THE EFFECT OF ND-NANOCLUSTER INTERACTION ON DE-EXCITATION OF ND-DOPED SILICON-RICH SILICON OXIDE

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Outline

- **Rare-earth doped nc-Si**
- **Excitation/de-excitation of RE in nc-Si**
- **Factors suppress thermal quenching (energy mismatch and others?)**
- **Probing thermal quenching with Nd-doping**
- **Nd excitation via exciton recombination of nc-Si**
- **Weak thermal quenching process in SRSO:Nd**
- **The lack of backtransfer due to weak copulings**
- **Implications**

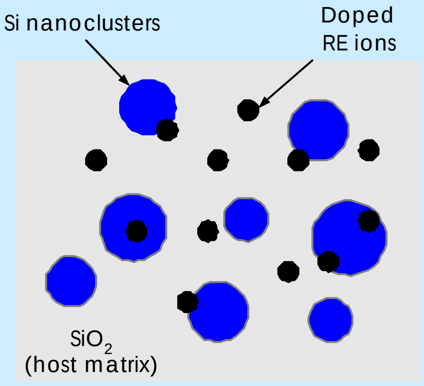
Rare-earth doped Si nanoclusters

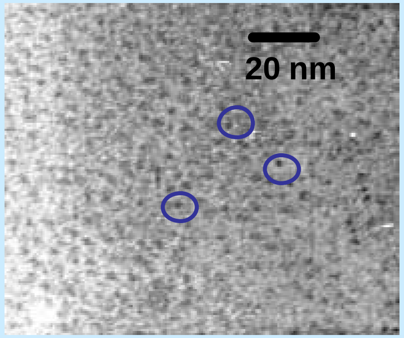
- **Rare-earth doped Si**
  - Si : well-developed infrastructure: from knowledge to technology
  - Rare-earth: stable and intrinsic luminescence by 4f transition independent of host and temperature

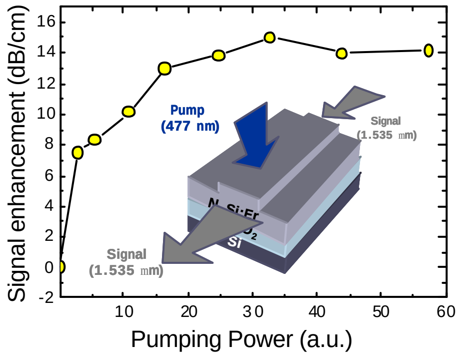
  ➔ Rare-earth doped Si: realization of silicon compatible micro-photonics

- **Silicon rich silicon oxide (SRSO):RE**
  - SRSO: Si nanoclusters embedded in SiO_2
  - Efficient RE luminescence environment
    (Wide bandgap, high excitation and luminescence efficiency)

  ➔ 1.54 μm optical gain from nc-Si:Er (Han *et al.* APL 2001)







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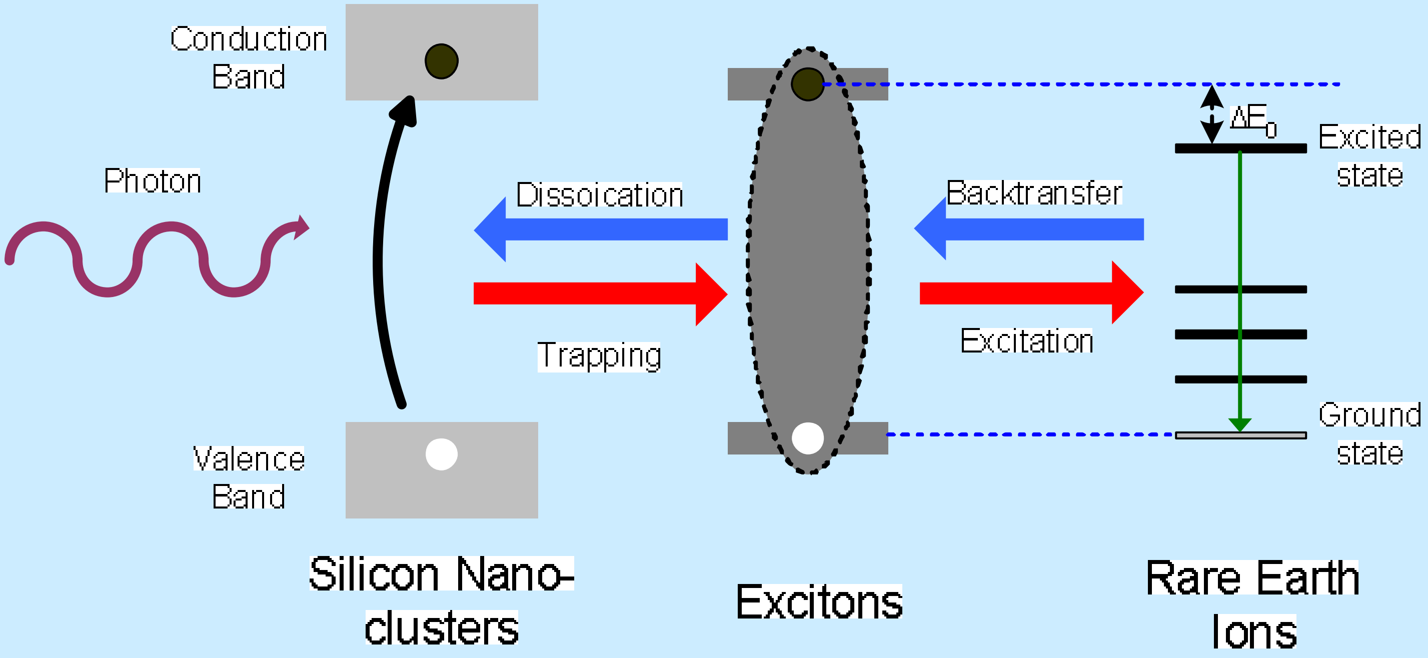
Excitation/De-excitation of RE in nc-Si



- **Exciton mediated excitation**
  - Similar to the RE excitation of bulk semiconductors (Schmitt-Rink PRL 1991)
  - Backtransfer: non-radiative decay of RE with help of phonons
    → Dominant thermal quenching mechanism
    → Plagues many RE-doped bulk semiconductors

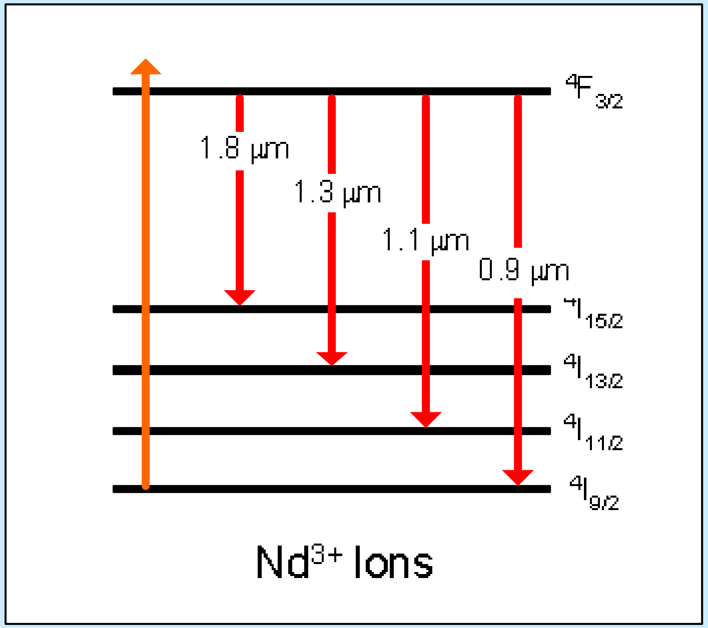
Suppression of thermal quenching

- **Wide bandgap:** increase of E_0 → increase of energy barriers (e.g. Favennec *et al.* MRS Proc. 1993)
  - Wide bandgap host: SiC, GaN
  - Quantum confinement effect of nano-clustering of narrow bandgap
    → Bandgap widening with decrease of nc size
    → The lack of thermal quenching for nc-Si (e.g. Seo *et al.* APL 1999)

- **Micro-structure: bandgap is not the whole picture**
  - Slight separation of RE away from nc-Si can also suppress thermal quenching
    → Different quenching behavior between Er-immersed and implanted porous Si (Wang *et al.* J. Luminescence)
    → Suppression of thermal quenching by buffer SiO_2 layer between nc-Si and SiO_2:Er (Shin *et al*. APL)

- **What factors other than bandgap contribute to suppression of backtransfer?**

Probing of backtransfer with Nd-doping



- **Nd^{3+} luminescence**
  - 0.9 mm ($^4F_{3/2} \rightarrow ^4I_{9/2}$) and 1.1 mm ($\rightarrow ^4I_{11/2}$)

    → within c-Si and nc-Si luminescence range

    → Sensitive probe of the interaction between nc-Si and RE ions
  - 0.9 mm luminescence can be detectable with Si photodiode

    → Integrating light emission and detection capabilities into single Si chip

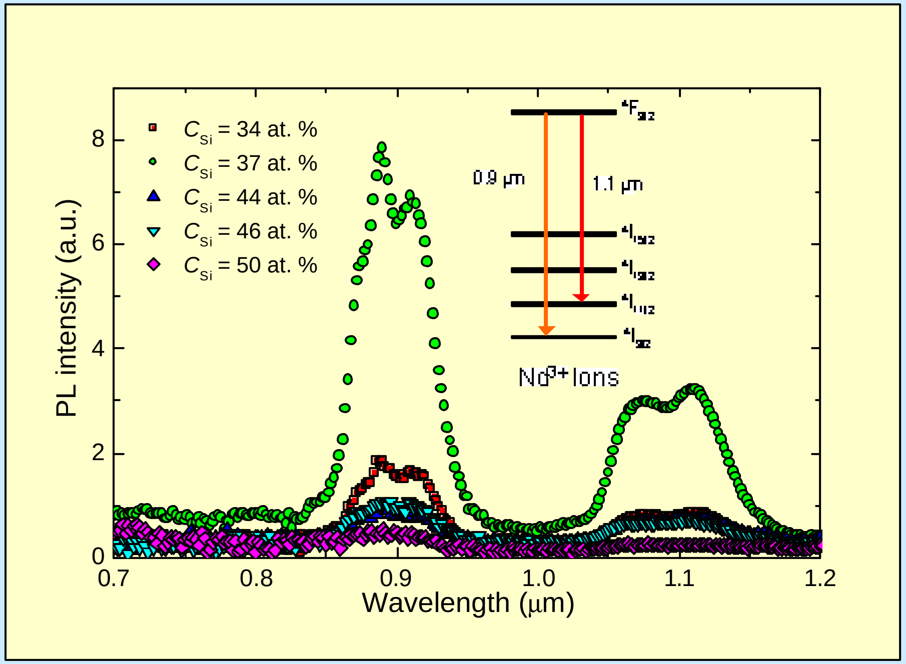
Experiment

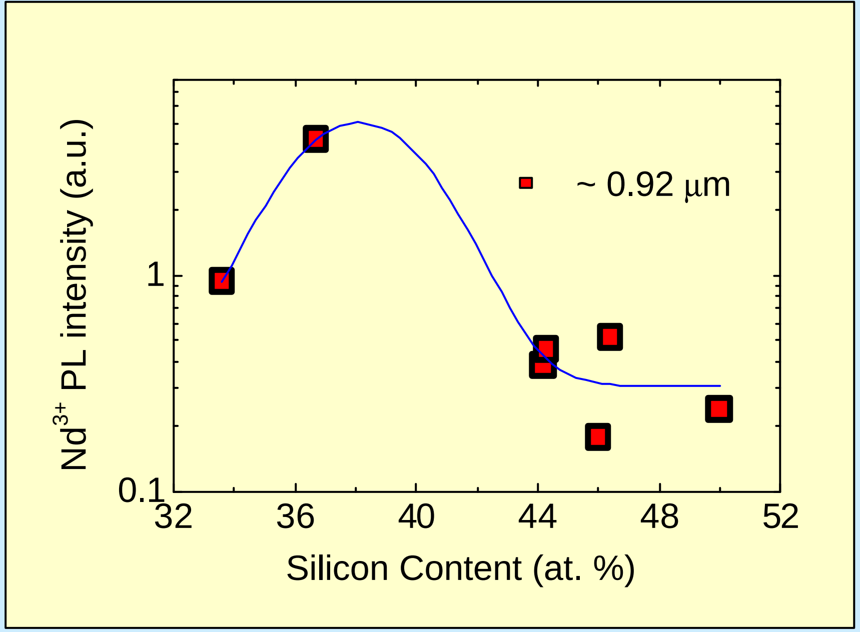
- **Film Fabrication**
  - ECR-PECVD with co-sputtering of Nd^{3+} target
  - Stoichiometric study : Si (34-50 at. %)
  - 1 mm thick with 0.14 at. % Nd

- **Post Treatment**
  - Rapid thermal anneal: 950 C for 5 min. for clustering of nc-Si

- **Luminescence measurement**
  - Detectors: Si photodiode + InGaAs photodiode
  - Pumped using **488 nm** line of Ar ion laser: not resonant with Nd^{3+} levels
  - Recording of luminescence decay traces with InGaAs(Cs) PMT and digitizing oscilloscope

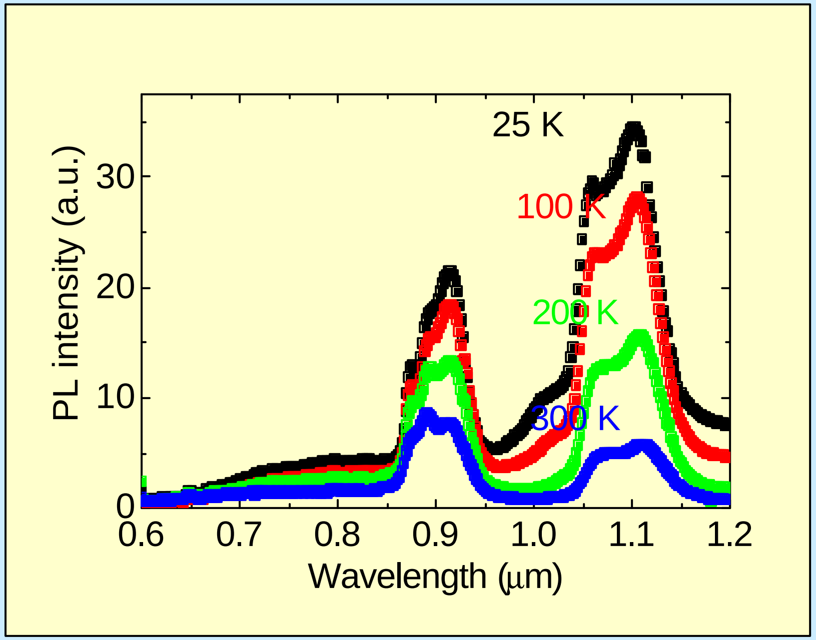
The size of nc-Si : crucial for Nd^{3+} excitation

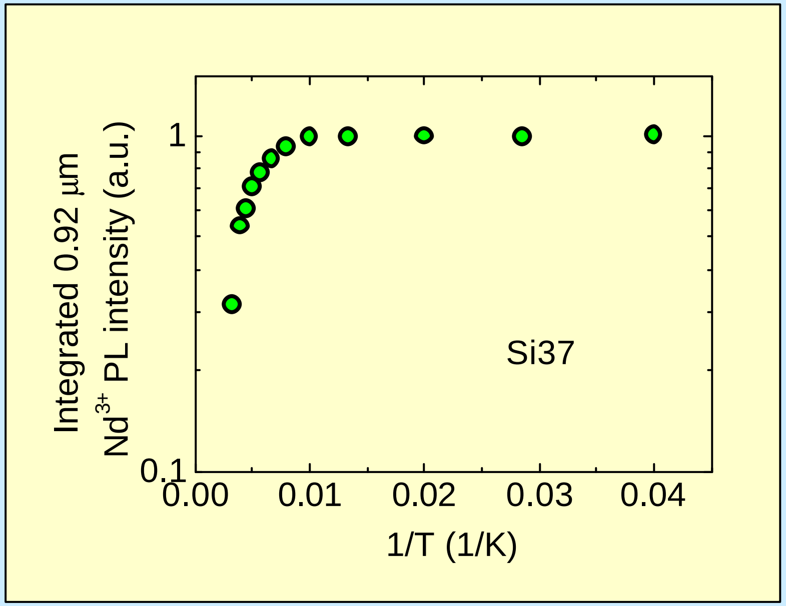




- **Clear Nd^{3+} luminescence at room-temperature**
  - Both 0.92 μm and 1.1 μm luminescence observed
- **Typical RE PL intensity dependence on Si content**
  - Nc-Si with > 44 at. % Si : too large not to have sufficient bandgap for Nd^{3+} excitation
  - The crucial role of quantum confinement effect of nc-Si for Nd-excitation
    → Nd^{3+} excitation via exciton recombination

Temperature dependence of Nd^{3+} PL





- **Intrinsic nc-Si luminescence at ~ 0.8 mm**
- **For 0.92 mm luminescence**
  - The superimposition of Nd^{3+} PL on the tail of intrinsic nc-Si luminescence
  - 3 folds decrease of Nd^{3+} PL intensity for Si37

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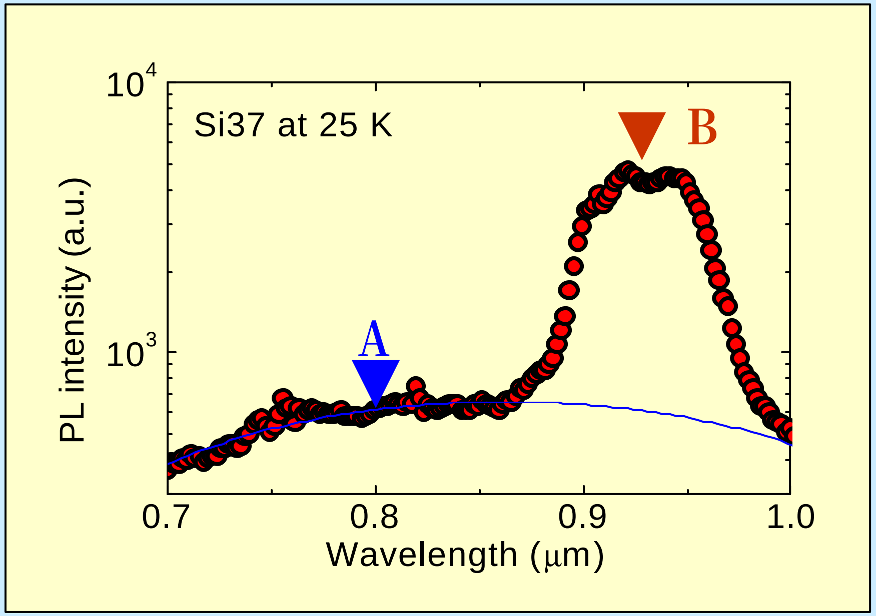
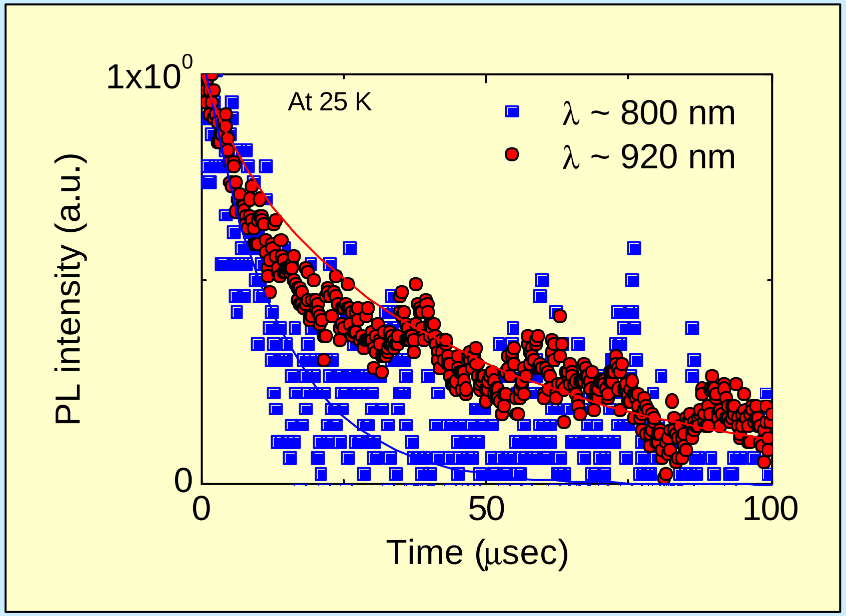
The determination of Nd^{3+} PL lifetimes

- PL decay traces

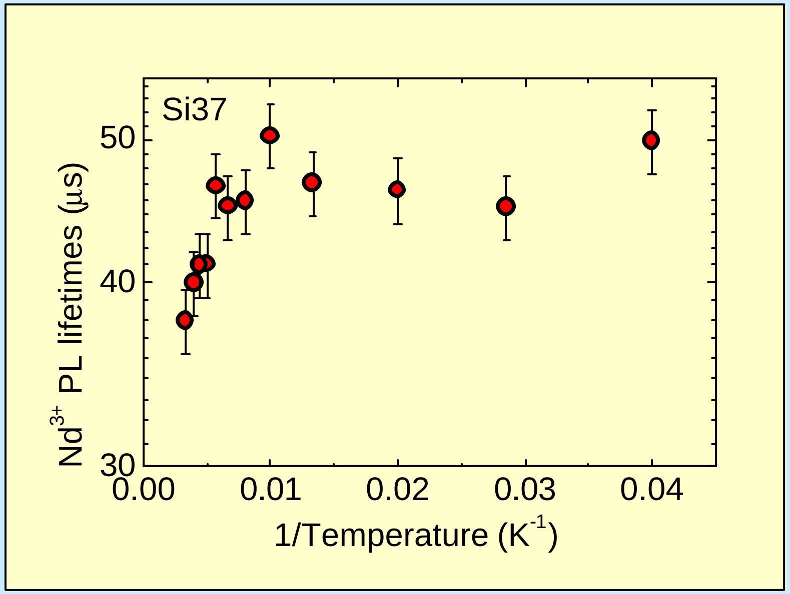




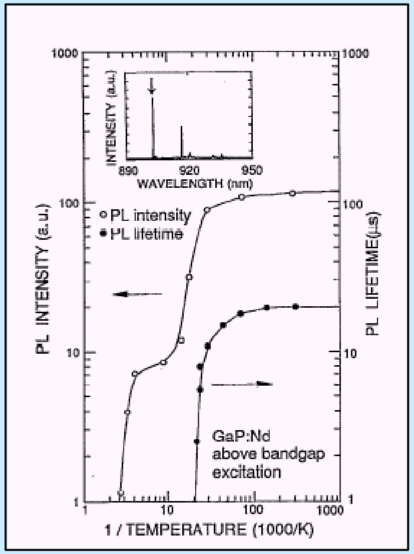
- **(A) Intrinsic nc-Si PL: exp($-W_{nc\text{-}Si}t$)**
  - $W_{nc\text{-}Si}$ < 10 μs : system response due to low temp anneal (950 ℃)
    (*c.f.* ~ 100 μs for high quality nc-Si after 1100 ℃ anneal)

- **(B) superimposition of Nd^{3+} on nc-Si PL**
  - $I\ (\lambda=0.92\ \mu m) \sim I_{nc\text{-}Si}\exp(-t/t_{nc\text{-}Si}) + I_{Nd}\exp(-t/t_{Nd})$

Wide bandgap effect on suppression still valid?

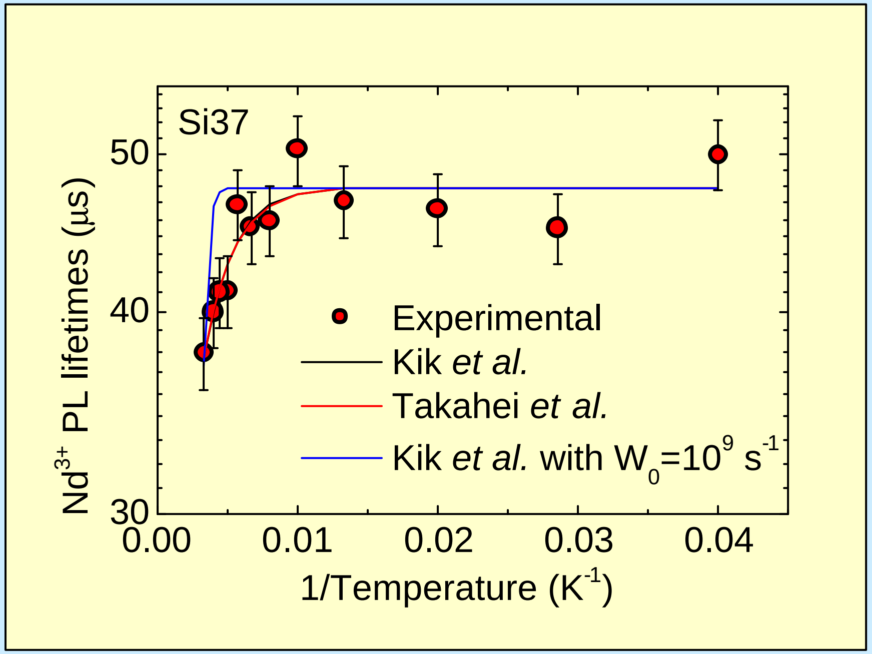


GaP:Nd (Taniguch et al. APL, 1991)



- The lack of backtransfer
  - t_{Nd}(T) 50 → 40 μs with T (cf. Nd-doped silica-based thin films)
  - The reason of 3 fold 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 thermal-quenching of Nd^{3+} luminescence

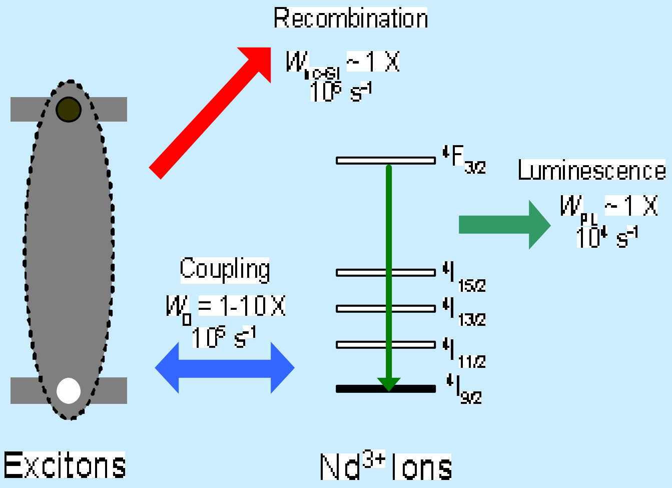
Weak couplings induce weak backtransfer



- 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}
  - Not explainable with W_0 ~ 10^9 sec^{-1} (for Er-doped bulk Si)

Efficient excitation with suppression of backtransfer



- Small energy mismatch but weak coupling
  - E_0 ~ 100 meV : agreement with previous results (Taguchi *et al.* JAP, 1996)
  - W_0 ~ 1-10 X 10^5 sec^{-1} : much smaller than 10^8-10^{10} sec^{-1} (for GaAs or GaP)
    → responsible for the lack of backtransfer
    → consistent with the excitation rate of Er^{3+} $\lesssim 10^6$ sec-1 (Watanabe *et al.* 2001)
  - Slight separation of RE from nc-Si weakens couplings suppressing backtransfer

Implications

- Nc-Si : efficient material for RE luminescence
  - High quality nc-Si by high temperature anneal : $W_{nc\text{-}Si} < W_{Nd} < W_0$

    → The excitons created via backtransfer can re-excite RE before dissociation or radiative recombination

    → Window for the efficient excitation ($W_{nc\text{-}Si} > W_0$) and the suppression of backtransfer : unique for nc-Si
  - Moreover, suppression of de-excitation of RE by Auger-excitation of free carriers (Seo *et al.* APL 1999)

  ➔ **Efficient excitation of RE w/o all possible de-excitation process**

- Slow excitation rate?($W_0 = 10^5$ sec^{-1}):
  - For example, 10 µm X 10 µm X 1 cm Er-doped waveguide based on nc-Si:Er with ~ 0.1 at % of Er

    → ~ 100 mW output power is feasible

    → Enough for the practical application

Summary and Conclusion

- **Excitons mediated Nd excitation in nc-Si**
  - The size of nc-Si: crucial for Nd excitation in SRSO:Er
- **3 fold quenching of Nd PL intensity**
  - Due to exciton dissociation
- **The lack of backtransfer**
  - Small energy mismatch (~ 200 meV)
  - However weak couplings between nc-Si and RE
    → responsible for site dependent quenching behavior
- **Efficient RE excitation avoiding thermal quenching possible**