学位論文（要約）

Effects of non-radiative recombination loss on multi-junction tandem solar cells
(多接合タンデム太陽電池における非輻射再結合損失の効果**)**

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Doctoral Dissertation

Effects of non-radiative recombination loss on multi-junction tandem solar cells

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by

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Abstract

Multi-junction (MJ) tandem solar cells, consisting of several series subcells with sequential bandgaps for receiving broader solar spectra, can potentially achieve the highest conversion efficiencies among present photovoltaic devices. Many researchers are now chasing realization of such high efficiencies. In this work, we have studied the effects of non-radiative recombination (NR) loss in MJ tandem solar cells. The NR is an inevitable process in semiconductor materials and plays a key role in the performance of the practical photovoltaic devices. It is dependent on material quality and direct measurements are difficult, which provides challenges to the development of MJ solar cells as well as sources of interesting problems of physics.

We list three problems, which are not solved satisfactorily in the preceding studies in the followings.

Firstly the theoretical efficiency limits (η_{sc}) and optimal sets of bandgaps so far obtained are for the case of no NR loss, which are too ideal to be practical design targets. The effect of NR on the detailed-balance-limit efficiency has been studied only in single-junction solar cells.

The second problem lies in the difficulty of characterization for each subcell. Since all subcells are grown in series on single substrates, we cannot directly measure the voltages of individual subcells. In the preceding studies, methods to evaluate such internal voltages from the external quantum efficiency (EQE) of electroluminescence (EL) and photocurrent have been developed based on the reciprocity relation [Roensch 2011, Kirchartz 2008, Hoheise 2013, Roensch 2011]. In those studies, however, the EL

intensity of each subcell is only given in a relative value, and as a result one needs adjustments (post-renormalization) of the subcell voltages to the total one. Since there exist possibilities of additional voltage-drops in tunnel junctions, ohmic contacts, etc., such adjustments often introduce errors and lead to erroneous conclusions about the optimal design.

The origin of the third problem is not in physics but in the habitual way of providing information in this field. Internal-luminescence quantum yield (y_{int}) serves as a good (anti-)measure of NR, because y_{int} is dependent on material quality but not on structural factors. However, the present formulations to estimate y_{int} need inputs of device structure parameters [Miller 2010, Miller 2012, Steiner 2013, Geisz 2013, Steiner 2012, Steiner JAP 2013]. However, the detailed device information, such as cell thickness and absorptive coefficient, are very often not disclosed in the photovoltaic fields.

The scientific works described in this thesis have been prosecuted for solving the above problems. Here, we summarize the objectives of the present study as;

1. to calculate realistic efficiency limits of tandem solar cells with taking the NR effect into account;
2. to establish a method to measure the NR recombination loss, external emission loss and junction loss in each subcell;
3. to find reliable way to estimate the voltage-drop across each subcell;
4. to propose a procedure to estimate quantum yields of external and internal luminescence in each subcell;
5. to characterize the degradations in respective subcells in proton-damaged InGaP/GaAs/Ge solar cells as an application of the above proposed methods.

The thesis is composed as follows.

In Ch.1, we introduce important backgrounds and give a literature review related to this work. Based on these, the objectives of the present work are addressed.

In Ch.2, we formulate the detailed balance limit of tandem solar cells with subcell y_{int} less than unity. We also derive formulas that relate measurable quantities and all subcell internal properties. An absolute EL measurement method for each subcell is described.

In Ch.3, for the simplest double-junction tandem solar cells, we formulate and numerically calculate the limit of conversion efficiency (η_{sc}) and the optimal set of subcell band-gap energy (E_g) under the condition that the quantum yields of top-cell and bottom-cell internal luminescence (y_{int1} and y_{int2}) are less than unity. In high-y_{int1} (y_{int2}) region, the limit in η_{sc} decreases drastically as y_{int1}(y_{int2}) decreases, while the reduction rate is comparatively low in low-y_{int1}(y_{int2}) region. When both y_{int1} and y_{int2} are less than 0.9, their effects on η_{sc}-limit are symmetric. Otherwise, they are asymmetric, *i.e.*, η_{sc}-limit is more sensitive to y_{int2} than to y_{int1}. When both y_{int1} and y_{int2} are less than 0.3, there exists a power law between η_{sc}-limit and the geometric mean, $y_{int}^{*}=\sqrt{y_{int1}y_{int2}}$.

In Ch.4, we investigate the effect of subcell y_{int} on the efficiency limit of n-junction ($n \geq 3$) tandem cells with introducing the geometric mean (y_{int}^{*}) of all subcell y_{int} as a measure of the subcell quality. We calculate the conversion efficiency limit and the optimized band-gaps of n-junction tandem solar cells under the 1-sun and 100-sun conditions with various y_{int}^*. We find that when y_{int}<0.3 holds for all the subcells, the η_{sc}-limits as a function of y_{int}^* does not depend greatly on the combination of (y_{int1}, y_{int2}, … , $y_{int\,n}$), hence can be represented by the special case of y_{int1} = y_{int2} =… = $y_{int\,n}$. It is found that as y_{int}^* decreases from 1 to 0.1, η_{sc} decreases drastically. And for y_{int}^*< 0.1, logarithmic relationships are found between η_{sc}-limit and y_{int}^*. With decreasing y_{int}^*, the optimized set of bandgaps shows significant blue-shifts. The present results provide realistic efficiency targets and bandgap designs.

In Ch.5, we apply the above formulation to the results of absolute subcell EL measurements in a satellite-use GaInP/GaAs/Ge 3-junction tandem solar cell, and characterize individual subcell properties. Each subcell y_{int} is obtained independently, and turned out to be strongly current dependent. The current-limiting subcell usually has a low $y_{int\,i}$ under maximum-output-power condition. Thanks to the absolute EL measurement, the subcell I-V curves are obtained without any ambiguous adjustment. The good agreement between the summation of the I-V curves obtained from EL measurement and the directly measured total I-V curve confirms the reliability and feasibility of the present method. This experiment also provides the detailed information on the energy losses, such as junction (JN) loss, NR loss, external emission (EM) loss, luminescence coupling (LC), and thermalization (TH) loss in the respective subcells, which provides intriguing insights into the characteristics of the 3-junction solar cell, though they seem very complicated at the first sight. The cell exhibits large NR and JN losses and is far from the detailed-balance limit. This is caused by low $y_{int\,i}$ at low-carrier-density regimes.

In Ch.6, we apply our formulation with absolute subcell EL experiments to twelve satellite-use GaInP/GaAs/Ge tandem solar cells damaged by proton irradiation with different energies and fluences. We examine the individual subcell degradations by using a sample with no damage as the reference. Firstly, we quantitatively evaluate the individual subcell I-V curves and efficiency in these samples. The sums of three subcell open circuit voltages (V_{oc}) are consistent with the total V_{oc} in the I-V measurements. Secondly, we quantify individual subcell y_{int} under AM0. We find that y_{int} is indeed a good indicator of radiation damages. Based on the y_{int} measurement, the effects of proton energy and fluence on material quality are analyzed, which quantitatively explain the individual subcell degradation. Finally, we confirm that the empirically obtained relation between the conversion efficiency and the geometric mean (y_{int}^*) of subcell y_{int} really holds for the measured values.

In Ch.7, a summary of the study and perspectives are presented.