

Peculiarities of dipolar ordering in mixed cation halide perovskites

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The methylammonium (MA) lead halides MAPbX₃ (where X = I, Br, Cl) are popular perovskite materials among scientists and industry due to their perspectives in effective and cheap solar cells [1]. During the past decade, the power conversion efficiency of cells based on these hybrid compounds exceeded more than 20%. A high performance of these materials results from several physical properties such as large absorption coefficient, optimal bandgap, long carrier diffusion length, low exciton binding energy, exceptional defect tolerance. However, a successful application of hybrid perovskite solar cells is mainly prevented by their lead toxicity and poor both thermal and water stability.

The most stable and efficient solar cells are obtained by using perovskites with mixed cations at the A-site. The most popular alternatives to MA are formamidinium and Cs⁺ ions. A Dimethylammonium (DMA) cation has been introduced recently as an alternative A-site modification for these compounds. Several investigations have shown that during certain synthesis procedures high quantities of DMA may be unintentionally introduced into MAPbI₃ and CsPbI₃. These modifications stabilize the preferable cubic phase of MAPbI₃ and leads to the enhanced performance at ambient condition.

In the field of classical inorganic perovskites it is well known that mixing may significantly perturb structure of resulting compound. Thus, the long-range order can be suppressed and frustrated phases may appear. The dielectric permittivity behavior of lead halides seem to be especially informative for the performance of the perovskite cells, as their relatively high value of the dielectric permittivity results in a pronounced defect tolerance and low exciton binding energy. However, a complete understanding of mixing effects on the dielectric permittivity dynamics and structural phase behavior is still absent. Here, we present a multitechnique experimental study of the mixed hybrid perovskite MA_{1-x}DMA_xPbBr₃. Our results show that structural phase transitions are significantly suppressed even for a low substitution of the DMA cations. For higher DMA levels, the long-range dipolar order disappears and dipolar glass dielectric behavior dominates in the dielectric spectra.