学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　　博士（情報科学）　　氏名　Ahmed Esmail Kamal Shalan

学 位 論 文 題 名

Optimization of nanoarchitectures for high-performance planar perovskite solar cells
（高性能ペロブスカイト太陽電池に向けたナノ構造の最適化）

Perovskite solar cells (PSCs) based on organometal halide light-absorbing materials have attracted enormous research interest for solar cell applications due to their many intriguing optical and electronic features, such as large absorption coefficient, large charge-carrier mobility, long carrier-recombination lifetime and unique defect physics. Since the first report on a solid-state PSC with a (power conversion efficiency) PCE of 9.7% in 2012, and a PCE of 22.1% was certified in 2016. To improve the net PCE value more, it is important to elucidate the essential factors influencing on the PCE. This thesis focused on several element technologies to enhance PCE and the external quantum efficiency (EQE) from the viewpoints of 1) the light-harvesting, 2) the hole extraction material, and 3) the hole blocking layer for the efficient charge separation. These studies were performed in order to give a better understanding and insight into the nanoarchitectures optimization for high-performance PSCs.

Although a lot of studies have been done related to PSCs, the EQE values are almost saturated especially in the longer wavelengths. Therefore, it is necessary to find a new way to improve the EQE at longer wavelengths for PSCs. Plasmonics is a highly promising due to its light-harvesting property. In this section, inverted planar PSCs with a p-i-n device configuration of indium-doped tin oxide (ITO)/NiO/$CH_3NH_3PbI_3$/[6,6]-phenyl-C_{61}-butanoic acid methyl ester (PCBM)/Ag with and without gold nanoislands (Au NIs) were prepared. The results demonstrated that the Au NIs increased the PCE to 5.1%, almost twice that of the samples without Au NIs. This result is due to the excitation of surface plasmons. Furthermore, we observed an enhancement of EQE at wavelengths shorter than the plasmon resonance. It is speculated that the plasmoelectric potential effect may contribute to the enhancement of EQE at the off-resonance region (Chapter 2).

A hole-extracting layer (HEL) is a key material which affects device performances of PSCs. CoO_x is a promising HEL for inverted planar PSCs with a p-i-n device configuration of ITO/CoO_x/$CH_3NH_3PbI_3$/PCBM/Ag. The devices fabricated according to a simple solution procedure showed the best photovoltaic PCE 14.5%, which is significantly superior to those fabricated with traditional HEL such as PEDOT:PSS (12.2%), NiO_x (10.2%) and CuO_x (9.4%), under the same experimental conditions. Photoluminescence (PL) spectra and the corresponding PL lifetime of perovskite deposited on varied HEL films were measured to obtain the hole-extracting characteristics, for which the hole-extracting times consistent with the trend of their PCE. The reproducibility and endurance of those devices were examined to show the outstanding long-term stability of the devices made of metal oxide HEL, for which the CoO_x device still had PCE of almost 12% for over 1000 h (Chapter 3).

A thickness of hole blocking materials also strongly affects the photovoltaic performances of PSCs.

The effect of the film thickness of the TiO_2 compact layer deposited by atomic layer deposition (ALD) techniques for mesoporous PSCs with an n-i-p configuration of fluorine-doped tin oxide (FTO)/ ALD coated TiO_2 (ALD-TiO_2)/mesoporous-TiO_2/$CH_3NH_3PbI_3$/2, 2', 7, 7' - Tetrakis [N, N - di (4-methoxyphenyl) amino]-9,9'-spirobifluorene (spiro-OMeTAD)/Ag, was investigated. Uniform and pinhole-free TiO_2 films with various thicknesses from 10-400 nm were deposited on a FTO substrate using the ALD technique. The device performance of PSCs showed a systematic trend on the thickness of the ALD TiO_2 compact layer and attained the best PCE 15.0% at the thickness of 200 nm. PL spectra and the corresponding PL lifetime for perovskite deposited on different ALD films were measured. It was elucidated that the effective PL quenching is due to the electron transfer from perovskite into the TiO_2 compact layer. The well-controlled TiO_2 compact hole blocking layer devices showed great stability and reproducibility, providing an alternative for high-performance PSCs (Chapter 4).

In summary, the element technologies of the device architecture of PSCs affecting the PSC performance have been investigated. The incorporation of Au NIs to the inverted planar PSCs improves EQE by enhancing the light-harvesting properties of PSCs. Furthermore, PSCs fabricated with CoO_x as a promising hole extraction material exhibited high PCE due to its great hole-extracting ability. Moreover, it was clarified that ALD-TiO_2 works as a compact hole blocking layer being useful for the efficient charge separation.