Tuesday Afternoon, October 30, 2012

Energy Frontiers Focus Topic
Room: 15 - Session EN+TF-TuA

Thin Film, Heterostructured, and Organic Solar Cells

Moderator: M. Filler, Georgia Institute of Technology

2:00pm **EN+TF-TuA1 Photonic Materials for Solar Energy Conversion at the Thermodynamic Limit**, ***H.A. Atwater***, California Institute of Technology **INVITED**

Ever since serious scientific thinking went into improving the efficiency of photovoltaic energy conversion more than 50 years ago, thermodynamics has been used to assess the limits to performance, guiding advances in materials science and photovoltaic technology. Photovoltaics have advanced considerably, resulting in single-junction solar cells with a record efficiency of 28.8% and multi-junction cells with an efficiency of 43.5%. As impressive as these advances are, these record efficiencies and also today's manufactured cell efficiencies in the 10–18% range fall far short of the thermodynamic limits. Why such a large gap? There is no fundamental reason, and in this talk, I will discuss methods for systematically addressing the thermodynamic efficiency losses in current photovoltaics that can enable a next phase of photovoltaic science and engineering – ultrahigh efficiency photovoltaics. This development takes advantage of recent advances in the control of light at the nanometer and micron length scales, coupled with emerging materials fabrication approaches, and will allow the development of solar cells with efficiencies in the 50–70% range.

Web resources:

http://www.lmi.caltech.edu/

http://daedalus.caltech.edu/

2:40pm **EN+TF-TuA3 Semiconducting Carbon Nanotubes as Polymer-Like Near-Infrared Bandgap Photoabsorbers**, ***M.S. Arnold***, *D.J. Bindl, M.-Y. Wu, M.J. Shea*, University of Wisconsin Madison **INVITED**

We are pioneering the exploration of semiconducting carbon nanotubes as the light-absorbing components of polymer-inspired solar cells and photodetectors.[1-2] Carbon nanotubes are conjugated polymer-like materials with built-in long-range crystallinity that gives rise to exceptional charge and energy transport characteristics, strong light absorption tunable throughout the visible and near-infrared spectra, and outstanding stability in air. We have discovered how to efficiently harvest photogenerated charges and excitons from optically excited nanotubes by pairing them in donor / acceptor heterojunctions with more electronegative electron accepting semiconductors. In particular, semiconducting nanotubes form a type-II heterojunction with C60 fullerenes and C60 derivatives with energy offsets sufficient to drive electron transfer from the optically excited nanotubes to C60, with an internal quantum efficiency (QE) for exciton dissociation and charge transfer > 75%, for nanotubes of diameter < 1 nm and gaps > 1 eV. Thus, we have identified the nanotube / C60 materials pair as a promising basis for future nanotube-based light harvesting devices.

In order to further guide the implementation of nanotubes in devices, we have also characterized exciton transport in nanotube films and shown that excitons can migrate in films by two mechanisms: (i) over short distances of ~ 5 nm via slow inter-nanotube diffusion and (ii) potentially over much longer distances via rapid intra-nanotube diffusion. As a proof-of-principal, we have fabricated both bilayer and blended nanotube / C60 heterojunction devices, which are analogous to polymer solar cells with nanotubes taking on the role of the semiconducting "polymer". Thus far, we have realized a peak external QE > 20% across 1000 – 1365 nm and a monochromatic power conversion efficiency of 7% at 1050 nm. Our results show that AM1.5G photovoltaic power conversion efficiency > 10% should be possible with future optimization of: (a) the nanotube bandgap (and diameter) distribution and (b) improved control over morphology.

[1] D. J. Bindl, M.-Y. Wu, M. S. Arnold, *Nano Letters* (2011).

[2] D. J. Bindl, A. S. Brewer, M. S. Arnold, *Nano Research* (2011).

4:00pm **EN+TF-TuA7 Understanding Vertical Stratification in Polymer:Fullerene Bulk Heterojunction Solar Cells**, *M.D. Clark*, Air Force Research Laboratory, *M.L. Jespersen*, University of Dayton Research Institute, ***B.J. Leever***, Air Force Research Laboratory

In the bulk heterojunction architecture of polymer-based solar cells (PSCs), the separate acceptor-donor phases form a bi-continuous inter-penetrating network by simultaneous casting from solution with morphological control stemming from external parameters such as thermal annealing, co-solvent inclusion, and drying conditions. While such treatments enhance device performance, a fundamental understanding of vertical concentration gradients within the fabricated active layer has been limited. In an effort to understand such morphological changes, several reports have explored 3D bulk heterojunction nanostructure using electron tomography,[1] ellipsometry,[2] neutron scattering,[3] and spectroscopic techniques.[4] This work, however, has yielded somewhat contradictory conclusions about fundamental network development and the origin of emerging concentration gradients. For example, some studies reported nearly equal blends at the PEDOT:PSS surface of annealed samples,[2,4c] while others found P3HT[1,4a,4e] or PCBM[3,4b,4d,4f] preferentially decorating the buried interface. Several groups[2b,3,4c,4f] further reported annealing causes PCBM diffusion towards the exposed surface, suppressing as cast vertical composition gradients. However, Xu et al.[4b] detected PCBM migration towards the PEDOT:PSS interface upon annealing, while Xue et al.[4a] suggested PCBM diffusion away from both interfaces. Here we report a combined experimental and theoretical analysis of phase segregation. The vertical stratification within a P3HT:PCBM bulk heterojunction solar cell is examined by depth profiling using both x-ray photoelectron spectroscopy (XPS) and time of flight secondary ion mass spectrometry (ToF-SIMS), with the effects of thermal annealing and P3HT:PCBM ratio being explored. In addition, the vertical phase stratification is predicted on thermodynamic grounds based on measured interfacial energies of the PSC constituents. Using these results, a fundamental understanding of the thermodynamic driving force for bulk heterojunction phase segregation and vertical stratification is then presented.

References

1 van Bavel, S.S. et al. *Nano Lett.***9**, 507 (2009).

2 (a) Germack, D.S. et al. *Macromolecules* **43**, 3828 (2010); (b) Campoy-Quiles, M. et al. *Nat. Mater.***7**, 158 (2008).

3 Parnell, A.J. et al. *Adv. Mater.***22**, 2444 (2010).

4 (a) Xue, B. et al. *J. Phys. Chem. C***114**, 15797 (2010); (b) Xu, Z. et al. *Adv. Funct. Mater.***19**, 1227 (2009); (c) Yu, B.-Y. et al. *ACS Nano***4**, 833 (2010); (d) Vaynzof, Y. et al. *ACS Nano***5**, 329 (2011); (e) Wang, H. et al. *Chem. Mater.***23**, 2020 (2011); (f) Germack, D.S. et al. *Appl. Phys. Lett.***94**, 233303 (2009).

4:20pm **EN+TF-TuA8 Novel, Single-Crystalline-like Silicon on Low-Cost, Flexible Substrates for High Efficiency Thin Film Photovoltaics**, ***V. Selvamanickam***, *P. Dutta, R. Wang, Y. Gao, M. Yang, G. Majkic, E. Galtsyan*, University of Houston

Thin film photovoltaics (PV) is being pursued by several institutions as a lower cost alternative to crystalline wafer technologies. The use of much less materials and roll-to-roll continuous processing in thin film technologies have been touted as the pathway to low-cost PV. However, the efficiencies of production thin film Si solar cells are about one half that achieved with crystalline silicon. Hence, achievement of single-crystalline-like silicon photovoltaics on flexible, low-cost substrates can be game changing by combining high efficiency with low cost. We are developing such a technology by creation of an architecture that yields single-crystalline-like thin films even on polycrystalline or amorphous substrates. This technology has been very successfully demonstrated and being commercialized in the superconductor field and inserted in the U.S. electric power grid [1].

The enabler that we have employed in this work is a single-crystalline-like thin film template of MgO made by Ion Beam-Assisted Deposition (IBAD). Such IBAD films have been successfully employed as templates for epitaxial growth of cube-textured superconducting films on polycrystalline substrates with critical current densities as high as those achieved on single crystal substrates [1].

MgO templates made by IBAD on flexible metal substrate have been used for epitaxial growth of germanium films using intermediate oxide layers. All layers were deposited by reel-to-reel magnetron sputtering and strongly (400) textured Ge films with an in-plane texture spread of just 1° FWHM

were achieved [2]. Optical properties of the germanium films are found to be comparable to that single crystal Ge and Hall mobility values over 700 cm2/Vs have been achieved. Epitaxial (400) textured silicon films have been grown by reel-to-reel magnetron sputtering on the Ge films. A continuous grading of germanium to silicon has been done to accommodate for the lattice mismatch. While excellent epitaxial growth has been achieved in Si and Ge on flexible metal substrates, the defect density of the films showed a high value of 10^8 per cm^2. Cross sectional TEM of the multilayer architecture showed concentration of threading dislocations near the semiconductor-oxide interface. Defect reduction strategies are being employed and recent progress in use of single-crystalline-like templates on low-cost, flexible substrates for high-efficiency silicon photovoltaics will be discussed in this presentation.

1. V. Selvamanickam et al. *IEEE Trans. Appl. Supercond.* **19** (2009) 3225.

2. V. Selvamanickam et al. *J. Crystal Growth* **311,** (2009) 4553.

4:40pm **EN+TF-TuA9 High-Efficiency Multijunction Solar Cells Employing Dilute Nitrides**, ***V.A. Sabnis***, *H.B. Yuen, M. Wiemer*, Solar Junction **INVITED**

Concentrating photovoltaic (CPV) systems have the opportunity to provide the lowest cost of electricity in hot, sunny climates. The advantages of CPV are based, in part, from the high performance offered by multijunction solar cells made from group III-V compound semiconductors. Unlike traditional PV, high concentration systems utilize mm-scale solar cells that comprise only 10-15% of the overall system cost. This low cost share means that increasing cell efficiency has significant leverage in driving down upfront capital costs and the levelized cost of electricity of a CPV project.

Production cell efficiencies for triple junction solar cells have reached 40% under concentration (25°C, AM 1.5D spectrum). We will review a number of exciting approaches for increasing cell efficiency that are being investigated world wide. Solar Junction has developed a set of dilute-nitride compound semiconductors that offer broad bandgap tunability over the infrared while retaining lattice matching to GaAs and Ge substrates. While significant efforts have been undertaken to develop dilute nitrides for multijunction solar cells over the last 15 years, these approaches have resulted in films that exhibited poor minority carrier properties resulting in low current drives and output voltages. Solar Junction has developed a molecular beam epitaxy process utilizing antimony as a surfactant that significantly enhances the minority carrier properties. Triple junction cells utilizing GaInNAsSb bottom junctions have achieved a world record efficiency of 43.5% under concentration. When used in conjunction with well known InAlGaP and AlGaAs compounds, GaInNAsSb films complete a lattice-matched epitaxial platform for enabling 4-, 5-, and 6-junction cells for achieving > 50% efficiencies in the coming years.

5:20pm **EN+TF-TuA11 Non-Radiative Carrier Recombination in InGaAs/GaAsP Strain-Balanced Superlattice Solar Cell**, ***T. Aihara***, University of Miyazaki, Japan

An inserting of the quantum wells (QWs) to GaAs p-i-n solar cells could be a promising candidate to solve the current matching issue in the multi-junction solar cells[1]. We have successfully obtained the non-radiative recombination process for excitonic and subband absorptions in the GaAs/AlAs multiple QWs (MQWs) by using PPT methods [2]. In this study, we investigate escape, radiative and non-radiative recombination mechanisms of photo-generated carriers in the strain-balanced InGaAs/GaAsP MQWs or superlattice (SL) inserted into GaAs p-i-n solar cell structure to improve the photovoltaic performance. We then evaluated above three processes by using the surface photovoltage (SPV), photoluminescence (PL), and piezoelectric photothermal (PPT) spectroscopies, respectively. A InGaAs/GaAsP MQWs absorbing layer that inserted into GaAs p-i-n junction was composed of 10 stacks of 7.4-nm-thick InGaAs well and 10.8-nm-thick GaAsP barrier. For SL absorbing layer, ultra-thin GaAsP barriers of 3.7 nm thickness with 0.56-nm-thick GaAs buffer were prepared. All the layers were grown by metal-organic vapor phase epitaxy on the GaAs substrate. The PPT detects a heat generated by the non-radiative recombination by the PZT directly attached to rear surface of the sample. Figures 1 and 2 show the temperature change of PPT spectra of MQWs and SL with GaAs thin buffer samples, respectively. For MQWs sample, three peaks were observed and A-peak was concluded to be due to the excitonic transition associated with the electron transition between first electron (e1) and heavy-hole subbands (hh1) in QW. On the other hand, B-peak was concluded to be the electron transition between 1st minibands in conduction and valence bands in SL. As the temperature decreased, peak intensities of A and B increased, whereas corresponding SPV peaks decreased. The temperature dependence of PL, PPT, and SPV signal intensities can be fitted with the Arrhenius equation. Figure 3 shows the fitting results of PPT A (MQWs) and B (SL) and SPV A peaks. As shown in Fig. 3, activation energy of SL was smaller than that of MQWs. This result implied that carrier escape from the QWs was enhanced for the case of SL . References [1] K. W. J. Barnham and G. Duggan, J. Appl. Phys. 67 (1990) 3409. [2] P. Wang et al.: Jpn. J. Appl. Phys. 46 (2007) 6857.

5:40pm **EN+TF-TuA12 Piezoelectric Photothermal Spectra and Carrier Nonradiative Recombination in InGaAs/GaAsP Super Lattice Structured Solar Cells**, ***T. Ikari***, *T. Aihara, Y. Nakano*, University of Miyazaki, Japan, *Y. Wang, M. Sugiyama, Y. Nakano*, University of Tokyo, Japan, *A. Fukuyama*, University of Miyazaki, Japan

Fabrication of multi quantum well (MQW) or superlattice (SL) structures embedded in an absorption layer of solar cell is a promising idea for developing higher efficient devices. This is because the quantum well can extend the absorption to longer wavelength region and enhance the short-circuit current. However, recombination centers for carriers are simultaneously generated at the boundaries, leading to the degradation of conversion efficiency. Although optical absorption and spectral response spectroscopy are usually used for investigating absorption and recombination mechanism in the solar cell, no direct technique for characterizing nonradiative recombination is presented. We have developed the piezoelectric photothermal (PPT) spectroscopy for detecting such nonradiative recombination in the QW [1]. Heat generated by a nonradiative recombination of photoexcited carriers were detected as PPT signal by using a piezoelectric transducer. In this paper, we report on the PPT spectra of InGaAs/GaAsP SL layer and show that this technique is sensitive and powerful to investigate the absorption spectra of SL. It is, then, becomes possible to discuss a recombination mechanism of the photo-excited carriers in the solar cell structure from the non-radiative transition point of view.

A strain-balanced InGaAs/GaAsP SL layer embedded into the intrinsic region of the GaAs *p-i-n* solar cell were prepared. The SL absorbing layer was prepared in the structure of InGaAs(3.7nm)/GaAsP(5.4nm) with 0.56-nm-thick GaAs buffer layer by MOVPE technique on the GaAs substrate [2]. PPT spectrum at 100K shows two dominant peaks. The conventional absorbance of SL and the PPT spectrum of the MQW (InGaAs(7.4nm)/GaAsP (10.8nm)) samples were also discussed for comparison. A signal from SL is more clearly observed in the PPT than the absorbance spectra. Although the step like signals accompanied with the exciton transition are well resolved for MQW samples, no step but broad peaks around 1.395 and 1.45 eV were observed for SL sample. The energies of these peaks were as expected from the calculation of the single QW without any interaction of the neighboring QW, i.e. tunneling. Since the wave function of quantized level spread into the next well for the SL structure, broad peaks were, then, observed. Although the step like density of states should appear even in the SL, drastic decrease of the PPT signal beyond the peak was observed. One possible reason is that the number of carriers that recombine nonradiatively inside the quantum well decreases by tunneling.

[1] T. Ikari, *et al*., Phys. Rev. B77 (2008) 125311.

[2] M. Sugiyama *et al*.: J. Cryst. Growth **315** (2011) 1.

Authors Index

Bold page numbers indicate the presenter