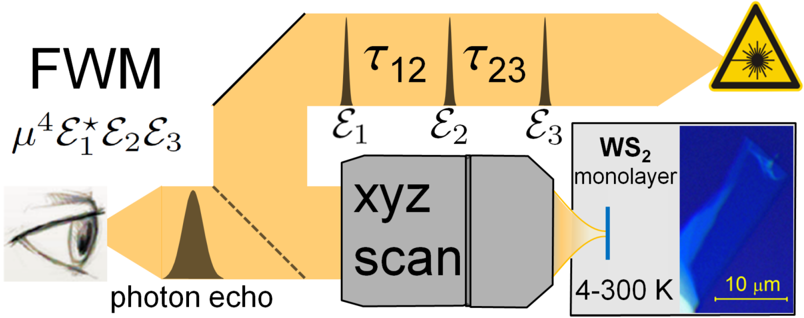
- Induced FWM response:

$$R^{(-1,2)}(t) \propto \mathcal{E}_1^*(t)\mathcal{E}_2^2(t) = e^{i\omega t}\sum_n A_1^*(t-n\tau)e^{in\theta_1\tau}e^{-2i\omega t}\sum_m A_2^2(t-m\tau)e^{-2im\theta_2\tau} =$$

$$e^{-i\omega t}\sum_n A_1^*(t-n\tau)A_2^2(t-n\tau)e^{-in(2\theta_2-\theta_1)\tau} = e^{-i\omega t}\sum_n A_{FWM}(t-n\tau)e^{-in\theta_{FWM}\tau}$$

FWM micro-spectroscopy ⇒ optical lock-in

W. Langbein et al.Optics Letters 31, 1151 (2006), **intensly exploited in Grenoble**



3-beam heterodyne detection & spectral interferometry

Measurement of the exciton polarization and density dynamics with an enhanced spatio-temporal resolution: (100 fs, 300 nm)

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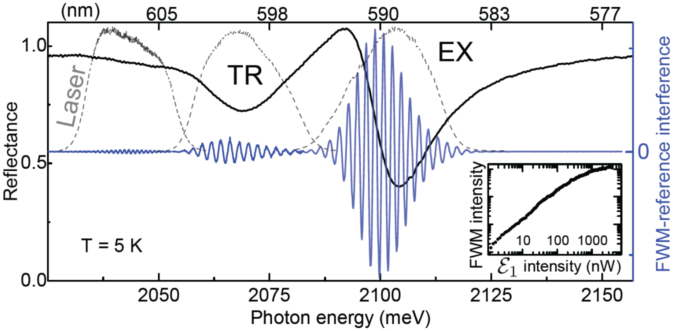

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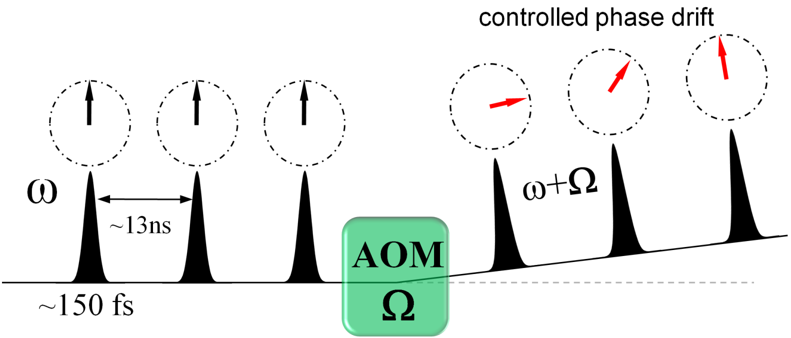
Measurement of the exciton polarization and density dynamics with an enhanced spatio-temporal resolution: (100 fs, 300 nm)

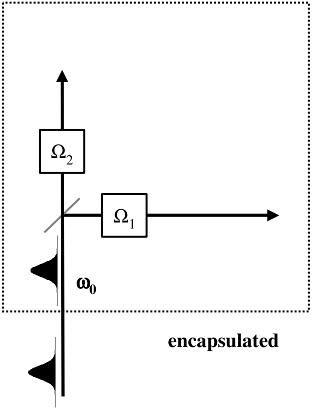
FWM micro-spectroscopy ⇒ in practice

Impact of an **A**cousto-**O**ptic **M**odulator on relative phases of the consecutive pulses within a pulse train



Experimental setup

Heterodyne FMW



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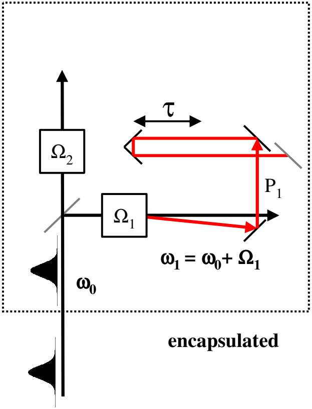

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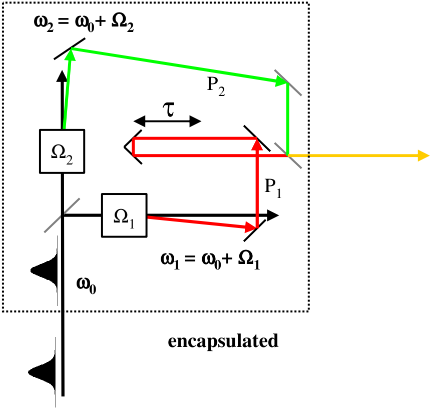

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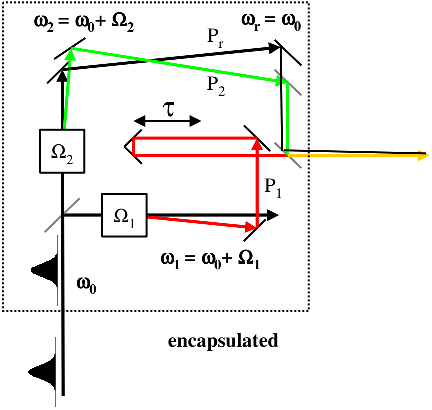

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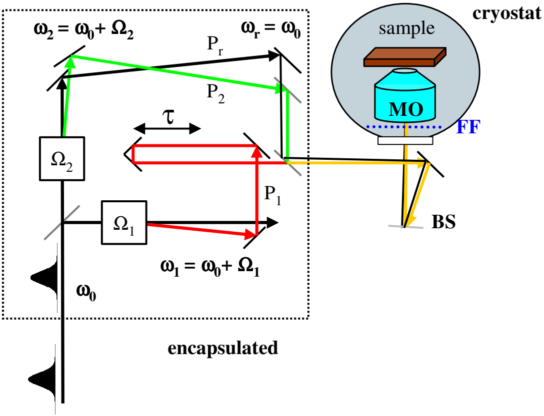

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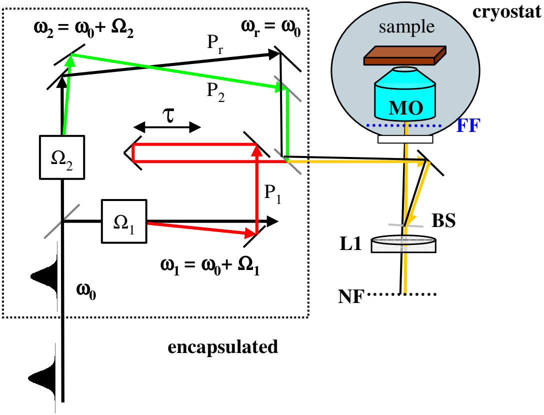

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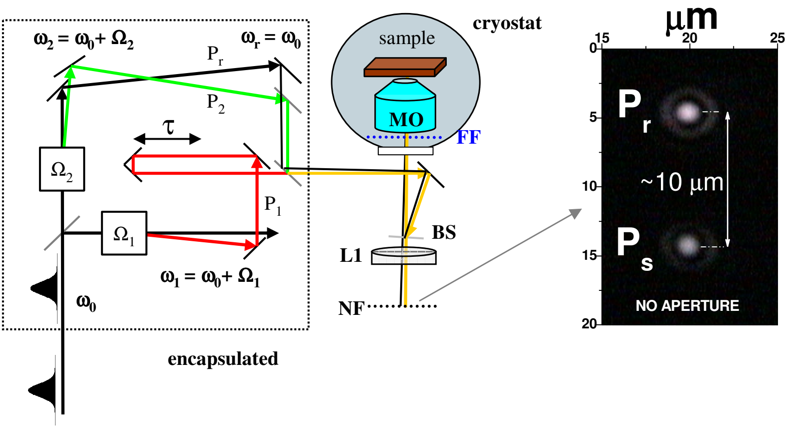

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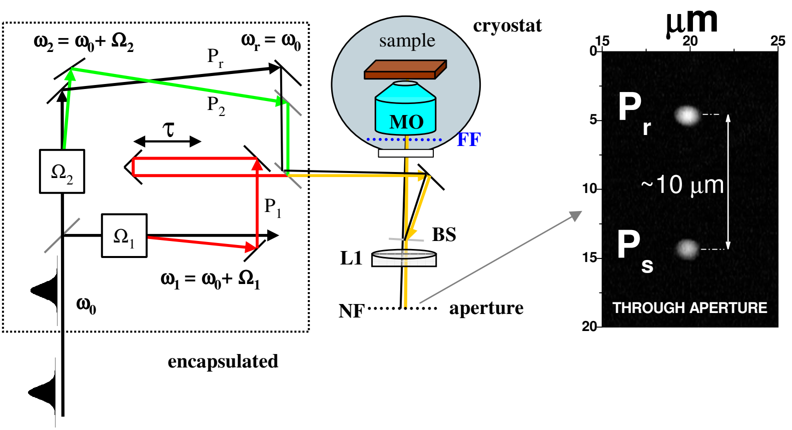

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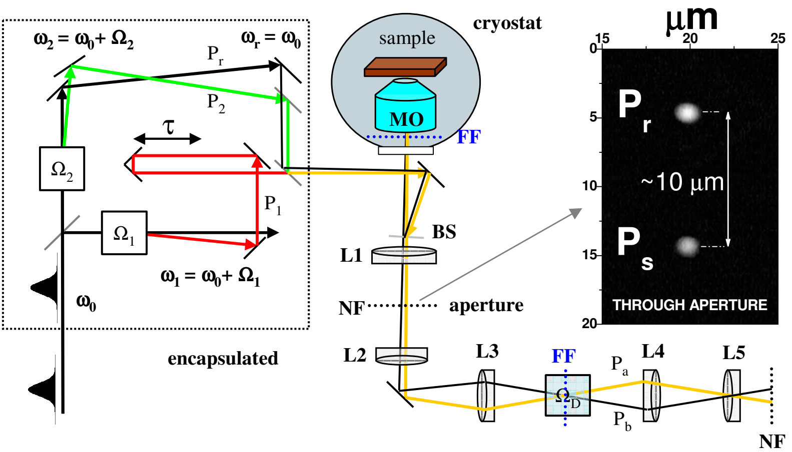

Experimental setup

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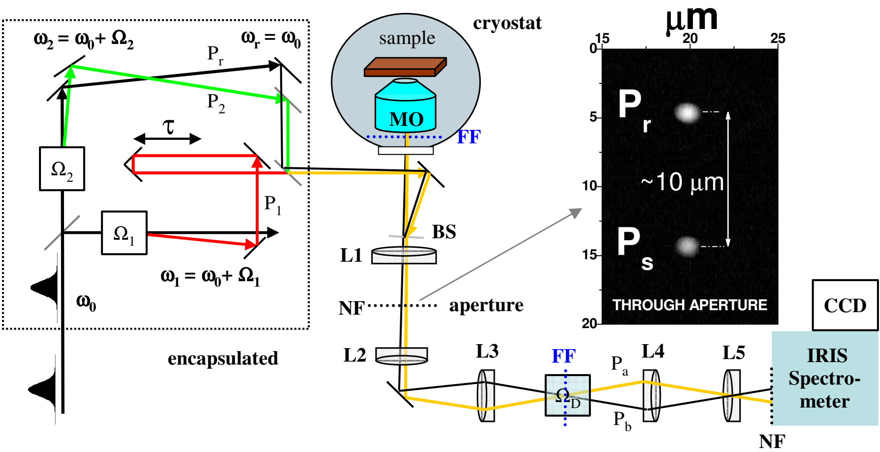
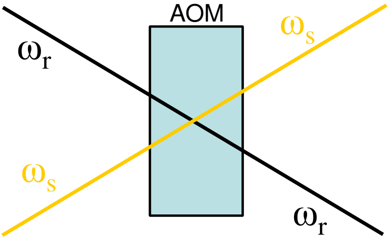


Detection Scheme

Spectral Interferometry with an AOM

$P_{a,b}$ contains interference term

$$2P_{a,b}(\omega, t) = |E_r|^2 + |E_s|^2 \pm 2\Re(E_r \cdot E_s^* \cdot e^{\imath\Omega_D t})$$



Balanced detection is filtering the signal at Ω_D

$$P_D(\omega) = P_a - P_b = 2\int_0^T \Re(E_r \cdot E_s^* \cdot e^{\imath\Omega_D t})dt$$

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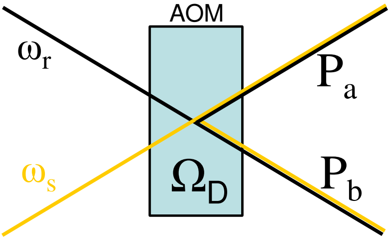

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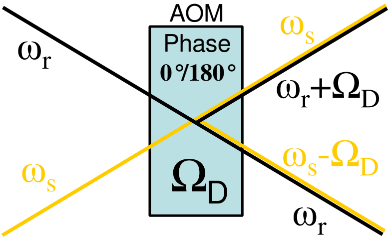

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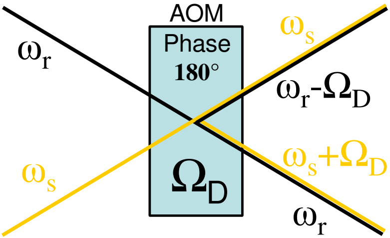

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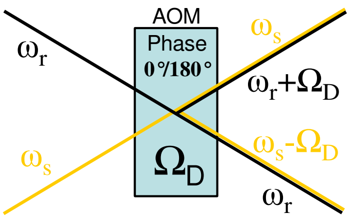
Detection Scheme

Signal selection in a mixing AOM



Ω_D

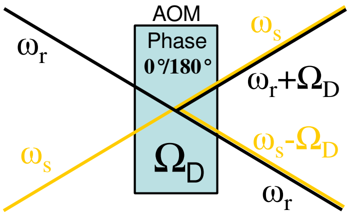
- Ω_1
- Ω_2
- $2\Omega_1 - \Omega_2$
- $3\Omega_1 - 2\Omega_2$...

Retrieves

- Pomp
- Probe
- FWM
- SWM...

Detection Scheme

Signal selection in a mixing AOM



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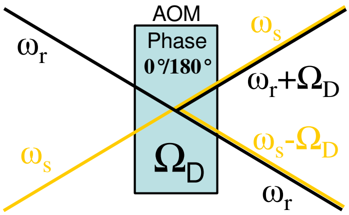

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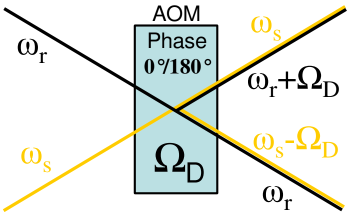

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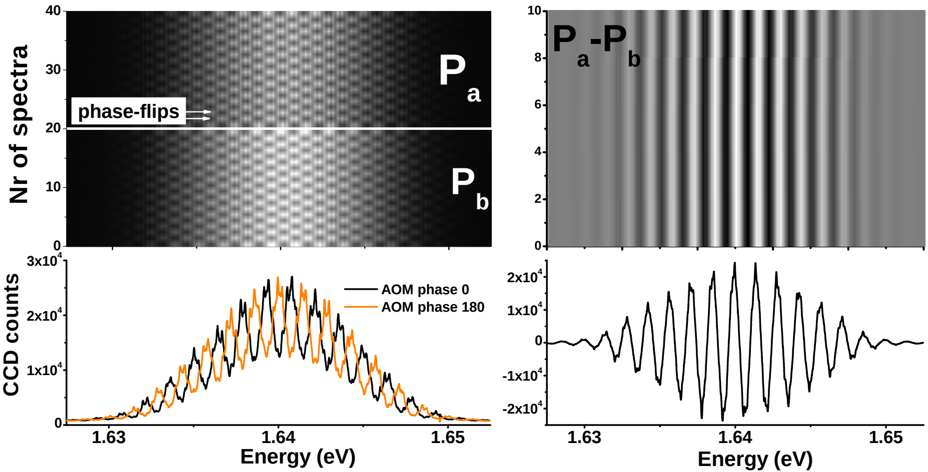
Detection Scheme

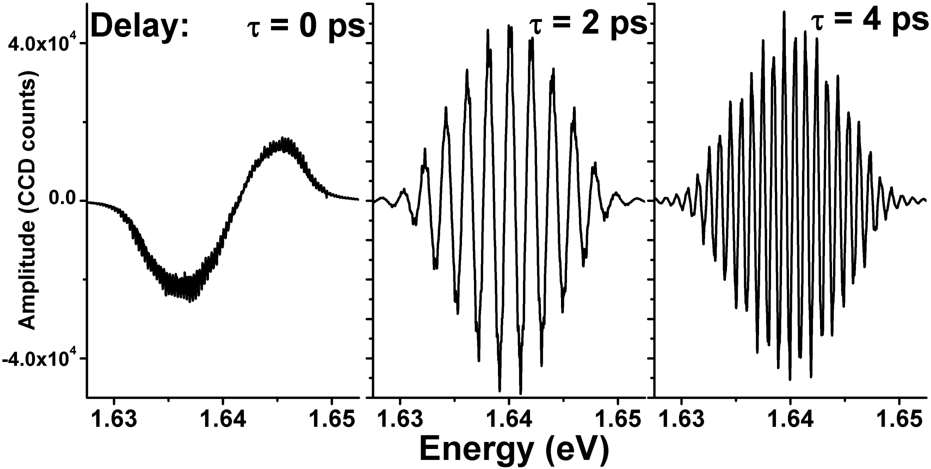
Balanced detection in a mixing AOM

pump-reference interference



Detection Scheme

Intereference period vs. delay, $\propto \tau^{-1}$



Detection Scheme

Signal retrieval in **amplitude** and **phase**

$$P_D(\omega) = 2\int_0^T \Re(E_r \cdot E_s^* \cdot e^{\imath \Omega_D t})dt \text{ jest rzeczywista}$$

Detection Scheme

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↓ Transformata Fouriera do t

$$F^{-1}(P_D) \text{ zawiera 2 czasowo odwrócone składniki}$$

Detection Scheme

Signal retrieval in **amplitude** and **phase**

$P_D(\omega) = 2\int_0^T \Re(E_r \cdot E_s^* \cdot e^{\imath\Omega_D t})dt$ jest rzeczywista

↓ Transformata Fouriera do t

$F^{-1}(P_D)$ zawiera 2 czasowo odwrócone składniki

↓ Zasada przyczynowości

Tylko dodatnie czasy są fizyczne: $\Theta(t)[F^{-1}(P_D)]$

Detection Scheme

Signal retrieval in **amplitude** and **phase**

$P_D(\omega) = 2\int_0^T \Re(E_r \cdot E_s^* \cdot e^{\imath\Omega_D t})dt$ jest rzeczywista

↓ FT to t

$F^{-1}(P_D)$ zawiera 2 czasowo odwrócone składniki

↓ Causality Principle

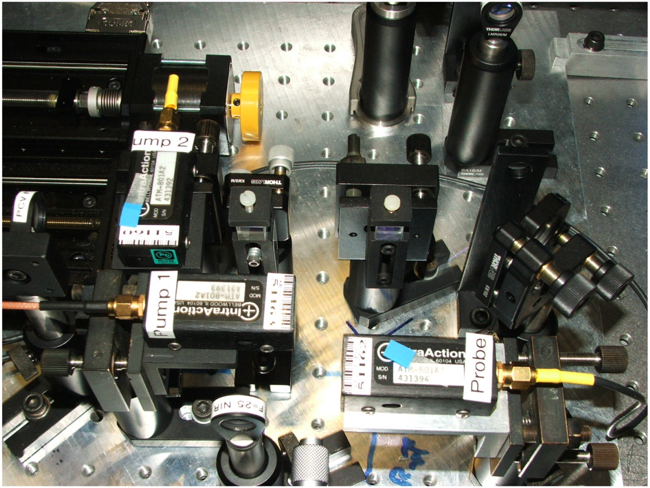
Tylko dodatnie czasy są fizyczne: $\Theta(t)[F^{-1}(P_D)]$

↓ Back FT to ω

$E_{sign}(\omega)$ **w amplitudzie i fazie**

Photos of the setup

Acousto-**O**ptic **M**odulators used as frequency shifters

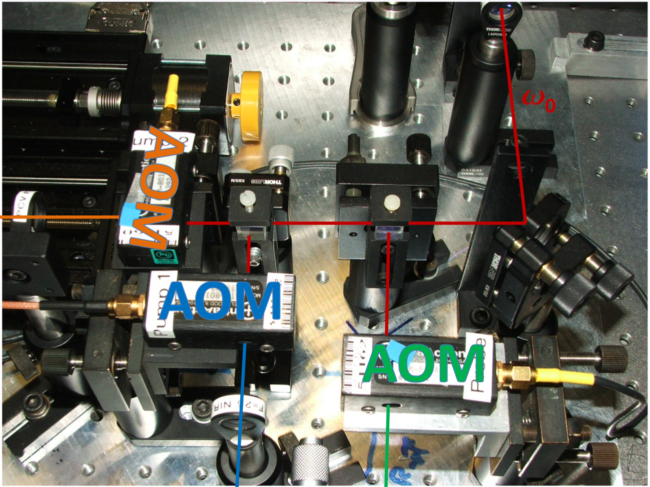


Photos of the setup

Acousto-Optic Modulators used as frequency shifters

ω_0

$+\Omega_3$



optical freqency → $\omega_0 + \Omega_2$ $\omega_0 + \Omega_1$ ← radio freqency

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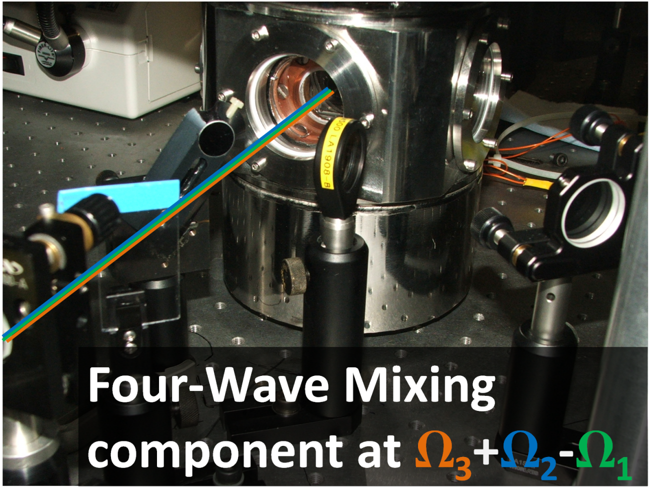

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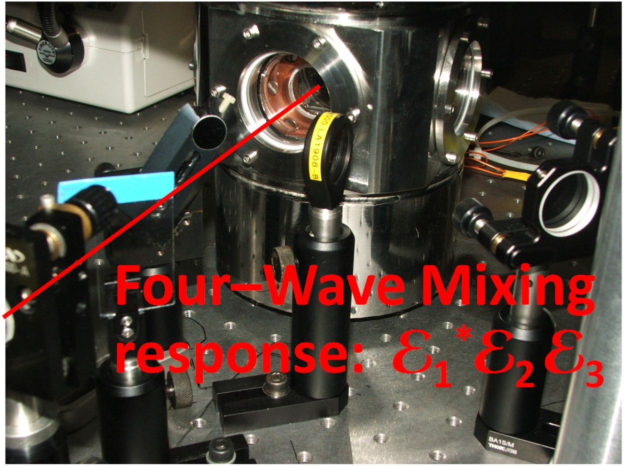

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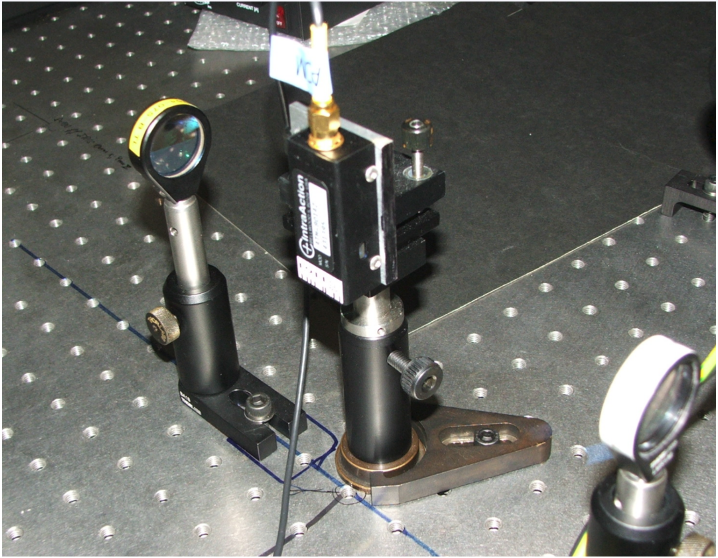

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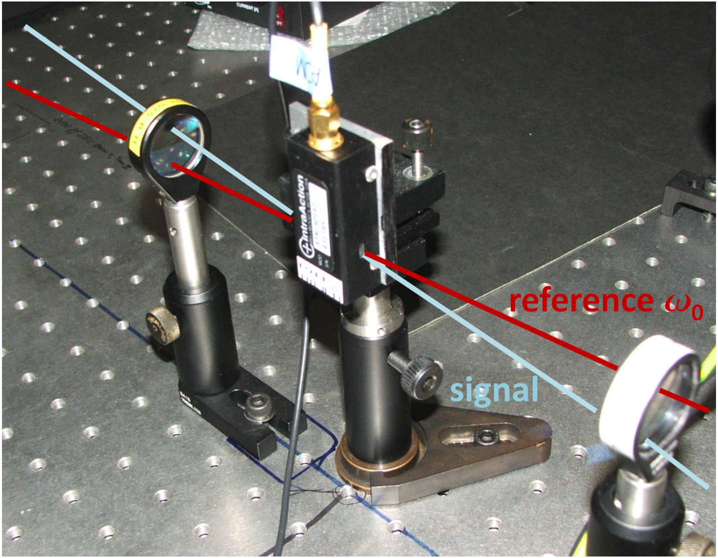

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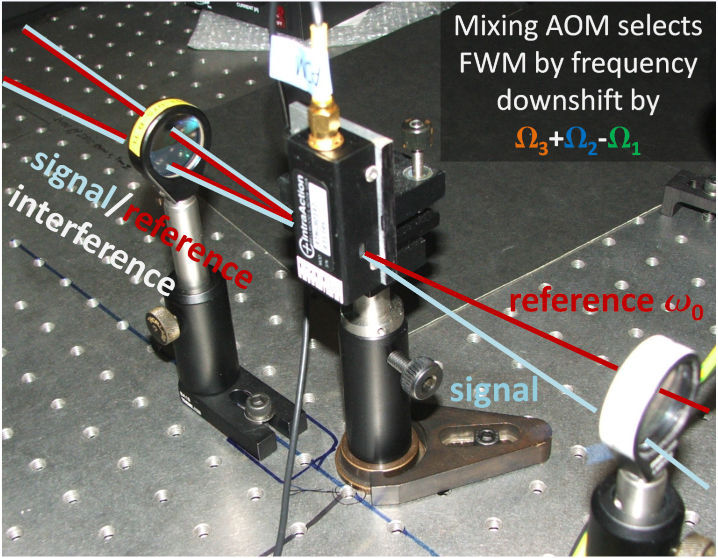

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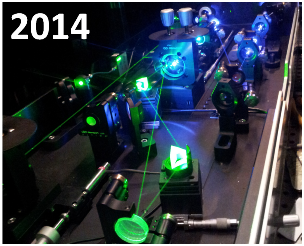

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