



Eidgenössische Technische Hochschule Zürich Swiss Federal Institute of Technology Zurich

Thin Film Physics Group ETH Zürich





Cover page:

Top: Flexible $Cu(In,Ga)Se_2$ solar cell on a polyimide sheet. Total thickness of the cells and substrate is 15 µm. Record efficiencies of 14.1% and 11.0% have been achieved for flexible $Cu(In,Ga)Se_2$ and CdTe solar cells, respectively.

Middle: Partial view of a monolithic staring photovoltaic PbTe-on-Si infrared focal plane array (IR-FPA) with $5.5 \,\mu m$ cut-off wavelength

Bottom: Spectral response of a RCED (Resonant Cavity Enhanced Detector).

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Academic guests:

Kaia Ernits (Tallinn University of Technology, Estonia) Christopher Hibberd (Loughborough University, UK)

General

The Thin-Film Physics group was with the Laboratory for Solid State Physics, ETHZ (head: Prof. Dr. H.-R. Ott), from October 2000 until September 2005, and is with the group of Prof. Dr. D. Pescia, also Laboratory for Solid State Physics, ETHZ since October 2005.

Before, the group was with the Institute of Quantum Electronics ETHZ (head: Prof. Dr. H. Melchior, retired in October 2000), and before 1997, the group was part of the organisation AFIF (Arbeitsgemeinschaft für industrielle Forschung) located at ETHZ. The group is financed exclusively by projects («Drittmittel»).

Sponsors

Armasuisse ETH European Space Agency Gebert-Rüf-Stiftung Swiss National Science Foundation Swiss Federal Office for Education and Science (BBW, for EU-projects) Swiss Commission for Technology and Innovation (KTI) Swiss Federal Office of Energy (BFE) Industries

Project Cooperation

Antec GmbH, Germany Arsenco, Altdorf, Switzerland Central Solar Energy Laboratory, Sofia, Bulgaria CIEMAT, Spain EKIPS, Norman, OA, USA EMPA, Dübendorf, Switzerland **ENSCP**, France EPFL, Lausanne, Switzerland HMI, Germany IAP, ETH Zürich INM, Germany INSAMET, San Sebastian, Spain Ioffe Physical-Technical Institute, St. Petersburg, Russia IQE, ETH Zürich ISOVOLTA, Austria Institute of Semiconductor Physics, Minsk, Belarus Lebedev Institute, Moscow, Russia South Bank University, UK St. Petersburg State Technical University, Russia Solaronix, Aubonne, Switzerland Tallinn University of Technology, Estonia Uni Stuttgart, Germany Uni Parma, Italy Uni Ghent, Belgium Uni Durham, UK Uni Montpellier, France Würth Solar, Germany ZSW, Germany

Research Activities

Science and technology of compound semiconductors

- Growth of molecular beam epitaxial (MBE) and polycrystalline layers of II-VI, IV-VI, I-III-VI₂, and III-V binary and multinary compounds. Applications for optoelectronic devices. Growth kinetics of heterostructures, superlattices and nano-structures (quantum dots). Phase formation and their identification.
- Structural properties of thin films, surfaces and interfaces. Crystallographic and microstructural defects. Lattice vibrational properties of semiconductors. Measurement and modelling of strain relaxation in thin films. Kinetics of dislocation-glide and -reactions in IV-VI-on-Si epitaxial layers. Recrystallization in semiconductors.
- Optical and electrical properties of thin films and heterostructures. In- and exsitu doping in semiconductors, electronic defects and transport properties.
- Growth, properties and applications of transparent conducting oxides (ZnO, ITO, FTO).
- Growth and properties of permeation barrier layers ("flexible glass" on plastics).
- Thin film growth processes like molecular beam epitaxy, e-beam evaporation, d.c. and r.f. sputtering, chemical bath deposition, electro-deposition, etc.

Infrared sensors and emitters on silicon substrates

- MBE growth of narrow gap IV-VI layers (lead chalcogenides) on Si-substrates.
- Fabrication of 1-d and 2-d IR sensor arrays; the Si-substrate may contain integrated read-out circuits.
- Development of microlithographic patterning techniques. Applications include thermal imaging and IR-spectrometry.
- Optically pumped edge emitting IV-VI lasers with 3-5 μ m emission wavelength, with cleaved or etched cavity mirrors.
- Optically pumped microcavity wavelength converter from 870 nm to 3-5 μ m
- Broad-band high reflectivity Bragg reflectors on flat and curved substrates
- Resonant Cavity Enhanced Detectors (RCEDs) with high quantum efficiency and narrow line width

Compound semiconductor thin film solar cells

- Solar cells based on Cu(In,Ga)Se₂ and CdTe (these materials yield stable and very high efficiency solar cells for economical production of solar electricity). Development of material technologies, fabrication processes, novel materials and processes for improved performance, and advanced tandem devices. Interface and transport properties of heterojunctions.
- Studies of basic material properties and heterostructures for large area and industrial production. Stability and reliability of devices. Terrestrial and space applications of lightweight and flexible thin film solar cells.

Some highlights

- Development of the first RCED (Resonant Cavity Enhanced Photodetector) for 4 to 9 µm center wavelength, exhibiting less than 1% width and above 60% quantum efficiency
- Development of 1- and 2-dimensional infrared sensor arrays for thermal imaging in epitaxial PbTe on Si-substrates which contain active circuits
- Development of optically pumped IV-VI lasers on Si-substrates with cleaved or etched cavity mirrors
- Development of optically pumped IV-VI microcavity wavelength converters on Si-substrates
- Flexible CdTe solar cells on polymers have been developed for the first time, and a record efficiency (11%) is achieved
- Long term stable and low resistance quasi-ohmic contacts on CdTe that yield 11% efficiency solar cells
- Lightweight and flexible Cu(In,Ga)Se₂ solar cells on polymer with a world record efficiency of 14.1%
- Electronic and structural comparison of Cu(In,Ga)Se₂ substrate and superstrate solar cells, 11% efficient superstrate solar cells have been obtained
- A low temperature deposition process for Cu(In,Ga)Se₂ solar cells; cells with 14% efficiency were achieved
- Highly transparent and conducting ZnO:Al layers with high deposition rates by RF magnetron sputtering

Awards

- Dominik Rudmann: Young Scientist Award (E-MRS 2004 Spring Meeting, Strasbourg, France, May 24-28, 2004)
- Daniel Abou-Ras: Graduate Student Gold Award (MRS 2005 Spring Meeting, San Francisco, USA, March 28-April 1, 2005)

Equipment

- 4 MBE-chambers with solid sources for CaF₂, Pb(Eu,Sn,Sr)Se, PbEuTe, Cu(In,Ga)Se₂, and CdTe
- 6 PVD for sample sizes up to $20 \times 20 \text{ cm}^2$, thermal and e-beam evaporation
- 3 sputtering systems, DC and RF (substrate size up tp 30x30 cm²)
- Complete photolithographic processing equipment
- Profilometer
- Light microscopy, SEM, XRD
- Electrooptic characterization for infrared sensors/emitters and solar cells
- Room temperature and low temperature Atomic Force Microscopy (AFM)
- Chemo-mechanical polishing

Self assembled PbSe quantum dots on PbTe/Si(111) with near equal sizes

Self-assembled PbSe quantum dots (QD) were grown on a few μ m thick PbTe(111) layers on Si(111). The QD form as pyramids with three (100) side faces and (111) base, i.e. with a very high aspect ratio. After overgrowth with 4 ML, about 100 dots μ m⁻² form with heights of about 16nm. The heights of a large number of dots were determined with AFM. Since all dots exhibit the same shape, their volumes are determined by one parameter (e.g. the heights) only. The observed distribution of these heights (and therefore the volumes of the dots) is extremely narrow: The standard deviation of the heights as measured by AFM can be as low as 2%. This seems to be the narrowest distributions ever observed for self assembled quantum dots.

Similar PbSe quantum dots with pyramidal shapes have first been described by Pinczolits et al. (Applied Phys. Lett. 73, 1998, p. 250). These authors used cleaved BaF_2 as a substrate. They observed two types of dots and with somewhat larger size distributions, however.

We explained the results on the basis of nucleation and growth theories.



Figures: AFM micrograph and size distributions of pyramidal quantum dots with 15 nm heights and size non-uniformities as low as 2%.

Reference: K. Alchalabi, D. Zimin, H. Zogg, Phys. Rev. Lett. 90(2), 26104 (2003)

Sponsor: Swiss National Science Foundation

Monolithic heteroepitaxial PbTe-on-Si infrared focal plane array with 96 x 128 pixels

A two-dimensional infrared focal plane array in a heteroepitaxial narrow gap semiconductor layer has been realized for the first time on a Si substrate containing the read-out electronics, and thermal images are demonstrated.

CMOS technology was used to fabricate the circuitries in the Si-substrate. Pitch is 75 μ m, each pixel contains a bare Si-area where epitaxial growth of the narrow gap layer occurs, and an access transistor. Addressing is performed line-by-line with a shift-register integrated on the chip.

The infrared-sensitive layer (PbTe for the 3-5 μ m wavelength range) is grown by molecular beam epitaxy at temperatures below 450°C, allowing fully processed and tested Si chips to be employed. Individual pixels are obtained by mesa-etching, and photovoltaic sensors are fabricated with standard photolithographic techniques.

Within the >97% operational pixels, high quantum efficiencies and differential resistances at zero bias with 4 M Ω mean value at 95K are observed. These values are much above the background noise limit for room temperature radiation.







Figures: Top: Schematic cross section, Bottom left: Part of the 2-d array, Bottom right: Demonstrational thermal image at 95K of the 96 x 128 PbTe-on-Si array with 5 µm cutoff wavelength

Reference: H. Zogg, K. Alchalabi, D. Zimin, K. Kellermann, IEEE Trans. Electron Devices ED50, 209 (2003).

Optically pumped IV-VI edge emitting IR-lasers

With the advent of compact high-power laser diodes with around 900 nm or 1500 nm emission wavelength, low-cost optically pumped lead chalcogenide IR-emitters for wavelengths above 3 μ m become attractive. In addition, the low Auger recombination of the lead chalcogenides is highly advantageous. DH (double heterostructure) and QW (quantum well) structures are grown by MBE on Si substrates. The structures typically consist of a bottom Pb_{1-x}Eu_xZ (Z = Se or Te) cladding layer, the active Pb₁_yEu_yZ layer containing PbSe QWs, and a top EuZ cladding layer which is transparent to the incoming 870 nm laser beam. If grown on (100) oriented Si-substrates, the roughly 2-3 μ m thick layers are lifted-off from the substrate by dissolving a previously grown BaF2 buffer layer, cleaved into pieces of e.g. 2000 x 300 μ m size and clamped between a heat-conducting substrate and a glass cover. When grown on Si(111)-substrates, edge mirrors are etched using dry processing.

The light of the 870 nm laser diode (typically up to 10W pulsed) is focused onto the structure and the lasing characteristics are determined. Up to now, we observed lasing up to about 250K with this limited power and without a lateral confinement.

These lasers exhibit high characteristic temperatures T_o up to 125K, although dislocation densities are as high as 10^8 cm⁻². We expect room-temperature operation if layers of higher quality are used.



Figures: Structure of an optically pumped edge emitting IV-VI IR-laser (top), the light-in-light-out characteristics (bottom) and a typical spectrum (right).

Reference: K. Kellermann, D. Zimin, K. Alchalabi, P. Gasser, N.A. Pikhtin, H. Zogg, J. Appl. Phys. 94 (11), 7053 (2003).

Vertical emitting optically pumped IV-VI IR-microcavity

A "wavelength transformer" downconverting part of the incoming 870 nm light to about 4 μ m wavelength was realized. It has a similar structure as a VCSEL (vertical cavity surface emitting laser), but with much lower reflectivity top and bottom mirrors: A $\lambda/2$ PbEuSe active layer containing PbSe QW (quantum wells) at the antinodes of the wave pattern is sandwiched between the two Bragg mirrors and illuminated from the top with a 870 nm III-V laser diode. The top Bragg mirror consists of e.g. 2 pairs of EuSe/BaF₂ which are transparent to the incoming light, the bottom mirror of e.g. 1 pair PbEuSe/BaF₂. Reflectivities are between 85% and 95%. The device works at RT in the subthreshold region. Its line width is given by the resonator design, about 6% for the present application for low-cost gas sensing of gases like CO₂, CO, CH₄. The layers are grown by MBE on Si(111) or BaF₂(111) substrates, the substrates are transparent to the output beam and also act as a mechanical protection.



Figures: Schematic structure of an optically pumped microcavity mid-IR-source and emission characteristics.

Reference: K. Kellermann, D. Zimin, K. Alchalabi, P. Gasser, H. Zogg, Physica E 20(3-4), 536 (2004).

Resonant Cavity Enhanced Photodetectors (RCEDs)

For multispectral infrared imaging and infrared spectroscopy infrared photodetectors are needed which are sensitive in a very narrow wavelength range only, but with high efficiencies. The preferred realization of such detectors is to use the Resonant Cavity Enhanced Detector (RCED) principle.

High efficiency resonant cavity enhanced photodetectors for the mid-wavelength infrared range from ~ 4 μ m up to more than 8 μ m have been realized for the first time worldwide. Spectral linewidths as narrow as 0.07 μ m (1.7%) at 4.13 μ m and 0.24 μ m (2.9%) at 8.41 μ m center wavelength, respectively, have been achieved. Peak quantum efficiencies are up to above 50 %.

A RCED consists of a Fabry-Pérot cavity defined by two mirrors and a thin photodetector which is placed in this cavity. In the cavity positive and negative interference occurs and standing waves are formed. The detector is primarily sensitive only at wavelengths at which positive interference leads to a high intensity in the absorber. The resonance wavelength is adjusted by the design of the detector.

With $Pb_{1-x}Eu_xSe$ and $Pb_{1-y}Sn_ySe$ detector materials a broad wavelength range from $< 3 \mu m$ to $> 10 \mu m$ is covered.

Simulation tools for RCEDs and other multilayer systems have been developed. These programs allow the calculation of light intensities and absorption at every position of the structure, the spectral response of any RCED and transmission and reflection of any multilayer system.



Fig. 1: Cross section of a IV-VI RCED for a design wavelength of 4.13 μm .

Fig. 2: Response of a RCED with a design wavelength of 4.13 μ m: experiment and simulation.

Reference: M. Arnold, D. Zimin, H. Zogg, Appl. Phys. Lett. 87, 141102 (2005)

Broad-Band High Reflectivity Infrared Bragg Reflectors

For laser applications high reflectivity reflectors are needed with reflectivities above 99.5 %. This cannot be reached with metal mirrors. Therefore Bragg reflectors are used. Bragg reflectors consist of alternating layers of two materials with a different refractive index, each layer with an optical thickness of a quarter of a wavelength $(\lambda/4)$.

We have realized broad-band Bragg reflectors made of alternating layers of PbSe and EuSe. These strucures show interesting features:

- High contrast in refractive index: EuSe: n = 2.3, PbSe: n = 5.0
- Reflectivity of 99.87 % with only 4.5 λ /4 pairs
- Broad stop band (Reflectivity > 99 % from $4.1\mu m$ up to $6.5 \mu m$)
- Grown epitaxially on Silicon
- Up to 3" (76 mm) diameter

To measure the absolute reflectivity of these mirrors a resonant cavity method is applied. Mechanically controlled by piezo positioners, a Fabry-Pérot cavity is formed. The optical finesse of this structure depends on the reflectivity of the mirrors. The finesse and therefore the reflectivity are derived from the transmission spectrum.



Fig. 1: TEM cross section of a 4.5 $\lambda/4$ pairs Bragg reflector.



Fig. 2: Reflectivity of a 4.5 $\lambda/4$ pairs Bragg reflector: Experimental (solid line), simulation (dotted line) and by the resonant cavity method (solid dots).

Reference: Diplomarbeit Ferdinand Felder, TU Wien, 2005

Towards the development of flexible Cu(In,Ga)Se₂ solar cells on polymer foils with efficiencies exceeding 15%

Development of Cu(In,Ga)Se₂ (called CIGS) solar cells on polymers is challenging because of the thermo-physical properties of layers and substrates. CIGS layers of suitable structural and opto-electronic properties should be grown at low temperature ($< 500^{\circ}$ C) as polyimides tend to degrade at higher deposition temperatures. Additionally, a method for controlled incorporation of an optimum amount of Na in CIGS is needed for high-efficiency cells since polyimides do not contain Na. We have applied a low temperature (450° C) CIGS deposition process by evaporation of elemental Cu, In, Ga and Se on commercially available polyimide films and Na from a NaF film was incorporated into CIGS layers with a post-deposition diffusion method that is suitable for in-line production of solar cells. With this method we were able to develop high-efficiency flexible CIGS solar cells.

Independent measurements at Fraunhofer ISE Freiburg, Germany, have confirmed 14.1% efficiency under AM1.5 standard test conditions. This is the highest efficiency reported to date for any kind of solar cell grown on polymer films. An average reflectance loss of about 13% was measured for these cells. Application of a commonly used anti-reflection coating would enable more than 15% efficiency flexible CIGS solar cells on polyimide foils.







Figures:



Top right: Current-voltage characteristics of a CIGS solar cell on a polyimide foil. The efficiency of 14.1 % under AM1.5 standard test conditions presents a world record for solar cells on polymer substrates.

Bottom: External quantum efficiency and reflectance of the front contact (ZnO:Al) surface. An average reflectance of about 13 % was measured.



Collaboration: Inst. f. Physikalische Elektronik, Univ. Stuttgart, Germany Sponsor: BBW (EU-Project), BFE

Flexible and lightweight Cu(In,Ga)Se₂ solar cells on aluminum foils

Cu(In,Ga)Se₂ (CIGS) solar cells on metal and polymer foils offer several advantages: They are flexible, lightweight and can be manufactured with roll-to-roll deposition processes. The roll-to-roll production is potentially low cost, while lightweight and flexible solar modules are attractive for a large variety of terrestrial and space applications. High efficiency solar cells on polymer and metal foils such as steel, Mo and Ti have already been developed. Aluminum is an interesting material because of low cost and weight, and it is used in several applications, especially in buildings. We have developed CIGS solar cells on coated aluminum foil from Akzo-Nobel.

CIGS layers of suitable structural and opto-electronic properties should be grown at low deposition temperatures (< 450°C), because of the difference in the thermophysical properties of layers and substrates and to avoid impurity diffusion from the substrate into the absorber layer (see figure). We have grown CIGS layers by evaporation of Cu, In, Ga, Se elements at different substrate temperatures and investigated the properties of the evaporated layers by different methods (SEM, SIMS, EDX). Due to the 3-stage process and low substrate temperature a strong band-gap grading was observed. The photovoltaic properties of small area solar cells were characterized by I-V and quantum efficiency measurements. An efficiency of 6.6% under simulated AM1.5 standard test conditions has been achieved. This is the first time that CIGS solar cells have been successfully grown on aluminum foils. Further improvements are expected with Na incorporation and by using an appropriate intermediate layer.



Figures:

Left: Current-voltage characteristics of a CIGS solar cell on Al foil under AM1.5 standard test conditions. No Na was incorporated and no AR coating applied. This is the highest efficiency reported to date.

Right: SIMS depth profile of CIGS/Mo layers in solar cells grown on Al foils at 500°C. Diffusion of Al through the Mo back contact and contamination of CIGS. The Ga-dip/In-hill is due to the 3-stage process and low In-Ga interdiffusion at low temperature.

References: D. Brémaud, D. Rudmann, M. Kälin, G. Bilger, H. Zogg, A. Tiwari, Pr Proc. 20th European Photovoltaic Solar Energy Conference, Barcelona, Spain, 2005.

Collaboration: Inst. f. Physikalische Elektronik, Univ. Stuttgart, Germany Sponsor: Swiss Federal Office of Energy (BFE)

Na incorporation into low-temperature-grown CIGS

In order to achieve high conversion efficiencies with Cu(In,Ga)Se₂-based solar cells, a small amount of sodium needs to be incorporated into the Cu(In,Ga)Se₂ (CIGS) absorber layer. Typical Na concentrations are on the order of 0.1 at.%, and traditionally the Na is allowed to diffuse from a soda-lime glass substrate into the absorber during CIGS growth. Hence, Na is present *during* CIGS growth, which is widely believed to have beneficial consequences on structural and electronic properties of the CIGS film. Therefore, on Na-free substrates such as polymers or metals, the deposition of a Na containing precursor film (e.g. NaF) is widely used in order to supply Na to the absorber during its growth. However, polymer substrates restrict deposition temperatures to below 500 °C, a temperature regime where the influences of Na on CIGS properties have received little attention.

We have developed a post-deposition treatment (PDT), where Na is diffused into asgrown CIGS absorbers. With the PDT, electronic properties of the CIGS films are still dramatically enhanced in a way typical for "traditional" Na effects. However, with the PDT, there cannot be an influence of Na on the CIGS growth kinetics, which indicates that the most relevant Na effects are less related to structural than to electronic absorber modifications. It is therefore likely that Na acts mainly by passivation of defects at CIGS grain boundaries.

We have compared the performances of solar cells processed with Na diffusing from the glass substrate during growth with those of cells with PDT Na incorporation. Thus, we obtain an indication of the significance of Na-induced modifications of CIGS growth kinetics for cell efficiency. When the absorbers were grown at low substrate temperatures (below 500 °C), we found that absorbers with PDT-Na performed better. This is attributed to an impeding influence of Na on CIGS phase formation, which became most apparent when the growth temperature was 370 °C. With higher substrate temperatures (around or above 500 °C), the performance of cells grown in the presence of Na became similar or superior, which might indicate a beneficial influence of Na on CIGS growth once a certain growth temperature is exceeded. With post-deposition Na incorporation, a cell efficiency of 13.8 % was achieved at 400 °C substrate temperature.



Figures: J–V characteristics of CIGS cells with Na incorporated using the PDT (no Na present during CIGS growth; solid lines) and by diffusion from the substrate (Na influences CIGS growth kinetics; dotted lines). The absorbers were grown at different substrate temperatures.

References: D. Rudmann, D. Brémaud, A. F. da Cunha, G. Bilger, A. Strohm, M. Kaelin, H. Zogg, A. N. Tiwari, Thin Solid Films 480-481, 55 (2005).
D. Rudmann, A. F. da Cunha, M. Kaelin, F. Kurdesau, H. Zogg, A. N. Tiwari, G. Bilger, Appl. Phys. Lett. 84, 1129 (2004).
Sponsors: BBW (EU projects), Swiss National Science Foundation, BFE, ETH

Band-gap grading in Cu(In,Ga)Se₂ solar cells

Flexible Cu(In,Ga)Se₂-based solar cells on polymer foils require low-temperature growth processes. Absorbers grown with the 3-stage process were used to prepare record cells with 14.1 % efficiency on polyimide foils. An apparent feature of such absorbers is a pronounced band gap grading, which is a consequence of the film deposition sequence and the lack of effective In–Ga interdiffusion at low substrate temperatures. The Ga/In concentration ratio, and therewith also the band gap, varies through the absorbers.

The influence of this band-gap grading on cell performance is not known. Therefore, we have grown absorbers with single-stage processes and have compared the effects of correspondingly graded and of ungraded band-gap profiles. No microstructural differences are apparent from SEM cross-section images. Cells prepared from graded absorbers at best match the performance of ungraded cells. However, more samples with varying grading are required to confirm these conclusions. Absorbers grown with the 3-stage process exhibit clearly larger grain sizes (see figure) and cells are superior to single-stage-grown cells. This may originate primarily from the different microstructure as a result of the different growth kinetics.



Figures: SEM cross-section images (P. Wägli, ETH) of $Cu(In,Ga)Se_2$ solar cells. Absorber (a) was grown with a 3-stage process and absorber (b) with a single-stage process.

References: D. Rudmann, D. Brémaud, M. Kaelin, H. Zogg and A. N. Tiwari, Proc. 20th European Photovoltaic Solar Energy Conference, Barcelona, Spain, 2005.
D. Rudmann, D. Brémaud, A. F. da Cunha, G. Bilger, A. Strohm, M. Kaelin, H. Zogg and A. N. Tiwari, Thin Solid Films 480-481, 55 (2005).

Sponsors: BBW (EU projects), Swiss National Science Foundation, BFE, ETH

Collaboration: Inst. f. Physikalische Elektronik, Univ. Stuttgart, Germany (G. Bilger, A. Strohm) Dep. de Física, Universidade de Aveiro, Portugal (A.F. da Cunha)

30x30cm² Cu(In,Ga)Se₂ Solar Cells Development on Glass and **Polyimide Substrates**

As a first step towards industrial production of thin film Cu(In,Ga)Se₂ solar cells, upscaling of all the processing steps to 30x30 cm² substrates is required. A new deposition reactor with 30 cm line sources and automatic substrate movement has been set up and is optimized for large area homogenous CIGS deposition. Further to that, front and back-contact layers from transparent conducting oxides or metals, as well as thin buffer layers of CdS or ZnS must also be applied homogenously on large substrate sizes. Special attention is focused on ultra-flexible plastic substrates which limit deposition temperatures to a maximum of ~400 °C. Therefore the deposition processes are optimized for low-temperature processing.



a)

b)



c)

d)

- a) $30x30 \text{ cm}^2$ CIGS deposition reactor. Figures: b) Evaporation line sources for the co-evaporation of CIGS. c) Chemical bath deposited buffer layer on $30x30 \text{ cm}^2$ glass substrates. d) Flexible solar cells rolled on a tube.
- Sponsor: Swiss Federal Office of Energy (BFE)

TiO₂/Cu(In,Ga)S₂ Nanocomposite Layers For Low-Cost Solar Cells

The nanocystalline dye-sensitized (or Grätzel-) solar cell has attracted a lot of interest since its breakthrough in the early 90's. Its great potential for low cost production by using inexpensive materials and processing techniques has resulted in many working prototypes which have the following concept: A thin porous layer of sintered TiO_2 nanocrystals, which are covered with a monolayer of light-absorbing dye, serves as photoactive electrode. This layer is typically deposited on SnO_2 :F coated glass where the remaining pores are filled with a redox electrolyte to close the electric circuit. The sealing of the cells against air, moisture and evaporation of the solvent are major issues for module lifetime improvement.

As an alternative, completely inorganic solid state Grätzel cell structures, also known as extremely thin absorber (ETA-) cells or 3D cells, have been proposed. In these cells an n- and p-type semiconductors form an interpenetrating network on nano-scale (Figure 1). Light absorption always takes place in near proximity of the p-n junction. This allows using low-quality absorber materials as bulk recombination in the absorber material is less important. Cu(In,Ga)S₂ (CIGS) being a very stable material and an optimal light absorber is a good candidate for the p-type absorber material. Due to the liquid state of our non-vacuum process precursors (dissolved Cu, In and Ga salts were used to prepare CIGS layers with solar cell efficiencies up to 6.7 %), infiltration of the pores between the TiO₂ nanocrystals is possible. The precursor solutions are deposited by doctor blade and converted in selenium vapor to form the interpenetrating CIGS/TiO₂ network.



Figure: Schematic drawing showing the $TiO_2/CIGS$ nanocomposite layer within the solar cell structure.

Preliminary results of solar cells showed reasonable V_{OC} values of over 400mV but low current densities. The infiltration may not have occurred completely. Also, the CIGS compound was directly grown on the TiO₂ nanoparticles. Substantial improvements of the photovoltaic characteristics are expected by applying ultrathin recombination barrier coatings prior to CIGS infiltration

Collaboration: Solaronix SA, Aubonne, Switzerland

Non-vacuum Transparent Conducting Oxide and Buffer Layer Deposition with Ultrasonic Nebulizer

Transparent Conducting Oxide (TCO) materials such as tin-doped indium oxide (ITO) and aluminium-doped zinc oxide (ZnO:Al) are interesting materials for use as front contacts in CIGS solar cells due to their good electric conductivity and good transparency to visible light. The sheet resistance of a good TCO layer is between 10 and 50 Ω /square and its optical transparency is greater than 90%. Indium sulphide (In₂S₃) is a promising material for use as a replacement buffer-layer in CIGS solar cells, avoiding the use of CdS. The band gap energy value of a buffer layer should be higher than 2 eV.

An ultrasonic spray system was used for growing TCO and In_2S_3 thin films. In the spraying system an alcoholic solution containing precursor-salts is nebulized by an ultrasonic actuator and then sprayed onto a heated substrate (Fig. 1). The challenge is to reduce the substrate temperature and the spray time as the absorber and the buffer layers degrade at temperatures above 250°C, due to diffusion process at the interface.

The sheet resistance achieved from a sprayed ITO (temp. 300° C) layer was 35 Ω /square, with transparency 91.6 % and thickness 0.24 µm. The minimum sheet resistance measured from a sprayed ZnO:Al (temp. 350° C) layer was 7400 Ω /square. this is still too high for using the layer as a TCO.

However, for CIGS solar cells with sprayed In_2S_3 (temp. 300°C) maximum efficiencies of 5.4 % were obtained. Efficiencies for the reference cell (with CBD CdS) were around 8.2 % (Fig. 2). The band gap values for In_2S_3 were 2.5–2.8 eV compared with 2.5 eV for CdS.



Fig. 1: Ultrasonic spray setup.



Fig. 2: I-V curves for solar cells with CBD

Microstructural and Chemical Study of Interfaces in Cu(In,Ga)Se₂ (CIGS) Thin-Film Solar Cells

Cu(In,Ga)Se₂ (CIGS) solar cells are produced by thin-film deposition and consist of a substrate (e.g. soda-lime glass), a back contact (e.g. Mo), the CIGS absorber, a buffer layer (e.g. CdS) and a window layer (e.g. i-ZnO/ZnO:Al); see Fig. 1. Interfaces play an important role in determining the solar-cell efficiency. Microstructural and chemical properties of different interfaces have been studied by means of bright-field (BF-TEM), high-resolution (HR-TEM) and energy-filtered transmission electron microscopy (EF-TEM), and also by selected-area electron diffraction (SAED), energy-dispersive X-ray spectroscopy (EDX) and scanning electron microscopy (SEM).

One topic is the study of interfaces between CIGS and In_xS_y buffer layers. Solar-cell efficiencies deteriorate considerably when the In_xS_y deposition temperature exceeds about 250 °C, independent of the In_xS_y deposition technique. The HR-TEM image in Fig. 2 shows an In_xS_y /CIGS interface where the In_xS_y layer was deposited at 300 °C by evaporation of In_2S_3 powder. A large density of planar defects is visible in the buffer layer. Copious Cu diffusion from CIGS into the buffer was found by EDX. SAED revealed that CuIn₅S₈ formed instead of In_xS_y . CuIn₅S₈ formation has been detected at various In_xS_y /CIGS interfaces. Its spinel-type crystal structure contains a large number of vacancies and the planar defects shown in Fig. 2, which may act as recombination centres at the p-n heterojunction of the solar cell and thus be responsible for the poor photovoltaic performance.

Further research topics are the study of Zn(O,S)/CIGS and CdS/CIGS interfaces, as well as the study of $MoSe_2$ for application as buffer layer between CIGS and any metal/semimetal back contact.



Fig. 1: TEM image of a ZnO/ buffer/ CIGS/Mo cross-section.

Fig. 2: HR-TEM image of an $In_xS_y/CIGS$ Interface. The In_xS_y layer was deposited at 300 °C.

Reference: D. Abou-Ras, D. Rudmann, G. Kostorz, S. Spiering, M. Powalla, A. N. Tiwari, J. Appl. Phys. 97(8), 084908 (2005)

Collaboration: Prof. Kostorz, Institute of Applied Physics (IAP), ETH Zurich.

Sponsors: BBW (EU projects); Swiss National Science Foundation

High efficiency flexible CdTe/CdS solar cells

Development of flexible and lightweight solar cells is interesting for many terrestrial and space applications that require a very high specific power (defined as the ratio of output electrical power to the solar module weight).

We have presented a novel process for developing CdTe solar cells on polyimide film substrates in superstrate configuration. The polyimide layer was spin coated on glass prior to the deposition of ZnO:Al, and the semiconducting layers are grown at low temperatures, around 400°C. Further development of the transparent conducting oxide (TCO) resulted in a record efficiency of 11% for CdTe flexible cell with Voc = 842 mV, Isc = 18.5 mA/cm2, and FF = 70.9%. For space applications it is desirable to avoid polyimides because of their possible degradation under UV light.

A new process for preparing substrate flexible CdTe solar cells has been developed to avoid these drawbacks. A layer of NaCl is deposited on glass prior to the deposition of TCO. Polyimide is spin coated on top of the layer so that it will still hold the solar cell but it will not be exposed to radiation.

After the deposition of the stacks, a lift-off process is applied. Solar cells of 7.3% efficiency with Voc = 692 mV, Isc = 21.5 mA/cm2, and FF = 49% were obtained.



Figures:

Top left: CdTe superstrate soloar mini module on polyimide

Top right: I-V characteristic of a 11% efficiency CdTe solar cell on polyimide (superstrate configuration) measured under AM 1.5 illumination.

Bottom: Schematic cross-section of the flexible CdTe solar cell on polyimide in superstrate configuration.

Stability of CdTe solar cells for terrestrial and space applications

CdTe/CdS solar cells have a promising potential for space application due to their high specific power (kW/kg) on thin and flexible substrates and excellent radiation hardness. We investigated the radiation tolerance of CdTe/CdS cells developed in our laboratory, by irradiation with protons (at the TANDEM accelerator of PSI/ EHTZ) and electrons (at the DYNAMITRON electron accelerator of the University of Stuttgart). The cells were irradiated with very high fluences of the particles of different energies in the MeV range, equivalent to the exposures of decades in space. The displacement damage dose formulation was applied to determine one single degradation characteristics as a function of damage dose, that contains the data of all particles, energies, and fluences. With this formulation it was possible to compare the CdTe solar cell technology in terms of radiation hardness to other technologies (see figure). Comparison of solar cell efficiency degradation curves clearly shows that CdTe is the most radiation tolerant solar cell.

While the efficiency of high efficiency Si and III-V solar cells drops to ~50% of the initial value, CdTe cells remain stable for a realistic dose in space (~1011MeV/g). The CdTe cells also recover fast from any damage, which implies that for operation in space no performance degradation due to radiation damage is expected. The stability of CdTe solar cells is still an issue and depends crucially on the impurities in the devices. SIMS measurements indicate impurity diffusion from the back contact material into the CdTe bulk and the CdS with accumulation in the CdS layer or near the CdTe/CdS interface. To correlate these observations of changed chemistry in the cell with the photovoltaic properties, i.e. general degradation, the technique of voltage dependent quantum efficiency measurement (referred to as Apparent Quantum Efficiency, AQE) was developed and applied to thermally stressed cells. In order to understand and interpret the AQE measurements a novel model was developed. The measurements indicate diffusion of Cu and Au into the CdTe cell resulting in degradation, whereas Sb and Mo may diffuse only in much smaller quantities and degrade the PV properties much less. Stable CdTe cells with Sb/Mo and Sb₂Te₃/Mo back contacts have been demonstrated.



Figure: Comparison of radiation hardness of different solar cell technologies. CdTe is the most radiation tolerant solar cell.

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- Guest co-editor: Thin Solid Films vol. 480-481 (2005) journal published by Elsevier Ltd.
- Guest co-editor: Thin Solid Films (to be published in 2006) journal published by Elsevier Ltd.
- Co-chair/Co-organiser of the European Materials Society-2004 Symposium on Chalcogenide Materials for Photovoltaics.
- Scientific advisory committee member of the European Photovoltaic and Solar Energy Conferences in 2004, 2005,.
- Co-chair/Co-organiser of the European Materials Society-2006 Symposium on Chalcogenide Materials for Photovoltaics.

Publications

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Marc Kälin

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Dmitry Zimin Growth and properties of optoelectronic structures based on IV-VI materials Diss ETH Nr. 15733, 2004 Examiner: Prof. Dr. G. Kostorz Co-examiners: Prof. Dr. M. Tacke, PD Dr. H. Zogg

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*** Paul Scherrer Institute, c/o ETH Zürich, Institute for Particle Physics, Switzerland

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* Dep. da Física, Univ. de Aveiro, Portugal

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