Radiation by out-of-equilibrium fluctuating currents: application to scintillators

Based on Roques-Carmes* & **Rivera*** *et al.* Science (2022).

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Scintillators are a ubiquitous tool to detect high-energy particles (ionizing radiation)

Core phenomenon

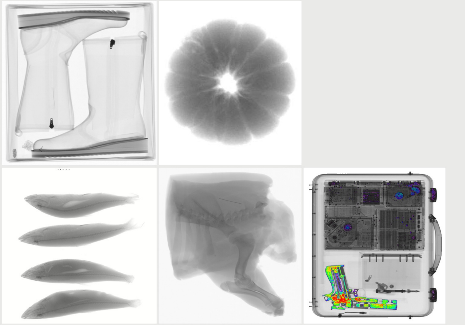
Ionizing radiation (e^-, X, γ)

Photons

Applications

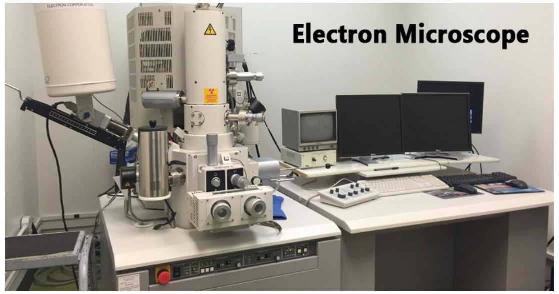
Medical imaging

Non-destructive testing



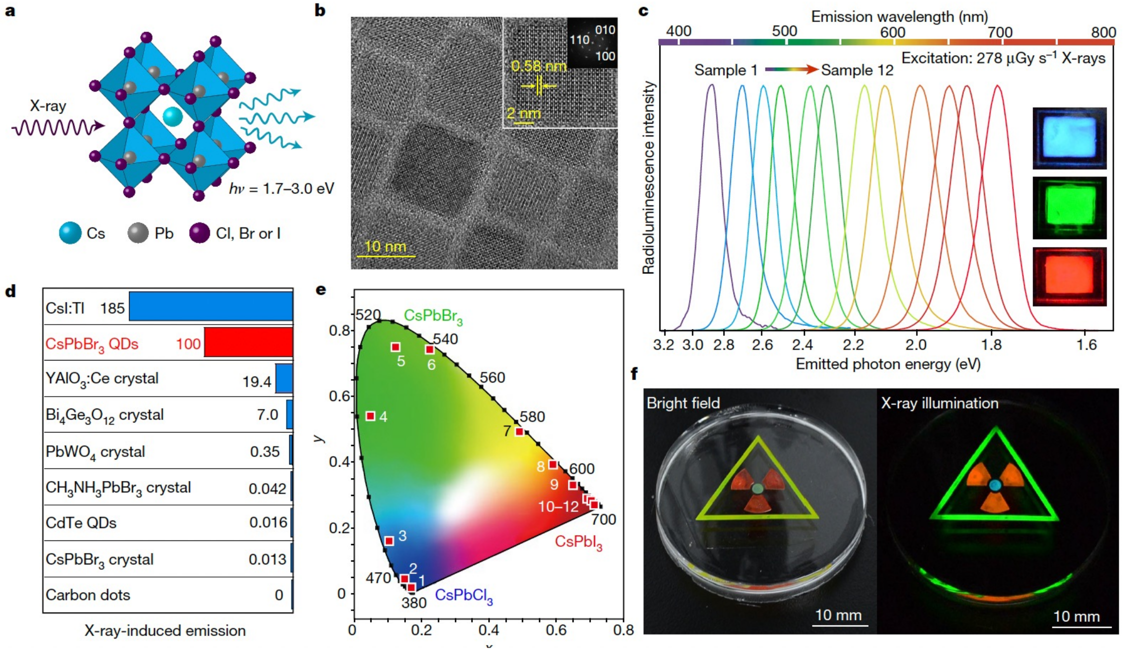
Radiation monitoring

Electron microscopy



Much knowledge (collected over decades) about scintillators in classic textbooks like Lecoq *et al.*

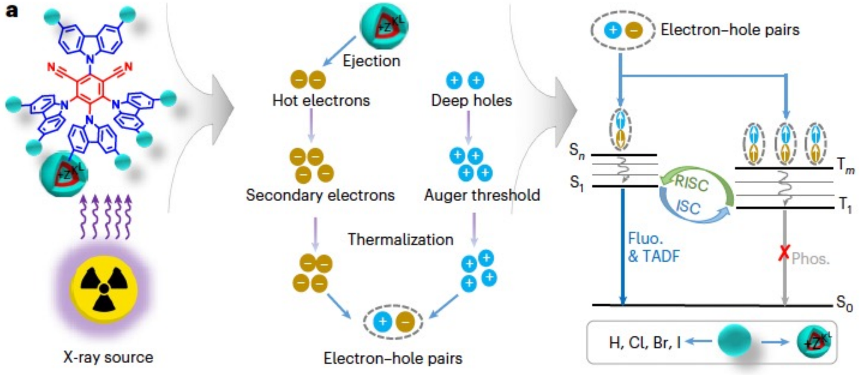
The quest for better scintillators is still on



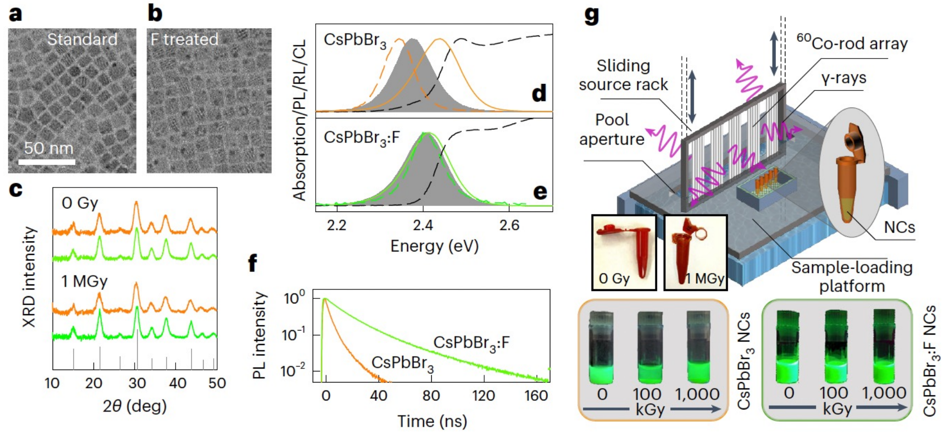
Chen *et al.* Nature (2018)

perovskites

heavy-atom engineered chromophores

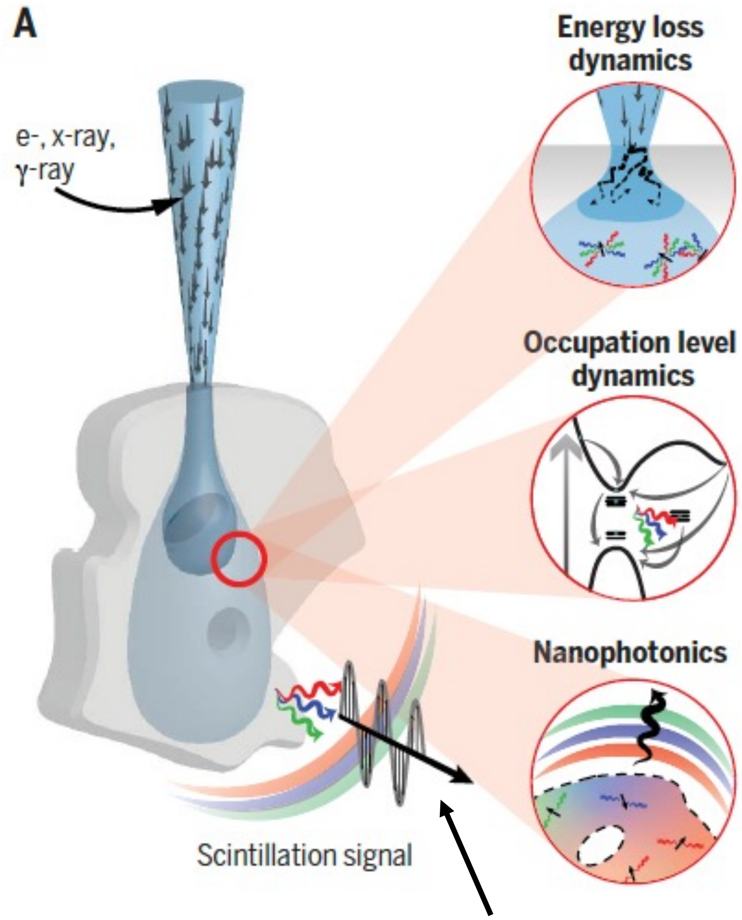


Wang *et al.* Nat. Photon. (2022)



Zaffalon *et al.* Nat. Photon. (2022; yesterday!)

A different approach: scintillation engineering by nanophotonic structures

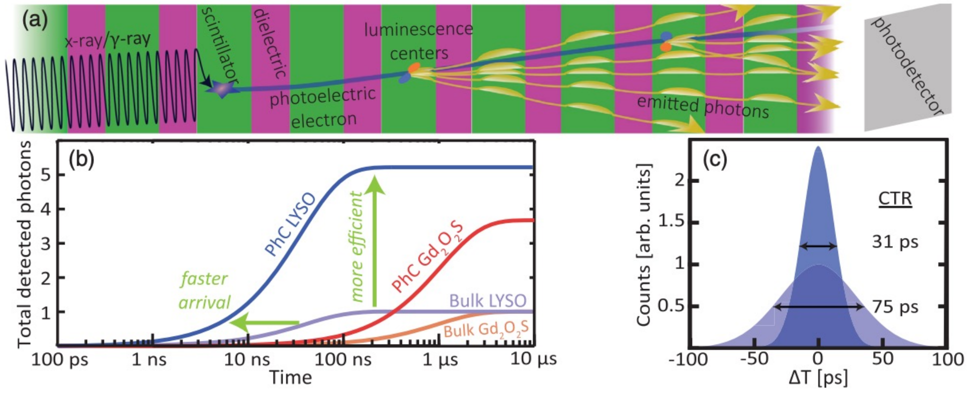


spontaneous emission!

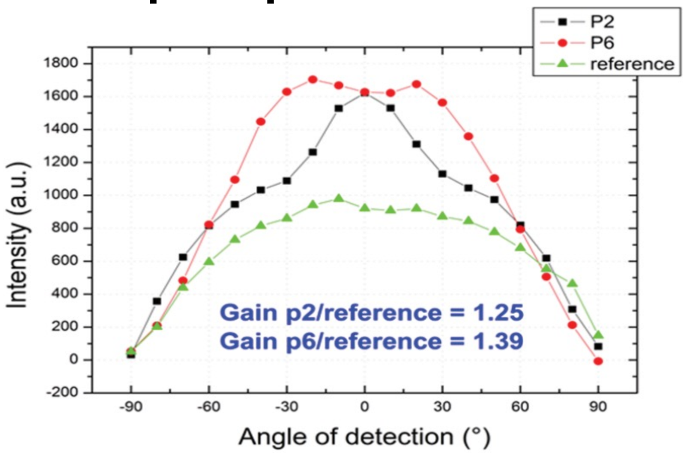
Controlling emission with nanophotonics

Purcell. Phys. Rev. (1946) [Purcell effect]
Yablonovitch. Phys. Rev. Lett. (1983) [Purcell in photonic crystals]
Gerard *et al.* Phys. Rev. Lett. (1998) [Purcell in a resonant cavity]
Greffet *et al.* Nature (2003) [thermal radiation]
Polman & Atwater. Nat. Mater. (2010) [solar cells]

Early work enhancing scintillation with pure photonics

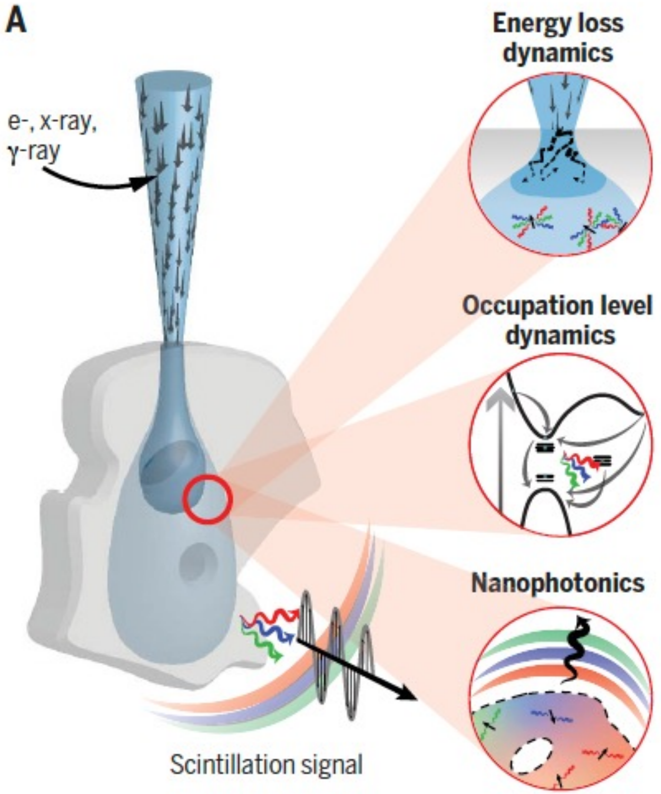


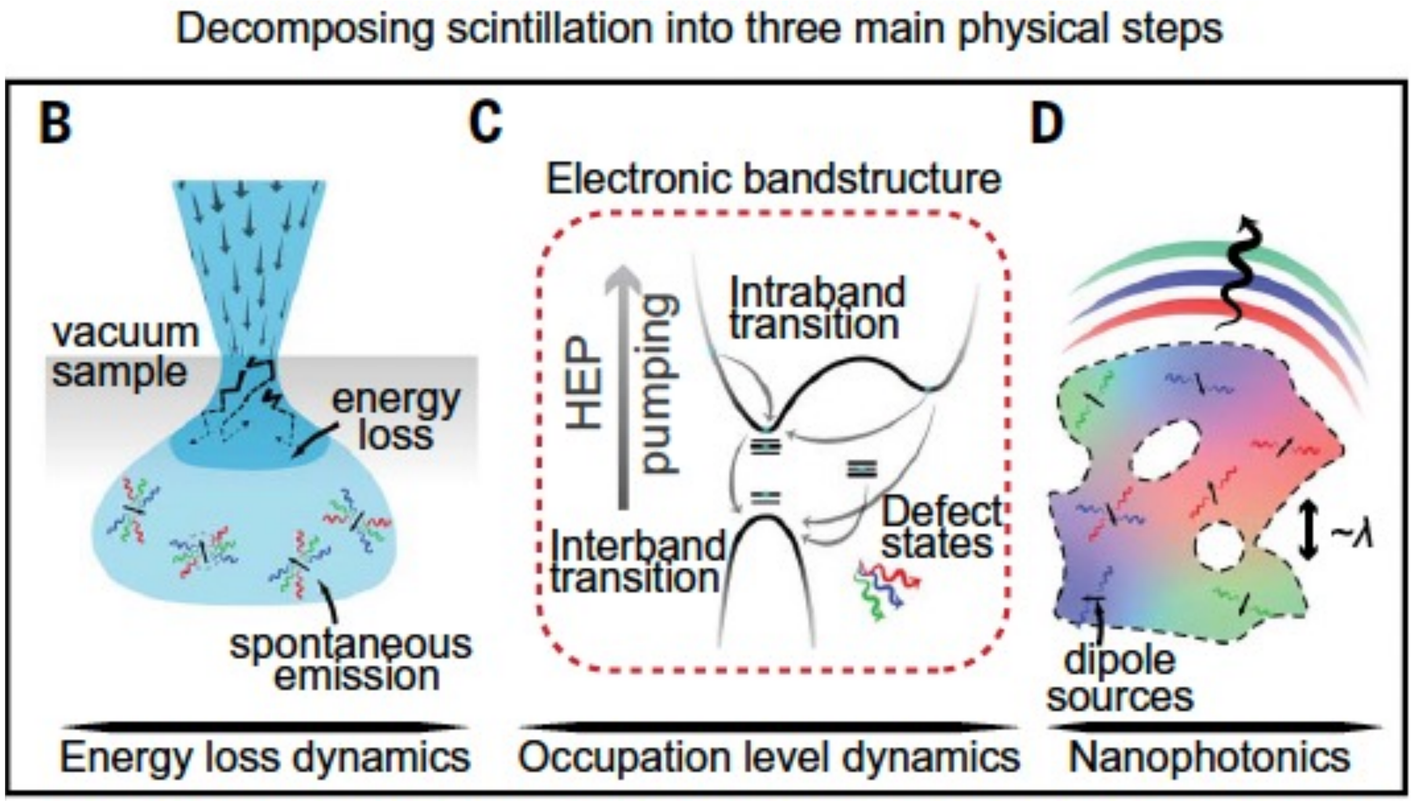
Kurman *et al.* Phys. Rev. Lett. (2020)
[Kaminer group]



Knapitsch *et al.* IEEE Trans. Nuc. Dev. (2010) [Lecoq group]

A general framework for nanophotonic scintillation





[One of our main contributions: general framework to model nanophotonic scintillation taking this picture into account.]

Roques-Carmes* & **Rivera*** *et al.* Science (2022).

A general framework for describing nanophotonic scintillators

- Spectral & angular density of emitted light in scintillation

[specified by Maxwell solutions]

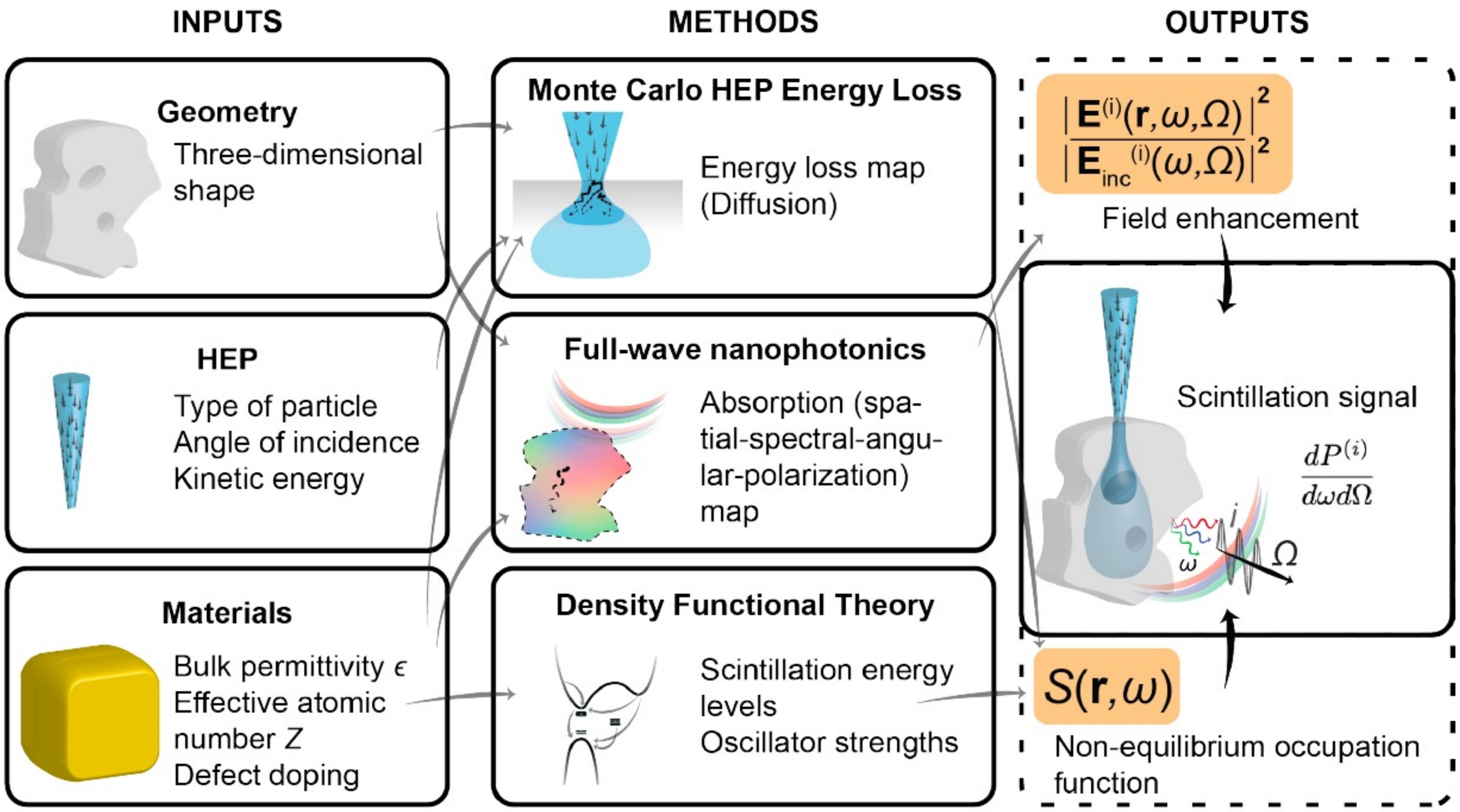
$$\frac{dP^{(i)}}{d\omega d\Omega} = \frac{\omega^2}{8\pi^2\epsilon_0 c^3}\int d\mathbf{r}'\,\frac{E_j^{*(i)}(\mathbf{r}',\mathbf{r},\omega)}{\left|\mathbf{E}_{\rm inc}^{(i)}(\mathbf{r}',\mathbf{r},\omega)\right|}\,\frac{E_k^{(i)}(\mathbf{r}',\mathbf{r},\omega)}{\left|\mathbf{E}_{\rm inc}^{(i)}(\mathbf{r}',\mathbf{r},\omega)\right|}\,S_{jk}(\mathbf{r}',\omega)$$

$$S_{jk}(\mathbf{r_1},\mathbf{r}_2,\omega) \sim \langle J_j^-(\mathbf{r}_1,\omega)J_k^+(\mathbf{r}_2,\omega)\rangle$$

[specified by microscopics: energies, matrix elements, occupations, energy loss]

- Strong similarity to description of thermal radiation (except for non-equilibrium current-current correlations) [essentially same tools as used to model thermal radiation.]

The end-to-end numerical pipeline for nanophotonic scintillation, summarized

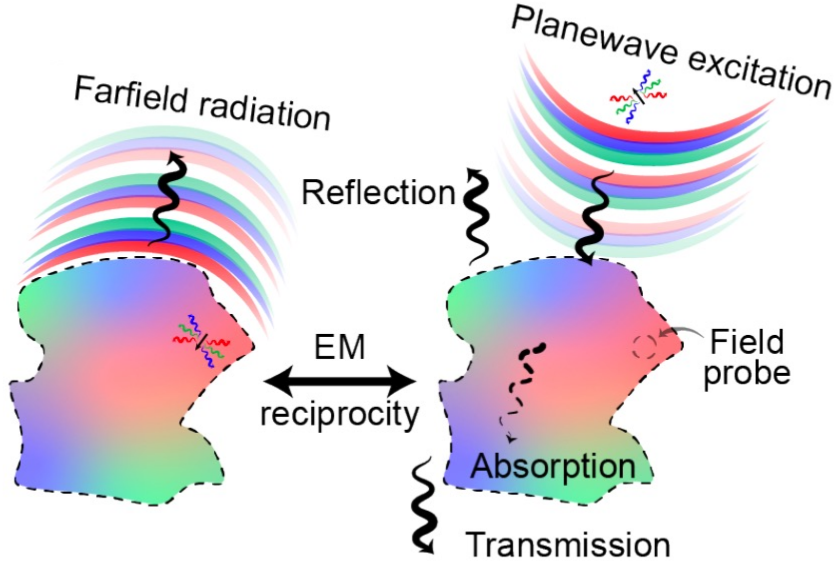


Designing a better scintillator = designing a better absorber

Reciprocity-based calculations in non-equilibrium settings
Schulz et al., Optics Express (2018)
Sheng et al., Nano Letters (2018)
Zhang et al., Synth. Metals. (2015)
Greffet et. al., Physical Review X (2018)

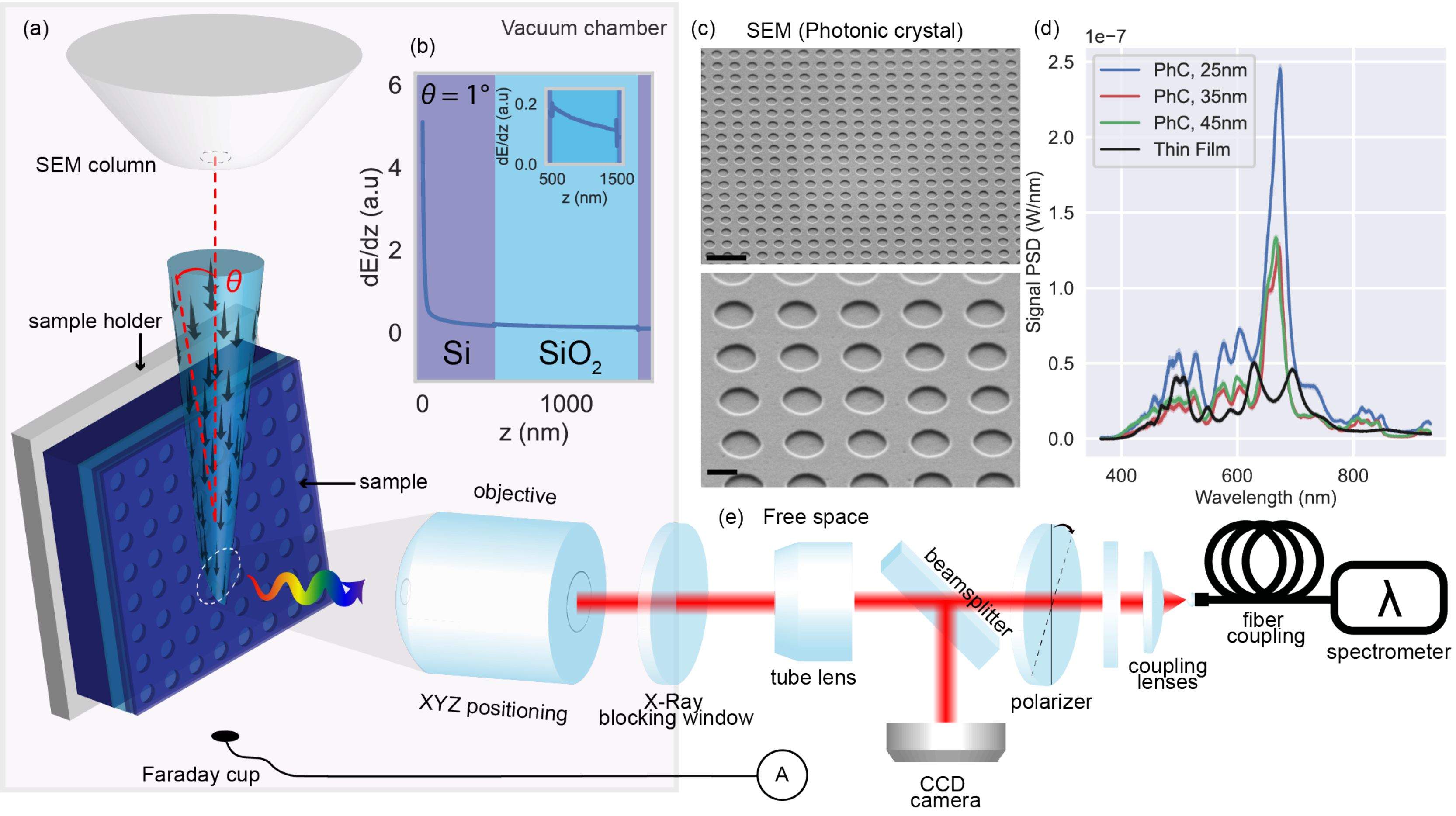
- In the case of a uniform spatial distribution of scintillating centers in some volume:

$$\frac{dP}{d\omega d\Omega} = \frac{\pi}{\epsilon_0\omega} \times \underbrace{S(\omega)}_{\text{spectral function}} \times \underbrace{(V_{eff}(\omega)/\lambda^3)}_{\text{proportional to “absorption”: } \sum_i \int d^3r' \left|\frac{\mathbf{E}(\mathbf{r}',\mathbf{r},i,\omega)}{E_{inc}(\mathbf{r}',\mathbf{r},i,\omega)}\right|^2}$$



- In this theory of scintillation, the power spectrum can be calculated (and optimized) readily from absorbed power in a volume!
  - Can now take ideas from solar cells, LEDs, thermal emission
  - Inverse design and optimization now become feasible for scintillators
  - Powerful way to understand Purcell from ensembles

Enhancement of electron-induced scintillation in photonic crystal slabs

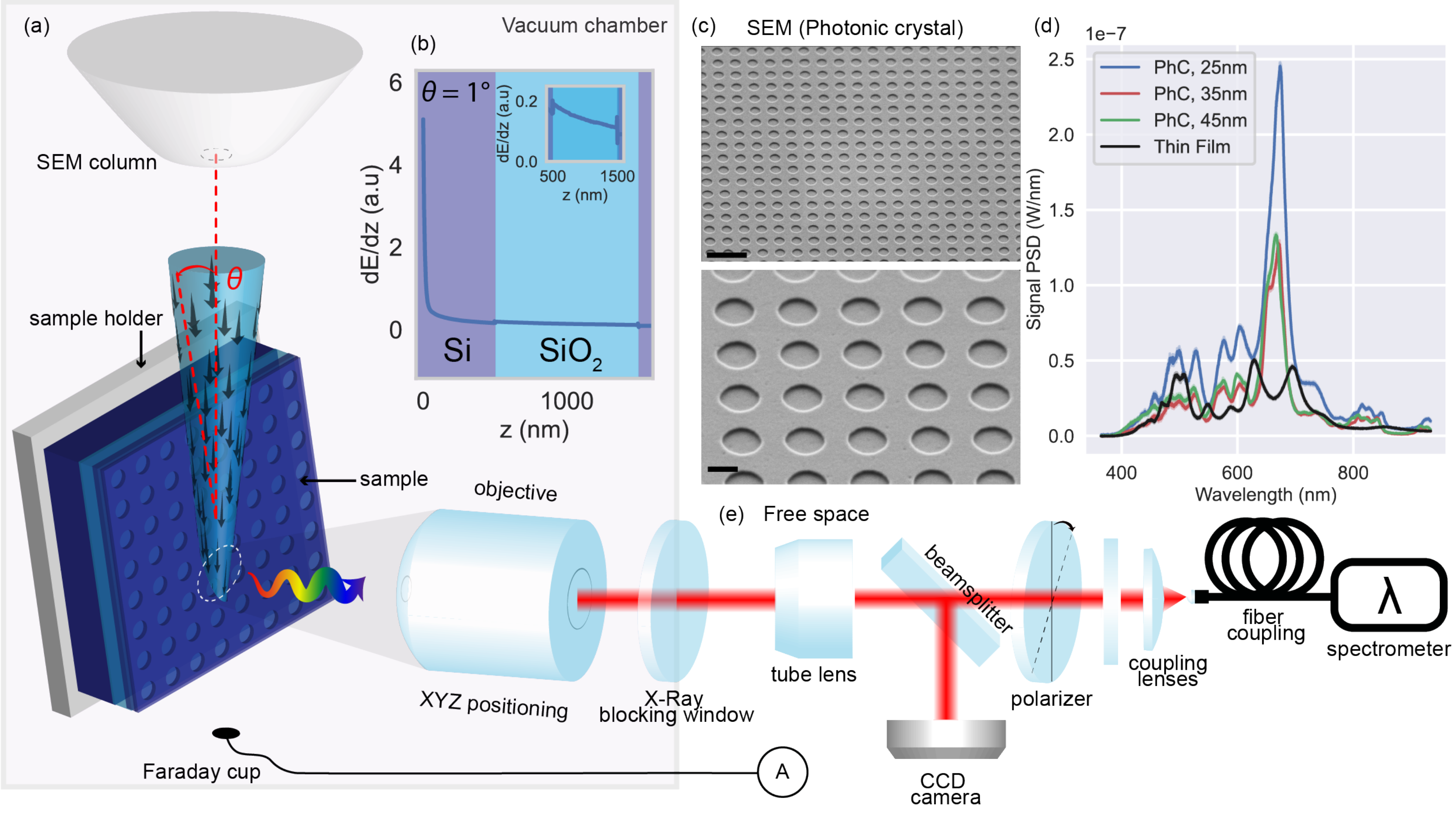


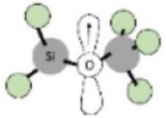
Charles Roques-Carmes
[postdoc at MIT/Stanford]

Nanophotonics in our SEM
Kaminer *et al.* Phys. Rev. X (2017)
Yang *et al.* Nat. Phys. (2018)
Roques-Carmes *et al.* Nat. Comm. (2019)

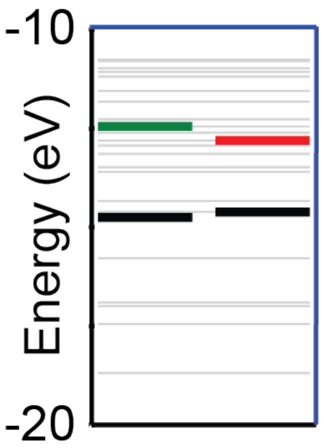
Roques-Carmes* & **Rivera*** *et al.* Science (2022).

Scintillation by self-trapped holes in silica

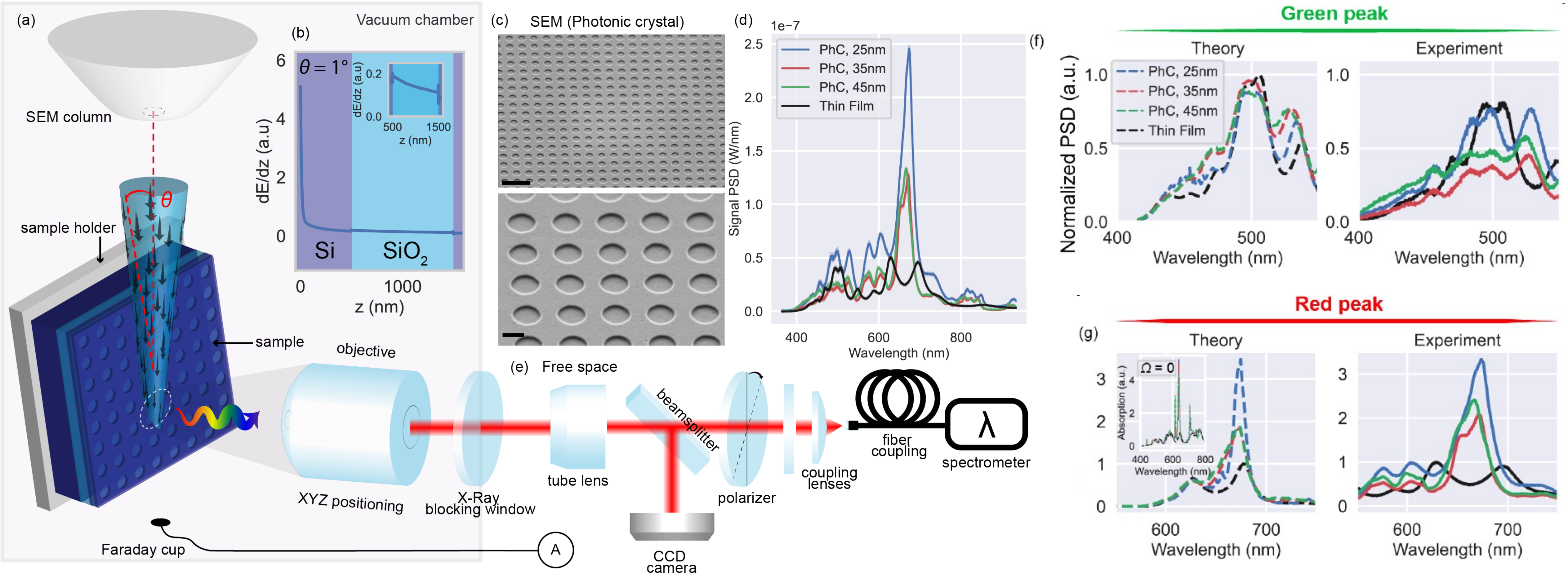


Girard et al., Reviews in Physics (2019)		**Optical transition (bandwidth) [eV]**
Self-trapped hole STH$_1$		2.61 (1.2) 1,88 (0.2–0.5)

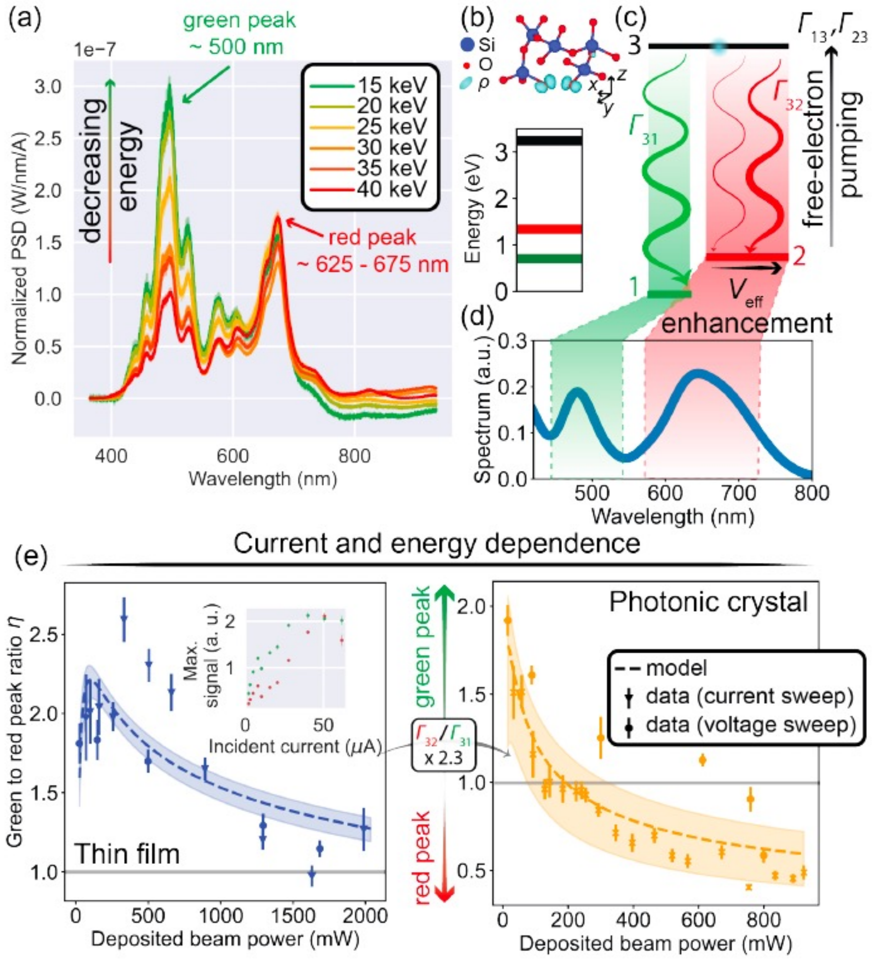
[DFT energy levels]



Accounting for scintillation spectra with the general framework



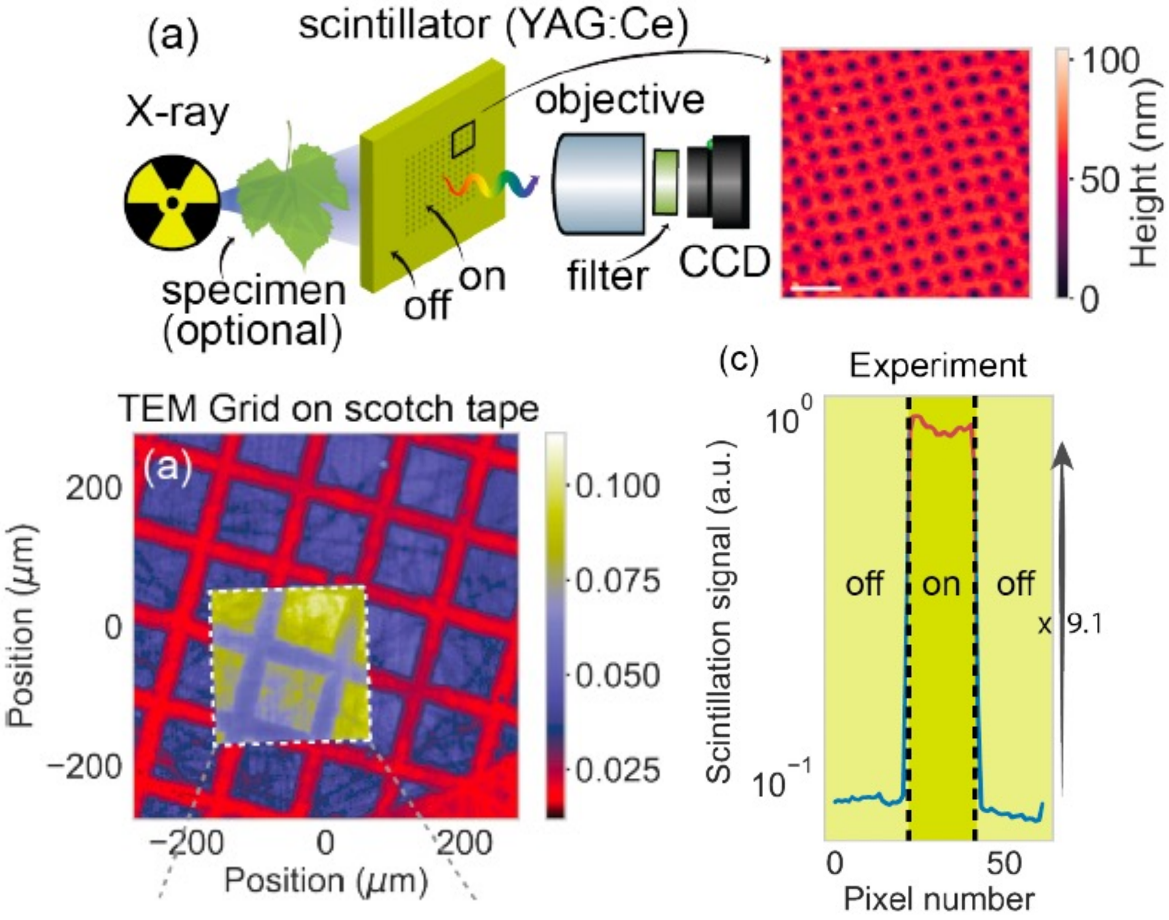
Out-of-equilibrium kinetics of defects controlled by nanophotonics



- Beyond predicting "nanophotonic" features the framework also, accounts for features requiring microscopics+nanophotonics
  - Nonlinear dependence of emission on pump + "cross-over" from green to red
    - 3-level dynamics of self-trapped holes

$$\begin{cases} \frac{dp_1}{dt} = -\Gamma_{13}\, p_1(1-p_3) + \Gamma_{31}\, p_3(1-p_1) \\ \frac{dp_2}{dt} = -\Gamma_{23}\, p_2(1-p_3) + \Gamma_{32}\, p_3(1-p_2) \\ \frac{dp_3}{dt} = \Gamma_{13}\, p_1(1-p_3) - \Gamma_{31}\, p_3(1-p_1) \\ \qquad +\Gamma_{23}\, p_2(1-p_3) - \Gamma_{32}\, p_3(1-p_2) \end{cases}$$

Nanophotonic-enhanced scintillation induced by X-ray bombardment

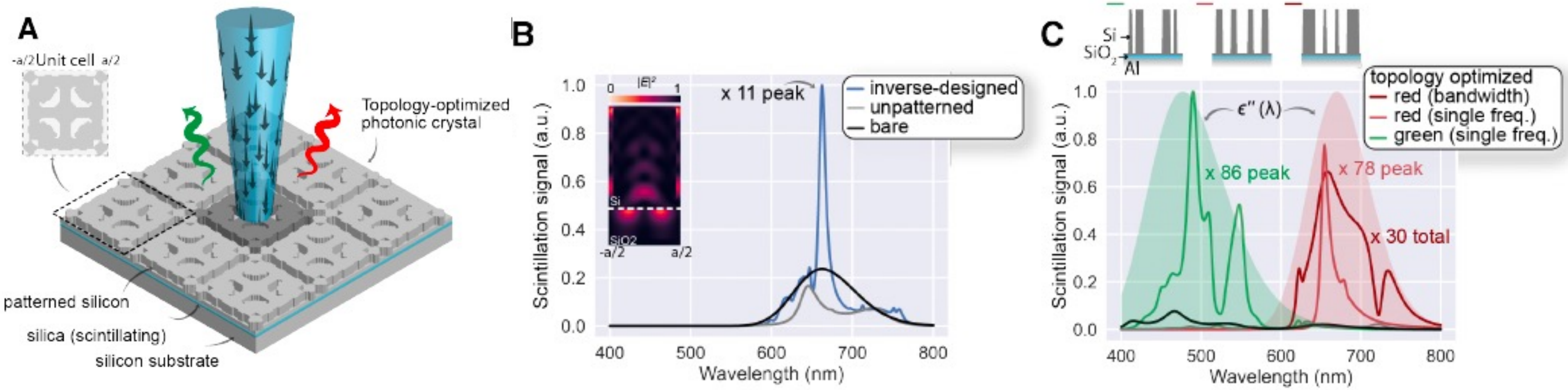


Roques-Carmes* & **Rivera*** *et al.* Science (2022).

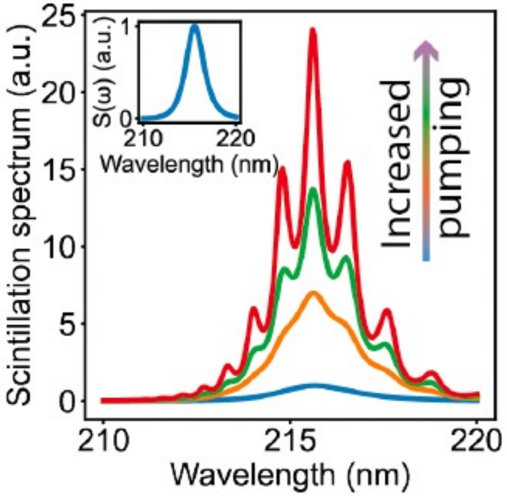
- Real-world example
  - Patterned photonic crystal into an X-ray scintillator
  - Put it into a micro-CT machine and look at light from patterned vs. unpatterned region
- **10x brighter image** from the patterned region

Outlook

Computational optimization of nanophotonic scintillators



Scintillation lasers



Watanabe *et al.*
Nat. Mater. (2004)