論文の内容の要旨

論文題目　Role of homogeneous luminescent coupling effect in
III-V based multijunction solar cells
（III-V族多接合太陽電池におけるルミネセンス結合と均
質化の効果）

氏　　名　ユヘコ　ベーニス　メー　フェタルベロ

Among the next-generation solar cells, the multijunction solar cell (MJSC) has demonstrated the highest solar energy to electric power conversion efficiency. The record MJSC power conversion efficiency attained so far was 47.1%, which was explicitly achieved by increasing the number of junctions under concentrated sunlight. Further increase can lower the production cost and ease market penetration. Particularly, increasing the conversion efficiency beyond 50% is the future target for photovoltaics community to achieve grid parity. To reach this target, more refinement needs to be done to minimize losses in solar cells such as carrier thermalization and electron-hole pair recombination resulting in light emission. Based on the latter, a possible strategy to improve the solar cell efficiency is by finding ways how to maximize the recycling of internal luminescence. By reabsorption or "recycling" of light emission due to electron-hole pair recombination in solar cells, the efficiency may increase by about 1.0%, which is considerably challenging to achieve recently.

The absorption of photons emitted by a higher bandgap subcell in an adjacent lower bandgap subcell of an MJSC is called the luminescence coupling (LC) effect. The LC effect is an internal phenomenon caused by photon emission due to radiative recombination from a higher bandgap subcell of an MJSC which then results to photocurrent generation in an adjacent lower bandgap subcell. This phenomenon allows carrier redistributions among subcells, thereby reducing current mismatch and wavelength sensitivity that helps boost the conversion efficiency of the MJSC. However, preliminary measurements showed that the LC current distribution is non-uniform or spatially mismatched. This causes an inherent reduction in energy conversion efficiency, implicitly suggesting that if the non-uniform LC current distribution is made uniform or spatially current-matched, the MJSC conversion efficiency may be

increased. Aside from this observation, no definitive discussion on the underlying physics of inhomogeneous LC effect can be found in the literature at the time of writing. With this, the need to find out its root cause arises, in the hope of materializing a possible absolute increase of 1.4% in MJSC power conversion efficiency when strong LC effect is made uniform. Furthermore, to the best of the author's knowledge, no study has attempted yet to find solutions to inhomogeneous LC effect techniques, making this discourse novel. Hence, possible techniques to spatially homogenize the LC effect are yet to be investigated.

In addressing the research problem stated above, the role and proof-of-concept in homogenizing LC effect to the power conversion efficiency of III-V MJSCs may be discovered. Specifically, this work aims to investigate novel MJSC designs that may generate spatially-matched or uniform LC current in a current-limiting MJ subcell that could lead to 1.0% higher MJSC conversion efficiencies. This work will be most interesting for people who work on concentrator MJSC systems as this showcases how to possibly improve the MJSC conversion efficiency through addressing the non-uniformity of the LC effect. As far as the author's knowledge, this has not been considered yet with currently existing III-V MJSC structures, therefore making this study original. Upon considering the non-uniform LC effect in novel MJSC structures, energy cost reduction may be achieved, which could mean better acceptance of solar energy technology implementation from electric power consumers. Experimenting on these designs may also help determine the physics behind non-uniform LC effect.

The role of homogeneous luminescent coupling (LC) effect in III-V multijunction solar cells (MJSCs) was probed. It was found that homogenizing the LC effect may increase the conversion efficiency of III-V MJSCs. This was experimentally demonstrated by electrical passivation, which garnered an efficiency increase of 1.05% to 1.18% with respect to a reference cell. This is close to the 1.35% efficiency increase predicted from modeling an MJSC with homogeneous LC current collection. The possibility was also demonstrated by photo-passivation. In this part, current redistribution onto lower bandgap subcells and more homogenized current collection on the benefitting subcells were observed. Theoretically, a homogeneous LC effect may be achieved by designing a plateau-stack of III-V MJSCs, in which the topmost subcell has the smallest lateral area. The larger the proportion between adjacent subcells, the lenient the choices for the material's energy gap will be, based on the detailed balance calculations made.

It was found that the laser beam induced current (LBIC) mapping is a useful technique in observing spatial LC current collection in III-V MJSCs. To get a perspective on the emission profile of adjacent subcells where the LC effect occurs, one may implement the electroluminescence/photoluminescence (EL/PL) mapping. It was found that the emission profile of the higher bandgap subcell where the LC effect originated was the same as that of the LC current collection in its adjacent lower bandgap subcell. Therefore, exploring these techniques while inducing the LC effect has led us to confirm that the LC current collection in a lower bandgap subcell will depend on the emission profile of its adjacent higher bandgap subcell.

The nonuniform LC effect in III-V MJSCs was observed among the adjacent subcells of Ge-based and Si-based samples characterized. The nonuniformity was found to get severe as the intensity of external irradiation upon a higher bandgap subcell was increased. Upon comparing the LC current collection in terrestrial- and space-grade samples, it was inferred that the higher bandgap subcell quality and design will affect the LC effect towards its adjacent lower bandgap subcell. By quality, it pertains to the number of defects acting as carrier traps or recombination centers present. On the other hand, design refers to which part of the solar spectrum the MJSCs can absorb more easily. Meanwhile, no change in LC effect homogeneity was observed for any cell temperature. Hence, it prevails as a problem in any cell temperature. In a broader perspective, this is an issue for concentrator systems, as terrestrial MJSCs typically operate at 85°C and beyond under high sunlight concentration [137–138]. As for various cell sizes, while the degree of LC nonuniformity did not vary at every higher bandgap subcell irradiation tested, the larger the cell area, the larger the losses observed due to the inhomogeneity of the LC effect.

To investigate the physics behind nonuniform LC effect, 3-dimensional distributed simulation program with integrated circuit emphasis (SPICE) electrical model, 2-dimensional Monte Carlo ray tracer, and a quasi-2-dimensional electro-optical prediction (Q2DEP) model developed in the course of this study were used. Before the implementation of these models, it was hypothesized that the nonuniform LC effect is possibly caused by the following physics: (1) lateral resistance effect resulting to nonuniform radiative emission within the MJ subcells at high carrier injection, (2) nonuniform material growth, (3) perimeter recombination, and (4) photon escape through the MJSC sidewalls. Although the nonuniformity also depends on the III-V or IV material defect distribution itself, the lateral resistance effects and perimeter recombination were identified to be the dominant causes of the inhomogeneous LC effect. On the other hand, low photon escape probabilities obtained from optical simulations of III-V and IV solar cell materials suggested that photon escape is not a prevalent cause of inhomogeneous LC effect.

Experimentally, it was found that perimeter recombination is mainly responsible for the inhomogeneity of the LC effect since this was observed to be alleviated by electrical passivation of the sidewalls of InGaP/GaAs/Ge triple-junction solar cells. The sidewall passivation was made possible by atomic layer deposition of thin Al_2O_3 film on full MJSCs with contacts and anti-reflection coating. Results revealed that this method increased the LC current collection in a limiting MJ subcell by 21.9% and enhance its uniformity by 7.2%. Furthermore, the luminescence homogeneity from the limiting MJ subcell was observed to improve by 37.5% after Al_2O_3 passivation at 2.5 V, when the MJSC was operated near its open-circuit voltage. Last, under standard global air mass 1.5 (AM 1.5G) condition at 1 sun concentration, the power conversion efficiency of the electrically-passivated III-V 3JSC increased by 1.05% to 1.18%; hence, close to the predicted value [170] of 1.35% and demonstrating a non-invasive way to improve current matching among MJ subcells.

Aside from electrical sidewall passivation, MJSC dimension modification was also probed as a possible solution to the nonuniform LC effect. This was investigated theoretically by the detailed balance calculation of a tandem solar cell, in which the top cell was designed to have a larger bandgap and a smaller active area than those of the bottom cell. Also, this was simulated using a 2-dimensional SPICE electrical model with a lumped series resistance model and Q2DEP model mentioned earlier. Simulation results revealed that if the active area of a current-limiting, lower bandgap MJ subcell is made at least 1.23 times larger than that of the light-emitting higher bandgap subcell, the nonuniformity of the LC effect may be eliminated. Furthermore, this may allow a larger current collection near the MJSC edges, which may then alleviate the perimeter recombination due to sidewall defects and oxidation over time.

Another potential solution explored was the deposition of all-inorganic metal halide perovskite quantum dots (PQDs) – an amorphous thermoplastic mixture on the selected area near the edges of a III-V semiconductor material by dip-coating technique. This was hypothesized to allow a larger amount of light emission towards the edges of a current-limiting lower bandgap subcell, which then may yield larger LC current generation specifically near the edges by photo-passivation or "photon healing" effect. At the same time, the amorphous thermoplastic provided a moisture barrier for the PQDs. It was found that the perovskite quantum dot-coated III-V substrates had larger photoluminescence peaks near the III-V substrate bandgap than the bare samples, suggesting that it can potentially induce LC effect when deposited on a III-V MJSC. With amorphous thermoplastic mixed with quantum dot solution, PL emission was observed to be sustainable for at least 10 weeks. On the other hand, without thermoplastic, the PL emission quenched within 3 hours after deposition. Hence, the weakness of perovskite against moisture, one of its greatest challenges, has been addressed in this work by the use of thermoplastics as a moisture barrier in the solution phase. Last, perovskite quantum dots were deposited on full III-V based MJSC devices, covering their entire front surface. From a fully dip-coated MJSC device with a moderate number of dipping cycles, desirable current redistribution among subcells was achieved; hence, a demonstration of current matching control among MJ subcells.

In summary, there are three pathways to possibly homogenize and increase current collection in a limiting cell of a III-V MJSC device – one is by growing smaller active area of the top cell, another is by electrical sidewall passivation using thin-film dielectrics, and the other is by photo-passivation of shunt defects in MJSCs using a PQD-amorphous thermoplastic film. The electrical sidewall passivation was experimentally observed to homogenize and increase current collection in a subcell benefitting from the LC effect. Hence, the electrical sidewall passivation of full MJSC devices is an additional step recommended in fabricating high-efficiency MJSCs with homogeneous current production, just after the fabrication of the semiconductor stack or after electrode fabrication, provided that the electrodes are masked during passivation layer growth. The PQD-amorphous thermoplastic films on a full III-V MJSC device successfully demonstrated current redistribution from the top cell into its lower subcells. Thus, for enhanced MJSC current matching, it is recommended to use a photo-assistive layer like PQD-amorphous thermoplastic films. Applying these solutions may reach the conversion efficiency increase by 1.35% in MJSCs, based on the estimation made in Chapter 3.