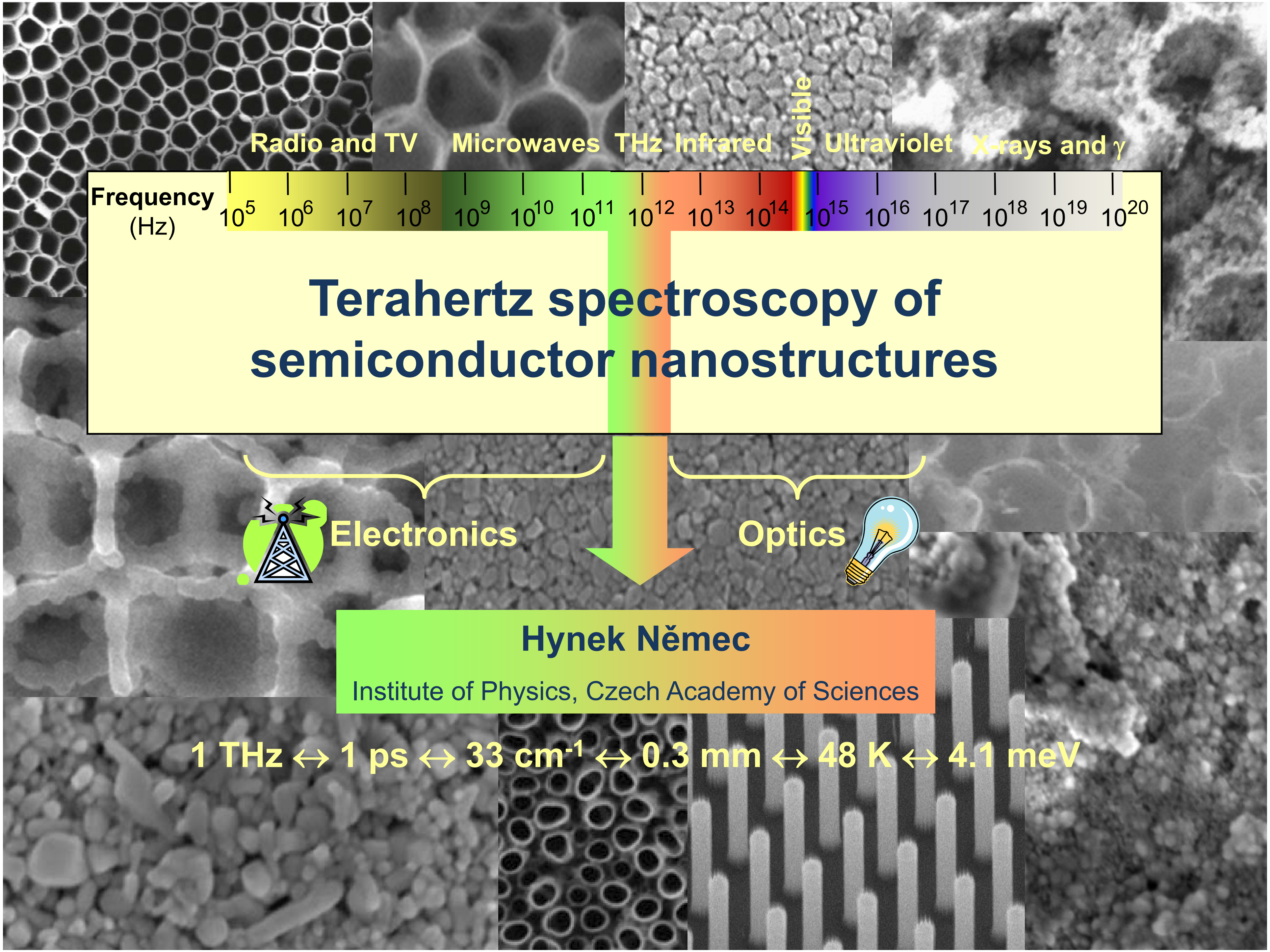


Terahertz spectroscopy of semiconductor nanostructures

Hynek Němec

Institute of Physics, Czech Academy of Sciences

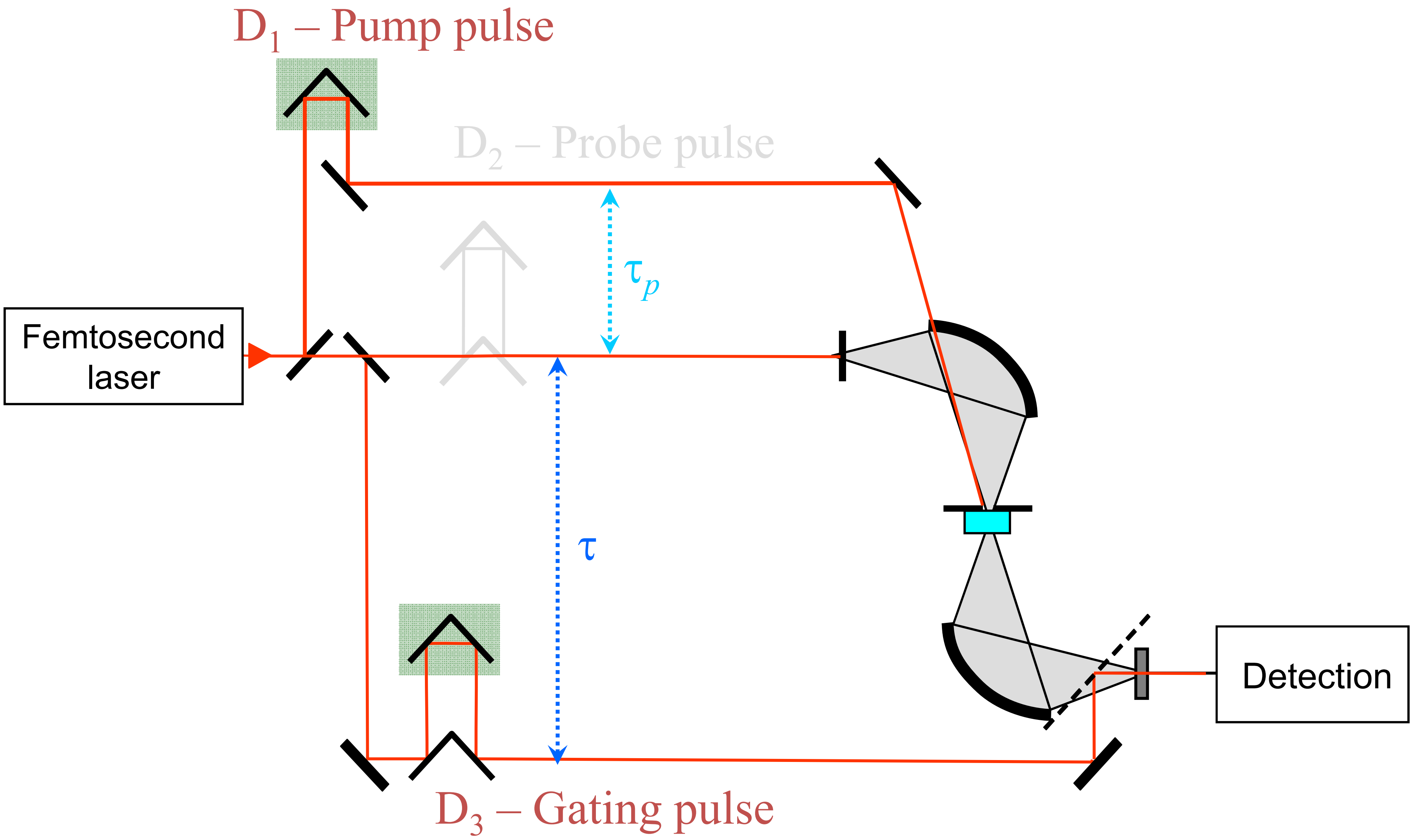
1 THz ↔ 1 ps ↔ 33 cm^{-1} ↔ 0.3 mm ↔ 48 K ↔ 4.1 meV

Outline

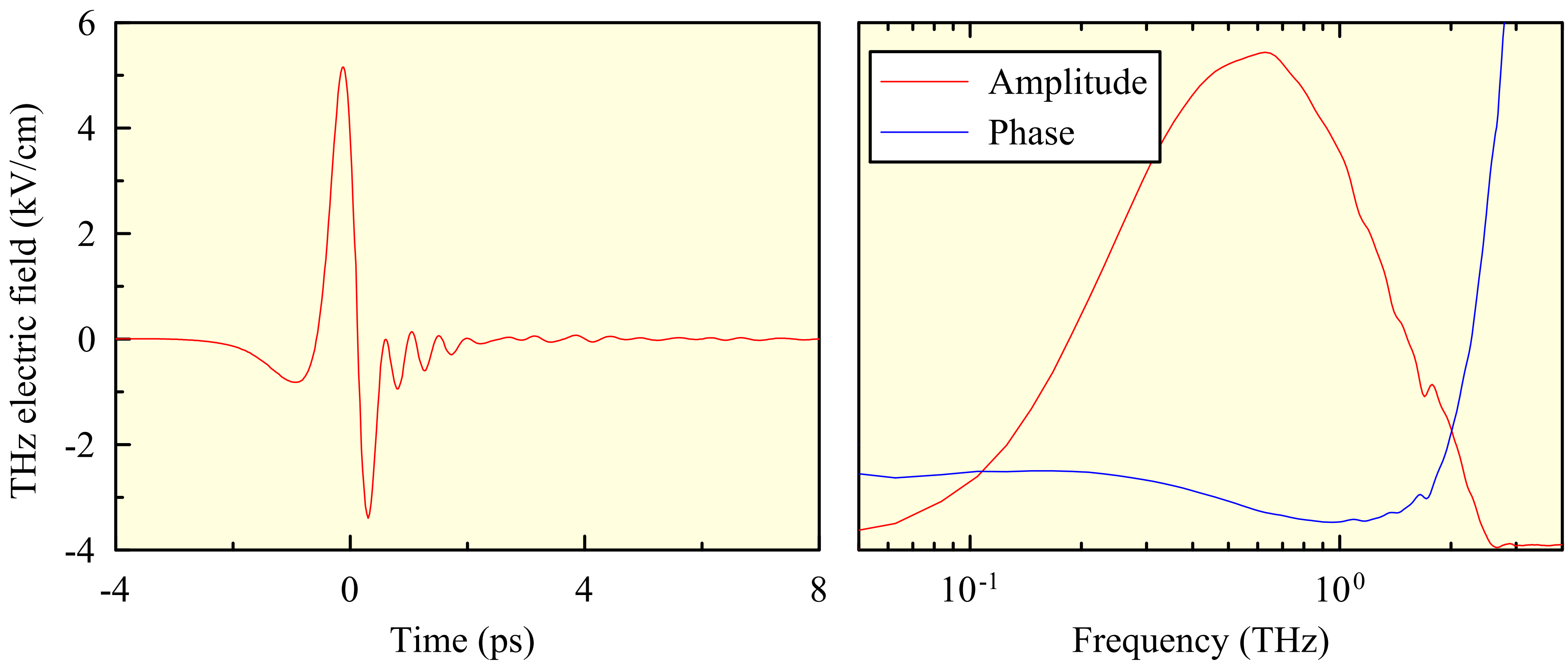
- Time-domain THz spectroscopy
  - Broadband THz pulses propagating in a free space
- Terahertz response of electrons/holes confined in semiconductor nanostructures ... to the electric field
  - Microscopic conductivity
  - Effective medium approximation (plasmon resonance)
- Experimental results: InP nanowires

Time-domain terahertz spectroscopy

Time-domain terahertz spectroscopy

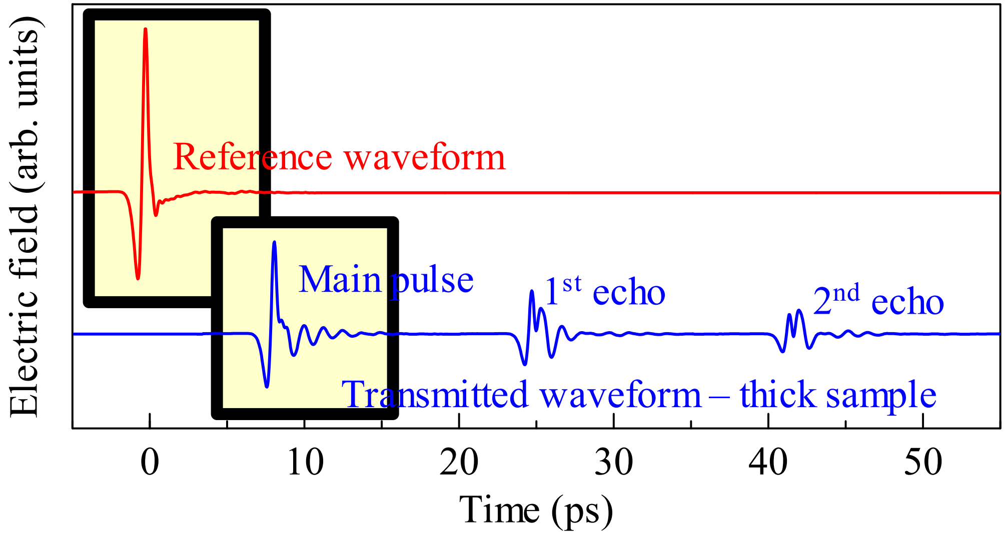


Time-domain terahertz spectroscopy



Time-domain terahertz spectroscopy

- Retrieval of material parameters

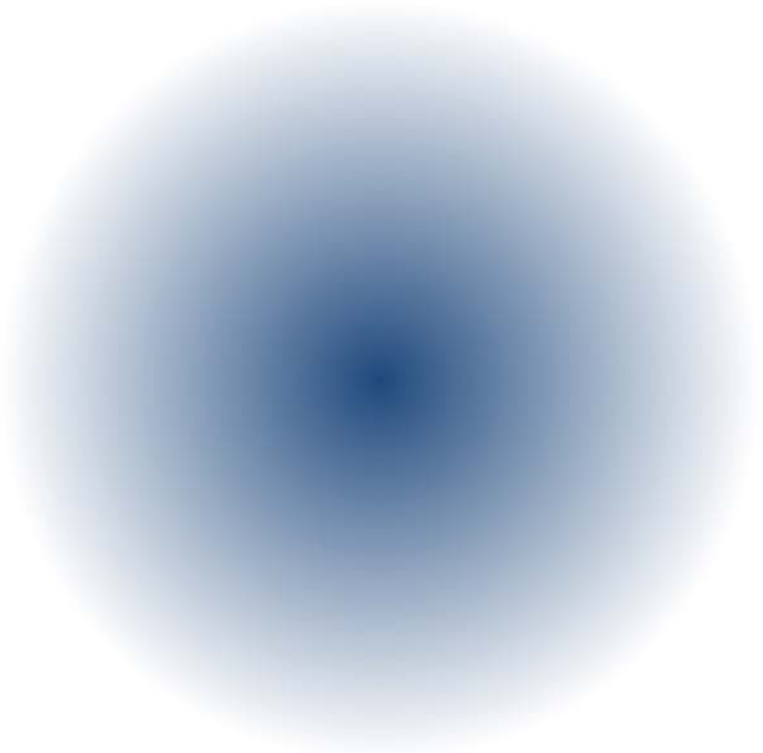


  - Time delay = 2π(*n* – 1)*d*/*c* → refractive index *n*
  - Attenuation = exp(–2πκ*d*/*c*) → absorption index κ

- Equivalence $(n+i\kappa)^2=\hat{\varepsilon}=\frac{i\hat{\sigma}}{\omega\varepsilon_0}$ → complex permittivity, conductivity or carrier mobility can be retrieved too → “ultrafast Ohm-meter” [T. Seifert]

Symbiosis of Tera- and nano- worlds

Symbiosis of Tera- and nano- worlds



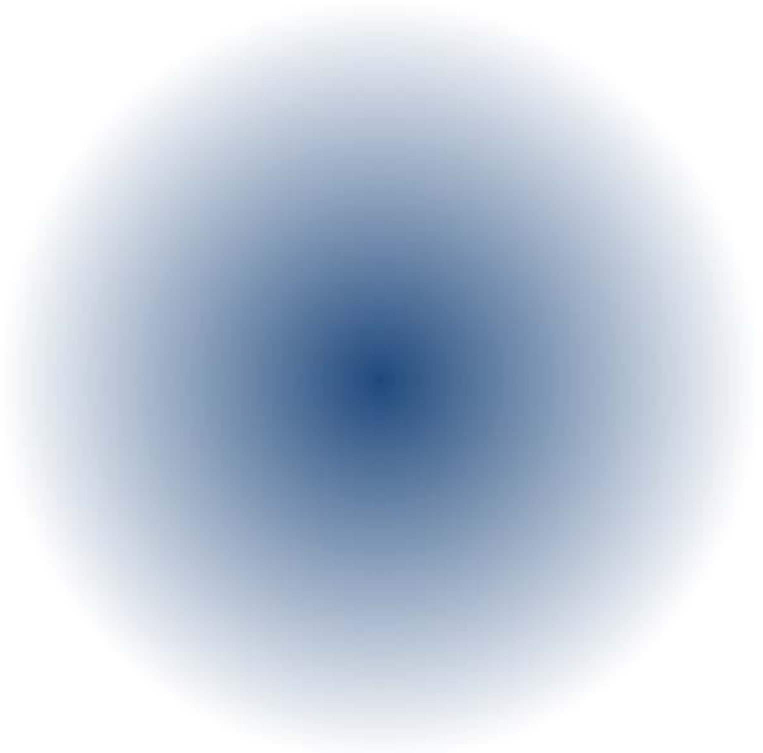
THz beam waist

> 300 μm

Single nanostructure

(< 1 μm)

Symbiosis of Tera- and nano- worlds



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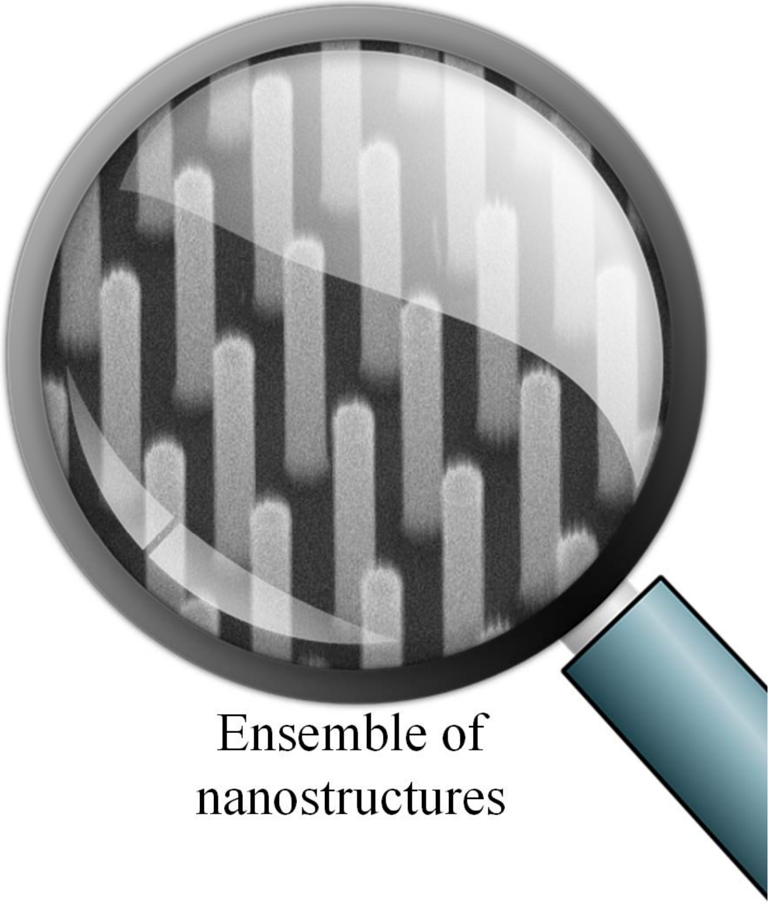
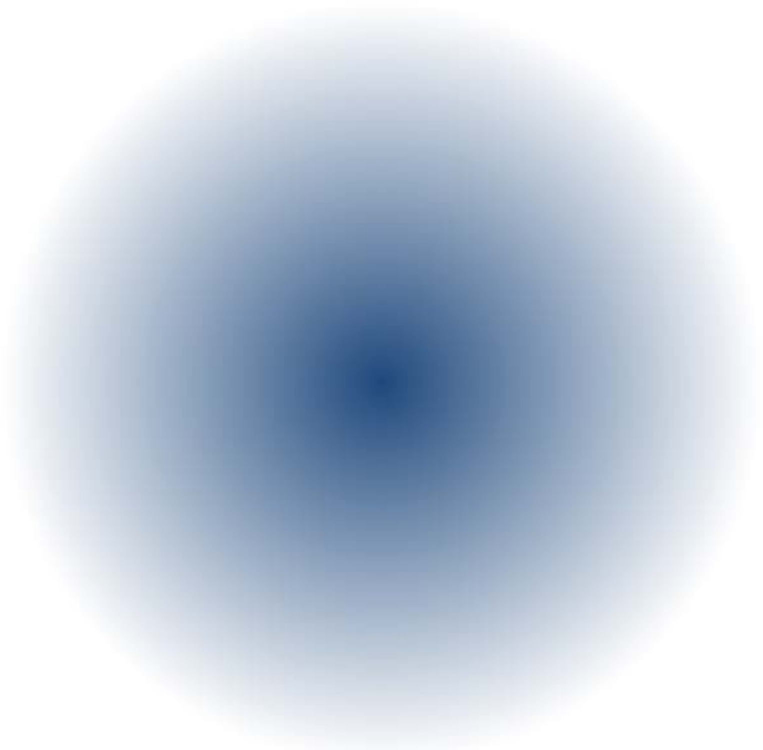


Ensemble of
nanostructures

Symbiosis of Tera- and nano- worlds

Benefit of using free-space radiation: we can probe also coupling among nanoparticles



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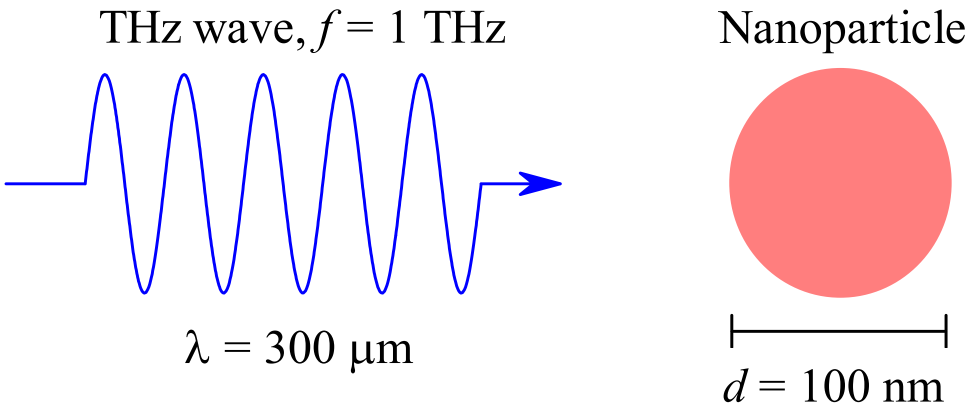
Ensemble of nanostructures

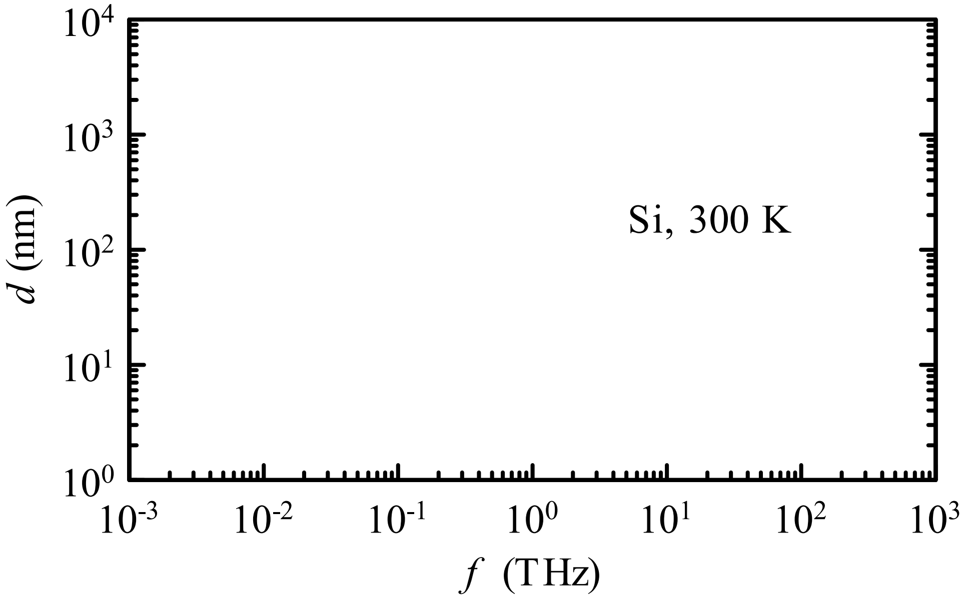
Symbiosis of Tera- and nano- worlds



What is the mobility μ of charges at a particular frequency f?

Symbiosis of Tera- and nano- worlds

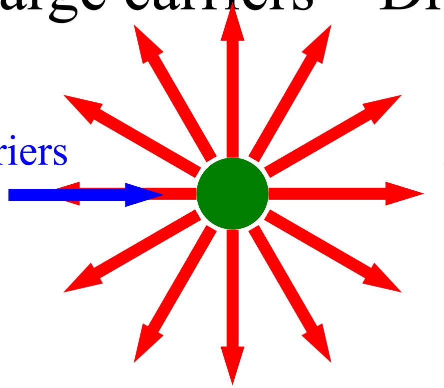
- Frequency *f* versus nanocrystal size *d*



Calculations of conductivity spectra

Models of electric response: Drude

- Response of free charge carriers – Drude model



Velocity of incident charge carriers

Velocity of scattered charge carriers

isotropic scattering

  - Relaxation of charge carrier drift velocity

$$\frac{dv}{dt} = -\frac{v}{\tau} + \frac{e_0 E(t)}{m_{\text{eff}}}$$

  - Mobility spectrum

$$\mu(f) = e_0 v(f) = \frac{e_0}{m_{\text{eff}}} \cdot \frac{\tau}{1 - 2\pi i f \tau}$$

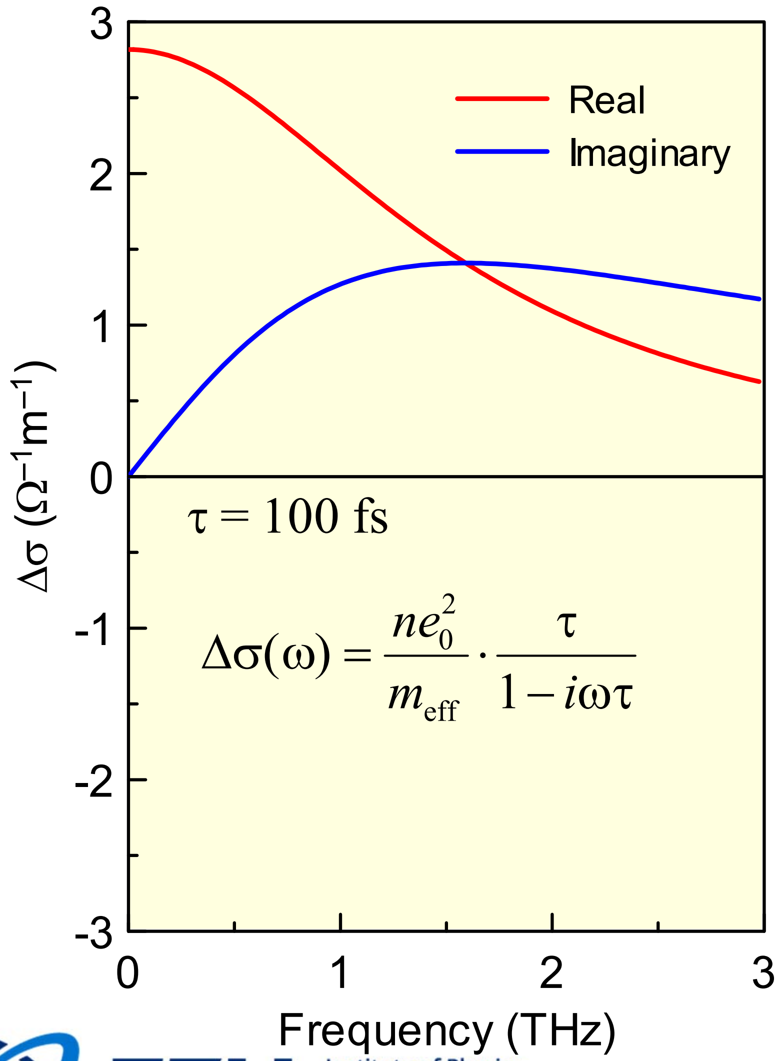
  - DC mobility within the Drude model

$$\mu_{\text{dc}} = \frac{e_0 \tau}{m_{\text{eff}}}$$

τ	Carrier scattering time
m_{eff}	Effective mass of charge carriers
e_0	Elementary charge (1.6×10^{-19} C)

Models of electric response: Drude

- Response of free charge carriers – Drude model



τ = 100 fs

$$\Delta\sigma(\omega) = \frac{ne_0^2}{m_{\text{eff}}} \cdot \frac{\tau}{1 - i\omega\tau}$$

Typical scattering times in semiconductors:

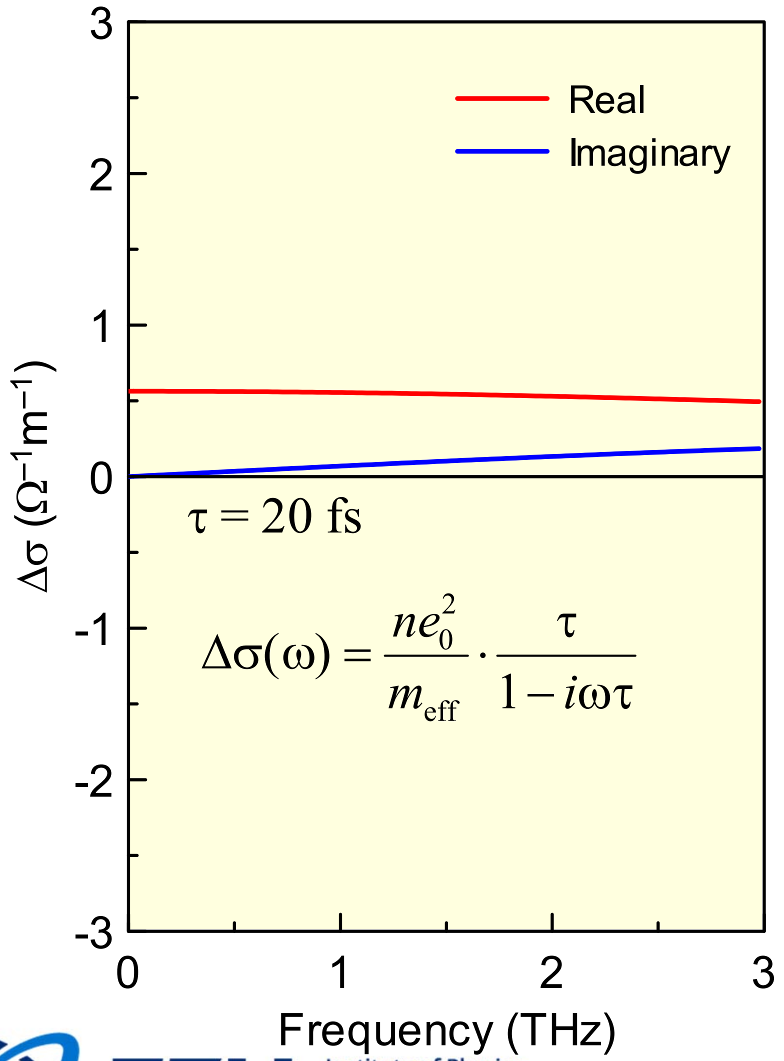
GaAs: $\tau \sim 270$ fs

ZnO: $\tau \sim 30$ fs

Fingerprints just in the terahertz spectral region

Models of electric response: Drude

- Response of free charge carriers – Drude model



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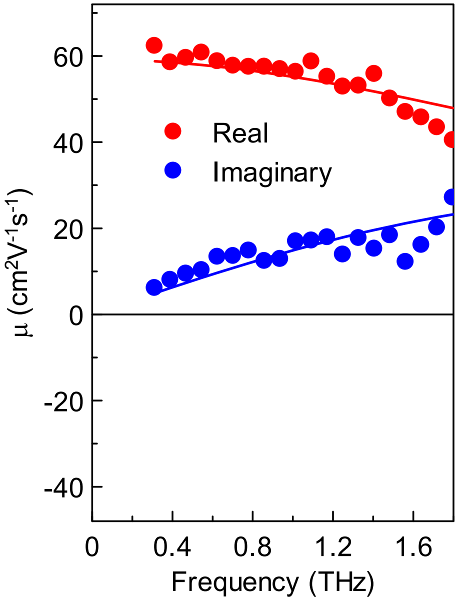
- ZnO crystal (bulk material)



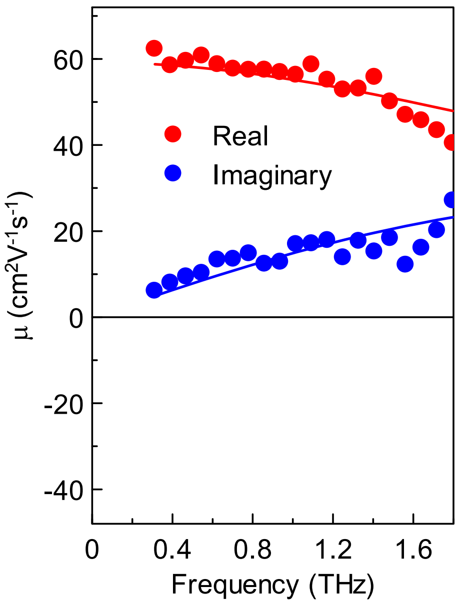
→ Drude behavior

→ Typical free charge carrier response

→ τ ~ 30 fs

Models of electric response

- ZnO crystal (bulk material)



→ Drude behavior

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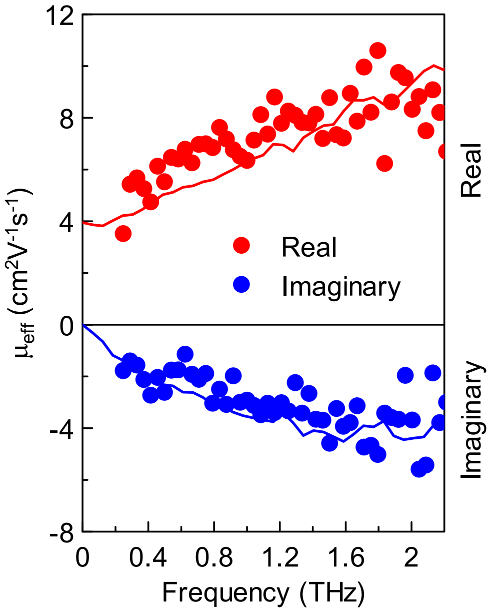
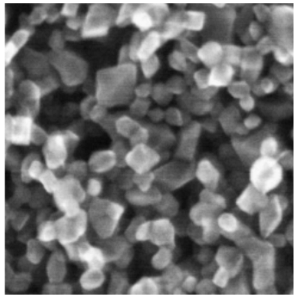
- ZnO nanoparticles





Nanoparticle diameter: ~15 nm

→ Used as electron transporting electrodes in Grätzel solar cells

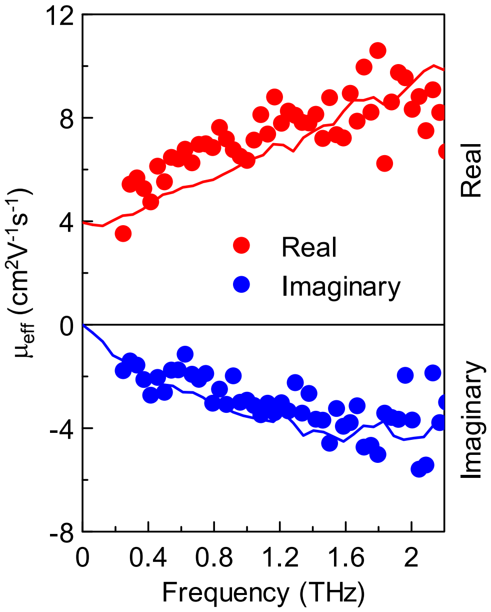
→ There should be free charges

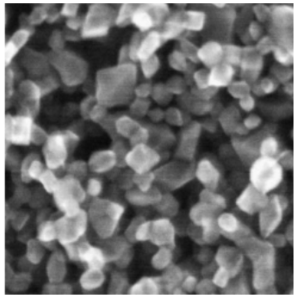
✗ The response is localized!!!

Models of electric response

- Trouble with fitting THz spectra: too many models may be employed
  - ~~Drude~~
  - Drude-Smith
  - Oscillator
  - Hopping
  - Plasmonic model + any response
- List of early works (since 2002): see references in [J. Photochem. Photobiol. A **215**, 123 (2010)]

- ZnO nanoparticles





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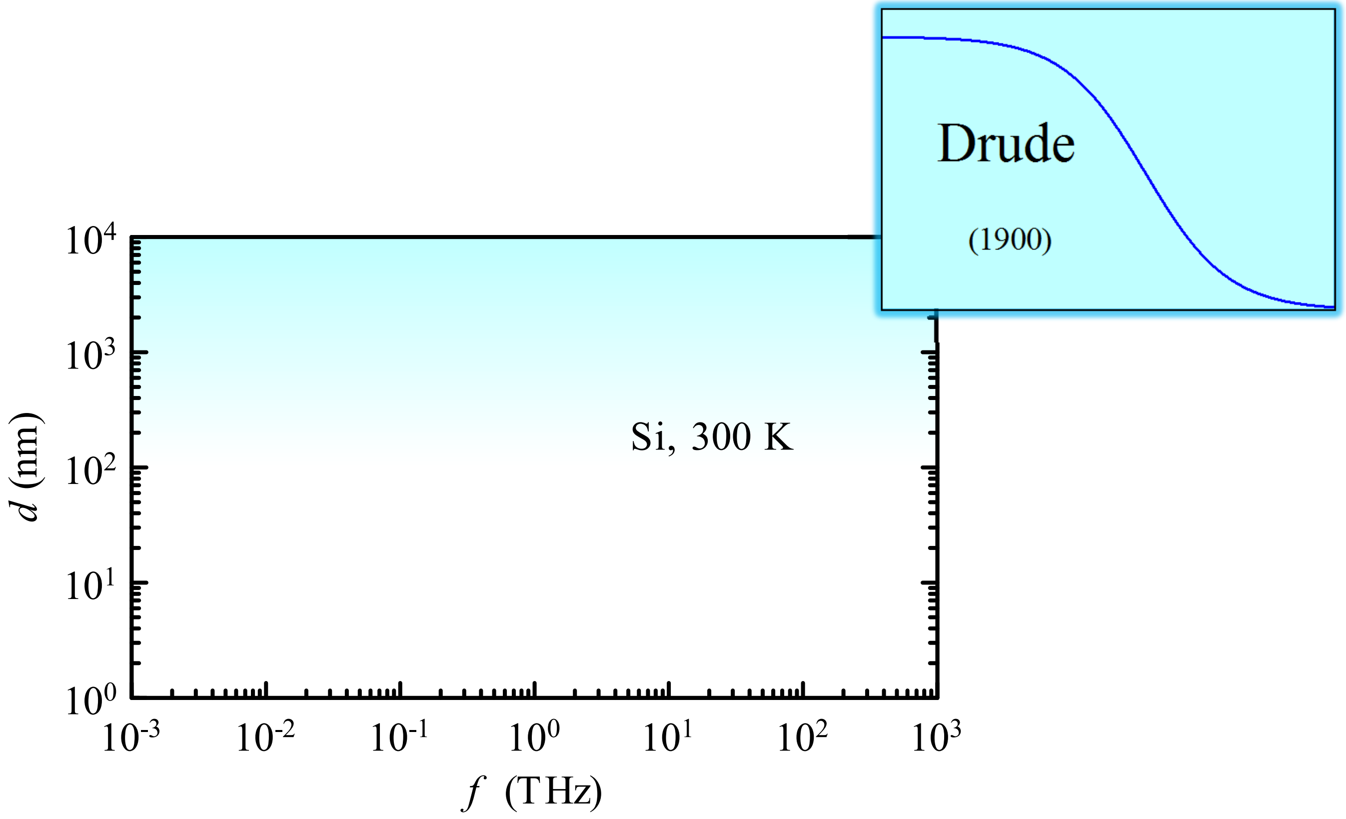

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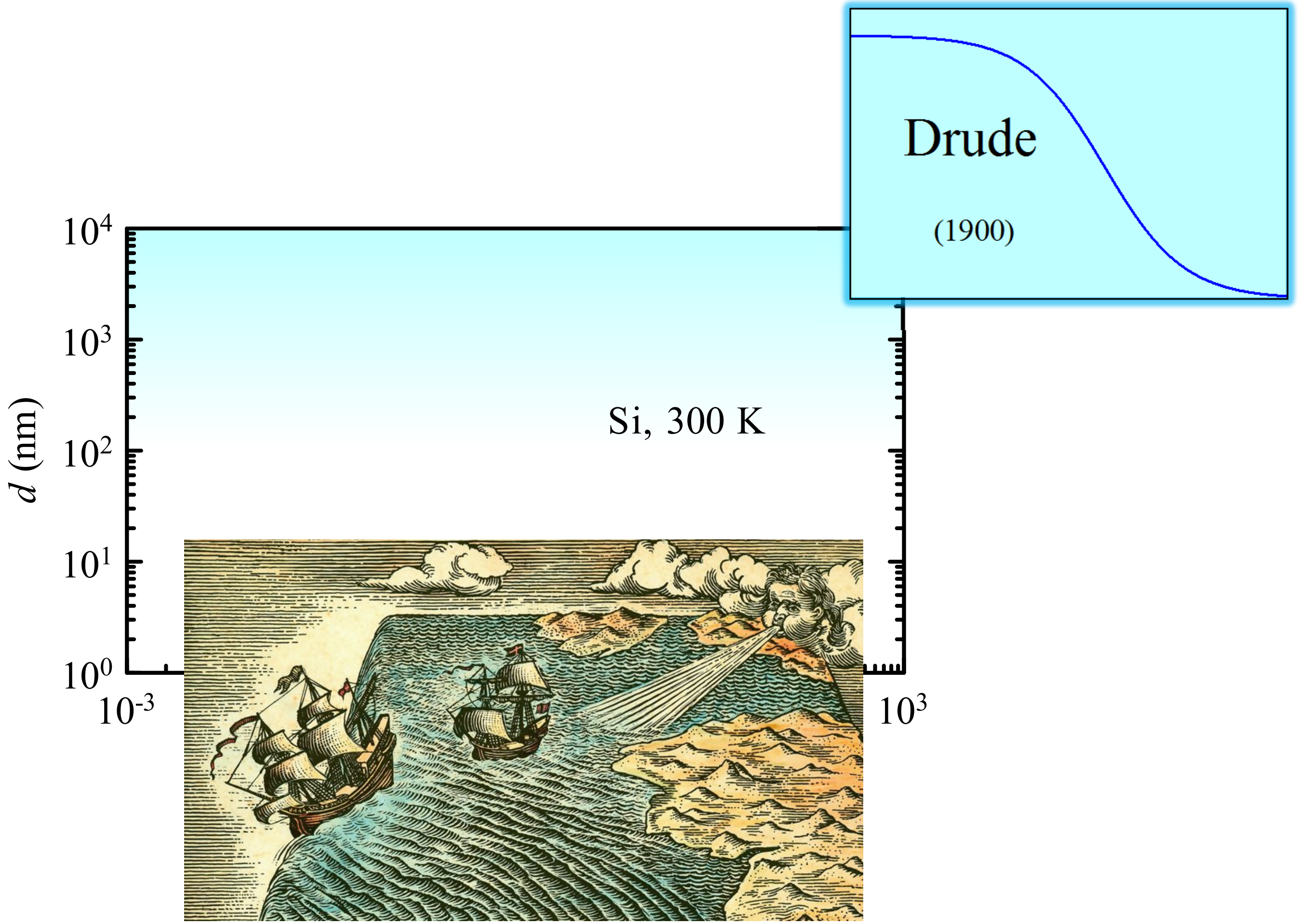
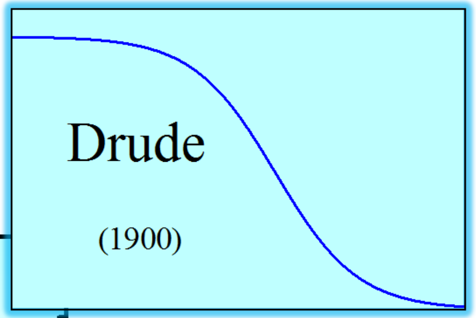


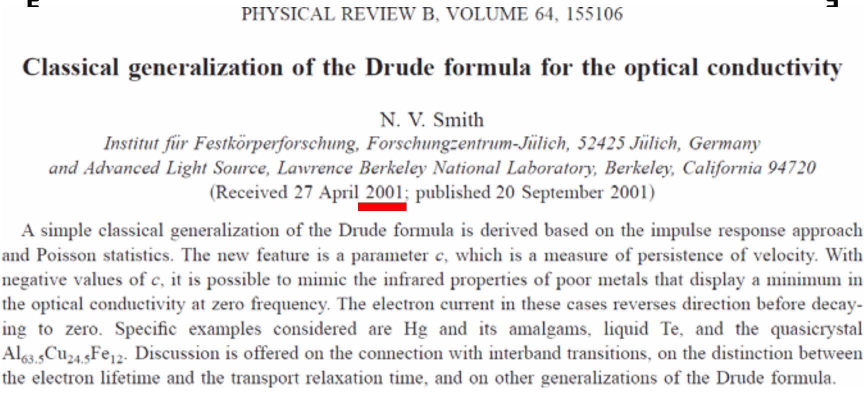
Illustration from https://www.npr.org/2017/11/11/561217363/terra-incognita-the-planet-factory-and-the-undiscovered-islands?t=1557220165821

Models of electric response

- Frequency *f* versus nanocrystal size *d*

$$\Delta\sigma(\omega) = \frac{ne_0^2}{m_{\mathrm{eff}}} \cdot \frac{\tau}{1-i\omega\tau} \cdot \left[1 + \frac{c_1}{1-i\omega\tau}\right]$$





PHYSICAL REVIEW B, VOLUME 64, 155106

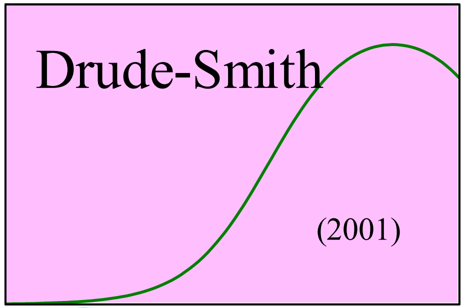
Classical generalization of the Drude formula for the optical conductivity

N. V. Smith

Institut für Festkörperforschung, Forschungzentrum-Jülich, 52425 Jülich, Germany
and Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720

(Received 27 April 2001; published 20 September 2001)

A simple classical generalization of the Drude formula is derived based on the impulse response approach and Poisson statistics. The new feature is a parameter *c*, which is a measure of persistence of velocity. With negative values of *c*, it is possible to mimic the infrared properties of poor metals that display a minimum in the optical conductivity at zero frequency. The electron current in these cases reverses direction before decaying to zero. Specific examples considered are Hg and its amalgams, liquid Te, and the quasicrystal $Al_{63.5}Cu_{24.5}Fe_{12}$. Discussion is offered on the connection with interband transitions, on the distinction between the electron lifetime and the transport relaxation time, and on other generalizations of the Drude formula.



Models of electric response

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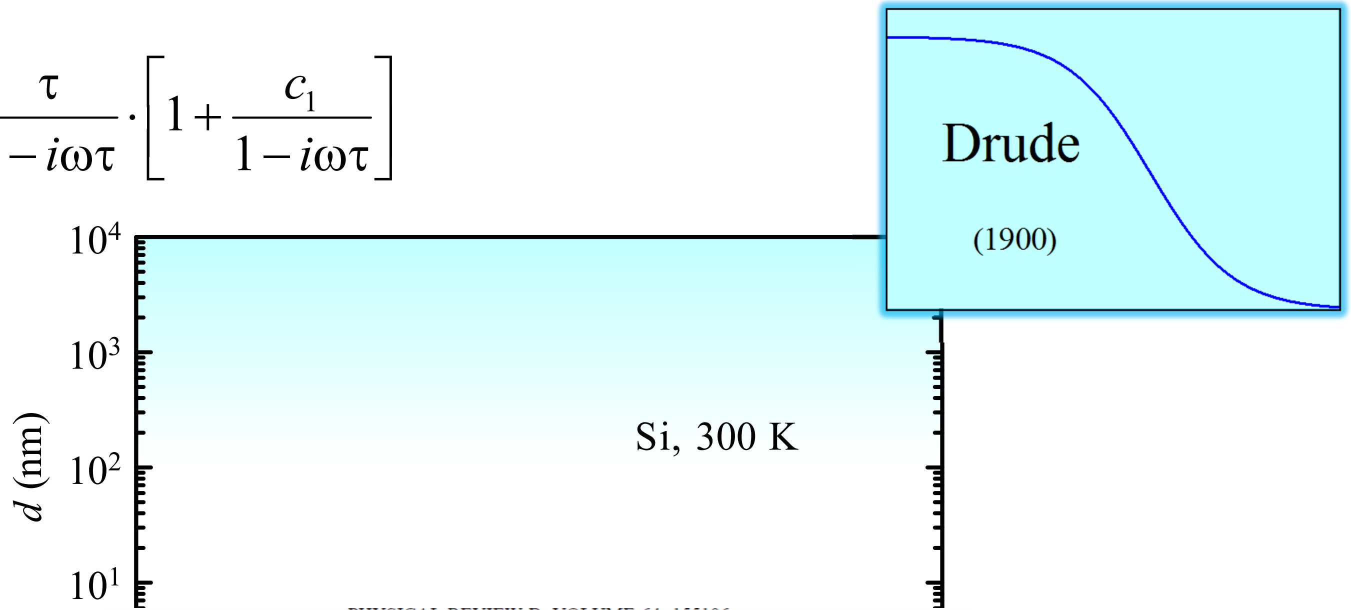

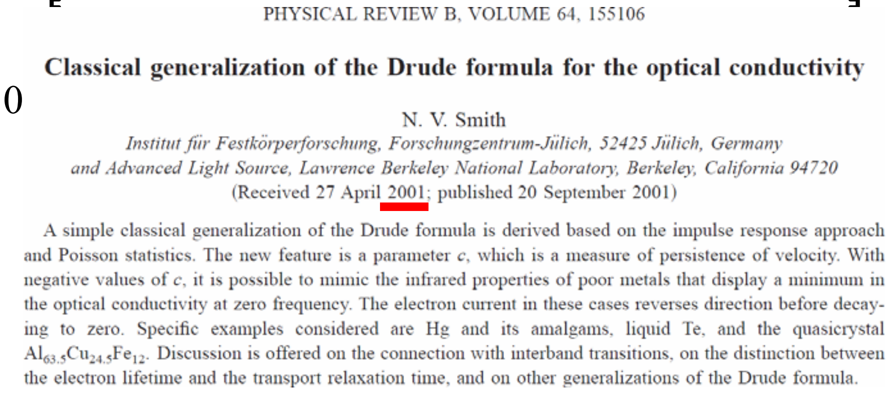

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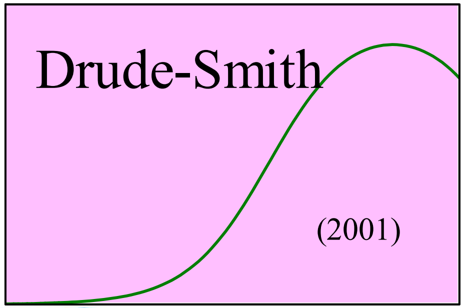
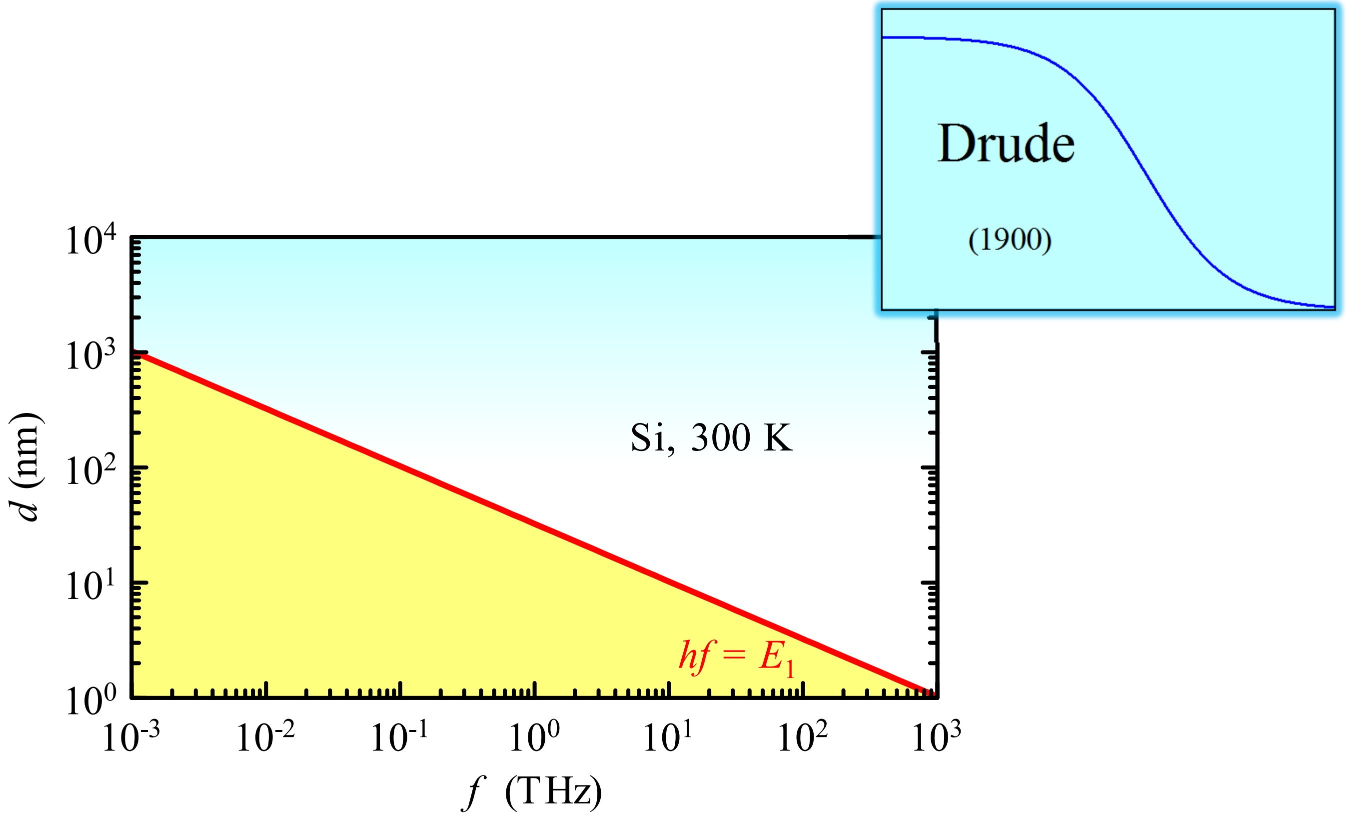


Deficiencies

- No *d*-dependence
- Unphysical

Models of electric response

- Lowest quantum transition hf vs. $E_1 \propto d^{-2}$



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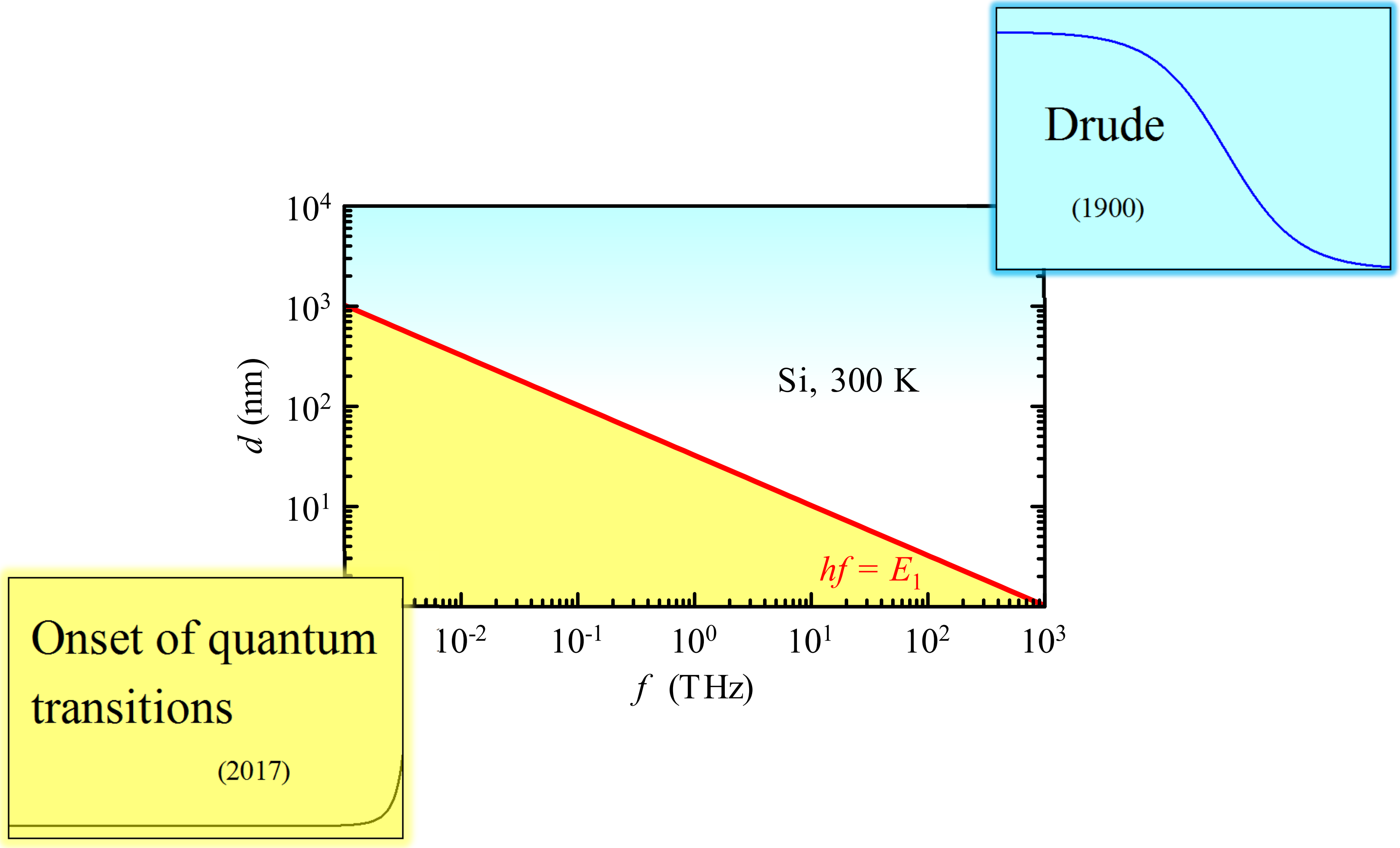

Models of electric response: Quantum calculations

- General Kubo formula

$$\sigma = \frac{iNe_0^2}{m\omega} + \frac{1}{\hbar\omega V}\int_0^\infty e^{i\omega t}\mathrm{Tr}(\rho_0[J(t),J(0)])\mathrm{d}t$$

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- Simplifications
  - Single-particle approximation
  - Relaxation time approximation

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$$\mathrm{Re}\,\sigma \propto \sum_{k,l}(f_k - f_l)\frac{\left|\langle l|p|k\rangle\right|^2}{(\omega+\omega_k-\omega_l)^2+\gamma^2}$$

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- Problem: non-zero DC conductivity in isolated nanoparticles

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- Simplifications
  - Single-particle approximation
  - **Relaxation time approximation**

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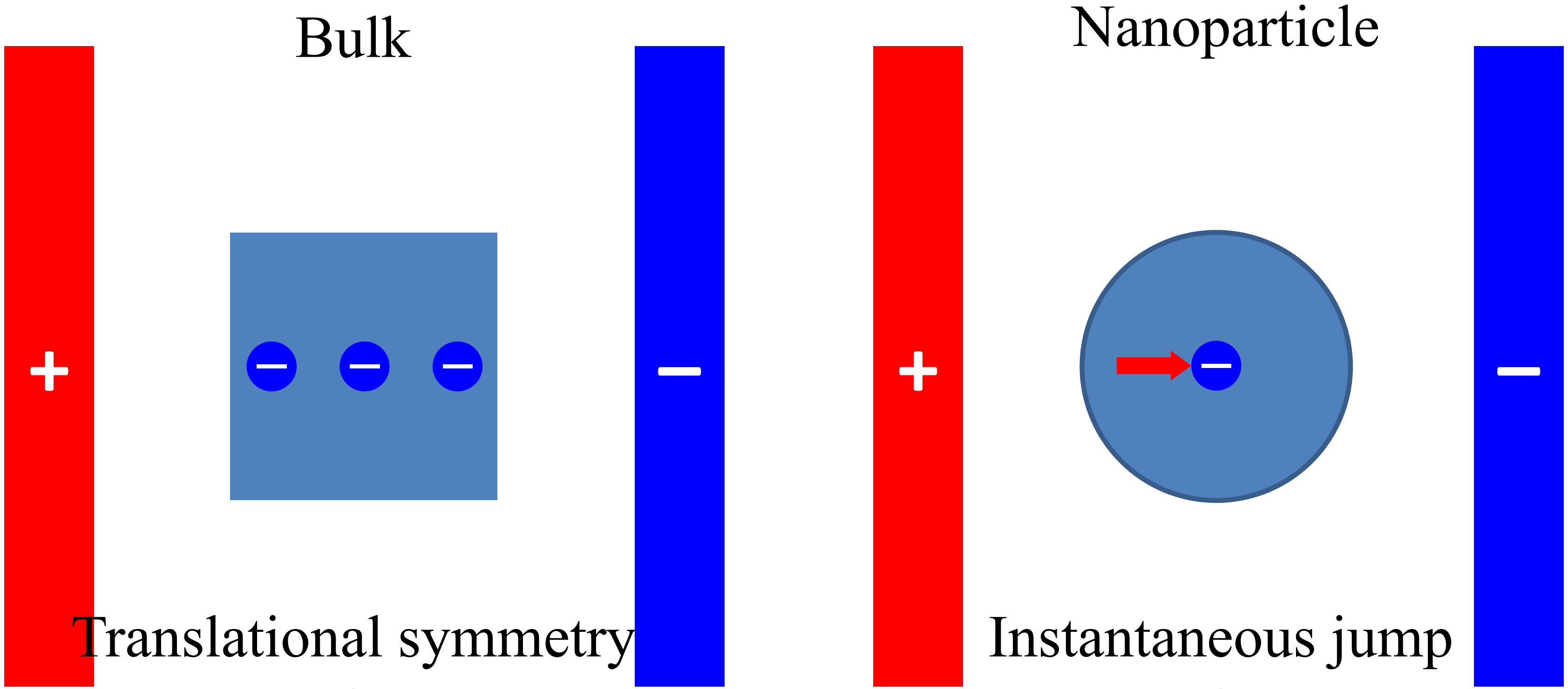
- Problem: non-zero DC conductivity in isolated nanoparticles

Quantum calculations – relaxation time approximation

- A charge displaced by the probing electric field relaxes back to its equilibrium position



Translational symmetry

↓

No net displacement upon relaxation

Instantaneous jump

↓

Extra current

Models of electric response: Quantum calculations

- Three phases of charge motion
  - Equilibrium

$$\hat{H} = \hat{H}_0 = \frac{\hat{\mathbf{p}}^2}{2m} + V(\hat{\mathbf{r}}) \quad \Rightarrow \quad \text{Density matrix } \rho_0 = \sum_k |k\rangle f_k \langle k|$$

  - Coherent regime

$$\hat{H} = \hat{H}_0 \underbrace{- e\hat{\mathbf{r}} \cdot \mathbf{E}}_{\Delta\hat{H}} \quad \Rightarrow \quad \frac{d\Delta\rho}{dt} = -\frac{i}{\hbar}[\Delta\hat{H}, \rho_0] - \frac{i}{\hbar}[\hat{H}_0, \Delta\rho] - \gamma\Delta\rho$$

Models of electric response: Quantum calculations

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  - Thermalization regime (diffusion equation with source)

$$\frac{dn_{\mathrm{th}}}{dt} = \gamma\sum_{k,l}\langle\mathbf{r}|k\rangle\langle k|\Delta\rho|l\rangle\langle l|\mathbf{r}\rangle \underbrace{+ D\nabla^2 n_{\mathrm{th}} + \nabla\cdot\frac{n_{\mathrm{th}}\nabla V}{m\gamma}}_{-\nabla\cdot\mathbf{j}_{\mathrm{th}}}$$

T. Ostatnický et al., PRB **97**, 085426 (2018)

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- Total current = coherent + thermalization

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Czech Academy of Sciences

T. Ostatnický et al., PRB **97**, 085426 (2018)

Models of electric response: Quantum calculations

- Example: cube-shaped nanocrystals

$$\sigma \propto \sum_{k,l} \frac{f_k - f_l}{\omega - (\omega_k - \omega_l) + i\gamma} \left[\sum_{n\,\mathrm{odd}} \frac{2D\gamma a S_{kln} x_{kl}}{D\pi^2 n^2 - i\omega a^2} - i(\omega_k - \omega_l) | x_{kl} |^2 \right]$$

D	Diffusion coefficient
a	Nanocrystal size
x_{kl}	Dipole matrix element $\langle k \mid x \mid l \rangle$
f_j	Occupation of the state j

$$S_{kln} = \delta_{k_y,l_y} \delta_{k_z,l_z} (\delta_{n,k_x-l_x} + \delta_{n,l_x-k_x} - \delta_{n,k_x+l_x})$$

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https://lts.fzu.cz/

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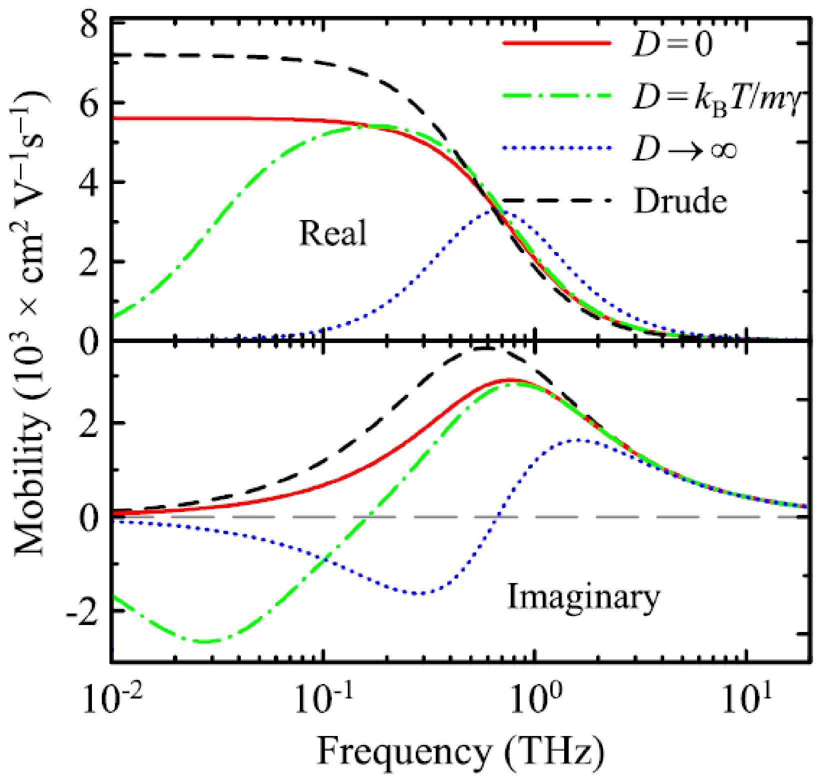

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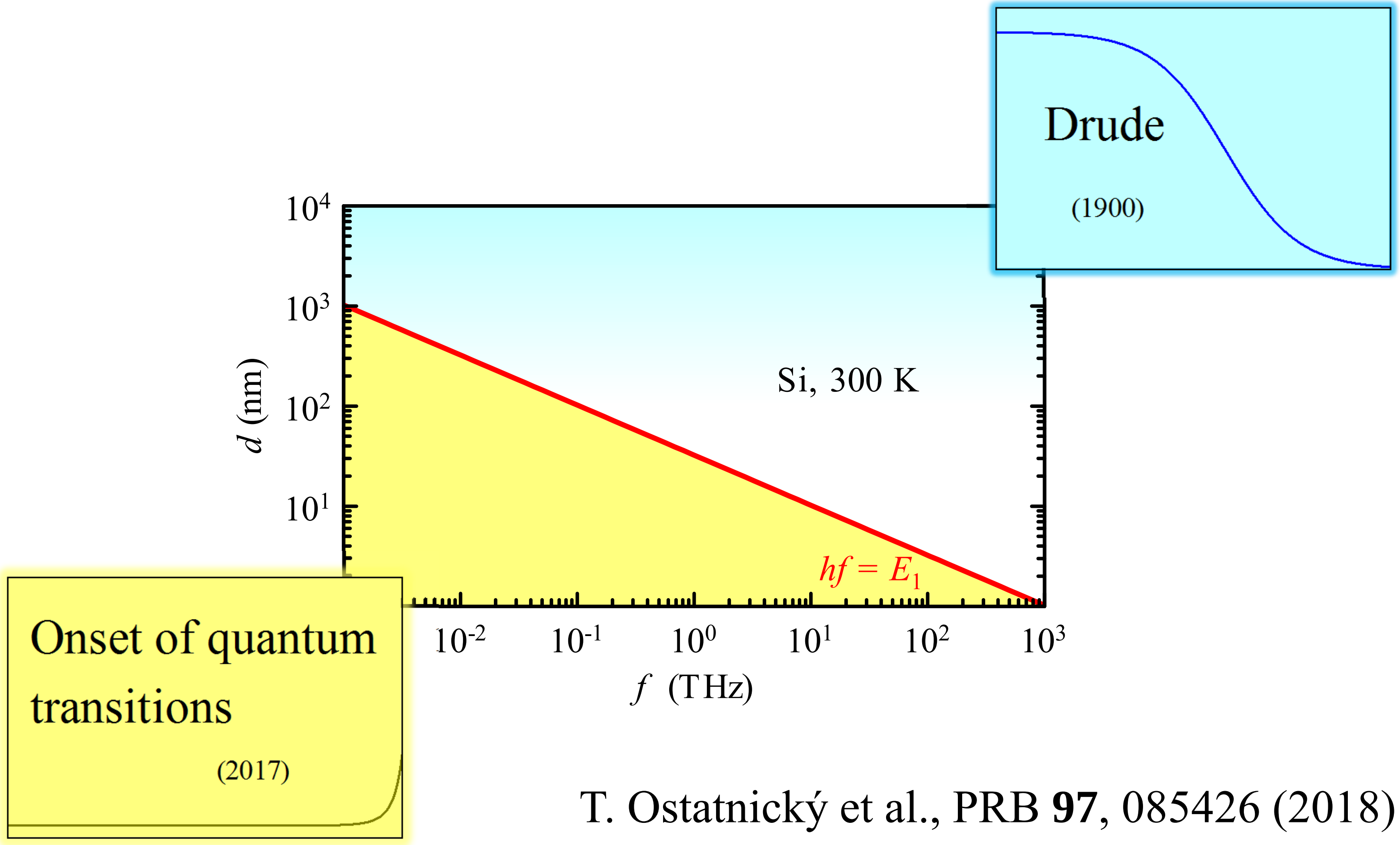
Large GaAs crystallites

$a = 1$ μm
$\gamma^{-1} = 270$ fs
$D = 180$ cm^2/s
$T = 300$ K
$n = 10^{16}$ cm^{-3}

Models of electric response

- Lowest quantum transition hf vs. $E_1 \propto d^{-2}$

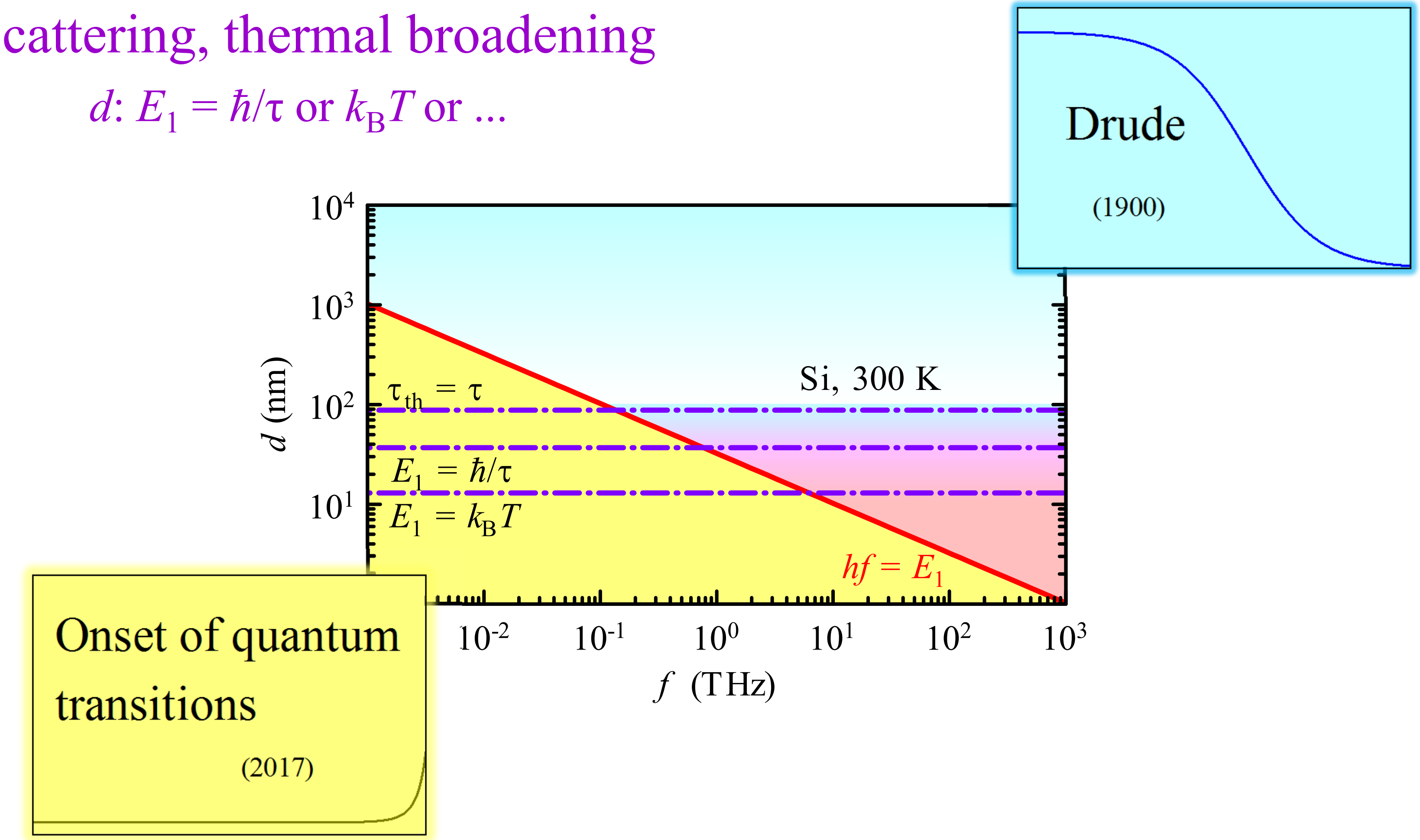


T. Ostatnický et al., PRB **97**, 085426 (2018)

Models of electric response

- Lowest quantum transition hf vs. $E_1 \propto d^{-2}$
- Scattering, thermal broadening

d: $E_1 = \hbar/\tau$ or $k_\mathrm{B}T$ or ...



Drude

(1900)

Onset of quantum transitions

(2017)

Models of electric response

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- Scattering, thermal broadening

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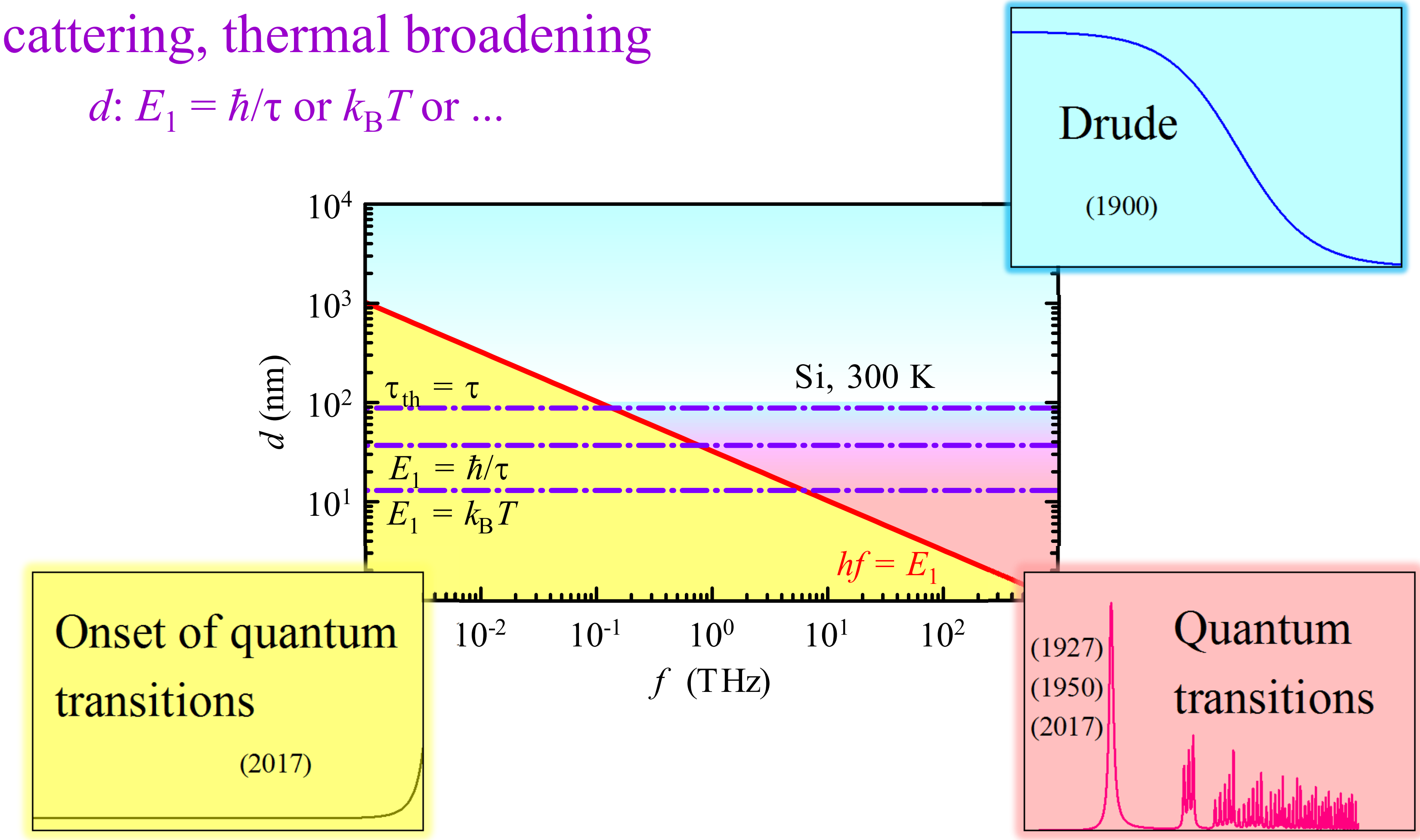
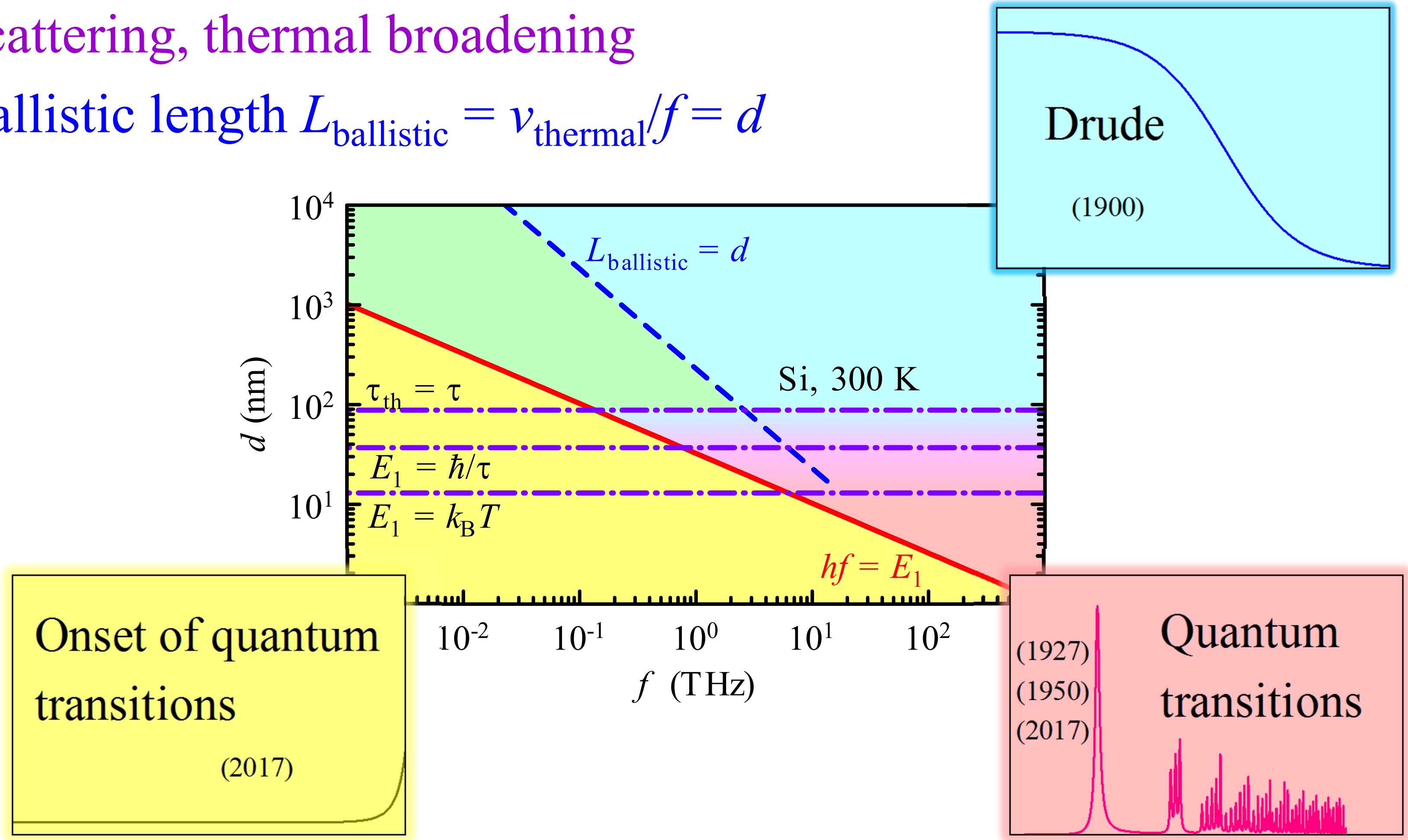


Models of electric response

- Lowest quantum transition hf vs. $E_1 \propto d^{-2}$
- Scattering, thermal broadening
- Ballistic length $L_{\text{ballistic}} = v_{\text{thermal}}/f = d$



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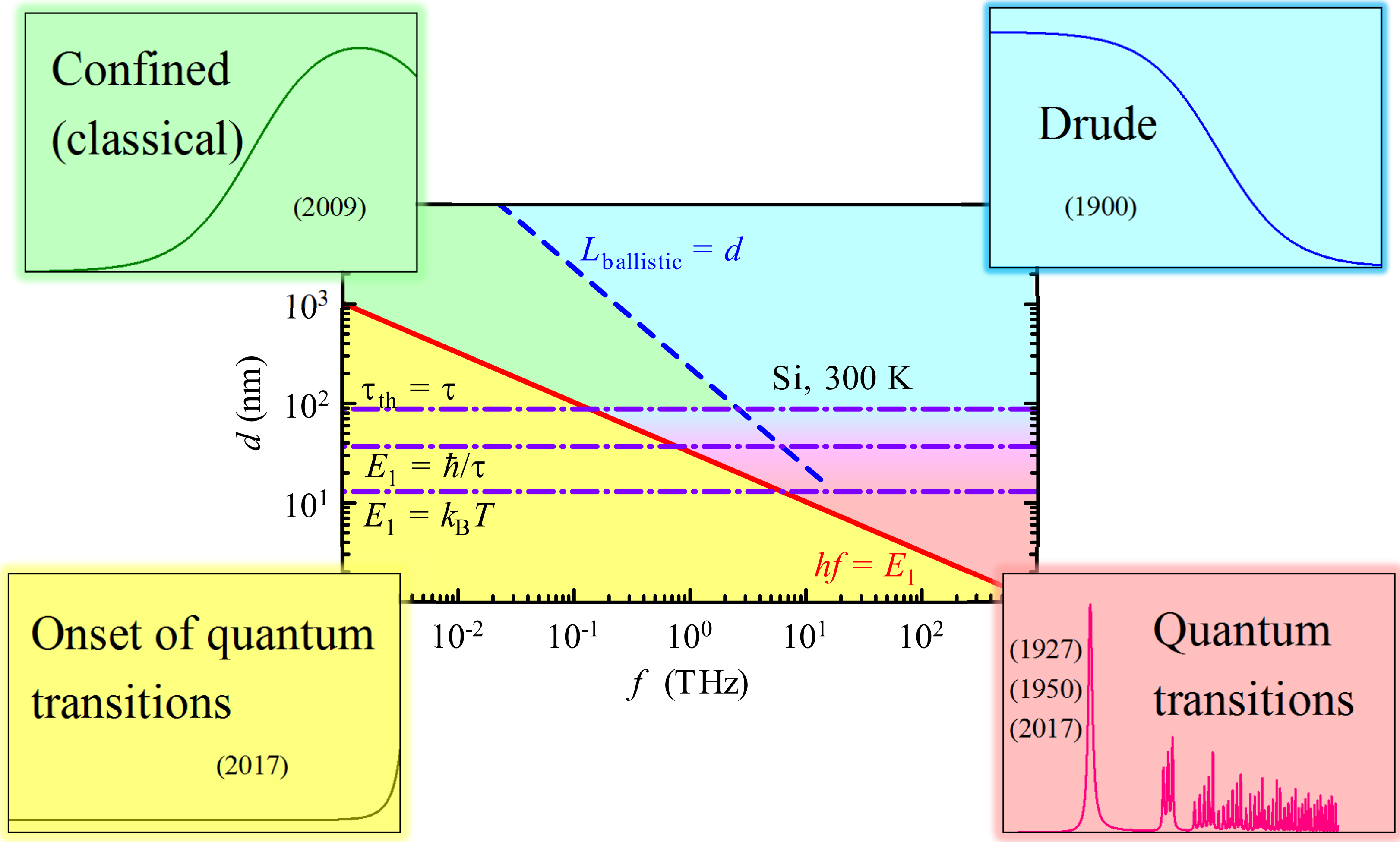
Models of electric response



Models of electric response: Monte-Carlo calculations

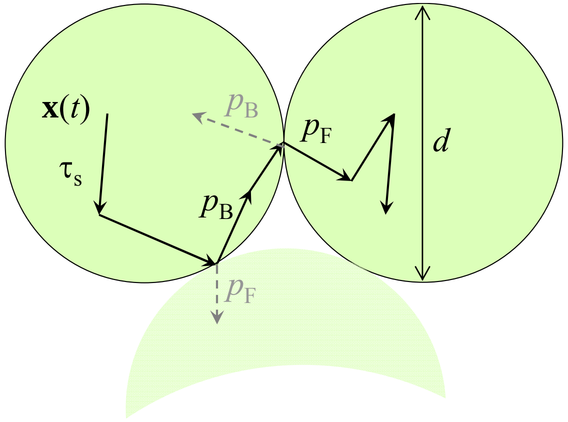
- Models of intrinsic conductivity
  - Starting point: Kubo formula (thermal motion gives the response function; no external field required for the simulations)

$$\mu(\omega) = \frac{e_0}{k_\mathrm{B} T} \int_0^\infty \left\langle v(t) v(0) \right\rangle \exp(\mathrm{i}\omega t)\, \mathrm{d}t$$

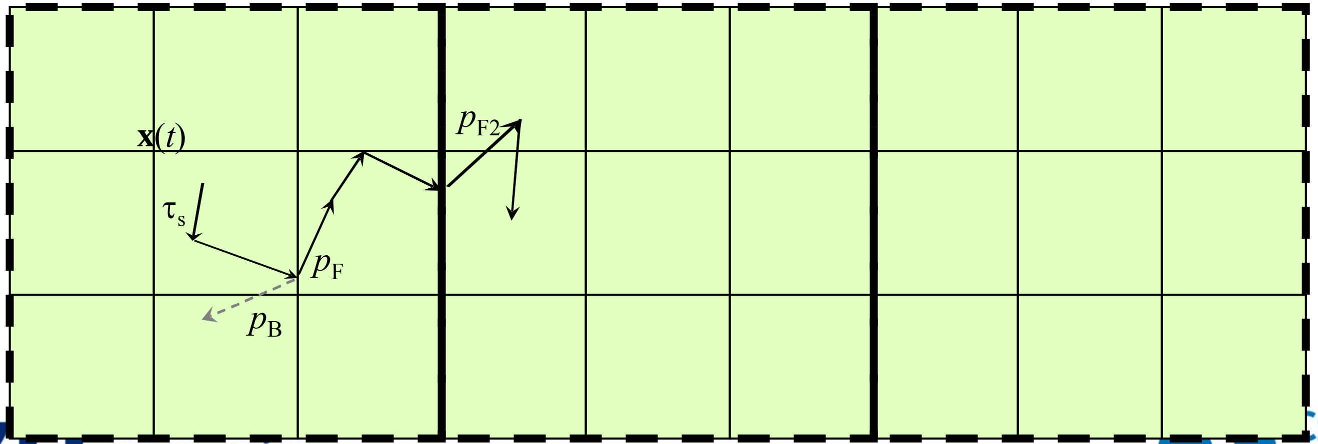
  - Parameters
    - Temperature T
    - (Position of the Fermi level)
    - Effective mass, scattering time
  - Semi-classical Monte Carlo simulations
    - Evolution of (thermal) velocity $v(t)$ of charge carriers with the statistical distribution f
    - Maxwell-Boltzmann distribution works for high temperatures and low excitation densities
    - Fermi-Dirac distribution needs to be considered otherwise

Models of electric response: Monte-Carlo calculations

- Band-like transport confined by nanoparticle surface



- More sophisticated models including e.g. two types of surfaces
  - Possibility to investigate aggregates of nanoparticles



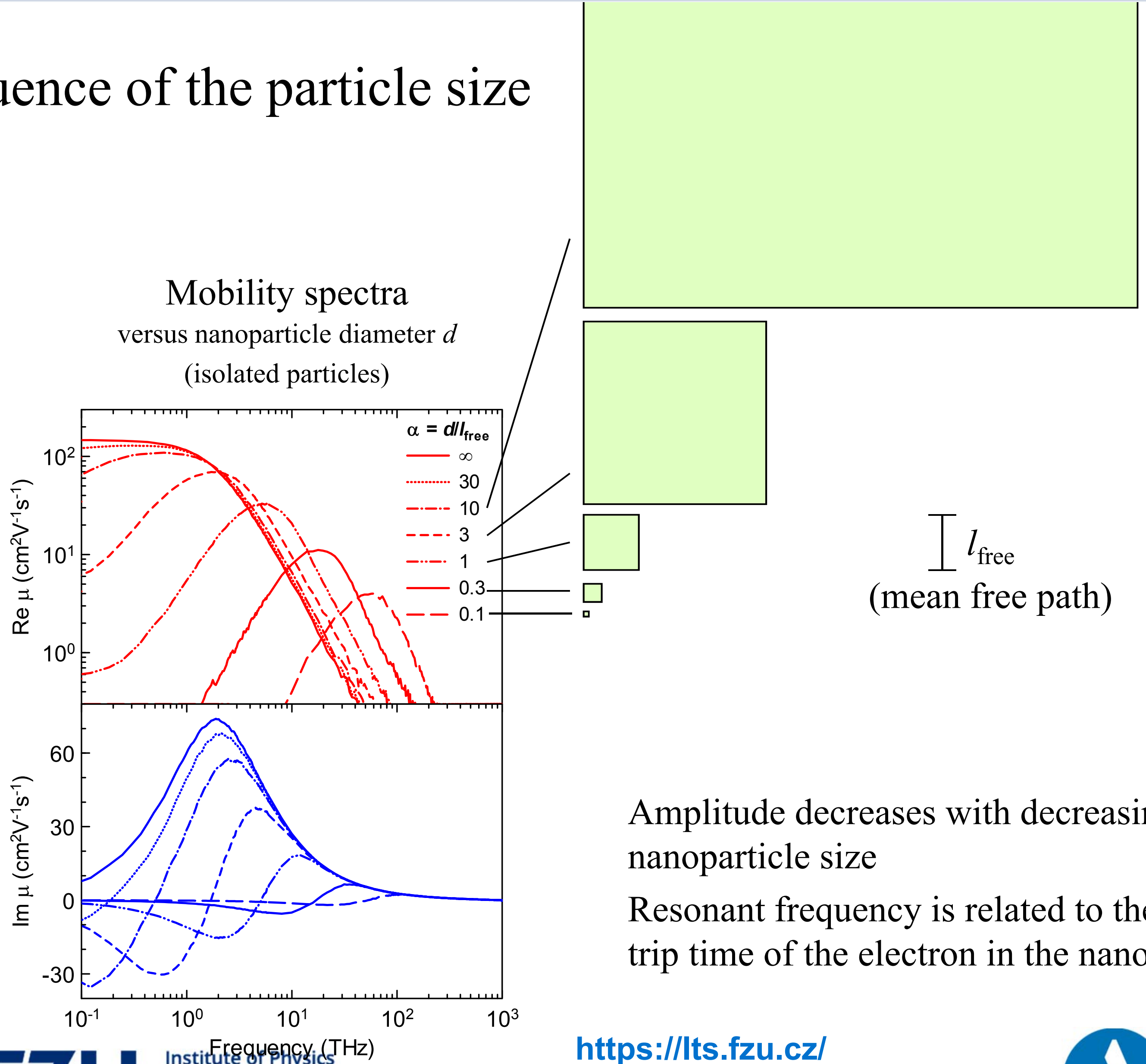
Models of electric response: Monte-Carlo calculations

- Influence of the particle size

Mobility spectra

versus nanoparticle diameter *d*

(isolated particles)



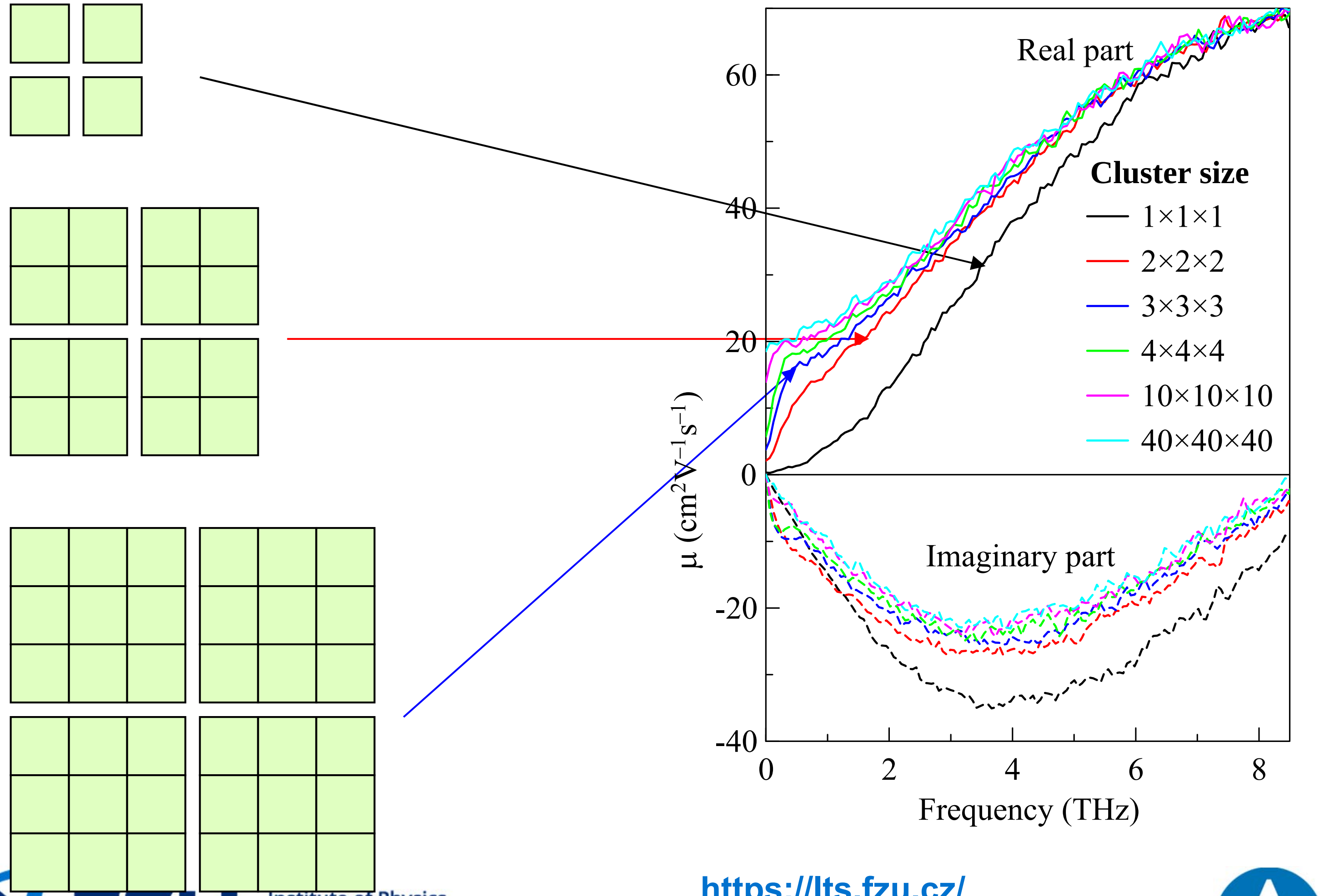
l_{free}
(mean free path)

Amplitude decreases with decreasing nanoparticle size

Resonant frequency is related to the round-trip time of the electron in the nanoparticle

Models of electric response: Monte-Carlo calculations

- Aggregation of nanoparticles
  - Existence of the second length scale appears as the second cut-off frequency



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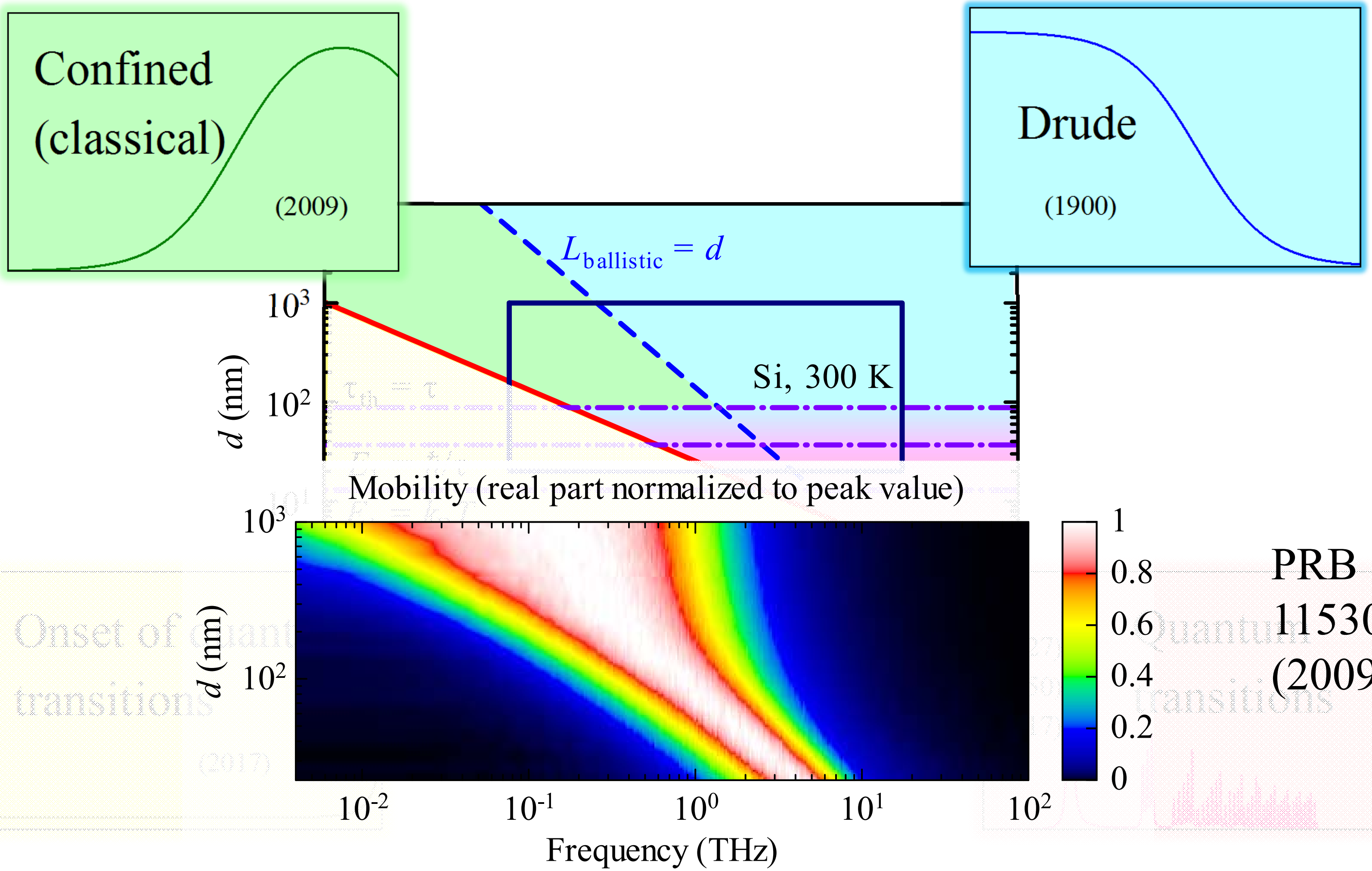
Models of electric response: Monte-Carlo calculations

- World of Monte-Carlo semi-classical calculations



PRB **79**, 115309 (2009)

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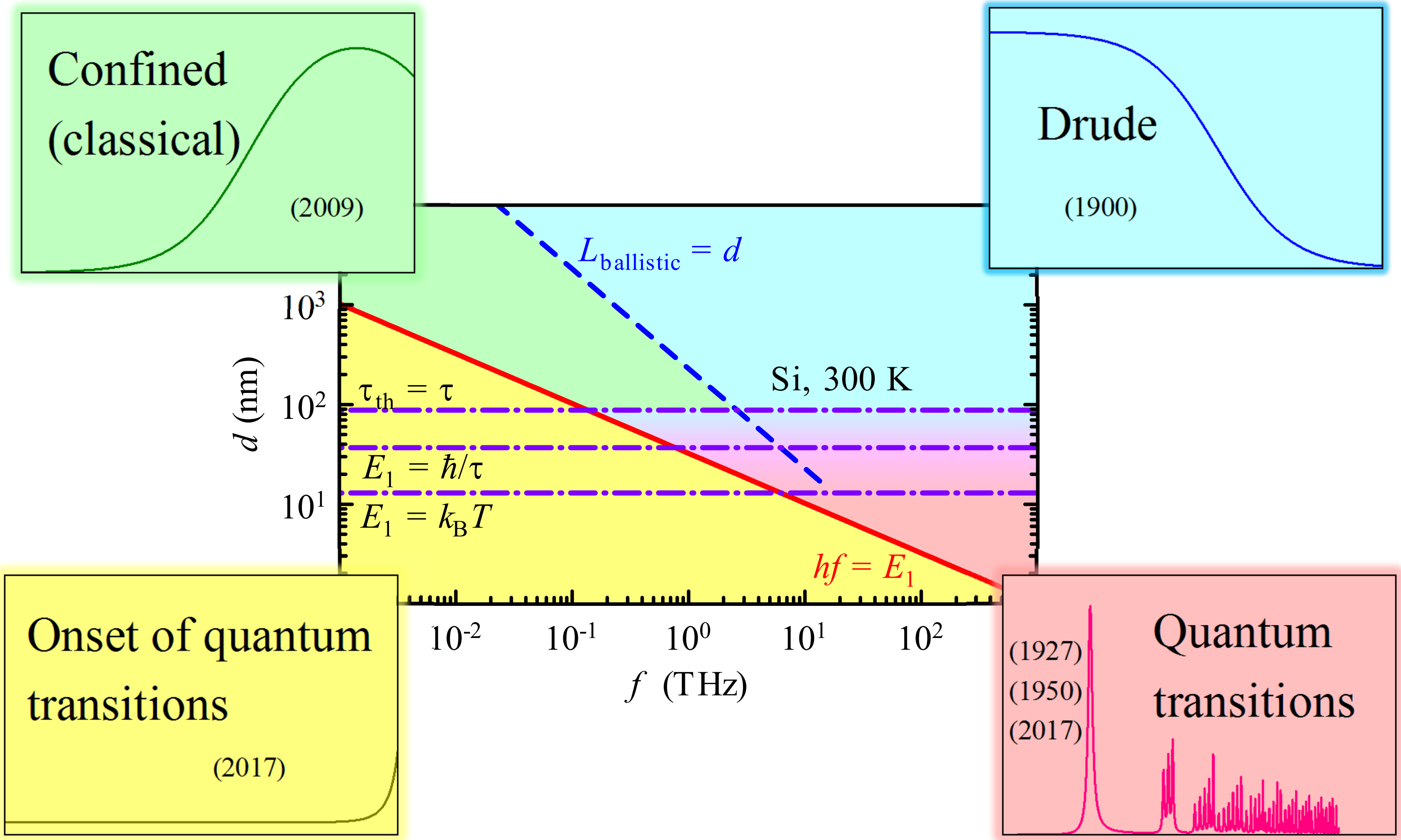
Models of electric response: Summary

DOI: 10.1002/adom.201900623



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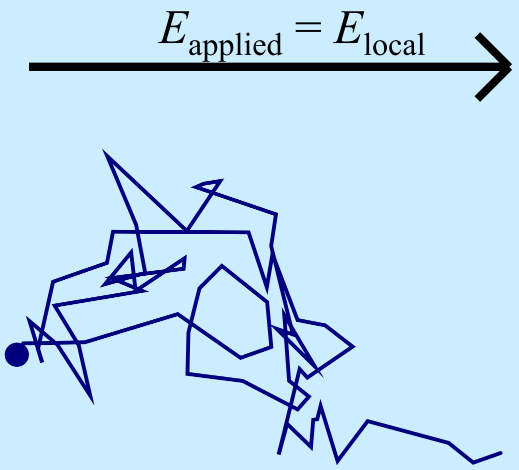
... 2nd part of the story: depolarization fields (effective medium approximation)

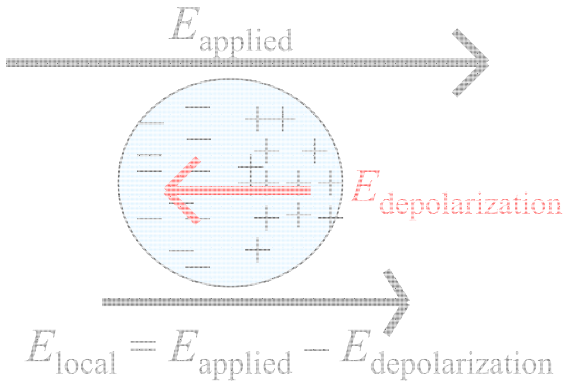
Conductivity of complex systems

- Homogeneous systems
  - local field = applied field
  - no interaction with surface

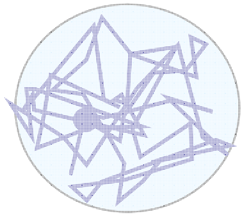


Charges undergo undisturbed random walk

- Nanostructured systems
  - local field ≠ applied field

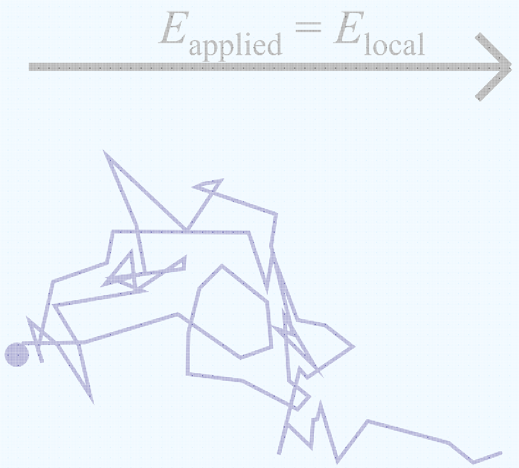


  - charges interact with surface
  - response to the *local* electric field



Conductivity of complex systems

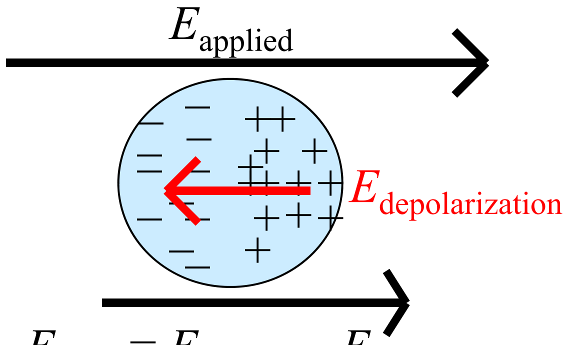
- Homogeneous systems
  - local field = applied field
  - no interaction with surface



$E_{\text{applied}} = E_{\text{local}}$

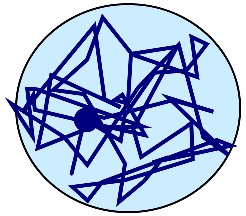
Charges undergo undisturbed random walk

- Nanostructured systems
  - local field ≠ applied field



$$E_{\text{local}} = E_{\text{applied}} - E_{\text{depolarization}}$$

  - charges interact with surface
  - response to the *local* electric field



Effective medium approximation

- Effective medium approximation
  - Permittivity profile ε(**r**) is approximated by a single value $\varepsilon_{\mathrm{eff}}$
  - Assumptions: characteristic dimensions << probing wavelength

- Transient permittivity
  - Permittivity ε(**r**) + Δε(**r**) gives a single value $\varepsilon_{\mathrm{eff}} + \Delta\varepsilon_{\mathrm{eff}}$
  - Formally: $\Delta\varepsilon_{\mathrm{eff}} \equiv \Delta\varepsilon_{\mathrm{eff}}[\varepsilon(\mathbf{r}), \Delta\varepsilon(\mathbf{r})]$

$\Delta\varepsilon_{\mathrm{eff}}[\varepsilon(\mathbf{r}), \Delta\varepsilon(\mathbf{r})]$ is a black-box which is evaluated as follows

- Electric field profile $E(\mathbf{r})$ is calculated numerically by finite-element method
- Effective permittivity is calculated from energy density:

$$\frac{1}{2}\varepsilon_{\mathrm{eff}}\langle\mathbf{E}\rangle^2 = \frac{1}{2V}\int_V \varepsilon(\mathbf{r})\mathbf{E}^2(\mathbf{r})dV, \quad \text{where } \langle\mathbf{E}\rangle = \frac{1}{V}\int_V \mathbf{E}(\mathbf{r})dV$$

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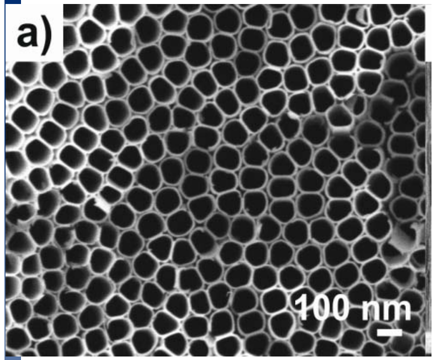
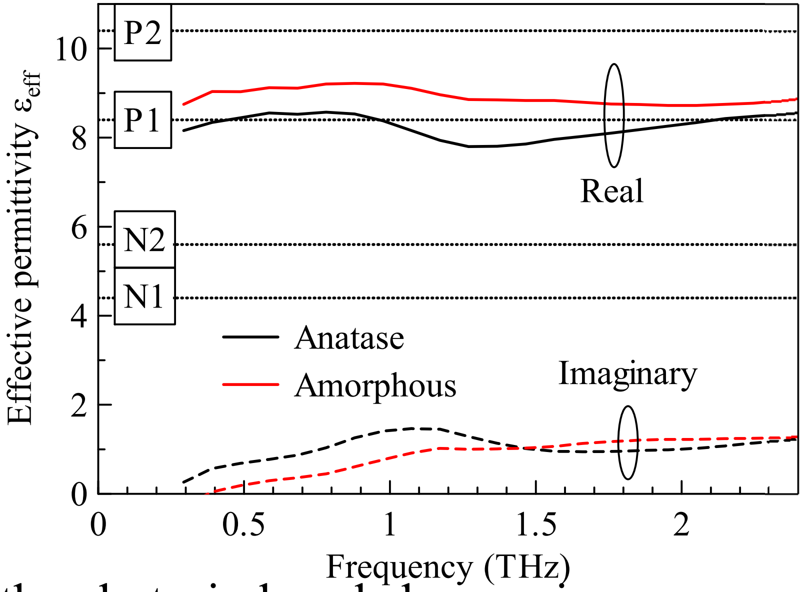
Effective medium approximation

- Implementation
  - Steady-state calculations: morphology + permittivities of components = effective permittivity
    - For simple morphologies, special approximations may be applicable (e.g. Maxwell-Garnett for sparse inclusions)
    - Numerical calculations for more complex structures are inevitable



Anatase nanotubes



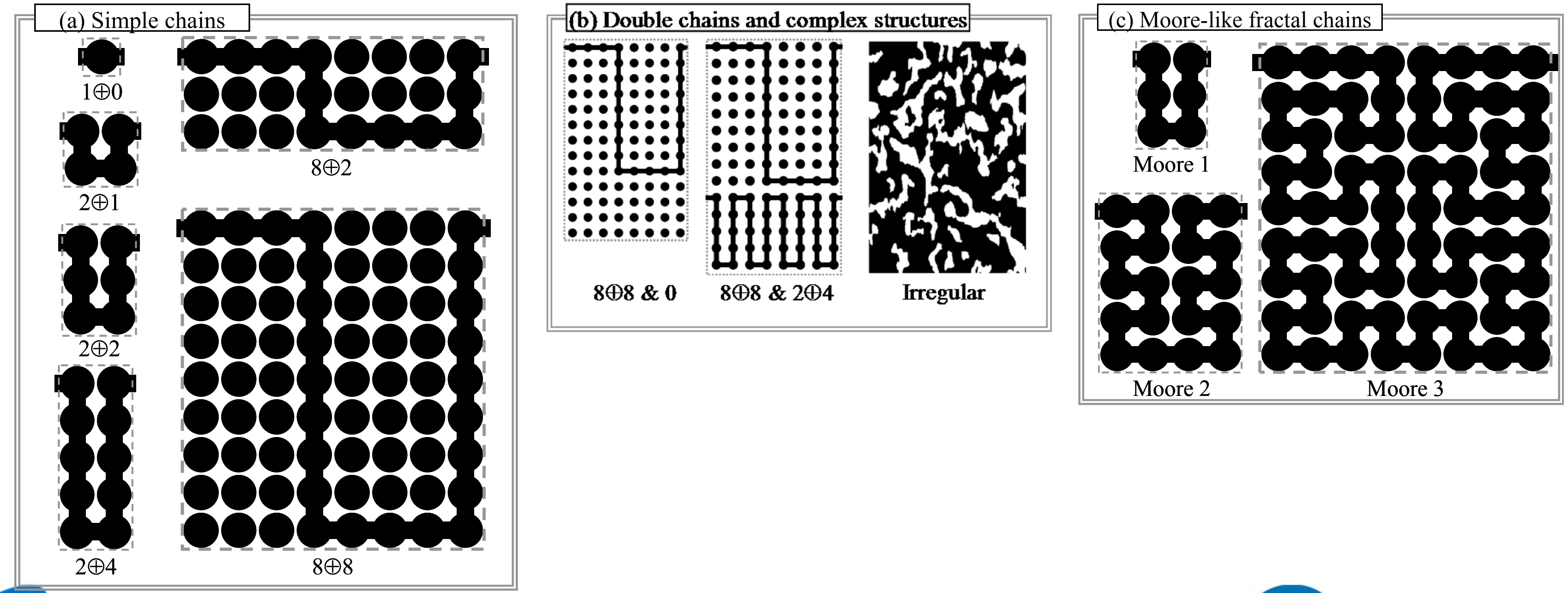
	structure	ε_{eff}
N1		4.4
N2		5.6
P1		8.4
P2		10.4

  - Can we treat the photo-induced changes in a more convenient way?

Jiří Kuchařík[1], Hanna Sopha[2], Milos Krbal[2], Ivan Rychetský[1], Petr Kužel[1], Jan M. Macak[2], and Hynek Němec[1]

Effective medium approximation

- Assumptions
  - Two-component system; only one component can be photoexcited
  - Purely real transient conductivity: Re Δσ ≠ 0, Im Δσ = 0 (i.e., purely imaginary transient permittivity)
- Investigated structures

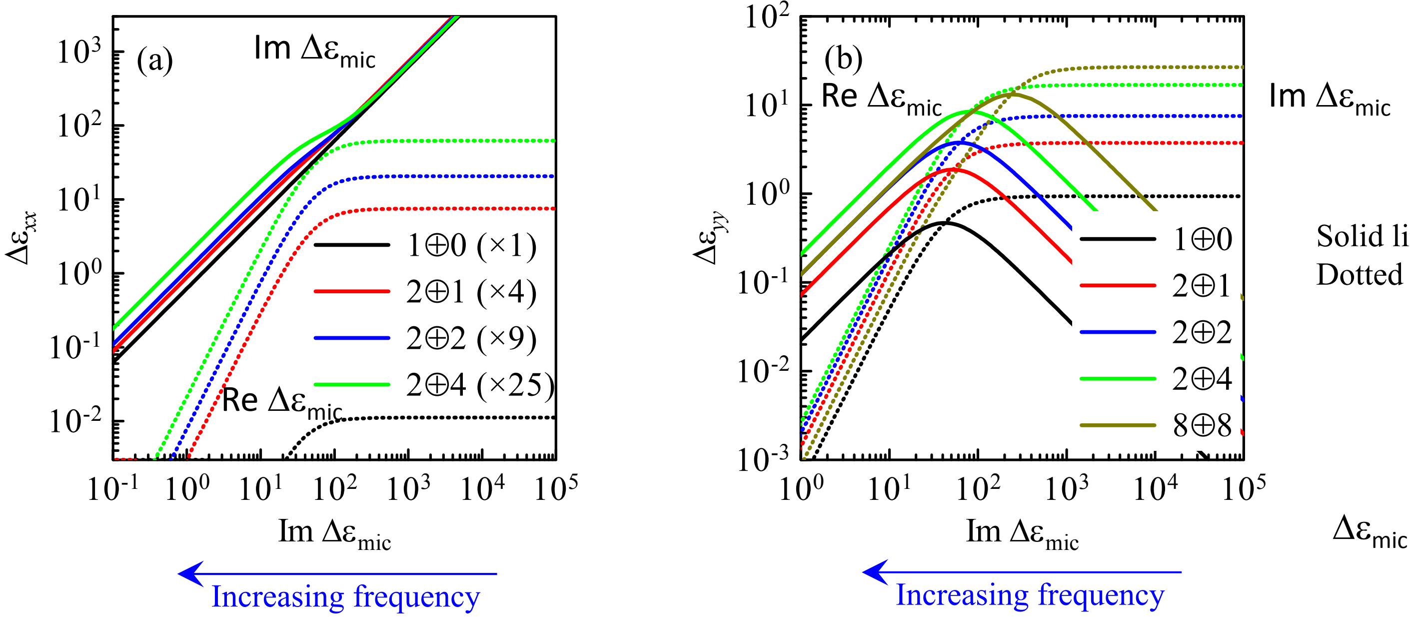


IEEE T. THz Science Technol. **3**, 302 (2013).

Effective medium approximation

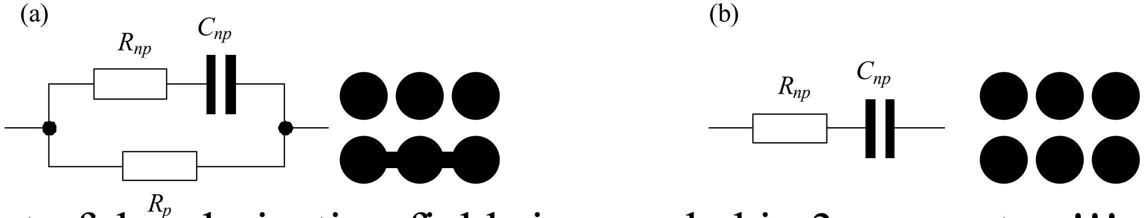
- Common properties

Percolated structures *Non-percolated structures*



Solid lines: imaginary part
Dotted lines: real part

$\Delta\varepsilon_{mic} = i n_{exc} e_0 \mu / (\omega \varepsilon_{vac})$



- The impact of depolarization fields is encoded in 3 parameters!!! (R_{np}, C_{np}, R_{p})

Effective medium approximation – summary

- Effective photoconductivity of inhomogeneous systems

$$\Delta\sigma = V\Delta\sigma_{\mathrm{mic}} + \frac{B\Delta\sigma_{\mathrm{mic}}}{1 + iD\Delta\sigma_{\mathrm{mic}}/(\omega\varepsilon_0)}$$

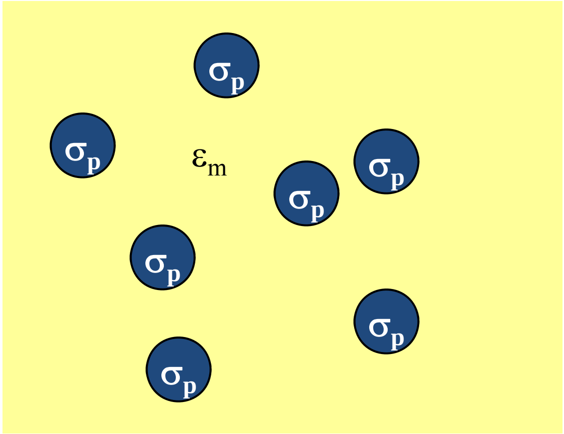
percolated

non-percolated

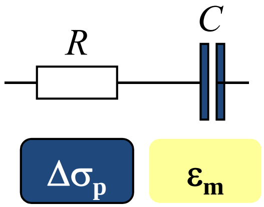
  - 3 independent parameters:
    - V … percolation strength
    - B, D …non-percolated part
  - Describes Maxwell-Garnett
  - Conform with Bergman representation
  - Pole in the denominator = localized plasmon resonance
  - Extensive 2D electrostatic simulations show that this model describes a large number of morphologies including complex percolation pathways
    - Morphology + ground state properties = parameters V, B, and D
  - <u>Assumption: components not conducting in the ground state</u>
    - Difference of conductivities needs to be calculated for conducting components. This has been discussed in detail in Joyce et al., Nanotechnol. **24**, 214006 (2013).
  - Assumption: subwavelength structures

Influence of depolarization fields

- *Non-percolated* photoconductor



Equivalent electrical circuit



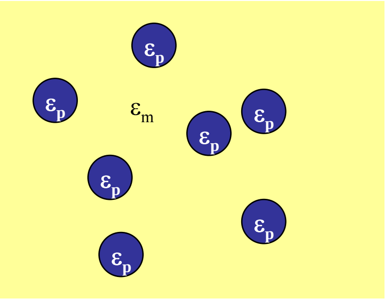
(a) Poor conductor (low $\Delta\sigma_p$):
Conductive response of particles limits the conductivity

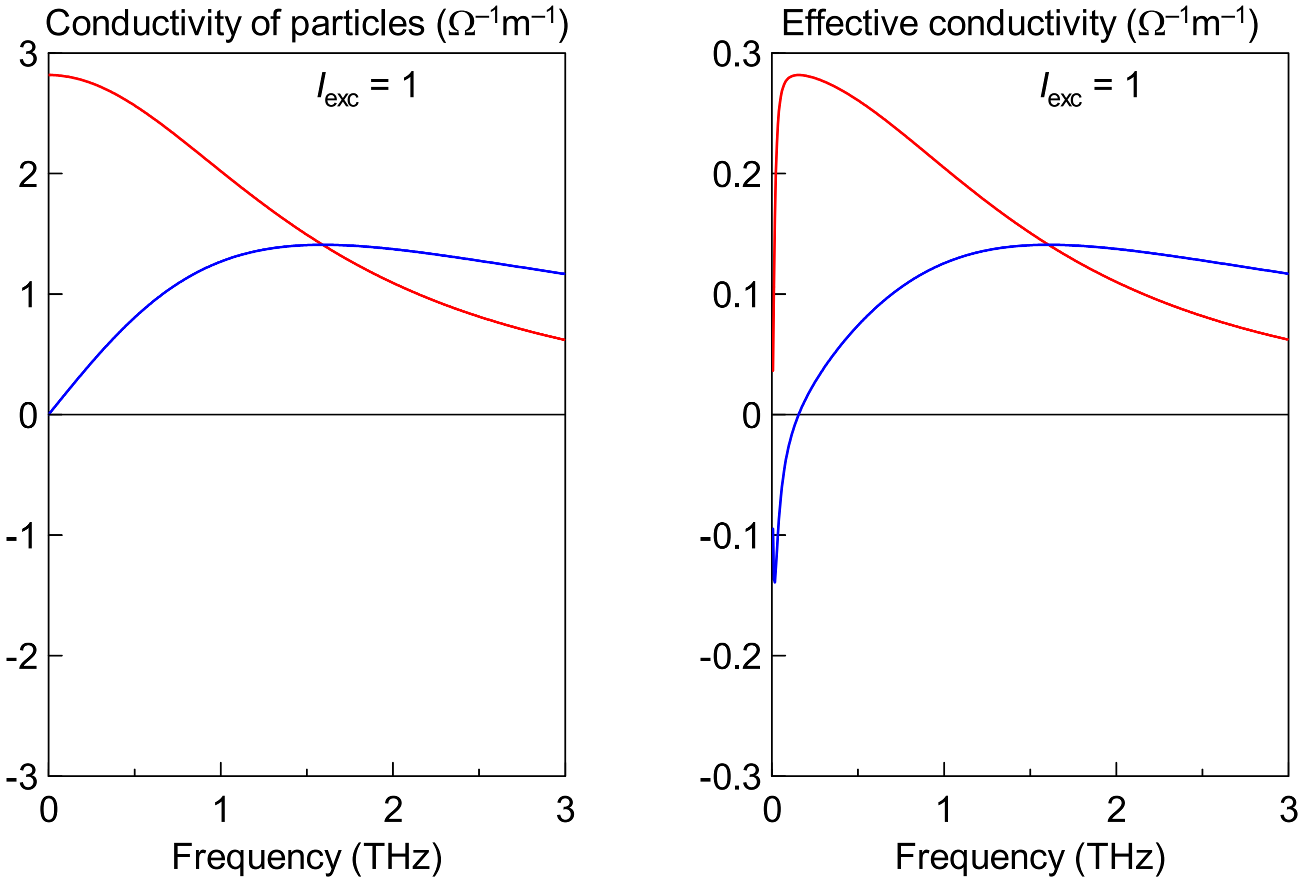
$$\Delta\sigma_{\text{eff}} \propto \Delta\sigma_{\text{p}}$$

(b) Good conductor (high $\Delta\sigma_p$):
Capacitance limits the conductivity

$$\Delta\sigma_{\text{eff}} \propto -i\omega\varepsilon_0\varepsilon_{\text{m}}$$

Influence of depolarization fields





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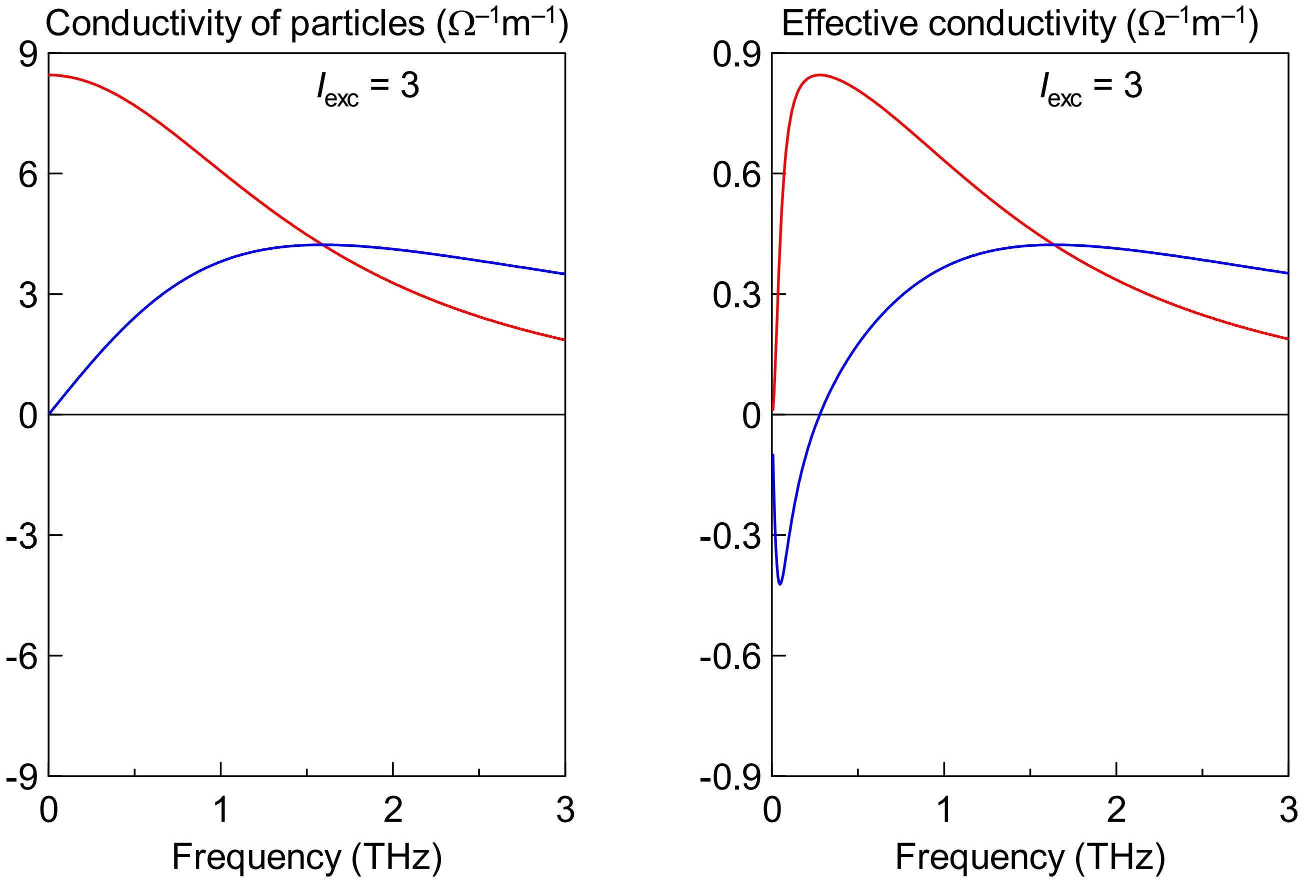
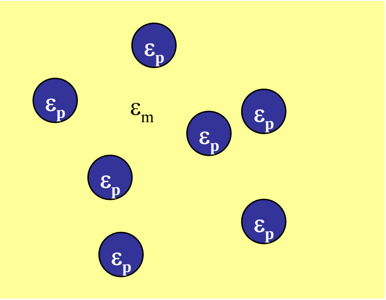
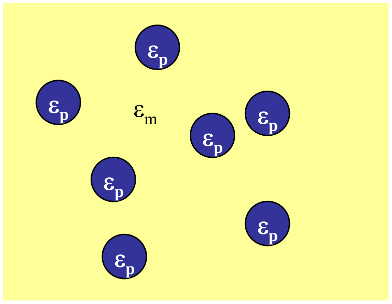
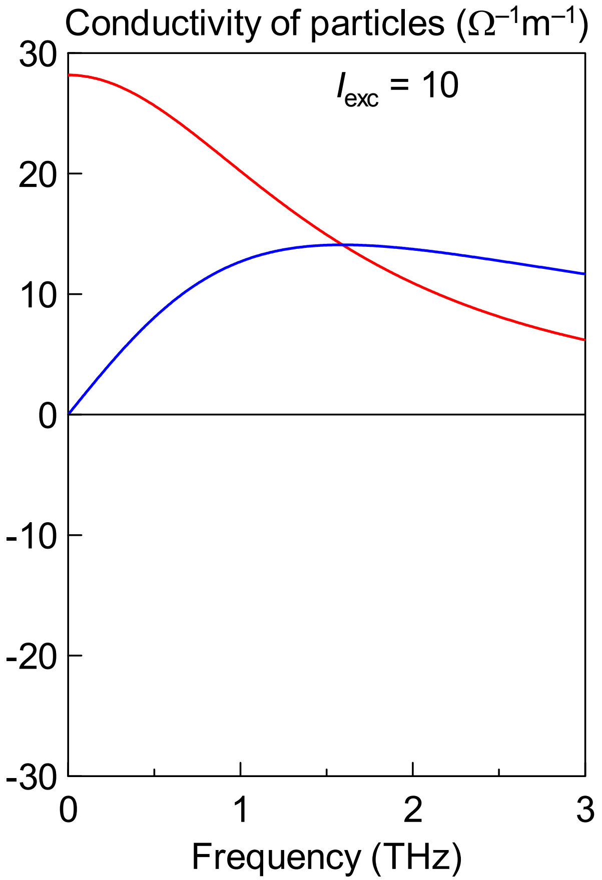
Influence of depolarization fields

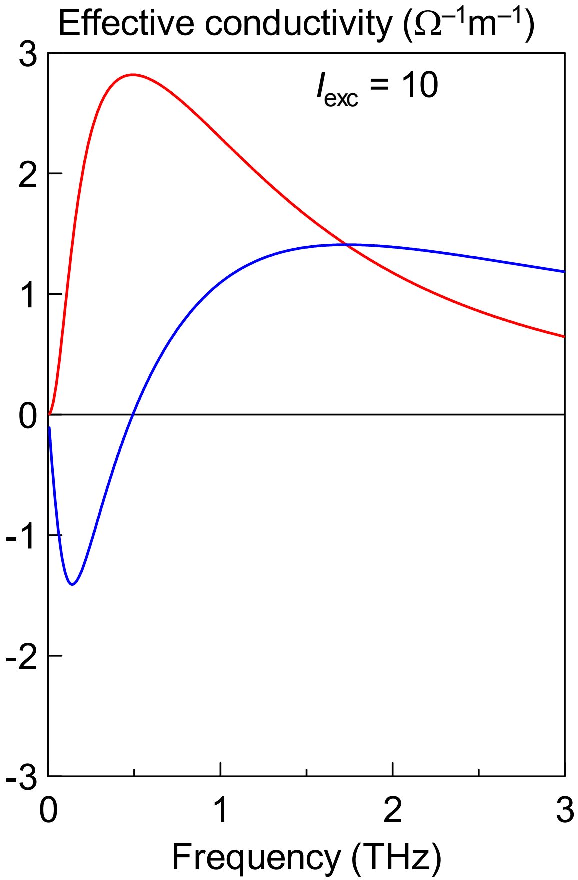




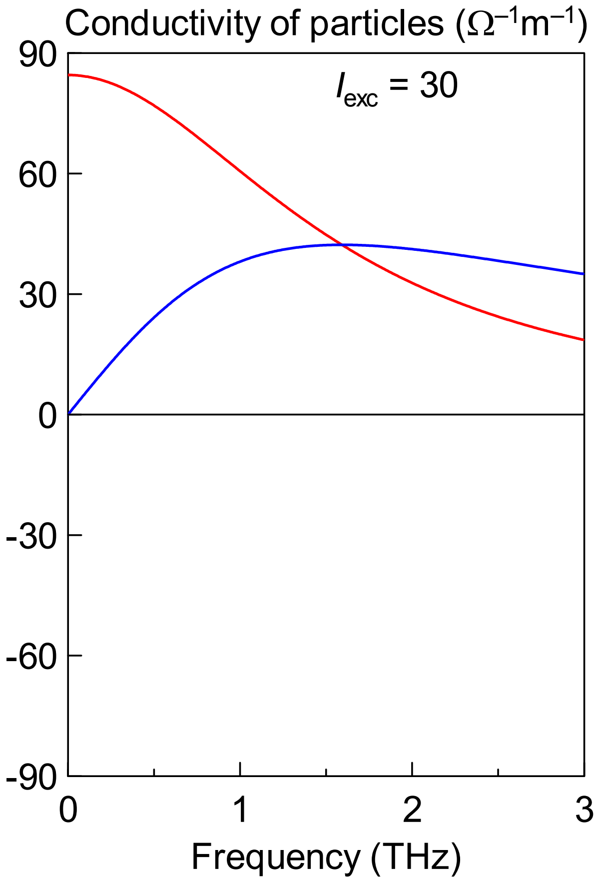
Influence of depolarization fields

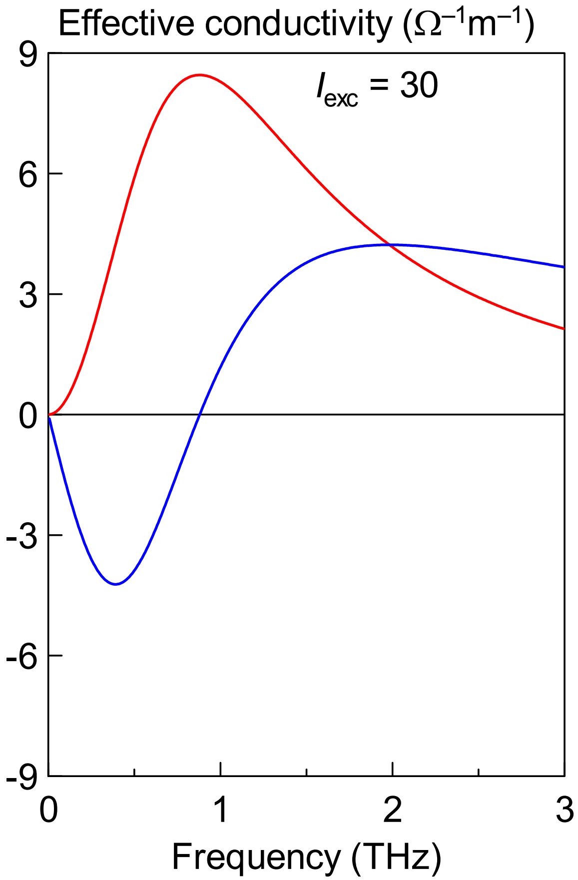


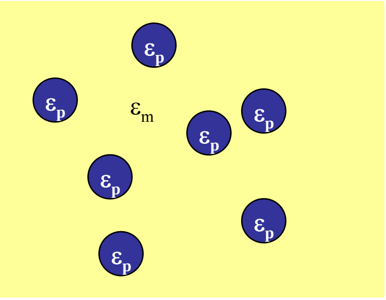

Conductivity of particles ($\Omega^{-1}m^{-1}$)


Effective conductivity ($\Omega^{-1}m^{-1}$)

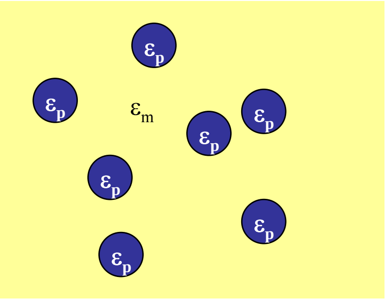
Influence of depolarization fields

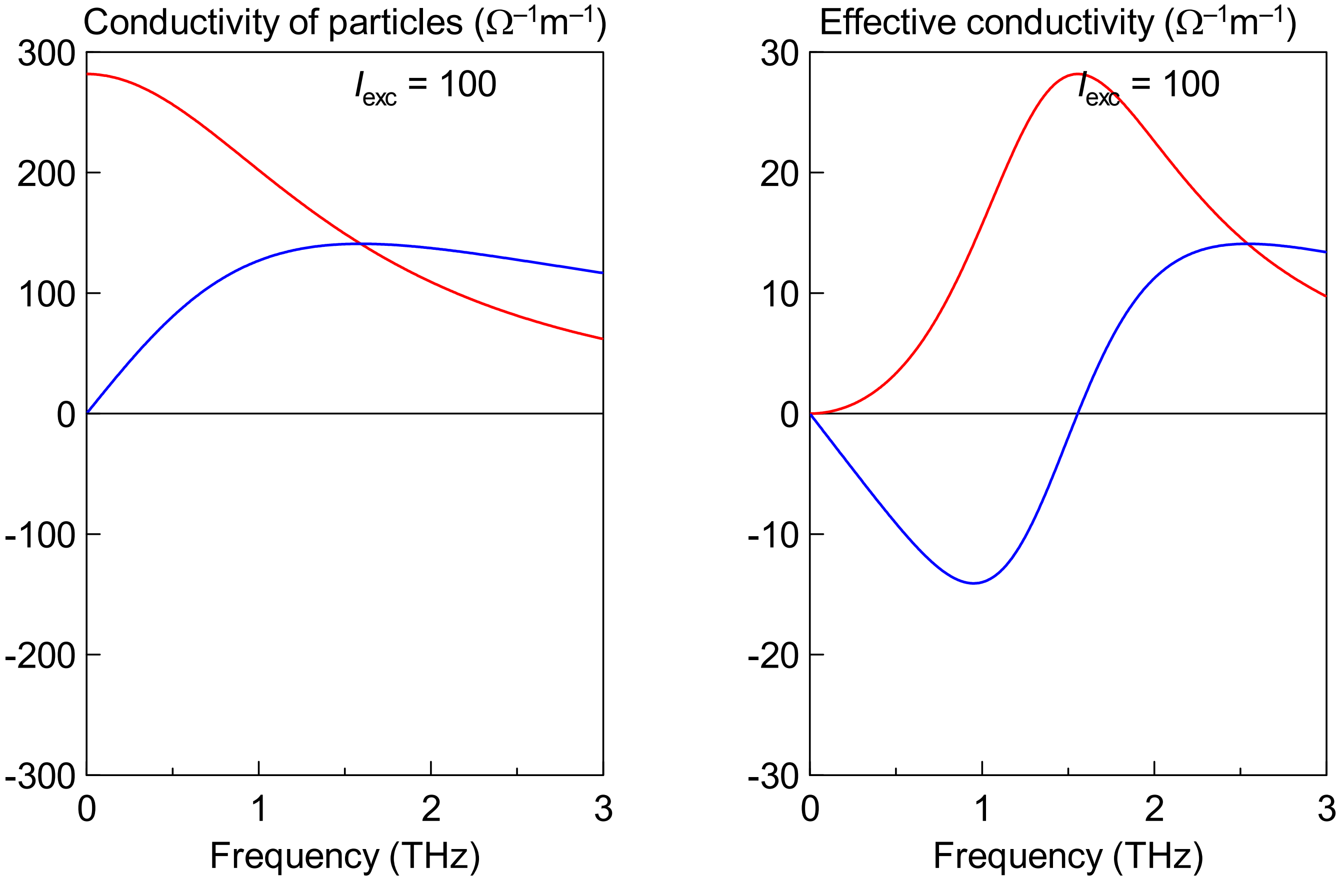






Influence of depolarization fields





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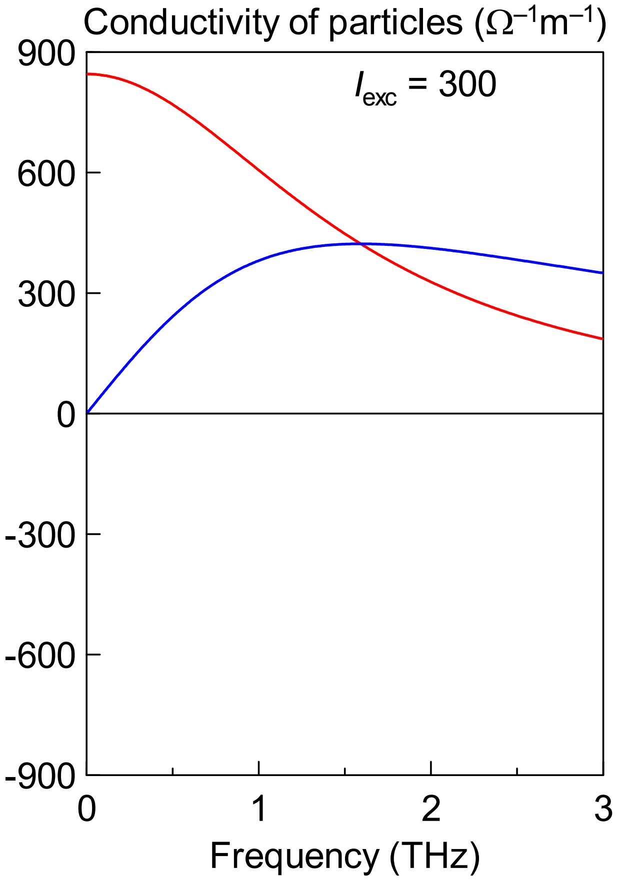
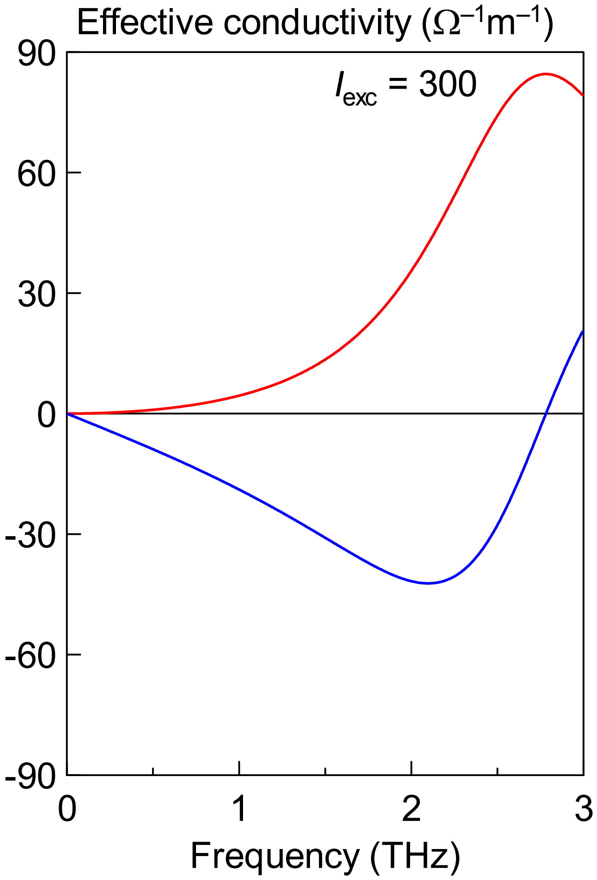
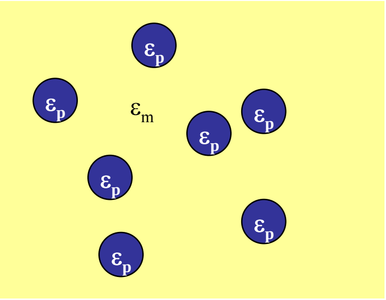
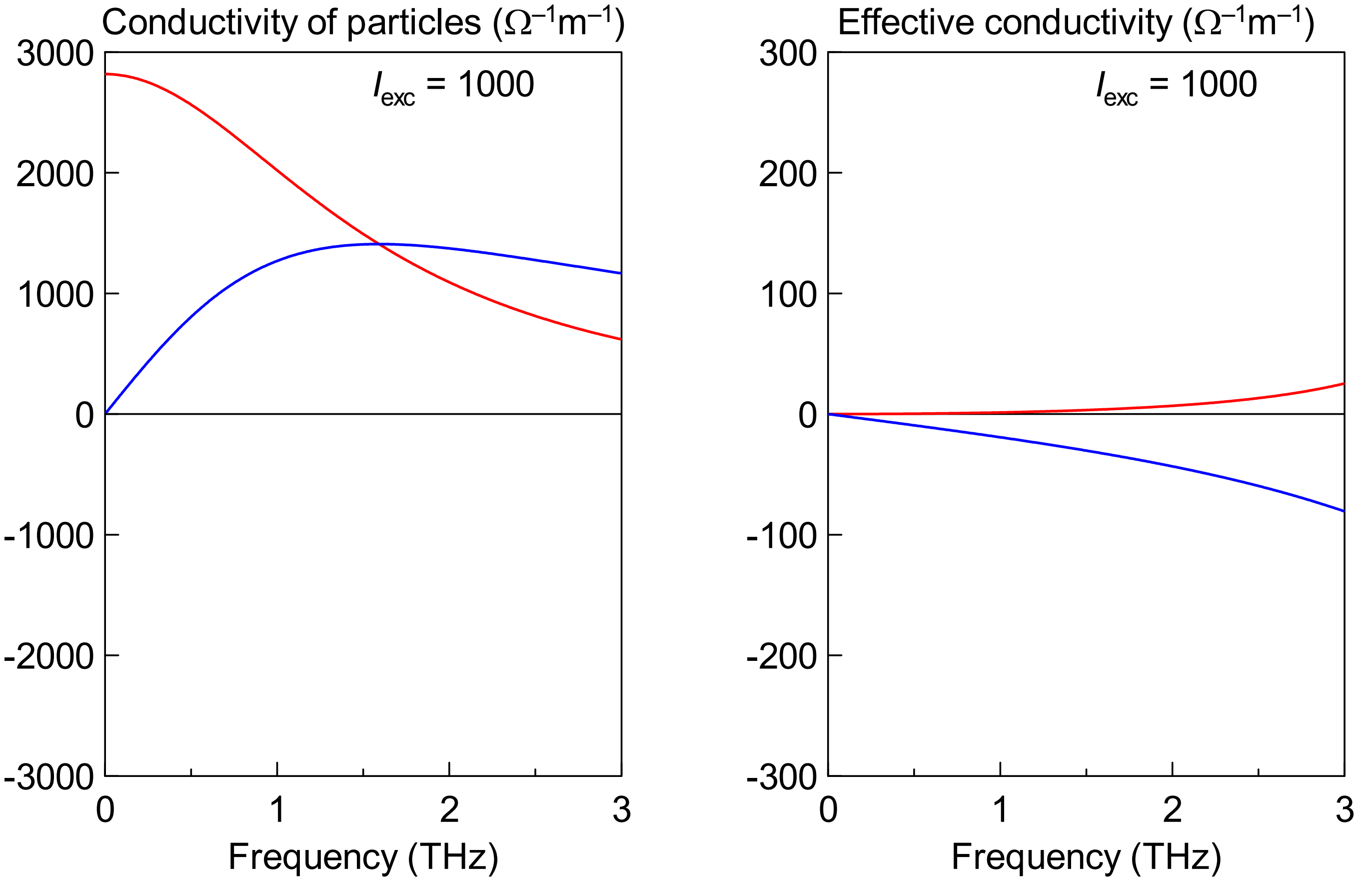
Conductivity of complex systems

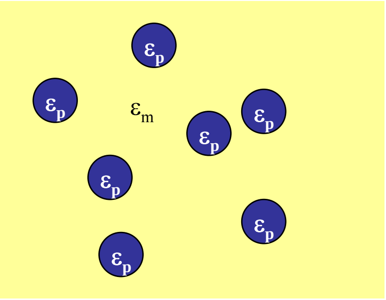






Influence of depolarization fields





Effective medium approximation – summary

- Effective photoconductivity of inhomogeneous systems

$$\Delta\sigma = V\Delta\sigma_{\mathrm{mic}} + \frac{B\Delta\sigma_{\mathrm{mic}}}{1 + iD\Delta\sigma_{\mathrm{mic}}/(\omega\varepsilon_0)}$$

	Percolated system	Non-percolated system
Low intensity	$\Delta\sigma = (V + B)\Delta\sigma_{\mathrm{mic}} \approx V\Delta\sigma_{\mathrm{mic}}$	$\Delta\sigma = B\Delta\sigma_{\mathrm{mic}}$
Medium intensity	$\Delta\sigma = (V + B)\Delta\sigma_{\mathrm{mic}} \approx V\Delta\sigma_{\mathrm{mic}}$	$\Delta\sigma = \frac{B\Delta\sigma_{\mathrm{mic}}}{1 + iD\Delta\sigma_{\mathrm{mic}}/(\omega\varepsilon_0)}$
High intensity	$\Delta\sigma = (V + B)\Delta\sigma_{\mathrm{mic}} \approx V\Delta\sigma_{\mathrm{mic}}$	$\Delta\sigma = -i\omega\varepsilon_0 \frac{B}{D}$

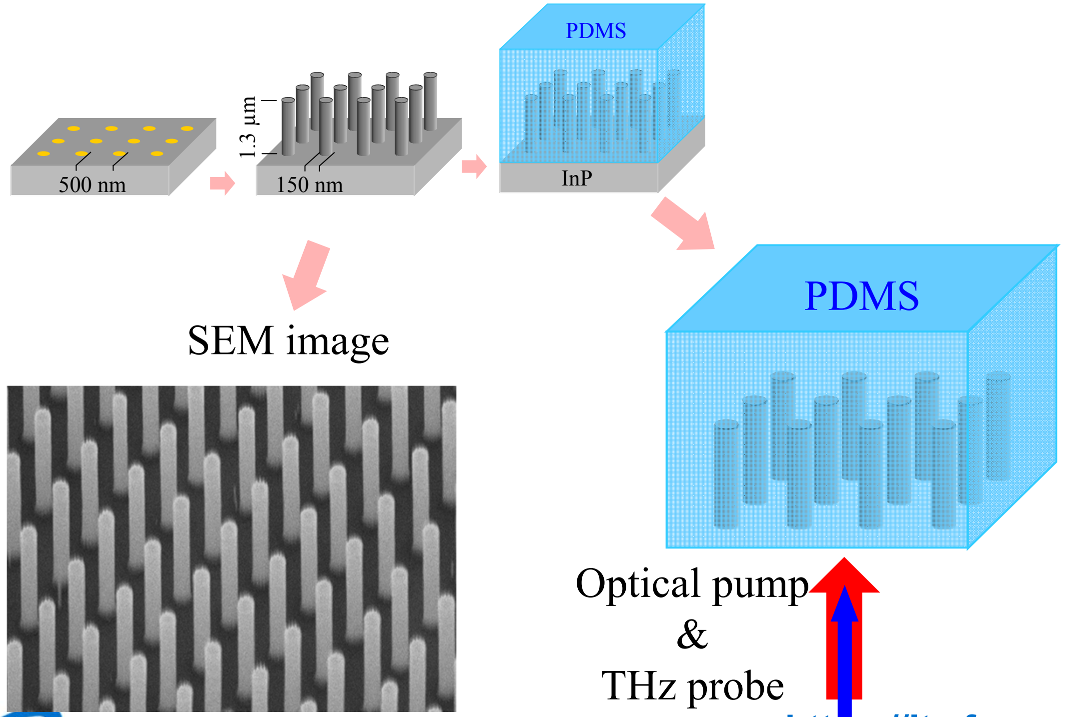
- Assessment of the percolation degree

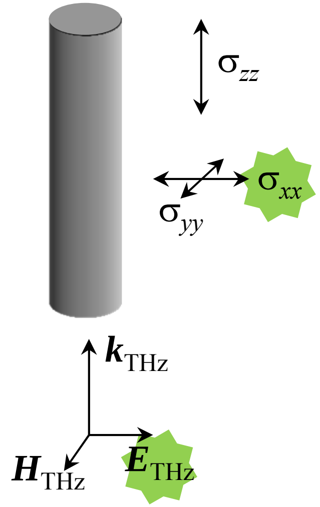
J. Phys. D: Appl. Phys. **47**, 374005 (2014)

Experimental results: InP nanowires

Example: InP nanowires

- InP nanowires – well-defined morphology
  - vertically aligned
  - mutually isolated
  - diameter 150 nm, period 500 nm, length 1.3 μm
  - *n*-doped by Sn to 2.5×10^{18} cm^{-3}



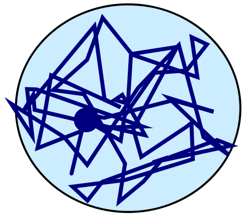


Transversal transport is probed

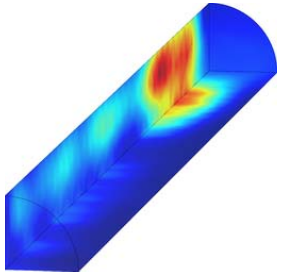
$$\begin{pmatrix} \sigma_{xx} & 0 & 0 \\ 0 & \sigma_{yy} & 0 \\ 0 & 0 & \sigma_{zz} \end{pmatrix}$$

Example: InP nanowires

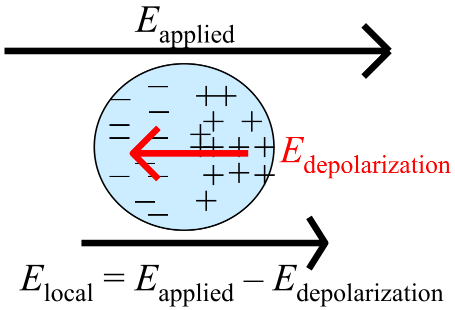
- Charge confinement effects
  - Characteristic parameters
    - Fermi velocity: 10^6 m/s
    - Times: electron scattering (100 fs) – probing period (1 ps)
    - ⇒ Lengths: 100 nm – 1 μm $\gtrsim$ diameter (150 nm)



- Waveguiding of the excitation beam
  - Excitation wavelength ≈ period & diameter ⇒ interferences
  - Analogy: photonic crystal lattice



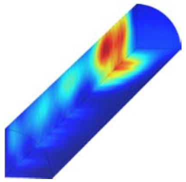
- Depolarization fields
  - Inhomogeneous system
  - Maxwell-Garnett approximation (low filling fraction)

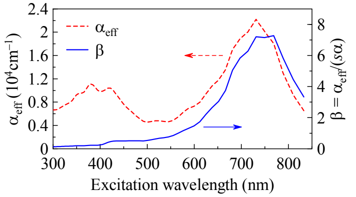


Example: InP nanowires

Excitation beam waveguiding

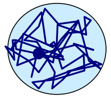
$n_{\mathrm{exc}}(z)$

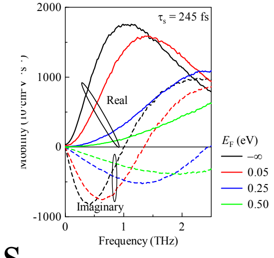




+ Charge confinement effects

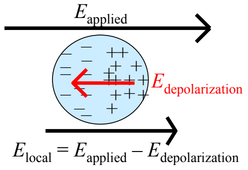
μ(ω)

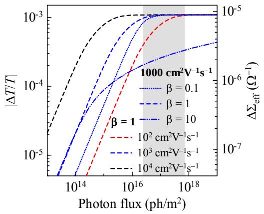




+ Depolarization fields

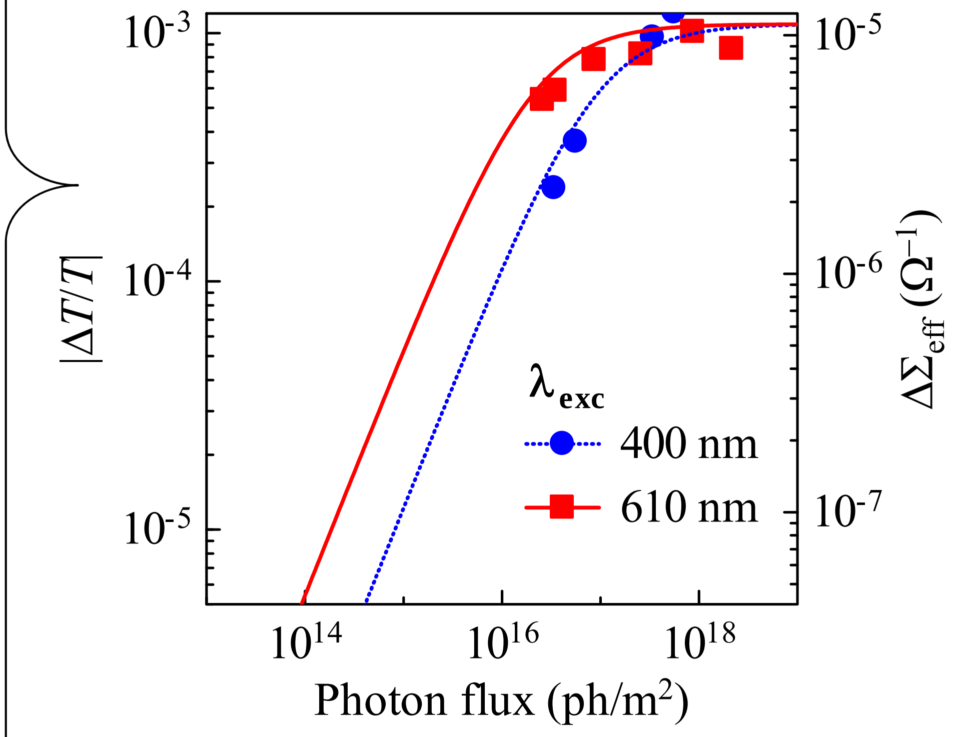
$\Delta\sigma_{\mathrm{eff}}(\omega,z)$





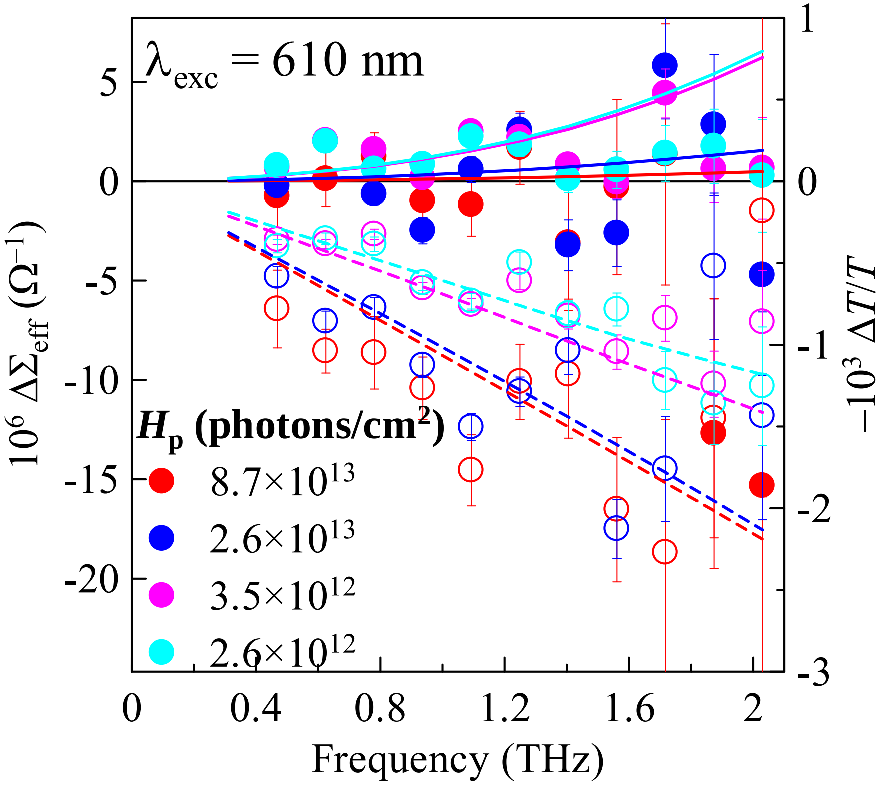
+ Only 2 unknown parameters

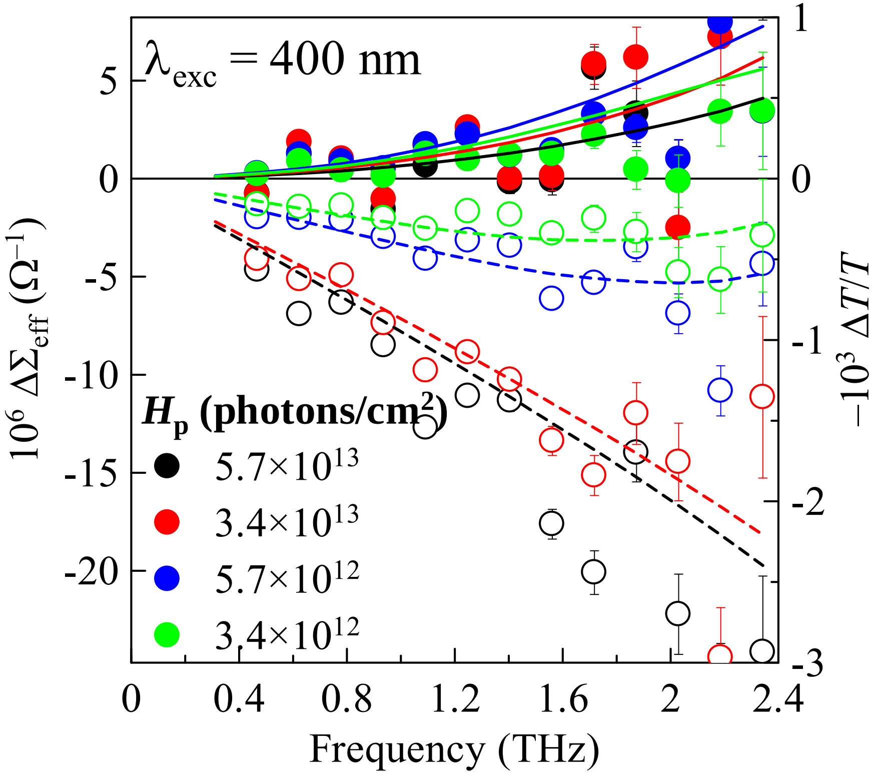
- Mobility (electron scattering time τ_s)
- Quantum yield of charges φ



Experiment: symbols
Theory: lines

Example: InP nanowires





Experiment: symbols
Theory: lines

$\tau_s \gtrsim 150$ fs

$\mu \gtrsim 3000\ \mathrm{cm^2V^{-1}s^{-1}}$

$\varphi = 65\%$

$\tau_s \gtrsim 150$ fs

$\mu \gtrsim 3000\ \mathrm{cm^2V^{-1}s^{-1}}$

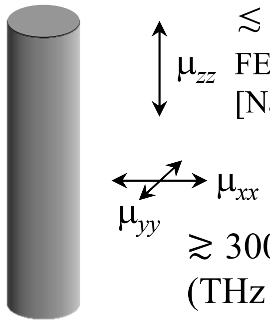
$\varphi = 14\%$

FZU Institute of Physics of the Czech Academy of Sciences

https://lts.fzu.cz/

Czech Academy of Sciences

Example: InP nanowires



$\lesssim 500\ \mathrm{cm^2V^{-1}s^{-1}}$

μ_{zz} FET mobility

[Nano Lett. 12, 151 (2012)]

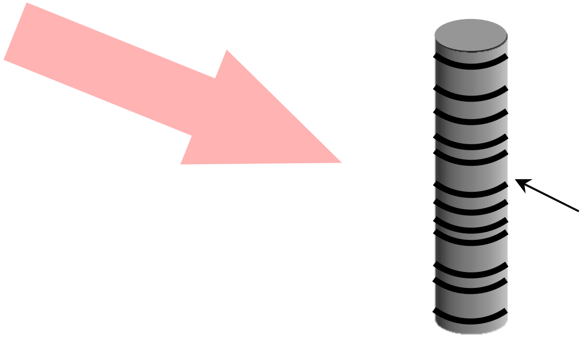
μ_{xx}

μ_{yy}

$\gtrsim 3000\ \mathrm{cm^2V^{-1}s^{-1}}$

(THz spectroscopy)

$$\begin{pmatrix} \mu_{xx} & 0 & 0 \\ 0 & \mu_{yy} & 0 \\ 0 & 0 & \mu_{zz} \end{pmatrix}$$



Stacking faults

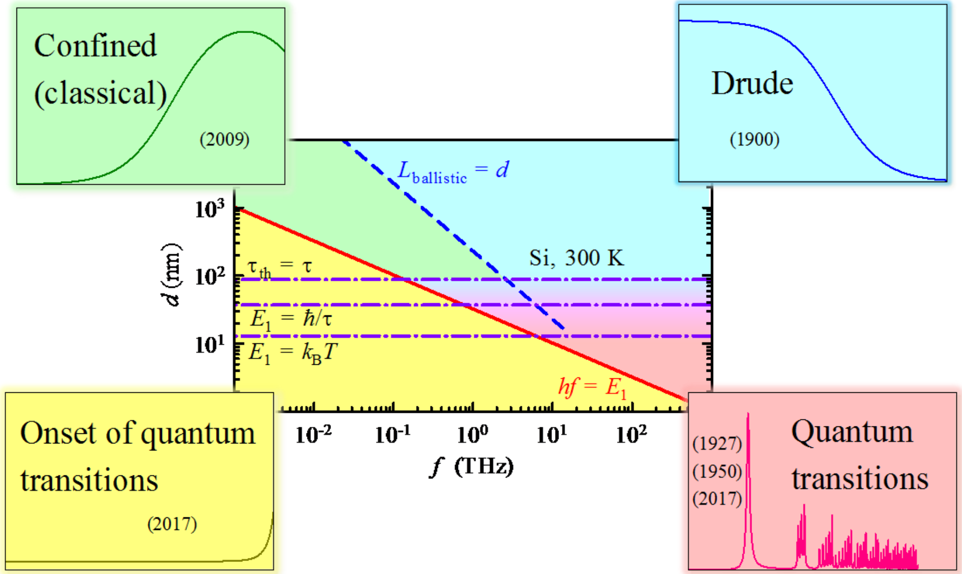
- Barriers for longitudinal charge transport
- Not affecting transversal charge transport

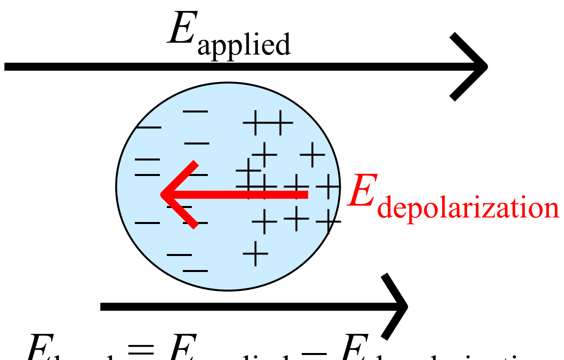
Summary

Microscopic calculations of THz response of confined charges (semi-classical/quantum) must be combined with effective medium approximation (VBD model applies for a large variety of morphologies)

Info encoded in THz spectra: Nanoparticle size, clustering, band-filling, presence of further forces, percolation degree

P. Kužel and H. Němec, Terahertz Spectroscopy of Nanomaterials: a Close Look at Charge-Carrier Transport. *Adv. Opt. Mater.* (in press)
DOI: 10.1002/adom.201900623





$$E_{\text{local}} = E_{\text{applied}} - E_{\text{depolarization}}$$