



Electronic Materials Science Challenges in Renewable Energy

Richard R. King

Spectrolab, Inc. *A Boeing Company*

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Electronic Materials in Renewable Energy



- Virtually all of the topics in the EMC Program have a potential impact on renewable energy technologies
- Electronic materials research can make or break many of these emerging technologies
- Challenge that is being picked up increasingly by Academic, Gov't, and Industry research centers:

"How can the basic materials science we have been working on for devices in telecom, computing, imaging, displays... be applied to photovoltaic cells, energy storage, and other renewable energy applications?"



A Few Key Growth Areas for Renewable Energy



This talk is 'focused' on the huge area of photovoltaics...

- Multijunction photovoltaic cells for grid-connected solar electricity
 - Metamorphic (MM) semiconductors to access new band gaps
 - III-V growth on Ge, SiGe, and silicon
 - Wafer bonding and engineered substrates
 - Narrow band gap semiconductors: antimonides and others
 - Wide band gap semiconductors: nitrides and others
 - Point and extended defects in MM materials
 - Low-dimensional structures: quantum dots, wires, and wells
- Flat-plate photovoltaics
 - Polycrystalline thin-film compound semiconductor solar cells (CuInSe₂, CdTe,...)
 - Microcrystalline silicon solar cells
 - Organic-inorganic hybrid photovoltaics



A Few Key Growth Areas for Renewable Energy

...but electronic materials impact many more aspects of renewable energy

General Applications

- Carbon nanotubes, graphene
- Zinc oxide
- Flexible and printed thin film electronics

Batteries, fuel cells for vehicles

- Porous, catalytic electrodes
- Ionic conductors
- Power conditioning, DC to AC conversion
 - Silicon carbide
- Thermoelectrics and thermionics

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- Global climate change and the solar resource
- Solar cell theoretical efficiency limits
 - Opportunities to change ground rules for higher terrestrial efficiency
 - Cell architectures capable of >70% in theory, >50% in practice
- Metamorphic semiconductor materials
 - Control of band gap to tune to solar spectrum
 - Dislocations in metamorphic III-Vs imaged by CL and EBIC
 - Metamorphic SiGe buffer and solar cell growth
 - III-V solar cells on Si substrates
- Polycrystalline Cu(Galn)Se2 and CdTe for flat-plate solar cells
 - Remarkable recombination inactivity for grain boundaries
 - Understanding energetically-favored defect formation
 - Defect energy levels, Fermi stabilization energy







Outline (cont'd)



- Nanostructures for high-efficiency photovoltaics
 - Nanorod solar cell arrays grown by VLS (vapor-liquid-solid) method
 - Quantum wells, wires, dots, low-dimensional structures in solar cells
 - Organic semiconductors for PV
- High-efficiency Multijunction terrestrial concentrator cells
 - Metamorphic and lattice-matched 3-junction solar cells with >40% efficiency
 - 39%-efficient cells at >1000 suns
 - *4-junction* metamorphic (MM) and lattice-matched (LM) concentrator cells
 - Inverted metamorphic 3- and 4-junction cells for terrestrial concentrators
 - Semiconductor bonded technology (SBT) for MJ terrestrial concentrator cells
- Concentrator photovoltaic (CPV) systems and economics









Global Climate Change



Temperature (°C)

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Climate and CO₂ Over the Last 400,000 Years



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(J.R. Petit, J. Jouzel, Nature 399:429-436)

Vostok Ice Core Data 4 2 0 -2 -4 -6 -8 -10

Antarctic ice core data allows for mapping of temperature and CO₂ profiles

Years Before Present

45000 4000 35000 3000 25000 2000 15000 1000 5000

Climate and CO₂ Over the Last 400,000 Years

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• Clear correlation between temperature and CO₂ levels



Climate and CO₂ -Recent History





Years Before Present

- CO₂ has reached levels never before seen in measured history
- If we do nothing, we allow this rising trend to continue at our own peril



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The Solar Resource

Solar power: 30,000 kilowatts per person



0.25% converted to wind (380 TW) Angus Rockett, Univ. of Illinois



The Solar Resource





 Entire US electricity demand can be provided by concentrator PV arrays using 37%-efficient cells on:

150 km x 150 km area of land or ten 50 km x 50 km areas or similar division across US

Concentrator Photovoltaic (CPV) Electricity Generation

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Map source: http://www.nrel.gov/gis/images/map_csp_us_annual_may2004.jpg

Higher multijunction cell efficiency has a huge impact on the economics of CPV, and on the way we will generate electricity.

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Solar Cell Theoretical Efficiency



Energy Transitions in Semiconductors







LM and MM 3-Junction Cell Cross-Section



ropcell

MidetesTunn

Middle Cell

Tunnel Junction

Bottom Cell



Lattice-Mismatched or Metamorphic (MM)

SPECTROLAB Photon Utilization Efficiency Description ABDEING COMPANY 3-Junction Solar Cells



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Energy Transitions in Semiconductors





- V = voltage of solar cell
 - = quasi-Fermi level splitting = $|\phi_p - \phi_n|$
- Not all of bandgap energy is available to be collected at terminals, even though electron in conduction band has energy E_g
- Only qV = q $|\phi_p \phi_n|$ is available at solar cell terminals
- Due to difference in entropy S of carriers at low concentration in conduction band, and at high concentration in contact layers: G = H - TS



Energy Transitions in Semiconductors





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Electronic Materials for Renewable Energy







Detailed Balance Limit of Solar Cell Efficiency



- 30% efficient
 single-gap solar
 cell at one sun,
 for 1 e⁻/photon
- 44% ultimate
 efficiency for
 device with
 single cutoff
 energy







- Assumptions for theoretical efficiency in Shockley and Quiesser (1961)
- Viewed from a different angle, these assumptions represent new opportunities, for devices that overcome these barriers

Assumption limiting solar cell efficiency	Device principle overcoming this limitation	
Single band gap energy	Multijunction solar cells	
	Quantum well, quantum dot solar cells	
One e⁻-h⁺ pair per photon	Down conversion	
	Multiple exciton generation	
	Avalanche multiplication	
Non-use of sub-band-gap photons	Up conversion	
Single population of each charge carrier type	Hot carrier solar cells	
	Intermediate-band solar cells	
	Quantum well, quantum dot solar cells	
One-sun incident intensity	Concentrator solar cells	



Maximum Solar Cell Efficiencies

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Measured Theoretical

 <u>References</u> C. H. Henry, "Limiting efficiencies of ideal single and multiple energy gap terrestrial solar cells," <i>J. Appl. Phys.</i>, 51, 4494 (1980). W. Shockley and H. J. Queisser, "Detailed Balance Limit of Efficiency of <i>p-n</i> Junction Solar Cells," <i>J. Appl. Phys.</i>, 32, 510 (1961). J. H. Werner, S. Kolodinski, and H. J. Queisser, "Novel Optimization Principles and Efficiency Limits for Semiconductor Solar Cells," <i>Phys. Rev. Lett.</i>, 72, 3851 (1994). 	95% 93%	Carnot eff. = $1 - T/T_{sun}$ T = 300 K, $T_{sun} \approx 5800$ K Max. eff. of solar energy conversion = $1 - TS/E = 1 - (4/3)T/T_{sun}$ (Henry)
 M. Green, K. Emery, D. L. King, Y. Hisikawa, W. Warta, "Solar Cell Efficiency Tables (Version 27)", <i>Progress in Photovoltaics</i>, 14, 45 (2006) R. R. King <i>et al.</i>, "40% efficient metamorphic GalnP / GalnAs / Ge multijunction solar cells," <i>Appl. Phys. Lett.</i>, 90, 183516 (4 May 2007). J. F. Geisz et al., "40.8% efficient inverted triple-junction solar cell with two independently metamorphic junctions," <i>submitted to Appl. Phys. Lett.</i> (2008). A. Slade, V. Garboushian, "27.6%-Efficient Silicon Concentrator Cell for Mass Production," <i>Proc. 15th Int'l. Photovoltaic Science and Engineering Conf.</i>, Beijing, 	72%	Ideal 36-gap solar cell at 1000 suns (Henry)
 China, Oct. 2005. R. P. Gale <i>et al.</i>, "High-Efficiency GaAs/CuInSe₂ and AlGaAs/CuInSe₂ Thin-Film Tandem Solar Cells," <i>Proc. 21st IEEE Photovoltaic Specialists Conf.</i>, Kissimmee, Florida, May 1990. J. Zhao, A. Wang, M. A. Green, F. Ferrazza, "Novel 19.8%-efficient 'honeycomb' textured multicrystalline and 24.4% monocrystalline silicon solar cells," <i>Appl.</i> 	56% 50%	Ideal 3-gap solar cell at 1000 suns (Henry) Ideal 2-gap solar cell at 1000 suns (Henry)
<i>Phys. Lett.</i> , 73 , 1991 (1998).	44%	Ultimate eff. of device with cutoff Eg: (Shockley, Queisser)
	43%	1-gap cell at 1 sun with carrier multiplication
3-gap GalnP/GalnAs/GalnAs cell at 326 suns (NREL) 40.8% 3-gap GalnP/GalnAs/Ge cell at 240 suns (Spectrolab) 40.7%	,) ,	(>1 e-h pair per photon) (Werner, Kolodinski, Queisser)
	37%	Ideal 1-gap solar cell at 1000 suns (Henry)
3-gap GaInP/GaAs/GaInAs cell at 1 sun (NREL) 33.8%	, D	
	31% 30%	Ideal 1-gap solar cell at 1 sun (Henry) Detailed balance limit of 1 gap solar cell at 1 sun (Shockley, Queisser)
1-gap solar cell (silicon, 1.12 eV) at 92 suns (Amonix) 27.6% 1-gap solar cell (GaAs, 1.424 eV) at 1 sun (Kopin) 25.1% 1-gap solar cell (silicon, 1.12 eV) at 1 sun (UNSW) 24.7%		





Growth on Ge or GaAs substrate, followed by substrate removal from sunward surface

Solar Spectrum Partition for **BOEING** 3-Junction Cell



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5- and 6-Junction Cells





Ref.: U.S. Pat. No. 6,316,715, Spectrolab, Inc., filed 3/15/00, issued 11/13/01.

SPECTROLAB Photon Utilization Efficiency Description ABDEING COMPANY 3-Junction Solar Cells



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SPECTROLAB Photon Utilization Efficiency Description ABDEING COMPANY 6-Junction Solar Cells



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3-Junction Cell Efficiency Losses from 100%









Metamorphic Semiconductor Materials



Metamorphic (MM) Semiconductor Materials



- Metamorphic = "changed form"
- Thick, relaxed epitaxial layers grown with different lattice constant than growth substrate
- Allows access to subcell band gaps desired for more efficient division of the solar spectrum in multijunction solar cells
- Also called lattice-mismatched
- Misfit dislocations are allowed to form in metamorphic buffer, which typically has graded composition and lattice constant
- Threading dislocations which can propagate up into active device layers grown on buffer are minimized as much as possible



Bandgap vs. Lattice Constant





Courtesy J. Geisz – NREL
Internal QE of Metamorphic GalnAs Cells on Ge





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Cross sectional TEM Ga_{0.44}In_{0.56}P/ Ga_{0.92}In_{0.08}As/ Ge

- Low dislocation density in active cell layers in top portion of epilayer stack:
 - ~ 2 x 10⁵ cm⁻² from EBIC and CL meas.

 Dislocations confined to graded buffer layers in bottom portion of epilayer stack



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High-Resolution XRD A BOEING COMPANY High-Resolution XRD Reciprocal Space Map (RSM)



- GalnP/ 8%-In GalnAs/ Ge metamorphic (MM) cell structure
- Nearly 100% relaxed stepgraded buffer → removes driving force for dislocations to propagate into active cell layers
- 56%-In GaInP top cell pseudomorphic with respect to GaInAs middle cell

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A BOEING COMPANY I.39-eV GalnAs Subcell



Growth on Ge or GaAs substrate, followed by substrate removal from sunward surface





Growth on Ge or GaAs substrate, followed by substrate removal from sunward surface





Growth on Ge or GaAs substrate, followed by substrate removal from sunward surface





Growth on Ge or GaAs substrate, followed by substrate removal from sunward surface



Dislocations in Inverted Metamorphic Cells – EBIC



1.39-eV ILM subcell

GalnAs comp.2% InLatt. mismatch0.1%Disloc. density 2.5×10^5 cm⁻²









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50 μm **0.97-eV IMM subcell** 33% In 2.3% 5.0 x 10⁶ cm⁻²





metal contact O.84-eV GalnAs base emitter transparent MM graded buffer layers nucleation and pre-grade buffer Ge substrate

EBIC images and dislocation density of inverted metamorphic cell test structures



Dislocations in Inverted Metamorphic Cells





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Solar Cell Voltage



Voltage depends on non-equilibrium concentrations of electrons and holes

$$pn = n_i^2 e^{qV/kT}$$

$$n_i^2 = N_C N_V e^{-E_g/kT} \qquad pn = N_C N_V e^{-(E_g - qV)/kT} = N_C N_V e^{-qW/kT}$$
$$V = \frac{kT}{q} \ln\left(\frac{pn}{n_i^2}\right) \qquad W \equiv \left(\frac{E_g}{q}\right) - V = \frac{kT}{q} \ln\left(\frac{N_C N_V}{pn}\right)$$

• Bandgap-voltage offset $W \equiv (E_g/q) - V$ is a useful parameter for gauging solar cell quality, especially when dealing with semiconductors of many different bandgaps

 Basically a measure of how close electron and hole quasi-Fermi levels are to conduction and valence band edges SPECTROLAB

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Band gap - Voltage Offset (Eg/q) - Voc



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Metamorphic III-V Growth on Si Substrates Using SiGe Buffers



SiGe metamorphic substrate engineering for III-V/Si PV





Steve Ringel, Ohio State, and Gene Fitzgerald, MIT



III-V/Si PV Results via SiGe Substrate Engineering



GaAs SJ cell on SiGe (Andre, et al., IEEE TED 2005) (Ringel et al., Prog. In PV, 2002)

GaInP/GaAs DJ cell on SiGe

(Lueck et al, IEEE EDL 2006) (Ringel, et al, 2004 Fall MRS; IEEE PVSC 2005)

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Polycrystalline Thin-Film Solar Cells:

Cu(In,Ga)Se₂



Cu(In,Ga)Se₂



\$0.5/kWh

Multijunction

CIGS



- Cost / watt equivalent to First Solar CdTe
- Performance matching the best multijunctions
- Issues:
 - Major defect contributing to carrier loss
 - Wide-gap devices to enable multijunctions?
 - Fundamental understanding of the device incomplete



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SIMS Atom Fraction (at.

1000 1500

Sputtering Time (sec)

2000

2500

0.0 0.2 0.4 0.6 0.8

Depth (microns)

1.0 1.2

500



The Real Device



Devices are thought to be limited by recombination in the depletion region, not by heterojunction recombination.

- What is the major recombination center?
- What do grain boundaries do?
- Why does CuGaSe₂ not work well?
- Why do some growth processes work better than others?

TEM image





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Thin-Film Cu(In,Ga)(S,Se)₂ - CIGSSe

Larry Kazmerski, NREL

Front Contact: 3.0 μm Al 0.05 μm Ni

 $\begin{array}{c} MgF_2 \\ Antireflection Coating \\ (ARC) (0.08-0.12 \ \mu m) \\ \hline ZnO \ Window (0.4-0.6 \ \mu m) \\ \hline CdS \\ Window (0.05 \ \mu m) \end{array} \begin{array}{c} Cha \end{array}$

CIGS Absorber (2-4 μm)

Mo Back Contact (1 μm)

Glass Substrate

Challenges

- Higher module efficiency
- Thinner aborber layers
 (< 1.0 μm)
- Alternative absorber production (processes)
- Gaps in efficiency between various absorbers
- Faster absorber processing
- Stability (water ingress)
- Materials availability/cost
- Uniformity and stoichiometry
- Standardization of equipment
- Environmental concerns
- Recycling, "Insurance"
- Substrates (glass, plastics)

Thin-Film Cu(In,Ga)(S,Se)₂ - CIGSSe





Chalcopyrite CIGS



- Disordering energy is low so there are many point defects
- CIGS is a polar compound so charged surfaces could be a problem





Yet:

- Extended defects inactive
- Polar surfaces most stable
- Hole mobility phonon limited for p to >10¹⁹ cm⁻³
- Polycrystalline devices work better than single crystals

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Self-Passivating Defects in Cu(Galn)Se₂



- Certain materials systems exhibit low minority-carrier recombination in spite of very high defect densities
 - e.g., GaN and InGaN devices, polycrystalline Cu(GaIn)Se₂ (CIGS) solar cells
- Need atomic level understanding of the mechanisms that render dislocations inactive in these remarkable materials
- CIGS solar cells
 - Cu vacancies (V_{Cu}) and cation antisite defects such as In_{Cu} have a low enthalpy of formation
 - spontaneously forms Cu-poor phases of CIGS
 - defects form complexes in which they can neutralize each other electrically
- Microchemical analyses show little composition change between grain boundaries and bulk in CIGS
 - suggests recombination activity self-passivating, due to nature of surface defects
- Can self-passivating behavior of polycrystalline CIGS can be extended to other semiconductors, additional device structures?
 - If so, opens possibility of high-efficiency MJ solar cells in low-cost thin-film configuration
- Understanding of self-passivating phenomena at defects and interfaces is essential for nanostructured photovoltaic cells, *e.g.*, quantum dot and nanowire solar cells
 - Very large interfacial area is natural consequence of nanoscale structures



(220)/(204) Oriented CIGS

- Layers facet spontaneously into **polar** (112) type planes
- Smooth facets alternate with rough facets •

Conclusion:

Indexing surface planes shows smooth planes are metal terminated

(220)/(204) epitaxial layer AFM image

Red: metal terminated Blue: Se terminated Somehow the polar surfaces are stabilized,

giving a very strong preference for these.

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(112)_A



CIGS solar cells

- Are heterojunction devices with a very strongly inverted junction (Cd doping overwhelms Fermi level pinning).
- Do not mind grain boundaries because they are highly faceted to extremely passive (112) surfaces.
- Heterojunction is made to these surfaces regardless of grain orientation.
- Point defects control doping in the bulk and are very consistent.
- Edge dislocations do not matter because they turn into (112) surfaces.

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Nanostructured, Organic, and Other Novel Solar Cells

Intermediate Band Solar Cells



A. Norman, M Romero, and M. Al-Jassim, (NREL), A. Luque, A. Marti, Spain Larry Kazmerski, NREL

MOCVD Growth of InGaAs/GaAs QD arrays on (113)B GaAs substrates for intermediate band solar cells



 QD arrays are being grown to test concept of intermediate band solar cell proposed by A. Martí and A. Luque

(311)B 50 period InGaAs/GaAs QD superlattice plan-view TEM



Chains of quantum dots along <110>



Nanostructures for Solar Cells





solar cell based on arrays of Si wires features:

- Orthogonalize light absorption and photocarrier collection
- Retain efficiencies competitive with planar, crystalline Si solar cells
- Compatible with low minority carrier diffusion length
- Si wire arrays formed by SiCl₄ chemical vapor deposition
- Can be formed into flexible arrays that are peeled off of template Si

Harry Atwater, Caltech



Nanorod Solar Cells



Large Area (>1 cm²) Si Wire Arrays



B.M. Kayes, M.A. Filler, et al., App. Phys. Lett. 91, 103110 (2007)

Harry Atwater, Caltech



- The "Gratzel" cell uses an electrolyte to conduct holes away from the chromophore.
- Issues
 - Energy loss at electrolyte/TiO₂ redox reaction
 - Energy loss at TiO₂ Ru complex interface
 - Hermetic seal to preserve electrolyte
 - Instability of Ru complex



Efficiency ~12% (record)

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Organic Photovoltaics



- Reasons for:
 - Flexible cells
 - Non vacuum/cheap substrate
 - Enables organic chemistry control
- Reasons against
 - Exciton binding energy
 - Mobility of carriers
 - Stability



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High-Efficiency Multijunction Cells



LM and MM 3-Junction Cell Cross-Section







Lattice-Matched (LM) Lattice-Mismatched or Metamorphic (MM)



High-Concentration 3-Junction Cell





Concentrator cell light I-V and efficiency independently verified by J. Kiehl, T. Moriarty, K. Emery – NREL



Metamorphic (MM) 3-Junction Solar Cell





• Metamorphic growth of upper two subcells, GalnAs and GalnP







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Metamorphic (MM) 3-Junction Solar Cell





 Metamorphic GaInAs and GaInP subcells bring band gap combination closer to theoretical optimum



Record 40.7%-Efficient Concentrator Solar Cell



Concentrator cell light I-V and efficiency independently verified by J. Kiehl, T. Moriarty, K. Emery – NREL R. R. King, 51st Electronic Materials Conf., University Park, PA, June 24-26, 2009

Inverted Metamorphic (IMM) 3-Junction Cell



Bottom ~1-eV GaInAs subcell is inverted and metamorphic (IMM)

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• Upper two GaInAs and GaInP subcells are inverted and lattice matched (ILM)

Inverted Metamorphic (IMM) *Second* 3-Junction Cell

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 Raising band gap of bottom cell from 0.67 for Ge to ~1.0 eV for IMM GaInAs raises theoretical 3J cell efficiency



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4-Junction Lattice-Matched Cell





- Current density in spectrum above Ge cell 4 is divided 3 ways among GalnAs, AlGa(In)As, GalnP cells
- •Lower current and I²R resistive power loss



0.67-eV Ge cell 4 and substrate

4-Junction Cell



 Lowering band gap of subcells 2 and 3, e.g., with MM materials, gives higher theoretical 4J cell efficiency

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Light I-V Curves Record Efficiency Cells

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 Light I-V curves for 3-junction upright MM (40.7%), 3J lattice-matched (40.1), 3J cell designed for >1000 suns (39.1%), and 4J LM cell (35.7%)



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Wafer bonding for multijunction solar cells

Low band gap cells for MJ cells using high-quality, lattice-matched materials

 Epitaxial exfoliation and substrate removal

 Formation of latticeengineered substrate for later MJ cell growth

 Bonding of high-band-gap and low-band-gap cells after growth

 Electrical conductance of semiconductor-bonded interface

Surface effects for semiconductor-to-semiconductor bonding







Larry Kazmerski, NREL







Concentrator Photovoltaic (CPV) Systems and Economics



Concentrator PV Systems with Multijunction Cells



- •1 football field of ~ 17% solar cells at 1-sun produces ~ 500 kW.
- By using MJ cells (> 35%) at concentration of 500 suns, same power is produced from smaller semiconductor area (or the football field produces 500 MW).



Combination of high efficiency & 500X concentration boosts output per semiconductor area by a factor of 1000.

MJ cells are replaced by less expensive optics and common materials.

Leads to reduced cost of energy despite paying extra for tracking & cooling.



- Solar cells = Semiconductor converting light into electricity
- Receiver = A collection of one or more solar cells mounted on substrates
- Module = Receiver + optics to concentrate the light
- BOS = Balance of System (everything else needed, e.g., tracker, inverter)



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III-V MJ cells give
56% measured
improvement in
module efficiency
relative to Si
concentrator cells

Courtesy of Solar Systems Pty. Ltd., Australia

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Balance of System Costs



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Economics for Device Physicists



Continuity equation:

$$\frac{\partial \rho}{\partial t} = qG - qR - \nabla \cdot J$$

...in \$\$ rather than charge carriers:



Terrestrial PV System Cost



R. R. King et al., 3rd Int'l. Conf. on Solar Concentrators (ICSC-3), Scottsdale, AZ, May 2005

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Summary



- Urgent global need to address carbon emission, climate change, and energy security concerns \rightarrow renewable electric power can help
- Electronic materials research has a huge opportunity to impact these challenges, by developing new materials, devices, processes to reach:
 - 45-50% efficient single-crystal multijunction cells
 - 25% polycrystalline thin-film PV
 - better fuel cells, batteries, power and other renewable energy devices
- Theoretical solar conversion efficiency
 - Examination of built-in assumptions points out opportunities to reach higher terrestrial PV efficiency
 - Theo. solar cell η > 70%, practical η > 50% achievable
- Metamorphic multijunction cells have begun to realize their promise
 - 40.7% metamorphic GaInP/ GaInAs/ Ge 3J cells demonstrated
 - First solar cells of any type to reach over 40% efficiency
 - Metamorphic 3J cells now over 41% efficiency
- Solar cells with efficiencies in this range can transform the way we generate most of our electricity, and make the PV market explode