L.1.1

Rare-earth doped nano-cluster silicon for silicon based photonic applications

Se-Young Seo and Jung H. Shin

(Silicon photonics laboratory, KAIST)

Outline

- Si based photonics: Needs? Breakthrough?
- Silicon nanostructures and limitations
- Rare earth ions for optical dopants in Si based photonics
- The advantages of nc-Si over bulk semiconductor for RE-doping
- The effect of exciton-RE coupling on backtransfer
- Efficient luminescence: optical amplification on visible non-resonant source
- Broad luminescence from co-doped RE system: RE-RE interaction and its control
- The expansion of luminescence to ‘real’ visible from RE-doped nc-Si

Fast but expensive

Broadband network: 'Backbone'

- Every home user want broadband
- Optical backbone level: fast but too bulky and expensive for home network
- The need development of low-cost, high-performance integrated optoelectronic devices

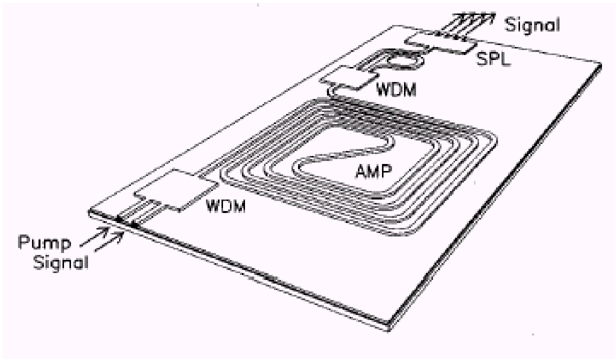
High speed home-network?

We need

- To miniaturize the equipment & To reduce the costs of the optical hardware

→ **Integrated Optical Circuit (IOC)**

:Chip integrated with several photonic capabilities such as light sources, photo-detectors, optical filter & waveguides



Example of IOC (Polman, Nature Mat.)

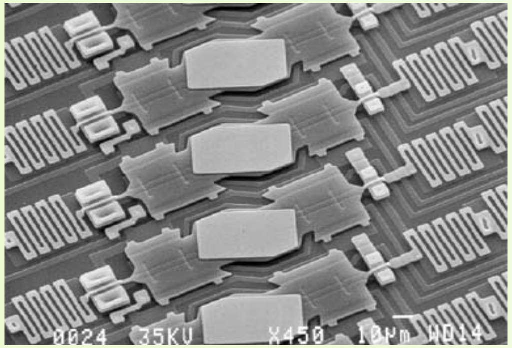
Si micro-photonics

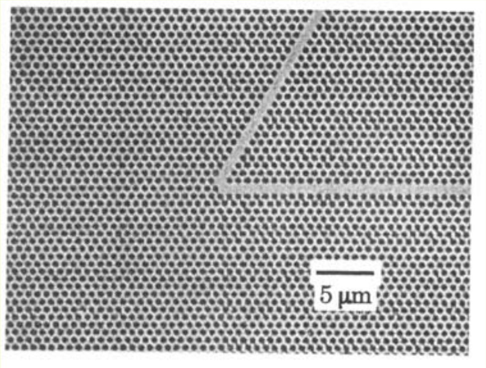
Si: promising candidate for micro-photonics

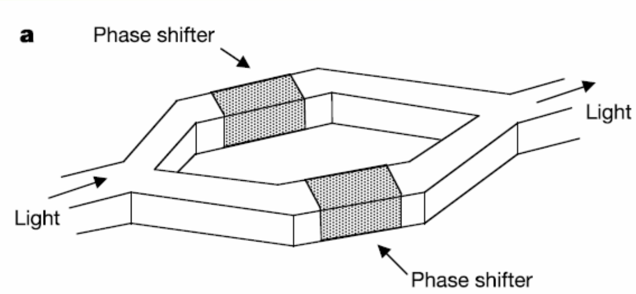
- Predominant material for electronic circuit and devices
- In addition, optically and electrically important related compound
  → Si, SiO_2, SiN_X and SiC
- The combination of well-developed infrastructure for Si and photonics

Key to success: ‘to siliconize’

- Various examples







1GHz modulator by Intel

Key breakthrough for Si photonics

All ready to open Si photonic Era?

→ Still need a few key breakthrough: **LIGHT EMISSON!!!**

- Si: is not efficient light emitter

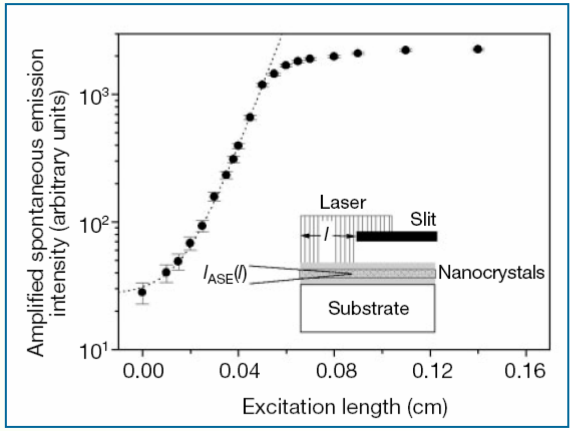
Teach silicon New tricks!!!

Silicon nano-structure

Training strategy: Si nanostructure

- Quantum confinement:
  → Increases the bandgap from 1.14 to 2-3 eV
- Physical confinement of electron-hole pairs:
  → Radiative transition rates are improved
- Physical isolation from quenching centers:
  → increased luminescence efficiency

➔Some fruitful results
E.g., Optical amplification from nc-Si (Pavesi Nature)

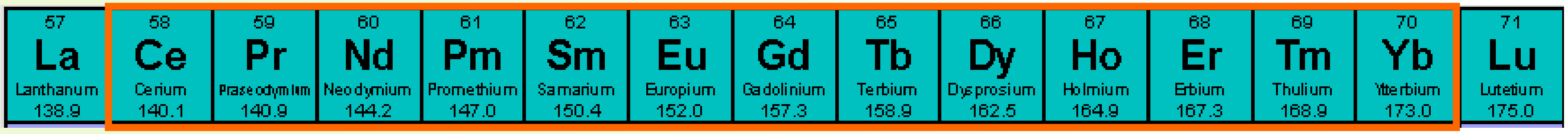


Limitations of nc-Si

- Broad luminescence from even a single nc-Si (Valenta APL 2002)
- Awkward luminescence position (generally near infrared)

Very strong Stokes shift –hard to exceed 2.0 eV) (Wolkin PRL 1999, Puzder JCP 2002)
Luminescence cannot be tuned down to IR range
→ Difficult to use telecommunication window of silica fiber

Rare earth ions



57	58	59	60	61	62	63	64	65	66	67	68	69	70	71
La	**Ce**	**Pr**	**Nd**	**Pm**	**Sm**	**Eu**	**Gd**	**Tb**	**Dy**	**Ho**	**Er**	**Tm**	**Yb**	**Lu**
Lanthanum	Cerium	Praseodymium	Neodymium	Promethium	Samarium	Europium	Gadolinium	Terbium	Dysprosium	Holmium	Erbium	Thulium	Ytterbium	Lutetium
138.9	140.1	140.9	144.2	147.0	150.4	152.0	157.3	158.9	162.5	164.9	167.3	168.9	173.0	175.0

Intra-4f transition from lanthanide series rare earth metals

- Incomplete 4f shells: Ce: [Xe].$4f^1$.$5d^1$.$6s^2$ ~ Yb: [Xe].$4f^{14}$.$6s^2$

How to overcome

Limitations of nc-Si

- Broad luminescence
- Awkward luminescence position
- High pump for lasing

Using RE ions

➔ Stable, atomic transition with environment insensitive luminescence positions

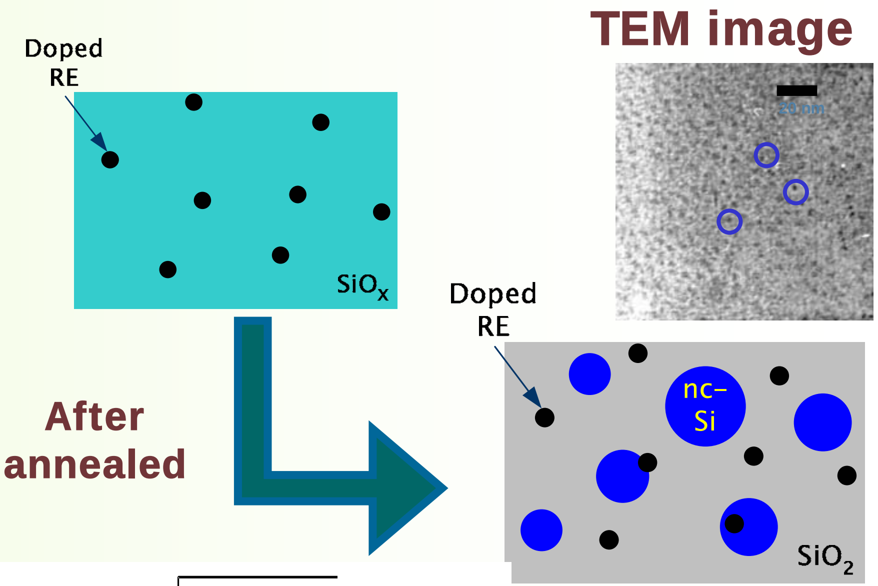
➔ Various visible to infrared luminescence

➔ Originally parity forbidden – allowed due to crystal field: long luminescence lifetimes and ease of population inversion in addition to multi level system

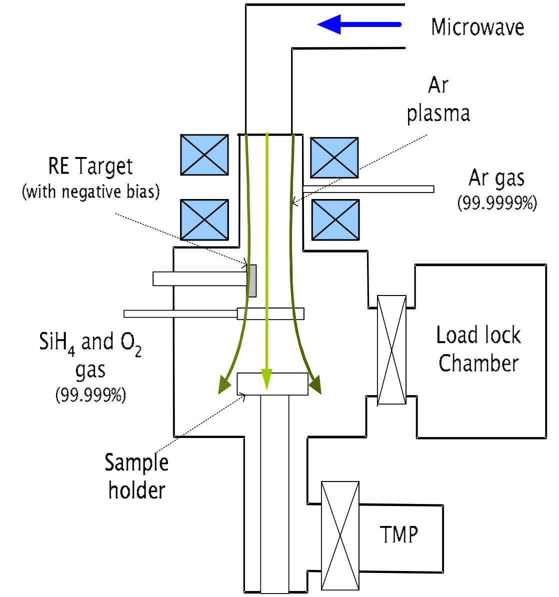
RE-doped silicon-rich silicon oxide

Silicon-rich silicon oxide (SRSO)

- 2-4 nm nc-Si embedded inside RE-doped SiO_2.
- Fabricated by depositing a-SiO_x ($x<2$) and annealing to precipitate nc-Si
- nc-Si size controlled by controlling the excess Si content
- Stochastic control over ‘nc-Si’ – RE coupling
- Rapid & dense formation of nc-Si



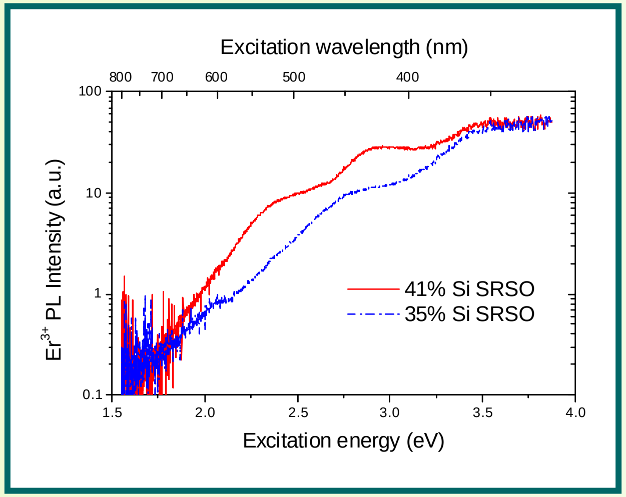
- **Sample deposition**
  - By ECR-PECVD
  - Flowing O_2, SiH_4 and Ar gases
  - Concurrent sputtering of RE targets

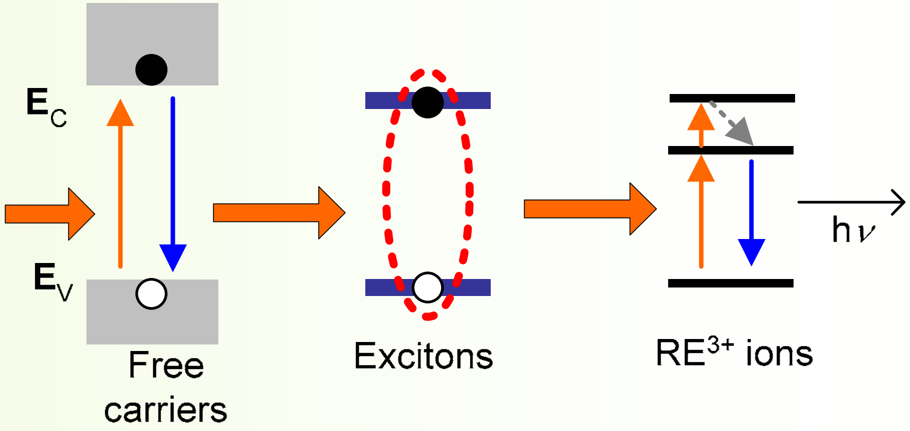


This presentation

- **The advantages of SRSO films for RE-doping**
- **Photonic Implications of RE-doped SRSO**
- **The extension of luminescence window of RE-doped Si nano-cluster by co-doping of impurity**

RE excitation via excitons in Si nanoclusters





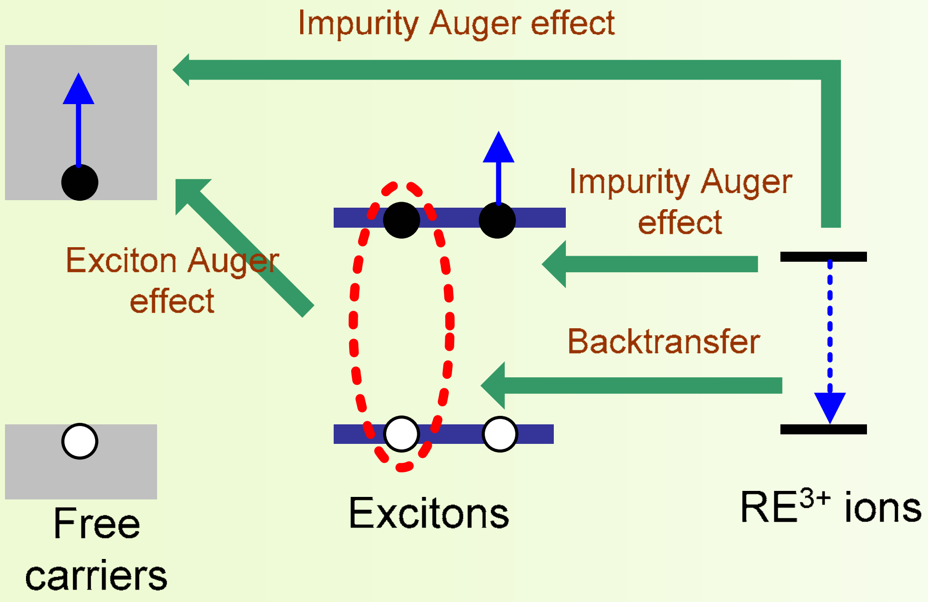
1.54 μm Er PLE spectra→ similar with absorption spectra of NanoSi

- Very efficient: effective excitation cross-section ~10^{-18} cm^2 for Er for nc-Si
  (c.f, 2 X 10-21 cm2 for silica)
- Continuous excitation cross section: broad band excitation.
  (c.f. direct optical absorption)

General for RE-doped semiconductor host

Problems of bulk semiconductor for RE doping

De-excitation processes



- Reversibility of physics :
Efficient excitation ↔ Efficient de-excitation

Problem of bulk semiconductor for RE doping

Thermal quenching

▪Energy backtransfer of excited RE ions

De-excitation of Auger effects

▪Always exist even at 0 K

▪Serious decrease in both excitation and quantum efficiency Population inversion is difficult

The advantages of nc-Si for RE-doping

The advantages of nc-Si for RE-doping: compared to bulk semiconductor,

- Efficient luminescence

  → Lack of all possible de-excitation path including backtransfer and Auger quenching (Seo et al. 1999 APL)

The lack of de-excitations of nc-Si: the origins

Backtransfer:

→ Bandgap: increases thermal barrier between RE and excitons and known to be predominant factor for ‘backtransfer;

→ Wide bandgap of nc-Si: the reason for the suppression of thermal quenching?

:Contrarily, strong thermal quenching of wide bandgap matrix (e.g, GaP:Nd)

Auger effect:

→ Auger effect has no relation with bandgap

→ The reason for the suppression of Auger effect?

The advantages of nc-Si for RE-doping

What is different?

- Nc-Si: inhomogeneous structure

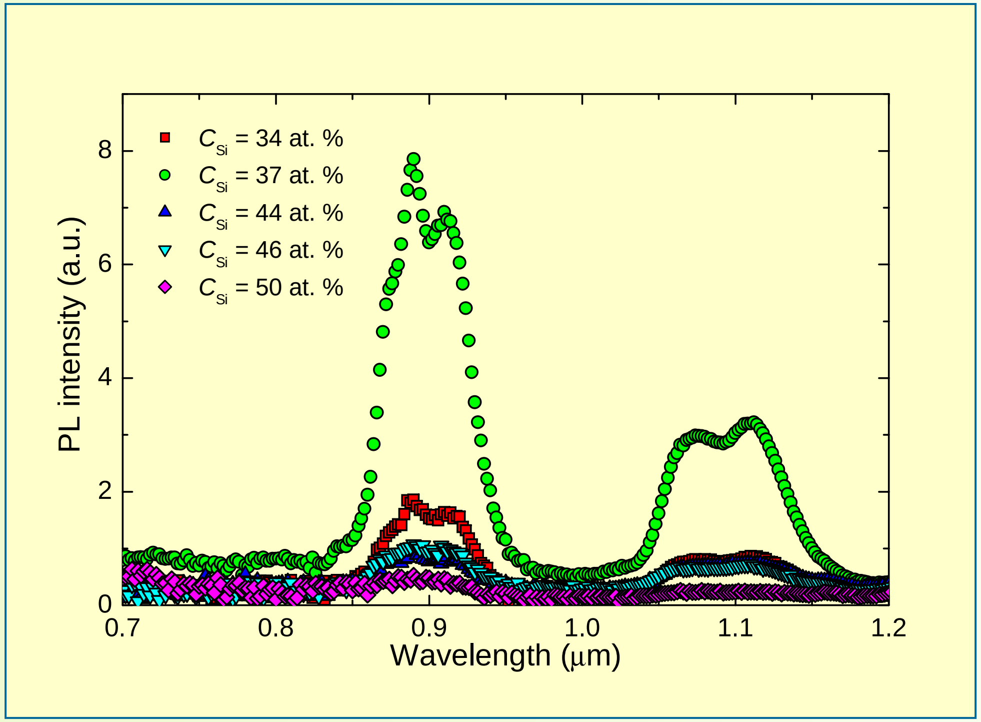
→ RE ions can be located in SiO_2 matrix, nc-Si, the interface between SiO_2 and nc-Si

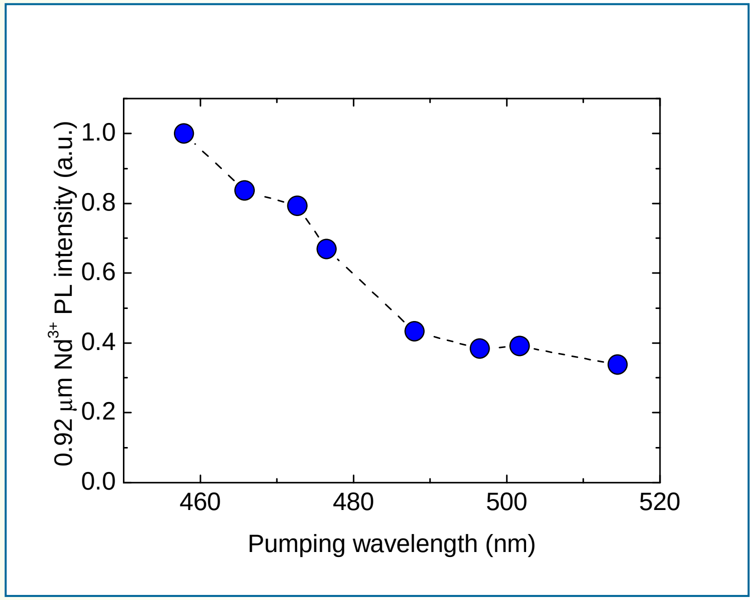
→ Various couplings may exist between excitons and RE ions

Thus exciton-RE coupling strength

- Determines Not only excitation of RE ions but also RE de-excitations such as Auger effect and backtransfer

Exciton-RE coupling strength:
Probing by Nd luminescence of Nd-doped SRSO films





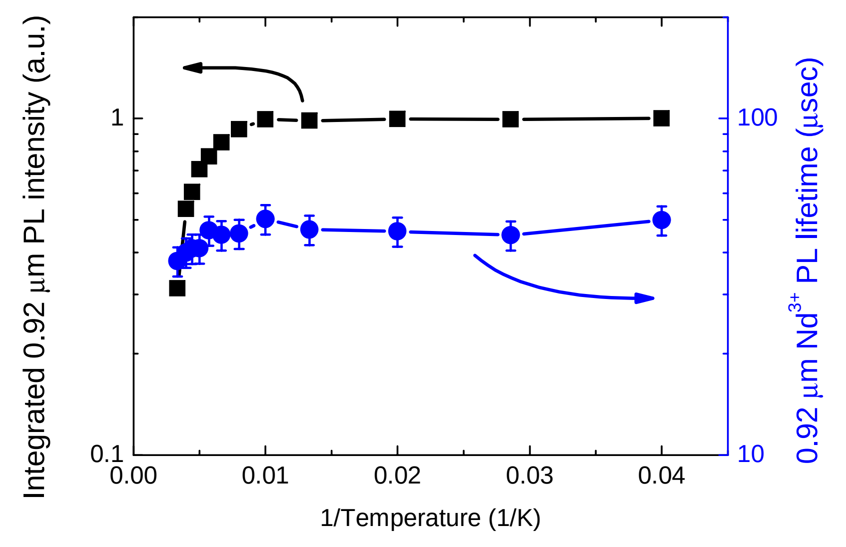
Nd^{3+} luminescence

- 0.9 um ($^4F_{3/2} \rightarrow {}^4I_{9/2}$) and 1.1 um ($\rightarrow {}^4I_{11/2}$)

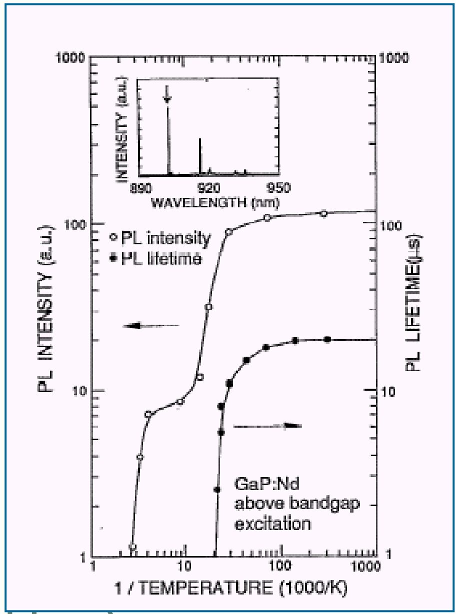
→ within c-Si and nc-Si luminescence range

→ Sensitive probe of the interaction between nc-Si and RE ions

Wide bandgap effect on suppression still valid?



GaP:Nd (Taniguch et al. APL, 1991)



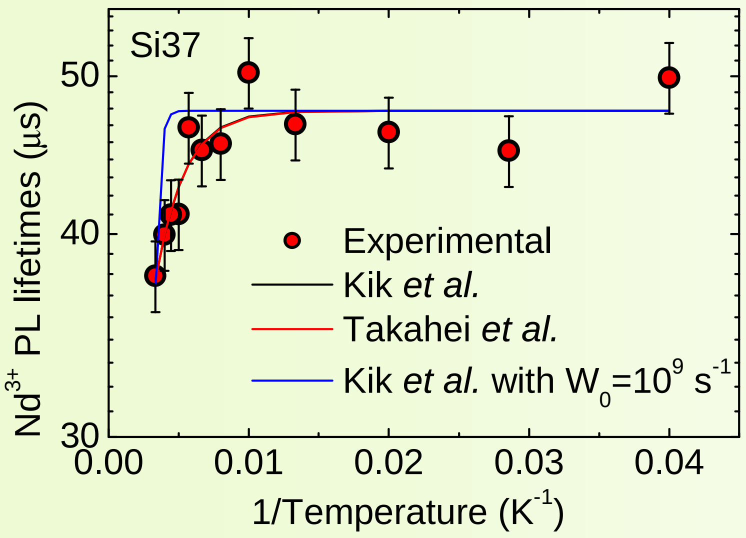
The lack of backtransfer

- t_{Nd}(T) 50 → 40 us with T (cf. Nd-doped silica-based thin films)
- The reason of 3 folds decrease of Nd PL intensity
  → not backtransfer but exciton dissociation

Only ~ 200 meV difference between nc-Si luminescence and Nd^{3+} excitation energy

- Not sufficient to suppress backtransfer (cf. bulk GaAs:Nd)
- Even GaP (E_g=2.26 eV) shows > 10 folds thermal-quenching of Nd^{3+} luminescence

Weak couplings induce weak backtransfer



- Kik’s
  - $W_{BT}(T) = W_0 \exp(-E_0/kT)$
- Takahei’s
  - $W_{BT}(T) = W_0 (n_q)^p \exp(-2n_q s)$

    $n_q=[\exp(\hbar\omega/kT)-1]^{-1}$

Analysis of temperature quenching of with

- Kik’s (Thermal dissociated) and Takahei’s (Multi-phonon relaxed) model
- Different physical basis but similar dependence on E0 and W0

    ➔ $W_{BT}(T) = W_0 \times f(E_0, T)$ (W_0: coupling prefactor)

Identical simulated results fit by both models with similar fitting value

- E_0 ~ 100 meV and W_0 ~ 1-10X10^5 sec^{-1}
- Much smaller than W_0 ~ 10^9 sec^{-1} for Er-doped bulk Si

The separation of RE from nc-Si

Different from bulk semiconductor

- Weak coupling between exciton and RE: responsible for suppression of backtransfer
- Slight separation of RE from nc-Si

→ Weakens the interaction between nc-Si and RE (Kimura et al.)

→ Uniqueness of nanocluster

This exciton-RE couplings are

Still strong enough for efficient excitation of RE

Weak enough for suppression of de-excitation

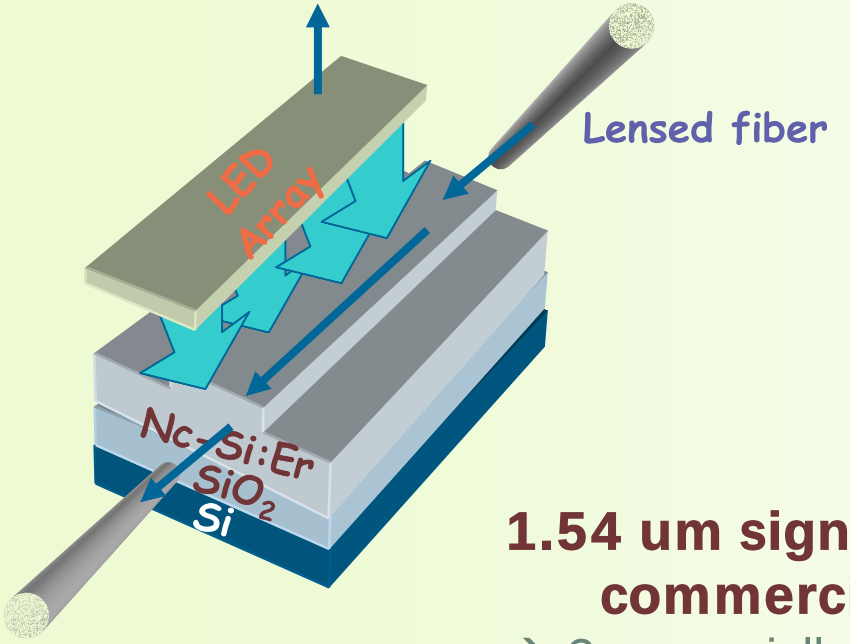
The suppression of de-excitation by Auger process and backtransfer

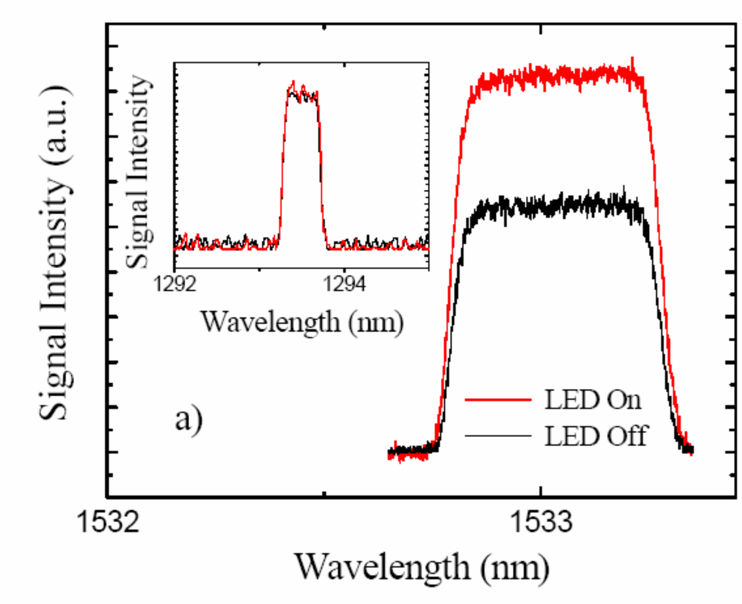
→ High concentration of excited RE ions at given pump power

→ Imply optical amplification!!!

Optical amplification pumping with LED Array

Turn-on state of LED





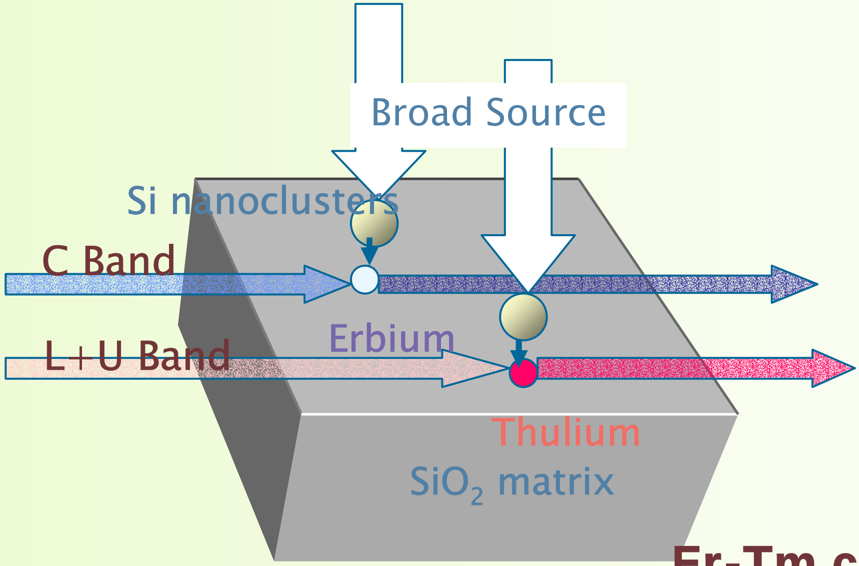
1.54 um signal enhancement even pumped by commercial blue LED array

→ Commercially viable

Si photonics lab. (KAIST)

Further extension bandwidth using RE-doped SRSO

Utilizing advantages of nc-Si sensitizing



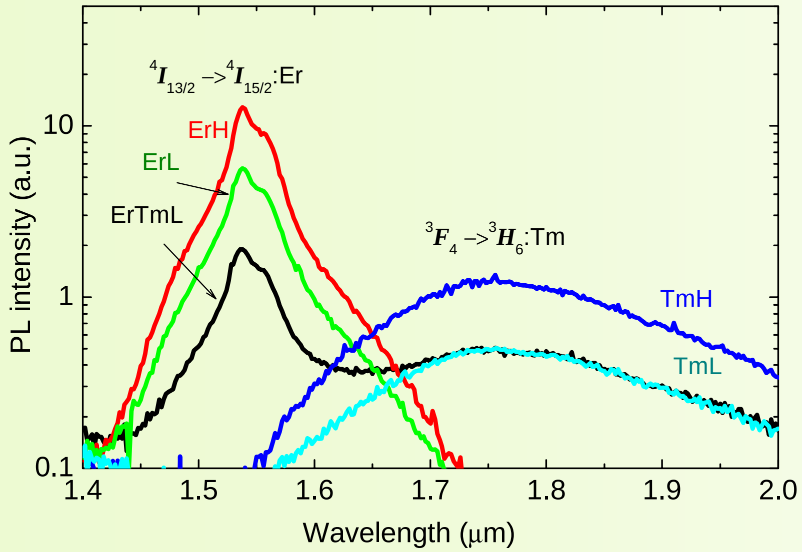
Er-Tm co-doped waveguide amplifier

Advantages:

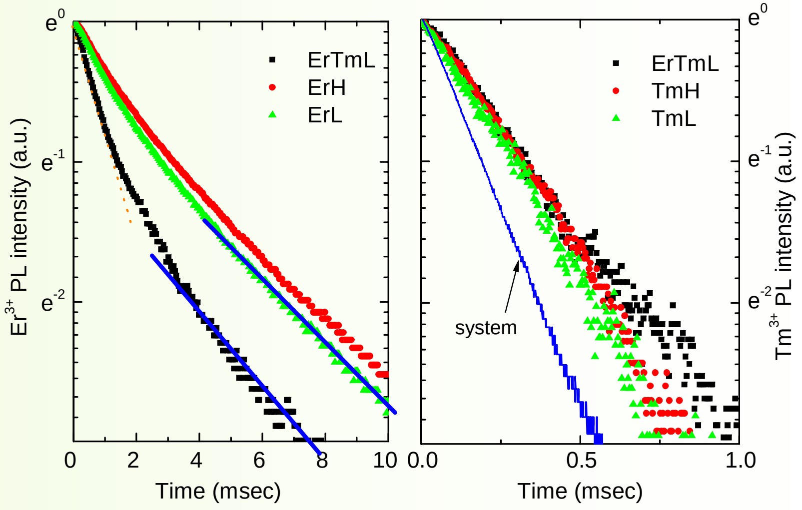
- Sensitized by nc-Si
- Dual pumping (Er and Tm) with single- broad source
- Simple scheme : Reduction in coupling loss

Broad and flat 1.4–1.9 μm luminescence from Er and Tm co-implanted SRSO

PL Spectra

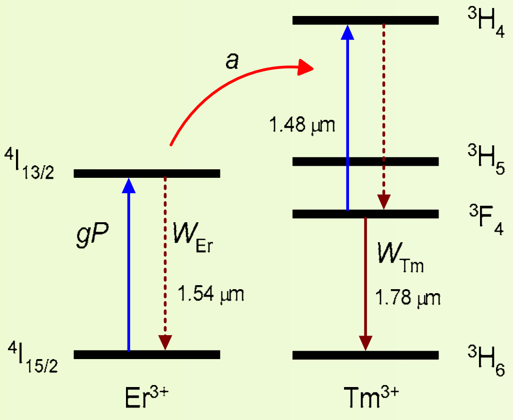


PL Decay traces

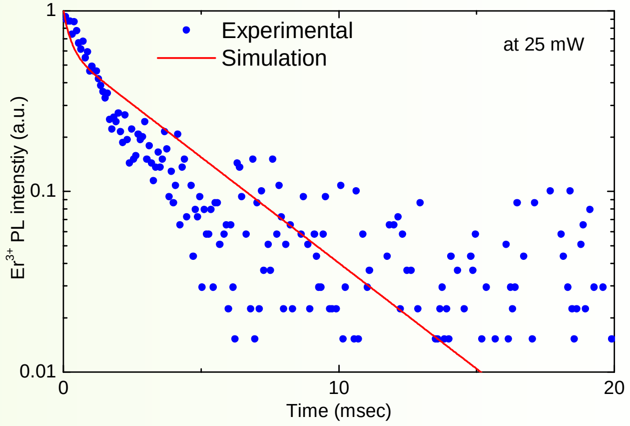


- **Er and Tm PL intensities increase with increasing RE content:**
  → PL intensity was limited by available RE
- **Tm co-doping: quenches Er PL intensity and PL lifetimes**
- **Er co-doping: has no effect on Tm luminescence**
  → Er and Tm interact – but how?

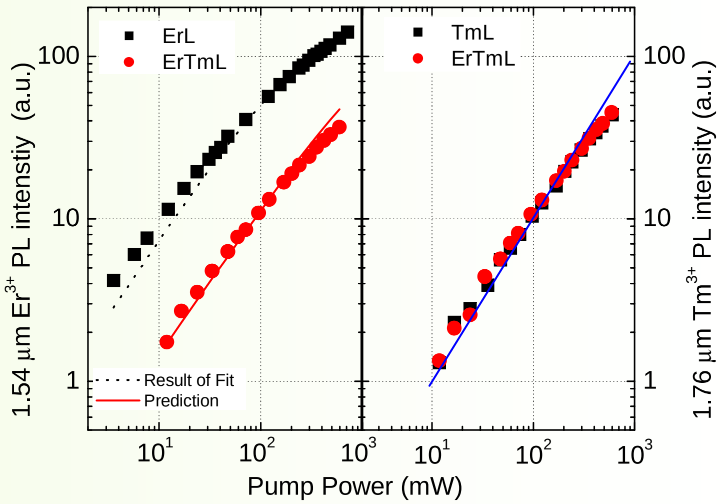
Interaction between Er and Tm: Co-operative up-conversion of Tm^{3+}



PL Decay traces

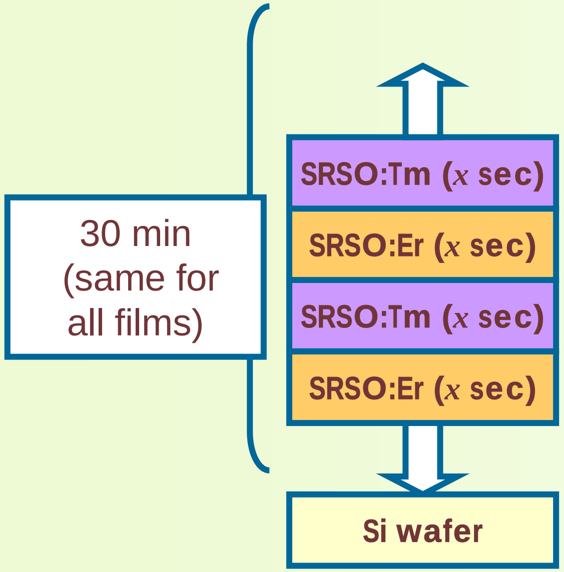


RE PL intensity as function of pump power

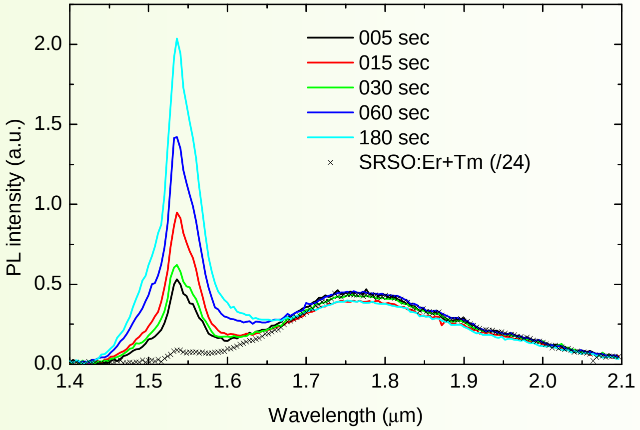


- **Almost exact agreement with experimental result → verifying interaction model**
- **Er-Tm interaction: Need to be minimized**

Control of Er-Tm interaction by spatial separation



Alternating SRSO:Er / SRSO:Tm layers



- **Er luminescence increases with increase in layer thickness**
- **The interaction between Er and Tm**

→ Can be successfully minimized by spatial separation

- **Judicious control and design of nc-Si size and RE distribution**

→ Enabling efficient luminescence minimizing undesired degradation

The realization of visible RE luminescence

Up to date, most RE-doped SRSO:

→ Investigated and demonstrated for photonics application in infrared range

And, need of the expansion to other wavelength:
RE ions for visible luminescence: implication for Si photonics

- Visible luminescence:

→ Open the feasibility of Si based full color display unit

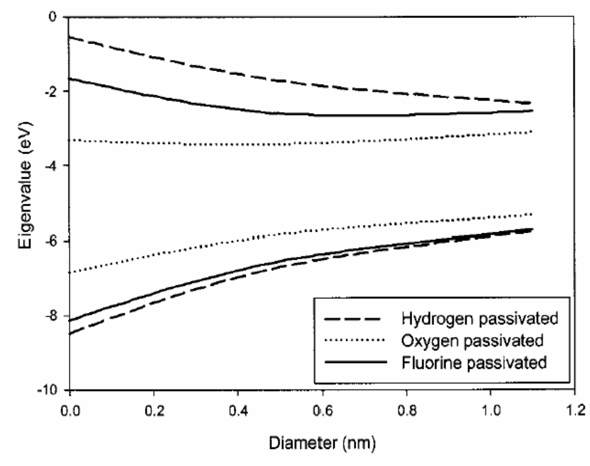
- Can be detected with Si photo-diodes

→ Applicable inter-chip communication using polymer fiber

HOWEVER,
The optical transition of oxide capped nc-Si

→ Strong Stokes shift limits below 2 eV

→ Difficult to excite RE onto visible 4f transition



(Puzder, JCP)

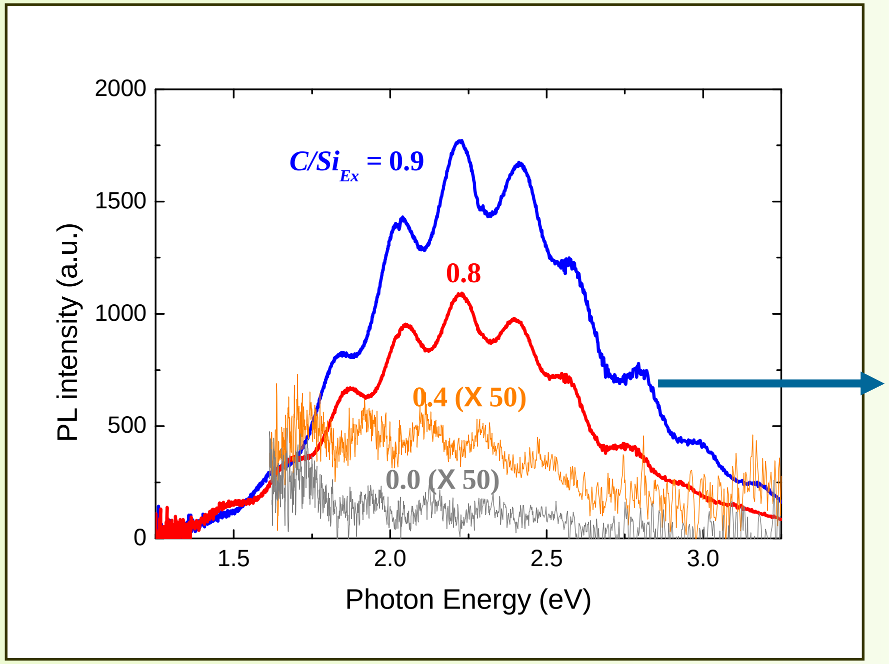
In order to realize visible RE luminescence from SRSO

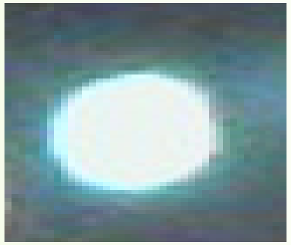
- The need of wide bandgap (blue-white)

→ by suitable impurity incorporation other than oxygen

Candidate of wide bandgap: 1. C-doped SRSO

(Seo et al. APL 2004)





C-doping into SRSO

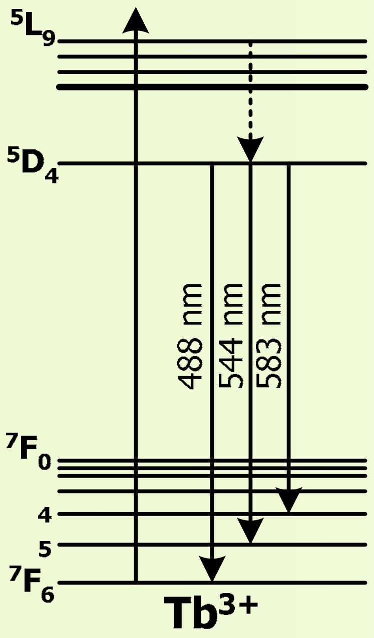
- Significantly increases blue-white luminescence
- Excitons in C-rich nc-Si is responsible strong visible luminescence

→ Only Si-C bonds exists among C related bonds

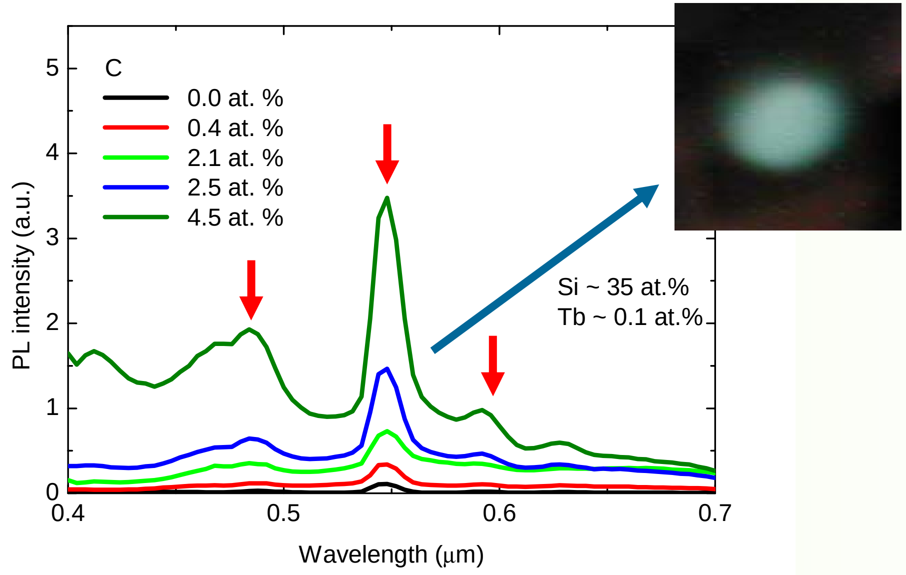
→ The dependence of Si content & anneal temp.: very similar to those of C free SRSO

- Can be utilized as wide bandgap material host for visible RE luminescence

Green luminescence from Tb-doped SRSO:C



(Seo et al. APL 2004, in press)



The enhancement of Tb^{3+} luminescence by co-doping of C

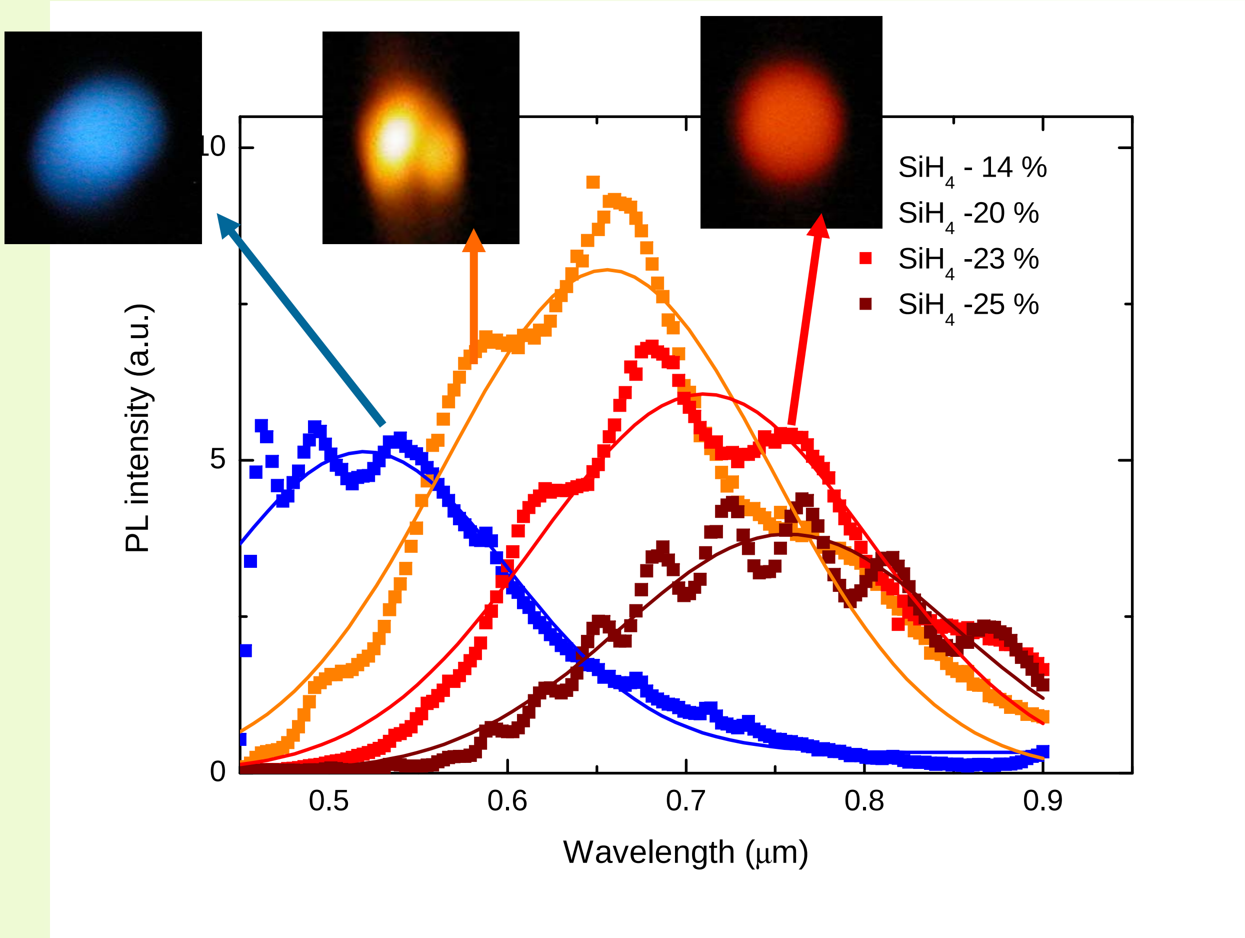
- C doping:

→ Increases the number of high energy excitons (which can efficiently excite Tb ions)

→ Strongly enhances Tb^{3+} luminescence

The details will be found in L.6.8

Candidate of wide bandgap: 2. Si rich Si nitride



Wavelength tuning by Si content

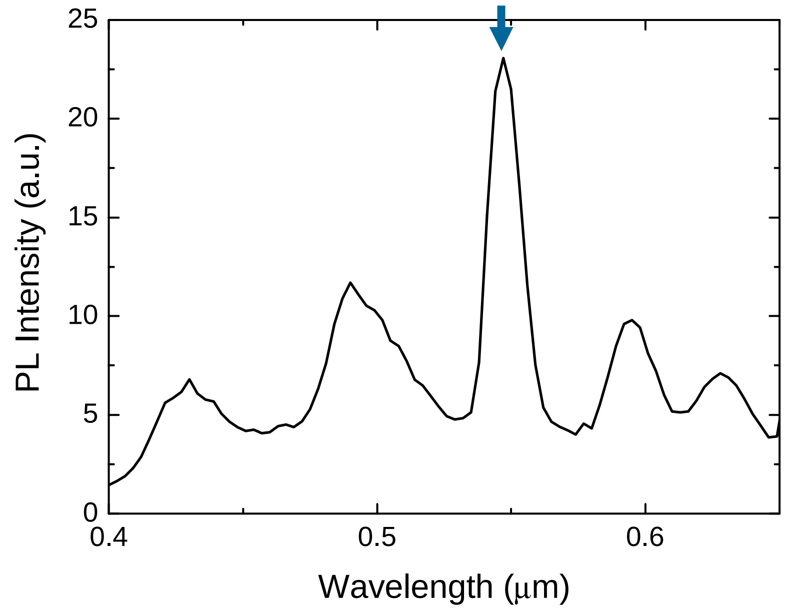
- Strong visible covering from RED to BLUE

→ Can be also utilized as host matrix of RE for visible RE luminescence? YES

MRS

Si photonics lab. (KAIST)

Tb luminescence from Tb-doped SRSN



Green Tb luminescence is also possible from Tb-doped SRSN
The sharp strong visible RE luminescence from nc-Si based wide bandgap such as SRSO:C and SRSN

→ Can be used for other RE ions (Eu, Er, Tm, Pr, and Sm)
→ Feasibility of RGB display pixel integrated on single Si chip

Summary & Conclusion

- **Advantages of nc-Si over bulk semiconductor for RE doping**

  →Separation of RE from carriers: suppress the de-excitation

  →But still holding benefit of nc-Si: efficient excitation cross-section

- **Photonic implication of advantages of nc-Si**

  →Suppression of de-excitations and efficient luminescence: optical gain from nc-Si sensitized Er-doped waveguide amplifier

  →Exciton mediated excitation: simultaneously exciting two different RE ions & pumping with non-resonant visible source

- **The expansion of luminescence band to visible from nano-cluster Si**

  → Suitable impurities can increase optical transition

  → Strong green luminescence can be accomplished from Tb-doping

Acknowledgement

Dr. Hak-Seung Han (Samsung Elec.)

Prof. A. Polman (FOM)

Prof. P. G. Kik (Univ. of Cent. Florida)

J. J. Penninkhof (FOM)

Kwan-Sik Cho, Hoon Jeong, Dai Gil Cha, and Jin-Ku Lee (Silicon Phonics Lab. KAIST)