



International Workshop

Energy Dissipation at Surfaces

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Energy Dissipation at Surfaces

3rd International Workshop
SFB 616

Bad Honnef
Aug. 25-28
2008

Ultrafast Dynamics
Ballistic Transport
Surface Reactions
Chemcurrent
Plasmonics
Friction

Impact Phenomena
Nanopotentiometry
Electromigration
Photoemission
Hot Electrons
Phonons

Deutsche Forschungsgemeinschaft
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SFB616

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Courtesy of F.-J. Meyer zu Heringdorf and S. Möllenbeck:
Surface Plasmon Polaritons in Ag Nanopillars by M. 2004

Two Photon Photoemission Microscopy Image of Surface Plasmon Polariton (SPP) waves at the Ag/vacuum interface. The dark areas in the image correspond to Ag islands that have self-assembled on a Si (111) surface. The Ag islands (dark) exhibit a periodic intensity modulation that constitutes a fingerprint of SPP waves: SPPs can be started by a $\lambda = 400$ nm femtosecond laser pulse whenever a component of the electric field of the laser pulse is perpendicular to the edge of Ag island. The SPPs then propagate across the islands in a direction perpendicular to the edge at which they were created. At the same time, the laser pulse that created the SPPs is also still present and propagates across the island as well. The superposition of the relevant electric fields while SPP and fs laser pulse travel across the surface results in a beat pattern, i.e. the periodic intensity modulation visible in the image.

(Courtesy of F.-J. Meyer zu Heringdorf and S. Möllenbeck
Reference for SPP imaging: L. Chelaru, F. Meyer zu Heringdorf, Surface Science **601**, 4541 (2007))

Monday, August 25

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9:00 **Electron Excitation and Emission for Grazing Impact of Fast Atoms on Metal and Insulator Surfaces** 32

Helmut Winter, Berlin, Germany

9:50 **Electron Dynamics at Surfaces at High Photon Energies Using the XUV Free Electron Laser (FLASH)** 34

Helmut Zacharias, Münster, Germany

10:40 Coffee

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Martin Weinelt, Berlin, Germany

12:00 Lunch

12:30 Tour

- Boat-trip on the Rhine river to Cologne with pastries and coffee
- Guided tour through Cologne and to the cathedral
- Dinner at the brewery "Sion" in the old town of Cologne

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9:00	The Excitation of Local Vibrations by an Electron Current: From Atomic Manipulation to Local Heating Nicolás Lorente, Barcelona, Spain	38
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Notes

Attosecond Electron Transport in Real Time

Adrian L. Cavalieri

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Electron transport on the attosecond timescale through several atomic layers has now been observed in single-crystal tungsten. This development was made possible by the extension of well-known attosecond spectroscopic techniques previously developed in gas-phase targets. In tungsten, an isolated attosecond XUV pulse was used to excite photoelectrons inside the crystal, while a waveform controlled, few-cycle NIR laser pulse was used to probe their precise time of emission. Photoelectrons originating from the 4f core-states were found to be emitted approximately 100 attoseconds after those from the delocalized conduction band states. These experiments, their results, and planned efforts for the future will be discussed.

Notes

Surprising Electronic Properties of Surfaces Covered with Organized Organic Layers

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A common tacit assumption in the scientific literature is that

the electronic properties of a close-packed organized organic monolayer adsorbed on solid substrate can be simply extracted based from the properties of the adsorbed single molecule.

This approach is of course very helpful when calculations are involved, since it limits the size of the system that has to be studied. The above-mentioned assumption is assumed to be verified by structural studies which usually indicate that there is no major change in the structure of the molecules upon adsorption. The weak coupling between the molecules in a monolayer seems to support this notion. This, then, is taken as a justification for using molecular-based calculations to predict the properties of the monolayer. Indeed, mostly only minor changes are observed in the vibrational spectrum of the molecules, upon formation of the close-packed monolayer.

Our recent experimental results suggest that the above –stated assumption can misguide our understanding in as far as the electronic and magnetic properties of the adsorbed layer are concerned. In the case that the length of the dipole of the molecules is larger than the distance between them, the layer can be considered as a dipole layer [1]. A dipole layer is characterized by two important phenomena:

*the field within the layer is very large, and
there is no field outside the layer. Hence, there is no image charge in the substrate.*

It will be shown that even in the case of relatively moderate dipole moment strength, minimization of the free energy requires, in this case, charge transfer between the layer and the substrate. This charge transfer is therefore a result of a cooperative effect and does not take place when a single molecule is adsorbed on the surface.

As will be shown, such charge transfer is indeed observed experimentally. It is the origin of new electric and magnetic properties [2]. Some of these new properties were investigated by us and the results will be presented.

- [1] Z. Vager and R. Naaman, Chem. Phys. **281**, 305 (2002);
Z. Vager and R. Naaman, Phys. Rev. Lett. **92**, 087205 (2004).
[2] I. Carmeli et al., Angewandte Chem. Int. Edition, **41**, 761 (2002);
I. Carmeli et al., Isr. J. of Chem. **43**, 399 (2003).

Notes

On the Influence of Nonadiabatic Electronic Transitions on Surface Chemical Dynamics

Daniel J. Auerbach

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There is a growing body of indirect evidence that such important processes as vibrational energy transfer and certain chemical reactions on metal surfaces involve a large, possibly dominant participation of nonadiabatic processes involving electron-hole pairs. Recently, we have obtained direct evidence of nonadiabatic generation of electron-hole pairs in chemical processes at surfaces. Specifically, we detect electron emission from a low work function surface resulting from collisions of vibrationally excited NO molecules. Electron quantum yield is very high, ~ 0.1 . Unlike all existing measurements or theories of nonadiabatic processes at surfaces, we find the seemingly paradoxical result that the quantum yield decreases with increasing velocity of the incident molecules. I will also discuss recent results for HCl (and DCI) inelastic scattering from surfaces.

Notes

Detecting Hot Electron Flow with Metal-Semiconductor Schottky Diodes during Exothermic Catalytic Reactions

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Atomic or molecular processes at the metal surface can generate a pulse of hot electrons with kinetic energy of 1-3 eV, and mean free path on the order of ~10 nm. Detection of hot electron flow could allow us to understand the mechanism of electronic energy dissipation and the role of charge transport through the metal-semiconductor interface in oxide-metal heterogeneous catalysis. To probe and utilize the hot electron flow in catalytic reactions, we fabricated metal-semiconductor Schottky diodes. The electron flow is detected as a chemicurrent if the excess electron kinetic energy generated by the exothermic reaction is larger than the effective Schottky barrier formed at the metal-semiconductor interface. We measure chemicurrent and turnover rate simultaneously as a function of temperature under CO oxidation, and found that the turnover rate and the chemicurrent are correlated. The molecular level mechanism of hot electron generation will be discussed. Recent results on detection of hot electrons generated on colloid metal nanoparticles with a nanoparticle-diode hybrid system will be also presented.

Notes

Nanoscale Thermal Transport and the Thermal Conductance of Interfaces

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Rapid progress in the synthesis and processing of materials with structure on nanometer length scales has created a demand for greater scientific understanding of thermal transport in nanoscale devices, individual nanostructures, and nanostructured materials. In this talk, I will emphasize a critical aspect of this growing field: the thermal conductance of interfaces. We have recently advanced the state-of-the-art of time-domain-thermoreflectance (TDTR) measurements of thermal transport and are using TDTR to study i) heat transport across individual interfaces with extremely high and low thermal conductance; ii) the thermal conductivity of nanoscale multilayers and disordered layered crystals that circumvent the lower-limits imposed by the minimum thermal conductivity; and iii) the dramatic reduction in the thermal conductance of interfaces with water that are created by changing the functionality of terminal groups from hydrophilic to hydrophobic.

Notes

New Devices for Ballistic Electron Emission

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The purpose of our research project is to make a device on which we can modify the reactivity of adsorbed molecules and intermediates by interaction with hot electrons as proposed by Gadzuk [1]. The aim is to get ballistic electrons in the energy range 2-3 eV above the Fermi level and thereby to get interaction with the empty anti-bonding states. It is expected that that a negative ion resonance will enhance the reactivity of otherwise difficult or sluggish reactions.

In this presentation we shall present our progress so far. Macroscopic devices with areas varying from 10^{-6} ... 1.0 cm^2 has been manufactured and tested for electron emission across a 5nm oxide barrier into a 5nm metal overlayer of gold or platinum. The emission of the devices is well understood [2]. By characterizing the emission under UHV conditions, it is possible to check the surface cleanliness by XPS and ISS and measuring the energy distribution of the emitted electrons into vacuum for higher energies than the 3 eV mentioned above. The latter is for example done as a function of modification of the surface with alkali metals. The cleanliness of the surface is not surprisingly very important and has strong impact on the obtained results. The progress of observing enhancement of reactivity under various conditions by this hot electron device will also be discussed.

[1] J. W. Gadzuk, Physical Review Letters **76**, 4234 (1996).

[2] L. B. Thomsen et al., Phys. Rev. B **76**, 155315 (2007).

Notes

Chemical Dynamics at Metal Surfaces: Electronic Excitations

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At metal surfaces nonadiabatic behavior is the rule rather than the exception. Electron-hole pair transitions, charge transfer and hot-electron-induced motion can be dominant pathways for energy flow. Recent experiments have demonstrated that molecular vibrational energy and reaction exothermicity can produce highly excited electrons, even resulting in electron emission. The conventional “molecular dynamics with electronic friction” (MDEF) theory, while quite successful in describing energy loss to electron-hole pairs, cannot encompass these recent observations. MDEF is based on a weak coupling assumption, resulting in single-quantum vibrational transitions; i.e., several weakly excited electrons rather than a single highly excited electron. In this paper we describe progress towards a unified theory of nonadiabatic dynamics at metal surfaces that is applicable beyond the weak coupling regime. We present initial applications to multi-quantum vibrational-to-electronic energy transfer in scattering of highly vibrationally excited nitric oxide from a gold surface, with comparison to experiment.

Notes

Simultaneous Spatial and Temporal Control of Nanooptical Fields

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Using time-resolved two-photon photoemission electron microscopy (TR-PEEM) we demonstrate simultaneous spatial and temporal control of nanooptical fields. For this purpose, we combine femtosecond polarization shaping and time-resolved two-photon photoemission electron microscopy on planar silver nanostructures manufactured by e-beam lithography. The experiments show that polarization pulse shaping can be utilized to manipulate the local field distribution in the vicinity of a nanostructure. Furthermore, the time-resolved cross correlation traces for different regions of the nanostructure show a clear variation of their relative intensities in the manually set time span of 400 fs within the polarization shaped excitation pulse. This confirms that the polarization-shaped incident laser pulse does indeed switch between two different excitation patterns within a time scale that can be controlled almost freely and is limited only by the spectral bandwidth of the used coherent light source.

Notes

The Dynamics of Elementary Excitations in Carbon Nanotubes and Graphene

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Carbon nanotubes and graphene represent the one- and two-dimensional forms of sp^2 -hybridized carbon in which every atom is at the surface. In this paper, we will review our current understanding of optical excitations in carbon nanotubes and graphene and of the pathways by which ultrafast relaxation and energy dissipation occurs in these systems.

Recent advances have permitted detailed investigations of both individual carbon nanotubes and monolayer graphene samples. Electronic transitions can be probed by optical absorption and scattering measurements, while the vibrational excitations are accessible using Raman spectroscopy. We will describe the understanding of the elementary excitations of these systems and their dynamics that emerges from these studies, as well as from characterization in the time-domain based on ultrafast spectroscopy.

Notes

Interface Controlled Epitaxy of Thin Organic Films

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In order to optimize the electrical contacts between organics and metals and improve a charge injection in organic electronic devices we need to understand the fundamental mechanisms that control both, the morphology and the electronic structure at the interface between metal and organic material. In general, the interfaces in organic films grown on variety of substrates are poorly ordered, but recent studies have revealed that organic films can grow epitaxially on metallic and semimetallic surfaces. We utilized a low-energy electron microscopy (LEEM) to understand the nucleation mechanisms and growth of organic films on semimetallic Bi(0001) surface. Our results confirm that the Bi(0001) makes a surprisingly good template for growing epitaxial, highly crystalline, bulk-like organic films, including pentacene and fullerenes. We concluded that the growth mode is determined by a competition between the interaction strength between organic molecules in the film, and between molecules and the substrate. Thus, careful substrate engineering may provide a pathway to the fabrication of organic devices with optimum performance characteristics.

Notes

Studying Irreversible Transient Phenomena on the Nanosecond and Nanometer Scale in the Dynamic Transmission Electron Microscope

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The use of pump-probe methods to investigate materials dynamics has typically utilized laser induced reactions/transitions coupled with either laser or X-ray interrogation methods. However, electron interrogation methods have the potential to provide higher sensitivity and resolution by utilizing bright electron sources, the ability to control the electron beam and the stronger interaction of electrons with matter. The potential for direct high spatial resolution imaging with ultrafast time resolution has recently been demonstrated with the single-shot dynamic transmission electron microscope (DTEM) concept employed at Lawrence Livermore National Laboratory (LLNL) on a JEOL 2000FX TEM. The important aspect of this approach is that one pulse of electrons is used to form the whole image, allowing irreversible transitions and cumulative phenomena such as nucleation and growth, to be studied directly in the microscope. Projects currently underway include determining the active sites in catalytic systems, the dynamics of phase transitions in materials, the defect interactions in irradiated metals, the mechanism of damage in laser machining, and the observation of many fundamental biomolecular processes. The operation of the DTEM, its current and potential future spatial and temporal resolution, and examples of materials applications will be presented.

This work is performed under the auspices of the U.S. Department of Energy, Office of Basic Energy Sciences by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344.

Notes

Electron Excitation and Emission for Grazing Impact of Fast Atoms on Metal and Insulator Surfaces

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Fast atoms with energies in the keV domain are scattered under a grazing angle of incidence from clean and flat metal and insulator surfaces. Making use of the coincident detection of the number of emitted electrons (biased surface barrier detector) and the dissipated projectile energy (time-of-flight method) provides clear-cut information on the relevant electronic excitation mechanisms. From our studies we can identify different interaction scenarios which explain the at first sight surprising feature of higher total electron yields for impact of fast atoms/ions on insulator targets compared to metals. Key features are the formation of transient negative ions for insulators and head-on binary collisions for metals concerning electronic excitations. We will show that details on the electronic surface potential for metals can be deduced from the threshold behaviour of electron emission. The pronounced dependence of electron emission on the azimuthal orientation of the target can be applied for ion beam triangulation studies on superstructures at surfaces of thin films and for controlled adsorption of atoms.

Notes

Electron Dynamics at Surfaces at High Photon Energies Using the XUV Free Electron Laser (FLASH)

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Recently, the XUV Free Electron Laser at Hamburg (FLASH) started operation and thereby provides intense pulses in the XUV and soft x-ray spectral region with femtosecond pulse duration. This makes high-lying electronic states of systems accessible or facilitates the participation of core levels in the reaction dynamics. Using photon energies of 38 eV and 57 eV we investigated the reaction and desorption of neutral H (D) atoms and of NO molecules from graphite surfaces, with internal state selective detection of products. For hydrogen direct atomic desorption is observed and the velocity distribution measured. For NO kinetic energy and rovibrational distributions have been measured and will be discussed. Besides these neutral products also various positive ions have been detected, with high kinetic energies. For NO and some positive ions a nonlinear desorption yield is observed, enabling in the future to observe two-pulse correlated reactions in the XUV.

Such a two-pulse correlator for the FLASH beam line has recently successfully been developed and tested by us, which allowed for the first time to measure the coherence time (7.5 fs) and pulse duration (about 35 fs) of the XUV pulses at 52 eV by linear and nonlinear autocorrelation, respectively.

Notes

Hot Spots and Spin Waves

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Recent experiments demonstrate that significant demagnetization of thin films of itinerant ferromagnets can be achieved within a few hundred femtoseconds upon optical excitation. Within this timescale, the excited electronic system and the underlying lattice are not in equilibrium and it seems that the transient hot electron population is responsible for the change of the magnetization. It remains controversial to date, which microscopic processes are fast enough to provoke femtomagnetism. To approach these problems we combined time-, angle- and energy-resolved two-photon photoemission with spin-resolved electron detection and investigated ultrathin iron and cobalt films on Cu(100).

In purely non-relativistic electron-electron or electron-phonon scattering a spin-flip is not possible because the Coulomb operator does not act on the spin part of the electron wave-function. Electrons only undergo a spin flip in the presence of spin-orbit coupling significantly enhanced at hybridization points in the band structure. We have identified these so-called spin hot-spots by linear magnetic dichroism. Initial bulk and surface states with minority spin-character are the source for the dichroic intensities and the apparent dichroic lifetimes of the image-potential states. Excellent agreement with *ab initio* fully relativistic calculations of the cobalt fcc band-structure allows us to precisely determine spin-orbit hybridization points close to the Fermi level.

Experimental access to the spin-dependent relaxation processes of low-energy electrons in *d*-band ferromagnets proves a challenge, as the dynamics occur on the femtosecond timescale and it is difficult to distinguish between refilling and the various scattering processes. For these purposes the well-defined dispersing image-potential-state electron can be employed as test charge or primary electron. While at the minimum of the image-potential band the decay rate is determined by the strongly spin-dependent density of *d*-states, the increase of the decay rate with energy above the band bottom is governed by spin-independent states only. We observe twice the increase in decay rate for minority-spin electrons than for majority-spin electrons on thin iron films. The factor of two is explained if electron magnon scattering is included. Then minority-spin electrons may also scatter into the majority-spin bands via magnon emission and thus gain twice the phase space of their majority-spin counterparts. These magnon-enhanced electron scattering processes allow for transfer of angular momentum of hot electrons on a femtosecond timescale.

Notes

The Excitation of Local Vibrations by an Electron Current: From Atomic Manipulation to Local Heating

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When an electron current is established through an atomic system, energy can be transferred to the ionic degrees of freedom, leading to vibrational excitation. In the case of a molecule chemisorbed on a surface under the tip of a scanning tunneling microscope (STM), local vibrations can be excited leading to control evolution of the molecular atoms. This has been shown for CO [1] and NH₃ [2] on metal surfaces. However, molecular control can only be achieved if a substantial intermode coupling is operational. I will show theoretical results on how to evaluate the intermode coupling, leading to a rationalization of the experimental findings [3]. Dehydrogenation of benzene has also been achieved by tunneling currents, however, our calculations show that the electron induced reaction correspond to a regime different from the vibrationally excited one [4].

- [1] T. Komeda, et al., *Science* **295**, 1055 (2002).
- [2] J.I. Pascual et al., *Nature* **423**, 525 (2003).
- [3] N. Lorente and H. Ueba, *Eur. Phys. J. D.* **35**, 341 (2005).
- [4] H. Lesnard et al., *J. Am. Chem. Soc.* **129**, 4298 (2007);
H. Lesnard et al., *J. Phys.: Cond. Matt.* June 2008.

Notes

Ultrafast Electron Dynamics and Electron-Phonon Coupling in Metals and Highly Correlated Materials

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Many physical properties of materials are governed by electron-phonon coupling, which gives rise to the temperature dependence of the electric conductivity in metals, the formation of charge density waves (CDW) in correlated materials or pairing in BCS superconductors. When an ultrashort laser pulse induces a non-equilibrium in the material the response and relaxation of the excited electrons and their equilibration with phonons occurs typically on different timescales.

Here we employ femtosecond time-resolved photoemission spectroscopy (TR-PES) to investigate the time evolution of electronic structure in conventional metals as well as correlated materials like Mott insulators or high T_c cuprates. Femtosecond TR-PES provides direct access to the cooling of photoexcited electron distributions in Ruthenium and Gadolinium [1]. These dynamics can be nicely described by a modified two-temperature model, which includes the contribution of non-thermalized hot electrons.

For the Mott insulator $1T\text{-TaS}_2$ we demonstrate that the photoinduced insulator to metal transition is driven directly by electronic excitation as revealed by the instantaneous collapse of the electronic gap [2]. A coherently excited lattice mode results in a periodic shift of the spectra (charge density breathing mode), which is lasting for 20 ps without perturbing the insulating phase. These findings clearly demonstrate that the metal-insulator transition in TaS_2 follows a Mott-Hubbard and not a Peierls-type mechanism

In the high T_c cuprate Bi-2212 we observe an ultrafast (<50 fs) electron thermalization and cooling of the electronic temperature on two distinct timescales. This observation of a bottleneck in the energy flow suggests that only a minor subset (20%) of all phonon modes contribute to the electron-phonon coupling with an interaction strength which is strongly anisotropic but weak [3]. Finally, I will briefly address some recent work on TbTe_3 , where we study the anisotropic electron dynamics in this CDW compound.

- [1] M. Lisowski et al., Phys. Rev. Lett. **95**, 137402 (2005);
M. Lisowski et al., Appl. Phys. A, **78**, 165 (2004).
- [2] L. Perfetti et al., Phys. Rev. Lett. **97**, 067402 (2006);
L. Perfetti et al., New J. Phys. (in press).
- [3] L. Perfetti et al., Phys. Rev. Lett. **99**, 197001 (2007).

Notes

Electronic Energy Dissipation of Fast Charged Particles in Matter

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The physics of the energy transfer from fast moving charged particles to solid matter is discussed. Starting from the basic ion-atom collision processes, the projectile-state variation is considered briefly including charge-state equilibration and primary energy-loss events. Important modes of the subsequent electronic energy dissipation are explained and special attention is devoted to the case of dense electronic excitations.

Notes

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