Resonant Multiple-Phonon Absorption Causes Efficient Anti-Stokes Photoluminescence in $CsPbBr_3$ Nanocrystals

Zhuoming Zhang[1†], Sushrut Ghonge[2†], Yang Ding[1], Shubin Zhang[2], Mona Berciu[3,4*], Richard D. Schaller[5], Boldizsár Jankó[2*], Masaru Kuno[1,2*]

[1]*Department of Chemistry and Biochemistry, University of Notre Dame, 251 Nieuwland Science Hall, Notre Dame, IN 46556, United States*

2. Department of Physics and Astronomy, University of Notre Dame, 225 Nieuwland Science Hall, Notre Dame, IN 46556, United States

3. Department of Physics and Astronomy, University of British Columbia, Vancouver Campus 325-6224, Agricultural Road, Vancouver BC V6T 1Z1, Canada

4. Stewart Blusson Quantum Matter Institute, University of British Columbia, Vancouver, British Columbia, V6T 1Z4 Canada

5. Department of Chemistry, Northwestern University, Evanston, IL 60208 and Center for Nanoscale Materials, Argonne National Laboratory, Lemont, IL 60439, United States

† contributed equally

berciu@phas.ubc.ca, bjanko@nd.edu and mkuno@nd.edu

.

Lead-halide perovskite nanocrystals such as CsPbBr3, exhibit efficient photoluminescence (PL) up-conversion, also referred to as anti-Stokes photoluminescence (ASPL). This is a phenomenon where irradiating nanocrystals up to 100 meV below gap results in higher energy band edge emission. Most surprising is that ASPL efficiencies approach unity and involve single photon interactions with multiple phonons. This is unexpected given the statistically disfavored nature of multiple-phonon absorption. Here, we report [1] and rationalize near-unity anti-Stokes photoluminescence efficiencies in CsPbBr3 nanocrystals and attribute it to resonant multiple-phonon absorption by polarons. The theory explains paradoxically large efficiencies for intrinsically disfavored, multiple-phonon-assisted ASPL in nanocrystals. Moreover, the developed microscopic mechanism has immediate and important implications for applications of ASPL towards condensed phase optical refrigeration.

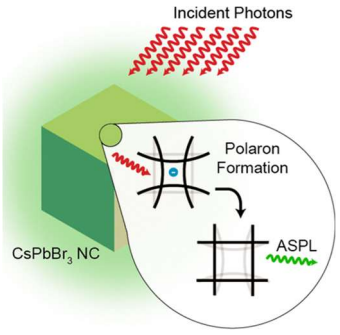

Fig.1. Schematic representation of phonon-assisted upconversion in $CsPbBr_3$.

References

[1] Z. Zhang *et al,* ACS Nano **18**, 6438 (2024). DOI: 10.1021/acsnano.3c11908.