Time-resolved kinetics of charge carriers trapping in chalcogenide glass irradiated by high-intensity laser pulses

E.A.Romanova[1], A.B.Seddon[2], T.M.Benson[2], S.Guizard[3]

[1]Saratov State University, Saratov, Russia
[2]University of Nottingham, Nottingham, UK
[3]CNRS-Ecole Polytechnique, Palaiseau, France

The **"Integrated Initiative"** of European Laser Infrastructures
in the 7th Framework Programme of the European Union

LaserLab Europe
«NONLINEAR PROPERTIES OF CHALCOGENIDE GLASSES», **2011**

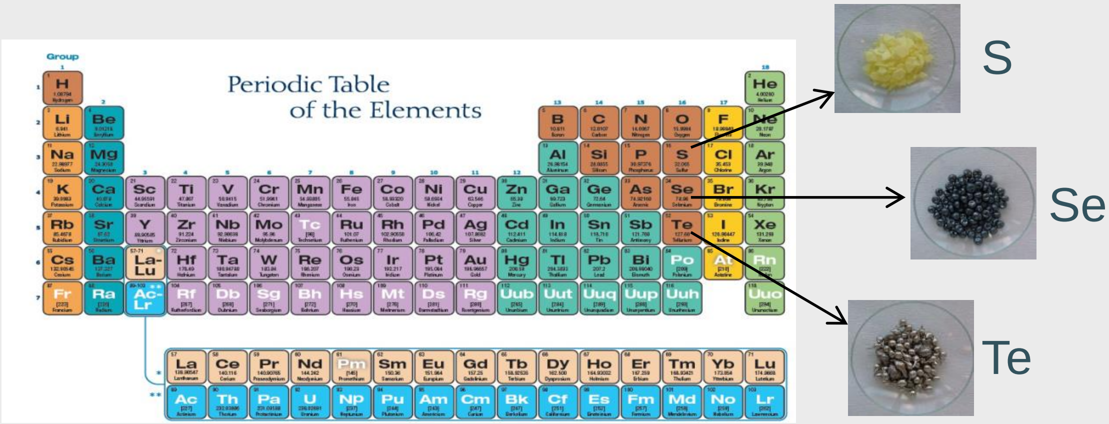
Laser Lab Europe
«STUDY OF SINGLE- AND TWO- PHOTON TRANSITIONS IN CHALCOGENIDE GLASSES», **2014**

Royal Society
«MID-INFRARED-TRANSMITTING OPTICAL FIBRE DEVICES AND SYSTEMS FOR MEDICINE»
2011-2012

Royal Society
«HIGHLY NON-LINEAR OPTICAL GLASSES FOR INFRARED PHOTONICS»,
2013 – 2014

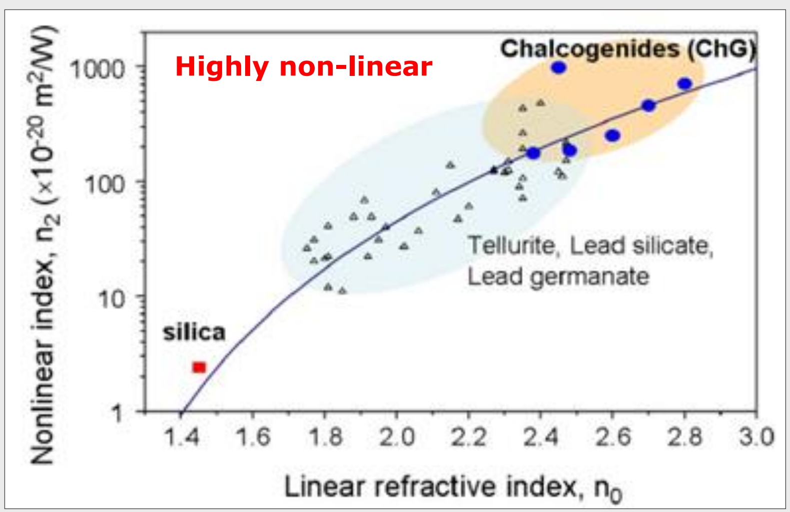
COST Action MP1401
«Advanced fibre laser and coherent source as tools for society, manufacturing and lifescience»,
2014 – 2018

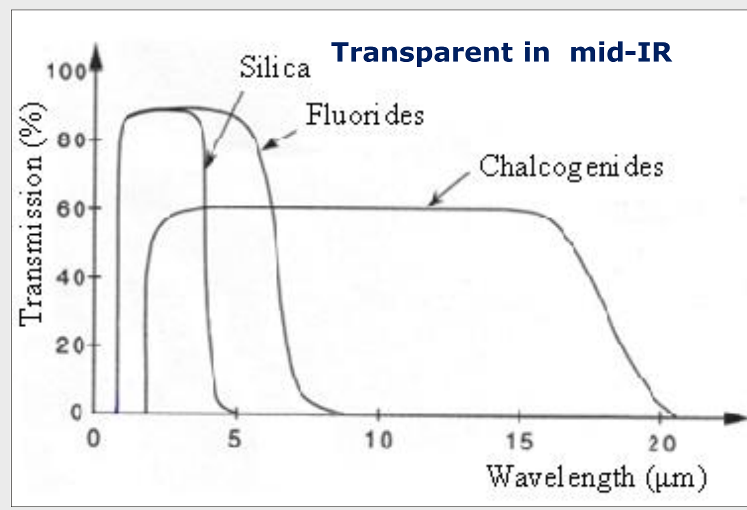
Chalcogenide glasses for non-linear photonics



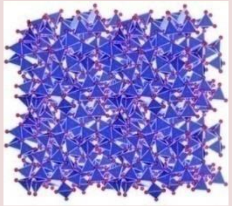
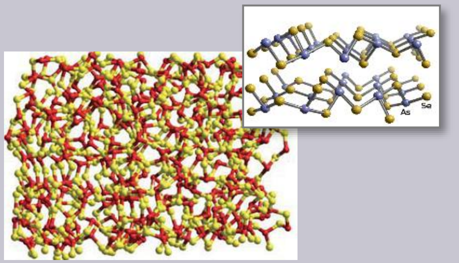
Typical compositions:

Ge - S
Ge - Se
As - S
As - Se
Ge - S - P
Ge - As - Se
Ge - Se - Te
As - Se - Te
Ge - As - Se – Te

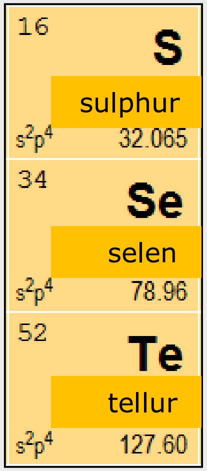


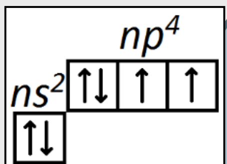


Photosensitivity of chalcogenide glasses

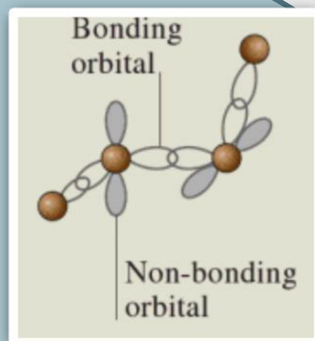
	Silica glass	Chalcogenide glass
Bangap energy	9 eV	1 - 3 eV
Glass network	rigid compact 3D 	layered 2D 
Photoinduced defects	Yes (colour centres)	Yes (excitons)
Photoinduced structural changes	No	Yes
Photoinduced refractive index change	Yes	Yes

Chalcogens: structural and electronic properties

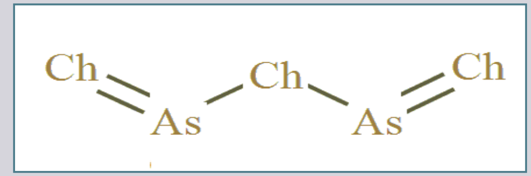




Lone-pair electrons

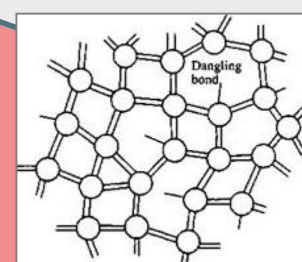


Stoichiometric composition:



Homopolar (“wrong”) bonds:
As-As, S-S, Se-Se etc.

Dangling bonds



Chalcogenide glasses – structural and electronic properties

Homopolar bonds

Dangling bonds

Lone-pair electrons

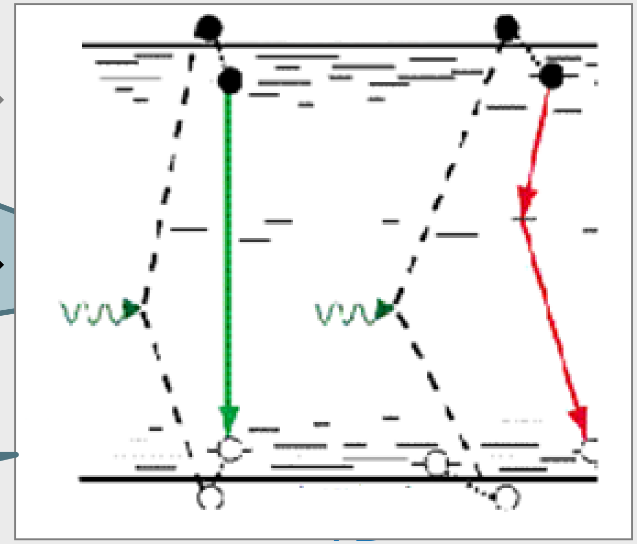
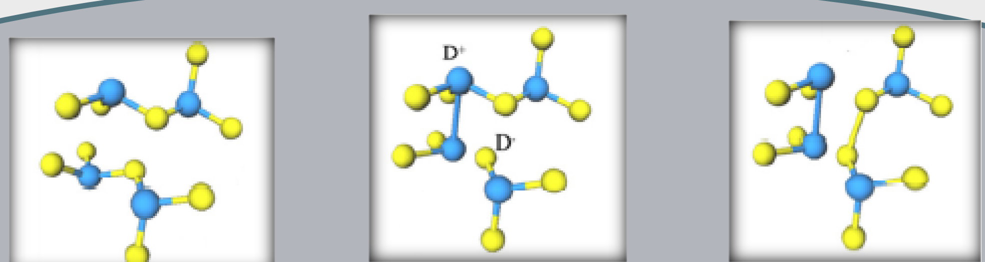


Photo-induced rearrangement of bonds



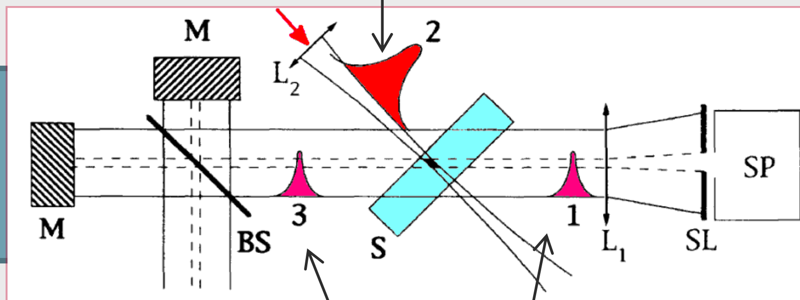
Outline:

- Interferometric pump-probe method for investigation of the nonlinear optical response of chalcogenide glasses
- Results of measurements of the non-linear optical response of the glass samples of the system As-S-Se near and far from the fundamental absorption band edge
- Numerical modeling and analysis of charge carriers kinetics upon a femtosecond pulse illumination and comparison with the experimental results

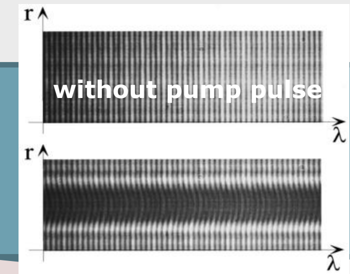
Measurement of the non-linear optical response of chalcogenide glasses

Wavelength: 800 nm; Pulse energy: 0.1-15 μJ; FWHM pulse duration: 50 fs

pump pulse



probe pulses



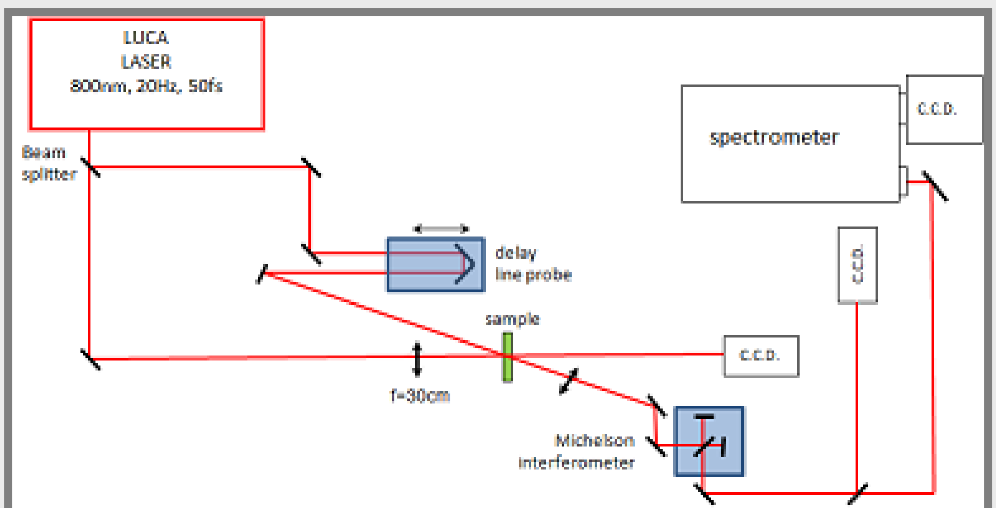
$$I(\omega) = I_0(\omega)\left(1 + T + 2T\cos(\omega\Delta t + \Delta\Phi)\right)$$

Phase shift

$$\Delta\Phi(r,t) = \Delta n(r,t)\cdot 2\pi L/\lambda$$

Absorbance

$$A = 1 - T$$

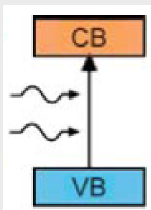


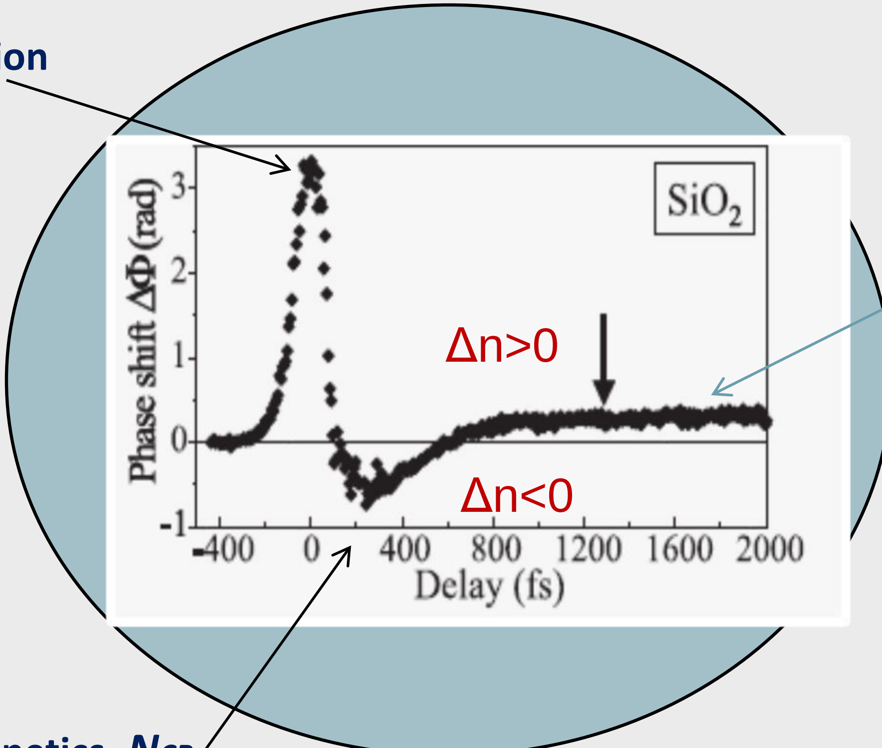
S. Guizard, *et al.*: Dynamics of femtosecond laser interactions with dielectrics, *Applied Physics,* A 79, pp.1695–1709, 2004.

Time-resolved non-linear optical response

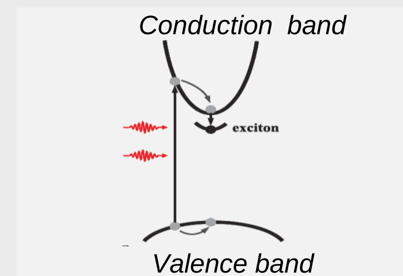
A single laser pulse irradiation for each time delay

Cross-phase modulation of the probe pulse





Trapped electrons, N_{tr}



Free electrons kinetics, N_{CB}

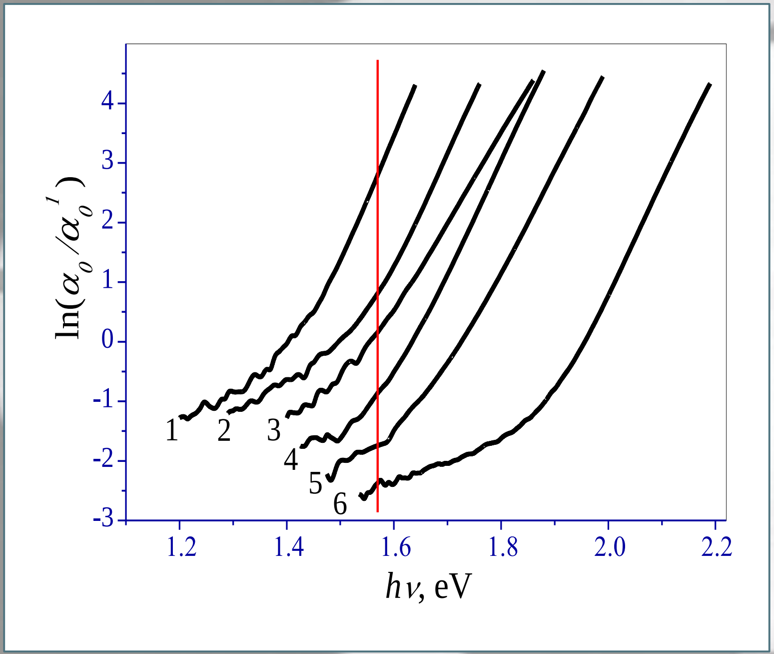
$$\Delta n(r,t) \approx n_2 I_p + \frac{e^2}{2n_0\varepsilon_0}\left\{-\frac{N_{CB} f_{CB}}{m^*\omega^2} + \frac{N_{tr} f_{tr}}{m(\omega_{tr}^2 - \omega^2)}\right\}$$

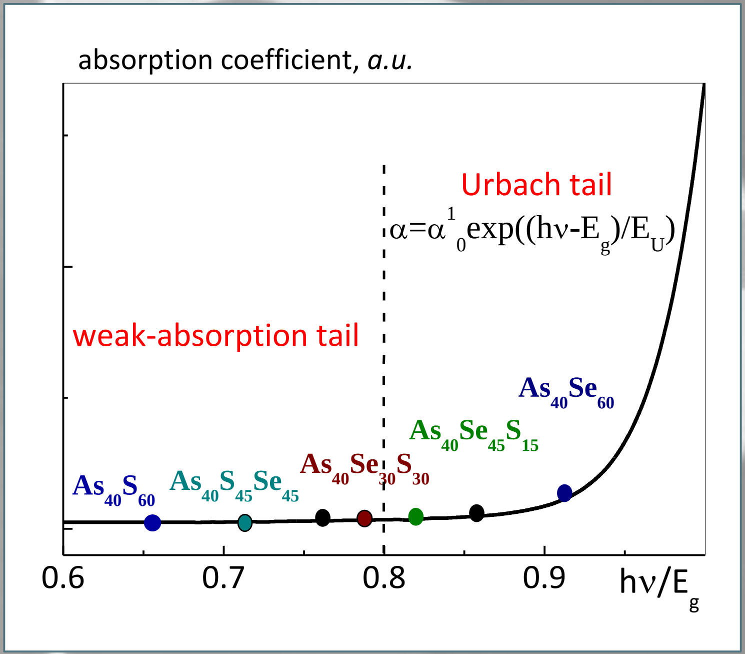
$$\Delta\Phi(r,t) = \Delta n(r,t)\cdot k \cdot L$$

S. Guizard, *et al.*: Dynamics of femtosecond laser interactions with dielectrics, *Applied Physics,* A 79, pp.1695–1709, 2004.

Chalcogenide glass compositions used in the experiment:

$As_{40}S_xSe_{60-x}$ (x = 0, 10, 15, 20, 30, 40, 45, 60)
University of Nottingham



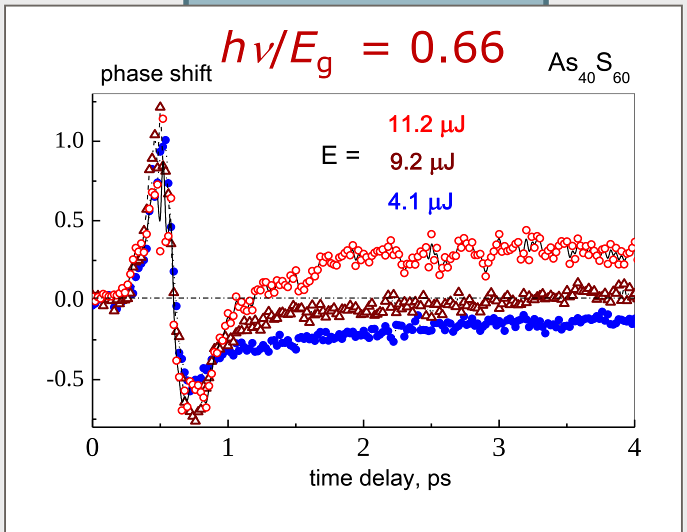


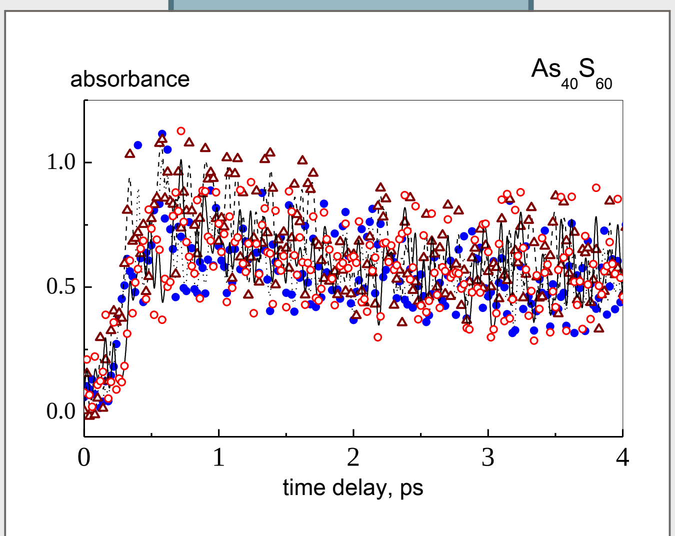
Эль Лисицкий. Проун "Большой круг". 1922-1923 гг.

Elena A. Romanova, Yulia S. Kuzyutkina, Andrey I. Konyukhov, Nabil Abdel-Moneim, Angela B. Seddon, Trevor M. Benson, Stephane Guizard, Alexandros Mouskeftaras, Opt. Eng. 53 (7), 071812, 2014

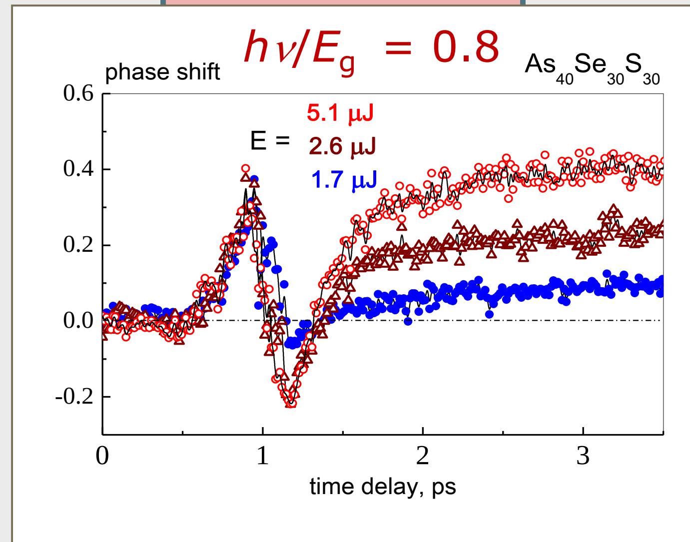
Time-resolved non-linear optical response

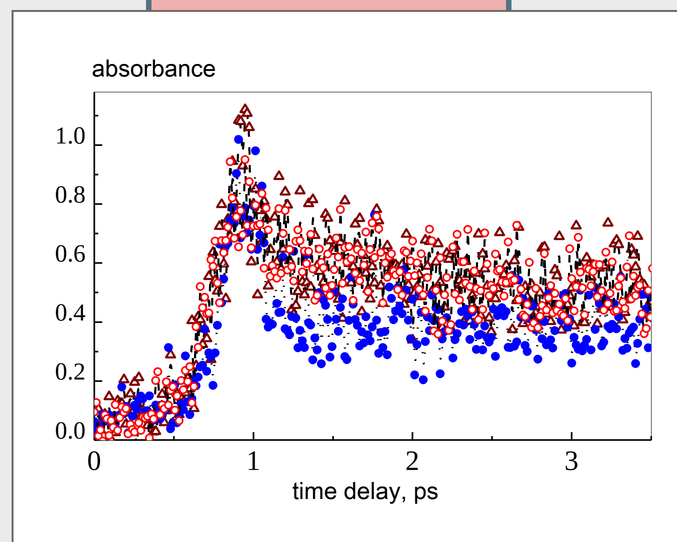
Far from the FAB edge, $h\nu/E_g < 0.8$



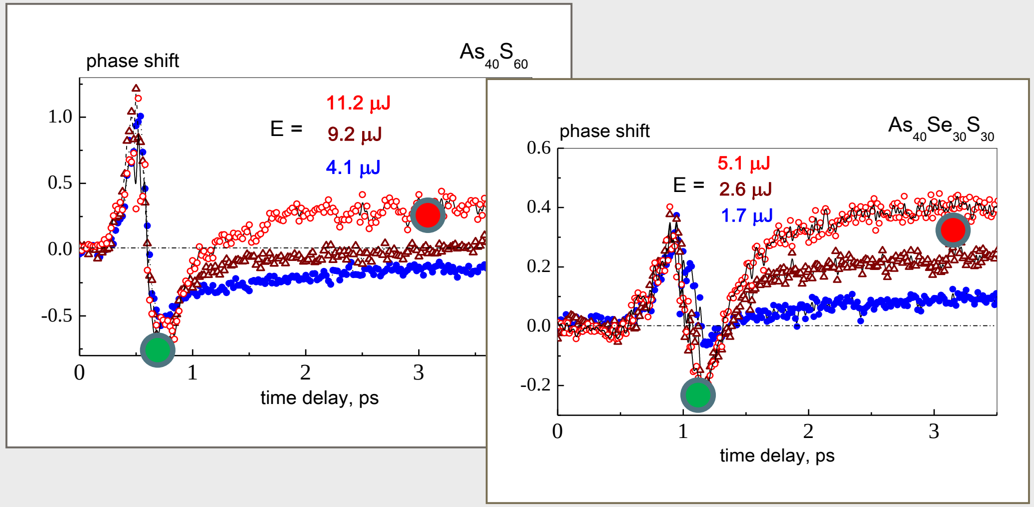


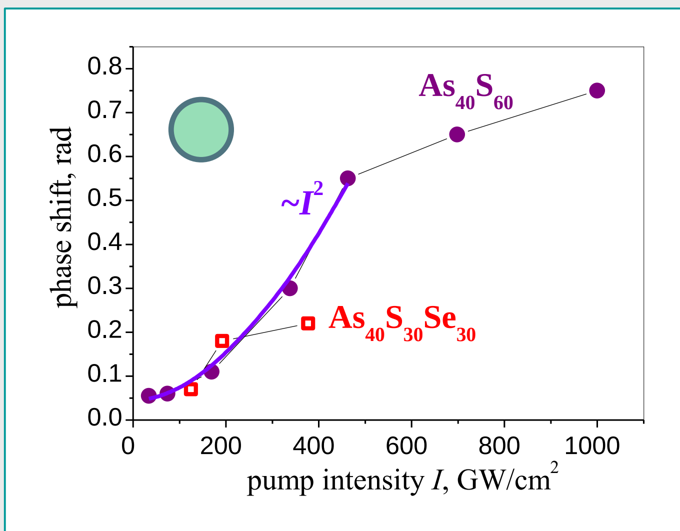
Near the FAB edge, Urbach tail

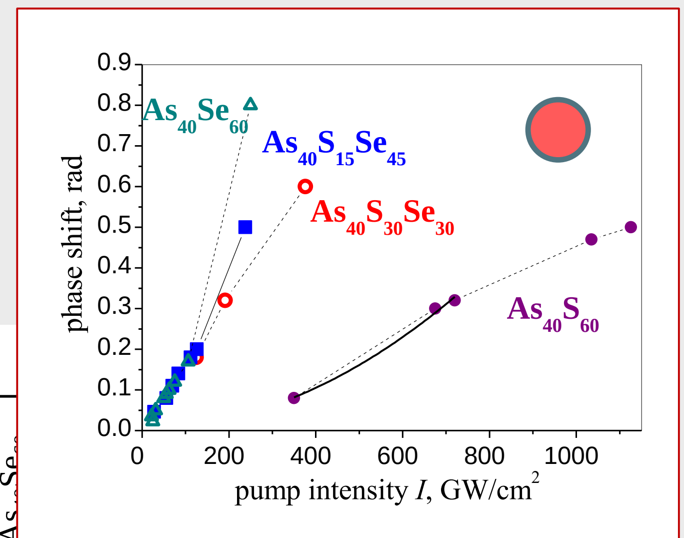




Time-resolved non-linear optical response







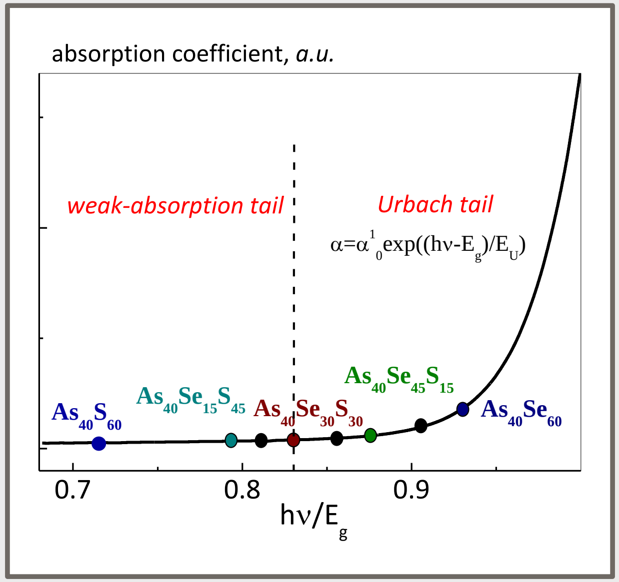
	A	$As_{40}Se_{15}S_{45}$	$As_{40}Se_{30}S_{30}$	$As_{40}Se_{45}S_{15}$	As Se
α, cm^{-1}	0.1	0.2	0.4	2.2	17

β_2 = 2-3 cm/GW

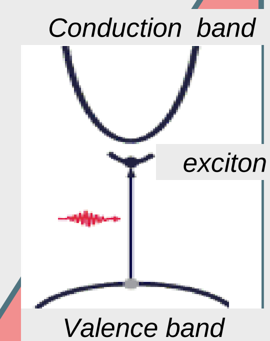
$$\beta_2 I^0_{pump} > \alpha$$

Charge carriers kinetics

Far from the FAB edge,
$h\nu/E_g < 0.8$



Near the FAB edge,
Urbach tail

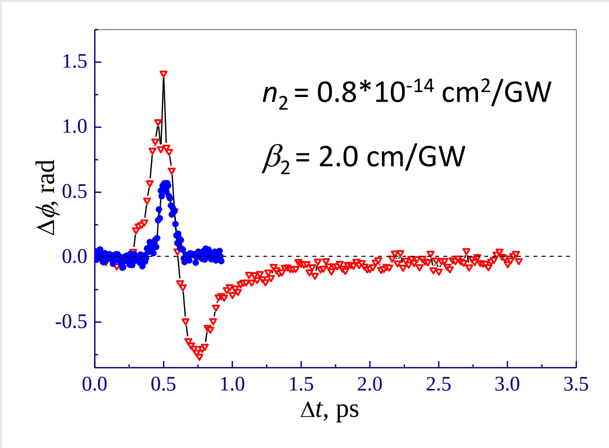


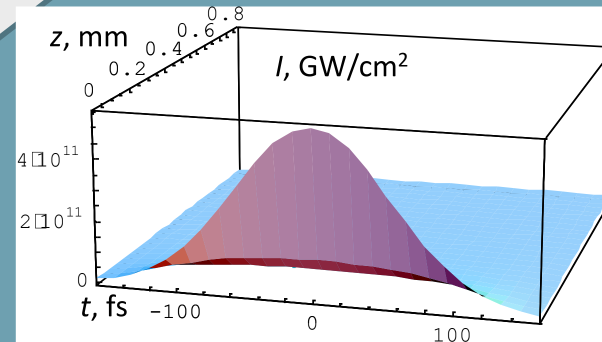
$$dn_{fh}/dt = \sigma_2 I^2/N_c - n_{fh}/\tau$$
$$dn_{th}/dt = dn_{fh}/\tau$$
$$dn_e/dt = \sigma_2 I^2/N_c - (\sigma_c/N_c)n_e(n_{th}-n_{tr})$$
$$dn_{tr}/dt = (\sigma_c/N_c)n_e(n_{th}-n_{tr})$$

$$dn_{fe}/dt = \sigma_2 I^2/N_c - n_{fe}/\tau$$
$$dn_{tr}/dt = \sigma_1 I + n_{fe}/\tau$$

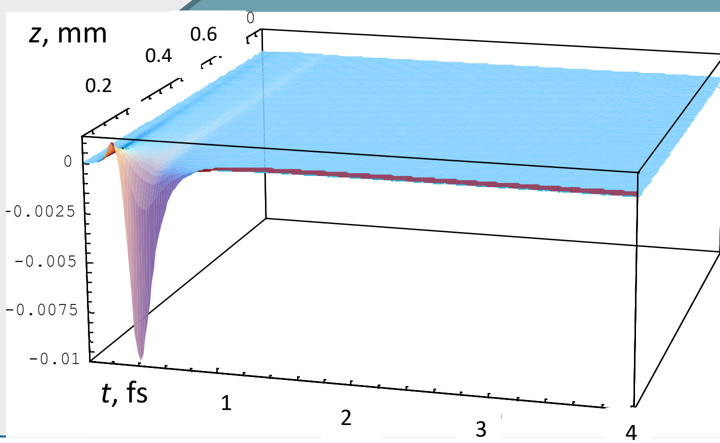
Charge carriers kinetics

Far from the FAB edge,
$As_{40}S_{60}$, $h\nu/E_g = 0.66$

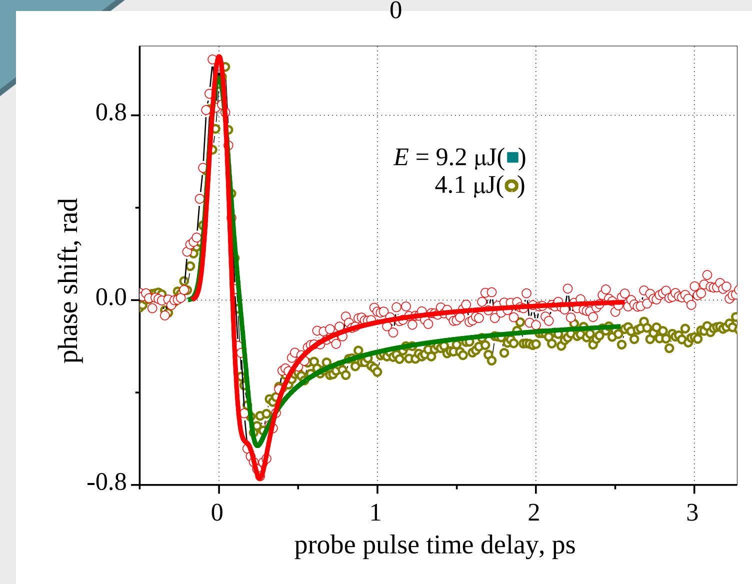




$$\Delta\varphi = k\int_0^L \Delta n(t,z)dz$$

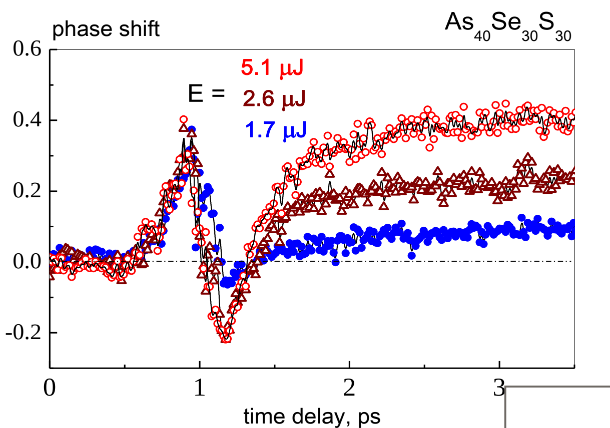


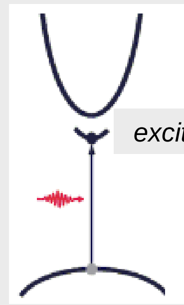
$$\Delta n(t,z) \approx n_2 I_p(t,z) + \frac{e^2}{2n_0\varepsilon_0}\left\{-\frac{N_{CB}(t,z)f_{CB}}{m^*\omega^2} + \frac{N_{tr}(t,z)f_{tr}}{m(\omega_{tr}^2-\omega^2)}\right\}$$



Time-resolved non-linear optical response

Near the FAB edge

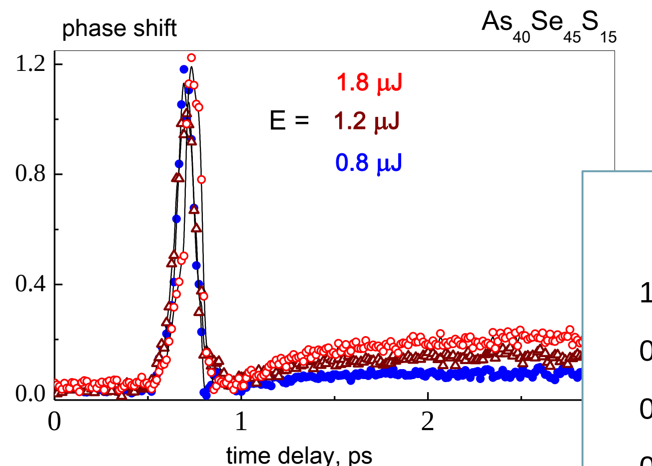




n_2=0.4*10^{-14} cm^2/W

β_2=2.7 cm/GW

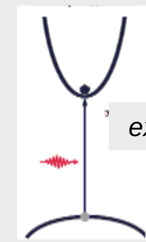
$h\nu/E_g$=0.8

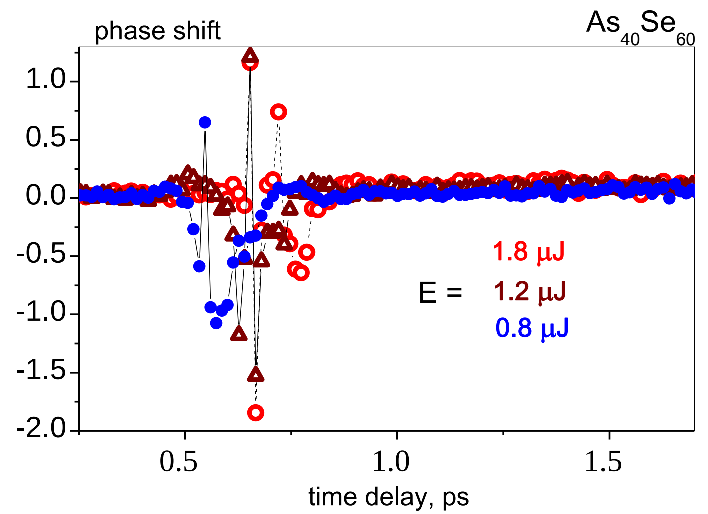


n_2=0.6*10^{-14} cm^2/W

β_2=4.0 cm/GW

$h\nu/E_g$=0.82



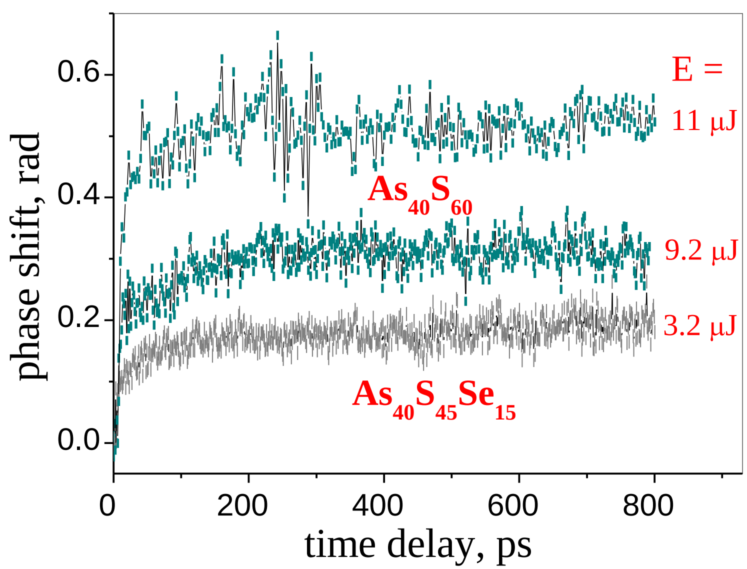


$h\nu/E_g$=0.88

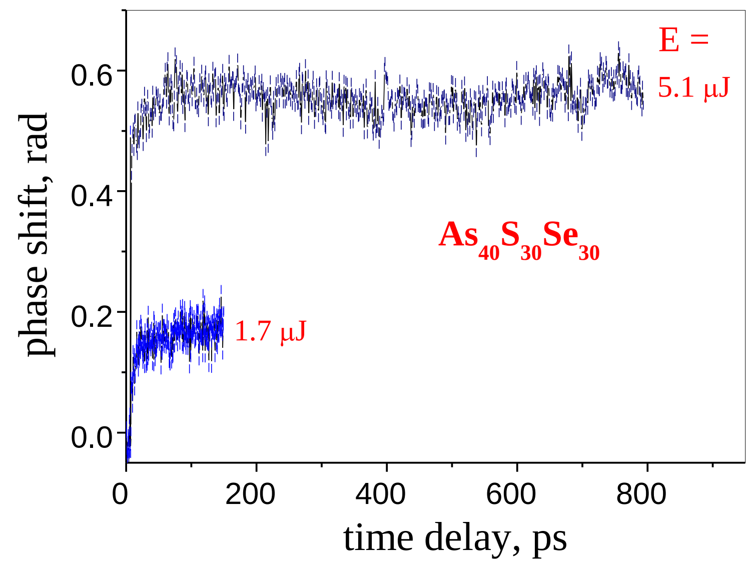
Time-resolved non-linear optical response

Long time-scale dynamics

Far from the FAB edge



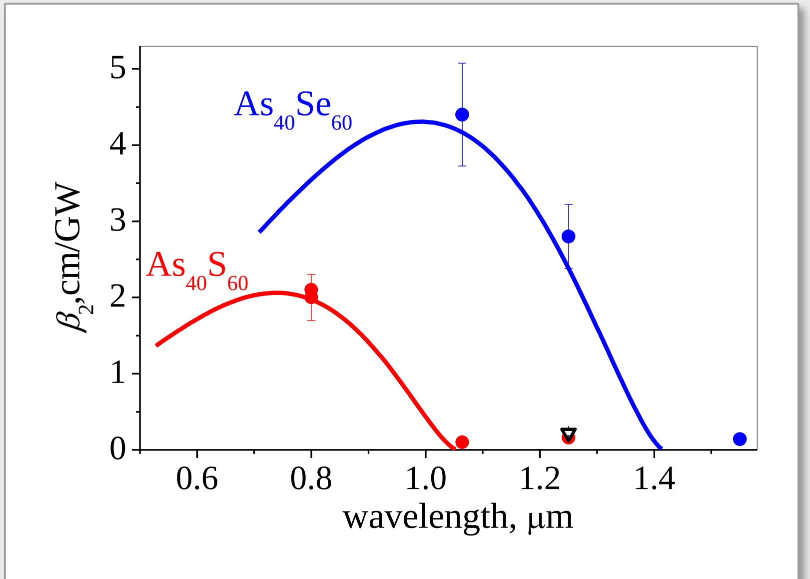
Near the FAB edge

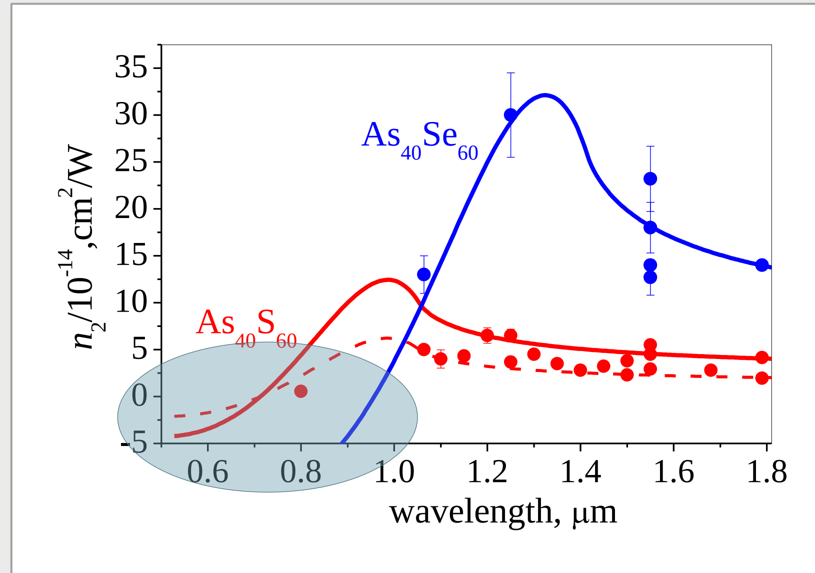


CONCLUSIONS

- The non-linear refractive index of chalcogenide glasses of the system As-S-Se is positive-valued near the fundamental absorption band edge.
- If the laser pulse frequency corresponds to the WAT range (far from the FAB edge), the time of electrons trapping depends on the pump pulse energy and is greater than the pulse duration. The charge carriers kinetics is well described by the two-stage process of holes trapping followed by electrons trapping.
- If the laser pulse frequency corresponds to the Urbach tail, the time of electrons trapping does not depend on the pump pulse energy and is decreasing with increase of the ratio of the laser photon energy to the bandgap energy.
- A “permanent” change of the refractive index has been registered over the range of the probe pulse delays up to 1ns. For irradiation far from the FAB edge, there is a threshold for such “permanent” modifications.

Non-linear optical response of chalcogenide glasses:
Comparison with the crystalline semiconductors





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