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PHOTON-RANDOM WALK MODEL FOR DIFFUSE TRANSIENT ABSORPTION SPECTROSCOPY

Modeling signals in diffuse transient absorption (TA) spectroscopy by photon random walk simulations

MOTIVATION

Functional material optical properties investigation via TA (e.g. absorption)

Solid-state → TA

Powder → diffuse TA → R,T data

Simulation

Lampbert-Beer law

$$\Delta\alpha(t)=\frac{1}{d}\ln\left(\frac{T_0}{T(t)}\right)$$

derive dependencies → ?

Solve reverse problem: predict scattering and absorption properties from diffuse TA experiments

METHODS

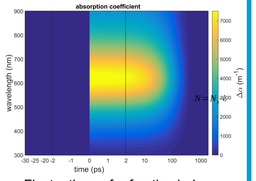
Random-walk model for photons with **scattering** and **absorption**

Photons assumed as energy carrying particles travel indepedently in sample.

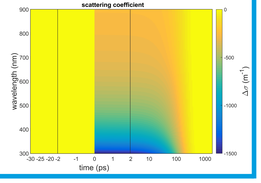
- input signal discretised both in time (N_t) and spectrum (N_s) axes; each time-spectrum bind can be cloned into (N_c) independent photons
- speed of photon depends on refractive index $v(x,\lambda)=\frac{c}{n(t,x,\lambda)}$
- scattering probability on each step $\rho(\Delta x)=1-\exp(-s(t,\lambda,x)|\Delta x|)$
- absorption reduces energy on each step $I(x+\Delta x)=I(x)\exp(-a(t,\lambda,x)|\Delta x|)$
- photon counted as *reflected* or *transmitted* after reaching the sample border
- simulation stops after reaching (1-ε) total energy absorbed, reflected or transmitted.

Total number of photons $N=N_t\cdot N_s\cdot N_c$

Lorentzian absorption band with monoexp. decay in *time* & *depth*



Fluctuations of refractive index



RESULTS

Workflow for simulation

Pulse: N(λ,t)

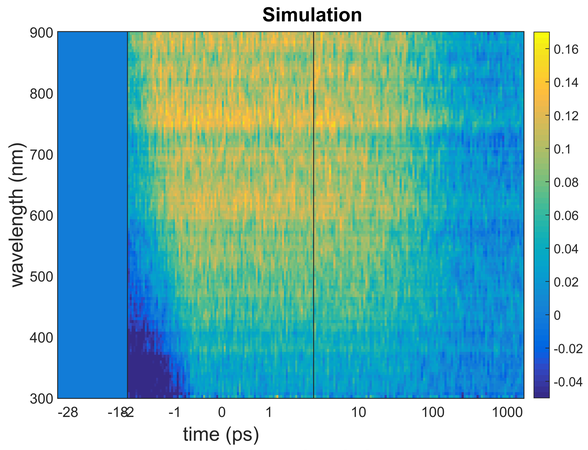
n(t,λ,x) – complex refractive index
s(t,λ,x) – scattering probability

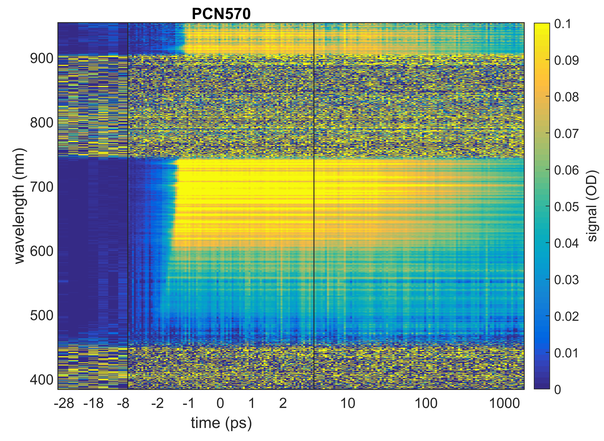
→ **Random-walk simulation**

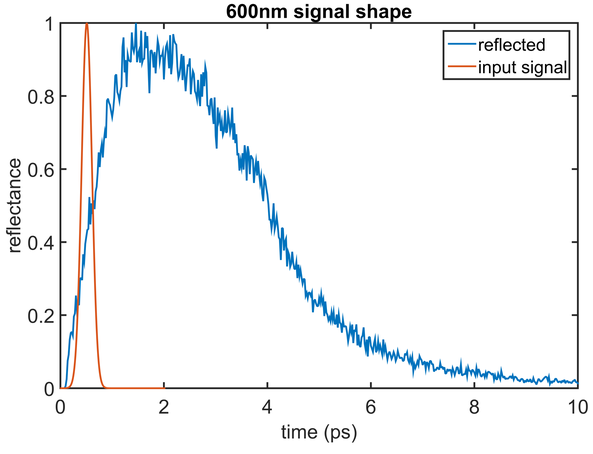
additional simulation parameters, e.g. sampling parameters

→ **Reflectance, Transmittance signals** R(t,λ), T(t,λ) **for each delay time**

Reflectance in simulation and experiment







DISCUSSION

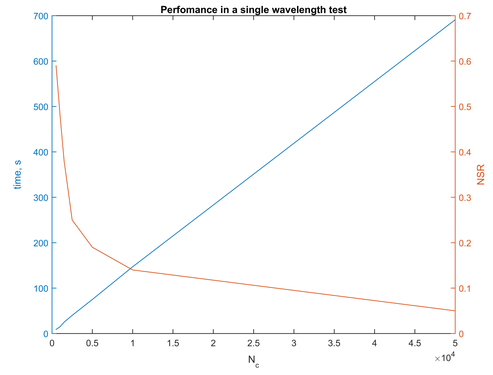
- reflected pulses are stretched by a factor of ~40
- transient spectra are smeared out in time
- ultrafast processes (< 1ps) are not resolved
- apparent signal strength is amplified compared to standard TA

PERFORMANCE

Estimating time of simulation $t\simeq N_{delays}\cdot N_s\cdot t_{single}(N_c\cdot N_t)$, where t_{single} is time in single wavelength test. Average time for largest N is 136 μs/photon.

Average t_{single} time in simulation for the whole spectrum was

$$40157\,s/128(N_s)/201(N_d)/100(N_c)/101(N_t)=153\ \mu s$$



Possible improvements

- use of GPU – almost every operation was vectorised
- further code simplification and vectorisation

CONCLUSION

Created perfomance-adjustable program for photon random walk simulation that can be applied for different n(t,λ,x) and s(t,λ,x).

HAS TO BE DONE

- Performance optimization should be continued
- Include fit routine for reverse problem
- Advanced excitation model for pump-probe experiment