

# A Direct Approach to Electromagnetic Quantum Fluctuations

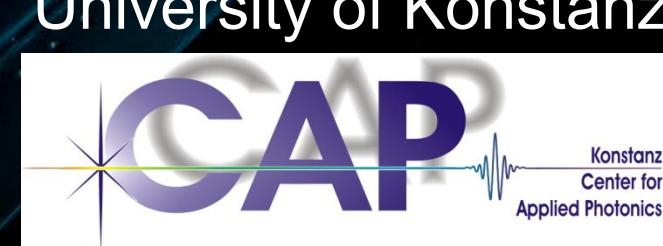
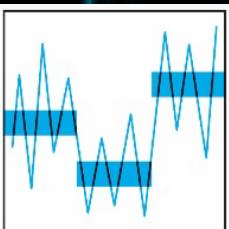
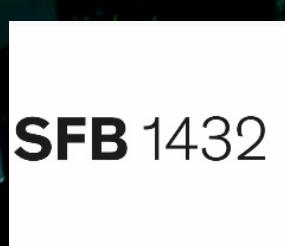
**Experiments:** C. Beckh, S. Haller, E. Hils, S. Hutter, H. Kempf,  
K. Oguchi, C. Riek, P. Sulzer, R. Tenne (now at TECHNION),  
D. V. Seletskiy (now at EP Montréal → UNM Albuquerque)

**Theory:** M. Kizmann, T. L. M. Guedes, E. Hubenschmidt, G. Burkard,  
A. S. Moskalenko (now at KAIST), S. Mukamel (UC Irvine),  
S. Onoe, T. C. Ralph (U Queensland)

Alfred Leitenstorfer

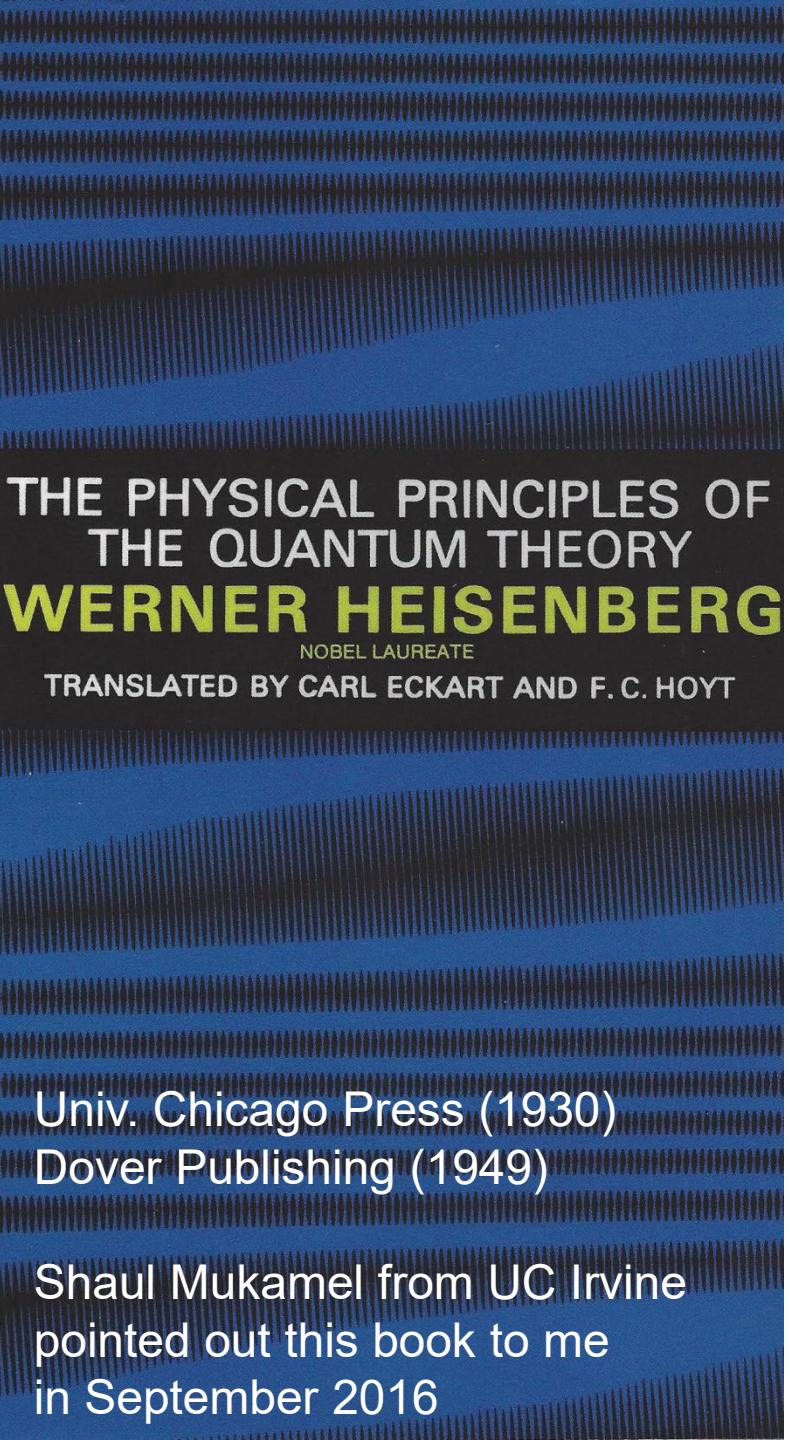
**OPO:** A. Muraviev,  
K. Vodopyanov (CREOL)

Department of Physics and  
Center for Applied Photonics



# Vacuum Fluctuations of Electromagnetic Field

- quantum physics: **uncertainty relations**, e.g.
  - position-momentum:  $\Delta x \cdot \Delta p \geq \hbar/2$  with reduced Planck constant
  - time-energy:  $\Delta t \cdot \Delta E \geq \hbar/2$   $\hbar = h/2\pi = 1.055 \cdot 10^{-34}$  Js
  - **Emmy Noether, 1915:**
    - symmetry** of system  $\leftrightarrow$  **constant of motion**, e.g.
    - homogeneity** of **space**  $\leftrightarrow$  conservation of **momentum**
    - homogeneity** of **time**  $\leftrightarrow$  conservation of **energy**
- **wave-like quantities**, e.g. electromagnetic field?



THE PHYSICAL PRINCIPLES OF  
THE QUANTUM THEORY  
**WERNER HEISENBERG**

NOBEL LAUREATE

TRANSLATED BY CARL ECKART AND F. C. HOYT

Univ. Chicago Press (1930)  
Dover Publishing (1949)

Shaul Mukamel from UC Irvine  
pointed out this book to me  
in September 2016

### III. §1. THE UNCERTAINTY RELATIONS FOR WAVES

Before proceeding to the subject proper, however, we must first discuss briefly what is meant by an exact knowledge of a wave amplitude—for instance, that of an electric or magnetic field strength. Such an exact knowledge of the amplitude at every point of a region of space (in the strict mathematical sense) is obviously an abstraction that can never be realized. For every measurement can yield only an average value of the amplitude in a very small region of space and during a very short interval of time. Although it is perhaps possible in principle to diminish these space and time intervals without limit by refinement of the measuring instruments, nevertheless for the physical discussion of the concepts of the wave theory it is advantageous to introduce finite values for the space and time intervals involved in the measurements and only pass to the limit zero for these intervals at the end of the calculations.

$$\Delta E_x \Delta H_v \geq \frac{hc}{\delta v \delta l} = \frac{hc}{(\delta l)^4}$$

# Vacuum Fluctuations of Electromagnetic Field

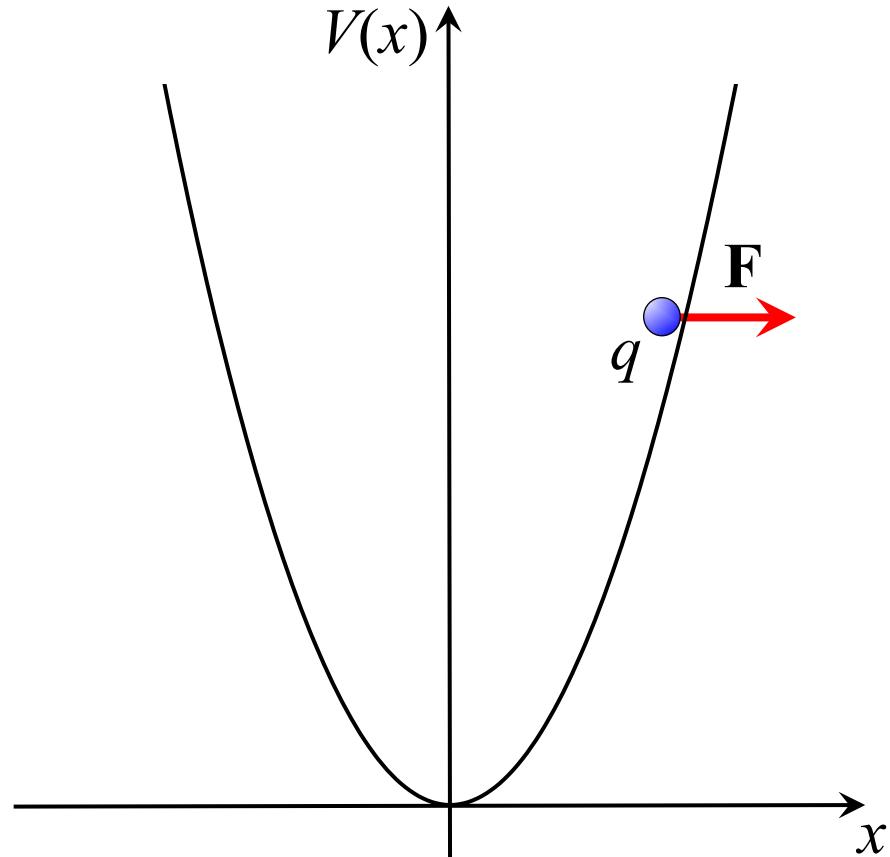
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  - time-energy:  $\Delta t \cdot \Delta E \geq \hbar/2$   $\hbar = h/2\pi = 1.055 \cdot 10^{-34}$  Js
  - **Emmy Noether, 1915:**
    - symmetry of system  $\leftrightarrow$  conservation law, e.g.
    - homogeneity of space  $\leftrightarrow$  conservation of momentum
    - homogeneity of time  $\leftrightarrow$  conservation of energy
- **wave-like quantities**, e.g. electromagnetic field?
  - **Heisenberg, 1929:**  $\Delta E_x \Delta H_y \geq \frac{hc}{\delta v \delta l} = \frac{hc}{(\delta l)^4}$
  - **Bohr and Rosenfeld, 1933:** general formulation including correlations in space and time according to Maxwell, Kgl. Danske Vid. Sels., Math.-fys. Medd. **XII**, 8 (1933)

# **Part I**

Are those vacuum fluctuations just an auxiliary quantity for theorists or are they real?

**Experiment!**

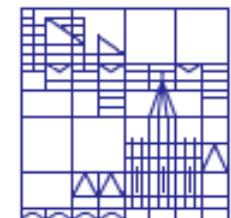
# Concept for Measuring Quantum Electric Field



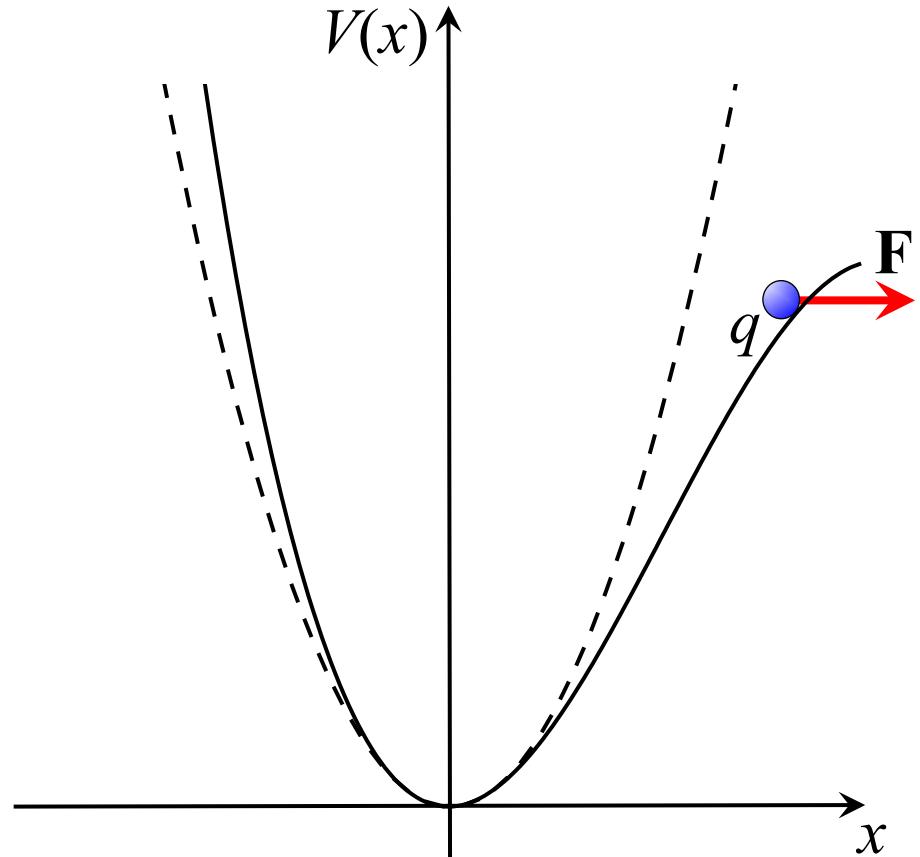
- fundamental tool: **test charge**  $q$
- **force** by electric field:  $\mathbf{F} = q\mathbf{E}$
- **bound charge** with  $\omega_{res} \gg \Omega_{THz}$   
displacement  $\mathbf{x} = k^{-1}\mathbf{F} = (q/k)\mathbf{E}$
- **vacuum field**  $\Delta E_{vac} \approx 20 \text{ V/cm}$   
for  $c\delta l \approx 10 \text{ fs} \rightarrow \sqrt{\langle x^2 \rangle} < 10^{-17} \text{ m}$   
in semiconductor like  $\text{AgGaS}_2$
- not accessible with elementary point charge (single electron!) due to uncertainty arguments

➤ average over **many electrons** in finite space-time volume  $(\delta l)^4$

compare discussions in L. Landau, R. Peierls, Z. Phys. **69**, 56 (1931) and in  
N. Bohr, L. Rosenfeld, Kgl. Danske Vid. Sels., Math.-fys. Medd. **XII**, 8 (1933)



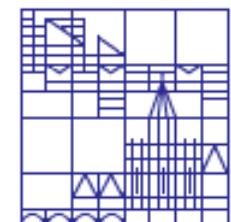
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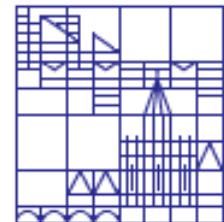
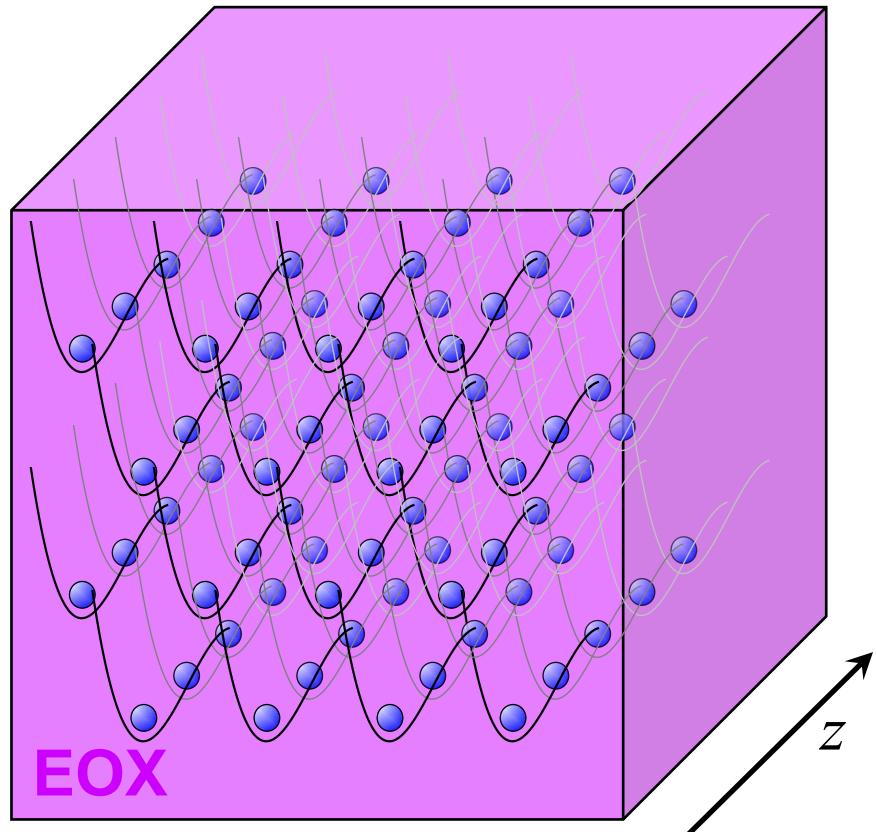
- average over **many electrons** in finite space-time volume  $(\delta l)^4$
- use **many photons** to probe e.g. refractive index change  
due to **anharmonicity** of electronic binding potential  $V(x)$

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# Detection of Electric Field: Transverse Pockels Effect

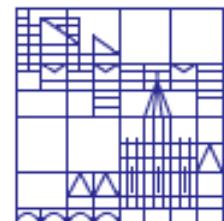
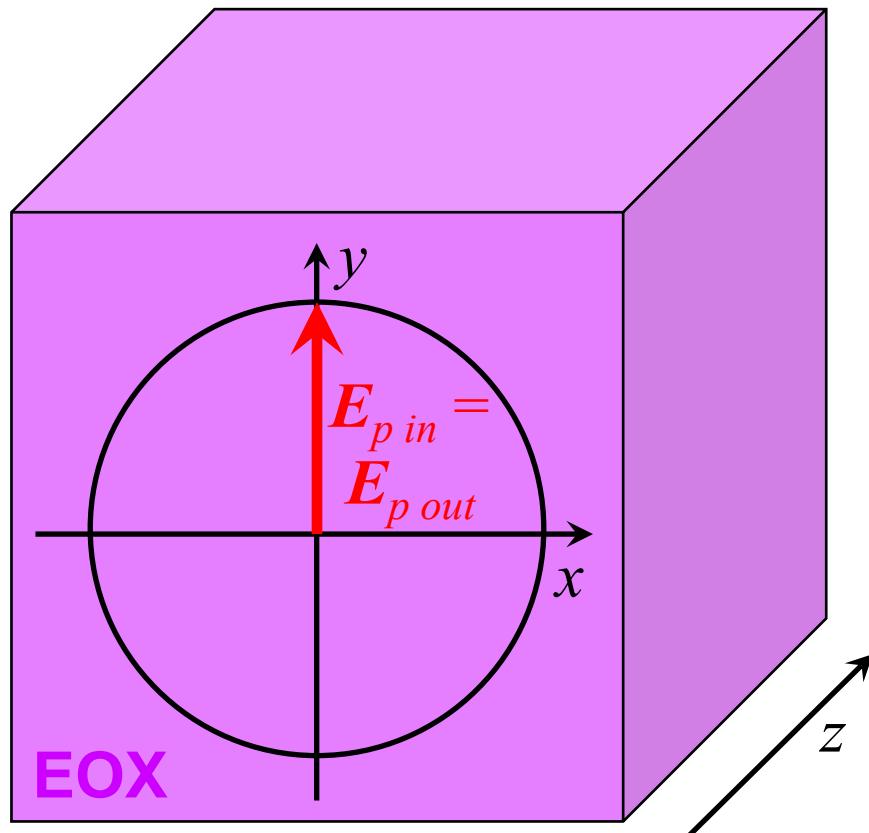
- sampling geometry with **electro-optic crystal EOX** ( $\text{ZnTe}$ ,  $\text{AgGaS}_2$ ):



S. Namba, J. Opt. Soc. Am. **51**, 76 (1961)

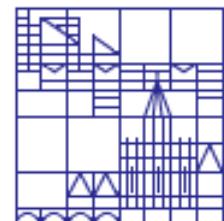
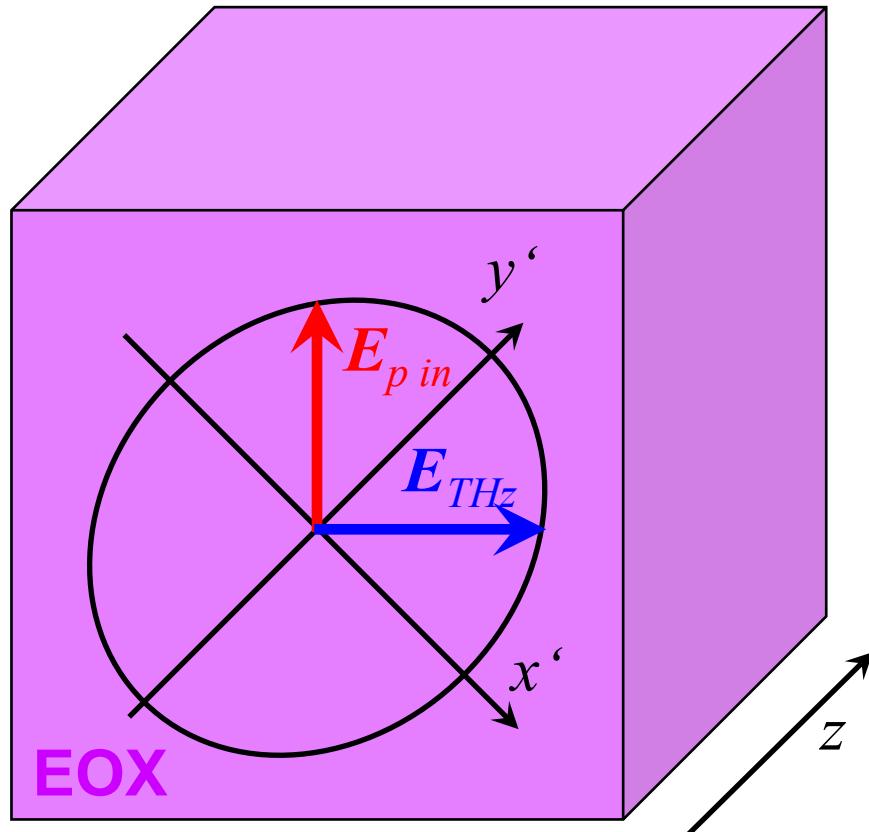
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no birefringence when  $E_{THz} = 0$ , i.e.  $n_x = n_y = n_0$



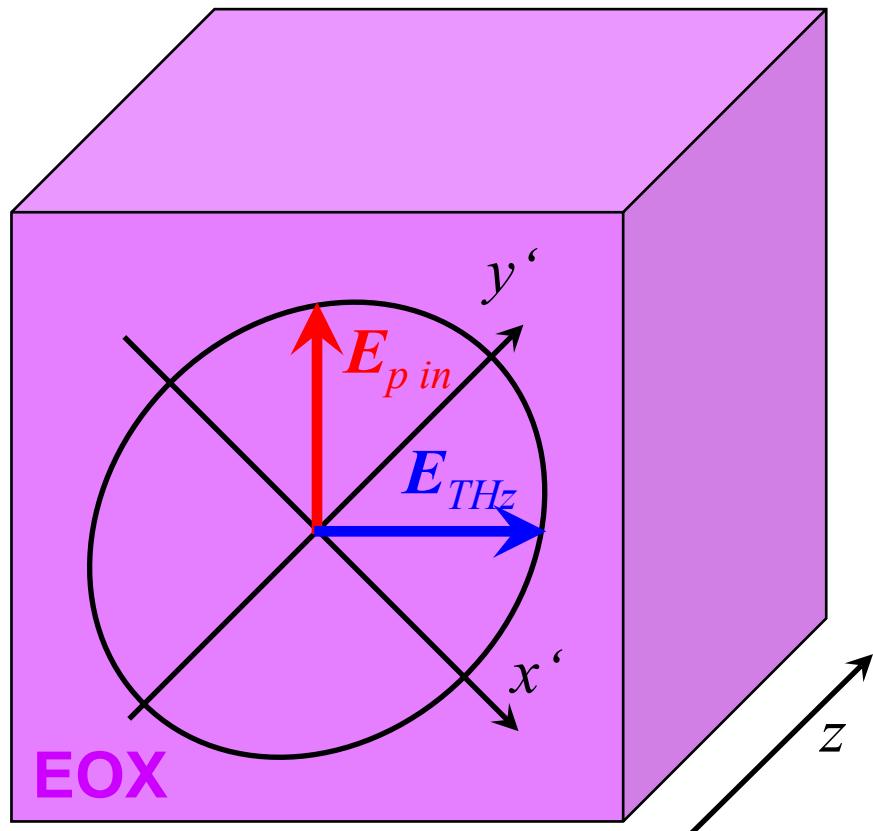
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  - when  $E_{THz} \neq 0$ : principal axes  $x'$ ,  $y'$ ,  $n_{x'} = n_0 - \Delta n$ ,  $n_{y'} = n_0 + \Delta n$

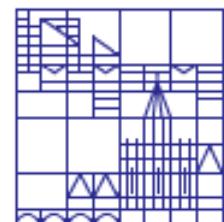


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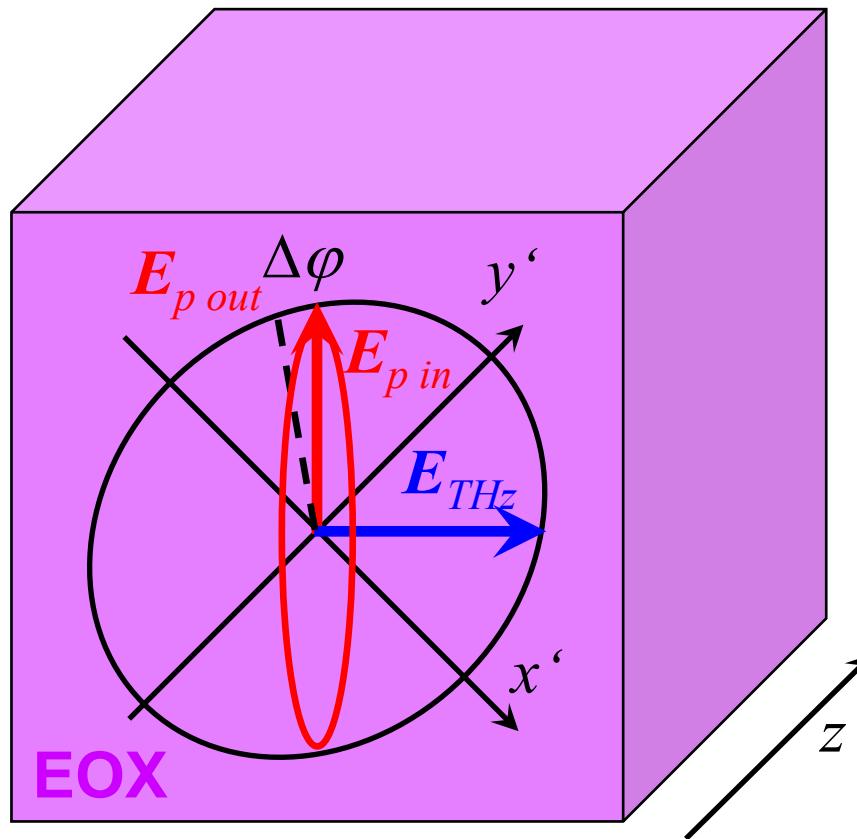


- electro-optic effect:  $\Delta n \sim r_{41} E_{THz}$
- co-propagation with  $E_{THz}$  in EOX:  
components of  $E_p$  pick up  
different **phase shift** in  $x'$  and  $y'$



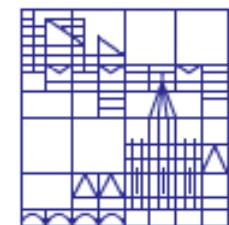
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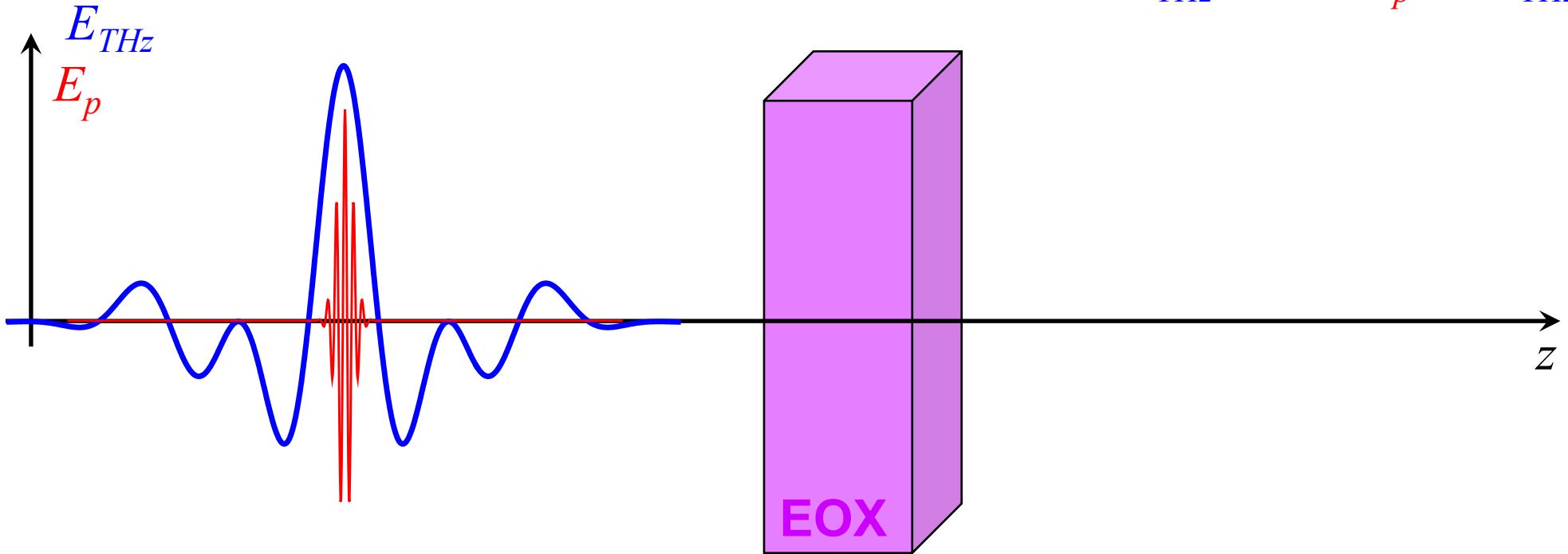
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components of  $E_p$  pick up  
**different phase shift** in  $x'$  and  $y'$
- after EOX: probe  $E_p$  elliptically  
polarized → ellipticity  $\Delta\varphi$
- ellipsometry with  $\lambda/4$  wave plate,  
Wollaston polarizer  
and optical bridge:

$$\Delta I_p/I_p \sim \Delta n \sim E_{THz}$$

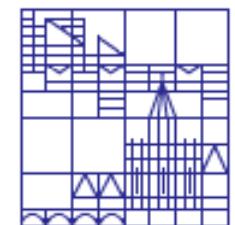


# Sub-Cycle Sampling of Electric Field

- ultrashort probe wave packet, intensity envelope  $\Delta t_p < (2 \nu_{THz})^{-1}$ : picks up electro-optic phase shift from local  $E_{THz}$ , note:  $\nu_p \gg \nu_{THz}$

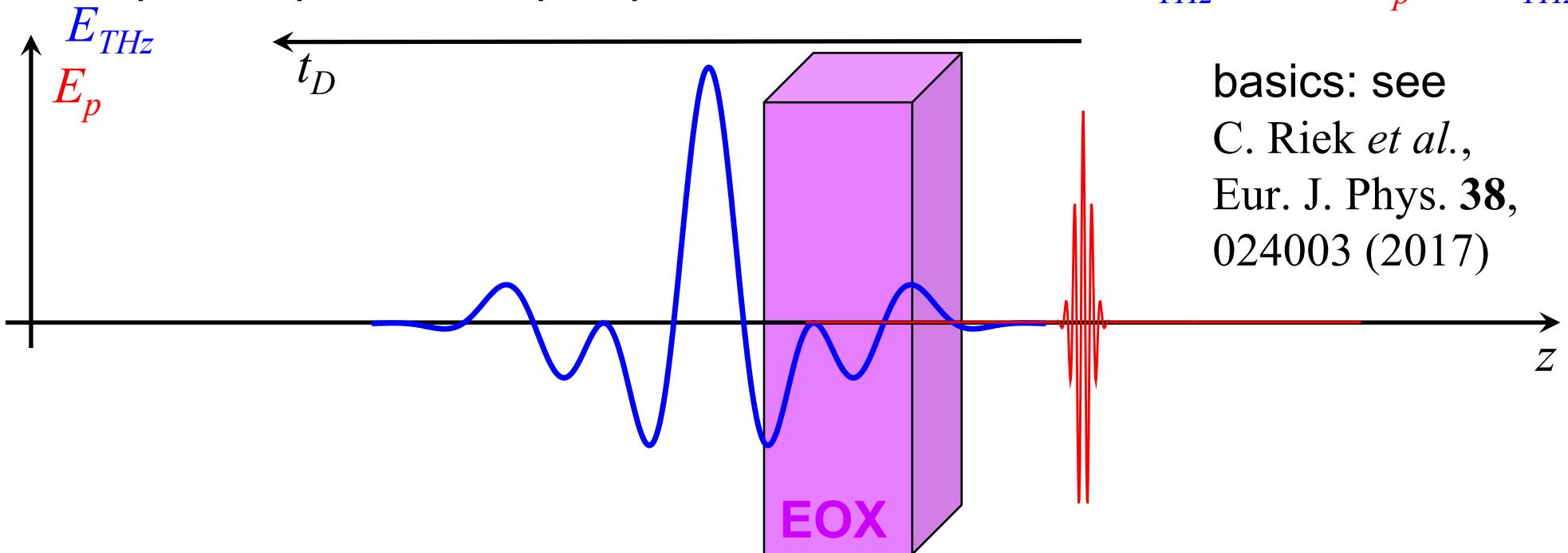


- broadband match group velocity probe – phase velocity signal:
  - use thin electro-optic crystal EOX (ZnTe, GaP...)
  - minimize dispersion  $\leftrightarrow$  work far from resonance
  - angle phase matching in uniaxial GaSe, AgGaS<sub>2</sub>...



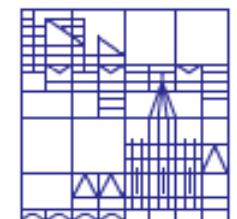
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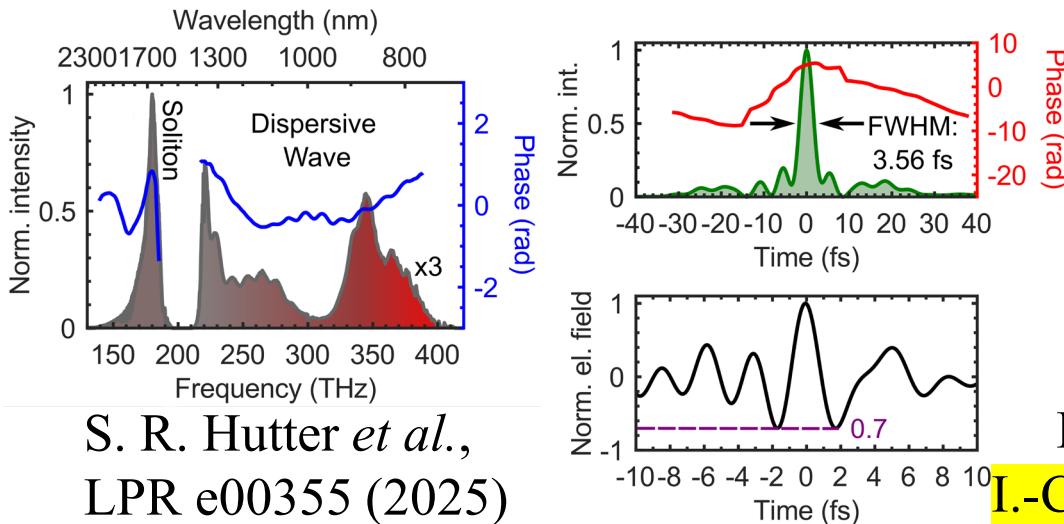
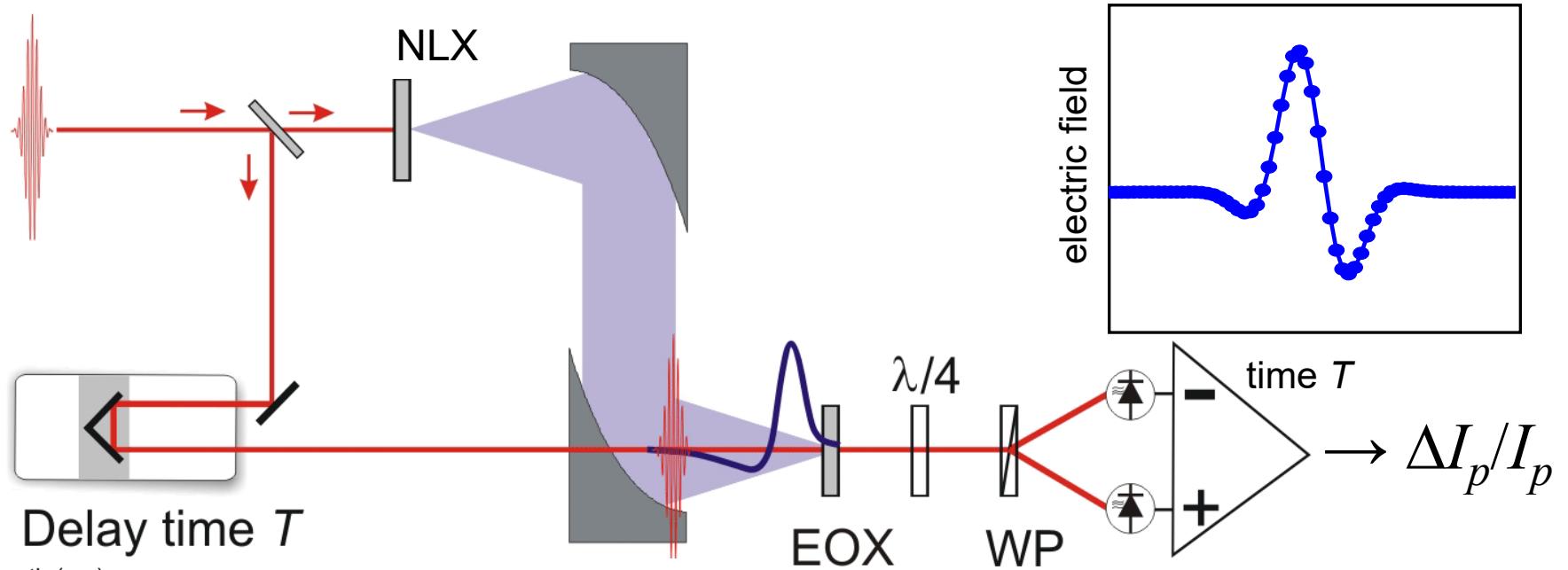


basics: see  
C. Riek *et al.*,  
Eur. J. Phys. **38**,  
024003 (2017)

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  - use thin electro-optic crystal EOX (ZnTe, GaP...)
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  - angle phase matching in uniaxial GaSe, AgGaS<sub>2</sub>...
- sample wave form by varying time delay  $t_D \rightarrow E_{THz}(t_D)$



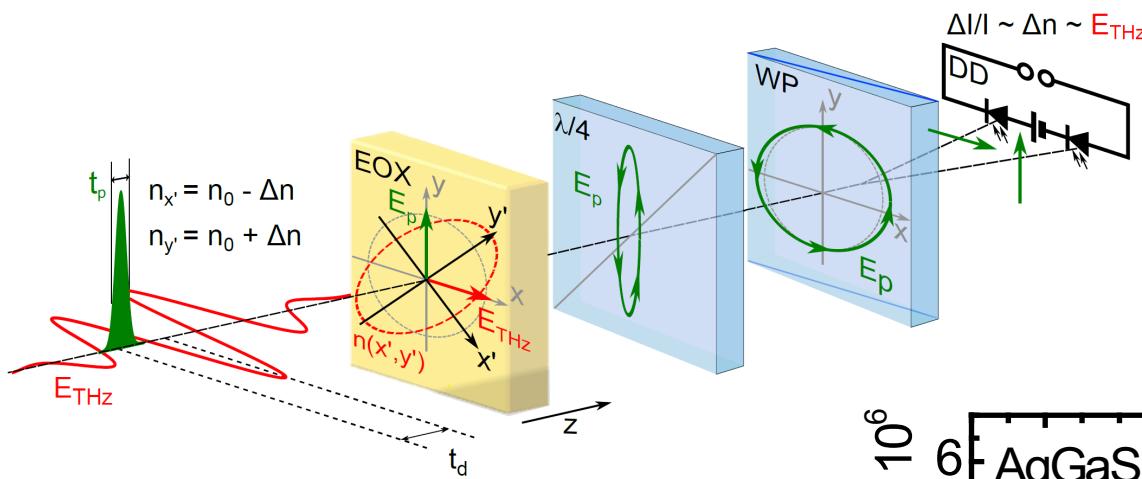
# Ultrabroadband Electro-Optic Sampling: Detect Electric Field with Few-Femtosecond Resolution



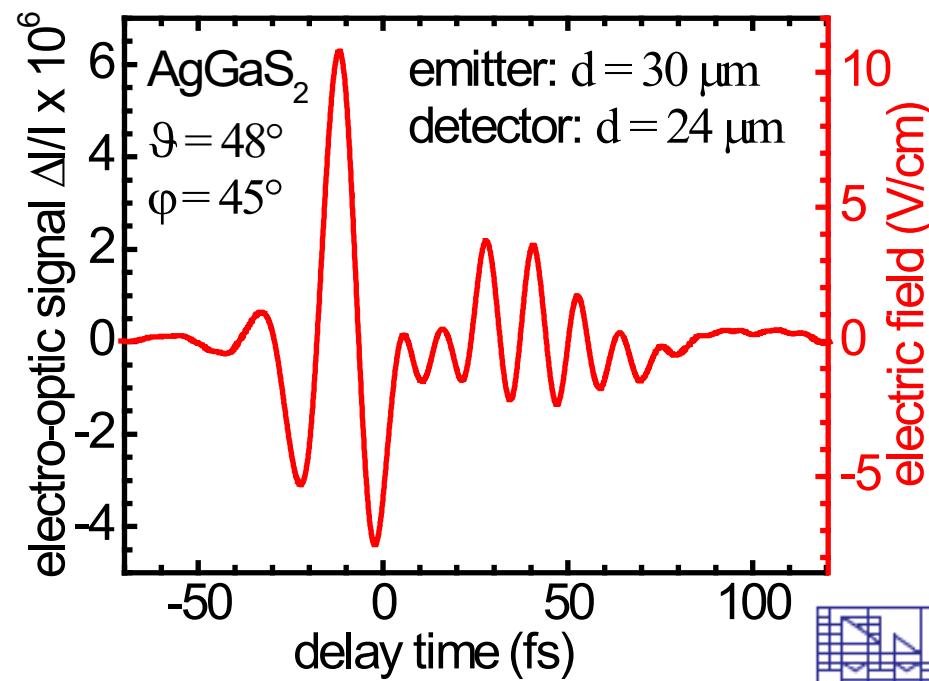
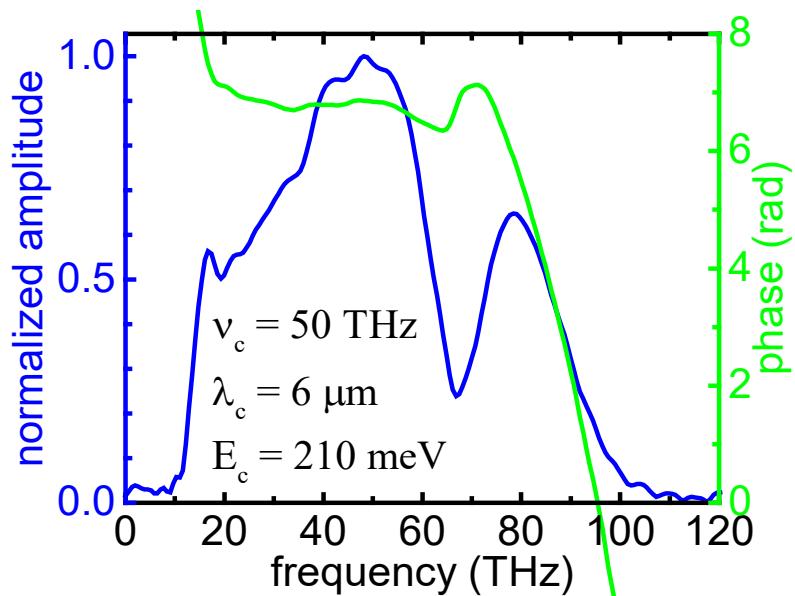
S. R. Hutter *et al.*,  
LPR e00355 (2025)

- Q. Wu + X.-C. Zhang, APL **71**, 1285 (1997)  
A.L. *et al.*, APL **74**, 1516 (1999)  
C. Kübler *et al.*, APL **85**, 3360 (2004)  
A. Sell *et al.*, APL **93**, 251107 (2008)  
P. Sulzer *et al.*, PRA **101**, 033821 (2020)  
E. Ridente *et al.*, Nat. Comms. **13**, 1111 (2022)  
I.-C. Benea-Chelmus *et al.*, Optica **12**, 546 (2025)

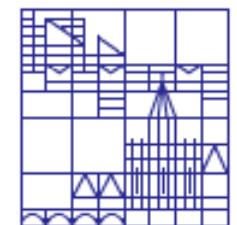
# Field-Resolved Detection of Sub-Photon Pulses



- average energy transient: 0.1 photons
- lock-in bandwidth: 1.8 Hz  
total acquisition time: 136 s
- signal-to-noise:  $>10^2$



- feasibility to directly analyze **local quantum noise** of electric field?
- **switch off pump branch** and search for signals from empty space...

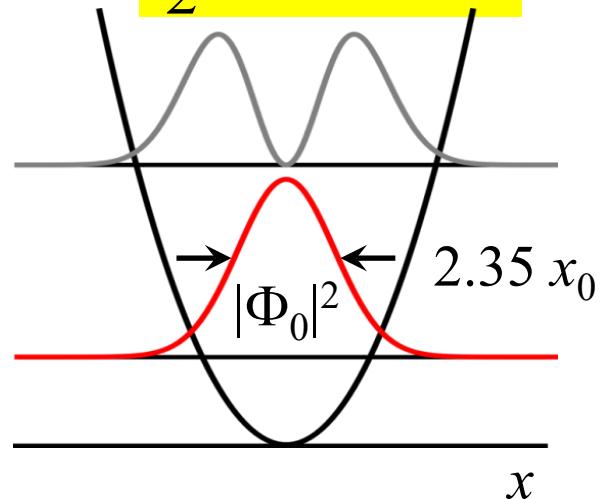


# The Quantum Vacuum: Single-Mode Picture

harmonic oscillator:

$$H_{HO} = \frac{1}{2} \left( \frac{p^2}{m} + m\omega^2 x^2 \right)$$

$$= \frac{m}{2} (\dot{x}^2 + \omega^2 x^2)$$



characteristic length:

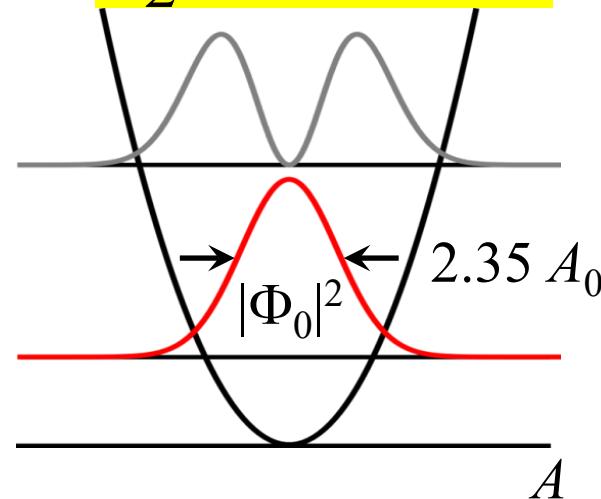
$$x_0 = (\hbar/\omega m)^{1/2}$$

rms amplitude of  
zero-point motion

electromagnetic field:

$$H_{RF} = \frac{\epsilon_0 V}{2} (E^2 + c^2 B^2)$$

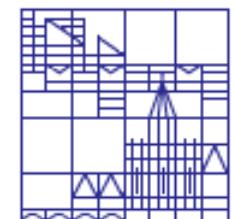
$$= \frac{\epsilon_0 V}{2} (\dot{A}^2 + \omega^2 A^2)$$



characteristic vector potential:

$$A_0 = (\hbar/\omega \epsilon_0 V)^{1/2}$$

rms amplitude of  
vacuum fluctuations



# An Operator for the Free-Space Electric Field

one-dimensional  
harmonic oscillator:

$$\hat{x} = \sqrt{\frac{\hbar}{2m\Omega}} (\hat{a}_\Omega e^{-i\Omega t} + \hat{a}_\Omega^\dagger e^{i\Omega t})$$

$\hat{a}_\omega$ : operator for annihilation,  
 $\hat{a}_\omega^\dagger$ : operator for creation  
of elementary excitation

Bosonic fields:  $[\hat{a}_\omega, \hat{a}_\omega^\dagger] = 1$

single-mode electro-  
magnetic vector potential:

mass  $m \rightarrow \varepsilon_0 V$ : mode volume,  
permittivity of vacuum

displacement  $x \rightarrow A$ :  
vector potential  
in Coulomb (transverse) gauge

electric field:  $\mathbf{E} = -\dot{\mathbf{A}}$

multimode electromagnetic field:  $\mathbf{A}(\mathbf{x}, t) = \sum_{\mathbf{k}} \mathbf{A}_{\mathbf{k}}(t) e^{i\mathbf{kx}}, \Omega = ck$

$$\hat{\mathbf{A}}(\mathbf{x}, t) = \sum_{\mathbf{k}, \lambda} \sqrt{\frac{\hbar}{2\varepsilon_0 V \Omega}} (\hat{a}_{\mathbf{k}, \lambda} \boldsymbol{\epsilon}_{\mathbf{k}, \lambda} e^{i\mathbf{kx} - i\Omega t} + \hat{a}_{\mathbf{k}, \lambda}^\dagger \boldsymbol{\epsilon}_{\mathbf{k}, \lambda}^* e^{-i\mathbf{kx} + i\Omega t})$$

$$\hat{\mathbf{E}}(\mathbf{x}, t) = i \sum_{\mathbf{k}, \lambda} \sqrt{\frac{\hbar \Omega}{2\varepsilon_0 V}} (\hat{a}_{\mathbf{k}, \lambda} \boldsymbol{\epsilon}_{\mathbf{k}, \lambda} e^{i\mathbf{kx} - i\Omega t} - \hat{a}_{\mathbf{k}, \lambda}^\dagger \boldsymbol{\epsilon}_{\mathbf{k}, \lambda}^* e^{-i\mathbf{kx} + i\Omega t})$$

$\boldsymbol{\epsilon}_{\mathbf{k}, \lambda} \perp \mathbf{k}$  ( $\lambda = 1, 2$ ):  
unit polarization  
vectors

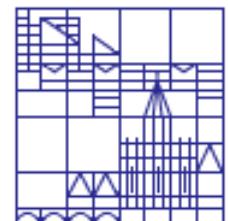
# Amplitude Vacuum Fluctuations in Free Space

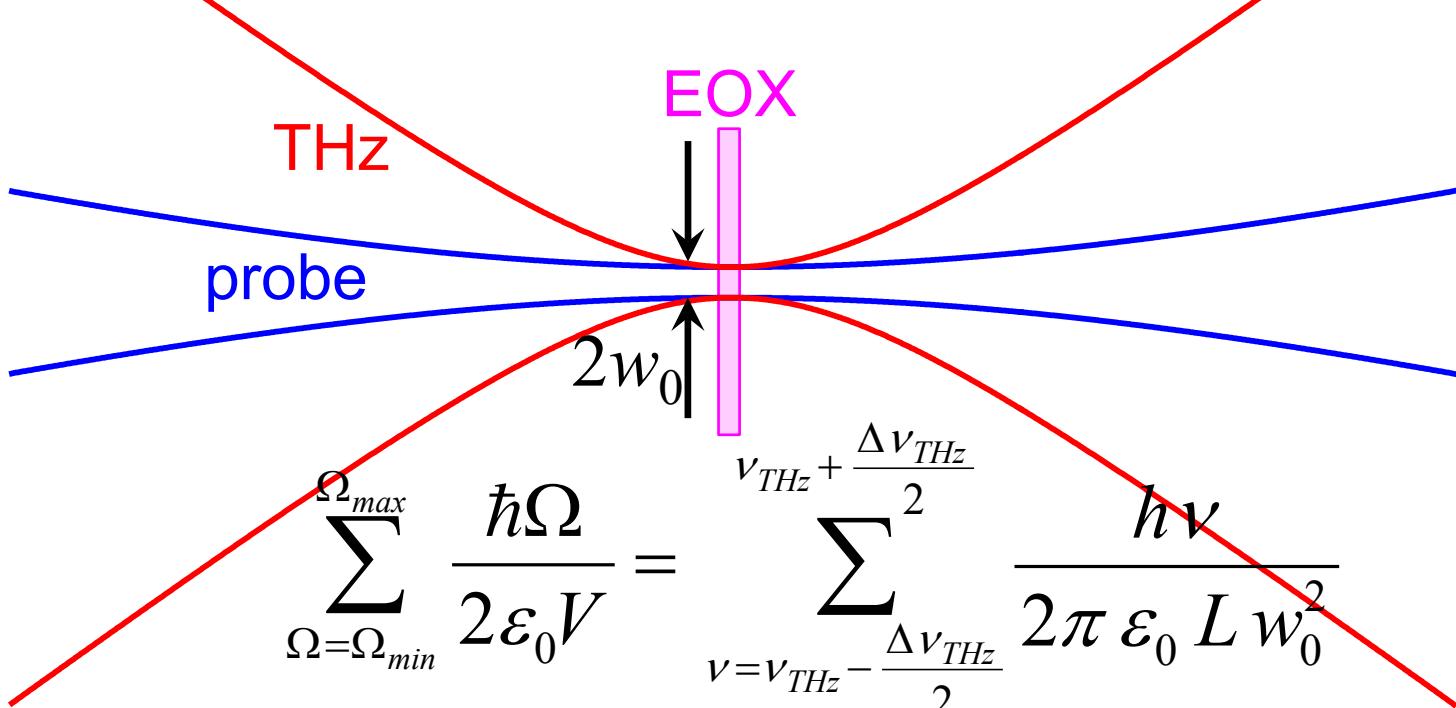
- physical meaning normalization volume  $V$  outside cavity?
- periodic boundary conditions: sum / **integrate over all frequencies**  $\nu = \Omega/2\pi$  the field measurement is sensitive to
- rms fluctuation amplitude in electric-field ground state:

$$\Delta \bar{E}_{vac}^{rms} = \sqrt{\left\langle \Phi_0 \left| \sum_{\Omega=\Omega_{min}}^{\Omega_{max}} \frac{-\hbar\Omega}{2\varepsilon_0 V} (\hat{a}_\Omega - \hat{a}_\Omega^\dagger)^2 \right| \Phi_0 \right\rangle} = \sqrt{\left\langle \Phi_0 \left| \sum_{\Omega=\Omega_{min}}^{\Omega_{max}} \frac{\hbar\Omega}{2\varepsilon_0 V} \hat{a}_\Omega \hat{a}_\Omega^\dagger \right| \Phi_0 \right\rangle} =$$

$= \sqrt{\sum_{\Omega=\Omega_{min}}^{\Omega_{max}} \frac{\hbar\Omega}{2\varepsilon_0 V}}$

$[\hat{a}_\Omega, \hat{a}_\Omega^\dagger] = 1$ ,  $\hat{a}_\Omega$ : annihilation,  
 $\hat{a}_\Omega^\dagger$ : creation of THz photon

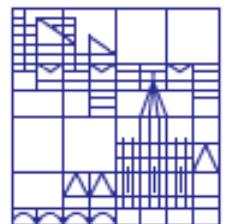




$$\sum_{\Omega=\Omega_{min}}^{\Omega_{max}} \frac{\hbar\Omega}{2\epsilon_0 V} = \sum_{\nu=\nu_{THz}-\frac{\Delta\nu_{THz}}{2}}^{\nu_{THz}+\frac{\Delta\nu_{THz}}{2}} \frac{h\nu}{2\pi \epsilon_0 L w_0^2}$$

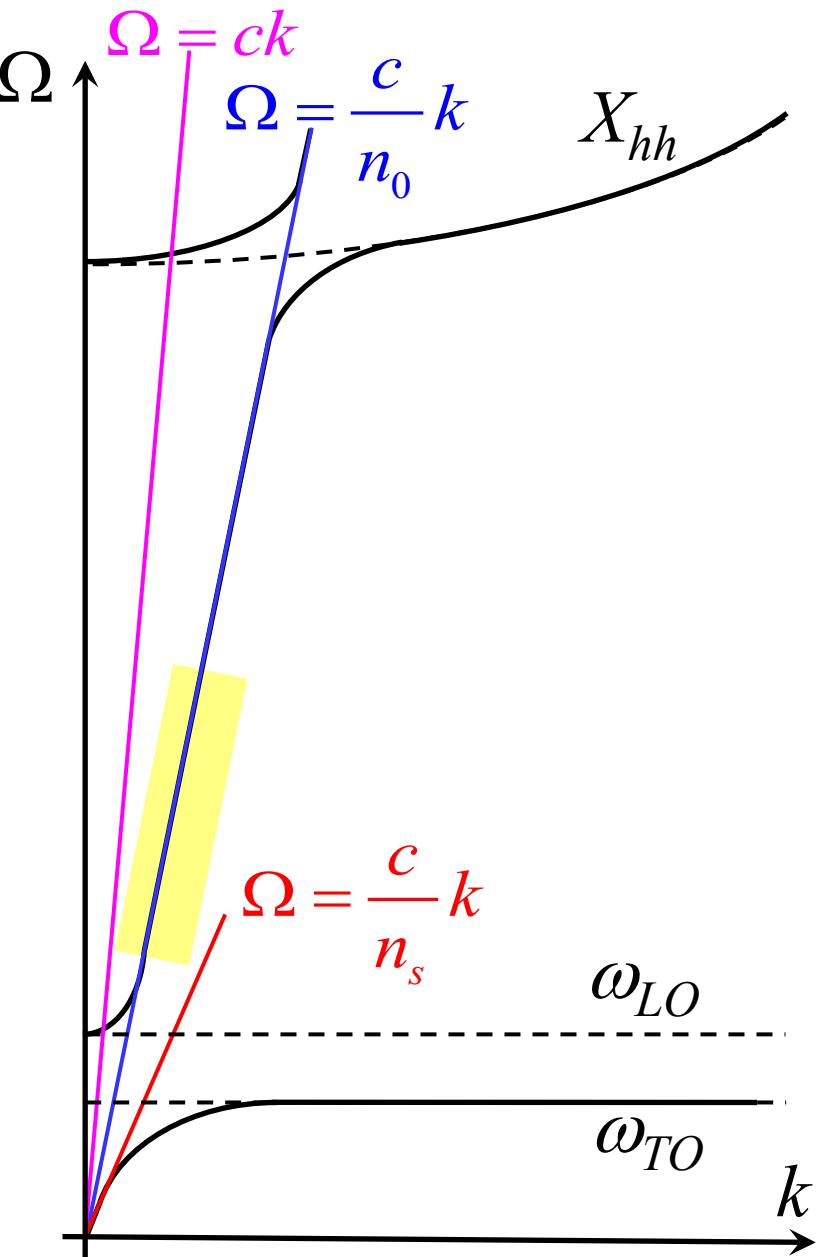
$$\rightarrow \sum_{\nu=\nu_{THz}-\frac{\Delta\nu_{THz}}{2}}^{\nu_{THz}+\frac{\Delta\nu_{THz}}{2}} \frac{h\nu}{2\pi n_0 \epsilon_0 L w_0^2}$$

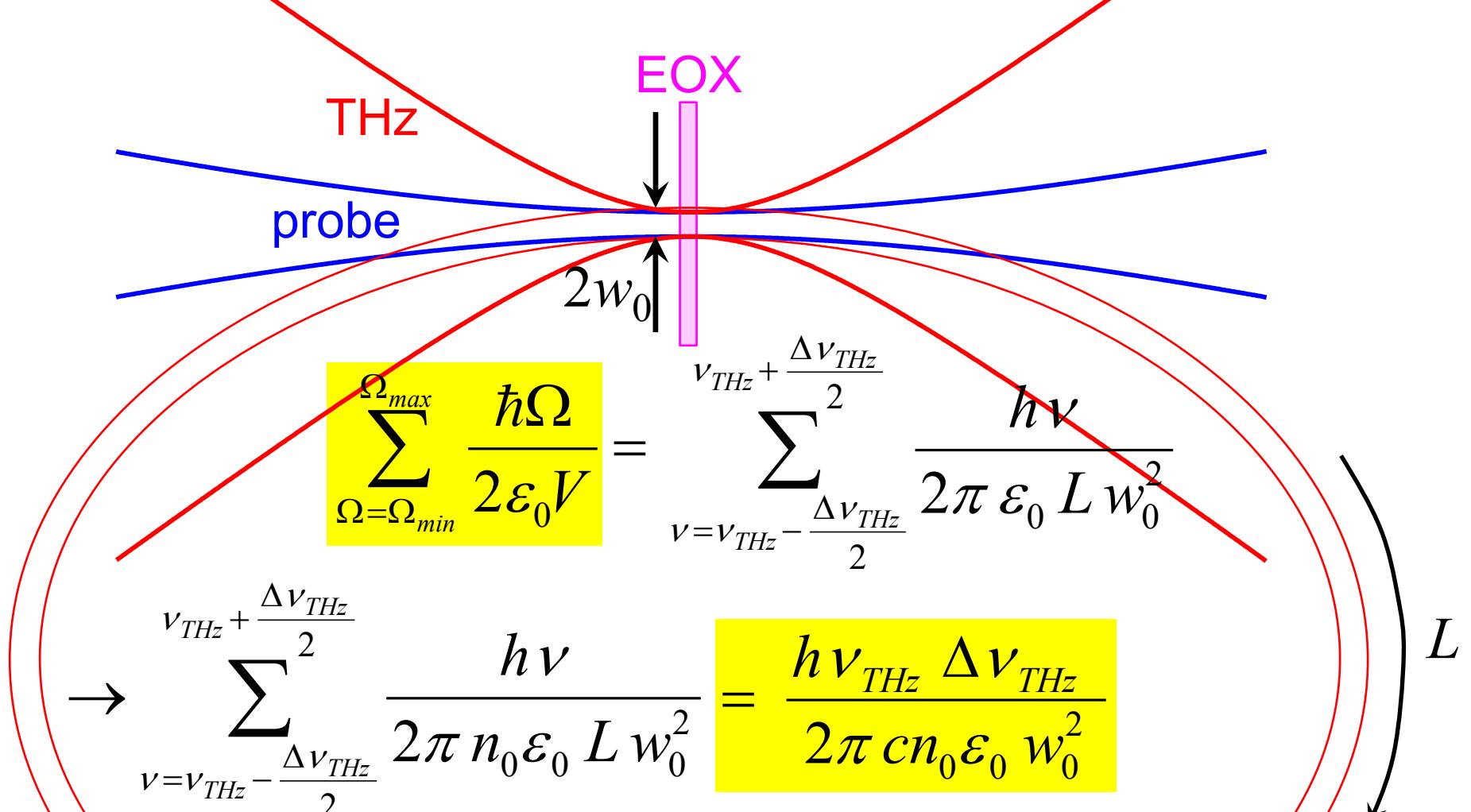
refractive index  $n_0$ : dielectric screening in EOX



# Photons in Free Space → Polaritons in EOX

- photon enters dielectric material:
  - strong coupling to polar eigenmodes: optical phonons, excitons...
  - phonon and exciton **polariton** branches
- our experiments: off resonance, frequencies far from phonon and exciton bands
- real-valued refractive index  $n_0 = \epsilon^2$ 
  - as **photon-like** as it gets in matter, **fully linked with free-space** electromagnetic field



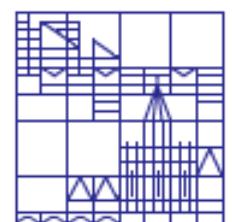


refractive index  $n_0$ : dielectric screening in EOX

unidirectional frequency spacing  $c/L \rightarrow$

$\Delta\nu_{THz} L/c$  modes

$L$  drops off  $\rightarrow$  geometry outside EOX irrelevant



# Amplitude Vacuum Fluctuations in Free Space

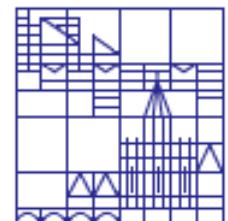
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- periodic boundary conditions: sum / **integrate over all frequencies**  $\nu = \Omega/2\pi$  the field measurement is sensitive to
- rms fluctuation amplitude in electric-field ground state:

$$\Delta \bar{E}_{vac}^{rms} = \sqrt{\left\langle \Phi_0 \right| \sum_{\Omega=\Omega_{min}}^{\Omega_{max}} \frac{-\hbar\Omega}{2\varepsilon_0 V} (\hat{a}_\Omega - \hat{a}_\Omega^\dagger)^2 \left| \Phi_0 \right\rangle} = \sqrt{\left\langle \Phi_0 \right| \sum_{\Omega=\Omega_{min}}^{\Omega_{max}} \frac{\hbar\Omega}{2\varepsilon_0 V} \hat{a}_\Omega \hat{a}_\Omega^\dagger \left| \Phi_0 \right\rangle} =$$

$\boxed{\frac{h\nu_{THz} \Delta\nu_{THz}}{2\pi c n_0 \varepsilon_0 w_0^2}} = \sqrt{\frac{\hbar}{\varepsilon \varepsilon_0 \Delta x \Delta y \Delta z \Delta t}}$

$[\hat{a}_\Omega, \hat{a}_\Omega^\dagger] = 1$ ,  $\hat{a}_\Omega$ : annihilation,  
 $\hat{a}_\Omega^\dagger$ : creation of THz photon

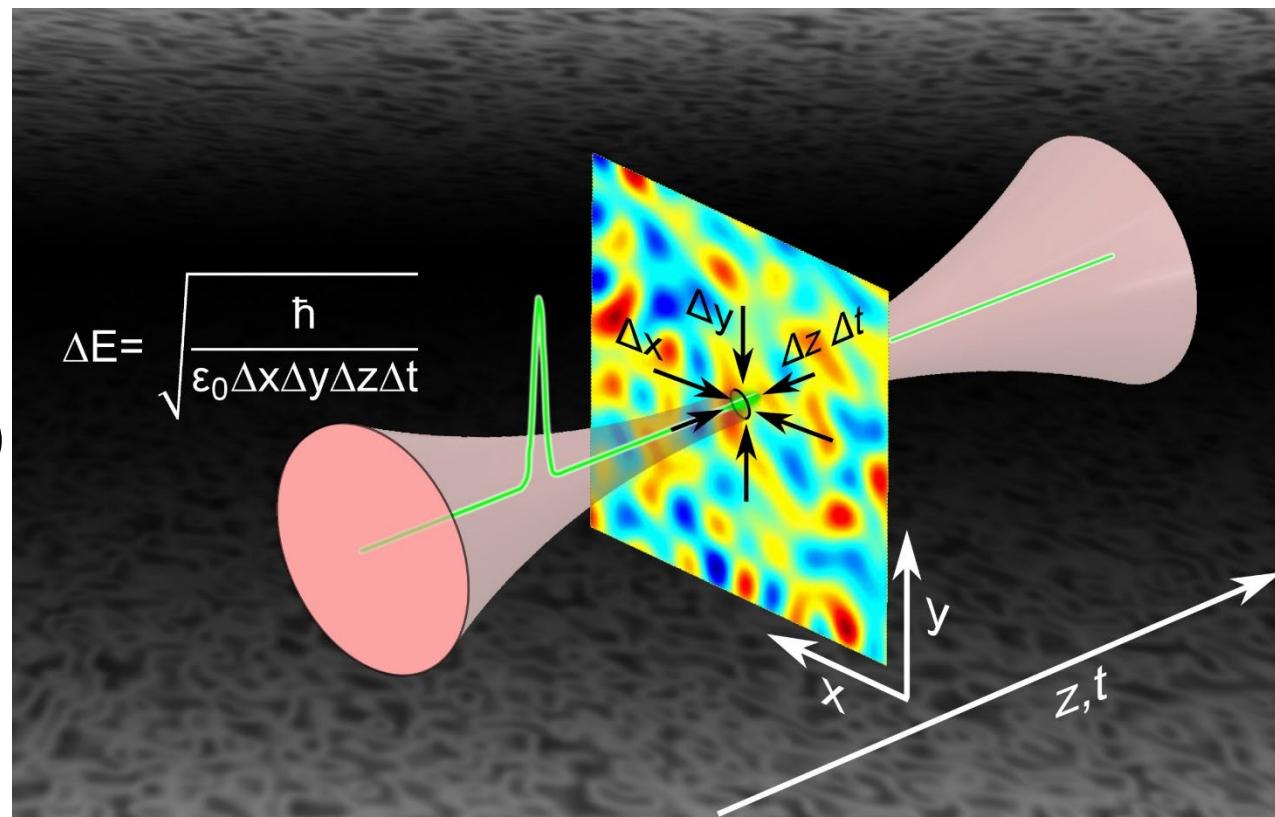
- vacuum amplitude depends on **four-dimensional space-time volume**  $\Delta x \Delta y \Delta z \Delta t$  over which probe pulse averages
- for our experimental parameters  $\Delta \bar{E}_{vac}^{rms} = 20.2 \frac{V}{cm}$



# Amplitude Vacuum Fluctuations in Free Space

- **Heisenberg:**  $\Delta E_x \Delta H_y \geq \frac{c}{n} \frac{\hbar c}{(\delta l)^4} = \sqrt{\frac{1}{\epsilon \epsilon_0 \mu \mu_0}} \frac{\hbar c}{(\delta l)^4}$
- **vacuum ground state:**  $\Delta E_{vac} \approx \sqrt{\frac{\hbar c}{\epsilon \epsilon_0 (\delta l)^4}} = \sqrt{\frac{\hbar}{\epsilon \epsilon_0 \Delta x \Delta y \Delta z \Delta t}}$ 
  - minimum uncertainty in both  $\Delta E_x$  and  $\Delta H_y$
  - Gaussian wave function (harmonic oscillator!)
  - for our setup:

$$\boxed{\Delta \bar{E}_{vac}^{rms} = 20.2 \frac{V}{cm}}$$



# Noise-Equivalent Field Electro-Optic Detection

- rms electro-optic signal due to shot noise from average number of  $N_p$  photons detected per probe pulse (coherent wave packet!):

$$\frac{\Delta I_{SN}^{rms}}{I_p} = \frac{\sqrt{N_p}}{N_p} = \sin\left(\frac{2\pi \nu_p r_{41} n_0^3 L_d}{c} \Delta E_{SN}^{rms}\right) \approx \frac{2\pi \nu_p r_{41} n_0^3 L_d}{c} \Delta E_{SN}^{rms}$$

- shot-noise equivalent field:

$$\Delta E_{SN}^{rms} = \frac{c}{2\pi \nu_p r_{41} n_0^3 L_d \sqrt{N_p} |R(\nu_{THz})|} = 65 \frac{\text{V}}{\text{cm}}$$

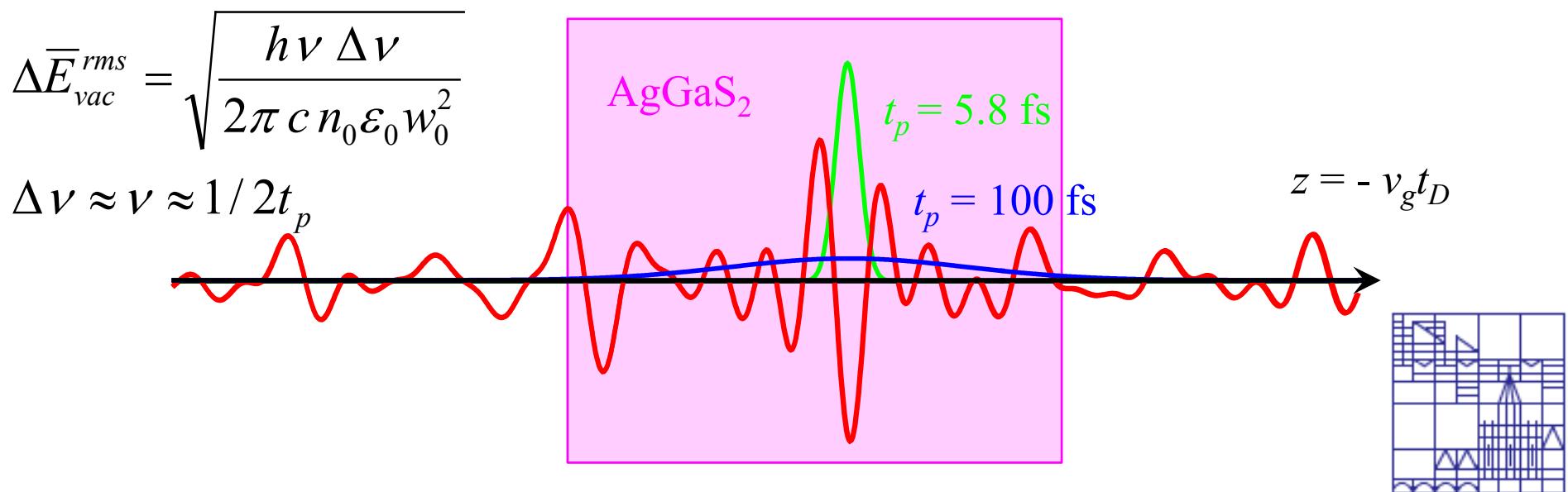
- detector thickness  $L_d = 30 \mu\text{m}$
- electro-optic coefficient  $r_{41} = 7.6 \text{ pm/V}$
- average number of probe photons detected per sampling pulse  $N_p = I_p/qR = 3 \text{ mA}/(1.6 \times 10^{-19} \text{ C} \times 40 \text{ MHz}) = 5 \times 10^8$
- probe center frequency  $\nu_p = 255 \text{ THz}$

- expected change total noise amplitude by vacuum fluctuations:

$$\frac{\Delta E_{total}}{\Delta E_{SN}} = \frac{\sqrt{\Delta E_{SN}^{rms}^2 + \Delta \bar{E}_{vac}^{rms}^2}}{\Delta E_{SN}^{rms}} = 1.047 \equiv 4.7\%$$

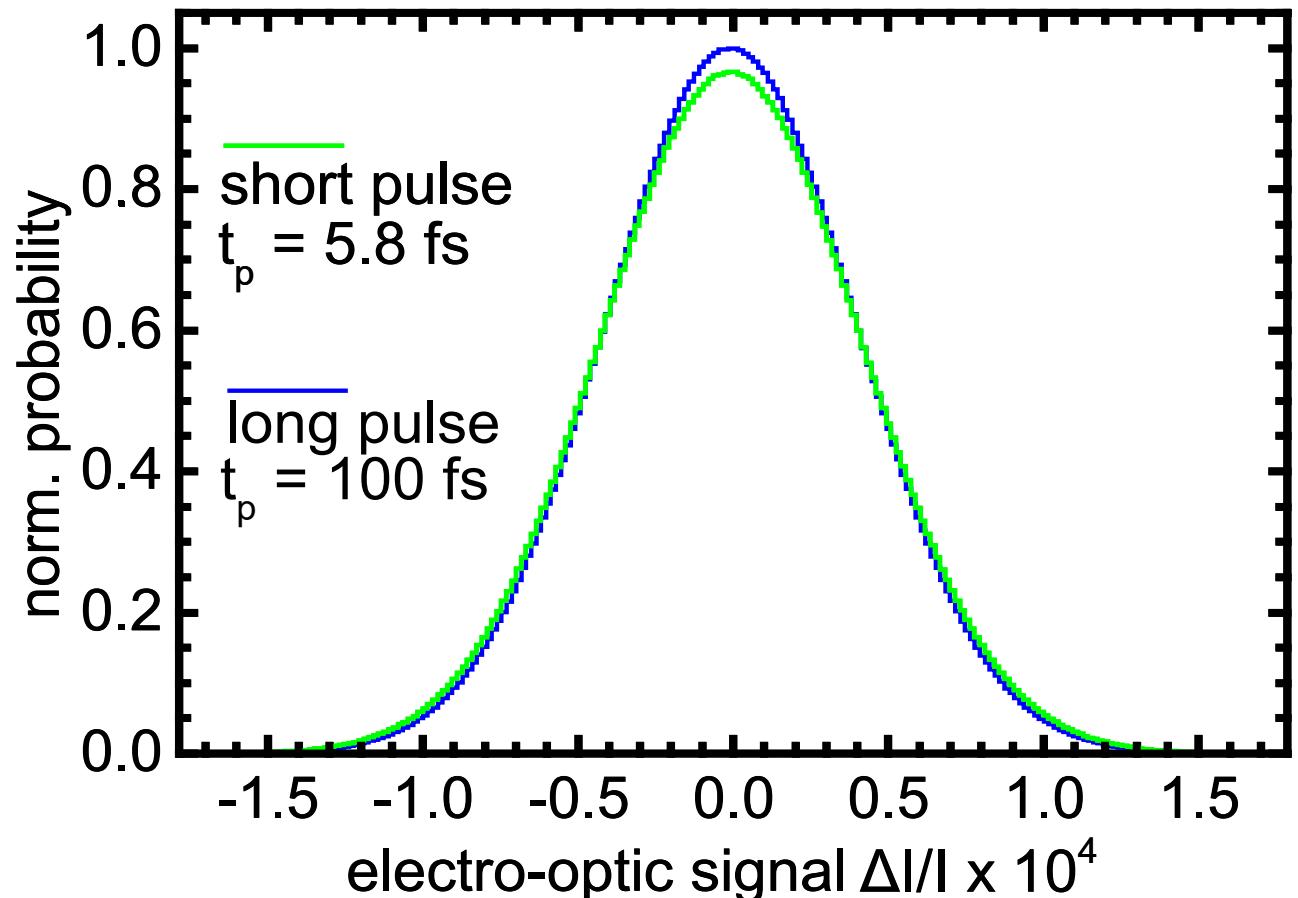
# Experimental Strategy to Detect Vacuum Noise

- How to prove that total noise measured in electro-optic detector contains component due to multi-THz vacuum fluctuations?
- **expand space-time volume** in which vacuum field amplitude is sampled while leaving other experimental parameters identical
- longitudinal option (along  $z = -v_g t_D$ ): **stretch sampling pulse**



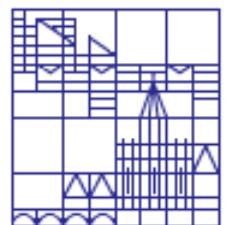
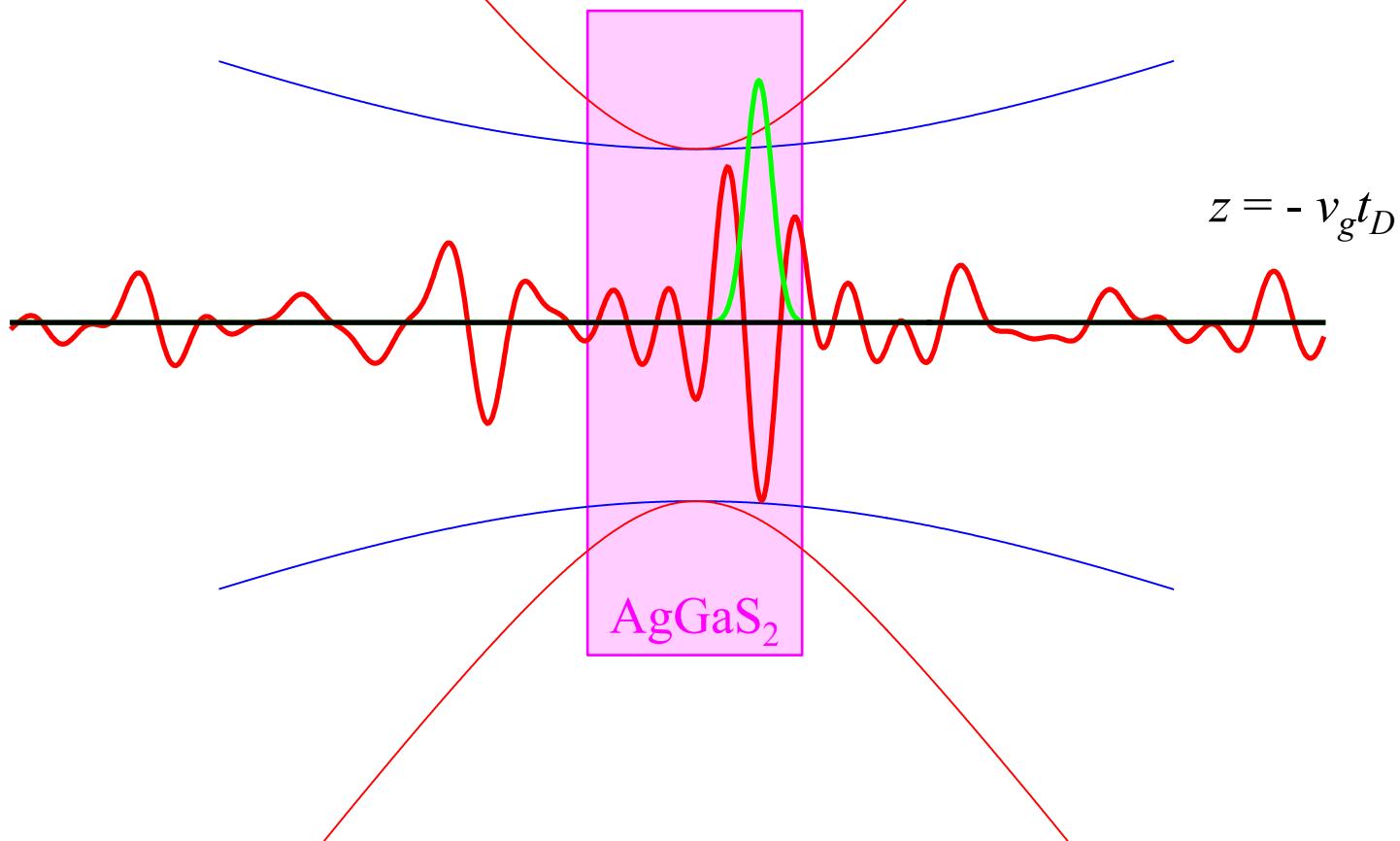
# Absolute Measurement of Vacuum Noise I

- longitudinal option (along  $z = -v_g t_D$ ): **stretch sampling pulse** from 6 fs to 100 fs by translating SF10 prism in compressor stage
- detect electro-optic signal with lock-in bandwidth of 1.6 MHz
- readout every 5  $\mu$ s and build up **histogram**
- change of total noise amplitude in experiment: 4%
- compare:  
4.7% predicted by theory



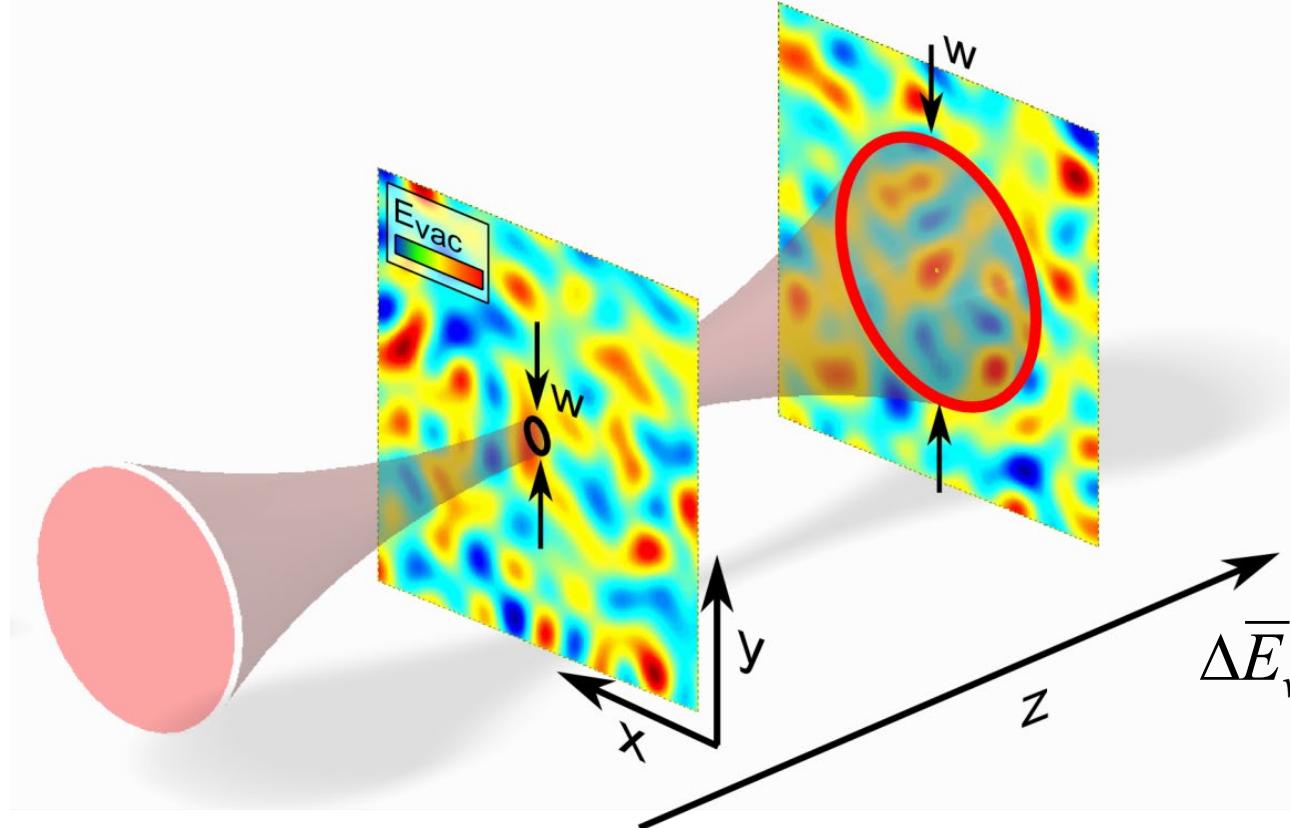
# Absolute Measurement of Vacuum Noise II

- transverse option (along  $x$  and  $y$ ): **translate electro-optic crystal** out of confocal region with  $z$ -averaged beam waist  $w_0 = 4.25 \mu\text{m}$



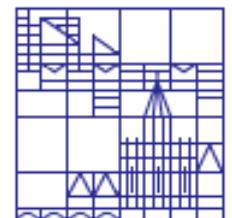
# Absolute Measurement of Vacuum Noise II

- transverse option (along  $x$  and  $y$ ): **translate electro-optic crystal** out of confocal region with  $z$ -averaged beam waist  $w_0 = 4.25 \mu\text{m}$



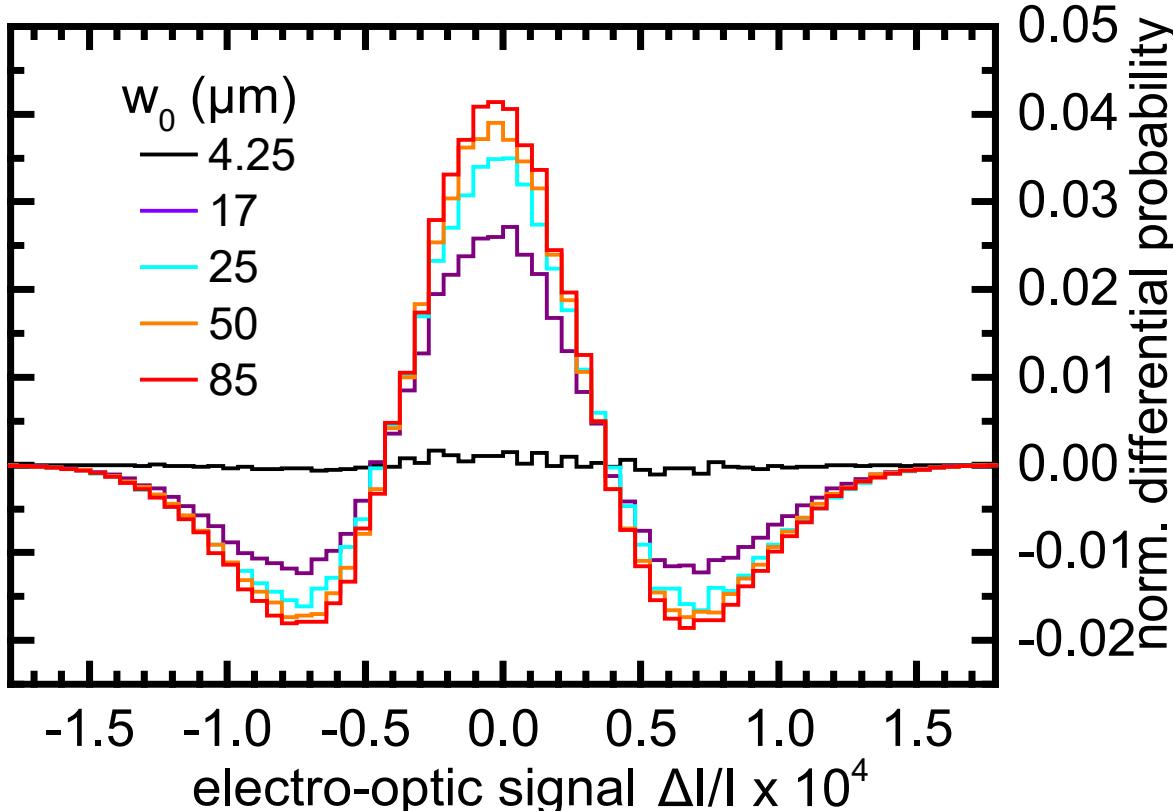
$$\Delta \bar{E}_{\text{vac}}^{\text{rms}} = \sqrt{\frac{h\nu \Delta\nu}{2\pi c n_0 \epsilon_0 w_0^2}}$$

- probe averages over lateral vacuum amplitude
- fluctuations on length scale  $\approx \lambda/2$  ( $\lambda_{\text{THz}} \approx 4.3 \mu\text{m}$ )  
→ integrated noise amplitude should decrease with  $1/w$

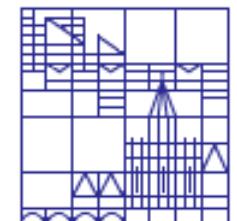


# Absolute Measurement of Vacuum Noise II

- transverse option (along  $x$  and  $y$ ): **translate electro-optic crystal** out of confocal region with  $z$ -averaged beam waist  $w_0 = 4.25 \mu\text{m}$
- take histograms at different beam spot radii  $w_0$
- plot deviations from confocal position as **differential histograms**

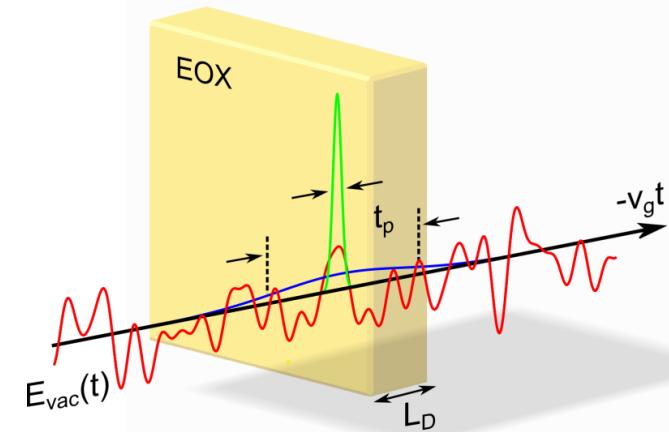
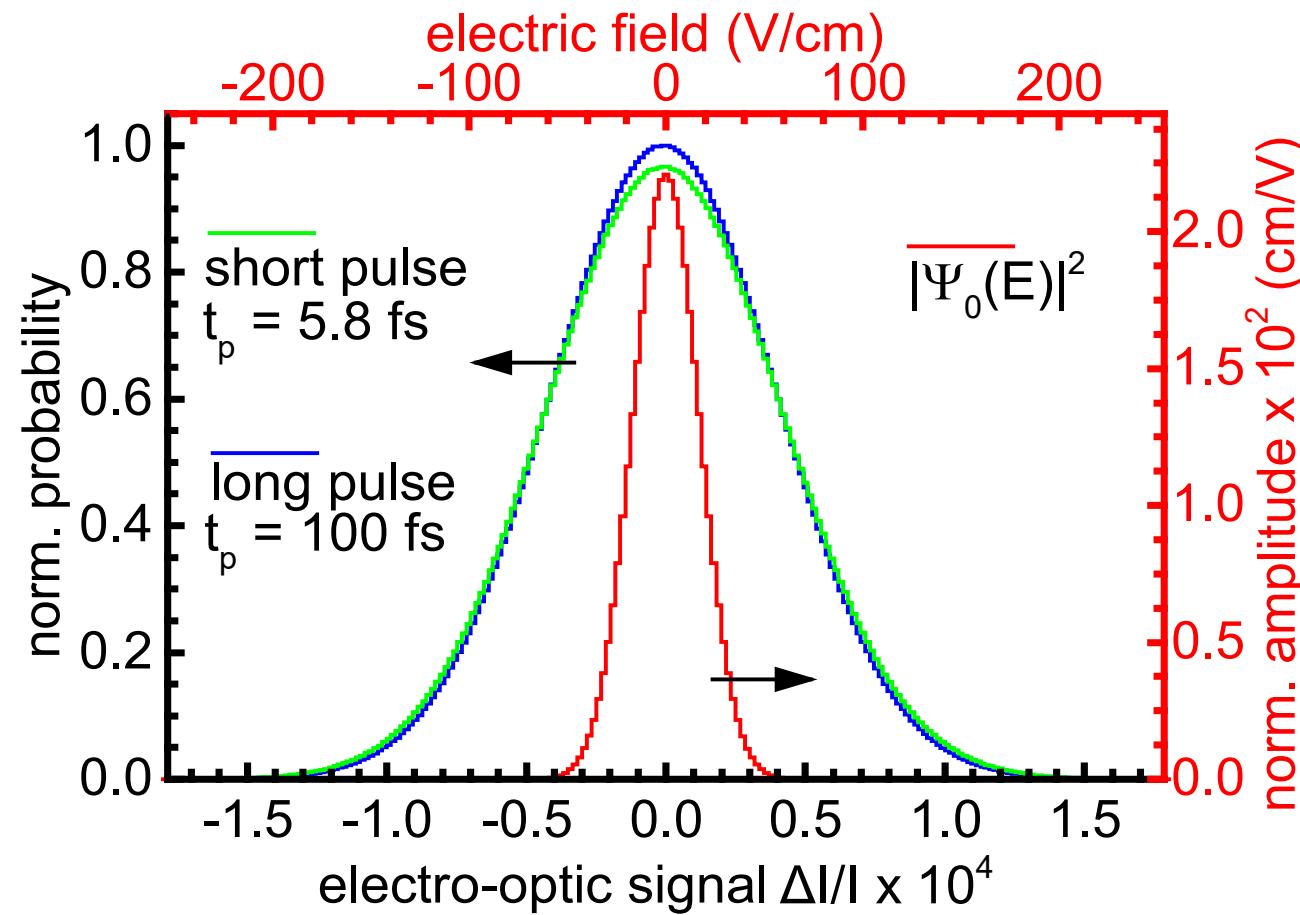


- maximum change total noise detected by experiment in transverse option: 4.2%
- compare theoretical result: 4.7%



# Retrieving the Ground-State |Wave Function|<sup>2</sup>

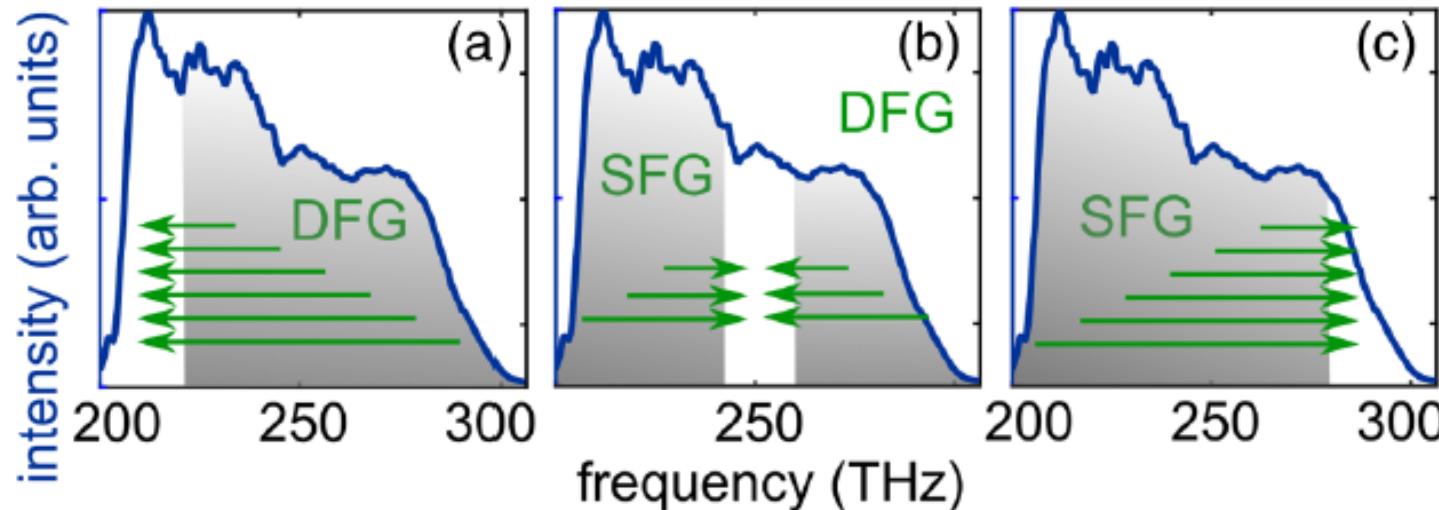
- de-convolute statistics with **bandwidth-limited** and **chirped** probe  
→ square of **ground-state wave function**  $|\Psi_0(E)|^2$  of electric field in probed space-time volume



- rms width of  $|\Psi_0(E)|^2$ :  
 $\Delta E^{rms} = 18 \text{ V/cm}$
- compare with theory estimate:  
 $\Delta \bar{E}_{vac}^{rms} = 20 \text{ V/cm}$

# Electro-Optic Sampling: Three-Wave Mixing

- second-order nonlinear optical process: ideally simultaneous difference- (DFG) and **sum- (SFG)** frequency generation



- near-infrared probe spectrum mixes with mid-infrared/THz photons
- **DFG: (parametric) amplification, SFG: annihilation** of MIR/THz
- NIR mixing product: **homodyne** detection within **broadband** probe
  - determine amplitude and **phase** of MIR/THz input wave packet

G. Gallot and D. Grischkowsky, J. Opt. Soc. Am. B **16**, 1204 (1999)

P. Sulzer, K. Oguchi *et al.*, Phys. Rev. A **101**, 033821 (2020)

# Quantum Theory Electro-Optic Sampling

- 2<sup>nd</sup>-order nonlinear mixing in zincblende geometry:

$$\hat{P}_s^{(2)}(t) = -\epsilon_0 d \hat{E}_{\text{THz},s}(t) E_p(t), \quad \hat{P}_z^{(2)}(t) = 0$$

- paraxial approximation, Laguerre-Gaussian mode functions for free-space quantum fields:

$$\left[ \Delta_{\perp} + 2ik_{\omega} \frac{\partial}{\partial r_{\parallel}} \right] \hat{\mathbf{E}}(\mathbf{r}; \omega) = -\frac{\omega^2}{\epsilon_0 c_0^2} \hat{\mathbf{P}}^{(2)}(\mathbf{r}; \omega)$$

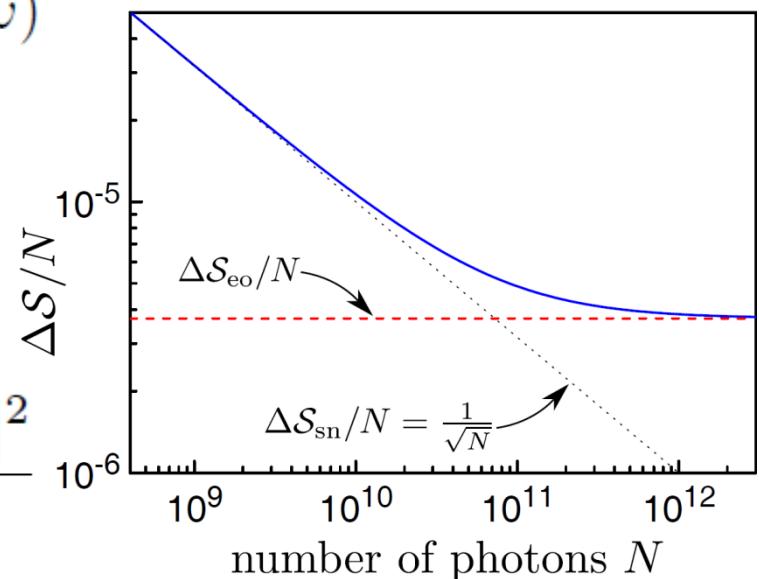
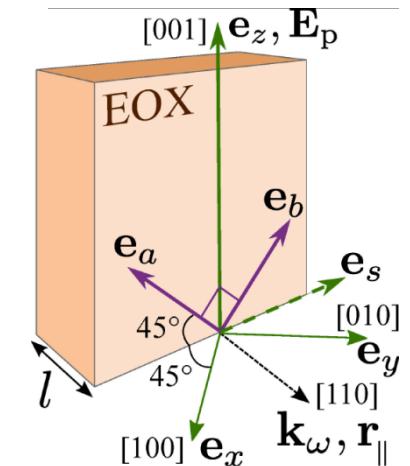
$$E_p(\mathbf{r}; \omega) = \alpha_p(\omega) LG_{00}(\mathbf{r}_{\perp}, r_{\parallel}; k_{\omega})$$

- two noise components in electro-optic signal:  $\langle \hat{S}_{\text{sn}}^2 \rangle = N$

$$\langle \hat{S}_{\text{eo}}^2 \rangle = N^2 \left( n^3 \frac{l \omega_p}{c_0} r_{41} \right)^2 \frac{\hbar \int_0^{\infty} d\Omega \Omega (n/n_{\Omega}) |R(\Omega)|^2}{4\pi^2 \epsilon_0 c_0 n w_0^2}$$

- longitudinal response function:

$$R(\Omega) = \text{sinc} \left[ \frac{l\Omega}{2c_0} (n_{\Omega} - n_g) \right] f(\Omega) \quad f_{\pm}(\Omega) = \frac{\int_0^{\infty} d\omega \eta(\omega) \alpha_p^*(\omega) \alpha_p(\omega \pm \Omega)}{\int_0^{\infty} d\omega \eta(\omega) |\alpha_p(\omega)|^2}$$

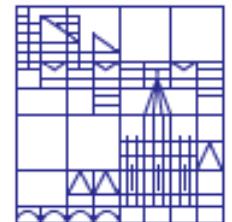
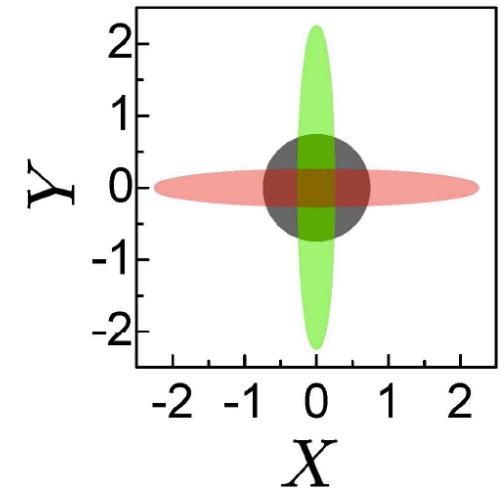
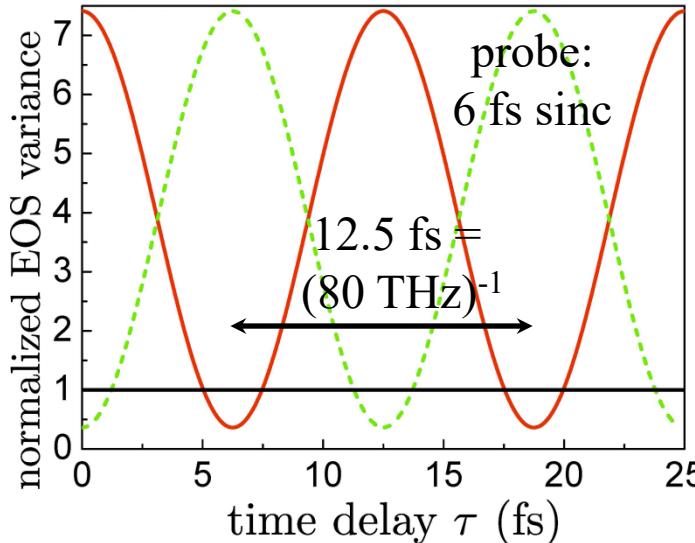


# Theory: Subcycle Quantum Analysis of Light

- apply continuum squeezing operator to bare vacuum state:

$$\exp \left[ \frac{1}{2} \int_0^{2\Omega_c} d\Omega \sum_{\alpha lp} (\xi_\Omega^* \hat{a}_{\alpha,l,p}(2\Omega_c - \Omega) \hat{a}_{\alpha,l,p}(\Omega) - H.c.) \right]$$

- example: constant squeezing  $\xi_\Omega \equiv \xi = re^{i\theta}$  in  $\pm 40$  THz interval around  $\Omega_c/2\pi = 40$  THz with  $\sinh r = 2$ , no squeezing outside
- error contours in complex amplitude plane for bare vacuum,  $\theta = 0$  and  $\theta = \pi$ :
- noise electro-optic signal depends on time delay probe



A. S. Moskalenko *et al.*,  
Phys. Rev. Lett. **115**,  
263601 (2015)

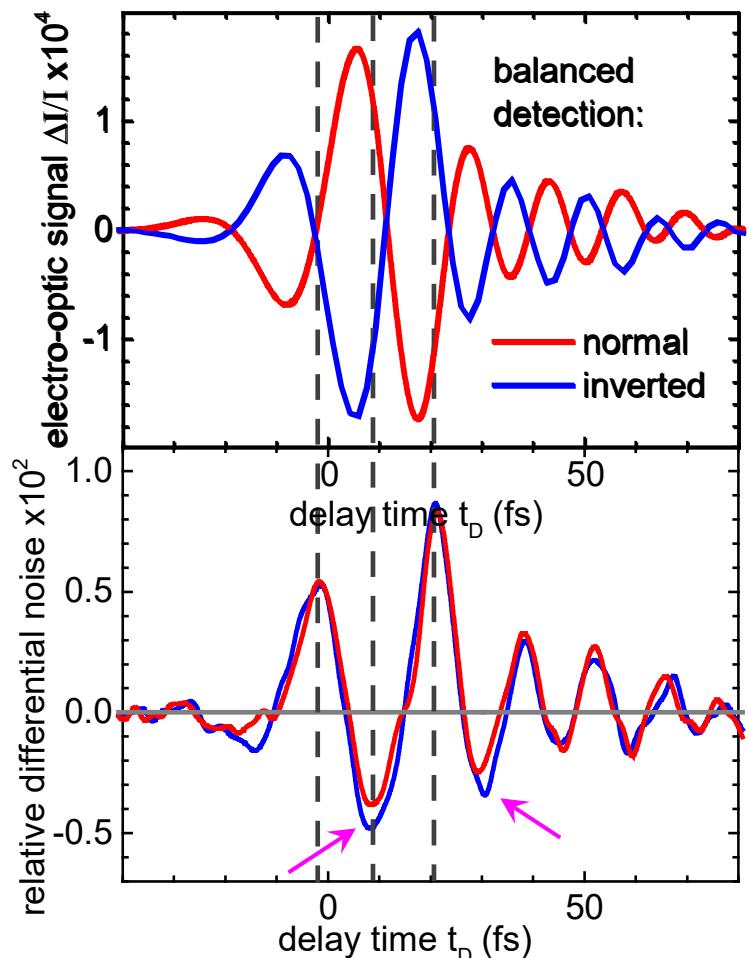
## Part II

### Squeezing!?

How to prepare other nonclassical states in the mid infrared and how to characterize them in the time domain?

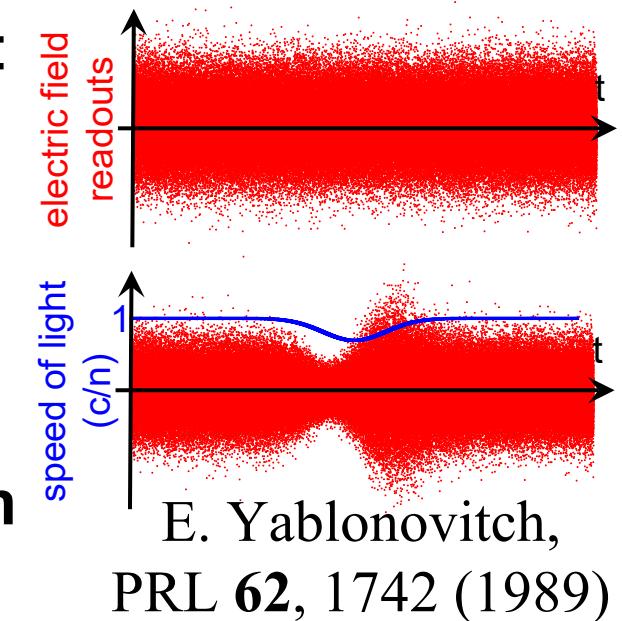
# Time-Domain Squeezing of the Quantum Vacuum?

- bare vacuum propagating with speed of light:
- local **acceleration of reference frame** by short pump pulse, e.g. via nonlinear refractive index (Kerr,  $\chi^{(3)}$ ):  $\Delta n \sim n_2 E_{\text{pump}}^2$



→ **synchronal  
squeezed vacuum  
experiment:**

- emitter: GaSe,  $d = 16 \mu\text{m}$ ,  $\Theta = 0^\circ$   
pump:  $t_p = 12 \text{ fs}$ ,  $E_p = 3.5 \text{ nJ}$ ,  $w_0 = 3.6 \mu\text{m}$
- detect **relative differential noise** with respect to bare vacuum input
- **negative signals: less noise in detector** due to squeezed vacuum from emitter!?



# Time-Domain Conjugate Variable to Electric Field

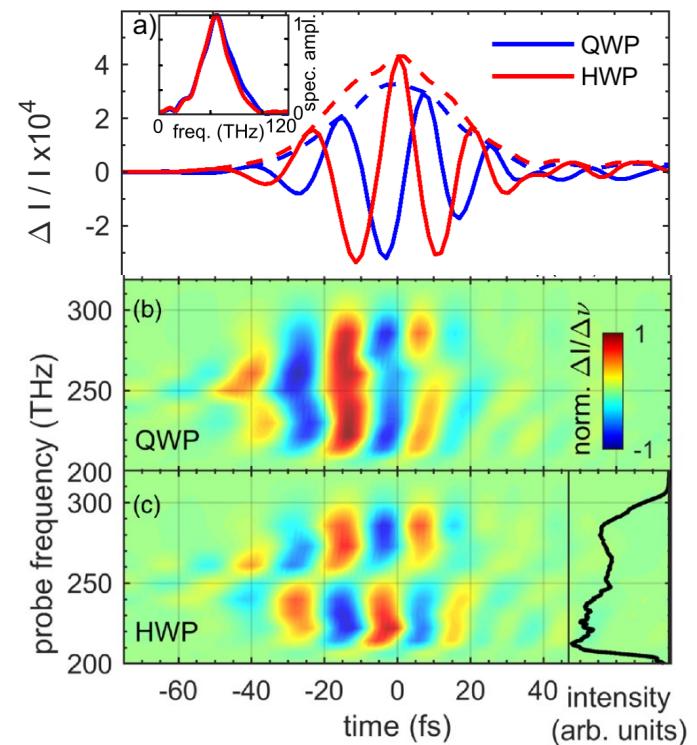
- single-mode case: G. Leuchs, Contemp. Phys. **29**, 299 (1988)

analogy to the mechanical case, we can write the electric field  $E(t)$  of a light wave in terms of the initial conditions for the electric field and its derivative  $dE/dt = \dot{E}$ , the displacement current density:

$$E(t) = E(0) \cos(\omega t) + [\dot{E}(0)/\omega] \sin(\omega t). \quad (3)$$

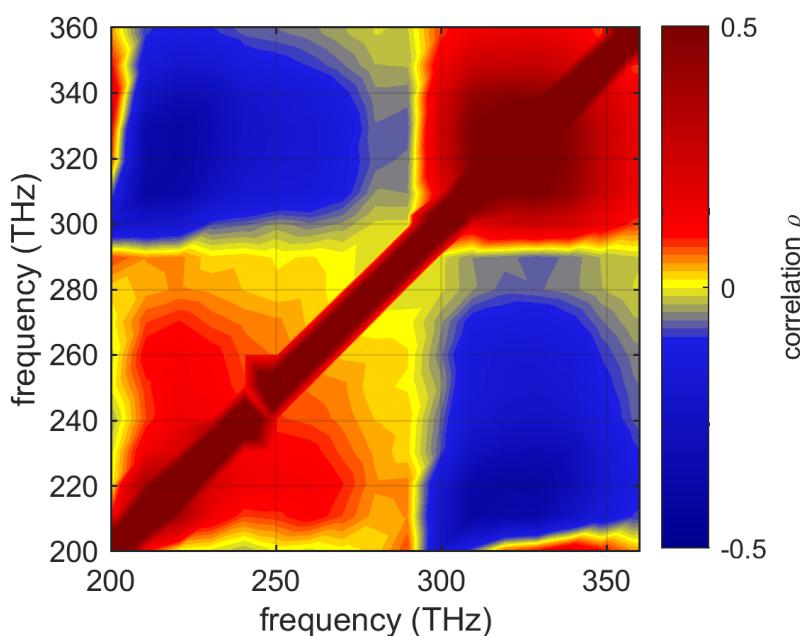
The displacement current in this expression may be replaced by the generated magnetic field. As a result, we can say that the amplitudes of the sine and cosine part of the electric field of electromagnetic radiation correspond to two conjugate variables whose variances obey the uncertainty relation.

- multi-mode case: **Hilbert transform**, phase shift by  $\pi/2$  at each frequency, keep amplitude spectrum identical
- in **electro-optic sampling**: replace  $\lambda/4$  in ellipsometer by  $\lambda/2$ , filter out low- or high-frequency part of probe spectrum

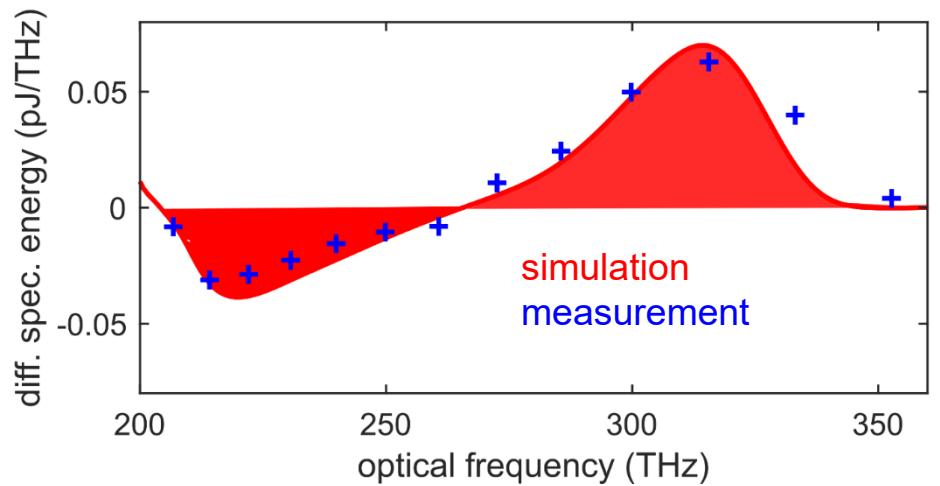
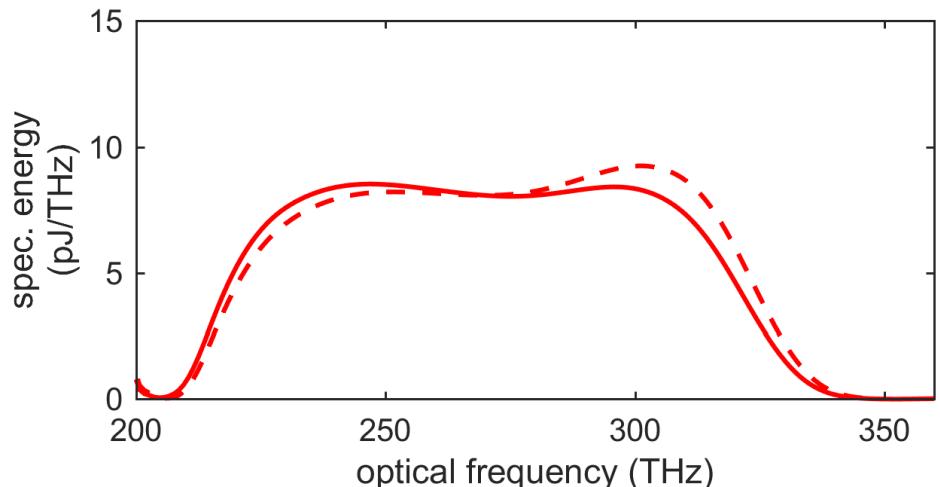


# Anti-Correlated Spectral Amplitude Noise of Probe

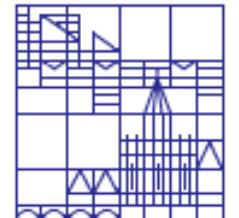
- spectrally integrated probe:  
close to shot-noise limit
- simulated spectra dispersive  
wave at different pump energies
- comparison with experiment



P. Sulzer *et al.*,  
Opt. Lett. **45**, 4714 (2020)

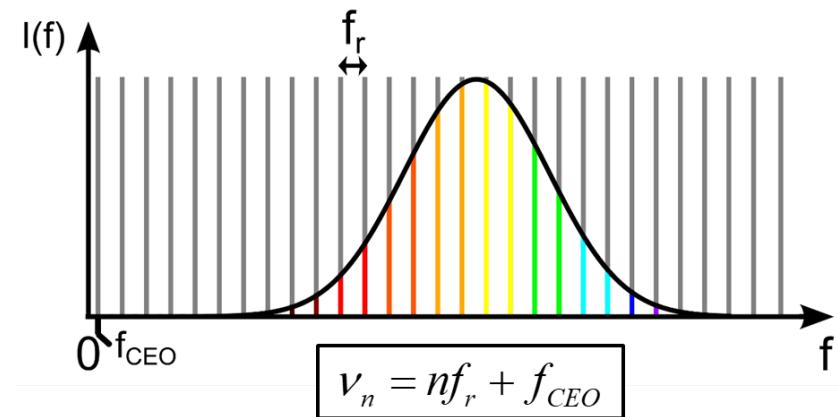
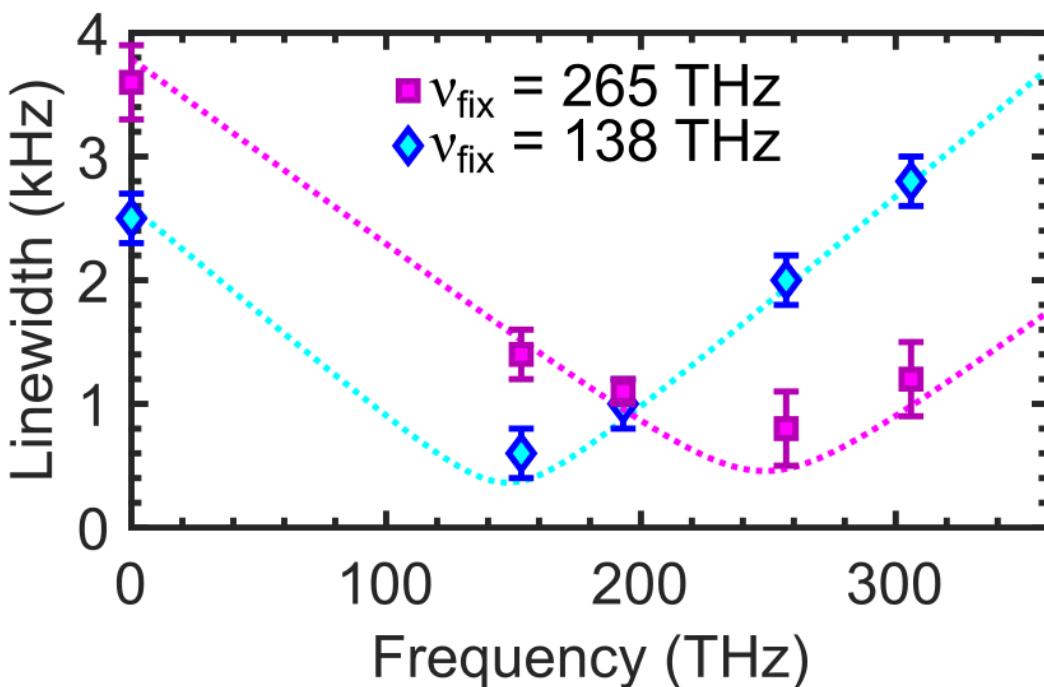


- 2D characterization of  
spectral noise correlations  
on 6-fs probe pulses

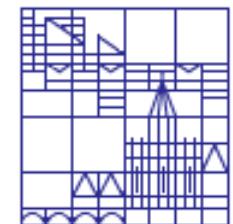


# Towards Time-Domain Squeezing I: Ultralow-Noise Modelocked Er:Fiber Oscillators

- timing jitter in fs pulse train from seed oscillator + Er:fiber amplifier  
→ enhanced pulse-to-pulse amplitude fluctuations  
→ anti-correlated spectral amplitude noise ( $<10^{-4}$ ) in few-fs probe
- design of Er:fiber oscillators with extremely narrow linewidth of femtosecond frequency comb over entire optical bandwidth

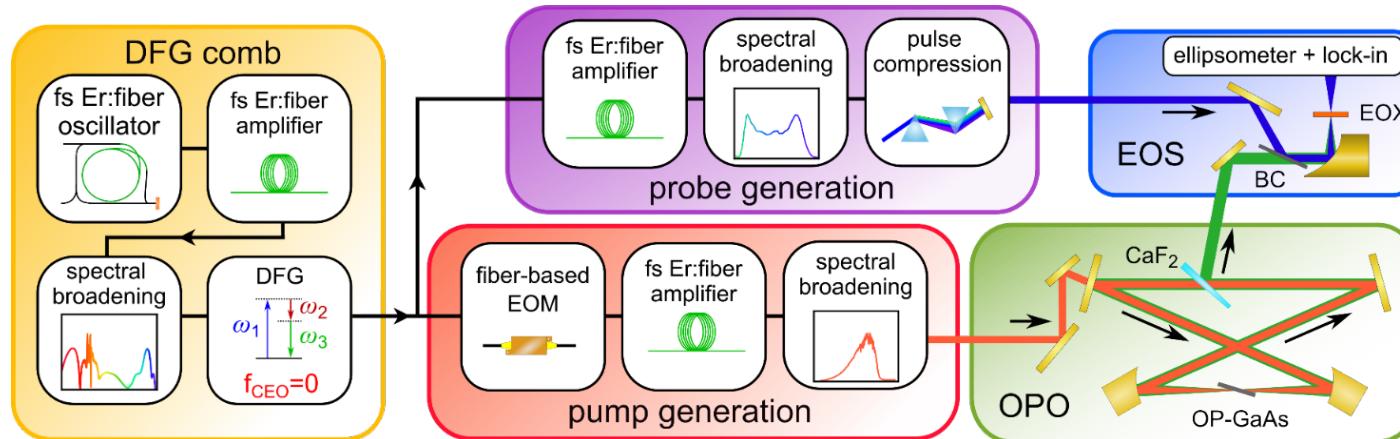


S. R. Hutter *et al.*,  
LPR **17**, 2200907  
(2023)

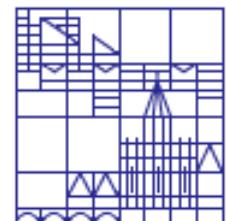
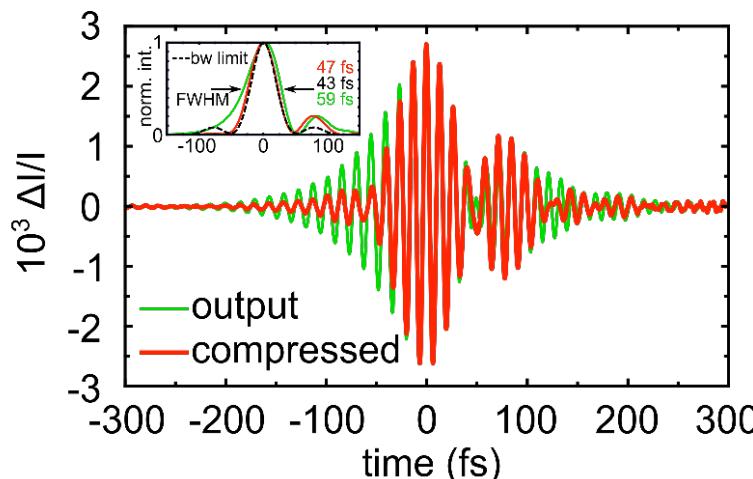


# Towards Time-Domain Squeezing II: Strong Pump from MIR Optical Parametric Oscillator

- optical-parametric oscillator based on OP GaAs pumped by passively phase-stable Er:fiber system

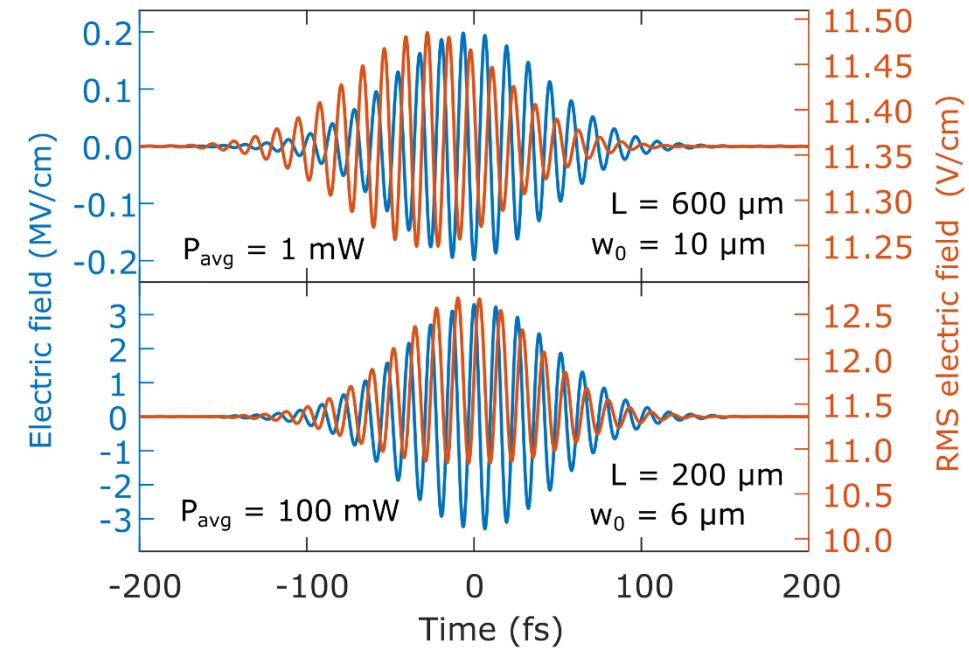
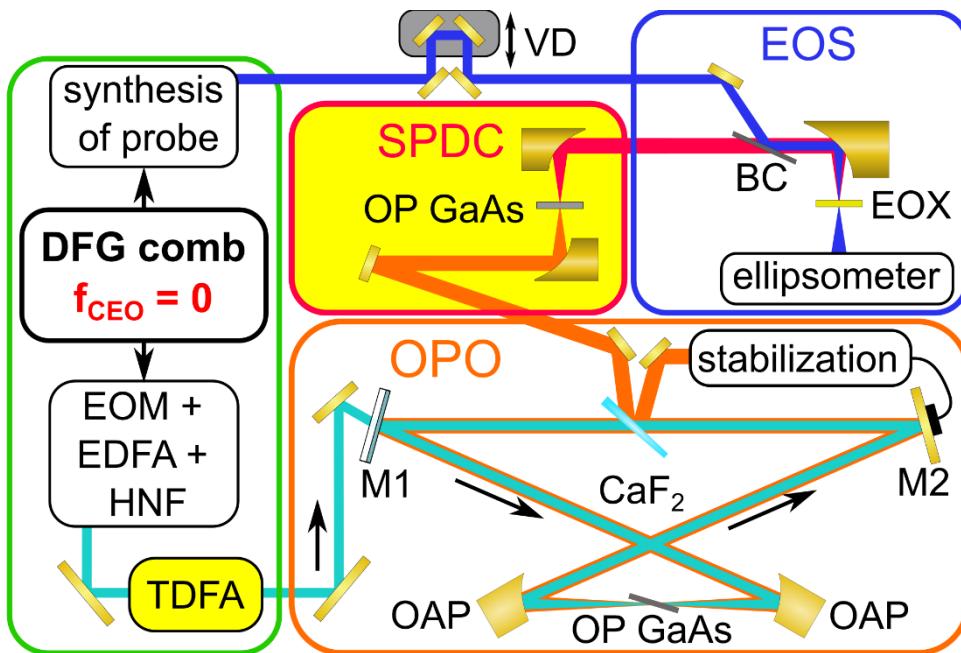


- field-resolved analysis  
of coherent output  
at 75 THz by  
electro-optic sampling

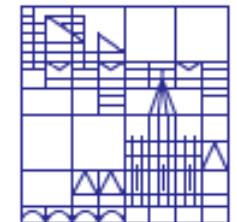


# Towards Time-Domain Squeezing III: Spontaneous Parametric Downconversion

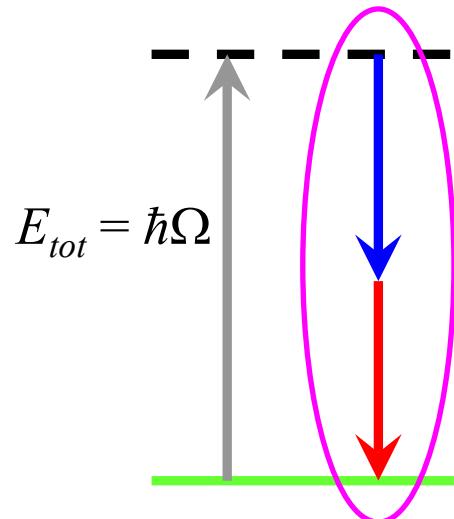
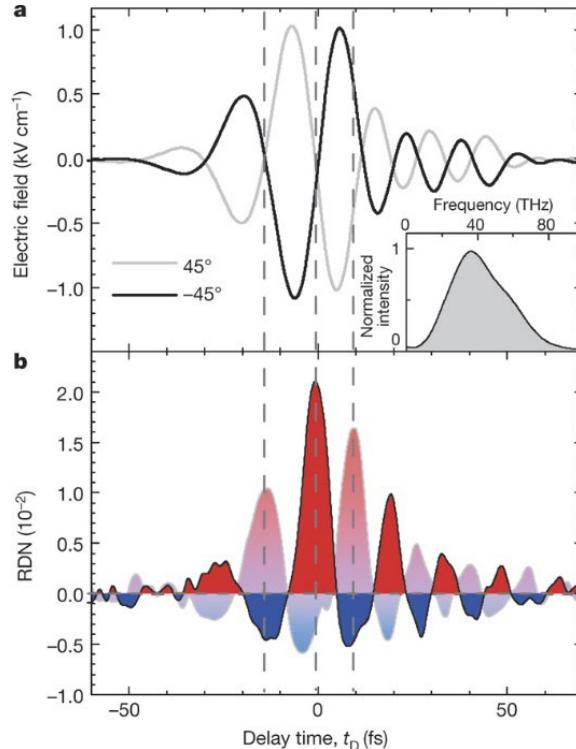
- pumping OP GaAs ( $d_{qpm} = 212.8 \mu\text{m}$ ) with MIR output from OPO  
→ generation of **squeezed vacuum** with **orthogonal polarization**



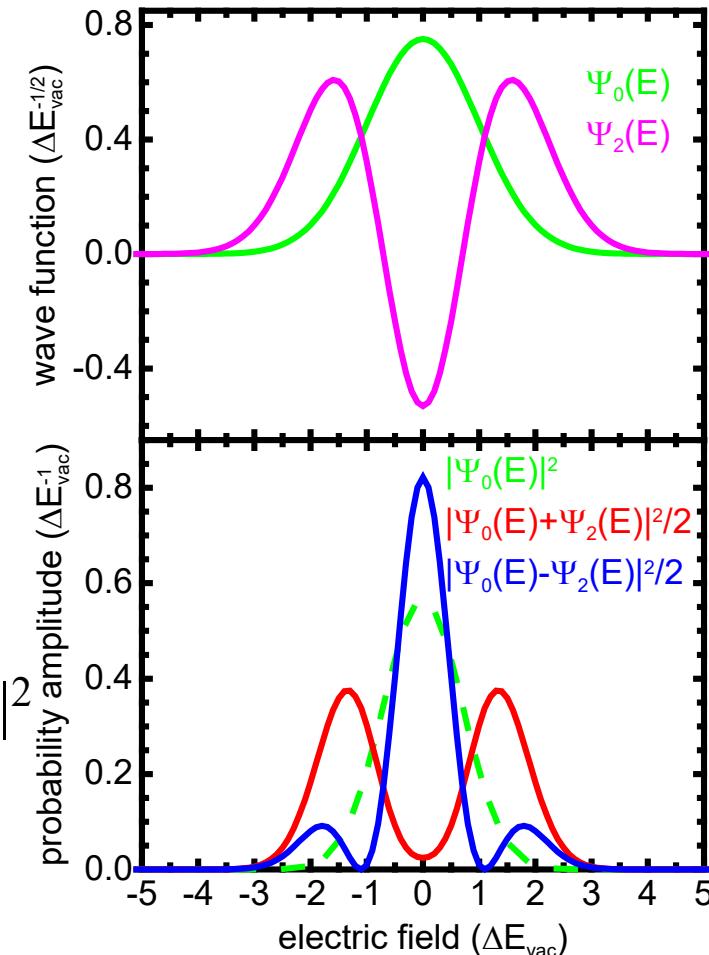
- quantum noise patterns and transmitted pump transients, calculated by first-order perturbation theory assuming geometries optimized for present and future performance



# Squeezed Vacuum $\leftrightarrow$ Photonic Entanglement



- correlated photons from **spontaneous parametric downconversion**  $\leftrightarrow$  quantum noise patterns from **Pockels distortions of reference frame**:
- superposition** of electric-field wave functions for **vacuum** and **2-photon** Fock states in probed space-time volume
- quantum statistics** evolves according to  $|\Psi_0(E) + \exp(-i\Omega t)\Psi_2(E)|^2$
- $\rightarrow$  local **squeezing** and **anti-squeezing** of probability amplitudes



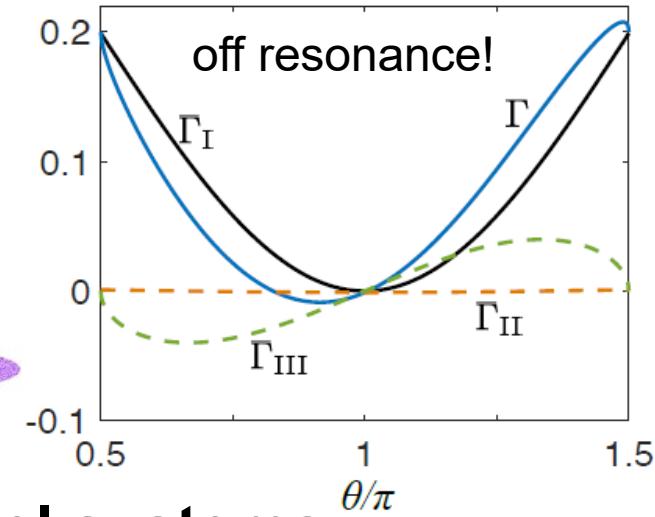
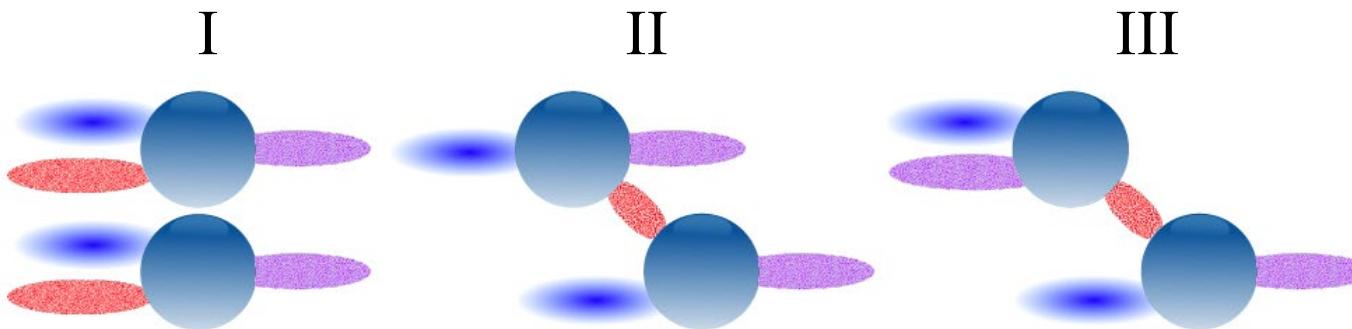
## Part III

# Theoretical Understanding: Overview

Lots of practical predictions  
for experiment to check out...

# Theory: Quantum Nonlinearity

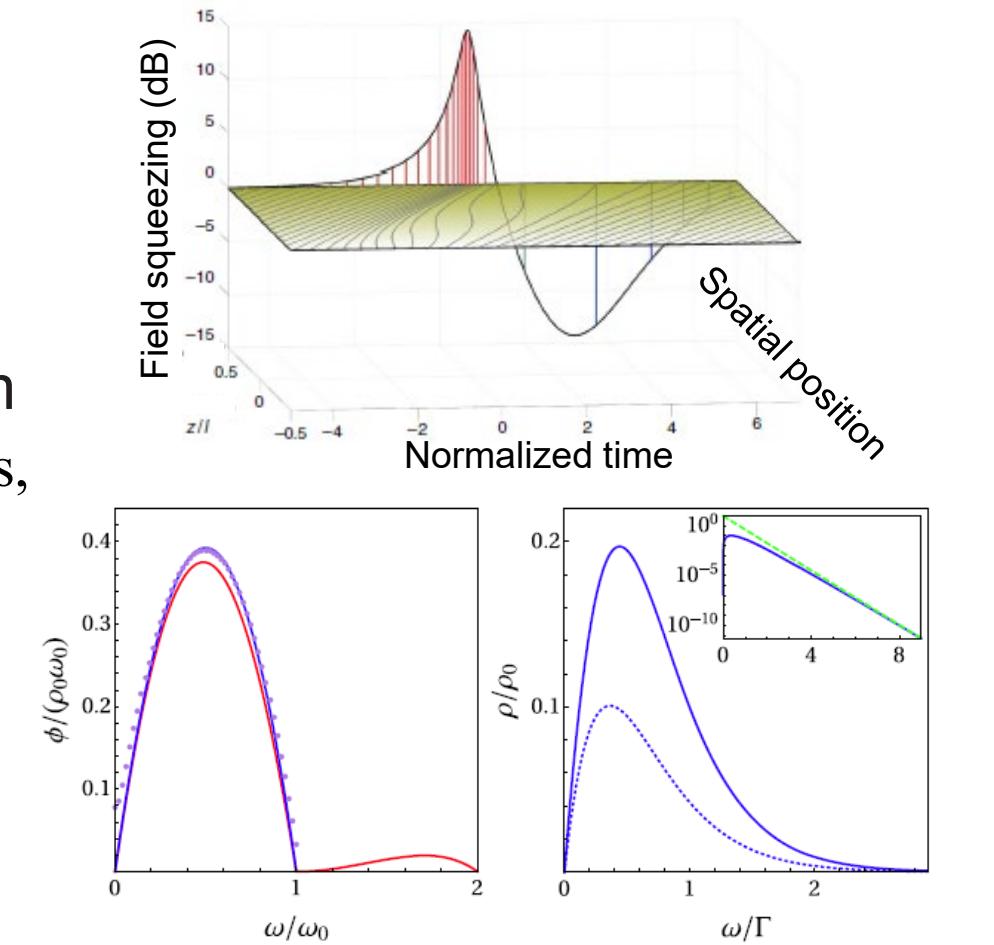
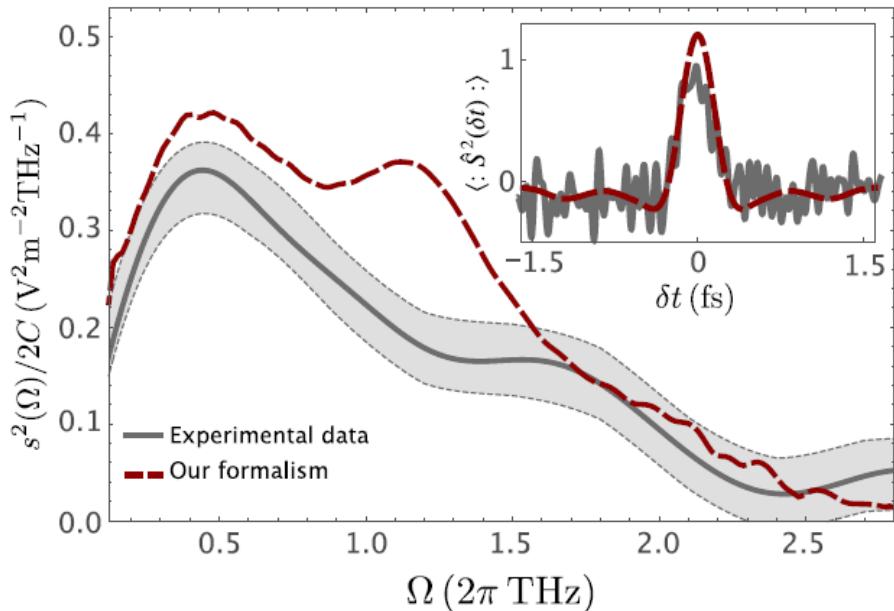
- nonlinear susceptibility electro-optic process: quantum treatment  
M. Kizmann, A. S. Moskalenko, A.L., G. Burkard, S. Mukamel,  
Laser Photonics Rev. **16**, 2100423 (2022)



- model: ensemble of **noninteracting three-level systems**
- superoperator formalism: include e.g. **anticausal processes**
- I: “conventional” part analogous to “**classical**” susceptibility → enhanced fluctuations due to **coupling with THz quantum vacuum**
- II+III: **quantum contributions** → **correlations** between molecules mediated by **THz** virtual or real photons, independent of THz input

# Theory: Time Flow and Accelerated Frames

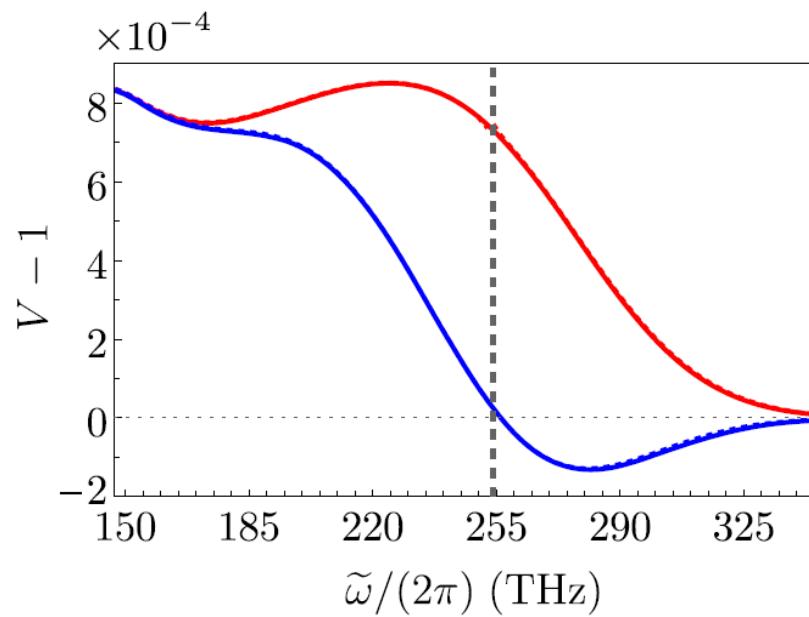
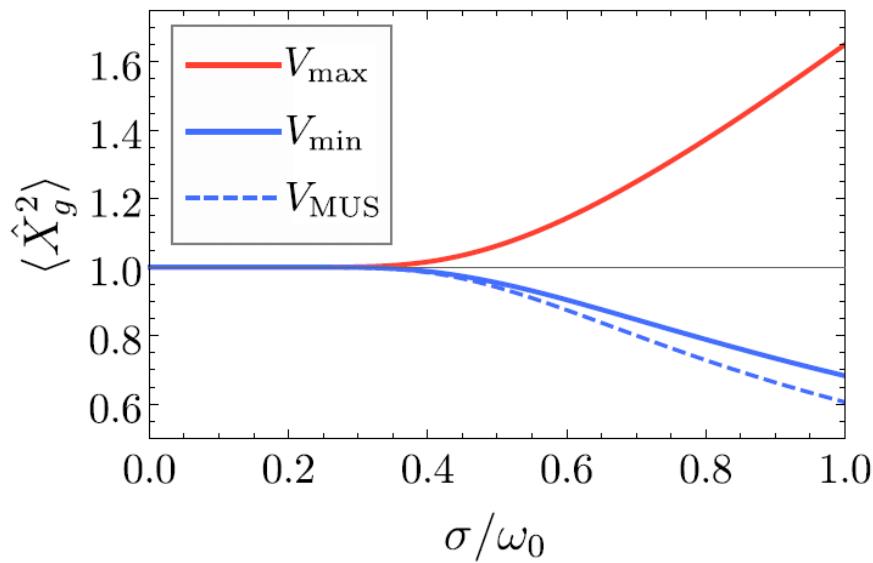
- time-flow picture of squeezing:  
M. Kizmann, T. L. M. Guedes,  
A. S. Moskalenko, G. Burkard *et al.*,  
Nature Phys. **15**, 960 (2019)
- spectra of quantum emission from  
accelerated frames: T. L. M. Guedes,  
M. Kizmann, G. Burkard, A. S. Moska-  
lenko *et al.*, PRL **122**, 053604 (2019)



- light-matter interactions in time-  
domain macroscopic QED:  
F. Lindel, R. Bennett, and S. Y. Buhmann,  
PRA **102**, 041701; NJP **22**, 093014 (2020)

# Theory: Unruh-DeWitt Detector

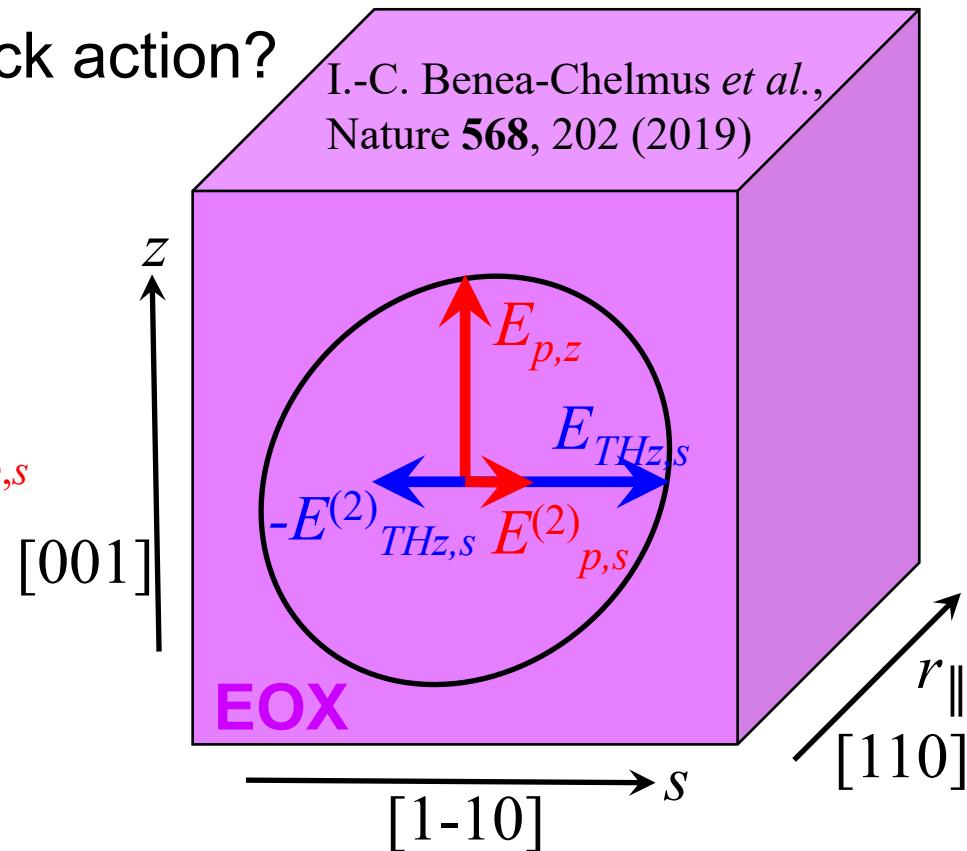
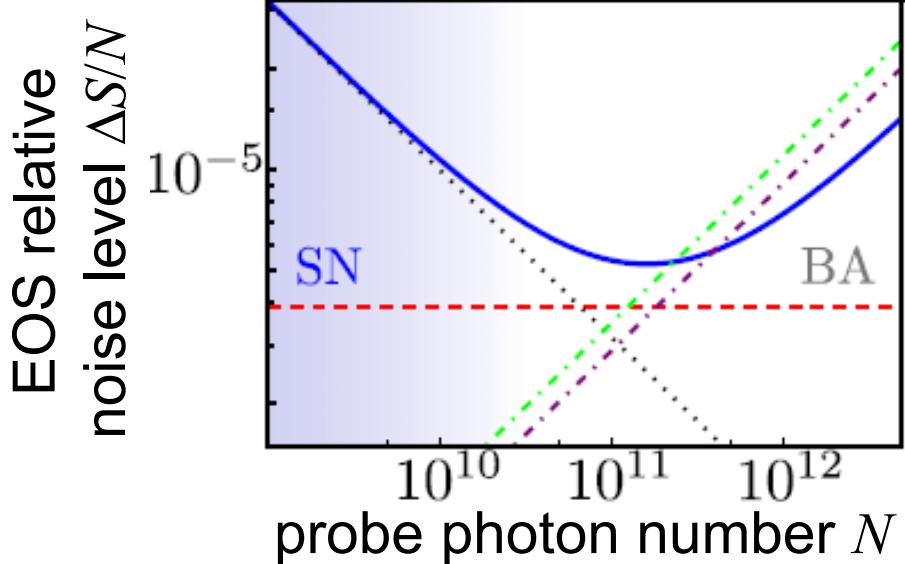
- relativistic quantum field theory of electro-optic sampling: solve Klein-Gordon equation for each field mode and polarization direction  
S. Onoe , T. L. M. Guedes, A. S. Moskalenko, A.L., G. Burkard, T. C. Ralph,  
Phys. Rev. D **105**, 056023 (2022)



- **subcycle activation** of electric-field detector maps **virtual photon-antiphoton** pairs  $\hat{a}_\omega \hat{a}_\omega^\dagger$  in quantum vacuum to real probe photons
- **electric field** ( $\lambda/4$ ) → enhanced fluctuations all over probe spectrum  
**Hilbert transform** ( $\lambda/2$ ) → squeezing above probe center frequency

# Theory: Back Action

- quantum electro-optic sampling: back action?
- second-order nonlinear mixing of THz field  $E_{THz,s}$  with probe  $E_{p,z}$   
→ electro-optic signal  $E_{p,s}^{(2)}$
- cascaded  $\chi^{(2)}$  mixing of  $E_{p,z}$  with  $E_{p,s}^{(2)}$   
→ new THz amplitude  $E_{THz,s}^{(2)}$



- small probe photon numbers  $N$ : shot noise  $\Delta S_{sn}$  dominates  
→ **weak measurement** regime
- when vacuum signal  $\Delta S_{eo} \rightarrow \Delta S_{sn}$ : onset of back action  $\Delta S_{ba}$ !

# Summary and Outlook

- **quantum measurement of the electric field:**
  - **test charge:** anharmonically bound valence electrons
  - **amplitude vacuum fluctuations** in free space:  $\Delta E_{vac} = \sqrt{\frac{hc}{\epsilon_0 (\delta l)^4}}$
  - **photon:** measure for **quantum statistics** in **space-time** complex
  - **quantum confinement in all four dimensions** (fully relativistic!): **energy + momentum** information  $\leftrightarrow$  scanning over **time + space**
- **towards time-domain manipulation of quantum field:**
  - acceleration local reference frame + uncertainty principle  
 $\rightarrow$  highly **correlated** and **synchronal** photon states
  - **relativistic quantum physics** in curved spacetime
  - mid-IR/THz quantum technology  $\leftrightarrow$  **correlated electrons...**

# Take Home...

- Heisenberg: nature can borrow **energy/mass** and **momentum** on short **time** and **length scales**
  - weak effects in **bare electromagnetism**: coupling constant  $\alpha_{em} = \frac{e^2}{4\pi\epsilon_0\hbar c} \approx 1/137$
  - electric-field **vacuum amplitude**:  $\Delta E_{vac} = \sqrt{\frac{hc}{\epsilon\epsilon_0(\delta l)^4}} t \sim \sqrt{\int d\Omega \langle \Phi_0 | a_\Omega a_\Omega^\dagger | \Phi_0 \rangle}$
  - **photon**: measure for **quantum statistics in space-time complex**
  - **intermediate coupling in condensed matter**, e.g. electron-phonon, electron-electron interactions: dynamics in semiconductor quantum dots, buildup of quantum correlations...
- **fun fact: we may ourselves be about 98.5% virtual!**
  - rest **mass nucleon** (proton, uud; neutron, udd): **1 GeV/c<sup>2</sup>**
  - approximate **mass 3 valence quarks**: **15 MeV/c<sup>2</sup>**
  - strong interaction:  $\alpha_{si} \approx 1 \rightarrow$  **virtual quark-antiquark pairs + gluons** dominate

[https://en.m.wikipedia.org/wiki/  
File:Quark\\_structure\\_neutron.svg](https://en.m.wikipedia.org/wiki/File:Quark_structure_neutron.svg)

