TAILORING CHEMICAL AND OPTICAL PROPERTIES OF 2D TRANSITION METAL DICHALCOGENIDES

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Single-layer of molybdenum disulfide (MoS_2) and other transition metal dichalcogenides (TMDC) appear to be promising materials for next generation applications (optoelectronic and catalysis), because of their low-dimensionality and intrinsic direct band-gap which typically lies in the visible spectrum. Several experimental groups have already reported novel electronic and transport properties which place these materials beyond graphene for device applications. MoS_2 is also known to be a leading hydrodesulfurization catalyst. Efforts are underway to further tune these optoelectronic and catalytic properties through alloying, defects, doping, coupling to a substrate, and formation of bilayer stacks (homo- and hetero-structures). In this talk I will present results from joint theoretical and experimental investigations [1-3] which provide a framework for manipulating the functionality of this wundermaterial and take us closer to the goal of rational material design. With emphasis on the chemical properties of defect-laden single layer MoS_2, I will examine modulations in its local atomic environment (see Fig. 1 for an example) under which it could serve as a catalyst for the conversion of synthetic gas (CO and hydrogen) to methanol. With regard to the optical properties, I will present results of our analysis of the excitation spectrum and the ultrafast charge dynamics in both single- and bi-layer TMDCs obtained through the application of combined time-dependent density functional theory and many-body theory approach. In particular, I will show how the reduced electron screening in these systems leads to surprising large binding energy of electronic bound states [4] - excitons (hundreds of meVs), trions (tens of meVs) and biexcitons, in rather good agreement with available experimental data. I will also show that ultrafast (10-100fs) transfer processes are possible in these materials as a result of strongly-delocalized hole orbitals. Possible technological applications will be discussed.

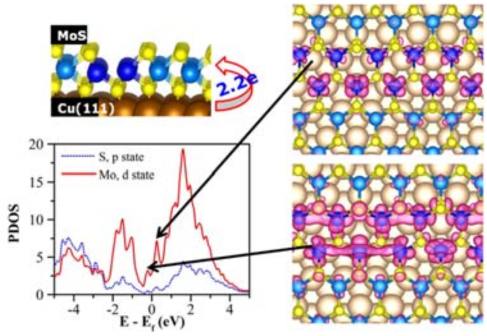

Fig. 1. Single layer MoS_2 grown on Cu(111)

Single-layer molydenum disulfide: transition metal dichalcogenide; exciton; catalysis

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A PERSPECTIVE OF QUASIPARTICLE STATE IN ORGANIC CRYSTALS

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Understanding the impacts of strong electron-phonon coupling as well as weak electronic interaction on the electronic state is required to discuss the rich of functionality in organic molecular materials. Angle-resolved ultraviolet photoelectron spectroscopy (ARUPS) is known to be a powerful technique to study the electronic states. However, the experimental study of fine features in the highest occupied molecular orbital (HOMO) state has not been progressed till recently due to difficulty in the sample preparation, damages upon irradiation and so on, though it can offer a wide variety of key information, that is essential to comprehend charge-hopping transport and small-polaron related transport in the ordered monolayer film [1] as well as to coherent band transport in the molecular single crystal [2,3]. We present recent findings regarding on the precise measurements of electronic states for large aromatic organic molecular materials by using high-resolution ARUPS. A quasiparticle state is appeared differently in the UPS spectrum depending on the strength of the intermolecular interaction which is confirmed by the width of the energy band.

Keywords: rubrene, electron-phonon coupling, small polaron, photoelectron spectroscopy

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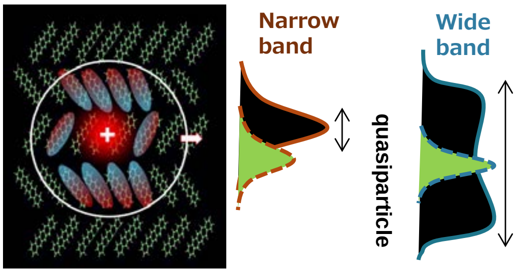

Fig. 1. HOMO-band features found as a polaron formation in organic crystals

SURFACE RESONANT RAMAN SCATTERING FROM THE BARE AND THE (2x1)-O-RECONSTRUCTED Cu(110) SURFACE

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In recent years Raman spectroscopy has been successfully employed to study surface vibrational modes of semiconductors via surface resonant Raman scattering. Metals like Cu, Ag, Au, on the other hand, which are important substrates for Surface Enhanced Raman scattering from rough surfaces, are not expected to show Raman signals, due to their crystal structure. As it is known from inelastic electron and He atom scattering the highly anisotropic Cu(110) surface exhibits a characteristic set of vibrational modes. Therefore, it is of particular interest that our surface Raman experiments show that the symmetry breaking on the metal surface enables indeed inelastic light scattering from surface phonon modes. This makes possible new high spectral resolution measurements of the same set of surface vibrational modes and a determination of their intrinsic lifetimes. Comparison of polarization resolved measurements with calculations of the surface vibrational modes provides indications to the enhancement mechanism responsible for the pronounced Raman scattering cross sections and the differences to particle scattering. Oxygen adsorption and the (2x1)O reconstruction of the Cu(110) surface gives rise to a distinct set of surface vibrational modes observable in Raman experiments. The coupling between the Cu(110) surface and the Cu-O chains in the topmost layer can be further clarified by comparison to detailed calculations.

Keywords: Surface Raman Spectroscopy, surface phonons, surface structure

Energy Level Control at ZnO/Organic Semiconductor Interfaces

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Hybrid inorganic organic systems (HIOS) are promising candidates for future (opto-)electronic devices by taking advantage of the complementary beneficial properties of two different material classes. However, inadequate interfacial energy level alignment is an intrinsic obstacle to superior device function. Hence, to design efficient HIOS devices, understanding and controlling HIOS energy level alignment is a key factor. Employing molecular electron donor or acceptor interlayers to tune the work function (ϕ) of a metal and thus the energy level alignment relative to an organic semiconductor (OSC) was reported previously [1].

In this contribution the concept is extended to HIOS interfaces comprising ZnO. The ϕ and energy level alignment modification caused by deposition of several donor and acceptor molecules onto different ZnO faces is investigated. ϕ reductions down to 2.2 eV by using the organometallic donor $[\text{RuCp*mes}]_2$ [2] and ϕ increases up to 6.4 eV using perfluorinated tetracyanoquinodimethane (F4TCNQ) [3] are demonstrated. For molecular acceptor adsorption, significant adsorption induced upward band bending of up to 0.9 eV is found within ZnO, whereas for donor adsorption no downward bending could be induced. Exploiting the huge ϕ modifications of ZnO, upward and downward HIOS energy level readjustments are shown, which realize ultimately low electron- or hole-injection barriers at the HIOS interface. Moreover, by using an OSC whose gap matches that of ZnO, the energy level offsets at the HIOS interface could be eliminated. This enables highly efficient, non-quenched energy transfer across the HIOS interface, which is usually inhibited by unfavorable energy level alignment.

Keywords: ZnO, energy level alignment, energy transfer

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Investigation of electronic structure at cathode interfaces in Subphthalocyanine acceptor based organic solar cells

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Subphthalocyanine (SubPc) acceptor based organic solar cells (OSCs) exhibited superior device efficiency of over 8% [1]. However, the energy loss at the cathode/organic hetero-interfaces, e.g., formation of energy barrier, has not been clarified yet. In this study, we investigated the electronic structures at SubPc/buffer/Ag heterointerfaces by means of synchrotron based ultraviolet photoelectron spectroscopy (UPS).

The SubPc/buffer/Ag heterostructures were formed by depositing buffer materials on Ag and subsequently depositing SubPc onto buffer/Ag stack structure in a step-by-step way in a vacuum deposition chamber. A series of pyridine based acceptor molecules (BCP, TPBi and TAZ) were applied as the buffer layers.

For all buffer/Ag stack structures, metal induced gap states within the HOMO-LUMO gap were observed. These states were located near the LUMO levels and their density of states reached to the Fermi level. Thus, the gap states act as shallow donors, which enhance the electron conductivity of the buffer layers. Since the work function of the buffer/Ag stack structures showed fairly small values (3.3-3.5 eV), which corresponds to the electron affinity of the SubPc layers (~3.3 eV), the buffer layers have a role of promoting a smooth carrier injection from cathodes to acceptor layers. Nevertheless, the energy difference between LUMO of SubPc and Fermi level of buffer is estimated to be ~0.6 eV. The carrier injection barrier of the system was quite huge as compared with that of C_{60}/buffer/Ag heterostructure (~0.2 eV). The difference in the barrier height might be caused by the density of disorder originated tail states in the acceptor layers and/or acceptor/buffer heterointerfaces [2] since structural disordering is easily introduced in non-planar aromatic systems (SubPc). We consider the disorder originated tail states, which were located at the deep energy position, caused the carrier injection barrier at the SubPc/buffer/Ag heterostructures.

Keywords: energy level alignment; organic solar cells; ultraviolet photoemission spectroscopy

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THE ENERGY LEVELS IN HOLE-DOPED MOLECULAR SEMICONDUCTORS

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Understanding the nature of charge carriers in organic thin films is crucial in order to achieve rational design in opto-electronic organic devices. In the present work, we rely on interface doping phenomena, as occurring when contacting an organic semiconductor with a high work function (modified) electrode, to study the electronic properties of a hole-doped molecular thin film [1,2]. The investigated system, consisting of a C_{60} film adsorbed Au(111) pre-covered with a thin MoO_3 layer, is studied by direct and inverse photoemission. A detailed analysis of the photoemission spectra, supported by theoretical calculations based on density functional theory, helps to precisely identify the corresponding density of states of the neutral and positively charged C_{60} coexisting at the surface.

The obtained results reveal a picture of the electronic structure which strongly differs from the traditionally conceived one, i.e. with the energy levels consisting of singly occupied molecular states lying within the energy gap of the semiconductor (Fig. 1. a) [3]. Instead, here we demonstrate that the on-site Coulomb repulsion splits the partially unoccupied frontier molecular level in the p-doped systems into two sub-levels (Fig. 1. b) [1]. The role of inter-site Coulomb interaction between molecular ions and neighbor molecules is further addressed and allows providing a complete picture of the energy levels of molecular semiconductors in the presence of excess charges [1].

Keywords: Organic semiconductors; photoemission; electronic structure; hole-doping

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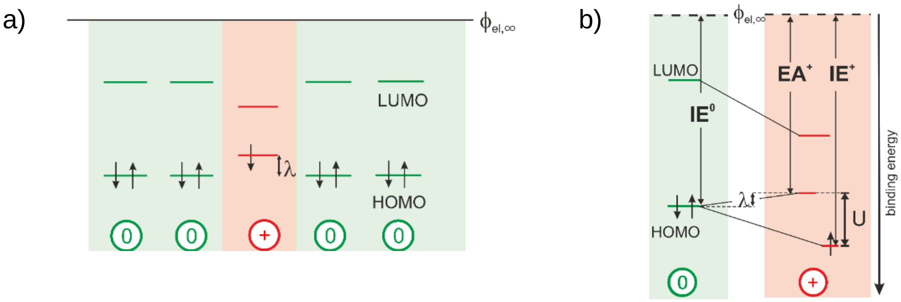

Fig. 1. a) Traditionally envisioned energy levels of positive polarons (red) embedded in a neutral organic matrix (green). b) Effect of the on-site Coulomb repulsion on the frontier energy levels upon hole doping as determined by (inverse) photoemisson.

Adsorption of organic molecules on transparent semiconducting substrates.

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The adsorption of organic molecules on semiconducting substrates is relevant to many applications from energy generation to drug delivery. Surface analysis using combined methods provides the most complete picture of surface structure, chemistry and electronic states, especially when a combination of methods is applied in-situ and in real time.

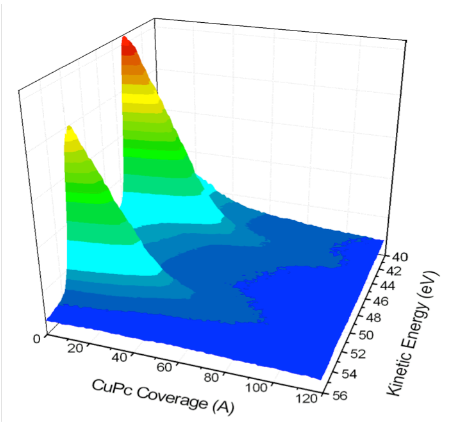

Fig. 1. Real-time monitoring of the growth of a CuPc film on a transparent oxide substrate.

Transparent oxides such as indium tin oxide (ITO) are almost exclusively used as substrates in photovoltaic (PV) devices for solar energy, usually as thin films on glass substrates. However, there is a growing interest in alternative and specialist substrates, for example UV absorbing materials for organic PV (e.g. ZnO) and ionizing radiation resistant substrates (e.g. diamond).

We have applied in-situ surface analysis to the growth of small organic molecules on several substrates to optimize the substrate, the thin film growth and the interface energetics[1]. Rapid screening and improved accuracy is achieved by data collection in real time as shown in Fig. 1 for CuPc growth on an ITO substrate. The attenuation of the substrate core level peaks provides the thin film growth mode (layer-by-layer in this case) while the energy position quantifies changes in band-bending and surface conductivity. Lineshape analysis reveals chemical changes and the optical gap of the organic semiconductor[2]. The complete interface energetics are provided by the excitation of core and bonding electrons using a range of sources from optical to UV to x-ray. For ZnO, this approach has enabled the high work function and 2-d conduction of the (0001) surface to be preserved at the inorganic-organic interface.

Keywords: Interfaces; Organic semiconductors; photovoltaics

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PURE AND DISORDERED GROUP III NITRIDE HETEROSTRUCTURES: GROWTH AND PHYSICAL PROPERTIES

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MBE growth of GaN and its alloys has advanced to the level where its is now the technique that has demonstrated the highest quality group III nitride layers and heterostructures. In this presentation we review our work on three main topics: (i) the realization of high electron mobility and high hole mobility bulk GaN by NH_3 MBE [1]. The work on p-GaN demonstrates the importance of *reduced* growth temperature to avoid natural compensation by native donors [2]. (ii) For lateral electron devices, we demonstrate the realization of pure AlN interlayers (via atom probe tomography) by plasma-assisted MBE for AlGaN/AlN/GaN and InAlN/AlN/GaN high electron mobility transistors [3]. The pure AlN interlayer allows the realization of record low sheet resistance 2DEGs. (iii) We demonstrate the surprising role that natural alloy fluctuations play in the vertical transport through nominal alloy heterobarriers [4,5]. In all of these studies, MBE has played an essential role in understanding the physical properties of the group III nitrides.

Keywords: MBE; Gallium Nitride; Transport

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TOWARDS AUTOMATED ANALYSIS OF X-RAY RECIPROCAL SPACE MAPS FOR THE CHARACTERIZATION OF GAN BASED HETEROSTRUCTURES

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Research efforts continue on the development of GaN based band gap engineered materials which find applications in HEMTs or LEDs. These devices contain epitaxial layered structures of increasing complexity. X-ray diffraction is one of the basic analytical methods that is routinely utilized for both materials research and quality control in production. X-ray scattering methods are non-destructive and deliver very detailed structural information on epitaxial layers and substrates including strain, strain relief, film thicknesses, crystalline quality and mosaic spread.

Maps around reciprocal lattice spots (RSM) can reveal additional information beyond that provided by single line scans such as high-resolution rocking curves. RSMs are typically used to aid the interpretation of peak displacement, peak broadening or peak overlap. However, they are considered too time consuming to collect and therefore are not frequently used.

Recent advances in X-ray area detector technology combined with smart positioning algorithms and data processing allow now RSMs to be recorded in similar timescales as rocking curves. These high-speed measurements find applications in the characterization in all crystalline advanced materials. Position sensitive detectors allow collecting many points at once and can be used in continuous mode, so less measurement and positioning steps are necessary to build up a given area of reciprocal space.

This contribution will discuss opportunities and limitations of this technique in conjunction with a feasibility study about automatic evaluation of RSM data from GaN based heterostructures. A fully automated workflow enables new possibilities of X-ray metrology in even high-throughput production environments.

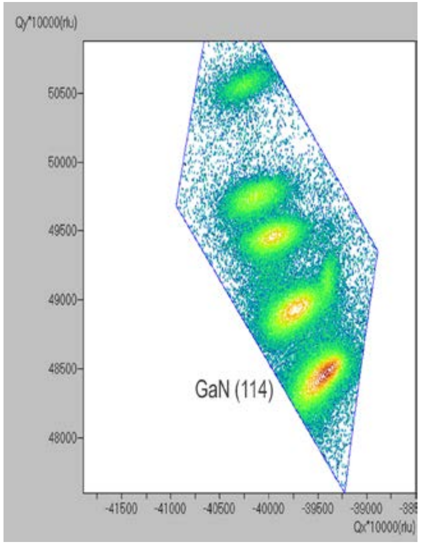

Fig. 1 Ultra-fast RSM collected in 30 seconds on a AlGaN/ GaN HEMT structure on Si (111)

Keywords: High-resolution X-ray diffraction; Reciprocal space map; GaN

TAILORING SINGLE-DOMAIN Si(100) SUBSTRATES FOR GaP GROWTH BY Ga-DRIVEN TRANSFORMATIONS OF SURFACE STEPS AND TERRACES

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For GaP-on-Si(100) heteroepitaxy, currently considered as a model system for monolithic integration of III-V semiconductors on Si(100), the surface steps of Si(100) have a major impact on the quality of the GaP film. Monoatomic steps cause antiphase domains in GaP with detrimental electrical properties. A viable route is to grow the III-V epilayer on single-domain Si(100) with biatomic steps, but preferably not at the expense of reduced terrace widths introduced by miscut substrates.

We have performed in situ investigations of the influence of Ga deposition on the surface terrace kinetics of Si(100) at elevated substrate temperatures by low-energy electron microscopy (LEEM). Starting from nearly equally distributed T_A- and T_B-terraces of a two-domain Si(100) surface, submonolayer deposition of Ga resulted in a transformation into a surface with prevailing T_A-terraces (Figure 1). By increasing deposition rate or decreasing temperature, we induced restructuring of Si(100) into a surface dominated by T_B-terraces as previously reported by Hara et al. [1]. The occurrence and mutual transformations of surface structures with different terrace and step structures in a narrow range of temperatures and Ga deposition rates are discussed (Figure 2).

The evolution of these surface structure transformations proceeds via three stages: (i) fluctuations of vicinal monoatomic steps, (ii) local merging of adjacent fluctuating steps and formation of a local biatomic step acting as nuclei for (iii) biatomic step growth along the direction of the steps (Figure 3).

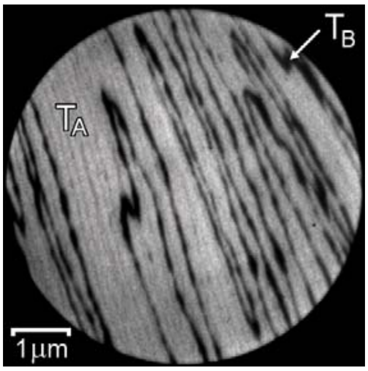

Figure 1. Dark-field LEEM image of a 90:10 distribution of T_A and T_B terraces on well-oriented Si(100) after 40 minutes of Ga exposure at 660 °C.

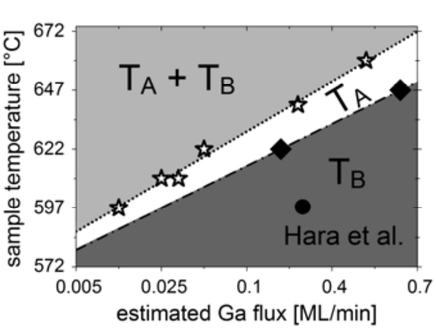

Figure 2. Existence regions for domain structures of Si(100) as a function of temperature and Ga flux. Stars and diamonds denote parameters for transformations into T_A- and T_B-dominance, respectively.

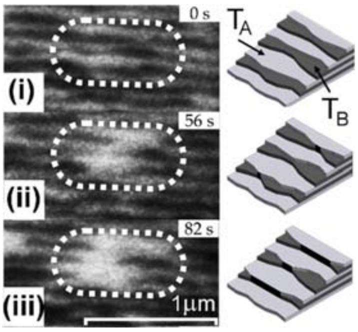

Figure 3. Ga-driven formation of biatomic steps and a local T_A domain.

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Surface processes during InGaN quantum well growth

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The active region of optoelectronic devices in the visible range based on group-III-nitrides is conventionally built from InGaN/GaN quantum well (QW) structures. For the performance of such devices control over the indium incorporation is mandatory. For long wavelengths emitters or solar cells layers with high indium concentrations ($x_{In} > 0.2$) are required. The more indium is to be incorporated the more such layers suffer from various effects such as high strain, defect formation and poor surface morphology as a consequence of the required low growth temperatures T_G. While at high T_G and lower values of $x_{In} < 0.2$ indium incorporation is limited by desorption, the growth process at lower temperatures is much more complex: Diffusion/cluster formation and desorption compete with indium incorporation. However, at least at low T_G indium incorporation takes place from a liquid-like adlayer [1,2]. The detailed structure of the adlayer and details of the interplay between indium incorporation - adlayer and adlayer formation - desorption are not yet understood.

To gain more insight into the indium incorporation process and the role as well as the properties of the adlayer we performed in-situ optical reflectivity experiments during MOCVD growth. Growth took place in a commercial Aixtron Aix200rf system with horizontal reactor using standard precursors. The system is equipped with a Laytec optical sensor which provides data on the curvature, the reflectivity at 633.2nm/950nm and the growth temperature by pyrometry.

We find evidence for indium on the surface during the time the precursor Trimethylindium (TMI) is supplied for low temperatures of about 970K (measured by a thermocouple). At these conditions the incoming flux is larger or equal to the desorption rate. The data reveal indium desorption and at least another loss mechanisms and possibly some reconfiguration of the surface (coverage). Furthermore, another source of indium is present delivering indium when TMI is already switched off.

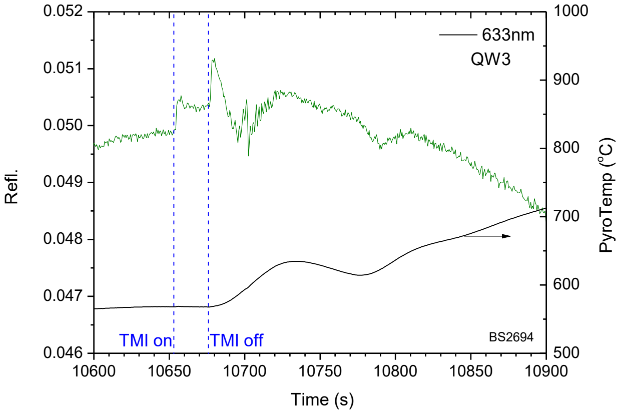

Fig. 1. Reflectivity transient taken during growth of the third QW in a 5xQW structure.

Keywords: Group-III-nitrides, reflectivity transient, adlayer

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BLACK SILICON AND ITS APPLICATION IN SOLAR CELLS

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Solar cell structure need low reflectance surface to maximize number of absorbed incident photons. Usually antireflection (AR) coatings are used in the solar cell industry. Their utilization is limited because they reduce reflection for narrow range of the light wavelengths. Jansen et al. [1] firstly demonst[illegible] replacement of conventional AR coating by black silicon layer. Nanocrystallin[illegible] Si is formed by electrochemical reaction utilizing catalytic mesh. The [illegible]ical solution penetrates through the space region of the nanocrystallin[illegible]sequently, the nanocrystalline Si layer grows. In this co[illegible]sent i) photoluminescence (PL) properties of black silico[illegible]mples (without RCA treatment) measured at 6K and [illegible]ar cell structures prepared on two black silicon sa[illegible]d on the BS sample with overlayer thickness of [illegible] reference samples. All samples were prepared o[illegible] samples with following different thickness of BS o[illegible] production of the corresponding solar cells: ~ 60 n[illegible] the photon energy region 1.3 eV – 2.5 eV [illegible] were not received on [illegible] reference sample after [illegible] RCA cleaning. On [illegible] other side ref[illegible] sample after [illegible] procedures, [illegible] generated [illegible] the ab[illegible] photon[illegible] Figure [illegible] signal of the [illegible] sample and [illegible] polished c-Si [illegible] recorded at ~ 6K in photon energy region 1.3 eV – 2.5 eV.

Keywords: Black silicon; Photoluminescence; Solar cell

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DEPTH-DEPENDENT STUDIES OF ELECTRON AND PHONON ULTRAFAST DYNAMICS IN FEMTOSECOND LASER INDUCED TRANSIENT STATES OF MATTER

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Ultra-fast laser pump-probe interactions with dielectric materials far from equilibrium provide a unique approach to both characterize and non-thermally modify these material systems [1, 2]. For example, short-pulse laser induced coherent acoustic phonons (CAP) provide a moving surface, indeed a moving mirror, which, when used in a simple reflection probe mode, has been shown to reveal strain, defect concentrations, and impurity concentrations as a function of depth with unparalleled sensitivity and depth resolution [3, 4]. This has opened exciting new avenues of exploration in the fields of nanoscale materials characterization and manipulation. To extend this approach, the realization that the CAP wave does indeed constitute an actual mo[illegible] surface, suggests that surface physics techniques that have been routinely appli[illegible]aracterize normal surfaces and interfaces may be applied to the moving CAP s[illegible]

In this presentation, we will discuss ongoing research [illegible]damentals of coherent acoustic phonon spectroscopy, (b) semicon[illegible]centration profiles measured using coherent acoustic phonon[illegible] ultrafast carrier and phonon dynamics of graphene on di[illegible]cilitating ultrafast laser induced low-temperature d[illegible]emes for accomplishing depth dependent materials [illegible]t states of matter.

Keywords: Coherent Acoustic Phonons; Pho[illegible]aphene; Second Harmonic Generation

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