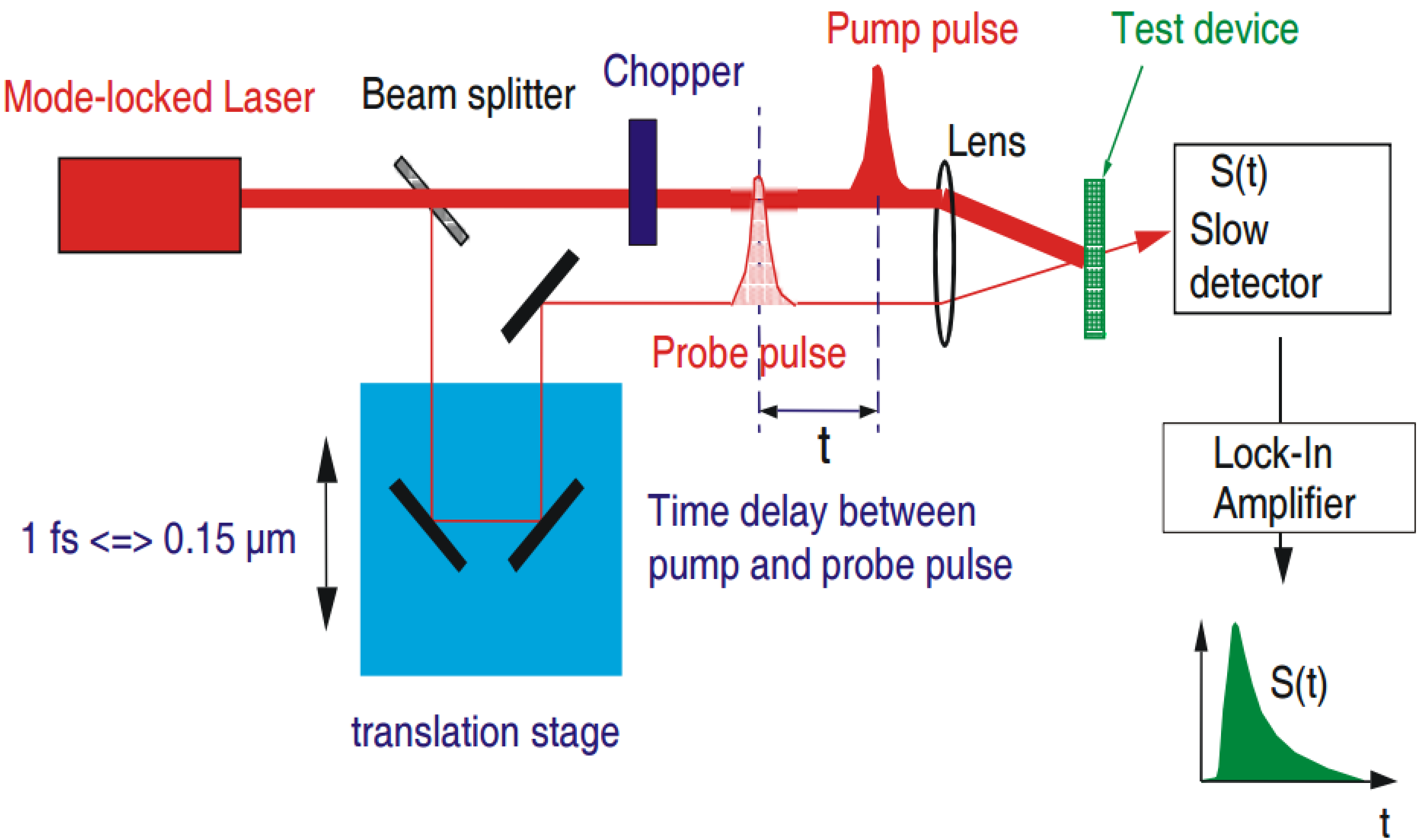
Dr. Katalin Kamarás (Wigner, SZFI) kamaras.katalin@wigner.hu
Dr. Sándor Bordács (BME, FT) bordacs.sandor@wigner.bme.hu

# Optical Spectroscopy in Materials Science
# Time-resolved spectroscopy

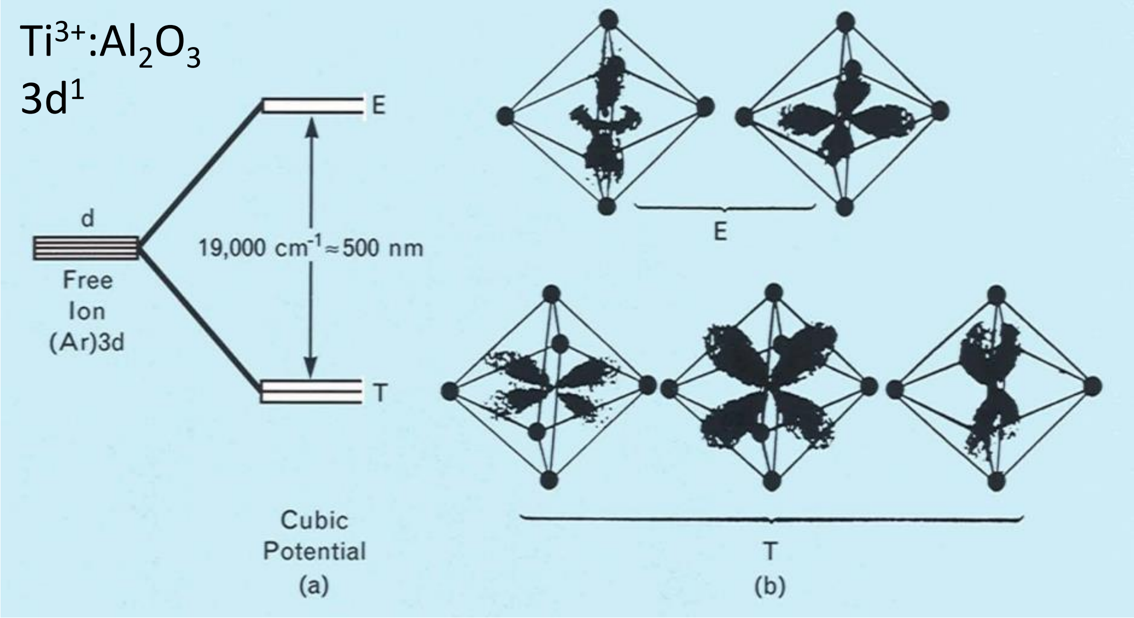
# Time-resolved absorption/reflectivity/luminescence ...

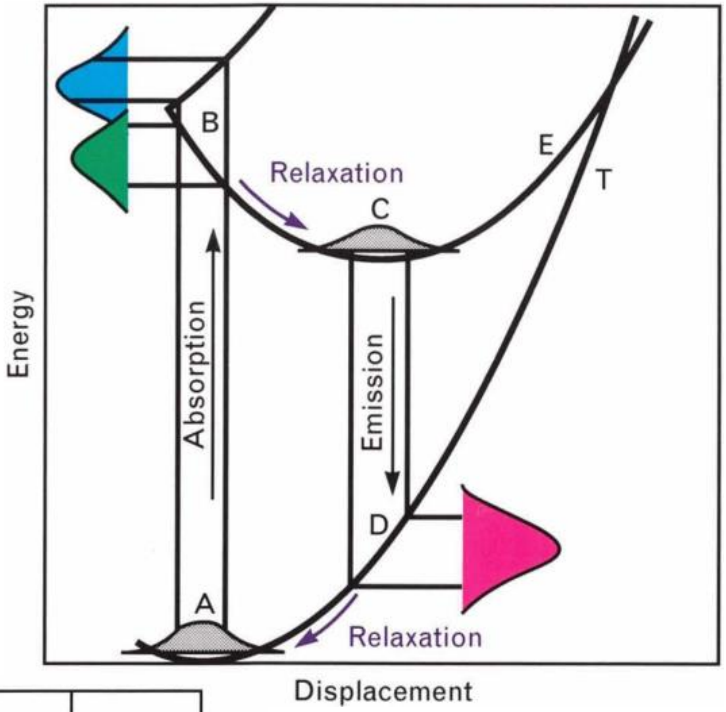


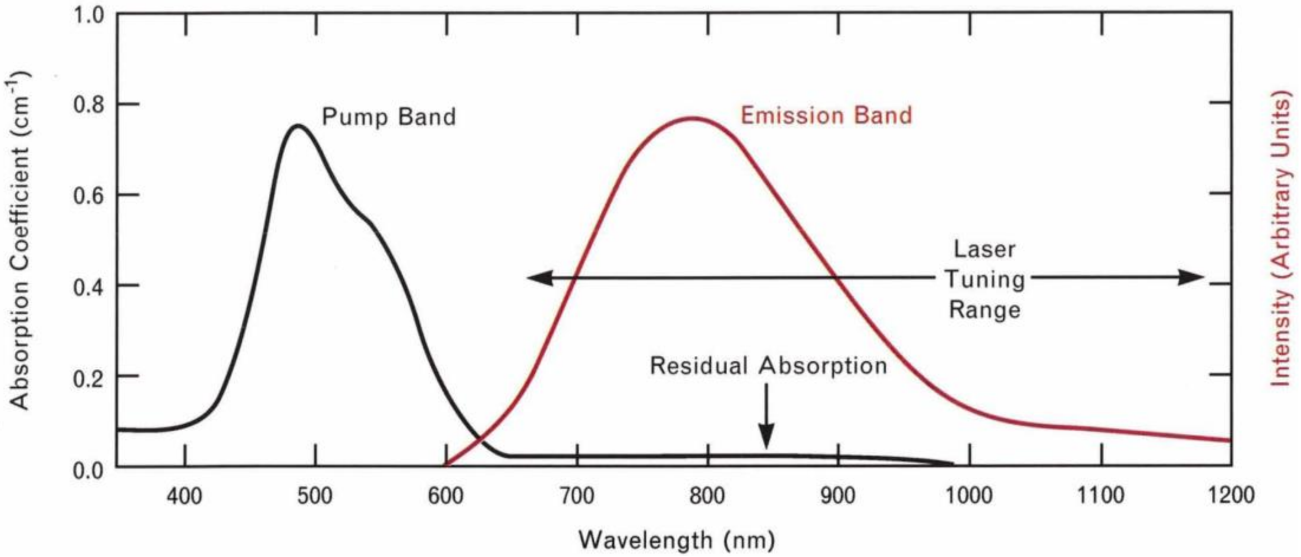

U. Keller ETH, Zürich

# Ti:sapphire LASER

$Ti^{3+}:Al_2O_3$
$3d^1$

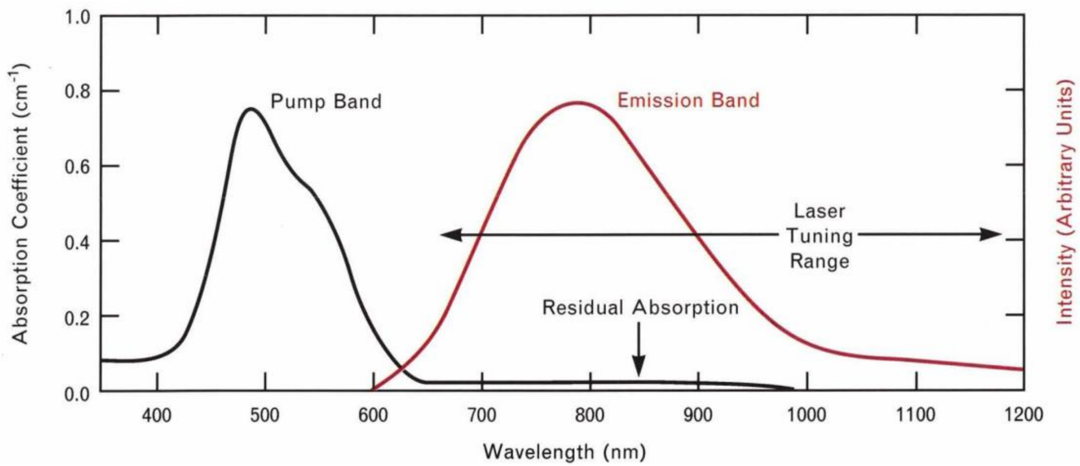


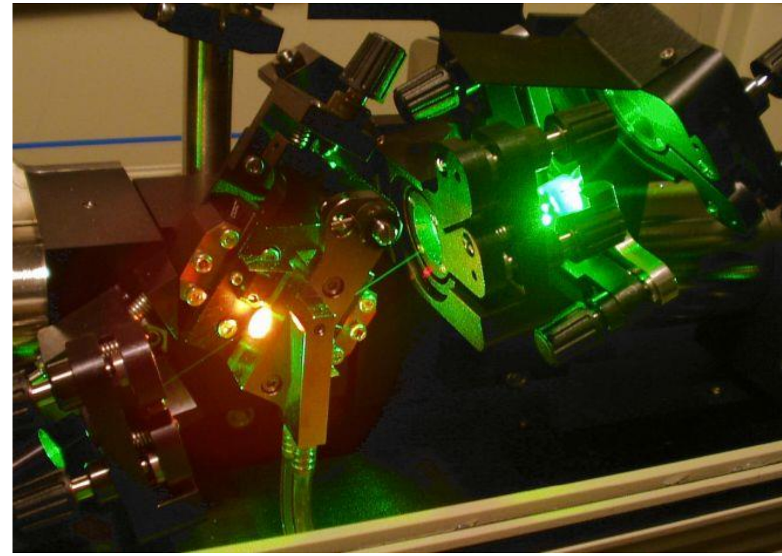



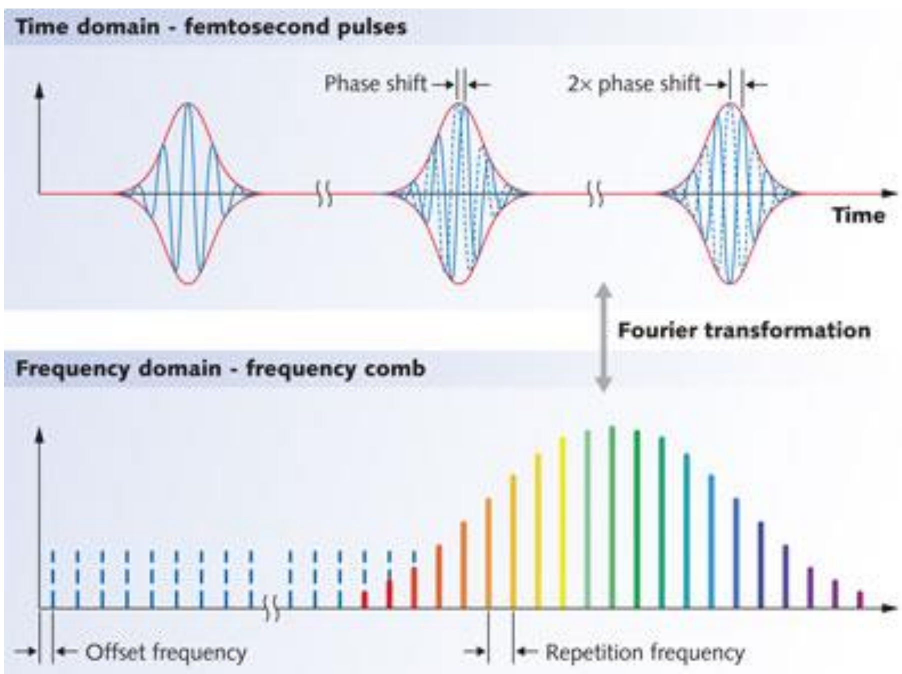

K.F. Wall and A. Sanchez. The Lincoln Laboratory Journal, 2 447 (1990)

# Ti:sapphire LASER




Pump: green, lasing: NIR

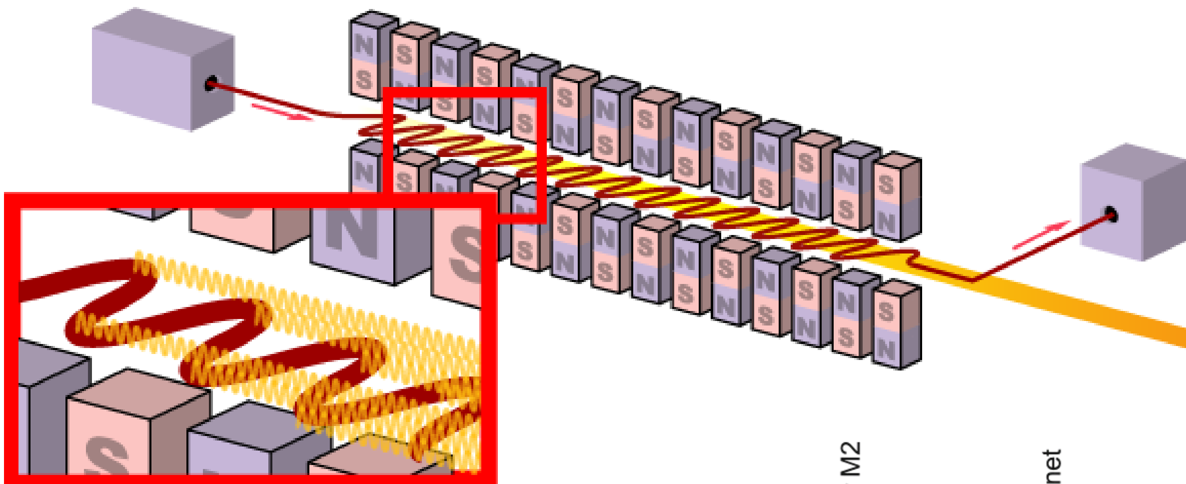


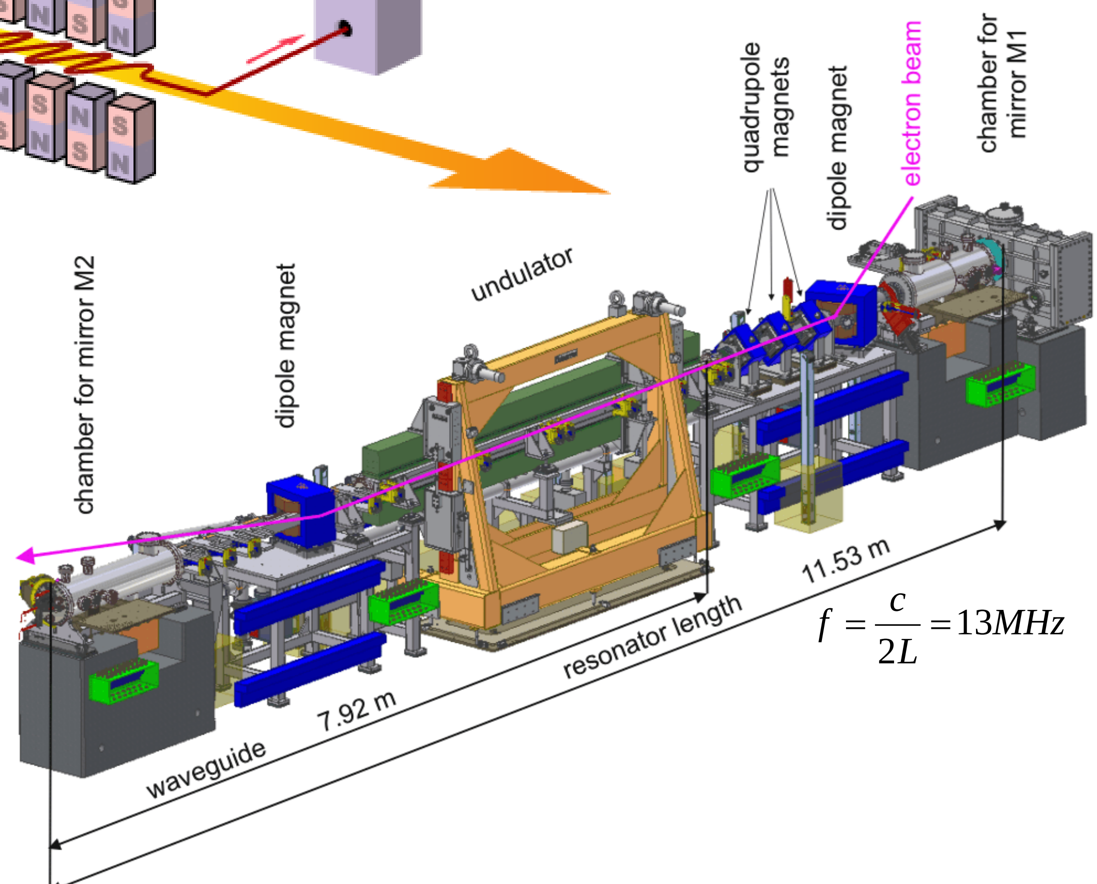



Cental wavelength: 800 nm (375 THz)
Pulse width: 10-100 fs
Repetition rate: 80 MHz ( $\frac{c}{2L}$ ), resonantor: ~2 m

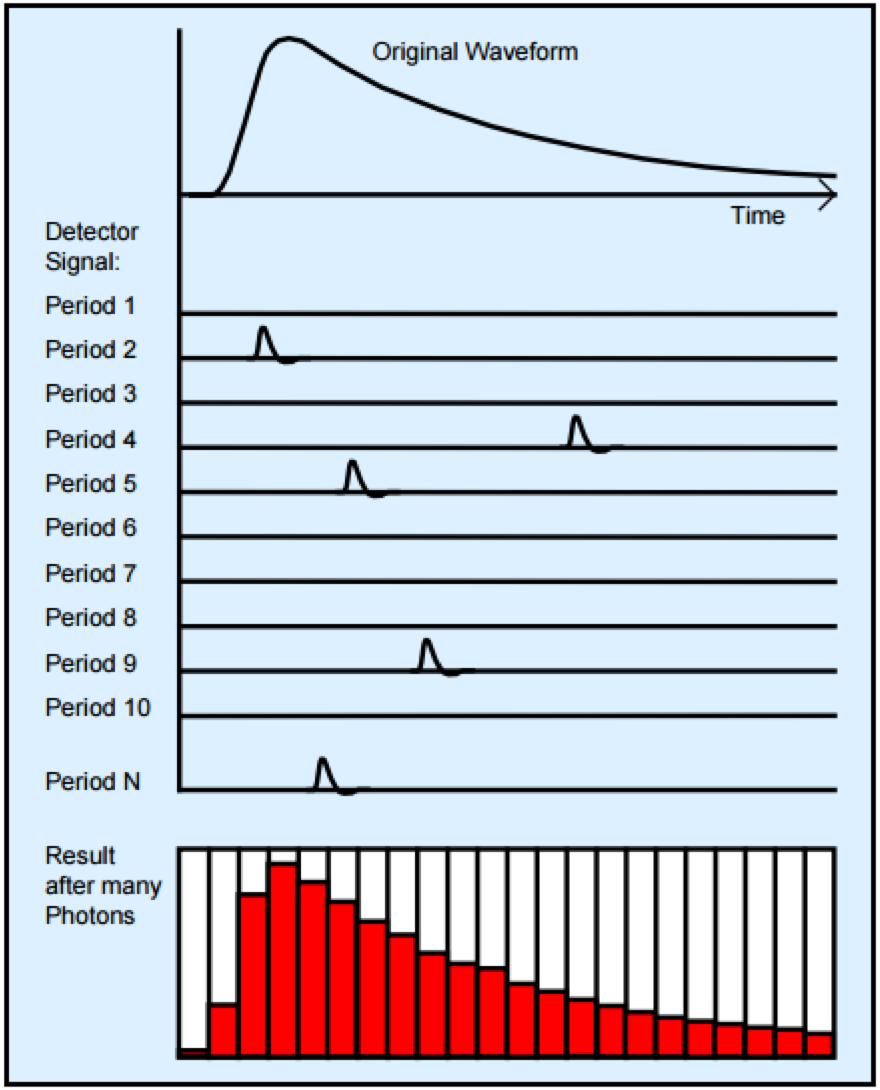
# Free Electron LASER (FEL)

Lasing medium: relativistic free electrons traveling in an undulator
Interaction between the electron beam and the E-field of the radiation leads to bunching and coherent radiation
Most widely tuneable LASER: from microwave to X-ray





# Time-Correlated Single Photon Counting



# Ahmed Zewail - Facts

Ahmed H. Zewail

**Born:** 26 February 1946, Damanhur, Egypt

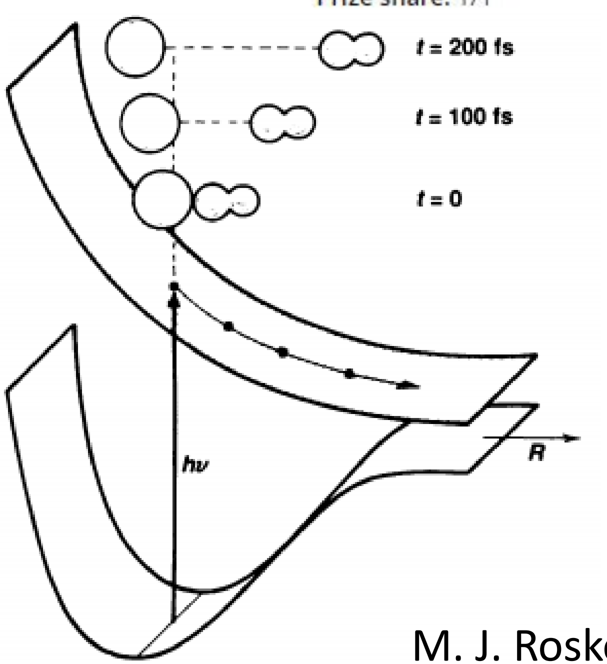
**Died:** 2 August 2016

**Affiliation at the time of the award:** California Institute of Technology (Caltech), Pasadena, CA, USA

**Prize motivation:** "for his studies of the transition states of chemical reactions using femtosecond spectroscopy"

**Field:** chemical kinetics, physical chemistry

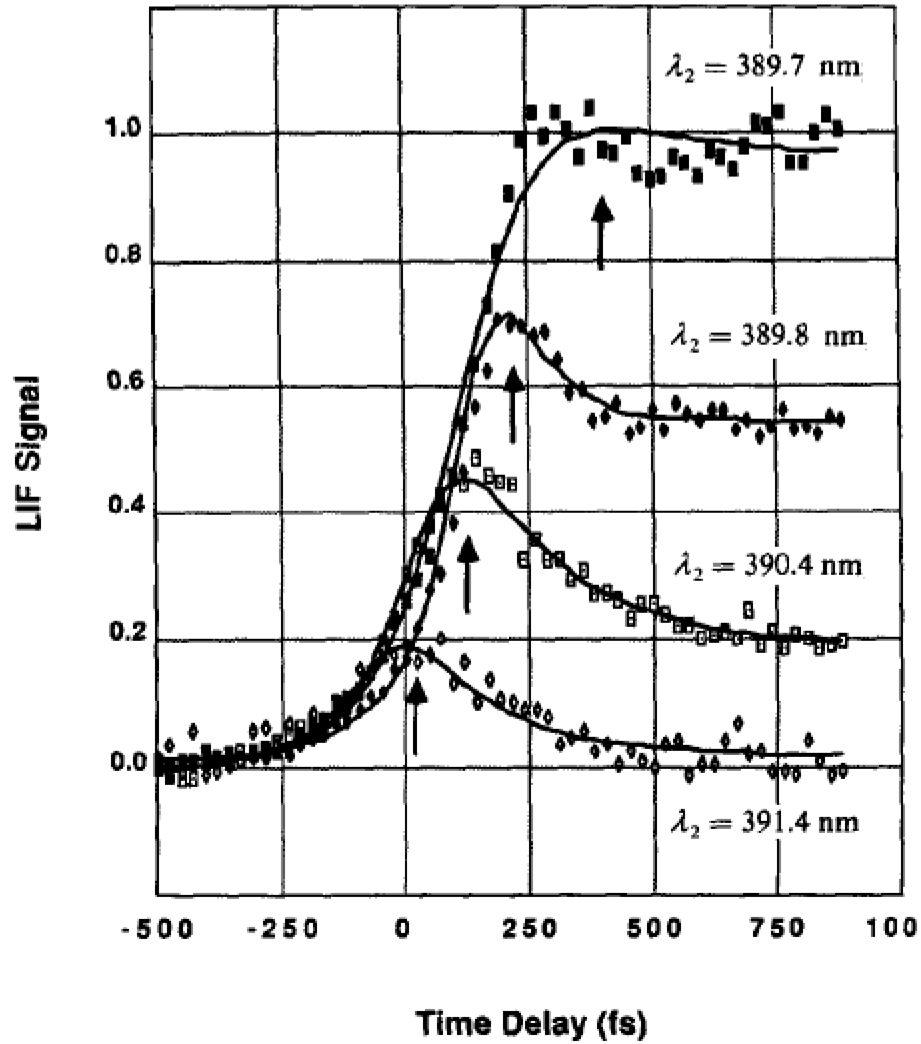
**Prize share:** 1/1




$$ICN^* \rightarrow [I \cdots CN]^{\ddagger *} \rightarrow I + CN$$

LIF: Laser Induced Fluorescence

Fluorescence signal is measured after absorption

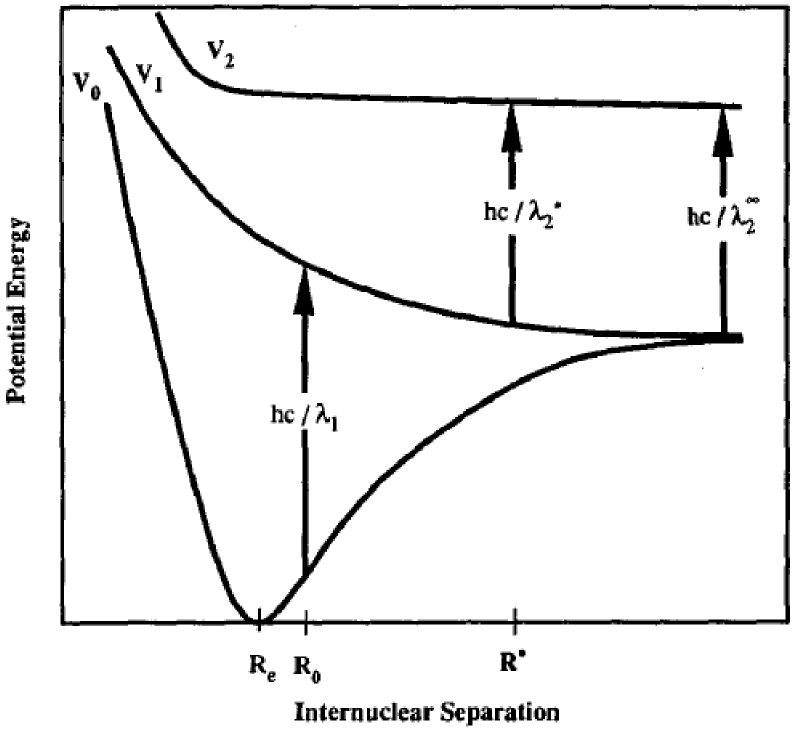


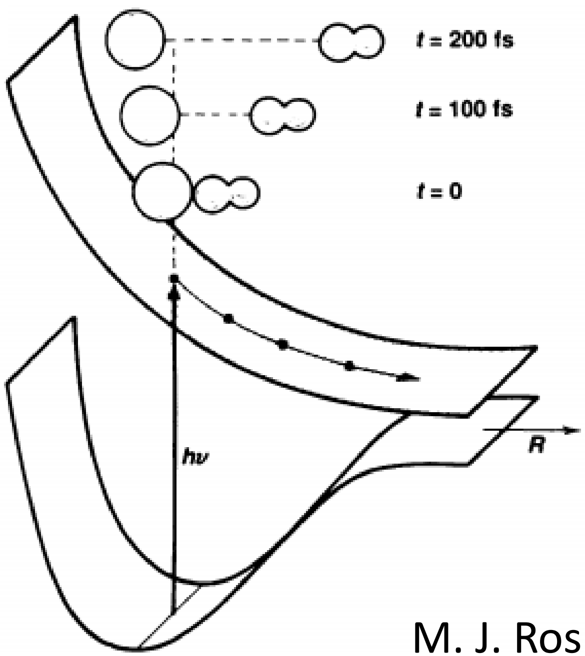

M. J. Rosker, M. Dantus, A. H. Zewail, J. Chem. Phys. **89**, 6113 (1988).

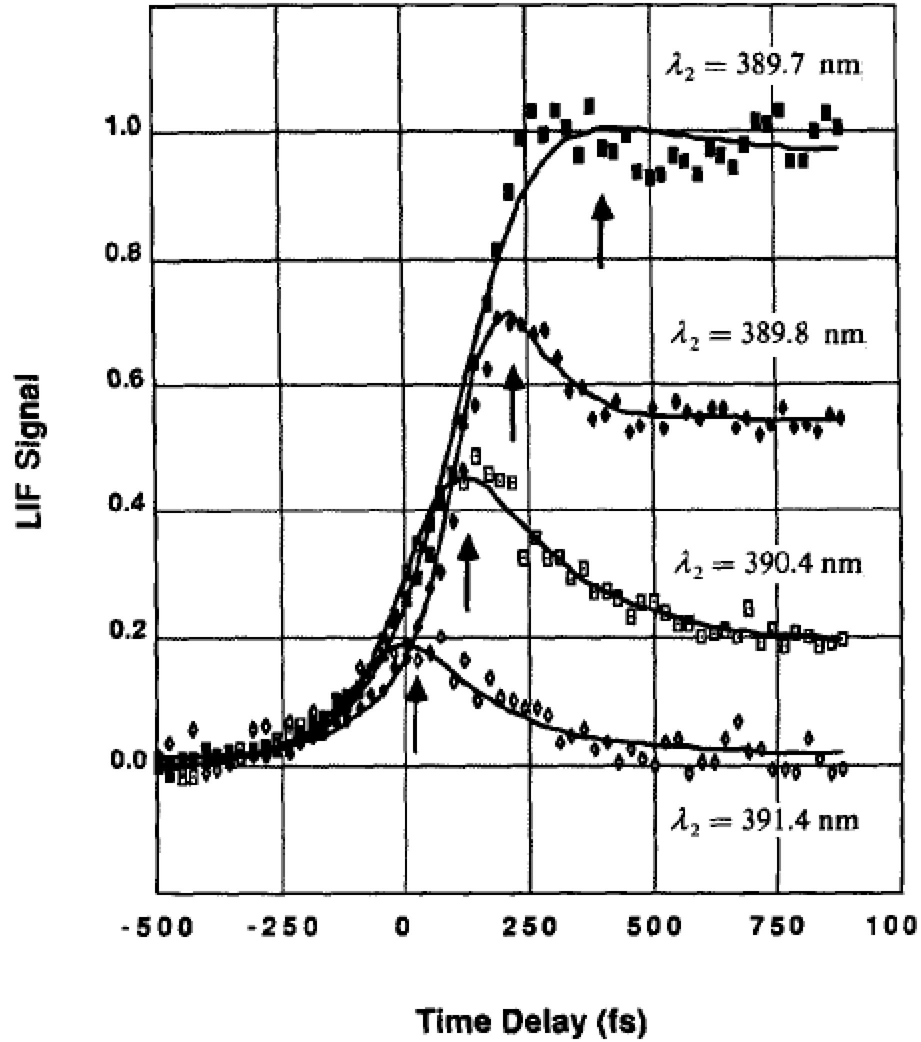
$$ICN^* \rightarrow [I \cdots CN]^{\ddagger *} \rightarrow I + CN$$

LIF: Laser Induced Fluorescence

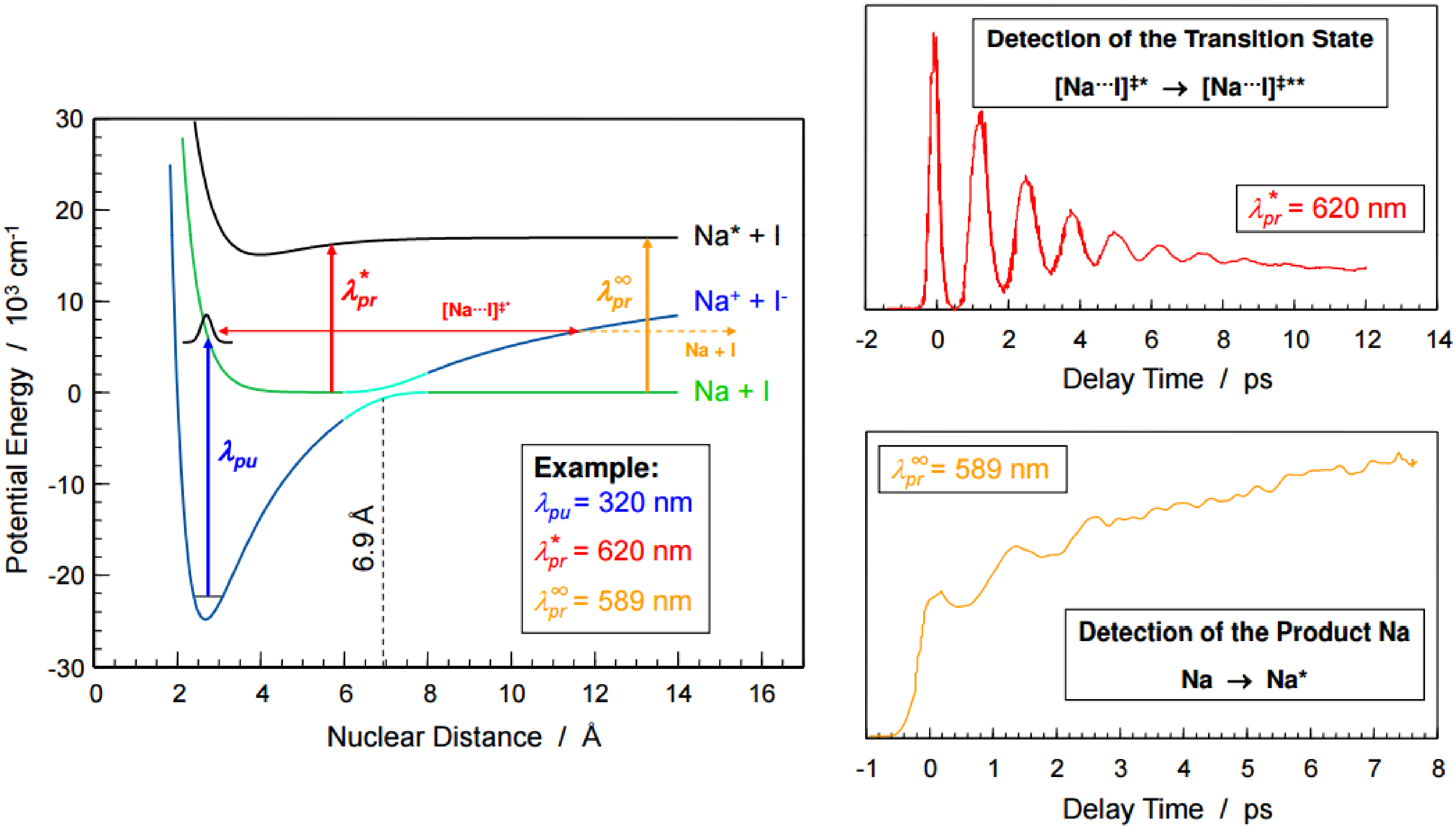
Fluorescence signal is measured after absorption



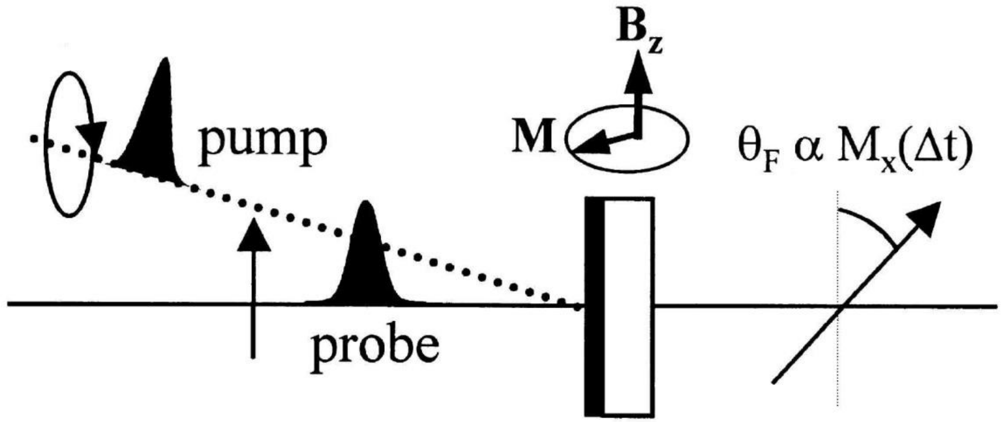

M. J. Rosker, M. Dantus, A. H. Zewail, J. Chem. Phys. **89**, 6113 (1988).



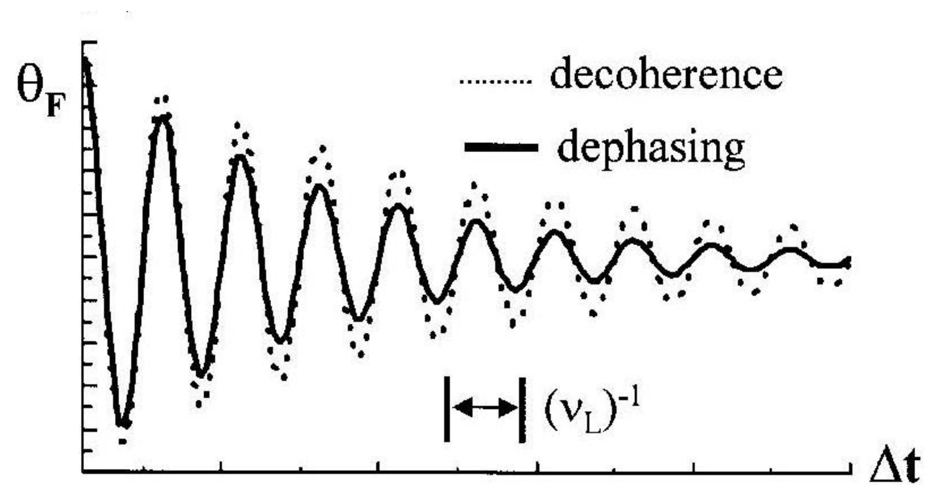


M. J. Rosker, M. Dantus, A. H. Zewail, J. Chem. Phys. **89**, 6113 (1988).
A. Materny, Jacobs University Bremen (2009).

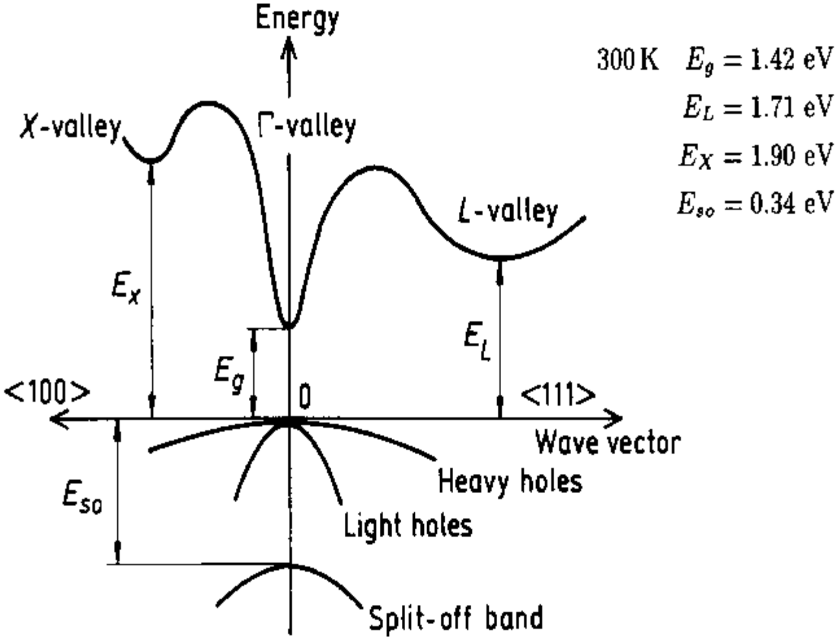
# Time-domain magneto-optical spectroscopy

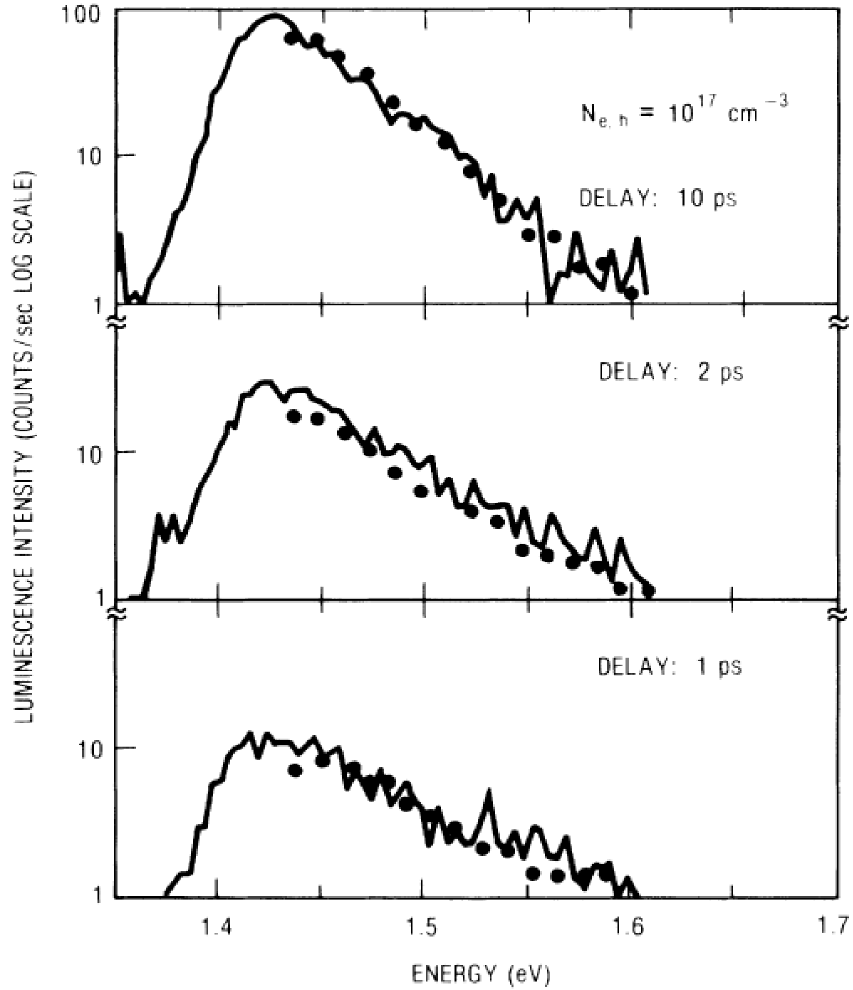



$$\theta_{\mathrm{F}}(\Delta t) = A\mathrm{e}^{-\Delta t/T_2^*}\cos(2\pi\nu_{\mathrm{L}}\Delta t)$$




J.M. Kikkawa et al., Physica E **9** (2001) 194.



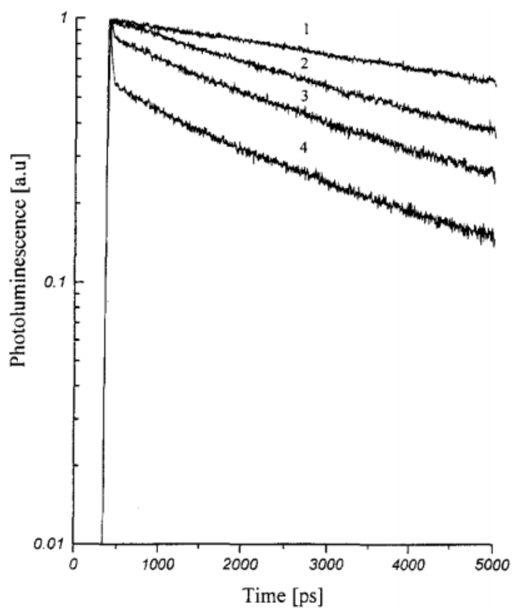



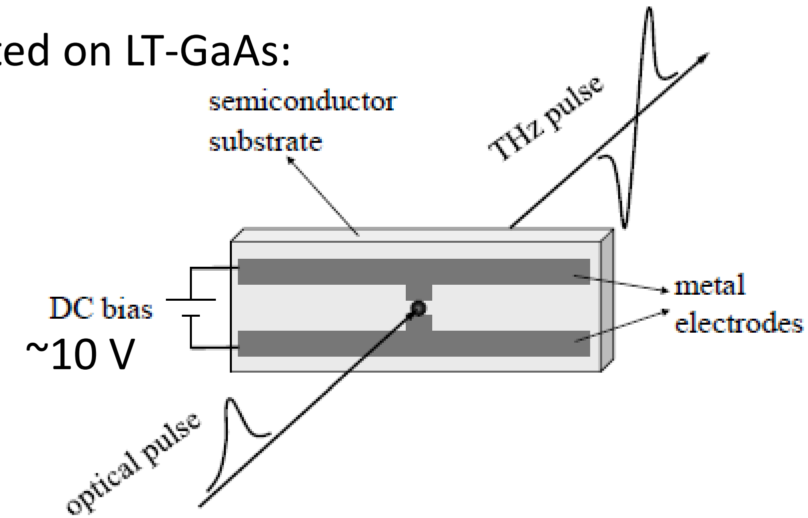

Fig. 1. Semilog plots of time resolved photoluminescence (PL) measured at various photon energies of 0.5 $\mu$m GaAs crystal. Curves 1, 2, 3 and 4 correspond to PL energy of: 1.43 eV, 1.49 eV, 1.57 eV, 1.65 eV, respectively.

Applied Surface Science 106 (1996) 457-465

J. Shah et al., Phys. Rev. Lett. (1987).

# THz spectroscopy: THz source

Dipole antenna fabricated on LT-GaAs:




LT-GaAs:
-recombination: $\tau_{rec}$=0.3 ps
-mobility: μ=200 $cm^2$/Vs
($\tau_{scat}$=30 fs)
-gap: $E_g$=840 nm

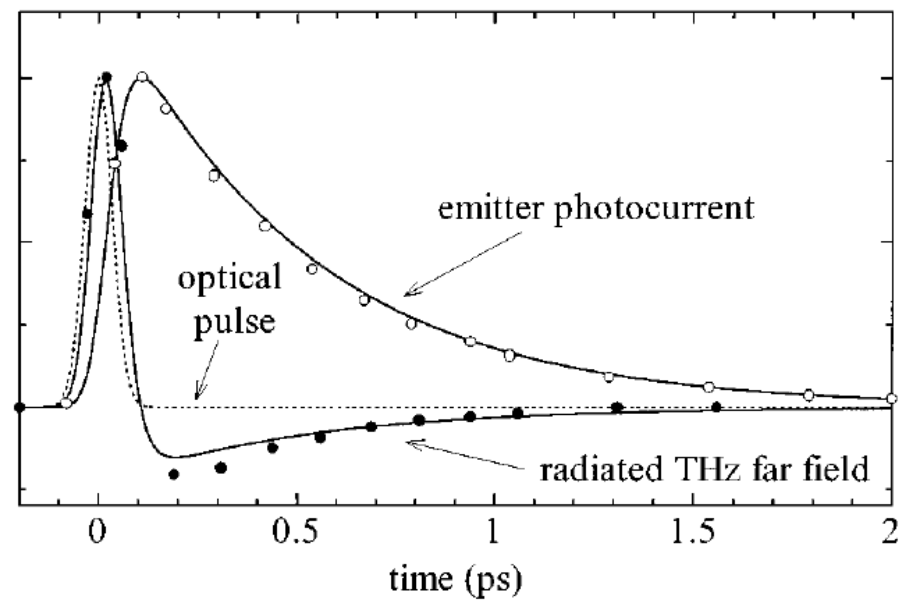
Ti:Sapphire LASER:
-central wavelength: λ=800 nm
-pulse width: τ<100 fs

$$E_{THz} \propto \frac{\partial j}{\partial t}$$

$$j(t) = \int P(t-t')[en(t')v(t')]dt'$$

$$[\ldots] = e\exp(-t'/\tau_{rec})\mu(1-\exp(-t'/\tau_{scat}))\frac{V}{d}$$

$$\nu_{Max} \sim \frac{1}{\tau_{rec}} = 3\text{ THz}$$





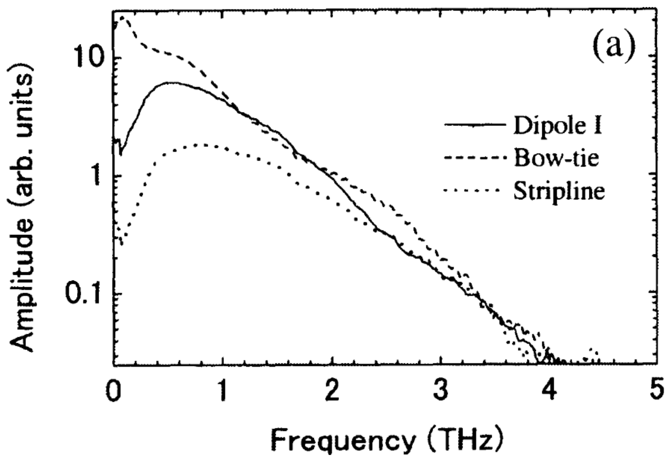
Y. S. Lee: Principles of Terahertz Science and Technology Springer, Berlin (2009)
L. Duvillaret et al. *IEEE J. Sel. Top. Quant. Electronics.* **7** 615 (2001)

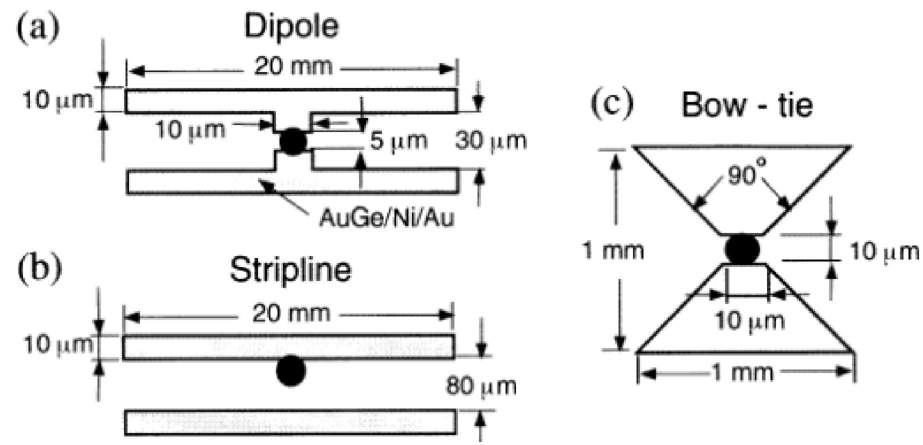
# THz spectroscopy: THz source

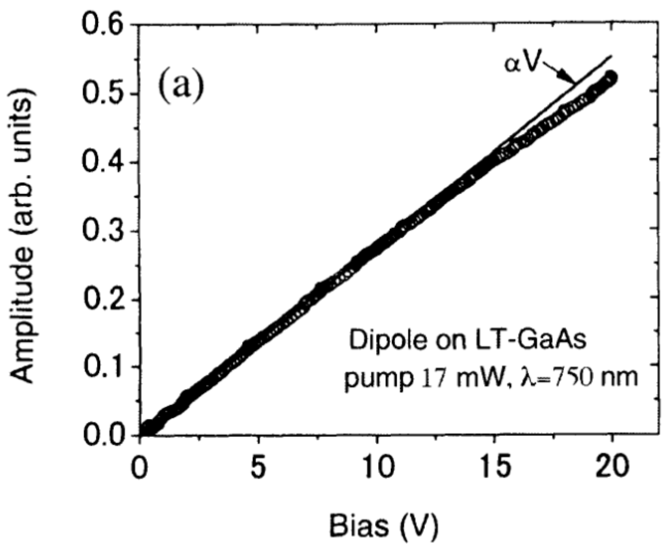
Ti:Sapphire
@ 800nm
20 mW

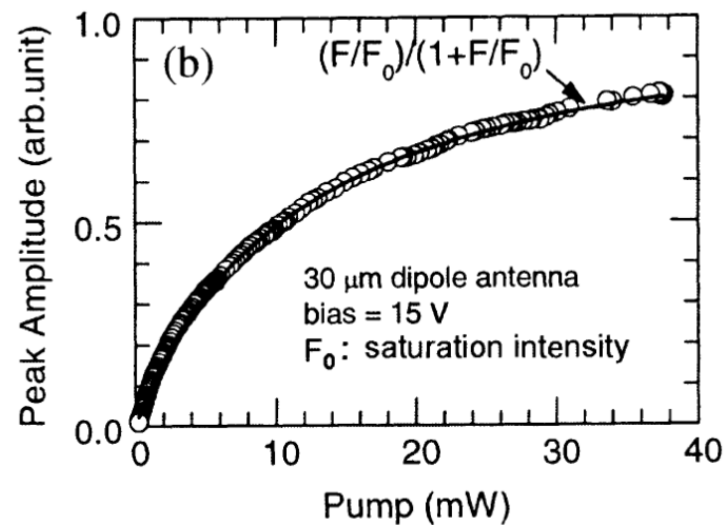
60 V →

THz radiation
2-3 μW



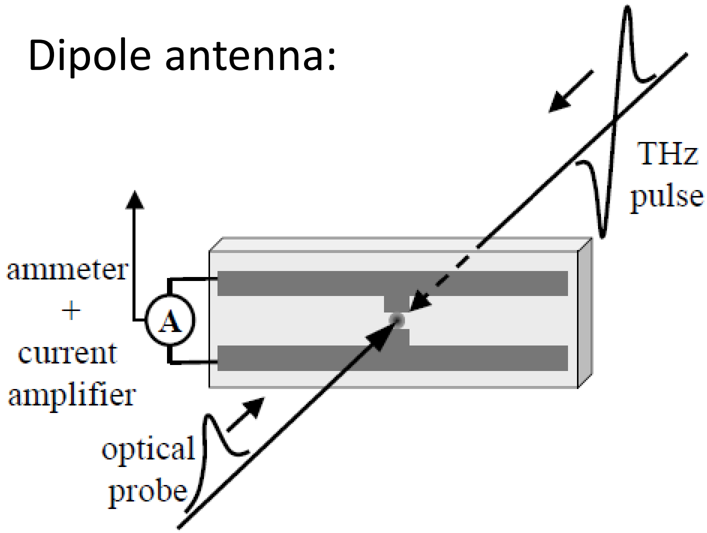



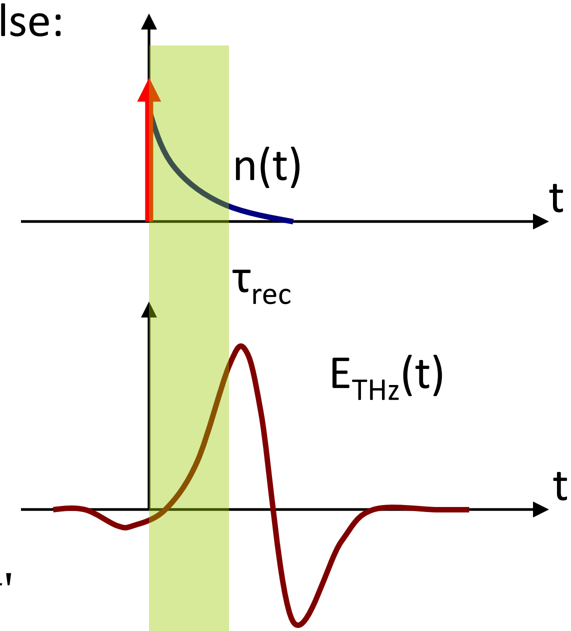

K. Sakai: Terahertz Optoelectronics Springer, Berlin (2005)

# THz spectroscopy: time-resolved detection

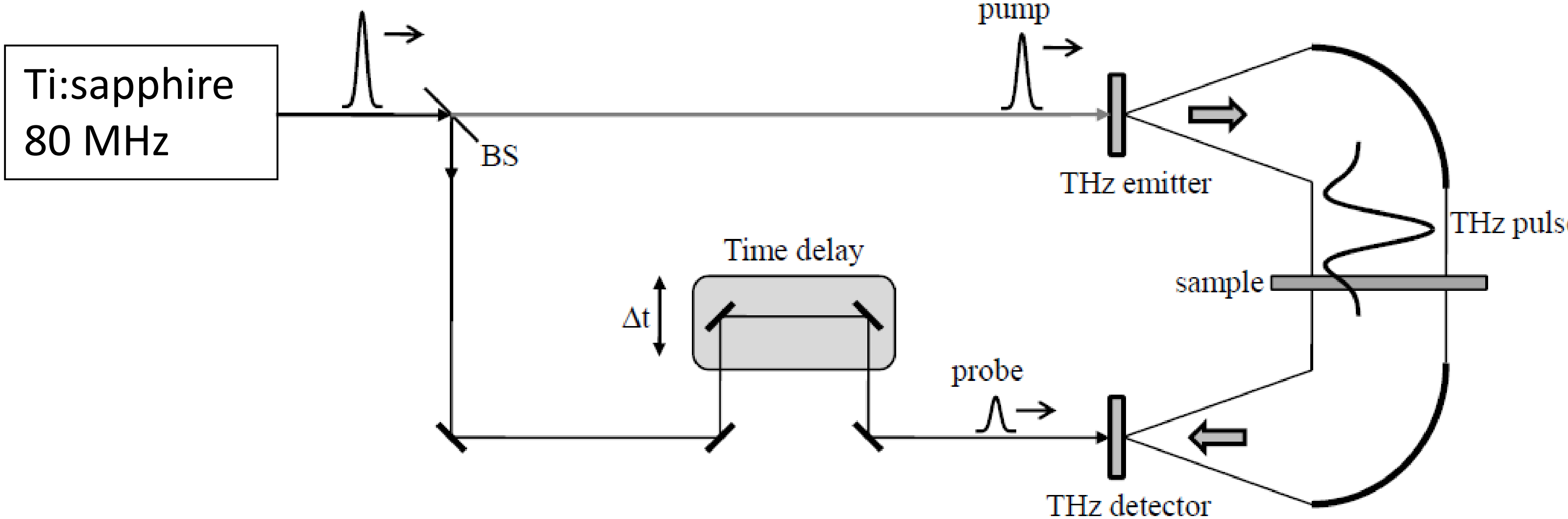
Dipole antenna:




δ-pulse:



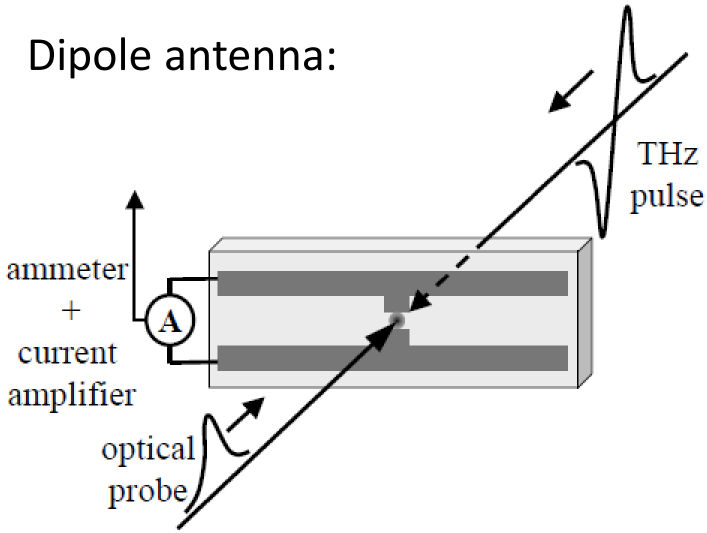

$$j_D(t) = e\exp(-t/\tau_{rec})\int_0^t \exp(-t'/\tau_{scat})\frac{e}{m}E_{THz}(t-t')dt'$$



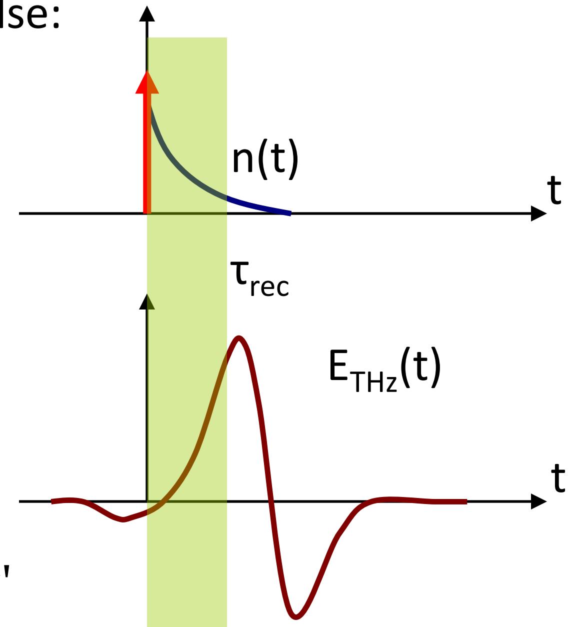

Y. S. Lee, Springer, Berlin (2009)

# THz spectroscopy: time-resolved detection

Dipole antenna:



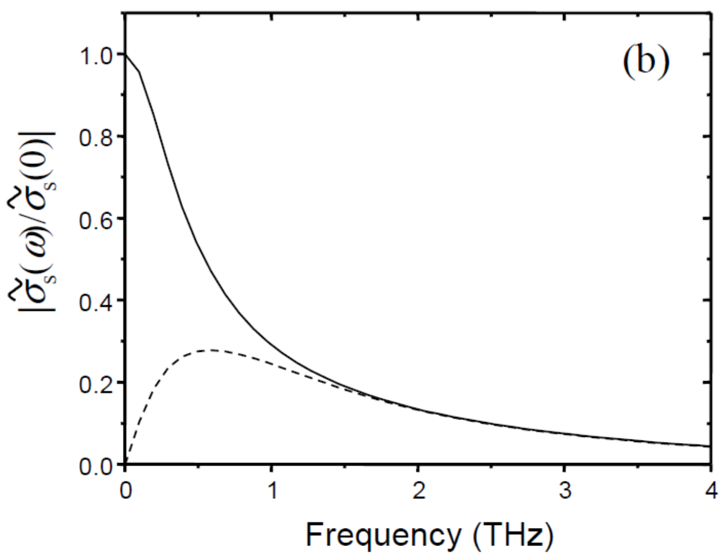

δ-pulse:




$$j_D(t) = e\exp(-t/\tau_{rec})\int_0^t \exp(-t'/\tau_{scat})\frac{e}{m}E_{THz}(t-t')dt'$$

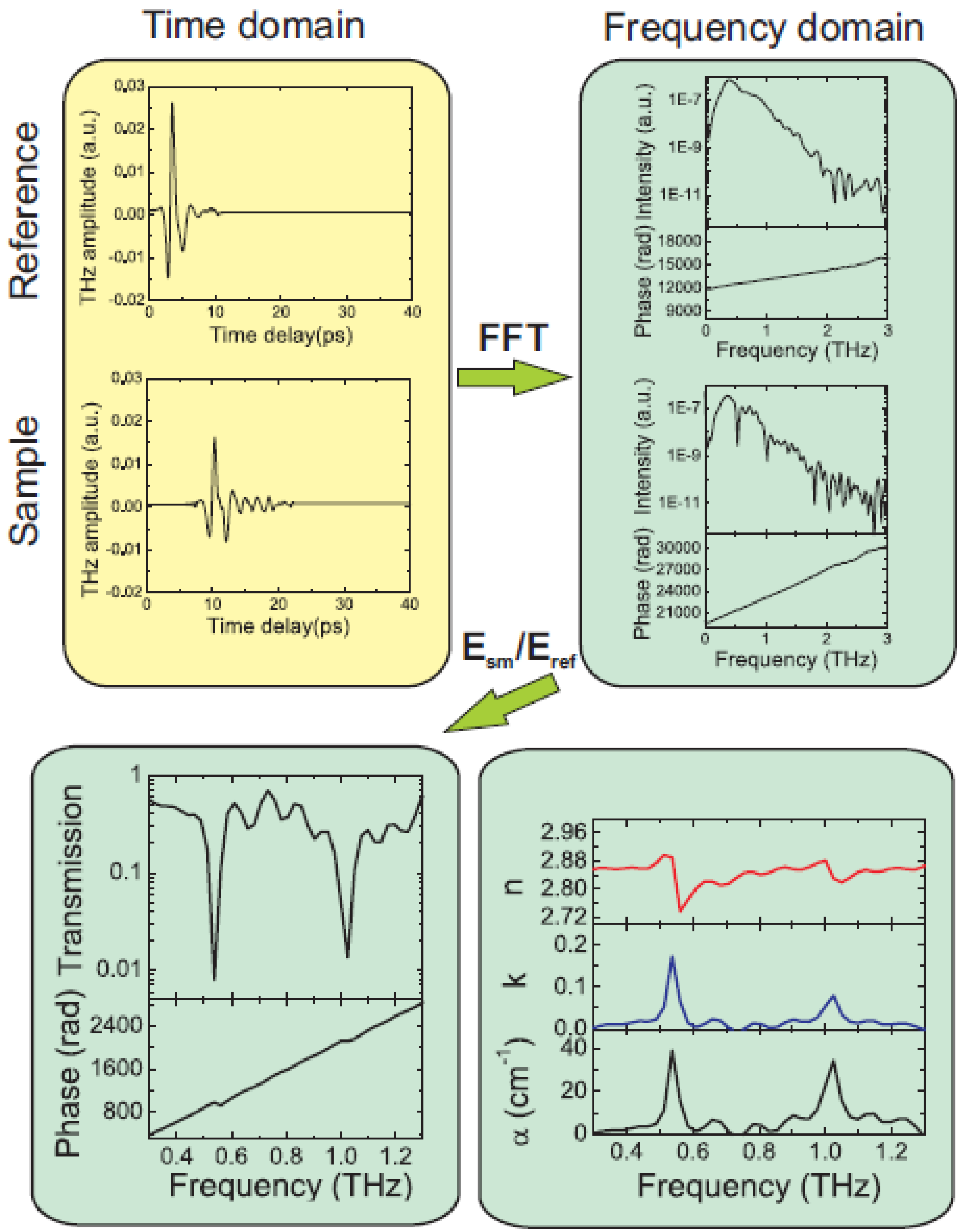
High-frequency cut-off: $1/\tau_{rec}$~3..7 THz

Low-frequency cut-off: diffraction limited




Y. S. Lee, Springer, Berlin (2009)

# THz spectroscopy: full complex THz spectrum

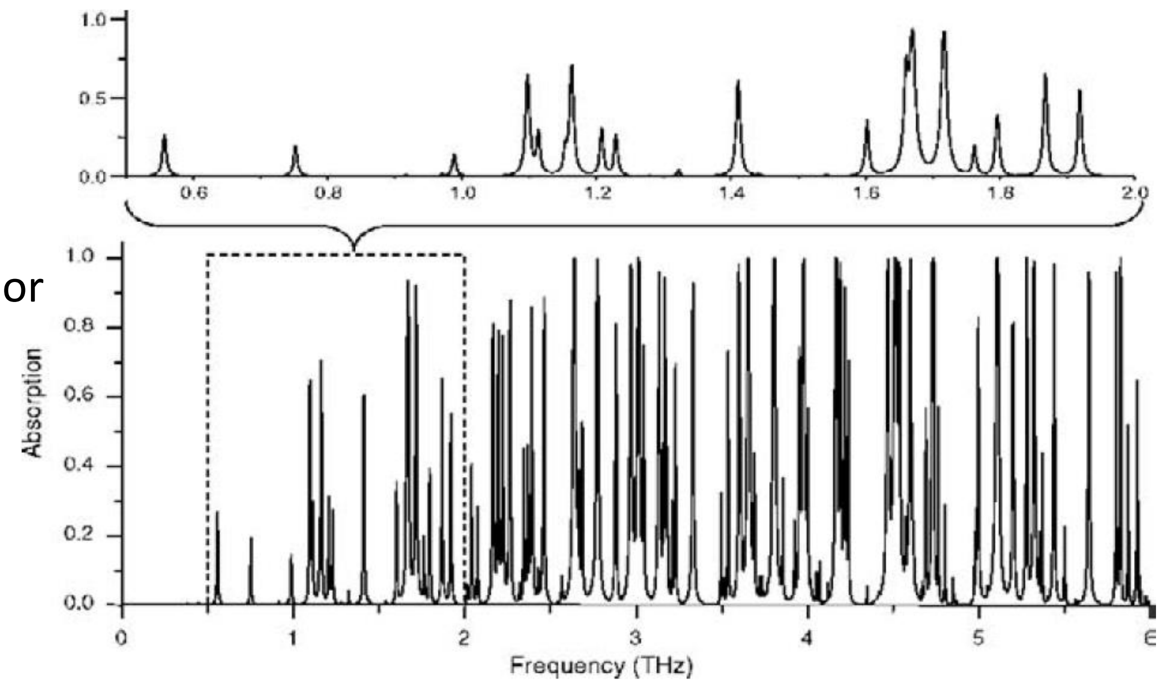


# THz fingerprints of molecules: rotational transitions

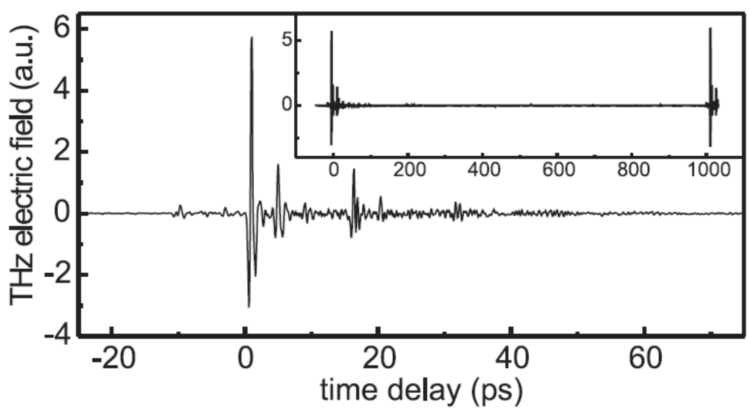
Molecular rotational energy:

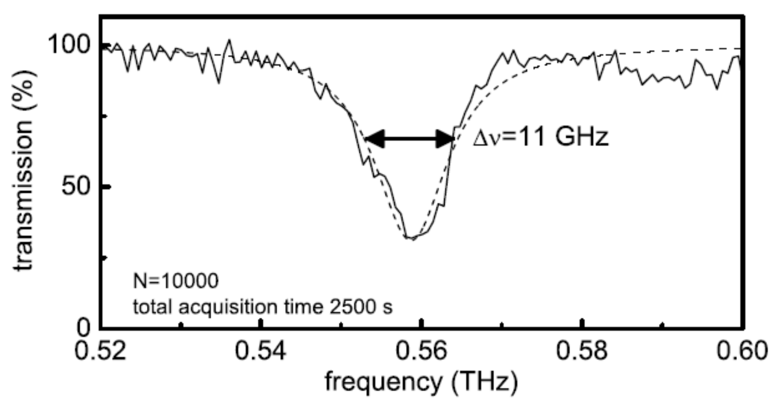
$$E \sim \frac{\hbar^2}{2MR^2} \sim 1THz$$

Rotational spectrum of water vapor




High-resolution (~1GHz)
THz spektroscopy:



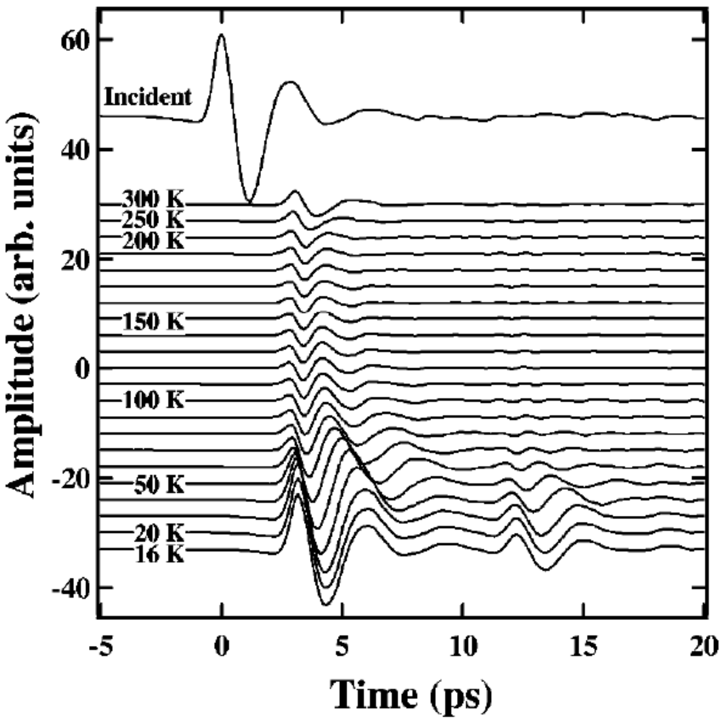



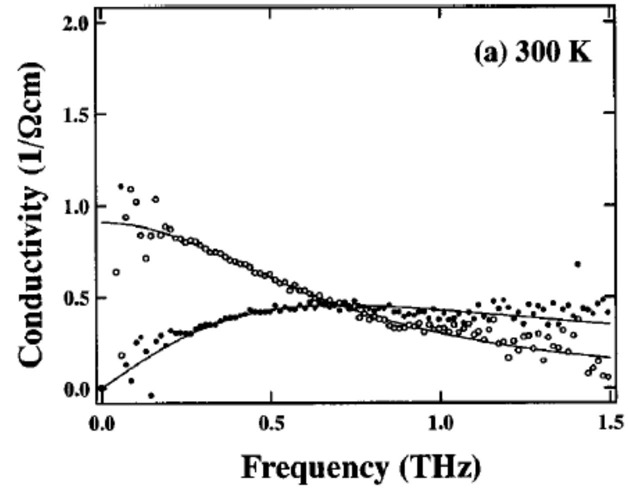


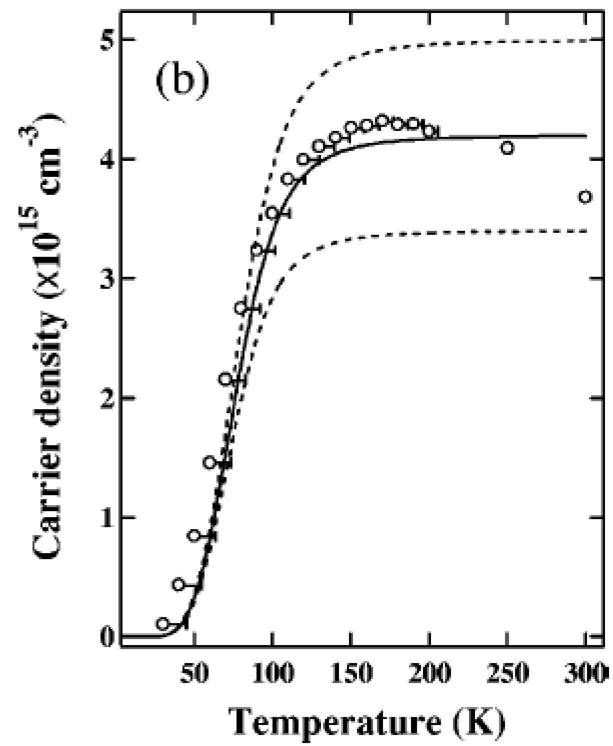
Y. S. Lee: Principles of Terahertz Science and Technology Springer, Berlin (2009)
A. Bartels et al., *Opt Exp* **14** 430 (2006)

# Wireless testing of dopped semiconductors

Dopped p-Si waffer




$N_D$=4.2×$10^{15}$cm$^{-3}$

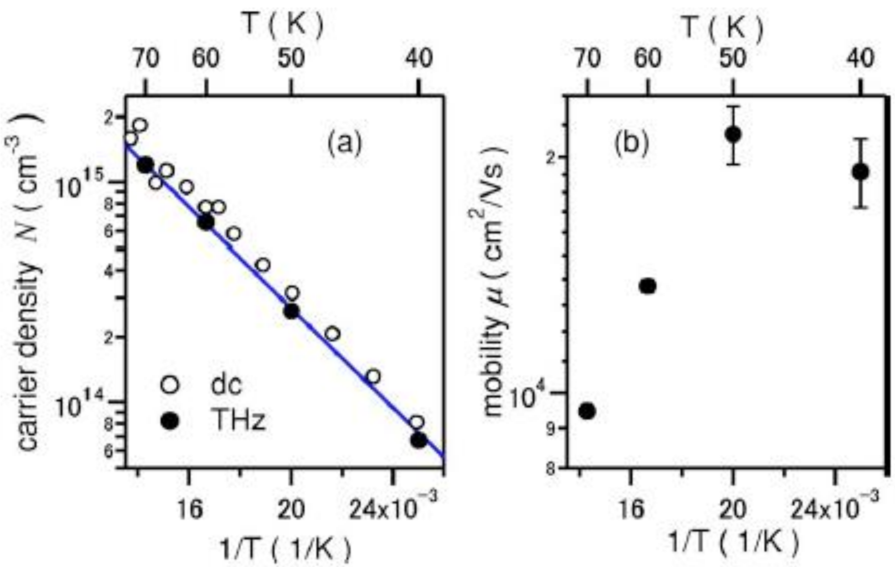
S. Nashima et al. *J. Appl. Phys.* **90** 15 (2001)

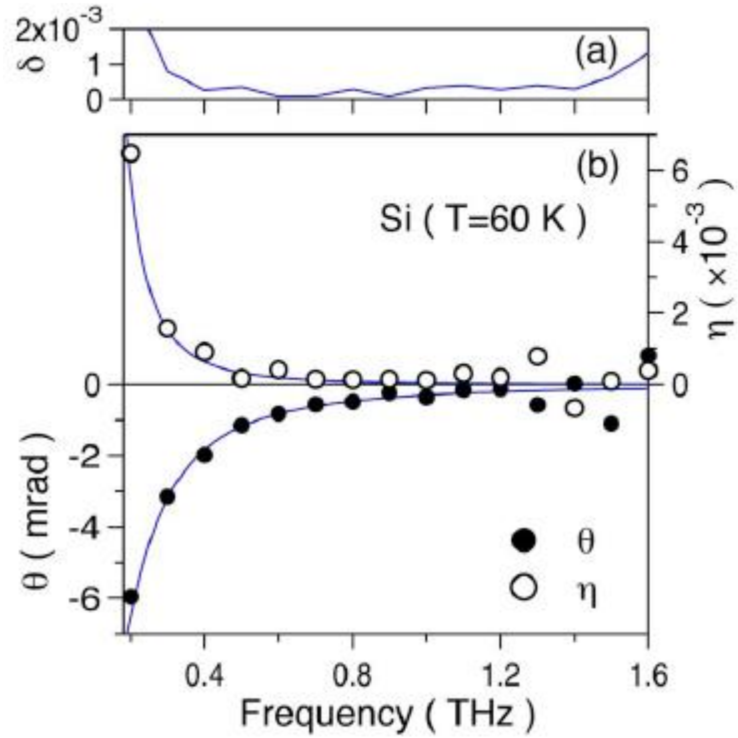
# Wireless testing of dopped semiconductors

Dopped n-Si waffer

$$E_y(\omega,B) \approx E_0(\omega)\exp\left[-i\omega\left(t-\frac{\hat{N}(0)}{c}d\right)\right]\times(\sin\theta + i\eta\cos\theta) = E_x(\omega,0)(\sin\theta + i\eta\cos\theta),$$

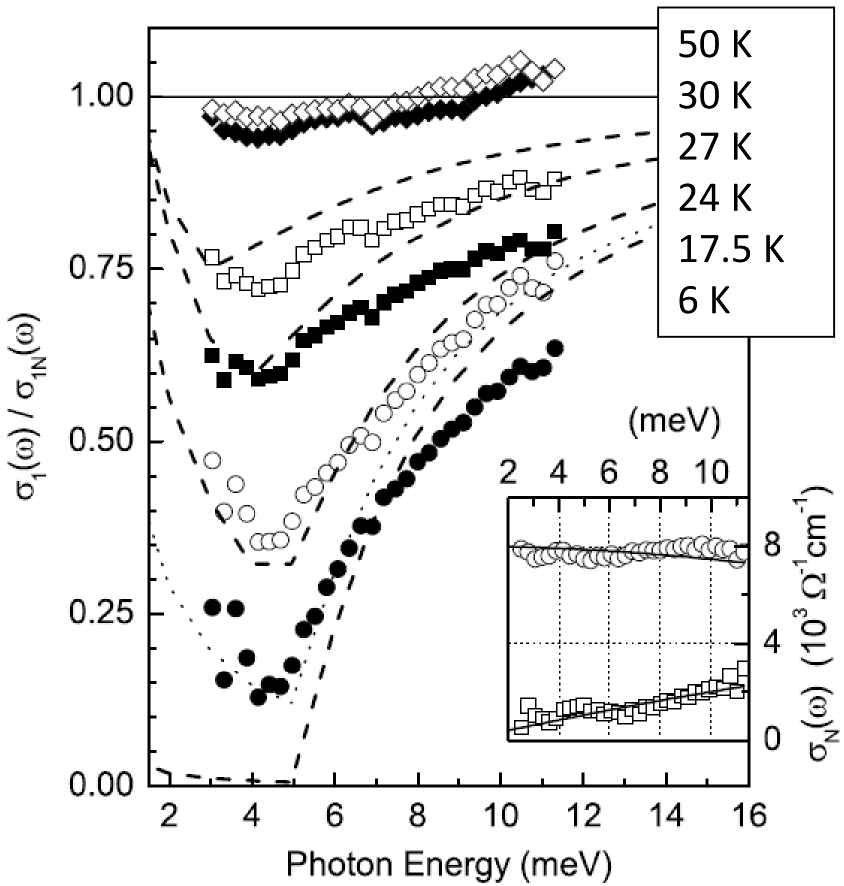
$$\theta + i\eta = -\frac{\omega}{2c}\frac{i\epsilon_{xy}}{\sqrt{\epsilon_{xx}}}d$$

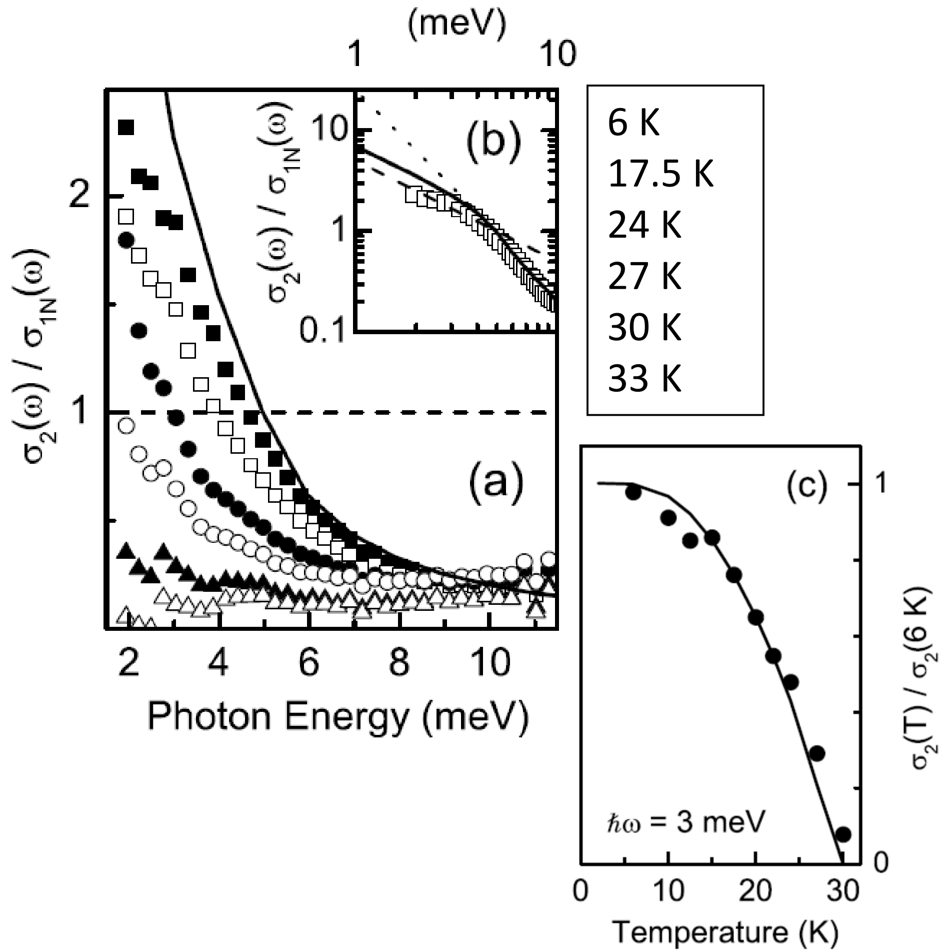



Appl. Phys. Lett. **92, 012111 2008**

# Electrodynamics of superconductors: $MgB_2$




BCS fit: $2\Delta_o$=5 meV

Weak coupling: $2\Delta_o=3.5k_BT_C$=9 meV

Quasiparticle excitations below the gap

R. A. Kaindl et al., *Phys. Rev. Lett.* **88** 027003 (2002)

# Metamaterials in the THz range

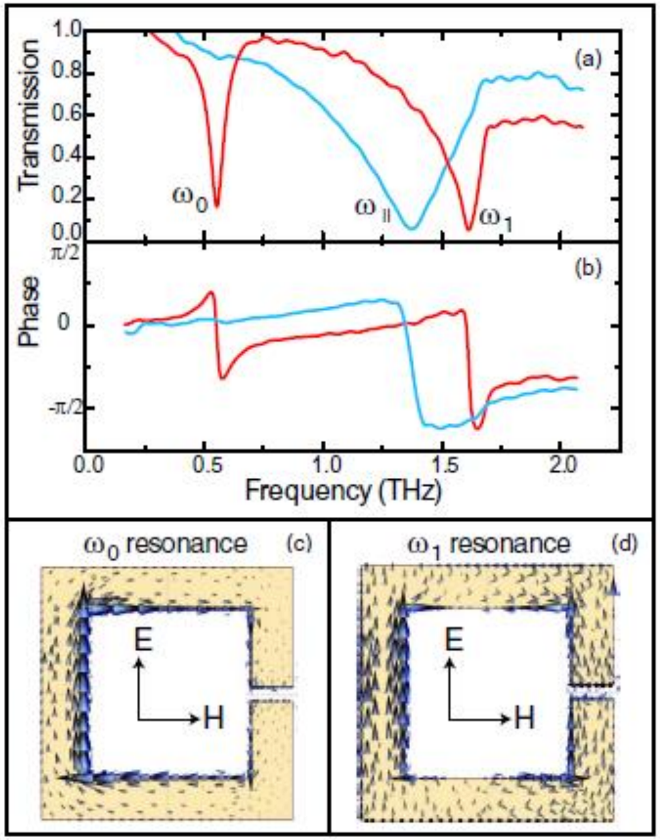
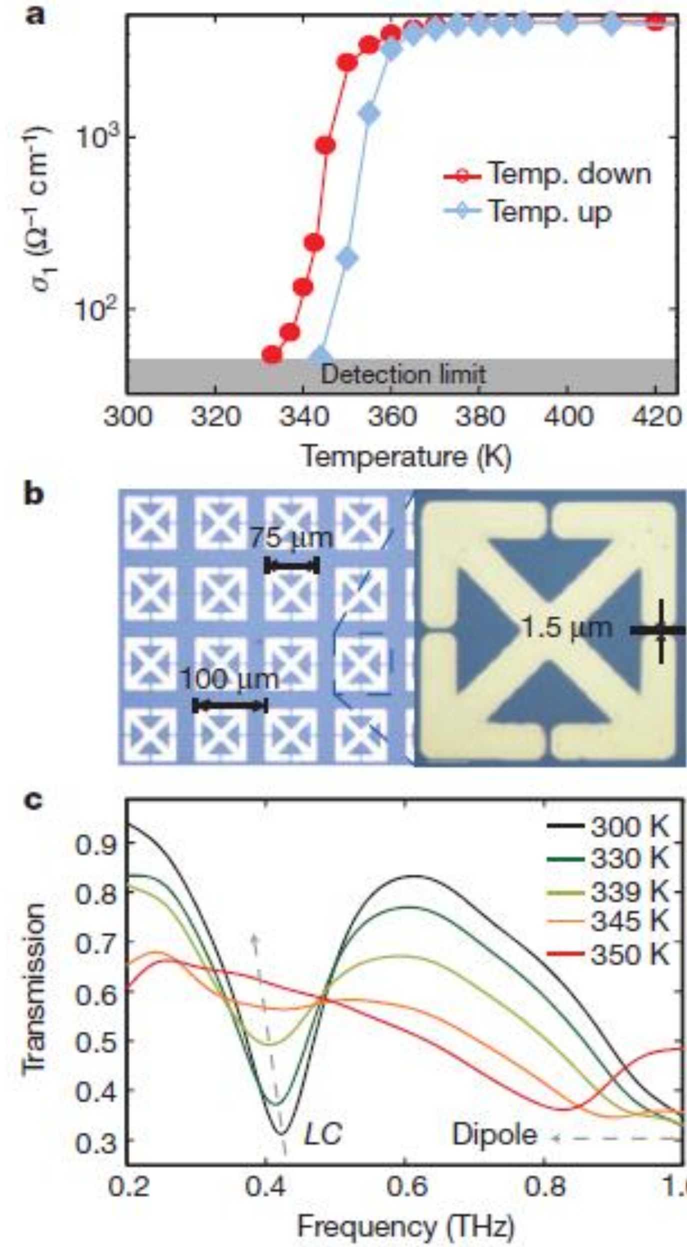



FIG. 1 (color online). The frequency dependent transmission spectra $T(\omega)$ of the SRR sample is shown in (a), and in (b) the corresponding phase of the transmission is shown. In (a) and (b), the electric field is perpendicular to the SRR gap [red (dark) curves] and parallel to the SRR gap [blue (light) curves] at normal incidence. (c) and (d) are the surface current densities for the $\omega_0$ (0.5 THz) and $\omega_1$ (1.6 THz) resonances, respectively, as calculated by simulation. See the text for details.

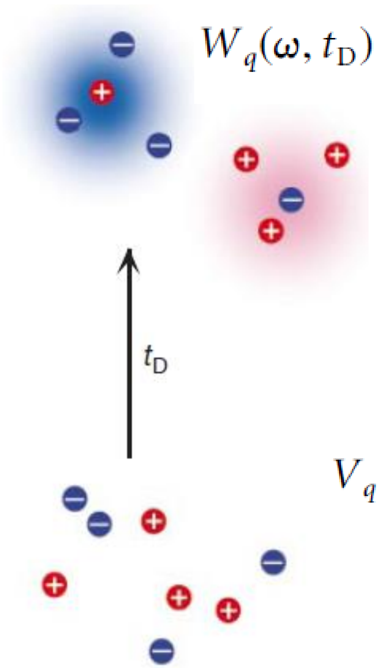
$VO_2$




PRL **96, 107401 (2006)**

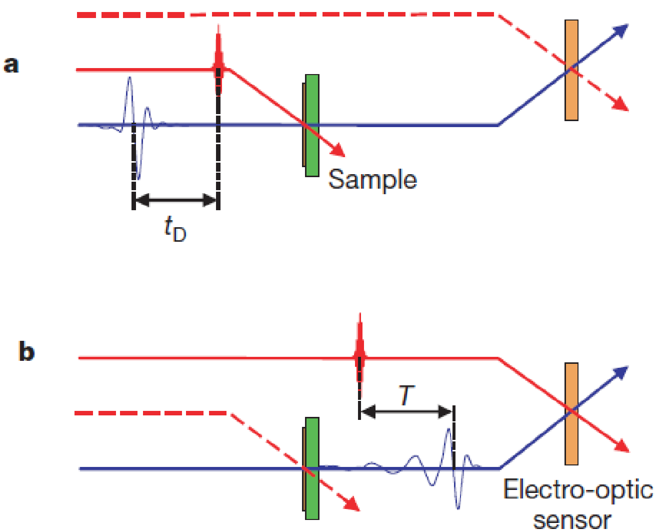
10.1038/nature11231

# Time-resolved study of a Coulomb gas

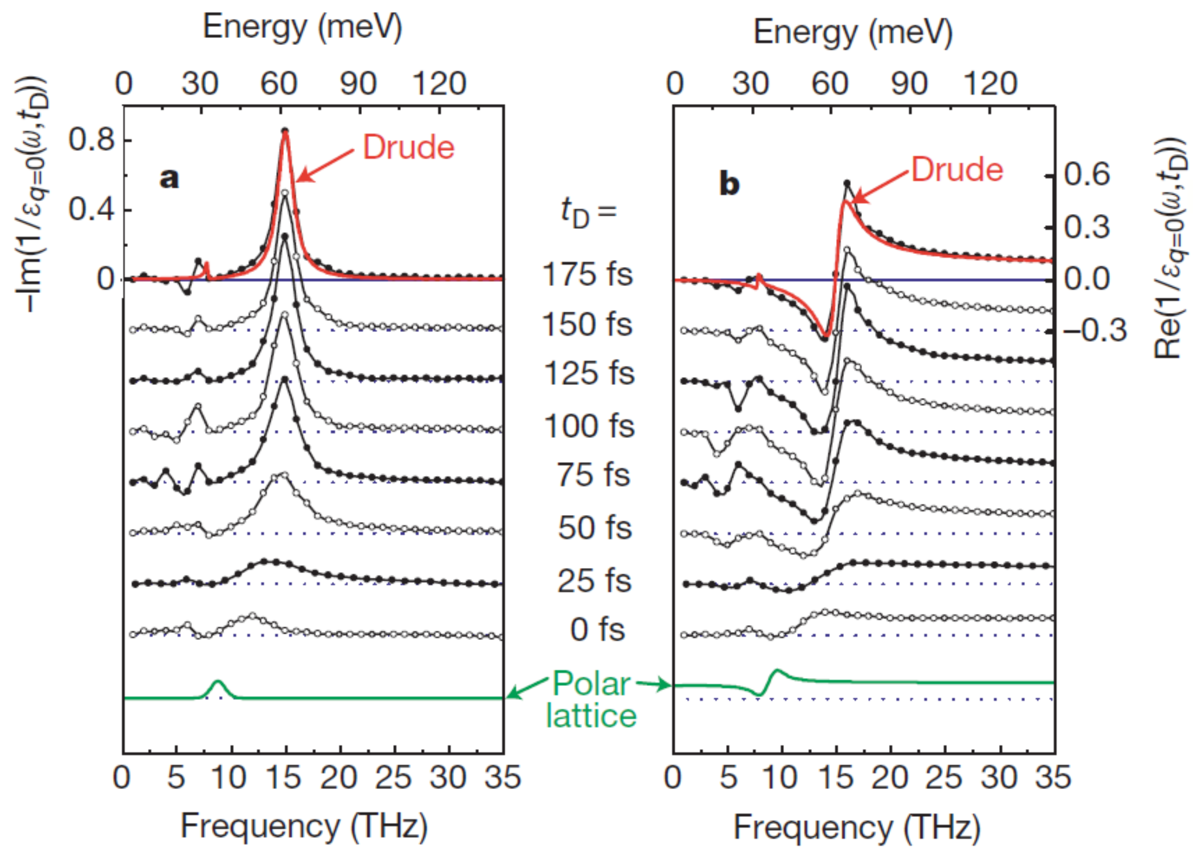



$$W_q(\omega, t_D) = \frac{V_q}{\epsilon_q(\omega, t_D)}$$

$$V_q = \frac{4\pi e^2}{q^2}$$



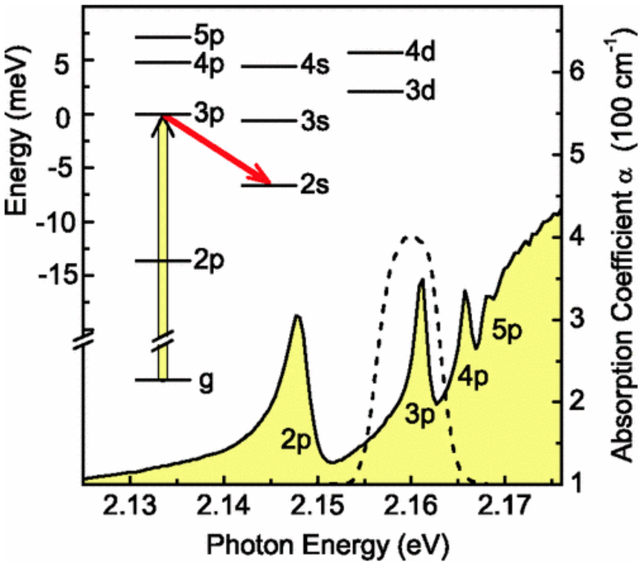

NIR pump – THz probe spectroscopy



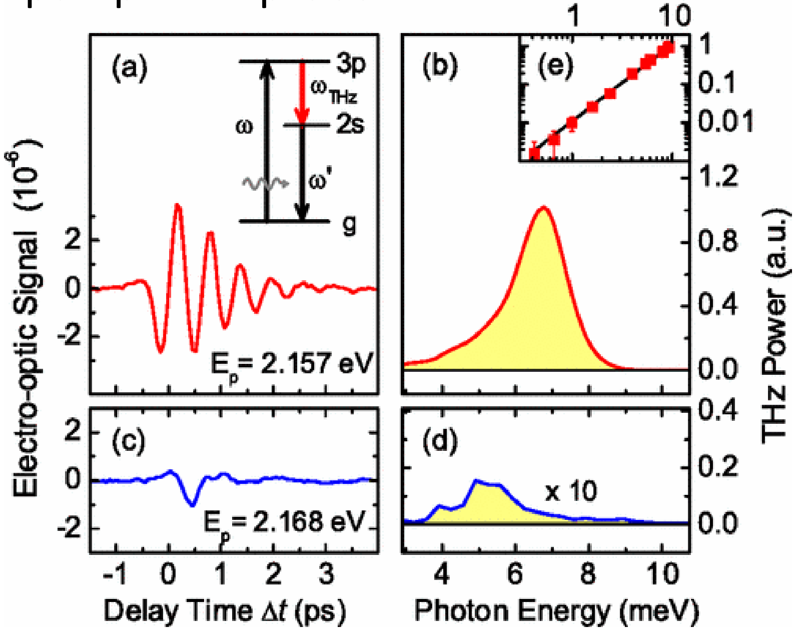

R. Huber et al., *Nature* **414** 286 (2001)

# Stimulated THz emission from excitons

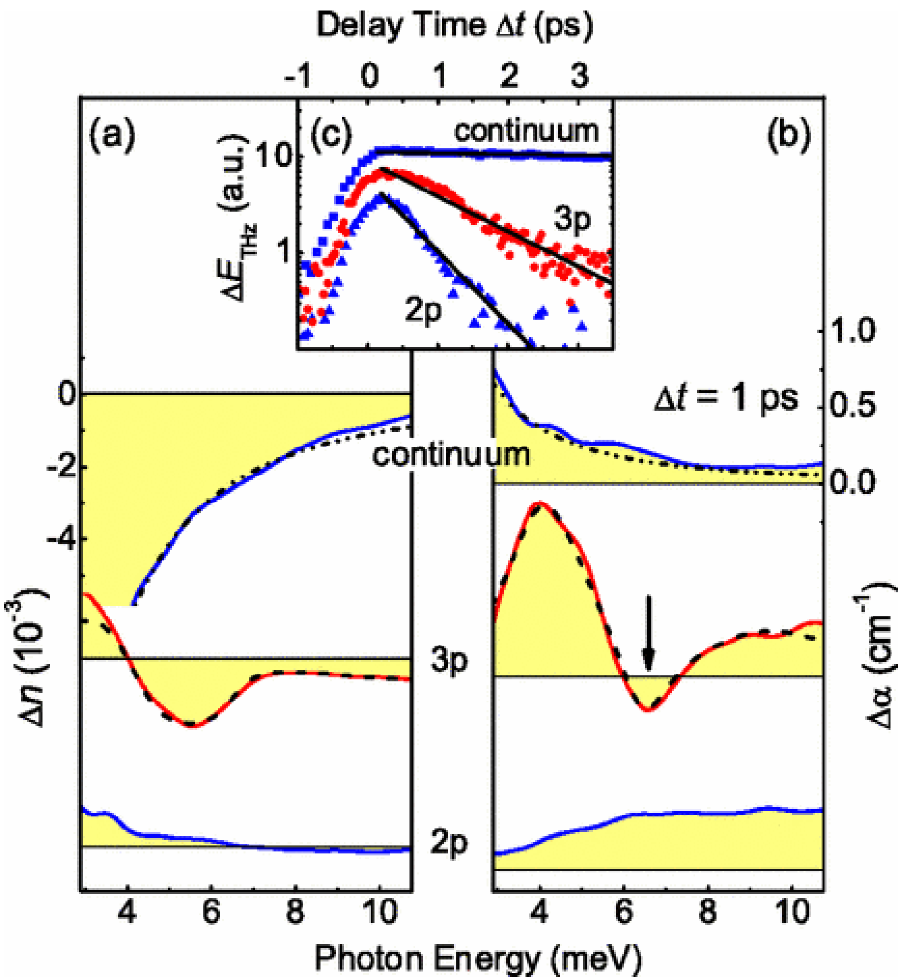
Excitions in $Cu_2O$




NIR pump - THz probe




THz response at Δt=1 ps
Stimulated emission:




Phys. Rev. Lett. **96**, 017402 (2006)