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נושא:

Near Unity Quantum Emitters: the role of thiocyanate on the surfaces of lead halide perovskite nanocrystals.

This research was performed under the supervision of
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ההרצאה תתקיים בחדר סמינרים הפקולטי

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Near Unity Quantum Emitters: the role of thiocyanate on the surfaces of lead halide perovskite nanocrystals.

Lead halide perovskite nanocrystals (NCs) are emerging materials for light emitting diodes, lasers, and solar cells due to their high photoluminescence quantum yields (PLQY). Treatment with thiocyanate (-SCN) has been known to improve the the power conversion efficiency of solar devices and their nanocrystalline counterparts treated by thiocyanate reach near-unity photoluminescent quantum yield.[1,2,3] However, the mechanism by which the SCN improves the PLQY of nanocrystals is still actively debated and microscopic details of the interaction between the SCN and NCs surface is not entirely understood. Practical difficulties arise for the treatment procedures themselves when introducing the -SCN salts into a colloidal solution of NC's in their native nonpolar (alkane) solvent, where the -SCN is insoluble. We use $CsPbBr_3$ nanocrystals as a model system to investigate the mechanisms hypothesized earlier for the PLQY enhancement.[4,5] In order to introduce -SCN more effectively, we used the treatment procedure based on the ionic liquid involving urea and ammonium-SCN (UAT).[6] Using multitude of structural and spectroscopic characterization techniques (XRD, TEM, EDS, EELS, PL, FTIR, 2DIR, and others), we found that while the crystal structure of the NCs is not affected by the -SCN treatment, the -SCN ions are surface-bound. We confirm thiocyanate is present on the surface of the nanocrystals through electron energy loss spectroscopy mapping. The integrity of the unchanged perovskite crystal structure confirms the mechanism of pseudohalide surface treatment over A site cationic doping. A holistic characterization of the colloidal suspension through ultrafast two-dimensional vibrational spectroscopy agrees with a surface bound species. The resulting modified CsPbBr3 surfaces are more stable to ionic transformation as demonstrated by anion exchange studies. These findings allow us to suggest that the mechanism responsible for the enhancement of PLQY involves passivation of shallow surface traps created by the bromine vacancies and that SCN passivation is possible because it binds to NCs in a less labile fashion than the native halides.

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