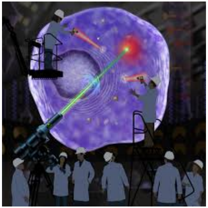
Nanoscopic thermometry with $\sim$ 30 mK precision: a quantitative study of cathodoluminescence in lanthanide-doped nanomaterials

Clarice D. Aiello

Inorganic Nanostructures Seminar @ Molecular Foundry, 03/16/17

Measuring temperature of tiny objects requires tiny sensor

- contact-based thermometry not applicable
- biology applications: cellular events marked by few-°C temperature changes
- industrial applications: temperature diagnostics for nano-electronics, materials...



http://scitechdaily.com/scientists-measure-and-control-the-temperature-inside-living-cells/

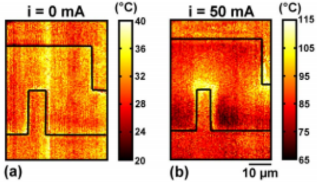


FIG. 4. (Color online) Reconstruction of the temperature map when (a) no current is passing through the structure and (b) a current of ~50 mA is circulating in the stripe. The contour of the stripe has been drawn in black for clarity. The image size is $45 \times 60\ \mu m^2$.

Aigouy et al., Appl. Phys. Lett. **87**, 184105 (2005)

Measuring temperature of tiny objects requires tiny sensor

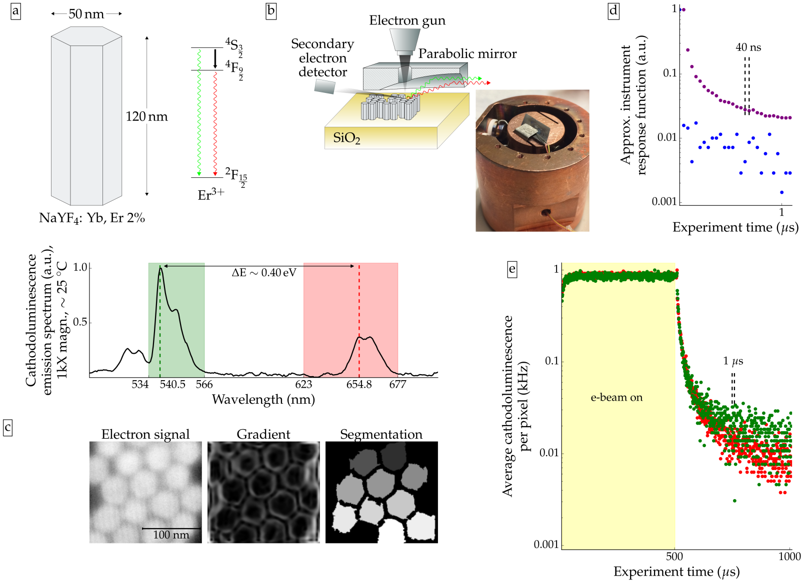
State-of-the-art:

- map how light from photostable nanomaterials changes as a function of temperature
- nanomaterials doped with lanthanide ions: sharp emission, long lifetimes, stability up to $\sim 1000\ ^\circ\mathrm{C}$ ✓
- optical excitation: works even for single nanoparticles... ✓ ...but diffraction-limited (we don't see the nanothermometer!) ✗

Idea:

- study cathodoluminescence of nanomaterials: nanoscopic localization (we see the nanothermometer!) ✓
- nanomaterial can form a 'film' or barrier to protect sample from radiation damage ✓

Overview of experimental conditions



Nanoscopic thermometry with $\sim$ 30 mK precision: a quantitative study of cathodoluminescence in lanthanide-doped nanomaterials

Intensity ratio thermometry

Lifetime thermometry

Nanoscopic thermometry with $\sim$ 30 mK precision: a quantitative study of cathodoluminescence in lanthanide-doped nanomaterials

Intensity ratio thermometry

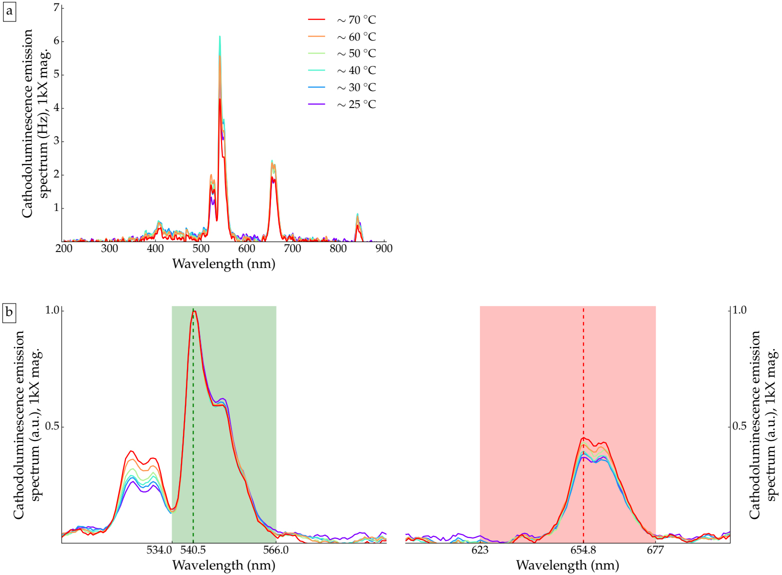
Lifetime thermometry

Can use a static signal to do thermometry

- study steady-state light while e-beam on

- signal is derived from ratio of intensity in spectral bands
- spectral peaks are linked thru phonon pathway:
  expect temperature dependence

Under cathodoluminescence, spectral behavior with temperature is akin to that under photoluminescence



Can use a static signal to do thermometry

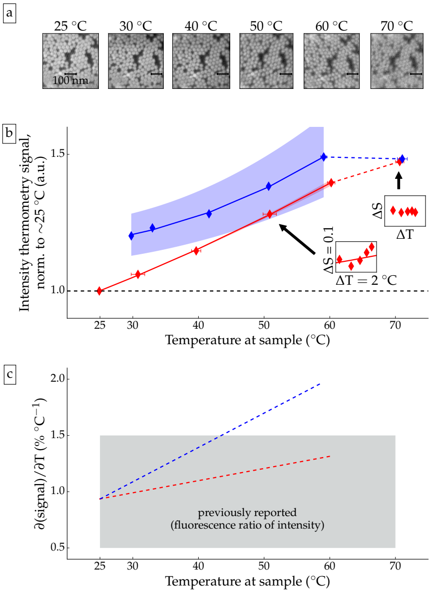
- study steady-state light while e-beam on

- signal is derived from ratio of intensity in spectral bands
- spectral peaks are linked thru phonon pathway:
  expect temperature dependence
- use empirical law

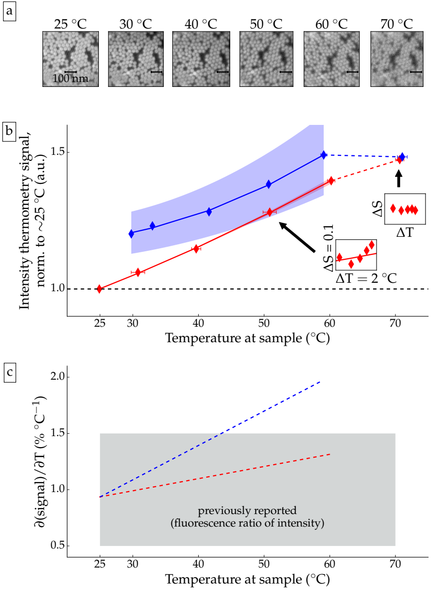
$$\mathrm{Signal}(t, T) = \frac{R(T)}{G(T)} \sim a \cdot T^2 + b \cdot T + c$$

- low signal-to-noise levels (all counts Poisson!),
  ratio is susceptible to fluctuations unrelated to temperature... ✗

Cathodoluminescence intensity ratio thermometry is on par with with optical...



Cathodoluminescence intensity ratio thermometry is on par with with optical... plus the nanometric localization



Drawbacks and advantages of these static signals to do thermometry

- measured signal dependence on pixel size (none),
  e-beam energy (?) and current ($\sim$ exponential)... ✓
- nanothermometer exposure to e-beam is prolonged ✗
- dependent on light collection fluctuations,
  local concentration of nanothermometers... ✗
- need to correlate observations of multiple spectral lines ✗

Nanoscopic thermometry with ~ 30 mK precision: a quantitative study of cathodoluminescence in lanthanide-doped nanomaterials

Intensity ratio thermometry

Lifetime thermometry

Can use a dynamic signal to do thermometry

- study transient light after fast blanking of e-beam
- extremely low signal-to-noise levels... ✗
  ...but noise averaged over by using
  transient light intensity as probability distribution ✓

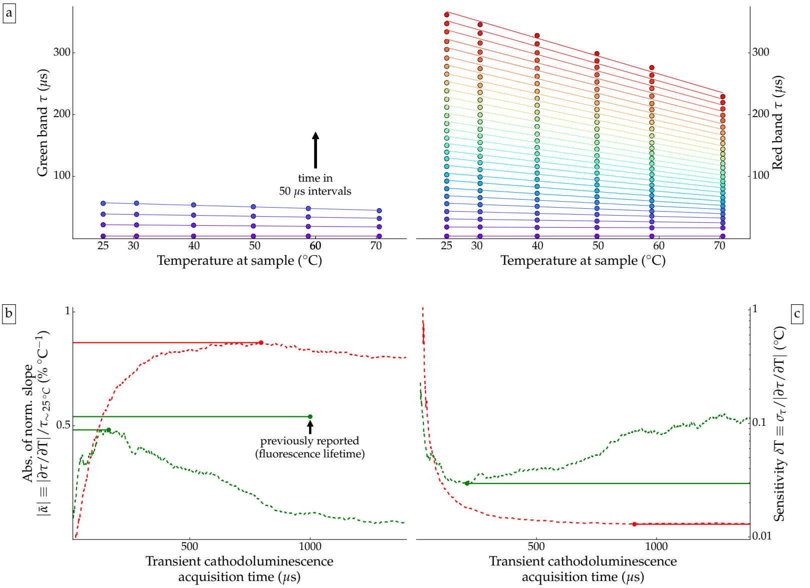
$$\text{Signal}(t, T) = \tau(t, T) \equiv \frac{\int_0^t I(t', T) t' dt'}{\int_0^t I(t', T) dt'}$$

- empirical model available

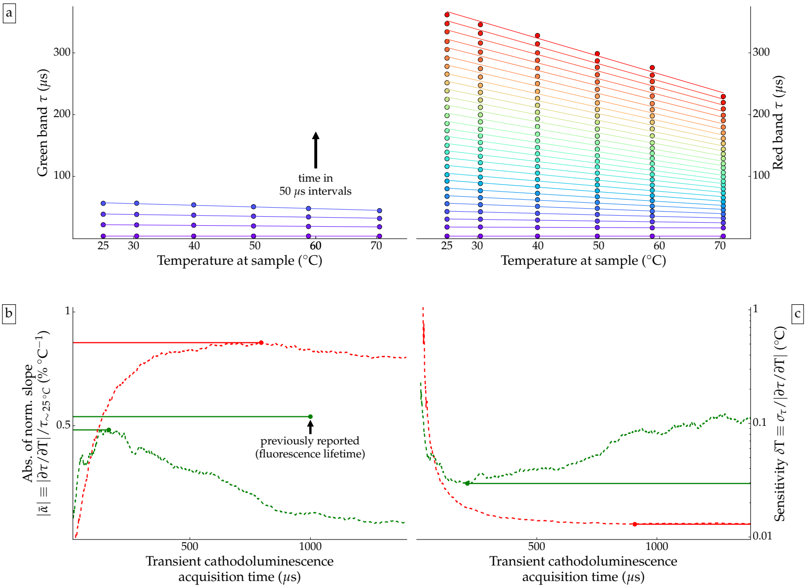
$$\tau(t, T) \sim \alpha(t) \cdot T + \beta(t)$$

- enables study of nanothermometer sensitivity
  as a function of acquisition time ✓

Cathodoluminescence lifetime thermometry is on par with with optical...



Cathodoluminescence lifetime thermometry
is on par with with optical... plus the nanometric localization



Drawbacks and advantages of this dynamic signal to do thermometry

- measured signal dependence on pixel size (none), e-beam energy ($\sim$ none) and current (strongly non-linear)... ✓
- nanothermometer exposure to e-beam is minimized ✓
- independent of light collection fluctuations, local concentration of nanothermometers... ✓
- only needs to measure one spectral line ✓

Nanoscopic thermometry with $\sim$ 30 mK precision: a quantitative study of cathodoluminescence in lanthanide-doped nanomaterials

Clarice D. Aiello

Group meeting, 04/17/17