Investigating Apparent Self-Heating of Individual Luminescent Nanoparticle Thermometers

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We have demonstrated a far-field optical thermometry technique based on the temperature-dependent luminescence of individual upconverting $NaYF_4$ nanoparticles doped with Er^{3+} and Yb^{3+} [1]. The technique utilizes the temperature-dependent ratio of luminescence from two thermally coupled levels in Er^{3+}. While lanthanide-doped upconverting nanoparticles are commonly used for thermometry, most measurements employ large ensembles of particles. Single particle measurements are much less common, and prior to this work the smallest isolated particle used for thermometry was several hundred nanometers in diameter. Although single particle measurements require excitation intensities many orders of magnitude higher than those used for ensemble imaging, self-heating effects are typically assumed to be negligible. However, when we measure the temperature-dependent ratio as a function of excitation intensity, we observe a linear increase that indicates a temperature rise of more than 60 K if interpreted as thermal.

To further investigate this unexpected apparent temperature rise, we employ a second thermometry method based on the temperature-dependent luminescence lifetime of the nanoparticles, and for the first time demonstrate this technique using individual upconverting nanoparticles. The apparent temperature rise as a function of excitation intensity measured using the luminescence lifetime is consistent with the results from the ratiometric approach. We also modulate the excitation laser and observe an apparent temperature rise as a function of modulation frequency that is consistent with a thermal interpretation. Though consistent, these results all contradict simple yet conservative thermal models that indicate the temperature rise should be less than 5 K. Consequently, we turn to more direct manipulation of the thermal environment: we place the nanoparticles on various substrates with thermal conductivities spanning three orders of magnitude, apply several conformal coatings to the particles, and alter the medium surrounding the particle. In all cases, there is no significant change in the apparent temperature rise, ultimately suggesting that the effects we observe are in fact not thermal in nature. These effects are calibratable, and thus do not limit the applicability of the thermometry technique. Nonetheless, our results highlight the need to carefully monitor the power output of the excitation laser, since even small fluctuations in excitation intensity can cause apparent changes in the measured temperature.

[1] J.D. Kilbane, E.M. Chan, C. Monachon, N.J. Borys, E.S. Levy, A.D. Pickel, J.J. Urban, P.J. Schuck, and C. Dames, *Nanoscale* **8**, 11611 (2016).