Optical nolinearities
as a tool for nondestructive diagnostics of advanced semiconductor materials

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Outline

1. Modulation of optical and electrical properties by light interference pattern:

- refractive index modulation by FC density and SC field
- absorption index modulation by recharged deep traps

2. Experimental configurations and study:
    - SC field impact on carrier dynamics
    - Determination of photogenerated carrier sign
    - Deep trap recovery rate: transient reflection grating

3. Implementation of TG techniques into devices

Laser-assisted nonlinear optical techniques can serve as a tool for **investigation of fast electronic properties of semiconductor materials by optical means.**

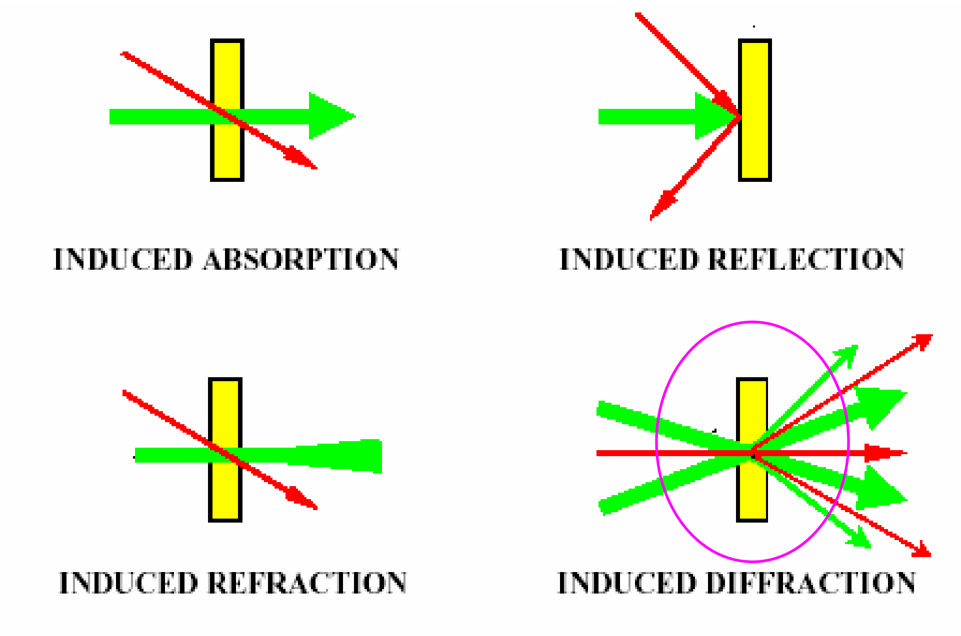
Do we need new optical diagnostics techniques?

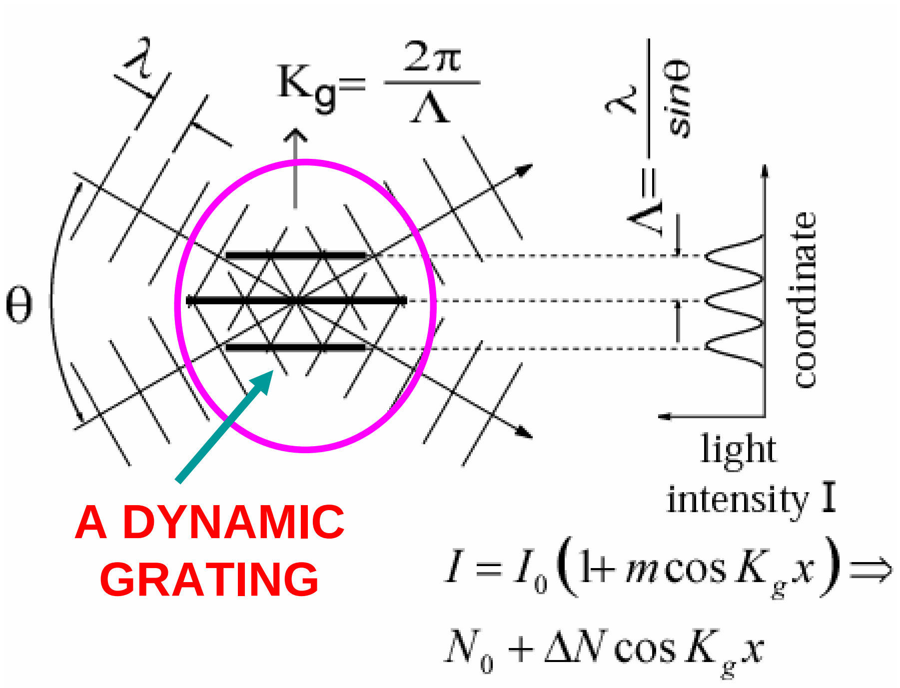
YES, because ---

the standard techniques, as *electrical*, *linear optical* spectroscopy and *photoluminescence* are not always applicable for materials characterization: they need contacts, reveal equilibrium properties *and* cannot be applied if radiative PL signal is very low *(e.g. SiC) !*

In most technologically important materials, **knowledge of carrier dynamics is the key issue.**

“Pump-probe” configurations for time-resolved nonlinear optical experiments





$$I = I_0\left(1 + m\cos K_g x\right) \Rightarrow N_0 + \Delta N \cos K_g x$$

Information about refractive index spatial and temporal modulation *Δn* (x,t) is read by a probe beam diffraction and will provide electronic parameters, as τ_R, D, L_D, s, τ_R (N_{FC}=B, C; N_{TD}=A), D(N), Esc, its origin …

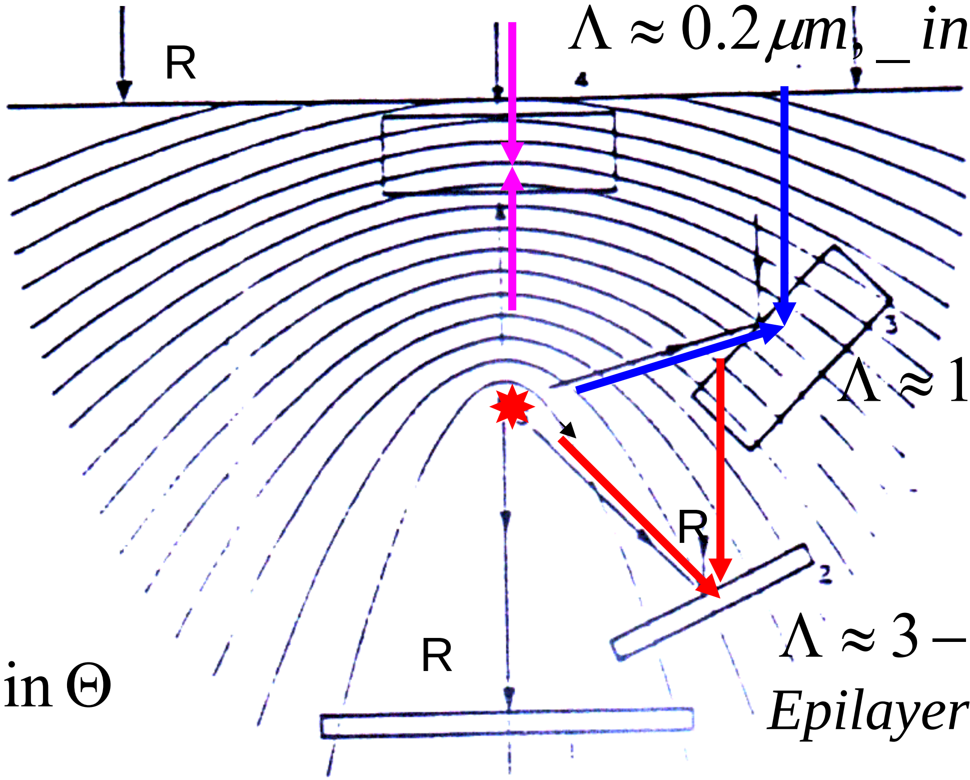
Merging the optical nonlinearities and photoelectric properties

Field	What knowledge it provides
Step 1. Dynamic holography	• Optical configurations to create and monitor the spatial modulation of real and/or imaginary part of the refractive index: $n(x,t) = n_0+\Delta n(t)\cos(Kx)$ $k(x,t) = k_0+\Delta k(t)\cos(Kx)$ • Type of light-induced diffraction grating: a phase, an amplitude, or mixed one • Relationships between the diffraction efficiency $\eta = I_1/I_T$ and modulation amplitude of Δn or Δk, e.g. $\eta \sim (\pi\Delta nd/\lambda)^2 + (\pi\Delta kd/\lambda)^2$
Step 2. Nonlinear optics	• Modulation mechanisms and their coefficients: $\Delta n_{FC} = n_{eh}\, \Delta N_{e,h}$ $\Delta n_{EO} = n_{eo}\, E_{SC}$ $\Delta k = \Delta a\lambda/4\pi$

Merging the optical nonlinearities and photoelectric properties

Step 3. Semiconductor physics	• Solution of a continuity equation provides the spatio-temporal evolution of modulated carrier density ($\Delta N(x, z, t)$), which is governed by carrier generation, transport, and recombination processes: $\frac{\partial N(x,z,t)}{\partial t} = \nabla[D(N)\nabla N(x,z,t)] - AN(x,z,t) - BN^2(x,z,t) - CN^3(x,z,t) + G(x,z,t)$
Step 4. Semiconductor technology	• Provides variety of modern materials – epilayers, heterostructures, bulk crystals - differently grown, doped, and processed (annealed, irradiated, etc.) for evaluation of metrological capability of nonlinear optical techniques • As a feedback, the transient grating technique can be exploited for ex-situ nondestructive testing

Optical geometries for a hologram recording



$\Lambda \approx 0.2\mu m, \_in'bulk, MQWs$

$\Lambda \approx 1-5\mu m, \_bulk$

$\Lambda \approx 3-30\mu m$

Epilayers, bulk, QWs

$\Lambda \approx \lambda / \sin\Theta$

Plane reference wave R interacts with the scattered one by an object S:

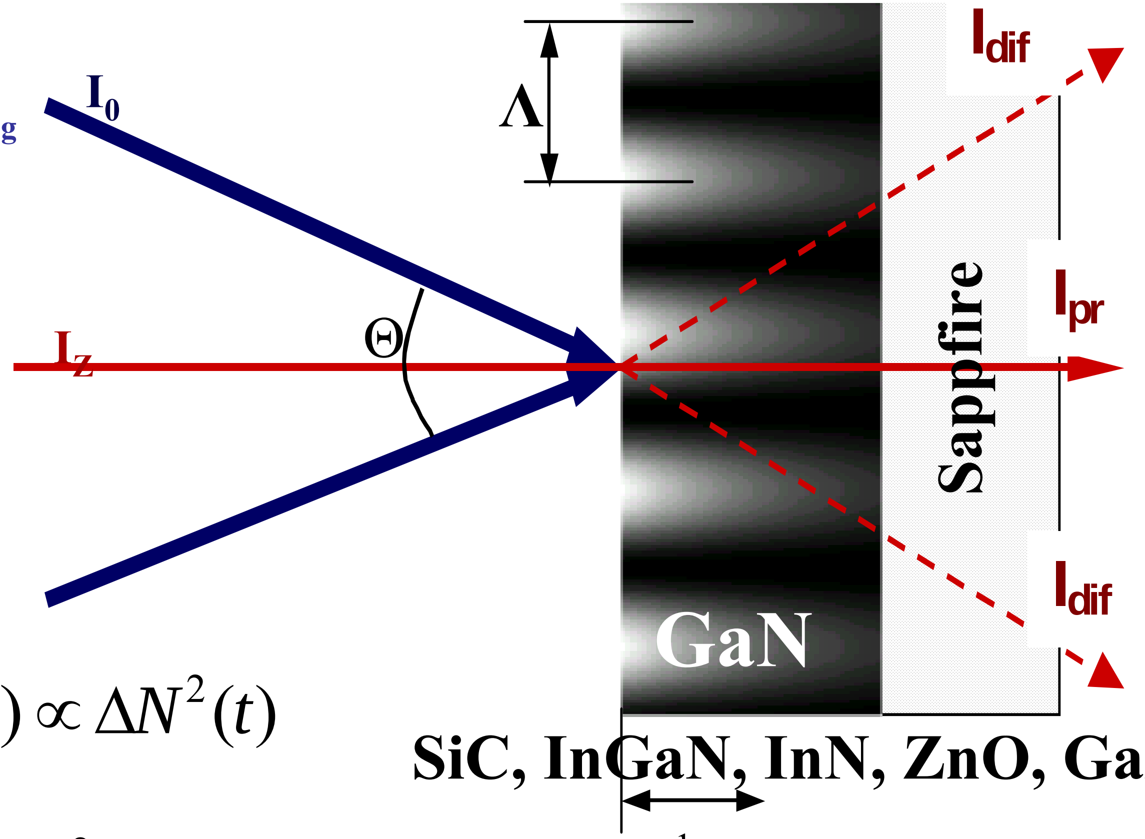
1 – axial geometry (D. Gabor,1951); **2 - Leith - Upatnieks** (1962), d<Λ, thin grating ; **3 – thick grating**, d>Λ, Bragg difraction; **4 – reflection** Bragg-**grating** (d>>Λ= λ/2n); here d is the thickness of a recording material

Thin grating at surface: recording and probing

Coherent ps light pulses from a YAG:Nd laser

Pump at hν>E_g
λ= 353 nm

Probe at
λ = 1064 nm



SiC, InGaN, InN, ZnO, GaAs,

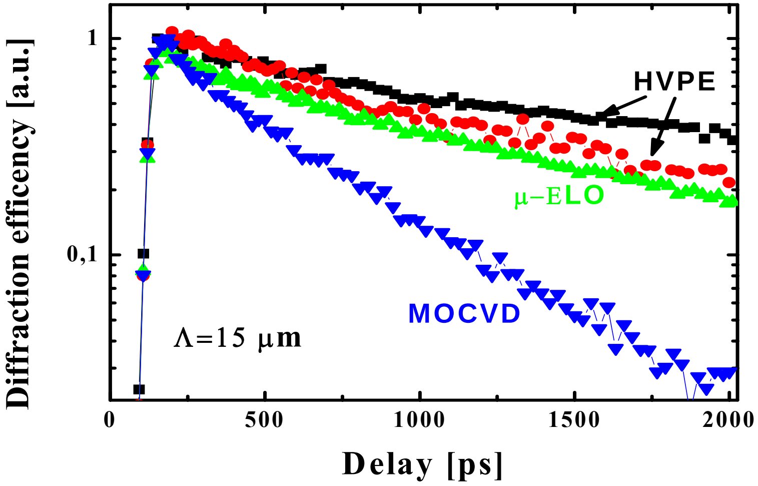
$$\alpha^{-1}=0.1\ \mu\text{m}$$

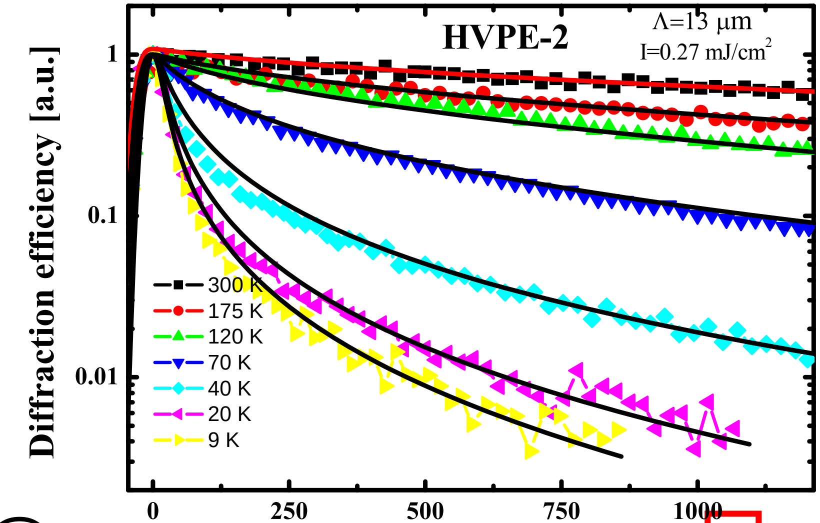
$$\Lambda \approx \lambda / \sin\Theta$$

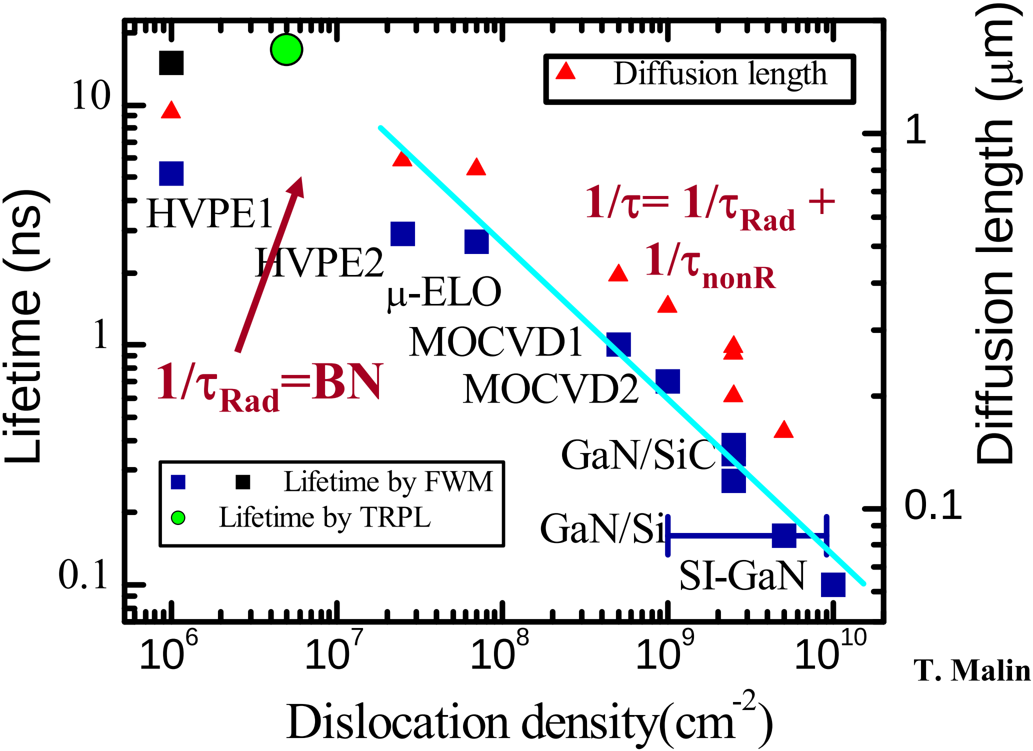
$$\eta(t)=\frac{I_{dif}(t)}{I_{pr}(t)}\propto \Delta n^2(t)\propto \Delta N^2(t)$$

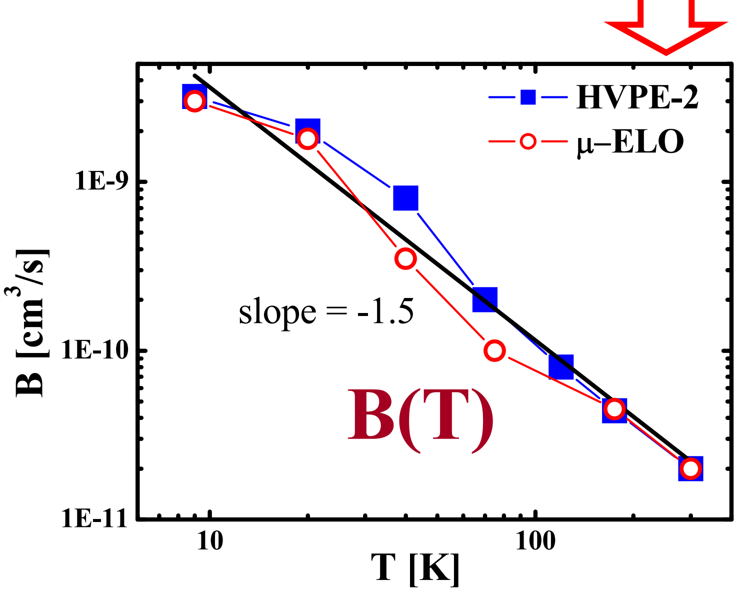
$$\eta(t)=f\left(s,D,\tau_R\right)\propto \mathrm{e}^{-\frac{2t}{\tau_G}}$$

Carier lifetime vs TD density and vs T in GaN



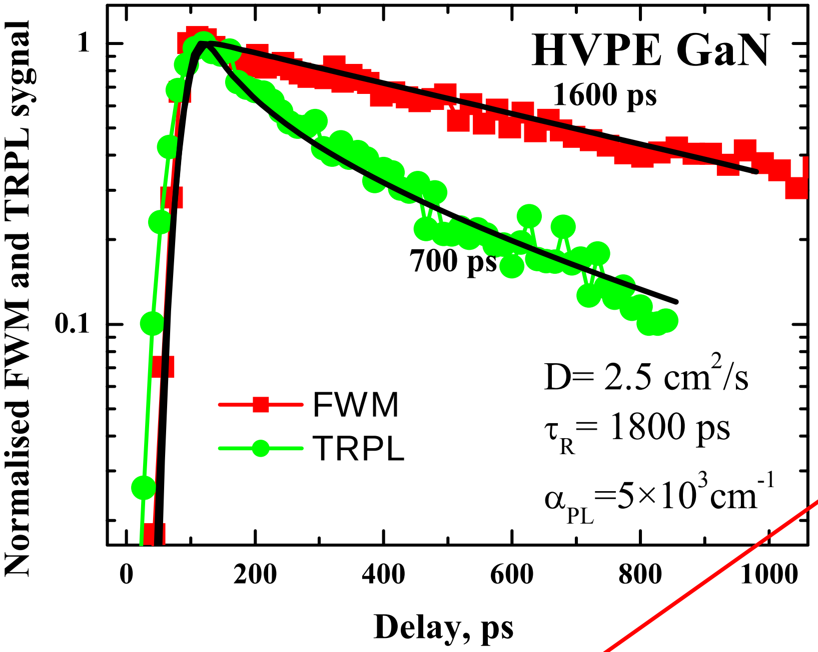


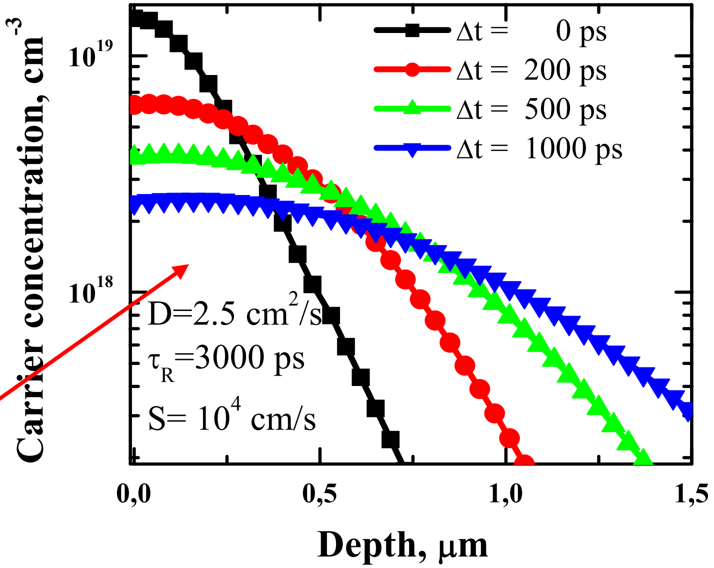




T. Malinauskas et al, phys. stat. sol. (b) 243, 1426-1430 (2006); J. Cryst. Growth 300 (2007) 223.

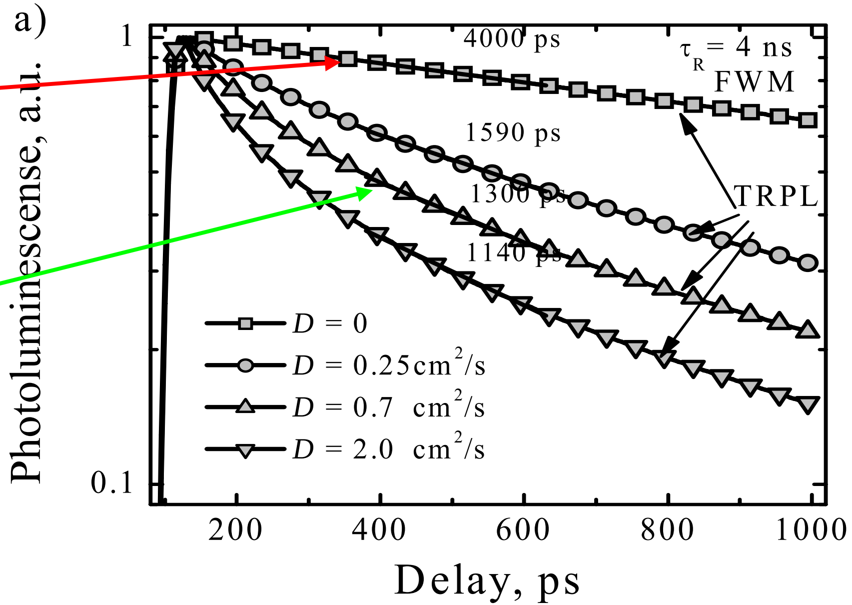
Carrier dynamics seen by TG and PL techniques in GaN





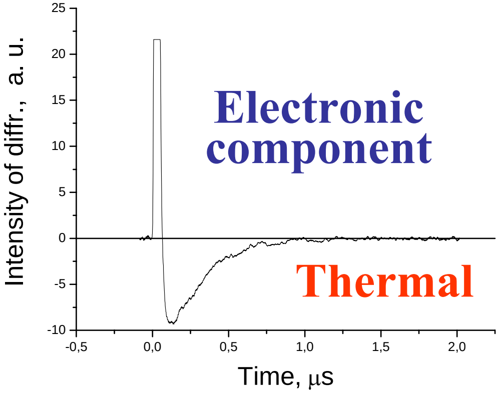
$$\eta_{FWM}(t) \propto \left[\int_0^d \left(\Delta N(z,t)dz\right)\right]^2$$

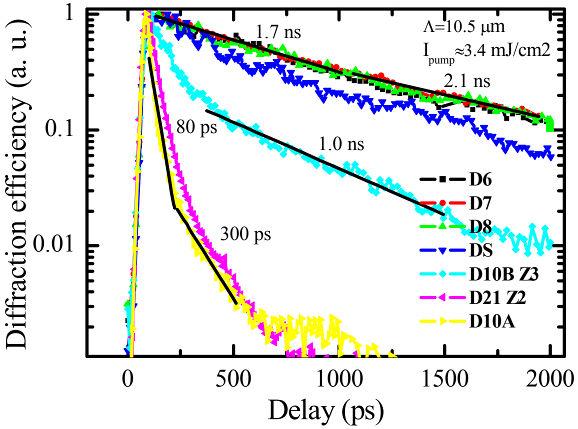
$$\eta_{PL}(t) \propto \int_0^d \left(\Delta N^2(z,t)\exp(-\alpha_{PL} z)dz\right)$$



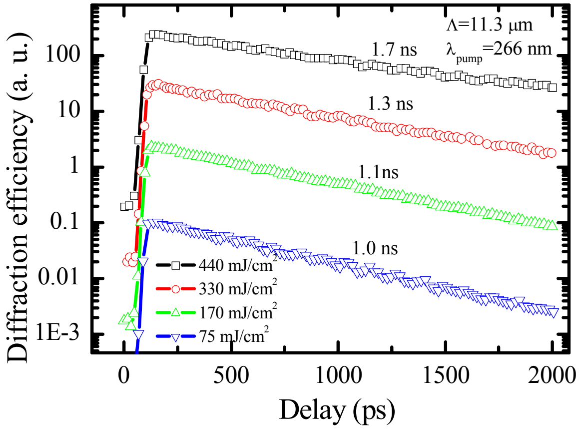
T. Malinauskas et al, APL 88 202109 (2006)

Synthetic diamonds: HPHT and CVD

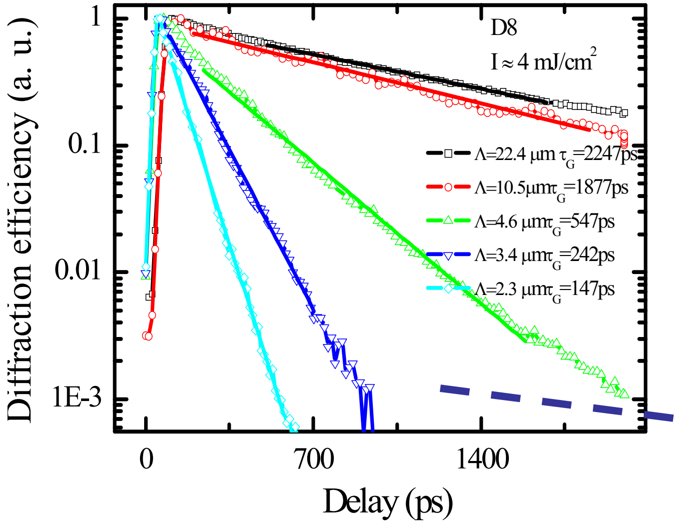




Ps-pump at 213 nm: grating in 10 μm thick layer

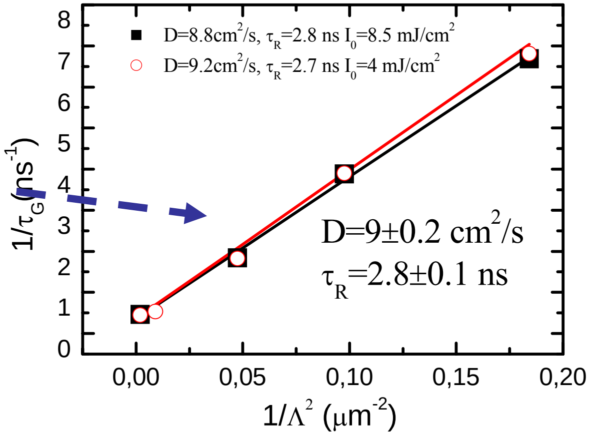


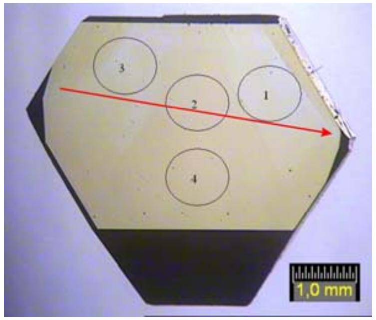
Pump at 266 nm: grating in the bulk



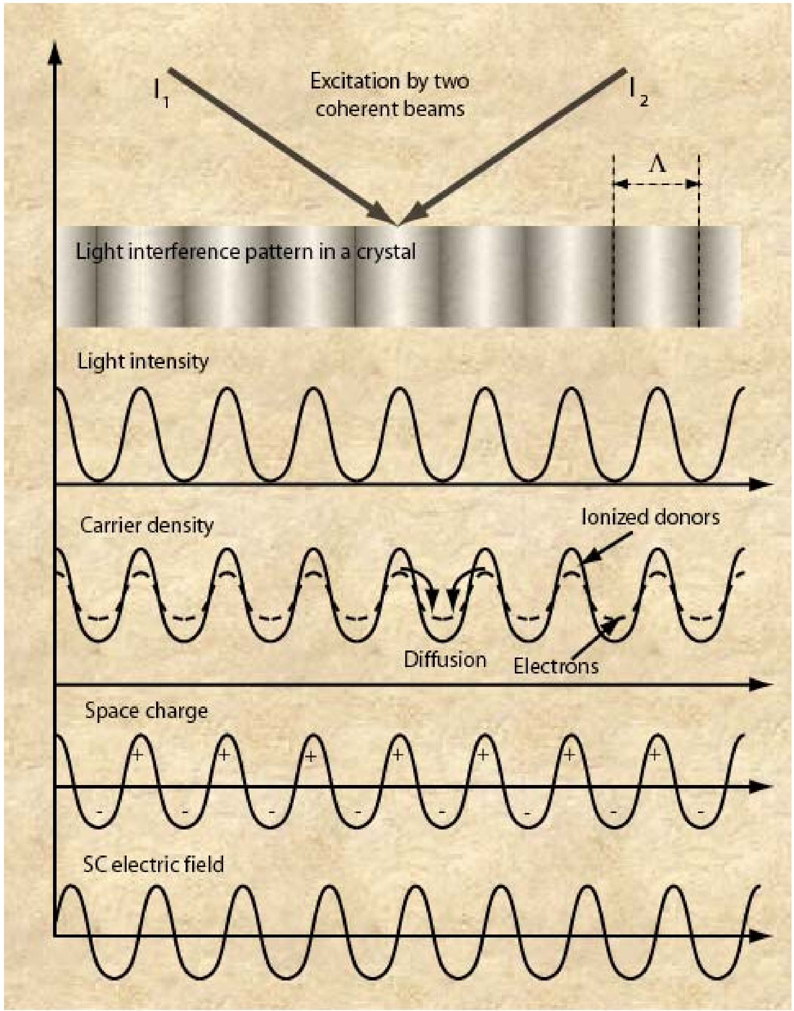
Parameters for the best IIa type HPHT diamond

Decay time of electronic grating correlates well with the nitrogen concentration in range 10^{17} -10^{19}





Gratings in the BULK crystals: deep trap photoexcitation



•Crystal excitation by a short pulse of light interference pattern for modulation of electrical and optical properties

• Carrier generation from deep impurity levels

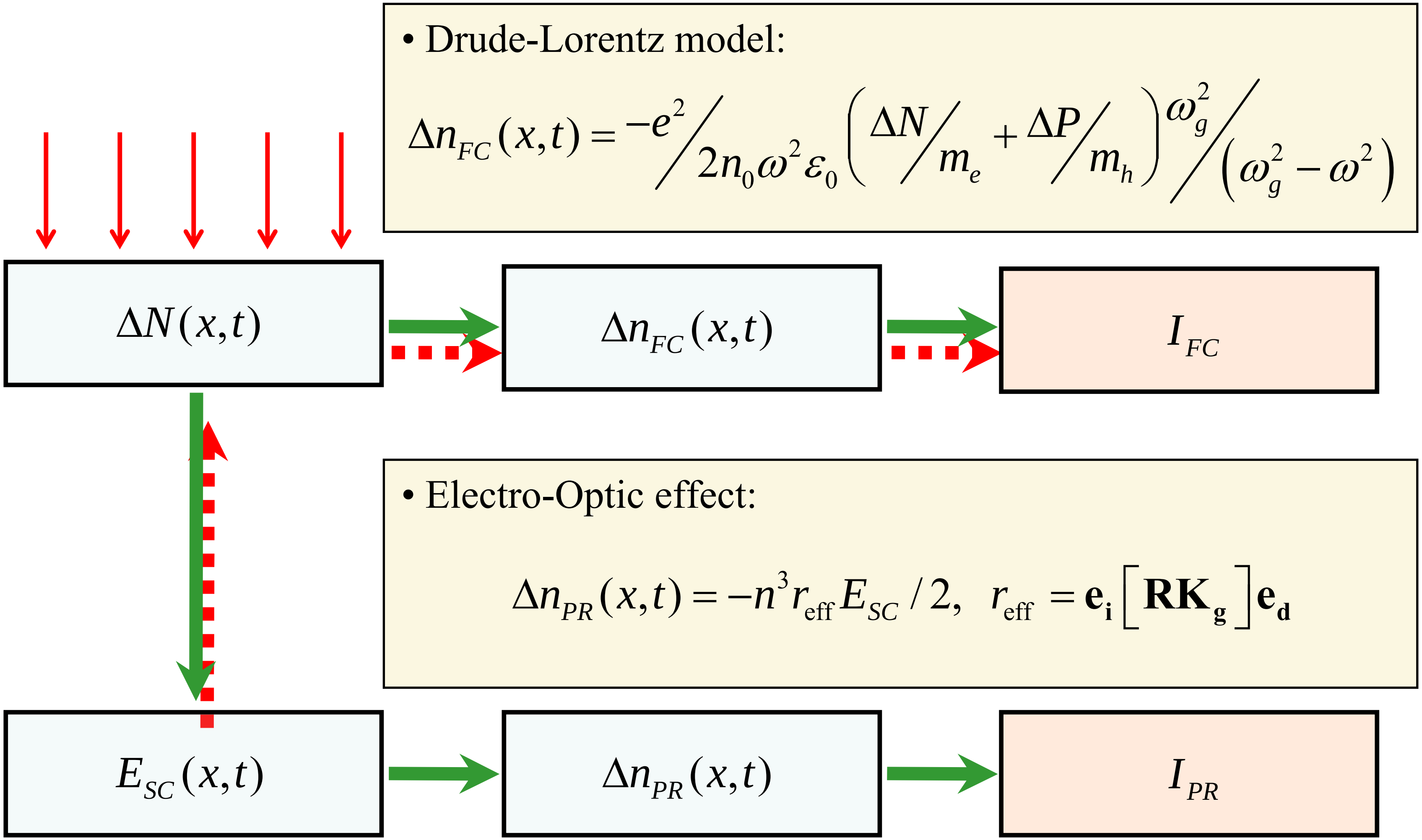
• Charge separation by diffusion of carriers toward the grating minima

• Build-up of the space-charge electric field and its impact to carrier transport

•Modulation of refractive index by FC, internal electric field

•Modulation of absorption index due to changes in deep levels occupation

Refractive index modulation mechanisms



- Drude-Lorentz model:

$$\Delta n_{FC}(x,t) = {-e^2}\Big/{2n_0\omega^2\varepsilon_0}\left(\Delta N\Big/m_e + \Delta P\Big/m_h\right)\omega_g^2\Big/\left(\omega_g^2-\omega^2\right)$$

$\Delta N(x,t)$

$\Delta n_{FC}(x,t)$

I_{FC}

- Electro-Optic effect:

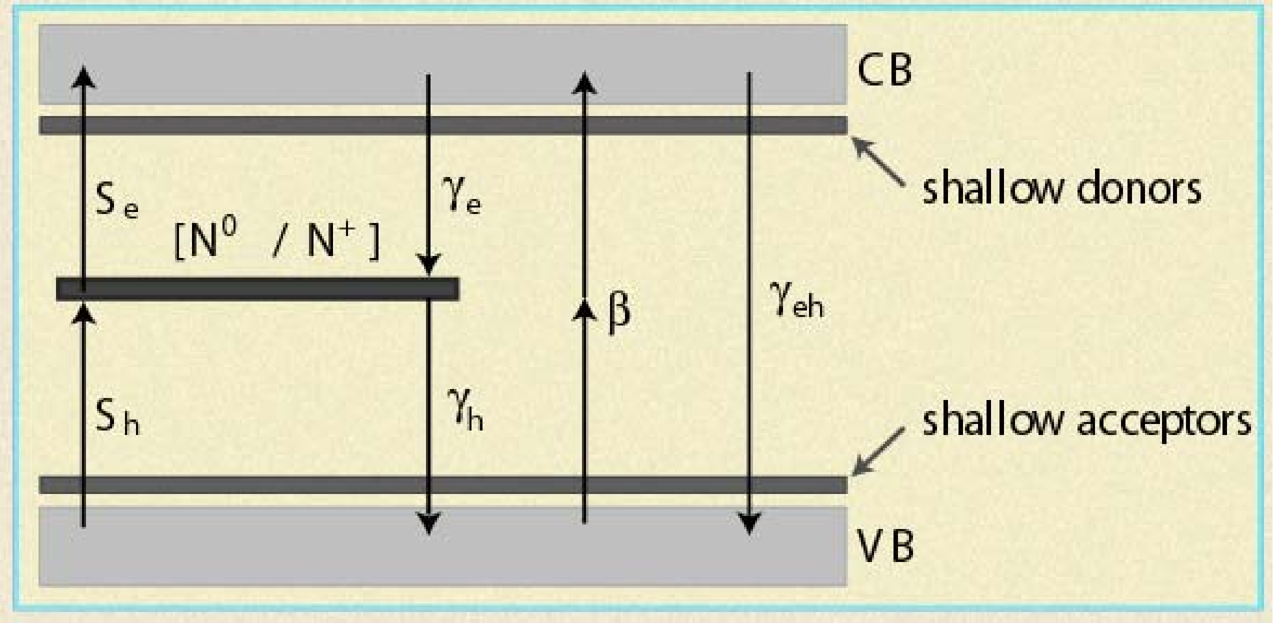
$$\Delta n_{PR}(x,t) = -n^3 r_{\text{eff}} E_{SC}/2,\quad r_{\text{eff}} = \mathbf{e_i}\left[\mathbf{RK_g}\right]\mathbf{e_d}$$

$E_{SC}(x,t)$

$\Delta n_{PR}(x,t)$

I_{PR}

Deep mid-gap donor recharge: <u>absorption index</u> modulation



Spatial photoexcitation of deep donor leads to **changes in the charge state of a deep trap**: $N_0 \Rightarrow N^+$ and $N^+ \Rightarrow N_0$.

This leads to the spatial modulation of recharged deep traps density:

$N_1(x) = \Delta N_0(x) - \Delta N^+(x)$ and to an **amplitude grating formation**:

$\mathbf{\Delta\alpha\ (x)} = S_e \Delta N_0(x) - S_h \Delta N^+(x) = 4\pi\Delta k(x)/\lambda$

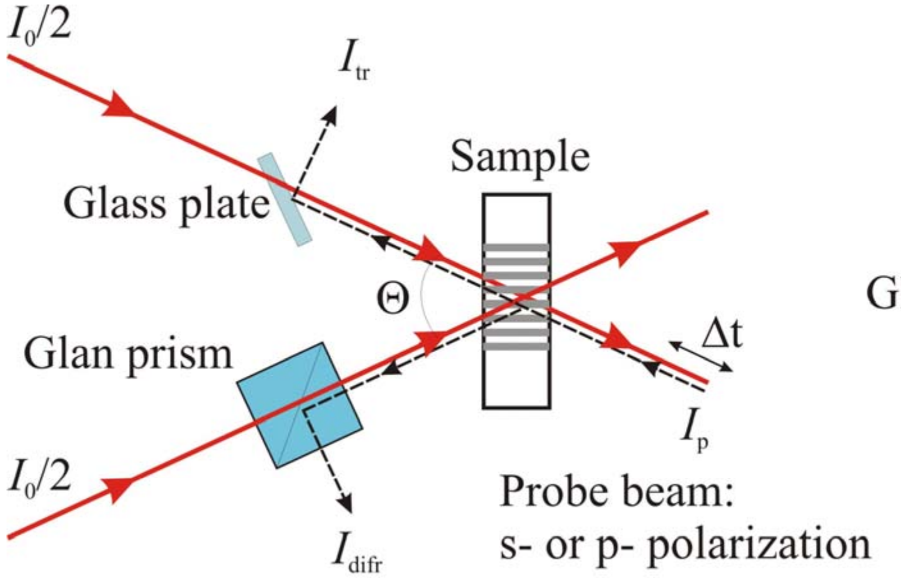
Usually **Δk < Δn,** thus the phase grating dominates- but not always!

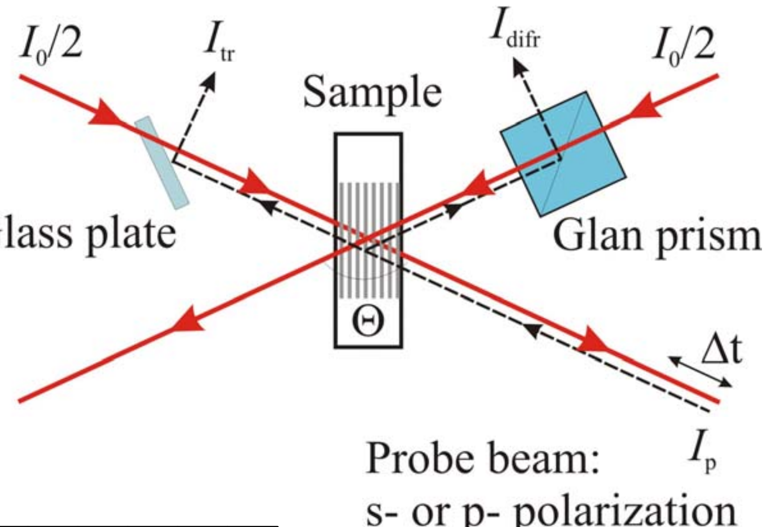
Experimental configurations: transient Bragg-gratings

Diffraction in transmission: $\Delta n(t)$

Diffraction in reflection: $\Delta\alpha(t)$

grating period is $\lambda/2n = 0.16$ μm

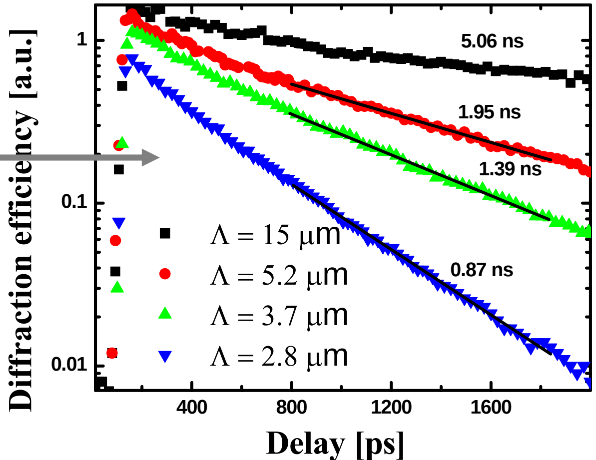




$$\eta(t) \propto [\Delta n_{FC}(t)]^2$$

$$\eta(t) \propto [\Delta n_{EO}(t)]^2$$

$$\eta(t) \propto \exp(-2t/\tau_e)$$



$$\eta(t) \propto \Delta a(t)^2$$

$$\frac{1}{\tau_e} = \frac{1}{\tau_R} + \frac{1}{\tau_D} \Rightarrow \tau_R, D;$$

Samples (*for defect engineering*)

- **Bulk semi-insulating II-VI: CdTe, CdZnTe, doped by V, Ge, Sn, Si, Bi, or ZnTe:V, codoped by Al, Sc**

- **Bulk semi-insulating III-V: InP (undoped and Fe doped), GaAs (EL2)**

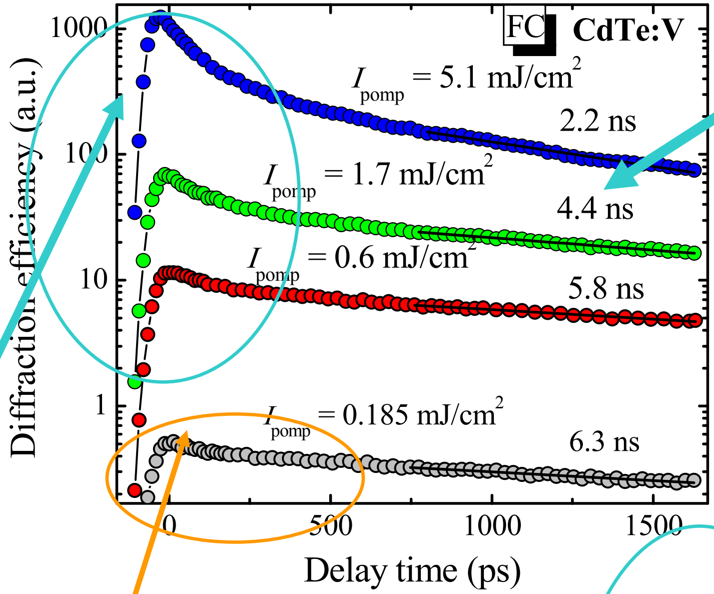
- **Goal – selection of a dopant able to create <u>only one</u> dominant deep trap level for compensation of free carriers.Then a proper density and charge state of deep imurity level will lead to efficient monopolar carrier generation and strong SC field formation**

Carrier and SC field dynamics in CdTe:V

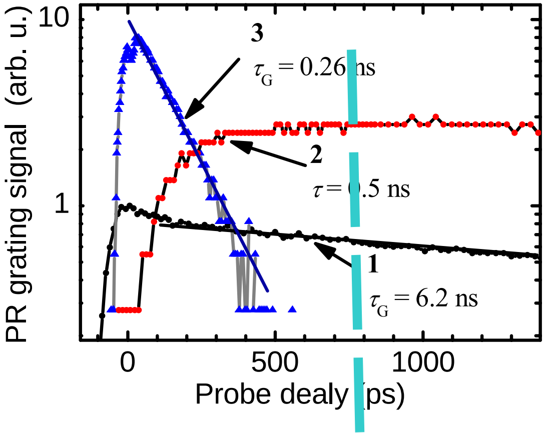
$\Lambda = 2\ \mu m$

E_c- 0.75 eV

FC grating



Recombination-governed decay



Diffusion-governed decay:

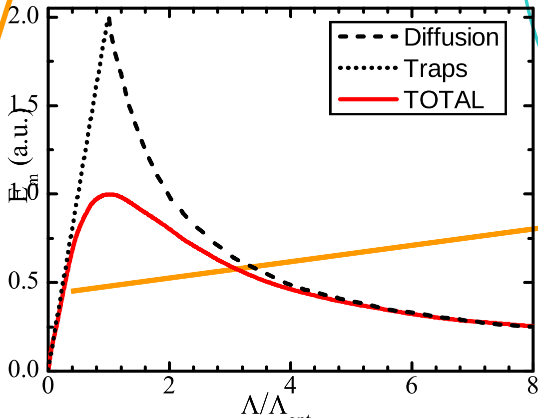
$$\tau_D = \Lambda^2/4\pi^2 D = = K^2 D$$

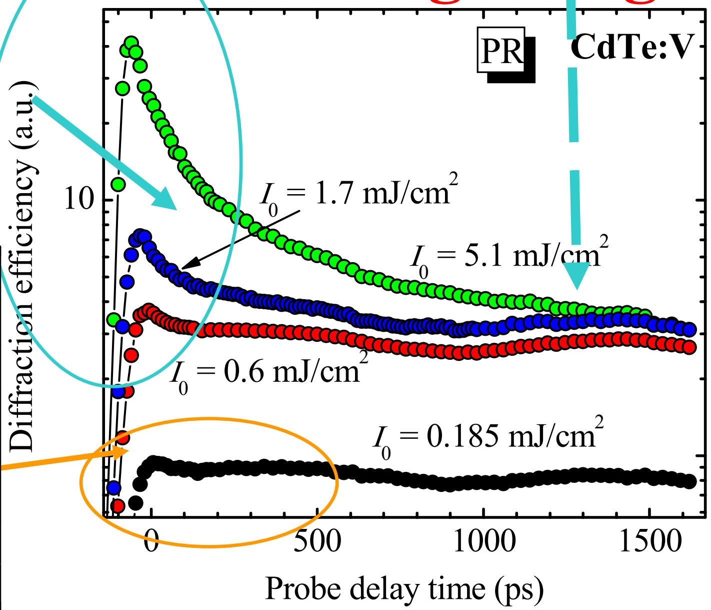
$$\tau_{Dr} = K/(\mu E_{sc})$$

$$E_{sc} = 2\pi kT/e\Lambda$$

PR grating

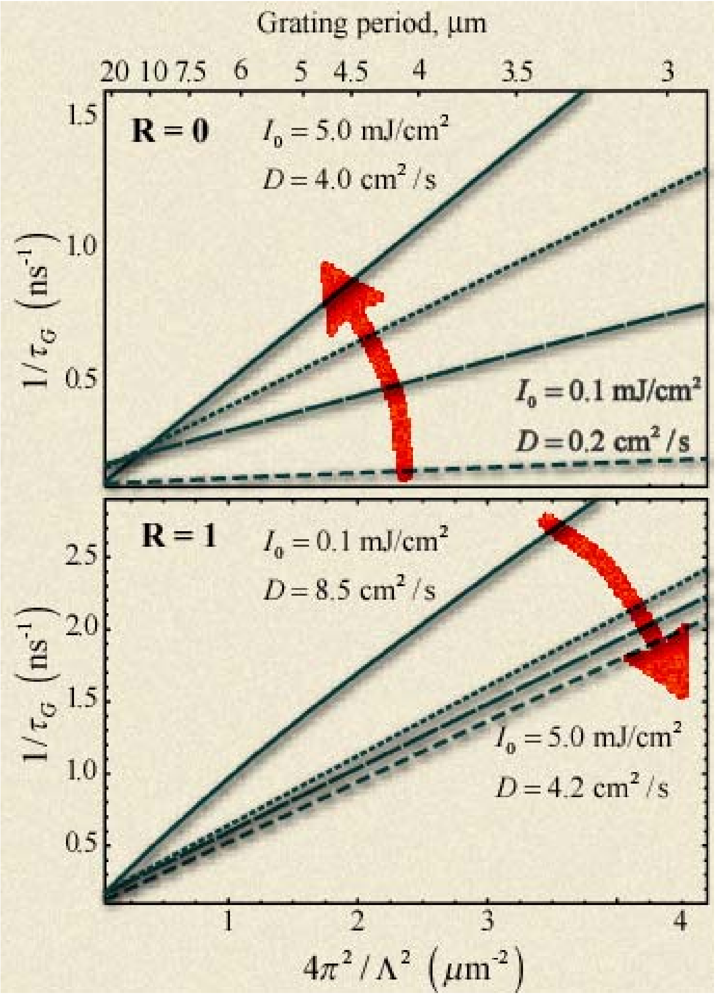
Diffusion-governed decay:





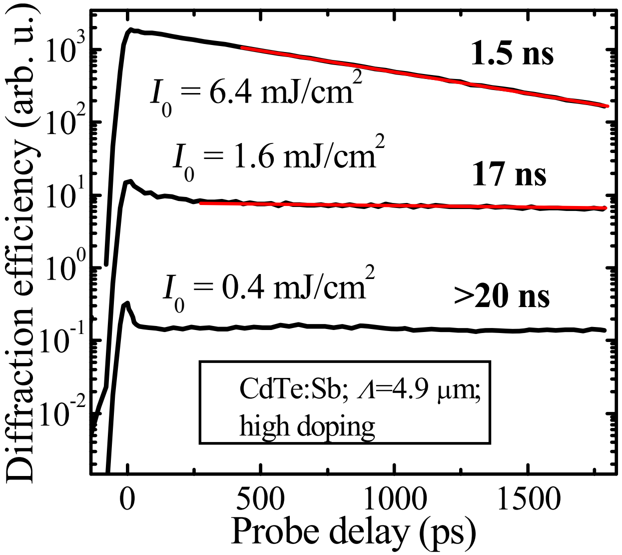
Electron diffusion compensated by drift in SC field

Determination of the photogenerated carrier sign in differently doped CdTe (1)

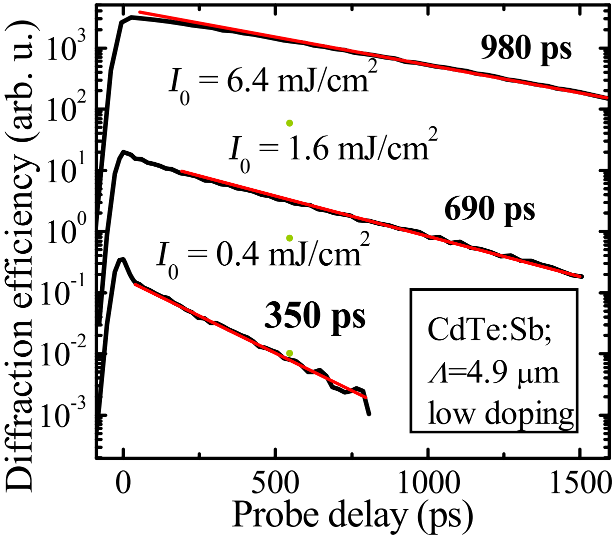


n>>p

$D_{eff} \rightarrow 0$

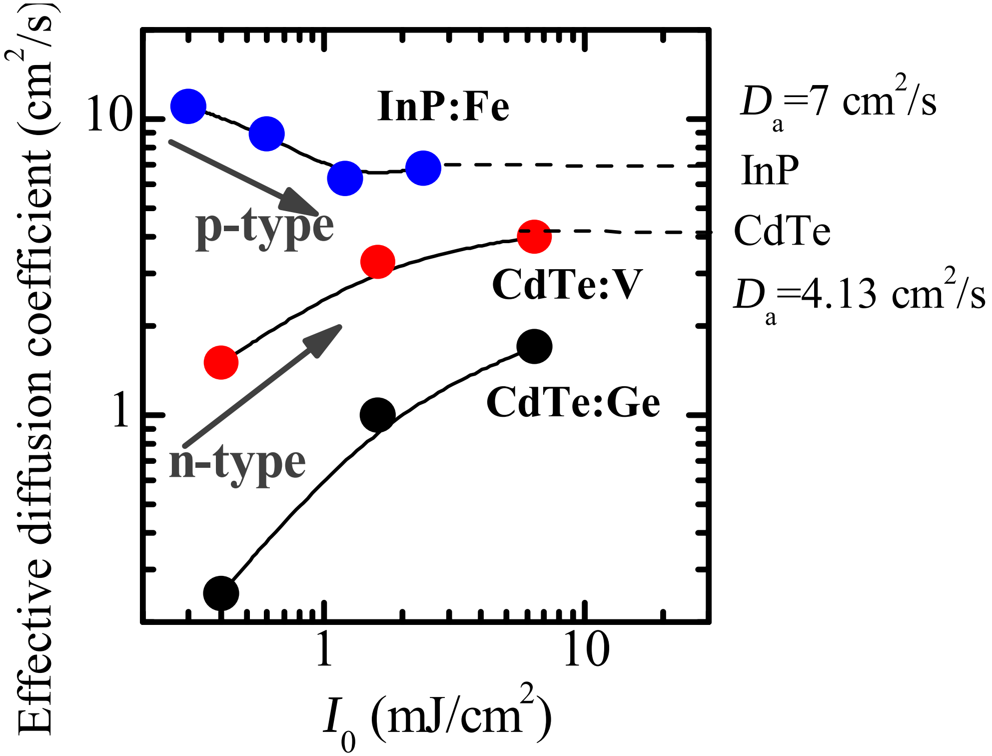


p >n, but never p>>n

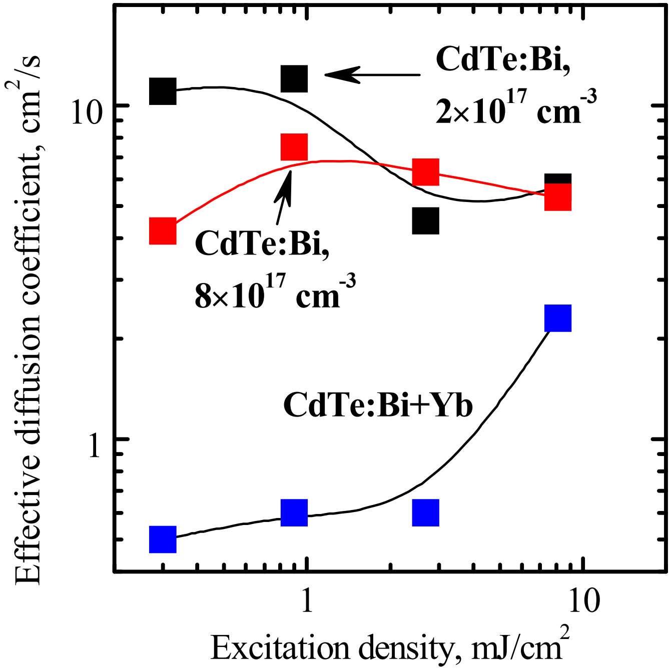


$D_{eff} > D_a$

Determination of the photogenerated carrier sign in differently doped CdTe (2)



CdTe:Ge = at 0.94 eV (Ge^+, deep acceptor) at 1.1 eV (deep donor complex X_0), at 1.22 eV (deep acceptor X^-), and at 1.35 eV (deep donor Ge_0). At 1064 nm (1.17 eV) generation both of holes via Ge+ and electrons from X_0 is possible.

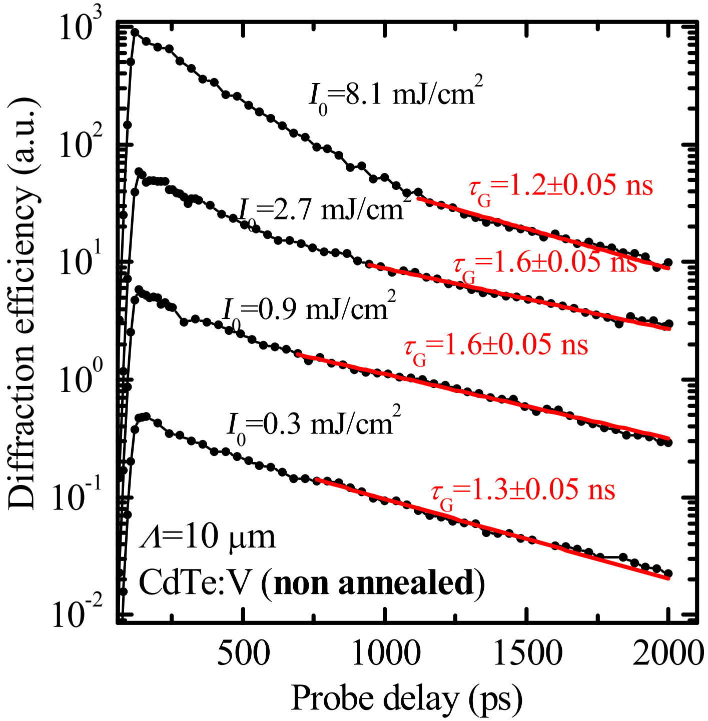


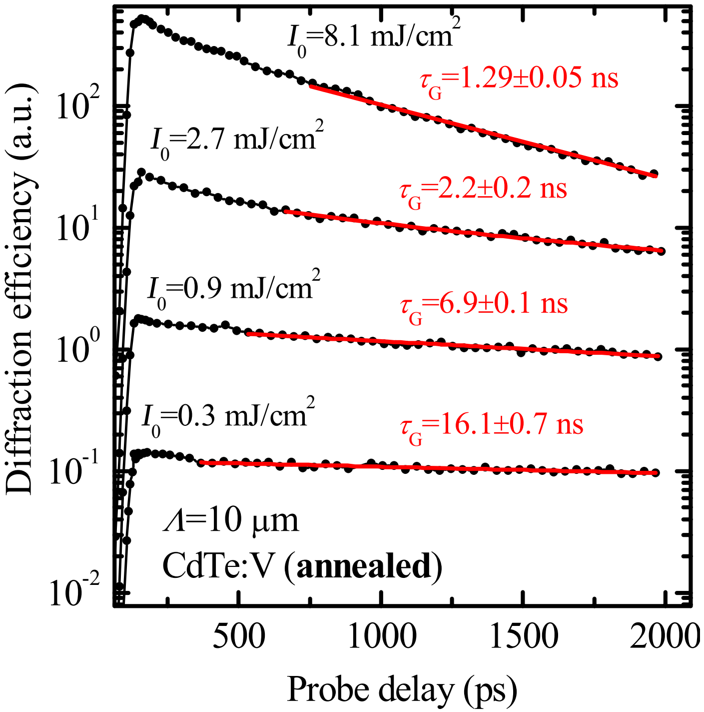
Bi(Cd) ⇒ deep donor

at E_v + 0.71 eV; N= 2 × 10^{17}

Becoming p-type with increase of Bi: **Bi (Te) ⇒ E_v+ 0.3 eV**

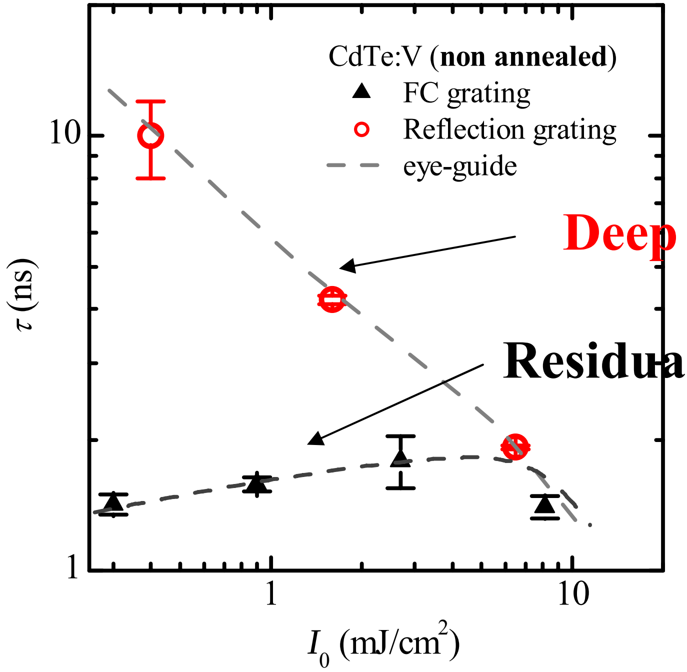
CdTe:V – as grown and annealed: diffraction on FC grating





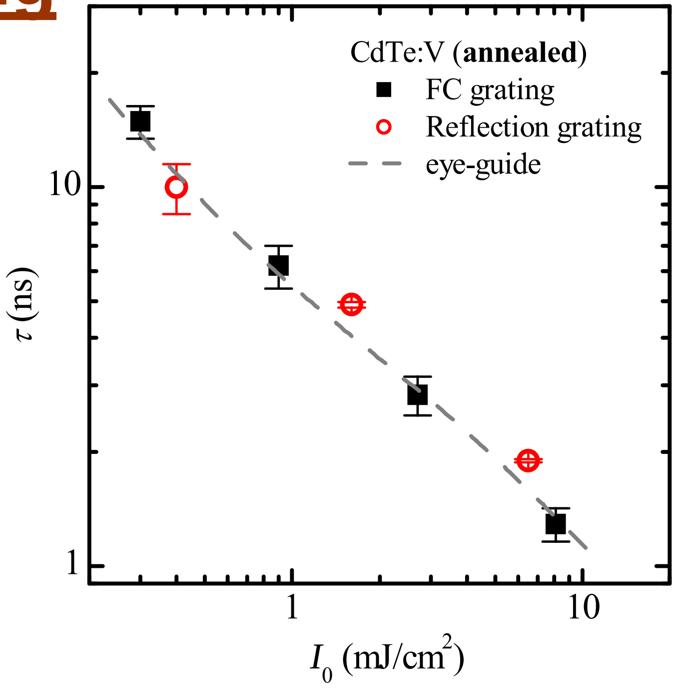
- Note different grating erasure times at low excitations: they indicate different carrier lifetimes due to higher recombination center density in as-grown crystal.
- After annealing, the crystalline quality increases due to healing the Te precipitates, filling Cd vacancies, reducing internal stress, thus carrier lifetime increases.

Optical discrimination of the deep impurity contribution to carrier trapping by reflection grating



Deep traps

Residual defects



By using the transient reflection grating technique, it becomes possible to reveal the role of deep traps in presence of various, even faster recombination channels

The similar lifetimes of grating in recharged deep traps in non-annealed and annealed samples show that after annealing the deep vanadium trap occupation remained nearly the same

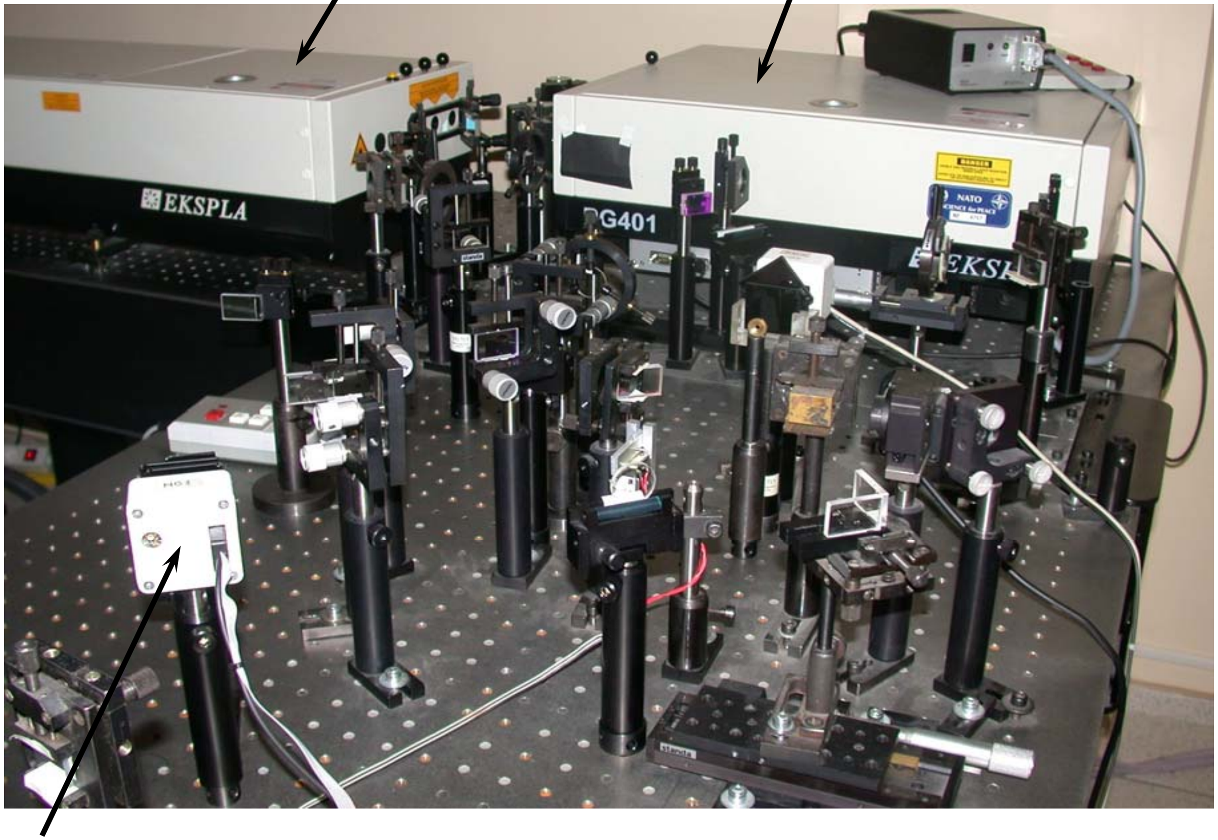
Implementation of transient grating techniques into devices

A HOLO-module

A set-up of picosecond FWM

ps laser PL2143

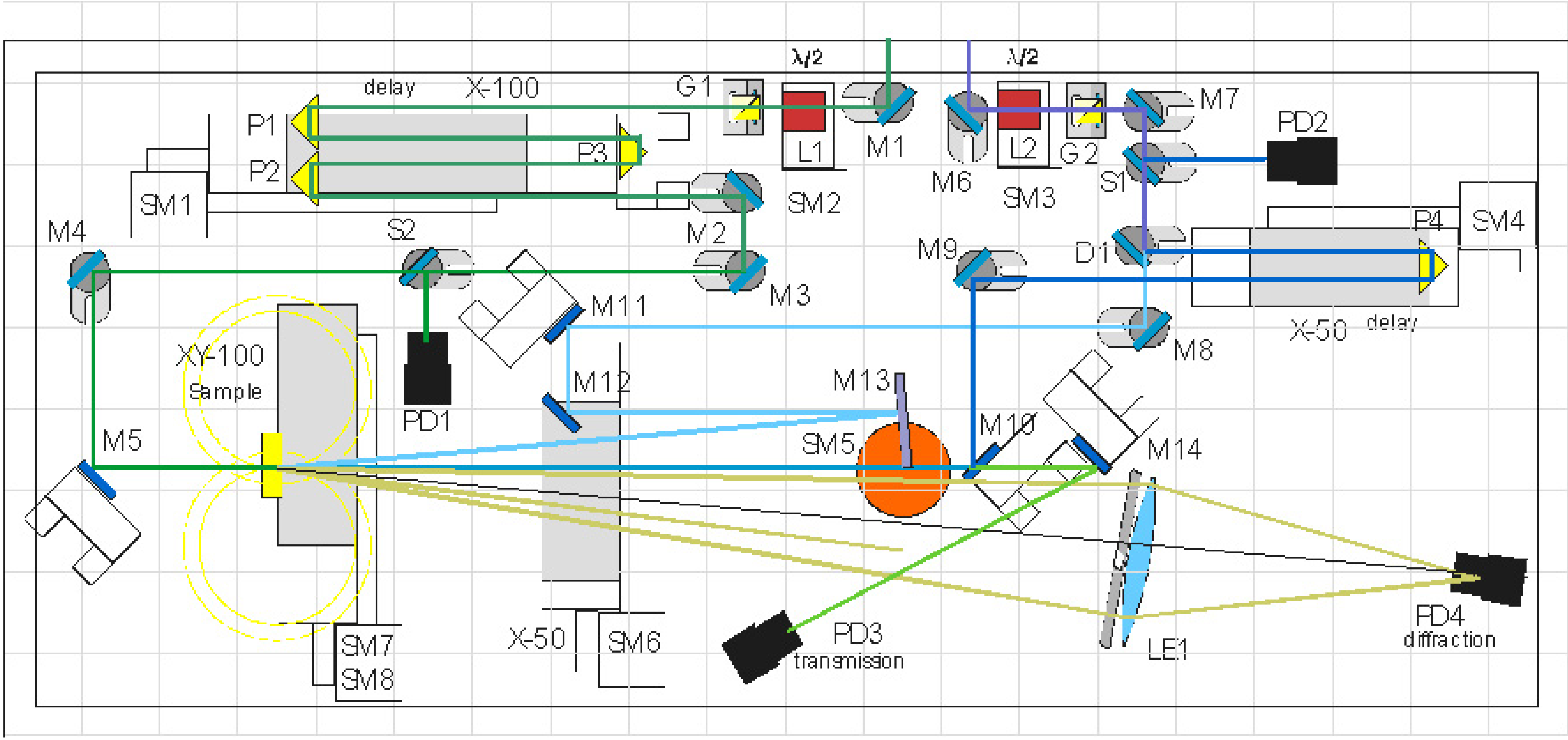
OPG PG401



Si detector

HOLO-1 module as a device* Ekspla UAB, Vilnius
(www.ekspla.com)

HOLO-2 optical scheme: nondegenerate picosececond FWM

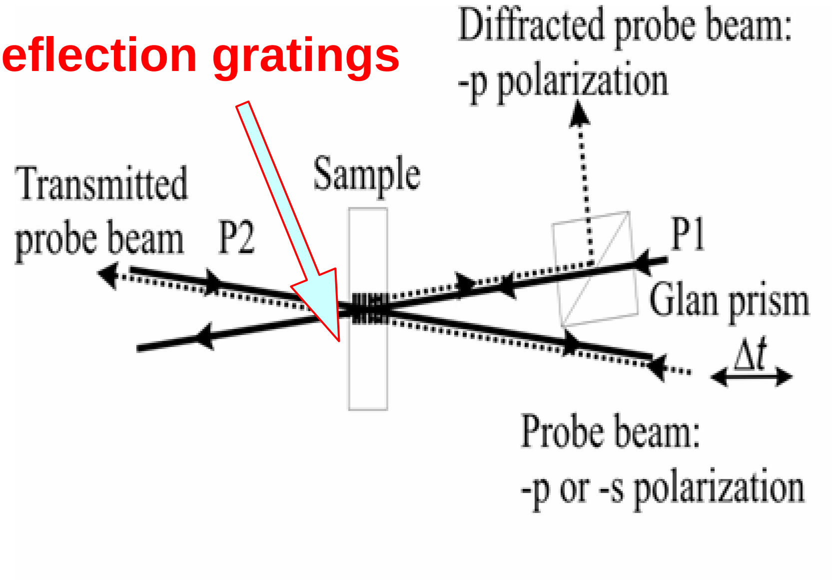


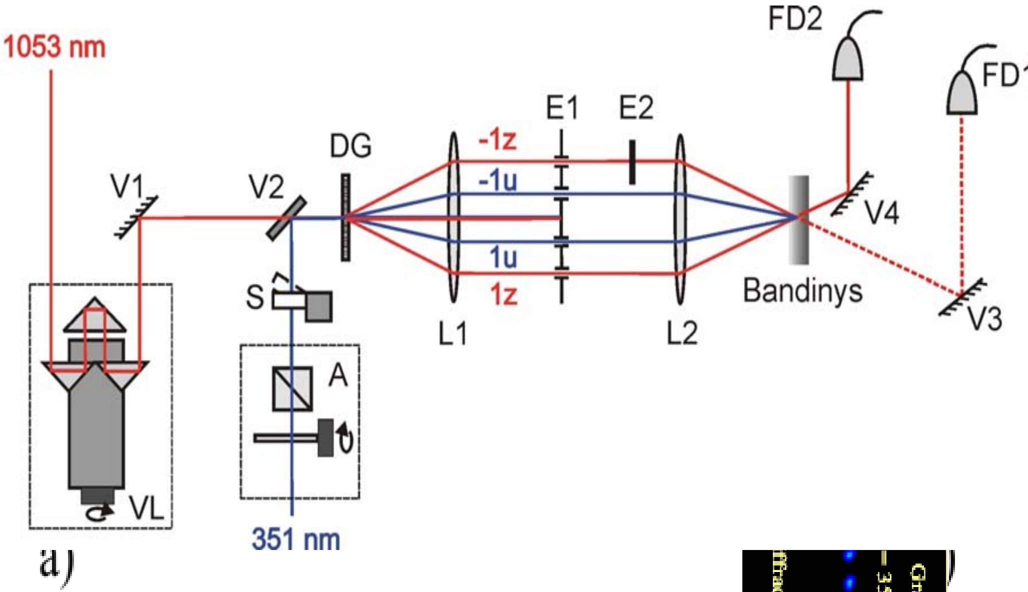
TG recorded by M13 and M10 beams, change of angle (grating period) is achieved by translation/rotation of mirror M13, delay line P4 adjustment for temporal overlapof the beams.

HOLO-2 based measurement system at Rentsellar Polytechnical Institute (USA)

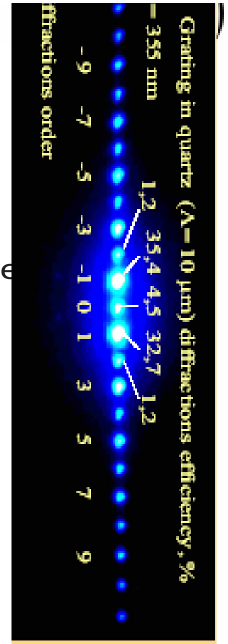
New optical schemes for dynamic holography

Reflection gratings

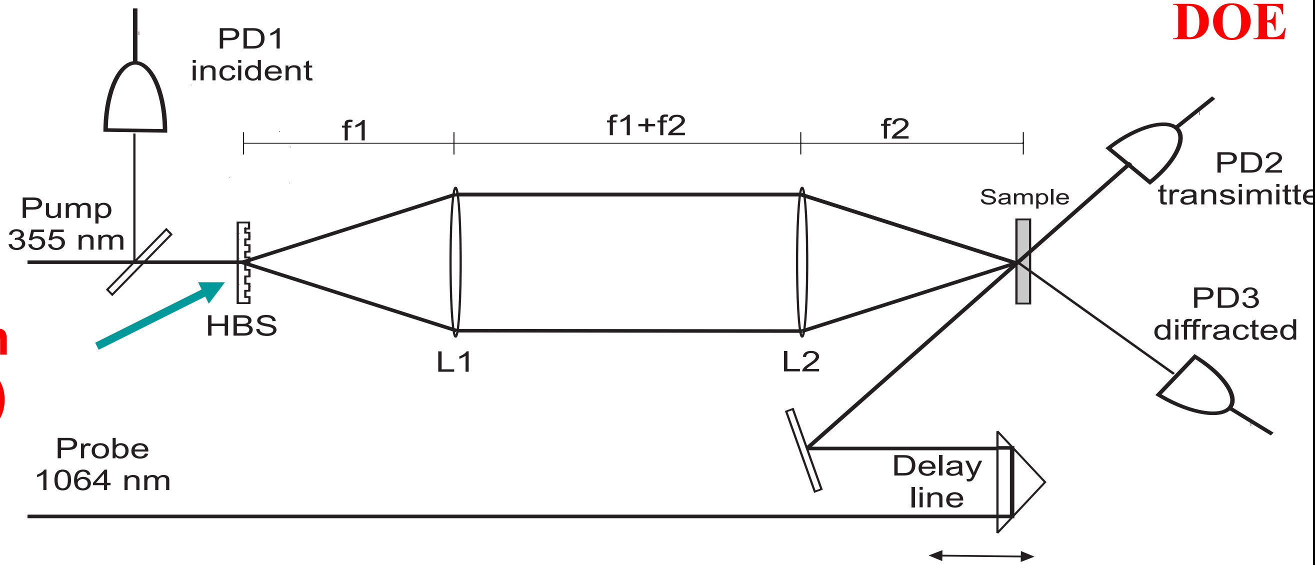




DOE



DOE as beam splitter (HBS)



Summary

We demonstrated various optical-holographic schemes of picosecond transient gratings which enable to explore free carrier, photorefractive, and absorptive nonlinearities for investigation of

- interband carrier generation, determination of carrier lifetime, D, S
- carrier generation from deep impurity centers in bulk crystals
- monitoring of carrier dynamics in presence of SC field
- direct monitoring of SC field – its buildup and decay
- determination of photogenerated carrier sign via effective diffusion coefficient
- determination of deep trap occupation ratio
- optical discrimination of the deep trap contribution to carrier capture

- Transient grating schemes, if implemented in devices, can serve as novel tools for determimation of electronic parameters of semiconductors

Thank you for your attention !