Conservation of photon rate in endothermic photoluminescence and its transition to thermal emission: supplementary material

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The Fluorescence Intensity Ratio Thermometry (FIR) method:

Using the FIR method, temperature can be measured by recording the intensity ratio of two adjacent optical emission peaks of a luminescent sample. Under the condition of full thermalization of electrons to the band edge, one can apply the Boltzmann population statistics, to the peaks ratio:

$$R = C \cdot e^{\left(-\frac{\Delta E}{k_b T}\right)} \quad (\text{S1})$$

where ΔE is the energy difference between the two emission peaks. The constant C is determined by the density of states and the matrix element for optical transitions between the states. Once C is known experimentally, the relation can be used to determine the sample's temperature during the experiment. In order to find C , we place the sample in a calibrated micro-furnace, which

Fig. S1. (a) The FIR measurement experimental setup (b) and the intensity ratio vs. temperature plot.

consists of a heated Quartz cylinder. From the same Neodymium (Nd^{+3}) doped Silica fiber as in the experiment, we cut a 5mm long fiber and connect (fuse) it, from both sides, to a long passive optical fiber in order to guide its emission (Figure S1a). The 5mm active fiber section is placed at the center of the furnace, while the fiber is pumped by 532nm laser from one side, and light is collected from its other side. A spectrum analyzer is used to measure the ratio between the 900nm and 820nm peaks as function of the furnace temperature. Figure S1b depicts the experimental intensity-ratio to temperature plot obtained from the measurement, and the fitted curve. The constant is found to be $C=2.6$. The inset shows the evolution of the peak ratio shift with temperature increase.

Calculation of the chemical potential from the experimental data:

The chemical potential can be calculated from equation (S1):

$$\mu(\hbar\omega, T) = K_b T \ln\left(\frac{R(\hbar\omega, T)}{\varepsilon(\hbar\omega) R_0(\hbar\omega, T)}\right) \quad (\text{S2})$$

Since μ is constant at the relevant emission band between 600nm-1000nm, it can be extracted by integrating the experimental emitted photon rate in the band under PL and thermal excitation presented in figure 2c (blue and red lines):

$$\mu(\hbar\omega, T) = K_b T \ln\left(\frac{\int_{\text{band}} R_{pl}(\hbar\omega, T) d\hbar\omega}{\int_{\text{band}} R_{thermal}(\hbar\omega, T) d\hbar\omega}\right) \quad (\text{S3})$$

For temperatures below 1150K, where the thermal emission signal is below our detection limit, the experimental data for the thermal emission was extrapolated to 300K as follows. The sample's relative emissivity was calculated as the ratio between the thermal emission in the experiment and the theoretical black body emission rate at the same temperatures:

$$\varepsilon(\hbar\omega) = \frac{R_{thermal}(\hbar\omega, T)}{R_0(\hbar\omega, T)} \quad (\text{S4})$$

The result is shown in figure S2.

Fig. S2. Measured relative emissivity of Nd:SiO2 sample.

The thermal photon rate for the temperature range 300K-1150K is then calculated by:

$$R_{thermal_extrapolated}(\hbar\omega, T) = \int_{\text{band}} \varepsilon(\hbar\omega) \cdot R_0(\hbar\omega, T) d\hbar\omega \quad (\text{S5})$$