Transient phenomena in scintillation materials

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Outline

- Motivation
- Experimental technique
- Results on GAGG:Ce
  - Experimental results on differential optical absorption in GAGG:Ce with and without Mg codoping
  - Modeling of carrier population kinetics
- Results on LSO:Ce and LYSO:Ce
- Conclusions

This research was focused on

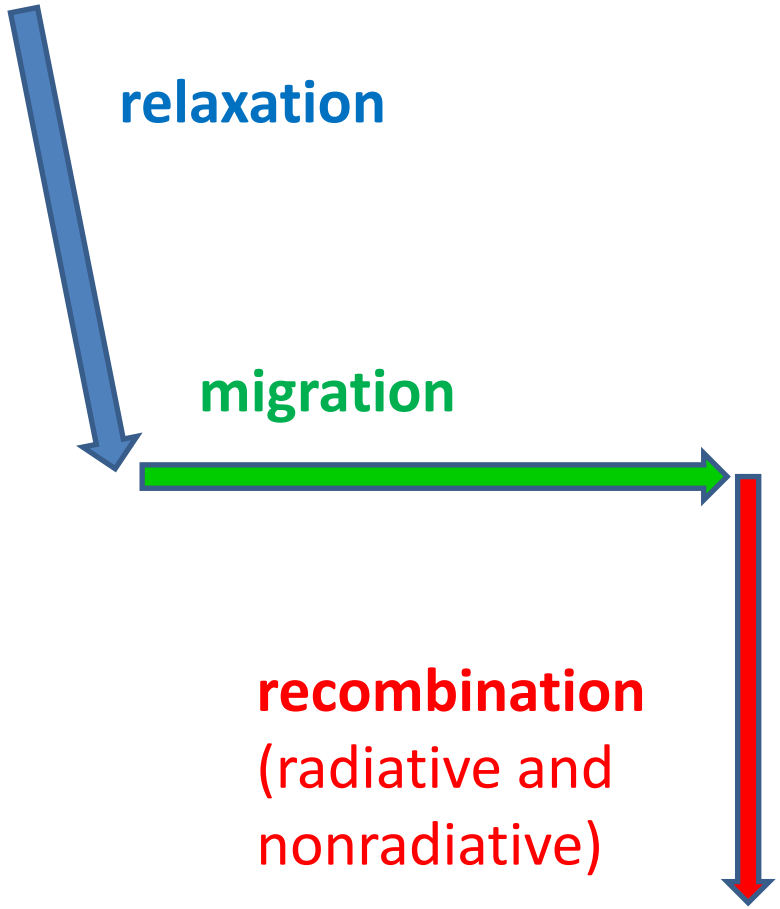
fast scintillators

CRYSTAL CLEAR

Let it be 10 ps

for
high-energy physics
and
medical imaging

Relaxation stages after excitation



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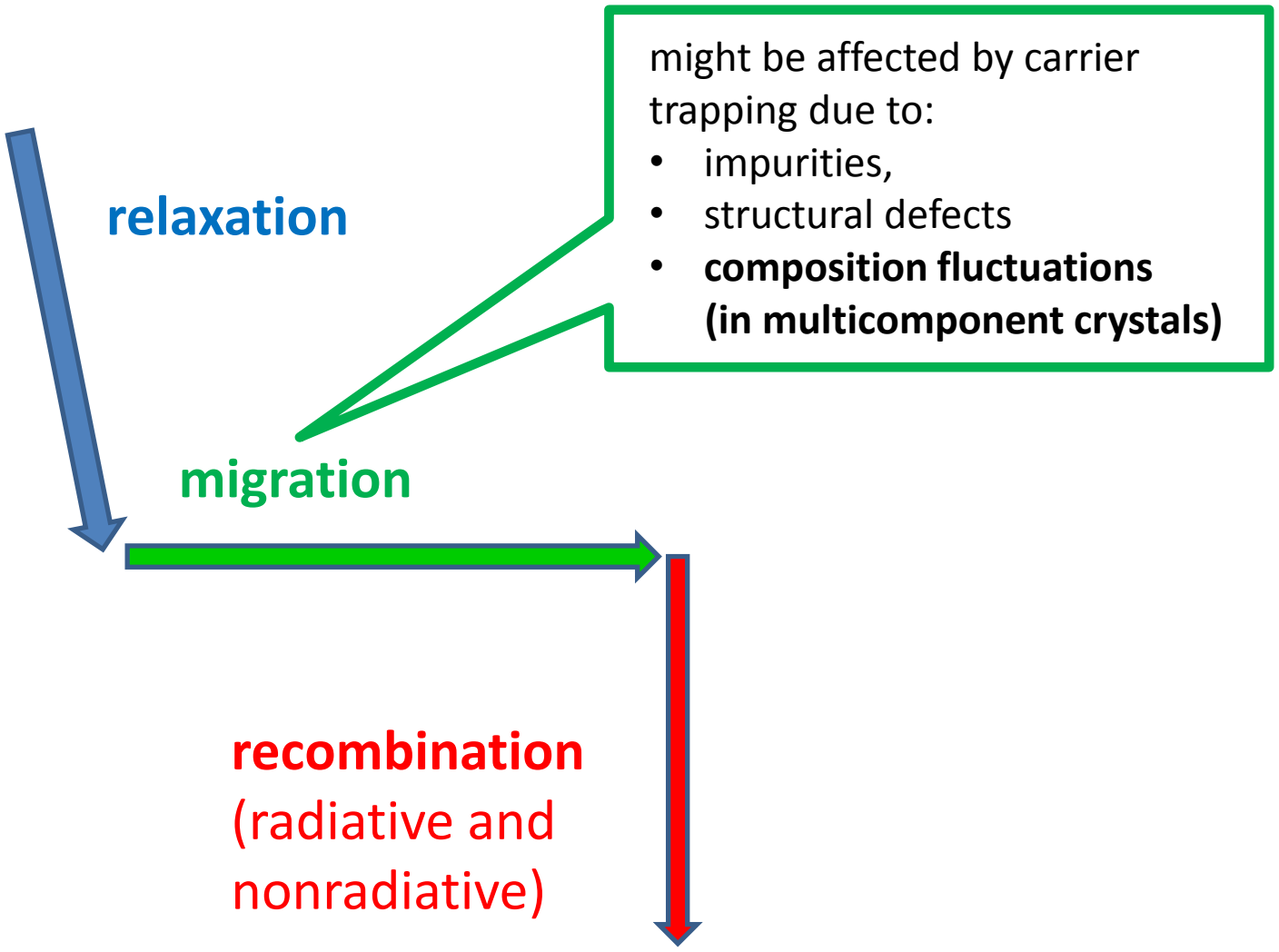

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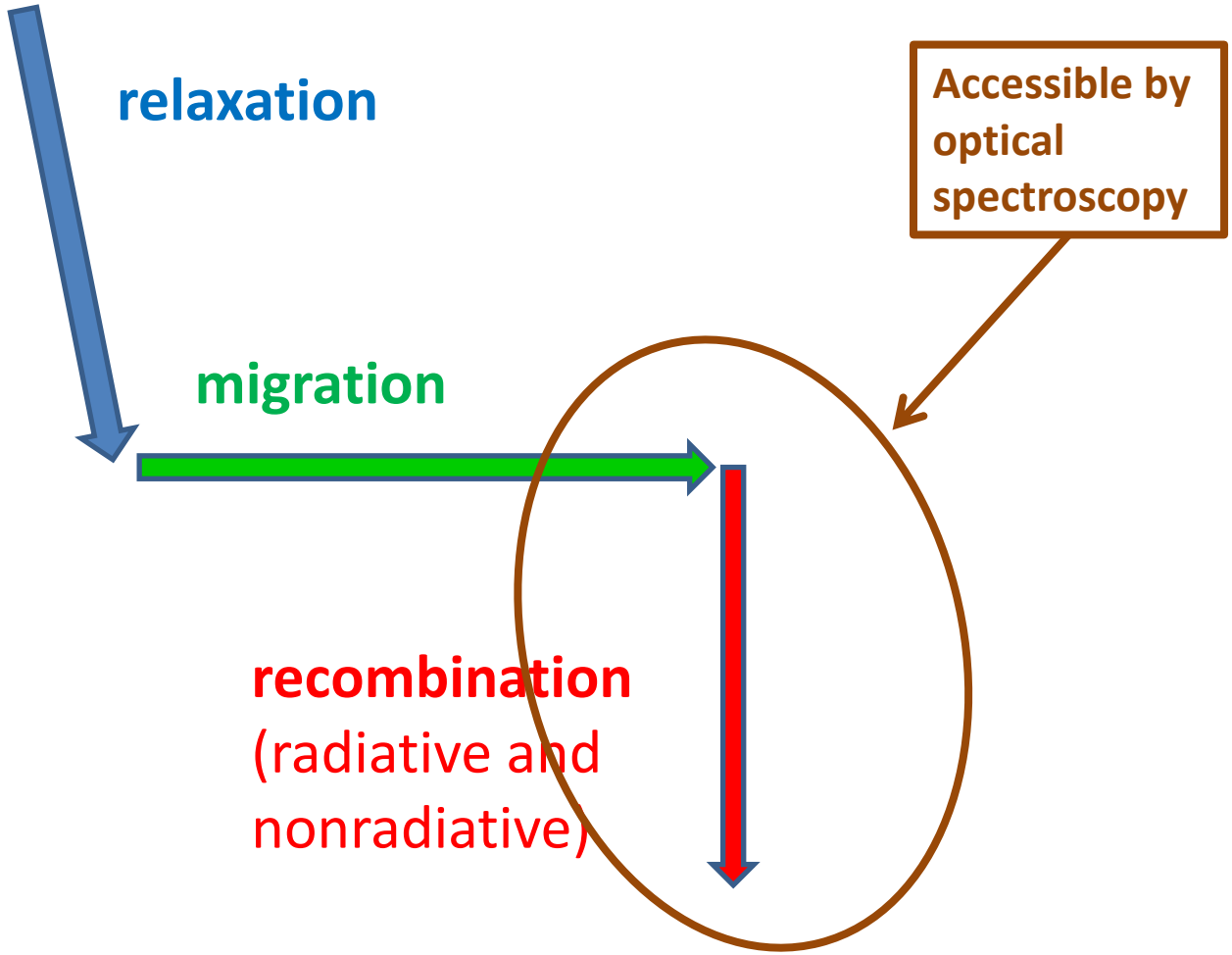
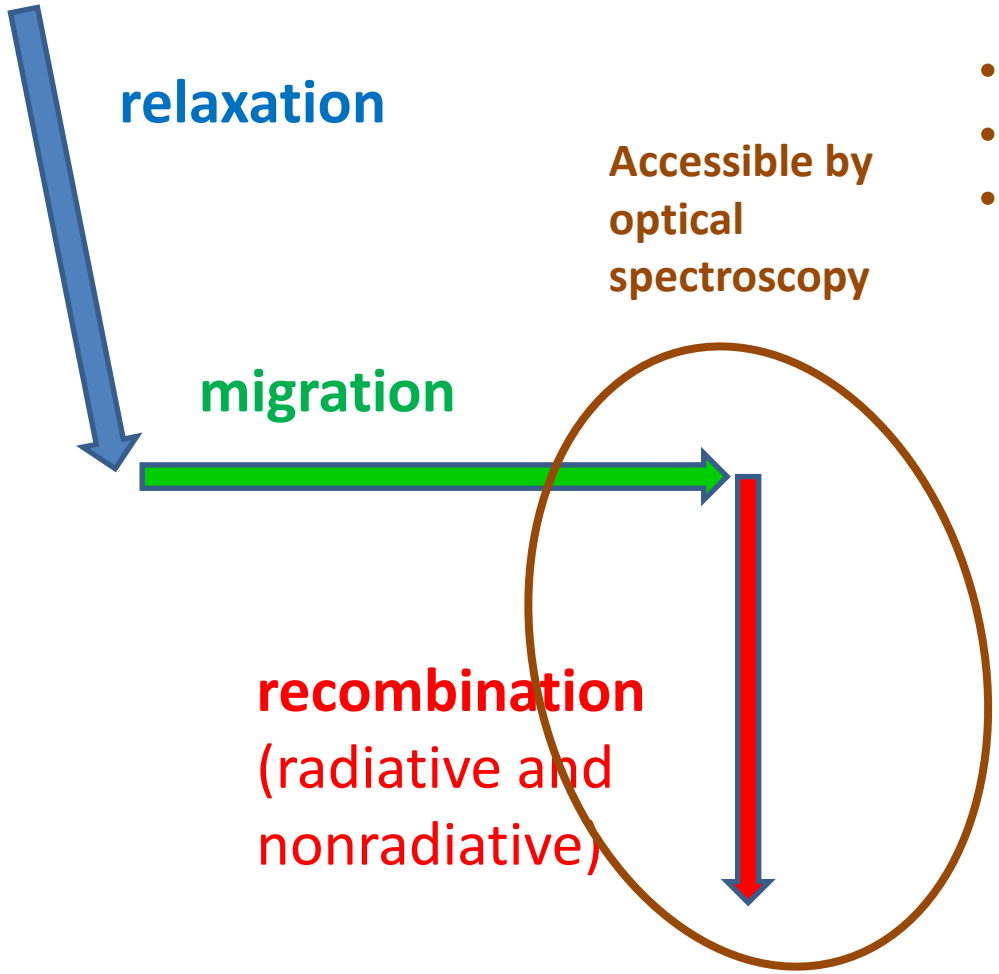
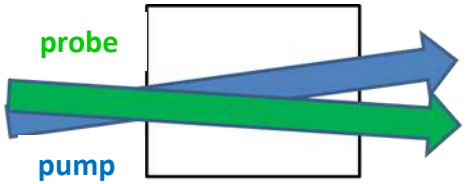


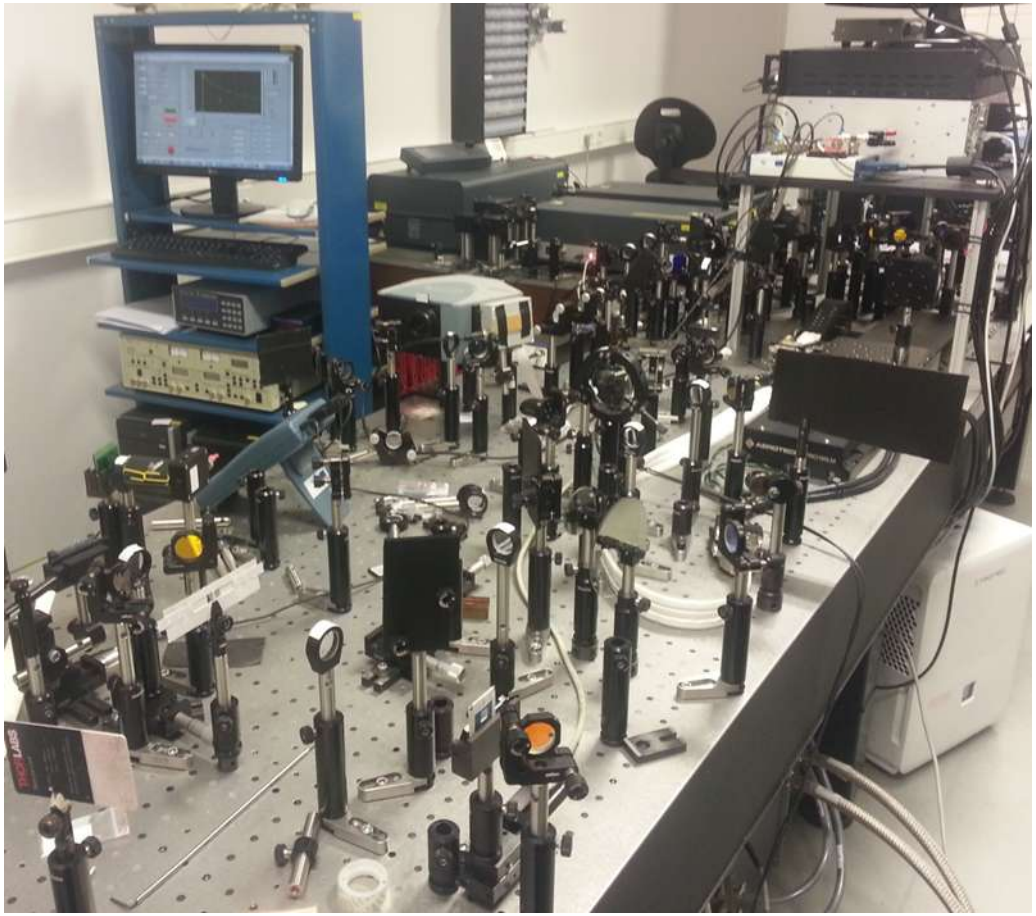
Relaxation stages after excitation



- optical absorption
- photoluminescence
- nonlinear optical absorption

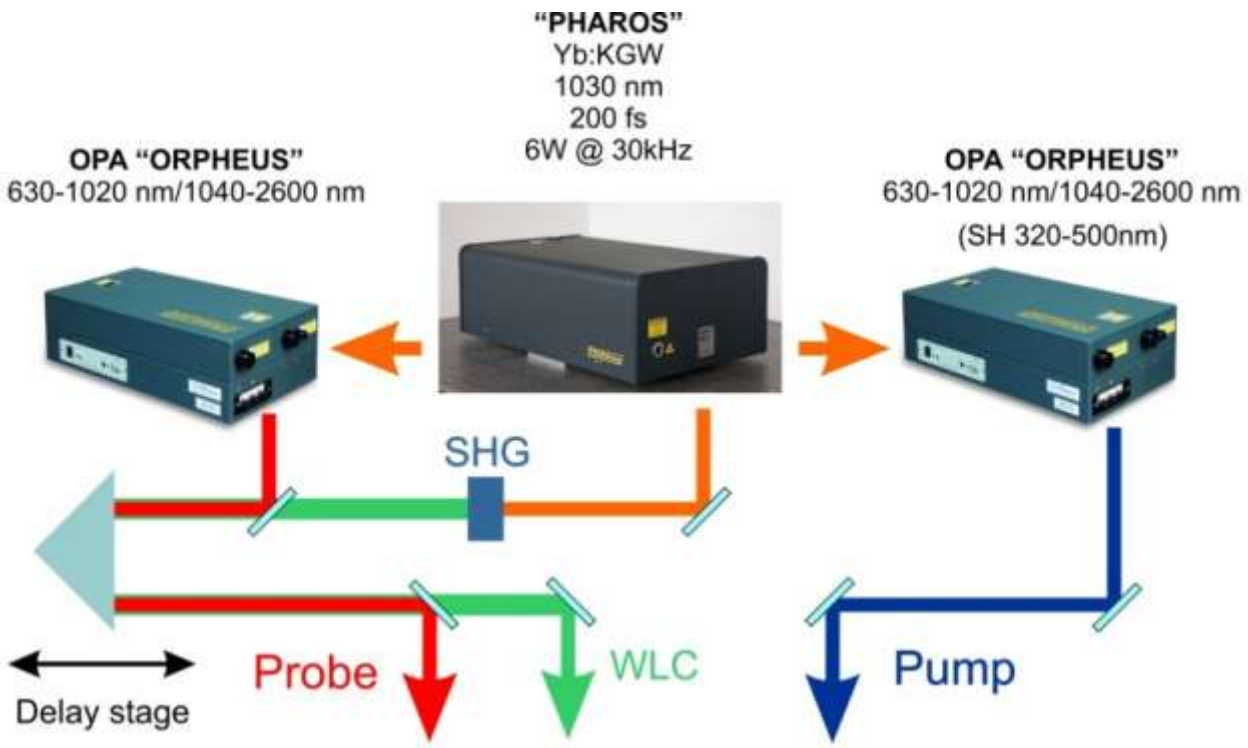
EXPERIMENTAL TECHNIQUE

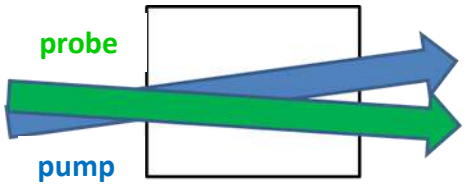




Optical pump and probe technique

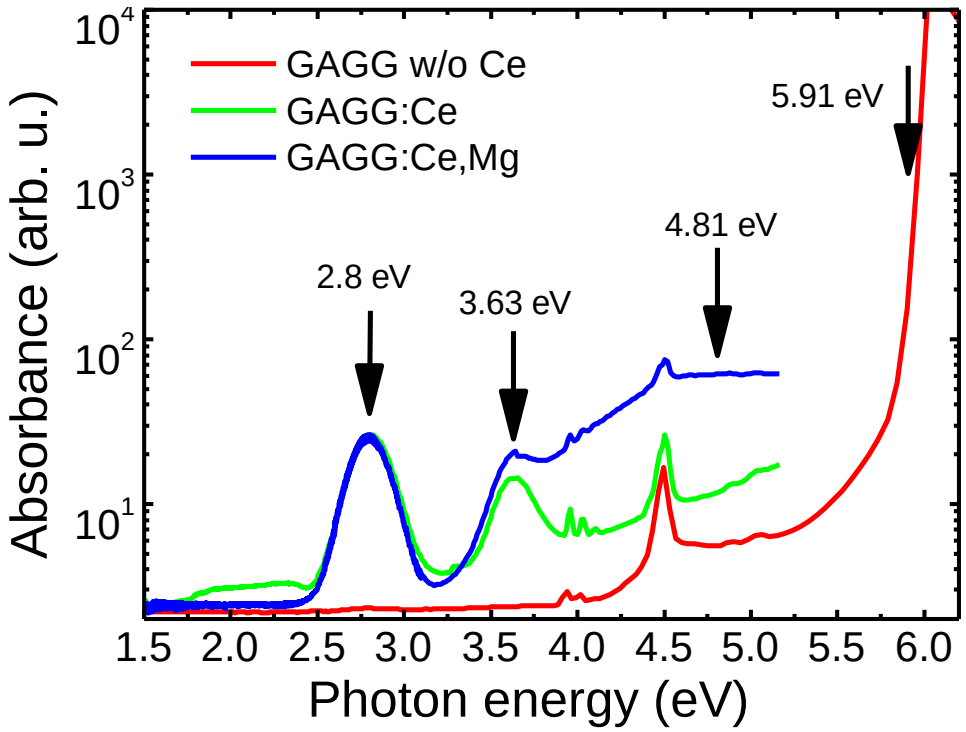
subpicosecond time resolution



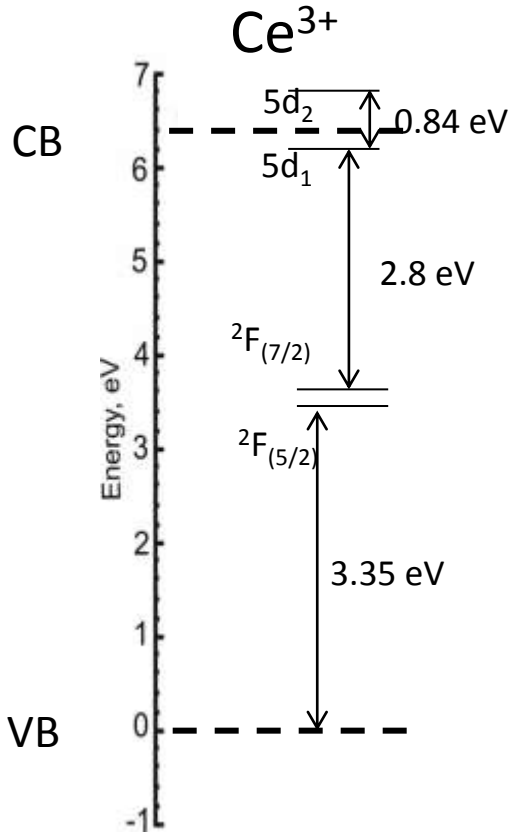


Optical pump and probe technique

selective excitation

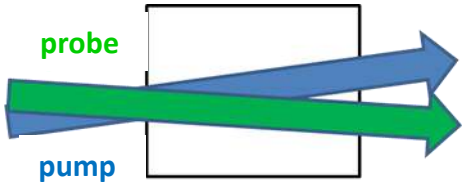


Absoption spectra of GAGG without intentional doping (courtesy of Dr. O.Sidletski), doped with Ce and codoped with Ce, Mg



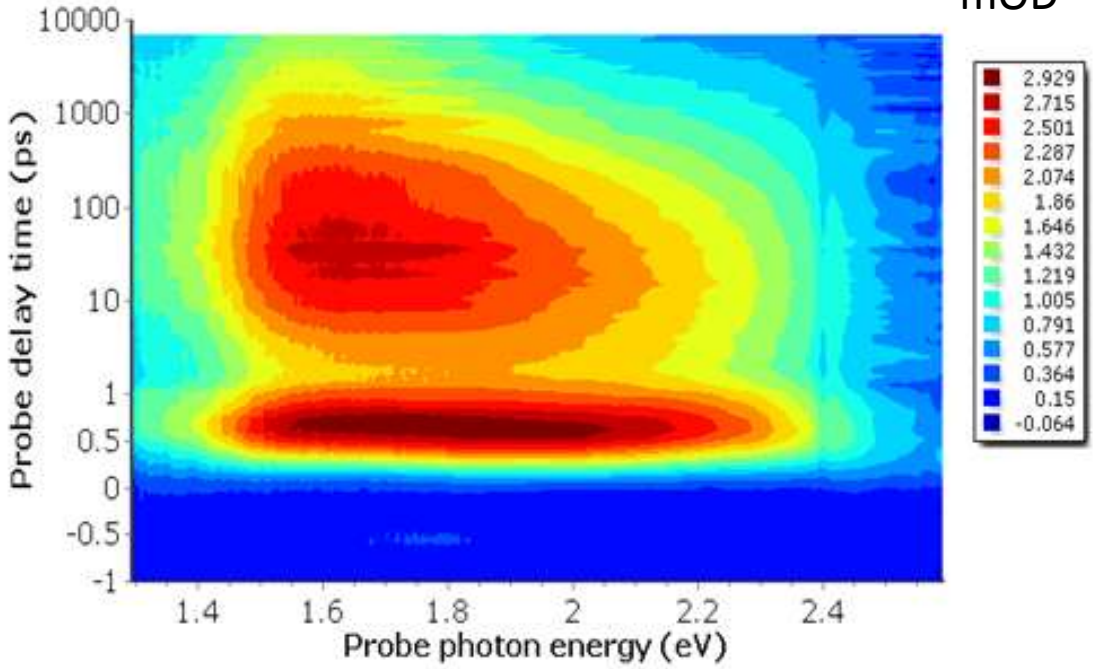
Energy levels of Ce^{3+} in respect of valence (VB) and conduction (CB) bands of GAGG

Optical pump and probe technique



revealing spectral and time features

Differential absorption, mOD

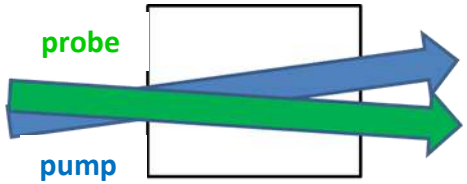


Differential absorption depends on:

- density of states
- absorption coefficient
- **density of nonequilibrium carriers**

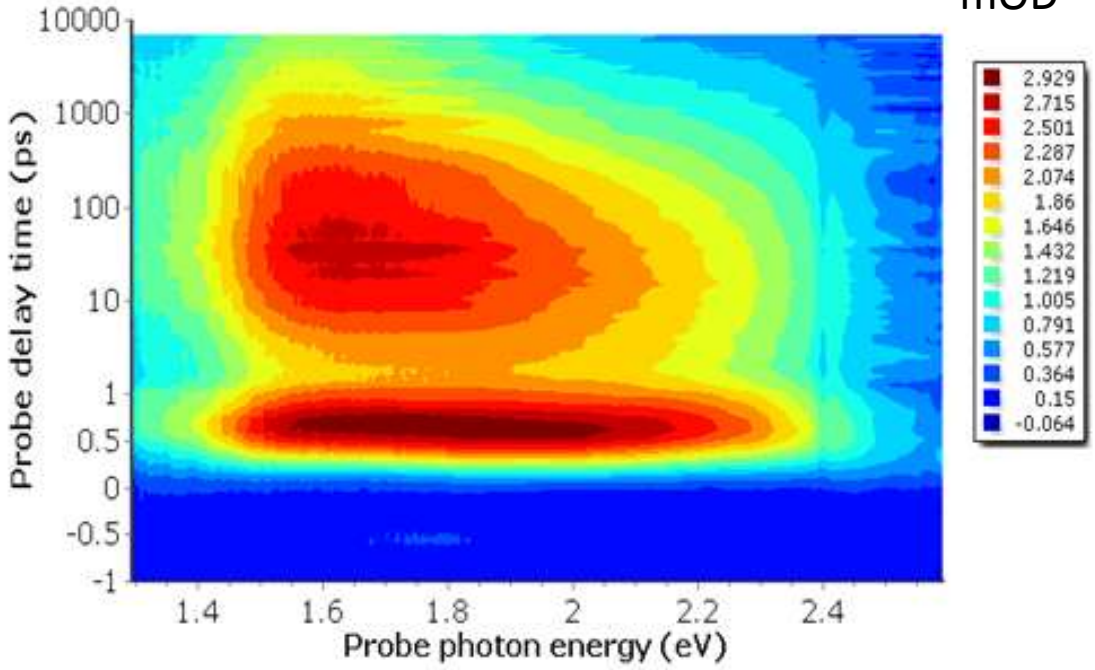
$$DAsignal = 1000 \times \log 10 \left(\frac{probeIntensitySampleUnpumped}{probeIntensitySamplePumped} \right)$$

Optical pump and probe technique



revealing spectral and time features

Differential absorption, mOD



Differential absorption depends on:

- density of states
- absorption coefficient
- **density of nonequilibrium carriers**

free,
trapped,
on emitting centers

$$DAsignal = 1000 \times \log 10 \left(\frac{probeIntensitySampleUnpumped}{probeIntensitySamplePumped} \right)$$


MATERIALS
STUDIED
MULTICOMPONENT

Multicomponent garnet-type scintillator

GAGG:Ce

gadolinium aluminum gallium garnet

$Gd_3Al_2Ga_3O_{12}$

fast
but might be
faster

garnet

Ceres,
a goddess of agriculture, grain crops, fertility and motherly relationships

Lutetium oxyorthosilicate

LSO:Ce

$Lu_2SiO_5:Ce$

fast
but expensive

Lutetium-yttrium oxyorthosilicate

LYSO:Ce

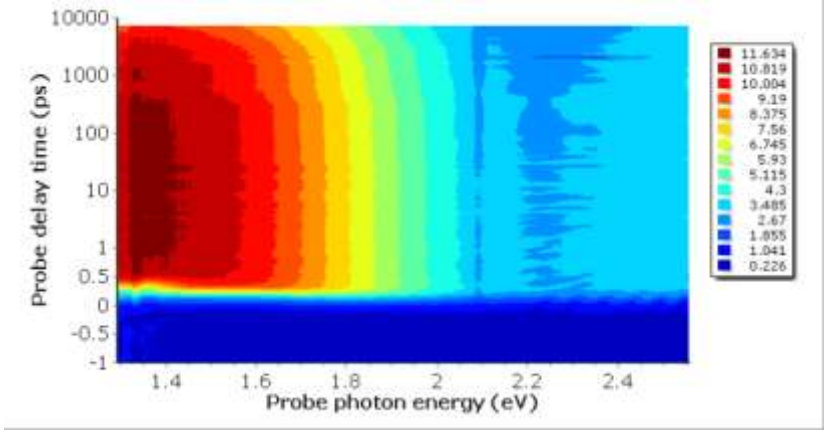
$Lu_{2(1-x)}Y_{2x}SiO_5:Ce$

less expensive
and probably
fast enough

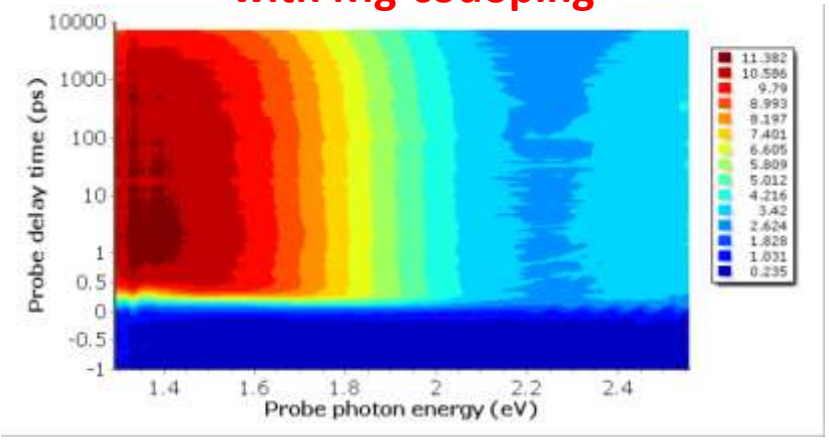
GAGG:Ce
without and with codoping

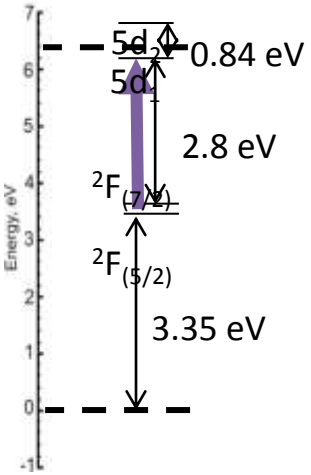
Differential absorption of GAGG:Ce as a function of probe photon energy and delay between pump and probe pulses at pump photon energy of 2.8 eV

no Mg-codoping



with Mg-codoping

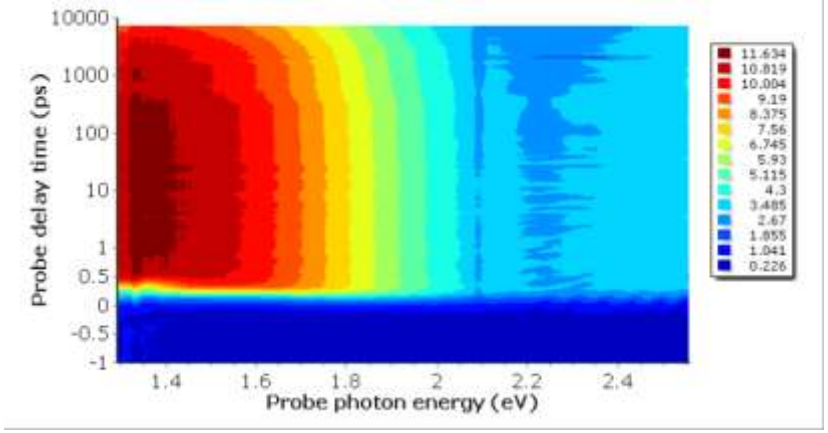




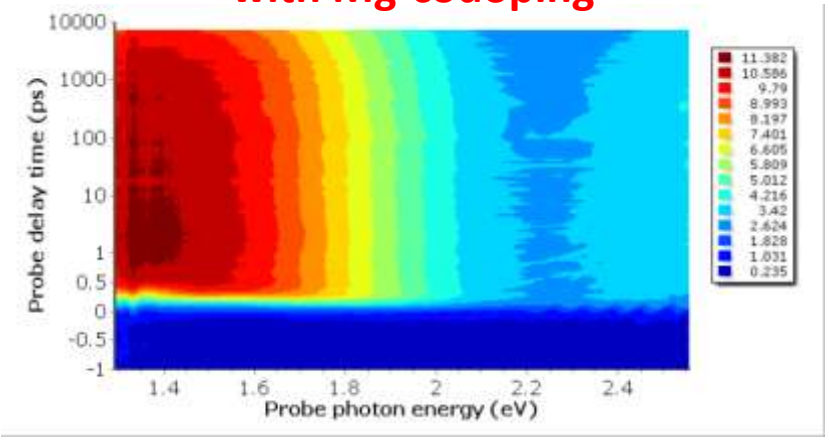
No DA in GAGG without intentional Ce doping

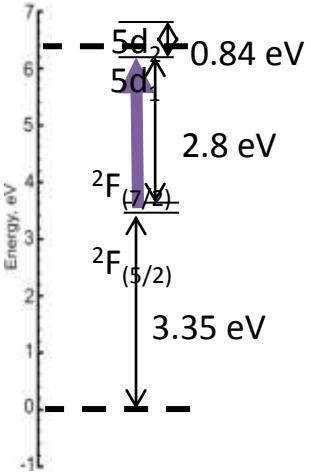
Differential absorption of GAGG:Ce as a function of probe photon energy and delay between pump and probe pulses at pump photon energy of 2.8 eV

no Mg-codoping



with Mg-codoping





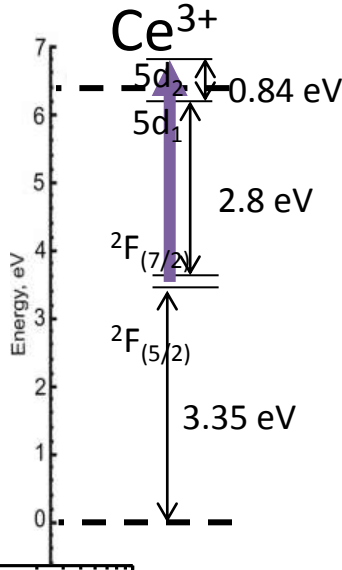
The differential absorption in the vicinity of 1.4 eV reflects the population of the lowest excited Ce^{3+} level

No DA in GAGG without intentional Ce doping

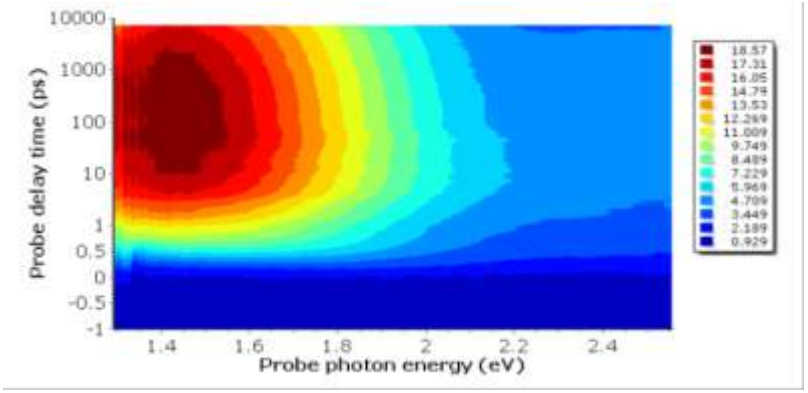
Differential absorption of GAGG:Ce as a function of probe photon energy and delay between pump and probe pulses

at pump photon energy of 3.63 eV

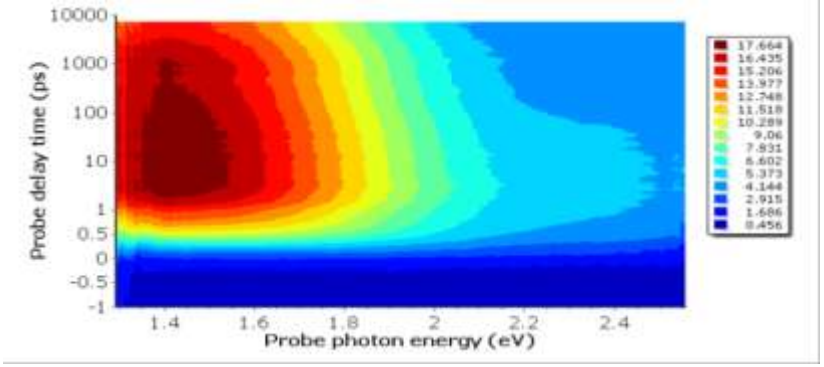
Excited electrons have two routes: intracenter relaxation and via crystal matrix

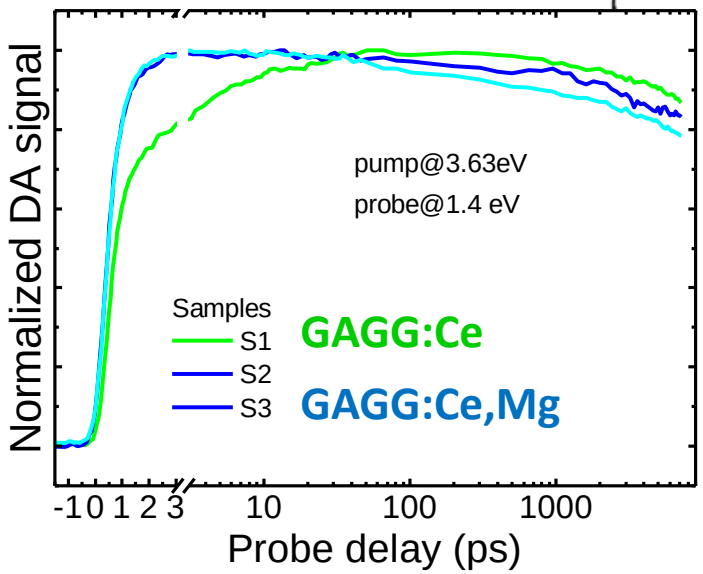


no Mg-codoping



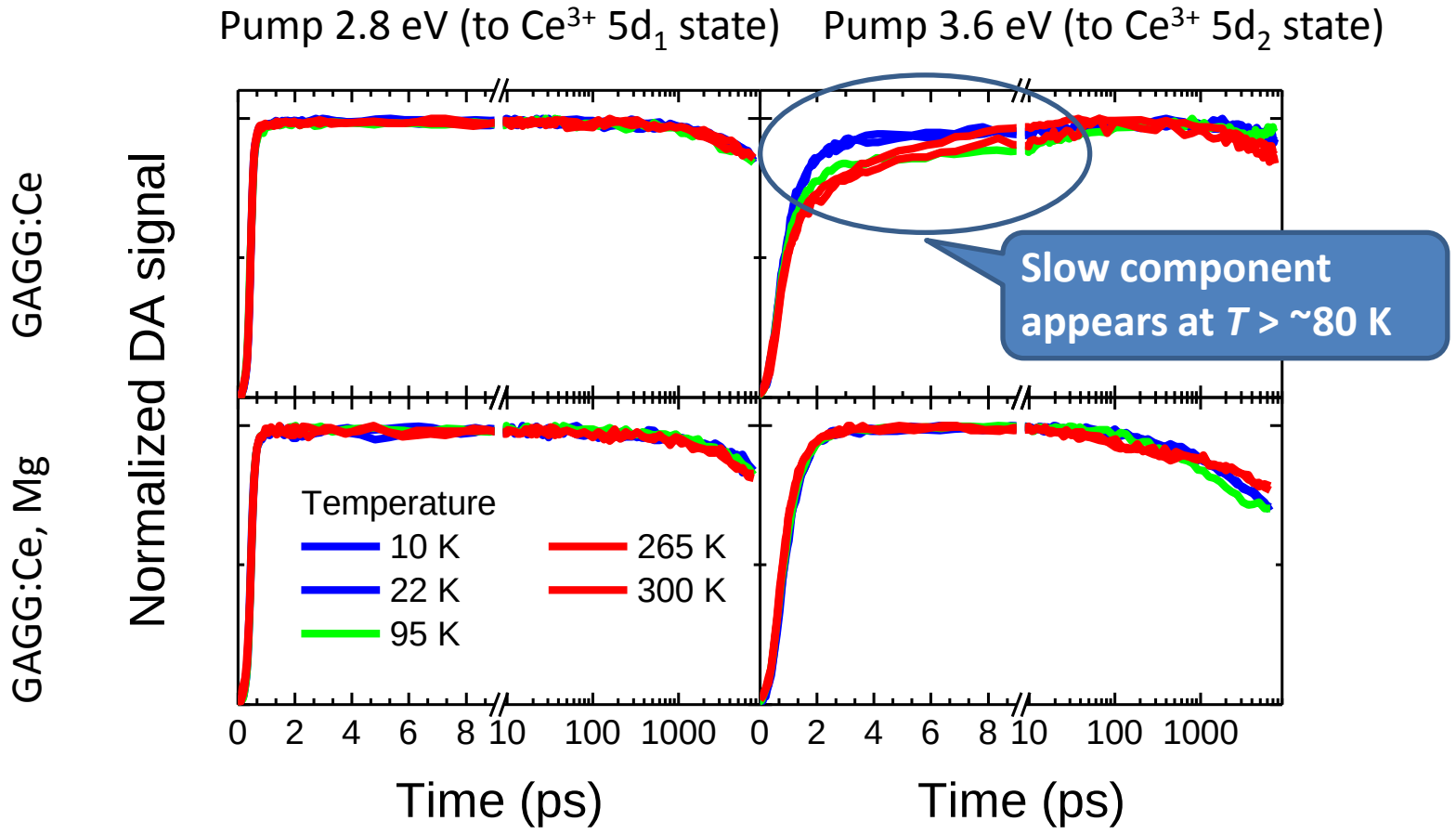
with Mg-codoping



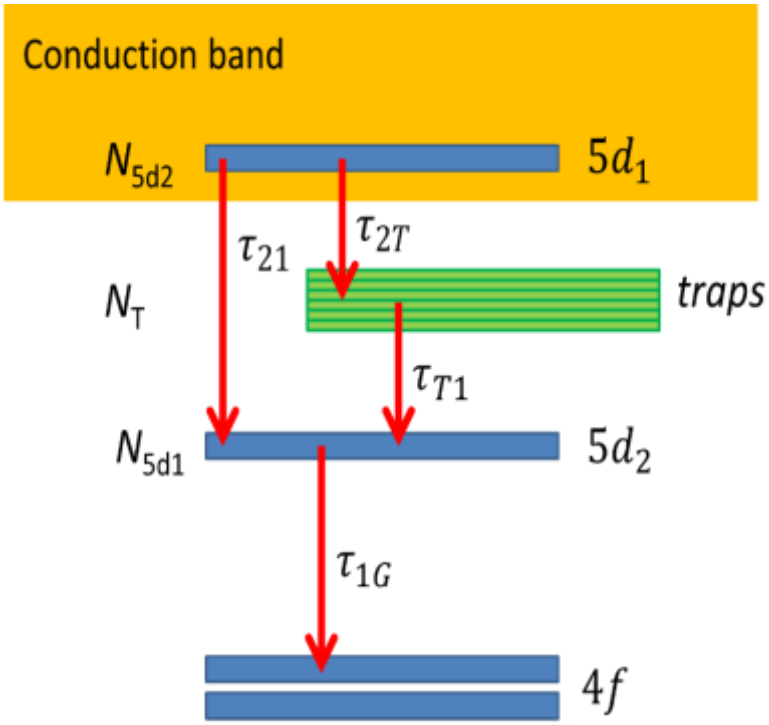


Kinetics of differential absorption at 1.4 eV in GAGG samples with different doping

Differential absorption kinetics in GAGG:Ce and GAGG:Ce,Mg at different temperatures



Simulation of population kinetics



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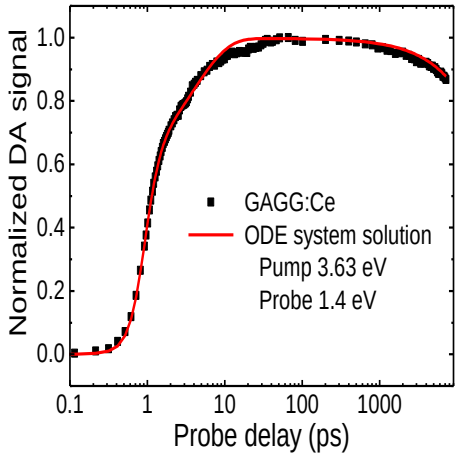
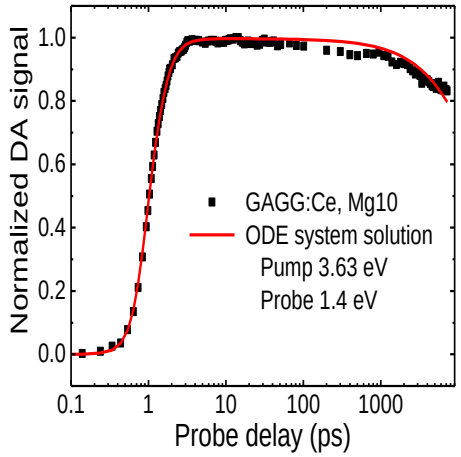
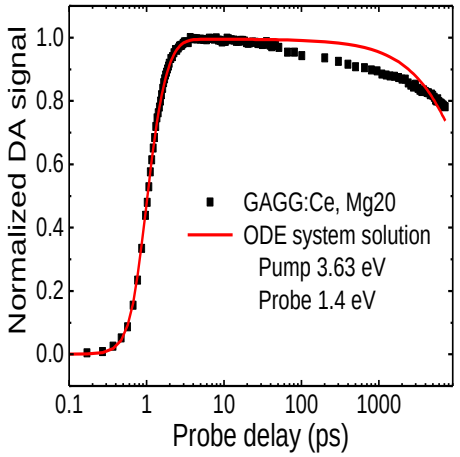
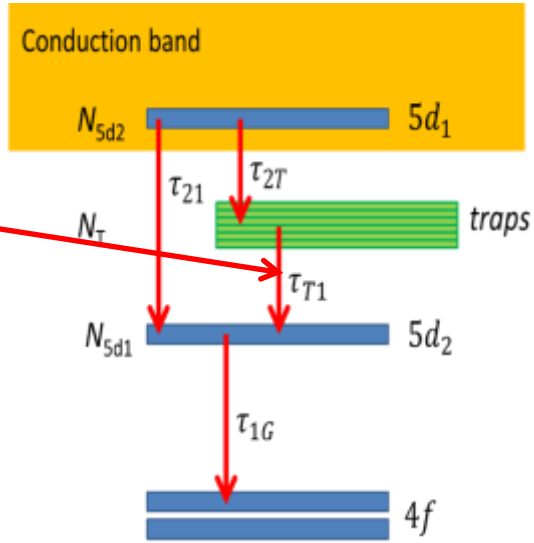






Sample	Mg content	T_{2T} (ps)	T_{T1} (ps)	T_{21} (ps)	T_{1G} (ns)
S1	-	1.05	4.09	0.52	56
S2	10	11.77	1.43	0.52	32
S3	20	89.27	1.00	0.52	24

Introduction of Mg decreases the influence of traps



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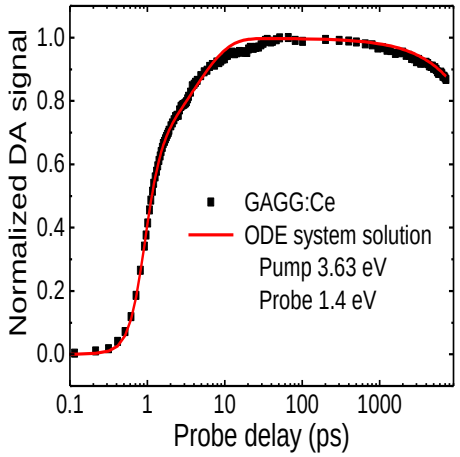

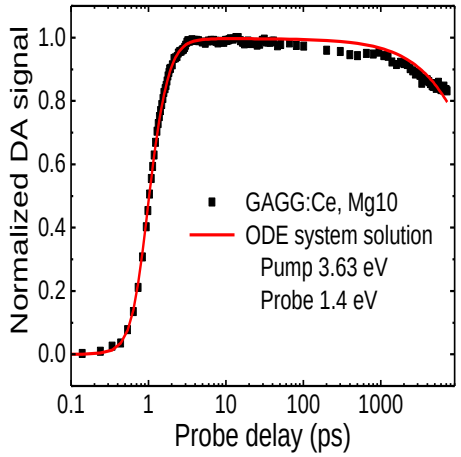

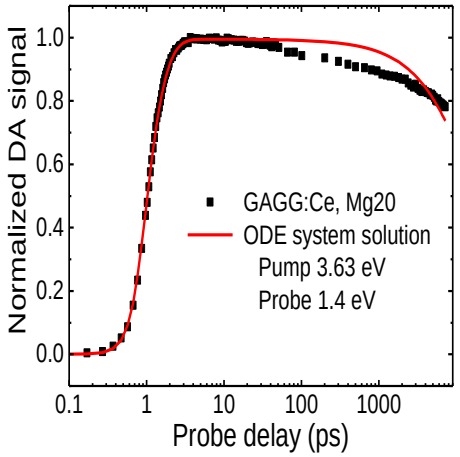

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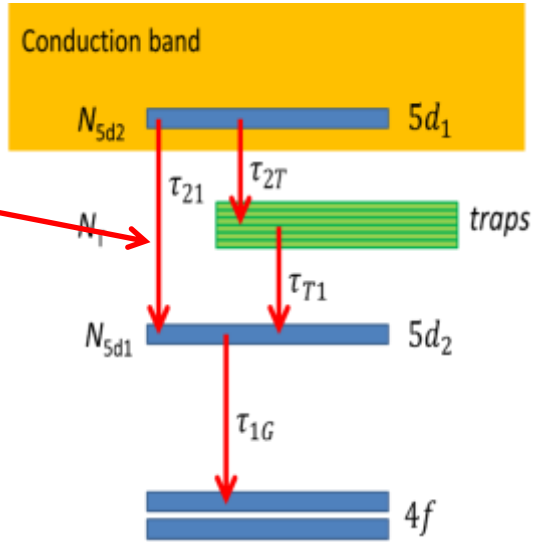
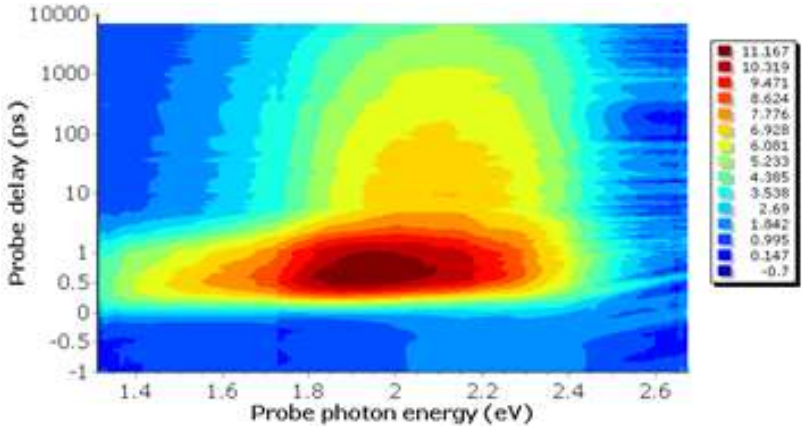


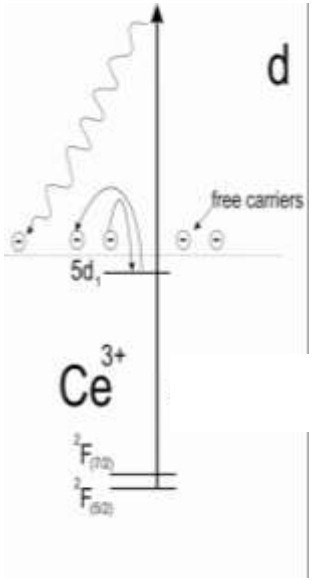
The time of intracenter $5d_2 - 5d_1$ relaxation at Ce^{3+} in GAGG:Ce equals 500 fs

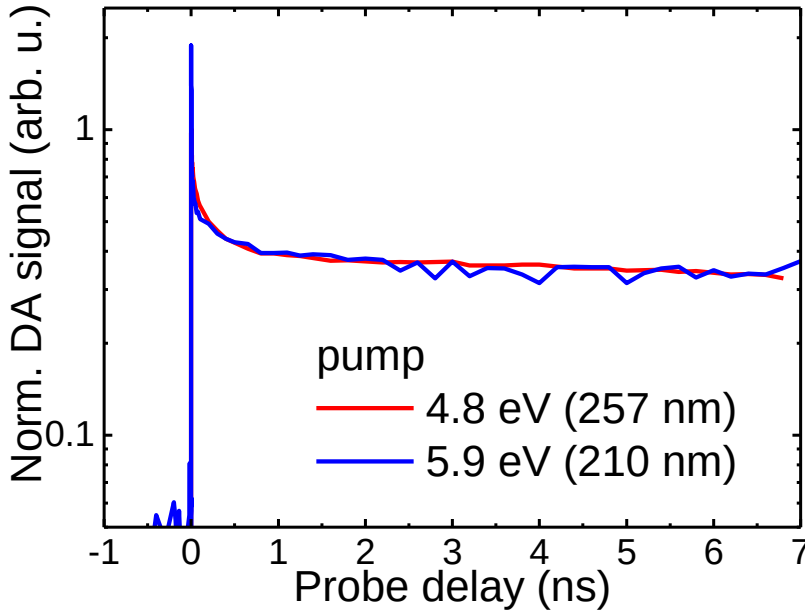
LSO:Ce

Differential absorption in LSO



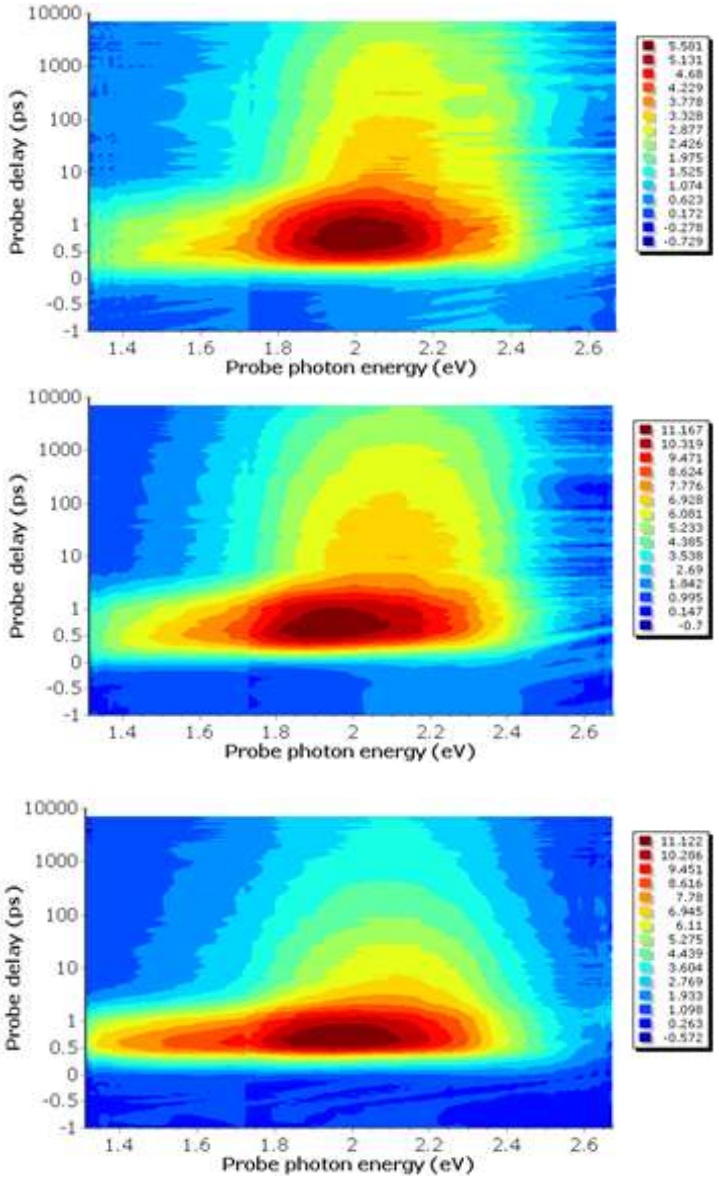
Conduction band





The fast component in the decay of differential absorption is caused by free electron absorption

Differential absorption of three LSO ingots

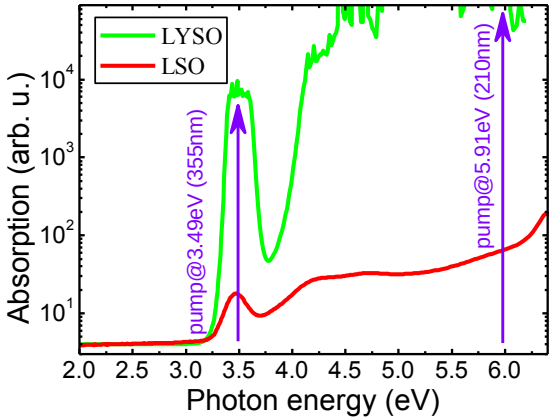


Are we able to identify
a clear and reliably measurable
figure of merit
for fast response?

Please attend the presentation
of Dr. Saulius Nargelas
October 10, 10 PM

Nonequilibrium carrier dynamics
LYSO:Ce versus LSO:Ce

Comparison of DA spectra & kinetics

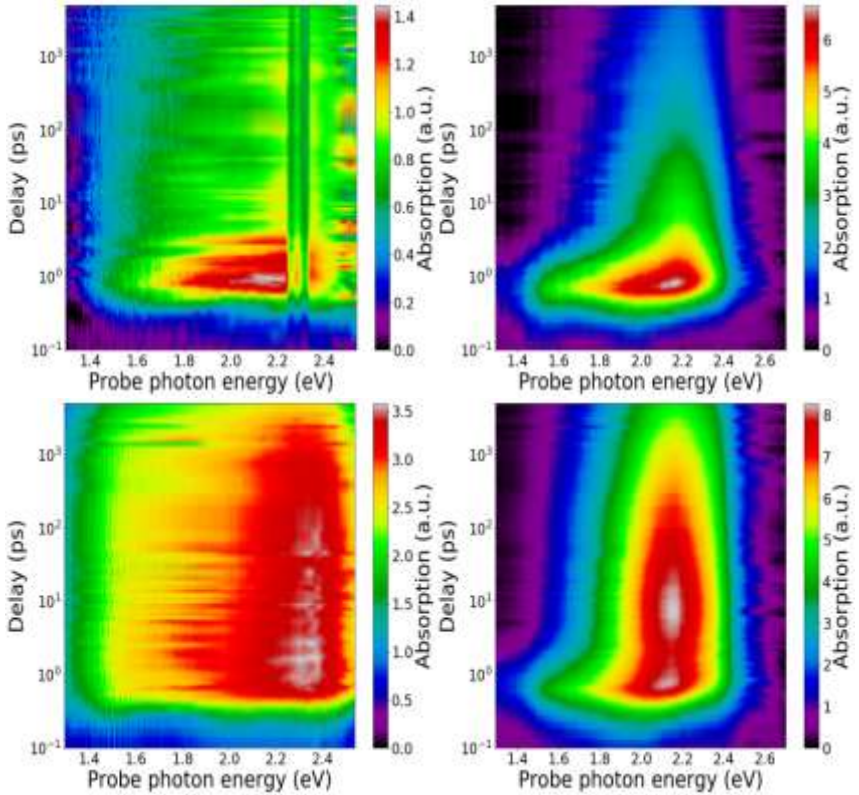


Pump @ 3.34 eV

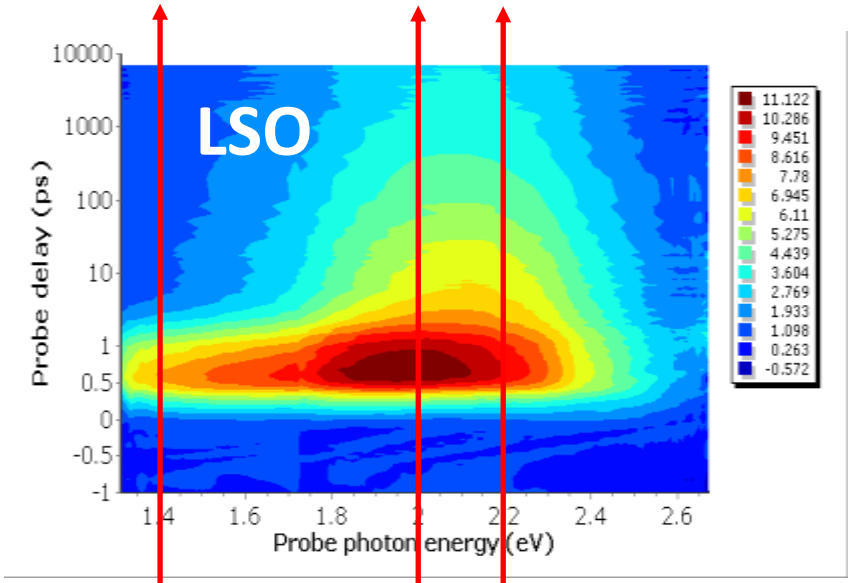
Pump @ 5.91 eV

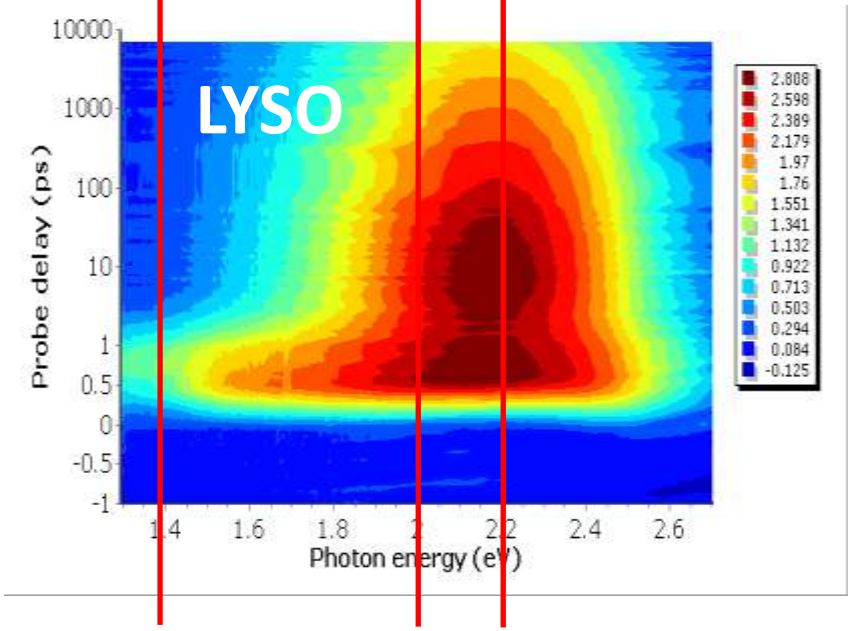
LSO

LYSO

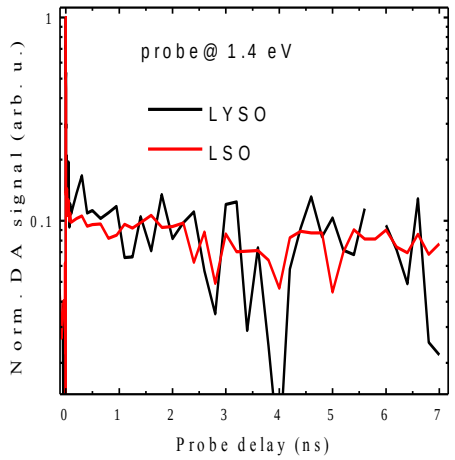


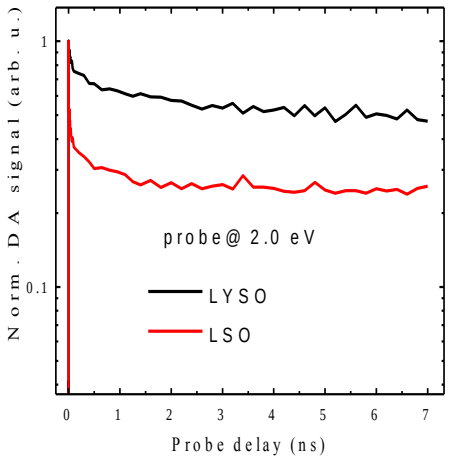
Comparison of DA spectra & kinetics

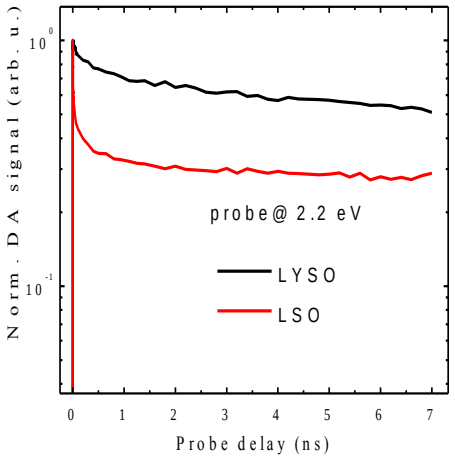




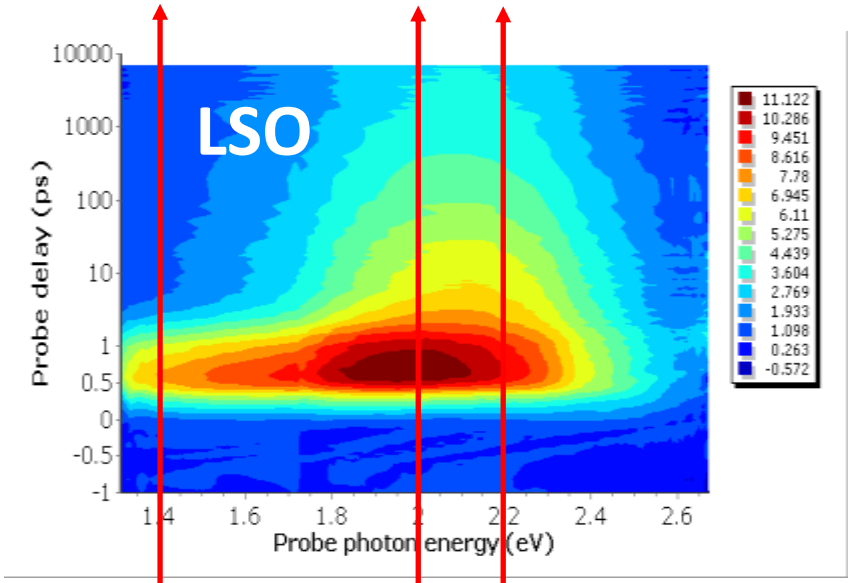
Nanosecond scale

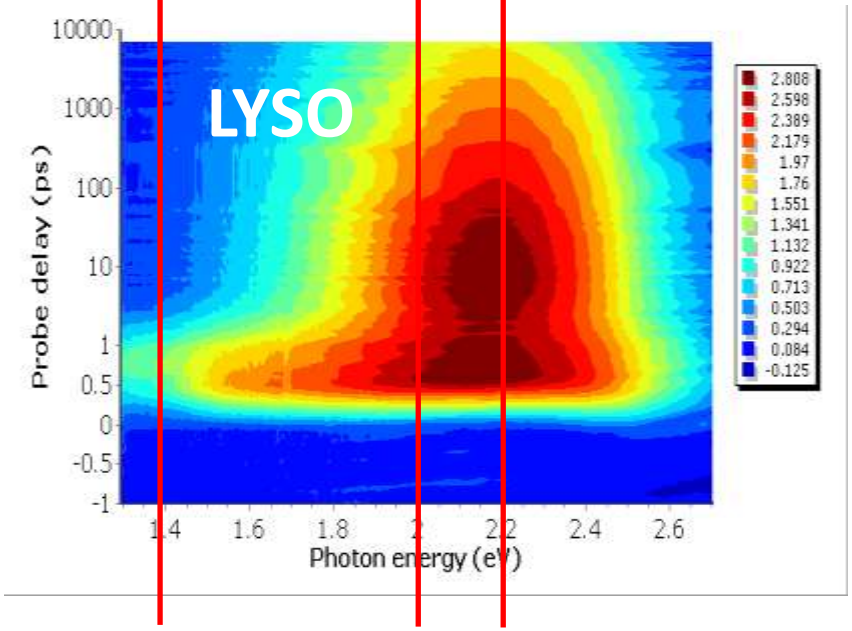




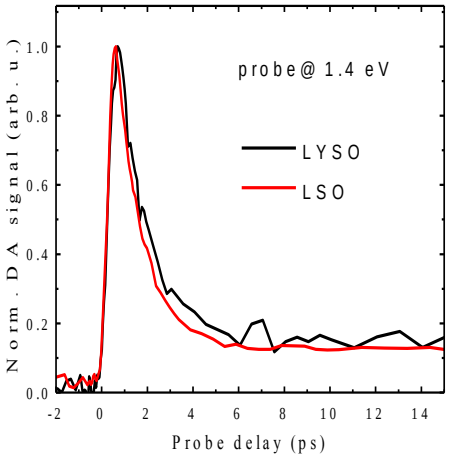


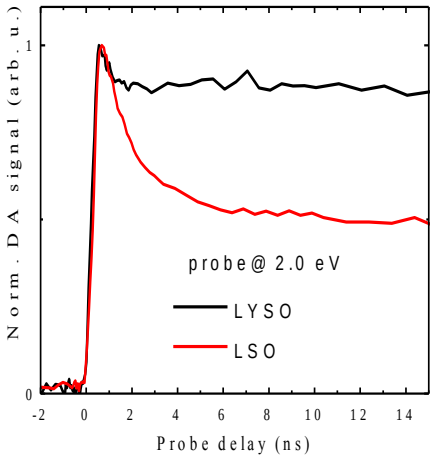
Comparison of DA spectra & kinetics

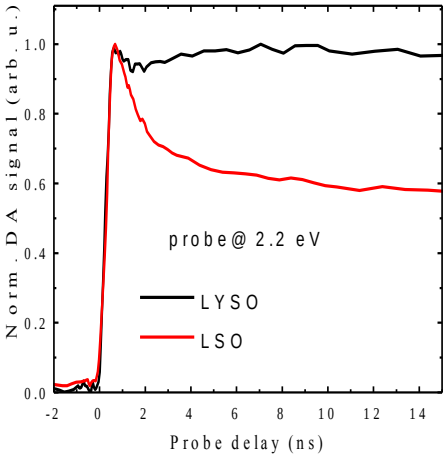




Picosecond scale

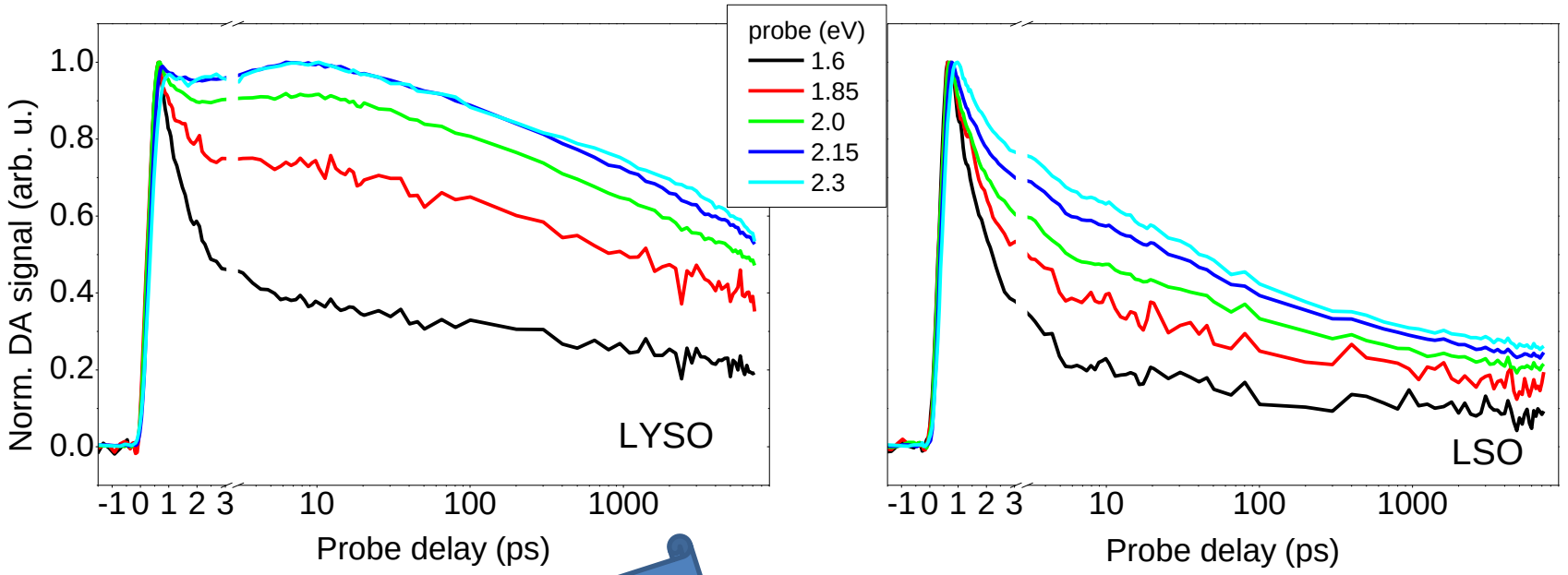






Comparison of DA spectra & kinetics

Absorption decay after 5.91 eV pump



MULTICOMPONENT

Conclusions on GAGG:Ce

- The rise of the differential absorption at resonant intracenter excitation of Ce^{3+} ions from the ground state into the first excited level $5d^1$ is instantaneous within the time resolution of 300 fs.
- The slow rise time component of differential absorption, observed when Ce^{3+} ions were excited into the second excited level $5d_2$, is due to **trapping** of the nonequilibrium electrons moving through the crystal matrix.
- **Intracenter $5d_2 - 5d_1$ relaxation time in Ce^{3+} equals 500 fs**
- Suppression of the slow component in the front of the differential absorption response is achieved by **codoping with Mg even at the codoping levels as small as 10 ppm**, which are insufficient to significantly change the valence state of cerium ions from Ce^{3+} to Ce^{4+}.

Conclusions on LSO:Ce and LYSO:Ce

- The initial rise time of differential absorption due to free electrons is in subpicosecond domain for both LSO:Ce and LYSO:Ce.
- Population of the emitting level of Ce^{3+} in LYSO:Ce **is delayed by several picoseconds due to migration of nonequilibrium electrons** through the matrix.
- The decay in population of the lowest excited Ce level proceeds at the same rate both in LYSO:Ce and LSO:Ce.

Thank you for your attention

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