Terahertz dielectric properties, ultrafast photocarrier capture/recombination and transport dynamics in graphene-mesoporous silicon nanocomposites

African School of Physics Seminars
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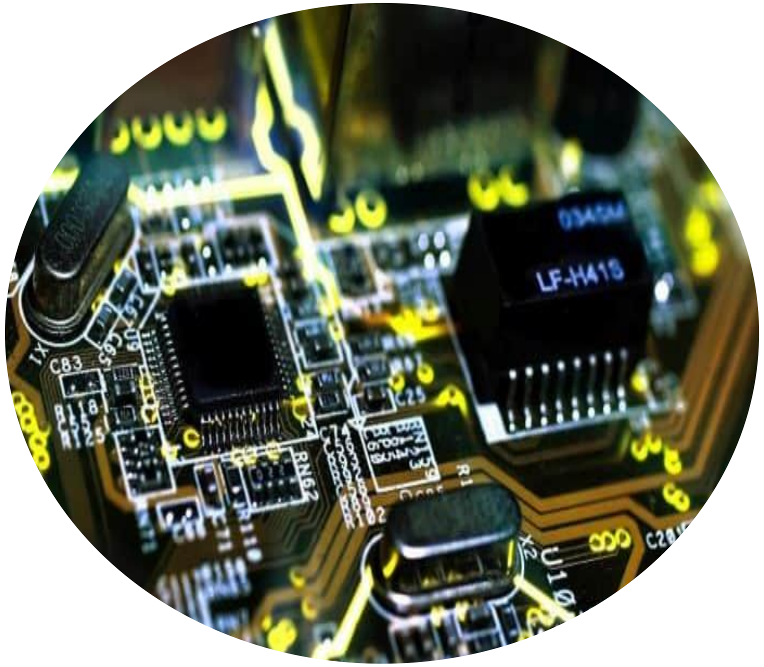
Plan of the presentation

- ❑Motivation and interest
- ❑Research methods
- ❑Results on investigating Graphene-Mesoporous Silicon Nanocomposites

Why studying electronic properties in semiconductor nanostructures ?

Nanoelectronics



Smartphone Computers

Flat screens Informatique

Optoelectronics

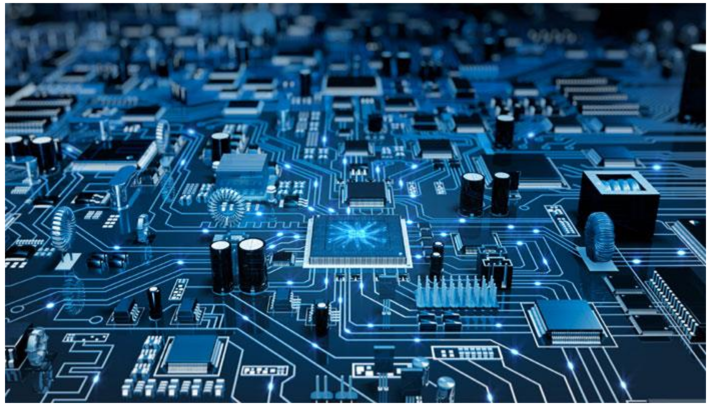
Photo-emitters Photosensors

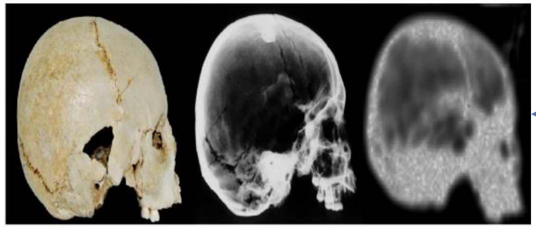
Quantum Technology

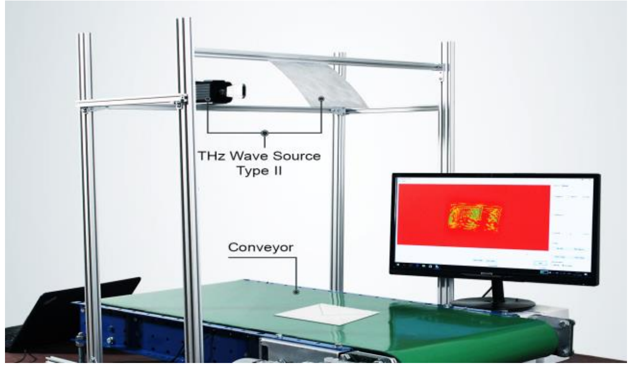
Light emitting diodes Fiber optics

THz technology

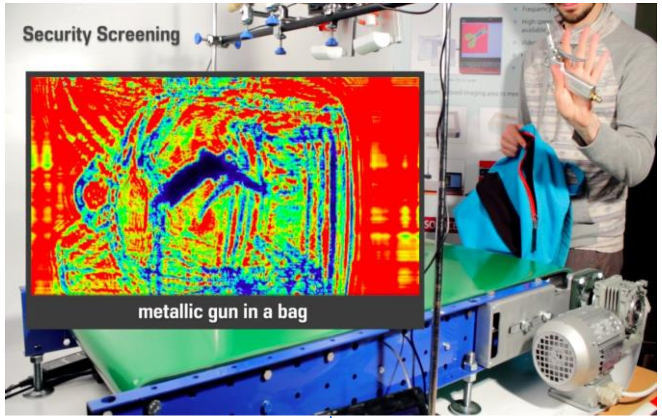
Electronic circuitory

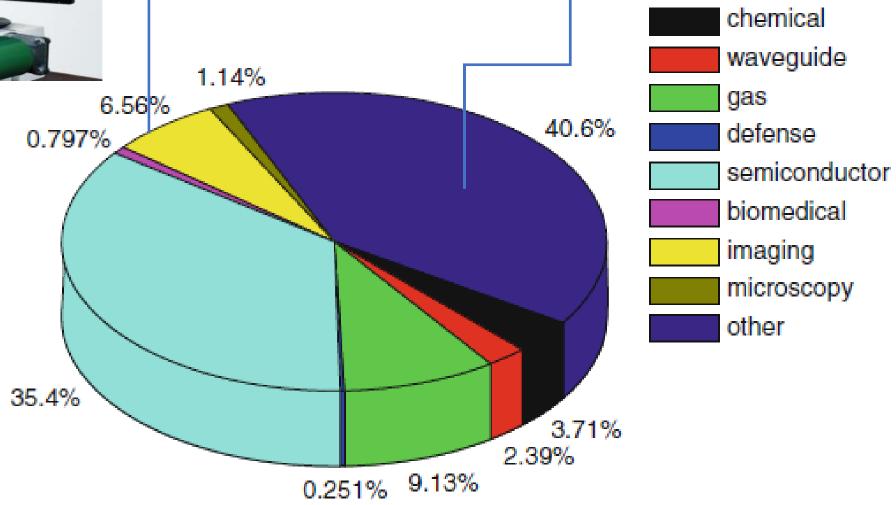




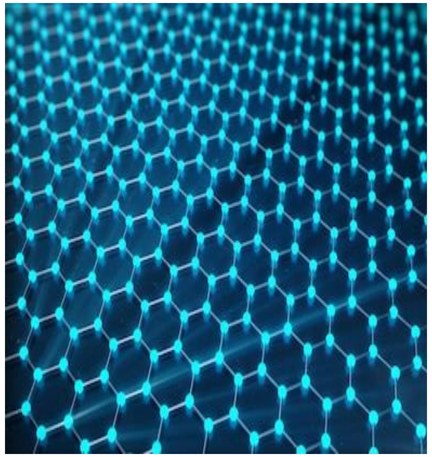


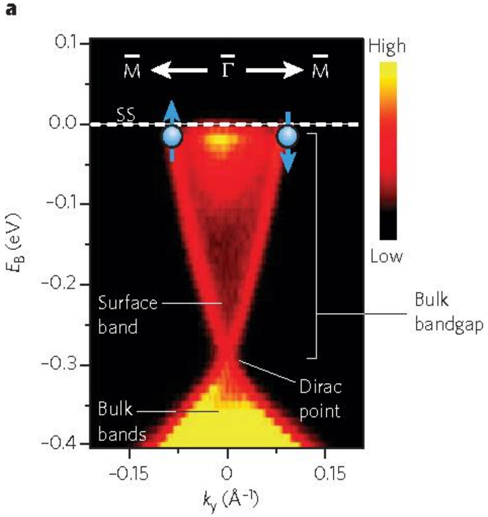
Imaging and security

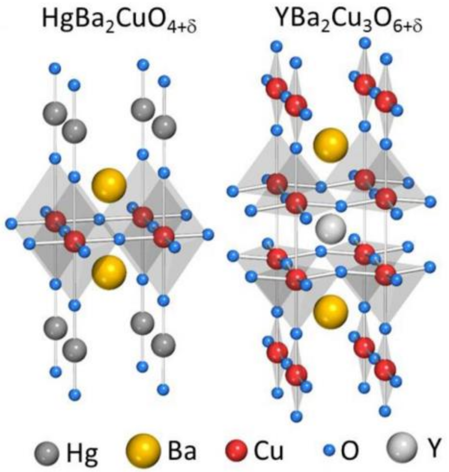




Quantum Materials







J.E. Moore, *Nature* **volume 464**, pages 194–198 (2010)

X.-C Zhang, Jingzhou Xu, Introduction to THz wave Photonics, 2010

Nature communications

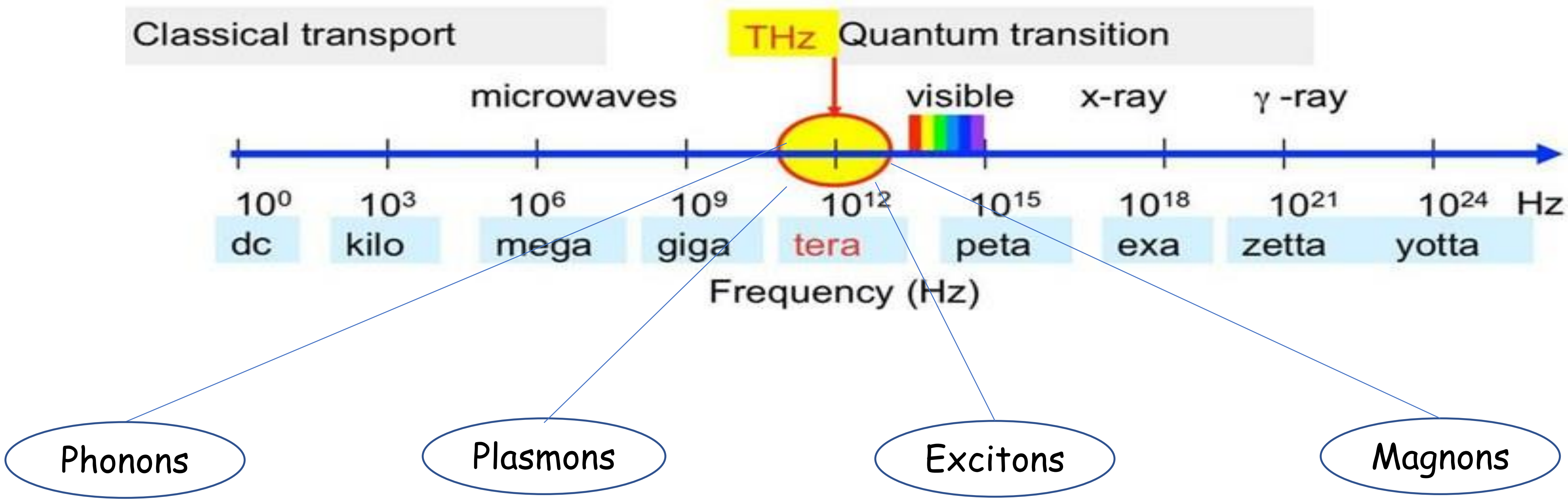
http:// terasense.com

http://zeenews.india.com

Terahertz (THz) radiation

0,1 – 30 THz

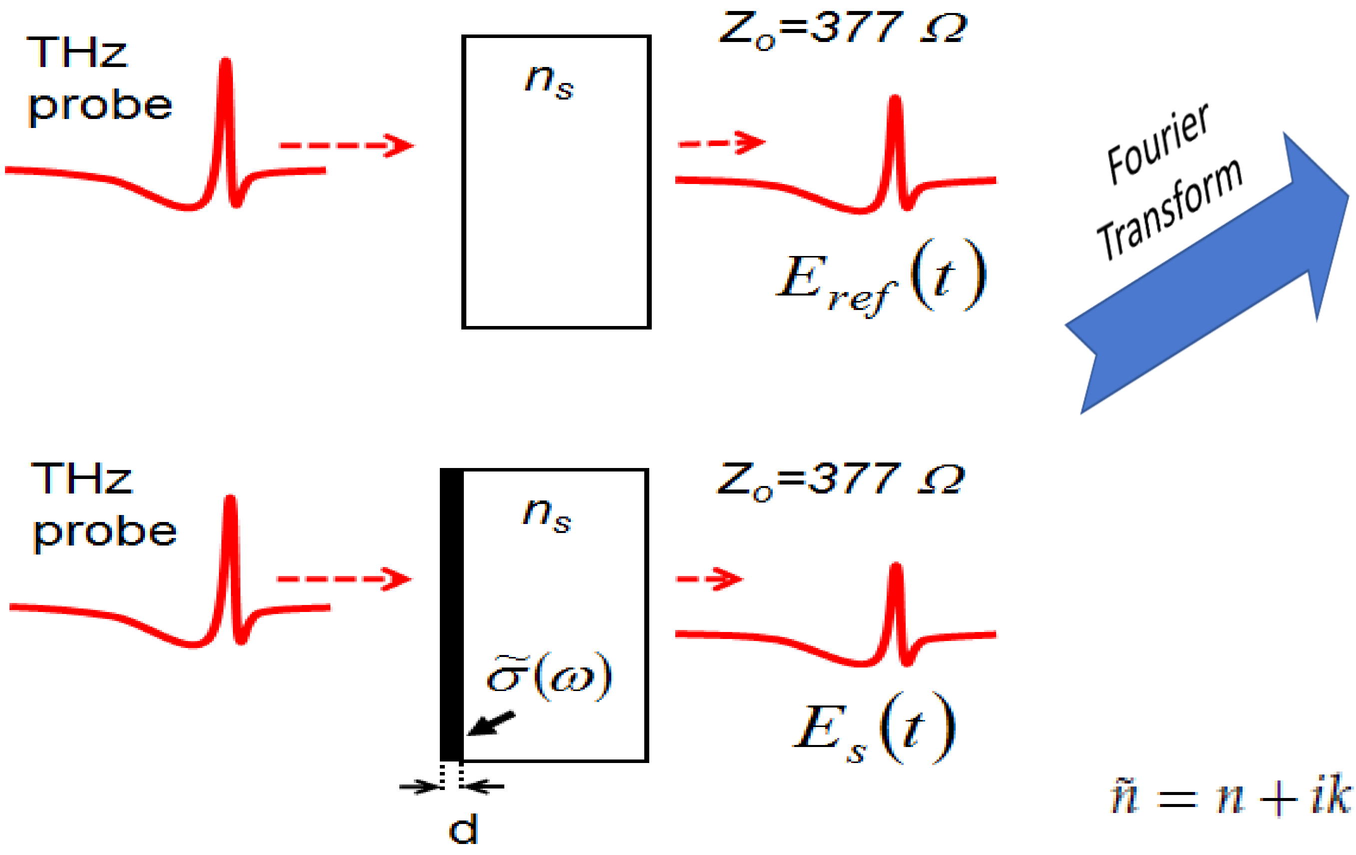
1 THz = 10^{12} Hz = 33 cm^{-1} = 4,1 meV = 48 K



Research Methods

- THz time-domain spectroscopy
- Optical Pump-Terahertz Probe spectroscopy

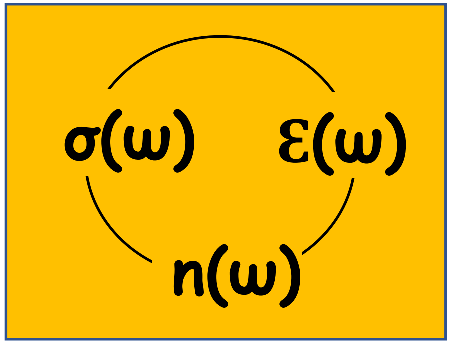
THz time-domain spectroscopy



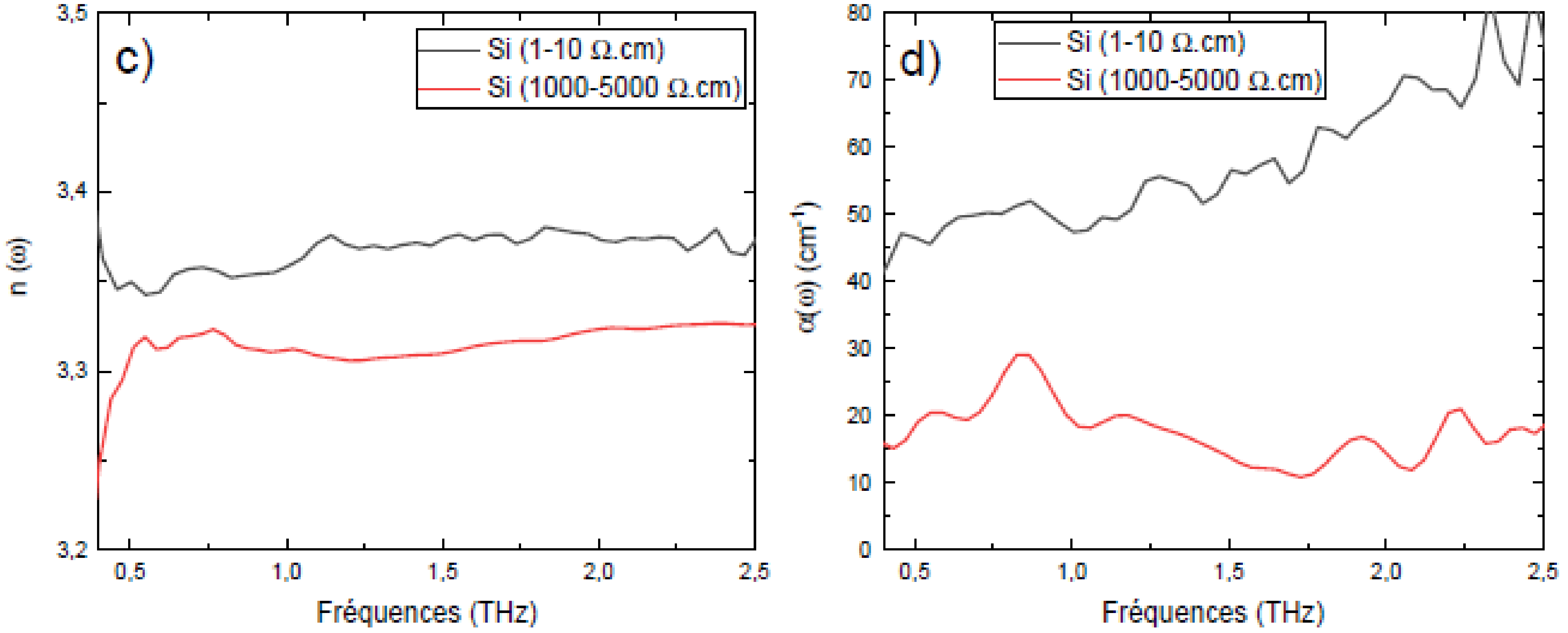
Amplitude and phase of $T(\omega)$ are function of n(ω) (σ ou ε)

Appropriate modelling of enables to extract dielectric or transport properties

$$\tilde{n} = n + ik = \sqrt{\left(\epsilon_r + \frac{i\tilde{\sigma}}{\epsilon_0 \omega}\right)}$$



Dielectric properties of Semiconductors

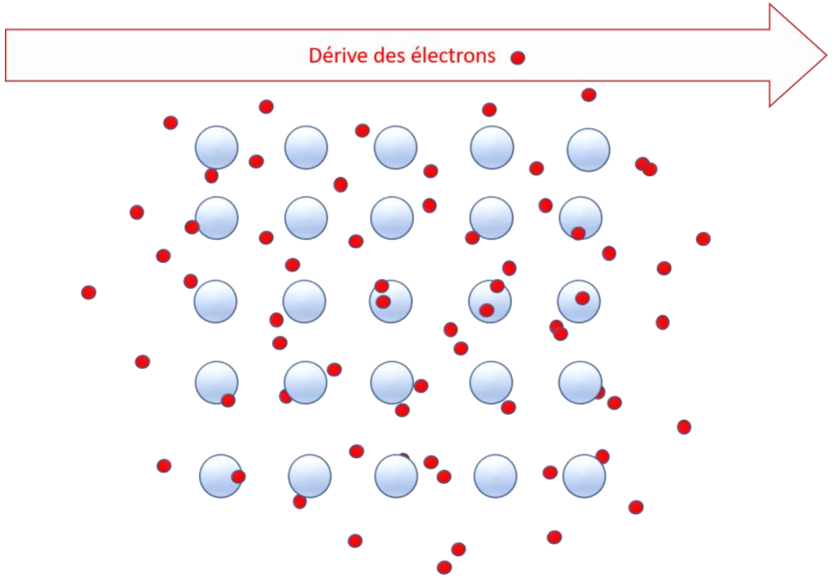


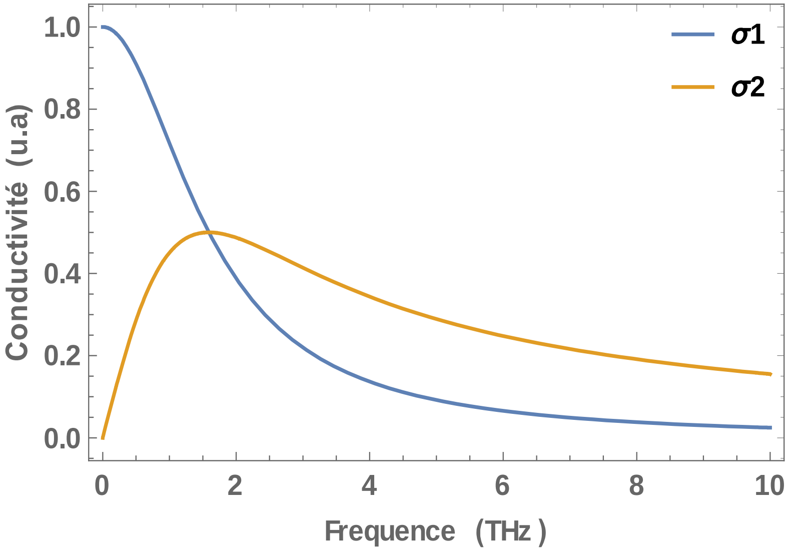
Défi Jr., PhD Thesis, Université de Sherbrooke

Semi-classical transport models

Drude-model

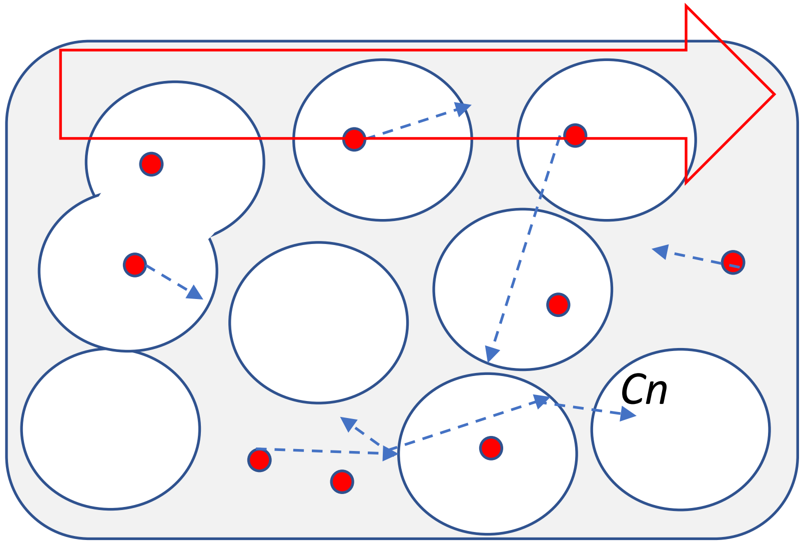
$$\sigma(\omega) = \frac{nq^2\tau}{m^*(1 - i\omega\tau)}$$

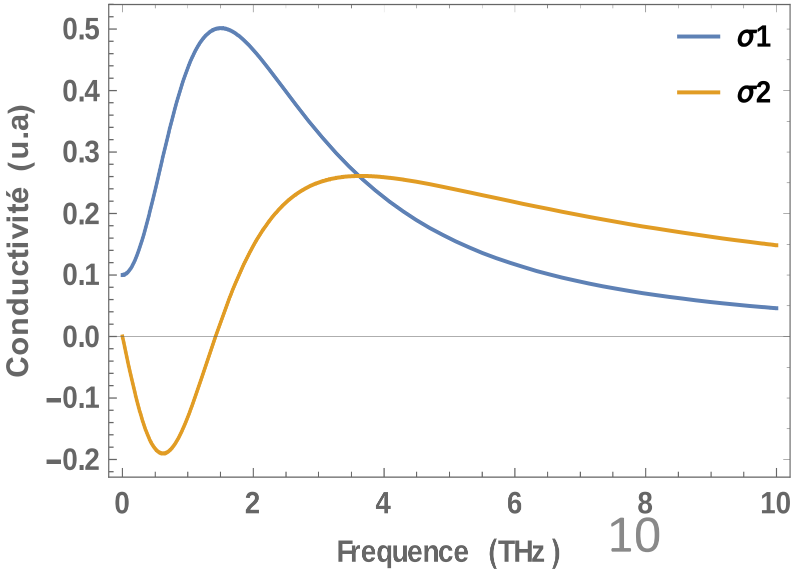




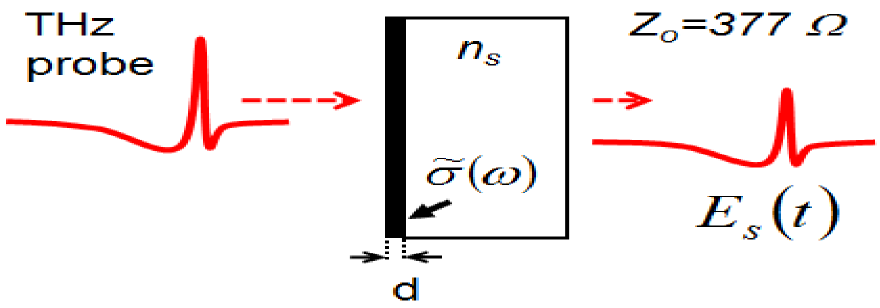
Drude-Smith model

$$\sigma(\omega) = \frac{\sigma_0}{1 - i\omega\tau}\left[1 + \sum_{n=1}^{\infty}\frac{c_n}{(1 - i\omega\tau)^n}\right]$$



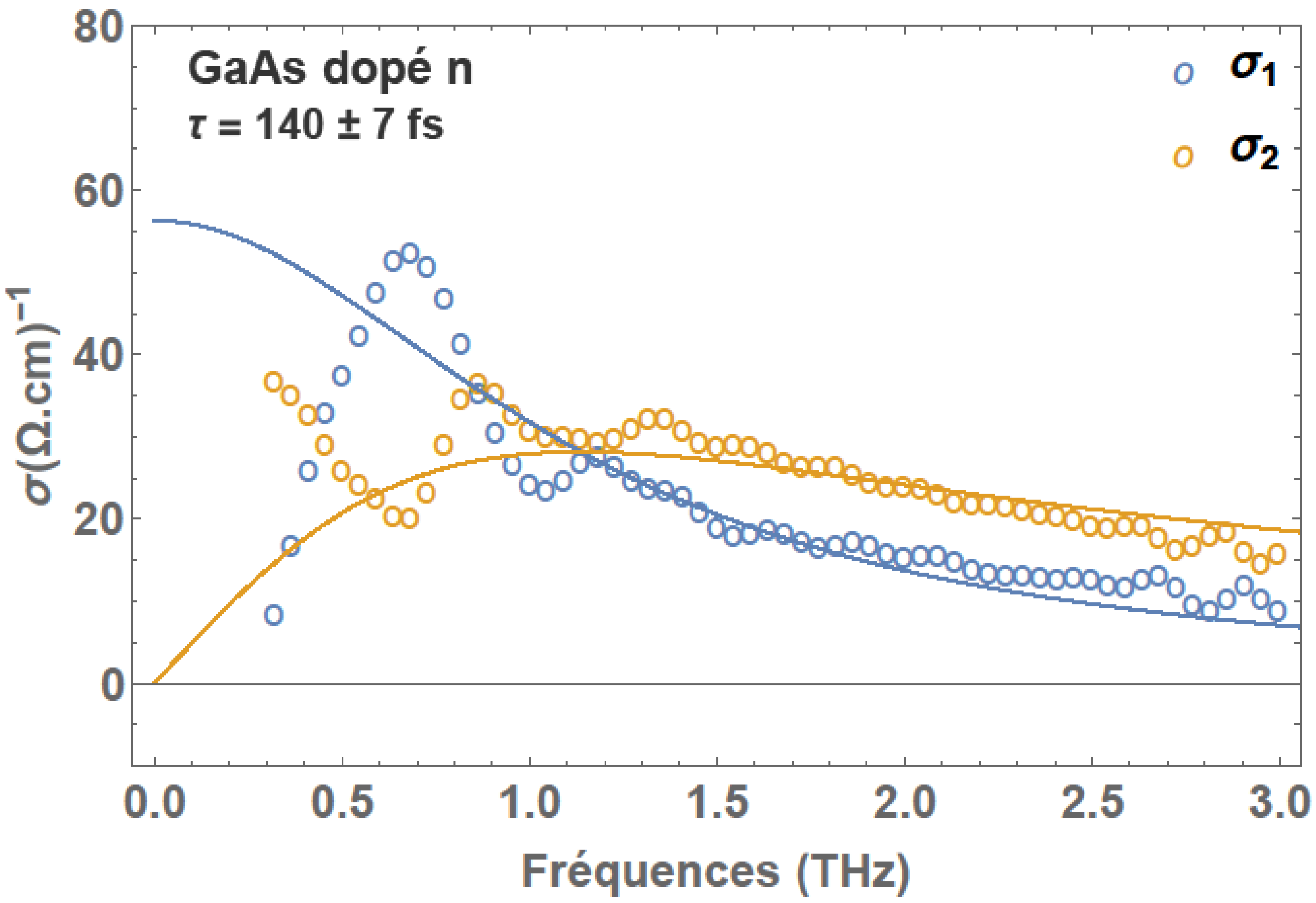


Classical transport in N-doped GaAs



Drude transport model

$$\sigma(\omega) = \frac{nq^2\tau}{m^*(1 - i\omega\tau)}$$



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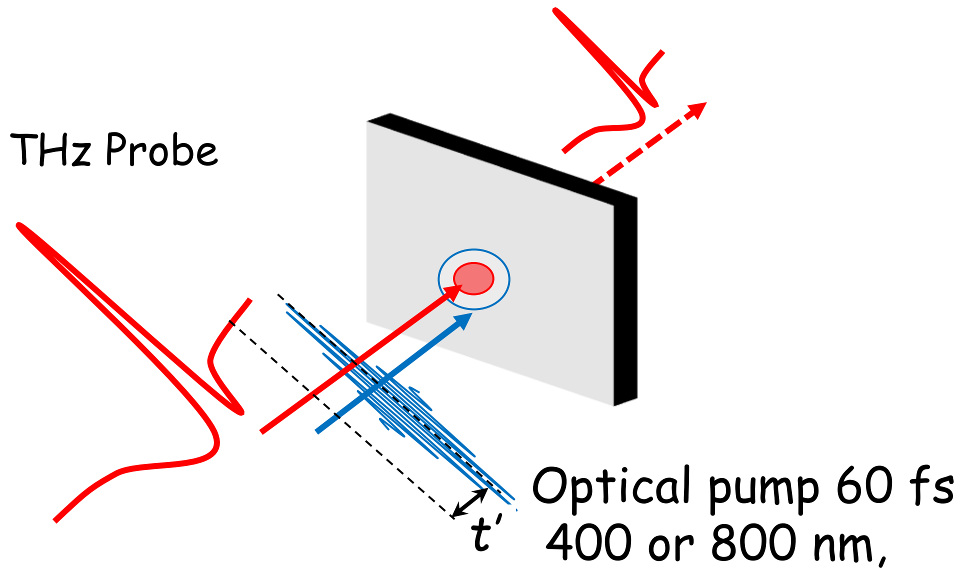
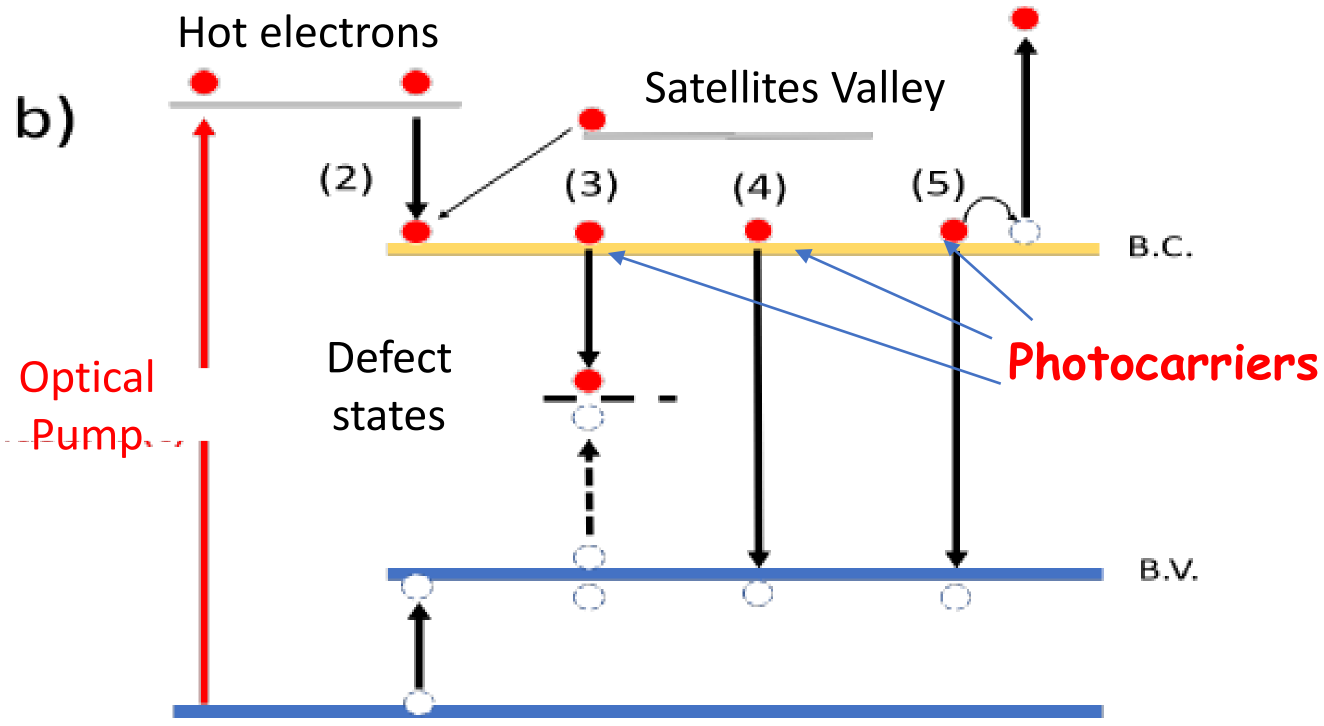
Free charge scattering time and density determined

Optical Pump THz probe spectroscopy





Time resolved photoconductivity

$$\frac{\Delta T(t)}{T} \propto \Delta\sigma(t)$$
$$= e\left(\mu_n \Delta n(t) + \mu_p \Delta p(t)\right)$$

Frequency resolved photoconductivity

$$\Delta\tilde{\sigma}_s(\omega) = \Delta\sigma_1(\omega) + i\Delta\sigma_2(\omega)$$

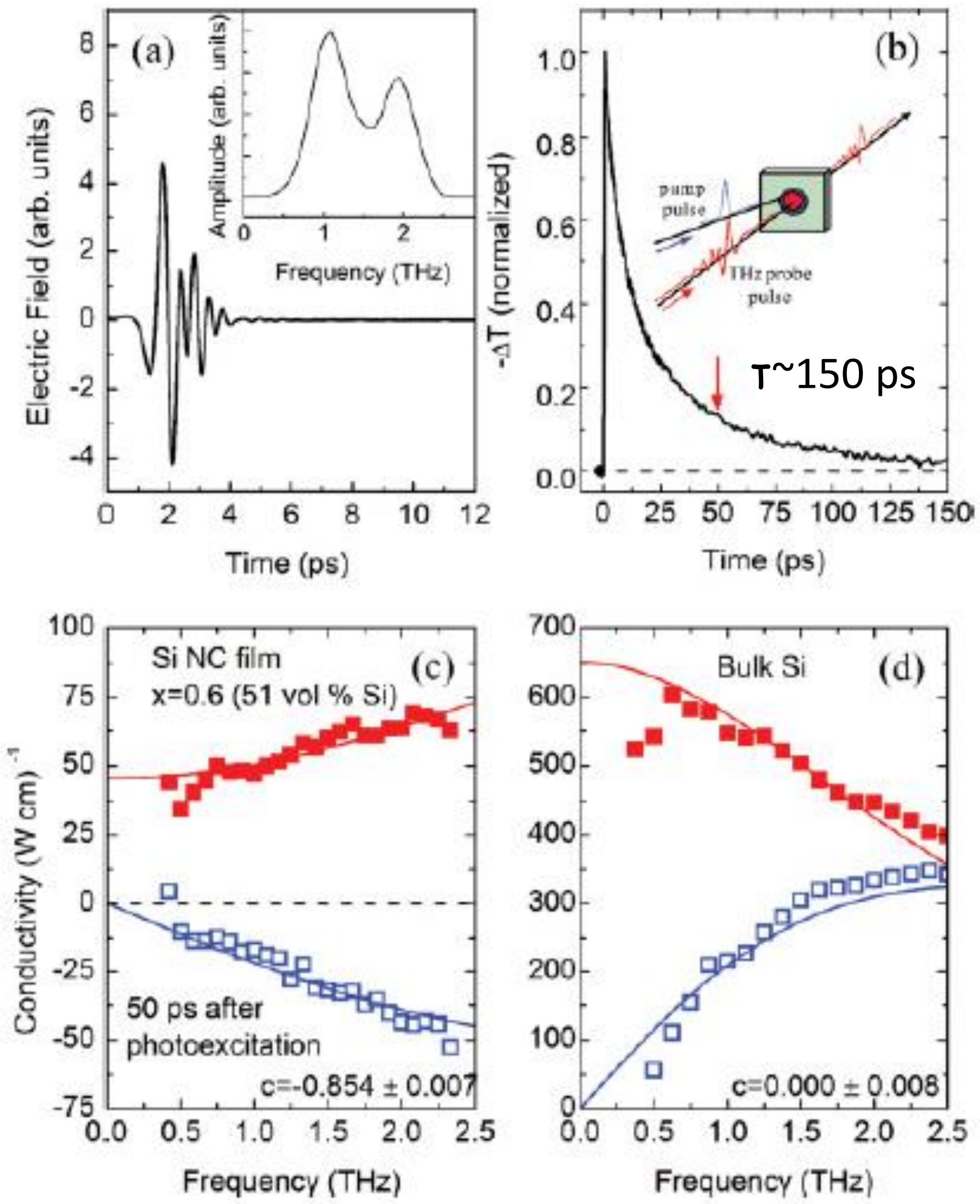
Photocarrier dynamics and transport in Si Nanocrystals

Drude transport in an unconfined system (d)

$$\sigma(\omega) = \frac{nq^2\tau}{m^*(1 - i\omega\tau)}$$

Drude-Smith transport type in Si nanocrystals (confined system) (c)

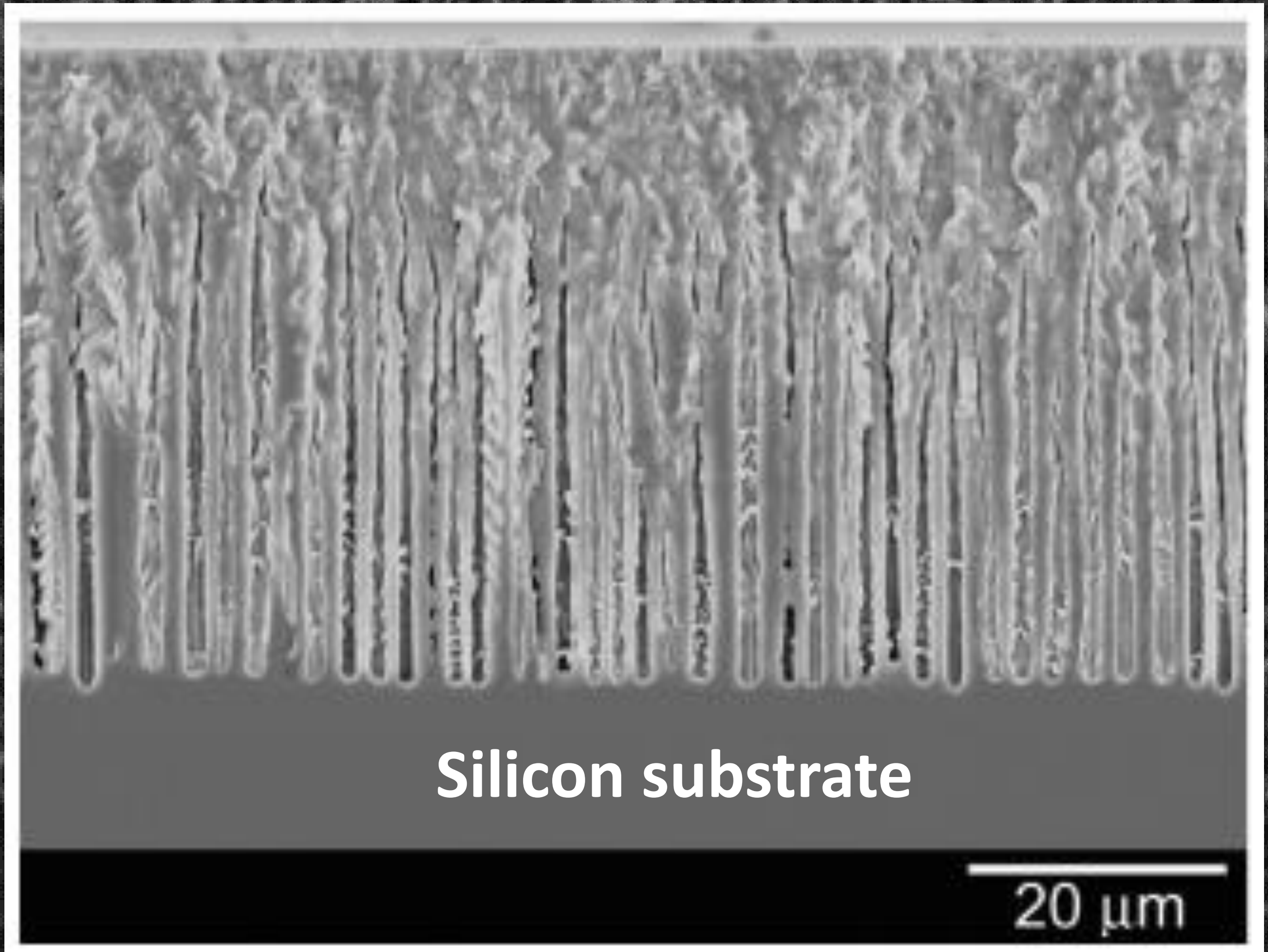
$$\sigma(\omega) = \frac{\sigma_0}{1 - i\omega\tau}\left[1 + \sum_{n=1}^{\infty} \frac{c_n}{(1 - i\omega\tau)^n}\right]$$



Titova et al., PHYSICAL REVIEW B **83**, 085403 (2011)

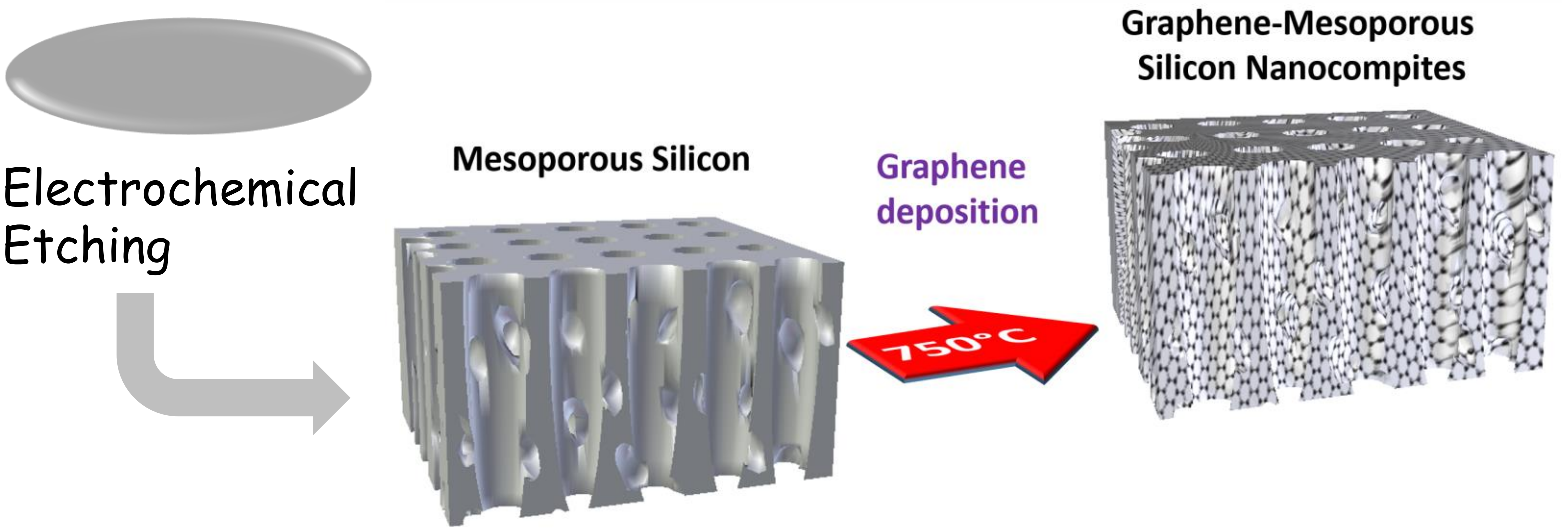
Any Questions so far?

Porous silicon

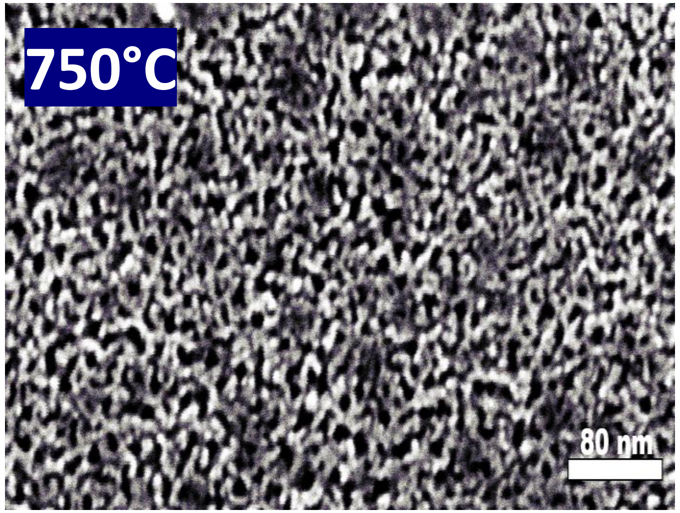


- ❑ Large surface-to-volume ratio
- ❑ Conserved Si crystalline structure
- ❑ Controllable pore size and orientation
- ❑ Controllable crystallite size

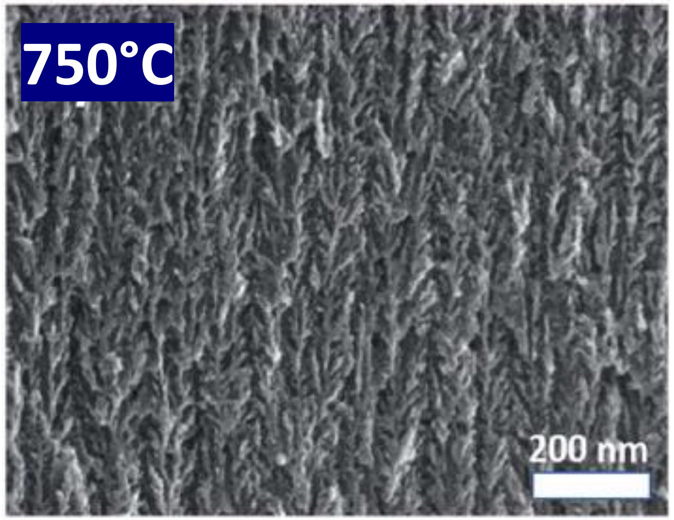
Graphene-Mesoporous Silicon Nanocomposites



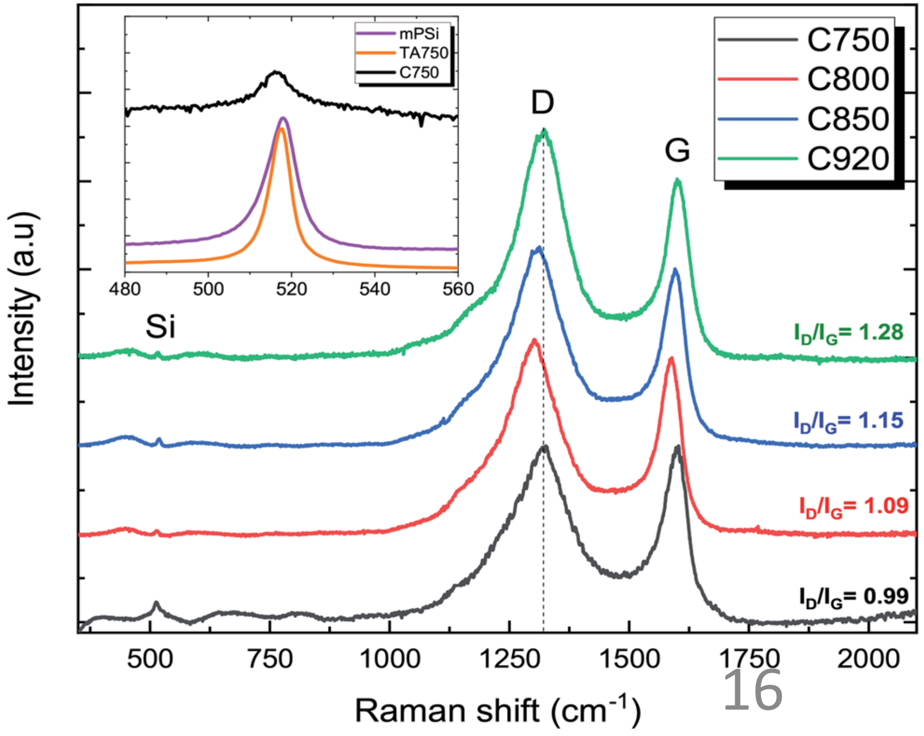
Pioneer Study
- Dielectric properties
- Charge dynamics
- Transport properties



In-plane view



Side view



THz time-domain spectroscopy

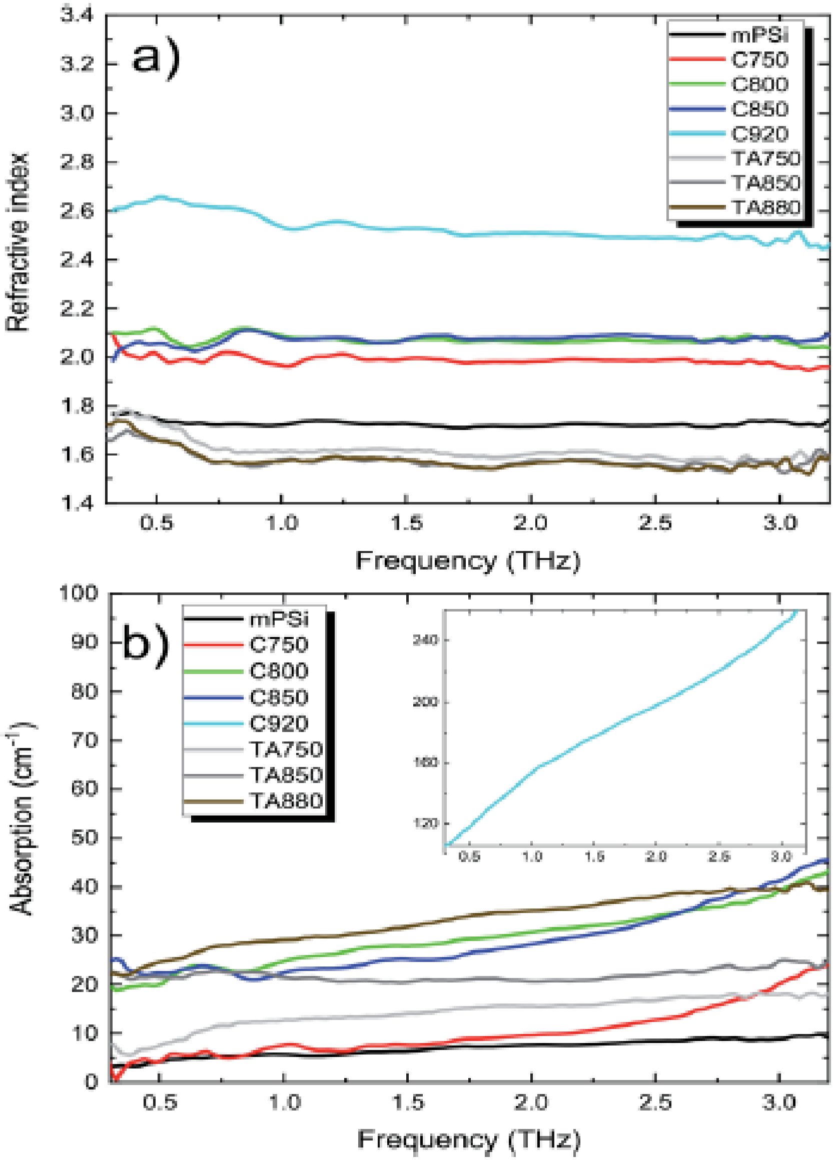


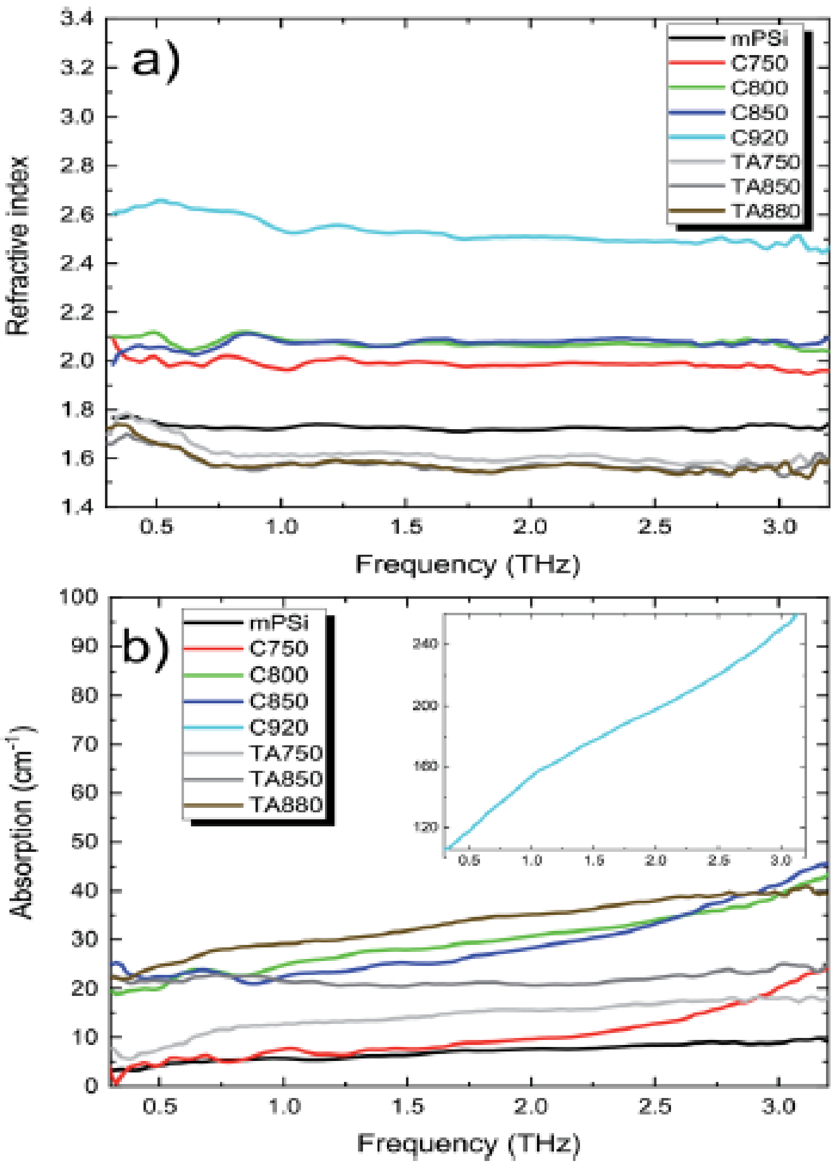
Table 1 Impact of graphene deposition temperature on refractive index and porosity as determined by Bruggeman model (eqn (6))

Samples	Temp (°C)	n_{eff}	Porosity p (%)	f_C (%)
mPSi	—	1.72	63.5	—
C750	750	1.96	61.5	7.7
C800	800	2.02	59.8	9.4
C850	850	2.07	59.7	9.5
C920	920	2.53	54.5	14.7

$$p\frac{{n_0}^2 - {n_{eff}}^2}{{n_0}^2 + 2{n_{eff}}^2} + f_{Si}\frac{{n_{Si}}^2 - {n_{eff}}^2}{{n_{Si}}^2 + 2{n_{eff}}^2} + f_C\frac{{n_C}^2 - {n_{eff}}^2}{{n_C}^2 + 2{n_{eff}}^2} = 0$$

D.J. Jubgang Fandio et al., Nanoscale Adv. 2020, **2**, 340

THz time-domain spectroscopy



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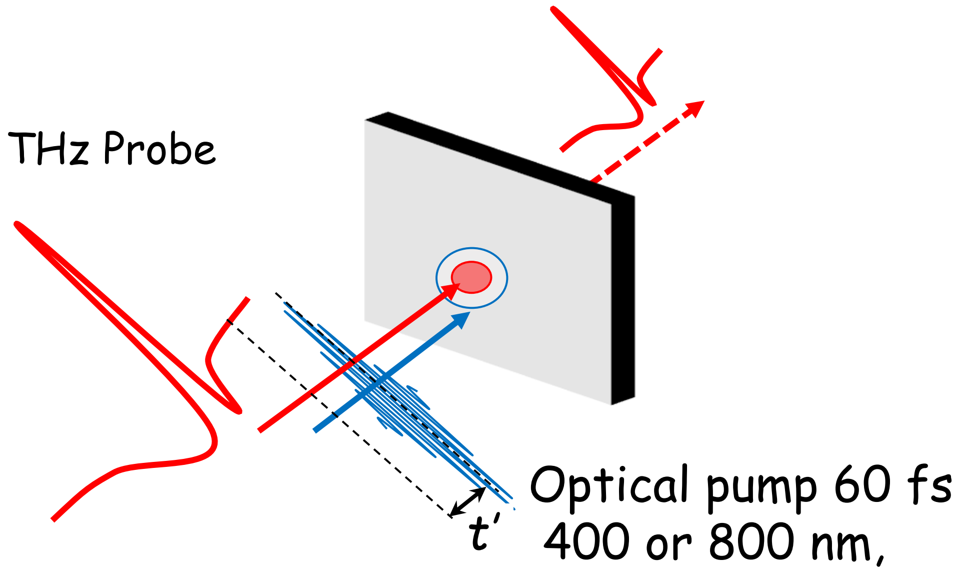
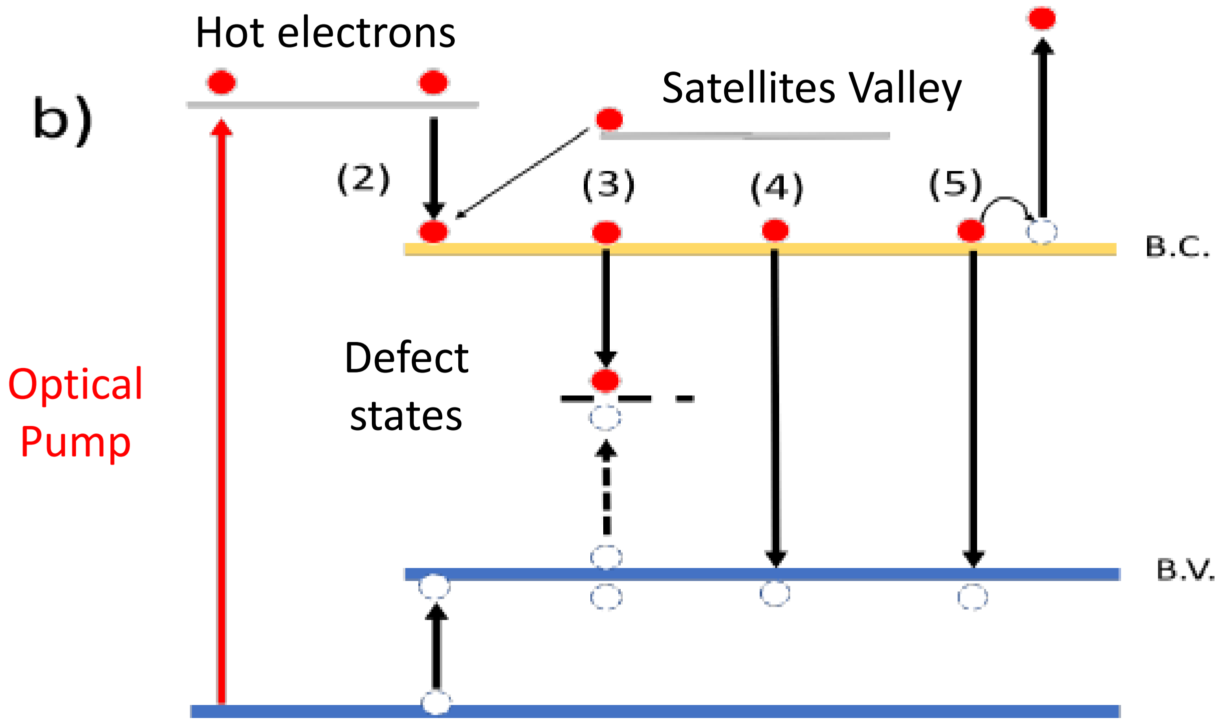
- ❑ Dielectric properties determined
- ❑ Porosity determined using an Effective Medium Theory
- ❑ Volume fraction of Carbon determined
- ❑ Increase in absorption level consistent with the presence of defect states (dangling bonds, impurities). Scénario also consistent with Raman spectroscopy.

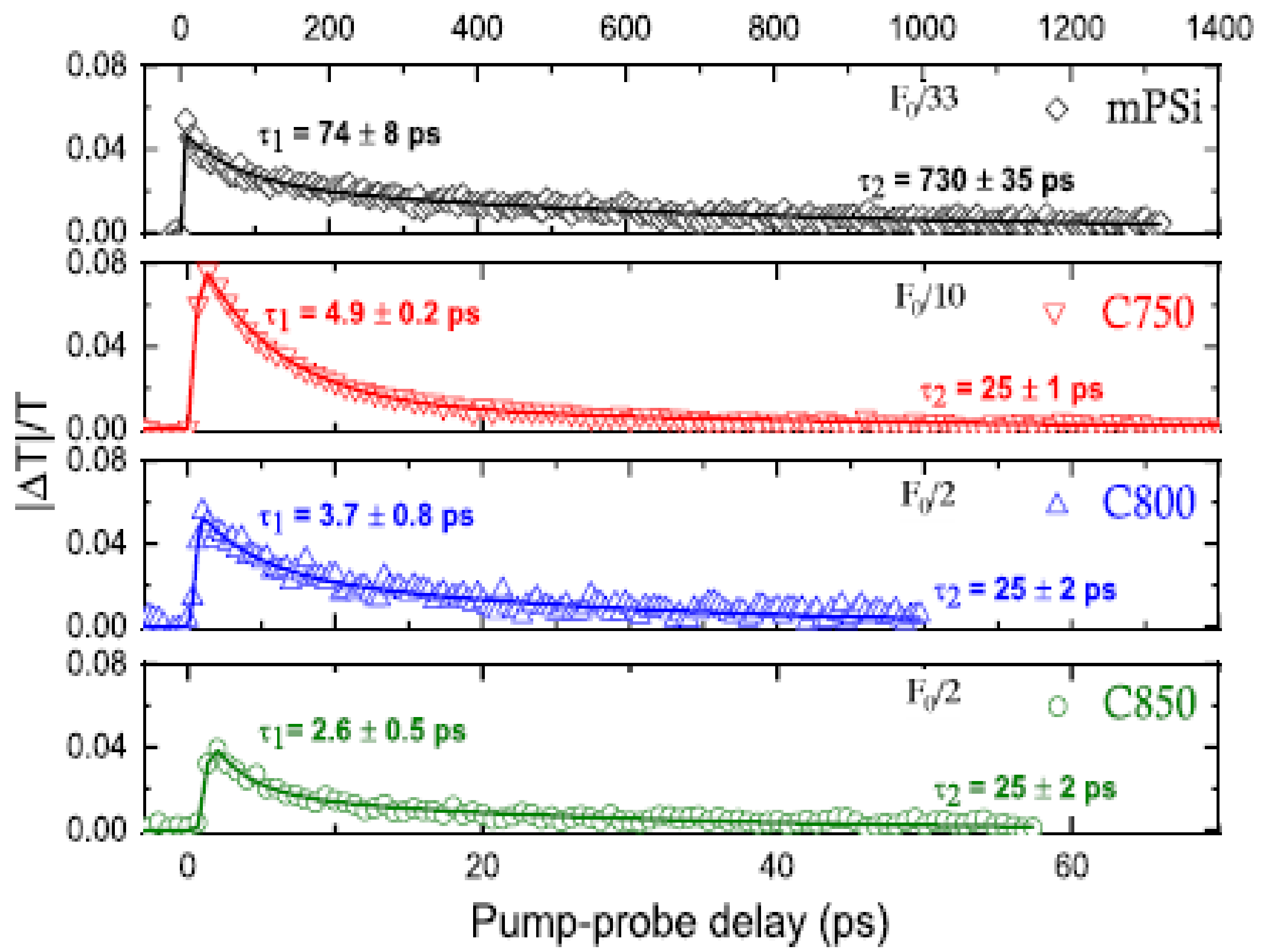
Optical pump THz probe spectroscopy





(2) Photocarrier cooling
(3) Photocarrier trapping
(4) Band-to-band recombination
(5) Auger recombination

Optical pump THz probe spectroscopy



D.J. Jubgang Fandio et al, PRB 102, 115407 (2020)

- ❑ Decrease of τ_1 and τ_2 with graphene coating.
- ❑ Decrease of τ_1 and of $\Delta T_{max}/T$ with the graphene deposition temperature
- ✓ τ_1 likely associated to the presence of surface defect states

Fitting function

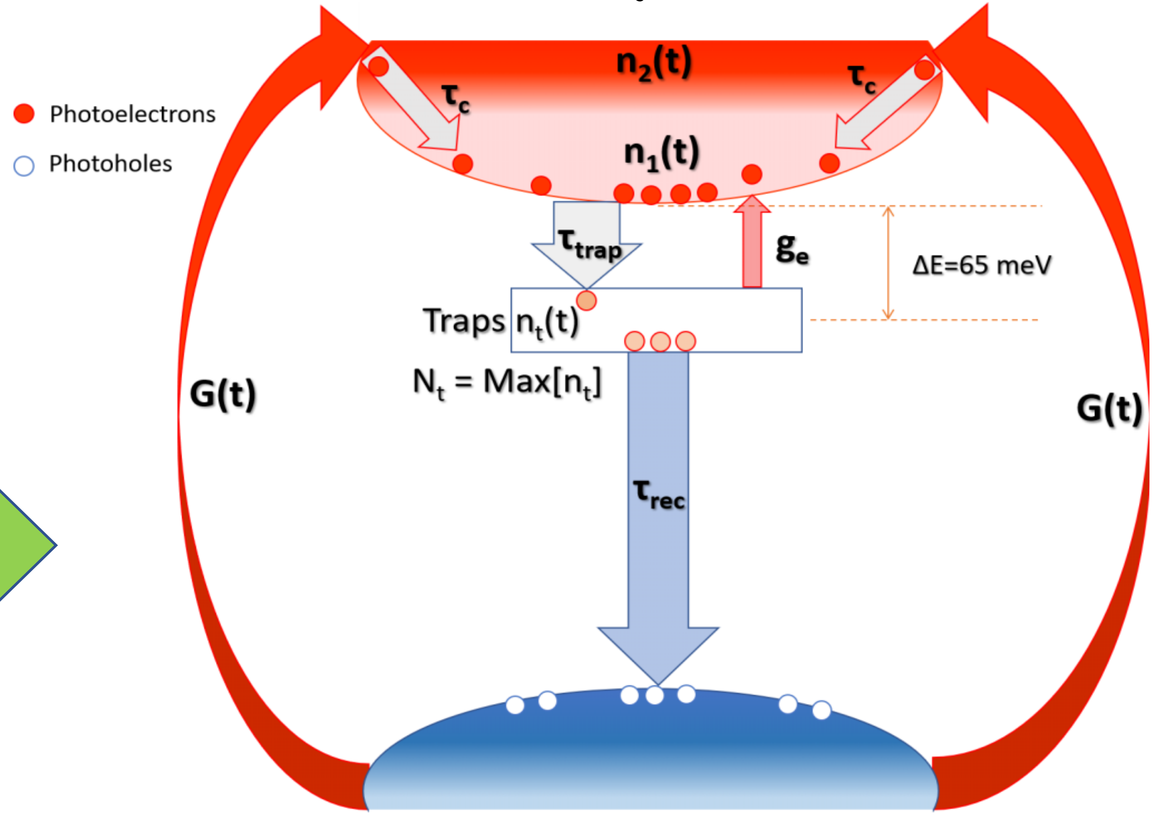
$$\frac{|\Delta T(t)|}{T} = erfc\left(\frac{-t}{\tau_r}\right)\left[a_1 \exp\left(\frac{-t}{\tau_1}\right) + a_2 \exp\left(\frac{-t}{\tau_2}\right)\right]$$

Capture / recombination dynamics

$$\frac{dn_2(t)}{dt} = G(t) - \frac{n_2(t)}{\tau_c},$$

$$\frac{dn_1(t)}{dt} = \frac{n_2(t)}{\tau_c} - \frac{n_1(t)}{\tau_{\mathrm{trap}}}\left(1 - \frac{n_t}{N_t}\right) + g_e n_t,$$

$$\frac{dn_t(t)}{dt} = \frac{n_1(t)}{\tau_{\mathrm{trap}}}\left(1 - \frac{n_t}{N_t}\right) - g_e n_t - \frac{n_t}{\tau_{\mathrm{rec}}},$$



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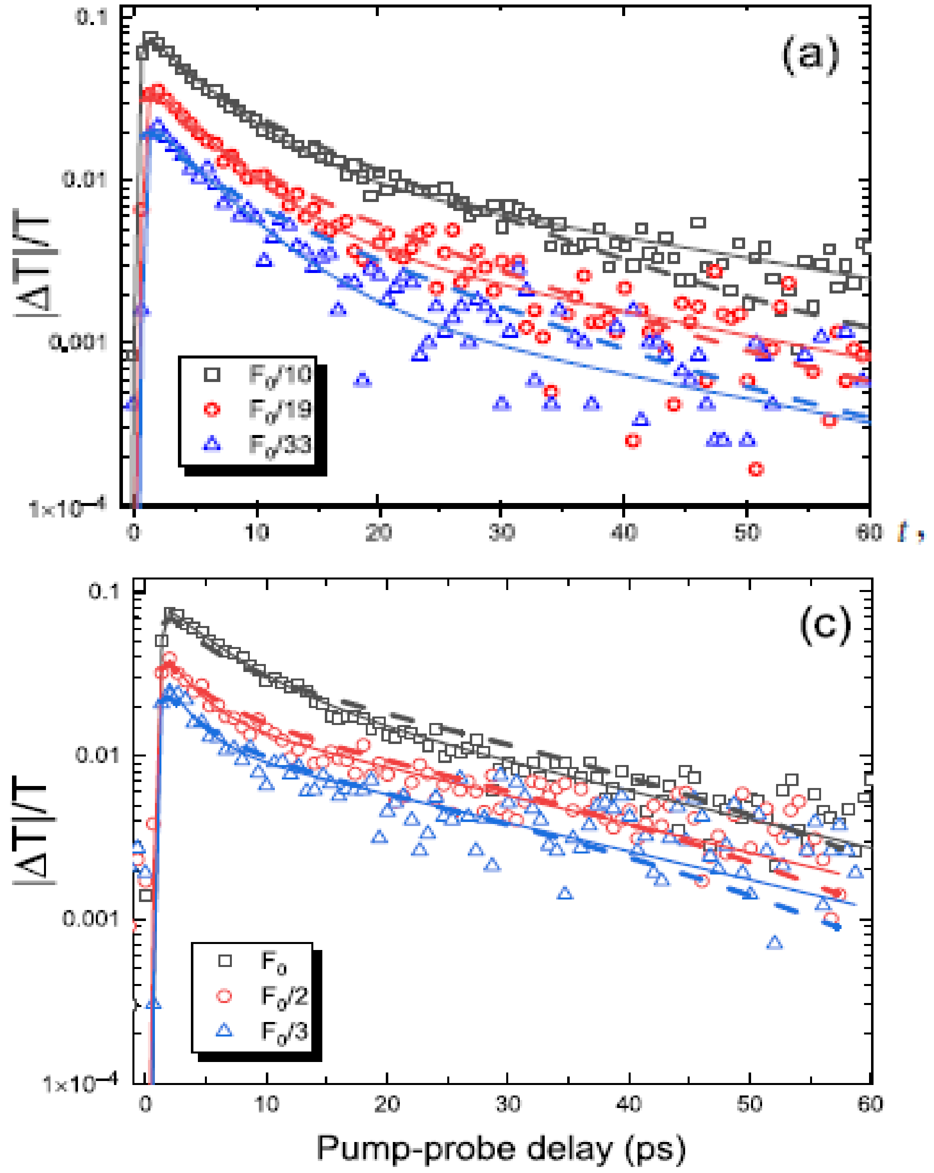
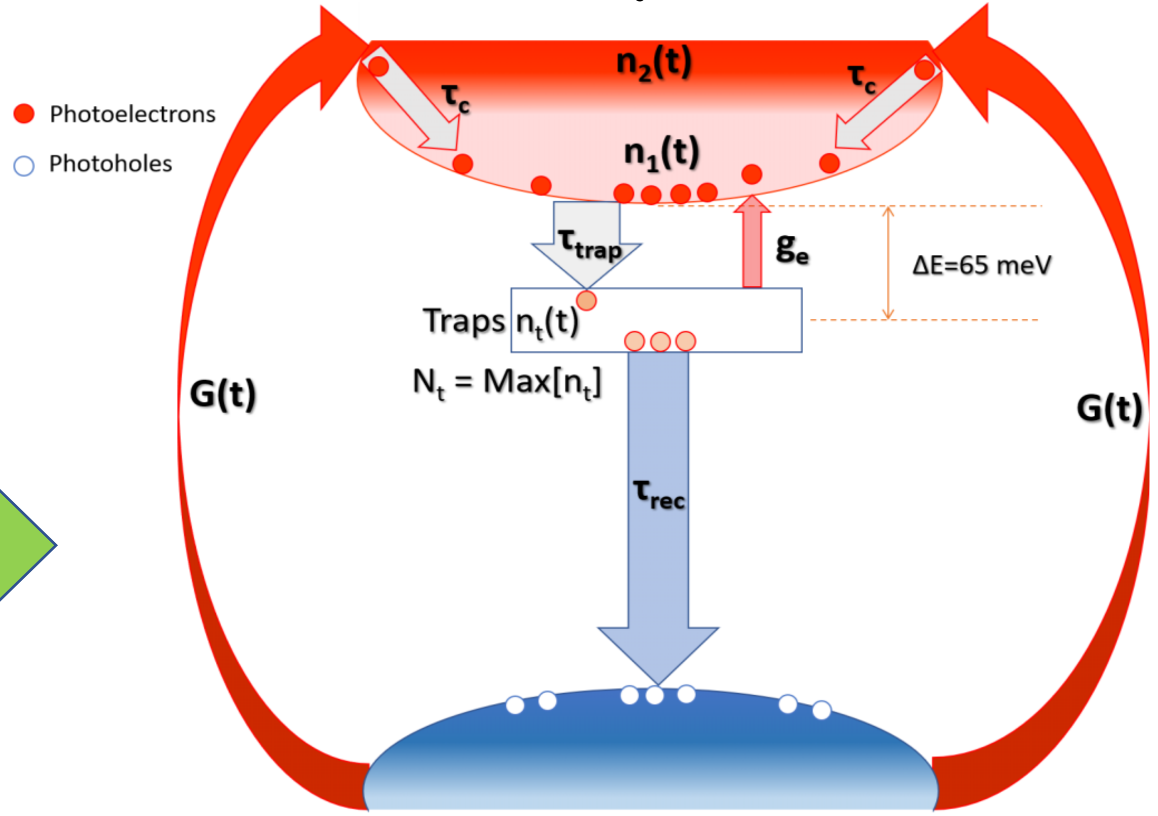
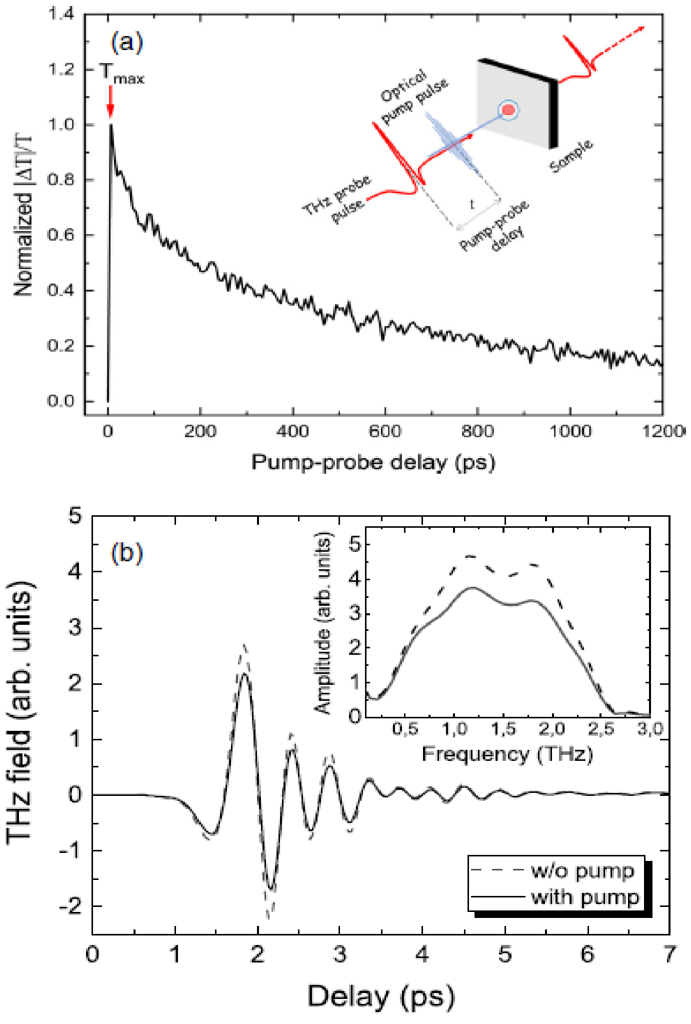




D.J. Jubgang Fandio et al, PRB 102, 115407 (2020)

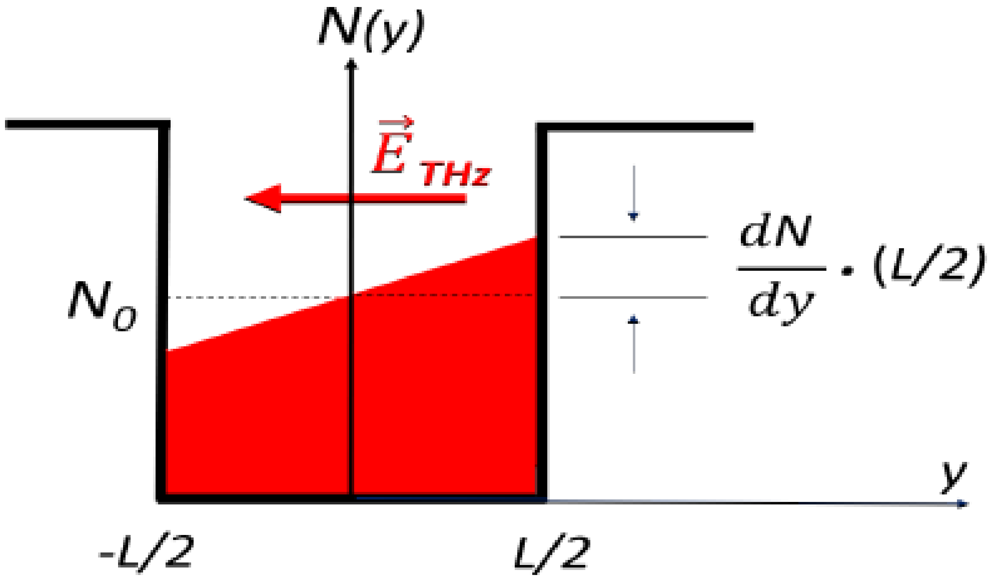
Échantillons	τ_{trap} (ps)	τ_{rec} (ps)	N_t (x 10^{15} cm^{-2})
C750	4,3 ± 0,3	110 ± 36	3
C800	4,1 ± 0,2	100 ± 26	20
C850	4,0 ± 0,1	90 ± 17	20

Frequency Resolved photoconductivity



Δσ(ω) Modified Drude-Smith model

$$\Delta\sigma(\omega) = \frac{Ne^2\tau'/m^*}{1 - i\omega\tau'}\left(1 + \frac{c}{1 - i\omega/a}\right)$$



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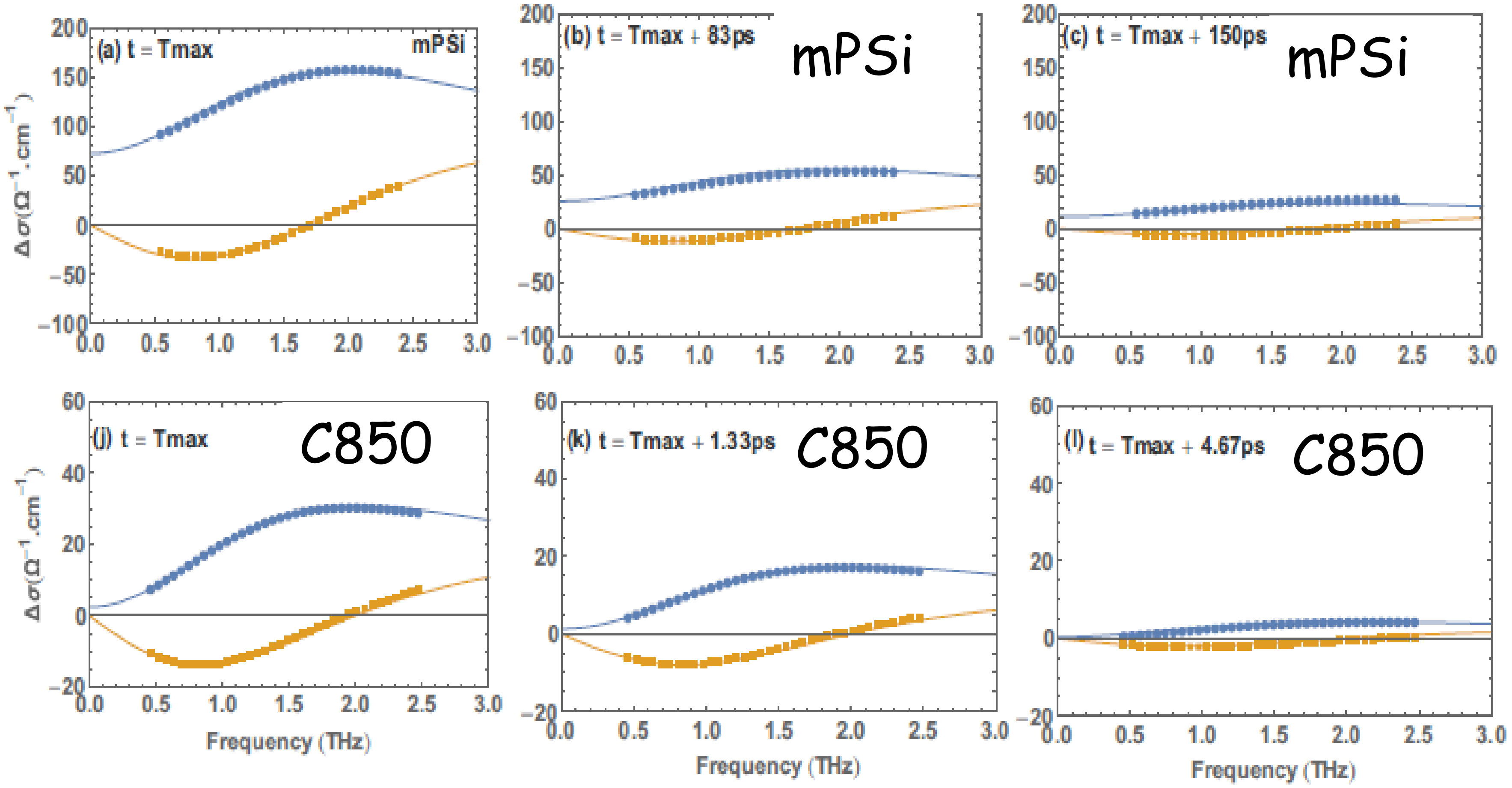
Charge transport properties



Effective mobility preserved ~ 446 cm²/(V.s)

Increase of charge confinement with temperature:

- C = -0.727 for mPSi
- C = -0.956 for C850

D.J. Jubgang Fandio et al, PRB 102, 115407 (2020)

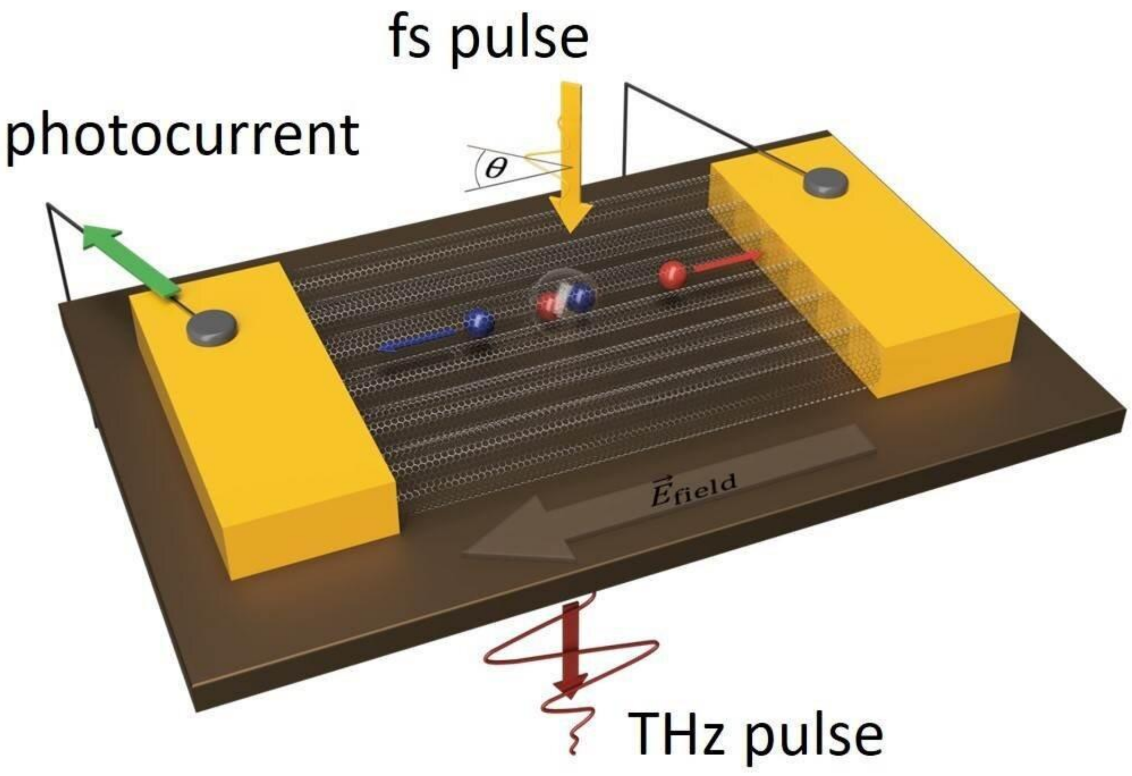
Conclusions

1. Dielectric properties established.
2. Relatively low photocarrier time(5 ps)

3. Confined charge carrier transport at Si/graphene interfaces by electrostatic barriers.

4. Good candidates for broadband THz emitters

Photoconductive antenna



$$\vec{E}_{THz}(z,t) = \frac{1}{4\pi\epsilon_0}\frac{A}{c^2 z}\frac{\partial \vec{J}(t)}{\partial t},$$
$$= \frac{Ae}{4\pi\epsilon_0 c^2 z}\frac{\partial N(t)}{\partial t}\mu \vec{E}_b,$$

Bagsican et al., Nanoletters, 2020,20,53098-3105

Acknowledgement

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THANKS YOU LORD JESUS