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Title:	EXPLORING THE PHOTOEXCITED CHARGE CARRIER DYNAMICS OF PEROVSKITE NANOSTRUCTURES THROUGH ULTRAFAST TRANSIENT ABSORPTION AND TERAHERTZ SPECTROSCOPY
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**Abstract:** "Perovskites: Bridging the gap between efficiency and affordability in solar energy" In the field of photovoltaics, inorganic metal halide perovskite solar cells have attracted much attention because of their high power conversion efficiencies (PCE) and affordable manufacturing. The great potential of perovskite solar cell technology has attracted the attention of researchers and industry experts due to this extraordinary advancement. In 2009, these novel materials debuted, displaying a PCE of 3.8%. Since then, there has been no turning back, and a staggering rise in these numbers has been seen, with current developments raising the efficiency of organic-inorganic perovskite-based tandem cells to 31.25%. Rapid charge carrier recombination, which causes energy loss, significantly hinders their overall efficiency. To get around this obstacle, scientists are working tirelessly to find ways to control carrier cooling, making it possible to collect and use the surplus energy currently lost as heat. Since the inherent relaxation mechanisms pursued by the heated carriers have a significant influence on their behavior. Therefore, gaining a comprehensive understanding of carrier cooling dynamics becomes imperative for advancing these fields. However, the thermalization and cooling of carriers occur in a femtosecond time scale, which can be understood using transient absorption spectroscopy. My thesis work primarily focuses on unraveling perovskite materials' photophysical properties by employing two key techniques: transient absorption (TA) spectroscopy and terahertz (THz) spectroscopy. By delving deep into the dynamics of carriers within perovskite materials, we aimed to provide valuable insights that can contribute to the development of more efficient and reliable devices. □ First work highlights that the shape of perovskite crystals can significantly impact carrier cooling in perovskite solar cells. The investigation reveals a captivating disparity in the hot carrier cooling rate between 2D Nanosheets (NSs) and 3D Nanocrystals (NCs), the carrier cooling is found to be faster in 2D NSs. However, this goes against the anticipated outcome since the cooling of the carrier was expected to be slow due to the decrease in the density of states in 2D NSs. The faster cooling rate observed in the 2D system can be attributed to the reduction in Fröhlich interaction, facilitated by the system's higher surface volume ratio, which leads to easier heat dissipation to the surroundings. □ In the next work, we tuned the facets and formed a newly synthesized dodecahedron cesium lead bromide (dodecahedron-CsPbBr<sub>3</sub>) perovskite and compared its properties with conventional cube CsPbBr<sub>3</sub> perovskite. Through transient absorption (TA) studies, we have observed that dodecahedron-CsPbBr<sub>3</sub> exhibits prolonged carrier cooling and a remarkable 20- 30% enhancement in biexciton yield compared to cube-CsPbBr<sub>3</sub>. Further, temperature- dependent TA studies revealed the rapid formation of polarons within 0.25 ps in dodecahedron- CsPbBr<sub>3</sub>, leading to an increase in polaron size compared to cube-CsPbBr<sub>3</sub>. Additionally, terahertz spectroscopic measurements have shown that the carrier mobility in dodecahedron- CsPbBr<sub>3</sub> at early time scales (few ps) is approximately 2 times lower than in cube-CsPbBr<sub>3</sub>, further supporting the faster polaron formation in dodecahedron-CsPbBr<sub>3</sub>. The slower cooling in dodecahedron-CsPbBr<sub>3</sub> places it as a better contender than conventional cube CsPbBr<sub>3</sub> in field of photovoltaics. □ In the third work, we investigated cesium lead chloride (CsPbCl<sub>3</sub>) which stands out for its remarkable blue emission. Also, the research carried out at low temperatures offers a more profound insight into the complex photophysical processes that are typically challenging to investigate at standard temperatures. This challenge arises from the increased system entropy caused by the phonon disorder. The underlying photophysics of CsPbCl<sub>3</sub> NCs is investigated using ultrafast TA and THz spectroscopy, focusing on temperature and excitation energy dependence. A direct dependence of carrier relaxation, carrier temperature, and energy loss on phase transition is observed. By analyzing the optical pump THz probe (OPTP) spectroscopy data, we have observed that decreasing the excitation energy leads to a slower decay in mobility, attributed to reduced Auger recombination and increased penetration depth. □ As the absorption and PL of CsPbCl<sub>3</sub> are restricted in the UV region, the PL could be extended up to the VIS region via doping. In the fourth work, we further doped rare earth ions (RE<sup>3+</sup>) i.e., Eu<sup>3+</sup> (Europium ion) in the lattice of CsPbCl<sub>3</sub> as it has revolutionized the field of optoelectronics, offering exciting possibilities to explore and manipulate optical, magnetic, and charge carrier transport properties. Herein, we have investigated the intricate mechanistic transfer processes from the band edge of the host material, CsPbCl<sub>3</sub>, to the dopant (Eu<sup>3+</sup>). We observe an intriguing defect-mediated cascading energy transfer phenomenon from CsPbCl<sub>3</sub> to Eu<sup>3+</sup>, accompanied by intriguing cross-relaxation interactions among different states of Eu<sup>3+</sup>, revealing two distinct characteristic traits, one of which is magnetic dipole transition and another one is electric dipole transition. Additionally, we observed two-photon absorption in our study, and intriguingly, we found no compromise in the efficiency of energy and charge transfer processes. □ In the final work, we prepared Eu-doped CsPbBr<sub>3</sub> in order to see how doping affects a different halide family member, i.e., CsPbBr<sub>3</sub>. In this study, we have investigated the spectroscopic properties of CsPbBr<sub>3</sub> and Eu-CsPbBr<sub>3</sub>. Our findings reveal that the doped counterpart exhibits significantly slower cooling which was opposite to what was observed in the case of CsPbCl<sub>3</sub>. At the same time, the formation and decay time of polaron remains unchanged. Low- temperature transient studies indicate that this is attributed to the absence of bound excitons resulting from the passivation of Br<sup>-</sup> vacancies. Additionally, OPTP studies demonstrate that this phenomenon also impacts carrier mobility and photoconductivity, leading to higher photoconductivity and slower mobility in Eu-CsPbBr<sub>3</sub>. The implications derived from our research indisputably demonstrate that the slower hot carrier cooling and enhanced photoconductivity in Eu-CsPbBr<sub>3</sub> position it as a superior candidate for photovoltaic applications.

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