有機金属ハライドペロブスカイト太陽電池における超格子と相共存の電子顕微鏡観察

Direct observation of self-organized superlattices and phase coexistence in organometal halide perovskite solar cell with transmission electron microscopy

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Recently, organometal halide perovskite solar cells (PSCs) have received great attention. The power conversion efficiency (PCE) of PSCs have shown a dramatic increase and certified PCEs in the case of mixed organic cations and halide anions have reached up to 22.7%. The remarkable achievements originate from their specific properties, such as long charge carrier diffusion length, large absorption coefficient and low recombination rate etc. Unfortunately, although photovoltaic characteristics of crystals such as the organometal halide perovskite are strongly concerned with their crystal structure and quality, microstructural analyses of the organometal halide perovskite have not been actively reported. Several analyses such as X-ray diffraction (XRD) have reported important features of the organometal halide perovskite like phase transition or overlap at transition temperature, however, because they cannot offer direct evidence about the configuration of crystal inside, precise and in-depth understanding in microstructural configuration of the organometal halide perovskite through transmission electron microscope observation.

For the crystallographic information of the organometal halide perovskite, planar type thin film perovskite solar cells (Au/Spiro-MeOTAD/MAPbI₃/TiO₂/FTO/Glass) were fabricated through spin-coating method with antisolvent.

Different from previous reports indicating each phase of the organometal halide perovskite solely exists (orthorhombic phase < 165K < tetragonal phase < 327K < cubic phase), it is observed that the tetragonal and cubic phases coexist at room temperature, and identified that superlattices composed of mixture of tetragonal and cubic phases are self-organized without a compositional change. Amazingly, the organometal halide perovskite self-adjusts configuration of phases and automatically assembles buffer layer between boundaries by introducing superlattice. We believe that the newly observed extraordinary structural characteristics of the organometal halide perovskite could be the origin of its unsolved problems and will shed new light on their new possibilities as promising materials for various applications.

This research has been supported by New Energy and Industrial Technology Development Organization (NEDO).