Energy transfer and lifetime measurements in laser crystals

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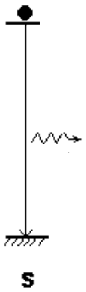
Outline

- Introduction
- Theoretical model
- Numerical results
- Experimental results
- Conclusions

Introduction 1: radiative & non-radiative decay

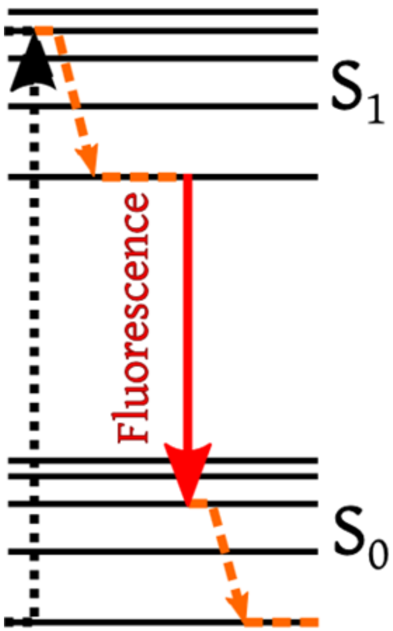
radiative decay



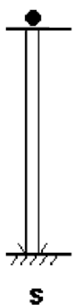
$$dN/dt = -\gamma N$$

$$N = N_o e^{-\gamma t} = N_o e^{-t/\tau}$$

γ: decay rate



nonradiative decay:

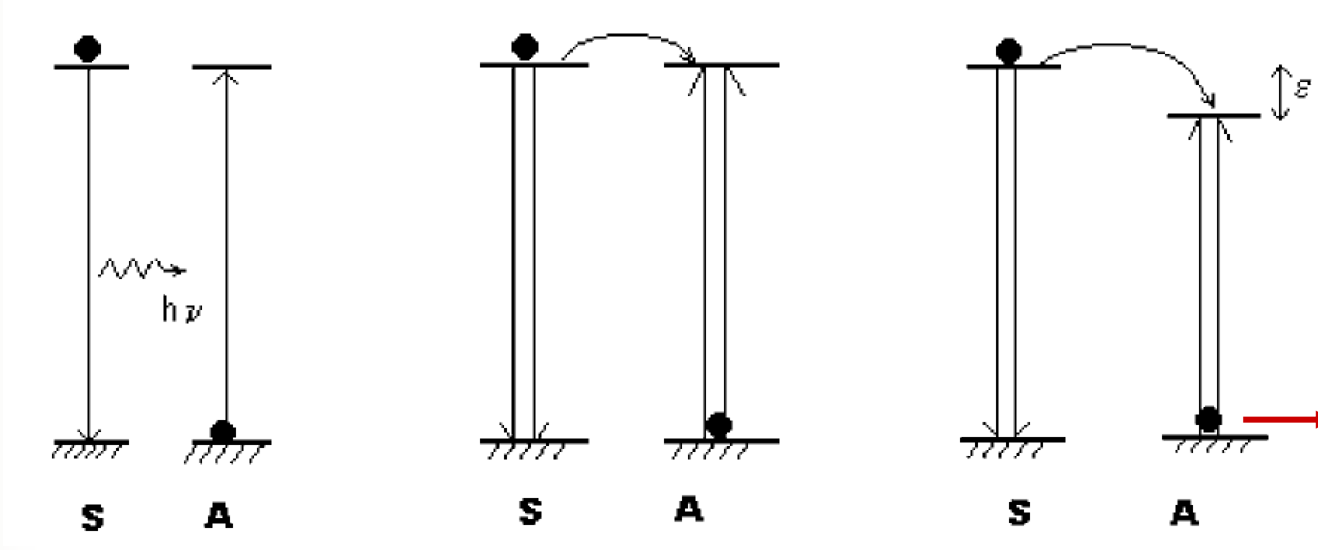


(1)collisions with other atoms

(2) returning to the ground state along a down-hill energy path that involves several coupled vibrational and electronic energy states

Introduction 2: energy transfer

Energy transfer
- radiative(radiation trapping, reabsorption): 與材料(光譜) 形狀 大小 濃度…有關
- non-radiative: dipole-dipole interaction …



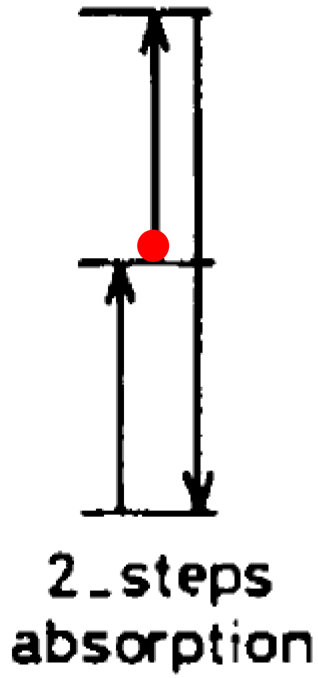
ground state

resonant

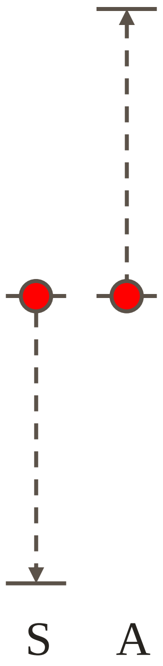
non-resonant
phonon-assisted

Introduction 2: energy transfer

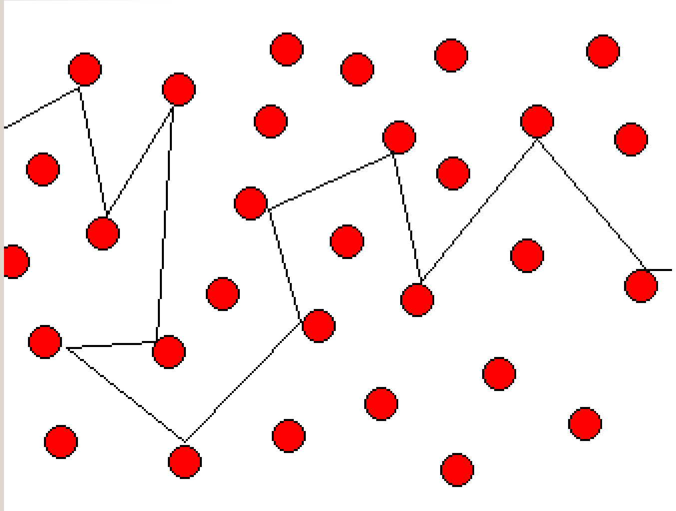
Excited-state absorption (ESA)



Energ transfer upconversion (ETU)



Introduction 3: Radiation trapping



$Compton:$

$$\frac{\partial n(\vec{r},t)}{\partial t} = D\nabla^2 n(\vec{r},t), \quad D = \frac{\ell^2}{3\tau}$$

$\ell : mean\ \ free\ \ path$

$\tau : lifetime\ \ of\ \ individual\ atom$

$$\text{Thermodynamics: } D = \frac{\ell\bar{v}}{3} \cong \frac{\ell}{3}\frac{\ell}{\tau}$$

$$\bar{v} = \frac{\ell}{\tau + \tau'} \cong \frac{\ell}{\tau'}$$ for material particles

$$\cong \frac{\ell}{\tau}$$ for photons energy transfer

$$\frac{dQ}{dt}=\left[\frac{n\bar{v}q(y_0-y')}{6}-\frac{n\bar{v}q(y_0+y')}{6}\right]_{y'=\ell}$$

$$\frac{dQ(y_0)}{dt}=\frac{1}{6}\left[\frac{\partial}{\partial y}n\bar{v}q\Big|_{y=y_0}(-2y')+O(y'^2)\right]_{y'=\ell}$$

$$=-\frac{1}{3}\ell\frac{\partial}{\partial y}\left[n\bar{v}q(y_0)\right]+O(\ell^2)$$

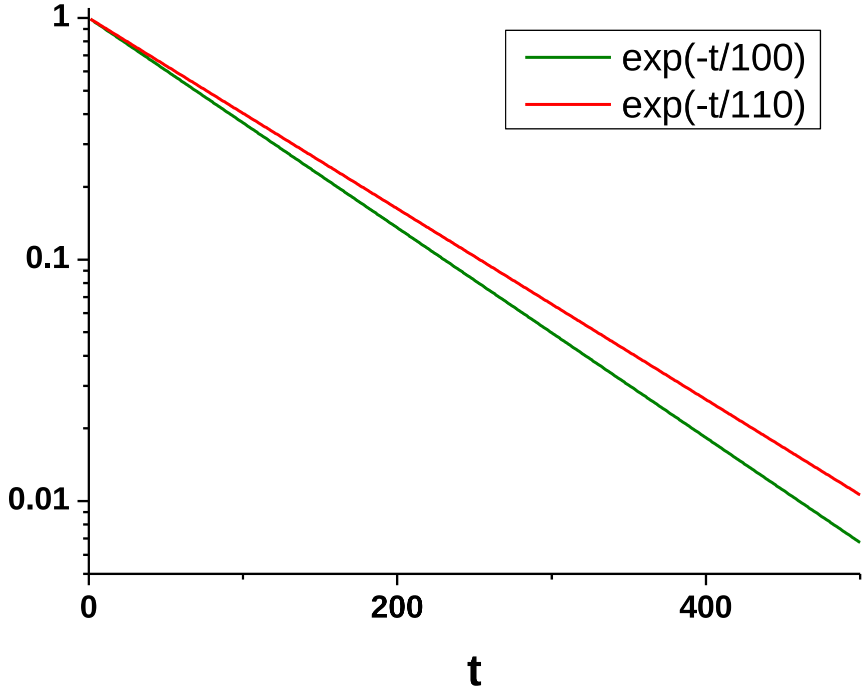
自擴散現象 $q = 1$,

$$J=-D\frac{dn}{dy}=\frac{dQ}{dt}=-\frac{1}{3}\ell\frac{\partial}{\partial y}(n\bar{v})=-\frac{1}{3}\ell\bar{v}\frac{dn}{dy}$$

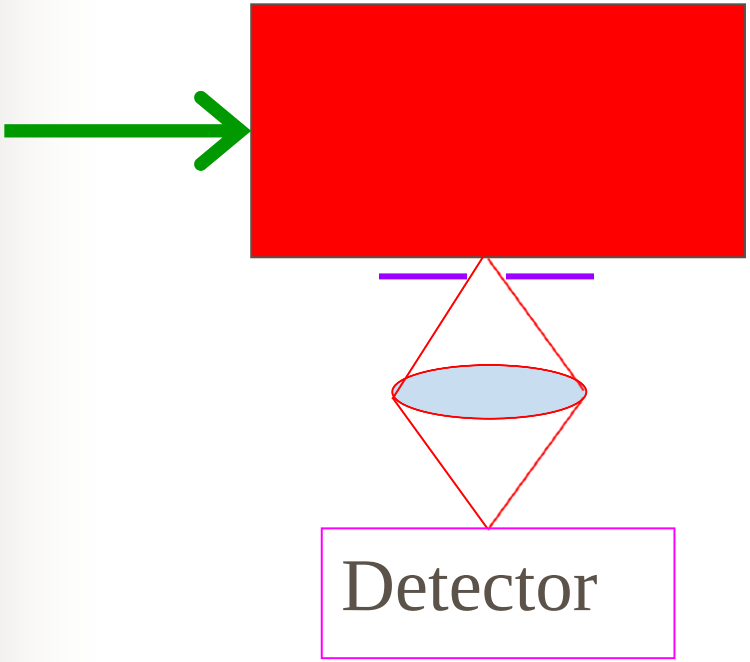
$$\Rightarrow D=\frac{1}{3}\ell\bar{v}$$

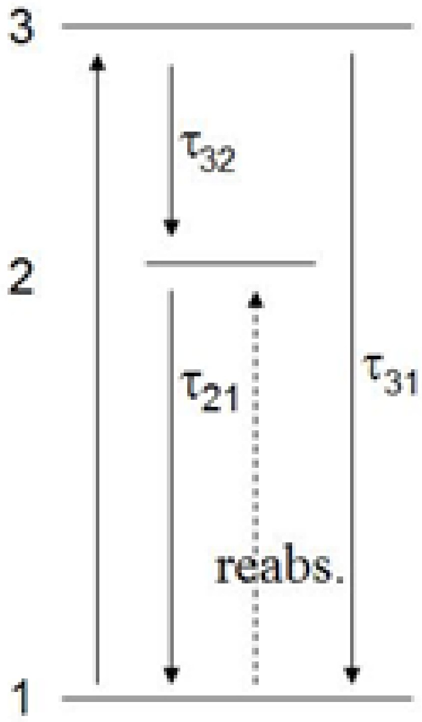
Lifetime lengthening due to radiation trapping

$$dN/dt = -\gamma_{sp}N + \alpha_{rea}N, \quad \gamma_{sp} > \alpha_{rea}$$



Lifetime measurement

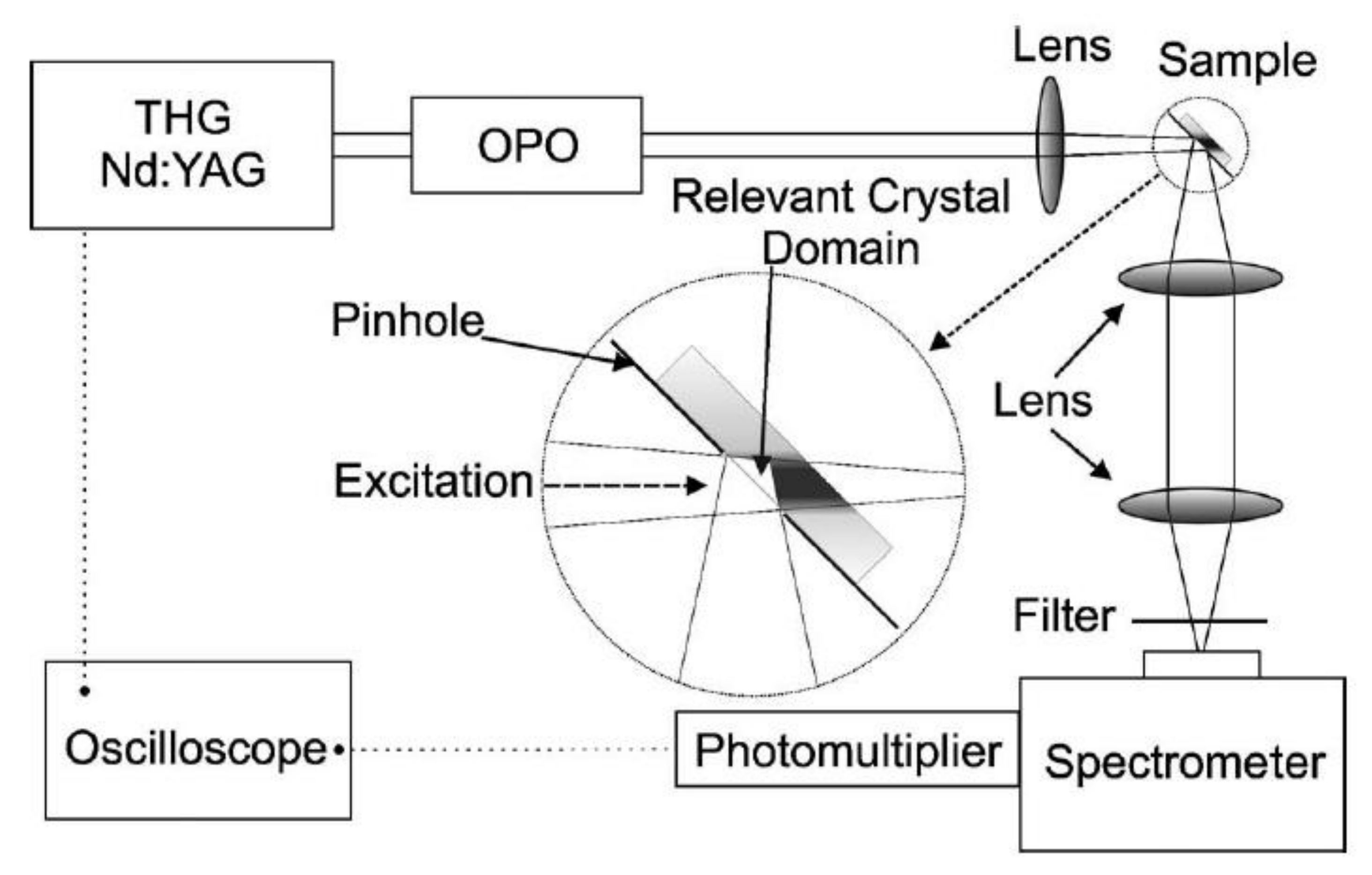




假設
n_g 不變

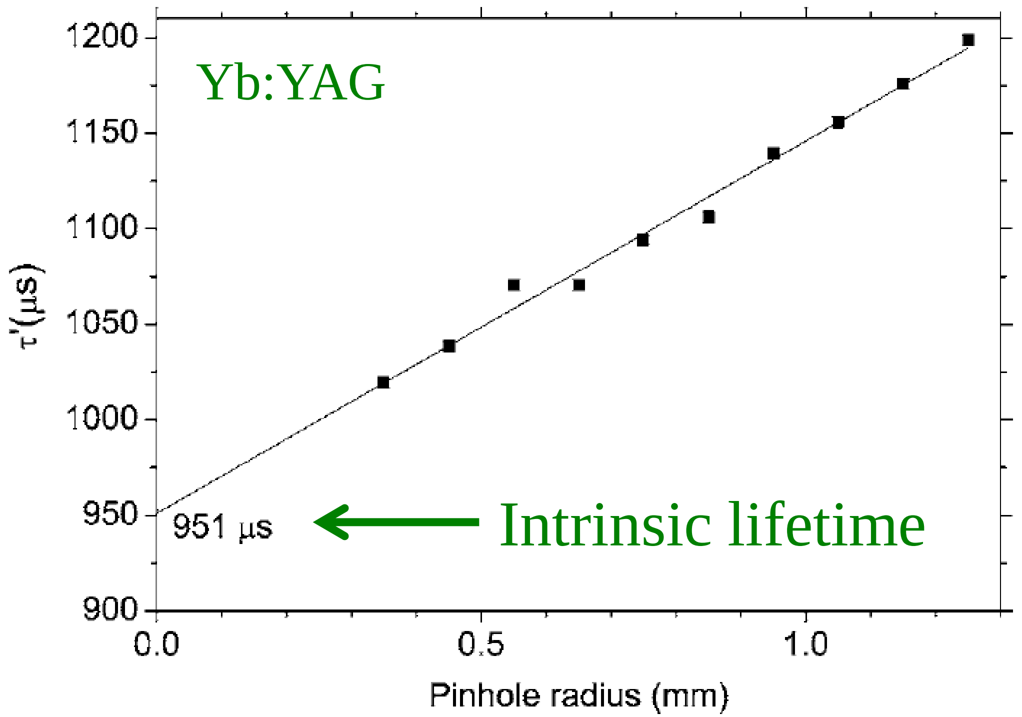
不管pump intensity, 量到就好

Measuring lifetime with pinhole method



H. Kühn et al. Opt. Lett. **32**, 1908-1910 (2007).

Problems using pinhole method



Q1: Using small pinhole needs strong pump.
Q2: Collecting light from unpumped region

Foundation of pinhole method

Consider (1) two-level system (2) excited state density $N \ll N_g$, neglecting stimulated emission

Suppose (1) one region with spatial homogeneous excitation (2)simulating loss by a factor $e^{-|\vec{r}-\vec{r}'|/\zeta}$, $\rho = r - r'$

$$\frac{\partial N(\vec{r},t)}{\partial t} = -\frac{N(\vec{r},t)}{\tau_{21}} + \sigma_g N_g \int \frac{N(\vec{r}',t)}{\tau_{21}} \frac{\exp[-\rho(\sigma_g N_g + 1/\zeta)]}{4\pi\rho^2} 4\pi\rho^2 d\rho$$

$$= -\frac{N(\vec{r},t)}{\tau_{21}} + \frac{N(\vec{r},t)}{\tau_{21}} \frac{\sigma_g N_g}{\sigma_g N_g + 1/\zeta}$$

$$= -\frac{N(\vec{r},t)}{\tau_{21}} + \frac{N(\vec{r},t)}{\tau_{21}} \frac{\sigma_g N_g \zeta}{\sigma_g N_g \zeta + 1}$$

$$= -\frac{N(\vec{r},t)}{\tau_{21}} + \frac{N(\vec{r},t)}{\tau_{21}} \frac{\alpha_g \zeta}{\alpha_g \zeta + 1}$$

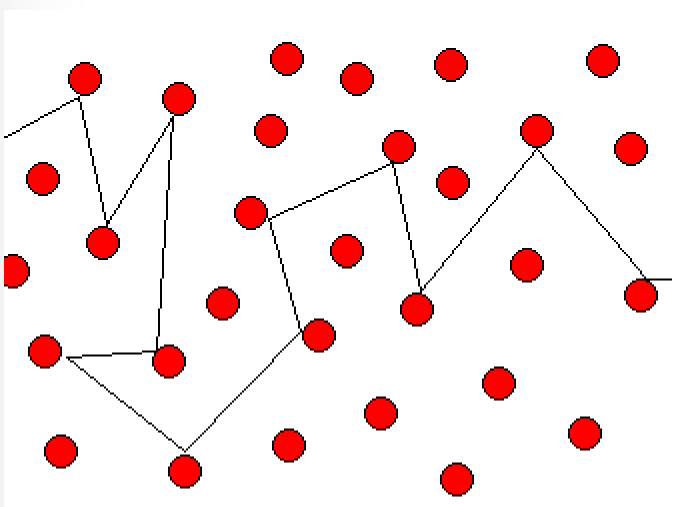
Reabsorption and τ’ depend on ζ that is proportional to pinhole diameter

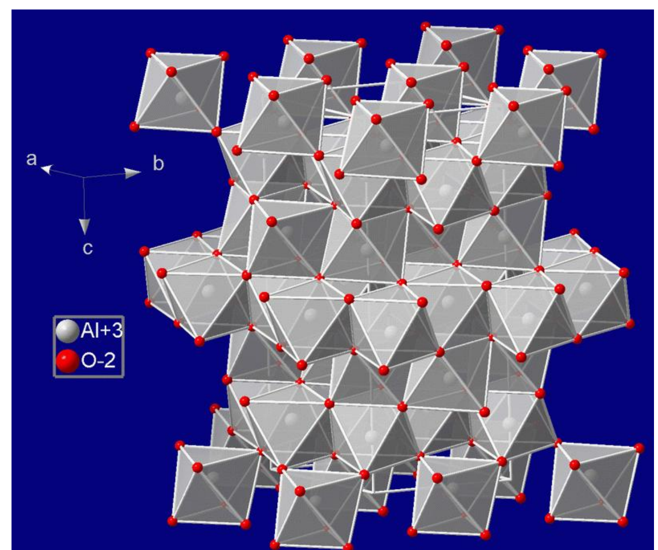
HB equation for radiation trapping

$$\frac{\partial N(\vec{r},t)}{\partial t} = -\frac{N(\vec{r},t)}{\tau_{21}} + W_r \int N(\vec{r}',t)\, G(\vec{r}',\vec{r}) d^3\vec{r}'$$

$G(\vec{r}',\vec{r})d^3\vec{r}$ 表示一個光子在 r’ 被發射而在 dr 附近的體積元素 $d^3\vec{r}$ 被吸收,

$$G(\vec{r}',\vec{r}) = -\frac{1}{4\pi\rho^2}\frac{\partial T}{\partial \rho},\quad T = \exp[-\rho(\sigma_g N_g)] = \exp[-\rho\alpha_g)]$$





Finding Kernel function G(r’, r)

定義光子在 ρ 和 $\rho + d\rho$ 被捕獲的機率爲 $K(\rho)d\rho$

$$K(\rho)d\rho = T(\rho) - T(\rho + d\rho) = -\frac{\partial T}{\partial \rho} d\rho$$

假設自發輻射各向同性, 則光子在 $d^3\vec{r}$ 被捕獲的機率爲

$$\frac{1}{4\pi} d\Omega K(\rho) d\rho$$

引進 $G(\vec{r}', \vec{r}) d^3\vec{r}$ 表示一個光子在 **r’** 被發射而在 **dr** 附近的體積元素 $d^3\vec{r}$ 被吸收, 則

$$G(\vec{r}', \vec{r}) d^3\vec{r} = G(\vec{r}', \vec{r}) \rho^2 d\rho d\Omega = \frac{1}{4\pi} d\Omega K(\rho) d\rho$$

$$\Rightarrow G(\vec{r}', \vec{r}) = \frac{1}{4\pi\rho^2} K(\rho) = -\frac{1}{4\pi\rho^2} \frac{\partial T}{\partial \rho}$$

Definition of transmission function

單一波長時

If the radiation energy transfer problem has mean free path, then the probability of transmission (not absorbed) after distance ρ is $T(\rho) = e^{-\rho/\ell}$

非單一波長時

$T(\rho) = \int P(\lambda) e^{-\rho\alpha(\lambda)} d\lambda,\quad \alpha(\lambda)$: absorption coefficient

HB eq. applied to two-region model

Consider (1) two-level system (2) energy transport governed by TIR (3) high symmetry geometry

Suppose (1) no correlation between emission point and absorption point (2) energy redeposition is unpredictable (3) two regions D&U → $G(\vec{r}', \vec{r}) = G = \dfrac{f}{V_s}$

$$\frac{\partial n(\vec{r},t)}{\partial t} = -\frac{n(\vec{r},t)}{\tau_{21}} + W_r \int n(\vec{r}',t)\, G(\vec{r}',\vec{r})\, d^3\vec{r}'$$

$$\frac{dN_D}{dt} = -\frac{N_D}{\tau_{21}} + W_r N_D \left(\frac{fV_D}{Vs}\right) \times 1 + W_r N_U \left(\frac{fV_U}{Vs}\right) \times 1$$

$$\frac{dN_U}{dt} = -\frac{N_U}{\tau_{21}} + W_r N_U \left(\frac{fV_U}{Vs}\right) \times 1 + W_r N_D \left(\frac{fV_D}{Vs}\right) \times 1$$

D to D

U to D

D to U

U to U

Solution of HB rate equation

Initial condition: $N(\vec{r}, t=0) = \begin{cases} N_D^0, r \in D \\ 0, r \notin D \end{cases}$

Solution:

$$N_D(t) = e^{-t/\tau} N_D^0 + N_U(t)$$

$$N_U(t) = (e^{-t/\tau^*} - e^{-t/\tau}) \frac{V_D N_D^0}{V_s}$$,其中 $\tau^* = \frac{\tau}{1-\eta f}$,

$$\eta = \tau W_r,$$

$$\frac{1}{\tau} = W_r + W_{nr}$$

double exponential function

S. Guy, Phys. Rev. B **73**, 144101 (2006).

Short-range coupling

$$G(\vec{r}',\vec{r}) = \frac{1}{4\pi\rho^2}K(\rho) = -\frac{1}{4\pi\rho^2}\frac{\partial T}{\partial \rho}$$

As $\rho \to 0$, G is large and not constant

$$\therefore G(\vec{r}',\vec{r}) = -\frac{1}{4\pi\rho^2}\frac{\partial T}{\partial \rho} + \frac{f}{V_s}$$

long-range coupling

short-range coupling

Suppose $n_D(\vec{r},t) = N_D(t) f_D(\vec{r}), n_U(\vec{r},t) = N_U(t) f_U(\vec{r}), \int f_D(\vec{r}) d^3\vec{r} = 1$

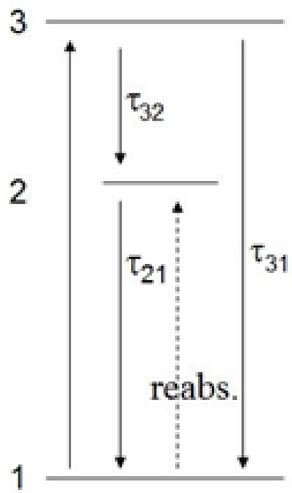
$$\frac{dN_D}{dt} = -\frac{N_D}{\tau_{21}} + W_r N_D(\bar{\alpha}\langle\bar{\rho}\rangle_D) + W_r N_D(\frac{fV_D}{Vs}) + W_r N_U(\frac{fV_D}{Vs})$$

$$\frac{dN_U}{dt} = -\frac{N_U}{\tau_{21}} + W_r N_U(\bar{\alpha}\langle\bar{\rho}\rangle_U) + W_r N_D(\frac{fV_U}{Vs}) + W_r N_U(\frac{fV_U}{Vs})$$

G. Toci, Appl. Phys. B **106**, 63-71 (2012).

Our rate equations for intermediate regime

Consider (1) three-level system (2) two regions (3) pump-dependent



$$\frac{dN_{D3}}{dt}=R_p-\frac{N_{D3}}{\tau_{31}}-\frac{N_{D3}}{\tau_{32}}$$

$$\frac{dN_{D2}}{dt}=\frac{N_{D3}}{\tau_{32}}-\frac{N_{D2}}{\tau_{21}}+\frac{N_{D2}}{\tau_{21}}\frac{\sigma N_{D1}\ell_a}{\sigma N_{D1}\ell_a+1}+\frac{N_{U2}}{\tau_{21}}\frac{\sigma N_{D1}\ell_b}{\sigma N_{D1}\ell_b+1}$$

$$\frac{dN_{D1}}{dt}=-R_p+\frac{N_{D3}}{\tau_{31}}+\frac{N_{D2}}{\tau_{21}}-\frac{N_{D2}}{\tau_{21}}\frac{\sigma N_{D1}\ell_a}{\sigma N_{D1}\ell_a+1}-\frac{N_{U2}}{\tau_{21}}\frac{\sigma N_{D1}\ell_b}{\sigma N_{D1}\ell_b+1}$$

$$\frac{dN_{U2}}{dt}=-\frac{N_{U2}}{\tau_{21}}+\frac{N_{U2}}{\tau_{21}}\left(\frac{\sigma N_{U1}\ell_{c1}}{\sigma N_{U1}\ell_{c1}+1}+\frac{\sigma N_{U1}\ell_{c2}}{\sigma N_{U1}\ell_{c2}+1}\right)+\frac{N_{D2}}{\tau_{21}}\frac{\sigma N_{U1}\ell_d}{\sigma N_{U1}\ell_d+1}$$

$$\frac{dN_{U1}}{dt}=\frac{N_{U2}}{\tau_{21}}-\frac{N_{U2}}{\tau_{21}}\left(\frac{\sigma N_{U1}\ell_{c1}}{\sigma N_{U1}\ell_{c1}+1}+\frac{\sigma N_{U1}\ell_{c2}}{\sigma N_{U1}\ell_{c2}+1}\right)-\frac{N_{D2}}{\tau_{21}}\frac{\sigma N_{U1}\ell_d}{\sigma N_{U1}\ell_d+1}$$

$$R_p=\frac{P_{inc}/h\nu}{\pi w_p^{2}\ell}\frac{N_{D1}}{N_t}$$

The cause using nonlinear coefficient

(1) $\sigma N_{D1}\ell_a > 1$ is possible

(2) $\sigma N_{D1}\ell_a > 1$ means reabsoption > spon. emission

unreasonable

(3) $0 < \dfrac{\sigma N_{D1}\ell_a}{\sigma N_{D1}\ell_a + 1} < 1$

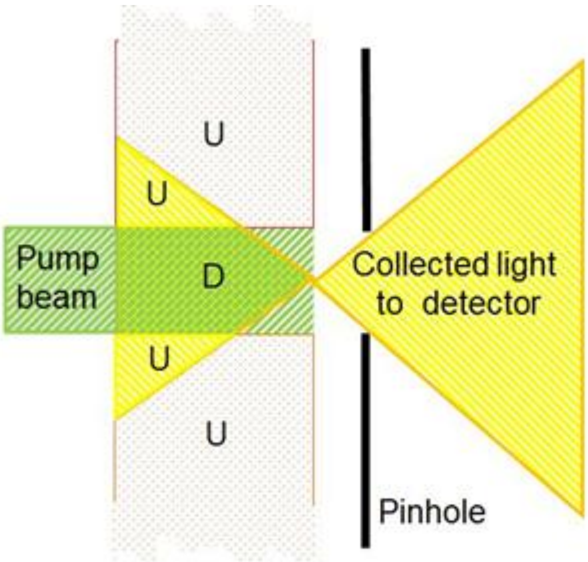
(4) As $\sigma N_{D1}\ell_a << 1$, $\dfrac{\sigma N_{D1}\ell_a}{\sigma N_{D1}\ell_a + 1} \cong \sigma N_{D1}\ell_a$

(5) As $\sigma N_{D1} l_a >> 1$, $\dfrac{\sigma N_{D1}\ell_a}{\sigma N_{D1}\ell_a + 1} \cong 1$

simulating reabs. due to TIRs

(6) $0 < \dfrac{fV_U}{Vs} \leq 1$, extending (3) to long-range coupling

Our collecting consideration



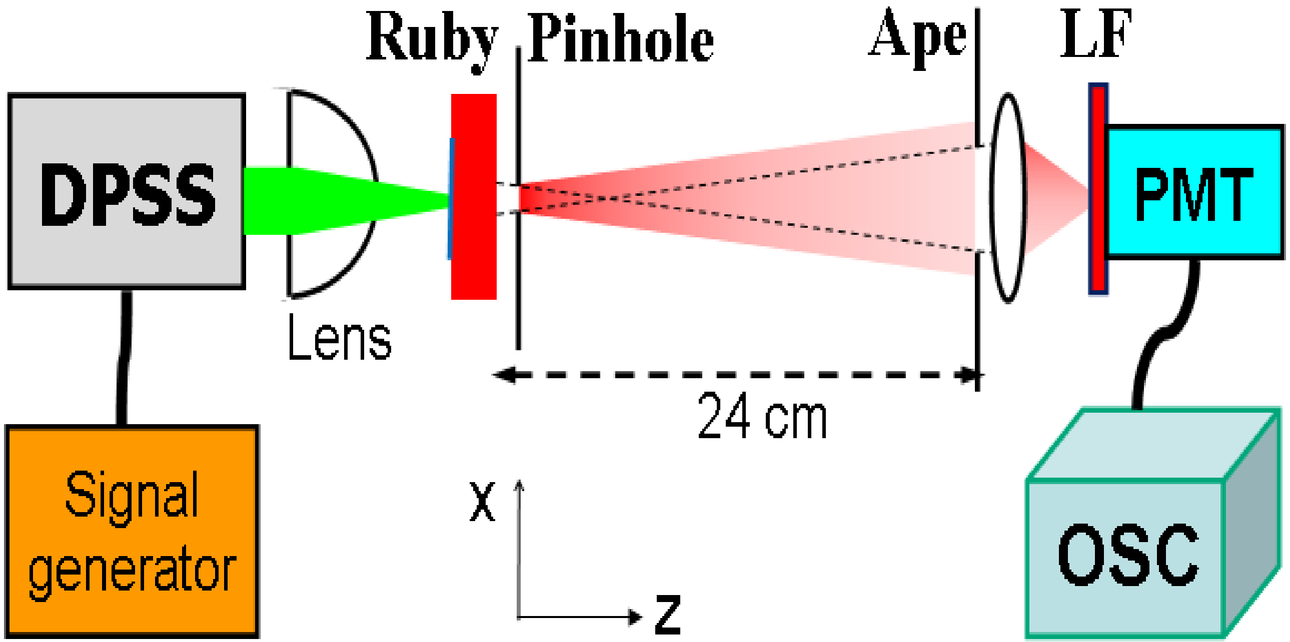
Parameters for 0.05at% ruby crystal

$N_t = 2.4\times10^{19}\ cm^{-3}$, $\tau_{31} = 3.3\times10^{-6}\ s$, $\tau_{32} = 5\times10^{-8}\ s$, $\tau_{21} = 2.8\times10^{-3}\ s$, $\sigma = 1\times10^{-19}\ cm^2$, $f = 0.5$,

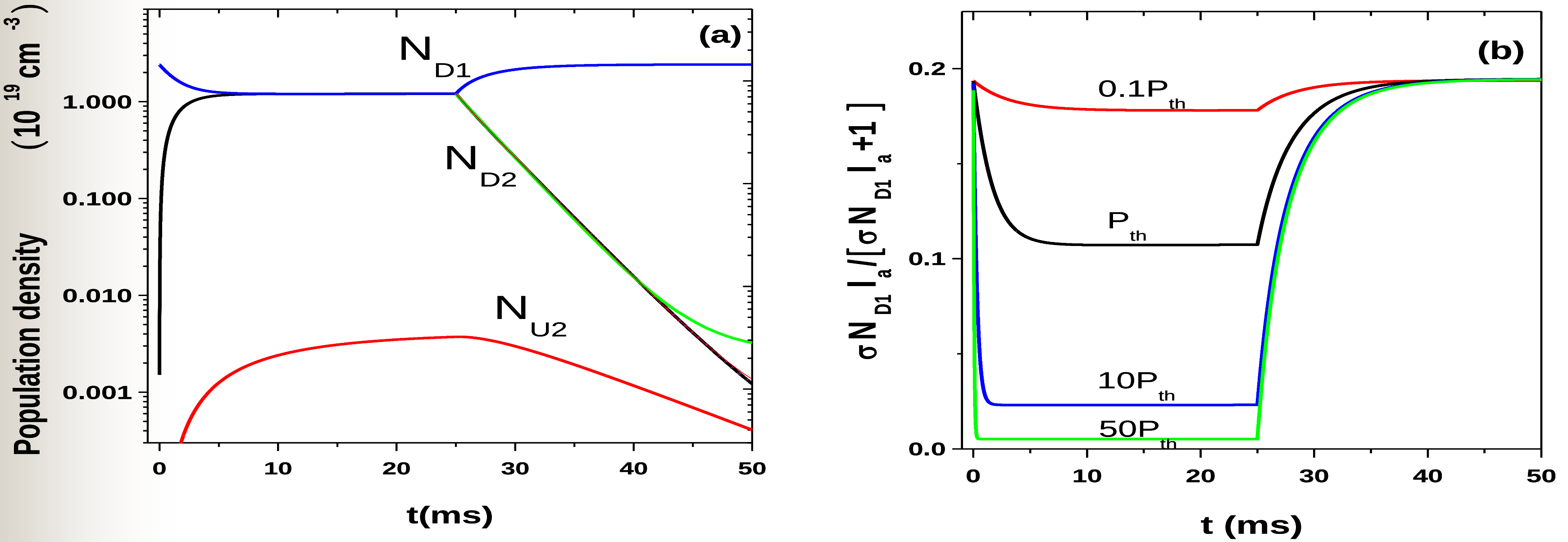
$V_U / V_S \approx 1,\ \ell_b = \ell_c = 4.25mm,\ \ell_d = 4.25\times10^{-3} mm,$

$\Delta t = 2\times10^{-8} s$

Experimental setup of **our axial pinhole method**



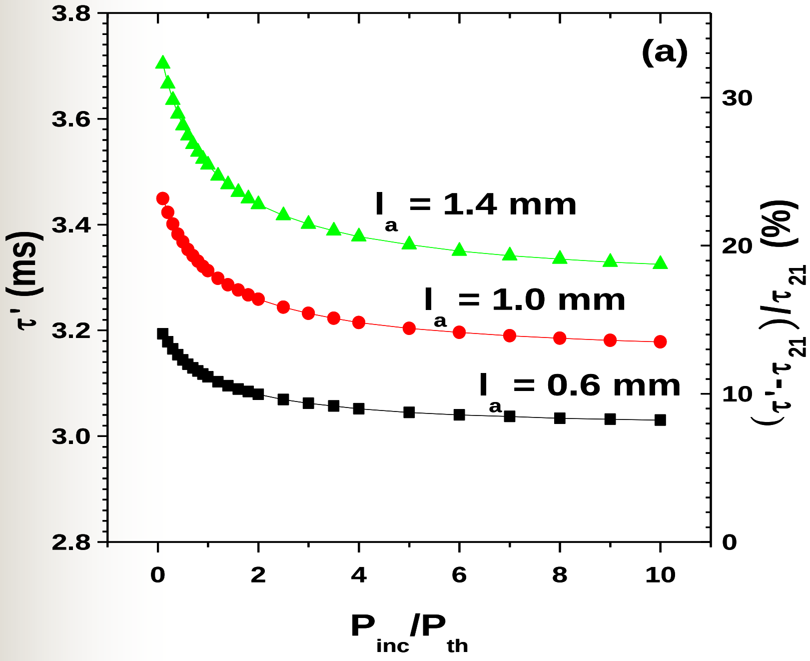
Result 1: saturation of radiation trapping

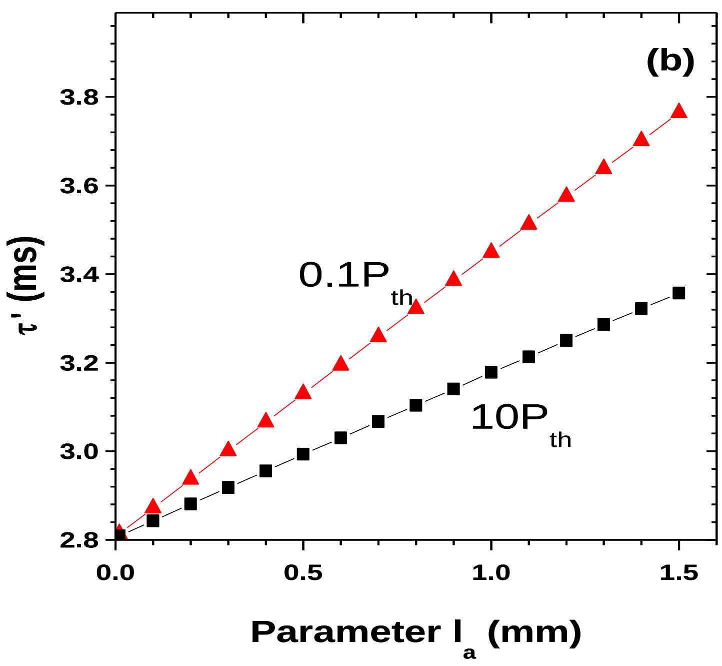


Fitted with single & double exponential function

$$A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2)$$

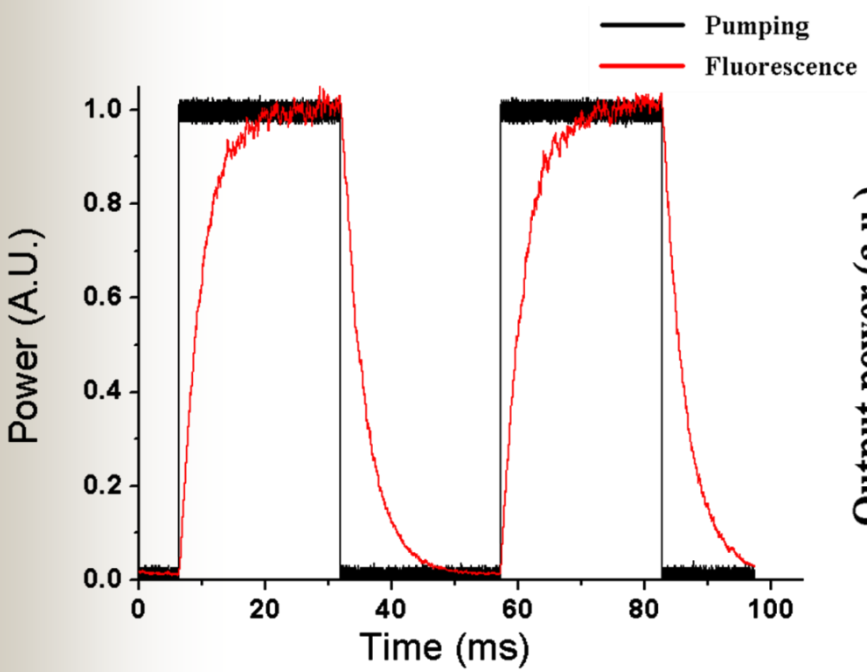
Result 2: lifetime clamping due to saturation of radiation trapping

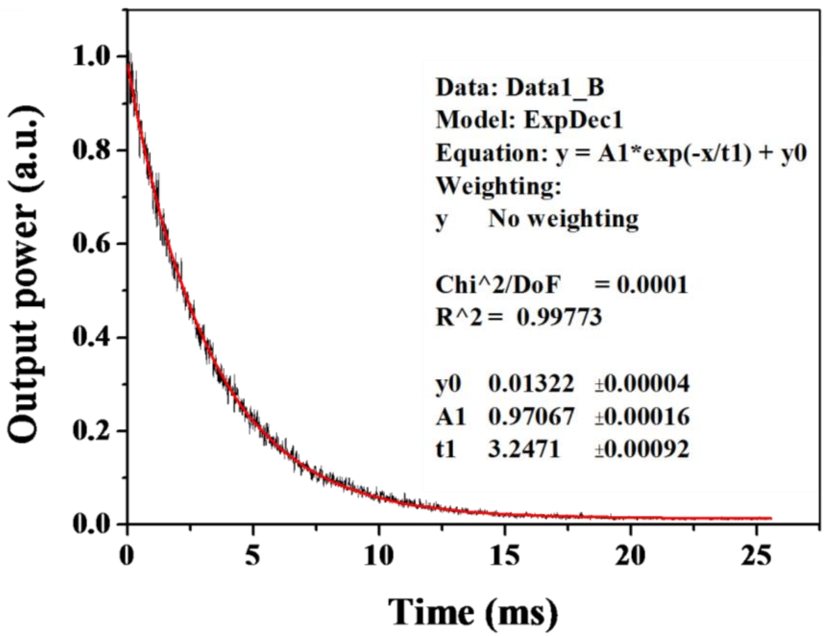




extending pinhole method

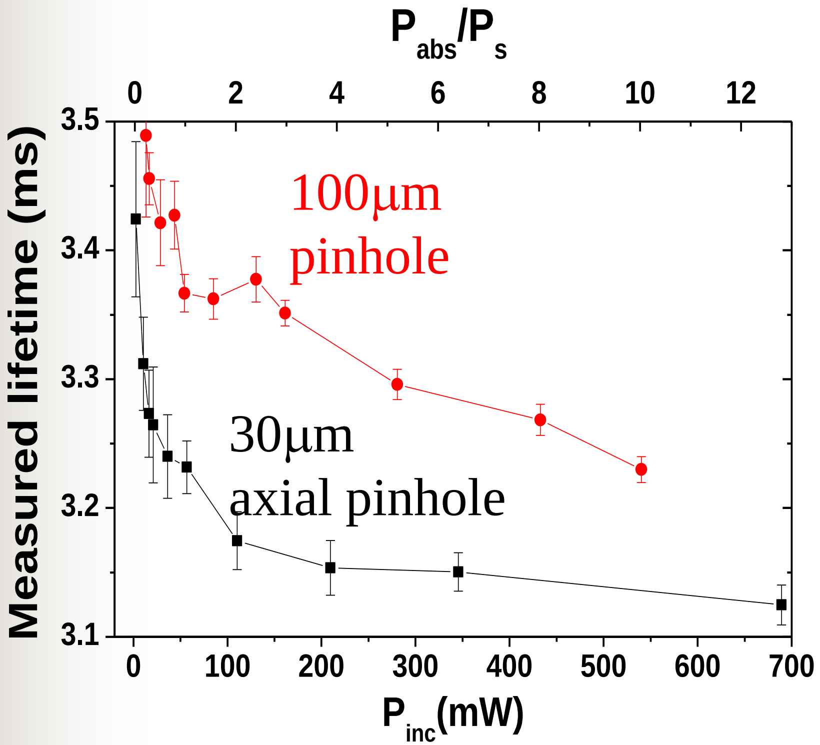
Exp. result: fluorescence decay & fitting



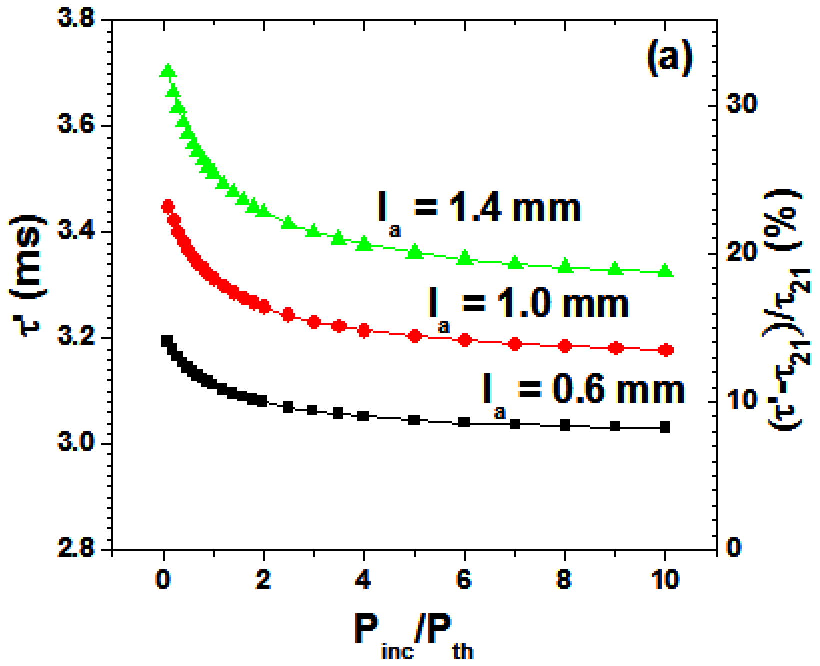


Exp. result: measured lifetime vs. pump

Experiment



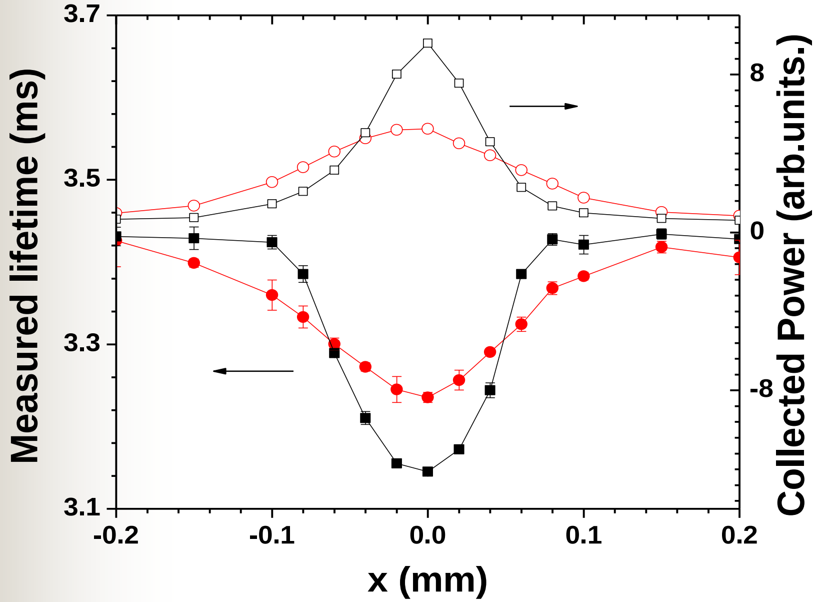
Simulation

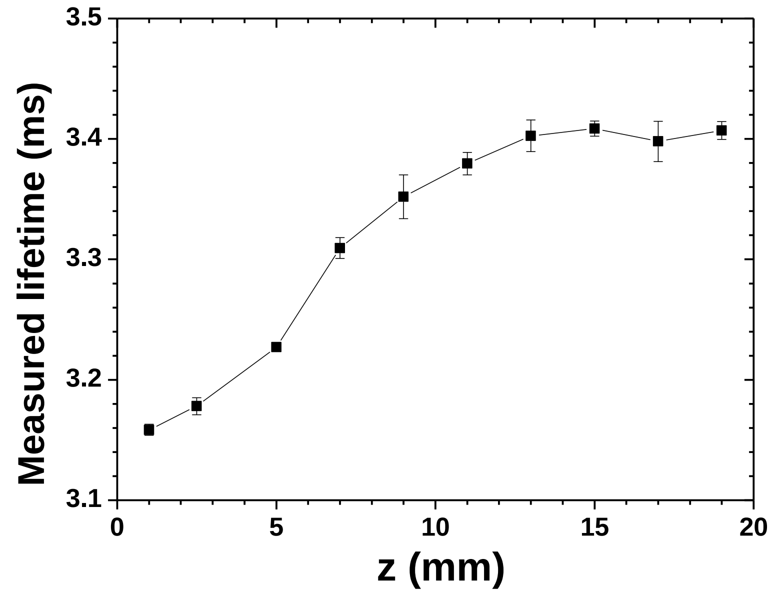


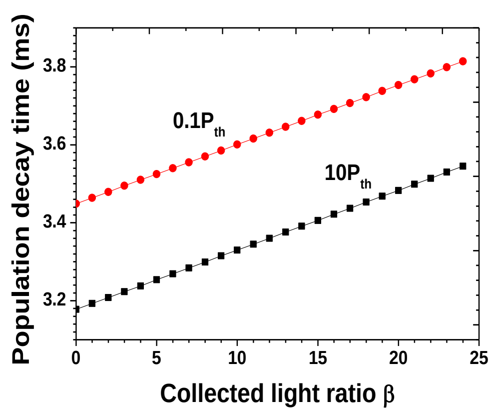
$$\Delta T = (\alpha P_{ph} e^{-\alpha\ell} / 2\pi K_c)\int_r^{r_b} (1-e^{-2r^2/w_p^2})dr / \mathrm{r} \approx 0.2°C$$

$$ASE\ term:\ -\sigma_e \mathrm{N}^2{}_{\mathrm{D2}}\ell_s / \tau_{21} \Rightarrow \text{small signal gain} < 1.06$$

Result: measured lifetime vs. x , z







Conclusions

1. The pump intensity was considered for the first time to give a theoretical description of radiation trapping. The intermediate radiation trapping regime $\alpha\ell \approx 1$ was treated.
2. Saturation of the radiation trapping was numerically verified under strong pumping and this saturation clamps the lifetime lengthening.
3. We experimentally verified the numerical results by an axial pinhole method on a ruby crystal.
4. Our model is confirmed valid for measured lifetime with the fluorescence from both the pumped and the unpumped regions.

Thanks for Listening