DESIGNING SPIN PROPERTIES IN TOPOLOGICAL, POLAR, AND MULTIFERROIC THIN FILMS

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The growth of high quality thin films has since long been known to be an effective method to control or enhance the electronic structure of materials. Here it will be shown that similar considerations can be applied to the spin texture of special classes of materials. For example, in topological insulator thin films the energy position and spatial localization of the spin polarized interface states can be controlled by the adequate choice of substrate and thickness [1].

Furthermore it will be shown that the highly polar surface of the transition metal oxide $SrTiO_3$(001) hosts a 2D electron gas that exhibits the spin signature of a Rashba-type spin splitting [2]. The properties of this state can be tuned by doping of thin films and transforms into a 2D hole gas for ferroelectric $BaTiO_3$ films.

The previous examples take place at the surface or interface, but the spin degeneracy can also be lifted in the bulk of a thin film if the crystal structure lacks inversion symmetry. In this respect ferroelectric materials bear large promise as in this case the inversion symmetry breaking can be controlled by an external electric field. In the polarized state of ferroelectric GeTe films the electronic structure consists of a plethora of spin-polarized bulk and surface states, whereby the latter can be suppressed by a capping layer [3].

In this work we will take this concept one step further and induce ferromagnetic order in ferroelectric GeTe by Mn-doping. By using a combination of spectroscopic techniques it will be shown that the films are both ferroelectric and ferromagnetic at temperatures below 130K and that the chiral spin texture remains even for Mn concentrations around 20%. Furthermore, our spin-resolved ARPES and soft X-ray ARPES measurements show that the ferromagnetic order induces a Zeeman gap and out-of-plane spin texture around the Dirac point [4]. By switching the magnetization it could be determined that the ferroelectric and ferromagnetic order are directly entangled. This opens up new pathways for the use of non-volatile spintronics applications and electronically switchable magnetic substrates.

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PROPERTIES OF METALLIC THIN FILMS GROWN ON PERIODICALLY CORRUGATED SURFACES

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Nanostructured thin films are of growing relevance for all kind of applications in photovoltaics, plasmonics, or as magnetic materials. Various methods have been used to fabricate nanostructured thin films with well defined morphology exhibiting tunable effective properties. Bottom-up, self-organized methods have been used extensively in the last years because of their fast and easy way of producing large-scale patterns with structures down to 10 nm.

Ion beam sputtering has proven to be a promising way to produce self-organized patterns on various surfaces [1]. Depending on the ion beam incidence angle, hexagonally ordered dot patterns as well as ripple patterns oriented perpendicular or parallel to the ion beam direction are formed during the continuous sputtering. Periodically corrugated surfaces can also be obtained via crystal surface reconstruction during annealing. The resulting surfaces provide templates for the growth of nanopatterned thin films. Depending on the surface and interface free energies these films can grow in a conformal way reproducing the surface topography or as nanoparticles on the substrate surface. Furthermore, depending on deposition angle, substrate temperature, beam flux, and deposition time, the nanoparticles can align parallel to the ripples, eventually coalescing and forming nanowires, thus tuning the physical properties of these structures via their geometrical dimensions.

Metal thin films grown in this way exhibit distinct optical properties due to localized surface plasmon resonance. Due to their alignment along the ripple structures the nanoparticles exhibit strongly anisotropic plasmonic resonances [2]. Furthermore, the magnetic properties of ferromagnetic thin films grown on rippled or faceted substrates are drastically changed by the presence of the periodic structures at the interface and on the surface [3].

Keywords: metallic thin films; magnetic properties; optical properties; surface plasmon resonance

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NANO-SURFACES : SURFACES OF NANOPARTICLES

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Surface Science is usually focused on planar, atomically flat surfaces, which are model systems far from common material specimens. A model system of other kind for surface exploration is the surface of (preferredly spherical nanoparticles (*Nanos*), which is more realistic in the variety of more or less imperfect surface structures, depending on special material choices and particle preparation methods.

They cover a broad field from planar facettes with edges, spikes and corners to disordered or non-crystalline boundaries without any Cartesian symmetry, etc
Such surfaces may be transferred into interfaces (or more precisely: three-dimensional “interlayers”) by foreign embedding media, which may be chosen from inert gases to passive or reactive liquids or to metallic, non-metallic or organic solids,

Their thickness may extend from sub-monoatomic layers to bulk-like surroundings. Those layers exhibit advantages compared to planar systems, since they may cover the interface completely with high uniformity and without any disturbing substrates.

Nanotechnology now-a-days allow the investigation of single such nanos..

Important advantages of this latter research route are extraordinary application sensitivities of many kinds of experiments., mainly due to the huge surface to inner volume ratio of nanos by which surface properties are scaled up. .

However, draw-backs are still to-day problems with sample preparation and characterization, and not all published experiments overcome them convincingly.

Among the most sensitive experiments are plasmonic surface investigations, which, in addition, give insight into the unique nano-size effects observed in the transition region between solid state nanos and molecular ones.
(General properties of surfaces can, however, only be extracted as long as an individual surface / interface can be clearly defined.)

Recent investigations have shown that in plasmonic excitations all electrons both of the inner particle volume and of the whole surface are involved stemming mainly from the conduction energy band but –to lesser extent- also from deeper lying bands and can be separated from each other with help of extensions of the fundamental work of Mie.

Since Mie’s basic paper, concerning spherical surface plasmon polaritons (SPP’s), it was usual to disregard separate surface material properties and to apply instead dielectric functions averaged over the whole particle. To-day we know that material properties of surface regions of nanos may differ essentially from those of the inner volume. Hence, to bring Mie’s description of the SPP’s into quantitative correspondence with experimental results, we require correction - and extension – effect-models concerning special nano-structure - and nano-surface – effects,. which have to be taken into account in the evaluation of the experimental data. .
Most efficient are various extension - effects to describe the observed strong surface induced relaxation and decoherence effects. One important example, the *Chemical Interface Damping* of the SPP’s , will be treated in this contribution to some detail.

It is the purpose of the present paper to compile several such nano-surface corrections which are useful to obtain more precise informations about nano-surfaces and ,moreover, about realistic surfaces in general.

General Reference:

Uwe Kreibig, Michael Vollmer : *Optical Properties of Metal Clusters /* Springer, Berlin 1995

HIERARCHICAL SELF-ASSEMBLY: THREE STEPS TO HIGHLY ORDERED ARRAYS OF UNIFORM METAL NANOSTRUCTURES

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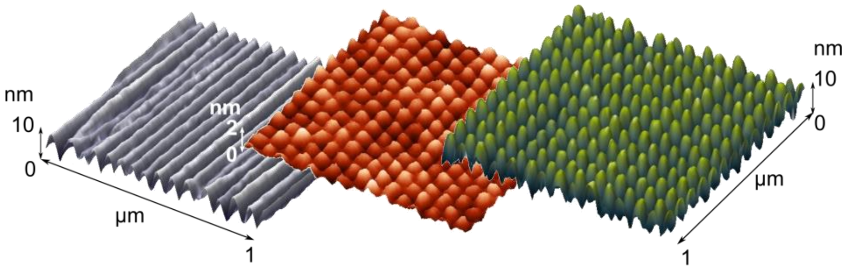
Nanopatterning via self-assembly has gained considerable interest as an alternative to lithography-based techniques for nanostructure fabrication. We propose a procedure for producing highly ordered arrays of uniform metallic nanostructures based exclusively on three subsequent self-assembly processes [1]: crystal surface reconstruction, copolymer microphase separation, and metal diffusion on chemically heterogeneous surfaces. The versatile approach allows for preparing nanostructures with scalable sizes and in a variety of shapes and materials. With this high-throughput technique, nanopatterns covering areas of several square centimeters can be fabricated easily.

We present results of in-situ structural and magnetic investigations of Fe nanodot arrays during formation by grazing incidence small angle X-ray scattering [2] and nuclear resonant scattering of synchrotron radiation [3], examining the dependence of the nanodot shape on deposition conditions and observing the evolution of magnetic moment dynamics during nanodot growth [4]. Possible applications of self-assembled nanopatterns could range from high-density magnetic data storage to catalysis or sensing based on surface plasmon resonance.



faceted α-Al_2O_3 substrate
copolymer template
self-assembled metal nanostructure pattern

Figure 1: AFM topography micrographs of the three self-assembling systems combined in the presented procedure. In hierarchical self-assembly, the morphological structures formed in one system guide the structure formation in the following one.

Keywords: self-assembly; diblock copolymers; metal nanostructures

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SPIN-SPLIT ONE-DIMENSIONAL SURFACE STATES ON VICINAL Si SURFACES

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Nanostructures that are supposed to be used in spintronics applications have to fulfill several conditions. Among most important is spin splitting of electronic bands at the Fermi level that is large enough at room temperature to prevent mixing of electrons with the opposite spins. In the present contribution we report on the existence of one-dimensional spin-split metallic states on vicinal silicon surfaces that reveal the largest splitting reported to now at room temperature.

A perfectly regular distribution of steps on a vicinal surface is prerequisite for a good quality of (quasi) one-dimensional nanostructures. The Si(553) surface has been used for this purpose as known to form an array of ordered steps even without any adsorbates [1]. However, in this case the electronic structure does not reveal dispersive and metallic bands, beside the surface state bands similar to those observed on Si(111)-(7x7). Quite different scenario occurs for the surface with the adsorbed Au or Pb atoms. They form either double atomic chains [2] (Si(553)-Au) or nanoribbons [3] (Si(553)-Pb) on each terrace of the vicinal surface.

The presence of the Au or Pb chain-like structures introduces parabolic-like one-dimensional surface states crossing the Fermi level. Due to the strong spin-orbit coupling the bands reveal considerable spin splitting of about 0.2 eV and 0.05 Å^{-1} in case of Si(553)-Au [4] and much larger - 0.6 eV and 0.2 Å^{-1} in case of Si(553)-Pb [5] at the Fermi level. The reason for the difference is a significant anisotropic electron charge distribution around Pb nuclei in the latter case which is caused by the unusual arrangement of the Pb atoms on terraces of vicinal surface [5,6]. It introduces a strong in-plane potential gradient what increases spin splitting of the surface states and the appearance of the out-of-plane component of the polarization vector. The weak interaction between Pb and Si atoms makes the spin-split states well-decoupled from the substrate what should assure pure spin currents in future spintronic devices.

Keywords: Spintronics; vicinal Si; Rashba effect

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QUASI-1D ATOMIC-CHAINS ON VICINAL SURFACES: THE ROLE OF DEFECTS AND CONSTRICTIONS TOWARDS ELECTRONIC TRANSPORT

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Metallic atomic chains on vicinal semiconductor surfaces are prototype quasi-1D ensembles for fundamental research, such as metal to insulator phase transition or Tomonaga-Luttinger liquid behavior [1]. Despite the intense studies over the last decades on several systems, e.g. Si(111)-4x1-In, Si(557)-Au and Si(553)-Au, the effect of native defects or induced by adsorption (e.g. O_2 and H_2) on the conductance of the wires, the interwire coupling or the transition temperature, are still under debate.

Here, we present a systematic study on the transport properties of In- and Au-chains on various Si(hhk) surfaces via a multi-tip STM/SEM system. The conductivities parallel and perpendicular to the wires is quantified by the rotational four-point-probe square method [2]. Additionally, an extremely higher sensitivity towards atomic imperfections and both conductivity components was reached by a spatial constriction of the electron path [2] (Fig. 1a). In detail, the Si(111)-4x1-In system was re-analyzed with greater attention. In contrast to previous studies, we showed in agreement with DFT calculations, that the interwire coupling can be gradually tuned by O_2 adsorption [3] (Fig. 1b). Moreover, the transport properties of various Au-phases on Si(hhk) surfaces were studied. While the 0.48 ML Au on Si(553) turned out to be extremely robust and almost unaffected (up to 20 Langmuir), the 0.2 ML phase on Si(557) reacts strongly as seen by the exponential decrease and the adsorbates induce even a crossover of its conductivity components. The adsorption behavior turns out to be strongly triggered by the presence of a Si-adatom chain, which is a characteristic building block for some Au/Si(hhk) chain systems [4].

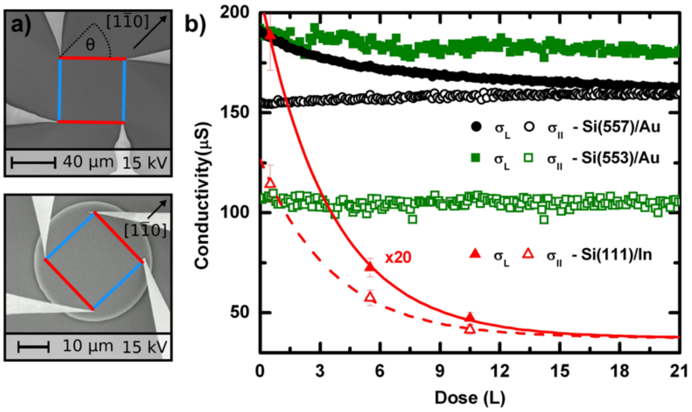

Figure 1: a) STM tips placed in square configuration on an unconfined In/Si(111) surface (above) and confined circular In/Si(111)-mesa structure (below). b) $\sigma_{||}$ and $\sigma_{\perp}$ versus O_2 dose for the Si(111)-4x1-In system, 0.48ML Au phase on Si(553) and 0.2ML Au phase on Si (557).

Keywords: Atomic-chains, metal-insulator transitions, interwire coupling, defects, lateral constrictions

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DENSITY FUNCTIONAL THEORY INVESTIGATION OF RARE EARTH SILICIDE NANOWIRES

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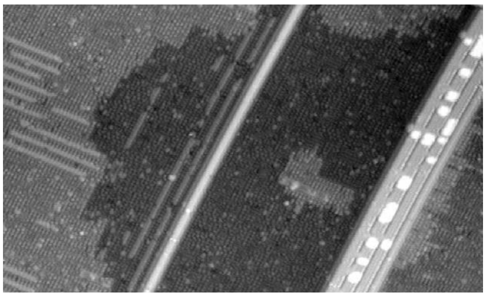

Fig. 1: STM image "showing two broad nanowires at the center and the right of the image as well as some remaining thin nanowires on the left" [1]

Quasi one-dimensional electron systems are of both fundamental interest because of their unusual physical properties as well as potentially interesting for devices on the nanometer scale. In this respect, rare earth (RE) silicide nanowires on silicon (001)-surfaces are of special interest, because the RE silicides exhibit useful physical properties such as low Schottky-barriers, thermal stability and low electrical resistivity one the one hand, and because the silicon (001)-surface is well known and very common in today's technology on the other.

One special feature of the RE silicide nanowires is their preparation, as they grow self-organized controlled by macroscopic parameters such as temperature, deposition amount, and atomic number of the RE. Under variation of these parameters, different systems of RE silicide nanowires have been studied experimentally [1].

Unfortunately, the exact knowledge of their atomic structure, which is crucial for understanding their physical properties, is still incomplete. As a first step towards understanding the nanowires structural and their electronic properties, we have investigated different wire models from first-principles within density functional theory (DFT). Thereby erbium is chosen as prototypical trivalent rare earth ion and its 4f-electrons are treated as frozen state in the atomic core.

The wire models base all on the hexagonal RE silicide structure suggested from the experiment [1]. They have been categorized corresponding to three degrees of freedom, namely width, height and depth of burial. After comparing the formation energy of different models within ab-initio thermodynamics, electronic band structures and densities of states are calculated for the stable wires. Structural and electronic properties are discussed and compared with available STM[1], STS[2] and ARPES[1] measurements.

Keywords: nanowires, rare earth, silicide, DFT

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SIZE-DEPENDENT PENETRATION OF GOLD NANOCLUSTERS THROUGH A NACL MEMBRANE

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CANCELED

Membranes and their size-selective filtering properties are universal in nature and their behavior is exploited to design artificial membranes suited for, e.g., molecule or nanoparticle filtering and separation. Exploring and understanding penetration mechanisms of nanoparticles in thin-film system[illegible]ay provide new opportunities for size selective deposition or embedding of the [illegible]cles.

Here, we demonstrate an unexpected phenomenon that nonporous atomically thin alkali halide films on a metal support can be exploited as a size-selective inorganic membrane for metal nanoparticles. In particular, relying on scanning tunneling microscopy techniques and density function[illegible] theory simulations, we find that NaCl [illegible] can filter out larger Au clusters, while [illegible] clusters are allowed to permeate [illegible] the NaCl onto the Au(111) su[illegible] can be tuned via the thickness of the NaCl film. We [illegible] of the filtering effect is the attractive energy/forc[illegible] substrate, which forces the clusters to pass throu[illegible] manner [1].

This mechanism [illegible]rmeation through any kind of barrier that separat[illegible] of attractive force. In particular, the membrane fi[illegible](111) system shows analogy to that of a living biol[illegible] lipid membrane acts as a barrier for ions and mo[illegible]tive force is governed by the osmotic pressure [illegible]onment. The here observed size-filtering mechani[illegible] understanding nanoparticle-film (membrane) interaction[illegible] nanoparticle filters.

Keywords: Metal [illegible]; penetration and transmission mechanisms; size-dependent membrane; thin nonp[illegible]alide film.

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ELECTRONIC STRUCTURE AND ELECTRON DYNAMICS IN NOVEL TWO-DIMENSIONAL MATERIALS

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Changing the dimensionality of a material results in significant modifications of its electronic properties. This is even the case if the parent material already has a layered structure with little interaction between the layers, as in the case of graphene and single-layer transition metal dichalcogenides.

Here we explore the properties of novel two-dimensional materials such as single layers of MoS_2, WS_2 and TaS_2 by scanning tunnelling microscopy angle-resolved photoemission spectroscopy (ARPES). The layers are grown epitaxially on Au(111), Ag(111) and graphene. For the semiconducting materials (MoS_2, WS_2), strong band gap renormalizations are observed due to the interaction with the substrate. For the metallic layers (TaS_2), we can study the effect of low dimensionality on electronic instabilities such as charge density waves and superconductivity.

While the static electronic properties of novel two-dimensional materials can be studied by standard ARPES, investigations of the ultrafast carrier dynamics require both time- and angular resolution and thus time-resolved (TR)-ARPES. There is, moreover, the technical requirement of high photon energies since the interesting part of the aforementioned materials' electronic structure (i.e. the (gapped) Dirac cone) is placed at the two-dimensional Brillouin zone boundary. Recently, it has become possible to probe states at such high k by TR-ARPES, thanks to the arrival of ultrafast high harmonic laser sources.

Keywords: Two-dimensional materials, electronic structure, ARPES, STM

FROM MONOLAYER TO MULTILAYER SILICENE

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Silicene has attracted a huge interest since publication of the archetype 3 × 3 reconstructed monolayer phase on a silver (111) substrate in 2012 [1]. It is a synthetic emergent two-dimensional Si-based material that might rival graphene for logic applications in electronics [2] : indeed, the first monolayer silicene device, a Field Effect Transistor with ambipolar characteristics, has been fabricated in 2015 [3].

Ordered hydrogenation of 3 × 3 silicene has been achieved recently in Beijing [4], next in our Lab in Marseille, with further High Resolution Electron Energy Loss spectroscopy results [5].

Multilayer silicene has been also synthesized ; it possesses an intrinsic $\sqrt{3} \times \sqrt{3}$ surface reconstruction, hosts Dirac fermions, and is self-protected in ambient air by its native oxide [6,7]. However, despite such evidences, its mere existence has been severely questioned.

Here, we will show thanks to novel *in situ* Photoelectron Spectroscopy data as well as *ex situ* Surface X-Ray Diffraction and Raman spectroscopy results that multilayer silicene is effectively grown in a rather low temperature regime (~200°C), while, instead, diamond-type silicon terminated by the well-known Si(111)$\sqrt{3} \times \sqrt{3}$-Ag superstructure is formed at higher temperatures.

Keywords: Silicene; 2D Phases; Structural and Electronic Characterizations

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Current Trends in Microscopic Characterization of 2D Materials with Spectroscopic Imaging Ellipsometry (SIE)

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Current developments in the field of 2D-Materials are focused on several areas. These areas include: microscopic devices based on stacking different 2D-materials into hybrid-structures, discovering new material configurations by exfoliating unconventional layered materials, optimizing CVD processes by minimizing crystallite boarders or using ideal shaped microcrystals as the starting point of a growth process and many more. These developments have in common that they require microscopic non-destructive characterization technologies.

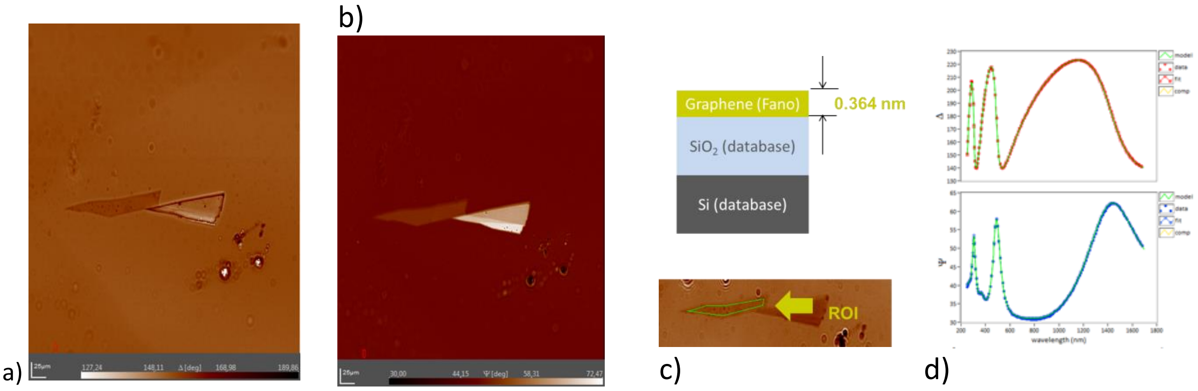

Fig. 1 Spectroscopic imaging ellipsometric measurement of a Graphene flake: Delta (a) and Psi (b) map – examples of a graphene flake, selected Region of Interest and Optical model (c) and wavelength spectra of Delta and Psi, (d) extracted for the ROI from Delta and Psi maps recorded for wavelengths in between 250 and 1700 nm (measured **data displayed as** points and model, line).

Conventional ellipsometry is well established in the field of thin film metrology due to the exceptionally high resolution in the z-axis, enabling very accurate thickness measurements for nano- and microfilms. However, conventional ellipsometry does not have a sufficient lateral resolution for a number of the described developments nor a direct microscopic visualization with highest contrast. The approach of spectroscopic imaging ellipsometry (SIE) differs from conventional ellipsometry in that the measurements are based on a series of micrographs taken at dedicated orientations of the optical components. The primary measurements are microscopic maps of the ellipsometric angles Delta and Psi (Fig. 1 a, b) or micro maps of dedicated elements of the Müller Matrix. Areas can be condensed by binning, averaging of selected pixels data (Region of interest, ROI, Fig. 1c) of equivalent areas and by using histogram data of inhomogeneous regions.

Keywords: Imaging Ellipsometry, Graphene, 2D Materials,

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CHARGE-TRANSFER BETWEEN PLASMONIC NANOPARTICLES AND SINGLE LAYER MoS_2

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Recently, intensive efforts have been made to control the electronic properties of MoS_2 monolayers by doping, including chemical doping[1], gate-bias tuning[2], and physical adsorption[3]. Plasmon-induced hot electrons generated by photo-excitation of Au nanoparticles can also lead to n-type doping of TMDC monolayers[4]. In this contribution, we report on the localized surface plasmons excitation in Au nanoparticles beneath single layer MoS_2. The subsequent changes in vibrational and optical properties of the MoS_2 monolayer were investigated via Raman spectroscopy and photoluminescence in order to evidence the effects of charge-transfer. We report for the first time the visualization as well as the intensity enhancement of the trion peak for this 2D/plasmonic system. Our findings can be effectively driven towards realizing the size and structure of the individual components of opto-electronic devices such as plasmonic field effect transistors.

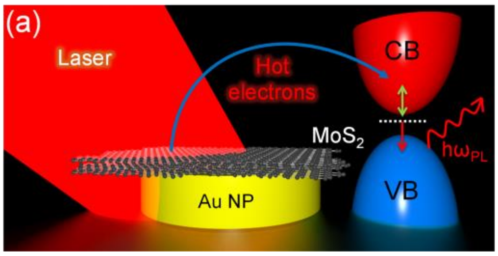

Fig. 1(a): Schematics of the interaction between the Au nanoparticle and MoS_2.

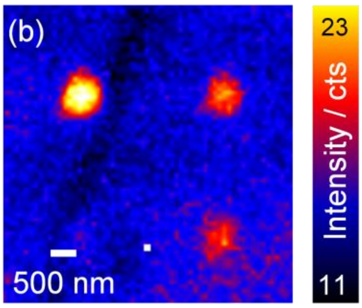

Fig. 1(b): PL intensity map of the 665 nm to 675 nm band showing higher enhancement in the MoS_2 single layer around the Au nanoparticles.

Keywords: MoS_2, plasmonics, hot electrons, two-dimensional, transition metal dichalcogenides, photoluminescence spectroscopy

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MODEL CATALYSIS WITH LIQUID ORGANIC HYDROGEN CARRIERS

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LOHCs are potential candidates for chemical hydrogen storage in future applications: The materials are high boiling organic molecules that can be reversibly hydrogenated and dehydrogenated in catalytic processes. [1] LOHCs consist of a pair of a hydrogen-lean and a hydrogen rich-molecule. The hydrogen-rich molecule is catalytically dehydrogenated at the time and place energy hydrogen/energy is needed, while the hydrogen-lean molecule is hydrogenated in times when excess energy is available.

In this contribution, two examples of the surface chemistry on dehydrogenation catalysts will be discussed, that is, the perhydro-N-ethyl-carbazole (H_{12}-NEC) [2] and dicyclohexylmethane (DCHM) [3]. DCHM and H_{12}-NEC were adsorbed by physical vapor deposition, and subsequently their reaction was monitored during heating. Although the molecules are quite complex, the individual reaction steps were identified by X-ray photoelectron spectroscopy. We will compare the behaviour of the two LOHCs in detail, concerning their dehydrogenation properties. At low temperatures, we find for both the adsorption of a monolayer, followed by formation of multilayers. Upon heating, the multilayers desorb and subsequently distinct dehydrogenation reactions occur, which will be discussed. At elevated temperatures, decomposition reactions are found; these also will be evaluated in detail, since they are detrimental to the storage cycle of the LOHCs.

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Keywords: Liquid Organic hydrogen carriers, model catalysis, photoelectron spectroscopy.

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NEAR-AMBIENT PRESSURE X-RAY PHOTOELECTRON SPECTROSCOPY OF IONIC LIQUID MULTILAYERS EXPOSED TO WATER; A FUNDAMENTAL STUDY OF GAS ABSORPTION IN IONIC LIQUIDS

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